Radioactive Waste Products 2002

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OPENING
Opening Address
Dr. Gerd Eisenbeiß
Member of the Board of Directors, Research Centre Juelich GmbH

First of all I welcome you very warmly here at Würzburg, a very beautiful baroque city located at the river Main and surrounded by vineyards.

I especially welcome our guests from abroad, from overseas. It is pleasure for me to open the 4th International Seminar on Radioactive Waste Products initiated by the Research Centre Jülich. Co-organizers are the Bundesamt für Strahlenschutz (BfS), Kerntechnische Gesellschaft (KTG), Gesellschaft Deutscher Chemiker (GDCh), European Network of Testing Facilities for Quality Checking of Radioactive Waste Packages (EN-TRAP), Commission of the European Union (CEC) in co-operation with Environmental Protection Agency (EPA), Washington DC.

This large number of institutions involved indicate the interest in the topic of this seminar for all countries with nuclear facilities.

Although in Germany by the Atomic Energy Act of this year the government decided to terminate the industrial nuclear energy production in twenty years, there still exists waste and even decommissioning of nuclear power plants after operation will produce radioactive waste which had to be treated and brought to disposal.

In about 10 years the exploration of an underground repository is planned. The start of the operation of the repository is envisaged for 2030. Up to this date all radioactive wastes have to be conditioned in a proper way for safe interim storage and final disposal. Since 1985 the Research Centre Jülich has hosted by contract the quality control group of BfS. After developments and installation of destructive and non-destructive test methods mainly for old waste packages nowadays new conditioning of generated radioactive waste is regulated by Process Control Quality Plan called "Sequence-of-Events-Plan" or Process Qualification Manual. This quality control in nuclear waste management implies more safety which is requested by government and public.

Development and conceptions of safety technology and safe working procedures, research and development devoted to minimisation of risks, including human errors, are typical examples of the research area of the Research Centre Jülich.

By the way, let me just add a few words concerning the Research Centre Jülich which was founded 1956. This Research Centre is member of the Helmholtz Association (HGF). With its 15 research centres and an annual budget of about 2.1 Billion Euro the HGF is Germany’s largest research pool.

Nuclear safety research including the issue of waste treatment and final disposal is part of the Helmholtz area "Energy research" together with the 3 other programmes dealing with

- renewable energies
- efficient energy conversion and
- nuclear fusion.

The Research Centre Jülich contributes with its interdisciplinary research structure to all four programmes.

Summarising, I think energy is a very topical subject. Safe and sustainable energy production is an international challenge. I hope this seminar will bring us towards a common understanding of the nuclear safety aspects and to some progress for better solutions, including minimising the risk of nuclear waste management.
Welcome Address
Henning Rösel
Vice-President of the Bundesamt für Strahlenschutz, Salzgitter, Germany

The 4th International Seminar on Radioactive Waste Products was organised by the Forschungszentrum Jülich in co-operation with the Bundesamt für Strahlenschutz and the European Commission. On behalf of the Bundesamt, I would like to welcome all participants of this scientific-technical meeting. I very much appreciate the participation not only of numerous German scientists, engineers and technicians as well as governmental and industrial representatives, but would particularly express my gratitude for the participation of many colleagues from abroad.

In continuation of the meetings which were held in 1985, 1990 and 1997, the 4th International Seminar on Radioactive Waste Products will again provide an international and interdisciplinary forum for presentations and discussions on recent technical and safety-related developments in waste acceptance requirements, waste package quality and waste characterisation. In particular, the seminar will focus on measures and procedures for quality assurance and quality control of waste packages to be disposed of. This main topic of the 4th International Seminar is of great importance and relevance to all institutions and/or organisations being responsible for the operation of interim storage facilities and repositories. I like to point out that especially the safety of such facilities in their operational and post-closure phase is decisively determined by a proper waste package quality, thus meeting the respective waste acceptance requirements of the particular site.

Radioactive waste management and disposal is a worldwide issue and international co-operation to support national programmes is therefore much appreciated. The international organisations provide, among other things, guidance to member countries on safe, economic and environmentally acceptable solutions for radioactive waste disposal. On a national basis respective programmes are developed, modified or successfully realized. Nevertheless, the challenge of radioactive waste management and disposal is no longer a scientific and technical exclusivity. The importance of ethical and social aspects, the dialogue with the public and transparency in decision-making processes increase more and more. Thus, when addressing safety-related key questions one needs to be as open as possible on scientific-technical aspects and to consider the involvement of the public requiring a clear, open-minded and transparent communication.

Having this in mind, I really do hope that the 4th International Seminar on Radioactive Waste Products will present an excellent opportunity to discuss radioactive waste related topics on an international basis. The common understanding of technical and safety-related issues as well as the useful exchange of ideas and experiences including efforts to increase public acceptance of and confidence in radioactive waste management and disposal should be the highlights of this seminar.
Inaugural Address
1. Introduction

Nuclear technologies are providing more and more significant contributions to the protection of man and his environment and to the improvement of the quality of life. The International Atomic Energy Agency has received the statutory mandate from the United Nations to develop and support the nuclear technologies and encourage their use through the dissemination of information, the provision of training, of equipment and specialized services, and the elaboration and issuance of safety standards.

This lecture will briefly report on this role of the IAEA and further elaborate on the achievements and current developments of nuclear technologies in the following fields:

- Nuclear medicine and human health
- Animal health and livestock managing
- Soil sciences and fertilizers
- Water resources and protection of the environment
- Plant breeding
- Sterile insect eradication of pests
- Various other applications as diverse as the removal of landmines and the management of the cultural heritage

2. The role of the International Atomic Energy Agency

The mission of the IAEA with respect to nuclear technologies is to contribute to sustainable development and protection of the environment by the use of nuclear sciences and their applications in human health, food and agriculture, industry, and the management of water resources. Those activities of the IAEA focus on the basic human needs and are subject to the restriction that the promoted nuclear technologies should have significant and competitive advantages over current alternative technologies.

Nuclear technologies are developed by the IAEA in the framework of coordinated applied research projects, which combine over several years the research contributions and achievements obtained by potential users of the technology in the industrial and developing world.

Available and mature technologies are transferred to and implemented in interested countries through technical cooperation projects, where the IAEA provides technical infrastructure and equipment, as well as training in the operational and safety aspects. The diversity of competitive nuclear technologies and variety of their application fields is a strategic strength for their future and also for the role of the IAEA as their promoter.

When focusing on opportunities to serve basic human needs, the following major problems can be identified: the world population reached about 6 billion people and increases every year by 80 million, which is equivalent to the total population of Germany. One billion people suffer from chronic malnutrition and from the lack of safe supplies of water. Every year 6 million
children in third world countries are dying from malnutrition and about the same number of adults are dying from cancer in the industrialized world. Further, every year, millions of people are affected and die from bacterial, parasite and viral infections. Malaria alone kills two million people annually all over the world. Human actions are polluting the environment and wasting natural resources, and many of them are in the longer term not sustainable.

The nuclear applications developed and supported by the IAEA outside the nuclear energy sector address therefore essentially

- the enhancement of a sustainable supply and security of food;
- the management of secure and renewable water resources;
- the improvement of human health;
- the protection of the environment through identification, diagnosis, prevention and remediation of sources of environmental pollution.

With time the priorities of those fields have been changing as can be seen in a survey of the assistance provided to IAEA Member states in the framework of the Technical Cooperation programme, displayed in Table 1.

3. Nuclear applications for improving human health

The most widespread diagnostic applications of ionizing radiation and nuclear techniques are the X-ray imaging in diagnostic radiology and the radioactive biological tracers in nuclear medicine. The dominant field for the therapeutic application of radiation in health is radiation oncology, which involves the delivery of curative radiation doses to targeted volumes of cancer affected locations in the patient’s body. This is usually done from an external source positioned outside the patient in teletherapy applications or by implanted sources in brachytherapy applications. Open sources, such as radioactive substances, have been frequently applied in the past for the treatment of thyroid cancer, but currently many new therapeutic applications of radioactive substances for specific treatments are emerging. An example is the development of Rhenium-188 labeled peptides for the treatment of liver cancer.

Computer assisted tomography has been a revolutionary development in diagnostic radiology and allows to display cross sections of selected body parts with their full anatomical and functional details. Nuclear medicine is applying radioisotopes in more than one hundred standardized methods for diagnostic, therapeutic and research purposes in the fields of oncology, endocrinology, cardiology, neurology and nephrology. Industrialized countries currently operate about 20 gamma cameras per million inhabitants, compared with a figure of less than one gamma camera available for a million inhabitants living in the large agglomerations of developing countries.

Molecular nuclear medicine is an emerging field of applications that deals with the imaging of a disease at the cellular or genetic level rather than at the gross level. Polymerase chain reactions are extremely sensitive diagnostic tools and can detect one abnormal cell in a million. They are used for the diagnosis of infectious diseases, cancer or syndromes and as of 2001, more than 25 in vitro tests have been licensed by the US Food and Drug Administration.

Radio-immuno-assay is one of the most important components of in vitro diagnostic techniques for the quantification of changes in proteins as well as other intermediate metabolites in disease
states. Worldwide sale revenues for those in vitro diagnostic products are expected to continue to increase from 20 billion dollar in 1999 to over 26 billion dollar in 2004.

Cancer is a major cause of death in industrialized countries, and the number of cases in developing countries is growing rapidly as life expectancy increases. About one half of the 10 million new cancer cases in the year 2000 occurred in the industrialized world and one half in the developing world. In 2015, it is projected that two thirds of the 15 million expected new cancer cases will occur in third world countries. Currently about 50 % of all cancer patients are requiring radiotherapy, and the survival rate of those receiving curative radiotherapy, possibly in combination with surgery or chemotherapy, in developed countries is around 45 %.

Teletherapy, using an external radiation source is most commonly applied, nowadays with advanced instruments, which deliver radiation doses with great accuracy to highly irregular tumor shapes while sparing the surrounding healthy tissue. In brachytherapy, with sources placed inside the body, implanted caesium-137 and iridium-192 sources are now replacing the radium-226 sources used in the past. Together with the world health organization, the IAEA is maintaining a registry of hospitals and clinical institutions that offer radiation therapy using radionuclides or high energy teletherapy equipment. Developing countries with 85 % of the world population have about third of the radiotherapy installations distributed over 2200 teletherapy machines, mainly with a cobalt-60 source, and 850 brachytherapy units. They have only one fifth of the electron accelerators dedicated to radiotherapy. Each installation can treat about 600 patients per year, which results in capacity for the treatment of 1.9 million patients of the total of 2.5 million patients, which would require and benefit from radiation treatment. To follow the expected demographic and cancer developments more than 5000 additional radiotherapy devices will be needed in the year 2015.

It is also worthwhile to mention that the IAEA, in cooperation with the WHO, is running a quality assurance program for radiotherapy installations with a capacity to test every year the operational protocols and the correct delivery of the prescribed doses in more than 600 hospitals in the third world.

Nuclear techniques using stable or radioactive isotopes are applied for diagnosis and research in the improvement of nutrition, in particular to identify deficiencies in vitamins, trace elements and micronutrients, which are the cause of anemia, growth retardation, blindness and impaired mental development. Techniques based on differential X-ray absorptiometry are currently applied as non-invasive methods for bone mineral density measurements in monitoring the evolution of osteoporosis.

4. Agriculture

Nuclear technologies are used in agriculture to improve and secure food supplies, to optimize livestock management, to prevent diseases and to eradicate pests. Biodiversity is the result of random mutations in living species, induced by cosmic and natural radiation sources and by copying errors in the genetic material. Applying radiation therefore can enhance and accelerate the occurrence and frequency of mutations to produce a lot of new plant variations in a relatively short period of time. Most of the induced mutations will lead to nonviable varieties or to varieties whose properties are less fitting to our needs than the original plants, but some mutations might show favorable characteristics and can be further cultivated and disseminated. A few examples will indicate the enormous impact this technology can generate in selected regions and individual countries. New rice species, fit to grow in alternating fresh and brackish
waters of the giant Asian river deltas constitute the basic nutrition for millions of people in Pakistan, China and Bangladesh. Huskless barley species, drought tolerant sorghum and rice, have been produced through radiation induced mutations and have been cultivated because they are better fitting to the geological environment and the prevailing climatic conditions in Peru or in Mali. Radiation induced mutations are most effective for the improvement of plants who are breeding through sterile cloning, such as bananas. In this way, an early flowering, delicious and pest resistant variety has been developed in Malaysia. The pest resistant species are not only producing greater yields, but they also require smaller quantities of pesticides or none at all, which results in lower market prices as well as a in cleaner environment. Bananas with an annual production of 100 million tons and rice are the main nutrition crops and agricultural products in the third world, but also the industrialized countries in the northern hemisphere are consuming large quantities of about two thousand products, whose characteristics have been improved or enhanced by radiation induced mutations. In Austria, the “Golden Haidegg” was developed as a luxury mutant variety of the Golden Delicious apple, with a longer shelf life, a brilliant color and absence of russetting. The current cultivation of mutant varieties of Japanese pears with radiation-induced resistance against the black spot disease needs only one fourth of the previously required quantities of pesticides.

Nuclear techniques are not only used for the improvement of foodstuffs, but also for their safety and to reduce post-harvest losses. In thirty countries, food irradiation is used to ensure safety and quality and to satisfy quarantine regulations in trade for some foods, including meat products, fresh fruits, spices and dried vegetable seasonings. Large commercial-scale irradiation of ground meat to ensure microbiological safety started in the USA in the year 2000 and the irradiated products have been widely sold in over 2000 supermarkets in 2001, apparently with no resistance from the consumers. Many other countries, especially those in Asia and the Pacific, in Latin America and in some Middle East and African countries could benefit immensely from using this technology to reduce the post-harvest losses, to control a number of food-borne diseases and to facilitate food trade. The Codex Alimentarius Commission, the worldwide authority for food safety has disseminated standard protocols, guidelines and recommendations to ensure the quality and safety of foods, including those processed by irradiation as well as those which may be contaminated by chemical and biological agents.

Radioisotopes are further used to increase livestock productivity by applying radioimmunoassay techniques for the analysis of hormones in milk, serum and plasma. The purpose is to optimize planning and diagnosis of pregnancy, to detect and correct reproductive health disorders, and for the timely identification and prevention of diseases, such as rinderpest pest, trypanosomosis, brucellosis or mouth and claw disease. With assistance of the IAEA, the veterinary laboratory in Dakar is distributing diagnostic kits to 34 African countries for the early detection of rinderpest and swine fever.

Nuclear technologies and isotopic tracer techniques of nitrogen, carbon and phosphorus are widely used for biological research on nitrogen-fixing bacteria. Similar techniques are applied to study the optimal use of fertilizers in terms of products, quantities and timing in a variety of climatic zones, geologies and nutrition cultures. They are also applied for minimizing the residual amounts of phosphates and nitrogen compounds in the environment and to assess the value of organic residues as sources of nitrogen for crops.

The eradication of pests using nuclear technologies is mainly based on the “sterile insect technique” or SIT. This technology consists of the large-scale production and systematic release
in the environment of radiation-sterilized male insects, such as fruit flies, Tsetse flies, screwworms and other pests. The released male insects behave in exactly the same way as the native species but are not producing any offspring. The next generation, already weakened by the treatment, is again treated with an injection of sterile males and this process is repeated until the complete eradication of the species, at least on a local or regional basis, is achieved. In contrast with the use of pesticides, this technique is extremely selective, eradicating only one single species without affecting the environment or disturbing the biological equilibrium. In particular the Mediterranean fruit fly is a notorious pest with a major economic impact and quarantine significance. The demand and production capacities for radiation-sterilized fruit flies have been increasing rapidly over the last decade from one billion flies to currently about three billion flies. The most successful fruit fly eradication programmes have been implemented in Central and Latin America, in the southern states of the USA, in Australia and in Japan. In 2001, a mass rearing facility capable of producing 12 million sterile male fruit flies per week was constructed with assistance of the IAEA in Sidi Thabet, Tunisia. Subtle genetic manipulations of the species allow the sterilization and selection of male insects in an early stage of development, such as the pupae. This technology, known as genetic sexing, results in reduced rearing costs and greater effectiveness of the released flies. This has allowed the application of SIT to reduce the fruit fly pests as an insecticide replacement rather than as a complete eradication tool. In the framework of an IAEA technical cooperation project a production plant for millions of sterilized tsetse flies has been built and this pest has been eliminated already from several regions in Africa. Further SIT applications are under development and operational projects are in progress against the apple codling moth in Canada, the cotton pink bollworm in the USA and date moths in northern Africa. The IAEA started an ambitious project for the eradication of Anopheles, one of the main malaria transmitting mosquitos. The economic benefits of this nuclear technology are significant. Maintaining Mexico free of Mediterranean fruit flies at an annual cost of about 10 million dollar has protected fruits and vegetables having a domestic and export market value of over one billion dollar. Similar results have been obtained in Chili, with the additional advantage that citrus fruits are produced there during wintertime in the Northern hemisphere, and exports to the industrialized world are booming.

5. Hydrology

Nuclear and isotopic tracer techniques are being used also in the management of water resources and climate studies. The natural fractionation and isotope enrichment processes mark the water bodies with distinctive isotopic fingerprints, providing information on the origin and age of the water. Isotopic techniques, based on the measurement of stable and radioactive nuclides in the water components, are used to model natural water systems. This is of particular interest in regions where water resources are scarce and valuable and the equilibrium between water consumption and renewal rates is essential for survival. Similar methods are used for identifying and tracing unwanted contamination of freshwater supplies with sewage water, as it frequently occurs in large metropolitan areas in developing countries.

The temperature dependence of the isotopic composition of water at the evaporation or condensation stages is used world-wide for studying climatic changes and greenhouse effects and thousands of years of climate history have been reconstructed on the basis of isotopic analyses of glaciers and Antarctic ice cores.
Examples to illustrate the economic and humanitarian importance of nuclear techniques in hydrology are easily found:

- the drinking water deficit of 260 million liter per day in Venezuela has been reduced by 30 % as a consequence of the exploitation of 50 new water wells, which have been located by isotopic techniques;
- isotopic measurements in groundwater in the Nile valley and in the Egyptian and Ethiopian deserts pointed to the presence of renewable water reserves in Nubian sandstone layers and allowed to increase the water supplies by 20 %;
- a suspected dam leakage in Venezuela could be refuted on the basis of isotopic measurements and resulted in saving 6 million dollar, which was the estimated cost for repair works on the dam;
- the exploitation of geothermal energy resources, supported and optimized by isotope hydrology techniques, results in millions of dollars of savings from oil imports in Central American and South East Asian countries.

6. Protection of the Environment

Nuclear technologies and measurement methods are widely used for environmental monitoring and research. Natural and artificial radionuclides in the atmosphere, the hydrosphere and the lithosphere are continuously monitored of course for radiation protection purposes. They are, however, also outstanding indicators for atmospheric and marine transport processes. They are excellent tracers for the behavior and transport of non-radioactive pollutants, for ecological and biological studies, and they are accurate timers for geochronological age determinations.

Starting from portable X-ray fluorescence equipment using isotopic sources to nuclear reactors for neutron activation analysis and up to giant accelerators and synchrotrons for the characterization of microscopic particles, we have at our disposal an impressive arsenal of nuclear techniques to search for harmful substances in the environment, for the accurate determination of their composition, and hence for the identification of their sources.

Nuclear techniques, however, are not only used for diagnosing harmful substances in the environment, but also for the prevention of their releases or the elimination of their consequences. In several countries, electron accelerators are installed in coal-burning power plants for the treatment of the off-gases. They destroy the sulfur and nitrogen oxides and induce them to react with added ammonia in order to produce clean effluents and a useful fertilizer as a byproduct. Electron beam technology has been demonstrated to be very effective for the destruction of toxic volatile organic compounds in off-gases, in particular at low concentrations. The key advantage of radiation treatments is that the toxic compound is eliminated and not merely transferred from one medium to another, such as is the case for scrubbers or absorbing media. This also applies to sewage waters, industrial wastewaters and sludge loaded with harmful toxic organic substances, which are treated with radiation sources and decontaminated or disinfected.

Nuclear techniques are also applied for restoring our environment from intentional and, regrettfully, sometimes criminal introduction of harmful substances and devices. About sixty million abandoned landmines in sixty-two countries each year cause thousands of deaths and injuries, mainly civilians including a large proportion of women and children. Nuclear techniques have been developed for the detection and identification of land mines. Equipment
currently available uses in-situ neutron activation combined with gamma ray spectrometry to measure concentration ratios of light elements, in particular hydrogen, carbon, nitrogen and oxygen for the detection of explosives in suspicious shallow-buried objects.

More recently, the large-scale application of electron accelerators is being considered for the irradiation of mail to destroy toxic substances such as botulism and anthrax spores.

7. Industry and Archeology

A wide variety of nuclear applications and techniques is also being used in non-nuclear industries, for example:

- transport- and construction companies using radiography for the examination of materials and structures,
- processing industries using isotopic sources for level and thickness gauging, neutron scattering for moisture measurements, and radiation induced polymerization, for example in the production of car tires,
- medical equipment suppliers using radiation sterilization for the elimination of viruses, bacteria and fungi,
- and many more, which cannot be covered within the scope of this presentation.

Interesting and maybe little-known applications of nondestructive nuclear techniques have been developed for archeological research and for the preservation of the cultural heritage. In Latin America, the IAEA recently coordinated an international project in this field by the promotion of interdisciplinary research, including several nuclear techniques. Each country participating in the project has been addressing unresolved questions with respect to ancient ceramics and potteries. Using a combination of X-ray fluorescence, neutron activation and particle accelerator techniques for trace analysis in the objects of interest, satisfactory answers could be obtained for many questions regarding regional archeological and historical issues.

I cooperation with Polish, Cuban and Austrian scientists, the IAEA developed a portable X-ray fluorescence device which has demonstrated an excellent performance for the non-destructive examination of objects of art. It is equipped with a laser pointer and computer steering and data processing unit, which enables it to produce within minutes a complete analysis of peculiar details, only a few square millimeters in size, on paintings, sculptures, polychromes, ceramics and coins. A prototype has been successfully been used in the Vienna Museum of Historical Arts for the authentification of 16th century paintings and the categorization of old Etruscan bronzes and coins.
8. Conclusions

Nuclear technologies, isotopic techniques and radiation applications are widely disseminated and provide an irreplaceable contribution to improve and sustain the current quality of life and to protect mankind and the environment. The extent of those applications for diagnostic and therapeutic purposes in the field of human health has been illustrated and the achievements and expectations based on current developments in computer tomography and molecular nuclear medicine have been described. A large increase in therapeutic applications is expected in particular in third world countries. Applications in agriculture, nutrition and food safety have reached maturity and a high level of public acceptance. They will continue to contribute to the improvement of crops, the eradication of pests, the prevention of diseases and the preservation of food, and they are now expected to provide solutions to more complex problems, such as malaria, trypanosomosis and myasis. Nuclear and isotopic techniques have already a long tradition and recognition in the management of water resources and the protection of the environment, and will be further developed to support climate studies and research in marine and terrestrial ecology.

It is worthwhile to remember again that the nuclear technologies in the fields described in this presentation have not been developed as scientific or fashionable curiosities, or because they were expected to receive preferential financial support. They have been developed and are still being used because in a comparative cost and benefits assessment, they consistently display convincing advantages with respect to the available alternatives.
TABLE 1

IAEA Support to Technical Cooperation Projects
(Percentage of Total Expenditures)

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References

International Overviews
RADIOACTIVE WASTE MANAGEMENT IN GERMANY

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Abstract

The Federal Republic of Germany intends to dispose of all types of radioactive waste in deep geological formations. This waste comprises spent fuel elements, vitrified fission product solution, nuclear power plant operational and decommissioning waste as well as spent sealed radiation sources and miscellaneous waste originating from small waste generators. The Atomic Energy Act gives the responsibility for the disposal of radioactive waste to the Federal Government with the Bundesamt für Strahlenschutz (BfS - Federal Office for Radiation Protection) as the legally responsible authority.

The Federal Government has made a pronounced change in energy policy since 1998, the most important feature of which is the abandoning or phasing out of nuclear energy. It is intended to irreversibly phase out nuclear energy use for electricity generation. Essential (basic) steps are the agreement which was achieved by the Federal Government and the utilities on June 14, 2000, and signed on June 11, 2001, and the April 2002 amendment of the Atomic Energy Act.

1. Introduction

Since the early sixties the German radioactive waste disposal policy has been based on the decision that all types of radioactive waste (short-lived and long-lived) are to be disposed of in deep geological formations.

The 1976 amendment of the Atomic Energy Act provided the legal basis for the disposal of radioactive waste. According to section 9a, disposal of radioactive waste was assigned to the Federal Government as a sovereign task with the Physikalisch-Technische Bundesanstalt (PTB-Federal Institute for Metrology) as the competent authority. On November 01, 1989, this competency was transferred by law to the Bundesamt für Strahlenschutz (BfS - Federal Office for Radiation Protection). Thus, BfS is now responsible for the establishment and operation of a repository, acting on behalf of the Federal Government. In this respect BfS is supervised by the Bundesministerium für Umwelt, Naturschutz und Reaktorsicherheit (BMU-
Federal Ministry for the Environment, Nature Conservation and Nuclear Safety) which, among other things, is responsible for nuclear safety and radiation protection in the Federal Republic of Germany.

All other radioactive waste management activities, e.g. spent fuel storage, reprocessing, conditioning, transportation and interim storage are within the responsibility of the waste generators. The Federal States must establish and operate regional depots for the interim storage of radioactive waste originating, in particular, from radioisotope application in industry, universities and medicine.

2. **Radioactive waste management**

A broad variety of different waste types and amounts are generated in Germany. At the end of 1999, an amount of approx. 33,000 m$^3$ of radioactive residues and of approx. 64,000 m$^3$ of conditioned radioactive waste with negligible heat generation (i.e., low-level waste (LLW) and intermediate-level waste (ILW)) was stored in engineered storage facilities. Due to a forecast into waste amounts to be expected in future, approx. 303,000 m$^3$ of conditioned waste with negligible heat generation and approx. 22,000 m$^3$ of conditioned heat-generating waste (i.e., high-level waste (HLW) and spent nuclear fuel (SNF)) are expected up to the year 2080.

**Current issues**

As a result of the 1998 federal elections a coalition of the Social Democrats and Alliance '90/The Greens came into power. The political aims of the Federal Government are given in the coalition agreement dated October 20, 1998. With respect to energy policy the most important feature is the abandoning or phasing out of nuclear energy. Thus, the Federal Government makes a pronounced change compared to the previous energy policy since 1998. It is intended to irreversibly phase out nuclear energy use for electricity generation. This has been performed in a stepwise procedure: in a first step so-called consensus talks with representatives of the utilities took place and in a second step respective legislative measures were taken.

The basic document on the future use of nuclear energy for electricity production in Germany was initialled on June 14, 2000, and signed on June 11, 2001. According to this document, the Federal Government and the utilities agree to limit the future utilization of the existing nuclear power plants. The most important agreements refer to operational restrictions. For each installation the amount of energy it may produce is calculated from January 01,
2000, until its decommissioning. In total, about 2,620 TWh (net) can be produced. According to this, the time of operation of a nuclear power plant amounts to 32 calendar years on average, starting at the beginning of commercial operation. The new policy is enforced by the latest amendment of the Atomic Energy Act which became effective on April 27, 2002.

New developments in spent fuel management

Up to now, spent nuclear fuel is either shipped to the French and British reprocessing facilities, stored on-site at the nuclear power plant or at centralized off-site interim storage facilities. HLW originating from reprocessing which is returned to Germany is stored at the Gorleben facility, too.

According to the agreement between the Federal Government and the utilities and to the April 2002 amendment of the Atomic Energy Act, the management of spent fuel will be restricted to direct disposal. Up to July 01, 2005, transports for reprocessing will be permissible. In addition, the nuclear power plant operators must provide interim storage facilities on-site. Subsequent to July 01, 2005, spent nuclear fuel may only be transported if no licensed interim storage capacity exists at the nuclear power plant site and if the operator of this site is not responsible for this situation.

As a consequence the utilities will construct and operate new engineered storage facilities at the sites of the nuclear power plants or near them. As far as necessary, additional storage places will be created on-site to bridge the time (up to 5 years) for licensing and erecting the proper interim storage facilities. Until these new facilities have been brought into operation, the utilities may transport spent nuclear fuel to the centralized interim storage facilities as well as to foreign countries until reprocessing will discontinue. As a result of the intended termination of reprocessing as well as the construction and operation of dry spent fuel interim storage facilities on-site, the number of shipments of spent fuel elements will considerably be reduced.

Regarding on-site engineered storage, starting in 1999 and continuing until fall 2000, applications were filed to initiate licensing procedures for the construction and operation of 12 interim storage facilities and 5 storage places with capacities in the range of 120 tons heavy metal (HM) to 2,250 tons HM and activities in the range of $7.6 \times 10^{16}$ Bq to $2.7 \times 10^{20}$ Bq. BFS is the competent licensing authority. All licensing procedures require a public hearing most of which were successfully held in 2001. There were significant regional differences in public interest and opposition. The number of objections varied between less than 2,000 against
the storage facility at Brunsbüttel (Northern Germany) and more than 75,000 against the storage facility at Gundremmingen (Southern Germany). All together 250,000 objections were raised against the on-site interim storage facilities. In addition, a public participation for citizens of the Republic of Austria was carried out for the six interim storage projects located in the south of Germany with regard to transboundary environmental impact assessment.

The licenses for the interim storage places at Neckarwestheim, Philippsburg and Biblis were issued by BfS on April 10, July 31 and December 20, 2001, respectively, as the first licenses for on-site interim storage places in Germany. The first license for an interim storage facility was issued by BfS on November 07, 2002; this facility is located at Lingen. It is intended to issue all other licenses in 2003. Operation of all new interim storage facilities is expected by 2005.

Disposal-related aspects

In the Federal Republic of Germany all types of radioactive waste are to be disposed of in deep geological formations. Up to now, according to the 1979 German radioactive waste management concept, two sites have been considered for disposal:

- The abandoned Konrad iron ore mine in the Federal State of Lower Saxony has been investigated for disposal of radioactive waste with negligible heat generation, i.e. waste packages which do not increase the host rock temperature by more than 3 Kelvin (K) on an average (LLW, ILW). At a depth of 800 m to 1,300 m the emplacement of up to 650,000 m³ waste package volume was planned. A total activity in the order of 10¹⁸ Bq and an alpha emitter activity of about 10¹⁷ Bq are anticipated in this facility. The licensing procedure was started on August 31, 1982.

- The Gorleben salt dome in the north-east of Lower Saxony has been investigated for its suitability to host a repository at depths between 840 m and 1,200 m for all types of radioactive waste, mainly for heat-generating radioactive waste originating from reprocessing and spent fuel elements (direct disposal). The accumulated inventory of beta/gamma and alpha emitters is planned to be in the order of magnitude of 10²⁵ Bq and 10¹⁹ Bq, respectively. Site-specific investigations were started in 1979.

Since 1971, short-lived low and intermediate level radioactive waste with an alpha emitter concentration of up to 4.0 x 10⁸ Bq / m³ originating from the operation of nuclear power plants and the application of radioisotopes in research, medicine and industry in the former German Democratic Republic was disposed of in the Morsleben repository, an abandoned
salt mine located near the village of Morsleben in the Federal State of Saxony-Anhalt. Since
German unity on October 03, 1990, the Morsleben facility has the status of a federal reposi-
tory in the sense of section 9a of the Atomic Energy Act. From 1971 through 1998 radioac-
tive waste with a total volume of about 37,000 m$^3$ and about 6,100 spent sealed radiation
sources were disposed of. The total activity of beta/gamma emitters amounts to 9.1 x
10$^{13}$ Bq, that of alpha emitters to about 8.0 x 10$^{16}$ Bq. According to a September 25, 1998,
court order BfS has to immediately stop further radioactive waste disposal in the so-called
eastern emplacement field of the Morsleben repository. Thus, last waste emplacement op-
erations were carried out on September 28, 1998.

3. Approach to Radioactive Waste Disposal

Persuant to the coalition agreement and the agreement between the Federal Government
and the utilities, the German radioactive waste management and disposal concept is being
reviewed and will be adopted due to political decisions, new findings and specific evalua-
tions. According to the agreements the most important issues are:

- The previous radioactive waste management concept has failed with regard to its content
  and no longer has a technical basis. A national waste management plan for the legacy of
  radioactive waste will be developed.
- A single repository in deep geological formations is sufficient for the disposal of all types
  of radioactive waste. The disposal of HLW by the year 2030 is the political objective for
  the disposal of all types of radioactive waste.
- There are doubts with regard to the suitability of the Gorleben site. Therefore, its explo-
  ration shall be interrupted and further sites in various host rocks shall be investigated for
  their suitability. The licensing procedure for the Konrad repository project shall be termi-
  nated; the Morsleben repository shall be decommissioned.

According to the coalition agreement, activities to develop a new national waste
management plan have been initiated. This document will comprise main issues and
important boundary conditions for the new approach of the Federal Government to
radioactive waste management and disposal. About three years ago the BMU set up a
special project group for its preparation. According to the work hitherto performed, the waste
management concept (including the principles, radioactive residues, the waste management
task, waste conditioning and quality assurance/quality control, interim storage and disposal),
inventory and prognosis of residues as well as waste management planning have basically
been elaborated. The national waste management plan will be presented within this
legislative period.
Nevertheless, the emplacement of waste packages into deep geological formations is still the preferred option for the safe disposal of all types of radioactive waste. Otherwise there is scientific evidence that a separate emplacement of radioactive waste with negligible heat generation as compared to the co-location of all types of radioactive waste in one single repository may have special advantage from a safety point of view, e.g. with respect to gas generation in the post-closure phase. Thus, the political aim to construct and to operate one single repository is still to be examined in detail focusing on safety-related aspects, specific issues of the waste management concept, and economical considerations.

Safety criteria

In the Federal Republic of Germany the "Safety Criteria for the Disposal of Radioactive Waste in a Mine" describes the basic aspects which must be taken into account to achieve the objective of disposal. They qualitatively specify the measures to be taken to achieve the protection goal of disposal and define the principles by which it must be demonstrated that this goal has been reached. The Safety Criteria were issued in 1983 and are at present being revised on behalf of BMU. The overall aim of this revision may be outlined as follows:

- Survey of the international status of the development of safety criteria for the disposal of radioactive waste and its evaluation as compared to the German situation.
- Consideration of respective activities being performed by international institutions such as the International Atomic Energy Agency (IAEA) (e.g., the RADWASS programme and the Joint Convention on the Safety of Spent Fuel Management and on the Safety of Radioactive Waste Management), the Organisation for Economic Co-operation and Development/Nuclear Energy Agency (OECD/NEA) and the International Commission for Radiation Protection (ICRP).

Status of Gorleben, Konrad and Morsleben

Though the Federal Government has expressed doubts with respect to the suitability of the Gorleben site, it is not considered to be unsuitable and will be included in the future site selection process. According to the agreement between the Federal Government and the utilities a further exploration of the Gorleben salt dome can contribute nothing to clarify the doubts of the Federal Government. For this reason the underground exploration will remain interrupted for at least 3, but at most 10 years (Gorleben moratorium); the moratorium became effective on October 01, 2000. A rapid clarification of these doubts has been initiated focusing on 13 research projects and studies:
These investigations aim to the clarification of conceptual and safety-related issues and are envisaged to be finished by the end of 2004.

The licensing procedure for the Konrad repository project was finished. On May 22, 2002, the competent licensing authority Niedersächsisches Umweltministerium (NMU – Ministry for the Environment of the Federal State of Lower Saxony) issued the license. Subsequently, the license was handed over to the licensee BfS on June 05, 2002, and published in the Gazette of Lower Saxony on June 12, 2002. According to the agreement between the Federal Government and the utilities BfS withdrew the application for immediate enforcement of the license on July 17, 2000, thus enabling court examination of the license. This withdrawal in particular means that the re-construction of the Konrad mine into a repository for all types of radioactive waste with negligible heat generation will only be possible after final court decision. The court cases are assumed to last for about four years; then further decisions on the Konrad project will have to be taken.

The Morsleben repository will not resume emplacement operations. An application for the licensing procedure for decommissioning was already filed on May 09, 1997. The assessment of the safety in the post-closure phase is of special importance. The site-specific safety assessment was originally based on a preliminary backfilling and sealing concept; more detailed site specific information and respective safety analyses proved this concept not to be acceptable. Thus, two alternative concepts have been developed. The sealing concept and the concept on extensive backfilling are being combined and the final concept is expected by the end of 2002. BfS is concentrating its activities on the licensing procedure and the preparation of respective documents. Latest important issues comprise the backfilling of the so-called southern emplacement field with crushed salt from November 18, 2000, until March 09, 2001, and the statement of BfS dated April 12, 2001, that the Morsleben repository will definitely never again be used for radioactive waste disposal (renunciation of those parts of the Morsleben repository operation license dealing with the emplacement of radioactive waste). In addition, BfS intends to advance backfill measures in
the central part of the Morsleben repository in order to maintain geomechanical stability and integrity. It is planned to start these backfill measures at the end of this year.

The siting process for a repository

According to the new approach to waste management and disposal, further sites in various host rocks shall be investigated for their suitability. The final site shall be selected upon a subsequent comparison of potential sites, including the Gorleben site. Thus, BMU set up a special expert group to develop repository site selection criteria and respective procedures on a scientifically sound basis. The criteria and procedures aim at finding the relatively best suited site in different host rocks in Germany. Such a set of criteria was not available when the Gorleben site was chosen in the seventies.

The principle objective of the site selection procedure to be developed is to identify - with public participation - potential disposal sites in a comprehensible and reliable way. Step by step and based on criteria which have to be defined beforehand, those areas, site regions and eventually sites shall be selected that offer particularly favourable conditions for the later demonstration of the site's suitability and its confirmation in a licensing procedure.

Of utmost importance for any site selection procedure is the question at which step of the procedure a decision on the host formation(s) has to be made. According to the expert group it is less this host formation but rather the integral geological setting at a site that provides for the necessary long-term isolation of the waste and for the justified assumption that even thereafter there should be no unacceptable releases. Therefore, the development of a site selection procedure is pursued which does not start with the fixing of host formations, but rather attempts to get host formations as a result of a selection process which is based on general criteria describing an integral geological setting with favourable properties for the disposal of radioactive waste.

The advantage of this approach is obvious: it avoids the never-ending protest of people living in the vicinity of, say, a salt site praising the merits of granite, and vice versa. Based on these considerations, the present status of the site selection procedure's structure is as follows:
Step 1: Sorting out areas with potentially negative geological conditions.
Step 2: Identification of areas with potentially positive integral geological conditions.
Step 3: Sorting out areas with unfavourable societal conditions.
Step 4: Narrowing down to regions for which best favourable geological conditions can be expected.
Step 5: Narrowing down to sites where public acceptance for further site investigations can be achieved (local positive interest).
Step 6: Site investigations without mining activities.
Step 7: Evaluation of siting alternatives considering requirements from the safety/environmental viewpoint and local concerns.

The expert group has so far worked on steps 1 to 3. Results were presented and discussed in two workshops held in fall 2000 and 2001, respectively. The final workshop will take place in October 2002.

Public acceptance is considered to be of central importance for the success of a site selection procedure:

- The site selection procedure needs a clear and transparent structure and must be based upon well founded criteria in order that progress, fairness and objectivity of the procedure can easily be followed and respective decisions are understood in the general public.
- The evaluation basis and criteria associated with the selection procedure must be fixed beforehand to avoid decisions which the public may perceive as not sufficiently justified or even arbitrary.
- Public participation is indispensable from the very beginning and in all phases of the selection procedure, particularly before and during the definition of the “rules of the procedure”.

The time needed for the development of repository site selection criteria and respective procedures is estimated to be about 3 years. In February 1999 work started; completion of work is envisaged for December 2002. The recommended criteria and procedures will subsequently be discussed in detail (including stakeholders, environmental groups, other interested initiatives and the general public) and, after all, legally be implemented. During this criteria development phase, new sites will neither be selected nor investigated.
Provided that the site selection criteria and procedures will be available in 2002 and their discussion as well as implementation performed in 2003/2004, it may be possible to nominate potential sites, carry out respective investigations and finally select the site by 2010. In this case, for detailed site investigations, repository planning, licensing procedure and construction of the repository approximately 20 years would be available in order to start operation by 2030.

The ultimate goal of the site selection procedure to be developed is that it is accepted by virtually everybody as suitable and fair before it will be applied and local interests become effective.

4. Conclusions

Having the present radioactive waste disposal-related situation in mind, it is to be recognized that future developments and decisions will particularly be determined by the agreement between the Federal Government and the utilities and the April 2002 amendment of the Atomic Energy Act. Regarding the on-site interim storage of spent nuclear fuel, applications were filed, the licensing procedures are in progress and three licenses for interim storage places as well as one license for an interim storage facility were already issued. With respect to waste disposal, first steps have already been taken. In particular, the rapid clarification of the doubts on the suitability of the Gorleben site raised by the Federal Government has been initiated. The next important issue will be the final preparation and publication of the site selection criteria and procedures at the end of 2002.
RADIOACTIVE WASTE MANAGEMENT IN THE UK

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Abstract
The UK has a significant nuclear legacy going back to the 1940s. The UK Government aims to develop, and implement, a UK nuclear waste management programme which inspires public support and confidence. The nuclear legacy, current waste management practices, regulatory arrangements and proposals for engaging the public are described.

1 Introduction
The UK has a significant nuclear legacy arising from Government funded work going as far back as the 1940s. The Government is proposing radical changes to current arrangements to tackle this legacy and clean-up nuclear sites[1]. These include the setting-up of a Liabilities Management Authority to provide strategic management and direction to a comprehensive and long-term clean-up programme. And proposals for developing a policy for managing solid radioactive waste in the UK[2]. This paper provides an overview of these developments.

2 The Nuclear Legacy
The nuclear legacy was largely created by Government funded work including:

- nuclear research and development programmes in the 1940s, 50s, 60s and 70s; and
- the development, construction and commissioning of the Magnox nuclear power stations and associated fuel cycle plants in the 1950s, 60s, and 70s.

In addition to facilities, such as fuel cycle plants and power stations, the legacy includes about 10,000 tonnes of intermediate level radioactive waste. These wastes are currently safely stored awaiting a decision on their long-term future. The quantity will increase to 250,000 tonnes when nuclear material currently in use is converted into waste. A further 250,000 tonnes of waste will arise during the clean-up of nuclear plant over the next century. Considerably larger amounts of low-level waste are disposed of at the special surface repository operated by BNFL at Drigg in Cumbria.

3 Current Waste Management Practices
The treatment and handling of radioactive waste has been determined by its characteristics. However, for management purposes, rather than for any regulatory need, radioactive waste is divided into four categories according to its heat generating capacity and activity content as detailed in Box 1.
Wastes which can be disposed of with ordinary refuse, each 0.1 m³ of material containing less than 400 kBq of beta/gamma activity or single items containing less than 40 kBq

Wastes containing radioactive materials other than those suitable for disposal with ordinary refuse, but not exceeding 4 GBq/te of alpha or 12 GBq/te of beta/gamma activity – that is, wastes which can be accepted for authorised disposal at Drigg, Dounreay or other landfill sites by controlled burial

Wastes with radioactivity levels exceeding the upper boundaries for LLW, but which do not need heating to be taken into account in the design of storage or disposal facilities

Wastes in which the temperature may rise significantly as a result of their radioactivity, so this factor has to be taken into account in designing storage or disposal facilities

Box 1: Waste Categories

Very low level wastes (VLLW)

Very low-level waste (VLLW) covers wastes with very low concentrations of radioactivity. It arises from a variety of sources, including hospitals and non-nuclear industry. Because VLLW contains little total radioactivity, it has been safely treated as it has arisen by various means, such as disposal with domestic refuse directly at landfill sites or indirectly after incineration.

Low level wastes (LLW)

Solid low-level waste (LLW) includes metals, soil, building rubble and organic materials, which arise principally as lightly contaminated miscellaneous scrap. Metals are mostly in the form of redundant equipment. Organic materials are mainly in the form of paper towels, clothing and laboratory equipment which have been used in areas where radioactive materials are used – such as hospitals, research establishments and industry.

Since the 1950s around 1,000,000 m³ of LLW have been safely disposed of, mainly at the shallow burial site at Drigg, Cumbria and to a lesser extent at Dounreay, Caithness. Figures for 1998 indicate 8,000 m³ of LLW in storage, about half of this is in temporary storage awaiting disposal. The rest is either unsuitable for disposal, or is being stored pending future treatment to make it passively safe. In addition to the LLW generated by BNFL, Drigg provides a UK-wide disposal service to a spectrum of customers including hospitals and universities.

Intermediate level wastes (ILW)

Intermediate level waste (ILW) arises mainly from the reprocessing of spent fuel, and from general operations and maintenance of radioactive plant. The major components of ILW are metals and organic materials, with smaller quantities of cement, graphite, glass and ceramics.

Over the period 1949 to 1982 73,530 tonnes of low and intermediate waste were disposed of by the UK to the North East Atlantic. Since 1982 ILW which would have been disposed to sea has been stockpiled. In addition some arisings from the late 1940s onwards have been stored on sites.

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Intermediate level waste (ILW) arises mainly from the reprocessing of spent fuel, and from general operations and maintenance of radioactive plant. The major components of ILW are metals and organic materials, with smaller quantities of cement, graphite, glass and ceramics.

Over the period 1949 to 1982 73,530 tonnes of low and intermediate waste were disposed of by the UK to the North East Atlantic. Since 1982 ILW which would have been disposed to sea has been stockpiled. In addition some arisings from the late 1940s onwards have been stored on sites.
Figures for 1998 indicate 71,000 m$^3$ of ILW in storage, 8,500 m$^3$ of which had been treated to achieve passive safety by forming stable packages for long term management. Stainless steel drums of 500 litre capacity are the main containers used. In order to avoid the additional radiological dose to workers and the very high costs that would be associated with re-packaging, conditioning is carried out in such a way as to anticipate the requirements for the future long-term management of the wastes. ILW, be it in raw or conditioned form, is mainly stored in shielded buildings, vaults or silos, mostly at the site where it arises.

High Level Waste

High Level Waste (HLW) is a heat-generating waste that has accumulated since the early 1950s at Sellafield and Dounreay as the concentrated liquid nitric acid product from the reprocessing of spent nuclear fuel. HLW comprises only about 2% of the UK’s total volume of stored radioactive waste, but about 90% of its radioactive content. HLW storage facilities have cooling systems to dissipate the heat that the waste generates, and massive concrete shielding to protect the operators. There are about 1300 m$^3$ of liquid HLW stored in water-cooled tanks at Sellafield. The equivalent of a further 900 m$^3$ of liquid HLW has already been converted at Sellafield into a solid and stable form by immobilising it in glass (vitrification) within stainless steel canisters of about 140 litre capacity. There is a smaller quantity (230 m$^3$) of less active HLW still in liquid form at Dounreay. Current Government policy is that vitrified HLW should be stored for at least 50 years to allow the heat to decline so as to make long-term management less complex.

4 Future Waste Arisings

Wastes of all categories will continue to arise from the remainder of the current UK nuclear power programme, continuing military activities and small users.

VLLW and LLW will be produced for the foreseeable future assuming radionuclides continue to be used in medicine, research and education. Landfill sites and incinerators will probably continue to deal with some of these wastes.

LLW will continue to arise from ongoing operations of the nuclear power industry and from military activities. The decommissioning of all today’s nuclear facilities once they come to the end of their lives will continue throughout this century and possible the next and will also produce much LLW. In total about 1,800,000 m$^3$ of LLW is expected by 2160, this is far more than has been disposed of to the Drigg site since it opened. Drigg only has a finite capacity in the currently consented area and may be full by about 2050, hence new arrangements will have to be examined. Successor plans will need to be in place before then to deal with arisings of LLW requiring long-term management.

About 210,000 m$^3$ of ILW will be arising almost equally from the continued operation of the current nuclear industry, and from decommissioning of existing facilities, that were not designed to modern standards for ease of decontamination. Storage buildings for this conditioned ILW will need to be constructed at the main nuclear sites whilst the long term management arrangements are determined. The arising of ILW from the non-nuclear industry is very small, but it is important that a route is available for the management of spent sealed sources not returned to manufacturers. Techniques are available for rendering ILW passively safe, and over time an increasing percentage of it will be conditioned. ILW from decommissioning will become more important as operation of ageing reactors and facilities cease.
About 1000 m$^3$ of conditioned HLW will arise from the completion of current reprocessing contracts by about 2015. Unless new reprocessing contracts are obtained at Sellafield no further HLW will be created there, other than a small amount from the decommissioning of the vitrification plants. The liquid HLW at Dounreay will also have to be converted to a more stable form for improved long-term management.

**Decommissioning Wastes**

Decommissioning wastes may comprise ILW, LLW and VLLW in varying proportions. They are different from operational wastes, as they are mostly building materials and larger items of plant and equipment. The major components are concrete, cement and rubble, metals and graphite. After 2020, most ILW will arise from decommissioning. At 2100, the conditioned volume of wastes generated is expected to be about 150,000 m$^3$. The rate of arising increases after 2100, as nuclear power station decommissioning is completed. In total, about 50% of all expected ILW will be from decommissioning existing facilities.

5 Other Potential Wastes

There are some radioactive materials that are not currently classified as waste, but for which future use is uncertain. These include plutonium, uranium and spent nuclear fuel. These potential wastes will be considered now within the development of a comprehensive waste management policy so as to avoid any late changes to plans which might prove costly.

6 Nirex

Storage will continue to be needed for radioactive wastes until a final management strategy is available. In the case of solid wastes with more than low levels of radioactivity, the implementation of a final management strategy is unlikely to be complete for several decades. Wastes requiring storage pending the development of a final management strategy will need to be maintained under conditions of passive safety with minimal need for human intervention. Some wastes will need to be processed and packaged to achieve this.

Proposals for the conditioning of wastes are put to Nirex which assesses them against the safety of storing, transporting, handling and possible disposing of the wastes. Following such assessments, Nirex provides formal advice to guide waste producer plans and future development. When satisfied that the proposals are consistent with Nirex standards and specifications, Nirex packaging principles and the Nirex phased disposal concept, (in particular that the packages would be safe in an underground facility for protracted periods both before and after any backfilling and sealing), Nirex will provide endorsement in the form of a Letter of Comfort. This is not an automatic outcome from the submission of waste packaging proposals, as Nirex is sometimes unable to issue a Letter of Comfort.

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Nirex was formed in 1982 and incorporated as a private limited company – United Kingdom Nirex Ltd – in 1985, to provide radioactive waste disposal services. Nirex is financed and owned by the main waste producers BNFL, UKAEA and British Energy in proportion to the volumes of waste they produce. In addition, the MoD contributes to the funding of Nirex.
7 Regulatory Arrangements

Over 99% of radioactive wastes are on sites licensed under the Nuclear Installations Act 1965 (nuclear licensed sites). The Health & Safety Executive (HSE) regulates radioactive waste management on these sites. The HSE may attach to site licences such conditions as it thinks fit with respect to the handling, treatment and disposal of nuclear matter – including radioactive waste.

The standard required by nuclear site licence conditions is that there should be no leakage or escape of radioactive materials from stored waste unless it is authorised by the Environment Agency in England and Wales or SEPA: the Scottish Environment Protection Agency in Scotland (the environment agencies). All new stores are regulated to this standard, but some older stores do not fully comply. In some of these cases, wastes are being retrieved using remote handling techniques.

The environment agencies are responsible for regulating, under the Radioactive Substances Act 1993, routine disposals of all forms of radioactive wastes (solids, liquids and gases). However, on sites that HSE licenses, the agencies have no statutory powers over waste storage until the licensee seeks permission to dispose of the waste. On sites that are not licensed, such as hospitals and universities, the agencies regulate storage of radioactive wastes as well as disposals.

However, HSE have a statutory requirement to consult the agencies on radioactive waste management issues before issuing, amending or varying nuclear site licenses, or attaching conditions to them relating to radioactive waste management. In addition to statutory consultation requirements, the HSE and the environment agencies have set down and jointly agreed their responsibilities and working arrangements on matters of mutual interest within a Memorandum of Understanding (MoU). The intention of the MoU is that the activities of the agencies and HSE in relation to licensed nuclear sites are consistent, co-ordinated and comprehensive. The possibility of conflicting requirements being placed on licensees, or others concerned, is eliminated, and duplication of activity is minimised.

In practice, a nuclear site operator wishing to process or store radioactive wastes must first seek permission to do so from HSE. The MoUs provide for the environment agencies to be consulted on the licensee’s proposals for construction, modification or decommissioning of plant. In accordance with the terms of the MoU, the HSE does not give the licensee permission to proceed with any proposal that has radioactive waste management implications unless the appropriate Agency has written to HSE to indicate its agreement.

Consideration is being given to strengthening the regulatory role of the environment agencies. The purpose of this would be to provide greater assurance that:

- the processing and storage of waste does not prejudice future management options, and
- that nuclear site operators give increased emphasis to long-term environmental considerations.

The overall aim is to establish an effective regulatory regime that will achieve the following:

- give proper emphasis to safety and long-term environmental considerations;
- maintain containment of radioactivity and prevent leakage so that wastes can do no harm to people or the environment;
• provide for any necessary steps to prevent unauthorised release of radioactivity into the environment to be taken promptly;
• where waste needs to be packaged or treated to achieve passive safety, to ensure that this is done promptly;
• provide assurance that processing and storage of wastes do not prejudice future waste management options;
• provide adequate opportunities for consulting, and informing the public;
• provide an appropriate balance between costs and benefits; and
• ensure transparency in setting standards and in the regulatory processes.

8 Public Debate

A successful waste management policy can only be achieved if it commands public support. Several techniques are being considered for engaging the public. These include:

• workshops;
• interactive panels;
• community advisory committees;
• citizen’s juries;
• consensus conferences;
• stakeholder dialogues;
• local Agenda 21 groups;
• regional sustainable development frameworks;
• economic valuation techniques;
• public meetings;
• public hearings and inquiries;
• open houses;
• deliberative opinion polls;
• research panels; and
• the internet.

Research has shown that:

• people can, will and want to engage with the issue of radioactive waste management. This includes engaging with the ethical debate. To facilitate this all stakeholders need to:
  • provide information in a neutral form outlining the pros and cons and including various people’s opinions;
  • use proactive techniques to allow access and space for people to discuss the issues;
  • demonstrate how people’s opinions have been taken into account;
  • involve people in the debate from the beginning and take their opinions into consideration;
• some people have negative associations with the word “nuclear” and do not distinguish between nuclear, radiation, bombs and waste; and
• most people are not familiar with current institutional arrangements, but seem to want an independent, inclusive body overseeing radioactive waste management, operating openly.
9 Acknowledgement

The author is pleased to acknowledge that this paper is largely based on "Managing Radioactive Waste Safely" a publication of the UK government departments listed at Reference 2.

10 References


Characterization of Radioactive Waste Products in the Context of the New German Radiation Protection Ordinance and of Relevant International Standards

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Abstract
A survey is given on the new German Radiation Protection Ordinance (RPO) and on the basis of the underlying system of radiation protection. Emphasis is laid upon the system of exclusion, exemption, and clearance in the context of radioactive wastes from practices and of production residues from work activities. The role of national and international standards in the process of harmonization of provisions of other ordinances and of guidelines with the new RPO as well as some open questions are discussed.

1 The Amendment of the German Radiation Protection Ordinance of 2001


The German Government has used the occasion of the amendment for a complete re-structuring of the RPO. The new RPO consists of five parts:

- Part 1 (§§ 1-3) - General provisions - gives the objective of the ordinance, defines its scope and contains detailed provisions on definitions. The objective of the Ordinance is the protection of man and of the environment, against the negative effects of ionizing radiation.
- Part 2 (§§ 4-92) deals with the protection of man and the environment against radioactive substances or ionizing radiation resulting from goal oriented uses in connection with practices.
- Part 3 (§§ 93-104) - Protection of man and the environment against ionizing radiation emanating from natural sources - covers certain types of work activities involving the presence of natural radiation sources leading to non-negligible exposures.
- Part 4 (§§ 105-110) deals with the protection of consumers in connection with the addition of radioactive substances to products.
- Part 5 (§§ 111-118) contains joint provisions applicable to all parts of the ordinance such as transitional and final provisions and administrative fees.

In this paper, some basics of the new ordinance and of the underlying system of radiation protection are discussed. Emphasis is laid upon aspects of exclusion, exemption and clearance in the particular context of radioactive waste from practices and of production residues from work activities. For a
more general discussion of the RPO see ref. [5]. In refs. [6-9], some special aspects of this paper are further elaborated.

The new RPO is a very complex, technical ordinance, comprising besides 118 provisions 14 annexes. As a result of this new RPO, many other ordinances, standards and technical guidelines need to be harmonized with the new provisions. In this context, the importance of recent national and international standards for the practical application of the RPO and of related ordinances and guidelines is discussed. Finally, some open questions are outlined.

2 The Current System of Radiological Protection

The fundamental principles of radiological protection are justification, limitation and optimization [10,2]. They are laid down in the first chapter of Part 2 of the RPO:

- § 4 - Justification - ensures that new types of practice resulting in exposure to ionizing radiation must be justified by their economic, social or other benefits in relation to the health detriment they may cause. Existing types of practice may be reviewed if there is new scientific evidence regarding their consequences.
- § 5 - Dose limits - enumerates the dose limits of the ordinance applicable to members of the public and exposed workers.
- § 6 - Dose reduction - makes it compulsory to avoid any unnecessary exposure. The principle of optimization requires that, even if a practice does not exceed the relevant dose limits, exposures have to be kept as low as reasonably achievable.

Given the omnipresence of natural radiation and radiation exposures, it is necessary to establish a concept of radiological protection in order to put the principles of radiological protection into operation. The concept used in the RPO is based on the terms exclusion, authorization and exemption. Any human activity or source which may lead to non-negligible radiation exposures need authorization unless they are excluded or exempted from regulation.

The concept of exclusion means that any exposure whose magnitude or likelihood is essentially not amenable to control is deemed to be excluded from regulation. It also comprises uncontrollable exposures which cannot be restricted under any conceivable means. Such exposures are for instance exposures originating from K-40 in the body, from cosmic radiation at ground level, from unmodified concentrations of natural radionuclides in most raw materials, and from fallout caused by past nuclear testing in the atmosphere. If the potential exposures are negligible they are excluded from regulatory control following the principle of the Roman law "de minimis non curat lex".

The concept of exemption determines what practices and sources (and their waste) may - and what may not - be freed a priori from all regulatory control. There are two reasons for exempting a source or an environmental situation from regulatory control:

- One is that the source gives rise to small individual doses and small collective doses in both normal and accidental conditions.
- The other is that no reasonable control procedures can achieve significant reductions in individual and collective doses.

Exemption is necessarily a source-related process, while the triviality of the dose is primarily individual-related. It follows the principle of the Roman law "de minimis non curat praetor".
However, exemption should not be granted to permit practices that would otherwise not be justified.

Practically, the source-relationship of exemption is put into effect by exempting materials from regulatory control if their total activities $A_i$ and activity concentrations $C_i$ are below exemption levels (Freigrenzen) $FG_i$. Values of the $FG_i$ are given in Appendix III of the RPO for both the total activities and the activity concentrations of the individual radionuclides. If several radionuclides are present it is required that

$$\sum_i A_i / FG_i \leq 1 \text{ and } \sum_i C_i / FG_i \leq 1$$

(1)

This to allow the material to be exempted.

The legal fixation of exemption levels requires the definition of a trivial dose. As discussed in ref. [7] the trivial individual effective dose is of the order of 10 to 100 μSv per year. This statement is based on considerations either of the acceptable annual individual risk taking into account the ICRP risk factors [11] or of the natural radiation background and its variability. Both approaches merely result in an order of magnitude for the trivial dose and not in a single numerical value as required for an ordinance. The RPO assigns a dose criterion for triviality of 10 μSv per year for a single source or practice in order to ensure compliance with the principle of trivial dose in case of several exempted practices.

After defining a dose criterion of triviality, exemption levels are the result of modeling exposure pathways on which exempted materials, practices, and sources potentially could lead to radiation exposures at the workplace or for members of the general public. The principles for exemption and methods for establishing exemptions levels are laid down in refs. [12,13]. A detailed account on the derivation of exemption levels and their application in the RPO is given in ref. [7].

2.1 What materials are radioactive?

In the system of exclusion, authorization, and exemption, the term radioactive substance is of crucial importance since on its basis the system is extended to cover the clearance of radioactive waste which in the new RPO [1] is extensively regulated for the first time on the basis of Article 5 of the Euratom Basic Safety Standards [2].

The term radioactive substance has been newly defined in a legal sense in § 2 of the amended Atomic Energy Act [4]. According to this definition, radioactive substances (nuclear fuels and other radioactive substances) are substances that contain one or more radionuclides and whose radioactivity or activity concentration with respect to nuclear energy or radiation cannot be neglected. In this legal definition of radioactive substances, the physical meaning of "radioactive" is confined to the term radionuclides. Radioactive substances are those that are subject to the Atomic Energy Act's protection and supervision regime and are those which are explicitly regulated by these provisions.

In general, radioactive substances within the meaning of the Atomic Energy Act are, thus, substances that contain

- man-made radionuclides or
- radionuclides of natural origin whose nuclear properties are to be used and whose radioactivity and radioactivity concentration exceed the exemption levels of the RPO.
i.e. substances whose handling will be subject to authorization.

Substances are not radioactive in the legal sense of the Atomic Energy Act if their activities "may be neglected" [10]. This is the case if they are cleared from regulatory control according to section 9 (§ 29) of the new RPO. The provisions of § 29 for the first time comprehensively stipulate the conditions of clearance of radioactive substances, the past RPO just gave in § 4 some regulations for radioactive waste.

2.2 Clearing of Radioactive Substances

In addition to provisions regulating delivery and storage of radioactive waste (§§ 72-79), the new RPO includes provisions on treatment and packaging of radioactive waste (§ 74). Thereby it is guaranteed that the authorities competent for the disposal of this waste are informed on the amounts of waste and the respective transports. Also, a loss of radioactive substances shall thereby be prevented.

According to the new RPO, a clearance procedure is possible for such radioactive substances with negligible radioactivity if they originate:

1. from use, treatment, handling of:
   a) man-made radioactive substances or
   b) radioactive substances of natural origin whose nuclear properties are used,
2. from practices subject to authorization within the scope of the Atomic Energy Act: (storage, treatment, processing, and other usage of nuclear fuels, operation, other possession, decommissioning, safe enclosure of a facility and dismantling of a facility or parts of a facility) or
3. from operation of accelerators.

The term clearance is defined in § 3 Nr. 15 as an act of state issued by an authority which releases solid or liquid radioactive substances and moveable objects, instrumentation, buildings and building rubble, excavated soil, and sites, which are activated or radioactively contaminated, from the regulatory control by the Atomic Energy Act and by the RPO. The RPO distinguishes between (unconditional) clearance and specific clearance depending whether these substances are subject to no restrictions regarding their future use, application, recycling, re-use or disposal or not. The Ordinance distinguishes different types and paths of clearance for particular materials and objects such as solid materials for disposal, materials in liquid form to be disposed by incineration in a corresponding plant, buildings to be demolished, and scrap metal to be recycled.

Clearance requires that surface contaminations and activity concentrations of such substances are below defined surface contamination limits and clearance values given in Appendix III of the new RPO and have received clearance. After clearance, such substances are no longer radioactive substances. They fall under relevant specialized law, especially the Closed Substance Cycle and Waste Management Act [14].

As stated in the Euratom Basic Safety Standards [2], the effective dose for individual members of the population resulting from clearance of radioactive substances shall be of the order of 10 µSv or less for any member of the public and the collective dose for the population shall be less than 1 man-Sv in a year. This is the basis for the clearing levels given in Appendix III of the RPO. For the derivation of the clearance values see refs. [15-17].

The exemption and clearance values of the new RPO differ considerably (e.g. table 1), since different models and exposure pathways underlie the derivation of exemption levels and clearance
values. When compared with the exemption levels, the clearance values tend to be the lower (table 1). This sometimes causes confusion and concern in the public since the differences can only be understood on the basis of knowledge about the underlying models and about the conservatisms implemented in these models. It would be desirable to have just one set of radionuclide-specific clearance and exemption levels to allow both exemption of practices and clearance of materials from regulated practices as it is discussed in the context of radionuclides in commodities, e.g. [18]. A plethora of different values leads to confusion.

Table 1: Comparison of exemption levels and (unconditional) clearance values expressed in specific activities for some selected radionuclides.

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Exemption levels in Bq/g</th>
<th>Clearance values in Bq/g</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-238sec</td>
<td>1</td>
<td>0.009</td>
</tr>
<tr>
<td>Pu-239</td>
<td>1</td>
<td>0.04</td>
</tr>
<tr>
<td>Pb-210++</td>
<td>10</td>
<td>0.02</td>
</tr>
<tr>
<td>Co-60</td>
<td>10</td>
<td>0.1</td>
</tr>
<tr>
<td>Cs-137</td>
<td>10</td>
<td>0.5</td>
</tr>
<tr>
<td>Sr-90+</td>
<td>100</td>
<td>2</td>
</tr>
<tr>
<td>I-131</td>
<td>100</td>
<td>2</td>
</tr>
<tr>
<td>I-125</td>
<td>1000</td>
<td>3</td>
</tr>
<tr>
<td>P-32</td>
<td>1000</td>
<td>20</td>
</tr>
<tr>
<td>Re-186</td>
<td>1000</td>
<td>1000</td>
</tr>
<tr>
<td>Cl-36</td>
<td>10000</td>
<td>8</td>
</tr>
<tr>
<td>Te-99</td>
<td>10000</td>
<td>10</td>
</tr>
<tr>
<td>C-14</td>
<td>10000</td>
<td>80</td>
</tr>
<tr>
<td>Fe-55</td>
<td>10000</td>
<td>200</td>
</tr>
<tr>
<td>S-35</td>
<td>100000</td>
<td>60</td>
</tr>
<tr>
<td>H-3</td>
<td>1000000</td>
<td>1000</td>
</tr>
</tbody>
</table>

1 if not noted otherwise the exemption levels are to be taken as clearance values for radionuclides with half-lives less than 7 d.

It is also problematic that the exemption levels given in the RPO are values rounded to orders of magnitude, revealing the character of the exemption concept, while the more-precisely given clearance values pretend a level of reliability, which cannot be derived from the underlying concept of clearance.

2.3 Practices and Work Activities

On the basis of the Euratom Basic Safety Standards [2] the scope of the new RPO was widely extended. It applies both to practices and work activities. Practices are defined as human activities that can increase the exposure of individuals to radiation from an artificial source, or from a natural radiation source where natural radionuclides are processed for their radioactive, fissile or fertile properties, except in the case of an emergency exposure. Work activities are defined as human activities which are not practices, but where the presence of natural radiation sources may lead to a significant increase in the exposure of workers or members of the public which cannot be disregarded from the radiation protection point of view.
In the new RPO, part III (§§ 93-104) deals for the first time with the protection of man and the environment against ionizing radiation emanating from natural sources as a consequence of work activities. As natural radiation is omnipresent, the protection concept differs considerably from the one concerning practices. In particular it does not contain a clause about justification. Three principal areas are subjected to new regulations:

- increased exposure of workers in specific working areas,
- exposure of aircraft operating personnel to cosmic radiation,
- increased exposure of members of the public due to production residues.

The new RPO explicitly lists possibly critical working areas in annex XI based on an examination by the German Radiation Protection Commission [19]. Those working places are subject to control, exposures must be estimated, and the competent authority has to be informed if it is possible that the exposure exceeds 6 mSv per year.

The protection of aircraft personnel against cosmic radiation is regulated in § 103 of the RPO. Aircraft crews must be informed on the risks of cosmic radiation, and the doses have to be monitored and communicated to the crew members if they wish. The assessed exposure has to be taken into account when organizing work schedules in order to avoid high doses.

Generally, § 93 states that for work activities the system on dose limitation developed in the chapters on practices applies.

2.4 Residues from Work Activities

With respect to the topic of this paper, the consideration of production residues is of importance. They are related to the frequently used terms naturally occurring radioactive materials (NORM) and technologically enhanced naturally occurring materials (TENORM). Up to now, the international practice is vague in relation to the controllability of exposures from other natural sources, including waste from industries processing NORM. As stated above as examples of exposures excluded from regulatory control it was referred to exposure from "unmodified concentrations of radionuclides of radionuclides in most raw materials" [10].

The IAEA Basic Safety Standards [10] discusses two approaches. One approach is to exclude respective industries unless the activity levels in materials used were such that the doses being received were sufficiently high to cause concern. The other approach follows from a decision that specified industries should be subject to regulation, i.e. that they constitute a practice in the context of these standards.

While the effects of radiation in the specified working areas listed in Annex XI of the new RPO on members of the public are negligible, the situation is different with regard to residues which might lead to a non-negligible exposure of members of the public. Therefore, it is laid down in the RPO that residues must be subject to supervision if their recycling or disposal could lead to an exposure of members of the public to more than 1 mSv as a guideline value (§ 97). The residues subject to supervision are listed in Annex XII. They have to be supervised and may only be released from supervision through a procedure which is modeled in accordance with the clearance procedure in § 29 (§ 98).

NORM and TENORM residues are exempted from supervision if the activity concentrations of each of the radionuclides of the U-238 and Th-232 decay chains are below 0.2 Bq/g. In addition, raw materials of the technological processes listed in Annex XII of the RPO, which contain
naturally occurring radionuclides are exempted from supervision. A graded system of limiting activity concentrations for residues from work activities is set up in Annex XII of the RPO which decide about release or not from supervision.

The release from supervision can be either unconditionally for further use, recycling, etc. or for disposal in the framework of the Closed Substance Cycle and Waste Management Act [14]. The Ordinance states under which circumstances this is the case; mainly, certain paths of disposal have to be followed. If residues cannot be released from supervision because of their specific activities the authorities can decide about further protection measures and the way in which these residues can be disposed.

3 Some Aspects of Quality

In the process of harmonization of the provisions of other ordinances and guidelines with those of the new RPO also other improvements will be made in order to update them with respect to the state of science and technology. Since practical measures in radiological protections always are relying on measurements of exposures and activities and since comparison of measurement results with legal limits, levels and guideline values provide the basis for actions to be taken in radiological protection as well as for decisions about clearance of radioactive substances and about release of residues from supervision, the quality of the measurements is of prime importance.

Since the previous RPO was issued in 1989, important developments in quality assurance and quality control have been made. Trace-ability of measurements and evaluations has become a conditio sine qua non and standardization of procedures, certification and accreditation of measurement laboratories have become top issues.

In particular with respect to the characterization of waste and residues, some aspects of these developments shall be discussed here, namely:

- **uncertainties in measurement** and
- **characteristic limits**, such as decision thresholds, detection limits and confidence limits.

Measurement uncertainties and characteristic limits have become fundamental data for the judgment about measurement results and for the characterization of measurement procedures. Increasingly, they are referred to in national and international ordinances and guidelines.

For the calculation of measurement uncertainties standardized procedures are laid down in the ISO Guide to the Expression of Uncertainty in Measurement (GUM) [20]. These procedures are in accordance with German national standards DIN 1319-4 [21] and DIN V ENV 13005 [22] and with recommendations by EURACHEM [23] for application in analytical chemistry.

The important point of these standards and recommendations is that for a measurement result a complete standard uncertainty has to be given which takes into account all known sources of uncertainties. On the basis of a Bayesian theory of measurement uncertainties [24] this includes contributions to uncertainty which can be determined from repeated or counting measurements (type A) as well as those which can only be obtained from other sources (type B). These type B uncertainties comprise all information available about previous measured data, experiences about the measurement procedures and processes or about the characteristics of relevant material, phenomena or instruments, specifications and information obtained from manufacturers, data from
calibration and other certificates as well as uncertainties which are attributed to data from hand-
books and compilations.

According to the state of science and technology, for each measurement its associated standard
uncertainty according to the GUM has to be given. With the standards cited [20-23] complete
standard uncertainties can be given for each measurement result obtained by any measuring
procedure in an unambiguous way.

On the basis of complete standard uncertainties according to refs. [20-23] also the characteristic
limits, such as decision threshold, detection limit and limits of the confidence interval can be de-

erived on the basis of DIN 25482-10 [25] and ISO 11929-7 [26], respectively, in a straight-

forward way.

These characteristic limits allow for the following statements or decisions:

- The decision threshold decides the question whether a result of a measurement indicates a
true value of the measurand larger than zero. In practice, this decides whether a dose or an
activity different from zero has been observed.
- The detection limit is the smallest true value of the measurand which can be reliably de-
termined and thereby qualifies the measurement procedure with respect to legal or other
requirements.
- The confidence limits enclose a confidence interval which contains the true value of the
measurand with a pre-selected probability.

The recognition that every result of a measurement has an uncertainty and that the capabilities to
measure any quantity is downwards limited by background effects or blanks causes some prob-
lems with respect to the practical application of the provisions laid down in the new RPO. Some
of them will be discussed in the next chapter.

4 Some Open Questions

One problem arises from the fact that materials to be cleared or released from supervision usually
are not homogenous. Thus, the specific activity on which a decision about clearance or release
has to be based is not a well defined quantity. It is not yet decided whether the specific activity of
each individual sample has to be below the respective limit or whether this has to hold for the
mean, the median or the expectation value of the distribution of specific activities in the material.
Since the clearance values and release limits are based on models of exposure pathways, the ex-
pectation value of the activity distribution may be the most adequate quantity to use for a deci-
sion. However, whether this will be the case is still under discussion.

A second problem arises from measurement uncertainties. If each measured result is uncertain,
how does uncertainty affect a decision about compliance with legal limits and, if confidence in-
tervals are used in demonstrating compliance, which confidence level has to be adopted? The
RPO and relevant technical guidelines do not yet give any advice in this respect. As discussed
elsewhere [27], first attempts have been made to cope with these problems in Austria by dealing
in standards the problems of legal limits and uncertainties [28,29]. In Germany and other coun-
tries, this is still an open problem.

A third problem deals with decision thresholds and detection limits. It appears frequently in ac-
counting of radioactive emissions from nuclear facilities and of the radioactive inventories of
waste when measured data are below the decision threshold. Frequently, then the detection limit or some percentage of it is used in the account. This is a complete misuse of the detection limit which has the purpose to qualify a measurement method, not to substitute missing data. If a result of a measurement is below the decision threshold, a true value of zero of the measurement must not be excluded and a zero value has to be adopted. If the assumption of a true value of zero matters, the detection limit, i.e. the sensitivity of the measurement has to be decreased as such that a true value of zero does not matter anymore.

5 Conclusions

The new German Radiation Protection Ordinance of July 20, 2001 is an ambitious piece of legislation, in particular with respect to the new provisions for the clearance of radioactive waste from practices and for the supervision of residues from work activities. International standards of metrology such cited in refs. [20-22,25,26] will have increasing impact on the characterization on waste and residues.

The multitude of innovations in the new RPO provide a sophisticated basis for an adequate protection of man and the environment against the dangers from ionizing radiation. For the characterization of waste and residues they are a challenge for both, the practical application and for the further development of legal regulations in radiological protection.

6 References

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Decommissioning and Standardization
Strategy of waste management
on the example of the decommissioning of a nuclear power plant

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Abstract
The dismantling of a nuclear power plant and the waste treatment is technically solved. The actual 'problem' of each new dismantling project is the proper management of the total project, in which the waste management plays an important role. The tasks turn out to be difficult, because the knowledge of other decommissioning projects can be used in some cases or only partly, i.e. it depends on the power plant itself. With the dismantling of the facility, the handling of the waste has to be organized and carried out.

The essential objective of the waste management is to minimize the expenses which are influenced by various factors i.e.: the waste itself, final or interim storage conditions, value of exemption level, treatment facilities, licensing conditions. All these factors have to be valued during the planning and the project development. This strategy of the waste management is the focus of the lecture.

1 Introduction
The dismantling of a nuclear plant in Germany is not an unknown territory and is technically solved. There is extensive know-how since more than 20 years with the already finished dismantling projects NPP Niederaichbach (KKN), Großwelsheim (HDR), nuclear ship Otto Hahn and the partly far advanced dismantling projects NPP Gundremmingen unit A (KRB A), testing nuclear plant Kahl (VAK), NPP Würgassen (KWW) as well as the experimental reactors FR 2, MZFR, KNK in Karlsruhe and further small reactors. The necessary techniques and methods exist for the dismantling of the components and the treatment of the residues and waste which arise. Regardless of this each new dismantling project has to be newly planned and organized, as each single plant has its own specific framework conditions, i.e.: high temperature reactors have a different activity inventory than light water reactors; with light water reactors considerable differences in the framework conditions for the planning and later realization can arise due to the type of reactor (BWR, PWR, WWER), due to different ways of usage, damages of fuel elements or accidents. The main task for each new dismantling project is the proper management of the total project, in which the waste management plays an important role. Waste management means the treatment of the radioactive residues from the dismantling to the final storage of the waste or to the free release.

2 Initial situation
The following waste groups exist after the final shut-down of the nuclear power plant:
- Spent fuel elements
- Operation media
- Operational waste
- Contaminated and activated components
The first waste groups, spent fuel elements, operation media and operational waste, can be disposed in the so called post-operational-phase in the scope of the effective operation licence. The operational equipment exists and is designed for this. The treatment of this waste shall not be handled further on.

3 General procedure of the dismantling

The dismantling of the facility contains the dismantling of contaminated and activated components, their working and treatment so that after the removal of the artificial radioactivity the structures of the building can be free cleaned and the facility can be discharged from nuclear supervision.

The dismantling of a nuclear power plant can be divided into the following dismantling stages:

Stage 1  Reducing of all systems and components for the concerns of the dismantling (remaining operation) and dismantling of the low and none contaminated components
Stage 2  Dismantling of contaminated components – especially of the reactor coolant system
Stage 3  Dismantling of activated components – reactor pressure vessel with fittings, activated parts of the biological shield
Stage 4  Dismantling of the remaining contaminated components, dismantling of the remaining operational facilities, decontaminating of the building, free clearance and discharge of nuclear supervision
Stage 5  Other usage of the buildings or demolition of the building structures

The realization of each stage 1 to 4 requires the permission according to the Atomic energy act and has to be checked in total for the environmental compatibility. Stage 5 can be handled within the framework of the building regulations.

The following illustration shows the nuclear licensing procedure in an overall view:

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**Figure 1:** Licensing procedure in Germany
Each stage requires a separate dismantling permission. The decommissioning, which means the final shutdown of the nuclear plant, will be applied with the first permission. According to the Atomic Energy act in connection with the act for the environmental compatibility the total procedure has to be carried out with public involvement. For this the altogether planned steps have to be described in a safety report and have to be proofed in view of the environmental effects. During the respective licensing procedure each stage has to be checked if the precaution steps for the effect restrictions stick to the safety report. It is important for the planning of the treatment of the nuclear residues to have information about the nuclear condition of the plant. This condition can only be determined after the shutdown or during the post-operational phase.

4 Objective

The most important objective of the waste management is to reduce the radioactive waste to a minimum so that the expenses for the interim and final storage are low. Therefore the total expenditure for the free clearance treatment of the radioactive residues has to be compared with the expenditure for the disassembly, packing of radioactive residues with the objective of radioactive waste. That means that in terms of economy it is not always useful to have a minimum of waste. The target is to reach a minimum of radioactive waste by a minimum of treatment of radioactive residues. This procedure is illustrated in a diagram as follows:

![Diagram](image)

Figure 2: Target for treatment of the radioactive residues

5 Effecting factors for the waste management

The waste management is influenced by various factors. The most important factors are:
- dismantling of the contaminated and activated components
- conditions of the final and the interim storage
- legal framework conditions
- available ways for waste disposal and treatment equipment
- special criterions of the plant.
For each factor examples of influencing criterions shall be named. The total of the influencing criterions is far more extensive.

5.1 Dismantling of contaminated and activated components
- Mass of the contaminated and activated components
- Materials and geometry
- Level of contamination and activation
- Type of contamination (air-launched, loose, stick to, slightly volatile)
- Nuclide mixture (amount, Alpha ejector, special nuclide)
- Results of the system decontamination in the post-operational phase

5.2 Conditions of final and interim storage
- Does a final storage exist? If yes: what are the terms of acceptance, what are the storage charges?
- Is there a suitable interim storage? If yes: is there enough space, what are the terms of acceptance and what is the charge?
- After this it has to be clarified, whether an interim storage, i.e. at the facility site has to be built and for what storage period (i.e. 40 years) it has to be planned. What are the criterions for the design of the interim storage?
- Is there enough space for an interim storage of material within the facility or on the site?
- Are there any possibilities for conventional waste disposal near the site, i.e. dumping site for rubble, isolations, asbestos etc.?

5.3 Legal framework conditions
- What are the clearance criterions (Radiological Protection Ordinance)
- Which output value for stack discharge and process drains will be approved?
- Which dose rate at the facility fence (<1mSv/a) has to be complied i.e. for the transportation availability on the facility site?
- What are the criterions for design (i.e. against earth quake) for a interim storage
- Transportation regulations.

5.4 Ways of waste disposal and equipment for treatment
- Which external equipment can be used and how are the specific expenses, i.e. for post-disassembly, decontamination, combustion, melting.
- How does the external treated facility parts have to be treated?
- What is the transportation fee to external equipment
- Is there any internal equipment that can be used (specific expenses, employees).
- Which internal equipment can be built (space requirement, investment, operational expenses, manpower-requirements.

5.5 Specific criterions of the facility
- Are there any other facilities in operation on the site?
- How many employees are available for the decommissioning
- Which personnel ability is available?
What is the amount of the available reserves and financial resources?
Are there any special objectives of the company?

All the influencing criteria have to be determined and considered in the planning of the waste treatment. In addition any further criteria resulting from the dismantling have to be also considered in the planning.

6 Overall planning of the waste treatment

The planning of the waste treatment is a substantial part of the overall planning of the decommissioning project and is subdivided into the following planning stages:
- Concept planning
- Preplanning
- Draft- and permission planning
- Completion planning
- Detail planning.

In the following the main planning topics of each planning stage will be examined regarding the waste treatment.

6.1 Concept planning

The concept planning is an iterative process which has to be carried out for different variations of the overall project after the basis data has been determined (masses, radiological condition):

![Diagram](image)

*Figure 3: Concept planning*
The result of the concept planning contains a description of the framework condition, of the concept and the estimated overall expenses.

For the waste treatment the following has to be basically regulated:
- Rough arrangement of the masses
- Rough arrangement in disposal destination
- Definition of internal/external treatment
- Evaluation of the final storage situation, concept planning interim storage

The rough arrangement of the masses is do be done on the basis of the following figure:

**Figure 4: Targets for the treatment of residues**

### 6.2 Preplanning

The preplanning also encloses the overall project and contains the concrete of the project on the basis of the chosen variation.

In the framework of the preplanning the following planning tasks are essentially important for the waste management:
- Arrangement of masses (contamination classes)
- Arrangement in disposal destination
- Pre-selection of procedures for the treatment of residues and waste (pre-classification)
- Planning of the internal treatment equipment
- Planning of the interim storage
For the pre-classification of the radioactive residues there are also a lot of criterions:

![Diagram of pre-classification of radioactive residues]

The result of the preplanning serves to illustrate the overall project in a safety report which is necessary in the scope of the public participation before receiving the first decommissioning and dismantling license.

### 6.3 Draft- and permission planning

The draft and permission planning will be carried out for each stage separate. For the waste management the following is planned:

- Timing of the residues to be worked on
- Type and number of equipment for the treatment of the residues and waste
- Definition of the packing / barrels
- Estimation of the necessary barrels
- Scope of the necessary reconstruction steps
- Definition of the framework conditions for the waste treatment
- Definition of the free release procedure
- Estimation of dose
- Estimation of expenses

The results of the draft and permission planning will be implemented in the enclosed documents of the application. Particularly the organizational flow of the steps will be determined as well as the corresponding framework conditions of the waste treatment. To optimize the flow in the planning and for the later processing suitable planning instruments should be designed and used. It is an obvious example to use a modular structured programme, like openRMS, for the waste management [1].
6.4 Completion planning
The completion planning will be carried out parallel to the permission procedure and serves the preparation for the realization of the steps of each stage. The following tasks are necessary for the waste management:

- Drawing up the equipment specifications for the treatment of the radioactive residues and waste
- Completion planning of reconstruction steps
- Specification of free release equipment
- Manpower planning
- Expense planning
- Setting up a detailed time schedule

6.5 Detail planning
The detail planning contains the concrete implementation of the steps in the facility after the decommissioning license has been given, i.e.:

- Drawing up job descriptions for the treatment, decontamination, transportation and free release
- Drawing up flow charts for the treatment of radioactive waste
- Drawing up flow charts for the release
- Follow up of the time schedule and expenses
- Preparation of the documentation.

Due to the demands for the documentation and the amount of data, the processing of the waste management cannot be useful handled without EDP anymore.

7 Acknowledgements
The waste management is a main and cost intensive part of the decommissioning of a nuclear power plant. Therefore in each planning step special attention has to be given to the planning of the waste treatment. Because of an iterative procedure, especially in the planning stages concept and preplanning, the various influencing factors can be valued, which secures an economic realization of the project. For the realization of the waste treatment enough flexibility is necessary, because the planning data, i.e. for the decontamination or for the management of the disposal destinations, cannot be exactly determined in advance. It needs to pay attention on this at the illustration of the waste management during the nuclear licensing procedure.

8 References
Waste Management and Analytics at VKTA

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Abstract

VKTA decommissions the old nuclear facilities of former GDR’s Central Institute of Nuclear Research and is responsible for fissile material and waste management, environmental and radiation protection and runs an accredited laboratory for environmental and radionuclide analytics.

Main objective is the minimization of radioactive waste. Therefore VKTA integrates its analytics in all decommissioning steps for radiological survey. Intensive analytical determination is urgent because of various nuclide vectors, consisted of alpha- beta- and gamma emitting nuclides. VKTA uses in-situ-technology and modern analytical technology for activity determination.

Solid wastes are treated in a treatment facility by mechanical methods, pressure beaming, ultrasonic or chemical decontamination, drying and pressing. Liquid wastes are treated chemically and by ion exchangers. For decay storage and waste for final disposal VKTA operates an interim storage.

As examples the treatment of the old reactor vessel and the solidification of fission product solutions are explained in the report.

1 VKTA’s tasks and decommissioning projects

Verein für Kernverfahrenstechnik und Analytik Rossendorf e.V. (VKTA) has been charged with decommissioning of all nuclear facilities of former GDR’s Central Institute of Nuclear Research, which was closed end of 1991. The Rossendorf research site is located in the east of the city of Dresden. The total amount of radioactive inventories was some E16 Bq including 951 irradiated high enriched (36% U-235) fuel elements of a Russian type 10 MW research reactor.

The period from 1992 to about 1997 was mainly characterized by obtaining the necessary licenses for decommissioning and developing a new infrastructure (i.e. waste treatment facility, interim storages for fissile material and waste, clearance monitoring facility).

Since that time the decommissioning work is in progress. The decommissioning projects are concentrated in three complexes:

- the reactors,
- the radioisotope production facilities,
- the former liquid and solid waste storage facilities.

Meanwhile two little zero power reactors are completely decommissioned and the dismantling of the 10 MW research reactor makes good progress.
The radioisotope production facilities were in operation for about 40 years. There a large variety of radionuclides with a total activity of about $4 \times 10^{14} \text{ Bq}$ was handled, what makes the decommissioning more complicated. The third complex contains the former waste treatment and storage facilities which were mainly located subterranean inside a fenced and controlled area.

Besides this VKTA takes care of fissile material and waste management as well as environmental and radiation protection for the complete Rossendorf research site and runs an accredited laboratory for environmental and radionuclide analytics.

One of the main objectives of decommissioning and waste management in Rossendorf is the minimization of radioactive waste. Therefore VKTA integrates its excellent analytics in all decommissioning steps for radiological survey. Actually we expect approximately 1300 m$^3$ radwaste and about 13000 m$^3$ material to be released. (Fig. 1)

2 Analytics

The analytical department is operating the accredited laboratory for environmental and radionuclide analytics and accompanies all the decommissioning steps from monitoring up to assessment and assists the decommissioning and the waste treatment with procedure recommendations. From the methodical point of view the main task is the analysis of artificial and primordial radionuclides. Conventional analytical methods rounds up the spectrum of possibilities.

To ensure the radiation protection at the several decommissioning objects at Rossendorf, intensive analytical determination is urgent. This is caused by the presence of very different nuclide vectors, consisted of alpha- beta- and gamma emitting nuclides. Therefore the radiological starting situation is examined very thoroughly. VKTA uses in-situ-technology as well as analytical examinations in the laboratories case by case. For example in the former radioisotope production facilities an area of about 10 000 m$^2$ was monitored with in-situ gamma spectrometry. (Fig. 2)

The most relevant radioisotopes at the VKTA decommissioning projects are U- and Pu-isotopes, Am-241, H-3, C-14, Fe-55, Ni-63, Sr-90, Co-60 and Cs-137. Analytical methods in a wide range of activity levels (from mBq up to GBq) and modern analytical technology is available for activity determination of this nuclides and enables VKTA to manage external service orders besides his own decommissioning tasks. (Fig. 3)

The detailed determination of the radiological starting situation leads to more accurate categorization of the residues, better estimation of expected waste amount and helps to optimize the decommissioning concepts by comparing the analytical data with the licensed limits.

For example we succeeded to minimize the amount of radioactive waste by intensive examination of contaminated soil during and in parallel to its remove. So the limit between material for restricted release and radioactive waste could be drawn much more precisely.

In this way the analytics of VKTA ensures to decrease the amount of radioactive waste to be stored in the interim storage at Rossendorf.

Based on its detailed analytical experience and in accordance to the de-minimis-concept, VKTA has also worked out a special concept for the release of premises with contaminated soil in deeper
layers under its surface, which actually is in the licensing process. Following this concept, we expect to reduce the amount of soil material which has to pass our mobile clearance facility from about 15 000 m$^3$ down to about 2700 m$^3$.

3 Waste management

Radioactive wastes of the VKTA decommissioning projects are characterized related to their physical, chemical and radiological conditions. The resulting data are recorded in a data file. The flow of low level waste leads from the decommissioning projects to the waste treatment facility, where the waste is conditioned in accordance with the requirements of long term interim storage. As solid waste treatment methods we usually use dismantling, cutting, milling, grinding or pressure beaming, ultrasonic or chemical decontamination, drying and pressing case by case. Low contaminated liquid wastes are treated chemically and by ion exchangers. Medium active liquid wastes were solidified with a special equipment.

The objective is to use the most effective decontamination and treatment method to reduce the volume of final radioactive waste and release as much material as possible. For volume minimization of waste for later disposal, VKTA uses high pressure pressing at external facilities.

After treatment the conditioned wastes are scanned by a gamma scanner for product control purposes according to the requirements of long term interim storage as well as final disposal.

VKTA’s waste recording records all steps of the waste material and activity flow from origin up to filled waste drums or arrangements.

4 Examples for correlation between analytics and waste management

The correlation between analytics and waste management will be shown with some examples:

4.1 The reactor and the reactor vessel
During decommissioning of the Rossendorf research reactor (Fig. 4) very different materials have to be treated, for instance a reactor vessel of aluminium alloy, a thermic column of graphite and a reactor top of cast steel. An extensive program of sampling and analysis delivered the basic data for the different matrices and enabled the determination of the relevant nuclide vectors. For the calculation of the neutron activation and nuclide compounds the radionuclide data were completed by the content of trace elements which were determined by ICP mass spectrometry. Analytically determined and calculated nuclide vectors corresponds well. This ensures that nuclide vectors for all different waste materials are available and the waste volume estimation becomes fairly precise. The data file of nuclide vectors is part of VKTA’s waste recording.

During reconstruction in 1987 the reactor vessel with a weight of about 4 Mg was removed from the reactor building and since that time stored in a pool in the former waste treatment area. Last year it was dried and cleaned and samples were taken and analysed. (Fig. 5) Because of financial reasons dismantling was decided to be done external at EWN GmbH Greifswald. The vessel was prepared for transportation and loaded into a shielded 40 ft. container, provided by NCS GmbH, which also transported it to EWN. After dismantling the components were separately pressed. As result 15 LAW drums, 3 MAW drums in concrete shieldings and 1 MAW drum in a MOSAIC-shielding were transported back to VKTA and are stored in the Rossendorf interim storage.
Based on this experience, we plan to treat the present reactor vessel the same way.

4.2 The fission product solution
As an example for liquid waste treatment, the solidification of liquid MAW should be explained. Before shut down of the nuclear facilities at Rossendorf, Mo-99 for production of Tc-99m-generators was separated by liquid-liquid-extraction in the AMOR complex. The residues of this process were fission product solutions as liquid MAW. Cs-137 and Sr-90 were the most relevant radioisotopes. Chemically the waste consists of highly acidic (nitric acid) solution containing aluminum nitrate. A total of 15 m³ had to be solidified for long term interim storage.

The radiochemical characterization of the solution has been performed by Institut fuer Sicherheitsforschung und Reaktortechnik of Forschungszentrum Juelich. This was necessary because the VKTA’s laboratory for higher activities was not yet in operation at this time.

For cementation of the fluid Westinghouse GmbH provided their mobile MOSS-200 facility. The campaign-specific qualified process was assessed by the product monitoring agency (PKS) Juelich and certified by Bundesamt fuer Strahlenschutz. (Fig. 6)

Under remote controlled conditions and with on line process parameter control and documentation the MAW solutions were solidified with cement into 200 liters drums. Within 6 month 307 drums were produced, which now are stored in concrete shieldings in the Rossendorf interim storage. (Fig. 7)

5 Rossendorf interim storage

The Rossendorf interim storage has a capacity of about 2000 m³ with an actual utilization of 20%. The licensed maximum activity is 5,0 E14 Bq with an actual utilization of 38%. (Fig. 8)

The storage concept considers safe long term storage of wastes for final disposal as well as decay storage for later release and includes a program of periodical control of stored waste arrangements. The objective of this concept is to minimize the waste for final disposal and to prevent additional conditioning measures by taking into consideration, that a final disposal in Germany will be available earliest in 30 years and that the requirements on conditions for final disposal are not fixed yet.
Decommissioning and Waste Reduction

Radiological Survey
- Monitoring
- Analytics

Analytics
- Leave
- Release
- Waste

- Calculation (10 μSv concept)
- e.g. Clearance monitor facility
- Declaration

Special features:
- Multiple nuclide vectors
- α-, β- and γ-radionuclides

Figure 1: Decommissioning and Waste Reduction

Figure 2: in-situ-gamma-ray spectrometry
Radiological survey: Radionuclide Analytics

α - particle spectrometry
- U- and Pu-isotops, 241Am

β - particle counting
- 3H, 14C, 65Fe, 60Ni, 90Sr

γ - ray spectrometry
- 60Co, 137Cs

Inductively Coupled Plasma Mass Spectrometry (ICP-MS) e.g. 238U, 235U

Radiochemical separation techniques

Figure 3: Radiological survey

Figure 4: Rossendorf research reactor (RFR)
Figure 5: Rossendorf reactor vessel

Figure 6: MOSS 200 (Westinghouse)
Figure 7: Solidification of fission product solution with MOSS 200 (Westinghouse)

Figure 8: Rossendorf interim storage
A Practical Approach to Volume Minimisation of Low Level Radioactive Waste at UKAEA, Winfrith

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Abstract
To fulfil the United Kingdom Atomic Energy Authority (UKAEA) policy on Best Practicable Environmental Option (BPEO), the requirements of the Radioactive Substances Act (RSA) 1993 and to meet the BNFL Drigg Conditions of Acceptance, the UKAEA Winfrith employ a range of techniques to minimise the volume of Low Level Radioactive Waste (LLW) consigned to BNFL Drigg for final disposal. To achieve this the UKAEA Winfrith Solid Waste Services Section commissioned a Plasma Cutting facility, a lead decomamination facility and the Winfrith Abrasive Cleaning Machine (WACM). These techniques coupled with regular supercompaction campaigns and a free release procedure ensure UKAEA Winfrith minimise the volume of LLW consigned to BNFL Drigg. This paper describes the waste minimisation techniques, the supercompaction campaigns and the free release monitoring programme employed at UKAEA Winfrith.

1 Introduction
Historically, the UKAEA pioneered the development of nuclear energy in the UK. Today, UKAEA are responsible for managing the decommissioning of the nuclear reactors and radioactive facilities in a safe and environmentally sensitive manner. As a result of the decommissioning programs large volumes of waste will be generated. UKAEA is committed to ensuring that the disposal of this waste is managed so that the volume of LLW sent for final disposal at BNFL Drigg is minimised. Waste volume minimisation is key to the UKAEA commitment to ensuring BPEO by preventing ineffective use of the BNFL Drigg facility and reducing the high costs and detrimental environmental impact associated with final disposal at BNFL Drigg. UKAEA achieve this by employing a process to segregate waste into items suitable for potential free release or LLW. This is in accordance with the Radioactive Substances Act 1993: SoLA (Substances of Low Activity) Exemption Order 1986 and waste containing radioactive materials not exceeding the LLW criteria respectively (4GBq/te α and 12 GBq/te β/γ) [1], [2]. UKAEA Winfrith has commissioned a waste processing facility designed to employ a suite of techniques to decontaminate various metal waste items which, in conjunction with the supercompaction of drummed waste, minimises the volume of LLW disposed of at BNFL Drigg. The methodologies employed are outlined in this paper.

2 UKAEA Winfrith Strategy to Minimise the Volumes of LLW Disposed of at BNFL, Drigg
UKAEA Winfrith have devised a strategy to minimise the volumes of LLW generated. This strategy encompasses volume minimisation at source through to the decommissioning of items using a purpose built facility. The processes involved are detailed below.

2.1 Free Release Monitoring Regime
The first stage in minimising the production of LLW is to determine, at the point of generation, whether a solid waste item is radiologically ‘clean’ or ‘exempt’ and therefore suitable for re-
cycling or free release consideration. 'Clean' materials are defined as those that are neither contaminated radioactive materials nor activated above background levels. 'Exempt' materials are defined as those materials containing man made radionuclides with specific activities less than those stated in the SoLA Exemption Order.

The SoLA order allows radioactive waste to be disposed of without authorisation provided that it is a solid, other than a closed source, which is substantially insoluble in water, the activity of which when it becomes waste does not exceed 0.4 Bq.g⁻¹. Naturally Occurring Radioactive Material (NORM) are exempt if the radionuclide activities are less than those stated in the Radioactive Substances Act (RSA) Schedule 1 or one of the exemption orders.

The criteria employed for items to be free released from the Winfrith site are:

- Specific Activity < 0.4 Bq.g⁻¹ and also
- Surface Contamination < 0.4 Bq.cm⁻² α, < 4.0 Bq.cm⁻² β/γ.

A planned strategy for a particular project includes the segregation of potentially 'clean' material at source based on knowledge of the area history. Material identified as being potentially clean, i.e. not contaminated or activated, is health physics monitored for re-assurance and if nothing is detected the material is segregated as suitable for free release. Contaminated materials are further segregated into those suitable for free release following decontamination and those suitable for disposal as LLW. Materials identified as LLW would be those considered unsuitable for decontamination, activated material or where radiochemical analysis has shown the specific activity to be > 0.4 Bq.g⁻¹. A suite of decontamination techniques are available at Winfrith to remove surface contamination on metal items therefore making them suitable for free release consideration following satisfactory health physics monitoring and compliance with the SoLA Exemption Order.

2.2 Supercompaction Campaigns

The volume of material segregated as LLW and requiring disposal at BNFL Drigg, is minimised by regular supercompaction campaigns at Winfrith. The BNFL Drigg Conditions of Acceptance (CoA) state that compatible waste is not acceptable. The majority of LLW items are packed into 220 litre drums at source. Larger items are packed into Half Height ISO (HISO) containers at source. Supercompaction campaigns are presently performed twice a year at Winfrith however the frequency is expected to increase with the increased production of waste from the decommissioning programs. The waste must not contain any free liquid, hazardous/special wastes (unless by prior agreement), pressurised containers, or vertical structural components which would not be compatible.

The supercompactor is a mobile facility manufactured by HANSA and owned by AEAT. It is contracted for use by UKAEA and managed by UKAEA personnel whilst at Winfrith. Two hundred and twenty litre drums are fed into the supercompaction chamber via a conveyor system. A sleeve is automatically placed over the drum to ensure its shape during supercompaction. The drum is compressed with a force of 2,000 tonnes, resulting in an average volume reduction of ~75%. The supercompacted 'puck' is automatically removed from the compaction chamber. The secondary liquid waste arisings from the process are collected and disposed of in accordance with the UKAEA Liquid Effluent Waste disposal procedures.

Approximately 250 supercompacted 'pucks' are loaded into an HISO container for grouting and final disposal at BNFL Drigg in comparison with the 36, two hundred and twenty litre drums that would be disposed of in HISO if supercompaction weren't performed. Typically, one thousand,
220 litre drums are processed in each supercompaction campaign, which equates to four HISOp for final disposal. During the last three year BNFL Drigg contract period, savings in excess of £300k have been achieved by supercompaction campaigns at Winsthth.

2.3 **Winfrith Plasma Cutting Facility**

The Winfrith decommissioning programs yield large metal waste items suitable for decontamination via the WACM facility or for disposal as LLW. Some of these items require size reduction to enable their processing through the WACM facility or disposal in HISOp container. A plasma cutting facility was commissioned at Winfrith in March 2000 to enable size reduction of such items. It is a hand operated 150 Amp facility capable of processing items of steel with a maximum thickness of 35mm. To date ~800 man-hours have been spent size reducing items. Large metal vessels and housings can be size reduced and flat packed for BNFL Drigg disposal thereby giving significant cost savings in Drigg disposal charges.

2.4 **Winfrith Abrasive Cleaning Machine (WACM)**

The extensive decommissioning program at UKAEA Winfrith is expected to produce significant quantities of contaminated fabricated structural steel. Normally, contaminated items are disposed of as LLW in 220 litre metal drums, with large items being double PVC bagged and disposed of in HISOp containers. The UKAEA Winfrith commitment to minimising the quantity of low level radioactive waste led to the commissioning of an abrasive cleaning machine to decontaminate these large steel items. The machine removes any painted surfaces and outer crud layer, effectively decontaminating the item, making it suitable for disposal either in accordance with the UKAEA free release criteria, to controlled land burial or for re-use. The items being processed through the WACM have been deemed suitable for free release following cleaning so the contamination is generally present in the painted or outer surface of the material. The WACM facility became operational in September 2001 and cost £250,000 (including filtration system and management costs).

The WACM consists of a contained and extracted cleaning machine that re-cycles and circulates the blast media at 1700kg/min. The blast media consists of a mixture of three grades of iron shot and grit. The larger grade shot provides a more aggressive cleaning medium to remove the majority of the painted/rusted surfaces. The finer grades of shot enable the removal of the paint and rust from within surface imperfections of the material. Less than 1% of the blast material is degraded through each decontamination process. The shot is re-cycled and topped up as necessary. The shot that is reduced to dust is segregated during the filtration process. The shot therefore undergoes a continuous self cleaning process that avoids the build up of residual contamination.

The item being decontaminated through shotblasting is loaded onto a conveyor system and is passed through a series of heavy duty curtain flaps into an inlet vestibule and then into a blast cabinet where it is subjected to the abrasive shot. The blasting operation starts automatically once the item enters the blast cabinet. From the blast cabinet the item feeds into an outlet vestibule through a series of heavy-duty curtain flaps and then to a 'blow-off' chamber where any remaining loose dust is removed. The blasting operation stops once the item leaves the blast chamber. Visual inspection of the item reveals any paint or crud that the system has failed to remove. The shot-blast process would be repeated for those items not visibly clean. On completion of the shot-blast process the item is monitored to determine if it meets the free release criteria as defined in section 2.1.

The shot-blast process is an air-less process i.e. the shot is not introduced into the chamber using compressed air, as would be expected, but is thrown into the chamber using 8 impellers. This
basically consists of four paddle blades with a hole in the middle with the shot passing through the hole and being accelerated towards the steel item by the motion of the rotating blades. A collection hopper located beneath the blast cabinet collects the abrasive material from the vestibules and blast cabinet. The abrasive material is removed to an air-wash separator where dusts and scales i.e. paint and rust, are separated from the re-cycled shot and collected into a 220 litre drum for disposal as LLW. The shot is continuously re-cycled with the quantity of shot within the process being monitored to ensure a constant level is maintained.

During operation, negative pressure within the vestibules and blast chamber is maintained by a two-stage filtered extract system. The shot blast process is a dirty process so if the primary filtration were a HEPA filter it would rapidly block with dust thereby making the process too expensive due to the frequent replacement of filters. The first filtration stage consists of a Donaldson-Torit (D-T) back-pulsing filter cleaning system. This basically consists of a cylinder housing eight inclined HEPA filters with extraction out of one of the ends. Dust particles stick on the filter surfaces so every 12 seconds, whilst the WACM is in use, compressed air is blasted into the D-T system to remove the particulates. The extract system removes the dust, which is collected in a catchpot for disposal as LLW. The second filtration stage consists of a HEPA filter to remove all particulates >0.6μm with 99.95% efficiency.

Waste items deemed suitable for processing through the WACM facility for potential free release must conform to the following criteria:

- Solid painted or surface contaminated metals which would otherwise be consigned to Drigg,
- No activated, hazardous or special waste,
- Specific Activity <0.4 Bq.g⁻¹,
- Loose Surface Contamination <0.4 Bq.cm⁻² α, <4.0 Bq.cm⁻² β/γ,
- Maximum β/γ contamination of 40Bq.cm⁻² and no hot spots >100Bq.cm⁻², maximum α contamination of 4Bq.cm⁻²,
- No assemblies,
- Cross-section not exceeding 450mm x 1,000mm; mass >2kg.

To date, approximately 20 tonnes of steel have been processed through the WACM. The projected throughput is >100 tonnes of steel per year operating on an occasional batch throughput. Throughput to date has not met the projected level due to issues relating to the presence of tritium ingress into the metal. The free release criteria specifies a total specific activity of < 0.4 Bq.g⁻¹. The Steam Generating Heavy Water Reactor (SGHWR) located at Winfrith is currently in a preliminary decommissioning phase and will be producing large quantities of steel. The steel from the secondary containment non-active services is considered suitable for free release following shot-blasting as any contamination has been shown to be present in the painted surfaces with no activation of the metal being present. Radiochemical analysis of the steel has however shown the presence of tritium in the metal at levels above the 0.4 Bq.g⁻¹ limit. The tritium has migrated into the metal following exposure to atmospheric tritium over the operational lifetime of the plant and not as a result of contamination. Therefore, the failure to meet the 0.4Bq.g⁻¹, due to the tritium content of the steel, has restricted the throughput of the WACM. The shot-blast process itself is currently being considered to determine whether sufficient surface material is removed to achieve the free release criteria. In addition a sampling and analysis program is being undertaken to determine the degree of tritium ingress into the metal.
One hundred tonnes of shotblasted steel would generate approximately 6-7 HISO containers, which would cost approximately £240,000 to £280,000 to dispose of at BNFL Drigg. The WACM machine itself cost £160,000 so in one year the cost of processing 100 tonnes of steel would cover the cost of the machine. Steel processed through the WACM after this point would result in a saving to UKAEA or to profit if the facility is used for commercial purposes. The projected cost savings associated with the WACM are £2m to £6m (discounted) over the 10 year estimated lifetime of the machine.

2.5 Lead Decontamination Facility

Large volumes of surface contaminated lead have been identified as being suitable for re-cycling or free release following decontamination. A lead decontamination facility was therefore commissioned in March 2000. This facility consists of a fibreglass modular containment with an extraction system similar to the one described for the WACM facility. The mechanical removal of the surface is performed by hand using simple powered wood planes. The UKAEA personnel performing the task are certified lead workers and wear full breathing apparatus during the operation. To date approximately 100 man-hours have been spent decontaminating approximately 4 tonnes of lead, predominantly small chevron blocks, which would have otherwise required final disposal at BNFL Drigg. Lead is considered to be special waste and would require additional justification for disposal at BNFL Drigg so volume reduction is essential.

3 Conclusions

UKAEA Winfrith minimise the volume of LLW sent for final disposal at BNFL Drigg by employing a range of radiological monitoring procedures, and waste processing and decontamination techniques. These processes are designed to demonstrate compliance with the UKAEA policy on BPEO, the requirements of the RSA 1993 and to meet the BNFL Drigg Conditions of Acceptance.

Radiological monitoring at source identifies ‘clean’ waste items to reduce the volume of waste requiring consideration for disposal as LLW. Regular supercompaction campaigns at Winfrith yield a 220 litre drum volume reduction of ~75% and a three year period saving for UKAEA Winfrith of >£300k in BNFL Drigg disposal costs. The plasma cutting facility enables large metal items and vessels to be size reduced giving significant cost savings to UKAEA in terms of BNFL Drigg disposal costs. The decontamination of metal wastes using the WACM facility enables their disposal as free release items reducing the volumes sent to BNFL Drigg for disposal in accordance with the UKAEA BPEO policy. One hundred tonnes of steel processed through the WACM facility saves £160,000 BNFL Drigg costs which equates to the cost of the machine. The facility could be operated at a saving to UKAEA or on a commercial basis for profit over its ten-year lifetime. The lead decontamination facility addresses the fact that BNFL Drigg imposes restrictions on the volumes of lead acceptable for disposal and decontaminates surface layers enabling free release of the bulk material.

These techniques ensure that UKAEA Winfrith minimise the volumes of LLW generated to prevent ineffective use of the BNFL Drigg facility and as a consequence reduce the costs associated with the final disposal of LLW.

4 References

STRATEGY ON RADIOACTIVE WASTE MANAGEMENT IN LITHUANIA

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Abstract
In Lithuania about 70-80% of all electricity is generated at a single power station, Ignalina NPP which has two non-upgradable RBMK-1500 type reactors. The Unit 1 will be closed by 2005. The decision on Unit 2 should be made in Lithuanian Parliament very soon taking into consideration substantial long-term financial assistance from the EU, G7 and other states as well as international institutions.

The Government approved the Strategy on Radioactive Waste Management in 2002. Objectives of this strategy are to develop the radioactive waste management infrastructure based on modern technologies and provide for the set of practical actions that shall bring management of radioactive waste in Lithuania in compliance with radioactive waste management principles of IAEA and with good practices in force in EU Member States.

Ignalina NPP is undertaking a program of decommissioning support projects, financed by grants from the International Ignalina Decommissioning Support Fund, administered by the European Bank for Reconstruction and Development. This program comprises also the implementation of investment projects in a number of pre-decommissioning facilities including the management of radioactive waste and spent nuclear fuel.

1 Introduction
There is only one nuclear power plant in Lithuania – Ignalina NPP. The Ignalina NPP operates two similar units with power rating of 1500 MW(e) and present power level of about 1250 MW(e) each. They were commissioned (first grid connection) in 12/1983 and 08/1987 respectively and provide approximately 70-80 per cent of the electricity produced in Lithuania. The original design lifetime was projected out to 2010-2015. The National Energy Strategy establishes that the first unit of Ignalina NPP will be shutdown before the year 2005, taking into consideration substantial long-term financial assistance from the EU, G7 and other states as well as international institutions. Regarding the second unit, the discussions are going on for earlier shut down till 2010.

Lithuanian Government approved strategy on management of radioactive waste on February 6, 2002 [1]. It defines activities for management of solid and liquid radioactive waste, and spent nuclear fuel at Ignalina NPP as well as radioactive waste from small producers. Ignalina NPP is undertaking a program of decommissioning support projects, financed by grants from the International Ignalina Decommissioning Support Fund, administered by the European Bank for Reconstruction and Development. This program comprises also the implementation of investment projects in a number of pre-decommissioning facilities including the management of radioactive waste and spent nuclear fuel.

State Enterprise Radioactive Waste Management Agency (hereafter referred to as “Agency”) has been founded in 2001 implementing the resolution of the Government. The Government also
approved the Agency activities program for 2002–2004. The main task of this program is to attain that the Agency performs all functions on radioactive waste management established by the Law on Radioactive Waste Management [2].

2 Strategy Objectives and Tasks

Objectives of the Lithuanian strategy on radioactive waste management are to:
- strive for implementation of proper radioactive waste management policy;
- develop the radioactive waste management infrastructure based on modern technologies;
- create the effective financing system for radioactive waste management;
- provide for the set of practical actions that shall bring management of radioactive waste in the Republic of Lithuania in compliance with the radioactive waste management principles of IAEA and with the good practices in force in EU Member States.

The tasks of this strategy are to:
- improve the legal basis for radioactive waste management;
- modernize a system of radioactive waste management at Ignalina NPP and to implement the new radioactive waste classification system according to the “Requirements on the Pre-disposal Management of Radioactive Waste at Nuclear Power Plant” approved by the order of the head of the State Nuclear Power Safety Inspectorate;
- be ready for the management of radioactive waste which will result from the Ignalina NPP decommissioning providing the plant with necessary radioactive waste management facilities;
- modernize the management infrastructure for radioactive waste generated by small producers;
- construct new repositories for radioactive waste in compliance with requirements of legal documents.

3 Strategy Implementation System

The Government of the Republic of Lithuania makes decisions on the design, construction, and decommissioning or permanent closure of specific storage facilities and repositories, sets aside in a prescribed manner plots of land for radioactive waste storage facilities and repositories, adopts legal documents for regulation of the acquisition, storage, transport and disposal of radioactive materials or entrusts the competent authorities with adoption of these legal documents in a prescribed manner.

Ministry of Economy submits to the Government proposals on the construction and decommissioning or permanent closure of storage facilities and repositories, organizes the international co-operation in the field of radioactive waste management, performs the functions of the founder of the Agency.

State Nuclear Power Safety Inspectorate controls and regulates the safety of radioactive waste management, storage and disposal facilities, and accounting of nuclear materials. Ministry of Environment, in co-ordination with the Ministry of Health Care, establishes the clearance levels and conditions for reuse and disposal of decontaminated materials, the maximal allowable levels of environment contamination with radionuclides and performs other functions established by
laws and other legal documents. Radiation Protection Center performs the state supervision and control of radiation protection.

Ignalina NPP is responsible for the safe management of radioactive waste produced during operation or accepted for storage or processing, and produced during decommissioning until this waste is transferred for disposal.

Agency is responsible for management and disposal of all transferred to it radioactive waste. Agency is the operator of the assigned to it storage facilities and repositories.

Radioactive waste management is financed by Ignalina NPP, small producers, state budget of the Republic of Lithuania, Ignalina NPP Decommissioning Fund, and International Ignalina Decommissioning Support Fund as well as bilateral co-operation and by other means.

4 Management of Solid Radioactive Waste at Ignalina NPP

Ignalina NPP produces about 1100 m$^3$ of solid radioactive waste per year. All solid wastes are divided into radioactive and nonradioactive (industrial) waste. All wastes in the controlled area are considered as radioactive until a dosimetric control is carried out. According to the old classification system from the former USSR solid wastes are considered to be radioactive if they meet one of the three criteria as follows:

- dose rate of gamma radiation is higher than $1 \times 10^3$ mSv/h at 10 cm of waste surface;
- specific activity is bigger than $7.4 \times 10^4$ and $7.4 \times 10^3$ Bq/kg for beta and alpha emitters, respectively;
- fixed surface contamination is higher than 500 and 5 particles/(min·cm$^2$) for beta and alpha emitters, respectively [3].

Former Soviet waste classification system was not suitable for proper waste segregation and preparation for disposal so new solid waste classification system based on the principles given by IAEA was adopted in Lithuania in 2001. Except for compaction of combustible low-level waste and cutting of long metallic pieces from reactor core and other high-active components, there are no conditioning processes for solid radioactive waste at Ignalina NPP now. Solid waste is stored at storage buildings in bulk. These storage facilities are above ground concrete structures and have a total storage capacity of 29000 m$^3$ solid waste. Long-term safety analysis of these storage facilities was performed and the waste retrieval concept was suggested. The stored waste should be retrieved for characterisation, classification and conditioning. Now these facilities are under the licensing process as storage facilities.

Solid radioactive waste generated during plant operation is collected and segregated into different groups depending on dose rate and composition (combustible and incombustible). Combustible 1st group (low level) waste is for most part wastes resulting from normal plant operation. Average annual generated amount is 260 m$^3$. It consists of overalls, personal protection means, paper and rags (about 40-50%), filters with a wooden frame (15-20%), timber and wooden trestle (15-20%), plastic waste, protective equipment, plastic film and rubber items (15-20%).

Incombustible 1st group waste consists of construction rubble (bricks, concrete) and other construction waste (35-40%), scrap metal (30-40%), insulation materials (15-20%), cable ends and other incombustible waste (5-10%). Average annual generated amount is 580 m$^3$. 


2\textsuperscript{nd} group waste (intermediate level) consists mainly of parts, units and consumable materials resulting from maintenance work on equipment of reactor coolant circulation circuit, gas circuit and turbine. 70 m\textsuperscript{3} of combustible and 110 m\textsuperscript{3} of incombustible 2\textsuperscript{nd} group waste are generated annually on average. Composition and constituent percentages of this waste and the 1\textsuperscript{st} group waste are virtually identical, only 2\textsuperscript{nd} group waste is more contaminated. Ventilation filters are collected in the transport corridor and packaged in to shielded transport containers or alternatively wrapped in plastic film and collected with other waste. Graphite waste, found mixed in Group 2 non-combustible waste, consists of graphite rings disassembled from spent fuel channels and CPS channels. The graphite rings are removed from the channels prior to cutting by using a special device which breaks the ring into pieces. Graphite waste must be treated separately because of the high contents of long-lived nuclides (mainly $^{14}$C).

Group 3 waste (long-lived), which is in general metallic (~90\%), is produced in the Hot Cells and Cutting Facilities which are special assemblies used to cut long metallic pieces from reactor core and other high-active components, to make the waste items fit shielded transport containers used in transport of waste of this kind. To protect the inner surfaces of transport container against contamination, a PVC bucket is used as a liner for Cutting Facility transport container.

Typical Group 3 waste items include central rods and tubes of spent fuel assemblies, sensors, joining pipes, Hot Cell filters, Control and Protection System (CPS) channels, CPS rods, fuel channels, additional absorbers, assembly channels, measuring channels, power detectors, emergency equipment, suspensions, etc.

According to the Lithuanian strategy, it is foreseen to modernize the management and storage of solid short-lived and long-lived radioactive waste of Ignalina NPP in 2002-2009 and to:

- reduce both the total activity and volume of radioactive waste, for such purpose to implement best available technologies;
- implement the new classification system for radioactive waste;
- arrange a licensed landfill for disposal of very low level waste;
- retrieve, characterize, to fulfill the conditioning of and to send back to storage facilities the solid short-lived radioactive waste accumulated in existing storage facilities;
- retrieve, and characterize the solid long-lived radioactive waste accumulated in existing storage facilities, fulfill its proper treatment, install proper interim storage facilities for long-lived radioactive waste, and store the long-lived radioactive waste in storage facilities without final immobilization until the final management methods are decided;
- create and implement the radioactive waste inventory record keeping system;
- strive for the direction of the radioactive waste for clearance as much as possible;
- perform investigations and prepare projects suggesting methodologies for calculation of conditional clearance levels and best management methods for materials with contamination exceeding unconditional clearance levels.

SKB-SWECO International-Westinghouse Atom Joint Venture with participation of Lithuanian Energy Institute has prepared a reference design of a near surface repository for short-lived low and intermediate level radioactive waste. This reference design is applicable to the needs in Lithuania, considering its hydro-geological, climatic and other environmental conditions and is able to cover the expected needs in Lithuania for at least thirty years ahead. Agency with participation of Ignalina NPP Decommissioning Projects Management Unit, Lithuanian Energy Institute and Swedish experts has already started drafting of preliminary Waste Acceptance Criteria for near surface disposal. State Nuclear Power Safety Inspectorate is drafting now the guide on siting of a near surface repository.
According to the Lithuanian strategy, it is foreseen to perform necessary investigations and draft recommendations on construction of a near surface repository until 2005.

5 Management of Spent Nuclear Fuel

The spent nuclear fuel is stored at reactors in the pools at least for 5 years. At Ignalina NPP site there is spent nuclear fuel dry interim storage facility for 72 casks. At present there are 20 ductile iron CASTOR-RBMK-1500 casks and 40 metal-concrete CONSTOR-RBMK-1500 casks in the dry storage facility.

According to the Lithuanian strategy, the best case would be to store the spent nuclear fuel in the dual-purpose storage systems applicable for both the long-term storage and transport. Until 2011, it is foreseen to expand the existing spent nuclear fuel dry long-term storage facility at Ignalina NPP or to construct a new facility in the vicinity.

Ready for storage spent nuclear fuel shall be removed to the dry storage facility in order that the Ignalina NPP decommissioning activities would be performed effectively.

Striving for the safe disposal of the spent nuclear fuel it is essential to:

- draft and implement the long-term research program “Possibilities to dispose of the spent nuclear fuel and long-lived radioactive waste in Lithuania”;
- analyze the possibilities to have in Lithuania a deep geological repository for spent nuclear fuel and long-lived radioactive waste;
- analyze the possibilities to create a regional repository taking joint efforts of a few countries;
- analyze the possibilities to dispose of the spent nuclear fuel in other countries, and to estimate the justification for a price of such disposal;
- analyze the possibilities to prolong the storage period in interim storage facilities for up to 100 and more years.

6 Management of Liquid Radioactive Waste at Ignalina NPP

The liquid waste is driven off and concentrated in an evaporation unit. The evaporator concentrate is fed to one of two bituminization units. The bitumen compound, having a salt concentration of about 40%, is transferred to the bituminized waste storage facility, which is an above ground concrete structure with 12 inside steel-coated canyons. It was determined that the existing storage facility may be acceptable for final disposal but further more detail analysis is necessary [4]. Now it is under the licensing process as storage facility. Ignalina NPP produces about 700 m$^3$ of bituminized waste per year.

The spent ion exchange resins and sludge of evaporator concentrates are currently being stored in the tank farm on site for liquid waste [5]. Ignalina NPP produces about 280 m$^3$ of spent resins plus filter aid per year.

According to the Lithuanian strategy, liquid radioactive waste shall be solidified and waste forms shall be enclosed into suitable containers as required for storage, transport and disposal in the near surface repository. Spent resins and sludge shall be cemented. Erection of the cementation facility of spent resins and sludge as well as new interim storage facility is now in phase of Basic
Design review by competent authorities. This cementation facility is expected to be operational by 2004. The liquid waste will be mixed with binding agent in an optimum ratio by a continuous operating cementation unit and directly filled into 200 l drums. After capping the drums will be loaded into a storage container (8 drums per container) or, if necessary, this container will be put into a shielding container. The throughput capacity of the cementation unit will be 450 m³/year. The waste package interim storage building for storage time of 60 years will be designed for a capacity of minimum 10 years of cementation plant operation.

Strategy indicates that investigations shall be performed and it shall be decided whether the bituminized radioactive waste storage facility can be converted into a repository or not. Depending on the decision, the bituminized radioactive waste storage facility shall be licensed as a repository or the bituminization technology shall be transformed in such a way that waste forms are enclosed into suitable containers as required for storage, transport and disposal in the near surface repository.

7 Management of Radioactive Waste Generated by Small Producers

Now small producers in Lithuania are generating only a few m³ of radioactive waste per year. Since the late 1980s radioactive sealed sources used in medicine, industry and research in Lithuania have been shipped to INPP. Over the years, until October 2000, the disused radioactive sealed sources were dumped, together with other wastes, into various storage areas of Buildings 155/1, 157 and 157/1 and they can be found in some compartments with all groups of waste. Low-intensity sources can be found packed in different kind of non-shielded packages mixed with other waste. Packing types include boxes (metal, wood, plastic, cardboard), drums, containers or plastic bags, etc. High-intensity sources are packaged into individual shielded containers or several sources have been packaged into one shielded container. These shielded containers can also be found mixed with other waste. In storages with Group 3 waste also unpacked sources can be found. Some of the gamma sources are installed to different kind of equipment.

Beginning October 2000, the disused sources have been stored separately from other waste in the compartment 18/3 in Building 157/1 reserved for this waste. All sources (> 17 000 pieces), still in their own-shielded packages, are loaded into 4 shielded cylindrical stainless steel containers for storage, which are then placed in the storage compartment.

According to the Lithuanian strategy, disused radioactive sealed sources shall be managed separately from other radioactive waste. These disused radioactive sealed sources that cannot be reused or sent back to the supplier shall be treated without the final immobilization until the acceptance criteria of a deep geological repository are established.

It is foreseen first of all to perform the safety analysis of existing “Radon” type radioactive waste facility near Maisiagala as for a temporary storage facility and, if the safety requirements are met, to perform the licensing. Later on, the investigations shall be performed and it shall be decided whether this facility can be converted into a repository or the site shall be after remediation released for the free use.

Radioactive waste generated by small producers and failed enterprises, the waste without owner and the illegitimate radioactive waste shall be collected, treated and temporary stored at Ignalina NPP storage facilities until a new near surface repository is constructed.
It is foreseen to modernize the technical basis for the treatment of radioactive waste generated by small producers. Now the Feasibility Study to establish a facility for the handling of institutional radioactive waste at Maisiagala is carried out.

8 Conclusions

1. Lithuanian strategy on radioactive waste management provides for the set of practical actions that shall bring management of radioactive waste in Lithuania in compliance with the radioactive waste management principles of IAEA and with the good practices in force in EU Member States.

2. Ignalina NPP is undertaking a program of decommissioning support projects which comprises the implementation of investment projects in a number of pre-decommissioning facilities including the management of radioactive waste and spent nuclear fuel.

9 References

Waste Acceptance, Requirement and Licensing
1. Introduction

The planning and construction of a repository require in particular the demonstration of its safety during the operational and post-closure phase. This process is predominantly determined by a site-specific safety assessment within the scope of which the required safety of a repository must be demonstrated. The results of the safety assessment are converted into quantitative waste acceptance requirements. In preparing such requirements the Bundesamt für Strahlenschutz (BfS-Federal Office for Radiation Protection) fulfills one of its obligations and, by this, provides guidance to radioactive waste treatment and conditioning. From a waste generator's and conditioner's point of view such requirements are of utmost importance for an appropriate selection of radioactive waste conditioning strategies and techniques.

2. Konrad and Morsleben Waste Acceptance Requirements

2.1 Derivation of Requirements

Since the early sixties, i.e. from its very beginning, the German radioactive waste disposal policy has been based on the decision that all types of radioactive waste (short-lived and long-lived) are to be disposed of in deep geological formations. Thus, BfS performed comprehensive safety-related planning work including the preparation of waste acceptance requirements for the Konrad and Morsleben disposal mines.

The Morsleben and Konrad waste acceptance requirements [1, 2] define the safety-related envelope or framework for radioactive waste intended for disposal in the respective repository. They do not include specific requirements on individually characterized or specified wastes. Both of them were prepared in such a way that a flexible system of requirements could be established, which is not only tailored to the radioactive waste presently generated but which also allows improvements and future developments in waste
conditioning techniques. Such a flexible system includes several alternatives and different options for the waste packages all of which ensuring the required level of safety for the repository in the operational and post-closure phase. The waste generators and conditioners thus need not meet “general” requirements but have the possibility of applying and fulfilling those requirements which are specifically applicable to the radioactive waste packages produced by them and intended for disposal. Naturally, such a flexible system of requirements is considerably extended and has become more complicated. The advantages offered to the waste generators and conditioners preponderate, however, over this apparent disadvantage.

2.2 The Konrad Waste Acceptance Requirements

The results of the Konrad site-specific safety assessment have been converted into both the design of the surface and underground facilities as well as in a system of waste acceptance requirements. This system is formulated in such a way that it first describes the general disposal-related aspects and the general requirements to be fulfilled by the waste packages and then develops into more specific requirements on the waste forms, the waste containers/packaging, the activity limitations on individual radionuclides, the documentation and the delivery of waste packages. A survey on the structure of the Konrad waste acceptance requirements is given in Table I.

Table I: Survey on the structure of the Konrad waste acceptance requirements

<table>
<thead>
<tr>
<th>1. General basic requirements on radioactive waste to be disposed of</th>
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<td>2. General requirements on waste packages</td>
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<td>- Surface contamination</td>
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<td>- Depressurized delivery</td>
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<td>3. Requirements on waste forms</td>
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<td>- Basic requirements</td>
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<td>- Waste form groups</td>
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</tbody>
</table>
4. Requirements on waste containers/packagings
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   - Waste container classes
   - Incident resistant packagings
   - Inner containers

5. Activity limitations
   - Permissible activities for individual radionuclides per waste package
   - Total activities
   - Declaration of radionuclides

6. Delivery of waste packages
   - Compliance with transport regulations
   - Permits
   - Marking of waste packages
   - Requirements on shipping units

2.3 The Morsleben Waste Acceptance Requirements

The structure of the Morsleben waste acceptance requirements is similar to that of the Konrad requirements. Nevertheless, there are two decisive differences:

(a) The operation of the Morsleben repository is regulated by the license issued on April 22, 1986, and by further documents pertinent to it. Thus, the Morsleben waste acceptance requirements prepared by BfS [2] include both boundary conditions prescribed in the license and additional regulations, in particular self-restrictions, results of supplementary safety assessments which keep to this framework and instructions by the self-surveillance of the Morsleben repository. According to this, at first sight, the Morsleben requirements appear to be rather complicated.

(b) The Morsleben waste acceptance requirements clearly distinguish between requirements on solid radioactive waste and on sealed radiation sources. Such a difference is not explicitly made within the Konrad requirements; they are formulated in a more general sense.

A survey on the structure of the Morsleben waste acceptance requirements is given in Table II.
Table II: Survey on the structure of the Morsleben waste acceptance requirements

1. Introductory remarks
2. Classification of radioactive wastes
3. General basic requirements on radioactive waste to be disposed of
4. Requirements on radioactive waste of type A1 (solid waste)
   - Basic requirements on waste forms
   - Activity limitations
   - Requirements on packagings
   - Packagings
   - Requirements on waste packages
   - Requirements on sealed radiation sources to be converted into type A1 waste
5. Requirements on radioactive waste of type A3 (sealed radiation sources)
   - Assignment of spent sealed radiation sources to waste types
   - Basic requirements on sealed radiation sources
   - Waste container classes
   - Packagings
   - Activity limitations
   - Surface contamination
   - Marking of waste packages
6. Declaration of radionuclides
7. Delivery of waste packages

3. Current Issues in Radioactive Waste Disposal

In Germany the Federal Government has made a pronounced change in energy policy since 1998, the most important feature of which is the abandoning or phasing out of nuclear energy use for electricity generation [3]. This has been performed in a stepwise procedure: in a first step so-called consensus talks with representatives of the utilities took place and in a second step respective legislative measures were taken. The basic document on the future use of nuclear energy for electricity production in Germany was initiated on June 14, 2000, and signed on June 11, 2001. According to this document, the Federal Government and the utilities agree to limit the future utilization of the existing nuclear power plants. The most important agreements refer to operational restrictions. The new policy is enforced by the latest amendment of the Atomic Energy Act which became effective on April 22, 2002.
According to the change in energy policy, the German radioactive waste management and
disposal concept is being reviewed and will be adopted due to political decisions, new
findings and specific evaluations [3]. The most important disposal-related issues are:

- A single repository in deep geological formations is sufficient for the disposal of all
types of radioactive waste. The disposal of high-level waste (HLW) by the year 2030 is
the political aim for the disposal of all types of radioactive waste.
- There are doubts with regard to the suitability of the Gorleben site. Therefore, its ex-
ploration shall be interrupted and further sites in various host rocks shall be investi-
gated for their suitability. The licensing procedure for the Konrad repository project
shall be terminated; the Morsleben repository shall be decommissioned.

Though the Federal Government has expressed doubts with respect to the suitability of
the Gorleben site, it is not considered to be unsuitable and will be included in the future
site selection process. According to the agreement between the Federal Government and
the utilities a further exploration of the Gorleben salt dome can contribute nothing to clarify
the doubts of the Federal Government. For this reason the underground exploration will
remain interrupted for at least 3, but at most 10 years (Gorleben moratorium); the morato-
rium became effective on October 01, 2000. A rapid clarification of these doubts has been
initiated; the respective investigations aim to the clarification of conceptual and safety-re-
lated issues. They are envisaged to be finished by the end of 2004.

The plan approval procedure for the Konrad repository project was completed. On May
22, 2002, the competent licensing authority Niedersächsisches Umweltministerium (NMU
- Ministry for the Environment of the Federal State of Lower Saxony) issued the license.
According to the agreement between the Federal Government and the utilities BfS with-
drew the application for immediate enforcement of the license on July 17, 2000, thus
enabling court examination of the license. This withdrawal in particular means that the re-
construction of the Konrad mine into a repository for all types of radioactive waste with
negligible heat generation (i.e., low and intermediate level waste) will only be possible
after final court decision. The court cases - at present five cases pending - are assumed to
last several years; then further decisions on the Konrad repository will have to be taken.

The Morsleben repository will not resume emplacement operations. An application for the
licensing procedure for decommissioning was already filed on May 09, 1997. At present,
activities are focussed on the preparation of respective documents to be handed over to
the licensing authority Ministerium für Landwirtschaft und Umwelt (MLU - Ministry for
Agriculture and Environment) in the Federal State of Saxony-Anhalt. According to the
present planning of BFS, this procedure might be finished in 2007.

According to the new approach to waste management and disposal, further sites in vari-
ous host rocks shall be investigated for their suitability. The final site shall be selected
upon a subsequent comparison of potential sites, including the Gorleben site. In Decem-
ber 2002, repository site selection criteria and respective procedures developed on a sci-
entifically sound basis were proposed by an interdisciplinary expert group. The criteria and
procedures aim at finding the relatively best suited site in different host rocks in Germany.
The principle of the site selection procedure to be developed is to identify - with public participation - potential disposal sites in a comprehensible and reliable way. Step by step and based on criteria which have to be defined beforehand, those areas, site regions and eventually sites shall be selected that offer particularly favourable conditions for the later demonstration of the site’s suitability and its confirmation in a licensing procedure.

4. Site-independent Waste Acceptance Requirements

4.1 Restructuring waste conditioning

Today in Germany there is no final disposal facility for any type of radioactive waste in operation. Due to this fact, there are no legally binding waste acceptance requirements, permitting final processing and packaging of radioactive waste. This will inevitably affect waste conditioning. On the one hand, the intention to start operation of a repository for all types of radioactive waste by the year 2030 means that the aspect of long-term engineered storage has to be taken into account in waste conditioning; on the other hand this situation will necessitate the current waste conditioning measures basically being planned and carried out in such a way that - depending on necessities and licensing-relevant framework - a certain flexibility should be granted with respect to future waste conditioning steps which cannot be established obligatorily at present. This should be understood as a contribution to reduce both the effort to meet future waste acceptance requirements and the additional radiation exposure of the personnel. Such consequences should mainly affect the conditioning of radioactive waste with negligible heat generation, as the conditioning of heat-generating radioactive waste - except that of spent fuel elements for direct disposal - is determined by the techniques and equipment installed in foreign reprocessing plants (COGEMA, BNFL). However, it cannot be excluded that even in these cases additional measures (e.g., with respect to packaging) might turn out to become necessary in future.

Thus, the long-term engineered storage above ground imposes certain safety-related requirements on the quality of intermediate or waste products and on the waste containers or packages, respectively, as well as in particular on the respective documentation. These requirements may in particular comprise

(a) Selection of suitable waste treatment and conditioning methods.
(b) Production of inherently stable, inert and dry intermediate and final waste products (depending upon the respective waste type).
(c) Application of corrosion preventive measures (drums and sheet steel containers).
(d) If necessary, additional measures aiming at reducing the residual humidity in a waste package.
(e) Use of suitable filters to prevent the formation of internal overpressures.

Keeping in mind that interim storage should be carried out in a safe manner - if possible, without additional maintenance and inspection measures - it cannot be excluded that
individual requirements will exceed, e.g., those defined in the Konrad waste acceptance requirements. This applies especially to residual humidity, the inner and outer corrosion prevention of steel packages as well as content and extent of the waste package documentation.

4.2 Recommendations Concerning Future Waste Conditioning Measures

As the Federal Government continues to pursue disposal of radioactive waste in deep geological formations, the concept and structure of waste acceptance requirements to be prepared in future should deviate only insignificantly from those for the Konrad repository [1]. It is to be expected that again a subdivision into basic requirements on radioactive waste to be disposed of, requirements on waste packages, waste products and waste containers as well as on acceptable radioactivity limits and on delivery of waste packages will be found.

Especially from the intended disposal of conditioned radioactive waste in a mine suitable for disposal it may be concluded that

(a) basic requirements on radioactive waste to be disposed of,
(b) requirements on waste packages,
(c) basic requirements on waste products and waste containers

will not be modified basically as compared to the Konrad waste acceptance requirements. At least, this should also apply to parts of the additional requirements which will be imposed on the quality of the waste product and the waste container from a safety point of view. Arguments supporting this position are expected to be deduced from the results of the research project SR 2379 "Development of site-independent requirements on the treatment of radioactive waste to be disposed of". This research project initiated by BMU and technically accompanied by BFS is performed on the basis of internationally available information about concepts for conditioning, transportation as well as interim storage and disposal of radioactive waste. The planned investigations shall support BMU/BFS in the assessment of basic safety-related problems concerning the requirements on different types of radioactive waste foreseen for interim storage and disposal as well as their conditioning under particular consideration of the international standard.

As long as compulsory waste acceptance requirements for a repository in deep geological formations do not exist or a repository is not available for operation, specific methods and techniques as well as the measures to be applied have to aim at achieving both those requirements which will ensure safe long-term engineered storage above ground and the basic requirements on radioactive waste, waste packages, waste products and waste containers including, if necessary, additional requirements on waste products and waste containers.

With regard to disposal it should be pointed out that conditioning of radioactive waste should take into account a certain flexibility. Care should be taken that in the future – if
necessary — additional (i.e., final) conditioning steps can be carried out as simply as possible avoiding significant radiation exposures to the personnel. Such a flexibility could be achieved by, e.g., only packaging intermediate and final waste products into suitable containers without grouting. From the current point of view grouting or immobilisation of waste products would only be advisable after obligatory waste acceptance requirements for disposal will have been prepared. On the other hand, it may become necessary for different reasons to store drums now or in the years to come in containers and to fill up the residual voids. In such cases the waste package quality assurance should have been performed successfully before filling up and comprise the assessment of the corresponding documentation with respect to completeness and comprehensibility.

The existing and continuously generated radioactive waste from operation and decommissioning of nuclear facilities has to be processed — as already stated — in such a way that intermediate products suitable for interim storage will result. At a later time these intermediate products will finally be conditioned to produce waste packages complying with future waste acceptance requirements. A comparable situation existed in Germany when radioactive waste which was stored until 1994 in interim storage facilities had to be re-treated and re-conditioned prior to acceptance and disposal in the Morsleben repository. Normally, sufficient information about composition, origin and radionuclide inventory was not available for radioactive waste older than five years. The re-qualification of this kind of waste before its final conditioning in order to fulfill the 1996 Morsleben waste acceptance requirements [2] partially meant considerable efforts. As contrasted to this, for a great number of waste packages the information concerning radionuclide inventory, waste product properties and waste origin could be extracted from the documentation and used successfully to demonstrate compliance with the Morsleben requirements. Therefore, information and data on radionuclide inventories, waste properties and waste origin including the various waste treatment and conditioning steps have to be collected and carefully documented to facilitate the future steps towards disposal without significant problems.

Thus, in order to bridge the gap between present necessities and future requirements, two different routes could be followed:

(a) A future repository in deep geological formations must be designed and planned in such a way that the already existing waste packages and those to be produced until the start of operation of this facility can be accepted for emplacement.

(b) Preparation of so-called "Site-independent waste acceptance requirements", i.e. a compilation of all presently known requirements on waste packages intended for disposal including a recommendation how to apply such site-independent waste acceptance requirements.

In the following the second route with regard to radioactive waste with negligible heat generation (i.e. low- and intermediate-level waste) will be described.

The "Requirements on radioactive waste to be disposed of (Waste acceptance requirements, as of December 1995), - Konrad Mine -" [1] had been prepared with respect
to disposal of radioactive waste with negligible heat generation in deep geological formations and were assessed intensively in the licensing procedure for the Konrad repository project. They compile in detail the basic requirements on radioactive waste to be disposed of, on waste packages, waste products and waste containers as well as additional requirements on the quality of the waste packages. Thus, with respect to an evaluation of the present situation, it is recommended to carry out the future conditioning of radioactive waste with negligible heat generation using the Konrad waste acceptance requirements (as of December 1995) as guideline. The main objectives for this recommendation are the following:

(a) Emplacement of radioactive waste in deep geological formation is still the preferred disposal route in Germany.
(b) Planning, construction and operation of a disposal mine is to be expected in future.
(c) Thus, only insignificant deviations from the basic requirements on waste disposal, waste packages, waste forms and waste containers/packagings are to be expected.
(d) At present, no other comprehensive and quantitative waste acceptance requirements are available in Germany.
(e) The Konrad waste acceptance requirements were approved by the competent licensing authority and their experts.
(f) The fulfilment of the Konrad waste acceptance requirements forms an integral part of various interim storage facility acceptance criteria.

In addition to the Konrad waste acceptance requirements (as of December 1995) flexibility in their application is to be ensured and additional requirements are to be taken into account. Since neither a site for a repository nor the results of a site-specific safety assessment are available, most of the additional requirements will be qualitative, not quantitative. Thus, the site-independent waste acceptance requirements still to be prepared may in particular comprise:

(a) Use of the Konrad waste acceptance requirements (as of December 1995) as basic guideline.
(b) Ensuring flexibility in the application of the Konrad requirements.
(c) Consideration of additional (qualitative) requirements:
   - Corrosion preventive measures for steel packages (drums, containers).
   - Gas generation (residual humidity content; inventory of metallic and organic constituents).
   - Treatment of organic complexing agents (e.g., EDTA and NTA).
   - Declaration of additional radionuclides (incident analysis; post-closure safety assessment – cf. the Morsleben waste acceptance requirements).
• Declaration of waste package constituents, i.e. relevant chemical substances constituting a potential contamination hazard for near-surface groundwater according to the Groundwater Ordinance of March 18, 1997.

• Compilation of an extensive and comprehensive documentation: characterization of waste products, waste containers and/or waste packages (reliable basis for a future verification of waste package properties).

• Additional requirements imposed by the competent authority within the Konrad licensing procedure.

5. Conclusions

Radioactive waste treatment and conditioning should in particular consider the requirements originating from the long-term engineered storage and the basic requirements on radioactive waste disposal, waste packages, waste forms and waste containers/packaging as given in the 1995 Konrad waste acceptance requirements. A flexibility within radioactive waste treatment and conditioning should in particular be maintained in order to avoid future re-conditioning measures and additional radiation exposures to the staff. The so-called "Site-independent waste acceptance requirements" should immediately be prepared and implemented to provide assistance and guidance to the waste generators and conditioners.

6. References


WASTE ACCEPTANCE: PROCEDURES, CRITERIA AND BACKGROUND

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Abstract

After an introduction to the Swiss waste management concept the waste acceptance procedures and the corresponding documents are briefly described. Then, the provisional waste acceptance criteria are discussed and the key requirements described. This includes quantitative and qualitative requirements on waste properties. The safety relevant properties (e.g. nuclide inventory, type of waste matrix and package, chemical properties) are assessed by means of a simplified safety analysis - which is applied on a case-by-case basis for each waste type. Finally, the paper summarises the experiences up to now and touches upon related activities of waste characterisation and documentation which are described in more depth in an accompanying paper.

1 Introduction and Overview

The Swiss nuclear disposal concept foresees two types of final waste repositories, namely:

- The L/ILW repository for low- and intermediate-level waste arising from the operation and decommissioning of Swiss nuclear power plants, and from medicine, industry and research, as well as for low-level technological waste from reprocessing. The repository will consist of mined caverns with horizontal access, located in a suitable geological formation.

After a site evaluation procedure lasting many years, in 1993 Nagra selected the Wellenberg (WLW) site in Canton Nidwalden for further investigation. In 1994, the "Genossenschaft für Nukleare Entsorgung Wellenberg" (GNW, Wellenberg disposal cooperative) - which was established specifically with the purpose of constructing and operating the L/ILW repository - submitted the application for the federal general licence for a L/ILW repository at the Wellenberg site, and a request for a mining-concession for the repository was made to the Canton of Nidwalden where the site for the proposed Wellenberg L/ILW repository is located. At a cantonal public referendum in June 1995 the granting of the mining-concession was rejected by a narrow margin (52 to 48%). Within the framework of the general licence application, the safety authorities review came to positive conclusions. However, because the project was blocked on the political level, the general licence procedure has been suspended since 1997.

In order to take into account public concerns (mainly monitoring / retrievability and public involvement in decision-making), the strategy for repository implementation has been adapted and a more step-wise approach is foreseen, where in a first step the concession will be restricted to an exploratory drift only and a concession for the repository will only be applied for after the results from the exploratory drift are available and positive. The repository project has also been modified to include a phase of long-term monitoring and more easy retrievability.

After a lengthy process, it became possible to submit an application for a concession for the exploratory drift at the end of January 2001. The government of the Canton of Nidwalden has issued the concession in September 2001. Several objections have been raised about the concession but these have been dismissed by the government and subsequently also by court. The cantonal public referendum on this concession takes place on 22 September 2002.
• The **HLW/TRU repository** for vitrified high-level and long-lived intermediate-level waste (primarily resulting from fuel reprocessing) and for direct disposal of spent fuel elements. The repository will be located in a deep geological formation and will consist of a drift system for in-tunnel emplacement of HLW and spent fuel and tunnels for long-lived intermediate-level waste, with access via vertical shaft and ramp.

Within the HLW repository programme, the crystalline bedrock has been investigated in the 80's and early 90's with a very extensive regional programme and the results have been summarised in a comprehensive evaluation in 1994. Currently, the main focus is on Opalinus Clay, a clay rich sediment. The next milestone of the HLW programme is to demonstrate that a safe repository can be implemented and that a corresponding site does exist ("Siting Feasibility"). As part of this milestone Nagra will also propose a potential siting area as a focus for future work for the HLW repository. The corresponding documentation will be submitted to the authorities at the end of this year for review which eventually will lead to a decision by the federal council on how to proceed. Disposal of HLW and Spent Fuel within the framework of a multinational project continues to be an option.

For planning purposes Nagra assumes approximately 80'000 m³ L/ILW, 6'500 m³ long-lived ILW and approximately 9'500 m³ HLW and spent fuel (volumes refer to packaged wastes). To date, only a small proportion of these wastes has arisen. Until the planned repositories become operational, all waste categories are held in interim storage.

Although the details on repository layouts have not yet been fixed definitively, it is nevertheless meaningful, at this stage, to prepare the way for future acceptance of wastes into the repositories: It has to be ensured that wastes arising today are conditioned in such a way that they can be disposed of in one of the planned repositories. It is also important that waste-related data which will be required for disposal purposes in the future are collected at the stage of waste conditioning to avoid the need for any subsequent (very costly) characterisation of the fully conditioned waste packages.

Today, a system is implemented which provides the waste producers with the boundary conditions to be followed when conditioning their wastes and that provides the repository planner with all necessary data up to the point of emplacement of the waste in the repositories. The system is structured in such a way as to allow the waste producers as much flexibility as possible: For example, the provisional acceptance criteria contain as few as possible binding technical requirements, allowing the waste producer and Nagra to define optimal case-specific requirements for each waste stream to be conditioned. The system as implemented also takes into account the provisions of the Swiss nuclear regulatory authorities, see HSK/R-14 Guideline [1].

The system requires good co-ordination between the repository operator (or in the present phase the repository planner, Nagra) and the waste producers. An agreement has been concluded in this respect. According to this agreement, the collaboration and mutual exchange of information between the repository planner and the waste producer begins at the stage of developing the waste conditioning process and continues up to the point of acceptance of the waste package into the repository. All these steps are carried out under the supervision of the Swiss nuclear regulatory authorities. A detailed description of the administrative steps can be found in section 2, while key technical issues of this process are described in section 3.
2 The Concept of Waste Documentation and Acceptance Procedures

Waste documentation has to contain all information which is required for planning the repositories and for providing a sound technical and administrative basis for future waste acceptance into the repositories. It consists of (see Fig. 1):

- waste type documentation, consisting of specification which takes into account the results of any prototype testing and issues from pertinent waste characterisation programmes
- individual waste package documentation, consisting of a waste package datasheet including the results of the quality control programme, an interim storage logbook and results from the waste characterisation programme

The waste package type specification is a technical document in which the waste producer describes and specifies the following aspects for a set of similar waste packages:

- Manufacturing conditions (conditioning procedures and plant)
- Structure and properties of the package and its components (nominal, band-widths, guarantees)
- Quality assurance measures
- Datasheet specimen

The datasheet contains selected parameters which are specific to the individual waste package. It documents the details of the conditioning process and, together with the corresponding waste package type specification, it provides an adequate characterisation of the individual packages.

The acceptance procedure consists of four steps (see Fig. 1):

- The provisional disposability certification procedure which is initiated following the development of a new conditioning process in cases where prototype testing is required. The result is a provisional disposability certificate, based on Nagra's provisional assessment of the conditioning process, the provisional waste package type specification (including datasheet specimen), the prototype testing programme and the draft quality control programme.
- The disposability certification procedure. In this procedure Nagra
  - definitively accepts various components of the waste documentation (waste package type specification, datasheet specimen)
  - concludes binding agreements with the waste producer regarding the quality control programme and the waste characterisation programme
  - confirms formally that the waste package type in question can be integrated into the Swiss disposal concept (provisional repository allocation).

The result takes the form of the disposability certificate with a corresponding technical assessment.

- The pre-acceptance procedure which is intended to confirm that individual packages conform to the waste package type according to their datasheet.
- The acceptance procedure which leads to actual emplacement of packages in the repository.

Formal disposability assessments and certifications have been carried out since 1989. To date, most of the important (in terms of quantity) NPP operational waste streams (resins, concentrates, compactable mixed wastes, incinerable mixed wastes) have been assessed. Nagra's evaluations of the waste specifications of the reprocessing companies were based on the same strategy.
Figure 1: Concept for waste package acceptance and documentation
The following tools were developed as part of the formal disposability assessment and are continually being updated to take account of new information:

- The computer-based information system for radioactive wastes and materials (ISRAM), which is used by the Swiss waste producers for
  - presenting the structure and characteristics of waste package types and datasheet specimens (appended to the waste package type specification)
  - creating and managing datasheets for each waste package produced
- Detailed procedural rules for disposability certification and pre-acceptance
- Repository-specific technical assessment standards (provisional waste acceptance criteria for both repositories, Simplified Safety Analysis (SISAN) used in the disposability assessments)

Still open at present are detailed concepts for the acceptance procedures.

### 3 Safety relevant Waste Properties and Contents of the Provisional Waste Acceptance Criteria

When discussing the safety relevant waste properties it is convenient to distinguish between the operational phase and the post-closure phase. The operational phase of a geological repository is in principle comparable to the operation of an interim storage facility. The most important safety relevant processes and the safety-determining factors for the operational phase of a repository are summarised in Table I.

For the Swiss repository concepts, in the post-closure phase groundwater is the most important pathway for any potential radionuclide releases. For the analyses of this release scenario it is convenient to divide the system into three components: the nearfield, the geosphere and the biosphere. The nearfield (consisting of the disposal rooms with the wastes and the engineered barriers and a few meters of directly surrounding host rock) ensures that only very small release rates will occur because of the very low water flow rates and retention by geochemical phenomena. Transport through the nearfield and the geosphere does take such a long time that the majority of radionuclides decays before they can reach the environment. The long transport times are again mainly due to the small water flow rates and the geochemical retention processes.

The good performance of the nearfield and the geosphere can in principle be perturbed by some of the waste properties:

- Some materials in some of the wastes (or their degradation products) may affect the geochemical conditions such that geochemical retention (sorption and/or low solubilities) will be significantly reduced
- Excessive swelling or shrinkage can lead to fissures in the engineered barriers and thus lead to preferential flowpaths which may lead to enhanced nuclide release rates
- Corrosion and degradation of some waste materials can lead to significant gas generation rates. Large gas generation rates can lead to enhanced radionuclide release rates e.g. due to expulsion of contaminated nearfield porewater.

Based on these considerations for both the operational and post-closure phase quantitative and qualitative criteria were developed for the preliminary waste acceptance criteria. Key criteria are summarised in Table II.
Table 1: Safety relevant processes and factors for the operational phase of a repository

<table>
<thead>
<tr>
<th>Process</th>
<th>Important factors</th>
<th>Waste package</th>
<th>Repository</th>
</tr>
</thead>
<tbody>
<tr>
<td>Normal operation</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Direct radiation</td>
<td>• Nuclide inventory</td>
<td>• Shielding, operational procedures, remote handling, etc.</td>
<td></td>
</tr>
<tr>
<td></td>
<td>• Shielding by package</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>• Shielding by waste matrix</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>• Number of packages</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Release of volatile nuclides</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>• Nuclide inventory (average)</td>
<td>• Ventilation</td>
<td></td>
</tr>
<tr>
<td></td>
<td>• Gas tightness of package</td>
<td>• Backfilling of disposal rooms</td>
<td></td>
</tr>
<tr>
<td></td>
<td>• Release of volatile nuclides from waste matrix</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>• Number of packages</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Incidents &amp; accidents</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Release of airborne nuclides</td>
<td>• Nuclide inventory (maximum)</td>
<td>• Operational procedures, handling equipment, transport container, disposal container</td>
<td></td>
</tr>
<tr>
<td>due to mechanical impact</td>
<td>• Dispersability of waste matrix</td>
<td>• Remote handling</td>
<td></td>
</tr>
<tr>
<td></td>
<td>• Mechanical protection by package</td>
<td>• Ventilation</td>
<td></td>
</tr>
<tr>
<td>Release of airborne nuclides</td>
<td>• Nuclide inventory (maximum)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>due to thermal impact</td>
<td>• Thermal stability of waste matrix</td>
<td>• Operational procedures, handling equipment, transport container, disposal container</td>
<td></td>
</tr>
<tr>
<td></td>
<td>• Thermal protection by package</td>
<td>• Remote handling</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>• Ventilation</td>
<td></td>
</tr>
</tbody>
</table>

Within the waste acceptance process (disposability certification, see Figure 1) the waste type specification is evaluated with respect to these criteria on a detailed case-by-case basis. The evaluation of the qualitative criteria (described as target (qualitative) in Table II) requires expert judgement. Most of these qualitative criteria, however, are also used to derive the input parameters for a simplified safety analysis (SISAN) in which compliance of waste-type specific dose rates for a spectrum of different scenarios is checked. These scenarios include both the operational and post-closure phase. The waste properties that directly feed into the input parameters for SISAN are summarised in Table III. Some of the input parameters cannot be directly taken form the waste type specifications but do require some pre-evaluation; these are summarised in Table IV. The subsequent dose calculations by SISAN are then used to assess the acceptability of the proposed nuclide inventory (both the average and the maximum) and to fix this inventory for the waste-type on a case-by-case basis. SISAN itself is an abstraction from a detailed model chain used in performance assessments for site and design evaluations. It is run on a PC and produces the required results within seconds.
Table II: Key criteria in preliminary waste acceptance criteria (examples, not comprehensive)

<table>
<thead>
<tr>
<th>Component</th>
<th>Property</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>Waste package</td>
<td>• No free liquids, no free gas</td>
<td>binding</td>
</tr>
<tr>
<td></td>
<td>• No explosive, pyrophoric or chemically unstable materials</td>
<td>binding</td>
</tr>
<tr>
<td></td>
<td>• Integrity of package not endangered due to gas generation</td>
<td>binding</td>
</tr>
<tr>
<td></td>
<td>• Stability/integrity of waste package until time of disposal</td>
<td>binding</td>
</tr>
<tr>
<td></td>
<td>• Maximum size</td>
<td>binding (quantitative value)</td>
</tr>
<tr>
<td></td>
<td>• Maximum weight</td>
<td>binding (quantitative value)</td>
</tr>
<tr>
<td></td>
<td>• Specific heat generation</td>
<td>target value (quantitative value)</td>
</tr>
<tr>
<td></td>
<td>• Specific gas generation</td>
<td>target value (quantitative value)</td>
</tr>
<tr>
<td></td>
<td>• Nuclide inventory (average, maximum), surface dose rate</td>
<td>binding (quantitative value fixed on case-by-case basis)</td>
</tr>
<tr>
<td></td>
<td>• Minimisation of swelling/shrinking materials</td>
<td>target (qualitative)</td>
</tr>
<tr>
<td></td>
<td>• Minimisation of voidage</td>
<td>target (qualitative)</td>
</tr>
<tr>
<td></td>
<td>• Minimisation of burnable materials</td>
<td>target (qualitative)</td>
</tr>
<tr>
<td>Waste matrix</td>
<td>• Solid/hard waste matrix</td>
<td>binding</td>
</tr>
<tr>
<td></td>
<td>• Compressive strength</td>
<td>binding</td>
</tr>
<tr>
<td></td>
<td>• Poor degradability by water</td>
<td>target value (quantitative value)</td>
</tr>
<tr>
<td></td>
<td>• Homogeneous distribution of nuclides</td>
<td>target (qualitative)</td>
</tr>
<tr>
<td>Package</td>
<td>• Compressive strength of filling material</td>
<td>binding</td>
</tr>
<tr>
<td></td>
<td>• Poor degradability of filling material by water</td>
<td>target value (quantitative value)</td>
</tr>
</tbody>
</table>
Table III: Input parameters for the simplified safety analysis (SISAN) used for disposal certification as a function of the different scenarios assessed

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Sceneario</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Operational phase</td>
</tr>
<tr>
<td></td>
<td>Release of volatile nuclides during normal operation</td>
</tr>
<tr>
<td>Waste volume</td>
<td>X</td>
</tr>
<tr>
<td>Nuclide inventory</td>
<td></td>
</tr>
<tr>
<td>Average</td>
<td>X</td>
</tr>
<tr>
<td>Maximum</td>
<td></td>
</tr>
<tr>
<td>Speciation</td>
<td></td>
</tr>
<tr>
<td>Leakage rate of package</td>
<td></td>
</tr>
<tr>
<td>Waste matrix class 1)</td>
<td>X</td>
</tr>
<tr>
<td>Package class 1)</td>
<td></td>
</tr>
<tr>
<td>Waste group 1)</td>
<td></td>
</tr>
<tr>
<td>Handling route 1)</td>
<td></td>
</tr>
</tbody>
</table>

1) The allocation of the waste package to the classes, route and group is determined by the factors summarised in Table IV

Table IV: Factors to determine the classes, group and route for a waste package as needed in Table III

<table>
<thead>
<tr>
<th>Property of waste package (or component)</th>
<th>Parameter</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dispersability of waste matrix ($\leq 60^\circ$C)</td>
<td>Waste matrix class</td>
</tr>
<tr>
<td>Burnability of waste matrix</td>
<td>X</td>
</tr>
<tr>
<td>Protection of waste matrix due to package (mechanical impact)</td>
<td></td>
</tr>
<tr>
<td>Protection of waste matrix due to package (thermal impact)</td>
<td></td>
</tr>
<tr>
<td>Inventory of substances with the potential to affect nuclide release</td>
<td></td>
</tr>
<tr>
<td>Form / size of waste package</td>
<td>X</td>
</tr>
<tr>
<td>weight of waste package</td>
<td>X</td>
</tr>
<tr>
<td>Surface dose rate</td>
<td>X</td>
</tr>
</tbody>
</table>
4 Summary and Conclusions

- Specific concepts have been formulated for the management of radioactive wastes arising in Switzerland from the use of nuclear energy and application of radioactive substances in the fields of medicine, industry and research. The strategy foresees two types of repository: one for low- and intermediate-level waste (L/ILW) and one for high-level and long-lived intermediate-level waste (HLW/TRU). For both repositories detailed concepts do exist.

- The safety analyses performed for the L/ILW and HLW repositories demonstrate that safe disposal is feasible in both cases.

- Although there is, as yet, no operating repository in Switzerland, waste packages are checked by Nagra on behalf of the future repository operators in terms of their suitability for disposal. This ensures (a) that an adequate planning basis for future repositories is available and (b) that documentation exists for each waste package which contains all information which will be required in the future.

- The formal disposability assessment and certification forms part of the procedure agreed between Nagra and the waste producers for waste acceptance. The disposability certification also fulfils the requirements of the HSK/R-14 Guideline as a precondition for official clearance of new conditioning processes.

- Those waste properties that are essential for the safety and smooth operation of a repository have been identified and assessed in detailed studies. The results of these studies have been the basis for formulating provisional waste acceptance criteria. These waste acceptance criteria are complemented by a calculational tool used for a simplified safety analysis (SISAN) to assess the safety of waste types on a case-by-case basis.

- Both the provisional waste acceptance criteria and the calculational tools used (the information system for radioactive waste and materials (ISRAM) and SISAN) have been successfully applied since the early 90's.

5 References


INTEGRATED SYSTEM FOR LONG-TERM RADIOACTIVE WASTE MANAGEMENT IN THE UK

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Abstract
Since the failure of Nirex application to build a Rock Characterisation Facility near Sellafield in 1997, Nirex has been applying lessons learnt from that failure. Some of the issues involved are generic and relate to the process by which legitimate authority can be gained for government policy development, the structure of the nuclear industry and the behaviour of institutions. Transparency must be central to the culture of organisations attempting to win public acceptance. In the UK, the Department for Environment, Food and Rural Affairs (DEFRA) have started a consultation process – Stage one completed in March 2002 – to consult about the safe management of radioactive waste.

Nirex has modified its approach to long-term waste management, using a concept of phased (stepwise and reversible) geological disposal. Nirex also provides waste producers with advice on, and endorsement of, the packaging and transport of wastes. Through these examples, this paper will demonstrate how Nirex is providing an integrated approach to the long-term management of radioactive wastes in the UK.

1 Introduction
The Nuclear Industry Radioactive Waste Executive (NIREX) was set up in 1982 to research, develop and operate radioactive waste disposal facilities on behalf of the nuclear power industry. In 1985 it became a limited company (UK Nirex Ltd.), with shares owned by the major waste producers, and a special share held by the Government to give it safeguarding powers. The Ministry of Defence contributes funding, but is not a shareholder. Nirex’s current purpose is to provide the UK with safe, environmentally sound and publicly acceptable options for the long-term management of radioactive materials. Given the history, Nirex has concentrated its consideration to intermediate-level waste and some low-level waste (ILW and LLW) and in support of this has developed its Phased Disposal Concept as one way of managing these wastes in the long-term.

In the UK, work on finding a solution to long-lived radioactive waste has been on-going since the 1970s. The decision to focus the search for a disposal site for intermediate-level wastes at a location close to Sellafield, Cumbria took place in 1991. This led on to a public inquiry in 1995-96 into the planning application made in 1994 to build an underground laboratory or Rock Characterisation Facility (RCF). The outcome of the planning inquiry was rejection of the Nirex application, which effectively stopped any further work on finding a solution. Since the rejection of the planning application in 1997, Nirex has been considering what went wrong so that any future attempt to find a solution to this difficult problem, can benefit from the mistakes of the past.

Nirex has not been alone in the UK in considering how we move forward. The House of Lords Science and Technology Committee undertook a study [1] on radioactive waste management and reported in 1999. One of their findings was that no consensus existed and for any solution to work, there must be a degree of agreement as to the solution. They also recommended an
integrated approach to waste management. One option for achieving this is the combined (co-
disposal) of high-level waste and spent fuel with solid intermediate-level and low-level waste
within a single facility [2]. Similarly the Radioactive Waste Management Advisory Committee
(RWMAC) has recognised that the "decide-announce-defend" approach to policy formulation is
inappropriate [3] and that a consensus building approach involving full and open discussion of
such issues must take place. The Government (Department of Environment, Food and Rural
Affairs (DEFRA) and devolved administrations) also seem to have adopted this view in the
consultation paper they issued in September 2001 [4].

This paper contains an analysis of the factors that contributed to past failures, and an outline of
the approach being adopted to address them (Section 2). It also contains an outline of the Phased
Disposal Concept and key technical activities (Section 3) and waste producer interactions
(Section 4), which are being implemented to provide the integrated system for long-term
radioactive waste management in the UK.

2 Lessons Learned

Nirex has analysed its failure in 1997 to gain planning permission for a RCF at the Sellafield site.
We started work on developing a transparency policy. This was agreed by its Board in 1998 and
states that Nirex is committed to achieving transparency through:

1. Fostering openness as a core value;
2. Listening as well as talking to people who have an interest;
3. Making information readily available under our Publications Policy and responding to
requests for information under our Code of Practice on Access to Information;
4. Making key decisions in a way that allows them to be traced so that people can see and
understand how they were arrived at; and
5. Enabling people to have access to, and influence on, our future programme.

The findings from the Internal Inquiry [5] confirm and reinforce this analysis. The results of
these reviews are far-reaching, but they can be summarised into three key issues: process,
structure and behaviour. The following sections outline the findings in these areas in more detail.

2.1 Process

Discussions with a wide range of stakeholders have revealed that in the years leading up to the
RCF Public Inquiry, decisions taken by Nirex were not transparent and that there was a lack of
stakeholder involvement. In particular, Nirex used a closed process whose pace was driven by
pre-determined deadlines and not by the needs of stakeholders. To address these issues in the
future there must be a clear, phased decision making process that:

- has been developed in consultation with all stakeholders;
- has clear decision points;
- explains how decisions will be taken;
- provides opportunities for stakeholders to make inputs.

The whole process must be transparent and inclusive. The pace of progress – the speed at which
the process moves from one phase to the next - should be determined by the time needed to
obtain stakeholder inputs, not by a project plan. The process should include 'checks and
balances', so that the behaviours of all the players can be analysed and reviewed.
2.2 Structure
Everyday experience demonstrates that the structure of an industry has a large impact on:

- the way the industry needs to be regulated;
- public confidence in the organisations involved;
- the clarity of the issues that need to be addressed;
- the ability of those in authority to make decisions.

Radioactive waste management involves short-term decisions that may have very long-term impacts. It is essential that these short and long-term issues are visible, to the regulators and to others who have to make decisions. One way of achieving this visibility would be to maintain a separation between the short and long-term roles by having a separate organisation that is focused on the long-term implications of radioactive waste management.

2.3 Behaviour
In addition to a properly instituted process and structure, delivery of a long-term implementable solution for radioactive waste management would still be dependent on the behaviour of those involved in the process. Research and experience [6,7,8,9] has shown that the behaviour must be:

- **Open** – the debate must take place in the public domain and there should be free access to all the relevant information. Those involved should be open to influence from different people with different opinions and perspectives.

- **Transparent** – the reasoning behind actions, deliberations and decisions should be made available. It must be clear from the outset how stakeholders and the wider public can be involved and how their opinions will be taken into account and used.

- **Accountable** – those responsible for the process should be accountable for their actions to all parties. This includes publicising the reasoning behind decisions and giving people feedback on how their views have been taken into account.

3 Phased disposal concept
Even though the UK does not have a long-term solution to radioactive waste management, the waste exists and must be packaged to make it passively safe. The rest of this paper explains how Nirex provides an integrated approach so that waste can be packaged in a way that will allow it to be safely managed in the long-term. This is illustrated with a number of key examples of activities that Nirex undertakes.

Following rejection of the RCF proposal, Nirex has modified its approach to long-term radioactive waste management, based around a concept of phased disposal [10]. The Phased Disposal Concept (PDC) is a stepwise and reversible approach to geological disposal made up of the following elements.

**The waste packages.** These consist of a wasteform within a waste container. The wasteform usually consists of the waste material encapsulated in a cementitious matrix. The containers are usually manufactured in corrosion-resistant stainless steel. Some waste producers have been producing and storing waste packages to Nirex specifications for more than ten years.

**A system for transporting the waste packages to the repository.** This takes account of all the regulatory requirements for transport in the public domain by rail, road and sea.
Receipt of transport packages at the repository and transport underground. Transport packages would be transferred to a vehicle for transport underground. Access underground could either be by drift (inclined tunnel) or vertical shaft.

Removal of unshielded waste packages from their re-useable transport containers. Waste packages would be removed from the transport containers underground in a shielded inlet cell.

Transfer and emplacement of waste packages in excavated vaults. It is planned that shielded and unshielded packages will be emplaced in separate vaults.

Extended underground storage. The waste packages would remain accessible and retrievable during this period, and would be monitored as part of routine operation of the facility.

Backfilling of the vaults. A specifically formulated cement-based material would be used, which provides alkaline conditions under which many important radionuclides have low solubility and are strongly adsorbed onto the backfill. The backfill therefore forms a long-term chemical barrier to the release of radioactivity.

Closure of the repository. Closure constitutes ultimate disposal of the waste, and would be achieved by sealing of the access ways. The decision to close the repository would be made at some appropriate time, by future generations.

Long-term geological isolation. This provides the necessary long-term protection of the accessible environment and human health. The host geological environment would be chosen to provide a stable setting, with low groundwater flow and isolation from inadvertent human intrusion or disruption of the repository by natural events.

As suggested by the sequential nature of the elements listed above, it is intended that the Phased Disposal Concept would progress in a stepwise manner. Each phase of the process would be reversible and time would be available to build confidence of society at each phase before moving on to the next.

3.1 Inventory of radioactive wastes

Nirex has been compiling and publishing the UK radioactive waste inventory, in conjunction with the UK Government environment department (currently DEFRA), since 1985. This major exercise, which is conducted every 3 or 4 years, provides the basis for the planning and design of waste management systems, and for the associated safety assessments. The provision of a national inventory, covering both existing and future wastes, has enabled Nirex to develop the PDC and provide waste owners with packaging advice in a consistent manner. It has also allowed the development of a good understanding of the key drivers for long-term waste management.

As noted above, Nirex provides guidance and advice to the UK nuclear industry. To ensure consistency and transparency, Nirex has developed a methodology that assists those organisations responsible for the packaging of wastes to identify the relevant radionuclides in their wastes, and to derive a comprehensive inventory of those radionuclides.

Studies have shown that the overall UK radionuclide inventory is dominated by waste from two principal sources: commercial reactor fuel and Stage 3 decommissioning of commercial gas reactors (Magnox and Advanced Gas-Cooled Reactors). Consequently, wastes from these two sources were studied in detail and a relevant radionuclide analysis undertaken for each. The application of this analysis to the dominant waste types produced a listing of 112 potentially relevant radionuclides [11].
3.2 Standards and specifications for waste packages
To complement the PDC and support waste owners in their efforts to condition wastes for long-term management, Nirex has developed the Waste Package Specifications and Guidance Documentation (WPSGD). This suite of documents defines standard waste containers and provides specifications for key aspects of their design and performance, including container parameters, wasteform composition, quality assurance and data records. It thus provides a firm foundation from which the waste owners can plan waste management strategies.

The current range of standard containers consists of the 500 litre Drum, 3 m³ Box, 3 m³ Drum and 4 metre Box. These containers have a variety of applications. For example the 500 litre Drum is a well-established, widely used container for packaging operational intermediate-level wastes, including supercompacted wastes. The 3 m³ Box is becoming widely used as an efficient way of dealing with a wide range of solid decommissioning wastes. The larger 4 metre Box, currently in the final stages of the container approval process, is being considered for many large-scale decommissioning projects.

Nirex have recently confirmed the requirements for the last in the range of standard waste containers, the 2 metre Box. This is a half-length version of the existing 4 metre Box, which could be used for packaging high density wastes or for use in restricted access areas which would preclude the use of the 4 metre Box. It is envisaged that both the 4 metre Box and the 2 metre Box will be approved as Industrial Package IP-2 containers under the IAEA Transport Regulations [12].

3.3 Interim storage of waste packages
Packages of conditioned waste are currently held in interim storage in surface stores. It is anticipated that the period of interim storage will be at least 50 years, prior to the availability of a disposal facility, followed by a second period of 50 years, during which the facility is operational, and then by a further period during which the facility remains open and waste packages can be monitored and retrieved if required. The key performance requirements are for waste packages: to maintain their integrity during interim storage; to provide containment of the radioactivity; and; to facilitate future handling and retrievability for future phases of waste management.

Nirex is developing a framework, in conjunction with the UK nuclear industry, which provides guidance on best practice in interim storage. The objective is to ensure that waste packages will be able to meet Acceptance Criteria for onward phases of waste management. The system is organised in three guidance documents, providing advice on maintaining, monitoring and checking (prior to transport) the integrity of waste packages [13].

3.4 Technical assessment of waste package production
The application of approved Quality Assurance arrangements to the production of waste packages in the UK is a Nirex requirement, supported by regulators. All aspects of waste packaging that could affect product quality must be covered, and as part of the assessment continuum, Nirex undertakes technical assessments. These focus on package production controls, package characteristics and measurement, and creation and maintenance of relevant information, including production records.

Nirex has developed the concept of the Waste Product Specification (WPrS) as a key document to be developed during the planning phase and subsequently implemented during production. This specification identifies and quantifies (where appropriate) the components of the manufactured package that require control during the production process. Confirmation of compliance with the WPrS during package production, thus making the finished article
acceptable for future phases of waste management, is an important outcome of the technical assessment process.

Technical assessment provides an opportunity for the waste owner to demonstrate to Nirex and other stakeholders the production of fully characterised waste products, and to provide on-going evidence of their storage under appropriate conditions. It also provides assurance that key waste package records and information are created and preserved. Thus the waste packages should meet the appropriate regulatory and safety requirements, consistent with future waste management options [14].

4 Interactions with Waste Producers

UK Government policy requires that the waste package should, so far as reasonably practicable, be designed to be compatible with all subsequent stages of waste management, from interim storage through to its final disposal. This minimises the possibility that wastes might need to be re-packaged at some later date since re-packaging (for whatever reason) would entail both additional radiation doses to workers and considerable additional costs. As noted previously, Nirex has developed the Waste Package Specifications and Guidance Documentation to help waste producers comply with this requirement.

4.1 Provision of advice

Nirex assists with the interpretation of, and checks on the application of, the Waste Package Specifications and Guidance Documentation through an advice process. This process is recognised by the main UK nuclear industry regulators: the Nuclear Installations Inspectorate [15], and the environmental Agencies (EA and SEPA) [16]. It is initiated by the submission of proposals for packaging of specific wastes by waste producers to Nirex for assessment.

The rigorous assessment process looks at all aspects of the properties and performance of the proposed packages, their compatibility with Nirex specifications, and consistency with the PDC throughout all its phases, as expressed in generic assessments of its operational, transport and post-closure safety. The assessment covers issues such as the chemical and physical properties of the wasteform, criticality safety, and the corrosion behaviour of the waste container. The assessment process also considers waste package performance under accident conditions, for example impact and fire. Key areas that are examined are the adequacy of the Quality Assurance regime to be applied to the development, production and storage of the waste packages, and the suitability of the records that are to be generated and maintained relating to the waste packages. As noted previously, it is largely based upon these records that compatibility with a disposal facility would be determined in the future.

Following completion of the assessment, a Letter of Advice (LoA) is provided to the waste producer outlining the results of the assessment. This may identify areas where the packaging proposals are not compatible with the Nirex standards and specifications of waste packages, and the Phased Disposal Concept, and may make recommendations for improvements or further research.

4.2 Endorsement through Letters of Comfort

A Letter of Comfort (LoC) represents Nirex endorsement that a specific packaging proposal is consistent with the PDC, Nirex's principles and its Waste Package Specifications. It is issued along with a LoA whenever this consistency can be demonstrated by the waste owner. The LoC signifies that, providing the packages will be stored in suitable conditions by the waste producer, then from Nirex’s perspective:
1. The packages are directly compatible with the needs for long-term management, or;
2. The packaging proposals may require further treatment to remediate limited deficiencies to make them compatible with long-term needs and these are possible using known technologies, or;
3. The proposed packages are interim packages and will definitely require further treatment, and are capable of being further worked to produce final disposal packages using known technologies.

In the first situation Nirex is able to issue a Letter of Comfort without caveats. In the second situation Nirex is only able to issue a Letter of Comfort with caveats. Such caveats would identify where an additional process may be necessary to mitigate the potential problem, should this be shown to be necessary. In the third situation Nirex is able to issue a Letter of Comfort without caveats for the initial process step. However, this is conditional on a future action being taken to convert these interim packages into final disposal packages.

Nirex cannot provide endorsement where major issues and uncertainties remain or where to mitigate the problem would require major re-working of packages or the application of new technologies. In such cases Nirex can only provide a Letter of Advice to inform the developer of where actions will be required to mitigate the issues.

In response to customer requests, Nirex has provided advice and endorsement for a wide range of specific packaging proposals. To date, this accounts for over 42% by volume of the UK ILW, and is supported by the issue of more than 80 Letters of Comfort.

5 Summary and Conclusions

Nirex has analysed its failure in 1997 to gain planning permission for a Rock Characterisation Facility at the Sellafield site. The issues include: the process of policy development and public acceptance of specific solutions; the structure of the nuclear industry, and; the behaviour of the institutions involved. It was concluded that transparency must be central to the culture of organisations attempting to win public acceptance for such significant proposals.

Nirex’s current purpose is to provide the UK with safe, environmentally sound and publicly acceptable options for the long-term management of radioactive materials. In support of this we have developed the Phased Disposal Concept (PDC) as one way of managing these wastes in the long-term. Nirex has a key role to provide waste producers with advice and endorsement (via the issue of a ‘Letter of Comfort’), on the packaging and transport of particular wastes. Waste producers seek advice from Nirex at each stage of the packaging project, from concept through to active commissioning.

A number of key activities are being undertaken by Nirex to develop the PDC, and to support waste owners in the conditioning and packaging of radioactive wastes for long-term management. These include the identification of key radionuclides in wastes; standardisation and specification of waste packages; maintaining, monitoring and checking their integrity, and; technical assessment of quality assurance arrangements. Taken together, these provide an integrated approach to the long-term management of radioactive wastes in the UK.
References

FROM WASTE PACKAGES ACCEPTANCE CRITERIA TO WASTE PACKAGES ACCEPTANCE PROCESS AT THE CENTRE DE L'AUBE DISPOSAL FACILITY

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1- Introduction

The Centre de l'Aube disposal facility has now been operated for 10 years. At the end of 2001, about 124,000 m³ of low and intermediate level short lived waste packages, representing 180,000 packages, have been disposed, for a total capacity of 1,000,000 m³. The flow of waste packages is now between 12 and 15,000 m³ per year, that is one third of the flow that was taken into account for the design of the repository. It confirms the efforts by waste generators to minimise waste production. This flow represents 25 to 30,000 packages, 50% are conditioned in the compaction facility of the repository, so that 17,000 packages are disposed per year. 54 disposal vaults have been closed.

In 1996 - 1999, the safety assessment of the repository has been reviewed, taking into account the experience of operation. This assessment was investigated by the regulatory body and, subsequently, a so-called “definitive license” to operate was granted to ANDRA on September 2, 1999 with updated licensing requirements. Another review will be performed in 2004.

To ensure a better consistency with the safety assessment of the facility, Andra issued new technical requirements for waste packages at the end of 2000. Discussions with waste generators also showed that the waste package acceptance process should be improved to provide a more precise definition of operational criteria to comply with in waste conditioning facilities. Consequently a new approach has been implemented since 2000.

2- Main features of Andra’s updated acceptance criteria for waste packages

2.1 General, radiological and chemical criteria

General criteria address to all packages that are delivered. They give requirements on the physico-chemical properties of waste: no free liquid, inert matter. They focus on the radiological characterisation of the package,
particularly on the identification of radio-nuclides that may be present. A list of 143 nuclides has been established. The purpose is to detect waste that could bring higher quantities of activity than taken into account in the safety assessment.

Activity limits are derived from the safety scenarios. Some limits are prescribed to avoid "hot spots" in the repository. This is the case for nuclides the impact of which is relevant not for a single package but for a group of packages or sometimes the whole repository. Long lived beta emitters belong to that category. Requirements are expressed in terms of specific mass activity limitation.

Table 1: radiological capacity of the Centre de l'Aube facility and derived activity limits

<table>
<thead>
<tr>
<th>Element</th>
<th>Radiological capacity of the facility TBq</th>
<th>Activity limit per package (bq/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{3}$H</td>
<td>$4 \times 10^{2}$</td>
<td>$2 \times 10^{3}$</td>
</tr>
<tr>
<td>$^{14}$C</td>
<td>$8.15 \times 10^{2}$</td>
<td>$9.2 \times 10^{4}$</td>
</tr>
<tr>
<td>$^{36}$Cl</td>
<td>$4 \times 10^{-1}$</td>
<td>$2.4 \times 10^{4}$</td>
</tr>
<tr>
<td>$^{41}$Ca</td>
<td>$1.25 \times 10^{1}$</td>
<td>$3 \times 10^{3}$</td>
</tr>
<tr>
<td>$^{60}$Co</td>
<td>$4 \times 10^{3}$</td>
<td>$1.3 \times 10^{5}$</td>
</tr>
<tr>
<td>$^{59}$Ni</td>
<td>$4 \times 10^{3}$</td>
<td>$1.1 \times 10^{5}$</td>
</tr>
<tr>
<td>$^{63}$Ni</td>
<td>$4 \times 10^{4}$</td>
<td>$3.2 \times 10^{5}$</td>
</tr>
<tr>
<td>$^{79}$Se</td>
<td>$3.72 \times 10^{2}$</td>
<td>$5.5 \times 10^{4}$</td>
</tr>
<tr>
<td>$^{90}$Sr</td>
<td>$4 \times 10^{6}$</td>
<td>$6 \times 10^{6}$</td>
</tr>
<tr>
<td>$^{92}$Zr</td>
<td>$4 \times 10^{2}$</td>
<td>$1.8 \times 10^{4}$</td>
</tr>
<tr>
<td>$^{94}$Nb</td>
<td>$2.01 \times 10^{1}$</td>
<td>$1.2 \times 10^{2}$</td>
</tr>
<tr>
<td>$^{95}$Mo</td>
<td>$1.02$</td>
<td>$3.8 \times 10^{4}$</td>
</tr>
<tr>
<td>$^{99}$Tc</td>
<td>$1.23 \times 10^{1}$</td>
<td>$4.4 \times 10^{4}$</td>
</tr>
<tr>
<td>$^{107}$Pd</td>
<td>$3 \times 10^{3}$</td>
<td>$3 \times 10^{5}$</td>
</tr>
<tr>
<td>$^{108}$Ag</td>
<td>$2.49 \times 10^{1}$</td>
<td>$1.4 \times 10^{3}$</td>
</tr>
<tr>
<td>$^{129}$I</td>
<td>$3.03 \times 10^{1}$</td>
<td>$1.4 \times 10^{3}$</td>
</tr>
<tr>
<td>$^{135}$Cs</td>
<td>$6 \times 10^{1}$</td>
<td>$2.6 \times 10^{5}$</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>$2 \times 10^{5}$</td>
<td>$3.3 \times 10^{5}$</td>
</tr>
<tr>
<td>$^{151}$Sm</td>
<td>$1 \times 10^{4}$</td>
<td>$4.5 \times 10^{5}$</td>
</tr>
<tr>
<td>total alpha emitters (after 300 years)</td>
<td>$7.5 \times 10^{2}$</td>
<td>$3.7 \times 10^{3}$</td>
</tr>
</tbody>
</table>

In italics: radioactive capacity prescribed in the Order of Creation of the Centre de l'Aube facility.

Another aspect that is focused in the new version of Andra's specifications is the identification of matters that might have a chemical impact. These matters are partly derived from regulations relevant for non radioactive repositories: lead, boron, nickel, chrome (total and VI form), antimony, selenium, cadmium, mercury, beryllium, arsenic, free cyanides, ammonia and asbestos. These matters must be quantified, generally using typical chemical spectra for waste. This inventory provides data to perform chemical impact calculations.
2.2 Different types of waste packages: containment requirements

Two main types of packages are distinguished: packages with a metallic envelope and packages with a long duration concrete envelope. To ensure the stability of the repository for a few hundred years, the first one are grouted inside the vaults to prevent corrosion. For the second category specifications were issued to give characteristics of the concrete to comply with.

The description of containment properties is pointed out. Containment may be obtained either by leaching performances of the matrix embedding the waste in case of "homogeneous" waste or by a "containment barrier" provided by a sufficient thickness of concrete around the waste. The diffusion coefficient of concrete (using tritium water as a reference material for diffusion) is specified depending on the thickness of the barrier.

![2 ways to achieve containment](image)

Leaching rates are in the range of $10^{-3}$ per year, diffusion coefficients in the range of $10^{-14} - 10^{-12}$ m²/s. It is focused that, in a case of a concrete envelope, minimum thickness is calculated to provide either mechanical strength and containment for a few hundred years.

3. The acceptance process

3.1 The agreement process

Following the 1989 order of creation of the Centre de l'Aube facility, prior to any delivery of a type of packages, Andra must issue an "agreement" for this waste package type. This agreement expresses that Andra considers that packages of this type to be produced will comply with the repository requirements.

At the end of 2001, about 130 agreements are "in operation". Some of them are generic, they are valid for different similar facilities. For instance there is one agreement that is valid for ion exchange resins generated by the PWR French power reactors. Finally there are about 300 sets of agreement – facility files in operation.

The agreement process has been formally implemented since 1985. Taking into account 15 years of experience, a review was performed with waste generators. Modifications aim to give a more explicit operational definition of a package complying with Andra's requirements, and then to reinforce the link between AQ procedures and the objective of compliance.

Actually some parameters that are specified by Andra can be monitored directly on the waste package or during the process. For instance the weight of a package can be measured and compared with the maximum specified weight, dose rate can be measured. For others it is not possible to control them directly during the fabrication process of the package. This is the case for containment parameters (leaching rate, diffusion coefficient). For these parameters it would be necessary to destroy the package and then to perform experiments that may last more than one year.
Therefore during the agreement process investigations are made to identify parameters of the package or of the conditioning process that can be monitored during the fabrication of the packages and that will ensure indirectly compliance with Andra’s requirements. For the diffusion coefficient example these operational parameters may be the composition of the cement used to make the diffusion barrier, its water content...

The choice of the operational parameters relies on a qualification step. Experiments are performed on prototype packages or samples and must demonstrate compliance with waste acceptance criteria. Note that the waste generator may choose to qualify his package in a wide or a narrow range.

Such an investigation is performed by the waste generator, with Andra’s support, for each technical requirement for disposal.

All these operational parameters are grouped in a list that gives a suitable description (or “specification”) of the package relevant with Andra’s requirements. This list, including the parameters and their values, is called “a contractual requirements sheet” or “CRS”. It gives the description of the package that the generator agrees to manufacture and that Andra agrees to take in charge in the disposal facility.

Supplying, fabrication or control procedures are then derived to ensure that actual packages will meet the CRS.

In that way the agreement process leads to an operational definition of the package complying with Andra’s requirements and to relevant AQ procedures.

<table>
<thead>
<tr>
<th>Andra requirement</th>
<th>Operational parameters</th>
<th>QA procedures</th>
</tr>
</thead>
<tbody>
<tr>
<td>No free liquid</td>
<td>No free liquid</td>
<td>Waste sorting or drying</td>
</tr>
<tr>
<td>Iodine 129 &lt; 1,400 bq/g</td>
<td>Cesium 137 &lt; 1,400 bq/g</td>
<td>Cs137 measurement</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Scaling factors verification program</td>
</tr>
<tr>
<td>Diffusion coeff. &lt; 10^{-13} m²/s</td>
<td>Grout, water content ...</td>
<td>Supplying the grout</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Conducting the process</td>
</tr>
</tbody>
</table>

Figure 3 : general flow chart of agreement process and examples

3.2 Packages quality surveillance by Andra

By the agreement process, Andra obtains confidence in the ability of the waste generators to produce waste packages. This confidence must be maintained by a surveillance by Andra of packages quality.

Different means are implemented for surveillance:

- computer check of packages declaration by the waste generator:
  
  Each package to be delivered to the Centre de l’Aube facility must be first declared by the waste generator in a computer tracking system. Identification of the package, type of the envelope, volume, mass, physical nature of waste, activity of nuclides are declared.

  The computer system checks the consistency of the declaration with the agreement file. An authorisation for delivery is then issued and the list of authorised packages is provided to the disposal facility.

- control of packages at delivery to the Centre de l’Aube facility.

  Controls are performed on dose rate and surface contamination of packages.
audits in waste generators facility

"Process/product audits" are performed by Andra in waste generators facilities. References for these audits are QA procedures derived from the Contractual Requirements Sheet. About 60 audits are performed per year, so main facilities are "visited" at least once a year. Deviations are analyzed according to the CRS with a follow up of corrective actions.

destructive and non destructive tests on actual packages

These tests are an important aspect of Andra's surveillance. Packages that are investigated are taken at delivery to the disposal facility. About 150 to 300 non destructive tests, 10 to 15 destructive tests are performed per year. With regard to the 25 to 30,000 packages that are delivered and to the 130 agreement files, such tests cannot have a statistical meaning. However data collected in the 1,500 non destructive tests and 200 destructive tests since 1984 provide significant information that may be used to adjust some conditioning processes or parameters evaluation (such as scaling factors).

A criterion to select packages to investigate is their contribution to the radioactive inventory. Tests also focus on recently agreed packages.

Depending on the type of packages non destructive tests consist on weight, dimensional controls, dose rate, surface contamination, gamma spectrometry, gamma-graphy, X radiography or neutron measurements.

Destructive tests are used to investigate the waste content of packages, general quality of conditioning. "Carrots" or samples are taken to perform measurements on mechanical strength, diffusion coefficient, leaching rate. Alpha or beta emitters (particularly long lived beta emitters) or chemical compounds are investigated.

So destructive tests provide important information on parameters that are used in the safety assessment for the industrial packages.

Figure 4 : non destructive tests (gamma spectrometry, tomography)

Figure 5 : destructive tests and "carrots" to perform strength or containment measurements
Long-term Interim Storage and Conditioning of LAW and MAW in Germany

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Johann B. Zech, TÜV Süddeutschland, Munich

0 Introduction

The aim of this presentation is to provide an overview of the current situation in Germany of long-term interim storage and conditioning of low- and medium-active wastes (LAW and MAW). Since the beginning of nuclear waste generation in Germany, no differentiation has been made between wastes only with short-lived and wastes also containing long-lived isotopes. Therefore all LAW and MAW have to be disposed in a final repository in deep geological formations to prevent any long-term radioactive release into the biosphere. On surface or near surface only interim storage is possible.

The simplified flow of radioactive wastes in Germany is shown in Figure 1. This diagram reflects all major sources of radioactive wastes, including those from industrial use and other areas of applications. The paths which are practiced today are represented by solid lines; future plans and developments are represented by dashed lines. On one hand the future final repository can be represented by a single final disposal site, whereby a differentiation is necessary between underground facilities for heat-generating wastes (i.e. HAW) and those for non-heat-generating wastes (i.e. MAW/LAW). Such a solution reflects both the political requirement of a single-repository concept and the technical debate on the suitability of such a concept. On the other hand a model with one site for the MAW/LAW final repository and another site for the HAW final repository is also a possible solution.

The current legal stipulations of the Atomic Energy Act (L 1) and the Radiation Protection Ordinance (L 2), and of recommendations of the Federal Ministry of Environment (BMU) and Reactor Safety Commission (RSK) (L 3 and L 4) also influence the current situation.

1 Current situation and future development

The political stipulations which have influenced the nuclear situation in Germany since 1998 have been laid down in the new amendment to the Atomic Energy Act (L 1) that went into effect in April 2002. The legal stipulations in this amendment mainly deal with the phase-out of nuclear power plants and handling of spent fuel (end of reprocessing). It has two effects on MAW and LAW:

- The phase-out of nuclear power plants reduces the amount of MAW and LAW produced through the operation of NPPs.
- The generation of MAW and LAW is reduced through the phasing out of reprocessing.

While both effects reduce the amount of wastes generated, they do not result in any fundamental change in the situation with respect to MAW and LAW. Table 1 shows an estimate of future MAW and LAW generation.

Below is an overview of the current final disposal situation:

- In June 2002 the license (plan-approval decision) for the planned Konrad repository for non-heat-generating wastes was given to the Federal Office for Radiation Protection (BfS) by the Lower Saxonian Environmental Ministry, the relevant licensing authority. The application was filed twenty years ago by the Federal Institute of Physics and Metrology (PTB) in
Braunschweig which was responsible at the time. The licensed capacity is 300,000 m³.

The political decision, whether the single final disposal concept shall be continued, is pending.

Although interim storage facilities on separate sites (Gorleben and Mitterteich) are currently available for LAW and MAW from nuclear power plants, the majority of those wastes are stored on-site at the nuclear plant locations. This also applies to wastes from fuel processing, decommissioning, back-fitting and partly from research. Wastes from other research areas, from the medical field, industry and trade are stored at the state collection depots ("Landessammelstellen"). This type of interim storage facility has to be provided by each state according to the German Atomic Energy Act. The actual status of all interim storage facilities is shown in Table 2.

Long-term interim storage of LAW and MAW thus affects many facilities at several locations. Table 3 in the annex provides an overview of the volume of these wastes as of December 31, 1999, which reflects the latest available data.

In the case of the previously existing specifications for conditioning wastes, it was assumed that the conditioned wastes would only have to be stored for a relatively short period of time (a few years), would only have to be handled during emplacement in the final repository, and that no radioactivity containment would be required afterwards. However, the actual situation has looked somewhat different for quite a while: until 1978, West Germany had Asse as a final repository, and East Germany, Morsleben until 1990 – both of these final repositories are former salt mines. Now there is no final repository available for LAW and MAW from the point of view of West Germany since 23 years. That is, with the exception of the short time in the 1990s when West Germany deposited a part of its wastes in Morsleben.

There are two possibilities for how things may develop in the future:

- If the single final repository concept continues to be pursued, a final repository viable also for LAW/MAW will not go into operation until around the year 2030. This means a storage period of approx. 40 years has to be reckoned with.
- However, even if the single final repository concept is not pursued, it would not be technically possible to dispose of the existing LAW and MAW in the near future. A realistic assessment of the currently licensed Konrad final repository is as follows:
  - it would take several years until the court decisions are final,
  - afterwards it would take another four to five years until the final repository is operational, and
  - once operation begins it would take many years until the current large volumes of wastes are disposed.

Therefore, regardless of scenario, it will easily take over 10 years for the interim storage of MAW and LAW.

Technical implementation is currently focused on measures for qualifying the stored waste containers (some which have already been stored for a long time) and adapting the storage facilities to the additional requirements of the significantly longer storage time.

2 Interim storage and conditioning of LAW and MAW

Large volumes of LAW and MAW are currently kept in interim storage facilities. Some of the wastes have already been stored for a relatively long time.

In general, a differentiation must be made between the different types of storage of residual materials and wastes:
- buffer storage until further conditioning or treatment
- decay storage for later clearance or later conditioning under more suitable radiological conditions or final disposal
- long-term interim storage in a conditioned form suitable for final disposal until material is actually placed in a final repository.

Most of the questions apply to the latter wastes due to their significantly longer interim storage time and need for conditioning before final disposal.

2.1 Protection objectives and concepts

Existing protection objectives and safety requirements involve safely containing radioactive substances and shielding the radiation to protect operating personnel and the environment. This entails the following measures:

- packaging radioactive residual materials and radioactive wastes
- conditioning radioactive residual materials and radioactive wastes
- interim storage
- internal handling and transport at the site and
- emplacement in the final repository.

For storing non-heat-generating wastes, a multiple barrier concept is generally used, in which, depending on the plant and chemical/physical properties of the radioactive materials, the barriers of the waste matrix, waste containers and - in some cases - the building, including its equipment, contribute in varying degrees to their safe containment.

The plants for interim storage of LAW/MAW and radioactive residual materials are generally designed for handling and storing enclosed radioactive materials; i.e. the waste containers are primarily responsible for ensuring safe containment of the radioactivity. In this case, the waste container has to ensure safe containment of the radioactivity during the entire interim storage period.

A storage concept for handling open radioactive materials is also permitted, however, in order to accommodate the assumed discharge of radioactive materials in the exhaust air and waste water, additional technical installations are required for targeted air guidance and for measuring emissions. These conditions are generally met if storage is within the control area of a nuclear power plant.

Transfer to a final repository demands additional measures, especially with regard to minimizing radiation exposure for staff personnel. The wastes should be stored in such a way that no extensive measuring is required, nor conditioning or packaging that could lead to unnecessary exposure of operating personnel when the wastes are ultimately transferred to a final repository.

2.2 Specifications of Radiation Protection Regulations

The Radiation Protection Ordinance (L 2), enacted in 2001, specifies a number of regulations that replace some of the previous specifications of the BMU guideline for waste flow control (L 8). These regulations are contained in Articles 72 to 79. They effect:
- Article 72 Planning of Accumulation and Storage of Radioactive Wastes: A report with information on the volume of radioactive wastes in a plant on December 31 and the expected waste volume to be generated in the following year is to be submitted by March 31 of the following year.
- Article 73 Data Acquisition: The volume of radioactive wastes is to be calculated based on a precisely prescribed scheme using an electronic accounting system.
- Article 74 Treatment and Packaging: The state collection depots and federal offices responsible for final disposal have the right to set down specifications concerning packaging and conditioning of wastes. The procedures used for producing waste packages suitable for final disposal must comply with the regulations of the Federal Office for Radiation Protection (BFS).

To date, there are only final waste disposal conditions for the Morsleben repository and Konrad mine; the Federal Office for Radiation Protection (BFS) has announced it will issue general requirements not specific to any particular final repository in 2002.
- Article 75 Responsibilities when Handing Over Radioactive Wastes: Defines the procedure for handing over wastes for transport or to another recipient (notice to authorities, declaration of acceptance)
- Article 76 Handing Over: Defines the handing over to plants of the federal government and to and from state collection depots
- Article 77 Exceptions from Obligation to Hand Over: No obligation to hand over applies to wastes for which an exception in accordance with Article 29 of the Radiation Protection Regulations (StrSchV) has been requested.
- Article 78 Interim Storage: Persons required to deliver wastes to final repositories must store the wastes in interim storage facilities until the respective final repository of the federal government goes into operation. Interim storage can also be operated together by several parties required to deliver wastes to final repositories.
- Article 79 Prohibition of Bypassing: It is not permitted to eliminate the wastes through dilution or dividing them up into volumes which are exempt.

Besides these special regulations for the disposal of radioactive wastes, the regulations for protecting the population and environment (Articles 46 to 48) must also be taken into account when designing and operating interim storage facilities for radioactive wastes. If the waste storage facility is part of a nuclear power plant licensed in accordance with Article 7 of the Atomic Energy Act or is one of the facilities of the federal government for safely storing radioactive wastes, the design specifications of Article 49 of the Radiation Protection Ordinance (StrSchV) are to be used for designing the facility's incident protection system. The specifications of Article 50 of the Radiation Protection Ordinance (StrSchV) apply to other plants; this article does not stipulate any explicit design specifications, but only refers to an administrative regulation of the federal government still to be enacted.

2.3 Current RSK Work on Safety Regulations

On request of the BMU, the Reactor Safety Commission (RSK) investigates the issues involved with the long-term interim storage of MAW and LAW. Two years ago, the RSK analyzed the incidents involving old wastes at the Geesthacht state collection depot and a conceptual study by TÜV Süddeutschland on the long-term interim storage of low- and medium-level radioactive
wastes in Mitterteich. The issues covered long-term maintenance of the radioactivity enclosure as well as reliable documentation of the stored radioactive materials – both in terms of compliance with licensing requirements for operating the storage facility and as the basis for the documentation required for later disposal in a final repository.

These discussions served as the point of departure for a full representation of the safety aspects for long-term interim storage of LAW and MAW, defining the issues involved in a uniform Germany-wide procedure, and recommending possible solutions.

The BMU's request for such deliberations stem on the one hand from the issues involved in the safe storage of the wastes over a considerably longer time than originally expected and on the other hand from its responsibility for a final repository for these wastes and the acceptance conditions to be complied with. Current storage and conditioning of wastes must be done in such a way that final disposal can occur without extensive additional measures. Interim storage should at least not involve any steps that would in any way hinder later final disposal.

The RSK committee "Supply and Disposal" is currently working on safety requirements for long-term interim storage of low- and medium-level radioactive wastes. This will involve two steps. The published report (L 4) of the first step outlines requirements and recommendations in a more general manner. The aim of this report was to define and assess all safety-related impact parameters for the interim storage of low- and medium-level radioactive wastes over a period of decades up to approximately 40 years. At the same time, all limiting conditions which, in terms of current knowledge, are to be clarified before placing wastes in an interim storage facility for ultimate disposal in a final repository should be considered. The requirements for wastes and waste packaging are an integral part of this report.

In a second step the RSK actually develops detailed guidelines called "Safety Requirements for the Longer Term Interim Storage of LAW and MAW", which deals with the following issues:

- maintaining the container's barrier function
- avoiding the development of significant volumes of gas from the waste products
- avoiding rotting, fermenting and radiolysis in the waste products
- avoiding other physical-chemical processes (e.g. development of acids) in the waste products that could have a negative effect on the integrity of the container,
- integrity of the waste container, taking into account mechanical damage and corrosion
- suitability of the sealing materials used (e.g. in terms of decomposition)
- handling and transporting the waste packaging
- taking into consideration the previously mentioned requirements already before interim storage, i.e. during the conditioning and documentation of the wastes
- avoiding the need to perform extensive measurements and re-measurements after conditioning
- sufficient documentation on waste handling, properties of the waste product and waste container, and activity inventory
- clarifying the requirements for conditioning the wastes (in compliance with the requirements for the Konrad final repository or other specifications). The BMU has asked the BfS to further develop the requirements for the Konrad final repository to come up with general specifications.

In May 2002 a symposium was held to discuss a first draft (L 7) of the "Safety Requirements" with the appropriate organizations. According to the timetable the "Safety Requirements" shall be adopted by the RSK by the end of 2002.
3 Summary

In Germany, LAW and MAW is to be disposed of in a final repository in a deep geological formation. This radioactive waste originates from the operation and decommissioning of nuclear facilities as well as from the use of radioactive substances in industry, medicine and research. In recent years, opportunities for final disposal of radioactive waste were only temporarily available. Consequently, LAW and MAW have been stored in interim storage facilities in the meantime. On December 31, 1999, the volume of conditioned wastes amounted to approximately 63,000 m³; raw residues and intermediate products amounted to approximately 34,600 m³. Some of this waste is already very old. From today’s point of view, a considerable span of time will pass, until opportunities for final disposal are available again. Therefore, with regard to future radioactive waste disposal in Germany, the focus will be on the long-term interim surface storage of large amounts of radioactive waste. This will be a main task in the near future.

Up until now, there have been no standardized specifications which define the requirements for the interim storage and conditioning for final disposal of LAW and MAW. The Reactor Safety Commission (RSK) is currently developing safety requirements, which shall be adopted at the end of 2002. In this contribution, essential parts of these safety requirements are described.

Literature

L 1 German Atomic Energy Act: April 26, 2002; www.bmu.de/fset1024.php

L 2 Ordinance on the protection against damage through ionizing radiation (Radiation Protection Regulations – StrlSchV) from July 20, 2001 (BGBl. I p. 1714); www.bmu.de/download/dateien/strahlenschutzv.pdf

L 3 RSK: Safety guidelines for dry interim storage of radioactive fuel assemblies in containers, approved of at the 338th RSK meeting on March 1, 2001; www.rskonline.de/Download/Leitlinien/LEITLINIEN050401.pdf


L 7 RSK: Safety Requirements for the Longer Term Interim Storage of LAW and MAW – Draft, May 16, 2002

L 8 BMU: Guideline for controlling radioactive wastes with negligible heat generation that are not stored at a state collection depot, from January 18, 1989, Bundesanzeiger No. 63a from Jan. 16, 1989, with supplements from June 26, 1989, Bundesanzeiger No. 124 from July 7, 1989
Figure 1:
Accumulation and disposal of radioactive wastes in Germany (simplified schematic diagram)

Table 1:
Accumulation of wastes with negligible heat generation until 2040 (m³, estimate of AkEnd, L 5)

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>EVU</td>
<td>23,000</td>
<td>31,000</td>
<td>46,000</td>
<td>73,000</td>
<td>22,000</td>
<td>195,000</td>
</tr>
<tr>
<td>Government</td>
<td>53,000</td>
<td>27,000</td>
<td>8,000</td>
<td>3,000</td>
<td>11,000</td>
<td>102,000</td>
</tr>
<tr>
<td>Total</td>
<td>76,000</td>
<td>58,000</td>
<td>54,000</td>
<td>76,000</td>
<td>33,000</td>
<td>297,000</td>
</tr>
</tbody>
</table>
Table 2: Present interim storage capacities, volume of conditioned wastes on Dec. 31, 1999 and utilization level of interim storage facilities for non-heat-generating wastes (data from L 6)

<table>
<thead>
<tr>
<th>Group</th>
<th>Utilization level (%)</th>
<th>Interim storage capacity (m³)</th>
<th>Volume on Dec. 31, 1999 (m³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Research (incl. customers)</td>
<td>61</td>
<td>75,760</td>
<td>45,960</td>
</tr>
<tr>
<td>Nuclear industry</td>
<td>36</td>
<td>4,914</td>
<td>1,772</td>
</tr>
<tr>
<td>Light-water reactors</td>
<td>21</td>
<td>23,164</td>
<td>4,865</td>
</tr>
<tr>
<td>Interim storage facility Nord (ZLN)</td>
<td>1</td>
<td>201,025</td>
<td>1,035</td>
</tr>
<tr>
<td>Decommissioned reactors (Dec. 31, 1999)</td>
<td>20</td>
<td>4,838</td>
<td>962</td>
</tr>
<tr>
<td>State collection depots</td>
<td>21</td>
<td>6,840</td>
<td>1,454</td>
</tr>
<tr>
<td>Other parties responsible for delivery of wastes</td>
<td>6</td>
<td>3,540</td>
<td>212</td>
</tr>
<tr>
<td>Interim storage facilities of power companies</td>
<td>16</td>
<td>42,477</td>
<td>6,796</td>
</tr>
</tbody>
</table>

Table 3: Overview of volume of radioactive residues (non-heat-generating wastes, data from L 6)

<table>
<thead>
<tr>
<th>Residue type</th>
<th>Volumes (m³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Untreated residues (recyclable residues and primary wastes):</td>
<td></td>
</tr>
<tr>
<td>- Volume at end of 1999</td>
<td>31,634</td>
</tr>
<tr>
<td>Intermediate products:</td>
<td></td>
</tr>
<tr>
<td>- Volume at end of 1999</td>
<td>2,944</td>
</tr>
<tr>
<td>- Accumulation in 1999</td>
<td>1,083</td>
</tr>
<tr>
<td>Conditioned wastes:</td>
<td></td>
</tr>
<tr>
<td>- Volume at end of 1999</td>
<td>63,712</td>
</tr>
<tr>
<td>- Accumulation in 1999</td>
<td>2,817</td>
</tr>
</tbody>
</table>
SAFE LONG TERM INTERIM STORAGE OF WASTE PRODUCTS AT THE KARLSRUHE RESEARCH CENTER

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Abstract
Due to the political situation in Germany there will presumably be no final disposal for radioactive waste in the next 30 years. This means, that the operators of nuclear facilities have to ensure a secure long term intermediate storage for radioactive waste products.

The Karlsruhe Research Center operated and cooperated with different nuclear research facilities and laboratories with hot cells, which are now dismantled. During operation and decommissioning of the nuclear facilities radioactive waste was produced. The Central Decontamination Department (HDB) of the Research Center Karlsruhe has been conditioning radioactive waste into waste products for final disposal. Until the opening of a final disposal the resulting waste products will have to be stored at HDB. To ensure secure long term storage, quality preserving measures will have to be taken. For example, the corrosion-preventing coating of the containers has to be kept intact. In case of damage the container has to be repaired or should be replaced. Another long term protection measure is the casting of drums in concrete inside the containers. This provides an additional barrier layer in case of drum corrosion.

At HDB 46,500 m³ of radioactive waste products are in intermediate storage. The main project of the next few years will be the realization of the quality measures. In this paper the different methods are described in detail.

1 Introduction
For almost 40 years, the Central Decontamination Department (HDB) of the Karlsruhe Research Center is conditioning radioactive waste into waste products suitable for final disposal. Customers of the HDB are:
- Research institutes of the Research Center
- Shut-down nuclear reactor prototypes, such as the Niederaichbach Nuclear Power Plant (KKN), the Karlstein Superheated Steam Reactor (HDR), the Research Reactor (FR2), the Multi-Purpose Research Reactor (MZFR) and the Compact Sodium Cooled Nuclear Reactor (KNK)
- Karlsruhe Reprocessing Plant (WAK) and the European Institute for Transuranium (both on the premises of the Research Center)
- Baden-Wuerttemberg state collection facility
- External customers

Before radioactive waste can be delivered to HDB, it has to be declared by the customer according to the acceptance criteria regarding its material characteristics, origin, mass, volume, dose rate and nuclide inventory as well as the total activity. Data on the radioactive waste, such as
pre-treatments, the conditioning and the waste products themselves are stored in the Karlsruhe Data Bank for Radioactive Waste (KADABRA).

HDB operates several facilities for the conditioning of radioactive waste:
- Solid and liquid burnable waste is incinerated and the resulting ashes are super-compacted, resulting in a waste product suitable for final disposal.
- Solid non-burnable waste is super-compacted or fixed in cement.
- Liquid non-burnable waste is, if necessary, either evaporated and solidified or dried.

The resulting waste products are either returned to the external customers or stored in the intermediate storage facility of the HDB until the hand-over to a final disposal. At the moment, about 46,500 m³ of radioactive waste products are stored there. Currently a minimum storage time of 30 years is considered as realistic and the conditioning and declaration of the waste products is done according to the preliminary acceptance criteria of the final disposal project KONRAD. Due to this facts, steps have to be taken to ensure a secure long-time storage of the waste products, as well as to bring the declaration in line with the acceptance criteria of KONRAD.

2 Re-Declaration

In the course of the changing repository situation in Germany from the research repository named ASSE up to the repository project called KONRAD, the requirements for the declaration of waste packages has changed completely. While a minimal declaration of only the nuclides and the total activity was sufficient for the delivery to the ASSE repository, it is necessary to declare a large number of nuclides for the KONRAD final disposal. According to the increasing standards for declaration the HDB has adjusted their acceptance criteria in 1995. Therefore, the older radioactive waste as well as the resulting waste products have to be re-declared. A procedure was developed to re-declare first the waste and afterwards the waste products.

For the determination of the new waste data, the delivering institutions are divided into three groups:
- Group 1: Customers for which a nuclide composition can be calculated supported by radiochemical analysis (research reactors)
- Group 2: Customers which handle only single nuclides (institutes of the Karlsruhe Research Center)
- Group 3: HDB as a producer of secondary waste, which occurs in the process of the waste treatment

About 26,500 resulting waste products have to be re-declared. Table 1 shows the original waste, which has to be re-declared in the first step.
Table 1: Overview of the radioactive waste in need of re-declaration

<table>
<thead>
<tr>
<th></th>
<th>Number of waste units</th>
</tr>
</thead>
<tbody>
<tr>
<td>Group 1</td>
<td>24,723</td>
</tr>
<tr>
<td>Group 2</td>
<td>39,288</td>
</tr>
<tr>
<td>Group 3</td>
<td>44,029</td>
</tr>
</tbody>
</table>

2.1 Preparation of the Original Data
Before 1990, HDB's acceptance criteria did not require the declaration of all separate nuclides, if the total alpha activity and the total beta activity were below 1.0 \times 10^7 Bq and 1.0 \times 10^8 Bq, respectively. If the total activity was below the above mentioned values and a declaration of separate nuclides existed, it was not entered into the KADABRA database. To facilitate a re-declaration based on all separate nuclides, about 180,000 covering letters originating between 1977 and 1990 were re-entered manually. These data were checked for typing errors and consistency of customer, accounting, nuclide, activity, measurement unit information and balance between mother and daughter nuclides. Since the nuclide compositions of group 2 customers are based on the date 31/12/1994, the corresponding decay calculations were done for this reference date.

2.2 Determination of Re-Declaration Nuclide Compositions

2.2.1 Determination of Re-Declaration Nuclide Compositions for Research Reactors (group 1)
Conservatively estimated nuclide compositions were determined by burn-up calculations and analytic methods for different time spans and reactors parts of the research reactors. Those compositions were assigned to the respective covering letters for the re-declaration.

2.2.2 Determination of Re-Declaration Nuclide Compositions for Institutes of the Karlsruhe Research Center (group 2)
The following data is collected and evaluated to determine the nuclide compositions for institutes which handle single nuclides:

- Monthly information about changes in the inventory of radioactive materials (required by law) between 1964 and 1994
- Inventory declaration of the institutes on 31/12/1994
- The accounting documents of the facility which centrally documents all in- and outgoing radioactive isotopes of the research center institutes

Based on this data, an upper limit for each institute's handled activity is estimated and used for the determination of the following conservative nuclide compositions:

- General nuclide composition without nuclear fuels
- Plutonium nuclide composition
- Uranium nuclide composition
- Thorium nuclide composition

2.2.3 Determination of Re-Declaration Nuclide Compositions for Secondary Waste of the HDB (group 3)
The majority of all activity treated at HDB originates from the Karlsruhe reprocessing plant. Therefore, the determination of the nuclide composition is based on the activation and the fission
products of the burn-up calculation. It is corrected by analyses of the effluents of each treatment facility.

2.3 Application of the Re-Declaration Nuclide Compositions

2.3.1 Application of the Re-Declaration Nuclide Compositions for the Waste of the Research Reactors (group 1) and the Secondary Waste of HDB (group 3)
If Cs-137 or Co-60 are declared in the covering letters, this nuclide is used as the key nuclide; the declared total activity is maintained and the other nuclides are calculated by their relation to the key nuclide in the nuclide composition. If no single nuclides are declared in the covering letters, the total activity is maintained and the nuclides are calculated by their percentage in the re-declaration nuclide composition.

2.3.2 Application of the Re-Declaration Nuclide Compositions for the Waste of the Institutes of the Research Center (group 2)
The re-declaration of group 2 waste results from the correlation of the total activity with the nuclide composition. Three different cases are distinguished:
Case 1: No declaration of separate nuclides
The activity of the separate nuclides is calculated by using the total activity and the general nuclide composition.
Case 2: Declaration of separate nuclides, but no declaration of fuel elements
The activity of all separate nuclides of the nuclide composition is calculated using the total activity. Missing nuclides are supplemented. The declaration of separate nuclides is utilized if the declared activity is greater or equal to the calculated activity. If the calculated activity is greater than the declared activity, the calculated activity is used. In the end, the new total activity is calculated by adding the single nuclides’ activity.
Case 3: Declaration of separate nuclides and fuel elements
In case of the re-declaration of non fuel element nuclides, the practice is the same as described in case 2. A re-declaration of the fuel element nuclides is done only if the fuel elements are not under EURATOM accounting supervision. The re-declaration of missing fuel element nuclides is done on the basis of a special fuel element nuclide composition. This special nuclide composition is created from the isotope ratio of the elements (U, Pu, Th) in the general nuclide composition.

According to the acceptance criteria of HDB it is always mandatory to declare fuel elements in the covering letters. Otherwise, no re-declaration is necessary.

2.4 Re-Declaration of the Waste Products
On the basis of the re-declared waste data the activity during all treatment steps and of the resulting waste products is recalculated. The calculation of the new values is done by the accounting system KADABRA, and a total computing time of about one year is currently estimated.
3 Requirements and Measures for Secure Long Term Intermediate Storage

3.1 The HDB Intermediate Storage for Non Heat Generating Waste

On the premises of the Karlsruhe Research Center HDB operates an intermediate storage for non heat generating waste.

Part I of the storage (building 519) serves as an intermediate storage for drums, concrete-shielded casks and cast iron casks. It is divided into several fork-lift operated areas. The different cask types are stored separately. Drums are stacked in 4 layers and shielding casks in 3 layers.

Areas A – C of part II of the storage (building 526) serve as a storage for steel and concrete containers. The containers are a special development in accordance with the acceptance criteria of the KONRAD repository and have a storage volume of 8 m³. The filled containers are brought into the loading zone by a fork-lift. From the loading zone they are lifted into the storage areas by a 250 kN crane. Each area accommodates 7×46 containers horizontally and 8 containers vertically. This provides 2,576 containers per area and a total number of 7,728 containers in all three areas.

Preparation for the secure long term storage and quality insurance will be done in the handling area D, after it is finished in June 2002.

The packing and unpacking of the drum-filled containers takes place in the packing area E. Measurements of the dose rate and activity as well as gas analyses are taken there.

![Fig. 1: Ground plan of the storage parts I and II](image)

The storage part I consists of six storage areas with a total storage volume of 15,600 m³. The halls A – C of storage part II have a total storage volume of 61,824 m³. The HDB intermediate storage for non heat generating waste has a storage volume of 77,424 m³ altogether. On 01/06/2001 occupancy was as shown below:
Table II: Storage capacity and occupancy in the storage part I and II

<table>
<thead>
<tr>
<th>Container types</th>
<th>Number of containers</th>
<th>Occupancy [m³]</th>
<th>Capacity [m³]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Waste products drums (150- up to 400-l-drums)</td>
<td>2,097</td>
<td>857</td>
<td></td>
</tr>
<tr>
<td>Concrete cask type I</td>
<td>4,001</td>
<td>6,402</td>
<td></td>
</tr>
<tr>
<td>Heavy concrete cask type II</td>
<td>3,046</td>
<td>4,874</td>
<td></td>
</tr>
<tr>
<td>Cast iron cask type II</td>
<td>166</td>
<td>266</td>
<td></td>
</tr>
<tr>
<td>MOSAIK container</td>
<td>40</td>
<td>86</td>
<td></td>
</tr>
<tr>
<td>Sum</td>
<td>9,350</td>
<td>12,485</td>
<td>15,600</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Container types</th>
<th>Number of containers</th>
<th>Occupancy [m³]</th>
<th>Capacity [m³]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Steel container type IV (FSC)</td>
<td>2,301</td>
<td>18,408</td>
<td>61,824</td>
</tr>
<tr>
<td>Concrete container type IV (NBC)</td>
<td>862</td>
<td>6,896</td>
<td></td>
</tr>
<tr>
<td>Heavy concrete container type IV (SBC)</td>
<td>1,143</td>
<td>9,144</td>
<td></td>
</tr>
<tr>
<td>Product steel container type IV (PSC)</td>
<td>745</td>
<td>5,960</td>
<td></td>
</tr>
<tr>
<td>Steel container type II</td>
<td>135</td>
<td>675</td>
<td></td>
</tr>
<tr>
<td>(Version 1 - 4)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sum</td>
<td>5,169</td>
<td>41,083</td>
<td>53,568</td>
</tr>
</tbody>
</table>

The accumulation of waste volume until 2080 results in a predicted total storage volume of 75,900 m³, without taking a possible reduction due to transports into an eventual final disposal into account.

Fig. 2: View of the partially filled storage area C of storage part II
Accordingly, the following requirements for the waste products and containers are established:

- There must not be a build-up of sizeable amounts of gases in order to avoid a pressurization of the waste packages or the creation of flammable or explosive gas mixtures.

Different mechanism may be responsible for the creation, release and destruction of gases inside waste packages. The main reasons for gas formation are:

- Metallic corrosion
- Bacterial decomposition of organic materials
- Radiolytical decomposition

It is shown by experience and experimental results that packages filled either with loosely inserted or super-compact mixed waste are especially prone to gas formation. The formation of $\text{H}_2$ is most important, while $\text{CO}_2$ and $\text{CH}_4$ can be neglected.

- The integrity of the container must not be diminished by mechanical damage or corrosion. Improper handling of the containers could damage the seal and the integrity could be no longer ensured.

The corrosion protection could be damaged by careless filling, by the transportation conditions and by dangerous substances (e.g. solvents) in the waste. During long term storage, it is possible that moisture in the waste or humidity from the surroundings will condense on the container walls. As a result, the damaged parts of the corrosion protection will start to corrode.

Metal cast in steel drums can develop contact corrosion, since the contact of different metals with each other will cause electrochemical reactions.

- The waste packages must remain transportable and must correspond to the acceptance criteria of the KONRAD repository.

3.3 Measurements for a Secure Long Term Intermediate Storage

Due to the long intermediate storage time of about 30 years, measurements have to be taken to ensure security during the whole time. Older waste products have to be controlled visually for changes caused by corrosion or gaseous build-ups during their previous storage time. To increase security and to avoid further corrosion, the drums are cast in containers with inactive concrete, according to the KONRAD acceptance criteria.

After casting the drums, further quality assurance or re-conditioning is no longer possible. Therefore, the necessary procedures have to be realized beforehand. To implement this, the documentation of the waste products has to be revised in order to correspond to the current standard. This documentation has to be checked by officials of the Federal Office for Radiation protection. Only then does the Federal Office for Radiation Protection allow the casting of the containers.
Before the release for casting, there are different activities necessary:

- Unloading of the older drums to inspect them for corrosion
  The containers are transported from storage area A – C to the handling area E before unpacking the drums from the containers. There, the containers are unloaded at ventilated working areas with security devices such as remote controlled handling. After unloading, the dose rate is measured on the surface and in 1 m distance from the drums, wipe tests are taken and a visual control is done. Damaged drums are taken away for later re-conditioning.

Fig. 3: Examples of corroded or damaged drums

- Repair of steel containers with damaged corrosion protection
  Steel containers with damaged corrosion protection or even actual corrosion damages are sorted out and repaired according to a special regulation.

- Sampling of gases for analysis
  The amount of sampled products is determined by the federal experts (normally 30% per treatment campaign). The gas samples give information about gas builds-ups due to chemical or biological reactions. To prepare for sampling, the lids of the drums are pierced under a nitrogen atmosphere in order to insert a special sampling device. The drums are locked airtight for at least 10 days before the sample is taken. The analysis of the samples gives information about the gas composition ($H_2$, $N_2$, $O_2$ and $CH_4$) as well as the pressure inside of the drums. If the results show a gas build-up ($H_2$-content greater than 4%), all resulting waste products of the same treatment campaign are either dried or silica gel is added to the drum contents.

- Spectrometric Measurements of older waste products
  Part of the waste quality control is the spectrometric measurement of the waste. Normally about 30% of all waste products are spectrometrically analyzed to verify the declared activity. This is done with a special spectrometer for 200-l and 400-l drums. 50 drums are measured automatically in one run to determine
  - the weight,
  - the medium and maximum dose rate on the surface and in 1 m distance,
  - gamma nuclides (qualitatively and quantitatively),
  - distribution of the activity.

- Sorting of older drums and re-packing into containers
  The drums are re-packed together in new or repaired containers grouped by customer in order to fulfill the regulations of final disposal and transportation. Before re-packing the drums, the waste package quality control such as gas analyses and spectrometry has to be performed. In addition, the dose rate in different positions is measured and pictures are taken of each drum.
3.4 Casting of the Containers

Waste products which pass all quality control steps and have a documentation accepted by the Federal Office for Radiation Protection as being in accordance with the KONRAD disposal acceptance criteria can be cast in containers. First, the containers are filled with inactive concrete. The casting is done in a campaign of 24 units daily. On the next day, the containers are transferred to a special storing area, where they remain for about 4 weeks until the concrete is completely set. Federal experts control the setting and give their permission to close the containers and to put them into storage.

4 Summary

Due to the increased storage time a number of preventive measurements have to be taken to ensure a secure long term storage. The enhanced requirements for the declaration of waste products make a re-declaration of older products necessary. To minimize radiation exposure, the re-declaration is done only by readjusting the original calculations. Because of the formerly low declaration requirements, a complete reproduction of the activity is not possible, so it is necessary to construct conservative nuclide compositions for the declaration. The nuclide compositions are used in coordination with the Federal Office for Radiation Protection.

Besides the re-declaration of the waste products, a visual control for corrosion and, if necessary, a re-conditioning has to be done. To get the certification that the waste products correspond to the KONRAD acceptance criteria, a number of product quality controls such as gamma spectrometry and gas analyses have to be done.

The final step for an additional security barrier is the casting of the waste products to avoid corrosion processes and therefore, the release of radioactivity.
CHARACTERISATION AND DOCUMENTATION OF RADIOACTIVE WASTE IN SWITZERLAND

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Abstract
Switzerland has radioactive waste from nuclear power plants and from medicine, industry and research (MIR). The characterisation and documentation of this waste has to meet the provisions given by the Swiss regulatory authorities in guideline HSK/R-14 including all information needed for a safe disposal in one of the future repositories. In order to meet these requirements we apply a sophisticated system of radiological measurements and extended activation calculations. In this paper we report on the inventory of waste packages with operational waste from the NPP by applying correlation factors and with activated components - including decommissioning waste - by a system of activation calculations. Finally we present results for reprocessing waste and spent fuel (MOX and UO2). All results are integrated into the computer-based "Information System for Radioactive Materials" (ISRAM), which was developed to document the whole spectrum of Swiss radioactive waste and materials. This allows a complete monitoring and tracking of all radioactive materials produced or used in Switzerland.

1 Basics

According to the guideline HSK/R-14 [1] of the Swiss Nuclear Safety Inspectorate (HSK) all radioactive waste (not only existing waste but also waste expected in the future) has to be specified. These specifications are checked in the framework of a disposability certification procedure (ELFB) with several steps by Nagra. Such certificates are prerequisites for the clearance of conditioning methods by HSK. The benefits of this procedure are on one hand that wastes produced at this time are conditioned in such a way that they can directly be disposed of in a future geological repository. On the other hand it is ensured that the pertinent data for waste management (interim storage, transport, disposal) are recorded during conditioning and do not have to be generated at later times, which would cause significantly higher expenditures. All important data describing a waste package that have emerged during the disposability certification procedure are saved in a database system that was especially developed for this purpose. Table I identifies typical information on waste types as contained in the "Information System for Radioactive Materials" (ISRAM). In addition, detailed characteristics of individual waste packages are appended to the database in the format of a datasheet. By means of the parameters "Date", "Transport data" and "Storage location" in Table I, the "life history" of a waste package can be described in the manner of a log-book.

This database system has been supplemented by several computational tools that perform statistical data evaluation, decay, activation and burnup calculations, and provide complementary information for nuclide inventories. These tools are to be addressed in the following sections.
Table I: Structure and declarations of waste packages and containers of radioactive materials

<table>
<thead>
<tr>
<th>Identification number</th>
<th>Operator number</th>
<th>Waste sort</th>
<th>Date</th>
<th>Preparation of conditioning</th>
<th>Final conditioning</th>
<th>measurement of dose rates and nuclides</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>Total mass</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Transport data</td>
<td></td>
<td></td>
<td>Storage location</td>
<td>storage place coordinates</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dose rates</td>
<td></td>
<td></td>
<td>Surface contamination</td>
<td>$\gamma$- and $\beta$-</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Materials</td>
<td>Masses of all known materials (raw waste and matrix)</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Calculated nuclides</td>
<td>via correlation factors or activation calculations (NPP-operational waste)</td>
<td>via activation- and burnup-calculations (reprocessing waste, spent fuel)</td>
<td></td>
</tr>
</tbody>
</table>

2 Characterisation of Radioactive Waste and Materials

2.1 Spectrum of Waste and Materials
Waste and materials to be characterised consist of:

- NPP operational waste
- activated reactor components
- waste from decommissioning of the Swiss NPPs
- waste from medicine, industry and research (MIR wastes)
- waste from reprocessing of spent fuel
- spent fuel assemblies (for direct disposal)
2.2 Radiological Characterisation

The methods applied for the radiological characterisation will be described in the following sections.

2.2.1 NPP operational waste

These wastes include resins, concentrates, slurries and (combustible) mixed wastes. From most waste sorts samples are periodically taken and analysed. The selection of nuclides that are measured is governed by the importance of nuclides with respect to operational and long term safety of the repository. The list of safety relevant nuclides that are measured is: H-3, C-14, Cl-36, Mn-54, Fe-55, Co-60, Ni-63, Sr-90, Tc-99, I-129, Cs-134, Cs-137, Ce-144/Pr-144, U-235, U-238, Pu-239/240, Pu-241, Am-241, Cm-244. From these measurements correlation factors to the key nuclides Co-60 and Cs-137 are determined enabling the calculation of inventories of waste packages by measuring the key nuclides or at least dose rates. As the list given above includes some nuclides, which are very difficult (and costly) to measure, the number of samples analysed must be kept small. Currently three representative and NPP-specific sets of correlation factors are determined for each waste sort, including periods with and without fuel leakage. In order to check the time dependence of the nuclide fingerprint, data for a limited number of nuclides are collected in short time intervals, and primary water samples, as well as smear-test samples from various regions, are analysed.

2.2.2 Activated reactor components

These waste components have been exposed to a significant neutron flux mostly over a period of several years. Typical representatives are BWR fuel channels, incore instrumentation and small parts from maintenance work. By activation of the impurities in these materials, a large number of short- and long-lived radionuclides is generated. In addition to nuclides usually considered, like Co-60, C-14, or Ni-63, also fission products and actinides from activation of uranium impurities in the base material are relevant. In the case of zircalloy from fuel channels, e.g., an uranium impurity of 1-2 ppm has been measured leading to specific activities of approximately 60 MBq/kg for Cs-137 and to a total α-activity of approximately 20 MBq/kg in the base material. These data have been validated by calculations using activation codes [2,3] designed for calculations of reactor components outside the fuel region ("infinite dilution"). Essentially all of this waste is characterised by these specific codes, and random samples are just taken to validate the codes.

2.2.3 NPP decommissioning waste

Assuming that all 5 Swiss NPPs have an operation time of 40 years, the first NPP decommissioning project in Switzerland would not start before the year 2010. The owners of nuclear facilities are required to make financial contributions to a joint decommissioning fund (assuming an operational lifetime of 40 years). The fund must cover all costs of decommissioning, including dismantling and disposal of decommissioning waste. Fund raisings are subject to periodic revision. With a view to updating the financial model, new decommissioning studies have been performed in the years 1998-2001. The activation codes mentioned in section 2.2.2 were used to determine the masses and inventories of the activated materials by dividing the whole reactor into volume elements, for each of which the neutron flux and nuclide inventory was calculated (Figure 1). Details of this method including some results can be found in Ref. [4].
Figure 1: Division of a NPP reactor into partial volumes for the activation calculations: E = reactor pressure vessel internals; R = reactor pressure vessel; B = Biological shield; D = Drywell
2.2.4 Waste from medicine, industry and research (MIR)
These wastes from various sources (industry, hospitals, sources used in research projects) are collected on a regulated basis. The corresponding material and nuclide inventories are extracted from a declaration sheet that has to be provided by the producer of the waste. These declaration sheets have a standardised form, and the producers are given advice in filling out the forms. Wastes coming from research facilities (like targets, beam dumps, activated shielding material, magnets) are described by application of sophisticated codes that calculate inventories after activation of materials by protons, neutrons or after spallation.

3 Reprocessing Waste
Currently only the vitrified high level waste in coquilles is transported to the Swiss interim storage facility ZWILAG. All QA measures with respect to the reprocessing process, the raw waste product and the coquille are taken by the manufacturer (COGEMA or BNFL), who also provides a basic list for the nuclide inventory in electronic form and on paper. Via an interface package-specific data can be directly imported into the ISRAM system. By the application of computer codes [3,5,6] the basic list for the nuclide inventory can be supplemented by those nuclides that are safety relevant for a later deep geological disposal. For each coquille the extended nuclide inventory list as well as the data of the storage container are added to ISRAM. Within ISRAM various parameters of the storage container can be automatically calculated from the data of the enclosed coquilles.

4 Inventories of Spent Fuel
4.1 Activities
Among the waste package types that have been implemented in the ISRAM system, those for spent fuel have the most extensive documentation. Essentially each new fuel assembly defines a new waste package type. The corresponding datasheet contains nuclide inventories of the fuel elements, which are calculated with various well established computer codes [3,5,6]. Like in the case of the reprocessing waste, specific variables of the storage container can be automatically calculated from the data of the enclosed fuel assembly.

4.2 Thermal power
The thermal power of a fuel assembly plays a crucial role in determining the earliest possible time for a direct disposal in a geological repository. There are various computer codes available to calculate the thermal power of spent fuel. In Table II a typical result obtained with the ORIGEN-ARP module of the SCALE-package [6] is shown. In the second row the total thermal power of UO2 fuel (with a burnup of 55 GWd/tHM) is shown in units of Watt/tHM for a number of cooling times after discharge. In the following rows those radionuclides are listed that give the dominant contributions to the decay heat. It can be seen that Cs-137, Sr-90 and their decay products provide most of the thermal power in that range of cooling times and determine the decrease in the heating power by their half-lives.
Table II: Thermal power of UO2 fuel (with a burnup of 55 GWd/tHM) in units of Watt/tHM.

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>35.0 yr</th>
<th>45.0 yr</th>
<th>55.0 yr</th>
<th>65.0 yr</th>
<th>75.0 yr</th>
</tr>
</thead>
<tbody>
<tr>
<td>total</td>
<td>1.22E+03</td>
<td>1.03E+03</td>
<td>8.77E+02</td>
<td>7.56E+02</td>
<td>6.51E+02</td>
</tr>
<tr>
<td>Ba137M</td>
<td>2.94E+02</td>
<td>2.32E+02</td>
<td>1.65E+02</td>
<td>1.47E+02</td>
<td>1.17E+02</td>
</tr>
<tr>
<td>Y90</td>
<td>2.97E+02</td>
<td>2.32E+02</td>
<td>1.65E+02</td>
<td>1.47E+02</td>
<td>1.17E+02</td>
</tr>
<tr>
<td>Am241</td>
<td>1.69E+02</td>
<td>1.36E+02</td>
<td>1.08E+02</td>
<td>1.08E+02</td>
<td>1.09E+02</td>
</tr>
<tr>
<td>Pu239</td>
<td>1.87E+02</td>
<td>1.73E+02</td>
<td>1.60E+02</td>
<td>1.47E+02</td>
<td>1.36E+02</td>
</tr>
<tr>
<td>Cs137</td>
<td>8.69E+01</td>
<td>6.90E+01</td>
<td>5.48E+01</td>
<td>4.34E+01</td>
<td>3.45E+01</td>
</tr>
<tr>
<td>Cm244</td>
<td>8.47E+01</td>
<td>5.78E+01</td>
<td>3.94E+01</td>
<td>2.68E+01</td>
<td>1.63E+01</td>
</tr>
<tr>
<td>Sr90</td>
<td>6.41E+01</td>
<td>4.76E+01</td>
<td>3.72E+01</td>
<td>2.90E+01</td>
<td>2.28E+01</td>
</tr>
<tr>
<td>Pu240</td>
<td>2.03E+01</td>
<td>2.03E+01</td>
<td>2.04E+01</td>
<td>2.04E+01</td>
<td>2.04E+01</td>
</tr>
<tr>
<td>Pu239</td>
<td>1.20E+01</td>
<td>1.20E+01</td>
<td>1.20E+01</td>
<td>1.20E+01</td>
<td>1.20E+01</td>
</tr>
<tr>
<td>Eu154</td>
<td>6.46E-00</td>
<td>2.89E-00</td>
<td>1.29E-00</td>
<td>5.73E-01</td>
<td>2.56E-01</td>
</tr>
</tbody>
</table>

In Figure 2 the cross sections of two conceptual fuel disposal container designs (with an approximate length of 500 cm and a diameter of 105 cm) by Nagra for 9 BWR and 4 PWR fuel assemblies are shown. There are two major restrictions for loading of such containers:

- there is only room for a maximum of 9 (4) fuel elements from BWR- (PWR-) reactors per container,
- the heat output is currently limited to 1500 Watt per container, because higher outputs would lead to temperatures in a surrounding bentonite barrier that could influence its positive insulating features.

Figure 2: Cross section (top view) of conceptual fuel container designs for 9 BWR and 4 PWR fuel assemblies

Figure 3 shows the thermal power of reprocessing waste and PWR spent fuel per disposal container for a number of different loadings as a function time. Adopting that a fuel assembly contains approximately 430 kg of initial heavy metal (IHM), the total mass of initial heavy metal in a container would sum up to roughly 1700 kg. While a container loaded with 2 standard coquilles could already be brought into a geological disposal 30 years after the delivery of the coquilles, containers filled with spent fuel at least require a cooling time of 45 years. However, this lowest cooling time is only valid for UO2 fuel assemblies with a moderate burnup of 48 GWd/tHM. In case of UO2 fuel with a higher burnup, or in case that also a MOX fuel assembly (which has a thermal power 2-3 times larger than UO2 fuel) is loaded into the container, the required cooling time increases up to 75 years after discharge. In order to avoid waiting for such long times, one would try to mix the "youngest" and "hottest"
fuel assemblies coming from a NPP with older and "cooler" fuel assemblies in such a way that the heat output limit is not exceeded. At this point the problem arises to:

- first calculate the thermal power spectrum of all fuel assemblies from a NPP, and
- then to find the best distribution of all these fuel elements into disposal containers, which is the distribution that requires a minimum number of the expensive containers.

This problem has to be solved for an adopted time for the disposal year, for a given operating time of a NPP, and for a burnup-scenario, because all these parameters determine the number and the cooling time of the fuel assemblies that are accumulated by a NPP. We solved this problem by generating a computer program that, subsequent for each calendar year, keeps track of the number of discharged fuel assemblies of a NPP and follows their cooling time until the year of geological disposal. At this time the thermal power spectrum of all fuel assemblies is determined and the combinatorial optimization (or minimization) problem to find the best distribution is solved, under the assumption that the fuel assemblies can still be arbitrarily accessed and put into disposal containers. By this way a complete "life history" (to some extent "life future") of a fuel assembly can be described up to the final assignment to a "best" disposal container in order to minimize the costs. When studying several burnup-scenarios, it was recognised that a change to very high burnups could significantly increase the number of needed disposal containers and with it the back-end-costs.

Figure 3: Thermal power of reprocessing waste and spent fuel for various burnups
5 Summary

For the management of radioactive wastes arising in Switzerland the database system ISRAM ("Information System for RAdioactive Materials") has been implemented and is used by the producers and by Nagra. Due to the modular structure of the system it can be easily adapted to new situations. The system fulfills both the requirements of the HSK/R-14 guideline and the needs of the producers of waste and of Nagra. It is equipped with various computational tools that allow to complement nuclide inventories and to perform statistical data evaluations, decay calculations, as well as activation and burnup calculations. As transport and storage location data are also recorded in the system, it is possible to follow the "life history" of a waste package in the manner of a log-book. If information on the NPP operating time, a disposal year, and the average burnup of fuel elements is given, even a cost optimised packing of fuel assemblies into disposal containers can be accomplished.

6 References

PROGRESS IN DOCUMENTATION STANDARD FOR LONG TERM INTERIM STORAGE OF RADIOACTIVE WASTE


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Abstract
For a long term interim storage of radioactive waste up to 40 years a specified documentation of the radioactive waste container or drum to be stored is essential. A typical documentation of a radioactive waste container contains a description of the features of the waste container and the content of waste, the origin of the waste, the waste-treatment and the verification of a stable waste-product. A calculation of activity is included as well. Based on this documentation it is assumed that all relevant information on the waste package characteristics is saved over the whole period of long term interim storage and gives a preparation for the delivery to the final repository.

1 Introduction
For a long term interim storage of radioactive waste up to 40 years a specified documentation of the radioactive waste container or drum to be stored is essential.

A detailed documentation of the radioactive waste has been developed with respect to the acceptance criteria of the sites for final disposal /1/, /2/ and interim storage /3/, /4/, /5/.

The requirements of a container documentation include all information concerning the waste, a verification of a stable waste product and a stable waste container for long term interim storage as well as a preparation of a documentation for a final storage in approx. 30 to 40 years.

2 General content of a documentation
A typical documentation of a container or drum filled with radioactive waste contains 10 chapters:

- Documentation sheet
- Plan of waste-treatment
- Detailed description of the conditioning procedure
- Calculation of the activity
- Certificates of radiological control
- Verification of a stable waste product suitable for long term interim storage
- Certificates of sealing of the container
- Documentation of the container properties
- Transport documents
- Various

The listed content of a documentation will be explained in the following chapter.
3 Content of a documentation in detail

3.1 Documentation sheet

A documentation sheet of two pages (page 1 is shown in Figure 1) summarizes the main characteristics of a waste container or drum.

It contains an identification of the waste container (e.g. KKW6500001) and a declaration of the raw waste, the conditioning procedure and the properties of the conditioned waste. Information about important data on activity e.g. $\beta/\gamma$-activity, $\alpha$-activity and special nuclides are given as well as a declaration of the mass, the dose rate and the contamination of the container.

The documentation sheet is usually signed by experts (e.g. TÜV) before approval by the authorities for delivery to the interim storage site. An additional documentation sheet is available for the future final repository Konrad in lower saxony (FRG).

Figure 1: Documentation sheet (Page 1)
3.2 Plan of waste treatment

Every waste treatment is based on a plan which is checked by experts and approved by the authorities. The plan of waste treatment contains in the first two sections all preliminary works (e.g. sorting of the waste in the nuclear power plant) and the transport to the site of conditioning.

The following section regulates the procedure of the waste treatment: combustion, cutting, compacting, drying, packaging etc. The verification of a stable waste product for long term interim storage is included as well. The last section concerns the documentation of the waste product and verification steps of the whole procedure by experts and approval by the authorities before storage. The transport to the interim or final storage is included as well.

3.3 Detailed description of the conditioning procedure

A detailed description of the conditioning procedure includes:

- Information concerning the amount, the features and the origin of the untreated waste.
- Detailed description of the waste-treatment: cutting, compacting, drying, packaging etc. Special characteristics during the conditioning-procedure are included as well.
- Documentation of the waste products: Each treated waste product within the container or drum can be related to its origin.
- Results of the conditioning procedure, e.g. amounts of released water or materials for the stabilization of the waste, documents of the operational-procedure of the waste conditioning system.
- Pictures of the waste container and waste products and check for conformity with the acceptance criteria of the sites for interim storage or final disposal.
- Overview of the whole campaign of waste treatment.

Figure 3 (see Appendix) shows a typical graphic of a documentation concerning the waste flow and waste conditioning of a campaign of disposal of scrap metal.

3.4 Calculation of the activity

The calculation of the activity is performed with respect to the acceptance criteria of the sites for interim storage or final disposal. In all acceptance criteria (1 to 5) the declaration of the β/γ-activity and the α-activity is required. In accordance to additional requirements in the acceptance criteria a various number of nuclides are declared as well. The present acceptance criteria of interim or final storage in Germany distinguish between nuclides to be declared regardless of their amount of activity and nuclides to be declared in dependence on their activity.

The calculation is also performed under consideration of the physicochemical characteristics of the waste. For example the enrichment of some nuclides in ion-exchange resins is taken under consideration. Inhomogeneity of the waste is also taken under consideration if possible.

For the derivation of the nuclide-vector used for declaration mainly
analytical measurements (γ-scanning)

- a consideration of eventual cross-contamination and
- utilization of special calculations of nuclide-distributions in nuclear-fuels and other materials after irradiation

are taken into account as well as special information received from the nuclear power plant. The information from the nuclear power plant, e. g. special nuclide-derivation in waste of different origin have priority over all other theoretical nuclide-correlations or correlations obtained from analysis of similar waste.

The first step in calculation of the total activity is the declaration of a few „key-nuclides“, e. g. Co-60, Cs-137 and total alpha. Then the calculation of the total activity is performed by using the elaborated distribution of nuclides in relation to the „key-nuclides“.

The last step is the important check of the activities for conformity with the acceptance criteria of the sites for interim storage or final disposal. In case of an exceed of limits for nuclide-concentrations or the total activity, estimations are given for the activity for decay and forseeable conformity of nuclide-limitations in the future. In case of a large exceed of limits for nuclide-concentrations a proposal is given for additional treatments of the waste to obtain a higher quality of the conditioned waste with higher limits of nuclide concentrations. This additional treatment could be the stabilization of the waste with concrete to obtain inflammability.

3.5 Certificates of radiological control

Each measurement of dose rate and contamination of a container or drum with radioactive material will be documented in special papers and signed by authorized members of the radiation protection. These papers (for example shown in Figure 2) give the evidence for the compliance of the waste-container with the acceptance criteria of the sites of interim storage or final disposal.

![Figure 2: Points of measurement of the dose rate of a waste container](image-url)
3.6 **Verification of a stable waste product suitable for long term interim storage**

A verification of the stability of the conditioned waste before long term interim storage is presently required. Three main sections of analysis can be summarized:

- Analysis of samples of the treated waste
- Results obtained from measurements of the humidity of the inner atmosphere of the container: No free water within the container.
- Gas analysis of the inner atmosphere (time dependant):
  - $\text{H}_2$: Corrosion within the container possible
  - $\text{CH}_4, \text{CO}_2$: Bacteriological reactions within the treated waste

All acceptance criteria of the sites for interim storage (1/3 to /5) or final disposal (1/1 to /2) request stable waste products with negligible gas-production rates or bacteriological reactions within the container.

Documents of additional analyses can also give evidence for a successful conditioning procedure. Based on these data it can be assumed that the waste product will be stable for a long term interim storage.

3.7 **Certificates of sealing of the container**

Container with radioactive waste originated from a nuclear power plant are sealed before transportation to a site of conditioning.

After the waste treatment the conditioned waste is packed into a container, a drum or a shielding box. Sealing of all container or shielding boxes takes usually place before transportation to the sites for interim storage.

Documents of removing of seals from container with raw radioactive waste and the sealing of a container filled with conditioned waste will be found in this chapter of the documentation.

3.8 **Documentation of the container properties**

A progress in documentation standards for long term interim storage also concerns the detailed documentation of the utilized container, drum or shielding box for long term interim storage.

The results of the manufacturing of the container, drum or shielding box must be shown in detail in this section of the documentation:

- Certificates of the material of the container, drum or shielding box
- Quality and thickness of the coating
- Certificates of the manufacturing
- Certificates of special tests of the container, drum or shielding box before loading of radioactive waste

Experiences from waste containers or drums who had encountered a long term interim storage up to 15 years before delivery to the final repository in Morsleben (FRG) show that a qualified selection of the drum material and the coating is essential for a successful long term interim storage.
3.9 Transport documents

Transport documents of the transport of the radioactive waste from the nuclear power plant to the different conditioning-sites are also included in the documentation. These transport documents verify the "history" of the treated waste from the origin in the nuclear power plant to the different sites of conditioning (see chapter 3.3).

3.10 Various

All documents who cannot be assigned to the above mentioned topics are collected in the last chapter of the documentation.

For example results of \( \gamma \)-scanning and tomographic measurements of the waste product or drum as well as results of special nuclide analysis can be found in this chapter. Documents upon preliminary treatments of the radioactive waste are included as well.

4 Conclusion

For a long term interim storage of radioactive waste a specified documentation is essential. The documentation of GNS/WTI has been developed with respect to the acceptance criteria of the sites for interim storage (/3/ to /5/) and final disposal (/1/ to /2/).

It can be assumed that future acceptance criteria of other final repositories will include similar requirements for quality of the waste products, the waste container and the calculations of activity.

Based on the documentation with detailed information on the waste products, the treatment, the container and the activity it can be assumed that all relevant information on the waste package characteristics is saved over the whole time of long term interim storage of approx. 30 to 40 years.

Before delivery of a container to the final repository thereafter only a visual control, a new measurement of the dose rate and an actualization of the activity declaration for decay will be necessary.

In case of a necessity for repacking of a waste container, this procedure will be simplified by having detailed information concerning waste content, waste treatment, activities and others.
5 References

/1/ Anforderungen an endzulagernde radioaktive Abfälle
      Endlagerungsbedingungen Schachtanlage Konrad
      Stand: Dezember 1995
      BfS-ET-1B-79

/2/ Anforderungen an endzulagernde radioaktive Abfälle und Maßnahmen
      zur Produktkontrolle radioaktiver Abfälle, Teil 1: Endlagerungsbedingungen
      Stand: September 1993 / August 1996
      BfS-ET-14/92-REV-2 / ET-1B-85

/3/ Brennelementlager Gorleben GmbH
      Technische Annahmebedingungen für das Abfalllager Gorleben
      Stand: 12/95

/4/ Kernkraftwerk Unterweser
      Technische Annahmebedingungen für die Einlagerung radioaktiver Abfälle in
      der externen Lagerhalle
      Stand: 24.10.2001

/5/ GRB-Sammelstelle Bayern für radioaktive Stoffe GmbH
      Annahmebedingungen für den Betrieb der EVU-Lagerhalle der Sammelstelle Mitterteich
      Ausgabe: Dezember 2001
Nuclear power plant (KKW)
Packaging of 32080 kg scrap metal into 80 drums
Loading of the drums into two 20'-Container
Number of campaign: KKW-2001-006/SRT

Conditioning site of GNS in Duisburg
Sorting of the scrap metal
Metal suitable for melting at SNU in Krefeld: 19.672 kg
Material for super compaction at GNS-Duisburg: 12.408 kg

Conditioning site of GNS in Duisburg
Supercompaction with the FAKIR
Result: 96 cartridges with supercompacted outsorted waste (not suitable for melting)

SNU Krefeld
Melting procedure

Figure 3: Waste flow and waste conditioning of a campaign of disposal of scrap metal (see chapter 3.3)
Activity Declaration and Assessment
ACTIVITY DECLARATION FOR RADIOACTIVE WASTE – METHODS AND APPLICATION

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Abstract
Activity determination for radioactive waste today requires nuclide specific data for a number of purposes. In particular, it is needed for activity declaration for waste storage and disposal. Requirements for these tasks are outlined. Available methods are described starting from direct measurement to purely analytical methods like burn-up calculations. Selection and application criteria for these methods are demonstrated for exemplary cases in German practice. Emphasis is given to more recent developments which aim at optimisation of methods, justification and qualification and the enlargement of the applicability range.

1 Introduction
Dealing with radioactive waste today requires knowledge about nuclide specific activity inventories in the waste. The purpose for this requirement may be quite different. In safety analyses the nuclides specific inventories are often an essential prerequisite. This may concern e.g. the analysis of waste handling activities for transport, storage and disposal, the long-term safety analysis for a repository or accident analyses. In all these cases inventories of radiologically relevant radionuclides have to be characterised to provide the basic input for the analyses.

Nuclide specific data may also be required to establish waste acceptance criteria. If these criteria are formulated in a waste specific manner, again an nuclides specific input is needed to fix reasonably realistic activity limits. If the activity limits are calculated backwards from primary dose limits e.g. for the public in a generalised manner the nuclide specific inventories may at least be helpful to assess their compatibility with the derived acceptance criteria.

Nuclide specific activity inventories may furthermore be necessary for designing and operating waste facilities e.g. with respect to radiation protection. Finally it has become a useful tool to apply nuclide specific compositions e.g. of α emitters as a kind of fingerprint for the waste by which its origin can be determined.

The most prominent purpose for determination of nuclide specific activity inventories, however, comes from the requirements for declaration of radioactive waste for storage and disposal. This will be the subject of the following presentation.

2 Methods for activity determination
A number of methods have been developed to determine the activity inventory of a waste package or a sample thereof for the purpose of activity declaration. According to their relation to the basic quantity to be declared – either activity or activity concentration – one can distinguish between direct and indirect methods.
2.1 Direct measurement

The most obvious method for activity determination is the direct measurement either on raw waste, the waste product or the final waste package. Without destructive procedures the method can only be applied for strong γ or neutron emitters because of the self shielding of the waste and the high attenuation coefficient for the other radiation types. If destructive sampling is performed a variety of radiochemical methods is available to determine any kind of activity if it is above the detection limit of the measurement equipment.

Obviously, the latter of these methods cannot be applied for activity determination on a routine base. The restriction of the other measurement methods to strong γ or neutron emitters leaves a large number of radionuclides that have to be declared but are not measurable in this way. Therefore indirect methods had to be developed in the past and are amply available today. However, measurement results from direct methods are a valuable if not essential input to the indirect methods to ensure their applicability.

2.2 Indirect methods

Indirect methods determine activities or activity concentrations by analytical calculations. To ensure that the results are in an appropriate agreement with reality an empirical component is introduced. This means that measurement results from direct measurements are used as basic input for the analytical calculations or for normalising calculation results.

The first and most simple approach is to determine by destructive sampling the activity concentrations for the radionuclides of interest on a sample that is considered as representative for a large amount of waste, e.g. a wastes belonging to the same batch or produced in the same conditioning campaign. This is often called the representative spectrum method since it determines the activity spectrum of a number of radionuclides, which represents the activity distribution for the whole batch, campaign or equivalent. In practice, this means that either the activity concentrations are assumed as equal for all waste to be declared or the total γ activity is measured on each waste package and the relative distribution of the representative spectrum is applied to determine the radionuclide specific inventories. In this case no consideration is given to the different behaviour of the individual radionuclides in generation and in transport to the waste, since it should be comparable for the whole amount of waste that is characterised. On the other hand only a limited number of samples are taken and measured directly. Thus only a limited check is possible on the assumption of homogeneous activity distribution in the waste.

To overcome the latter restriction or to apply the semi-empirical approach to non-homogeneous waste or a combination of different waste types and to establish a broader statistical base to check consistency of data the scaling factor method was developed. The idea is that radionuclides originating from the same source and showing the same physico-chemical behaviour on its transport to the waste should have a constant ratio of their activity concentrations. This ratio is evaluated from a number of direct destructive measurements and averaged. If one of these comparable nuclides is easily measurable the other radionuclide can later on be determined from the activity of the first nuclide by calculation with the predetermined ratio. The easily measurable radionuclide is usually called a “key nuclide”. The most common examples are Co 60 and Cs 137.

Special semi-empirical methods had to be developed for Tritium, which does in general not correlate to common key nuclides. One approach is to determine average concentrations of H-3 in different waste streams. A slightly more differentiating method is the derivation of Tritium con-
centrations in waste streams from the Tritium concentration in the primary coolant by means of transfer factors. The transfer factors describe the transition ratio of Tritium in different conditioning steps (e.g. reactor coolant cleanup system, liquid radwaste treatment system). Comparable methods sometimes are applied for C 14 and Cl 36.

If a radiologically relevant radionuclide is not measurable at all, all semi-empirical methods fail. In this case analytical mathematical methods may offer an alternative. If e.g. the build-up of the radionuclide is dependent on the burn-up of nuclear fuel in a reactor the activity can be determined by burn-up calculations, for which computer codes are available. Since burn-up calculations provide only information on the generated activity the transport behaviour of the radionuclide of interest has to be considered separately.

A comparable approach is the calculation of nuclide specific activities from activation calculations. Here, the build-up by activation is calculated for two radionuclides, from which one has a measurable activity. The determination of Fe 59 from Fe 55 or Zr 93 from Zr 95 may serve as examples.

Finally decay calculations may be used for the activity evaluation of radionuclides that appear as daughters of measurable radionuclides but cannot be measured themselves. Transient or secular equilibria between mother and daughter nuclides fall into this category.

Today in Germany all these methods are applied. The majority of the radionuclides to be declared are determined by a further developed version of the scaling factor method. The relationship between the radiologically relevant radionuclide and the key nuclide is derived by regression analysis and instead of the measurement data their logarithmic value is used to open the method for non-linear relationships between radiologically relevant and key nuclide. Figure 1 shows an example for the evaluation of measurement results from radiochemical analyses by this method.

![Figure 1: Correlation of Am 241 to Pu 239 for PWR waste streams](image-url)
3 Application for Activity Declaration

3.1 Selection criteria

All the methods described above have been developed for individual radionuclides or groups of radionuclides, for individual waste streams or groups of waste streams and for different scenarios by which the activity has been generated. Therefore, if a method is selected for application it has to be checked if the original assumptions and conditions behind it are still fulfilled. Table 1 shows an overview.

Table 1: Methods for activity determination and their prerequisites

<table>
<thead>
<tr>
<th>method</th>
<th>selection criteria</th>
<th>examples</th>
</tr>
</thead>
<tbody>
<tr>
<td>direct (non-destructive)</td>
<td>hard γ or n emitter routinely measurable from outside a waste package</td>
<td>Co 60, Cs 137 (key nuclides), Co 58, Mn 54, Cc 144</td>
</tr>
<tr>
<td>representative spectrum</td>
<td>homogeneous waste, representativity of sample, comparable nuclides generation and transport history</td>
<td>ion exchange resins from the same treatment system, evaporator concentrates from the same liquid waste source</td>
</tr>
<tr>
<td>scaling factor</td>
<td>hard-to-measure radionuclides that can be correlated to easily measurable nuclides</td>
<td>activated corrosion product, fission products, some α emitters</td>
</tr>
<tr>
<td>mean concentration in waste stream</td>
<td>homogeneous waste, low fluctuation in source</td>
<td>H 3, C 14, Cl 36</td>
</tr>
<tr>
<td>burn-up calculation</td>
<td>generation by activation of nuclear fuel or as fission product, comparable transport behaviour</td>
<td>α: Np 237, Am 243, Cm 245, Cm 246, Pu 242, Th 230, Th 228, U 234, U 236, FP: Eu 152, Eu 155, Sn 126, Sm 151</td>
</tr>
<tr>
<td>activation calculation</td>
<td>generation by comparable n-flux in reactor or its environment, comparable transport behaviour</td>
<td>Fe 59, Zr 93, Sb 124, Zn 65, Ca 41</td>
</tr>
<tr>
<td>decay calculation</td>
<td>mother-daughter relation, ( t_{1/2}(\text{mother}) &gt; t_{1/2}(\text{daughter}) ), comparable transport behaviour</td>
<td>Ra 224 (Th 228), Nb 95 (Zr 95)</td>
</tr>
</tbody>
</table>

3.2 Evaluation of data and application

To put the above methods into practice in most cases numerical tools have to be provided to determine the input parameters for activity declaration. In Germany, this is achieved for the semi-empirical methods mainly by a code named ELSA II, which can deal with the measurement data bases and offers different data selection and evaluation methods. Figure 2 demonstrates the numerical output of the evaluation of a correlation, which can be used for routine declaration. The selected data come from some western European countries including Germany. The waste streams considered are evaporator concentrates and ion exchange resins from pressurized water reactors.
For burn-up calculations public domain or commercial programs are at hand, which can be used for a wide range of purposes [1]. However, it has to be checked if the libraries behind the calculations are appropriate for the application purpose. A less broad selection is available for activation calculations. These codes are mostly restricted to certain types of installations and designs. Furthermore, they mostly were elaborated for other purposes than activity determination on waste. Therefore their application usually requires more efforts than other methods. Consequently, this method is restricted to a few application areas, which cannot be covered by other determination methods. The most common example is the activity determination for core internals.

Figure 2: Evaluation results for the correlation of C 14 to Co 60 for European PWR

The results of all the activity determination methods described have to be converted in a tool, that enables routine activity declaration for individual waste packages. The basic principles in the development of such tools commonly are:

- activity determination by direct measurement should be kept to a minimum
- results of direct measurements have the highest priority
- a ranking for the available methods is performed
- the activity inventories of key nuclides for scaling factor methods or analytical methods is established e.g. by
  - direct measurement
  - measurement of dose rate and application of conversion functions
  - total γ activity measurement and fixed relation between key nuclides and
any combination of the above methods

In all countries where radionuclide specific activity determination and declaration is required more or less sophisticated computer codes have been developed for this purpose. In Germany, the AVK code and its derivative the ReVK code, both developed by ISTec are the most often applied codes for this purpose [2].

4 Advanced application

In recent years further developments of methods for activity declaration have taken place. They have been directed to the application of different methods in combination, the application in new areas, the improvement of evaluation tools and to better justification of the assumptions and criteria behind the application.

4.1 Combination of methods

Since burn-up calculations provide only information on the generated activity the transport behaviour of the radionuclide of interest has to be considered separately. This difficulty can be overcome if measurable radionuclides with comparable physico-chemical behaviour exist, that can be calculated by burn-up calculations, too. In this case a ratio between the measurable radionuclide as an auxiliary nuclide and the nuclide of interest can be evaluated from the burn-up calculations. The activity of the auxiliary radionuclide can be determined either by direct measurement or by semi-empirical methods (e.g. via key nuclide). The activity of the relevant radionuclide is then calculated from the ratio determined by burn-up calculations.

Typical examples for this methods are α emitters or higher actinides that can be related to U 238, Pu 239, Cm 244 or total α emitters [3] or Ru 103 has been determined from Ru 106 [4]. Figure 3 demonstrates the quality of the method for the example of Pu 241.

![Figure 3](image-url)  
Correlation of Pu-241 to Co-60 for PWR determined by correlation of measurement data and burn-up calculations
The activity determination was first performed by deriving a ratio for Pu 241 and Pu 239 from burn-up calculations and then correlating Pu 239 to Co 60. The result is shown as a dotted line in figure 3. The solid line is from the direct correlation of Pu 241 to Co 60. Since in this case both methods were applicable, the comparison could be used as a qualification of the new method. As figure 3 shows, an excellent agreement between directly (measurement data) and indirectly (burn-up calculations) calculated correlations exists.

A comparable approach is the calculation of nuclide specific activities from activation calculations. Here, the build-up by activation is calculated for two radionuclides, from which one has a measurable activity. The determination of Fe 59 from Fe 55 or Zr 93 from Zr 95 may serve as examples.

4.2 Improvement of evaluation tools

With respect to recent developments in disposal policies in Germany there is a recommendation to declare radionuclide inventory down to very low levels, e. g. some Bq/package. This results in an increasing interest in short lived radionuclides which are generated as decay products.

This trend requires the development of an upgrade for the validated activity calculation procedures to consider complete decay chains. In the past activity build up processes were calculated by use of numerical integration. The respective calculation parameter were defined as constants limiting the application to a fixed number of radionuclides.

The new approach introduced by ISTec is based on the Bateman solver using variables as parameter. These variables are stored in external tables which can be accessed and updated by the user. Thus, a significant enlargement of flexibility is gained. Various waste producers applying this new activity calculation procedure are enabled to consider a wider nuclide spectrum. A comparative view on the results of the former calculation versus this new procedure justifies this improvement: Radiological properties determined by this new approach meet the practical reality more detailed without changing the user's efforts.

4.3 Improved justification

In the past the application of the scaling factor method based on correlations often was faced with the questions how many samples should be taken to assure representativity of the results and to minimise statistical fluctuation. This question was often treated in a empirical manner. The log-log plot of a correlation and its data points was assessed qualitatively and as a general rule it was concluded that the higher the correlation coefficient was the fewer samples are needed for representativity.

Kashiwagi et al [5] have proposed a statistical criterion to answer the above question. Without going too much into details the method was based on the following principle. From a given pool of data for a certain combination of radionuclides a specified number of samples are selected randomly. These data are correlated and the correlation coefficient and the lower confidence limit are determined. This procedure is repeated for a certain number of times and the results are averaged. If the number of samples for the evaluation is increased steadily and the same averaged values are calculated each time a dependency of the correlation coefficient from the number of samples should appear.

Figure 4 shows an example for the correlation of Am 241 to Co 60. Obviously, beyond about 30 samples there is no additional gain in statistical accuracy, which could justify to increase the number of required samples. The correlation coefficient of the correlation based on all available
data was 0.82. Other results would be achieved for correlations with different scatter of the measurement data. The lower confidence limit was chosen for a 95 % coverage of the selected data in accordance with the general practice in Germany for the evaluation correlations. The required number of samples can either be assessed in a qualitative manner as above. Another possibility is to use the slope of the lower confidence limit curve, for which a limit can be set.

![Graph showing the dependence of correlation coefficient on sample size](image)

**Figure 4:** dependence of correlation coefficient from sample size

### 5 Conclusion

In conclusion activity declaration in Germany has gained a high degree of experience. Tools for automated and computer based evaluations and application are available. Qualification and justification of the methods applied have reached a considerable level of sophistication. Further developments should follow this path to increase quality while achieving results with increasing effectiveness and accuracy.

### 6 LITERATURE


STATUS OF OPERATION OF RADIONUCLIDES ASSAY SYSTEM IN KOREAN NUCLEAR POWER PLANT

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ABSTRACT
In Korea, 17 nuclear power plants composed of 13 Pressurized Water Reactors and 4 CANDU Reactors are currently in operation. The cumulative amounts of low and intermediate level radioactive waste in nuclear power plant reached 58,718 drums (unit: 200 liter) in 2001. Efforts to construct LILW disposal facility are continued and its first operation is planned in the year 2008. Its first stage capacity is assumed to be 100,000 drums and total capacity will reach to 800,000 drums. Radwaste disposal site selection is an urgent national project at present time. Regulations and guidelines require detailed information about the radioactive waste package and its contents prior to the transport to the disposal sites. The Enforcement Decree of the Korean Atomic Energy Act (Articles 234-17) requires the Minister of Science and Technology (MOST) of Korea to establish regulation for the waste acceptance (MOST Notice.1196-10). It requires detailed information about the radioactive waste package and its contents such as activity of radionuclides, total activities, types and characteristics of waste. For the measurement of the concentrations and activities of radionuclides in radwaste drum, a radionuclides assay system is installed at Korean nuclear power plant (KORI site) in 1996. The waste drum can be measured in the vertical direction with eight vertical segments while in the radial direction also with eight segments. Using this measurement method, homogeneous and non-homogeneous waste drum can be measured. Scaling factor methods have been played a dominant role in the determination of the radionuclides concentration in this system. For corrosion product, generic scaling factors were used due to the similarity and better-characterized properties of Korean analyzed data as compared with the worldwide data base of PWR industry. For fission product and TRU nuclides, it is not easy to determine the generic scaling factors. Thus simple model reflecting the operation history of power plant and nuclear fuel condition is used. 17 gamma-emitting nuclides, 7 beta-emitting nuclides and total alpha (summation of alpha-emitting nuclides) are analyzed in the radionuclides assay system. The more R & D efforts for the declaration of radionuclide inventory in radwaste packages are required and planned on in Korea. The KINS (Korean Institute of Nuclear Safety), Korean regulatory authority, is going to develop the waste acceptance criteria containing target nuclides to be declared, its declaration method, limits on radionuclide inventories and concentrations for the more detailed requirements. Continuous R & D plan to develop the more advanced radionuclides assay system are recommended. In Line with this, efforts to the improvement of the old radionuclide assay system for the updating of data and detection method are under discussion. It will enhance the declaration of activity inventories for individual waste packages.

1. INTRODUCTION

Status of nuclear power generation and generated radioactive waste. In Korean, 17 nuclear power plants are currently in operation. 3 more units are under construction, 6 units in preparation for construction and 2 units in planning [Table 1]. The cumulative amount of low and intermediate level radioactive waste in nuclear power plant reached 58,718 drums (unit: 200 liter drum) in 2001, and total available storage capacity on site is 99,990 drums. The capacity and cumulative amounts of each site is listed at Table 2. Radio Isotope generated from hospitals, research laboratories and industry, and so on,
has reached 4,217 drums (Unit: converted into 200 liter drum). Storage and management of Radio Isotope is done by NETEC (Nuclear Environment Technology Institute). Its available storage capacity is 8,800 drums (Saturation year; 2010). However in this paper, it is focused to radiwaste generated from nuclear power plants.

Efforts to construct LILW disposal facility are continued and its first operation is planned in the year of 2008. Its initial stage capacity is assumed to be 100,000 drums and total capacity can be reach to 800,000 drums. Operation period is planned about 50 years. The type of disposal facility may be Engineered-Vault or Rock-Cavern type. Radwaste disposal site selection is an urgent national project at present time. The responsibility for the selection of candidate sites rests with KHNP (Korean Hydro and Nuclear Power Co., Ltd.), and KHNP should discuss with Local Governments for the candidate sites.

Regulations and guidelines require detailed information about the radioactive waste package and its contents. An environmentally sound and safe management of low-level radioactive waste requires knowledge of the characteristics and the inventories of radionuclides in the radioactive waste package. Potential risks during the transportation and disposal of radioactive waste are based on the assessment of radiological risk associated with the various nuclides in radioactive waste package. Accordingly, regulations and guides developed in various countries also require detailed description of the radioactive waste package and its contents. In Korea, the Enforcement Decree of the Korean Atomic Energy Act (Articles 234-17) requires detailed information about the radioactive waste package and its contents.

Measurement of concentration and total activity of radionuclide contained in radwaste drum is very important for the accurate and efficient management of radioactive waste in NPP. To achieve this most important objective, non-destructive prediction methods have been improved to reduce the radiation exposure for workers and to increase the reliability of measurement during the last twenty years. The determination of compositions of radioactive waste package raises a number of problems. Sometimes, representative sampling is not possible. And even if it is possible, it poses a considerable radiation exposure to sampling personnel. Furthermore, many radionuclides, whose declaration is required for the shipping with the gamma emission, must be analyzed.

In this study, prediction method for scaling factors that can account for plant-operation history is suggested for the Korean nuclear power plant (KORI #2). The operational results are presented and the suggestions for the more improved next-generation radionuclide assay system in Korea are provided.

2. OPERATIONAL STATUS OF RADIONUCLIDE ASSAY SYSTEM

2.1 Background and History of Radionuclide Assay System

At the end of 1993, Korea Electric Power Research Institute (KEPRI) organized the overall project to design and install the radioactive waste assay system with partially cooperated with Korea Atomic Energy Research Institute (KAERI) and Korea Advanced Institute of Science and Technology (KAIST).

A number of consultations and discussions among these three research institutes and modifications of final project goals during thirty months lead to:

A) Collection of the data for non-destructive assay method
B) Collection and analysis of the operational history of NPP
C) Development of non-destructive assay technique for the measurement of the radioactivity emitted from various radwaste streams
D) Design of model drum to evaluate the assay system
E) Development of the integrated control program for radioactive waste assay system
F) Development of chemical analysis method of radioactive waste with the waste sampling technique
G) Development of scaling factor determination program
H) Establishment of the normal and abnormal operation guideline of radioactive waste assay system

The established waste characterization program measures the concentrations of all relevant nuclides
either directly or indirectly by relating Difficult-To-Measure (DTM) radionuclides (Target nuclide) to other Easy-To-Measure (ETM) radionuclides (Key nuclide). So, the application of scaling factors is followed. Previous studies [1, 2, 3 and 4] have suggested the statistical scaling factors. Statistical scaling factors are generated by the use of sampling data that are collected from the radiochemical analysis of waste samples from different NPPs in off-site laboratories. These statistical scaling factors give somewhat generic and passive tendency for the related Nuclear Power Plant (NPP).

Accordingly, generic scaling factors might show some disadvantages such as following:

(A) Generic scaling factors cannot account for the overall operation history of NPP in detail.
(B) Since sampling procedures must be carried out with substantial operational change or modification of NPP environment, generic scaling factors cannot be coped with the detailed regulations and have some difficulties with sampling procedures and radiation protection.

With careful considerations, Kori NPP was selected as the candidate site for utilization and installation of this project. Radionuclide assay system was installed and started operation during mid 1996.

2.2 Analysis of Waste Drum

Waste drums generated from nuclear power plant are divided into four categories. The type of waste drum and the position of generation are summarized in Table 3. It is designed that the waste drum assay system installed in the nuclear power plant can utilize the scaling factor method to predict the radionuclides concentrations for each waste drum. In the selection of assay target radionuclides, the following conditions are considered;

A) Costs for radiochemical analysis
B) Radiological and biological toxicity
C) Radionuclides listed in regulations and safety assessments.

With these considerations, the following radionuclides are selected; H-3, C-14, Ni-63, Fe-55, Co-60, Sr-90, Nb-94, Tc-99, Cs-137 and gross alpha [TRU].

2.3 Applied Method for Scaling Factor

Gamma-ray detection method and scaling factor method are used to declare the radionuclides. Gamma emitting radionuclides are detected by gamma-ray detection system. Selected beta & alpha emitting radionuclides are calculated by the corresponding scaling factor method.

A. SCALING FACTOR FOR CORROSION PRODUCTS

H-3 doesn’t show a good correlation with Co-60 and Cs-137 in worldwide industry data set, since H-3 is generated by fission in the fuel and neutron activation in the coolant. However, with Korean Analyzed data, it shows a fairly reasonable correlation with Cs-137 [Fig. 1]. Each scaling factor for corrosion product radionuclides such as C-14, Ni-63, Fe-55 and Nb-94 is analyzed based on a worldwide data base of PWR experience [1,2,3,4] and shows good correlation with the key nuclide (Co-60) (for example, Ni-63 [Fig. 2]). The analyzed values of Korean nuclear power plant show very close similarity to the worldwide industry data set. For the corrosion products, it confirmed that general industry wide data set and the analyzed values for Korean power plant predict fairly reasonable scaling factors for the Korean PWR operation practice.

As sampling for the various radionuclides are accumulated, updated scaling factors of corrosion product will be produced. From the analysis of current data, it is concluded that the scaling factor and characteristic of worldwide industry data base is very similar to that of Korean nuclear power plant. Thus for the five general corrosion products [H-3, C-14, Ni-63, Fe-55 and Nb-94], known general scaling factors are used to give concentrations of radionuclides assay system.
B. SCALING FACTOR FOR FISSION PRODUCTS AND TRU NUCLIDES

From the known worldwide database [3, 4], it is concluded that these data sets do not give reasonable generic relationships for the fission products (FPs) and TRU nuclides. The reason is that these two types of nuclides depend on operation history of power plant and nuclear fuel conditions. Representative fission products such as Sr-90 and Tc-99 are selected as nuclides of concern and gross alpha activity is measured for the TRU scaling factors respectively.

Due to the difficulty of the data base for the general scaling factors, a simple model [5] is suggested to represent plant specific scaling factors for these types of nuclides.

\[
SF_{FP}^{\text{KEY}} = \frac{R_{1-131}}{R_{\text{KEY}}} \times \bar{A} \times C_{ko} \times \left[ \frac{C_{FP}}{C_{1-131}} \right]_{\text{ORIGENPC-2}} \quad \text{Eq. (1)}
\]

\[
SF_{\text{TRU}}^{\text{KEY}} = \frac{R_{1-131}}{R_{\text{KEY}}} \times \bar{A} \times C_{ko} \times \left[ \frac{C_{\text{TRU}}}{C_{1-131}} \right]_{\text{ORIGENPC-2}} \quad \text{Eq. (2)}
\]

\(SF^{\text{KEY}}_{\text{FP/ TRU}}\) = Scaling factor for Fission Products and TRU nuclides (in reactor coolant).

\(R_i\) = release rate of isotope i from tramp fuel.

\(\bar{A}\) = fractional release of I-131 from tramp fuel.

\(C_{ko}\) = fractional contribution of I-131 released from tramp fuel by knockout mechanism.

\([C_i/C_{1-131}]_{\text{ORIGENPC-2}}\) = activity ratios of C compared to C_{131} in the fuel calculated by ORIGENPC-2 code.

\(\gamma_i\) = fission yields of isotope i.

The above model has been derived with the following assumptions:

1. For the release sources of fission product and TRU nuclides, these nuclides are assumed to be released from tramp uranium only.
2. Fission product nuclide is released dominantly by recoil release mechanism and however small amounts of the fission product also released by knockout release mechanism (Eq. 1). TRU nuclide is released only by knockout release mechanism (Eq. 2).
3. In the knockout release mechanism, release ratio of the DTM (FP or TRU) nuclide to Key nuclide is proportional to the corresponding activity ratio within the nuclear fuel.
4. Iodine nuclide is released both from tramp fuel and defective nuclear fuel cladding through all of the diffusion, knockout and recoil release mechanism.

In the knockout release mechanism, the ratio of the release rates of the corresponding radionuclides is directly proportional to corresponding activity ratio within the nuclear fuel, in particular for the Sr-90/I-131, Tc-99/I-131 and Pu-239/I-131. Once the release rate of I-131 from tramp fuel by knockout is determined, the release rates of the DTM radionuclides are determined from the inventory ratios of the radionuclides in fuel as by eq. (1) and eq. (2).

Concentrations of iodine that is sampled daily from reactor coolant are used to determine the fractional release of I-131 from tramp fuel (\(\bar{A}\)). Then the fractional contribution of I-131 released from tramp fuel by knockout is calculated separately based on a release model suggested by previous publications [5, 6] ORIGENPC-2 computer code is utilized to calculate activity ratio between the DTM (FP or TRU)nuclide and I-131 nuclide.

C. VALIDATION

To validate the calculated results, radiochemically analyzed values for KORI Unit 2 were compared to theoretically calculated values by eq. (1) and eq. (2). Three scaling factors such as Sr-90/Cs-137, Tc-
2.4 Operation Results and Discussion

The operational results of radionuclide assay system are summarized in Table 4. The activity data is analyzed and collected for the years of 1998 to 2000 including detailed drum numbers and corresponding positions of drum generation at the Kori site.

Non-gamma radionuclides in the activity data sheet which are selected at the time of installation are H-3, C-14, Ni-63, Nb-94, Fe-55, Sr-90, Tc-99, and TRU. The other data are followings:

A) The name of operator
B) Drum identification code
C) Surface dose rate
D) Drum weight
E) Detection date
F) Activities of gamma radionuclides (Co-58, Co-60, Cs-137, Mn-54, Ag-110m, Cs-134, Ce-141, Nb-95, Cr-51, Ce-144, Fe-59, Rb-86, Ru-103, Ba-140, Zn-65 and Sb-125).

Table 5 shows analyzed results for radionuclide activity of DAW for the case of without concrete shielding, with concrete shielding, concentrate bottom and spent filters generated from Unit 1 at the Kori site. Here, due to the relatively small number of analyzed data, spent filter is averaged over for the all Kori NPPs.

The activity data are statistically analyzed and showed that

A) Activity of DAW with concrete shielding is 2-3 order of magnitude higher than that of DAW without shielding.
B) In generally, the activities of Fe-55, Ni-63, H-3 and C-14 show higher values than that of Sr-90, TRU, Tc-99 and Nb-94.
D) Activity of Sr-90 shows highest values in concentrate bottoms.
E) For spent filter, the trend of strength of radionuclide activity shows slightly different tendency than that of other waste type.

3. CONCLUSION

An indirect method to determine concentrations of important radionuclides has been set up to support radionuclides assay system installed within Korean nuclear power plant. For scaling factor of corrosion products, analyzed sample values show very similar trend to the worldwide industry data set. Thus the corresponding generic scaling factors can give reasonable estimates to determine the concentrations of corrosion products. In the case of fission products and TRU nuclides, a newly derived simple model is applied and validated with the analyzed sample value. Thus the plant-specific and operation history dependent scaling factor have been generated to give reasonable results for the estimates of corresponding radionuclides concentration.

An additional and frequent sampling procedure must be implemented to update the performance of Korean nuclear waste management. KHNP (Korean Hydro & Nuclear Power Co. Ltd.) plans to carry out the more research efforts to construct radionuclide assay systems for PHWR (CANDU-type reactor in Wolsong site) and establish the analyzed method for the cumulated radwaste drums in all other sites. Next-generation assay systems for the Korea NPPs are expected to be improved and upgraded with the experiences of design, manufacturing, installation, and operation of the radionuclide assay system at KORI site.

ACKNOWLEDGEMENT
This work was performed by the financial support from the Korea Institute of Nuclear Safety (KINS) and “Mid-and-Long-term Nuclear R&D Program”.

4. REFERENCES


Table 1. The present and future of nuclear power plants

<table>
<thead>
<tr>
<th>Classification</th>
<th>Name of Plant</th>
<th>Unit</th>
<th>Capacity (MW (%))</th>
</tr>
</thead>
<tbody>
<tr>
<td>In operation</td>
<td>Kori (#1,2,3,4)</td>
<td>17</td>
<td>14,716 (53.87)</td>
</tr>
<tr>
<td></td>
<td>Yonggwang (#1,2,3,4,5)</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Ulchin (#1,2,3,4)</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Wolsong (#1,2,3,4)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Under construction</td>
<td>Yonggwang (#6)</td>
<td>3</td>
<td>3,000 (10.98)</td>
</tr>
<tr>
<td></td>
<td>Ulchin (#5,6)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>In prep. For construction</td>
<td>Shin-Kori (#1,2,3,4)</td>
<td>6</td>
<td>6,800 (24.89)</td>
</tr>
<tr>
<td></td>
<td>Shin-Wolsong (#1,2)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>In planning</td>
<td>Undecided</td>
<td>2</td>
<td>2,800 (10.26)</td>
</tr>
<tr>
<td>Total</td>
<td></td>
<td>28</td>
<td>27,316 (100%)</td>
</tr>
<tr>
<td></td>
<td>By 2015 (33%)</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 2. The capacity and cumulative amounts of radioactive waste (Dec, 2001)

<table>
<thead>
<tr>
<th>Site</th>
<th>Storage capacity</th>
<th>Cumulative amount</th>
<th>Saturation year</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kori</td>
<td>50,200</td>
<td>31,744</td>
<td>2014</td>
</tr>
<tr>
<td>Yonggwang</td>
<td>23,300</td>
<td>10,382</td>
<td>2011</td>
</tr>
<tr>
<td>Ulchin</td>
<td>17,400</td>
<td>11,342</td>
<td>2008</td>
</tr>
<tr>
<td>Wolsong</td>
<td>9,000</td>
<td>5,250</td>
<td>2009</td>
</tr>
<tr>
<td>Total</td>
<td>99,900</td>
<td>58,718</td>
<td></td>
</tr>
</tbody>
</table>

Table 3. Type of waste drum and the location of generation

<table>
<thead>
<tr>
<th></th>
<th>Aux. Building (KORI Unit 3)</th>
<th>Aux. Building (KORI Unit 4)</th>
<th>Radwaste Building</th>
</tr>
</thead>
<tbody>
<tr>
<td>Spent Filter Drum</td>
<td>√</td>
<td>√</td>
<td></td>
</tr>
<tr>
<td>Spent Resin Drum</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Evaporator Bottom Drum</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dry Active Waste Drum</td>
<td></td>
<td>√</td>
<td></td>
</tr>
</tbody>
</table>
Table 4. Information of the analyzed radioactive waste drum, radionuclide and applied scaling factor.

<table>
<thead>
<tr>
<th>Analyzed waste drum</th>
<th>Dry Active Waste (2428 drums)</th>
<th></th>
<th>1998–2000</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>a. without concrete shielding: 2,227 drums</td>
<td>b. with concrete shielding: 201 drums</td>
<td>(total: 2,602 drums are analyzed)</td>
</tr>
<tr>
<td></td>
<td>Concentrate Bottom (142 drums)</td>
<td>Spent filter (16 drums)</td>
<td>HIC: High Integrity Container</td>
</tr>
<tr>
<td></td>
<td>Spent resin (HIC: 16)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Radionuclide</td>
<td>Gamma emitting nuclides (17)</td>
<td>Beta emitting nuclides (7)</td>
<td>Alpha emitting nuclides: TRU (total alpha)</td>
</tr>
<tr>
<td></td>
<td>(Co-58, Co-60, Cs-137, Cs-134, Ag-110m, Mn-54, Ce-141, Ce-144, Cr-51, Zr-95, Nb-95, Fe-59, Rb-86, Ru-103, Ba-140, Zn-65, Sr-125)</td>
<td>(H-3, C-14, Ni-63, Nb-94, Fe-55, Sr-90, Tc-99)</td>
<td></td>
</tr>
<tr>
<td>Scaling factor</td>
<td>Key nuclides: Co-60, Cs-137</td>
<td>Target nuclides: H-3, C-14, Ni-63, Nb-94, Fe-55, Sr-90, Tc-99, TRU</td>
<td></td>
</tr>
</tbody>
</table>

Table 5. Statistical results of radionuclide activity of waste type

<table>
<thead>
<tr>
<th>Unit 1</th>
<th>Unit Bq/drum</th>
<th>DAW without concrete shielding</th>
<th>DAW with concrete shielding</th>
<th>Concentrate bottom</th>
<th>Spent filter</th>
</tr>
</thead>
<tbody>
<tr>
<td>H-3</td>
<td>Mean</td>
<td>7.74E+06</td>
<td>5.71E+09</td>
<td>3.39E+09</td>
<td>9.66E+08</td>
</tr>
<tr>
<td></td>
<td>Max</td>
<td>6.40E+08</td>
<td>2.20E+10</td>
<td>3.80E+10</td>
<td>2.40E+09</td>
</tr>
<tr>
<td>C-14</td>
<td>Mean</td>
<td>6.51E+06</td>
<td>4.80E+09</td>
<td>1.84E+08</td>
<td>3.10E+09</td>
</tr>
<tr>
<td></td>
<td>Max</td>
<td>5.10E+08</td>
<td>1.80E+10</td>
<td>2.20E+09</td>
<td>1.20E+10</td>
</tr>
<tr>
<td>Ni-63</td>
<td>Mean</td>
<td>1.55E+07</td>
<td>1.16E+10</td>
<td>3.19E+09</td>
<td>7.99E+09</td>
</tr>
<tr>
<td></td>
<td>Max</td>
<td>1.40E+09</td>
<td>4.90E+10</td>
<td>3.70E+10</td>
<td>2.80E+10</td>
</tr>
<tr>
<td>Nb-94</td>
<td>Mean</td>
<td>3.76E+03</td>
<td>2.75E+06</td>
<td>4.04E+05</td>
<td>1.55E+06</td>
</tr>
<tr>
<td></td>
<td>Max</td>
<td>2.80E+05</td>
<td>9.80E+06</td>
<td>4.70E+06</td>
<td>4.80E+06</td>
</tr>
<tr>
<td>Fe-55</td>
<td>Mean</td>
<td>1.71E+07</td>
<td>1.25E+10</td>
<td>3.65E+08</td>
<td>7.05E+09</td>
</tr>
<tr>
<td></td>
<td>Max</td>
<td>1.30E+09</td>
<td>4.40E+10</td>
<td>2.00E+09</td>
<td>2.20E+10</td>
</tr>
<tr>
<td>Sr-90</td>
<td>Mean</td>
<td>7.28E+05</td>
<td>1.28E+08</td>
<td>5.30E+08</td>
<td>5.18E+06</td>
</tr>
<tr>
<td></td>
<td>Max</td>
<td>1.10E+08</td>
<td>6.70E+08</td>
<td>1.60E+10</td>
<td>4.80E+07</td>
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<tr>
<td>Tc-99</td>
<td>Mean</td>
<td>6.46E+04</td>
<td>1.25E+07</td>
<td>3.67E+06</td>
<td>4.85E+05</td>
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<tr>
<td></td>
<td>Max</td>
<td>9.90E+06</td>
<td>5.90E+07</td>
<td>1.10E+08</td>
<td>4.00E+06</td>
</tr>
<tr>
<td>TRU</td>
<td>Mean</td>
<td>3.14E+05</td>
<td>5.03E+07</td>
<td>4.67E+07</td>
<td>1.11E+06</td>
</tr>
<tr>
<td></td>
<td>Max</td>
<td>4.90E+07</td>
<td>2.90E+08</td>
<td>1.40E+09</td>
<td>9.20E+06</td>
</tr>
</tbody>
</table>
Fig 1. Concentration relation of Corrosion Product [H-3/Cs-137]

Fig 2. Concentration relation of Corrosion Product [Ni-63/Co-60]

Fig 3. Comparison result with Measured and Calculated Scaling Factor
ASESSMENT OF LILW PACKAGE ACTIVITY IN SPAIN

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Abstract
The different methodologies used in Spain for calculation of LILW package activity depend on the nature of the package (solid wastes or solidified homogeneous wastes) and on the isotopes to be measured. $^{60}$Co and $^{137}$Cs are the easily measured isotopes to be recorded by the producer on each package, the other isotopes of significance for the El Cabril Disposal Facility being evaluated by ENRESA by means of different methodologies. The methodologies currently used are gamma spectrometry, radiochemical techniques, theoretical methods for calculation of the activity of gamma-emitting isotopes, and finally the application of scaling factors for the calculation of difficult to measure emitters.

1.- Relevant aspects requested by the regulatory authority

All LILW waste packages to be disposed of at the El Cabril Disposal Facility are required to have an associated acceptance process, which will be approved depending on a series of requirements applied to such packages [1].

The acceptance process for packages of a given type generated by a producer consists basically of drawing up of the acceptance documentation [2] (Process Book and Acceptance Dossier), supported by information provided by the producer and by different laboratory tests aimed at demonstrating suitability for definitive disposal at the El Cabril D.F..

The requirements applied to the packages refer fundamentally to the following physical and chemical characteristics:
- Identification and quantification of the radionuclides contained in the package.
- Resistance to radionuclide release to the external environment.
- Mechanical requirements and those associated with durability.

The main objective is the identification and quantification of isotopes of significance from the point of view of the El Cabril disposal facility. All LILW packages are required to have an activity value identified for each.

Given the different activity determination methods in existence, these have to be evaluated, approved and included in the associated acceptance documentation.

The first step for the acceptance of an LILW type package is submittal by the producer of the Package Description Document (PDD), which indicates the production process used for the packages whose acceptability is to be evaluated. This document must include the activity determination methodology used for $^{60}$Co and $^{137}$Cs as well as for other gamma emitters having significant activity in the packages.

This methodology will be studied by ENRESA, checking the acceptability of the method in accordance with the corresponding activity calculation specifications [3].
2. Appropriate LILW package activity evaluation methods

The different types of LILW package activity evaluation methods are described below, depending on the isotope to be determined and the nature of the wastes:

Used fundamentally by the producer for the measurement of both $^{60}$Co and $^{137}$Cs and other gamma emitters with significant activity. This technique is applied fundamentally to the following:

- Samples of non-conditioned liquid wastes: the activity of the final package is determined by the number of litres of waste included in the package. A final correction for the Contact Dose Rate of each package is carried out for all those making up the batch, with samples considered to be representative, due to possible lack of homogeneity in the production process.
Evidently, this technique is applied exclusively to packages of homogeneous moist wastes incorporated in hydraulic conglomeratant.

- Whole package: there are currently various measurement techniques of this type. If the efficiency calibration is accomplished on the basis of a standard, the essential difficulty is to be found in manufacturing the latter. If, on the other hand, this calibration is performed using statistical simulation methods (Montecarlo), waste homogeneity is hypothetically assumed.
This technique is applied fundamentally to homogeneous waste packages, although homogeneity is assumed hypothetically if it is applied to other waste types.

2.2. Theoretical methods (TM).
Used by both the producer and by ENRESA for the evaluation of activity in both $^{60}$Co and $^{137}$Cs and other gamma emitters with significant activity. The following generic expression is applied for this technique:

$$A = \frac{D}{\sum_{i=1}^{n} d_i \cdot f_i}$$

where:
- **A**: Activity contained in the package (MBq/Package).
- **D**: Contact dose rate representative of the package (µGy/h).
- **n**: Number of isotopes considered.
- **f_i**: Isotopic fraction, in unit terms of isotope i.
- **d_i**: Activity Pass Factor – contact Dose Rate (µGy/h per MBq/Package).

For one same type of package, the Pass Factor of an isotope will change only with its density. For this reason if the isotopy and corresponding Pass Factors are available for each isotope and density, the use of shielding calculation codes [4] will make it possible to assess the activity of the package from the contact dose rate.

The determination of the contact dose rate representative of the package is accomplished in accordance with the following method:

- The contact dose rate is measured at the generatrix of the drum closure bolt (0°), noting down the maximum value obtained.
- This same operation is repeated at the other three generatrices, located at 90°, 180° and 270° with respect to the former.
- The arithmetic mean of the four maximum values obtained is taken, this being the representative dose rate of the package.
Once the total activity of γ emitters has been obtained, the activity of each \((A_i)\) must be calculated, for which:

\[
A_i = f_i A
\]

This technique is applied mainly to the following:

- Heterogeneous compactable waste packages.
- Heterogeneous non-compactable waste packages.
- LILW packages with concrete walls, containing fundamentally cartridge filters or desiccated sludges.

Whole package gamma spectrometry is difficult to apply to these wastes, as has been indicated above.

Figure 1 shows the Activity Pass Factors – contact Dose Rate of \(^{60}\text{Co}\) for filter type packages with concrete wall thicknesses of 5, 10, 15 and 22 cm, in addition to the Pass Factors for homogeneous packages (0 cm concrete wall).

2.3. **Destructive assessment (DA). Radiochemistry.**

Used by ENRESA mainly to measure pure Beta and Alpha emitters. This technique is applied fundamentally to the following:

- Samples of non-conditioned wastes (Ciemat laboratory and currently the ENRESA Waste Quality Verification Laboratory), used to create a database for application of the Scaling Factors methodology.
- Samples of test piece trepanation dust from packages of conditioned wastes (Waste Quality Verification Laboratory and Ciemat), used to check the levels of activity of these isotopes, present in packages received at El Cabril and to extend the aforementioned database.

2.4. **Scaling Factors (SF).**

By using the data compiled from radiochemical samples, correlations have been established between difficult to measure and easily measured isotopes, the aim being to establish a Scaling Factor to be applied, initially, from the measurement of \(^{60}\text{Co}\) or \(^{137}\text{Cs}\) in an LILW package. This technique is applied to practically all LILW packages.
3.- Isotopes of significance at the El Cabrif disposal facility

Fundamentally, two groups of isotopes are established for determination [5]:

- Evaluated by the producer, easily measured and known as key isotopes: $^{60}$Co, $^{137}$Cs.
- Difficult to measure and evaluated by ENRESA:
  - $^3$H, $^{14}$C, $^{59}$Ni, $^{63}$Ni, $^{94}$Sr.
  - $^{99}$Tc, $^{95}$Sr, $^{129}$I.
  - $^{238}$Pu, $^{239}$Pu, $^{241}$Pu, $^{241}$Am, $^{244}$Cm.

Various lines of research are open in relation to the possibility of adding several isotopes more, depending on their radiotoxicity, half-life and amount present in the waste, arisen from Safety Assessment revisions.

4.- Types of LILW packages

The following nuclear power plants are in operation in Spain:

- Almaraz NPP, 2 units (PWR).
- Ascó NPP, 2 units (PWR).
- Cofrentes NPP (BWR).
- Garoña NPP (BWR).
- José Cabrera NPP (PWR).
- Trillo NPP (PWR).
- Vandellós 2 NPP (PWR).

The operation of these plants produces the following types of LILW waste packages, divided into two major groups:

- Homogenous solidified wastes.
  - Grain resins.
  - Powdered resins (Cofrentes NPP).
  - Evaporator concentrates with boric acid (PWR’s).
  - Evaporator concentrates without boric acid (BWR’s).
  - Moist sludges.
  - Desiccated sludges.

- Heterogeneous solids, immobilised or otherwise:
  - Cartridge filters (PWR’s).
  - Sludge filters (BWR’s).
  - Compactable heterogeneous solids.
  - Non-compactable heterogeneous solids.

All of these wastes are incorporated in drums, mainly with a capacity of 220 litres, although other sizes may be used (290 l, 400 l and 480 l).

The Vandellós 1 nuclear power plant is currently being dismantled and in addition to the wastes indicated above, others specific to this operation have been generated. In view of the large quantities of wastes generated, the decision has been taken to use other types of packaging of a larger size and different geometry.

Each package type has an associated activity calculation methodology that depends on the type of waste, the isotopes present and the geometry of the source considered, as indicated in point 2.
5.- Scaling Factors

As has been indicated above, radiochemical techniques are required for assessment of the activity of isotopes having difficult to measure half-lives:

- Measurement of homogeneous non-conditioned wastes at external laboratories: CIEMAT and currently the El Cabril WQVL.
- Measurement of homogeneous conditioned wastes at external laboratories: El Cabril and CIEMAT.

In Spain, Scaling Factors have been calculated for difficult to measure isotopes for each waste stream and nuclear power plant, using the methodology currently approved by the regulatory authority [6]. This methodology is based on the Normal distribution apparently followed by the logarithms of isotope concentrations in the waste streams [7]. If the difficult to measure isotope correlates with the easily measured one, there is a linear relationship between them and a Scaling Factor (SF) may be established, as shown in Figure 2.

If, on the other hand, no relationship may be established between the difficult to measure and easily measured isotopes, the activity value proposed for the difficult to measure isotope in the package is the mean activity concentration (MAC), as shown in Figure 3.

Statistical tests are used to verify the existence or otherwise of a relationship between the correlated isotopes, with a view to using a Scaling Factor or the Mean Activity Concentration.
The factor used (SF or MCA) corresponds to the geometric mean of the data used in its calculation, this being a good approximation to the median as well as a consistent estimator, minimising the variation of individual data.

If a sufficient number of statistically established data is not available for a waste stream (at least 15), the data will have to be linked to others for another stream from the same plant, stream linking statistical tests being used for this purpose. If the test fails, data for streams for other nuclear power plants and having similar characteristics should be used, the linking tests also being applied. The final objective is to remove the linked data as data for the stream under study become available.

Scaling Factors have also been established for the nuclear power plant currently in the dismantling phase, Vandellós 1 [8]. Given its application to potentially declassifiable materials, an associated uncertainties calculation methodology has had to be developed by ENRESA, for the establishment of an upper limit of 95% for the range of confidence corresponding to the activity value of the isotopes present in the material, and for comparison with the Levels of Declassification established by the Regulatory Authority.

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Swedish Waste Disposal Concept for Operational Waste

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Abstract:

The Swedish waste disposal system for low and intermediate level waste consists of an underground final repository SFR 1 and shallow land burials located close to the nuclear power stations.

The first waste with ion exchange resin conditioned in cement was produced in the early 1970’s. At that time the final disposal was not yet defined and consequently the packages were very robust to fit “any” disposal system. These packages were also relatively uneconomical and modification has been performed ever since the repository site was established.

Since 1988 operational low and intermediate level waste had been disposed of in the central underground repository (SFR). The waste originates from the nuclear power plants and Studsvik research facility.

A new updated safety analysis for SFR 1 was finalised in June 2001. Development of waste acceptance criteria is in practice an iterative process concerning existing waste, repository development, safety assessment, new waste types (processes) and volume of waste.

Since a couple of years Ringhals nuclear power plant has been studying the possibility to mix bead and powder on exchange resins (IXR) in the same waste matrix. This question has been raised due to that fact that there sometimes are more or less of the different types of IXR available at the plant for processing. With this new strategy there is a possibility to optimise the waste load at any time in the waste packages.

Another issue has also been to minimise chemical additives to the waste matrix such as plasticers and other compounds. There is always a goal to avoid or to minimise organic complexing compounds in waste dedicated to SFR. This concerns the fact that organic chelates could reduce the sorption capacity in the final repository. Results from studies of those new recipes at the Älvkarleby concrete laboratory and full scale tests at Ringhals NPP in Sweden have shown that mixing bead and powder in different proportions can give a product acceptable for final disposal in the SFR facility. Some changes had to be done in the stirrer function for the “in-drum” mixing when processing the waste in full scale, compared to lab scale test The waste packages have after this stirrer modification been giving a suitable final product that meets the waste acceptance criteria.

When developing new recipes for Ringhals NPP the main criteria to be fulfilled were:

- The compressive strength for the cement matrix should be $\geq$ 10 MPa to meet the acceptance criteria in SFR.

- Reaching a homogeneous product without the use of organic additives
1. **The Swedish system for disposal of LILW**

Short-lived waste from operations and maintenance from nuclear power is after 40 years approximately 63000 m³. Long-lived waste is estimated to about 20000 m³ from the Swedish program including decommissioning waste, and some waste from medicine, research and other industry. Decommissioning of the Swedish reactors including central interim storage, encapsulation plant and Studsvik research centre is estimated to generate 170000 m³ radioactive waste. Classification of radioactive waste in Sweden coincides closely with that system developed by IAEA [1]. The responsibility for the management of the radioactive waste from nuclear power production, lies with the operators of the nuclear power plants. The utilities have jointly formed SKB, the Swedish Nuclear Fuel and Waste Management Company, to safely manage the radioactive waste from the reactors to final disposal. The task of SKB is thus to plan, construct, own and operate the systems and facilities necessary for transportation, central interim storage and final disposal. SKB has developed a system that ensures the safe handling of all kinds of radioactive waste from the Swedish nuclear power plants for a long time period ahead. The keystones of this system when handling the low and intermediate level waste are (figure 1):

- Nuclear power plants, research, industry, medical care
- Shallow land burials adjacent to nuclear power plants
- Transportation M/S Sigyn
- Future repositories for low and intermediate level waste, SFR3 short-lived decommissioning waste, SFL3-5 long-lived waste
- SFR1 operational low and intermediate level waste
- CLAB - interim storage of spent fuel and some core components

![Diagram of the Swedish waste management system for low and intermediate level waste](image)

Figure 1. The Swedish waste management system for low and intermediate level waste

- A transport system which has been in operation since 1983.
- A central interim storage facility for spent nuclear fuel and core components, CLAB, in operation since 1985.
- A final repository for short-lived, low and intermediate level waste, SFR, in operation since 1988.

At the sites adjacent to the nuclear power plants there are established shallow land burials for disposing off very low level waste. The remaining components of the system that are now being planned are:

- A repository for short-lived waste from decommissioning
- A repository for long-lived low and intermediate level waste

The costs for the management of all radioactive residues from the nuclear power plants, as well as for the decommissioning of the plants is borne by the operators of the reactors. To cover all future costs and ensure that means will be available a fee is levied on nuclear electricity and is paid to the Nuclear Waste
Fund. Costs for operational waste generated during operation of the nuclear power plants is paid directly by the nuclear power plants.

2. Guidelines for waste acceptance criteria in Sweden for SFR 1

Back in 1987 a first version of guidelines from the authorities came out in the area of waste acceptance criteria for radioactive waste for disposal in SFR 1. In 1991 a new version was updated, in a short while a new updated version is coming.

This document from the Swedish authorities [2] is in practice directly addressed to SKB on the first hand and to the nuclear power operators on the second hand. It regulates what kind of information that is needed to specify for different kind of waste types.

All types of waste should have an approved waste type description (WTD) including all information of what kind of waste acceptance criteria that has to be fulfilled.

This description is subdivided in five different stages;

1) Conditioning of the waste
2) Intermediate storage of the conditioned waste package
3) Transportation to SFR
4) Handling and operation in SFR
5) Final storage after disposal in SFR

In all these stages every single waste acceptance criteria shall be evaluated if it is of importance.

The waste acceptance criteria consists of a so called functional requirement. Coupled to this functional requirement it is possible to evaluate a so called quality criteria on the individual waste type.

2.1 Qualitative waste acceptance criteria for waste type to be disposed of in SFR 1 (authority guidelines)

General

Design, geometry and dimensions: The design, geometry and dimensions of a waste package shall be in compliance with the systems for handling and transportation and with the appropriate disposal part of the repository.

Weight: The weight of a waste package must not exceed the limits set by the handling and transport systems. The distribution of mass within a waste package must not jeopardize the stability during handling operations such as lifting and stacking.

Marking: Each waste package, which might consist of an assembly of primary packages in a common enclosure during transport and disposal, shall bear a unique identification marking. The marking shall be documented in the waste producer's register and in the repository register. It shall also enable the package to be located in the repository.

Radiological properties

Radionuclide inventory: The contents of gamma emitting radionuclides shall be known in terms of species and quantities for each waste package. An account of the contents of alpha and beta emitting radionuclides shall be given. The accuracy of this inventory shall be sufficient to assure compliance with given limits for different kinds of packages and different emplacement cavities in the repository.
**Surface dose rate and dose rate at a certain distance:** Maximum external dose rate, measured and reported as surface dose rate and/or dose rate at a certain distance (normally 1 m) from the outer surface of the package, shall be lower than the limits applicable for the facilities where the packages are stored and the equipment used for their handling. Limits for transportation shall be taken into account.

**Surface contamination:** Transferable contamination by radionuclides, i.e. contamination that might be released from the outer surface of the waste packages during normal handling or pouring of water for a short time, shall be kept within authorized limits.

**Internal radiation:** Internal dose rates as well as the internal integrated radiation dose, must not be as great that processes induced by radiation, e.g. radiolysis, affect the properties of the waste form, packaging and the barrier functions in the repository to an unacceptable extent.

**Homogeneity:** Solidified liquid and wet waste shall be homogeneously distributed to an extent that radionuclides never occur in such concentrations that the above mentioned radiological properties will be affected to an unacceptable extent. During package and grouting of solid waste, components etc., the active material shall be emplaced in the packaging in such a way that the activity becomes distributed throughout the packaging as homogeneously as possible.

**Chemical properties**

**Composition and structure:** The chemical composition and structure of the waste form and its packaging shall be known and defined to such an extent that allows an assessment of the material properties of the waste package.

**Homogeneity:** Solidified liquid and wet waste shall be homogeneous distributed to such an extent that the mechanical and physico-chemical properties assigned to the waste form from the aspects of safety and radiation protection are not jeopardized and can be assessed with sufficient accuracy.

**Liquids:** Waste packages are not allowed to contain free liquid that, due to leakage, might lead to unacceptable radiological consequences.

**Corrosion resistance:** Waste packaging shall have a durability against external and internal corrosive attacks that is sufficient with regard to conditions before backfilling around the package or sealing of the repository cavity.

**Gas formation:** Gas formation rate and volume, caused by the composition and structure of the waste form or package, shall not jeopardize the safety before closure of the repository or, after that, give rise to unacceptable disturbance of the barrier functions of the repository. Different mechanisms and processes for gas formation shall be regarded, e.g. radiolysis, biological decomposition, metal corrosion and other possible transformation processes. The content of organic substances and other biodegradable materials and metals whose corrosion might give rise to gas evolution shall be specified with sufficient accuracy to ascertain assessment of the consequences with respect to radiation protection. Wastes in the form of compressed gases are not to be disposed of in the repository.

**Combustibility and fire-resistance:** Combustible waste and waste forms shall be packaged in such a manner and have such characteristics that the risk of self ignition is negligible. Combustible waste shall be sufficiently well characterized and specified in terms of quantities and composition to permit the necessary precautions to be taken. Any fire shall be prevented from spreading through appropriate measures. Explosive materials are not allowed in the waste.

**Chemical reactivity:** Waste packages shall not contain substances that, due to their nature and quantities, might jeopardize the stability of the waste packages or the barrier functions of the repository to an unacceptable extent. Complexing agents shall be avoided as far as possible.
Mechanical properties

**Mechanical strength against external stresses:** The mechanical strength of the waste packages against external stresses such as pressure, strain, bending and impact, shall be sufficient to preclude unacceptable releases of radionuclides during foreseeable incidents and accidents. The waste form shall have a structure and homogeneity that is in compliance with this requirement.

**Mechanical stability:** The structure and volume of the waste shall be such that they do not deteriorate to an extent, leading to unforeseen release. Example of such processes are swelling of the waste form under pressure build-up and degradation of mechanical strength caused by changes in temperature.

### 3. SFR facility

SFR, The Swedish Final Repository for Radioactive Waste, is a central disposal facility for short-lived low and intermediate level radioactive waste from the operation of the nuclear power plants. The storage capacity of SFR is at present 63000 m³. Today, SFR is receiving operational waste from nuclear power plants such as ion exchange resins conditioned in cement or bitumen matrix, scrap metals and rubble. Also similar waste from the use of radioisotopes in medicine, research and industry is disposed of.

#### 3.1 Repository design

SFR is built in bedrock at about 50 meters depth. The repository has been designed so that the waste will be isolated to prevent any escape of toxic components into the environment in harmful quantities even after the facility has been sealed and abandoned. No institutional control is foreseen after sealing. Isolation is accomplished by different barriers surrounding the waste. In SFR, the waste is disposed of in different rock caverns that have been adopted to the different waste types and their different demands on handling and barriers [3]. The most active waste is disposed of in a concrete silo built in a cylindrical rock cavern. The silo is about 50 m high and has a diameter of 25 m. The thickness of the concrete wall is about 0.8 m. Bentonite clay is placed between the silo wall and the rock. The design of the other four caverns is different and depends on the type and dose-rate of the waste packages. The caverns are about 160 m long and 14-18 m wide. In one cavern, for intermediate level waste, remote handling is used as in the silo, while in the other caverns a forklift truck is used for handling. The handling method depends on the dose rate and the origin of the waste packages.

#### 3.2 Long-term safety

The long-term safety of the facility is ensured by the combination of good waste packages, engineered barriers and the isolation provided by the rock [4]. The only conceivable escape mechanisms for the radionuclides from the repository are by moving groundwater or by a well intruding into the repository. Most of the radioactivity, up to about 90%, will be deposited in the silo, which has been equipped with the most extensive barrier system. These are the immobilized waste, waste package, chemical sorption on the material in the waste and in the silo, concrete wall of the silo, bentonite clay and the rock mass. In the other caverns the isolation is provided by the waste itself, the rock mass and in some cases by concrete structures around the waste. Due to the land uplift in Sweden; about 6 mm/year in the area, the seabottom above SFR will become dry land in 1000 years time, and the hydraulic regime in the rock will change. At that time, however, only small amount of long-lived radionuclides like nickel-59, chlorine-36 and plutonium will remain. The total amount of plutonium in the repository is expected to be less than 0.5 kg distributed in a large volume. Dose calculations to the biosphere are calculated up to 10000 years ahead.
3.3 Safety analyses of SFR

At least every tenth year a new safety performance assessment is performed according to operational permits for the facility. The latest safety performance assessment was finalised in June 2001 [5]. There have been significant improvements in the analyses as compared with the earlier analyses – from 1993. In general, there are better tools to perform detailed model calculations, not only for various processes, but also for the emission calculations. If these more detailed calculations are to be meaningful, they require input data with a considerably higher degree of detail than before. Examples of the significant improvements that have been made include:

- The scenario and system analysis has been improved. More experience has been acquired, and new working models developed, since 1993. The analysis started with a review of what SFR 1 and its surroundings look like, and of the various factors that affect its development. This has been done without preconceptions, and has been checked against international experience/databases.
- The waste inventory is now better known. Earlier analyses were based on estimates that were originally made in the early 1980s. The new safety report now includes ten years' of operating experience from SFR 1. Most of the radionuclide and material estimates are based on measured values.
- The near field analyses have been improved, primarily in respect of concrete degradation, gas evolution and gas transport, but also in connection with such aspects as the creation of complexes and the properties of bitumen.
- Hydrogeological modelling has been carried out with a considerably greater degree of detail, with greater numerical precision and with a realistic description of transient processes.
- The biosphere has been modelled with a new and considerably more comprehensive description. There have also been significant improvements in the description of expected biosphere development. The calculations of radionuclide emissions have been carried out in a more consistent manner. The earlier approach, for example, involving two periods (the 'inland' and 'salt water' periods) has been abandoned, giving way to a process that better allows for the fact that the system would change gradually. Calculation of radionuclide transport in the repository is also based on the detailed hydrogeological analysis, which means that the previously used simplified approaches have been abolished.

3.4 Operation of SFR

Active operation in SFR started in April 1988. So far (mid 2002) about 29000 m³ of waste have been disposed of. The facility is operated by a staff about 12 man-years/year, working day shift only [6]. Much of the handling in SFR is done by remote control from a central control room. Only during transport of the containers, handling of low-level waste and some closure operations are the operators exposed to radiation from the waste. The doses to the personnel have thus been very low, in the order of a few mrem/years.

4. Improvements of WAC for SFR 1

When operation started of SFR 1 back in 1988 it was based on the types of waste and waste treatment processes that was invented back to the beginning of 1980-ies and even earlier. Design of SFR was fitted to the existing and planned waste types. Improvements of WAC have after the start of operation of SFR, been performed stepwise according to the safety analyses 1987, 1993 and the latest one 2001.

When developments are suggested they must always be confirmed to be feasible with respect to the above mentioned general waste acceptance criteria and the requirements derived from the safety analysis and the safety performance assessment.

Generally a lot of suggestions are coming from the waste producers to optimise the waste volume in different waste packages. Also suggestions are coming for optimising the content of radionuclides in each waste package and development of new waste packages. Some examples of improvements that have been done for waste to be disposed of in the SFR 1 facility are:
- Development of new types of waste packagings, suitable for odd waste.
- Cracked cubical concrete moulds have been placed in steel overpacks in order to avoid reconditioning.
- Possibility to deposit large components such as a reactor lid from a PWR reactor from Ringhals as a whole component in rock vault BTF.
- Optimising of waste load in each waste package with conditioning with cement, up to 1200 kg (corresponding to approximately 250-300 kg dry weight) of ion exchange resin per steel mould instead of approximately 500 kg in a concrete mould. Consequently, the external radiation has been raised.
- Optimising waste load in bitumen matrix
- Development of new transport casks that could take waste packages with an external radiation up to 500 mSv/h.
- Reevaluating the mechanical strength for waste packages. As an example, the requirement for the mechanical strength for the 42 waste packages stacked in the SILO used to be 40 MPa, but it has now been reevaluated that 10 MPa is enough.

5. Example from development of new recipes for conditioning of spent ion exchange resins in cement matrix.

The first waste conditioned in cement was produced in the early 1970’s. At that time the final disposal was not yet defined and consequently the packages were very robust to fit “any” disposal system. These packages were also relatively uneconomical and modification has been performed ever since the repository site was established. When processing the waste there has been established a praxis in Sweden to use an “in-drum lost stirrer” principle.

Normal backbone in the structure of the IXR is styrene and divinylbenzene with either sulphonate or quarternary amine as functional groups, depending on what kind of ions or products are to be removed from different process system in the NPP’s.

Nuclear power plants that generate radioactive waste have developed new recipes for conditioning of ion exchange resins in cement matrix over the last years in Sweden. The goals are to optimise the waste load, reduce the use of plasticisers and to be able to mix powder and bead resins in the same waste package. The reason for mixing different kinds of ion exchange resins in the same waste package is to optimise the waste load depending on availability of resins or dose rate limits. Ringhals nuclear power plant has at the Älvkarleby concrete laboratory finalised a study in the year 2000 [7]. The aim of this development project in Älvkarleby was to evaluate alternative recipes for conditioning of spent resins in cement matrix.

Waste acceptance criteria from SKB is as described in paragraph 2 above. A detailed criteria for the matrix mechanical strength is that must have a strength of at least 10 MPa without the use of any organic plasticisers.

The development program was performed in three steps,

1) 4 smaller tests with a volume of approximately 2.4 litre where performed in order to do some introductory tests. These tests showed that it was possible to mix the different types of ion exchange resins (powder and bead). All this tests were performed with an additive named Sikament 10 (a carboxylated plasticiser).
2) Test without Sikament 10 in laboratory scale which gave promising results.
3) Full scale tests, which verified that it is possible to mix different kinds of resins.

Full scale tests have shown that it is possible to produce a proper final waste package conditioned with cement to fulfill the demands of 10 MPa as a quality criteria. This is in most cases possible to achieve without the use of plasticisers or superplasticisers that could act as complexing agents in a final repository like SFR 1. In practice at Ringhals nuclear power plant they have to modify the stirrer that
is used in the in-drum mixing system when conditioning this kind of waste types to reach best compressive strength and homogeneity.

6. References


SAFETY ASSESSMENT ASPECTS OF MATERIALS FOR RADIOACTIVE WASTE CONTAINERS

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Abstract
In design testing and research BAM had investigated various aspects of material behaviour to ensure safety of RAM waste packages under operating and accident conditions:
- Strength and fracture toughness of cubic ductile cast iron containers in handling accidents,
- Thermal behaviour of ion exchanger resin material in a full-scale fire test of a cubic DCI container,
- Long-term, high and low temperature behaviour of VITON gaskets.

1 Handling Accident Impact Assessment of Cubic Ductile Cast Iron (DCI) Containers

Since 1992 BAM has investigated cubic DCI containers for disposal of radioactive wastes in drop tests with full-scale casks [1, 2, 3, 4]. In difference to a usual type B package transport container design the waste packages for interim storage or final disposal do not have additional bolt-on impact limiters that are constructed to reduce the mechanical forces acting on the package structure. While the regulatory 9 m drop test (transport) onto an essential rigid ("unyielding") target can be endured in most cases only with such impact limiters (sometimes called shock absorbers), the storage or disposal site related handling accident has to be endured by the packages usually not equipped with such impact limiters. This situation requires specific investigations concentrated on target behaviour and target/cask interaction during an impact, because the behaviour of the target rules the penetration of a container into this target, and the resulting impact force-time dependence that leads to related stress/strain-time relations in the container.
structure. In drop tests simulating the most rigid Konrad repository foundation BAM used a reinforced concrete slab placed upon a few cm thick compressed wet sand layer upon the unyielding target (see Figure 1). Since the early beginning of the drop tests with cubically shaped DCI containers BAM identified that the most critical drop position of the container is the flat horizontal drop onto the largest side of the cube; a drop onto the corner or onto the edge of the cube leads to larger penetrations into the target and subsequently to lower decelerations, impact forces and container stresses (see Figure 2).

Since then only flat bottom drops had been performed with the containers to investigate the rather critical stresses at designated points on the inner container surfaces (centre of the bottom plate, hollow groove positions between bottom and walls) caused by the hard impact including stress waves and container wall bending. For regular qualities of ductile cast iron (i.e. mainly ferritic structure with limited perlite content of up to 20 %) it was shown that with artificial crack-like flaws inside the container walls at the highest stressed locations this hard impact could be withstand [3]. In a research project BAM develops an advanced analysis method for this impact situation including a fracture mechanics assessment for the container if the material is melted by adding scrap metal from the dismantling of nuclear installations. In that case the material is mainly perlitic, and becomes much more brittle than the mainly ferritic DCI. Meanwhile
BAM has developed a reasonable applicable calculation method concerning the exact dynamic stress analysis of a container under these hard impact conditions [4, 5, 6, 7, 8] by using the finite element method (FEM). BAM has performed first fracture mechanics evaluations of dynamically loaded surface cracks. Figure 3a shows the FEM model and Figure 3b a comparison of calculated and measured strains at the centre of the container bottom side.

It has to be considered that stresses in a hard dynamic impact – as it has to be investigated in our case – shall not be calculated in a manner often used for relative soft impact analyses of casks with impact limiters or casks dropping onto “soft” targets, that is to take the maximum peak deceleration as basis for a quasistatic stress analysis. In case of a very dynamic impact leading to high peak stresses, and high stress rates as in the flat bottom drop of cubic containers with low penetration into the target only the dynamic FEM stress analysis is applicable. In further analyses BAM calculated the stress intensity factors at cracks in the bottom centre, and in the hollow grooves between bottom and walls (Fig. 4). Those stress intensity factors calculated for different
crack geometries can be compared with the fracture toughness values of the cask material tested at the dynamic strain rates as estimated for the container impact to judge on the integrity of the container structure, and the quality assurance measures to exclude crack-like defects bigger than

Figure 5: Critical depth of a crack in the centre of a plane container wall

the allowable ones. Figure 5 shows a preliminary result of the investigations presenting the relation between critical crack sizes in the centre of the container bottom and maximum principal stresses for some typical fracture toughness values of DCI materials melted with scrap additions. These crack-stress-relations meanwhile have been calculated in refined models also for the cracks in grooves. Results of the improved investigations will be presented soon [9].

2 Ion Exchanger Resin in Fire Testing

In the regulatory assessment of the cubic DCI container Type VI the question came up concerning the behaviour of ion exchanger resins inside the container in a 1 hour fire test according to the preliminary requirements for the German Konrad Repository. After laboratory tests with that material in autoclaves it was clear to BAM that this question could not be solved by calculations and/or small-scale tests. Mainly the problem concerning the heat transfer from the container wall to the bulk, and inside the bulk was not clear. For that reason a full scale fire test was performed with an original container filled with the spherical ion exchanger resin “LEWATIT S 100 KR/H chlorfrei” (manufacturer BAYER) with around 50 % moisture content. The test was carried out on the propane-burning BAM open fire test site [10] with flame temperature in the vicinity of the container surfaces of above 800 °C in average. During the fire the inner pressure increased but reached its maximum of 0.93 MPa after 5 h. No discontinuity of the pressure curve occurred. After four days the internal pressure decreased to about 0.43 MPa. The maximum inner wall temperature inside the container reached 392 °C after 73 min. Figure 6 shows the temperature distribution inside the ion exchanger resin along a horizontal symmetric axis. It shows a delayed heat transfer and a big influence of the temperature distribution by mass transfer, see, for example, the thermocouple at 215 mm distance from the container wall where the temperature increased in a very short time to about 100 °C. The maximum temperatures of ion exchanger resin will be at the edges and corners of the cubic container and were estimated to be more than 400 °C.
3 Long-Term, High and Low Temperature Performance of Viton Gaskets

For the investigation of the long-term behaviour of VITON gaskets a cask manufacturer’s (GNS) program was agreed and witnessed by BAM that included leak-tightness measurements of selected casks in one to two years intervals, gasket investigations of those and additional casks, and investigations of gaskets manufactured in recent time. For that examination MOSAIK casks were selected which were stored between 4.3 and 10.6 years. The results of the leak-tightness measurement at room temperature for two different gasket material mixtures (VITON MK 682 and MK 634) demonstrated the existing function with leakage rates $< 10^{-5}$ Pa m$^3$/s. To investigate the temperature influence compression set measurements at temperatures between 200 °C and -40 °C with the two VITON materials have been performed. The function limit is reached at a compression set of 100 % where no decompression of the gasket occurs after load relieve. Both materials had compression sets of around 35 % at 200 °C; MK 682 reached 100 % at -20 °C, MK 634 (a mixture designed for lower temperatures) reached 100 % compression set at -40 °C. In this condition the gasket has a hardness of 96 Shore A. Because it is doubtful, whether the gasket is able to compensate any geometrical deformation of the sealing system during use at these temperatures it was the intention to assess the behaviour of such a system in additional investigations. For that reason sealings were tested with nearly original dimensions. The elastomere seal was mounted in the dove-tailed groove of the lid and the lid was screwed to a flange of a vacuum chamber. This test design was positioned within a climatic chamber for the adjustment of the different test temperatures. During the cooling of a MK 634 seal a slow increase of permeation rate of helium through the VITON seal was recorded. Between -51 °C and -53 °C the leakage rate increased significantly as a result of forming a gap. At this temperature (Tg) the elastomere is losing its ability for elastic deformation. But this process is reversible. After warming up the sealing system the gap was closed again. In principle irradiated seals show the same behaviour. After an irradiation with appr. 16 Gray the opening of the gap began between -47 °C and -50 °C. Based on these test results the conclusion is allowed that special VITON
mixtures can be used down to -40 °C without the need for taking into account the special physical and chemical nature of the contents of the package for activity release considerations.

References


APPROACHES TO A REALISTIC DETERMINATION OF GAS GENERATION IN RADIOACTIVE WASTE

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Abstract
For final repositories, gas generation and its transport into the geosphere constitutes an important issue of the safety analysis. Especially after repository closure, gas generation might cause a pressure build-up and/or an acceleration of the nuclide transport with the groundwater which can be of importance for the results of a safety analysis.

Present methods to determine gas generation in radioactive waste sometimes lack a degree of realism, which cause design measures not justified on realistic assumptions. To improve this situation different approaches are characterised, which have been developed throughout the last years based on measurements on real wastes. The paper mainly deals with low and intermediate level waste but can be applied to high level waste as well.

The approaches developed will be outlined and examples will be given taken from experiences in licensing procedures. Finally, recommendations will be presented to enlarge the applicability of theses approaches and further qualify their basic input parameters.

1 Introduction

Gas is expected to be generated in underground repositories for radioactive wastes. The generation rates and quantities of gas that may be produced will vary with waste type and repository concept. However, issues arising from gas generation need to be addressed in safety cases for all repositories, since they cannot be ruled out a priori for any repository type or concept. As a consequence, gas generation may have an influence on design and operation of a repository and especially on its closure concept for ensuring long-term safety. Therefore, a strong incentive exists to determine the gas generation rate and the total amount of gas eventually generated in a repository with a reasonable degree of realism to avoid unnecessary and unjustified countermeasures.

The approaches that have been applied for this purpose and the present efforts for improvement are outlined in the following sections.

2 Generation processes

It is widely accepted that anaerobic corrosion and microbial degradation provide the major contribution to gas generation for low and intermediate level waste (L/ILW). Only in case of high dose rates as for high level waste (HLW) and spent nuclear fuel (SF) radiolysis may become relevant as well. Further processes like hydrolysis, thermal degradation or radioactive gas generation by decay are limited in their contribution to special types of waste (e.g. Magnox fuel) or special waste constituents (e.g. Aluminium, Zinc) or can be neglected at all (e.g. α decay, radioactive gases from spontaneous fission).

Especially the two most prominent contributors to gas generation, i.e. corrosion and microbial degradation, depend strongly on the environmental conditions like pH value, temperature, humidity, oxygen supply and on the waste itself. Corrosion rates for steel for example may vary over several orders of magnitude depending on the pH value while gas generation by corrosion is effectively suppressed if sufficient oxygen is available to maintain aerobic conditions.
If the environmental conditions in the post-operational phase of a repository cannot be assumed to be constant, time dependent generation rates have to be assessed either for individual processes or for the total gas generation rate. Furthermore, these conditions may even be influenced or controlled by the design of the repository. As an example the physico-chemical conditions in the near field of the high level waste disposal concept in Sweden is controlled by the bentonite backfill around the waste containers. By this way the copper surface of the containers remains nearly undisturbed as long as the environmental conditions can be kept constant.

3 Potential consequences of gas generation

The main problems associated with gas generation concern the following issues:

- Some of the generated gases are ignitable and thus induce the risk of fire or explosion. This may either apply to emplacement areas during the operational phase of a repository or to the release of these gases to the environment in the post-operational phase.
- Generated gases may incorporate radioactive isotopes (e.g. $^3$H$_2$O, $^{14}$CO$_2$, $^{14}$CH$_4$). Especially long-lived radionuclides like $^{14}$C may thus be released more rapidly as a gas than as a groundwater contaminant.
- Pressure build-up due to gas generation may impede the integrity of the engineered or geological barriers. This may open new flow paths for groundwater movement into the repository or for contaminated groundwater outflow.
- Pressure build-up by gas generation may interact with the lithostatic pressure of the host rock. The latter is responsible for the compaction behaviour of waste, backfill and seals. Since compaction may enhance the sealing capacity of the engineered barriers gas generation may have an impact on the effectiveness of these barriers.
- Finally, gas generation may introduce a second phase in flow processes. The resulting two-phase flow interacts with the geomechanical effects described above and is dependent on the water supply, which is essential for the gas generation processes. The impact of this two-phase flow and its interactions may be beneficial e.g. if a gas cushion forms, which delays or avoids water inflow to the repository. In contrast, it may accelerate contaminated groundwater movement and increase activity release rates to the biosphere. Because of these interactions the flow behaviour is non-linear and consequences therefore are difficult to predict.

For the treatment of gas generation in safety analyses this means, that a high gas generation rate may not always be a conservative rate, i.e. the highest radiological consequences may not be associated with the highest gas generation rates. This implies, that the results of a safety analysis should be based on the most realistic values for gas generation instead of upper bounds. In addition, of course the scenario analysis for the safety case should identify all relevant fep's induced by gas generation and a sensitivity analysis should ensure that sensitive parameters have been investigated sufficiently close to frame their possible impact.

4 Assessment of gas generation

4.1 Waste characterisation

To put assessment of gas generation into practice one starts with the characterisation of the waste. For LILW corrosion and microbial degradation have been identified as the most important gas generating mechanisms (see above). Accordingly, the determination of the metallic constituents
and the organic material in the waste are of primary interest. For some more or less homogeneous waste forms like ion exchange resins or evaporator concentrates this is rather easy to realise. Most of the LLW, however, appears as mixed waste with varying metallic and organic content. In these cases a careful assessment of the average metallic and organic portions in the waste is necessary.

If mixed kinds of metal may be present in the waste, additional consideration should be given to local corrosion effects from the formation of electrochemical elements. Special metallic constituents like aluminium, zinc or magnesium have to be characterised separately, because they can react intensely under gas formation depending on the chemical conditions. Hydrolysis of aluminium in high pH environment is a prominent example.

Gas generation rates are normally given as volume gas generated per unit of surface area and time. To assess gas generation rates therefore a reacting surface has to be determined. This poses special problems in mixed waste. Some values of experience may help but have to be adjusted to the waste type considered [1,2].

The determination of the organic contents in the waste is mostly handled less detailed. The lack of gas generation data for long-term assessment restricts the possibilities for differentiation considerably. Commonly, it is sufficient to distinguish between cellulose type materials and other organic constituents.

The chemical environment in the waste package (e.g. pH value) is first dependent on the waste itself. If the waste is fixed with grout, cement or similar a high pH environment (pH ~ 12) is prevailing. This holds until the contact of the grout with groundwater alters the chemical state or degrades the cementitious material.

For HLW and spent fuel corrosion takes place under rather well defined conditions. The materials are well known and their corrosion or gas generation rates are determined in extensive laboratory tests for most materials, repository concepts and host rocks. Furthermore, the reacting surfaces can be determined without problems. Organic materials normally will not be expected in these wastes. An exception may be if paraffin is used as a neutron absorber in spent fuel containers.

The assessment of gas generation from radiolysis for HLW or SF is necessary only if thin-walled containers are used or if a contact of the waste with groundwater cannot be excluded. The latter will in general take place not before some hundred or thousands of years when radioactive decay has reduced the radiation source strength considerably. Nonetheless, the radioactive inventory of HLW and spent fuel should be known with sufficient detail already from other necessities.

4.2 Gas generation potential

The most straightforward method for gas generation assessment is to calculate the total gas generation potential. For this purpose it is first assumed that the total metallic contents of a waste package is corroded by anaerobic corrosion. Secondly, the whole organic material in the waste is taken as degraded with the maximum gas output. Other contributors to gas generation like radiolysis or hydrolyses are either excluded because of their proven negligible contribution or are assessed in the same global manner.

Of course, this method provides strongly overestimated results for gas generation. Therefore, its applicability is restricted to situations, where the gas generation potential is very low and influences or interactions from outside the emplacement area can largely be excluded. The emplace-
ment of HLW or spent fuel in tight backfill (e.g. bentonite) or host rock material (salt, clay) may be an example. As long as container integrity can be assumed and gas transport through the engineered or geological barrier is restricted to diffusion rather than advective flow the gas generation potential gives sufficient information. The requirements to ensure these conditions (low gas generation, low pressure build-up, tight near field enclosure) are accordingly higher in this case.

If at least diffusional transport is needed to balance the low gas generation from the waste, time dependent gas generation has to be determined. This can be calculated from the gas generation potential using estimates of the upper bound of corrosion rates respective gas generation rates. In this simple situation the highest value for the rate can be considered as conservative.

4.3 Gas generation rates

In most cases the assessment of gas generation requires more detailed investigation and determination. For L/ILW e.g. different types of waste have to be treated. Gas generation has to be assessed for the individual waste package based on averaged values or for wastes with comparable gas generation behaviour. Environmental conditions may be assumed constant within a range of tolerance, if this can be justified from the chosen disposal concept. Last but not least is has to be checked if interactions with gas transport, geomechanical or geochemical effects have to be envisaged. If so, it has to be decided if a realistic or a conservative assessment is required for the safety analysis.

In this situation, mainly two approaches have been chosen in the past. The first and most often used relies on the determination of gas generation rates for each contributing process, for each waste to be considered and for each environmental condition. The second derives gas generation rates from statistical analysis of available measurement results from real waste packages. The evaluation can be differentiated for different types of waste. Since measurements are mainly performed under interim storage conditions some adjustment has to be made for adaption of the results to the repository situation. This is performed by purposeful experimental investigations which allow for interpolating or extrapolating their results to the prevailing conditions.

Both methods will be shortly outlined in the following sections.

Assessment of individual gas generation processes

In this approach the individual gas generating processes have to be assigned to the waste types to be considered. Table 1 gives an example for some LLW types. The main constituents are classified according to their corresponding gas generation mechanism. Radiolysis has been omitted because of its negligible contribution.

For each scenario identified in the scenario analysis the environmental conditions have to be characterised. At least the following parameter should be considered: oxygen supply (aerobic, anaerobic), pH value (in backfill or groundwater) and the type of groundwater (e.g. crystalline rock with low mineralisation, crystalline rock with high mineralisation, clay, undersaturated saline, saturated saline).

Correspondingly, the environmental conditions within a waste package have to be characterised. For waste without fixation neutral conditions can be assumed according to the common requirements for conditioning. For grouted waste a pH level of 12 typically has to be considered. Bituminization is applied mostly for concentrates. For this waste product gas generation can be ne-
neglected because the gas generating agents (metal, organics except bitumen) and the necessary water to start gas generation are missing. The bitumen itself shows only very low degradation rates if at all, which can be neglected.

Table 1: Assignment of gas generating mechanisms to waste types

<table>
<thead>
<tr>
<th>Waste type</th>
<th>main constituents</th>
<th>Relevant mechanisms</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ion exchange resin</td>
<td>No metal, Other organics</td>
<td>- M</td>
</tr>
<tr>
<td>Evaporator concentrates</td>
<td>- no organics</td>
<td>-</td>
</tr>
<tr>
<td>Metallic (only Fe)</td>
<td>Iron, steel, - no organics</td>
<td>- C</td>
</tr>
<tr>
<td>Metallic (mixed)</td>
<td>Iron, steel, Al, - no organics</td>
<td>Al C, H</td>
</tr>
<tr>
<td>Mixed waste (org.)</td>
<td>Cellulose, Other organics</td>
<td>- M</td>
</tr>
<tr>
<td>Ashes</td>
<td>Fe, Al</td>
<td>Al, carbides C, H</td>
</tr>
</tbody>
</table>

For the combinations of interior and exterior environmental conditions that apply for the investigated scenario the gas generation rate has to be quantified for each relevant gas generation mechanism in the last column of Table 1.

A large amount of measurement results are available for corrosion rates and corresponding gas generation rates under a variety of environmental conditions. Therefore sufficient knowledge and tools are available in most cases to provided realistic rates. Figure 1 shows an example two fitted curves for the relationship of the gas generation rate by corrosion from the pH value [3].

Figure 1 Dependence of the gas generation rate by anaerobic corrosion from the pH value for highly mineralised groundwater

For microbial degradation less detailed data are available and the determination of gas generation rates has to some extent to rely on qualitative assessments and plausible arguments. Nonetheless, the available measurement results enable some differentiation for environmental conditions and
material. The characterisation of individual metabolic processes and the transfer to repository conditions, however, remain a rather speculative exercise at the moment. The available experimental evidence shows that microbial gas generation happens with much lower rates than those that can be derived from conventional waste in landfills. Similarly it can be concluded that the theoretical gas generation potential by far overestimates the amount of microbial gas to be expected in a repository.

Finally, the assessment of individual gas generation processes may incorporate balances for the contributing media. This concerns e.g. the consumption of metals by corrosion, or of organic material by degradation. It may include also a balance for the corrosive medium if water supply is limited.

**Statistical evaluation of measurement results**

The previous paragraphs have focused on detailed investigations of certain gas generation mechanisms in laboratories and on measurements using individual waste containers (e.g. drums) with more or less well-characterised contents. The physico-chemical environment for gas generation in a L/ILW container in a repository is in general only to a limited extent comparable with laboratory conditions. Furthermore, the limited knowledge about the governing conditions, e.g. waste constituents contributing to or influencing gas generation, sometimes hamper transfer of laboratory measurements to real waste. Therefore, it has been considered helpful, especially for low-level waste, to complete existing knowledge about individual effects by measurements on real waste. The experience gained from these measurements can be used to create a validated link between laboratory data and real waste situations, which aids in a prognostic characterisation of gas generation for the post-operational phase of a repository.

First results of a comprehensive evaluation of measurement data from low-level waste have been reported in [2]. Measured gas generation rates were differentiated for the type of waste, waste age and temperature. Figure 2 demonstrates the temperature dependence derived from routine as well as deliberate measurements for two types of waste. Gas generation follows an Arrhenius type temperature function, which is waste type specific [4].

*Figure 2.* Temperature dependence of gas generation for different LLW types
4.4 **Time dependent rates**

For the assessment of gas generation in a safety analysis a further degree of sophistication may be required, if time dependent environmental conditions and correspondingly time dependent gas generation rates have to be considered. In this case, different phases are derived for which more or less constant conditions can be assumed. For each phase the same parameters have to be determined as for a scenario in the last section.

Typically, one can distinguish an initial aerobic phase in dry mine atmosphere. This phase forms the interface between the operational and the post-operational phase. Although no gas generation is awaited from corrosion under these conditions the material consumption in this phase may be important for the further steps of the analysis.

After sealing of the emplacement room or the whole repository aerobic conditions will continue until the oxygen is consumed. This time period has to be determined to provide the starting point for an anaerobic dry phase. Since in most repository concepts a flooding of the emplacement area by inflowing groundwater is expected, the dry conditions of the previous phase may convert to inundated conditions at a later time. Depending on the waste constituents and their interaction with the groundwater chemical condition may change thereafter again.

Thus the previously assumed constant conditions throughout a scenario are split in this case into several phases. In addition, it may be taken into account, that for corrosion metal parts may have a different thickness. This induces another time dependent feature, because with time thinner metal parts are completely corroded and contribute no longer to gas generation.

4.5 **Saturation limits**

As has been indicated before, microbial degradation at present is assessed with higher uncertainty margins than other mechanisms. Therefore, activities have started to assess a realistic gas generation potential from natural analogues. The existence of organic material for geologic time periods suggests, that the degradation process does not follow the energetically optimum path down to the last mol, but is stopped at a certain limit leading to a kind of saturation behaviour.

Just recently, first results from investigations on these subject have been achieved and will be published in the near future. The results indicate, that the gas generation potential is at least an order of magnitude lower as are long-term gas generation rates from microbial degradation.

5 **Conclusions**

In conclusion, characterisation of gas generation for the long-term safety assessment of a repository has become increasingly demanding throughout the last years. Sophisticated tools and investigations have been provided to cope with these requirements. Today a number of approaches can be offered according to the individual situation and the degree of detail required. Further improvements are possible, but have to be accompanied by according efforts for qualification.

6 **Literature**


Quality Assurance and Control
PRODUCTION CONTROLS (PC) AND TECHNICAL VERIFICATION TESTING (TVT). A METHODOLOGY FOR THE CONTROL AND TRACKING OF LILW WASTE PACKAGE CONDITIONING.

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Abstract

As part of its low and intermediate level radioactive waste (LILW) characterisation and acceptance activities, ENRESA has set up a quality control programme that covers the different phases of radioactive waste package production and implies different levels of tracking in generation, assessment of activity and control of the documentation associated therewith. Furthermore, ENRESA has made available the mechanisms required for verification, depending on the results of periodic sampling, of the quality of the end product delivered by the waste producers. Both processes are included within the framework of two programmes of complementary activities: Production Controls (PC) and Technical Verification Testing (TVT).

1 Production Controls (PC)

These controls consist of a type of auditing through which ENRESA verifies compliance with aspects included in the PDD (Package Description Document drawn up by the producer), of significance to ensure that the waste packages produced are similar to those studied during the characterisation processes. These PC's, which require qualification, as chief auditors, of the personal in charge of performance, may be of three types:

1.1 Resources PC (Type 1)

This type of PC, applied following approval by ENRESA of any new waste conditioning methodology proposed by the producers, includes the 'in situ' verification of the availability of equipment and documentation suitable for waste package generation, the fundamental objective being to guarantee that the producer possesses the technical resources required for waste conditioning to provide an end product in keeping with the contents of the documentation submitted for approval.

Prior to drawing up the PC, the producer submits to ENRESA the support documentation for the new methodology - the type package PDD - and ENRESA prepares the corresponding "Checklist", an action guideline that contains the aspects to be verified during tracking. These lists contain the following for each type of waste considered:

a Generic considerations:
✓ Resources possessed by the producer for the qualitative and quantitative determination of materials whose presence is restricted by the acceptance criteria relating to safety in disposal: pyrophoric materials, explosives, chelating agents, oils, etc.
✓ Calibration of the equipment used for physical, chemical and radiological determinations.
b Specific considerations:

✓ Homogeneous wastes (ion exchange resins, evaporator concentrates and sludges)
✓ Waste temporary storage, homogenisation and sampling systems.
✓ Cement dosing systems.
✓ Conditioning matrix agitation systems
✓ Chemical, physical and radiological determination resources.

✓ Heterogeneous solid wastes (compactable and non-compactable)
✓ Resources for the segregation of materials, elimination of humidity and suitable preparation of the wastes in the packaging.
✓ Equipment used for volume optimisation and void refill.
✓ Calculation equipment and methods for physical and radiological determinations.

✓ Filter and dispersible solid type wastes subjected to drying (desiccated sludges, incinerator ashes, etc.)
✓ Resources and materials used for the H.C. mould in metallic packaging.
✓ Resources for introduction and centering of the wastes in the pre-concreting mould.
✓ Equipment used for immobilisation mortar dosing.
✓ Calculation equipment and methods for physical and radiological determinations.

This PC is completed with the issuing of the corresponding report, which informs the producer of the observations and recommendations made as a result of tracking. If this report is favourable, it implies approval of the corresponding PDD.

1.2 Activity PC (Type 2):

This type of controls serves to check correct application by the waste producers of the activity determination methodology approved for each type package in the corresponding acceptance documents. The first step consists of a check by ENRESA, applied to the entire production of LILW, of the coherence of the activity of gamma emitters with respect to the dose rate of each package. These checks, performed using computer applications developed by ENRESA specifically for each producer and type package, are based on the basic principles of shielding physics.

The producers periodically report to ENRESA on their waste package production, including the physical and radiological determinations performed with respect to them (mass, waste type, conditioning, activity, dose rates and dates of generation and measuring, etc.). This information, transmitted by means of computer resources in a previously agreed format, is automatically processed by the aforementioned applications, which reflect any relevant deviation $D$ (1) equal to or greater than 200, whenever the producer determines the activity of the package by spectrometry on whole packages or non-conditioned samples, or $D \geq 10$ if the activity has been determined using the theoretical methods developed by ENRESA, or jointly with the producer (fig. 1).

\[
D = 100 \frac{A_{\text{producer}} - A_{\text{ENRESA}}}{A_{\text{ENRESA}}} \cdot \alpha (A_{\text{producer}} > A_{\text{ENRESA}}) 
\]

\[
D = 100 \frac{A_{\text{ENRESA}} - A_{\text{producer}}}{A_{\text{producer}}} \cdot \alpha (A_{\text{ENRESA}} > A_{\text{producer}}) 
\]

where:

$A_{\text{producer}}$: Activity indicated by the producer on the Package Docket.

$A_{\text{ENRESA}}$: Activity assessed by ENRESA on the basis of theoretical methods.

Following analysis of the causes and scope of the deviations detected, and as long as these are due to the following:
The activity of isotopes with a half-life of more than 5 years (the only ones of significance at the Disposal Facility)
Methodologies, procedures or processes used to determine activity (not for dose rate measurement or equipment involved therein)

all the additional information that the producer might provide on the systematic approach used and on the status and validity of the equipment calibrations and eventual calculation processes is requested. In parallel to the above, the producer is notified of the temporary suspension of the documentary acceptance of the type package, as well as of the possible removals scheduled, and is requested to provide one or more packages of those involved in the anomaly for Technical Verification Testing (TVT) (see section 2) at the Waste Quality Verification Laboratory (WQVL) of the El Cabril Disposal Facility.

Following reception of the results of the radiological waste package determinations, a check is made of possible relevant deviations \( D \) in accordance with the criteria indicated in figure 1, and corrective measures guaranteeing the correct evaluation of the activity of the packages to be produced, and re-evaluation of those generated, are implemented.

\[ \Omega \geq 50 + 100 \left( \frac{2\sigma_{BL} + 2\sigma_{r0}}{A_{r0} - A_{rca}} \right) \]

\[ \Omega \geq 75 + 100 \left( \frac{2\sigma_{BL} + 2\sigma_{r0}}{A_{r0} - A_{rca}} \right) \]

\[ D_1 \geq 200 \]

\[ D_2 \geq 200 \]

\[ D_3 \geq 10 \]

Figure 1: Relevant deviations versus the type of activity evaluation performed.
1.3 Process PC (Type 3):
The objective of this type of PC is to check *in situ* all aspects relating to the waste package generation process and the adequacy of these aspects with respect to the acceptance documentation. These are generally performed annually for each producer and type package.

As in the case of Type PC's, a "checklist" is drawn up on the basis of the information contained in the PDD, this allowing all the aspects of the type package generation process to be verified. The issues to be checked are as follows, depending on each type of waste:

- **Homogeneous wastes**
  - Waste agitation and homogenisation process and sampling.
  - Gamma spectrometry and chemical analysis of wastes.
  - Dosing and materials used in waste conditioning.
  - Setting, in the case of conditioning in an H.C. matrix, and waiting period prior to covering of packages.
  - Absence of free liquid and volume of occupation.
  - Weight and dose rate.
  - Activity assignment methodology.
  - Validity of cement and drum quality certificates.
  - Validity of calibration of spectrometer, mass determination scales, dosing system control instrumentation and radiation detectors for dose rate measurement.
  - Status of current revision of documents relating to conditioning.
  - Records generated during waste conditioning.

- **Heterogeneous wastes**
  - Segregation of materials and suitability of those with a high degree of rigidity or expansive power in the case of compactable wastes.
  - Elimination of humidity.
  - Package mass and dose rate.
  - Reference isotopy and/or activity assignment methodology.
  - Validity of quality certificate for metallic packaging.
  - Validity of calibration of weighing instrumentation and radiation detectors.
  - Revision of conditioning-related documentation used.
  - Records generated during waste conditioning.

- **Filters of liquid circuits and desiccated solids (Sludges, incinerator ashes, etc.)**
  - Generation of outer packaging H.C. mould.
  - Determination of mass prior to introduction of waste.
  - Type of waste to be conditioned.
  - Determination of mass following introduction of waste.
  - Absence of free liquid.
  - Dosing used for mould hydraulic conglomerant and for immobilisation conglomerant.
  - Volume of occupation of waste, checking that the space between the waste and any point of the packaging allows for a thickness of immobilising agent greater than that established by the applicable acceptance criteria.
  - Weight and dose rate of resulting package.
  - Checking of adequate distribution of blocking concrete and of degree of package filling.
  - Activity assignment methodology.
  - Validity of cement and packaging quality certificates.
  - Validity of calibration of weighing instrumentation and radiation detectors.
  - Revision of conditioning-related documents used.
  - Records generated during waste conditioning.

Following the control, a report is drawn up describing the checks performed, the documents examined, the non-conformities detected and the recommended corrective actions.
If the non-conformities are considered to be relevant, ENRESA may decide to temporarily suspend documentary acceptance of a group of packages or the waste stream in question, until such time as pertinent corrective actions are implemented.

2. Technical Verification Testing (TVT):

These are a set of checks and laboratory tests performed on packages with their acceptance processes approved (Process Book published) and that use non-destructive and destructive techniques for the determinations required. The objectives of TVT are as follows:

- Checking of package activity and similarity with that declared by the producers.
- Compliance with the package properties associated with the generation methodology.
- Chemical checks on aspects of significance for disposal facility safety.
- Compliance with objectives relating to the quality of the conditioned wastes.

The aspects relating to the quality of the characteristics of the conditioning carried out on the wastes centre initially on homogeneous moist wastes incorporated in a solid matrix of hydraulic conglomerant, the main objective of the TVT performed. At present, other waste streams are being tested, following the perfection of the corresponding measuring techniques. As in the case of production controls, a distinction is made between three types of TVT’s (figure 2) by gamma spectrometry.

### TVT Based on Type of Test Requested:

- **TYPE 1**
  - Non-destructive testing
  - Includes the following aspects:
    - Type 1
    - Radiocchemical and chemical determinations

- **TYPE 2**
  - Includes the following aspects:
    - Type 2
    - Removal of test pieces from package for subsequent checking

- **TYPE 3**

![Figure 2 Types of TVT](image)

### 2.1 TVT Type 1

The main objective of this type of TVT is to verify the physical and radiological parameters of the package with respect to the declarations made by the producers. Non-destructive techniques are used, among them the following:

a) Physical checks
   - Dimensional control.
   - Package mass.
   - Visual and photographic inspection of the structural status of the package.

b) Radiological checks
2.2 TVT Type 2
In addition to what is indicated for Type 1 TVT, these tests include the acquisition of solid samples by destructive techniques for subsequent chemical and radiochemical checks. The tests additional to those performed for Type 1 are as follows:
- Determination of weak β emitters
- Determination of α emitters
- Chemical determinations

2.3 TVT Type 3
In addition to what is indicated for TVT Types 1 and 2, these tests include the acquisition of test pieces by destructive package trepanation techniques for mechanical checks. The tests additional to those performed for Types 1 and 2 are as follows:
- Checking for absence of free liquid
- Checking of correct setting
- Mechanical matrix resistance tests.

Figure 3 summarises the tests contemplated for each type of TVT.

The following tests are considered, depending on the type of TVT:

<table>
<thead>
<tr>
<th>TESTS</th>
<th>TVT Type 1</th>
<th>TVT Type 2</th>
<th>TVT Type 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Visual and photographic inspection</td>
<td>x</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>Determination of dose rate</td>
<td>x</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>Determination of package homogeneity</td>
<td>x</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>Dimensional control</td>
<td>x</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>Chemical spectrometry</td>
<td>x</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>Degree of homogeneity</td>
<td>x</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>Absence of free liquid</td>
<td>x</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>Correct setting</td>
<td>x</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>Radiochemical determinations</td>
<td>x</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>Mechanical tests</td>
<td>x</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>Other determinations</td>
<td>x</td>
<td>x</td>
<td>x</td>
</tr>
</tbody>
</table>

Figure 3 Tests included in TVT

2.4 TVT scheduling
The annual scheduling of TVT is carried out on the basis of the following:
- Number of typified waste streams with acceptance process completed.
- Characteristics of such streams.
- Capacity of the WQVL.
- Period between TVT’s for one same stream.
- Results of production controls and/or previous TVT’s.
- Waste transport programme.

TVT’s are documented by way of two types of reports:
a) Results report: This is an internal document issued by the WQVL and describes the actions performed on the package subjected to TVT and the results obtained, with the pertinent documentation attached.

b) Annual report: This is drawn up by the ENRESA LILW Engineering Department and includes summaries of the results, evaluations and conclusions obtained from all the TVT’s carried out during the year.

Once the reports for each TVT have been published, the results are evaluated (checking of activity, absence of free liquid and mechanical testing). If significant discrepancies are encountered with respect to the parameters indicated in the PDD and/or Package Dockets, the producer is informed and additional information is requested or, where appropriate, suitable corrective actions are proposed.

2.5 Knowledge acquired and feedback of experiences.

Depending on the knowledge acquired as a result of the PC’s and TVT’s, ENRESA established a flow of information with the producer (feedback of experiences), which allows progress to be made as regards know-how and improvement of the different techniques, materials and methodologies for waste conditioning, this in turn allowing better documented, higher quality products to be obtained, with subsequent optimisation of such products and a positive effect on the operation and safety of the Disposal Facility.

5. References

[1] ENRESA; 031-ES-IN-0053 "Clasificaciôn, metodologia y criterios de aceptaciônde los bultos de residuos radiactivos sólidos o solidificados, procedentes de la operaciônde las instalaciones nucleares y radiactivas, para su acondicionamiento en los contenedores de hormigôn de la instalaciôn nuclear de almacenamiento de residuos radiactivos de sierra de Albarrana". (Classification, methodology and acceptance criteria for packages of solid or solidified radioactive wastes from the operation of nuclear and radioactive installations, for conditioning in the concrete containers of the Sierra de Albarrana nuclear installation for radioactive waste disposal). Rev. 2 September 1996.


Quality Assurance Measures for Radioactive Waste Products to be Returned from Abroad

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Abstract
The sharing of installations for the conditioning of radioactive waste between multiple waste producers is common practice. In case of facilities, which accept radioactive waste from foreign countries to be conditioned and the waste products to be returned, special considerations have to be taken into account regarding QA. One prominent example is the COGEMA/La Hague site in France, where spent nuclear fuel from several countries is reprocessed and the residues being conditioned for return.

According to a contract between COGEMA and the German utility companies, spent fuel from German nuclear power plants is transported to the La Hague site, reprocessed and conditioned. The first cask containing 28 canisters with vitrified high active concentrate arrived back in Germany in 1996. Independent quality assurance measures in addition to the already implemented quality management system had to be developed to make sure, that the waste products comply with the national regulations defined for transport, interim storage and final disposal in Germany.

In general, two ways exist to demonstrate the fulfilment of waste acceptance requirements:
- Process qualification with subsequent inspections or
- random checks of the final product.

In the case of vitrified waste products with high levels of dose rate, process qualification with subsequent process inspections was chosen.

During the process qualification procedure, the vitrification process has been assessed by independent experts (here: Produktkontrollstelle, PKS) on behalf of the "Bundesamt für Strahlenschutz" (Federal Office for Radiation Protection, BfS), the competent regulatory authority for disposal of radioactive waste in Germany, for its suitability to produce an appropriate product for final disposal. The qualification of the vitrification process for high active concentrate from reprocessing of spent fuel from light-water reactors at the COGEMA UP2 and UP3 plants has been approved by BfS in 1996. Regular inspections are carried out to ensure, that the qualified process conditions are met. These inspections are accomplished in two levels:
- Continuous process inspections by independent (from the service provider) experts (for the COGEMA/La Hague site: Bureau Véritas, BV) on behalf of the customers and
- regular (about twice per year) inspections and audits of the on-site inspector by independent experts on behalf of BfS.

For every canister to be returned to Germany, the documentation is checked by PKS on behalf of BfS. In addition to the control measures performed by the on-site inspector during production, direct measurements of every canister, including gamma- and neutron-dose rate, are carried out before loading the canisters into casks for transport to Germany.

Process qualification with inspections proved to be a suitable method to ensure the quality of radioactive waste products, even in the case of conditioning in foreign countries. In total 420 canisters with vitrified residues from reprocessing of spent fuel in 15 casks have been returned from France to Germany since 1996.
Introduction

In Germany, radioactive waste arises, e.g., from the operation of nuclear power plants, from research centres, from the reprocessing of spent nuclear fuel abroad or from the decommissioning of nuclear installations. These wastes have to be treated and packaged (i.e. conditioned) for long-term storage and disposal.

Prior to disposal of radioactive waste in Germany, the compliance with the regulations for transport, interim storage and final disposal has to be demonstrated. In particular, independent control measures are required.

The fulfilment of the waste acceptance requirements (AR) [1,2] for final disposal may be demonstrated within the quality control procedure by

- qualification of conditioning processes with subsequent inspections or
- random checks on existing waste packages.

Process qualifications can be done per campaign or campaign independent.

In addition to the Federal Office for Radiation Protection (Bundesamt für Strahlenschutz, BfS) as the responsible authority for radioactive waste disposal in Germany the supervisory authorities of the resp. facilities are involved in the conditioning process. In 1994 the competent authorities agreed on a harmonized quality control procedure for radioactive waste (see fig. 1):

- Authorized experts evaluate the process control quality plan (PCQP) on behalf of the BfS and the supervisory authority.
- The supervisory authority and the BfS agree on a common PCQP.
- Accompanying inspections defined in the PCQP are carried out by an authorized expert on behalf of or coordinated with the supervisory authority.
- Based on a test report written by this authorized expert the BfS confirms the fulfilment of the AR or approves the waste for final disposal; in parallel to this the approval from the supervisory authority for the interim storage of the waste is given.

Because of the federal structure of Germany variations of this scheme are possible.

The quality assurance measures for radioactive waste disposed of in the Morsleben repository (ERAM) between 1994 and 1998 were successfully performed according to this scheme [3,4].

QA/QC for German Waste from Processing Abroad

Reprocessing of spent nuclear fuel from German power reactors takes place at the COGEMA / La Hague plant in France and at the BNFL reprocessing plant at Sellafield / UK. The fission product concentrate is vitrified for return to Germany. For this "return waste" the German quality assurance procedure (see fig. 1) must be applied accordingly.

The QA/QC procedures for vitrified high level waste from reprocessing at the COGEMA / La Hague plant are described as an example.

Approval Procedure

The first step for acceptance of radioactive waste products from processing in foreign countries is the approval of the waste specification by the German Federal Ministry of Environment, Nature Conservation and Reactor Safety (BMU). The approval procedure is outlined in fig. 2.

The final specification for the vitrified waste product was issued by COGEMA in 1986. After an agreed period of two years for assessments by experts (here: Reactor Safety Commission, RSK) the specification was approved by BMU in 1988.
Fig. 1: The Quality Control Procedure in Germany
Process Qualification

For the vitrification process at the COGEMA facilities in La Hague a campaign independent process qualification was carried out. The process qualification is based on the Handbook for Process Qualification, which describes the process and the QA/QC measures performed by the producer. The procedure for process qualification is outlined in fig. 3.

The purpose of the process qualification is to make sure, that the waste product complies with the national regulations for final disposal. Since a final decision on the repository site for heat generating radioactive waste in Germany is not taken, the assessment on the suitability for final disposal is based on a list of 16 properties and characteristic values of HAW glass products relevant for disposal (see table 1). These 16 properties and characteristic values were determined by an expert group on behalf of the German federal ministry of research and technology (Bundesministerium für Forschung und Technologie, BMFT) [5].
Fig. 3: Process Qualification

<table>
<thead>
<tr>
<th>No.</th>
<th>Relevant Property</th>
<th>No.</th>
<th>Relevant Property</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Total activity</td>
<td>9</td>
<td>Mass Fractions (Waste, frit, additives)</td>
</tr>
<tr>
<td>2</td>
<td>Activities of rel. Nuclides</td>
<td>10</td>
<td>Mixing</td>
</tr>
<tr>
<td>3</td>
<td>Criticality Safety</td>
<td>11</td>
<td>Product Conditions (physical)</td>
</tr>
<tr>
<td>4</td>
<td>Thermal Properties</td>
<td>12</td>
<td>Hydrolytic Stability</td>
</tr>
<tr>
<td>5</td>
<td>Dose Rate</td>
<td>13</td>
<td>Quality of Container</td>
</tr>
<tr>
<td>6</td>
<td>Surface Contamination</td>
<td>14</td>
<td>Package Mass</td>
</tr>
<tr>
<td>7</td>
<td>Quality of Fixation Material</td>
<td>15</td>
<td>Stackability</td>
</tr>
<tr>
<td>8</td>
<td>Composition of Raw Waste</td>
<td>16</td>
<td>Marking of Package</td>
</tr>
</tbody>
</table>

Table 1: Relevant Properties and Characteristic Values
Continuous Process Inspections

In 1987, before startup of the vitrification plants in La Hague, the reprocessing customers agreed to charge one on-site inspector (for COGEMA/La Hague: Bureau Veritas, BV) with continuous surveillance of the vitrification process. The main duties of the on-site inspector are:

- Audits according to ISO 9002,
- surveillance of the process,
- inspections of the relevant facilities,
- document checking for the waste packages,
- certification for all conform products/containers/waste packages.

Independent Control by PKS

The on-site inspector is working on behalf of the customers and is therefore not regarded to be completely independent as required by the German regulations. To compensate for this "dependency" additional control measures by independent experts (here: PKS) are implemented on behalf of the competent German authority (BfS). The scheme for quality assurance is outlined in fig. 4. To ensure the independence of the on-site inspector and to verify the fulfillment of the qualified process conditions PKS performs in particular the following tasks:

- Regular (about twice per year) audits of the on-site inspector,
- regular (about twice per year) inspections of the relevant facilities,
- document checking for the waste packages to be returned to Germany.

During the audits of the on-site inspector the following points are addressed:

- Changes within the on-site inspector’s organization,
- changes in the working procedures of the on-site inspector,
- changes of instrumentation and procedures for the vitrification plant,
- results of the surveillance activities,
- results of document checking,
- discussion of additional relevant topics.

The inspection programme of PKS for the vitrification facilities contains the following main aspects/items:

- Site-visit,
- inspection of external subcontractors,
- discussion with the operator of the reprocessing and vitrification plants,
- discussion with the responsible QA/QC section of the operator,
- random inspection of the ongoing conditioning process,
- follow-up of identified deviations,
- checking of relevant documents (such as working procedures – applicable or descriptive documents – or production records).
**Direct Measurements of the Final Product**

The radionuclide inventory of a canister is determined by the compositions of the high active liquor and the fines solution and the resp. amount. The gamma and neutron dose rates are calculated during production from the radionuclide inventory.

As an additional control measure direct measurements of the gamma and neutron dose rate of each canister before loading the canisters into casks for transport to Germany were implemented on demand of PKS. These measurements are performed under surveillance of independent experts on behalf of the supervisory authority for the Gorleben interim storage facility (Niedersächsisches Umweltministerium).

The measured dose rates can be compared to the dose rates calculated from the radionuclide inventory. Figures 5 and 6 show the distribution of the ratio between measured and calculated dose rate for the gamma resp. neutron dose rate. The comparison shows, that measured and calculated dose rates are in good agreement. This result proves, that the relevant gamma and neutron emitters were determined correctly.
Fig. 5: Gamma Dose Rate for 19 loadings each of 28 canisters

Fig. 6: Neutron Dose Rate for 19 loadings each of 28 canisters
Conclusion

Process qualification with subsequent inspection is accepted to be a suitable method to ensure the quality of radioactive waste products. It proved also to be applicable for radioactive waste from reprocessing in foreign countries.

Continuous surveillance of the conditioning process by the on-site inspector whose activities are monitored and assessed by German experts forms a sound basis for the quality assurance measures on behalf of the German authorities.

The comparison between measured gamma and neutron dose rates and calculated values based on the determined inventory of the canisters proved to be a reliable quality control parameter. The dose rate measurements of the final product can therefore be used as an additional checking procedure to assure the quality of the process and the product.

The described QA/QC concept applied to foreign reprocessing and conditioning of HLW containers can also help to enhance the public acceptance for the waste return in Germany.

Up to now 420 canisters in 15 casks have been transported from France back to Germany in the last years. Another 12 casks are already loaded with 336 canisters and are awaiting the transport to Germany in La Hague.

References


MELTING OF QUALIFIED HLW GLASS - THE PROCESS AND PRODUCT CONTROL OF THE KARLSRUHE VITRIFICATION FACILITY (VEK)

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Abstract
Vitrification of about 70 m³ of high-level waste (HLW) solution with a total activity of 7.7E17 Bq is a major step within the decommissioning program of the former Karlsruhe pilot reprocessing plant WAK. The immobilization will be carried out in a new vitrification plant designated VEK. The technology applied is based on a liquid-fed ceramic melter developed by Forschungszentrum Karlsruhe (FZK).
Licensing of the waste product requires the qualification of the VEK process for production of glass canisters the specifications of which have to be in accordance with a set of 16 specified final disposal-relevant parameters. In support of construction, operation and licensing of VEK and the glass product as well, a nonradioactive prototype vitrification test facility was operated from 1998 to 2000 at the Institut für Nukleare Entsorgung (INE) of FZK. Within the scope several long-term test runs it could be demonstrated that the VEK process is suitable to produce qualified canisterized glass.

Introduction
As a result of twenty years of reprocessing operation, about 70 m³ of high-level waste (HLW) concentrate are currently stored on the site of the former Karlsruhe pilot reprocessing plant (WAK). This facility which is under decommissioning since 1991 will be dismantled and the site converted to green field until the year 2010. An essential step in the decommissioning program is the conditioning of the stored HLW and its subsequent transportation to the German intermediate storage site at Gorleben. The conversion of the HLW into a stable waste form will be carried out by vitrification in a new plant designated VEK (Verglasungseinrichtung Karlsruhe), which is now under construction on the WAK site. Hot operation of the plant is scheduled from 2004 until 2005. Owner of the plant is FZK, which is also responsible for the management of the vitrified waste.

Product Licensing
There are two independent licensing procedures for the glass product and for construction and operation of the VEK plant. Licensing of the plant is the duty of the government of the federal state of Baden-Württemberg, whereas the licensing of the product is under the responsibility of the federal government of Germany. The institution in charge is the federal office for radiation protection (Bundesamt für Strahlenschutz, BfS).
The basis for acceptence of the HLW product is formed by a set of 16 parameters which are considered to be relevant for final disposal. These specified parameters listed in Table I can be grouped into three categories, related to the radioactivity, the chemical and physical properties of the glass and to the quality properties of the stainless steel canister. The composition of the glass
The product as the most important property is determined by parameters No. 7-9, other parameters (No. 1-5, 11 and 12) are also governed by the composition.

A main step of the licensing procedure for the product is the qualification of the vitrification process applied in VEK. All the required data, control steps and quality assurance measures that are necessary to produce glass canisters qualified for final disposal are composed in a file named ‘Process Qualification Manual’ which is part of the license application documentation. The process-relevant data given in this document are based on the experimental testing of the vitrification technique in a 1:1 scale nonradioactive prototype test facility (see below). The licensing procedure has actually proceeded to an advanced status, finalization of the process qualification will be performed within the scope of the cold test operation of VEK, scheduled for 2004.

**Table 1: Parameters/Properties of glass canisters relevant for final disposal acceptance**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Parameter</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 Total activity</td>
<td>9 Mass ratio waste oxides to glass frit</td>
</tr>
<tr>
<td>2 Activity of relevant nuclides</td>
<td>10 Homogeneity</td>
</tr>
<tr>
<td>3 Safety against criticality</td>
<td>11 Physical conditions</td>
</tr>
<tr>
<td>4 Thermal properties</td>
<td>12 Hydrolytic stability</td>
</tr>
<tr>
<td>5 Dose rate</td>
<td>13 Quality of the canister</td>
</tr>
<tr>
<td>6 Surface contamination</td>
<td>14 Mass of the glass canister</td>
</tr>
<tr>
<td>7 Quality of glass frit</td>
<td>15 Stackability and handling properties</td>
</tr>
<tr>
<td>8 HLW composition</td>
<td>16 Identification of the glass canister</td>
</tr>
</tbody>
</table>

**HLW and Glass Product**

Currently, the waste is stored in two tanks, featuring different chemical compositions, radioactivity levels and volumes. Prior to their processing, the contents of the two tanks will be mixed, so that a uniform waste solution can be expected to be delivered to the VEK plant. The nominal composition of the waste solution to be vitrified – based on sampling results from the storage tanks and calculation of the resulting mix – is given in Fig. 1. The waste solution is characterized by a total oxide yield of 121 g/l and a high specific activity of 1.2E13 Bq/l. Considerable elements are sodium, which appears in the highest concentration of all elements and the noble metals ruthenium, palladium and rhodium which altogether amount to a concentration of more than 5 g/l. Sodium has an important function in the glass network.
structure, whereas the noble metals play a special role in nuclear waste glass melting because of their insolubility in the glass [1]. Following the international state-of-the-art the HLW will be immobilized in borosilicate glass. The development of a proper glass matrix was carried out by INE of Forschungszentrum Karlsruhe from 1987-1989 and characterization took place from 1990 until 1995 in the scope of an extensive technology program established for vitrification of the WAK waste solution [2]. Table II shows the composition of the reference HLW glass product based on target waste oxides loading of 16 wt.%. The table also gives the data of electrical resistivity and viscosity essential for the melting process. The production of glasses within a range of 13-19 wt.% is tolerated. Glasses beyond these limits are considered as out-of-specification product.

### Table II: Composition and melting properties of the reference HLW glass product.

<table>
<thead>
<tr>
<th>Oxide</th>
<th>wt%</th>
<th>Melt temperature</th>
<th>Viscosity (dPa·s)</th>
<th>El. resistivity (Ω·cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO₂</td>
<td>50.40</td>
<td>950°C – 1150°C</td>
<td>530</td>
<td>21.0</td>
</tr>
<tr>
<td>B₂O₃</td>
<td>14.78</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>2.60</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Li₂O</td>
<td>2.94</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Na₂O</td>
<td>5.96</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CaO</td>
<td>4.45</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>MgO</td>
<td>1.85</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>TiO₂</td>
<td>1.01</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Waste oxides</td>
<td>16.00</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

### VEK Vitrification Technology

The vitrification technology applied in VEK is based on the liquid-fed ceramic melter (LFCM) process, which has been developed by INE of Forschungszentrum Karlsruhe. The core process comprehends the steps of HLW reception from the storage tanks, the melter feeding with HLW solution and glass frit, the waste glass melting and glass pouring, the subsequent glass canister treatment, the wet and dry off-gas treatment and the recycling of the secondary liquid wastes after their concentration in an evaporator system. A detailed description of the VEK vitrification process is given elsewhere [2].

Due to the limited HLW stock and, hence, operation time VEK has been designed as a compact-size vitrification plant. Key component of the process is a cylindrical-shaped Joule-heated ceramic melter, which is schematically shown in Fig. 2. The melt tank consists of a high temperature refractory surrounded by a back-up refractory and several layers of different insulation ceramics. Electric current between one pair of air-cooled power electrodes heats the glass pool. Feeding of the melter with HLW solution (continuously) and bead-shaped glass frit (batchwise) is carried out by two separate lines through the central inlet pipe in the melter ceiling. The materials conversion processes take place in a process zone located on top of the molten glass pool. In the upper layer of this zone evaporation and drying processes take place. In the layer below the resulting nitrate salts are converted to oxides (calcination), which then enter into glass melting reactions with the glass beads in deeper layers with increasing temperature. The cool process zone (‘cold cap’) has an important barrier function against escape of semi-volatile compounds from the melt. Therefore the extent of the cold cap is an essential parameter which has to be strongly controlled during operation in order to maximize the portion of materials immobilized in glass.

Glass pouring is performed by means of a bottom drain system acting as a thermal valve. It mainly consists of a metallic glass channel inside a thick-walled Inconel 690® pin. For the purpose of pouring is inductively heated by a 10 kHz mean frequency (MF) heating, thus melting
the solid glass inside the channel. Adjusting of the MF power input performs control of the pouring rate. Stop of glass discharge is achieved by discontinuation of powering. The melter is designed to manage waste glasses with high noble metals concentrations. As the noble metals are not soluble in borosilicate glass melt, they form separate particulate phases, which due to their high density tend to settle to the melter floor. If remaining in the system for longer time these sediments may become immobile. Accumulation then can lead to increasing distortion of the electrical field in the melt with strong impact on processing behaviour. The noble metals compatibility is achieved by combination of an inclined bottom area of the melt tank and the bottom drain system. The sloped walls causes the collection of the sediments in the discharge area from where they are removed in the early phase of the glass pouring. No water-cooled mechanical stirring is needed like in other process techniques.

![Schematic view of the VEK glass melter](image)

**Figure 2: Schematic view of the VEK glass melter**

The relatively low capacity of the glass pool which amounts to about 160 l / 420 kg of melt allows pouring batches of 100 kg only. As a consequence the filling of a canister with a target glass capacity of 400 kg requires four batches. Based on an experimentally verified nominal glass production rate of approximately 7 kg/h the time period between the pouring batches is almost 15 h and the time for production of a complete canister takes about 60 h. During the complete filling procedure the canister remains unheated. Some characteristic canister production data are compiled in Table III.

**Table III: Canister Production Data**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Data</th>
</tr>
</thead>
<tbody>
<tr>
<td>Canister capacity</td>
<td>400 kg</td>
</tr>
<tr>
<td>Mass per pouring batch</td>
<td>100 kg</td>
</tr>
<tr>
<td>Time between pourings</td>
<td>15 h</td>
</tr>
<tr>
<td>Canister production time</td>
<td>60 h</td>
</tr>
<tr>
<td>Pouring rate</td>
<td>100-120 kg/h</td>
</tr>
<tr>
<td>Time per pouring</td>
<td>1.5 h</td>
</tr>
<tr>
<td>Glass pouring temperature</td>
<td>1100°C</td>
</tr>
</tbody>
</table>
Process and Product Control

Besides ensuring a safe operation process control also serves for maintaining process conditions, which are necessary to produce a waste glass with specified properties. Parameters belonging to this category are the melting temperature, the homogeneity (mixing) as well as the glass pouring conditions.

Melting temperature
The melt temperature is continuously monitored and recorded by means of a thermocouple, which is deeply immersed in the glass melt. The temperature is controlled by adjusting the electrical current-controlled power input that the target range of 1150-1200°C is maintained at the monitoring point.

Glass homogeneity
Homogeneity is mainly the result of a sufficiently long residence time and intensive mixing. Both requirements have been implemented in the melter design. Based on the nominal feed rate of 10 l/h and a corresponding glass melting rate of about 7 kg/h an average residence time of approximately 60 h is reached. This considerably long mixing time in combination with the intensive thermal convection occurring in the upper melt tank area (confirmed by numerical 3D modeling) ensures the production of a refined and well homogenized glass.

Glass pouring
During the pouring procedure a pouring rate in the range of 100-120 kg/h is to be maintained in order to achieve a moderate melt level decrease inside the melter and to attain a uniform spread of the glass melt in the canister. The glass flow rate is deduced from the weight increment measured by weight loading cells.

Glass composition
With respect to glass product properties main attention has to be paid to the correct feeding of the melter. This means that the ratio of the input streams of glass frit and waste solution (i.e. stream of waste oxides based on a given oxide yield) has to be adjusted to ensure a product within tolerable deviations from the target waste oxide loading. Glass product out of this range has to be treated as out-of-specification product, as rework is not practicable.

The melter feeding system of the VEK plant is illustrated in Fig. 3. It contains the two 3 m³ receipt tanks, a 300 l feeding vessel and a glass frit feeding system. The feeding vessel is the central equipment for control of melter feeding. The level variation of the feeding vessel controls the continuous HLW stream from the feeding vessel to the melter, maintained by a one-stage airlift as well as the glass frit stream from the glass frit feeding system. The feeding vessel works within a defined volume range to ensure – besides a stable airlift feeding characteristic – especially the transfer of a constant HLW volume (25 l) to the melter between refillings from one of the two receipt tanks. This volume forms the basis for the glass frit addition. At minimum level, the refilling is activated. When reaching the upper level, refilling is terminated and the volume of 25 l is subsequently transferred to the melter. Reaching of the upper level limit simultaneously initiates the addition of the glass frit adjusted to the volume of 25 l of HLW. This kind of linked control assures an always constant ratio of HLW volume and quantity of glass frit. Moreover, the link is independent on the HLW feed rate. A change in the feed rate merely entails a change in the frequency of the feeding vessel’s refilling and the linked glass addition. Control of the precision of the melter feeding is performed by means of a protocol, which contains information about the materials input during the period between two following refillings, defined as feeding vessel cycle. Among these data there are the transferred HLW volume, the mass of added glass frit and the calculated waste oxide loading. Considerable deviations from the
target value would lead to a new adjustment of the glass frit mass per feeding vessel cycle. In addition to the short-term control (check approx. every 2.5 hours) another control step is performed by balancing the materials input into the melter over 10 feeding vessel cycles. If this long-term check (every 20-25 h) reveals deviations of the waste loading from the target the glass frit addition is corrected. The way of melter feeding and of twice control of material input ensures the production of glass within the desired composition range (see below).

The control steps to achieve correct melter feeding are:

Step 1: Determination of the total oxide yield as well as that of specific activity-related and process-related elements by chemical analysis from sampling of the receipt tanks. Fixing of the waste oxide loading of the glass product according to the analysis results. Adjustment of the glass frit mass addition necessary for 25 l of HLW solution transferred during a feeding vessel cycle. Control of material input per feeding vessel cycle and comparison with target value. In case of considerable deviation, correction of the glass frit mass input immediately after termination of the feeding vessel cycle.

Step 5: Materials input balance over 10 feeding vessel cycles to provide long-term control of the glass composition. If a systematic deviation is detected, it is compensated by a corrected glass frit addition.

Prototype Testing and Experimental Results

In support of planning, construction, operation and also licensing of VEK a nonradioactive prototype vitrification test facility was operated from 1998 to 2000 at the Institut für Nukleare Entsorgung (INE) of FZK. Fig. 4 shows a photo from the remotely equipped melter cell. Besides the melter it contains the feeding vessel and also a dust scrubber and a condenser belonging to the melt off-gas treatment system. Within the scope of five long-term test runs 26 m³ of HLW simulate was converted to 18 metric tons of waste glass during 3000 h of feeding time. This full-scale operation has also been used to promote the licensing procedure of glass product as well. For this purpose nearly 1000 samples were taken from the pouring glass stream and analyzed with respect to their chemical composition and waste oxide loading. These properties are decisive for several parameters given in Table I.

Main question to be answered in this context was whether the process and its control features have the ability to produce a waste glass composition within specified ranges. Another question
in the focus concerned the impact of the multiple pouring on the integrity of the glass product inside the canister.

**Simulation of HLW and glass product**

In respect of representative operational and glass product results, the waste solution was simulated as close as possible to composition of the genuine waste [3]. Except from technetium and the actinide elements, which where substituted by manganese and lanthanum, respectively, as they do not exist as stable isotopes all the other constituents were used as found in the HLW solution. The glass frit applied was the same as foreseen for hot operation. Thus, the melting properties of the resulting glass product have been expected to be comparable to these of the active glass.

![Figure 4: View into the melter cell of the Prototype Test Facility (PVA)](image)

**Waste glass loading**

The target waste oxide loading of the glass is fixed at 16 wt% with a tolerable bandwidth of ± 3 wt%. The aim of the melter feeding control for all the test runs also when using simulates with non-reference HLW compositions was to demonstrate its effectiveness to run the system at target line of 16 wt%. The average waste loading achieved during all test runs varied between 16.1 and 16.4 wt%, which reflects the accuracy of the melter feeding control philosophy.

**Composition of the glass product**

In order to confirm the effectiveness of the melter feed control, samples were taken from the pouring glass stream according to a defined sampling sequence. The samples then were analyzed with respect to their chemical composition by use of X-ray fluorescence and induction-coupled plasma emission spectroscopy (ICP-AES). Fig. 5 shows as a typical example the analysis result of the first test campaign. The diagram contains the concentration of SiO$_2$ as the main glass structure-forming compound originating from the glass frit and of Na$_2$O, originating from both the HLW solution and the glass frit as function of the sequence of samples taken during the test run. Each 20 samples cover the production of one canister (canister No.1: sample 1 to 20, canister No.2 sample 21 to 40 and so on) composed of 4 pouring batches with 5 samples per pouring. In total 6.5 canisters were produced. The equivalent production time was nearly three weeks. The following issues are obtained from the diagram for both oxides: (1) there are straight concentration curves indicating a constant HLW to glass frit melter feeding over the whole test
run, (2) if compared to the target indicated on the right side of the diagram the concentration curves confirms the correct control of the melter input streams of HLW and glass frit.

**Figure 5: Concentration of SiO₂ and Na₂O determined from glass samples from pouring taken during the complete PVA-1 test run**

These results were found in all the tests also for other elements except the noble metals. Their segregating behavior and the melter design used lead to a nonuniform concentration in the pouring stream. Due to their sedimentation and collection in the discharge area of the melter (see Fig. 2) their discharge characteristics shows high concentrations in the initial phase of the pouring. As a consequence there is also a nonuniform vertical distribution in the canister with higher concentrations near the interfaces between the glass batches. However, this has no effect on quality issues as the noble metals due to the HLW characteristics are of no radiological relevance.

**Condition of the canisterized glass**

As discussed above, the limited glass inventory of the melter requires the filling of a canister by four pouring batches. The question arising is to what extent this procedure affects the integrity of the canisterized glass. Due to the comparatively long time between the pourings and the abandonment of a canister heating there is a potential that the glass near the surface of a poured batch cools too much down so that the heat capacity of the following batch is not high enough to remelt this area, leading to separated batches. Temperature measurements in the interface area between the batches inside the canister could not clear up the situation. The temperatures in the interfaces after reheating by the following batch were found to be in the range of the softening temperature of 520-540°C. Further clarification was expected from non-destructive investigation of canisterized glass carried out by Bundesanstalt für Materialprüfung (BAM) in Berlin. Application of digital radiography (DR) and computerized 3D-tomography (CT) allowed to get access to information about the conditions of the glass inside the canister, especially near the batch interfaces. Results are given in Fig. 6, which shows the DR-image of a complete canister. Whereas the upper half of the glass block shows a monolithic appearance, the lower one reveals inhomogeneities in the interfaces between first/second and second/third batch as well. Additionally, in the center of the lower part a sharp-edged void can be observed. It is obviously the result of a shrinking process. An image of a CT-slice shows the symmetric radial extent of this void (see right side of Fig. 6). The other slice images contained show the radial voids and fissures distribution found in inhomogeneous areas near the batch interfaces. From all the 3D data obtained the surface increase (sum of outer and inner surface) relative to the surface of a
monolithic block was calculated to be 7. The comparison to the recommended limit of 20 shows that the multiple pouring canister delivers a well acceptable integrity of the canisterized glass.

![Image showing canisterized glass and pouring batches](image)

**Fig. 6.** Images from nondestructive investigation of canisterized glass by digital radiography and computerized tomography. Canister filled by 4 pouring batches of 100 kg each.

**Summary and Conclusions**

The new vitrification plant of Forschungszentrum Karlsruhe will produce about fifty tons of highly active waste glass poured into European standard canisters. The licensing of the waste glass canisters, which is under the responsibility of the federal German government, requires the qualification of the waste glass and of the vitrification process. The acceptability of the waste glass package is determined by a set of 16 selected parameters/properties, the bulk of which is related to the composition and waste loading of the glass product. The qualification of the vitrification process, developed by Forschungszentrum, has been supported by operation of a 1:1 scale prototype test facility. In the scope of several long-term tests the ability of the process to produce waste glass with composition and waste loading within specified ranges could be successfully proven. Moreover, it could be shown that the condition of the canisterized glass poured in four batches is acceptable and in compliance with the requirements.

**References**


Assessment of the compatibility of vitrified high-level waste with geological disposal in a Boom Clay formation: remaining uncertainties

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Abstract
At SCK•CEN, the compatibility of vitrified high-level waste with the candidate geological disposal in Boom Clay is assessed in an experimental programme. This paper discusses the contribution of this programme to performance assessment, the uncertainties associated with the extrapolation of the experimental data to longer timescales, and other uncertainties affecting the compatibility of the HLW glasses with the geological disposal. The experimental programme has provided estimations for the long-term glass dissolution rate and mobile radionuclide concentrations in media representative for the geological disposal conditions. Long-term glass dissolution rates in Boom Clay slurries are between 10 and 100 times smaller than the initial dissolution rate in pure water, and depend on the glass composition. Very small mobile radionuclide concentrations are observed in reducing conditions at the lowest temperature considered (40°C) ~5x10⁻⁹ M for Np-237 and Tc-99.

1 Introduction
In Belgium seven nuclear power plants (NPP) are operated, which together generate a total installed capacity of 5.5 GW(e). About 55% of the total electricity need is actually provided by the NPP's. Besides several nuclear facilities are in operation in Belgium, including fuel fabrication, materials research reactors, etc. To cope with the long-living and high-level nuclear waste that is generated from the various nuclear activities, already in the late 70's screening studies were initiated by SCK•CEN on the Boom Clay formation as a potential host for that type of waste. An underground research laboratory (URL) in Boom Clay was built in the early 80's at a depth of ~220m. Today this URL infrastructure has been extended with a second shaft, and the total length of the underground gallery is more than 200 m now. This URL has enabled to investigate the possibilities to construct underground galleries in Boom Clay at that depth, and to perform various kinds of experiments (mechanical, corrosion, migration, etc) in in situ conditions. NIRAS/ONDRAF, the national agency for fissile materials and radioactive waste, has proposed a design for geological disposal, basically consisting of a central stainless steel disposal tube (in which the HLW canisters are inserted), backfilled with a clay/sand mixture. NIRAS/ONDRAF recently completed SAFIR2 (Safety Assessment and Feasibility Interim Report 2), that summarizes the knowledge accumulated over the past ten years [1]. SAFIR2 will serve as a basis for the Belgian government to evaluate the national research, development and demonstration programme on geological disposal of long-living and high-level radioactive waste. In Belgium two types of HLW glasses are to be considered. Some 2200 packages (total volume of 241 m³) PAMELA glass were produced by BELGOPROCESS, to condition the high-level reprocessing waste generated by the former EUROCHEMIC plant. The second type of HLW glass is produced by the COGEMA R7T7 facility, and refers to the reprocessed HLW from the NPP spent fuel. Presently either 420 or 3915 packages of 0.18 m³ are taken into account, depending on the partial or full reprocessing option [1]. In total four glass compositions are to be
considered: two borosilicate glasses (the R7T7 SON68 and the PAMELA SM513) and two high Al$_2$O$_3$ (~20 wt%) borosilicate PAMELA glasses SM527 and SM539.

The experimental approach is linked with the assessment of the performance (PA) of the disposal design [2], by providing input data for PA calculations. PA uses in particular the estimations for the glass matrix dissolution rate observed in experiments in conditions relevant for the geological environment. The radionuclide concentrations that are measured in solution in such experiments are taken into account for the selection of solubility limits for radionuclides in PA calculations. The experimental programme further aims to consolidate the scientific basis for the parameter values provided to PA, by interpreting the experimental results in terms of mechanisms, amongst other by modeling. In our experimental programme, we did not spend large attention to processes that we considered as being sufficiently documented in the international literature [3,4], such as radiation damage in the glass, thermal effects, mechanical properties, and other.

This paper reports some of the main conclusions from the R&D programme performed over several years time, focusing on the glass matrix dissolution rate and radionuclide concentrations in the interstitial claywater, and discusses the remaining uncertainties.

2 Experimental approach

We performed a parametric experimental study of the dissolution of the mentioned HLW glasses in media they will potentially come into contact with. First of all, we opted for static interaction tests (no flow of the solution), because of the extremely small hydraulic conductivity of the Boom Clay formation ($10^{-12}$ m/s). We let the following parameters vary:
- temperature (various values between 40°C and 150°C)
- glass surface area to solution volume (S/V; between 10 m$^{-1}$ and 10000 m$^{-1}$)
- interacting medium (pure solutions, e.g. Boom claywater, slurries of claywater with Boom Clay, and slurries of claywater with candidate near field materials)

The programme includes tests in relatively realistic conditions (low temperature, low S/V, relatively high Boom Clay concentration, negative redox potential), in dissolution-accelerating conditions (to obtain data for the long-term in short-term tests), or in intermediate situations. Temperature and S/V are typical interaction accelerating parameters. Take as an example the accidental situation of water ingress in the void space in upper side of a glass canister: this would yield an S/V of 30 m$^{-1}$ (geometrical surface of the upper glass block surface). S/V ratios in our programme are up to 10000 m$^{-1}$, so the exposed glass surface in our tests is up to ~300 times as large as in that hypothetical "reality". This parametric approach also offers the best chance to gather an understanding of the dissolution processes that will occur over long periods of time (geological timescale) in the geological disposal environment, that yet can't be specified/identified now.

We also performed corrosion tests in the underground laboratory in contact with Boom Clay, to verify if the data obtained in laboratory conditions can be confirmed in "real" in situ condition.

3 Experimental results

We will highlight two cases that provide input data for the performance assessment study of the geological disposal of HLW glass in Boom Clay. Detailed data on these and other tests, and on the on-going modelling studies can be found in [5-7].
The first case is the determination of a long-term dissolution rate of the glass matrix in the geological disposal environment, based on the principle that performance assessment should not use dissolution rates that are lower than can be demonstrated in representative experimental conditions. Having identified that amongst different potential backfill materials (the near field in the early stage of the disposal) and Boom Clay (the far field, and expected to be the environment in the long-term) the latter is one of the most corrosive ones, we report on dissolution data in Boom Clay media. We derived a long-term dissolution rate for glass SON68 of about $10^{-2}$ g.m$^{-2}$.d$^{-1}$, this is about 10 times lower than the initial dissolution rate at 40°C in pure water, and about 100 times smaller than the initial dissolution rate at 90°C in pure water [8]. This data was obtained from a test at 40°C, at S/V of 2500 m$^{-1}$, in a slurry of Boom clay with claywater, in reducing conditions. The dissolution rate was almost constant with time between 200 and 700 days duration. A statistical evaluation of the data led to the value of $10^{-2}$ g.m$^{-2}$.d$^{-1}$, which was considered as the best estimate value for input in the performance assessment study. The difference with the dissolution rate in similar tests at 90°C was statistically not significant. Other experiments, with e.g. lower clay concentration or with addition of glass frit to the clay have suggested lower dissolution rates. These are however not (yet) used in PA calculations, because these environments are less representative for the current Belgian disposal design.

This best estimate dissolution rate of $10^{-2}$ g.m$^{-2}$.d$^{-1}$ yields a lifetime of the glass block of ~160000 years, based on a simplified model [6]. The minimum lifetime of the glass block for acting as a barrier in the performance assessment is 100000 years. We expect that the lifetime of a SON68 glass block will be far longer than these 100000 years, based on the 160000 years calculated above, and based on the expectation that the dissolution rate will further decrease with time. Strong uncertainties exist at the present in the international literature, as to the value of the "final" dissolution rate, and about the rate controlling mechanism. Diffusion processes (water in glass [9], silica through clay [10]) might very well control the long term glass dissolution – resulting in decreasing rates with time. Final dissolution rates $10^{-4}$ times smaller than the initial rate were measured by Vernaz in pure water at 90°C [11].

Figure 1: Total mass losses upon dissolution of glass SON68 (left) and SM539 (right) in a clay slurry (1140 g Boom clay per liter clay water), with an S/V ratio of 2500 m$^{-1}$, at 40°C [5].

Figure 1 shows the experimental data for the test reported above, together with the data for similar tests on glass SM539. The best estimate rate here is about $10^{-1}$ g.m$^{-2}$.d$^{-1}$ [5], about ten times larger than for SON68 (and accordingly a 10 times shorter lifetime). This high value is probably related with the occurrence of secondary minerals that form during the glass dissolution, either within the leachate or on top of the glass. The generation of secondary minerals is believed
to suppress the saturation of the solution with respect to typically Si and Al, and thus allowing further dissolution of the glass [12]. If we can’t demonstrate that the long term dissolution of SM539 falls strongly down beyond the observed one, this glass couldn’t be considered as a barrier in the disposal concept.

The second case deals with the leaching behaviour of radionuclides from the HLW glasses when coming into contact with potential disposal media. We intensively studied the leaching behaviour of Np-237 and Tc-99, since they were calculated by the performance assessment studies to be amongst the most critical radionuclides in a geological repository in Boom Clay. The glasses doped with varying concentrations of Np-237 and Tc-99 (see [5,7]) were corroded in an oxidized "short-term" medium (consisting of FoCa clay candidate backfill material and claywater, and a reducing "long-term" medium (including various solids (Boom Clay, FoCa clay, steel corrosion products and claywater). All tests were done in N2 atmosphere. The short term medium is dominated by the backfill material, whereas in the long term medium the Boom Clay (=the disposal rock) is believed to dominate the near field. The duration of the oxidized conditions depends on the disposal design that will be chosen, but it may range from hundreds till thousands of years. Leaching tests were performed at 40°C and 90°C, S/V of 100 m⁻¹, and for different periods up to 2 years. Full details are given in [5, 7]. The released radionuclides were separated in a mobile fraction (mobile through Boom Clay, i.e. having a size smaller than ~5 nm), and an immobile fraction (colloids larger than 5 nm, sorbed on the clay).

We currently observed steady state values for the mobile Np-237 concentration in solution within a few months or less. The mobile Tc-99 concentrations did achieve steady state concentrations in fewer situations. We show the average mobile concentrations over the two years duration obtained at 40°C in Table I. We were able to propose a speciation for both radionuclides [7]:

- Np and Tc in the reducing medium have the (IV) valence state, and both appear to be poorly soluble. The mobile Np-237 concentration agrees with the solubility of Np(OH)₄, which is 5x10⁻⁹ M. We expect that Np-humate complexes have formed as well in the claywater. However, because more than 99% of the organic matter is immobile in Boom Clay, Np will be essentially complexed and retained by this fraction. The mobile Tc-99 concentration agrees with the solubility of TcO₂.xH₂O.

- Np and Tc in the oxidized medium are respectively in the (V) and (VII) valence state. Their mobile concentrations are substantially larger than in the reducing medium ~10 times for Np, ~10000 times for Tc. The Np concentrations have not yet reached the solubility of NpO₂OH. Np-carbonate complexes, observed in tests reported in [13] are probably present as well. The Tc concentrations in the oxidizing medium appear to correspond with the solubility of TcO₄, that does not form any complexes.

Table I Mobile concentration in solution of Np-237 and Tc-99 upon interaction of SON68 glass in two clay slurries at 40°C, S/V of 100m⁻¹. The Np and Tc content in the glass was 0.3, resp. 0.025 wt%.

<table>
<thead>
<tr>
<th>Medium</th>
<th>[Np] (in M)</th>
<th>[Tc] (in M)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Short term medium (oxidizing)</td>
<td>5x10⁻⁸</td>
<td>2x10⁻⁵</td>
</tr>
<tr>
<td>Long term medium (reducing)</td>
<td>5x10⁻⁹</td>
<td>5x10⁻⁹</td>
</tr>
</tbody>
</table>

The redox condition of the medium appears to have a very strong effect on the radionuclide concentration in solution – reducing conditions favoring low concentrations. The other parameters (temperature, Np and Tc concentration in the glass, glass composition) have a smaller effect [7].

Even if we are able to propose values for the steady state Np and Tc concentration and a speciation in the media considered, strong uncertainty remains on the radionuclide speciation and
on the long-term mobile concentration. We know for instance that Np humate complexes will form, possibly mixed hydroxy/humate complexes. Maybe other cations may be part of the complexes as well. But because of the extremely low Np concentration in solution in the reducing medium, no direct speciation techniques are applicable. Finally, the values given in Table I are steady state concentrations (kinetically controlled), rather than thermodynamic equilibrium values.

4 Remaining uncertainties

4.1 The long term (years to geological timescales) glass dissolution rate.
There is strong evidence in the international literature that the rate of glass dissolution in closed systems (no water flow; limited water volumes) will decrease fairly soon – within days in accelerating conditions [4, 8]. Reasons are that upon saturation of silica (the main glass constituent) in the solution glass dissolution slows down. Diffusion processes (for instance, water into glass) and processes like precipitation from solution will induce dissolution continuing beyond the silica saturation stage. In fact the glass dissolution will never stop completely, but one may assume that the long term dissolution rate will be lower than 10^{-2} g.m^{-2}.d^{-1} as concluded above in case of glass SON68. Values of 100 times smaller were reported in pure solutions [11], and might be expected in clay media as well, once the sorption sites of the clay are saturated - typically with silica. Glass dissolution in clay media (such as Boom Clay) will therefore achieve saturation much later, and glass will dissolve to a much higher extent than in pure solutions. We need evidence about the value of the dissolution rate in Boom Clay medium that will prevail in the long term, and we must identify the processes controlling this final rate.

4.2 The long term release of the radionuclides of importance.
We have not achieved the thermodynamic solubility concentrations for the Np and Tc species in our laboratory studies. So, will these thermodynamic equilibrium concentrations be achieved indeed in the long term? We have not established a full speciation of the Np and Tc species either. Continued research is needed to identify the species formed, and to gather basic thermodynamic data. As an example we mention Np-humate (HA) complexes. It has been demonstrated that Np readily complexes with HA [13], but in in situ Boom Clay condition they will be present in a very minor extent, because only 0.05% of the organic matter in Boom Clay is mobile. It is highly probable that most of the Np will react with the immobile fraction of organic matter, retarding the Np migration. Therefore, association/dissociation reactions should be considered. It will be in addition of prime importance to know the evolution of the near field around the glass canister with time, because its geochemistry will strongly affect the speciation of the actinides and of Tc. How fast will the near field become reducing (consumption of the oxygen), what will be the concentration of the complexants (humic acids, carbonates, etc) in the near field in function of time? Will co-precipitation of different radionuclides present in the fully active glass affect their solubility? Finally, the performance assessment of the geological disposal of HLW glass may generate new insights or conclusions. Recently other radionuclides were found to become critical for the long-term safety as well, such as Sc-79, Sn-126, Zr-93 and Pd-103 [2]. Data on their mobile concentration in disposal media have to be obtained as well.

4.3 The effect of integrated conditions on the glass dissolution and the radionuclide release.
In the real geological disposal situation different processes will occur simultaneously: we mention, glass dissolution, migration of released elements through the near field, sorption on the solids (canister/container corrosion products, backfill & far field materials, complexation, precipitation effects, radiolysis effects (γ and α radiolysis). In several R&D programmes integrated situations are being considered [5, 14-15]. An in situ test is underway in Belgium,
which integrates all processes mentioned above [16]. The database for the effect of radiolysis in integrated conditions is quite limited – its effects may be essentially to lower the solution pH and to oxidize the near field. The main uncertainty associated with the integrated effects however may be to identify the main processes occurring in this situation, and defining the parameter values for these processes, so as to be able to elaborate an analytical model for the glass dissolution in integrated conditions.

4.4 The initial state of the glass blocks that will eventually come in contact with the near field. In case of the HLW glasses considered in Belgium large efforts have been done to know the quality of the glasses, and to guarantee the quality of the real glasses, amongst others by characterizing the glasses (reference as well as real ones), by qualifying the vitrification process, and by quality assurance during the process [17, 18]. In the design phase of the vitrification processes the glass compositions were optimized, and databases of glass properties were established. But the as produced high-level waste glass blocks will evolve with time. We mention the following uncertainties:

- The structural incorporation of the long-living radionuclides should be documented. Uncertainties exist in general presently regarding the valence/coordination state of e.g. Tc and the actinides in the HLW glasses. Their structural incorporation might change after production due to radiation effects. This knowledge is necessary in order to get a full understanding of the radionuclide release from the glasses.
- The calculated chemical and radiochemical inventory in the HLW glasses should be validated by measurements, especially for the critical long-living radionuclides. Measurements by chemical and radiochemical analysis have been reported on fully active glass samples [e.g.,19]. However, long-living radionuclides such as Se-79, Sn-126, Zr-93 and Pd-107 were identified as critical radionuclides for the long-term performance. Validation of their calculated inventory by measurements is recommended.

5 Concluding remarks

A large database has been established on the long-term dissolution behaviour (matrix dissolution and radionuclide release) of candidate high-level waste glasses in Boom Clay media - the candidate disposal rock in Belgium - and considerable understanding has been achieved. This is confirmed by the evolutions at the international level. Analytical models have been proposed and elaborated that might enhance the understanding of the glass dissolution [20, 21], and enable to make predictions to the longer term (geological timescale). But we need more evidence of the parameter values and underlying processes of the long-term glass dissolution. We should succeed to decrease the best estimate dissolution rate for the R7T7 glass of $10^{-2}$ g.m$^{-2}$.d$^{-1}$, for example by applying longer test durations or by using experimental set-ups with a better precision. Also the glass dissolution in the integrated conditions of the geological disposal system needs to be known in more detail. The status of the HLW glass block that will eventually come into contact with the geological medium must be characterized in more detail as well.

6 Acknowledgements

The authors gratefully acknowledge the financial support by NIRAS/ONDRAF and the EC for most of the R&D discussed in this paper.
7 References

1. Introduction

Regulation of low level radioactive waste (LLW) disposals in the UK requires the waste producing organisation to have quality control procedures in place to ensure that waste meets the transfer and disposal authorisations set out by the Environment Agencies*. For large volume producers this means measuring the radioactivity content of waste drums prior to disposal. These measurements are then used to make a declaration of the radioactivity content of the waste consignment. Since a variety of methods may be used to assay the LLW, it was proposed by the Agency that a programme of quality intercomparison testing should be established to monitor the relative performance of the assay systems employed and provide a check on site operator’s quality control procedures.

A review of the potential for on-site checking using suitable standard waste packages was carried out at the Waste Quality Checking Laboratory (WQCL) in 1998.

As a result of this review, two drums of simulated radioactive waste have been prepared and developed at WQCL and these have been used as the basis of a programme of on-site

intercomparison tests similar to the Round Robin Test carried out by EN-TRAP partners in 1996 [1]. Sites and operators are selected on a random basis and the system operators are asked to assay the drum as they would their own waste arisings. Results are collected on the day of the test and a short report is prepared for each.

In general the waste drum gamma assay systems used by site operators fall into two categories,

1. High resolution gamma spectrometry (HRGS) systems using high purity Ge detectors of low to moderate efficiency and high spectral resolution.
2. Low resolution gamma spectrometry (LRGS) systems using high efficiency NaI detectors of low spectral resolution.

We report typical results achieved by both types of assay systems when analysing the WQCL reference drums.

2 Review and Package Development

An initial review examined the key variables and design requirements for the standard waste packages needed for the intercomparison testing programme in order to represent typical UK low level waste streams. The key parameters were:

- The number of reference drums needed to fully meet the requirements.
- Traceability of sources
- The waste matrix materials to be included in the drums.
- The radionuclides to be included and their distribution within the drums.
- The activity content of the reference standards used.
- Packages to be readily transportable by road.
- Packages to provide a realistic check on the different monitoring systems in use.

In carrying out this review NNC collated data from two main sources. Firstly, historical UK consignment data records held at WQCL were examined and data were compiled on typical waste matrix densities (Figure 1) gamma emitting radionuclide content and radioactivity from a variety of waste producers. Secondly, a survey was conducted to gather information on the types of LLW gamma assay systems in use on operators’ sites.

The review concluded that two reference drums should be used The first, RD15, was the reference drum used in the EN-TRAP Round Robin Test thereby adding to the database of existing measurements on this drum and providing a broader base from which to draw comparisons. The drum contains six radioactive reference sources distributed within simulated mixed density waste material. The total gamma emitting radioactivity of this drum is close to the average (4.6 MBq) as calculated from measurements made at WQCL on thirteen consignments of real LLW (814 drums) over seven years of routine operation. These consignments originated from a number of different sites and represent a variety of UK waste streams. The radionuclides chosen; Mn-54, Co-60, Sb-125, Ba-133, Cs-137 and Am-241 represent both activation and fission products and are fairly typical of those found in real waste.
Figure 1  Distribution of drum weights checked at WQCL in previous consignments

The second drum, RD16, was designed and built by NNC specifically for this intercomparison testing programme. The philosophy behind the design of the drum was to produce a modular waste package, which can be assembled in a variety of configurations and can be adapted to meet specific testing requirements. The drum (Figure 2) contains twelve discrete modules or 'cheeses' which fit closely together and may be filled with materials of different density. Three different materials were chosen to fill the modules these are vermiculite (density 0.1 g.cm\(^{-3}\)) 'spilkene' (a proprietary absorbent material with density 0.73 g.cm\(^{-3}\)) and pea gravel (density 1.48 g.cm\(^{-3}\)).

The modules may also contain reference sources and are arranged in three layers, top middle and bottom, containing four modules in each layer. Sources have been prepared at the Laboratory to simulate real waste in that they are dispersed in sealed packages rather than point sources. The sources can be added to selected modules and the configuration of the drum can thereby be tailored to simulate a variety of UK waste streams. The number of radionuclides used in this drum so far has varied between three and eleven in three separate configurations.

3  Testing Programme

A programme of site testing commenced in December 1999 and to date a total of eleven sites have been included. Sites may be selected at random or at the request of an Agency Site Inspector. The process is initiated by a letter from the Agency to the site operator notifying them that they have been selected to take part in the programme and requesting their cooperation. WQCL is informed and the Laboratory Manager makes contact with the site to make the necessary arrangements. Prior to the test the standard waste packages are loaded into overpack containers for transport to the selected site. The overpacks, which are
Type A, IP2 containers, are used in order to comply with the regulations for transporting radioactive materials by road in the UK. Seals are placed on the overpacks to ensure the integrity of the reference standard drums and they are then transported to the test site in readiness for the test.

**Figure 2**  *Schematic diagram of RD16 construction*

On-site checking is conducted by the site operator's staff as for a normal drum of LLW. The measurements are supervised by WQCL staff and normally witnessed by the Environment Agency Site Inspector. Relevant details of the assay system, such as, count times used density correction if applicable and any calibration and performance monitoring performed, are provided. The results together with any relevant calibration date are collected on the day of the test and a report to the Environment Agency on the site test is prepared on return to WQCL. A copy of the test report is provided to the Agency Programme Manager, the Site Inspector and the Site Operator.

### 4 Results and Discussion

The results of the tests are described below. The radionuclides detected and quantified in each of the drums together with their reference radioactivities (decay corrected where appropriate) are compared and the differences between the true and reported activities for each radionuclide present are expressed as a percentage deviation from the reference value using the equation below.

\[
\text{% Deviation} = \frac{\text{Measured Radioactivity (Bq)} - \text{Reference Radioactivity (Bq)}}{\text{Reference Radioactivity (Bq)}} \times 100
\]

The percentage deviations for the different type of system and reference package are shown in Figures 3 to 6. Of the eleven sites tested to date six sites operate HRGS systems (coded H1 to H6 in Figures) and five sites operate LRGS systems (coded L1 to L6 in Figures). The results are described below.
4.1 HRGS Systems

Figure 3 shows the results for drum RD15. The deviations from the true activity reported at each of the test sites for each of the six gamma emitting sources in this drum are shown together with the mean results obtained from the WQCL SGS system (WQCL) and the mean results obtained in the Round Robin Test (RR).

![Graph showing deviations from true activity for different gamma emitting sources in drum RD15](image)

**Figure 3 HRGS System Results for Reference Drum RD15**

All of the sites with HRGS systems were able to identify all of the sources within the drum with the exception of Am-241, which was not identified by any of the site systems. Am-241, which is a low energy (59.6 KeV) gamma emitter, was however identified and quantified by the WQCL SGS and the majority of the Round Robin Test partners. With the exception of sites H4 and H6, most of the sites underestimated the activity of the sources detected. All measurements lie within the range reported for this drum in the Round Robin Test with the exception of the Cs-137, Mn-54 and Sb-125 values for site H4.

All the HRGS systems tested used drum rotation and were able to locate sources within the waste package by making measurements on individual slices or segments (usually 8). The positions of the sources as determined by the systems agreed well with their true location. Most of the scanners had a single high purity Ge detector, however where high throughput is required multiple detectors are employed to reduce the scan time.

In general, some form of correction is made for the density of the waste in the drum. This was normally a simple mass over volume correction but some of the systems made use of a transmission source or the differential peak attenuation method.
Comparison of results for drum RD16 is complicated by the fact that three different drum configurations have been used so far in the testing programme. Each configuration however was measured using the WQCL SGS to provide a benchmark measurement for each test. The results are given in Table 1 and are illustrated in Figure 4.

Table 1  
RD16 Results for HRGS systems - % Deviations from True Activities

<table>
<thead>
<tr>
<th>Site Code</th>
<th>Configuration</th>
<th>Co-60</th>
<th>Cs-137</th>
<th>Cs-134</th>
<th>Sb-125</th>
<th>Co-57</th>
<th>Am-241</th>
</tr>
</thead>
<tbody>
<tr>
<td>H1</td>
<td>RD16/3</td>
<td>-5.7</td>
<td>135.2</td>
<td>-6.4</td>
<td>N/D</td>
<td>N/D</td>
<td>N/D</td>
</tr>
<tr>
<td>WQCL</td>
<td>&quot;</td>
<td>-1.8</td>
<td>21.8</td>
<td>-57.3</td>
<td>24.1</td>
<td>61.8</td>
<td>25.5</td>
</tr>
<tr>
<td>H2</td>
<td>RD16/2</td>
<td>N/P</td>
<td>N/P</td>
<td>-62.5</td>
<td>6</td>
<td>N/P</td>
<td>N/D</td>
</tr>
<tr>
<td>H3</td>
<td>&quot;</td>
<td>N/P</td>
<td>N/P</td>
<td>3.9</td>
<td>7.4</td>
<td>N/P</td>
<td>N/D</td>
</tr>
<tr>
<td>H4</td>
<td>&quot;</td>
<td>N/P</td>
<td>N/P</td>
<td>8.8</td>
<td>42.7</td>
<td>N/P</td>
<td>N/D</td>
</tr>
<tr>
<td>WQCL</td>
<td>&quot;</td>
<td>N/P</td>
<td>N/P</td>
<td>-53.1</td>
<td>17.6</td>
<td>N/P</td>
<td>-54.6</td>
</tr>
<tr>
<td>H5</td>
<td>RD16/1</td>
<td>N/D</td>
<td>N/D</td>
<td>-62.5</td>
<td>6</td>
<td>N/P</td>
<td>N/D</td>
</tr>
<tr>
<td>H6</td>
<td>&quot;</td>
<td>47.1</td>
<td>N/D</td>
<td>-14.3</td>
<td>N/D</td>
<td>N/D</td>
<td>N/D</td>
</tr>
<tr>
<td>WQCL</td>
<td>&quot;</td>
<td>-39.6</td>
<td>-44.7</td>
<td>26.4</td>
<td>N/D</td>
<td>N/D</td>
<td>-57.2</td>
</tr>
</tbody>
</table>

N/P = Source not present  
N/D = Source not detected

There was a noticeable tendency for the WQCL and H2 systems to underestimate the activity of Cs-134 when it is present. This was attributed to the location of this source within modules containing high density material. Experience with the WQCL system has shown a systematic trend in the uncertainty associated with sources located in high density matrices, which, can give rise to an underestimation of their radioactivity.
Co-60 and Cs-137 were not always identified within this drum although they are key constituents of most site operator's waste streams, this is attributed to the generally lower activity of the sources used and the higher mass of the RD16 standard waste package. As with drum RD15, Am-241 was only detected by the WQCL SGS system.

4.2 LRGS Systems

Five of the sites that have taken part in the intercomparison testing so far, use low resolution (LRGS) drum scanners. These systems use high efficiency NaI detectors (normally 3) to make rapid measurements of the key gamma emitting radionuclides within the waste. Drum rotation is used and a mass measurement is taken to make a gamma attenuation correction. A disadvantage of these systems is that only one or two peaks can be detected due to the low spectral resolution of the instruments.

The majority of the systems so far examined were configured to measure only the Co-60 in the waste, values for other radioactive components would then be inferred by the operator from a waste stream fingerprint. The results for the Co-60 measurements expressed as a deviation from the true radioactivity on the date of the test for these systems is shown in Figure 5.

![LRGS results for Co-60 in both Reference Drums](image)

**Figure 5** LRGS results for Co-60 in both Reference Drums

The configuration of RD16 used at site L1 did not contain Co-60, however the measurement of the Co-60 in RD15 at this site differed from the true activity by only 0.6% and was the closest of all measurements made so far.

With the exception of site L3, the results for RD15 are better than those for RD16. This is attributed to the homogeneity of the simulated waste within this drum. All the LRGS instruments checked used a simple mass correction for gamma attenuation, however, since there is a wide range of material density within this one drum (RD16) this can lead to errors in the quantification of the Co-60. A larger deviation from the true Co-60 activity was
anticipated at site L3 because a check source measured prior to the tests gave a result that was just outside the site's tolerance band. The site were however advised to proceed with the test.

The overestimation of the Co-60 in drum RD16 reported by some LRGS systems may be due to the Cs-134 in this drum. Cs-134 has gamma rays at 1168 keV and 1365 keV which are near to the gamma lines of Co-60 at 1173 keV and 1332 keV respectively which these LRGS systems may not be able to resolve. Even though the Cs-134 and Co-60 sources are in different layers of RD16 which were scanned by different detectors in the LRGS system, it is the sum of the spectra from the three detectors that is used to quantify the Co-60 in the drum.

![Figure 6 Co-60 Results for all Sites](image)

A comparison of the Co-60 results from all sites for drum RD15 is shown in Figure 6. The majority of the sites tested including those that measured this drum as part of the Round Robin Test slightly underestimated the Co-60 radioactivity, the major exception being site H4. In general the L sites (LRGS systems) performed slightly better than the H sites (HRGS systems).

5 Conclusions

A review of the potential for on-site checking of site operators' drum monitoring equipment was carried out at WQCL in 1998 under the Environment Agency's LLW monitoring contract. As a result of this review two drums of simulated waste have been prepared at WQCL. These standard waste packages form the basis of an ongoing programme of on-site intercomparison tests on site operators' waste drum gamma assay instrumentation. The use of reference drums containing defined radionuclides of known radioactivity allows the
Agency to assess the adequacy of operators’ arrangements for assaying drummed LLW destined for disposal at the BNFL Drigg repository.

A programme of site testing commenced in December 1999 and we have described some of the results obtained. The results have been used to compare and contrast the two main types of gamma assay instrumentation currently in use in the UK. Most of the sites visited as part of this programme relate measurements of key radionuclides in the waste to a wastestream fingerprint which is then used to infer the activity of the full range of alpha, beta and gamma emitting radionuclides in the waste drums. Site operators may have a number of these fingerprints relating to different areas on the site having LLW arisings. In recent years there has been a trend for site operators to move away from HRGS systems and towards LRGS systems for the assay of LLW. This offers a number of advantages i.e. the systems are robust and reliable and do not require cryogenic cooling, drums can be scanned in five minutes instead of one hour and they take up less room than the HRGS systems. The precision of the high efficiency detectors in determining e.g. Co-60 is good but relies on the accuracy of the operators’ wastestream fingerprint in characterising the waste consignment from this measurement. HRGS systems as operated by WQCL and most of the EN-TRAP partners that took part in the Round Robin Test have the disadvantage of being larger, more complex to operate and requiring longer count times due to the low efficiency of the Ge detectors. Their advantage, however is that they are able to detect a larger number of gamma emitting radionuclides in the waste and are therefore able to monitor for any unexpected components or variance in the wastestream at the operators’ site.

Where wastestreams are inhomogeneous errors can arise from differences in the attenuation of the emitted gamma rays compared with that calculated from a simple mass over volume density correction.

The programme to date has demonstrated the value of this type of intercomparison testing in the quality control of radioactive waste disposal. It is the Environment Agency’s intention to utilise the results obtained to date in order to refine the protocols for future tests. The Agency also intends to extend this programme to other areas of non-destructive testing in the future (i.e. neutron counting).

6 Acknowledgements

The authors acknowledge the support and cooperation of all the site operators included in this programme.

7 References

Non-Destructive Analysis
NON - DESTRUCTIVE ASSAY OF RADIOACTIVE WASTE PACKAGES

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Abstract

According to a relevant contract between the Austrian Government and the Austrian Research Centers Ges.m.b.H. ARC, all radioactive waste arising in Austria is treated, conditioned and interim stored at ARC.

In comparison with other countries without nuclear industry, ARC is well equipped with treatment and conditioning technology for Radwaste, including incineration, high force compaction, water treatment and storage facilities.

Ashes from incinerated waste, sludges from precipitation and other Radwaste suitable to be mixed with cement as to from concrete, was and in certain cases still is the option for conditioning.

Records of the results of such samples were examined recently. It was found that gammaspectroscopic measurements carried out in the past were based on a calibration system applying incomplete assumptions.

1 Introduction

In the fifties Austria had the intention to join the countries using nuclear energy for electricity production. Therefore, as a first step, a research organisation - Austrian Research Centers Seibersdorf (ARC) - was established in 1956 and a research reactor was built, sited at Seibersdorf, about 35km south of Vienna and was taken in operation in 1960. Two other research reactors were built in Vienna and Graz taking up operation a few years later. During the seventies a power reactor was built and ready for service in 1978, but after a referendum it never went in operation.

At the same time an agreement was signed between the Ministry of Health and Environment and ARCS to establish and operate a centralized facility for treatment, conditioning and interim storing of radioactive waste. It was finally built on the site of the ARCS. These facilities include a water treatment facility, an incinerator for the treatment of burnable Radwaste, high force compactor for non burnable material, cementing equipment for conditioning and interim storage halls for unconditioned and conditioned Radwaste.

Not all facilities were erected to the same time, there were different start up dates, for example the storage halls were built in the seventies, the incinerator was taken in operation in 1980 for a test phase and commissioned for radioactive material 1983 followed by routine operations with Radwaste since then. The high force compaction unit started operation in 1995.
Planning, building and operation of all facilities were carried out in accordance with the relevant regulations and laws in close cooperation with the licensing authorities. In the absence of a nuclear industry in Austria, only low and intermediate level radioactive waste, collected from medical, research and industrial applications of radioactive material is treated, conditioned and stored. Only Austrian's waste is collected and treated, with the exception of one project where contaminated ion-exchange resins from an Italian power plant were treated and conditioned. All sent back to Italy for interim storage there.

2 Radioactive Waste Management in Austria

As mentioned above ARC is acting for the country of Austria as a centralized facility for treatment, conditioning and storing of low and intermediate level radioactive waste. The following graph illustrates the process.

![Diagram](image)

*Figure 1: Scheme waste management in Austria*

A strict presorting is required and done by the producer. Transport is carried out to our facility according the relevant regulations. After arrival of the packages at ARC they are controlled and labelled. Measurements (Alpha-Beta gross counting and laboratory Gammaspectroscopic
assay) and checks are carried out with respect to the state of the material. Results of the measurements together with customer specific data are put into a data processing system. The next step after an interim storing in special storage buildings is segregation if necessary. Dependent on the category of the material - combustible or not combustible - a treatment process in the incinerator or the high force compaction unit is applied.

Ashes from the incinerated waste as well as special slurdes resulting from the precipitation of contaminated water are conditioned using cement. From each batch a 1 litre sample is taken for measurements. Pellets of compacted non combustible material (scrap, glass, PVC, dried precipitation sludge, filters, etc.) are loaded into 200l drums and measured with the drum scanner. Finally all drums containing conditioned waste are transferred into the interim storage building for interim storage.

Each step of the processes are accompanied by records carefully carried out and put into an electronic data processing system.

Additional tests are performed in some cases to investigate the leaching behaviour of conditioned material, samples for compressive strength tests are taken in order to ensure appropriate concrete quality and for special analyses an X-ray-fluorescence-analyser is available.

3 Available measurement systems for radiological assay

In order to be able to meet the requirements of QA/QC the following measurement systems are available and applied for radiological assay at ARC:

- Alpha-Beta gross counting
- Various Gammaspectroscopy systems as shown in the table below:

<table>
<thead>
<tr>
<th>System</th>
<th>Detector</th>
<th>Calibration</th>
<th>Samples</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lab. Gammaspec.</td>
<td>HPGe 47.5%</td>
<td>Empirical</td>
<td>Filter, Vol. Samples up to 1 litre</td>
</tr>
<tr>
<td>Drum Scanner</td>
<td>GeLi 7%</td>
<td>Empirical</td>
<td>Drums</td>
</tr>
<tr>
<td>ISOCS</td>
<td>BEGe 34%</td>
<td>Characterization</td>
<td>Multipurpose</td>
</tr>
<tr>
<td>Exploranium</td>
<td>Nal</td>
<td>Empirical</td>
<td>Mobile</td>
</tr>
</tbody>
</table>

4 Proceedings

It was discovered that gammaspectroscopic measurements were carried out in the past based on incomplete calibration assumptions. As a result of that, activity of packages may be considerably under- or overestimated, depending on the matrix characteristic and factors due to the measurement geometry.

At the moment a project to re-analyse samples of Radwaste conditioned in the past many years ago but still available, is being carried out.
As a first step a calibration standard for cemented sludges and ashes was created having a density of 1.8 g/cm³. It simulates conditioned ashes and sludges. The size of that sample standard is 1 litre, a volume typically used since such samples are measured using the laboratory gammaspectroscopic system. Measurements in the past were carried out using a calibration representing aqueous solutions with density 1.0.

A comparison of the efficiency curves obtained is shown on the graph below:

![Graph comparison efficiency curves density 1.0 vs. 1.8](image)

*Figure 2: Graph comparison efficiency curves density 1.0 vs. 1.8*

As a result the activity content was underestimated in the past by about 20% at high energies and up to a factor of 2.3 (²⁴Am) at low energies, as shown in figure 3 and table 2:

![Difference density 1.0 versus 1.8](image)

*Figure 3: Deviation due to different matrix density: 1.0 versus 1.8*
Table II: Underestimation of activity content in the past

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Underestimation of activity content [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{57}$Co</td>
<td>57</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>31</td>
</tr>
<tr>
<td>$^{60}$Co</td>
<td>23</td>
</tr>
</tbody>
</table>

As a further step 1 litre samples measured in the past were reanalyzed and the results were intended to be compared with results of the relevant drums. Those drums were generally not measured in the past due to lacking equipment. Because of the storing of drums in compact form only a few drums are accessible, but such drums are not from the past they are rather recently placed into the storage hall. It was decided that due to unacceptable handling efforts for sorting out specific drums from the storage hall - which is already full - and for radiation protection reasons, not to touch such drums now, but to take some drums from the hall presently in use.

A picture of the interim storage is shown below:

![Interim Storage Facility presently in use](image)

A comparison of measurements on recently conditioned material with associated 1 litre samples should result in an improved information on the radiological inventory of the old drums, when the still existing relevant samples are measured.

1 litre samples were analysed with the laboratory gammaspectroscopic system and also with ISOCS. The associated drums were measured with the drum scanner and with the ISOCS system. The results were compared.
A typical example, using key nuclides, is shown on the graph below:

![Graph showing typical results on a radiological content, drum EF9221](image)

Looking at the results of the 1 litre sample it is to observe in general a slight difference between laboratory gammaspectroscopy and ISOCS which is within the range of the uncertainty of the calibration and measurement.

Further it is to notice a considerable underestimation of activity considering the bar which represents the gamma scanner. Measurements were carried out assuming a calibration with density 1 as done in the past. But it is evident that a cemented sample requires a calibration using density 1.8. It seems to be clear that the activity content is considerably underestimated for this reason.

Another problem was identified and may be seen on that graph. Using this 7% GeLi and a measuring time of 3600 seconds the $^{241}$Am peak could not detect with the Drum Scanner.

The following tables show the results:

**Table III: Comparison lab. gammaspec. vs. ISOCS, 1 litre samples**

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Average Deviation Activity Conc. [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{60}$Co</td>
<td>+4.7 1)</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>-1.1</td>
</tr>
<tr>
<td>$^{241}$Am</td>
<td>+0.1</td>
</tr>
</tbody>
</table>

1) Including 1 sample where inhomogenous distribution of activity was identified.

**Table IV: Comparison lab. gammaspec. vs. gamma scanner**

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Average Deviation Activity Conc. [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{60}$Co</td>
<td>-28.5 1)</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>-37.8 1)</td>
</tr>
<tr>
<td>$^{241}$Am</td>
<td>-65.3 2)</td>
</tr>
</tbody>
</table>

1) 2 drums removed where inhomogeneous distribution of activity was identified by dose rate measurement.
2) 2 drums removed >MDA.
Figure 5 shows one of the drums removed from the average value:

Marked with the arrow a strong deviation can be observed by about a factor of 2 between 1 litre sample and drum measurement by ISOCS. The reason was a hot spot of $^{60}$Co activity, which was identified by manual dose rate measurement. The material investigated was conditioned sludge from precipitation.

Measurements on unconditioned precipitation sludges were carried out. Several unconditioned precipitation sludges were examined just in the same way carried out in the assay of the conditioned sludges.

A typical example is shown in the graph below:

Figure 5: Graph drum EF9236

Figure 6: Graph typical sample 287
The typical overestimation of the laboratory Gammaspectroscopy against measurement done by ISOCS (effective density in this case was 0.67) may be observed. Another observation is the strong overestimation of the drum scanner because of using incomplete calibration (inappropriate drum size, density 1.0).

5 Conclusions and plans

It is clear but not surprising that the use of correct parameters at radiological measurements are of great importance. Establishing a 1.8 g/cm^3 density sample for calibration purposes, representing conditioned ashes and sludges for laboratory gammaspectroscopy, improved the accuracy of information on the radiological inventory on conditioned radioactive waste stored at ARC.

It is intended to install a detector with higher efficiency for low level measurements of waste packages to replace the 7% GeLi which leads to more economic assay time. The future use of the GeLi for packages with high activity is possible and planned.

Extended investigations have to be carried out in order improve informations on the content of materials containing low energy gamma emitting nuclides. The difference on the $^{241}\text{Am}$ content measured in the 1 litre samples and the drum measurements carried out with the ISOCS system should be minimized.

Establishing appropriate calibration procedures for the drum scanner for drums containing conditioned waste by supercompaction is a further task to be worked on in detail.
Independent Quality Checking of UK LLW for Plutonium using the SCK•CEN Hexagon 2000 Passive Neutron Coincidence Counter

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Abstract

Checking low level wastes requires the detection of sub-milligram levels of plutonium. This is an extreme challenge for passive neutron coincidence counting as the levels of true signal are close to the background signal caused by cosmic ray interactions. The origin of cosmic-induced background is discussed.

We describe the Hexagon 2000 passive neutron coincidence counter that has been designed and built for waste quality checking of low-level wastes in the UK for the Environment Agency. The instrument uses computed neutron coincidence counting and improved filtering of high multiplicity cosmic-induced events. The instrument also correlates background signal with atmospheric pressure to further increase sensitivity.

Preliminary measurements of background data and of test drums containing small amounts of plutonium are described.

Keywords: Non-destructive assay; Waste assay; Neutron Coincidence Counting; Neutron Background

1. Introduction

The Waste Quality Checking Laboratory (WQCL) at Winfrith carries out independent monitoring of UK low level radioactive waste (LLW) disposals at BNFL’s Drigg facility in Cumbria. Recently the laboratory has acquired a passive neutron coincidence counter in order to monitor the plutonium contents of LLW in 200 litre drums.

The allowable level of plutonium in LLW destined for Drigg is 0.1 GBq/te plutonium alphas. For waste, at unit density and derived from a typical nuclear reactor, this corresponds to about 1.5 mg of plutonium per 200 litre drum, or about 0.6 mg $^{240}$Pu$_{eq}$, where $^{240}$Pu$_{eq}$ is the amount of $^{240}$Pu equivalent to the total mass of all the isotopes of plutonium in a drum.

Neutron coincidence counting has been commonly applied to the non-destructive assay of alpha bearing wastes and counts the coincident neutrons emitted by the spontaneous fission of the even numbered isotopes of plutonium. The real coincidence count rate, i.e. the coincidence count rate corrected for accidental coincidences, is proportional to the mass of fissile material. Unfortunately there is always a background neutron coincidence count rate caused by cosmic rays. Even in well designed instruments, this background count rate is comparable with the real coincident neutrons emitted by the weights of plutonium mentioned above.
Cosmic rays appear as showers of coincident particles and the direct detection of these results in a coincident count rate. In addition, spallation caused by the interaction of a cosmic particle with a nucleus of the materials of construction of the instrument or the waste drum may generate additional bursts of neutrons. In the past statistical treatment of the measurement data has been used to eliminate such bursts that result in high multiplicity events. To achieve this the actual measurement time is subdivided into a number of shorter measurements and the standard deviation of all sub-measurements is then used to reject outliers.

The SCK•CEN Hexagon 2000 instrument installed at WQCL uses computed neutron coincidence counting [1-4], which is a software approach to the conventional neutron counting and has a refined rejection filter for better performance. Further, the instrument is coupled to a pressure measurement that records the atmospheric pressure, as the cosmic ray induced background is known to depend on atmospheric pressure. This correction for pressure further characterises the variation of the background.

We report below the measured background rates for different simulated waste matrices and also measurements using sub-milligram sources of plutonium.

2. Neutron Background in a Passive Neutron Assay System

The coincident neutron background recorded in a neutron assay system originating from cosmic rays is two-fold. On one hand it is composed of the neutrons of a cascade that directly penetrate in the assay cavity, where they may thermalise and be detected. These neutrons are coincident in time. On the other hand, a nuclear cascade may be provoked inside the cavity giving birth to a burst of particles among which are many neutrons. This effect, known as "the multiplicity effect", was overlooked in the early stage of cosmic ray research where it was responsible for an over estimation of the flux. Which of the processes that occur is very much dependent on the energy of the incident particles.

2.1 Quantifying the Variation of Neutron Background

The intensity of cosmic rays at sea level varies with time. The fact that the background counts are not constant in time introduces an additional uncertainty in the background subtraction process. If these evolutions can be quantified and predicted then the uncertainty on the background can be decreased and, with that, the detection limits. Variations in the cosmic ray fluxes at sea level depend on [5,6]:

a) Atmospheric pressure.

b) Height of the main muon production level.

c) Extra-terrestrial factors.

a) The influence of the atmospheric pressure is easily understood. When the pressure increases the cosmic rays must penetrate a denser atmosphere. This results in a decreased flux. A linear relation can describe the relation between a pressure variation and the particle flux. For the neutron flux this relation is given by [6]:

\[ \Delta I = -7.20 \times 10^{-3} I_0 \Delta p, \]  

(1)

where \( \Delta I \) is the fractional change in neutron flux due to the pressure variation \( \Delta p \) and \( I_0 \) is the mean intensity at sea level.
A nuclear cascade process which produces neutrons can be provoked by muon capture. The muon flux also changes with pressure according to the relation:

$$\Delta I_\mu = -1.58 \times 10^{-3} I_{\mu 0} \Delta p,$$

where $\Delta I_\mu$ is the fractional change in muon flux and $I_{\mu 0}$ is the mean muon flux at sea level. The numerical constants in both equations have units of $\text{hPa}^{-1}$.

b) The main muon production occurs at a specific altitude. The flux of particles that reaches the earth’s surface is a function of the height through the survival probability of the muons. The mean height of muon production is not constant, but changes seasonally with an amplitude of 3 to 5 km. It is high in the summer and low during winter. The following relation describes this seasonal variation:

$$\Delta I_{\mu H} = -0.05 I_{\mu 0} \Delta H,$$

where $\Delta I_{\mu H}$ is the change in muon flux and $\Delta H$ is the deviation in kilometre from the mean muon production height corresponding to 100 hPa. The numerical constant has units of $\text{km}^{-1}$.

c) There is also an 11-year periodic variation of the cosmic ray intensity caused by the modulation effect of the solar activity.

Solar flares are accompanied by a decrease of the cosmic ray activity (Forbush effect).

### 2.2 Background Measurements using the Hexagon 2000

We used the passive neutron waste assay system Hexagon 2000 developed at SCK\_CEN to study the influence of the neutron background. The cavity of the Hexagon 2000 has a hexagonal section. Each side of the hexagon contains eight $^3\text{He}$ detector tubes with an active length of 1000 mm. The top and bottom slabs of the cavity contain detector tubes of active lengths 300 mm and 450 mm arranged in such a way that they fit in the hexagonal inner section of the cavity. The top slab contains 16 detector tubes and the bottom slab 14 detector tubes. All detector tubes are 25 mm diameter and are filled to 4 atmosphere $^3\text{He}$. The detector slabs are made of high density polyethylene and are composed of a 25 mm thick moderator slab, a 25 mm deep area where the detector tubes are positioned and a 150 mm reflector and shielding slab. No cadmium lining is used in the interior of the cavity. The volume averaged detection efficiency for an empty drum is 22%.

The software package NeuTICS was used to determine the real coincidence count rate using a time interval analyser (TIA). The TIA records and time stamps all pulses received from the Hexagon 2000 neutron detector. NeuTICS uses a rejection filter to reject high multiplicity bursts.

### 2.3 The Rejection Filter

The multiplicity rejection filter applied in the NeuTICS software uses a statistical $n$-sigma rejection algorithm. For this purpose the recorded pulse train is filtered by comparing the number of pulses in relatively small windows of length $W$ to the average value, $N$, plus a user-defined number, $n$, of standard deviations, $S$. For background measurements window widths of only a few seconds are used. The window width is determined by assuming that the uncertainty on the
number of pulses follows Poisson statistics and by computing the window width for which the average number of pulses equals a number of user-defined pulses. The width of a window therefore increases with increasing \( N \). We typically used windows that on average contain 15 pulses. The rejection level at which a window will be rejected when \( n = 3 \) is then 27. The filtering is applied by evaluating the filter criterion for the observation window starting with each pulse. If a window is rejected all pulses in this window are ignored and the evaluation continues with the next pulse following the rejected window. For each rejected window the measurement time is decreased by an amount equal to the window width.

For a pulse rate of the order of 3 n/s and for \( N = 15 \) the observation window is of the order of 5 seconds. This is significantly shorter than what is used in the more conventional approach where a measurement is physically subdivided into a series of shorter sub-measurements, typically lasting several minutes. Each sub-measurement is then analysed and the outliers are rejected following an \( n \)-sigma decision level. Filtering with shorter observation windows results in a more efficient detection of bursts caused by, for example, cosmic rays. Moreover, since such a burst typically lasts only about 100 \( \mu \)s, the rejection filter is more efficient in only rejecting the affected part of the pulse train. The better efficiency of the rejection filter with shorter observation windows is illustrated in Figure 1. This Figure shows an example of the rejection probability with a curve that corresponds to the required mean number of pulses, \( N \), used to define the width of the window, \( W \), in order to detect and reject a burst of \( i \) pulses added to a window containing \( N + 2S \) pulses. From Figure 1 it is clear that a burst of a small number of events can only be detected if the mean number of pulses is sufficiently small.

Figure 2 shows an example of filtered background as a function of the mean number of pulses used to define the observation window. The error bars represent the computed uncertainty (\( k = 2 \)). For small \( N \) the filtered real rate is lower, indicating that parts of the pulse train were rejected. For larger values of \( N \) the obtained filtered real rate seems to be stable. This shows that outliers can only be detected with the smaller values of \( N \).
2.4 Background Measurements

The filtered coincident background has been measured with 200 litre drums containing the following matrices:

- Polyethylene Shavings (density 0.15 g/cm³)
- Mixed Waste (density 0.5 g/cm³)
- Concrete Rubble (density 1 g/cm³)

The counting time was fixed at 4 hours for all measurements and data were collected for several days in order to accumulate data over a range of atmospheric pressures. The data obtained are shown in Figure 3, from which the pressure dependence is clearly observed. The regression lines through the three data sets are parallel with a slope of about 0.00076 (1/hPa). This value is smaller than the constants given in the equations 1 and 2 in Section 2.1.
Another feature of the data in Figure 3 is that the mean background rate appears to depend on the waste matrix in the counter. Figure 4 shows that this variation is, to a first approximation, a linear function of the mass of the drum in the cavity. Figure 4 was obtained by taking the mean values of the background real rate at 1013 hPa.

High-Z materials are well known to produce neutrons in spallation interactions with cosmic rays. From Figure 4 we see that other materials apparently also show this effect. One could argue that high energy neutrons are likely to be produced in the metal of the drum itself and that the moderation of the matrix enhances their detection. However, the polyethylene matrix and the mixed matrix have more or less equal moderating properties and yet a clear difference is observed between these drums. Therefore, we argue that appropriate background corrections in waste assay should take into account both the atmospheric pressure and the drum mass. However, given the limited number of matrices considered above, it is probably too early to generalize on these observations. Further measurements on other matrices are in hand.
3. Measurements Using Plutonium Sources

Detection limits were experimentally checked measuring plutonium sources with masses comparable with the predicted detection limits. For this purpose, a set of low-mass sources was obtained from the Institute for Transuranium Elements, Karlsruhe. These sources were placed in test drums containing the desired matrices and which were fitted with re-entrant tubes to accommodate the sources.

The data obtained by measuring the Pu sources in the three different matrices are shown in Figures 5 to 7. The data were obtained over a 4 hour measuring time. The sources were placed in a tube at 15 cm from the centre and at mid height of the drum.

It can be seen in all three cases that the data points lie above a line at three standard deviations above the background, which was taken to be the detection limit for these tests.

A further series of tests using eight hour counts on the matrices confirmed lower detection limits, as expected from improved counting statistics. Figure 8 shows the results obtained for the polyethylene drum.

![Figure 6](image6.png) 4 hour counts on Mixed Waste Drum using 1.0 mg $^{240}$Pu$_{eq}$ (Key as per Figure 5).

![Figure 7](image7.png) 4 hour counts on Concrete Drum using 0.7 mg $^{240}$Pu$_{eq}$ (Key as per Figure 5).
4. Conclusions

We have presented experimental data of neutron background data obtained with a passive waste assay system used on 220 litre drums filled with different matrices. These data were obtained using a new approach in waste assay by neutron counting that consists of the use of a rejection filter that acts in very short time sequences and a background subtraction that accounts for the variation of the neutron background with atmospheric pressure.

The proposed method of background subtraction is especially meaningful when relatively long measuring times are used to detect small amounts of plutonium. During such protracted measurement sequences, involving both background and actual measurements, variations of the atmospheric pressure are likely to occur. In the absence of such a correction the variation of background with pressure may be of the order equivalent to more than a milligram of $^{240}\text{Pu}_{eq}$.

We further observed that the background signal measured with a blank drum appears to depend on the drum's gross weight. Not accounting for this variation of the background will similarly result in a systematic over-estimation of the plutonium mass.

A broad quantification of the responses to the plutonium sources shown in Figures 5 to 8 indicates a detection limit between 0.06 and 0.2 GBq/te of plutonium alphas in simulated waste.

In this paper we have discussed managing uncertainties in neutron coincidence counting by taking into account atmospheric pressure and drum mass. Other uncertainties pertain to the technique, including any uncertainty associated with add-a-source matrix correction techniques and fundamental uncertainties with respect to an unknown source distribution. All such factors need to be taken into account in an overall uncertainty budget.
5. Acknowledgements

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6. References


WASTE CHARACTERIZATION METHODS AT BELGOPROCESS
AND THE IMPORTANCE OF NDA

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Abstract
Waste characterization in the end cycle becomes more and more important. Several methods are available for a radiological characterization: from copying the waste producers declaration over a calculation based on known characteristics or measured values to combinations of several techniques. The decision on what technique(s) to be used will be based on several criteria. One also has to evaluate at what stage of the waste treatment process the characterization has to be performed. Recently Belgoprocess has performed larger efforts and investments to assure a good waste characterization. These are concentrated in studies on historical and recent waste, resulting in isotopic vectors and the purchase of several NDA devices in order to cover the whole scale of waste the company treats.

The measuring results always need to be integrated with isotopic vectors.

1 Introduction
Governments and the nuclear sector realize more and more the importance of waste characterization in the end cycle. Its role is clear when one considers all aspects [1]:

- Legal: requirements of the entrusted agency (NIRAS/ONDRAF in Belgium) by the national authorities (full characterization of each waste package) as one of the acceptance criteria;
- Safety: not to exceed the exploitation permits of treatment installations and storage/disposal buildings;
- Economical: optimization of waste streams before the waste treatment (the correct waste type in the suited installation);
- Quality control: verify third party producers declarations;
- Social: information for future generations, especially in view to long term storage and disposal.

2 Methods for radiological characterization
The characterization of the waste can be performed at several stages of the waste treatment process (see Fig 1).

For example, the process of combustible waste:
- Samples of the waste are taken. This information is used to calculate the expected activity (1).
- The raw combustible waste is drummed and measured at the moment of reception (2).
- After incineration the ashes are sampled and the 200 l ash drums can also be measured (3).
- The drums are compacted and the pallets are put in a 400 l drum that can also be measured (4).
- Conditioning is done by pouring concrete in the drum and results in a new geometry (5).
- Before disposal (not operational in Belgium yet) a final control (6) can be performed.
Several techniques are available for a radiological characterization:
- Administrative: a copy of the waste producers declaration;
- Calculation: a calculation based on known characteristics or measured values such as dose rate;
- Sampling and measurement of alpha, beta or gamma totals or spectrometry;
- Destructive analysis of samples;
- Non-destructive analysis, such as active or passive gamma- or neutron spectroscopy, tomography, ...
- Combinations of those techniques.

3 Advantages and disadvantages of the used techniques

Each technique has his advantages and disadvantages in regard to the definition of the operational range based on the waste characteristics and the knowledge of them: solid, fluid or gas, size, material, homogeneity, activity distribution, dose rate, activity type, activity level, isotopic composition, chemical form, ...

Some typical dangers of applied methods are:

3.1 When using administrative techniques
Since the price of the waste treatment and disposal is function of the activity level, waste producers could tend to underestimate their declarations. Waste characterization is often expensive and requires investments in systems, manpower and knowledge. Especially small waste producers often don’t possess sufficient means and are not able to determine the activity of their waste with enough precision.

3.2 When using mathematical techniques
Mathematical techniques have two weak points: the way the correlation was derived and the quality of the input value. One has to take care that the reference points (especially when using empirical methods) are equal to the real waste. This means all parameters, such as isotopes, age, material, burn-up, ... need to be the same or compensated for. The range where one can expect the real waste to be in, should be covered.
with enough calibration data points. An independent verification using other techniques is required. Of course the errors valid for measurement techniques could have occurred in the determination of the basic data.

Choosing the input parameter is also important. Dose rates for instance are very sensitive for the matrix material and inhomogeneity and for the activity distribution. The dose rate will vary much with the location on the drum surface.

3.3 When using sampling

Sampling is the perfect solution for homogenous liquids. Taking samples from solid materials such as concrete blocks or metals is only allowed when one is sure about a fairly homogenous contamination. Piping and instrumentation have the risk of activity concentration at certain locations. For all waste great care has to be taken that the sampling is representative. A practical example is the drums filled with ashes coming from a waste incinerator. Knowing that the ashes are transported with a screw and that samples are taken at different heights and different radii, one could assume good representativity and limit itself to sample taking. Figure 2 shows that this is not so evident as it seems. This figure clearly indicates the presence of hot spots, causing a possible error of a factor 10 (proven practically).

Figure 2: Drum with ashes

3.4 When using NDA

The less parameter one has pre-knowledge about, the more risk of significant errors there will be. The main parameters are:

- The matrix characteristics: homogenous/heterogeneous, voids, fill height, internal shielding, chemical form, presence of radiation absorbing and moderating materials (boron), ...
- The activity distribution: homogenous/heterogeneous, lump effect, spatial distribution (radius and height), ...
- The isotopic composition: radiation type, energy, composition, one or several compositions, dominant isotopes, ...

Another practical example is shown in Figure 3, taken from [2]. In a drum containing γ-waste, a lead container with α-waste is hidden. Using gamma measurements, even with transmission sources, will never reveal this. The container itself can be detected with X-rays or tomography. The source can only be seen with advanced neutron measurements. This case can be found in real waste where for instance a piece of piping or machinery is contaminated and the radiation is shielded.
4 Looking for the optimal solution

From the above it is clear that the error made on the assay depends on the knowledge of the waste characteristics. The more unknown parameters there are, the larger the error will be. Before defining what method will be used, one should consider the need for precise knowledge and take into account the related cost.

The final choice of a suited method depends on several company and country depended parameters:

- The waste type:
  - The gamma and alpha activity
  - The presence of fissile and fertile isotopes
  - The isotopic composition (or the lack of knowledge about it)
  - The matrix composition (liquid/solid, absorbers, moderators, density, …)

- The required output:
  - Detection limit
  - Accuracy
  - Reports in mass or activity, isotopic specific or not

- The cost per measurement:
  - Number of packages to be assayed
  - Investment and operational cost
  - Return on investment

A first way to improve the result is the upgrading of basic systems. As an example we can take the case of gamma spectrometry on waste drums. The basic configuration would be an open geometry, thus a detector looking at the whole drum. Matrix effects can be compensated for by upgrading the system by making the drum rotate and by moving the collimated detector so that it only sees portions of the drum.

A second way is to combine systems into integrated combined systems or combine the information from several systems [3] Some examples:
Adding gamma measurements to a passive neutron system can, at high enough count rates, convert the measured Pu mass to an activity. The transmission results of an active neutron measurement can provide the necessary matrix corrections for the passive neutron measurement. Visual techniques such as X-rays and tomography can bring extra information about the matrix, but have no quantitative use yet.

No matter what technique is used, there still will be isotopes that cannot be measured, thus the end evaluation will always include the use of isotopic vectors. Ideally each measurement should be analyzed by a skilled operator, testing different assumptions. For companies dealing with vast waste streams this is not feasible. They should rely on a maximum automation, a good data keeping and logging, and a conservative approach in chosen the implemented algorithms and calculating the accuracy.

One notices that almost 30 years of research on active measurement systems has a limited effect on the industry. Due to the limited market for that kind of assay systems, innovations seldom are worked out completely nor applied in commercially available systems. A lot of parameters, having serious consequences on the assay results, still cannot be determined by the measurement systems. Lots of efforts have been done, and still can be done on the statistical treatment of data, the matrix corrections, multiplicity analysis, the imaging, the practical combining of different techniques (including visual techniques such as X-ray and tomography), ...

Alternatively, one can accept the error made. It is important to have a good idea about the possible error and its occurrence. Even state of the art systems can not 100% accurately characterizes unknown waste.

5 Radiological waste characterization at Belgoprocess

To our knowledge active neutron and combined systems are in Europe only used in industrial exploitation for the characterization of waste packages in the UK, France and Belgium. Other applications are found in safeguards and the weapon industry. Most other institutes will use a gamma system in combination with isotopic vectors, allowing large uncertainty on the isotopic results. The involved companies choose their system based on the national limits for geological disposal and shallow land burial.

Recently Belgoprocess, the Belgian waste treatment facility, has performed large efforts and investments to assure a good waste characterization. Lots of studies on historical and recent waste have been performed, resulting in isotopic vectors and several NDA devices were purchased in order to cover the whole range of waste this company treats.

Since no system or combination of systems is able to determine all present isotopes, the integration of the measuring results with one or more isotopic vectors is important. NDA can also contribute in determining which isotopic vector to select.

The figure 3 below shows the principle of radiological waste characterization at Belgoprocess: Based on the producer’s information, a vector is selected from a database of available and validated vectors. This vector will be decayed to compensate for the wastes age. Data from measurements such as NDA ($^{239}$Pu and $^{240}$Pu equivalent masses, isotope specific activities, total activities, …), dose rate measurements at several heights and angles, sample analysis and the beta over alpha ratio will be used to rescale the vector to the final one.

Then the soft will combine all information in the final report. The basic rule is that measurements are preferred over calculations.

The calculations are all performed within the Belgoprocess WCS (“Waste Characterization Software”) module.

Important is that each method, each vector, each measuring system and each calculation has to be validated by both the Health Physics Department and Niras/Ondraf.
6 NDA systems at Belgoprocess

Belgoprocess possesses over some state of the art assay systems and is in the process of purchasing more systems in order to cover all needed measurement types. QA-QC is implemented for each system to provide the necessary guarantee that the quality of the information complies with the applicable acceptance criteria and requirements. Each system is known by an acronym. An overview:

6.1 NDA-IPAN/GEA

The IPAN system is a highly automated system performing integrated active neutron, passive neutron, passive coincidence neutron and passive gamma measurements on 28, 150, 200 and 400 l waste packages [4]. The aim is to determine the Pu-content and the alpha activity. The so-called “imaging technique” reports the probable activity distribution in the matrix. The system can receive 24 drums at once and needs only a push-button start and some limited off-line analysis. It is being used in two modes:

a. With the original software (“BELGO”) for A2X, or alpha suspected waste. ISO 9001 certification was obtained for running the system in routine. External experts are now judging the validation of the integrated system for a very broad range of matrices and isotopic compositions. [5]

b. With new software (“PANCALC”) for A3X, or alpha contaminated waste. At higher count rates the IPAN BELGO software becomes unstable and certain detection elements get saturated. In order to overcome this, a new project was developed. New software was written that is based on the passive multiplicity data [7]. This was done for two types of waste: (a) non-combustible waste packed in baskets in 400 l drums and (b) combustible waste packed in 200 l drums. The range is from a few to several hundreds of grams of Plutonium. The system was constructed in 1992-1993. A new project (“REVIPAN”), now in its definition phase, is a total revision of the system. This revision will bring the system back to a state-of-the-art status. It aims at more modern algorithms, revising the mechanical and electronically components, add extra functionality, adapt to new operating systems, ...
A very rude and simple algorithm was developed to quickly get a graphical report on the distribution of waste and activity in the drum. This tool ("VISIPAN") will have limited use.

### 6.2 NDA-GaPu
The GaPu system is a gamma drum scanner in open geometry in order to obtain the Pu-content (grams level) with a very simple system and method. The current demands on accuracies for characterization are so high that there is little use for the system left.

### 6.3 NDA-HRA-Solarium1
This installation is in project phase. The system, probably passive gamma and passive neutron measurements, will be used to characterize conditioned waste at different activity levels. Effective start of the project is estimated to be mid 2003.

### 6.4 NDA-HRA-Solarium2
This device is related to the same main project and is in project phase too. The system, based on passive gamma measurements, will be used to characterize conditioned waste at different activity levels. The project group is now finalizing the contract. Delivery is planned in the summer of 2003.

### 6.5 NDA-LAVA(R)
The LAVA(R) unit will be realized end 2003 to measure low gamma activities in immobilized Ra-waste. Possibilities towards free release will be examined. It will be housed in 20ft container, allowing bringing the system to the waste instead of the waste to the system. This and the limited use of utilities together with the reduction of the controlled area will be cost effective.

### 6.6 NDA-HAVA
NDA-HAVA was a system used in a project to determine the Pu-content of high active material from head and top ends of fuel rods in tight baskets. After treatment of this unique waste, the system was dismantled and parts were used again in other applications.

Table 1 illustrates the compliance matrix with Figure 1. As one can see, it will be possible to carry out all needed characterization measurements at each process step.

<table>
<thead>
<tr>
<th>System</th>
<th>Waste type</th>
<th>Geometry</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
</tr>
</thead>
<tbody>
<tr>
<td>NDA-IPAN/GEA</td>
<td>Alpha</td>
<td>28l, 150 l, 200 l, 400 l</td>
<td>X</td>
<td>X</td>
<td>X</td>
<td>X</td>
<td>X</td>
<td>X</td>
</tr>
<tr>
<td>NDA-GaPu</td>
<td>Alpha</td>
<td>28 l, 200 l</td>
<td>X</td>
<td>X</td>
<td>P</td>
<td>p</td>
<td>p</td>
<td>p</td>
</tr>
<tr>
<td>NDA-HRA-Sol 1</td>
<td>Gamma + alpha</td>
<td>400 l, others maybe possible</td>
<td>p</td>
<td>p</td>
<td>X</td>
<td>X</td>
<td>X</td>
<td>X</td>
</tr>
<tr>
<td>NDA-HRA-Sol 2</td>
<td>Gamma</td>
<td>400 l, others maybe possible</td>
<td>p</td>
<td>p</td>
<td>X</td>
<td>X</td>
<td>X</td>
<td>X</td>
</tr>
<tr>
<td>NDA-LAVA(R)</td>
<td>Gamma</td>
<td>200 l, 150 l</td>
<td>X</td>
<td>X</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>NDA-HAVA</td>
<td>Gamma</td>
<td>Tight baskets</td>
<td>X</td>
<td>X</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

X: suited  
p: possible after extra calibrations

In order to ensure the correct handling of the data, great care has to be spent on QA/QC. That's why Belgoprocess applied for qualification/validation and ISO 9001.
7 Conclusion

NDA is important, but only a part of the techniques needed to correctly characterize nuclear drummed waste. The choice of the system will depend on many parameters and often a combination of different NDA techniques will be needed. The possible error will still be large. More advanced and integrated systems have far better results but are very expensive. Belgoprocess uses NDA techniques combined with destructive analysis and isotopic vectors. State of the art assay systems such as IPAN and transmission gamma spectroscopy are applied.

8 References

Characterization of Waste
Qualified Waste conditioning in New Waste Conditioning Facilities at FZJ

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Abstract
The Operation Division – Decontamination (B-D) at Forschungszentrum Juelich GmbH (FZJ) the Research Centre Juelich is in charge of the centre's waste management activities including radioactive waste treatment and interim storage. It is equipped with the necessary facilities.
During the past years the treatment facilities for LAW have been upgraded to meet current technical standards in radiation protection, to reduce final waste volume and to meet acceptance criteria developed for German final disposal sites and for long-term interim storage by Bundesamt für Strahlenschutz (BfS) the Federal Office for Radiation Protection.
Key item of the acceptance criteria are the detailed declaration of radionuclides and in case of the Konrad site of non-radioactive constituents, which are regarded as hazardous for ground water in the final waste package and the qualification of the waste conditioning processes.
Therefore
• the design of the new process lines considers nuclide detection,
• the process and sampling methods undergo BfS qualification,
• B-D contributes to an assessment program commissioned by BfS to identify toxic chemicals in radioactive waste products.

1 Introduction
The Decontamination Department (B-D) part of the Operations Division at Forschungszentrum Juelich GmbH (FZJ) the Research Centre of Juelich is in charge of the centre’s waste management activities including radioactive waste treatment and interim storage.
There are three main process lines for treatment of radioactive materials and conditioning of radioactive waste:
• dismantling and decontamination
• incineration of low-level radioactive solid and liquid waste
• evaporation and drying of low-level radioactive waste water and sludge

Figure 1 shows how the different process lines are linked together and their throughput per year working one shift per day.
Due to the amendment of the Radiation Protection Ordinance and to more stringent standards related with air pollution control almost the entire equipment for waste treatment had to be re-constructed. On this occasion they have been adjusted to new parameters, as there are the changes in waste generation, acceptance criteria for final disposal, current acceptance criteria for long-term interim storage and strict control on waste management. Furthermore cementation the process generally being used for waste immobilization in the past has been replaced by super compaction in order to reduce final waste volume and by this disposal costs.

In the case of the ERAM site, located at Morsleben the disposal fee had been 12,500 DM/m³, approximately 6,400 €/m³ this site has been closed in 1998. For a future site the disposal fee is expected to be two or three times higher.

The amount of waste to be incorporated in cement is limited for reasons of product chemistry. Therefore only about 60 kg of dry waste, i.e. salts or ashes, can be mixed into 200 litres of cement product, but up to 360 kg of super compacted dry waste fits into the same volume. Drying 74 tons of concentrates and sludge per year produces about 24 tons of dry waste. Super compaction of this amount saves about 1.5 – 4 millions Euro per year calculated according to the above given figures in terms of disposal fees.
2 Treatment of Burnable Radioactive Waste

Figure 2 shows the flow sheet of the new Juelich Incinerator, which is a combined incinerator for burnable solid and liquid radioactive wastes.

![Flow Sheet of Incineration](image)

Solid waste collected in 200 litre drums and HEPA-filters are fed into the preheated gasification chamber via glove box and lock chamber without further pre-treatment. The waste sink down to a pair of flaps, which serve as a grate and separate the gasification chamber from the primary burner. While sinking down the waste is heated up to about 800°C. At the grate level it is partly burned partly gasified by adding understoichiometric amount of air. Temperature and power control is performed by injection of water vapour into the gasification chamber. Additional amount of air is blown through the flaps into the primary burner where the temperature rises to 1100°C. The gases pass sideways to the secondary burner. The ashes fall down to be collected on a second pair of flaps where burnout is completed. Finally the ashes are discharged into 180 litre drums via cooling funnel. These drums fit into the super compactor.

The main advantage of this process developed at Juelich Research Centre is that gasification and burning are performed under very stable constant conditions. The temperature doesn’t oscillate and the off gas composition doesn’t either. The carbon monoxide content is very low. Comparatively small amounts of fly ash are generated.
The ceramic liner of the primary burner is worn by abrasion and mechanical stress. The flaps are made of heat resistant steel. An additional steel lining protects the primary burner walls.

The entire off gas line including secondary burner has been replaced to meet the requirements for air pollution control. Design of the new secondary burner allows residence time of two seconds at 1200°C.

Radioactive burnable liquid waste is directly being fed into the secondary burner. Behind the secondary burner the off gas enters a two-stage scrubber, acid and neutral stages, and cools down very quickly. The scrap solution is being neutralized precipitated and conditioned in the wastewater treatment line.

The cool saturated off gas passes are a moist activated carbon column in order to adsorb dioxins and other hazardous substances. It is an important safety feature of this type of column that moisture prevents possible ignition of the activated carbon.

The last stage of the gas purification line consists of two parallel HEPA-filters.

Significant advantages of the new off gas line are:

- Quick temperature drop in the scrubber prevents dioxin recombination.
- The activated carbon reduces the dioxin content in the purified off gas below the required limit of 0.1 nanogram per cubic meter.
- Outages caused by deposition and condensation of substances in the former tube bundle off gas cooler has been eliminated.

The incinerator nominal throughput is 50 kilograms per hour based on a 20,000 kilojoules per kilogram energy content of the waste. The yearly throughput is limited by the license to 140 tons put together of 120 tons solid waste and 20 tons liquids.

3 Treatment of Radioactive Waste Water

Radioactive wastewaters and sludge are converted into thermodynamically stable solids. The wastewater treatment line consists of storage tanks, evaporator and two stage drier.

The wastewater store houses 17 storage tanks with 825 cubic meters total volume. The large number of storage tanks allows the separate treatment of different wastes.

Wastewaters from different laboratories usually delivered in small portions often contain significant amounts of mercury. In these cases mercury will be precipitated prior to evaporation.

The feed is held in two parallel feed vessels. While being pumped into the evaporator, nitric acid is added to adjust the pH to 3 – 4. After evaporation has been completed sodium hydroxide will be added to the evaporator in order to neutralize the concentrate. Five tons of water are evaporated per hour until the solid content in the batch amounts to a maximum of 40 %. The distillate is pumped back into a clean vessel in the wastewater store. It can be recycled into the evaporator for further decontamination if necessary. The concentrated batch is released into a buffer vessel.
All kinds of radioactive concentrates, precipitates and sludge, for example also radioactive sludge from the ponds of the sewage treatment plant, will be dried and heat-treated in the two-stage drier as shown in figure 3.

Figure 3: Principle Flow Sheet of Drying

The main design characteristics of this unique equipment guarantee:

- homogeneity of the product
- constancy of product composition throughout processing
- representative sampling
- dry thermally stable compactable final product

In the 6 m³ supply vessel of the drier the feed will be homogenized by circulation and stirring to maintain homogeneity and constancy of product composition.

Samples are taken out of the supply vessel in order to obtain radiological data such as easily detectable main radionuclides and the scaling factor necessary for the calculation of the undetected radionuclides.

The homogenized feed enters the fluidised bed drier where 0.08 m³ per hour is evaporated. Heated air with its oxygen content reduced to 4% is both the fluidising and heating medium.
The dry granules produced in this stage will be heated to 350°C in the subsequent pipe-type fluidised bed heater. At this temperature residual organics and nitrates in the granules will react and be converted into the thermally stable final product. The final product passes through a cooler into the collecting vessel.

Unlike cemented dry concentrates, not cemented ones are thermodynamically unstable. Laboratory experiments have shown that a self-sustaining exothermal reaction starts as the product’s temperature exceeds about 200°C. This has been proved by thermal of non-heat-treated and heat-treated products.

Thermal stability is an essential for the acceptance criteria for final waste package which include the demand to resist 300°C. Consequently non-stable products have to be insulated against heat, for example by concrete so that the size and weight of the waste package are increased. Production of stable products avoid additional costs for vessel fabrication transport and final disposal fees.

Thermal stability is further important during the treatment process because frictional heat could ignite a thermally non-stable product in the super compactor.

4 Super compaction

Ashes and heat-treated dry concentrates will be super compacted using the FAKIR super compactor, which is operated by the company Gesellschaft für Nuklear-Service mbH (GNS) in the process building of B-D.

5 Packaging

The waste conditioning process ends with the packaging of the final waste products. Various types of standardized cylindrical or box-shaped containers are accepted by the German Federal Office for Radiation Protection (BfS) for final disposal. Consequently these packaging also will be used for the long-term interim storage. All waste packaging must meet general requirements:

- External dimensions, weight limits
- Stack ability
- Corrosion resistance
- Specified tightness

6 Quality assurance and quality control

Being responsible for the operation of the final repository for radioactive waste BfS set up quality standards for the waste products and packaging and requires detailed declaration of the radionuclides contained in the final waste package. This declaration is part of the waste package documentation submitted for approval prior to waste shipment to the final disposal site.

To ensure the required quality of the packaging only those are used which has been fabricated on the basis of a quality assurance program. Quality control of the containers comprises graded measures depending on the different requirements, in the scope of self-imposed control by the manufacturer up to external control by independent experts.
To ensure the required quality of the waste product the conditioning processes undergo a qualification procedure in agreement with BfS. All relevant according items are laid down in the process handbook. The handbook contains the chain of responsibility, characterization of the primary waste, process description, the classification of the waste packages, the waste form and packaging, the process instrumentation, the control measurements, the bandwidth and limiting values for process and product parameters, the beginning and end of a conditioning operation, faults and maloperations, malfunctions and a documentation of process parameters and waste package documentation.

7 Declaration of radionuclides

Identification of radionuclides listed in the acceptance criteria for disposal in the centre's waste is difficult and for some nuclides accurate identification is almost impossible. During the past versatile research program a mixture of waste charges from different institutes with very different nuclide compositions have been collected. The resulting nuclide composition is overlapped with the specific ones from the high temperature reactor AVR and from the uranium enrichment laboratory of URENCO.

The nuclide declaration is therefore a result of:

- Measurements on samples of the final product or on samples of interim products taken on key points of the process.
- Calculations on the basis of a number of scaling factors reflecting the origin and history of the particular waste batch.
- Plausibility control.

Prepared this way the nuclide declaration is valid as an average for a particular waste batch or for the collective of the resulting waste packages. It is quite accurate for the whole collective but might differ in a wide range for one individual out of this (see table 1).

Table 1: Comparison of measured (γ-scan) total γ-activity with total γ-activity values based on sample analysis

<table>
<thead>
<tr>
<th>Drum-No.</th>
<th>Drum Mass kg</th>
<th>Activity Bq Co 60</th>
<th>Activity Bq Cs 137</th>
<th>Activity Bq Sb 125</th>
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</thead>
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<tr>
<td>8700</td>
<td>153</td>
<td>7,07 E 5</td>
<td>1,78 E 6</td>
<td></td>
</tr>
<tr>
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<td>122</td>
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<td>8841</td>
<td>166</td>
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<td>9970</td>
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<td>4,57 E 7</td>
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<tr>
<td>∑ Activity γ-scan</td>
<td>-</td>
<td>4,99 E 8</td>
<td>4,18 E 7</td>
<td>6,88 E 5</td>
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<tr>
<td>∑ Activity Sample Analysis</td>
<td>7,33 E 8</td>
<td>5,0 E 7</td>
<td>3,77 E 6</td>
<td></td>
</tr>
</tbody>
</table>
The comparison between predicted data prepared by the above described procedure and measurement on waste drums shows the accuracy of the declaration.

Sampling procedure including sample preparation is due to qualification by BfS.

In case of incineration the samples are taken out of the drums filled with the ash resulting from one campaign. Three samples are taken out of each drum (see fig. 4) using a three-chamber sampling tube (see fig. 5) penetrating to the drums bottom. All samples are mixed to prepare a homogenous representative sample related to the particular campaign. A portion out this will be milled and analysed. The representative sample will be kept until the waste's shipment to the repository.

Figure 4: Sampling
Figure 5: Sampling tube

Regarding inorganic liquid waste the samples of the final dry granules are taken in the same way. But in contrary to the incineration there is an additional or alternative possibility to take homogenized representative samples out of the loop of the supply vessel.

At present sampling of the ash has been documented and demonstrated under the control of BfS and of an independent expert appointed by BfS. Approval and qualification are expected for the near future.

Qualification of the processes are under way whereby the optimum for control measures must be found and agreed upon with BfS. External control of the entire process is not necessary and must be limited to process parameters which have an influence on the relevant product quality.
CONDITIONING OF PLUTONIUM WASTE FROM MOX-FUEL FABRICATION FOR LONG-TERM INTERIM STORAGE

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ABSTRACT

In the period since Germany's experimental final repository ASSE was closed in 1978, around 5000 drums of conditioned plutonium-bearing radioactive waste from mixed-oxide (MOX) fuel fabrication have accumulated in the interim storage facilities of Siemens AG's MOX fuel fabrication plant in Hanau, Germany – formerly ALKEM GmbH, now Siemens Decommissioning Projects (Siemens DP). Another 5000 drums will arise in the course of decommissioning and dismantling the MOX plant which has now been underway for some months. The German Federal Government estimates that a geologic repository will not be needed for at least another 30 years. Therefore Siemens is taking the necessary steps to enable radioactive waste to be safely stored in aboveground interim storage facilities for a prolonged period of time.

Conditioning of MOX waste by cementation in drums was started in 1984. Permission to keep the drums in interim storage for a longer period of time in their current form would be extremely difficult to obtain as their corrosion resistance would have to be demonstrated for another 30 years.

The present goal is therefore to create a waste form suitable for interim storage which needs no maintenance over a long-term period, incorporates state-of-the-art technology and satisfy the requirements currently specified for final storage at the “Konrad” repository [1]. The goal can be attained by immobilizing the cemented waste drums in concrete inside steel “Konrad Containers” (KCs). The KCs themselves and the concrete backfill represent two further barriers which not only serve as radiation shielding but also protect the drums against corrosion as well as any possible release of radioactive materials in the event of accidents occurring during interim storage.

1 REQUIREMENTS TO BE MET BY WASTE PACKAGES

The requirements that must be met by the waste packages derive from the three purposes for which they are to be used; namely, interim storage, shipment and subsequent final storage. One common set of requirements applicable to all three is that the waste package must be structurally intact, must be free of contamination on its exterior surfaces, and must be unpressurized. Further, it must not exhibit any damage from corrosion and must, if necessary, be easy to decontaminate on the outside. In addition, mechanical and thermal loads must not lead to any release of radioactive materials.

Other requirements derive from the present necessity for long-term interim storage in aboveground facilities. To ensure that the containers will need absolutely no maintenance during interim storage, the provision made to protect the KCs on the inside and outside against corrosion...
must satisfy more stringent requirements than those applying for final storage. Furthermore, they
must not contain more than 15 g of fissile material per 100 kg of waste product in order that the
stipulations of Section 3 of the German Radiological Protection Ordinance (StrlSchV) for the
storage of "other radioactive substances" are met.

As soon as a final repository becomes available, the KCs will have to be shipped there. Hence, in
addition to the criteria described above, they also have to satisfy the requirements stipulated for
shipment as Class III LSA (Low Specific Activity) Material in an IP-2 industrial package [2] as
well as those associated with the safe transport of the waste packages within a final repository to
their location of emplacement.

With regard to the final storage, the waste packages must meet the acceptance criteria specified
for the Konrad repository [1].

2 FABRICATION OF WASTE PACKAGES

Fabrication of a waste package must comply with all of the above requirements. The packages
are manufactured according to manufacturing and examination sequence plans approved by the
German Federal Office for Radiation Protection (BfS). This ensures that they satisfy all criteria
for final storage at Konrad as well as for long-term interim storage. Compliance with the re-
quirements applicable for shipment of an IP-2 industrial package is monitored and confirmed by
German Lloyd and the authorized inspection agency as part of the type test carried out as per
ISO 1469, Part 1 [3].

2.1 Waste Containers

The waste containers comprise various models of type-tested steel containers certified as safe for
on-site transport operations within the Konrad repository. To prevent any buildup of pressure
inside the containers due to the formation of alpha radiolysis gases, the containers are equipped
with sintered metal filters that serve as an alpha barrier.

The steel containers are protected on the outside and the inside by special coatings which guar-
antee that they will need absolutely no maintenance during storage periods of at least 40 years.
The internal coating comprises a layer of polyurethane at least 3 mm thick. Those containers
which are to be filled with contaminated demolition rubble have an extra inner layer of polyure-
thane hard foam at the bottom which is at least 6 mm thick to protect them against mechanical
damage while they are being filled. The outside surface of the container is protected by a triple-
layer coating system that has a total film thickness of 240 μm and is easily decontaminable.

The polyurethane coating used for the liner has already demonstrated its long-term resistance to
corrosion and mechanical loading on sheet pilings in some German sea ports where it was ex-
posed over a period of many years to extremely aggressive conditions. This performance has
been confirmed in accelerated aging tests conducted at elevated temperatures which demon-
strated the coating's durability for at least 40 years.

For the protective coating used on the exterior of the KCs, the coating system manufacturer
guarantees that, provided the coating materials are applied properly, the KCs will be capable of
being stored in non-air-conditioned rooms for 30 years without suffering any deterioration.
2.2 Waste Products
The waste products normally placed in the KCs are 200-l drums containing waste that has been immobilized in cement or volume-reduced in a high-efficiency compactor. When the KCs are filled, care is taken to ensure that only waste coming from the same conditioning batch – or at least waste that has been subjected to the same conditioning technique – is packaged together. This permits all waste products packaged in any one KC to be assigned to the same waste product group according to the Konrad acceptance criteria. The actual number of drums packaged in a KC depends on the specific type of container and its design. In addition to meeting the requirements regarding waste product groups, care is also taken during filling of the KCs to ensure that the activity and fissile material limits specified per waste package for final storage at Konrad are likewise complied with.

2.3 Waste Packages
The waste drums are immobilized in concrete in the KC according to a sequence plan approved by the Federal Office for Radiation Protection. In a Type VI KC, for example, 10 drums are arranged inside the container in such a way that a layer of concrete at least 6 cm thick will be formed adjacent to the inside wall. The figure 1 below shows how the 10 drums are positioned inside this KC. Tests have verified that the waste package can survive being dropped from a height of 5 m onto a rigid plate without any radioactive materials being released from the drums. Thus one of the most important requirements for final storage at Konrad is fulfilled.

Figure 1: Waste Package with 10 Waste Drums Immobilized in Concrete inside a Type VI Konrad-Container

3 POTENTIAL SAVINGS IN FINAL STORAGE VOLUME THROUGH BACKFILLING WITH RADIOACTIVE MATERIALS
After the drums have been placed in the KCs, the containers still have a free internal volume of up to around 50% (depending on their specific design) which must be backfilled with concrete. The Konrad final storage requirements permit this otherwise wasted storage volume to be utilized for low-level waste. As long as the limit on total waste package activity has not already been reached with the drums alone, the concrete used for backfilling can be mixed with contaminated
liquids and/or radioactive noncombustible solid aggregates such as demolition rubble, slag or granulated metal. The mass fraction of aggregates in the concrete may total up to 25% without causing the compressive strength of the concrete backfill to be below its minimum specified limit. In this way it will be possible to dispose of all of the rubble arising during dismantling of the MOX fuel fabrication plant by placing it in KCs along with waste drums, thus preventing any need for additional final storage capacity for this refuse.

4 NEW INTERIM STORAGE FACILITY AT HANAU

The new storage building in which the conditioned waste is to be placed is built right next to the Siemens site in the industrial park of Hanau-Wolfgang. The building is now open for storage since October 2002. The building has been designed to accommodate a maximum of 1000 containers stacked in up to five tiers on top of each other. Transportation of the containers to the building via public roadways is not necessary. The building is 50 m long, 24 m wide and 17 m high. The storage building itself as well as the stacks of containers have been designed to retain their structural stability under seismic loading and is ventilated by natural convection.

Due to the fact that the waste packages will mainly contain alpha emitters (plutonium and uranium and daughters), the overall level of radiation inside the storage building will be only very low. The 30-cm-thick walls of the building are thick enough to guarantee a maximum annual dose of less than 1 mSv on the building exterior. Occupancy of the area outside the building is therefore unrestricted. The building itself is classified as a "monitored area" according to the Radiological Protection Ordinance.

The heavy weight and large dimensions of the KCs represent an additional safeguard against theft. For this reason, the only form of surveillance that will be needed is a burglar detection system with a direct link to the nearest police station.

Storage of the conditioned plutonium-bearing waste requires a license according to Section 7 of the Radiological Protection Ordinance. Interim storage is a purely stationary and maintenance-free operation. This means that apart from random visual checks of the containers' outer surface protection performed once a year, no other efforts or expense will be required for safe storage over a period of 30 years.

5 CONCLUSION

Radwaste conditioning based on the requirements specified for final storage at the Konrad repository provides a solution to current problems connected with the disposal of already existing waste and provides the prerequisites for safe long-term interim storage without any need for maintenance. By immobilizing this waste in concrete inside high-quality containers designed for long-term corrosion resistance, a waste package incorporating two further barriers is created which not only ensure the highest possible degree of safety during interim storage but also satisfy the requirements for final storage at the Konrad repository.
6 REFERENCES


   Series 1 Freight Containers – Specification and Testing – Part 1: General Cargo Containers for General Purposes
DETERMINATION OF NUCLIDE INVENTORIES IN ACCELERATOR RADWASTE

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Abstract
In Switzerland, the location of two of the largest accelerator facilities, CERN and PSI, the authorities requested for a valuation of the radionuclide inventories in accelerator radwaste. In the first phase, model calculations should be verified by radioanalytical analyses. At PSI, the radioactive contents were measured after a careful chemical separation, by γ-spectrometry, α-spectrometry, low-level counting and accelerator mass spectrometry, respectively. Examples: (i) The copper beam dump of Target E was analyzed; its activities were in the range between 1·10^3 Bq/g for ^60Co and 1·10^2 Bq/g for ^56Fe. (ii) In shielding concrete, more than 30 mBq/g ^239,240Pu were found which is higher than the exemption limit. (iii) In graphite targets, at end of bombardment 2·10^11 Bq/g ^7Be were detected. (iv) In an EU project, the know-how of the radiochemical separation procedures was used for determination of transmutation-relevant nuclear reaction cross sections.

1 Introduction

In Switzerland two of the largest European accelerator facilities are operated, CERN near Geneva and the Paul Scherrer Institute (PSI) in Aargovia Canton. In both centers, radioactivity is induced by interactions of accelerated protons and secondary particles (protons, neutrons, mesons etc.) with several materials (targets, beam dumps, mechanical parts, shielding materials). With regard to the future interim or final disposal of radioactive waste originated from these nuclear reactions, its nuclide inventory has to be determined. As used in disposal of nuclear power waste, the strategy will be to find out a nuclide vector, which allows to measure easily the amount of a few key nuclides but to determine the residual inventory, based on the measured data, by model calculations. On the other hand, this vector should differ remarkably from that of nuclear power waste, since the latter is formed by fission products mainly, while in the accelerator-induced waste fission products should play a marginal role only. In the first phase, these model calculations have to be validated by measurements, this means by radiochemical analyses of the waste nuclides. In the present paper, some examples of such radioanalyses are outlined, which were elaborated at Paul Scherrer Institute.

2 Beam Dump of Target E

2.1 History
In rotating Target E, consisting of graphite, α-mesons are produced by interaction of 590-MeV protons with light nuclei (carbon). The slightly energy-degraded beam was stopped in a water-cooled massive copper block of 4m x 1m x 1m ("beam dump"). It was planned, however, to increase the beam current up to 2 mA and to supply the neutron spallation source SINQ (at that time under construction) with this beam. In course of this constructions, the old beam dump must be decommiss-
The dismantling was performed in beginning of 1990. The beam dump was in use from 1973 to December 1989; the beam current was increased in this time stepwise up to 1.2 mA; the overall accumulated dose was assumed to be 5.95 Ah. The diagram of the beam dump is shown in Fig. 1. The arrangement consisted of the copper block mentioned above, a shielding plate of cast iron and some mechanical parts. As shown in Fig. 1, copper samples were taken from the proton beamspot about 2 cm below the surface (maximum activated part) and from a position ~ 1.5 m off, which was activated by secondary particles (No's. 6 and 9, respectively). Furthermore, a cast iron sample was taken from the shielding plate (No. 12); a stainless steel (No. 15) and a Steel 37 sample (No. 16) were taken from upper roof. After taking the samples, the beam dump was cast into concrete and stored in interim disposal place.

2.2 Experimental

The borings, taken from the metal samples, were dissolved in 7N HNO₃. Silver was separated by precipitation as iodide and purified. Then, from 6N HCl solution, iron (and antimony) were separated by extraction with di-isopropylether. The separation of tritium and ¹⁴C is described in a special paper in this conference [1]. By a special column technique (cf. below), plutonium was separated which can be seen as a representative for α-emitters. The chemical yield was determined by atomic absorption spectrometry (AAS).

The overall γ-spectra, taken before the chemical treatment, showed that ⁶⁰Co is the dominating radionuclide in any sample. Beside ⁶⁰Co, the nuclides ⁵²Na, ⁵⁴Mn and ⁴⁴Ti/⁴⁴Sc could be detected simultaneously in the same spectra without chemical separation. In the separated silver fraction, ¹⁰⁸ᵐAg was measured, likewise by γ-spectrometry. In the iron fraction, ⁵⁵Fe was measured by low-level counting (LLC), ⁶⁰Fe was measured by accelerator mass spectrometry (AMS) [2], and ¹²⁵Sb by γ-spectrometry. ³H and ¹⁴C were separately measured by LLC [1], and the α-emitters by α-spectrometry.

2.3 Results and Discussion

The results of the measurements are shown in Table 1. In order to get information about the further development of the radionuclide inventory, the corresponding data at time of "end of bombardment" (EOB) are given, and the data 200 years after EOB as well (if activity > 1·10⁻⁵ Bq/g). The activity data cover 12 orders of magnitude; their measurement consequently requests a palette of radioanalytical methods.

The dominating radionuclide is ⁵·₂₇·⁶⁰Co. Based on the measured data, the ⁶⁰Co content in the beamdump was estimated roughly to be 5·10¹² Bq at EOB. For the determination of the nuclide vector, it should be an useful key nuclide. In contrast to ⁶⁰Co, the activities of the other nuclides are remarkably dropping; by simple measurement they are not detectable beside ⁶⁰Co. On the other hand, ⁶⁰Co should not cause problems in final disposal; after 200 years it is dropped and should not play a role in the future waste composition.

The second class are the shorter-lived nuclides. They are useful for elaboration of the chemical separation schemes and for validation of model calculations as well. Additionally to the nuclides mentioned in Table 1, in earlier measurements traces of ²⁷²·d ⁵⁷Co and ²⁵⁰·d ¹¹⁰ᵐAg were found, but these nuclides are dropping rapidly, already to date their content is infinitely small.

The longer-lived radionuclides are important for final disposal. It seems that ⁴⁴Ti and ¹⁰⁸ᵐAg are the dominating nuclides in a foreseeable future, it should be looked out for these nuclides more precisely than done in the past. Particularly their behavior in living systems should be studied. It is obvious that extremely long-lived radionuclides like ⁹²Fe will become dominating in the far future; from this reason it is planned to extend the measurements to the nuclides ¹·₆·10⁻⁰·⁹¹Be, ⁷·₂·10⁻⁵·²⁶Al, ³·₇·10⁻⁴·³⁶Cl, ⁷·₅·10⁻⁴·⁵⁹Ni and ¹·₆·10⁻⁷·¹²⁹I. All these nuclides are measurable by AMS.
Table 1. Nuclide inventories in samples from beam dump of target E, calibrated to EOB (A),
July 1st, 2002 (B), and 200 years after EOB (C) (in Bq/g)

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Half-life</th>
<th>Measurement Method</th>
<th>Time</th>
<th>Sample 6 Copper Activated by Protons</th>
<th>Sample 9 Copper Activated by Secondary Particles</th>
<th>Sample 12 Shielding Cast Iron</th>
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<td>12.2 a</td>
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<td></td>
<td></td>
<td>C</td>
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<td>C</td>
<td>--</td>
<td>--</td>
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<td>--</td>
<td>--</td>
</tr>
<tr>
<td>$^{55}$Fe</td>
<td>2.7 a</td>
<td>LLC</td>
<td>A</td>
<td>$1.00\times10^8$</td>
<td>$4.05\times10^6$</td>
<td>$2.02\times10^6$</td>
<td>$2.62\times10^3$</td>
<td>1.56$\times10^5$</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>B</td>
<td>1055</td>
<td>43</td>
<td>8.17$\times10^4$</td>
<td>1.06$\times10^4$</td>
<td>6303</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>C</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td>$^{60}$Fe</td>
<td>1.5$\times10^6$ a</td>
<td>AMS</td>
<td>A</td>
<td>2.38</td>
<td>$6.51\times10^3$</td>
<td>$5.47\times10^5$</td>
<td>$1.06\times10^5$</td>
<td>$1.91\times10^5$</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>B</td>
<td>2.38</td>
<td>$6.51\times10^3$</td>
<td>$5.47\times10^5$</td>
<td>$1.06\times10^5$</td>
<td>$1.91\times10^5$</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>C</td>
<td>2.38</td>
<td>$6.51\times10^3$</td>
<td>$5.47\times10^5$</td>
<td>$1.06\times10^5$</td>
<td>$1.91\times10^5$</td>
</tr>
<tr>
<td>$^{60}$Co</td>
<td>5.27 a</td>
<td>Ge(Li)</td>
<td>A</td>
<td>$2.97\times10^7$</td>
<td>$5.73\times10^6$</td>
<td>$1.53\times10^5$</td>
<td>$3.05\times10^5$</td>
<td>5.89$\times10^5$</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>B</td>
<td>3432</td>
<td>663</td>
<td>$2.95\times10^4$</td>
<td>3.00$\times10^4$</td>
<td>1.91$\times10^5$</td>
</tr>
<tr>
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<td></td>
<td></td>
<td>C</td>
<td>$2.2\times10^5$</td>
<td>--</td>
<td>--</td>
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<td>--</td>
</tr>
<tr>
<td>$^{108}$mAg</td>
<td>418 a</td>
<td>Ge(Li)</td>
<td>A</td>
<td>27.2</td>
<td>0.76</td>
<td>$&lt;\text{d.l.}$</td>
<td>0.38</td>
<td>$\leq 0.55$</td>
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<td></td>
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<td>B</td>
<td>26.6</td>
<td>0.74</td>
<td>$&lt;\text{d.l.}$</td>
<td>0.38</td>
<td>$\leq 0.54$</td>
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<td></td>
<td></td>
<td></td>
<td>C</td>
<td>23</td>
<td>0.64</td>
<td>$&lt;\text{d.l.}$</td>
<td>0.33</td>
<td>$\leq 0.47$</td>
</tr>
<tr>
<td>$^{125}$Sb</td>
<td>2.77 a</td>
<td>Ge(Li)</td>
<td>A</td>
<td>91.3</td>
<td>4.0</td>
<td>1004</td>
<td>199</td>
<td>36.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>B</td>
<td>4.0</td>
<td>6.9</td>
<td>44</td>
<td>8.7</td>
<td>1.6</td>
</tr>
<tr>
<td></td>
<td></td>
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<td>C</td>
<td>--</td>
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<td>--</td>
<td>--</td>
</tr>
<tr>
<td>$\alpha$- emitters</td>
<td>sum</td>
<td>$\alpha$- spectrometry</td>
<td>A</td>
<td>$\leq 2.4\times10^2$</td>
<td>$\leq 3.5\times10^3$</td>
<td>$\leq 8.7\times10^3$</td>
<td>$\leq 5.7\times10^3$</td>
<td>$\leq 8.6\times10^3$</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>B</td>
<td>$\leq 2.4\times10^2$</td>
<td>$\leq 3.5\times10^3$</td>
<td>$\leq 8.7\times10^3$</td>
<td>$\leq 5.7\times10^3$</td>
<td>$\leq 8.6\times10^3$</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>C</td>
<td>$\leq 2.4\times10^2$</td>
<td>$\leq 3.5\times10^3$</td>
<td>$\leq 8.7\times10^3$</td>
<td>$\leq 5.7\times10^3$</td>
<td>$\leq 8.6\times10^3$</td>
</tr>
</tbody>
</table>

d.l. detection limit
Preliminary comparison with model calculations [3] gave indication that the bored sample from beamspot (No. 6) was not originated from exact center of beamspot but rather from its flank.

3 Determination of Plutonium in Shielding Concrete

3.1 Origin of the samples
Due to technical deficiencies, in December 1998 the target BX2 was dismounted and replaced by a newly constructed target. BX2 was used as a logging target for the 72 MeV beam, some meters behind the beam exit of the Injector II cyclotron. Every 10 minutes it has got the proton beam for some seconds in order to perform a measurement; subsequently the beam was shifted back into the main beamtube. Target BX2 was in use from the start of Injector II operation in 1985 up to November 1998. During this time, the proton beam current was increased from 100 μA up to 1.2 mA, the overall accumulated integrated current at BX2 was 1.93 Ah.

During the reconstruction, the opportunity was availed to take two samples of shielding concrete. The first sample was located in beam direction, 11 cm from target, behind 8 cm copper and 3 cm lead shielding (Sample 1). The second sample was a 20 cm long boring column of 5 cm diameter, taken 4 cm beside the beam direction, but shielded by 8 cm copper only. Its distance to the target was 12 cm. From this boring column, five samples were taken in distances of 5 cm each, vertically from the surface (Samples 2.1 to 2.5).

3.2 Experimental
First, the γ-Spectra of all samples were taken by a Ge(Li) Detector.

The separation scheme for uranium and plutonium is shown in Fig. 2 [4]. The concrete was fine pulverized in a mill. In order to detect the chemical yield, defined amounts of $3.75 \times 10^5$ a $^{238}$Pu and $68 \times 9$ a $^{232}$U carriers were added. The powder (~ 1 g) was disintegrated with 3-5 ml 40 % HF/4-5 ml 7N HNO$_3$. The solution was oxidized by H$_2$O$_2$, and plutonium and uranium were separated by two consecutive columns, BIORAD 1-X2 and UTEVA, respectively. The measuring samples were prepared by electrodeposition. The four nuclides $^{239,240}$Pu, $^{242}$Pu, $^{238}$U and $^{237}$U were measured by α-spectrometry in the respective fractions. The purity of each fraction was checked by γ-spectrometry using a Ge(Li) detector.

3.3 Results and Discussion
The radionuclide inventory of shielding concrete (Sample 1), measured by γ-spectrometry with a Ge(Li) detector and calibrated to July 1st, 2002, is shown in Table 2. The uranium and plutonium contents of Sample 1 likewise were included into Table 2. The uranium and plutonium contents of Sample 2, together with a blank (raw concrete) are shown in Table 3.

**Table 2: Nuclide Inventory of Sample 1, calibrated to July 1st, 2002**

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Half-life</th>
<th>Activity (Bq/g)</th>
<th>Nuclide</th>
<th>Half-life</th>
<th>Activity (Bq/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{152}$Eu</td>
<td>13.3 a</td>
<td>2756</td>
<td>$^{133}$Cs</td>
<td>2.06 a</td>
<td>321</td>
</tr>
<tr>
<td>$^{54}$Mn</td>
<td>312 d</td>
<td>2726</td>
<td>$^{40}$Sc</td>
<td>83.8 d</td>
<td>311</td>
</tr>
<tr>
<td>$^{60}$Co</td>
<td>5.27 a</td>
<td>669</td>
<td>$^{137}$Ba</td>
<td>10.5 a</td>
<td>16.4</td>
</tr>
<tr>
<td>$^{22}$Na</td>
<td>2.6 a</td>
<td>379</td>
<td>$^{239,240}$Pu</td>
<td>2.4 \times 10^7 a</td>
<td>0.0264</td>
</tr>
<tr>
<td>$^{154}$Eu</td>
<td>8.8 a</td>
<td>330</td>
<td>$^{238}$U</td>
<td>4.46 \times 10^7 a</td>
<td>0.0172</td>
</tr>
</tbody>
</table>
The radionuclides in shielding graphite are formed mainly by impurities; the europium nuclides and $^{46}$Sc from Rare Earths and their homologues, $^{60}$Co and $^{54}$Mn from iron, $^{23}$Na possibly from magnesium and $^{133}$Cs and $^{134}$Ba from barium. The amount of $^{152}$Eu is far beyond the exemption limit; the shielding blocks must be generally handled as radioactive materials.

The relation of plutonium in comparison to uranium is shown in Fig. 3. Despite the remarkable differences in the radioanalytical results, the quotient shows a closed line, nearly a straight one. This indicates a formation of plutonium from the uranium impurities, mainly by thermalized neutrons, but it shows also strong inhomogeneities of the uranium content in the sample. Even the $^{239,240}$Pu content is beyond the exemption limit, a respective strategy for deposit of Pu-containing materials should be elaborated.

4 Beryllium-7 in Graphite Targets

The Target E of PSI is used for production of intense meson beams. It consists of ultrapure graphite; the mesons are formed by interaction of high-energy protons with light nuclei, here with carbon. The target is a rotating wheel, it is described and shown in detail elsewhere [1].

Table 4. Radionuclide Inventory in Dismounted Graphite Targets, Calibrated to E.O.B. (Bq/g)

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Half-life</th>
<th>Name of Target</th>
<th>E 71 (3/96)</th>
<th>E 72 (9/96)</th>
<th>E 76 (2/97)</th>
<th>E 78 (12/98)</th>
<th>E 82 (4/01)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^3$H</td>
<td>12.3a</td>
<td></td>
<td>1.78·10^6</td>
<td>5.84·10^7</td>
<td>3.84·10^9</td>
<td>6.3·10^8</td>
<td>3.84·10^8</td>
</tr>
<tr>
<td>$^7$Be</td>
<td>53.3d</td>
<td></td>
<td>2.26·10^11</td>
<td>1.53·10^11</td>
<td>1.75·10^11</td>
<td>8.95·10^10</td>
<td>1.37·10^11</td>
</tr>
<tr>
<td>$^{14}$C</td>
<td>5730a</td>
<td></td>
<td>632</td>
<td>519</td>
<td>384</td>
<td>1749</td>
<td>357</td>
</tr>
<tr>
<td>$^{22}$Na</td>
<td>2.60a</td>
<td></td>
<td>39.8</td>
<td>76.2</td>
<td>193.8</td>
<td>271.2</td>
<td>943</td>
</tr>
<tr>
<td>$^{44}$Ti-Sc</td>
<td>60.4a</td>
<td></td>
<td>28.6</td>
<td>78.2</td>
<td>14.8</td>
<td>17.0</td>
<td>--</td>
</tr>
<tr>
<td>$^{46}$Sc</td>
<td>83.8d</td>
<td></td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>8.61</td>
</tr>
<tr>
<td>$^{54}$Mn</td>
<td>312d</td>
<td></td>
<td>1041</td>
<td>1.18·10^4</td>
<td>3690</td>
<td>7044</td>
<td>1790</td>
</tr>
<tr>
<td>$^{57}$Co</td>
<td>271d</td>
<td></td>
<td>284</td>
<td>2.4·10^4</td>
<td>7194</td>
<td>179</td>
<td>1.57·10^4</td>
</tr>
<tr>
<td>$^{60}$Co</td>
<td>5.27a</td>
<td></td>
<td>6.8</td>
<td>162</td>
<td>59</td>
<td>60</td>
<td>52</td>
</tr>
<tr>
<td>$^{88}$Y</td>
<td>107d</td>
<td></td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>2.52</td>
</tr>
<tr>
<td>$^{134}$Cs</td>
<td>2.06a</td>
<td></td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>2.38</td>
</tr>
<tr>
<td>$^{173}$Hf</td>
<td>1.87a</td>
<td></td>
<td>217</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td>$^{177}$Lu</td>
<td>1.37a</td>
<td></td>
<td>301</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
</tr>
</tbody>
</table>
The targets are cut into small pieces. The γ-spectra were taken by a Ge(Li) detector, without chemical treatment. The measured activity data are shown in Table 4. The activity values were calibrated to the time of E.O.B. ("end of bombardment").

The formation of \(^7\text{H}\) and \(^1\text{4C}\) in the graphite targets is discussed elsewhere [1].

The high amount of \(^7\text{Be}\) likewise is formed via direct reactions, nuclear reactions of the type of \(X(p,^7\text{Be})Y\), mainly via \(^{12}\text{C}(p,\alpha)^7\text{Be}\). Literature data [5] are in fair agreement with the data given in Table 4. We can assume that during the irradiation no amount of \(^7\text{Be}\) is released, all the formed nuclide remains in the graphite. The determination of \(1.6\times10^6\ \text{a} \ ^{10}\text{Be}\) will be carried out in future. Both beryllium radioisotopes are produced in amounts far beyond the exemption limit.

The heavier nuclides are formed in remarkably lower amounts; they are generally originated by impurities in graphite. On the surface of the target some sputtered metal traces are precipitated. Thus, the formation of \(^{25}\text{Na}\) may be interpreted as from an aluminum source, \(^{24}\text{Mn}\) and \(^{60}\text{Co}\) as from copper, \(^{172}\text{Hf}\) and \(^{173}\text{Lu}\) as from lead. The amount of \(^{88}\text{Y}\) came into the sample material during the handling of the dismantled target. Consequently, the reaction channels leading to the nuclides in the graphite target can be, in some limits, defined.

For the interim or final disposal, however, the graphite targets should not pose great problems. By storage of a few years, the amount of \(^{10}\text{Be}\) is allowed to decay, and the possible handling of tritium and \(^1\text{4C}\) is described elsewhere [1]. The role of the extremely long-lived \(^{10}\text{Be}\) is still open.

In a scientific cooperation project between CERN and PSI, it was tried to use the \(^7\text{Be}\) in the graphite targets as target material for measuring the cross section of the reaction \(^7\text{Be}(p,\gamma)^8\text{B}\), which is an important reaction in the "solar neutrino puzzle". It is assumed that via this reaction, in the sun the most higher-energy neutrinos should be formed. The active graphite was cut into small pieces and filled in a heating tube of a target corpus useful for producing a radioactive beam of the respective radionuclide.

With this equipment, a \(^7\text{Be}\) beam intensity of > 300 nA could be achieved, which is some orders of magnitude higher than the present "world record" for \(^7\text{Be}\) beams. ISOLDE could split the beam into \(^7\text{Be}, ^9\text{Be}, ^{10}\text{Be}\) and (highly enriched \(^7\text{Li}\)), in agreement with the expected relations. With a part of this activity, the requested cross section could be measured; it is shown in Fig. 4. The measured cross section data presently are presenting the most exact indication about this part of the solar neutrino puzzle. The details of the technical equipment [6] and the cross section measurement itself [7] are to be published.

5 Acquisition of Nuclear Data for Accelerator-Driven Systems (ADS)

An accelerator-driven system consists of the coupling of a high-energy intense proton beam (~ 1 GeV) with a spallation target and a subcritical reactor core. The proton beam that is incident to the ADS target (e.g. lead, lead-bismuth eutectics etc.) will create a large amount of spallation products with energies covering the full range up to the GeV region. Also other light ions as well as radioactive residual nuclei will be created in the spallation processes. The objective of this project is to contribute to the understanding and modeling of nuclear reactions in the 20-2000 MeV region, which is specific to ADS. The lack of concerning data is the largest barrier for detailed conception of transmutation processes and possible construction of equipment. In order to overcome this deficiency, we are measuring cross sections of the formation of long-lived residual nuclei via relevant proton and neutron induced reactions on (i) iron, (ii) lead, (iii) bismuth and (iv) uranium. Special attention is given to the determination of \(^{36}\text{Cl}, ^{29}\text{Al}, ^{10}\text{Be}\) and possibly \(^{127}\text{I}\).
For this purpose, these nuclei are separated and purified in preparation of AMS measurements (or low-level counting or \( \alpha \)-spectrometry, resp.). The chemical procedures necessary for the separation, are nearly identical to those used for analysis of radioactive accelerator waste.

As a model for ADS facilities, the target of the PSI spallation source SINQ may be useful. The present target is constructed as an experimental target which can be filled with different materials (Fig. 5). After the irradiation, the content should be analyzed with regard to the formation of the long-lived radionuclides. To date, the target is operated with a beam current of 1.7 mA of nearly 590 MeV protons for several months, possibly up to a year. These targets may cause the next big problem in handling radioactive waste at PSI.

6 Acknowledgments

The authors are grateful to Professor H. W. Gäggeler for his constant support of this work, Dr. H. E. Beer for excellent cooperation with regard to PSI radwaste, Professor R. Michel for cooperation in cross section measurements, Dr. Sabine Teichmann for providing the most samples, Dr. J. Eikenberg and Mr. S. Bajo for analysis of plutonium, Mr. F. Atchison for model calculations, Dr. U. Köster, CERN, for cooperation in the \( ^{7} \)Be field, and Mr. L. Wyer and Mr. S. Stallone for many technical help.

7 References

8 Figures

Figure 1. Dismounted beam dump of Target E with location of the samples

Figure 2. Flow-sheet of the separation procedure of plutonium and uranium from concrete
Figure 3. $^{239,240}$Pu in Relation to $^{238}$U in Shielding Concrete

Figure 4. Cross Section of the Solar Neutrino Reaction $^7$Be(p,$\gamma$)$^8$B (taken from [7])

Figure 5. SINQ-Target, schematically (above), photograph (bottom)
CHARACTERIZATION OF ALPHA LOW LEVEL WASTE IN 118 LITRE DRUMS BY PASSIVE AND ACTIVE NEUTRON MEASUREMENTS IN THE PROMETHEE ASSAY SYSTEM

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Abstract
This paper deals with the PROMETHEE (PROMpt, Epithermal and THErmal interrogation Experiment) waste assay system for alpha Low Level Waste (LLW) characterization. This device, including both Passive and Active Neutron measurement methods, is developed at the French Atomic Energy Commission (C.E.A.), Cadarache Centre, in cooperation with COGEMA. Its purpose is to reach the requirements for incinerating alpha waste (less than 50 Bq[a], i.e. about 50 pg of Pu per gram of raw waste) in 118 litre-«European» drums.

The PROMETHEE development and progress are performed with the help of simulation based on the Monte Carlo code MCNP4 [1]. These calculations are coupled with specific experiments in order to confirm calculated results and to obtain characteristics that can not be approached by the simulation (background for example). This paper presents the PROMETHEE measurement cell, its current performances, and studies performed at the laboratory about the most limiting parameters such as the matrix of the drum – its composition (H, Cl...), its density and its heterogeneity degree – the localization and the self-shielding properties of the contaminant.

1. Introduction
The development of alpha Low Level Waste (LLW) characterization methods is currently in progress at the French Atomic Energy Commission according to the French radioactive waste management program. Assay methods and systems are studied and developed in order to determine the best packaging and disposal ways for nuclear waste.

The purpose of this paper is to present the development, in cooperation with COGEMA, of an active neutron method-based, non-destructive assay prototype being able to reach the incinerated alpha LLW criterion. Criteria for alpha LLW are divided into three parts regarding to their activity level:

- drums containing between 185 MBq and 3.7 GBq [α] per ton of packaged waste (about 10 mg of plutonium after 300 years) can be wrapped and surface disposed of (note that a mean value of 0.37 GBq [α] per ton of packaged waste for the whole surface disposal, evaluated after 300 years, is also required);
- drums containing less than 185 MBq [α] per ton of packaged waste (about 0.5 mg Pu) can be compacted and blocked and then surface disposed of (evaluation at 300 years);
- drums with less than 50 Bq [α] per gram of crude waste (about 50 µg Pu) at the treatment date can be incinerated.

Passive and active neutron measurements of the radionuclides contained in 118 litre-«European» drums (460 mm in diameter, 750 mm high) may then allow us to identify alpha low level «incinerated-waste». Partial information are then available and may be complemented using other techniques (gamma-ray spectrometry, induced photon interrogation...).
Passive neutron assays involve the detection of spontaneous fission neutrons emitted by even nuclides \( ^{238}\text{Pu}, ^{240}\text{Pu}, ^{242}\text{Pu}, ^{242}\text{Cm}, ^{244}\text{Cm} \ldots \) and of neutrons resulting from \((a,n)\) reactions with light nuclides (O, F, Be...). The latter is conditioned by the presence of high \( \alpha \)-activity radionuclides \( ^{238}\text{Pu}, ^{240}\text{Pu}, ^{241}\text{Am} \ldots \) and light elements (depending on the chemical form of the contaminant). However, passive measurements do not lead to sufficient Detection Limits and won't be further develop in the present application.

The principle of the Active Neutron Interrogation method (according to the DDT (Differential Die-away Time) technique\[2,3\]), can be described in four phases:

- a pulsed, 14 MeV neutron generator emits neutrons at periodic intervals (repetition rate);
- neutrons are moderated and thermalized within the graphite and/or the polyethylene walls of an assay system. The neutron energy falls down to the thermal region \((\approx 0.025 \text{ eV})\);
- thermal neutrons induce fissions in the fissile nuclides \( ^{233}\text{U}, ^{235}\text{U}, ^{239}\text{Pu}, ^{241}\text{Pu} \ldots \);
- fast prompt and/or delayed neutrons resulting from thermal fissions \((\approx 2 \text{ or } 3 \text{ prompt neutrons per fission, one hundred times less delayed neutrons})\) are counted within the detection system.

In the time interval of counting, they are discriminated from the thermal interrogating neutron flux by cadmium and/or boron carbide filters. The detected signal is theoretically proportional to the fissile content of the radioactive waste drum.

Three measurement methods can be used: the total counting, in which all detected neutrons are taken into account, is the simplest. However, large uncertainties may occur, due to the chemical form of the contaminant or to the presence of moisture; the active neutron coincidence counting leads to the detection of pairs of neutrons; the active neutron multiplicity counting for the determination of time-correlated neutron groups. Results mainly depend on the generator neutron emission (to be maximized), the active background (to be minimized), the lifetime of the interrogating neutron flux in the irradiation cell (to be maximized), and the same parameters as in passive measurements.

A full description of the PROMETHEE (PROMpt, Epithermal and THErmal interrogation Experiment) waste assay system and the preliminary results obtained with four different matrices of interest and a neutron source intensity of \(1.6 \times 10^8 \text{ s}^{-1} \ [4\pi]\) are available in reference\[4\]. A brief description is given in paragraph II, and the last and best performances obtained by increasing the neutron source intensity by a factor of 15 are presented in paragraph III. Outcoming conclusions are finally discussed in the last paragraph.

2. Brief description of PROMETHEE 6

Over the last 15 years, the PROMETHEE cell is developed and improved step by step; the sixth version is currently in progress since 1996\[5\]. Figure 1 shows schematically the irradiation and measurement cell.

The general form is a \( L \times 1 \times H = 1900 \text{ mm} \times 1400 \text{ mm} \times 1600 \) parallelepiped mainly composed of high purity graphite. The six faces of the cell are covered with a 100 mm polyethylene shield.

The cavity cell is \( L \times 1 \times H = 550 \text{ mm} \times 550 \text{ mm} \times 900 \text{ mm} \) in dimensions. The graphite walls are covered with a 2 mm thickness of high purity aluminium. A turntable allows sectorial or uniform rotation measurements.

The usual neutron source is a 14 MeV, D-T neutron generator (GIN 26, SODERN) that can produce high level neutron intensities between \(3.7 \times 10^7 \text{ s}^{-1} \ [4\pi]\) and \(1.8 \times 10^8 \text{ s}^{-1} \ [4\pi]\), with a repetition rate of 125 Hz and a pulse duration of 15 μs. The generator is set up within graphite located in opposite to the door.
The PROMETHEE cell contains a total of 88 $^3$He-proportional counters divided into three detection units placed in the two lateral faces and in the door of the cell, at a distance of 100 mm from the measurement cavity. Each detection unit is composed of polyethylene covered with 1.5 mm Cd on the side towards the cavity, and 10 mm B$_4$C on the five other sides. An external bare counter, 150 mm active long and filled with $^3$He at a pressure of 6 bars, is used to monitor the generator neutronic emission.

![Graphite $^3$He, Neutron generator, Polyethylene, B4C carbide, Cd, $^3$He detector, Cadmium, Graphite](image)

**Figure 1** Schematic draw of the PROMETHEE 6 neutron waste assay system.

### 2.1 Electronics and data processing

The detector signals are amplified and shaped in 24 charge amplifiers (ACH, EURISYS MESURES). The ACH modules are placed close to the detector outputs, inside the irradiation hall. Each amplifier output is transmitted out of the irradiation hall through 50 Ω cables and fed to multichannel scalers (MCS, NOVELEC). The signal from the monitor is transmitted out of the irradiation hall through a shielded 50 Ω cable to an ADSF (EURISYS MESURES) amplifier module and then to a multichannel scaler. The detection of a signal is synchronized with the generator pulse and recorded between each sweep. Contributions of each irradiation-sweep are then added to improve counting statistics. The twenty-four measurement banks are connected to a passive and active coincidence or multiplicity counting card HISTOC using twelve inputs (NOVELEC, CEA patent). Data are recorded in a dedicated PC. Finally, a second PC equipped with a specific software, developed at the laboratory and using a Multi Channel Scaler, is dedicated to the total neutron counting.

### 2.2 Physical characteristics of the PROMETHEE cell

Table 1 presents the main characteristics of the PROMETHEE cell. According to the DDT method, the discrimination between the interrogating thermal neutron flux and the fast prompt neutrons from fission is allowed by using the cadmium/B$_4$C thermal absorbers wrapping the detection blocks. Furthermore, the existing large difference between the lifetimes in the cavity (1120 μs) and in the detection banks (19.4 μs) allows a clear distinction between the fast interrogating neutron flux and the fast prompt neutrons from fission. The detection efficiency (25.7 %) particularly contributes to good results in neutron coincidence and multiplicity counting. The total prompt signal is measured between 658 μs and 5600 μs after a 15 μs generator pulse with a 125 Hz repetition rate. Coincident prompt neutrons are detected between 1200 μs and 7284 μs after a pulse. These intervals were chosen in order to optimize detection limits. The coincidence gate is 72 μs wide.
Table 1 Main characteristics of the PROMETHEE 6 waste assay system.

<table>
<thead>
<tr>
<th></th>
<th>PROMETHEE 6</th>
<th>PROMETHEE 6</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lifetime in the detection blocks (μs)</td>
<td>19.4</td>
<td>19.4</td>
</tr>
<tr>
<td>Lifetime in the cell (μs)</td>
<td>1120</td>
<td>1120</td>
</tr>
<tr>
<td>Active detection efficiency (%)</td>
<td>25.7 ± 0.3 (1σ)</td>
<td>25.7 ± 0.3 (1σ)</td>
</tr>
<tr>
<td>Total passive background (s⁻¹)</td>
<td>1.35 ± 0.06 (1σ)</td>
<td>1.35 ± 0.06 (1σ)</td>
</tr>
<tr>
<td>Coincident passive background (s⁻¹)</td>
<td>0.117 ± 0.023 (1σ)</td>
<td>0.117 ± 0.023 (1σ)</td>
</tr>
<tr>
<td>Total active background (s⁻¹)</td>
<td>70.5 ± 0.4 (1σ)</td>
<td>70.5 ± 0.4 (1σ)</td>
</tr>
<tr>
<td>Coincident active background (s⁻¹)</td>
<td>0.095 ± 0.020 (1σ)</td>
<td>0.095 ± 0.020 (1σ)</td>
</tr>
<tr>
<td>Active net signal (²³⁹Pu eff) (s⁻¹.mg⁻¹)</td>
<td>12.2</td>
<td>12.2</td>
</tr>
<tr>
<td>Coincident active net signal (²³⁹Pu eff) (s⁻¹.mg⁻¹)</td>
<td>1.8</td>
<td>1.8</td>
</tr>
<tr>
<td>Passive background (s⁻¹)</td>
<td>1.35 ± 0.06 (1σ)</td>
<td>1.35 ± 0.06 (1σ)</td>
</tr>
<tr>
<td>Coincident passive background (s⁻¹)</td>
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</tr>
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<td>1.8</td>
</tr>
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</tr>
<tr>
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<tr>
<td>Coincident active net signal (²³⁹Pu eff) (s⁻¹.mg⁻¹)</td>
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<td>1.8</td>
</tr>
<tr>
<td>Passive background (s⁻¹)</td>
<td>1.35 ± 0.06 (1σ)</td>
<td>1.35 ± 0.06 (1σ)</td>
</tr>
<tr>
<td>Coincident passive background (s⁻¹)</td>
<td>0.117 ± 0.023 (1σ)</td>
<td>0.117 ± 0.023 (1σ)</td>
</tr>
</tbody>
</table>

3. Performances of PROMETHEE 6

The last and best performances are obtained by increasing by a factor of 15 the neutron source intensity, thus using the 14 MeV, D-T neutron generator GENIE 36 (SODERN) emitting about 2.4×10⁹ s⁻¹ with a repetition rate of 125 Hz, a pulse duration of 200 μs and an applied high voltage of 140 kV. The measurement time, mostly of 5 min, is extended to 15 min for low fissile mass samples. The total prompt signal is measured between 679 μs and 5782 μs after a 200 μs generator pulse with a 125 Hz repetition rate. Coincident prompt neutrons are detected between 1000 μs and 7126 μs after a pulse. The coincidence gate width is shortened from 72 μs to 48 μs to reduce the accidental coincidence counting rate.

Table 2 gives the characteristics of the plutonium oxide samples used in this study. The ²³⁹Pu eff parameter is the equivalent mass of ²³⁹Pu (equation 1) which should be used to give the same result as the current sample. It depends on the fissile isotopes of the PuO₂ sample (mass, thermal neutron cross section, v):

\[ M(²³⁹Pu eff) = \frac{6\%²³⁹Pu + 1.38\%²³⁷Pu}{100} M(Pu) \]  

This parameter is a convenient way to compare measurement signals of samples which don’t have the same isotopic composition. The self-shielding coefficients are evaluated using the MCNP Monte Carlo code [1] and the physical and chemical characteristics of the samples. Note that in the case of incinerated waste, less than about 1 mg Pu is researched and self-shielding effects should be negligible.

Figure 2 presents the variation of the total and coincidence net signals versus the effective Pu mass in a cellulose matrix of density 0.14 g.cm⁻³. Figure 3 shows similar results in a PVC matrix of density 0.253 g.cm⁻³. Each curve allows the determination of the following calibration coefficients (Table 3). Given uncertainties corresponds to 2σ.

Finally, these measurements lead to the evaluation of the PROMETHEE performances in terms of Detection Limit Mass (α and β risks taken equal to 2.5 %). The Detection Limit Activity is calculated considering the example of a plutonium isotopic composition of a PWR 17x17 nuclear fuel, having an initial enrichment of 4 % and a burn-up of 33000 MWd.t⁻¹ (cooling time of 3 years). Four configurations are given (Table 4) : the source is at the centre of the drum or homogeneously spread in it ; the counting is total or coincident. For the easier comparison of the Genie 36 performances, results relating to the GIN 26 neutron generator are also indicated.
Table 2 Main characteristics of the Pu samples.

<table>
<thead>
<tr>
<th>Isotopic composition (% weight)</th>
<th>Sample</th>
<th>Pu mass (mg)</th>
<th>(M(\text{Pu}_{\text{en}})(\text{mg}))</th>
<th>(M(\text{Pu}_{\text{en}})) uncertainty (mg)</th>
<th>Self-shielding coefficient</th>
</tr>
</thead>
<tbody>
<tr>
<td>238Pu</td>
<td>0.84</td>
<td>44</td>
<td>44.2</td>
<td>32.5</td>
<td>0.3</td>
</tr>
<tr>
<td>239Pu</td>
<td>66.34</td>
<td>43</td>
<td>18.0</td>
<td>13.2</td>
<td>0.2</td>
</tr>
<tr>
<td>240Pu</td>
<td>23.98</td>
<td>42</td>
<td>11.7</td>
<td>8.63</td>
<td>0.12</td>
</tr>
<tr>
<td>241Pu</td>
<td>5.29</td>
<td>41</td>
<td>4.53</td>
<td>3.32</td>
<td>0.04</td>
</tr>
<tr>
<td>242Pu</td>
<td>3.55</td>
<td>40</td>
<td>1.29</td>
<td>0.95</td>
<td>0.02</td>
</tr>
<tr>
<td>241Am</td>
<td>2.01</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>238Pu</td>
<td>1.30</td>
<td>F</td>
<td>8.312</td>
<td>6.034</td>
<td>0.016</td>
</tr>
<tr>
<td>239Pu</td>
<td>64.93</td>
<td>E</td>
<td>0.9194</td>
<td>0.6675</td>
<td>0.0015</td>
</tr>
<tr>
<td>240Pu</td>
<td>24.00</td>
<td>D</td>
<td>0.5570</td>
<td>0.4043</td>
<td>0.0018</td>
</tr>
<tr>
<td>241Pu</td>
<td>5.13</td>
<td>C</td>
<td>0.1870</td>
<td>0.1558</td>
<td>0.0016</td>
</tr>
<tr>
<td>242Pu</td>
<td>4.63</td>
<td>B</td>
<td>0.1305</td>
<td>0.0947</td>
<td>0.0015</td>
</tr>
<tr>
<td>241Am</td>
<td>4.85</td>
<td>A</td>
<td>0.0466</td>
<td>0.0338</td>
<td>0.0015</td>
</tr>
</tbody>
</table>

Figure 2 Total and coincidence net counting rate versus \(M(\text{Pu}_{\text{en}})\) in a density 0.14 g.cm\(^{-3}\), cellulose matrix.

Figure 3 Total and coincidence net counting rate versus \(M(\text{Pu}_{\text{en}})\) in a density 0.253 g.cm\(^{-3}\), PVC matrix.
Table 3 Calibration coefficients in a cellulose and a PVC matrix.

<table>
<thead>
<tr>
<th>CALIBRATION COEFFICIENT</th>
<th>Cellulose d = 0.14 g.cm⁻³</th>
<th>PVC d = 0.253 g.cm⁻³</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total counting</td>
<td>148.5 ± 3.6</td>
<td>2.94 ± 0.12</td>
</tr>
<tr>
<td>Coincident counting</td>
<td>15.54 ± 0.29</td>
<td>0.269 ± 0.012</td>
</tr>
</tbody>
</table>

Table 4 Performances of the PROMETHEE 6 waste assay system (DLM : Detection Limit Mass ; DLA : Detection Limit Activity).

<table>
<thead>
<tr>
<th>Matrix</th>
<th>Total counting</th>
<th>Coincidence counting</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>DLM (mg ²³⁹Pu)</td>
<td>DLA (Bq.g⁻¹)</td>
</tr>
<tr>
<td></td>
<td>DLM (mg ²³⁹Pu)</td>
<td>DLA (Bq.g⁻¹)</td>
</tr>
<tr>
<td></td>
<td>DLM (mg ²³⁹Pu)</td>
<td>DLA (Bq.g⁻¹)</td>
</tr>
<tr>
<td></td>
<td>DLM (mg ²³⁹Pu)</td>
<td>DLA (Bq.g⁻¹)</td>
</tr>
<tr>
<td></td>
<td>DLM (mg ²³⁹Pu)</td>
<td>DLA (Bq.g⁻¹)</td>
</tr>
<tr>
<td>Empty</td>
<td>0.11</td>
<td>---</td>
</tr>
<tr>
<td>Cellulose</td>
<td>0.096</td>
<td>0.12</td>
</tr>
<tr>
<td>0.144 g.cm⁻³</td>
<td>0.024</td>
<td>0.03</td>
</tr>
<tr>
<td>0.180 g.cm⁻³</td>
<td>1.5</td>
<td>0.53</td>
</tr>
<tr>
<td>0.253 g.cm⁻³</td>
<td>3.4</td>
<td>0.70</td>
</tr>
<tr>
<td>PVC</td>
<td>1.1</td>
<td>0.23</td>
</tr>
<tr>
<td>0.253 g.cm⁻³</td>
<td>0.18</td>
<td>0.19</td>
</tr>
<tr>
<td>0.264 g.cm⁻³</td>
<td>0.30</td>
<td>0.25</td>
</tr>
<tr>
<td>Metal</td>
<td>0.30</td>
<td>0.23</td>
</tr>
<tr>
<td>0.263 g.cm⁻³</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* Emission source: 2.4·10⁷ n.s⁻¹ in lieu of 1.6·10⁷ n.s⁻¹

Considering only those latter results (GIN 26), Table 4 shows important differences in the values according to the matrix composition. The best results are obtained in neutron coincidence counting, except for the EVA¹ (poly(Ethylene-Vinyl-Acetate)) matrix in which the high hydrogen density induce a decrease of the detection efficiency from 25.7 % (empty cavity) to 12.1 % (source at the centre of the EVA matrix). The minimum DLA is about 60 Bq.g⁻¹ at the centre of a cellulose matrix. A minimum effect is observed on the measured DLM in a cellulose matrix compared with an empty cavity because of the low H content and the absence of Cl. For three matrices (PVC, metal), the influence of the composition is less important in homogeneous distribution than in the case of a point source located at the centre of the drum. Furthermore, the large impact of the Cl content is illustrated by the variation of the DLM measured with the two PVC matrices : the DLM (and thus the DLA) values increase when the Cl content increases.

Using the GENIE 36 generator which is a neutron source 15 times higher than the GIN 26 one, the best results are now obtained in the total counting mode. This is due to the variation of the accidental counting rate in the coincidence background : when this rate is preponderant among the background components, the detection limit becomes approximately constant even though the neutron source intensity increases (saturation effect). This is not the case for the total counting detection limit which is inversely proportional to the square-root of the neutron source intensity (see equations 2 and 3 for a simplified detection limit calculation). Thus, the relative

¹ The EVA matrix is used for the same purpose as the PVC one but contains no chlorine.
performances of both counting methods are varying when the interrogating neutron source is modified.

\[ DL_{\text{Low}}(s^{-1}) = \frac{1}{t \cdot k_T \cdot (1 + 2t(B_a + B_b))} \]  
\[ DL_{\text{Cone}}(s^{-1}) = \frac{1}{t \cdot k_C \cdot (1 + \sqrt{2R_a + 4A_t + 2R_0})} \]

\( t \) is the time measurement; \( k_T \) and \( k_C \) are the total and coincidence calibration coefficient, respectively; \( B_a \) and \( B_b \) are the active and passive backgrounds in total counting; \( R_a \) and \( R_0 \) are the real active and real passive backgrounds in coincidence counting; \( A \) is the accidental background.

The detection limits are improved by using a higher neutron source intensity. In total counting, the gain is of about a factor of 4 in a cellulose matrix, and of 3.1 in a PVC matrix. In coincidence counting, these factors are 1.8 and 1.7.

The criteria for incinerating waste is reached with a GENIE 36 neutron generator in the case of the cellulose matrix. The chlorine content of some matrices unfortunately limits the potential performances of the method (e.g. PVC, \( d = 0.253 \text{ g cm}^{-3} \) matrix).

4. Conclusions

The PROMETHEE cell is a Passive-Active Neutron waste assay prototype developed for the determination of the fissile mass in 118 litre-«European» drums in the field of alpha-contaminated Low Level radioactive Waste. The aim is to reach the level of 50 Bq[α] (50 µg Pu) per gram of crude waste. Preliminary detection limits of less than 0.024 mg and 1.1 mg of effective \( ^{239}\text{Pu} \) at the centre of a cellulose matrix and a PVC matrix, respectively, can be currently measured (total counting, measurement time of about 15 min, neutron emission of \( 2.4 \times 10^9 \text{ s}^{-1} \)). Those values become 0.03 and 0.23 mg of \( ^{239}\text{Pu}_{\text{eff}} \) for a homogeneous distribution of the matter. The incinerated criteria is reached in the case of a non-absorbing matrix (e.g. cellulose), but further improvements will have to be made in the other cases. These envisaged studies are, for instance, the increase of the neutron source intensity (by the use of a new neutron generator or an electron linear accelerator), the improvement of the electronic modules for a better response to high neutron fluxes, the use of borated shields in front of the detection units or the reduction of the distance between the detection blocks and the measurement cavity.

Furthermore, the Active Neutron Interrogation PROMETHEE 6 waste assay system gives reliable performances as long as the hydrogen content and the heterogeneity of the measured matrices are not too important. The measured matrices must also be close to the calibration ones. Otherwise, several effects may lead to large uncertainties on the declared fissile mass: 1- the nature of the matrix, i.e. its composition, its density and its heterogeneity degree, and 2- the contaminant itself, whose composition (U/Pu), distribution in the matrix and self-shielding properties may have to be taken into account. Because of the strong interaction between all these effects, they can not be treated separately.

An algorithm for the reconstruction of the distributed mass using the Non Negative Least-Square (NNLS) method, applied to the different signals resulting from all detection banks, has been initiated. Assuming the hypothesis of a known matrix type, the algorithm allows the reduction of the uncertainties due to localization effects. Actually, in the case of PVC matrices in which the fissile mass is higher than three times the detection limit, the DLM can be reduced to less than 30%.

Furthermore, the variation of the measured signals with the matrix composition shows the need of a matrix correction algorithm. Most of the matrix correction studies published during the last 15
years present methods based on the use of flux monitors placed either inside the measurement cavity or near the assay system [6]. Moderation and absorption coefficients resulting from the analysis of the detected signals lead to the classification of matrices in families. Flux monitors may also lead to the determination of global correction methods that do not separate moderation and absorption factors.

Preliminary matrix correction studies in the PROMETHEE 5 assay system consisted in determining an absorption coefficient (ABS) using three proportional $^3$He, cadmium collimated flux monitors placed inside the cavity, superimposed on a vertical axis at one of the four corner of the cell [7]. This kind of method may lead to good results if calibration coefficients are well-known. Once a matrix is recognized, a global correction factor may be applied.

Other axes of research are also investigated at the laboratory (Ph.D. thesis) in order to study the potentialities of the neutron capture reaction ($n$,y) to identify and to quantify the main constituents of the matrix [8]. This method should be more sensitive than the previous one because of the use of the gamma-ray spectrometry allowing discrimination between components of interest (H, Cl, B...). The net gamma-ray areas are combined in a specific algorithm in order to correct the declared fissile mass.

5. References

[5] J. Romeyer Dherbey, C. Passard, A. Mariani, ENS class I meeting on research facilities for the future of nuclear energy, Bruxelles, Belgium (June 1996).
SELF-INDUCED X-RAY FLUORESCENCE : AN INNOVATIVE METHOD FOR URANIUM CHARACTERIZATION IN BITUMINIZED WASTE DRUMS

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Abstract
This article describes the feasibility study of a new non-destructive measurement method for uranium in bituminized radioactive waste drums. The principle consists in using the emission of fluorescence X-rays, uranium being irradiated by radioisotopes present in the drum, particularly $^{137}$Cs. This technique has two determining advantages. First of all, it is definitely more sensitive than the usual gamma-ray spectroscopy and therefore allows us to achieve a detection limit ranging between 0.25 kg and 1 kg of uranium (according to the drums) in approximately 300 kg of bituminous mix, with a measurement time of 20 minutes. In comparison, the detection limit of gamma-ray spectrometry is between several kilograms and dozens of kilograms for drum contact dose rates ranging between 0.01 and 0.3 Gy/h.

The second advantage is that it is a passive method which does not require any external source of irradiation in order to induce the fluorescence.

1 Introduction

Within the framework of the cleaning operation of UP1, the spent fuel reprocessing plant located in Marcoule, the CEA (French Atomic Energy Commission) has developed a measurement assay system for 225-liter radioactive waste drums [1]. These drums contain bituminized radioactive sludge resulting from effluent treatment. The aim of this system is to sort out the packages in order to determine precisely where they must be sent. The packages which meet the admission criteria of the French surface disposal site, at the Aube Center are to be sent there. The others will be dispatched to an interim storage site in Marcoule. The device consists of an active gamma imaging station, a gamma-ray spectroscopy station and a passive and active neutron measurement cell. This work was carried out for the CODEM, which is an economic interest group made up of the CEA, the EDF and COGEMA, in charge of UP1 dismantling operations and especially waste retrieval operations.

The total alpha activity of the bituminized waste drum must comply with a maximum acceptability limit for surface disposal at the Aube Centre. Since the total expected alpha activity in these packages is very close to this acceptability criterion, reducing the uncertainty associated to the quantity of its major components, plutonium and $^{241}$Am is very important. Plutonium is assessed by passive ($^{240}$Pu) and active ($^{239}$Pu) neutron measurements, and $^{241}$Am by gamma-ray spectroscopy with the 59.5 keV peak.

However, these packages contain uranium in variable and significant quantities (approximately a few kilograms) which leads to a great photon attenuation at low energy and therefore to high
uncertainties in $^{241}$Am quantification. Uranium also induces interfering signals in neutron passive and active assay, which disturbs plutonium quantification. Uranium characterization is thus of the utmost importance as far as measurement uncertainty reduction is concerned.

Unfortunately, standard gamma spectrometry measurement, based on the use of the 1001 keV gamma-ray of $^{234}$Pa ($^{238}$U radioactive decay tree), does not detect quantities of about a kilogram of uranium in the 20 minute time limit allotted to characterize each one of the sixty thousand bituminized waste drums present in Marcoule. In order to satisfy these requirements, we have presented a feasibility study performed by numerical simulation of a new measurement technique based on the assay of uranium X-rays. These radiations are due to the fluorescence induced by radioisotopes contained within the bituminized matrix, which explains the use of the term, "self-induced X-ray fluorescence method". We also describe an experimental validation of the numerical model, which uses the Monte Carlo transport code MCNP [2], based on the analysis of measurement results performed using 20 actual bituminized waste drums.

2 Principle of the method

The prevailing gamma emitter in these drums is $^{137}$mBa, the direct radioactive daughter of $^{137}$Cs, the main gamma-ray energy of which is 661.7 keV. Part of the low energy photons resulting from Compton scattering in the bituminous mix is absorbed in uranium by a photoelectric effect. The fluorescence X-rays thus emitted have characteristic energies allowing uranium identification. Providing we have:

- the activity of the fluorescence source, i.e. the $^{137}$Cs contained in the barrel, which can be determined by standard gamma spectrometry,
- the sensitivity coefficient $C_X$ linking the X-ray net area to the uranium mass and to the $^{137}$Cs activity (unit of $C_X$ is counts s$^{-1}$ GBq$^{-1}$ $^{137}$Cs kg$^{-1}$U)).

This parameter is firstly numerically calculated (feasibility study) and then validated experimentally.

The uranium is then quantified using the X-rays net areas.

3 Feasibility study by numerical simulation

3.1 Modelling

Using MCNP, the industrial gamma spectrometry device has been modeled in order to estimate the detection limit of the method in a measurement time of 20 minutes. The statistical convergence is improved (reduction of computing time) by splitting the calculation into two steps.

1. The first step consists in evaluating the photon flux at the measurement distance with the "Flux at a point detector" tally F5 of MCNP. A mono-energetic photon source of 661.7 keV is homogeneously sampled in the bituminous mix. The physical treatment includes the Compton scattering of source photons in the bituminized matrix, the photoelectric absorption of the low energy scattered photons in uranium, which is uniformly distributed in the matrix, and finally X-rays emission and transport outside the matrix to the detector point.

2. The second step consists in simulating the gamma-ray spectrum in the high purity germanium detector (planar design: 25 mm. in diameter and 13 mm. thick) using the "Energy distribution of pulses created in a detector" tally F8 of MCNP. A more realistic comparison between
calculated and experimental data can be achieved by the introduction of the measured energy resolution in MCNP calculations.

Thus we obtain the useful signal (uranium X-rays) and background noise (Compton continuum).

3.2 Detection limit of the method
The calculated detection limit of the method is found to be lower than 0.25 kg of uranium in a measurement time of 20 minutes, with a bituminous mix density of 1.3 and constituents with atomic numbers lower than 30. However, some drums can contain significant quantities (more than a kilogram) of barium (Z=56) or uranium (Z=92). These elements have a high photon absorption coefficient below 200 keV. Their effect is thus significant on the useful signal of fluorescence, but fortunately they also attenuate the Compton continuum background noise. Consequently, they are not too detrimental to the detection limit, which remains lower than a kilogram of uranium in the most absorbent bituminous mixes, but it is of prime importance that they be taken into account for uranium quantification.

3.3 Correction of the signal attenuation due to high-z elements
It clearly appears that the effect of the high-Z elements must be taken into account. For instance, Figure 1 shows a strong attenuation effect of uranium on its own fluorescence signal.

![Graph](image)

*Figure 1. Self-attenuation of uranium fluorescence signal (98.4 keV, Kα line).*

A correction method based on the analysis of the low energy Compton continuum of the gamma-ray spectrum [3, 4] can be applied. This continuum, which is partially due to scattering within the bituminous mix, is very sensitive, below 200 keV, to the content in high-Z elements because of their high photon absorption cross section. Its variation allows us to detect the presence of heavy elements and to correct their effect on the fluorescence signal. With the help of this correction, which is still under development, we hope to achieve a relative uncertainty of ± 40 % on the uranium mass assessed by self-induced X-ray fluorescence.
4 Experimental validation of the modeling scheme

In order to validate the previous calculations, 20 real bituminized waste drums have been modeled and the simulation results compared to the experimental ones. These drums have various densities, filling heights and chemical compositions. The measurements are carried out using a coaxial detector having a relative efficiency of 10%. Paragraph 4.1 presents the comparisons of the measured and calculated spectra. Paragraph 4.2 is an analysis of the possible causes of deviations.

4.1 Comparison of experimental and calculated spectra

Figure 2 shows the experimental vs. calculation comparison for drum #6 (mix density of 1.26, measurement distance and time : 5.3 m – 80,000 s).

<table>
<thead>
<tr>
<th>Drum #</th>
<th>Experimental / calculated X-ray net areas</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.03</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>0.95</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>1.16</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>1.12</td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>0.95</td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>1.24</td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>0.92</td>
<td>14</td>
</tr>
<tr>
<td>8</td>
<td>1.38</td>
<td>15</td>
</tr>
<tr>
<td>9</td>
<td>0.98</td>
<td>16</td>
</tr>
<tr>
<td>10</td>
<td>1.29</td>
<td>17</td>
</tr>
<tr>
<td>11</td>
<td>1.13</td>
<td>18</td>
</tr>
<tr>
<td>12</td>
<td>1.06</td>
<td>19</td>
</tr>
<tr>
<td>13</td>
<td>0.94</td>
<td>20</td>
</tr>
</tbody>
</table>

Table 1. Ratios between experimental and calculated net areas of the 98.4 keV uranium X-ray.

Visually, the calculated and experimental net areas of uranium X-rays seem to be in good agreement. Furthermore, one can notice the absence of some peaks in the calculated spectrum due to a simplified numerical model: 59.5 keV gamma ray of $^{241}$Am and fluorescence X-rays from the detector lead shield are not modeled. The most intense peak for uranium is the K$_{\alpha}$ line with an energy of 98.4 keV. Table 1 presents the experiment vs. calculation comparison of its net area for the 20 waste drums.
Experimental and calculated results are in good agreement regarding the uncertainties on the main input data of the numerical model: the density and the chemical composition of the bituminous mix, the uranium mass and the $^{137}$Cs activity. In particular, the chemical composition of the first six drums is known with much better accuracy than the others, as it was determined by radiochemical analyses (destructive method requiring sampling). The other drum compositions are extrapolated thanks to dates and production series.

4.2 Modeling uncertainties
Here we study the various possible uncertainties associated with the modeling in order to explain the observed differences between the experimental and calculated data.

4.2.1 Uncertainty concerning the bituminous mix density
The density is determined by a mere weighing of the drum and a $^{60}$Co gamma-ray radiography to measure the filling height and thus the volume of bituminous mix. The relative uncertainty associated with the deduced density is about 10% with a confidence level of 95%. This uncertainty impacts on two levels in our modeling.

First, any variation of the modeled density implies a variation of the self-induced fluorescence sensitivity coefficient $C_X$. This decreases as the density increases because of the absorption in the bituminous mix of part of the interrogating flux (i.e. the scattered photons originating from $^{137m}$Ba which induce fluorescence in uranium) and also part of the fluorescence X-rays.

Second, the density influences the $^{137}$Cs and $^{238}$U assay through traditional gamma-ray spectrometry, these nuclides being the basis radioactive input of the model ($^{238}$U quantification leads to the total uranium mass). Indeed, the determination of these activities requires a knowledge of the density for the detection efficiency calculation. Activities increase as density does (and conversely). As a matter of fact, the density rise induces a detection efficiency decrease, which raises activities since they are obtained by dividing the signal by the efficiency.

The uncertainty induced by the density on the calculated X-ray net area is lower than 10%, as shown in Table 2. This low impact can be explained by the opposite effect of density variations on the sensitivity coefficient $C_X$ and on the activities of $^{137}$Cs and $^{238}$U.

Table 2. Effect of density on calculated uranium X-ray at 98.4 keV for drums #1 and # 6.

<table>
<thead>
<tr>
<th>Density variation</th>
<th>Radioactive input data of the model</th>
<th>Calculated net area of the 98.4 keV X-ray</th>
<th>Radioactive input data of the model</th>
<th>Calculated net area of the 98.4 keV X-ray</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$A(^{137}$Cs) $A(^{238}$U) $\rightarrow$ M(U)</td>
<td>$A(^{137}$Cs) $A(^{238}$U) $\rightarrow$ M(U)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$-10%$</td>
<td>0.91</td>
<td>1.04</td>
<td>0.91</td>
<td>1.04</td>
</tr>
<tr>
<td>$+10%$</td>
<td>1.09</td>
<td>0.92</td>
<td>1.09</td>
<td>0.92</td>
</tr>
</tbody>
</table>
4.2.2 Uncertainty concerning the chemical composition

As shown in paragraph 3.3, high-Z elements have an influence on the net fluorescence signal. However, the attenuation effect of the other components in the bituminous mix (such as H, C, N, O, Si, S, Ca, Fe, ...) must not be ignored. In order to estimate the uncertainty due to variations of the chemical composition, we compared two simulation results. On the one hand, the drum #6 which is of well-known composition. On the other, a simplified composition using the correct uranium description and a mean value for the other components. The effect on the net area of the 98.4 keV X-ray is close to 10 %. This value constitutes only an estimate of the uncertainty due to the poor knowledge of the detailed chemical composition of the bituminous mix. Actually this uncertainty is difficult to assess accurately since compositions of the 60,000 drums are very variable.

4.2.3 Uncertainty concerning the drum filling shape

The filling shape is modeled as being a horizontal plane surface, but a retraction of the bituminous mix is sometimes observed during cooling, as shown in Figure 3. Here we study the influence of the filling shape on calculated results.

![Figure 3. Gamma-ray radiography (60Co) of drum #9 and MCNP model.](image)

As in the case of the previous study (density effect), taking into account the real shape of the filling leads to an opposite variation of the Cx coefficient and of the 137Cs and 238U activities. These compensated effects explain the low variation of the calculated net area of uranium X-ray, as shown in Table 3.

<table>
<thead>
<tr>
<th>Ratios between the parameters calculated with the actual filling shape and those calculated with the simplified plane shape</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A^{137\text{Cs}}$</td>
</tr>
<tr>
<td>0.95</td>
</tr>
</tbody>
</table>

4.2.4 Uncertainty concerning the radioactive input data of the model

In order to evaluate the total uncertainty on the activities of $^{137}$Cs and $^{238}$U, the following causes must be taken into account:

- simplifications of certain physical parameters (density, filling shape, chemical composition, etc.),
• uncertainty about the intrinsic efficiency of the detector (calibration measurement uncertainty),
• uncertainties about the measurements of the drums (counting loss corrections, extraction of the net area for the peaks presenting a low signal to noise ratio, etc.),
• uncertainty associated with gamma-ray intensities (basic nuclear data).

The total uncertainty concerning the activities of the \(^{137}\text{Cs}\) and \(^{238}\text{U}\) is then estimated to about 20% for each with a confidence level of 95%, which leads to an uncertainty of roughly 30% on the calculated uranium X-ray net area.

4.2.5 Uncertainty concerning the radioactive distribution

At the end of the effluent treatment process, the radioactive sludge is incorporated into the bitumen within an extruder to obtain a homogeneous mixture which is poured at hot temperature into the drums and where it spreads homogeneously. Radial heterogeneity is thus not very likely. However, it is possible to find axial heterogeneity since drums are filled in several different steps.

Therefore, we study here the effect of \(^{137}\text{Cs}\) and uranium axial heterogeneities in the case of drum #2. We model three bituminous mix layers (low, medium, high) of the same height. Uranium and \(^{137}\text{Cs}\) are distributed differently in each one of them according to the data acquired using two independent measurement methods. The first one is a collimated gamma-ray spectrometry performed at three heights, in the middle of each layer. The second one is a radiochemical analysis carried out on samples taken from each of the three layers. The relative activity levels and calculated X-ray net areas are shown in Table 4. The effect of these axial heterogeneities is less than 20% for this drum which is one of the most heterogeneous drums among the 20 measured.

Table 4. Effect of axial radioactive heterogeneities on calculated uranium X-ray at 98.4 keV.

<table>
<thead>
<tr>
<th>Homogeneous model</th>
<th>Heterogeneous model based on radiochemical analysis of samples</th>
<th>Heterogeneous model based on collimated gamma-ray spectrometry</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{137}\text{Cs})</td>
<td>(^{137}\text{Cs})</td>
<td>(^{137}\text{Cs})</td>
</tr>
<tr>
<td>U</td>
<td>U</td>
<td>U</td>
</tr>
<tr>
<td>High layer</td>
<td>0.33 0.33</td>
<td>0.15 0.46</td>
</tr>
<tr>
<td>Medium layer</td>
<td>0.33 0.33</td>
<td>0.35 0.27</td>
</tr>
<tr>
<td>Low layer</td>
<td>0.33 0.33</td>
<td>0.50 0.27</td>
</tr>
<tr>
<td>Ratio between 98.4 keV X-ray net area to the homogeneous model</td>
<td>0.83</td>
<td>1.11</td>
</tr>
</tbody>
</table>

4.2.6 Total uncertainty of the self-induced x-ray fluorescence method

The various sources of uncertainty studied in the previous paragraphs allow us to explain the fluctuations of the experiment vs. simulation ratio in Table 1 within a range of approximately ± 40%. This value also reflects the uncertainty level we want to reach for the determination of uranium by self-induced X-ray fluorescence. The modeling uncertainties evaluation shows that this goal can be achieved, provided the signal attenuation by high-Z elements is taken into account.
5 Conclusion

This study shows the feasibility of uranium quantification in bituminized waste drums using fluorescence X-rays induced by other radioisotopes present in the waste. The detection limit of this method, called "self-induced X-ray fluorescence", in a measurement time of 20 minutes, is estimated by numerical simulation to be ranging between 0.25 kg and 1 kg of uranium depending on the drums.

We also validated the numerical model used for the previous feasibility study by comparing calculated data to experimental data for 20 real waste drums. This allowed us to identify the main sources of uncertainty in the numerical modeling. It also enabled us to estimate the total relative uncertainty associated with the uranium mass determined by self-induced X-ray fluorescence to be at approximately ± 40 % with a confidence level of 95 %.

However, this value can be obtained only by taking into account the signal attenuation by the elements of high atomic number present in the matrix. We now propose to study a correction method based on the analysis of the Compton continuum of the gamma-ray spectrum [3, 4]. This work is now in progress with the aim of rendering this new measurement method operational before the industrial station is started up.

6 References


CHARACTERISATION OF RADIOACTIVE GRAPHITE FROM NPP DISMANTLING

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ABSTRACT

Vandellós I, a Spanish Nuclear Power Station that used natural uranium fuel, graphite moderator and CO₂ cooling, was definitely shutdown in 1990 and after performing its dismantling, its radioactive graphite had to be managed as radioactive waste and therefore characterised.

The radioactive graphite has different characteristics to other radioactive wastes due to its physical and chemical properties and also to the presence of $^3$H and $^{14}$C as mainly radioactive contaminants.

Taking into account the characteristics of radioactive graphite, a radiochemical methodology in order to characterise this radioactive waste has been developed.

In this paper the procedures to analyse high energy gamma-emitting radionuclides that are measured by direct gamma-ray spectrometry without separation; the radiochemical procedures to analyse pure beta-emitting radionuclides; low energy gamma and X-ray emitters and the most significant alpha-emitting radionuclides are shown.

In addition, the main data obtained together with the conclusions about the operational behaviour of the core graphite are presented.

1. INTRODUCTION

Vandellós I Nuclear Power Plant consisted of a gas cooled type reactor moderated and reflected by graphite and cooled by carbon dioxide. It had a nominal throughput of 460 MW, and produced about 56000 GWh from its first coupled to the grid on 6th May 1972 to the last operation day on 19th October 1989, when a fire in conventional installations and without radiological consequences occurred.

The Spanish Government declared Vandellós I in definitive shutdown in July 1990 because of the high cost involved in recovering the plant to modern security standards. The responsibility of the plant was transferred from the original owner (Hifrensa) to ENRESA for carrying out decommissioning activities after the unloading of the reactor, removing of nuclear fuel and conditioning and removing operating wastes.

Dismantling activities, until decommissioning Level 2, began in February 1998 and was scheduled for finishing at the end of 2002. As a consequence of those activities only the 20% of the site will remain as a regulated area including the reactor containment in a newly constructed
outdoors-protective structure. A latency period of 25 years is scheduled for the starting of the final decommissioning activities until Level 3.

Quite important amounts of radioactive graphite arise in this type of nuclear reactors both during normal operation (tubular sleeves associated to each individual fuel element) and during dismantling (structural graphite pile acting as reflector and moderator and containing control rod and fuel channels).

Vandellós I moderator graphite pile is a vertical cylinder, 10.2 m height and 16.6 m diameter with a total weight of about 2680 tons. The total amount of graphite sleeves used during Vandellós I operation were about 181000 with a total weight of about 1000 tons.

From the waste management point of view, it is important to identify and quantify nuclides present in graphite waste. For this purpose, CIEMAT has developed a radiochemical characterisation project within the framework of CIEMAT-ENRESA co-operation agreement where more than 200 powdered samples, taken during the conditioning activities of graphite sleeves, has been characterised and partial results were shown in a Spanish Nuclear Society Annual Meeting [1].

Previously to the end of the dismantling activities until Level 2, a drilling procedure was used for obtaining 58 cylindrical samples (50 mm height and 20 mm diameter) from graphite pile channels. Approximately all the samples were taken from channels located in the same semi-plane (between the central axis and the west part of the moderator graphite pile).

The aim of this paper is to present the determination procedures for characterising alpha, beta and gamma emitters in graphite samples and correlate the main results with the position in the graphite pile of the reactor.

Summarised results obtained from the radiological characterisation of these samples are given in this paper. Positions of the reference samples are shown in Table I.

<table>
<thead>
<tr>
<th>Height (m)</th>
<th>0.0 D</th>
<th>0.1 D</th>
<th>0.2 D</th>
<th>0.3 D</th>
<th>0.4 D</th>
<th>0.5 D</th>
</tr>
</thead>
<tbody>
<tr>
<td>7.7</td>
<td>G-13</td>
<td>G-21</td>
<td>G-29</td>
<td>G-37</td>
<td></td>
<td>G-57</td>
</tr>
<tr>
<td>6.3</td>
<td>G-12</td>
<td>G-20</td>
<td>G-28</td>
<td>G-36</td>
<td></td>
<td>G-56</td>
</tr>
<tr>
<td>5.1</td>
<td>G-11</td>
<td>G-19</td>
<td>G-27</td>
<td></td>
<td></td>
<td>G-55</td>
</tr>
<tr>
<td>3.9</td>
<td>G-2</td>
<td>G-5</td>
<td>G-10</td>
<td>G-18</td>
<td>G-26</td>
<td>G-34</td>
</tr>
<tr>
<td>3.3</td>
<td>G-9</td>
<td>G-17</td>
<td>G-25</td>
<td>G-33</td>
<td></td>
<td>G-53</td>
</tr>
<tr>
<td>1.5</td>
<td>G-1</td>
<td>G-4</td>
<td>G-8</td>
<td>G-16</td>
<td>G-24</td>
<td>G-32</td>
</tr>
<tr>
<td>0.4</td>
<td>G-7</td>
<td>G-15</td>
<td>G-23</td>
<td>G-31</td>
<td></td>
<td>G-51</td>
</tr>
</tbody>
</table>

Table 1. Positions and references of the samples extracted from the graphite pile
2. EXPERIMENTAL

Radioactive graphite presents special characteristics regarding to the radioactive waste characterisation process used in CIEMAT. Firstly the physical and chemical properties of graphite solid as well as powder that leads to develop special sample preparation processes and secondly the special radioactive content where majority components are $^3$H and $^{14}$C.

Cylindrical graphite samples were taken from the pile at several heights but in the same radial position. As it can be observed in Table I, most of the time two samples were taken from neatly the same position (e.g. at 0.2 D and 9 m).

For extracting a powdered aliquot from the solid graphite cylinders a special method was adopted. The method consists on milling the cylinder in such conditions that dispersion of powder and consequent contamination are avoided.

Sample preparations required special techniques for homogenising the graphite powder in the ribbed planchets for gross alpha-beta determination, the graphite powder was spread on a filter paper, put on the planchet, using a cotton steak. In this way the powder stand on the filter due to the static electricity forming a homogeneous thin film that both adjust to the calibration geometry and avoid self-absorption correction.

In order to get a stable and homogeneous counting geometry for direct gamma spectrometry a specific geometry based on a suspension of graphite powder in a gel formed from Instagel® plus water, conditioned in a liquid-scintillation counting glass vial was used. This counting geometry assesses an adequate sample homogenisation along the sample counting time.

First characterisation step performed on all samples involved gross alpha and beta activity determination by proportional counter; $^1$H and $^{14}$C separated by catalytic combustion oven and determined by LSC. High-energy beta-gamma emitters such as: $^{22}$Na, $^{24}$Na, $^{44}$Mn, $^{58}$Co, $^{59}$Fe, $^{60}$Co, $^{65}$Zn, $^{96}$Nb, $^{95}$Zr, $^{106}$Ru, $^{106m}$Ag, $^{108m}$Ag, $^{125}$Sb, $^{137}$Cs, $^{144}$Ce, $^{152}$Eu, $^{155}$Eu and $^{241}$Am were directly measured.

Second characterisation step was performed on some selected samples (G-5, G-13, G-16, G-23, G-30 and G-37). These samples were solubilised in acidic media and radiochemical processes for pure beta emitters: $^{36}$Cl, $^{41,43}$Ca, $^{6}$Ni, $^{85,90}$Sr and $^{241}$Pu beta-gamma emitters: $^{55}$Fe, $^{59}$Fe, $^{59}$Ni, $^{93m}$Nb and $^{94}$Nb and the most significant alpha-emitting radionuclides: $^{238}$Pu, $^{239,240}$Pu, $^{241}$Am, $^{242}$Cm, $^{244}$Cm and $^{234,238}$U were performed.

2.1 Determination of Tritium and Carbon-14

Samples of graphite powder were placed into a combustion oven where the sample combusts in a stream of oxygen gas at 900°C, it then passes the combustion products through a series of ceramic materials at 680°C that trap the volatile beta-gamma emitters present in the sample. Metals, salts and materials with very high melting points remain in the boat. The gaseous sample is passed through a section of teflon tubing where most of the $^3$H$_2$O vapour condenses on the walls. The gaseous products are then passed through a vial containing tritium trapping scintillation solution where the remaining $^3$H$_2$O vapour is condensed and trapped. The tritiated water droplets remaining on the walls of the teflon tubing are then washed down into the vial with tritium trapping scintillation solution.

$^{14}$CO$_2$ passes through the tritium vial without reacting with the tritium trapping scintillation solution. The gas is then bubbled through the vial containing carbon trapping scintillation solution where it reacts almost instantaneously with the solution. Both vials are then purged with
nitrogen to carry off any free oxygen that will produce a reduction in counting efficiency due to chemical quenching. Samples are then ready to be counted by liquid scintillation counting.

2.2 Determination of High Energy $\gamma$-Emitters

High-energy $\gamma$-emitters were determined by direct high-energy $\gamma$-spectrometry, without radiochemical separations, using a conventional system that consists of a MCA S-35 from Canberra System and a coaxial HPGe detector with relative efficiency 20%. Spectra were analysed using Spectran-AT program and the efficiency curves for the specific counting geometry were obtained with a QCY-48 standard solution from Amersham.

Some radionuclides such as $^{59}$Fe and $^{94}$Nb, need a specific radiochemical separation process, prior to their determination by $\gamma$-spectrometry although they are high-energy $\gamma$-emitters. The radioactive concentration of these nuclides in the samples was some orders of magnitude less than the concentration of the other $\beta-\gamma$ emitters whose emissions disguise $^{59}$Fe and $^{94}$Nb lines [2, 3]. The radiochemical separation of $^{59}$Fe is described together with $^{55}$Fe and the separation process of Nb is described below.

2.3 Determination of Pure $\beta$ and $\beta-\gamma$ Emitters

In order to analyse the non-volatile radionuclides one aliquot of the original graphite powder sample was dissolved by an acid treatment with $\mathrm{H}_2\mathrm{SO}_4$, $\mathrm{HNO}_3$ and $\mathrm{HClO}_4$ acid, obtaining a final solution that was prepared in order to obtain the solution in 4M HCl.

Iron-$^{55}$/59. Ferric ions are precipitated as hydroxides by ammonia after addition of known amounts of stable iron as carrier. The precipitate is dissolved in concentrated nitric acid. The final solution obtained is 0.5M HNO$_3$. The measured is carried out once the possible gamma interferences are checked by gamma spectrometry. $^{55}$Fe is determined by liquid scintillation counting by the measured of Auger electrons which are consequence of radioactive decay by electron capture. $^{59}$Fe is measured by gamma-ray spectrometry. The chemical yield is determined by spectrophotometry from the initial iron added as carrier.

Nickel-$^{59}$/63. Nickel is precipitated with dimethylglyoxime, after addition of known amounts of stable nickel as carrier for determination of the chemical yield by spectrophotometry. A liquid-liquid extraction of nickel with chloroform and re-extraction with 0.5M HCl is carried out. The final solution obtained is 0.2M HCl. $^{63}$Ni is determined by the measured of its beta emission by liquid scintillation counting once the possible gamma interferences are checked by gamma spectrometry. $^{59}$Ni is measured by X ray spectrometry with a planar Ge detector.

Strontium-$^{89}$/90. Strontium is separated by adsorption on an Eichrom Sr-Resin column after addition of known amounts of stable strontium as carrier and after conversion of the solution to the nitrate form. The rinses are performed with 8M HNO$_3$, 8M HNO$_3$-0.5M oxalic acid and 8M HNO$_3$. The elution of strontium is carried out with 0.05M HNO$_3$. The solution obtained is evaporated to dry, then it is added HNO$_3$ and again is evaporated to dry in order to be sure that all sample is in the nitrate form. Finally the residue is weighed and the weight corresponding to Sr(NO$_3$)$_2$ form is obtained in order to determine recovery of carrier. $^{89}$Sr is determined by the measured of its beta emission by liquid scintillation counting once the possible gamma interferences are checked by gamma spectrometry. Two counting are made, one immediately after the separation and the other one with a minimum time interval of 8-10 days to correct the influence of $^{89}$Sr.

Calcium-$^{41}$/45. Calcium separation is based on selective precipitation reactions by hydroxides, carbonates and chromates after addition of known amounts of stable calcium. At the end of the
radiochemical separation procedure the calcium carrier, together with the radionuclides of calcium, is precipitated as calcium carbonate, which is dried and weighed to determine recovery of carrier. Next, the precipitate is dissolved in hydrochloric medium in order to measure the Auger electron emissions, which are consequence of radioactive decay by electron capture, from $^{41}\text{Ca}$ and the beta emissions from $^{45}\text{Ca}$. The measured is performed once the possible gamma interferences are checked by gamma spectrometry, by dual label measurement in a liquid scintillation counter.

**Niobium-93m/94.** The radiochemical separation procedure of niobium is based on its selective precipitation as $\text{Nb}_2\text{O}_5$ in 4M HCl medium after addition of known amounts of stable Nb(V) as carrier. The precipitate $\text{Nb}_2\text{O}_5$ is dissolved by HF, forming the stable complex $\text{NbOF}_5^2-$. The chemical yield of the separation process is obtained by spectrophotometric measurements of the stable purple complex Nb-PAR. Then, $^{94}\text{Nb}$ and $^{93\text{m}}\text{Nb}$ were measured by high-energy and low-energy $\gamma$-spectrometry respectively.

**Chlorine-36 (Volatile radionuclide).** Chlorine separation is based in an oxidation technique performed in the graphite powder sample. The graphite sample is dissolved with HNO$_3$, HClO$_4$ and H$_2$SO$_4$ acid. The gasses formed during the dissolution pass through a series of NaOH bottles where chlorine (36Cl) and carbon dioxide (14C) are trapped. The basic solution obtained is neutralised in order to measure the 36Cl beta emissions by liquid scintillation counting. The measured is carried out by the dual label technique.

2.4 Determination of $\alpha$-Emitters

The determination by alpha spectrometry requires the separation of the $\alpha$-emitters from fission products, rare-earth and other elements that interfere in the preparation of the radioactive source and its measurement. All alpha emitters sources are prepared by electro-deposition and measured by a 450 mm$^2$ ion implanted silicon detector.

**Plutonium.** The separation of plutonium is performed by anion exchange chromatography. Fission products and rare-earth are eliminated washing the column with nitric-methanol. Americium and curium are eluted with HNO$_3$ and finally, the column is rinsed with HCl solution and plutonium is eluted with HCl/HI solution. To calculate the chemical recovery, $^{242}\text{Pu}$ is used as tracer.

**Americium and Curium.** The solution of americium and curium, obtained in the separation of plutonium, are extracted with 0.45M Di(2-ethylhexyl)phosphoric acid in n-heptane. Americium and curium are reextracted with HCl. Finally, americium and curium are purified by anion exchange chromatography. To calculate the chemical recovery, $^{243}\text{Am}$ is used as tracer.

**Uranium.** The separation of uranium from plutonium, americium, curium, rare earth and fission products is carried out by liquid-liquid extraction with TBP-n-heptane. Uranium is extracted and purified by anion exchange chromatography. To calculate the chemical recovery $^{232}\text{U}$ is used as tracer.

3. RESULTS

For the first step of characterisation some gamma emitters determined by direct $\gamma$ spectrometry shown all values under minimum detectable activity (MDA): $^{24}\text{Na}$, $^{54}\text{Mn}$, $^{58}\text{Co}$, $^{59}\text{Fe}$, $^{65}\text{Zn}$, $^{95}\text{Zr}$, $^{106}\text{Ru}$, $^{108m}\text{Ag}$, $^{110m}\text{Ag}$, $^{125}\text{Sb}$, $^{144}\text{Ce}$ and $^{152}\text{Eu}$. 
The results of characterisation, in terms of maximum and minimum activity determined, average specific activity and their standard deviation as well as the ratio of data up to minimum detectable activity are collected in Table II.

Specific activity variation in function of height from the pile base and approximately distance from the pile centre in each vertical section is plotted in figures 1 to 4 for \(^3\)H, \(^{14}\)C, \(^{60}\)Co and \(^{137}\)Cs. Data from double sample at the same position (same distance to the centre and height) are represented as average value of specific activity.

**Table II.** Ranges of specific activity (Bq/g) up to MDA from nuclides determination at 1st Characterisation step

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>&gt; MDA (%)</th>
<th>Maximum (Bq/g)</th>
<th>Minimum (Bq/g)</th>
<th>Average (Bq/g)</th>
<th>STD (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^3)H</td>
<td>100.0</td>
<td>1.69E+06</td>
<td>2.30E+04</td>
<td>3.57E+05</td>
<td>79.3</td>
</tr>
<tr>
<td>(^{14})C</td>
<td>100.0</td>
<td>1.56E+05</td>
<td>8.54E+03</td>
<td>7.10E+04</td>
<td>54.5</td>
</tr>
<tr>
<td>(^{22})Na</td>
<td>65.5</td>
<td>7.45E+02</td>
<td>3.24E+01</td>
<td>2.19E+02</td>
<td>73.9</td>
</tr>
<tr>
<td>(^{60})Co</td>
<td>100.0</td>
<td>9.79E+04</td>
<td>1.25E+02</td>
<td>2.08E+04</td>
<td>108.0</td>
</tr>
<tr>
<td>(^{64})Nb</td>
<td>6.9</td>
<td>1.65E+02</td>
<td>2.38E+01</td>
<td>7.77E+01</td>
<td>70.5</td>
</tr>
<tr>
<td>(^{133})Ba</td>
<td>62.1</td>
<td>1.01E+03</td>
<td>3.34E+01</td>
<td>3.28E+02</td>
<td>73.2</td>
</tr>
<tr>
<td>(^{134})Cs</td>
<td>50.0</td>
<td>1.18E+03</td>
<td>6.09E+01</td>
<td>3.00E+02</td>
<td>67.7</td>
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<tr>
<td>(^{137})Cs</td>
<td>44.0</td>
<td>3.83E+03</td>
<td>1.33E+01</td>
<td>4.20E+02</td>
<td>179.7</td>
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<td>(^{154})Eu</td>
<td>22.0</td>
<td>1.76E+03</td>
<td>2.64E+02</td>
<td>7.54E+02</td>
<td>51.6</td>
</tr>
<tr>
<td>(^{158})Eu</td>
<td>35.0</td>
<td>7.15E+02</td>
<td>9.27E+00</td>
<td>2.17E+02</td>
<td>66.7</td>
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<tr>
<td>(^{241})Am</td>
<td>26.0</td>
<td>1.04E+02</td>
<td>3.30E+00</td>
<td>2.33E+01</td>
<td>94.5</td>
</tr>
</tbody>
</table>

**Figure 1.** \(^3\)H specific activity distribution
Figure 2. $^{14}$C Specific activity distribution

Figure 3. $^{60}$Co Specific activity distribution
The evolution of activity along two vertical lines (0.1 D and 0.4 D) are represented in figure 5 for the main 5 nuclides ($^3$H, $^{14}$C, $^{60}$Co, $^{137}$Cs and $^{241}$Am).

The same representation for samples taken at two pile heights (3.9 m and 9.0 m) along the radius is shown in figure 6.
Six samples were selected for performing the second step characterisation process. These samples were G-5, G-13, G-16, G-23, G-30 and G-37.

The specific activity determined for $^{56}$Fe, $^{59}$Ni, $^{93m}$Nb and $^{94}$Nb are under minimum detectable activity. Maximum, minimum and average specific activity of the six samples and the ratio of the data considered for calculations are collected in Table III.

The same parameters for the nuclides of the first step of characterisation in these six samples are collected in Table IV.

Table III. Ranges of specific activity (Bq/g) up to MDA of nuclides determined at 2<sup>nd</sup> Characterisation step

<table>
<thead>
<tr>
<th></th>
<th>&gt; MDA (%)</th>
<th>Maximum (Bq/g)</th>
<th>Minimum (Bq/g)</th>
<th>Average (Bq/g)</th>
<th>STD (%)</th>
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</thead>
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<tr>
<td>$^{41}$Ca</td>
<td>100.0</td>
<td>4.65E+02</td>
<td>7.06E+01</td>
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<td>63.9</td>
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<tr>
<td>$^{56}$Fe</td>
<td>100.0</td>
<td>1.56E+04</td>
<td>1.34E+03</td>
<td>7.83E+03</td>
<td>70.3</td>
</tr>
<tr>
<td>$^{63}$Ni</td>
<td>100.0</td>
<td>3.28E+04</td>
<td>1.25E+03</td>
<td>1.43E+04</td>
<td>84.5</td>
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<tr>
<td>$^{89}$Sr</td>
<td>100.0</td>
<td>1.04E+01</td>
<td>1.04E-02</td>
<td>3.86E-02</td>
<td>87.6</td>
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<tr>
<td>$^{90}$Sr</td>
<td>100.0</td>
<td>7.65E+02</td>
<td>7.04E+01</td>
<td>2.60E+02</td>
<td>97.0</td>
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<tr>
<td>$^{238}$Pu</td>
<td>100.0</td>
<td>1.31E+02</td>
<td>6.44E+00</td>
<td>4.47E+01</td>
<td>92.0</td>
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<tr>
<td>$^{240}$Pu</td>
<td>100.0</td>
<td>2.06E+01</td>
<td>3.53E+00</td>
<td>8.79E+00</td>
<td>65.0</td>
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<tr>
<td>$^{241}$Pu</td>
<td>100.0</td>
<td>1.94E+03</td>
<td>2.87E+02</td>
<td>7.21E+02</td>
<td>79.3</td>
</tr>
<tr>
<td>$^{241}$Am</td>
<td>100.0</td>
<td>6.40E+01</td>
<td>1.33E+01</td>
<td>2.60E+01</td>
<td>68.5</td>
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<tr>
<td>$^{242}$Cm</td>
<td>100.0</td>
<td>6.10E-02</td>
<td>6.93E-03</td>
<td>2.90E-02</td>
<td>73.1</td>
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<tr>
<td>$^{244}$Cm</td>
<td>100.0</td>
<td>5.79E+02</td>
<td>1.75E-01</td>
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<td>134.6</td>
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<tr>
<td>$^{238}$U</td>
<td>100.0</td>
<td>2.42E+00</td>
<td>5.64E-02</td>
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<td>101.7</td>
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<tr>
<td>$^{235}$U</td>
<td>100.0</td>
<td>2.31E+00</td>
<td>5.90E-02</td>
<td>7.10E-01</td>
<td>106.6</td>
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</table>
Table IV. Ranges of specific activity (Bq/g) up to MDA on six selected samples of nuclides determined at 1st Characterisation step

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>&gt; MDA (%)</th>
<th>Maximum (Bq/g)</th>
<th>Minimum (Bq/g)</th>
<th>Average (Bq/g)</th>
<th>STD (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>³H</td>
<td>100.0</td>
<td>5.11E+05</td>
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<tr>
<td>¹⁴C</td>
<td>100.0</td>
<td>1.22E+05</td>
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<td>²³Na</td>
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<td>7.45E+02</td>
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<td>57.1</td>
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<tr>
<td>⁶⁰Co</td>
<td>100.0</td>
<td>4.36E+04</td>
<td>1.59E+03</td>
<td>1.62E+04</td>
<td>87.0</td>
</tr>
<tr>
<td>⁹⁰Nb</td>
<td>33.3</td>
<td>8.26E+01</td>
<td>2.38E+01</td>
<td>5.32E+01</td>
<td>55.3</td>
</tr>
<tr>
<td>¹³³Ba</td>
<td>83.3</td>
<td>1.01E+03</td>
<td>1.31E+02</td>
<td>5.47E+02</td>
<td>59.4</td>
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<tr>
<td>¹⁴⁷Cs</td>
<td>100.0</td>
<td>7.12E+02</td>
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<td>3.57E+02</td>
<td>56.0</td>
</tr>
<tr>
<td>¹⁴⁷Cs</td>
<td>100.0</td>
<td>3.83E+03</td>
<td>3.05E+02</td>
<td>1.42E+03</td>
<td>89.6</td>
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<tr>
<td>¹³⁵Eu</td>
<td>83.3</td>
<td>1.76E+03</td>
<td>5.20E+02</td>
<td>1.11E+03</td>
<td>43.3</td>
</tr>
<tr>
<td>¹⁵⁸Eu</td>
<td>100.0</td>
<td>7.15E+02</td>
<td>2.26E+02</td>
<td>4.00E+02</td>
<td>41.4</td>
</tr>
</tbody>
</table>

Relative activity of alpha, pure beta and beta-gamma emitters are plotted in bar diagrams as are shown in figures 7, 8 and 9 respectively. The relative specific activity per nuclide is calculated as the ratio between activity of each sample and average activity of the six samples in order to get comparable activities values to study the tendency.

![Figure 7. Evolution of alpha emitters](image-url)
4. DISCUSSION

The procedures applied for characterising the graphite samples are already implemented in the routine operation of the Waste Characterisation Project in CIEMAT as for other operational waste streams, however the special physic-chemical characteristics of graphite implied to apply a specific sample preparation process.
These processes are:
- Milling the graphite cylinders avoiding dispersion as well as contamination
- Homogeneous distribution of the obtained powder in order to fix a measurement geometry
  with adequate characteristics according to the standards for gross alpha-beta measurement
  and direct high energy gamma spectrometry.

Results from characterisation (Table II) show some important direct conclusions:

Tritium activity appears as the higher radioactive nuclide in the graphite (around $10^5$ Bq/g in
average) that means one order of magnitude higher than the other two majority components $^{14}$C
and $^{60}$Co. This could be produced by the activation of $^3$H and $^2$H bonded to the organic molecules
associated at graphite and minor contribution from contamination coming from fuel during
operation. Distribution of $^3$H in figure 1 looks heterogeneous (Std = 79 %) regarding to both radial
and vertical positions in the pile and presents higher activity concentration at height up to 7.7 and
radial position in the range 0.2D to 0.1D.

Activity of $^{14}$C shows (figure 2) pretty regular trend distribution along the pile radius and a
maximum concentration between 3.3 to 5.1 m height (central zone of the pile). Production of $^{14}$C
comes from activation of $^{12}$C and being this nuclide the structural component of the pile, its
concentration should be linked to the integrated neutron flux in each zone.

It is important to pay attention on the behaviour of activity ratio $^3$H/$^{14}$C regarding to $^{14}$C
concentration. It is observed that as much as $^{14}$C concentration grows up the ratio decrease faster,
that could be in relation with the degradation of the structure of the graphite by the integrated
neutron flux. When the structure is degraded there are more possibilities for $^3$H to escape from
the graphite structure and consequently reduce its activity. This behaviour was also observed in
previous R+D works [4, 5] on leaching studies of graphite sleeves from the same plant when
probes with higher activity concentration of $^3$H presented lower leaching rates of $^3$H.

Cobalt-60 present similar profile along the radius of the pile that $^{14}$C with maximum activity
concentrations in each radial distance at 3.3-3.9 m height. The maximum values were registered
for 0.4D in opposition to the behaviour observed with $^{14}$C in which the maximum concentration
appears at lower radial distance. It is remarkable that the main source of $^{60}$Co are the devices
made of stainless steel wire used for fixing the fuel element into each graphite sleeve.

Cesium-137 activity distribution (figure 4) shows random distribution in both senses, so that we
can conclude that its presence it is not in connection with the composition of the pile and neutron
flux.

These distributions can be checked clearly in figure 5, where it is plotted the profile for five
nuclides at radial positions 0.4D and 0.2D. It is possible to check the relation of activity
concentration of these nuclides as well as the trend of $^{241}$Am and $^{137}$Cs that does not look in the
same form that $^{14}$C profile, so these nuclides are not correlated with integrated neutron dose. On
the other hand the profile of $^{60}$Co has the same tendency that $^{14}$C. These remarks can be also
checked in the profile of activity in function of radial position for 1.5m and 3.9 m height
(Figure 6).

The study of activity distribution concerning the six selected samples for destructive
characterisation shows that the activity concentration is higher in the samples that range from
1.5 m and 3.9 m height and no significant variance in function of the radial position of sample in
the pile.
For alpha emitters the activity distribution presents the behaviour already mentioned for every nuclide tested (figure 7).

In the case of pure beta emitters (figure 8) besides of $^3$H and $^{14}$C all nuclides determined present practically the same tendency except for $^{63}$Ni that presents a maximum at higher vertical level (7.7 m).

In the case of beta-gamma emitters characterised (figure 9) by direct measurement of the graphite powder, the behaviour is the same for every nuclide except for $^{137}$Cs and $^{60}$Co already discussed and $^{134}$Cs whose tendency is the same than $^{137}$Cs as was expected. We can also observe in these nuclides another local maximum in the sample collected at 9.0 m height.

5. REFERENCES


FREE RELEASE WASTE CHARACTERISATION DURING THE DECOMMISSIONING OF WINDSCALE PILE 2 CHIMNEY.

A.J. Frith (NIRAS), E.D. Dickson (BNFL), J.A. Hewetson (BNFL), K McCormick (BNFL)

Abstract:

The decommissioning of Windscale Pile 2 Chimney resulted in the removal of one of most historically significant and prominent features of the Sellafield site. The project was the first large scale concrete "free release" operation to be undertaken on the Sellafield nuclear site, producing 4500 tons of concrete of which 3000 tons was demonstrated to be "free release".

The paper describes the radiometric techniques employed in the characterisation and segregation of the concrete into Low Level Waste (LLW), Very Low Level Waste (VLLW) and Free Release categories. It examines the robust solutions that were developed to meet the technical and regulatory challenges of the project, which included the definition of free release, the selection of averaging volumes, the testing and validation of the monitoring systems employed for ton quantities of concrete and the identification and removal of small numbers of fuel particles from the free release waste stream.

As a result of interest shown by other BNFL and UKAEA decommissioning projects at Sellafield, the paper finishes by discussing ways in which the system may be developed to assay other waste streams.

1. Introduction

The two Windscale Piles were the first production reactors to be built in the UK. With construction commencing in 1947, the first reactor (Pile 1) went critical in 1950, with Pile 2 going critical in 1951. Concerns over the safety and supply requirements of the water-cooled reactors being built and operated in the United States led to the adoption of an air-cooled reactor design. Key to the operation of these reactors was the supply of cooling air at a rate of one ton of air per second, that was fed into the reactor by two large fans on either side, passed through the core and was exhausted via a 425 ft (130m) tall chimney (Figure 1 and 2).

After a fire in the core of Pile 1 reactor in 1957 resulted in the world's first major nuclear accident, both reactors were shut down. The two reactors and chimneys remained in care and maintenance until the 1990's, when work began on decommissioning the reactors and concerns over the future structural stability and the ease of decommissioning the deteriorating chimneys led to the decision to decommission the chimneys. The decommissioning of Pile 2 chimney began in 1996.

Unlike Pile 1 Chimney, which suffered gross contamination due to the 1957 fire, Pile 2 Chimney was contaminated at lower levels with fission products that resulted from the failure of small numbers of the 700,000 aluminium fuel cartridges contained within the reactor core. In addition, the identification early in the waste characterisation project of "hot particles" containing alpha emitting radionuclides, led to a closer examination of the Pile 2 reactor operating records. This identified a number of incidents that involved the oxidation of fuel in failed fuel cartridges. These incidents were linked at the time to the release of fuel particles into the environment, and the Project Team concluded that this was the source of the "hot particles" within the chimney.

Therefore, in addition to the considerable engineering challenges posed by the decommissioning of Pile 2 Chimney, there was the characterisation of the contaminated concrete, which is the subject of this paper.

Figure 1. A sectional diagram through a Windscale Pile Reactor
2. Waste categorisation

Initial measurements indicated that the activity concentration of the concrete on the inner surface of the concrete chimney was consistent with the Low Level Waste (LLW) categorisation used in the UK, i.e.
- less than 4 GBq/t alpha (approximately 0.1 Ci/ton)
- less than 12 GBq/t beta/gamma (approximately 0.3 Ci/ton)

But what of the remaining concrete within the walls of the chimney? Would the activity concentration of the concrete within the body of the walls of the chimney be sufficiently low to permit the disposal or re-use of the material without regulatory control (i.e. "free release")?

Calculations, supported by activity concentrations determined on a number of cores taken though the sides of the chimney, indicated that significant quantities of the concrete would potentially meet "free release" criteria. As the cost of LLW disposal from BNFL Sellafield is currently £1,100/m³ (EUR 1,700, US$ 1,650) and there is an acknowledged need to conserve the capacity of the UK's only LLW depository by preventing the disposal of inappropriate waste, there were strong commercial and environmental reasons to embark on a "free release" project. The main objectives of the project would be to address the requirement to monitor bulk quantities of concrete at low activity concentrations, and to segregate material falling within different activity concentration categories.

3. "Free release" criteria

The first step of the work involved the definition of "free release". This would enable the performance parameters of the monitoring system to be established.

The disposal of radioactive waste from a site licensed under The Nuclear Installations Act 1965 (NIA) (1) is controlled by The Radioactive Substances Act 1993 (RSA) (2). The "free release" criterion commonly applied in the UK to solid material that is substantially insoluble in water is given in The Radioactive Substances (Substances of Low Activity) Exemption Order 1986 (SoLA) (3). This defines an activity concentration of 0.4 Bq/g below which the keeping and use of material and the accumulation and disposal of waste is exempt from control under the RSA.

Whilst SoLA specifies an activity concentration in terms of grams, it is acknowledged by the Regulator that the testing of each gram is not required, and that it is permissible to average activity concentration over a larger increment. Averaging volumes in the UK tend to range from 0.1 m³ to 10 m³ (4).

Therefore, at the outset, the project defined "free release" as being material or waste with an activity concentration less than 0.4 Bq/g when averaged over a project disposal increment that would lie within the 0.1 m³ to 10 m³ range.

During the latter stages of the work it became apparent that the concrete may be re-used as construction material on a nearby Nuclear Licensed site. As radioactive material or waste on a nuclear licensed site is controlled by the conditions of the license issued under the NIA rather than the RSA, an extensive revision to the "free release" criteria had to be made. This involved the adoption of risk-based criteria, which is discussed in Section 9.

4. Radionuclide fingerprint

In general, the monitoring of ton quantities of material at activity concentrations of the order of 1 Bq/g is only practicable where gamma-emitting radionuclides are present.

As the operational nature of the reactor, the 40 year period that had elapsed between reactor shut-down and decommissioning, and the fingerprint obtained from the steel lining plates within the chimney clearly indicated that cesium-137 would be one of the primary contaminants, it was decided to make use of the penetrating nature of the gamma radiation emitted by Cs-137/88-137m in the monitoring of this material.

In order to relate the Cs-137 activity concentration to the total activity concentration, it was necessary to determine the radionuclide fingerprint of the concrete. Fingerprint analysis was undertaken on material of
higher activity concentration than would have been admitted to the "potentially free release" waste stream in order that the analytical uncertainties would represent a smaller fraction of the determined value. Confirmation that there was no significant variation in fingerprint at lower activity concentrations, due to factors such as the different mobility of contaminants, was confirmed by analysis of concrete of lower activity concentration. This work also demonstrated the absence of measurable activity concentrations of gamma emitting radionuclides, other than Cs-137, and those which are part of the U-238 and Th-232 decay series.

The radionuclide fingerprint used by the project (Table 1) is such that the multiplication of a Cs-137 activity concentration by 1.89 will give the total contaminant activity concentration.

Table 1: The radionuclide fingerprint of the concrete in Pile 2 chimney.

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Activity Concentration</th>
</tr>
</thead>
<tbody>
<tr>
<td>H-3</td>
<td>0.50 %</td>
</tr>
<tr>
<td>Sr-90</td>
<td>46.28 %</td>
</tr>
<tr>
<td>Cs-137</td>
<td>52.96 %</td>
</tr>
<tr>
<td>U-234</td>
<td>0.05 %</td>
</tr>
<tr>
<td>U-238</td>
<td>0.05 %</td>
</tr>
<tr>
<td>Pu-239+240</td>
<td>0.14 %</td>
</tr>
<tr>
<td>Am-241</td>
<td>0.01 %</td>
</tr>
</tbody>
</table>

5. Considerations for the prototype system

The first issue to be addressed in the design of the system was the avoidance of deliberate dilution and dispersal to achieve "free release", an approach that is not permitted in the UK. The system must therefore prevent the mixing of contaminated concrete with "free release" concrete, as far as practicable. This was achieved by in-situ monitoring with health physics instrumentation, and targeted mechanical removal of areas of elevated activity concentration. Thus, concrete with an elevated activity concentration was removed from the "free release" waste stream at the outset. This ensured that the concrete forwarded for "free release" assessment had a low activity concentration and could be classed as "potentially free release", within the limitations of activity concentration measurements made at this level with hand-held instruments in an active area.

Further measures to avoid dilution and dispersal included preventing transfer of material between increments once the "potentially free release" material was loaded into uniquely identifiable increment containers.

The second issue was the form in which the "potentially free release" concrete should be processed. The majority of concrete was broken off the chimney using hydraulic breakers (Figure 3) and was consequently in a form that was difficult to monitor and would be difficult to store efficiently. These difficulties were overcome by crushing the concrete to a particle size of 14 mm or less using an industrial crushe. This produced a locally homogenised material that was better suited to making activity concentration measurements at higher levels of confidence, being the same approach used by analytical laboratories. Crushing brought the additional benefit of efficient packing, achieving a bulk density of 1.5 g/cm³, and converted the concrete into a form whereby, once free-released, it could be used in construction projects.

Figure 3: Concrete being broken off the chimney using a hydraulic breaker.
The third issue was to determine the increment over which the activity concentration should be averaged. In general, there are advantages in adopting the largest averaging increment that can reasonably be justified, and it was decided that 10 m³ skips would be suitable.

What remained was the development of a monitoring system that would be able to demonstrate that the activity concentration of the locally homogenised "potentially free release" crushed concrete, was less than 0.4 Bq/g, averaged over a 10 m³ increment.

The first operation was to examine the uniformity of activity concentration of the material within a skip by taking ten 1.5 kg samples at 1 m³ intervals during the filling of each skip. These were assayed in a Marinelli geometry for gamma activity using a 2" x 2" sodium iodide detector, housed in a substantial lead shield and operated at the base of the Pile 2 chimney.

Skips where the uniformity of activity concentration had been demonstrated were to be passed out of the Sellafield Separation Area to a low background "non-active" area. The gamma activity was determined using an "infinite depth" counting technique by positioning a single 2" x 2" sodium iodide detector centrally above the skip. The gamma activity was to be related to the total activity via the radionuclide fingerprint.

This was thought sufficient for the highly uniform, low activity concrete anticipated, and trials were undertaken to test the efficacy of this approach.

6. Trial Processing

Trials were undertaken on material from the first stages of decommissioning at the top of the chimney, an area that housed the filter galleries and had been identified at the outset as an area with the highest levels of contamination. It was fortuitous that this material was processed first, as a number of problems became apparent.

The first problem was that the activity concentration of the "potentially free release" material, whilst low, was variable. This meant that in taking an averaging increment of 10 m³, very few skips would pass the uniformity criteria adopted by the first stage of monitoring, and the activity concentration of many would exceed 0.4 Bq/g.

This problem was addressed by the adoption of a smaller averaging increment, being a standard 1 m³ nylon bag used in the construction industry, and termed an intermediate bulk container (IBC). Whilst the number of process increments was increased ten-fold, with the associated handling costs, it was clear that this reduction in averaging increment would optimise the segregation of "free release" and LLW, producing an overall benefit.

The second problem was the discovery of small numbers of millimetre-sized particles with activities of the order of kBq (1 x 10⁸ Ci) within otherwise low activity concentration concrete. The low frequency of occurrence of these particles made little difference to the average activity concentration of the increment, but their presence raised important questions relating to the "free release" of such material. Subsequent radiochemical analysis of a number of particles showed that these were fragments of irradiated fuel, probably relating back to one of the fuel oxidation incidents during the Pile 2 reactor history, which has been noted previously.

This problem was addressed by adopting the best available technology for the identification and removal of these particles. This required the introduction of a further monitoring stage, which would take place after the ten-sample uniformity monitoring process but before the final sentencing monitor. Here, the whole contents of a 1 m³ IBC was put into a hopper and was fed along a conveyor belt in a layer of crushed concrete approximately 5 cm in depth and less than 30 cm width. A 30 cm caesium iodide strip detector was placed across the conveyor in a lead shield, and the radiometric systems were interfaced with the conveyor drive mechanism, such that the triggering of the count rate alarm would stop the conveyor and permit the removal of the particle(s). After the optimisation of parameters such as shielding and conveyor speed, it was demonstrated that a particle activity of 600 Bq Cs-137 would be intercepted in the worst-case position at the edge of the belt and that particles of 600 Bq Cs-137 would be intercepted near the centre of the conveyor, where the counting geometry is optimum.

Whilst the "hot particle monitor" relied primarily on the detection of the gamma photon emitted by the primary contaminant Cs-137 / Ba-137m, testing demonstrated that the way in which the detector was set up meant that the system was sensitive to beta radiation, via lead X-rays resulting from interactions with the shielding and through bremsstrahlung generated by interactions with the shield and the detector itself.
7. Operational system

The salient components of the refined system used in the processing of the concrete produced by the decommissioning of Pile 2 chimney are identified in Figure 4 and are described briefly in this section.

Figure 4    A schematic of the waste characterisation system.

7.1 In-situ screening

The monitoring of concrete at the work face within the chimney was undertaken using portable health physics instruments. Whilst the limitations of such instrumentation to indicate activity concentration, especially within the gamma radiation fields that existed within the chimney, are acknowledged, this approach was suitable to identify material of elevated activity concentration and to permit its removal. The remainder was forwarded to the "free release" monitoring system as "potentially free release" material.

7.2 Stage 1 Uniformity Monitor

The Stage 1 Uniformity monitor continued to operate on ten 1.5 kg samples, but with samples now taken at 0.1 m³ intervals. IBC's were only allowed to continue the "free release" process if the average activity concentration of the ten samples was less than 0.4 Bq/g and no single sample exceeded 1.0 Bq/g. IBC's that did not meet these criteria were sentenced as VLLW and were fed into another waste stream.

7.3 Stage 2 Hot Particle Monitor

After transfer from the Sellafield Separation Area to a low background area on the Sellafield site, each individually numbered IBC was loaded into the Stage 2 hopper (Figure 5), and was fed along a conveyor passing beneath the 30 cm caesium iodide strip detector (Figure 6), before being returned to the original IBC. This operation permitted any particles with an activity of greater than 800 Bq Cs-137 to be intercepted and removed.

Figure 5.    The Stage 2 Hot Particle Monitor
It must be noted that whilst this process introduced further adventitious mixing of the contents of an IBC, no material was transferred between IBCs. In addition, as the Stage 2 monitor also responds to the activity concentration the entire contents of an IBC is monitored as it passes beneath the detector of the bulk concrete, thus ensuring a greater degree of confidence in the uniformity of activity concentration of the product than had been practicable with Stage 1. It was determined that the mass of concrete to which the detector was sensitive during the time integration period used by the instrument was approximately 5.2 kg. The alarm would trigger where the total contaminant activity concentration of that mass exceeded 0.6 Bq/g.

Figure 6. Concrete being passed beneath the shielded detector

7.4 Stage 3 Final Sentencing Monitor

As a result of the operations of Stages 1 and 2, it was possible to present 1 m$^3$ of crushed concrete whose uniformity of activity concentration had been demonstrated to a high degree to the Stage 3 Final Sentencing Monitor (Figure 7).

Each IBC was assessed for activity concentration over a 10-minute count period, with an environmental background measurement being made before and after the IBC measurement in order that a statistical test could be applied to confirm the background stability. Those IBCs that passed this final assessment were forwarded to temporary storage, where upon verification that all the measurement records and quality control records were present and correct, the material was finally categorised as "free release".

Figure 7 The Stage 3 Final Sentencing monitor
8. Results

Decommissioning of the Pile 2 chimney continued until the structure had been reduced to the below the level of the reactor building and within 5 m of the air duct (Figure 8). This work generated 4500 tons of concrete of which 3000 tons was shown to be "free release" based on the 0.4 Bq/g criterion.

Figure 8. Pile 2 chimney upon completion.

9. Final free release

During the latter stages of the work a requirement for large quantities of such material for new construction at the nearby BNFL Drigg site was identified. The use of this material for this purpose would remove the cost and environmental impact associated with importing new material.

This introduced a final problem for the project; the 0.4 Bq/g "free release" de-minimis activity concentration is not recognised by the NIA, nor does this Act have any alternative de-minimis activity concentration. Confronted with the prospect of having to manage this material as if it were "radioactive", an alternative approach to the problem was required.

Drawing on the team's experience of nuclear site de-licensing, the criterion for de-licensing within the NIA was examined. The vague statement contained within this Act that de-licensing could occur only when "there had ceased to be any danger from ionising radiation from anything on the site" has been more rigorously defined by the Nuclear Installations Inspectorate (NII) as being a risk rate of $10^{-6}$ deaths per year. This equates to an annual dose of approximately 30 μSv.

The case was presented to NII that the placement of material on a nuclear licensed site that would not subsequently prejudice the de-licensing of that site should not require control as if it were radioactive. This argument was accepted and it remained to demonstrate that the annual dose resulting from the placement of this material would meet this dose rate criterion.

As the radiological hazard is not only dependent on activity concentration, but on total activity, and the radionuclides present, the radiological risk was modelled for the whole material. Using the radionuclide fingerprint, it was possible to apportion the total activity which had been calculated for comparison with SoLA into its constituent radionuclides to enable the annual dose resulting from each radionuclide to be calculated. The exposure models used identified that external radiation was the primary exposure route associated with the placement of this material.

Fortunately, the majority of the concrete previously sentenced as "free release" under the 0.4 Bq/g criterion was well below this activity concentration, (Figure 9 shows the number of bags within given activity concentration limits for a typical production run). Therefore it was possible to demonstrate that the placement of this material met the risk-based criterion without the need for the removal of any IBC's that had previously been declared "free release".

After all had been put before the Regulator, consignments of "free release" concrete were shipped from the Sellafield site by rail (Figure 10), for re-use in new construction.
10. Conclusion

This project demonstrates a commitment to a robust "100%" monitoring process, which has enabled a high degree of confidence to be assigned to the "free release" categorisation of ton quantities of material. Confidence has been gained by peer review and scrutiny by the Regulators and their consultants.

Whilst the system was specifically designed to process potentially free release concrete from Pile 2 Chimney, where the primary contaminant is caesium-137, the predominance of this 30 year half-life gamma emitting fission product in many of the contaminated construction materials at the Sellafield site makes the System eminently suitable for handling other waste streams of similar radionuclide fingerprint.

The system is currently being used to process 850 tons of concrete that originated from a shield wall surrounding a dry waste silo on site, that is currently being decommissioned and where caesium-137 is again a primary contaminant.

In addition, the possibility of developing the system to handle waste streams where the primary contaminants are radionuclides other than caesium-137 is also being examined. In particular, waste streams where uranics and transuranics contaminants are present represents a technical challenge for the team.

11. References

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3. The Radioactive Substances (Substances of Low Activity) Exemption Order 1986, Statutory Instrument No 1002, HMSO

12. Acknowledgements

The authors would like to acknowledge the contribution made to this project by all the members of the Pile Chimney's Team, namely (in alphabetical order), S. Cowper, M. Hetherington, M. Howarth, and P. Salmon.

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Poster Presentations
Status and Developments in Design Testing and Licensing of Non-Heat Generating Radioactive Waste Containers in Germany

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Abstract
Containers for non-heat generating waste are used and qualified in accordance with the transport regulations and with the requirements for interim storage respectively the provisional requirements for final disposal in Germany (KONRAD repository). The KONRAD requirements are the basis for container design qualification at present and they will have to be considered for any finally realised repository. At the same time very similar requirements are the basis for the interim storage container qualification.

BAM as the competent authority for design testing and quality assurance measures for transport containers as well as for final disposal containers on behalf of the Federal Institute for Radiation Protection (BfS) is involved in several licensing applications by different container manufacturers. The industry is increasingly interested in demonstrating the accordant of their containers with the current final disposal requirements. This paper shows examples and experiences from design testing of containers made of steel, concrete/granular iron or ductile cast iron. It also shows results from a research project investigating cubic ductile cast iron casks.

1 Introduction
Containers for non-heat generating waste are increasingly designed, fabricated and operated for later final disposal considering the current final disposal requirements for the German KONRAD repository. The licensing and test requirements were explained in detail in a former presentation of BAM at RADWAP 1997 [1]. It was evident that such containers are normally developed to be operated under transport, interim storage and final disposal conditions. Therefore it was important to analyse and compare the requirements from the three fields of operation and to show a wide similarity of requirements but with some specific differences in detail [1].

A short summary of the KONRAD requirements is given in Figure 1. The impact velocity of 4 m/s corresponds to a drop test height of 0.8 meters. Additionally to the design test requirements the container manufacturer has to demonstrate that his fabrication works on an appropriate quality assurance program and finally each container type has to be fabricated on the basis of certified “Inspection and Test Plans”. If one compares the requirements from the transport regulations [3], [4] it is obvious that most of the therefore necessary basic proofs are suitable directly or with slight modifications for final disposal certification of at least KONRAD Class 1 (ABK I) containers.
Basic Requirements:
- Container geometry (defined container types)
- 6 m stacking heights
- Corrosion resistance of steel containers
- no mechanical or corrosive damages impairing container leak-tightness and integrity during handling and stacking

Specific Requirements:

<table>
<thead>
<tr>
<th>Waste Container Class I (ABK I)</th>
<th>Waste Container Class II (ABK II)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Impact with 4 m/s velocity and fire test (800°C, 1 hour)</td>
<td>Drop test from 5 m heights:</td>
</tr>
<tr>
<td>→ limited oxygen supply, so that waste with Tₚ&gt;300°C doesn’t burn but pyrolyses</td>
<td>→ leakage rate ≤10⁻⁴ Pa·m⁻³/s</td>
</tr>
<tr>
<td>Impact with 4 m/s velocity</td>
<td>and fire test (800°C, 1 hour)</td>
</tr>
<tr>
<td>→ leakage rate ≤10⁻⁵ Pa·m⁻³/s</td>
<td>→ leakage rate ≤10⁻⁴ Pa·m⁻³/s</td>
</tr>
</tbody>
</table>

„Accident-Safe“ design: Waste is fixed in stable form:
Drop test from 5 m heights: → Integrity of inner packages or the fixing is kept, no activity release

Figure 1: Summary of final disposal requirements (KONRAD facility)

2 Tasks of BAM Design Testing

On behalf of the Bundesamt für Strahlenschutz (BfS) as the competent authority for final disposal container licensing in Germany BAM is responsible for the assessment of container design testing and quality assurance measures referring to the licensing process. This includes the detailed examination of each container construction relating to material qualification, corrosion prevention and mechanical and thermal qualification for operational and accident conditions. The assessment of the applicant’s quality assurance measures includes the quality management system fixed by the quality management handbook as well as the quality assurance measures for container fabrication and operation.

During the last years several different container design by different manufacturers have been applied for a KONRAD license. At first there are several types of cubic steel containers of the KONRAD types IV and VI. Secondly special concrete containers with granular iron have been developed. These containers are cubical shaped or of cylindrical design. And thirdly there are cubical and cylindrical ductile cast iron containers. The applied qualification level depends on different waste forms which for the container constructions are developed and includes ABK I up to ABK II and in some cases the “Accident-Safe”-design (see Figure 1).

The following Figure 2 shows a typical test configuration, a 0.8 m drop test with a concrete container onto its long edge. In this case you find only slight deformations in the container’s impact area but no loss of integrity. Other test configurations are stacking pressure tests, lifting tests and fire tests. In all cases it is necessary to ensure that the prototype test container represents all relevant properties of the later serial production containers including the effects of the container contents.

The final result of BAM design testing and assessment of quality assurance measures is in the positive case a “BAM Test Certificate” which confirms that the container design is in accordance with the KONRAD requirements. This certificate is then the basis for the official license given by BfS.
3 Research and Development

In addition to the daily tasks of design testing for container design certification BAM as the Federal Institute for Materials Research and Testing always investigates in the development of new analytical and experimental safety assessment methods for cask designs.

During the last years BAM concentrated on the investigation of the impact behaviour of cubic ductile cast iron casks. Generally such cubic casks show more critical stresses compared to cylindrical casks because of their flat walls connected by rectangular edges and corners. On the basis of a comprehensive research project sponsored by the Federal Ministry of Education and Research (BMBF) and in co-operation with the industry BAM has been developing methods of modelling complex container – foundation structures for finite-element analysis [2]. A main task of this work is the correct determination of highly dynamic stress conditions of containers in the case of hard impacts without impact limiters onto real foundations of storage facilities. The determination of adequate material properties especially for the foundation and the choose of proper contact conditions between the container and the foundation are difficult questions within the frame of this task. Figure 3 shows an example of a 5.5 m drop onto the container bottom with only slight variation of the impact angle but with significant influence on stress level over time. For necessary verification of the calculation results extended drop test series with original prototype containers have been performed. On this basis BAM develops now extended fracture mechanics safety assessment methods for ductile cast iron materials melted with scrap additions. The main idea in this case is to use a higher content of scrap metal additions from decommissioning of nuclear installations. Altogether such investigations lead to a better understanding of complex dynamic container behaviour under hard impact conditions and help to determine safety margins more precise.
There is an increasing demand for qualification of waste containers referring on the current KONRAD requirements which are expected to be the basis for any future repository in Germany. Nuclear installations want to operate qualified waste containers during at-site interim storage covering the later final disposal aspects. Several container manufacturers want to get licences for their different container types which means an increasing demand for design testing by BAM. Research activities enable more efficient use of materials from decommissioning of nuclear installations for fabrication of ductile cast iron containers.

5 References


Monitoring of Induced Seismicity: Tool for Control Stability of Underground Nuclear Waste Repository

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ABSTRACT: Requirements and character of operation of induced monitoring system according to the two-stage model of destruction are considered.

INTRODUCTION

The creation of underground nuclear waste repository causes change of rock massif properties. For maintaining of longevity safety of environment is necessary to carry out monitoring of its strained and deformed condition. It is realized with the help of monitoring systems, which allow estimating the stability of the rock massif, detect zones of preparation of dynamic events and forecast strong dynamic events.

The full monitoring of a rock massif is carried out by the complex system which accepts, registers and processes all the information describing stress and strain changes in rocks and also destruction processes shown as radiation signals of the acoustic emission and microseismicity (AE/MS).

Thus, the induced monitoring system is a subsystem of the complex monitoring system with the following goal function – on the basis of observable elastic oscillation signals accompanying brittle destruction processes of rocks to ensure continuous automated monitoring, estimation and forecasting of the condition of the observed rock massifs. The reaching of this purpose depends on the correspondence of used technical means and procedures of receiving, registration and data processing of AE/MS events to physical regularities of rock destruction processes, which are described by models of this physical phenomena.

TWO-STAGE MODEL OF FRACTURE

Kinetic concept of stability of solids revealed the principle possibility of the forecasting of the macrocrack according to which the destruction is stipulated not by the limit condition of the solid under a load but by the process explicating in time [1].

In the correspondence with the two-stage model of destruction of heterogeneous bodies [2, 3], the structural heterogeneity of the material leads to uneven distribution of the affixed exterior load. As a result there are elements, which probability of destruction by virtue of reduced thermoactivation barrier is higher than the average probability for the whole solid. Such cracks accidentally distributed in space originating from destruction are stabilized on the boundaries of heterogeneity. Thus, the first stage of the process consists in multiple dispersible accumulation of noninteracting cracks, which size is determined by the given level of structural heterogeneity J. In the process of accumulation of cracks in some area their concentration casually exceeds the average concentration for the whole solid. In some areas called nucleus of destruction, where local cracks reach the threshold concentration between cracks, interaction arises that stimulates further cracking. The conditions are formed, at which the nucleus zone loses its stability and the effect J + 1-st rank appropriate to the following size of heterogeneity is formed. If in the material there is a hierarchy of heterogeneity sizes intrinsic to rocks, the model assumes the similarity of the fracture process at all existing scale levels. The peculiarity of the given model is its invariance to the scale of rock destruction process.
The casual character of rock destruction has defined statistical methods of its exposition. Cracks arising during loading may be conceived as a stream of discrete events, each of them is characterized by a coordinate on time axis and in space as well, and by the size of the corresponding crack. Such flow can be correlated by the sequence of registered AE/MA where energy of signals are characteristics of crack sizes. Reformulating the two-stage model from positions of statistics of the stream of discrete events we can consider the first stage satisfying the conditions of the quasi-stationary Poisson process and the disturbance of these conditions is a criterion of formation of the destruction nucleus.

As parameters describing spatially-temporally singularities of destruction, use average values of space ($\Delta r$) and temporal ($\Delta t$) intervals between chronologically sequential events and their factors of variation ($V_{\Delta r}, V_{\Delta t}$).

On the first stage ($t<T_1$) of uncorrelated formation of cracks their stream is Poisson with intensity $\lambda$ and $\Delta r$. When cracks reach the threshold concentration in the observing volume of material ($t=T_1$), conditions arise for their interaction and cracking is stimulated by it, that breaks conditions of the Poisson process. In the Fig.1 the diagrammatic illustrations of the destruction nucleus [3] are represented.

This stage is shown in increasing of parameters $V_{\Delta t}$ and $V_{\Delta r}$ and decreasing $\Delta t$ and $\Delta r$. As a result of loss of stability by the destruction nucleus at the moment $t=T_2$ a crack of the following level of heterogeneity is formed. The moment $T_2$ not necessarily completes the second, non-stationary stage. As a result of the nucleus zone relaxation beginning from the moment $T_3$ inverse tendencies can arise, i.e. decreasing $\Delta t$ and $\Delta r$ and increasing $V_{\Delta t}$ and $V_{\Delta r}$.

In the common case, fracture is realized simultaneously at different scale levels and at each of them one stage follows another one. Therefore detection of considered trends is possible at performance of three conditions [2]. It means that analyzed sample should include only events that come from the area of fracture nucleus preparation at the smaller interval of time than that of the beginning of the relaxation process. Energies of these events are corresponding to the certain rank of the fracture process. Practically, performance of these three conditions consists in triple (spatial, time and energetic) selection of event flow, where criterion is revealing and optimization of parameters trends that described above (Fig.1) [4].

The analysis of regularities of formation of the rock burst nucleuses and earthquakes [5] has allowed to relate energy AE/MS with ranks of events. The ratio on energy at creation of cracks of the given hierarchical level to previous does not exceed 4-th is order, i.e. $\frac{\lg(E_{j+1}/E_j)}{\Delta t}=C$, where constant parameter $C$ varies in limits 4±0.5.

REQUIREMENTS FOR A SEISMIC MONITORING SYSTEM

Described conditions allow formulating the clear requirements to an induced seismicity system of monitoring. In this case two-stage model of destruction represents itself as a criterion of a completeness and sufficiency of an accepting events stream for a realization of the forecasting macrocrack. That can be used for the setting of limit characteristics of means and procedures of signals AE/MS registration of monitoring system. Briefly it looks as follows.
The seismic monitoring system should conduct continuous registration of a stream of discrete events AE/MS, ensuring sensitivity permitting correctly to register events \( J \) of a rank. The energy of an event \( J \) of a rank is defined as \( \lg E_j = \lg E_{j+1} - 4 \), where \( E_{j+1} \) — energy appropriate to a rank \( J+1 \), which select at a formulation of the monitoring task for concrete volume of rock massif. So at selection of the rock burst nucleus with an energy \( E = 10^8 \) J it is necessary to analyze a seismic events stream with an energy beginning from \( 10^7 \) J

Basing on sensitivity of a seismic monitoring system in terms of a rank (minimum energy) AE/MS event is possible to determine many characteristics of system such, as real minimum volume and volume of controlled space in whole, a frequency bandwidth, dynamic range and others.

**Sensitivity.** Sensitivity of the seismic monitoring system defines from the task of the forecasting of dynamic events, since events of a given rank. Agrees [2] to realize prediction of events energy rank \( J+1 \), the system of seismic monitoring should conduct continuous registering of a discrete AE/MS events stream, ensuring sensitivity permitting correctly to register events smaller on an energy \( J \) of a rank. The event energy of \( J \) of a rank is defined as

\[
\lg E_j = \lg E_{j+1} - 4, \quad (1)
\]

where \( J+1 \)— energy to a rank \( J+1 \). At selection of the rock burst nucleus with an energy \( E=10^9 \) - \( 10^8 \) J it is necessary to analyze a seismic events stream, with an energy beginning from \( 10^7 \) - \( 10^6 \) J. At forecasting of events with an energy \( 10^2 \) - \( 10^7 \) J, that corresponds to weak dynamic manifestations of rock pressure, it is necessary to register events with an energy \( 10^{-6} \) - \( 10^{-3} \) J.

**Dynamic range.** The largest experienced events (at rockburst) have energy \( 10^{10} - 10^{13} \) J. The smallest events that are useful in determining the state of the rock have \( 10^{10} - 10^9 \) J. In laboratory conditions the modern technology allow to fix events and with a smaller energy (till \( 10^{12} \) J). However, at first, the registering of events with similar values of energies to a rock condition is extremely hampered, and on more or less acceptable distance (even \( 30-60 \) m from an event source) practically is impossible; secondly, has not almost not practical value, since reflects the destruction rock process in the most initial stages of destruction far from real creating of the nucleus of a significant dynamic event.

Usually dynamic range of seismic monitoring systems measure not in relations of energies (or classes, or magnitudes) seismic events, and, as well as for any electronic measuring system, in relations of a minimum and maximum input signal - ratio of the amplitudes of the maximum measurable signal \( A_{\text{max}} \) and the noise level \( A_{\text{noise}} \) in decibels:

\[
DR = 20 \log \left( \frac{A_{\text{max}}}{A_{\text{noise}}} \right) \text{ [dB]}, \quad (2)
\]

Observations and the theoretical reasons show that measurable the range in velocities of ground motions in an installation site of a seismic sensor should be between \( 10^{-6} - 10^{-7} \) m/s (a noise level) and \( 5 \times 10^6 \) m/s [5, 6]. Thus, the seismic transducers should have a dynamic range at least \( 132 \) dB; in the whole system of seismic monitoring, of course, is not worse.

**Frequency bandwidth.** A frequency bandwidth of a system of seismic monitoring defines on an corner frequency \( f_0 \) of expected signals AE/MS. The corner frequency is predominant frequency of a spectrum of a seismic wave in a point of installation sensor, which is determined as frequency, on which are intersected a low-frequency and high-frequency asymptote of a spectrum. For a given event, frequencies up to \( 3f_0 \) must be recorded for accurate energy determination and frequencies down to \( f_0/2 \) for seismic moment [6]. Therefore for a frequency bandwidth will be executed

\[
FRE = f_0/2, \quad 5 \cdot f_0, \quad (3)
\]

where \( f_0 \) and \( f_0/2 \) corner frequencies for the least and greatest event appropriate to a required range.

The regional seismic system (\( E=10^8 \) - \( 10^9 \) J) should have a frequency range from \( 0.2 \) up to \( 4000 \) Hz, and local system (\( E=10^3 \) - \( 10^4 \) J) — from \( 100 \) up to \( 50000 \) Hz. Such frequency range is accessible now and is realized in separate research systems. However in systems of routine operating, use an a lower upper limit of a frequency bandwidth for a drop of cost of a system.
Usually suppose, that for a regional seismic system the good frequency bandwidth makes 0.5-1000 Hz, and for local system – 1-20000 Hz [6].

**Size of monitoring zone.** The size of monitoring zone of a seismic system of by one-package sensors corresponds to a size of area of registering (definition of coordinates) microseismic events sources with energy, exceeding the given $E_{min}$. The size of monitoring zone is determined mainly by three factors: by attenuation of an seismic wave energy due to transmission through rock; by sensitivity of measuring transducers; by a required accuracy of the definition of coordinates of AE/MS events registered in monitoring zone by one package sensors.

In practice the monitoring zone defines from experimentally obtained relations connecting maximum a detection distance with energy or a corner frequency of an AE/MS signal. The detection distance names a distance at which a given magnitude event may be recorded.

The relations between energies, corner frequency and detection distance are shown in a Table 1. The Table 1 is building on base of analysis of experimental associations attenuating of signals AE/MS radiating in a rock obtained Hardy [7] and Mendecki [6].

### Table 1

<table>
<thead>
<tr>
<th>Energy, log $E$ [J]</th>
<th>-3</th>
<th>-2</th>
<th>-1</th>
<th>0</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
<th>8</th>
</tr>
</thead>
</table>
| Corner frequency, $f_0$ [Hz] by stress drop $\Delta \sigma$ = 1 [MPa] | 4·10$^4$ | 2·10$^5$ | 10$^5$ | 8·10$^4$ | 3·10$^3$ | 2·10$^2$ | 10$^2$ | 6·10$^1$ | 4·10$^0$ | 2·10$^0$ | 10$^0$ | 6 $^0$
| Detection distance [m] | 2·10$^2$ | 3·10$^2$ | 4·10$^2$ | 7·10$^2$ | 9·10$^2$ | 10$^2$ | 1·5·10$^2$ | 2·10$^2$ | 4·10$^1$ | 6·10$^1$ | 8·10$^1$ | 10$^1$

**CONCLUSION**

An approach to the definition of characteristics of a seismic monitoring according the two-stage model of destruction is considered. The given approach allows optimising the process of seismic monitoring systems creation, removing many indeterminations at a choice of parameters of monitoring device.

**REFERENCES**

1. Introduction

The role of Nirex is to provide the United Kingdom with safe, environmentally sound and publicly acceptable options for the long-term management of radioactive materials. This includes the development of coherent concepts providing a range of options for the management of radioactive waste. One such option is the phased deep geological disposal of wastes.

The Phased Disposal Concept [1] is designed to meet all safety standards and ensure long-term environmental protection and is underpinned by on-going research. It can be defined as a number of stages in the waste management process including storage, handling, transport and future potential disposal of radioactive wastes. Based on this Concept, Nirex has developed the Waste Package Specification (WPS) for Intermediate Level Waste [2]. The waste package constitutes a combination of a wasteform (typically waste immobilised in a cement matrix) and a waste container (typically a stainless steel canister). The WPS defines standard waste packages and provides firm foundations for the waste packagers so they can plan efficiently and with confidence, their waste management strategies. The WPS includes requirements for waste container parameters and design guidelines for lifting equipment.

The WPS is constantly monitored and regularly updated in order to provide the waste packagers with up-to-date information and to reflect changes occurring within the industry (e.g. regulatory changes). It is currently used throughout the UK nuclear industry to produce passively safe waste packages meeting the needs of a range of stakeholders, including prolonged storage.

2. Range of Containers

The current range of containers had been developed through close collaboration with customers and consists of two categories for packaging of wastes, as follows:

- storage and disposal containers - the 500 litre Drum, 3 m$^3$ Box and 3 m$^3$ Drum;
- storage, transport and disposal containers - the 4 metre Box and 2 metre Box (currently under development).
All containers in the current range are manufactured in stainless steel in order to minimise corrosion, thus ensuring a long-lasting performance.

2.1 Storage and Disposal Containers

Storage and disposal containers include the 500 litre Drum, 3m³ Box and 3m³ Drum. They are intended for storage and ultimate disposal of the waste. They are not designed to provide any radiation shielding in themselves, hence, when filled with waste they often need provision of remote handling equipment. For transport operations they will be placed in a reusable-shielded transport container, that is designed to contain either 4 x 500 litre Drums a 3m³ Box or 3m³ Drum.

The 500 litre Drum is a well established, widely used container for packaging operational intermediate-level wastes, including supercompacted wastes, as shown in Figure 1. However, the flexibility of constraints in Nirex’s approach to packaging has enabled a range of designs based on the 500 litre drum to be developed reflecting the various wasteform types. The design differences generally apply to the internal features, thus the container shown in Figure 1 features grout annulus on its circumference and an anti-floatation plate preventing the supercompacted 'pucks' from floating to the top of the package during grouting operations. In contrast, drums used for encapsulation of liquid waste will feature a mixing paddle, and drums containing encapsulated solid waste may be equipped with a centralising feature such as a basket.

Figure 1: - 500 litre Drum with supercompacted waste

Figure 2: - Nirex 3m³ Drum

The 3m³ Drum is used primarily for mixing larger volumes of liquid wastes. An example of the 3 m³ Drum is shown in Figure 2.

The 3m³ Box, shown in Figure 3 during prototype testing stage, is becoming more widely used as an efficient way of dealing with a wide range of solid decommissioning wastes. It is important to note that the external dimensional envelopes and handling features of the 3m³ Box and 3m³ Drum are the same. Container standardisation provides real benefits in terms of efficiency and cost of future waste management, through optimisation of handling equipment and the envisaged handling operations. Wastes are currently being processed and packaged at a number of locations around the United Kingdom. Nirex's drive for container standardisation ensures that future waste management will be streamlined with reduced exposure time, improved efficiency and reduced cost, without detriment to safety of operations.
2.2 Storage, Transport and Disposal Containers

In addition to the containers described in the previous section, Nirex has been working on the development of standard waste containers which are also Transport Containers in their own right and can be used for storage, transport and eventual disposal. The current philosophy is to develop two complementary containers, based originally on the standard ISO freight container codes. These are colloquially known as the 4 metre Box and the 2 metre Box. It is envisaged that both the 4 metre Box and the 2 metre Box will be approved as IP-2 containers under the IAEA Transport Regulations.

To ensure that the limits of IAEA Transport Regulations are met, the design of both containers includes provision for casting various thicknesses of reinforced concrete shielding inside the stainless steel cladding. The shielding thickness is determined, based on the activity of the payload, by the waste packager. Such a flexible approach allows the packagers to utilise these containers in the most efficient way in terms of mass and volume of packaged waste.

Again, a degree of standardisation is applied during the development work on both containers. As suggested by their designation, their length will vary. They will however be of the same width and height, and will share the same handling and tie-down features. The larger 4 metre Box, shown in Figure 4, is currently in the final stages of the container approval process, and is already being considered as the potential standard waste package for a number of leading large-scale decommissioning projects.

Figure 3:- 3m³ Box prior to drop test

Figure 4:- Prototype 4 metre Box in transport
The need for development of the last in the range of standard waste containers, the 2 metre Box, has recently been confirmed through interaction between Nirex and the waste packagers. The 2 metre Box, the outline of which is shown in Figure 5, is envisaged as a half-length version of the existing 4 metre Box. When in service it will be used for packaging high density wastes or for use in areas with limited access where the use of the 4 metre Box would not be practicable or possible.

2.3 Practical Experience and Packaging Advice

The practical experience gained from the design, development, prototype manufacture and testing of the various containers is used by Nirex to provide practical advice to the waste packagers on various aspects of container development. One example of this approach is the recently published Best Practice Guide - Welded Joint Design and Manufacture for Stainless Steel Containers [3]. This is intended as a reference document for waste container designers, manufacturers and waste packagers alike. It contains practical advice on the best approach to fabrication with stainless steel, covering the whole process from the initial joint design to the final weld testing. For example full penetration butt welds, where crevices which act as corrosion sites are avoided, are the preferred joint type for the fabrication of structural members and the containment boundaries. Additionally the integrity of this type of joint is readily verifiable using a range of NDT methods, e.g. radiographic inspection would be suitable for welds in materials less than 6mm thick, e.g. container skin. On the other hand ultrasonic inspection is typically restricted to welds in materials thicker than 6mm, and is therefore likely to be used in structural rather than containment areas.

Figure 5: Outline of the 2 metre Box

The knowledge gained from research and development of waste containers is also used to support packaging advice given to customers as part of packaging proposal assessment during the Letter of Comfort process as described in [4]. For example, the advice given to customers includes recommendations for weld integrity most suited to any particular welded joint. The advice given might also reflect the view that the extent of welding on waste containers should be reviewed and minimised by, for example, the use of standard sections or pressed component parts.

3. References

DEVELOPMENT OF A SYSTEM TO MAINTAIN, MONITOR AND CHECK WASTE PACKAGES DURING INTERIM STORAGE

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Abstract
This paper describes the development of a systematic approach to the maintenance, monitoring and checking of the integrity of waste packages during interim storage. The approach consists of: the control of environmental conditions in stores; monitoring of waste package condition during storage; and; application of test techniques prior to transport. Because of the long timescales over which waste packages will be stored, application of best practice is considered prudent. Observations to date indicate that these measures are robust. Monitoring of inactive waste packages will continue, and will be supplemented by information from coupons and dummy packages in active stores. This should enable guidance to be refined and updated in the future.

1. Introduction

The role of Nirex is to provide the United Kingdom with safe, environmentally sound and publicly acceptable options for the long-term management of radioactive materials. This includes operational and decommissioning wastes from nuclear power plants and other wastes from industrial, defence and medical applications involving radioactivity. To facilitate conditioning and packaging of radioactive wastes into a form that meets requirements for passive safety and long-term management including potential disposal, Nirex has defined packaging standards and specifications.

Packages of conditioned waste are currently held in interim storage in surface stores. It is anticipated that there will be a period of interim storage lasting several decades, prior to the availability of a disposal facility, followed by a second period of similar length, during which the facility is operational, and then by a further, possibly longer period during which the facility remains open and waste packages can be monitored and retrieved if required. The key performance requirements are for waste packages: to maintain their integrity during interim storage; to provide containment of the radioactivity; and, to facilitate future handling and retrievability for future phases of waste management.

Nirex conducts independent assessments of proposed waste packaging activities and provides waste producers with advice to guide package design and associated research and development. Where appropriate the assessment may lead to endorsement of the packaging plans. Nirex is developing a framework, in conjunction with the UK nuclear industry, to provide guidance on best practice in interim storage. The objective is to ensure that waste packages will be able to meet Acceptance Criteria for onward phases of waste management.

2. Guidance on Storage Environmental Conditions

In the UK, the primary material used for manufacture of intermediate level waste (ILW) containers is stainless steel. The environmental conditions to which the filled packages are
exposed are important in determining their corrosion performance, which will affect their long-term integrity. Nirex has therefore developed guidance for store designers and operators regarding the factors that should be considered in respect of environmental conditions in conditioned waste stores. This is intended to form an input into the development, by waste producers, of strategies for the storage of radioactive waste, packaged in stainless steel waste containers. It is focused on the above-ground storage phase of radioactive waste management but it is also relevant to environmental conditions during subsequent phases of waste management.

To provide context, the guidance on environmental conditions includes a discussion on the influence of environment on package integrity by addressing:

- **Materials**: The most commonly used stainless steels for radioactive waste packages are the standard austenitic grades 304L and 316L (1.4307 and 1.4404 respectively).
- **Atmospheric corrosion mechanisms**: The passive film on stainless steels restricts general or uniform corrosion over the whole surface and the corrosion resistance in normal atmospheric conditions is therefore high. On a free surface, localised corrosion takes the form of small depressions or pits in the surface (pitting corrosion) or, under certain conditions, could be manifested as cracking in regions of tensile stress (atmospheric stress corrosion cracking (ascc)); it can also occur at crevices between mated surfaces (crevice corrosion).
- **Environmental factors affecting atmospheric corrosion**: The main factors for stainless steel are:
  - temperature and relative humidity: these affect the amount of water that will be available on the surface of containers to support corrosion;
  - atmospheric pollution: pollutant aerosols and gases may enhance stainless steel corrosion rates; chloride is an aggressive ion and is transported in air far from the sea or in discharges from industrial processes;
  - microbial activity: microbially influenced corrosion results from their ability to change the nature of the local environment, such that it promotes corrosion of the underlying metal;
  - radiation fields: the main effect may be to produce nitric acid from atmospheric nitrogen, oxygen and water.

The guidance focuses on the control of environmental conditions in waste stores. This is based on surveys of the literature on atmospheric corrosion of stainless steel and experimental studies of corrosion mechanisms commissioned by Nirex. It is supported by monitoring of waste packages and environmental conditions in an inactive storage building [1]. The key factors are:

- **Moisture and temperature control**: It is important to guard against exposure of waste packages to liquid water, since localised corrosion of stainless steel is more likely if a water film is present, particularly if the water were to penetrate into crevices. Studies of ascc have identified ranges of relative humidity where the risk of initiation is greatest, due to absorption of moisture by deliquescent chloride salts.
- **Air filtration**: Stainless steels should have clean surfaces to offer optimum corrosion resistance; store designers should consider the design of store buildings, filtration and ventilation systems and the effect these may have on internal chloride levels.
- **Care of stainless steel**: Ferrous particles from tools or handling equipment can become embedded in the surface of stainless steel at pressure points and suffer from corrosion, which can initiate corrosion of the underlying stainless steel. Surface contamination or application of coatings during storage should be avoided.
- **Transient conditions**: Consideration should be given as to how transients (e.g. rapid changes of humidity or temperature) might affect the corrosion behaviour of stored waste containers.
3. **Guidance on Monitoring**

Monitoring the condition of ILW packages during storage is an important element of a strategy to ensure that they can provide the longevity required [2,3]. In particular, an appropriately designed monitoring programme can:

- provide early warning of changes to the structural and containment elements and allow actions for any necessary treatments to be put in place;
- provide feedback to waste container and package designers and store designers and operators to reduce the potential for repeat occurrence of failures;
- satisfy licensing body requirements and demonstrate legal compliance;
- confirm performance remains consistent with design intent;
- provide public reassurance that waste packages will provide the necessary physical containment.

Nirex has developed guidance on the monitoring of waste packages in waste stores. A number of techniques are available, or could be extended for remote application, which could be used for monitoring the condition of stainless steel packages within a waste store, including:

- container (visual examination, sacrificial coupons, corrosion indicators);
- seals and filters (micro-hardness, pressure drop, etc.)
- wasteform (dimensional changes, dummy packages, NDT techniques, direct observation).

Baselining should be considered, to establish a starting point against which changes can be assessed. An alternative to using real packages would be to place dummy packages within the waste store, i.e. a package with identical design features to a real package subjected to representative environmental conditions. The corrosivity of environments in which radioactive waste containers are stored and the external corrosion rates of containers could be assessed by exposing standardised coupons of the same material to the storage environment and monitoring their condition.

Monitoring techniques should be worked-up into a practical monitoring strategy. Pre-requisites for the monitoring strategy will be specification of the storage environmental conditions and an assessment of the performance of waste packages under these conditions. The different stages in the development of a condition monitoring strategy may be summarised as:

- Identification of potential degradation mechanisms.
- Identification of monitoring techniques to detect signs of degradation.
- Estimation of package reliability based on degradation rate data.
- Determination of monitoring frequency as a fraction of the time between the point of detection of degradation and the expected time of waste package failure.
- Identification of a means of selecting a representative sample of packages to be monitored.

The derivation of a monitoring strategy requires consideration of several other factors relating to the feasibility of its implementation, such as:

- The costs and radiological doses associated with different monitoring operations;
- The potential for damaging waste packages during monitoring operations, including physical retrieval for inspection;
- The practicalities of operating and maintaining monitoring equipment.
4. **Guidance on Quality Checking before Transport**

The transport of radioactive material through the public domain, within the UK, is governed by the relevant IAEA transport regulations [4], which are enforced by the Department for Transport (DfT), the UK Competent Authority. In carrying out that function they perform both an approval and a compliance role. Only those transport package designs which have been approved by a Competent Authority, and comply with the terms of the approval certification, may be used for transporting radioactive material. Compliance with the terms of the approval certification must be demonstrably assured at the time of transport or transport cannot take place. All these tests and checks will be closely linked to, and governed by, condition monitoring results.

Nirex has developed guidance to waste producers, store designers and operators on the inspection, testing and recording requirements which will have to be implemented at the time of transport to allow the transport of waste packages from their storage facility. It therefore provides an input to the development of strategies for the storage of radioactive wastes. It is recognised that there could potentially be many changes to the transport regulations before the packages are transported, and that these could affect the content of the guidance. Topics addressed in the guidance include containment, shielding, heat generation, gas generation, dose rates, package condition, and lifting and handling requirements. Each package needs to have an unique identification number, and systems put in place to ensure each package can be identified with its own records.

5. **Summary**

A systematic approach to the maintenance, monitoring and checking of the integrity of waste packages during interim storage has been developed and described. In support of UK waste packaging operations Nirex has developed guidance on:

- control of environmental conditions;
- monitoring of waste package condition, and;
- test techniques prior to transport.

Because of the long timescales over which waste packages will be stored, application of best practice is considered prudent. Observations to date indicate that these measures are robust. Monitoring of inactive waste packages will continue, and will be supplemented by information from active stores (e.g. from coupons and dummy packages). This should enable guidance to be refined and updated in the future.

6. **References**


THE IDENTIFICATION OF RADIONUCLIDES RELEVANT TO 
LONG-TERM RADIOACTIVE WASTE MANAGEMENT IN THE UK

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Oxfordshire OX11 0RH, United Kingdom

Abstract
The means of deriving a list of relevant radionuclides for waste management in the UK from
the 2345 known is described and the list reported. The basis for this list is a conservative
estimate of the total inventory for wastes, reviewed against criteria derived for safety
relevant situations. This listing is kept under review to take account of developments in
regulations and the Nirex generic concepts.

1 Introduction
The role of Nirex is to provide the United Kingdom with safe, environmentally sound and
publicly acceptable options for the long-term management of radioactive materials. These
materials include operational and decommissioning wastes from nuclear power plants and other
wastes from industrial, defence and medical applications involving radioactivity. Nirex provides
guidance and advice to the UK nuclear industry in the optimisation of packaging operations. To
ensure consistency and transparency, Nirex has developed a methodology for identifying the
radionuclides relevant to future phases of radioactive waste management.

As there are a very large number of radionuclides that could arise in radioactive wastes, Nirex has
undertaken a programme of work to determine their significance. This paper describes the
derivation of the list of relevant radionuclides. This has been developed through consideration of
their safety impact on a range of transport, repository operation and post-closure situations.

2 Process for Identifying Relevant Radionuclides
There are a number of internationally recognised databases of known radionuclides. The JEF-2.2
radionuclide database identifies a total of 2345 known radionuclides, of which 273 radionuclides
have half-lives in excess of ten days (this cut-off being significantly less than any conceivable
cooling period prior to waste management operations) [1]. To screen this list of 273 potentially
relevant radionuclides, the following step-wise process was used:

- produce conservative estimates of the total inventories of all 273 potentially relevant
  radionuclides in UK radioactive wastes;
- identify and define an appropriate range of safety relevant situations;
- determine the concentration of each potentially relevant radionuclide that would have a
  significant impact on safety for each situation (the limiting concentration);
- screen the inventory against limiting concentrations to establish those radionuclides for
  which there is a significant risk of exceeding a safety limit (the relevant radionuclides).

The methodology is based on consideration of average radionuclide concentrations, derived from
the inventory of each radionuclide and an appropriate volume of waste.
3 Derivation of Inventory Data

In the derivation of the total inventories, the following sources of waste were considered:

- commercial reactor fuels;
- decommissioning wastes from commercial reactors;
- 'exotic' radionuclides produced in spallation neutron source targets;
- naturally-occurring long-lived radionuclides;
- other non-fuel cycle wastes.

The study focuses on reactor fuels and decommissioning wastes. The inventories for reactor fuels and decommissioning wastes were derived as follows.

3.1 Calculations for Commercial Reactor Fuels

Calculations were performed for commercial reactor fuel elements to predict the inventory associated with Magnox, AGR, BWR and PWR reactor fuels, fuel cladding and other related wastes. Losses of activity both in the reactor and during reprocessing were not considered, thereby providing a conservative estimate of the total activity associated with wastes. The inventories of fission products and actinides arising from the fission of uranium and the higher actinides, such as plutonium, were calculated using the FISPIN7 code [2]. The inventories of activation products and any fission products and actinides generated from trace amounts of thorium were calculated using FISPACT-97 [3].

3.2 Calculations for Reactor Decommissioning Wastes

Calculations were performed for the main sources of decommissioning waste from UK commercial reactors. All calculations used FISPACT-97 to model activation and the generation of fission products and actinides from traces of thorium and uranium in the materials. Representative calculations were performed for each of the following six waste types:

- Magnox reactor moderator and reflector graphite;
- Magnox reactor mild steel (ILW);
- Magnox reactor stainless steel (ILW);
- AGR moderator and reflector graphite;
- AGR mild steel (ILW);
- AGR stainless steel (ILW).

The total activities associated with each of the six waste types were based on the total mass of material expected to arise from all UK power stations.

4 Definition of Safety Relevant Situations

A wide range of possible situations may result in safety-related effects, the magnitude of which will depend on the activity involved. A review of regulatory requirements and the Nirex generic concepts identified a total of 67 separate situations or requirements. This initial set of situations was further reviewed to eliminate those situations that were either encompassed by other situations or which did not impose a direct numerical limit on any radionuclide. In this manner, a total of 21 key situations were identified, categorised as follows:

- requirements imposed by IAEA Transport Regulations (6 situations);
- situations arising from the Nirex generic operational safety assessment (6 situations);
- Nirex waste package specifications (4 situations);
- situations arising from the Nirex generic post-closure performance assessment and related regulatory requirements (5 situations).
5 Testing for Relevance

The basis for establishing the relevance of a radionuclide is the limiting concentration, defined as the concentration of that radionuclide that would give a safety impact equal to the limit for the situation being considered (for example a heat output of 50W). For each of the 21 key situations, the limiting concentrations have been derived from the appropriate regulations and specifications or by consideration of the various generic safety cases. Where appropriate, the derivations were based on the consideration of Nirex standard waste containers.

For each situation, the average concentration of each radionuclide, derived from the estimated total inventory and the volume of waste, is scaled by the limiting concentration. The scaled values are then rank-ordered and compared with a defined insignificance factor (IFn) to judge which radionuclides are relevant for that situation, as shown in Figure 1. This method takes into account the presence in a waste of a range of different radionuclides.

If the scaled value exceeds unity, then the safety limit has been reached. Conversely, to allow for uncertainty, the demonstration of non-relevance requires that IFn be a small fraction of unity. Consequently, for situations related to transport and operations, an IFn of 10⁻² was adopted, whereas for post-closure situations an IFn of 10⁻³ was used.

6 Definition of Relevant Radionuclides

Package types requiring transport in an IAEA Type B container ("Type B") and industrial packages (IP) were considered separately. This resulted in two slightly differing lists of relevant radionuclides, which were merged to form a single listing of relevant radionuclides. A total of 112 relevant radionuclides have been identified, as listed in Table 1. Of these, seven are short-lived daughters that will be in equilibrium with the parent radionuclide. For each radionuclide, Table 1 also identifies the type of situation for which the safety limit has been judged to be the most demanding. These situations are identified separately for Type B and IP packages (some radionuclides are significant only for one or other of the package types, and some for both).
Table 1: List of 112 Relevant Radionuclides (short-lived daughters shown italicised) and situations for which they are most significant (Type B package/IP package)

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<th>O/O</th>
<th>Ba133</th>
<th>O/O</th>
<th>Pb210</th>
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</table>

T – situations related to Transport safety and regulations
O – situations related to Operational safety
P – situations related to Post-closure safety
dash – not significant for the relevant package type

7 Summary and Conclusions

The means of deriving a list of relevant radionuclides from the 2345 known has been described and the list has been reported. The basis for this list is a conservative estimate of the total inventory for wastes, reviewed against criteria derived for safety relevant situations. This listing is kept under review. Currently, further sources of waste, for example experimental reactors, are being examined to ensure that any additional relevant radionuclides are identified. The definitions of the safety relevant situations also will be reviewed to take account of developments in regulations and the Nirex generic concepts.

8 References

Radioactive beryllium from fission and fusion reactors constitutes a special waste stream for which at present no standardised management route exists. In this paper, we discuss the advantages and drawbacks of five conditioning methods in the framework of final geological disposal: the option of ‘no conditioning’ or direct disposal, cementation, bituminisation, vitrification, and phosphatisation. Because of the high chemical toxicity of beryllium metal and its unfavourable corrosion characteristics at extreme pH’s, conditioning methods that leave beryllium in its metallic state are to be avoided. Incorporation of the beryllium waste in a silicate glass or in a phosphate matrix is therefore preferred.

1 Introduction

Both existing fission reactors and future fusion reactors are sources of large quantities of radioactive beryllium waste. The management of this beryllium waste will either result in the recycling and reuse of the beryllium in the nuclear industry, or in the permanent disposal of the conditioned waste. To provide technical and scientific support for the beryllium waste management, SCK•CEN has initiated a research programme with a twofold goal: (i) to characterise existing beryllium waste from the Belgian materials testing research reactor BR2, and (ii) to propose conditioning methods for beryllium waste from both fission and fusion reactors. Beryllium waste from fusion reactors has been discussed elsewhere [1, 2]. This paper will focus on the conditioning of beryllium waste from a materials testing reactor.

2 Characterisation of beryllium waste

The BR2 (Belgian Reactor 2) is a beryllium-moderated and water-cooled high flux materials testing reactor operated at the Belgian Nuclear Research Centre. The lifetime of a beryllium moderator assembly (matrix), taking into account the BR2 operation scheme and the fluence limit, is about 15 years. After this time, the metal embrittles and cracks due to the production of helium by neutron irradiation. Up to now, two replacements of the matrix were performed [3]. The irradiation conditions of the beryllium during its lifetime inside the reactor core are well documented [4]. The chemical composition of the beryllium was analysed for two matrices, before and after irradiation. From these measurements, we can conclude that the concentration of trace elements in the beryllium does not significantly change during 15 years of irradiation, even in contaminated surface samples [4].

The helium content of the second beryllium matrix was determined by the hot vacuum extraction technique [5]. This allowed to experimentally verify the fast neutron fluence received by the moderator at the specific sampling location of the analysed beryllium samples, as the produced quantity of He (and 3H) in the irradiated beryllium is a linear function of the fission fluence [6]. We performed a radiological characterisation of the beryllium waste using γ-spectrometry and liquid scintillation counting (for 3H). The results of activation calculations for the most important activation products were compared to measured values, and usually a good agreement between the measured and calculated values was observed. The most important nuclides contributing to the specific activity of the beryllium are 3H, 60Co, 65Ni, and 137Cs. Table 1 summarises the specific activities of the most abundant radionuclides.
Table 1. Specific activity of the second BR2 beryllium matrix at the maximum flux plane for selected radionuclides (the fission fluence at the maximum flux plane was 5.23 \times 10^{22} \text{n.cm}^{-2})

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Specific activity at maximum flux plane (Bq.g{\text{-}1})</th>
</tr>
</thead>
<tbody>
<tr>
<td>{\text{^{60}Co}}</td>
<td>4.3 \times 10^{7}</td>
</tr>
<tr>
<td>{\text{^{63}Cu}}</td>
<td>7.5 \times 10^{6}</td>
</tr>
<tr>
<td>{\text{^{85}Sr}}</td>
<td>6.1 \times 10^{5}</td>
</tr>
<tr>
<td>{\text{^{134}Cs}}</td>
<td>3.6 \times 10^{4}</td>
</tr>
<tr>
<td>{\text{^{137}Cs}}</td>
<td>2.0 \times 10^{6}</td>
</tr>
</tbody>
</table>

3 Interim storage and direct disposal of beryllium waste

At present, beryllium waste is most often stored in the pools of the reactors, as no standard route exists for its management because of its toxic nature and high tritium content. The beryllium waste from the first moderator of BR2 (535 kg of beryllium) was placed in PAMELA containers\(^1\), using dry clean quartz sand to fill the voids between the metallic parts and the container. The sand ensures the mechanical stability of the packed waste. The hollow geometry of the beryllium waste results in a very low filling degree: only 7-10 % of each canister can be used for beryllium packing. The filled PAMELA canisters are stored under ventilated conditions to prevent tritium build-up. Although this solution is viable for interim storage, it is not usable for the final geological disposal of beryllium waste\(^2\). The main problem with the direct disposal of metallic beryllium is anaerobic corrosion with associated hydrogen gas production. At the pH conditions presented by for example a clay disposal environment, the anaerobic corrosion of beryllium can generate large amounts of hydrogen, which could eventually lead to gas-driven transport of radionuclides through the clay layer\(^7\). Furthermore, the chemotoxicity of beryllium is of concern in the case of direct disposal. To avoid these problems, appropriate conditioning methods have to be found. In this paper, we consider the following options: cementation, bituminisation, vitrification, and phosphatisation.

4 Cementation

Cementation is a proven technique for the conditioning of diverse waste streams\(^8\). It consists of immobilising waste with a cement matrix and has been applied during the last decades to low- and intermediate-level radioactive waste. The basis of many types of cement used for waste conditioning is ordinary Portland cement (OPC), but often additives such as fly ash and blast furnace slag, are incorporated in the cement matrix to improve its characteristics. Cementation is a simple, relatively inexpensive technique, resulting in a non-flammable matrix with good durability. The main drawback in the case of beryllium however, is the high pH (12,5) of the cement matrix. In strongly alkaline conditions, metallic beryllium is subject to accelerated general corrosion (for details, see [2]). As mentioned above, the associated production of \(H_2\) could lead to gas-driven transport of radionuclides through the clay layer.

---

\(^1\) The PAMELA container is a stainless steel (AISI 304) 200 litre drum.

\(^2\) At present, no acceptance criteria for radioactive beryllium waste exist in Belgium and there is no formal basis on which to conclude that the beryllium waste should be disposed in a geological repository. In this paper, we consider the disposal in the Boom Clay as a working hypotheses, without any implication for the actual disposal option for radioactive beryllium waste.
5 Bituminisation

This process has been applied to the conditioning of low-level and intermediate level sludge, evaporator concentrates and ion-exchange resins [9], but never to beryllium. The bituminisation process is technically complex. The encapsulation of beryllium in bitumen could present practical problems. The beryllium from fusion reactors will probably be in the form of large metal pieces, and hot bitumen will rapidly harden at contact with the cold metallic pieces, preventing bitumen from completely filling the voids between the beryllium parts. Furthermore, as beryllium remains in its metallic form, corrosion and associated gas generation cannot be excluded. However, since bitumen does not impose extreme pH values, the corrosion rate will be lower than in cement.

6 Vitrification

Vitrification is currently utilised for the conditioning of highly active fission product residues resulting from the reprocessing of spent fuel from the Belgian nuclear power plants [10]. One of the reference processes used today consists of two steps, starting with the isolated fission products from the reprocessing plant, which are in the form of concentrated nitric acid solutions. The nitric solution of fission products is fed continuously to a rotating calciner, in which the solution is evaporated, and the residue dried and partially calcinated. The calcinated residue is mixed with other glass constituents to reach the desired composition. It is subsequently heated, melted, and the liquid glass is then poured into a canister [10]. A vitrification process for the beryllium waste from fusion reactors could be based on this type of process and consist in dissolving the beryllium in nitric acid, calcining, and mixing the thus obtained beryllium oxide with molten glass.

The use of glass as an immobilising matrix for the deep geological disposal of high-level radioactive waste in clay layers has been extensively investigated at SCK•CEN [11], including the characterisation of the vitrified waste forms [12]. The long-term behaviour of beryllium in the glass matrix is expected to be very favourable. As beryllium is fully oxidised during the vitrification process, corrosion of metallic beryllium and the associated hydrogen production will be suppressed during geological disposal. The dispersion of beryllium is controlled by the corrosion resistance of the glass. The vitrified waste is very resistant to leaching in Boom Clay, with corrosion rates as low as 1 μm/year [13].

7 Phosphatisation

Phosphate ceramics are currently developed at IPN Orsay (France), for the conditioning of uranium, plutonium, and neptunium. It is a wet process in which the dissolved species are mixed with phosphoric acid, evaporated, and sintered at 300-800 MPa and 1250°C [14]. Beryllium might be a suitable material to be conditioned in phosphate. Indeed, Be may substitute for calcium on the Ca\(^{2+}\) sites in Apatite (Ca\(_6\)(PO\(_4\))\(_3\)(F,Cl,OH)). As beryllium and calcium are both alkaline earth metals, the synthesis of Be phosphates might then be possible.

8 Conclusion

Because of the high chemical toxicity of beryllium metal and its unfavourable corrosion characteristics at extreme pH’s, conditioning methods that leave beryllium in its metallic state are to be avoided. Incorporation of the beryllium waste in a borosilicate glass or in a phosphate matrix is therefore preferred. In view of the large industrial experience with the vitrification of high-level radioactive waste, we will further evaluate vitrification as the preferred conditioning option for radioactive beryllium.
References


EXTENTION OF THE MEASUREMENT RANGE OF THE IPAN/GEA SYSTEM FROM mg Pu TO 200 g Pu FOR THE NON-DESTRUCTIVE ASSAY OF 200 l AND 400 l WASTE PACKAGES

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Abstract
Belgoprocess experienced the need for an extension of the operational range of its IPAN system from the sub gram to several hundreds of grams of Pu content in waste packages.
A new analysis method is being developed. This method results in a new software that uses the raw data from the system and only one parameter from the original software.
Calibrations were performed on mock-up drums and validation tests on real waste.

1 Introduction
The Belgoprocess IPAN/GEA system is a combined passive-active neutron and gamma system designed in the early nineties to measure Pu-contents at the mg to g level. Today, a new waste treatment project of alpha contaminated waste ("A3X-project") expressed the need for a characterization up to 200 g of Pu. The company strategy was to extend the operational range of the existing system over purchasing a new system.

2 Requirements and conditions
To use this system for the assay of waste packages containing grams of plutonium we were faced with some hard- and software limitations. Apparently new calibrations were necessary since tests showed that no meaningful results were obtained with the originally installed calibrations. Since the IPAN calibrations are used in combination with different algorithms containing empirically determined parameters stored in the software itself, we were faced with the need to start from scratch to set up new calibrations and a corresponding analysis method.
The IPAN/GEA system software is written in C++ and runs still in a DOS environment. We did not want to alter the original software and neither were we in favour of having different versions of this software running. So we decided to develop a completely new analysis software that would run independent of the IPAN software for the analysis of the data and could be used offline. Moreover this approach would give full flexibility to tailor the analysis method.
The communication channel between the new analysis software and the IPAN system are the data files that were obtained from the IPAN software. These files contain all raw counting data from which coincidence count rates can be computed. Since matrix correction is an indispensable requirement in neutron assay of waste packages with varying matrices we aimed at using the parameter "Moderator Index" (MI) computed by the IPAN code. However tests showed that the MI parameter is biased by the plutonium loading in the drum. For this problem we also didn't want to alter the settings for the passive measurements since this would require to alter the settings each time one switches between passive and active measurements, with all corresponding risks and effects on the calibrations. Another approach was developed to get around this problem.
3 Working method

We chose a straightforward calibration and analysis method. The measurement quantity used to relate to the plutonium mass is the REALS rate computed from the multiplicity distributions recorded by the Programmable Multi-Channel Coincidence Counting Module (PMCCM) of the IPAN system. The PMCCM is a typical shift register based multiplicity counter that records the histograms of the detected pulse multiplicity in two small time windows (gates). The first window is the (R+A) gate and starts counting pulses some microseconds after each pulse. The second window is the Accidentals gate (A) gate and is triggered by clock pulses. The histogram gives the multiplicity up to 32. From these sets of numbers the REALS rate \( R \) is readily computed using the relation:

\[
R = \frac{1}{t_m} \sum \frac{(R+A) - (A)}{(R+A) + (A)} \frac{1.28 \sum \frac{I_i}{t_m}}{}
\]

Where \( p_n \) and \( q_n \) are the normalised counts in respectively in the (R+A) gate and in the (A) gate:

\[
p_i = \frac{(R+A)}{\sum{(R+A)_i}} \quad \text{and} \quad q_i = \frac{(A)}{\sum{(A)_i}}
\]

\( t_m \) is the measurement time. The second factor between brackets gives the total count rate. The factor 1.28 accounts for the fact that the accidental window is triggered each 128 \( \mu \)s while it is 100 \( \mu \)s long.

The aim of the calibrations is to relate \( R \) to the Pu-240eq mass, accounting for matrix effects. One calibration \( R \) (Pu-240eq) is used as the reference calibration. Matrix effects are accounted for by using the MI provided by the IPAN software. MI is computed from the normalised counts in the flux monitors in two time windows respectively corresponding to the detection of epithermal neutrons, shortly after a neutron burst has been fired by the neutron generator, and the thermal neutrons, some long delay after the burst was fired.

A second calibration is set up \( CR \) (MI) where \( CR \) is a correction factor expressing the ratio between the REALS rate obtained in one matrix compared to the REALS rate in the reference matrix. The calibration measurements were made using fuel pins of SCK’s VENUS reactor. These fuel pins had been used in the past for optimising MOX fuel and was sufficiently well characterised for this calibration. Up to 15 fuel pins were used to span the mass range to 40 g Pu-240eq. For the calibration measurements, the fuel pins were inserted in vertical tubes in the calibration drums using configurations that simulate the homogeneous source distribution.

4 Results and discussion

Basic calibrations \( R \) (Pu-240eq mass) were determined for different matrices. The reason for making the calibrations in function of mass was to investigate if neutron multiplication is present. No substantial effects of neutron multiplication where observed and the calibration curves could be approximated sufficiently well with a straight line. The figure 1a and b shows the calibrations for respectively the 400 L drum geometry and the 200 L drum geometry.
While monitoring the precision of the MI we discovered that it is a non negligible function of the plutonium content in the waste drum and hence is no unique indication of the matrix moderation which is required to make a sound matrix correction. In figure 2 the MI for different matrices and as a function of the plutonium loading is shown.

It is clearly observed that the MI computed by the IPAN software decreases with increasing plutonium loading. The relation between MI and Pu-240eq mass is well approximated by a straight line, and it turns out that the slope is a linear function of the plutonium loading. Faced with these findings we opted for an iterative determination of the Pu-240eq mass in an analysis. The uncorrected moderator index is first used to determine a first estimation of the Pu-240eq mass. This first estimation of the Pu-240eq mass is then used to correct the MI from which a new estimate of the Pu-240eq mass can be made. This process is repeated until the Pu-240eq mass converges to a fixed value. Typically we used 10 iteration steps.

Another problem was encountered while setting up the calibration CR (MI) for the 200L configuration. It turned out that no smooth calibration curve can be obtained and that the data, apparently, are subdivided in different classes. We can clearly identify a curve for the concrete matrices and another for the PVC and bitumen matrices (see figure 3). The apparently relatively large scatter in the data, obtained if one assumes a single calibration curve, that fits all data
points, will result in a large uncertainty on the computed Pu-240eq mass. For the moment we do not have a clear idea what could cause this non-smooth evolution of CR (MI). Since measurements are aimed for relatively low MI we opted for a calibration curve based on the PVC matrices.

![Figure 3 Duality of the calibration data CR (MI).](image)

An uncertainty budget was set up accounting for the most prominent uncertainties. The main source of uncertainty is an unknown source distribution deviating from the homogeneous source distribution simulated in the calibration experiments.

Based on these calibrations test measurements were initiated on real waste drums for which declared values exist. The declared values could be reproduced within an uncertainty range of 50% with some outliers.

5 Conclusion

A new software and analysis method were developed to analyse the raw count rate data of the IPAN system computing the REALS rate from the multiplicity data. Two basic calibrations are set up per configuration: the reference calibration expressing the relation between REALS rate and the Pu-240eq mass for the reference matrix and a calibration expressing the correction factor CR as a function of the moderator index. The latter turned out to be also a function of the Pu-240eq mass and an iterative approach was necessary to determine the Pu-240eq mass from the REALS rate and the unbiased moderator index. For the range of waste matrices aimed to be analysed with this approach we estimated the uncertainty to be generally less than 50%.
SRWGA: a Comprehensive System for Gamma Characterisation of Radioactive Waste Packages

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Abstract

The SEA Radioactive Waste Gamma Analyser (SRWGA), here described, is the gamma assay system of the ENEA Laboratory for Waste Characterisation, which started the operation in 1996 as a simple gamma scanner and was under a continuous improvement process to become a multi-techniques system as it is now.

In the following a brief description of the techniques implemented is given: Open Geometry, Segmented Gamma Scanning (with multi-transmission correction), Angular Scanning, Low Resolution Emission and Transmission Tomography.

At the end is reported a comparison of the results obtained measuring with each technique available a Waste Package Reference Standard where gamma sources in different radial-azimuthal positions have been located and some consideration on the flexibility of the system.

1 Description of the System

The SRWGA (Fig. 1) was designed for the assay of radioactive waste drums containing gamma emitting nuclides. It accommodates drums up to 1500 kg and 220, 400 and 500 litres capacity.

The system operates with an high resolution HPGe coaxial detector, 40% relative efficiency, liquid nitrogen cooled, shielded by means of a lead cylinder (10 cm thick) with a rectangular collimation window (2.5 X 10 X 20 cm).

Three load cells are installed under the turntable for measuring the drum mass and one mixed transmission source is provided (In$^{114m}$ plus Ag$^{109m}$, 100 and 10 mCi respectively).

Two synchronised computer units control the mechanics (four engines allowing vertical lift of the detector, vertical movements of the transmission source, rotation and longitudinal movements of the turntable) and the data acquisition and analysis system.

2 Featured Techniques

The flexibility of the system allows to apply four different measuring techniques, each one with his peculiar field of application:

Open Geometry (OG, [1]), for homogeneous wastes. The collimation and the detector to drum distance are such that the drum can be seen fully by the detector, while the drum is rotating. The measuring procedure is constituted by one transmission measurement to calculate the gamma attenuation factor and one emission measurement to calculate the activity inside the drum. This measuring technique relies on two basic hypotheses: uniform activity distribution and homogeneous density distribution for the whole drum.

Fig. 1 - SEA Radioactive Waste Gamma Analyser
Segmented Gamma Scanning (SGS, [1]), for nearly homogeneous wastes. The collimation and the detector to drum distance are such that only a slice of a drum can be seen by the detector, while the drum is rotating. The measuring procedure is constituted by 11 transmission measurements (with the drum rotating and the detector and transmission source lifting at the end of each step) to calculate the gamma attenuation factor for each segment (a logarithmic curve of transmission vs. energy giving the transmission factor for each energy of the emission spectra) and 11 emission measurements to calculate the activity (corrected for the attenuation) for each segment: the sum of all contribution gives the total activity inside the drum.

The basic hypothesis are uniform activity distribution and homogeneous density distribution for each segment in which the drum is subdivided.

Angular Scanning (AS, [2]), for wastes with non-uniform activity distribution. The geometrical configuration is the same as SGS. The measurement procedure is the following: the rotation of the sample is partitioned into \( n \) discrete angular steps \( \Delta \theta \), such to have \( n \Delta \theta = 2\pi \), and for each of these steps a gamma spectrum is recorded. The analysis of these spectra allows to plot peak count rates against the angular displacement. The shape of this plot depends on the spatial distribution of the nuclide activity dispersed in the volume of the involved segment: it gives the experimental profile, from which number, position and activity of eventual hot-spot(s) can be recognised.

The basic hypothesis is the uniform density distribution for each cylindrical segment of the drum.

Transmission and Emission Computerised Tomography (TCT-ECT, [3]), for every kind of wastes. Transmission Computerised Tomography, by means of combined axial and rotational movements of the drum with respect to the Detector-Transmission Source direction, generates slices of the two-dimensional distribution of attenuation values of the matrix within each segment; the actual procedure foresees 11 axial steps, each one subdivided into 18 rotational spectra acquisition, so that a spatial resolution of 5 X 5 cm is achieved. Relying on results obtained by the TCT, the three-dimensional distribution of nuclide activity can be reconstructed by means of ECT. The same measuring procedure as for TCT is implemented, except that the drum has to be partitioned into cubic volume elements (voxels).

3 Experimental set-up

In order to make a comparison between the different techniques, a drum containing a neoprene homogeneous matrix and radioactive point (certified) sources placed in two different geometrical configurations has been set-up.

The configurations selected for the experimental measurements (as example, in fig. 2 Configuration A is reported) have been designed to evidence the problems which characterize measurement techniques such as interferences or "overlaps" between sources placed in different segments or in different radial positions. The final goal is to reproduce in this way the dishomogeneities of the activity of real wastes and the relative effects. The configurations are the following:

**Configuration A:** \(^{152}\text{Eu}\) (three sources in the 2\(^{nd}\) segment at radial position A, C and D, total activity 3072 kBq), \(^{137}\text{Cs}\) (one source in the 8\(^{th}\) segment at radial position D and one source in the 7\(^{th}\) segment at position F, total activity 3382 kBq), \(^{60}\text{Co}\) (one source in the 7\(^{th}\) segment at radial position E, total activity 1868 kBq).

**Configuration B:** \(^{152}\text{Eu}\) (one source in the 5\(^{th}\) segment at radial position F and one source in the 4\(^{th}\) segment at position C, total activity 2324 kBq), \(^{60}\text{Co}\) (one source in the 5\(^{th}\) segment at radial position F and one source in the 6\(^{th}\) segment at position D, total activity 5356 kBq).
4 Comparison between techniques

In the following figure the ratio between measured and certified activity for Open Geometry, Segmented Gamma Scanning and Angular Scanning is reported.

As can be seen, in both configuration Open Geometry technique response - based on the hypothesis of uniformity of the package - is really far from the certified activity; the Segmented Scanning behaviour is a little better with a very good result for Eu in configuration B (less than 10% lower than the certified activity): this is mainly due to the position of the source (the detection efficiency corresponding to that position is near to the detection efficiency for an "homogeneous source").

The Angular Scanning presents almost always errors lower than 20%; these results are so appreciable mainly because the matrix is really homogeneous and the correct height of the sources has been predetermined. As a matter of fact this measuring technique may suffer for an incorrect choice of the scanning quote. In this case the quote has been chosen by interpolating with a parabola the cps (counts per second) obtained with Segmented Scanning for different segments and using the parabola maximum as the measuring quote.

The worst result (Co for configuration B) is due to the interference between the sources in the 5th and 6th segments, that gave some problem to the correct identification of the quote to which perform the Angular Scanning.

Figure 3 - Comparison between techniques

Figure 4 shows tomographic images of the sources distribution reconstructed with the ECT software: a quantitative evaluation of activity is not yet possible because the analysis software is...
The map was reconstructed with 198 projections, each of 15 seconds (less than one hour per segment).

5 Conclusions

In the actual situation SRWGA, with only one detector and one transmission source, allows to perform almost all the main gamma measurements techniques for waste characterisation. The flexibility of the system allows to choose the more suitable measuring procedure for each drum to be characterised (timeliness vs accuracy principle), so that the measuring procedure can be selected depending on:

• waste known characteristics as origin, physical and/or chemical parameters, homogeneity, etc;

• accuracy required, because the results accuracy increases going from OG to ECT-TCT through SGS and AS;

• available measuring time: this is one of the main parameter because to characterise a drum are approximately needed 40 minutes for OG, 6 hours for SGS, from 30 minutes to 6 hours (depending on number of segments) for AS, and about 18 hours for a complete TCT and ECT.

Actually, at ENEA laboratory of Casaccia is applied a general measurement procedure that foresees the following steps:

1. SGS is performed as basic procedure: the spectra acquired in emission and transmission are analysed and a “first hypothesis” activity is calculated based on the proper assumption.

2. AS is performed on eventual “suspect segments” in which the activity distribution seems to be not uniform; the results obtained are used to correct the activity measured with SGS.

3. TCT and ECT is performed only on waste drums which origin leads to exclude the possibility of homogeneity and on drums already measured with the other techniques but which results are not reliable.

6 References


Abstract

Ten years ago EN-TRAP was founded on the initiative of the European Commission. Its aim was to promote a European collaboration between the national regulators, licensing authorities and laboratories carrying out checking of radioactive waste packages. Its members are 12 laboratories from different EU member states. The Network is operating via a Steering Committee and Working Groups, that meet on the average between once and twice per year.

The main objectives of EN-TRAP are information exchange and harmonisation between the laboratories. EN-TRAP works on different aspects of quality checking of nuclear waste packages. This paper discusses the main achievements made by the working groups of EN-TRAP.

1 Introducing EN-TRAP and its Working Groups

EN-TRAP is the acronym for European Network of Testing Facilities for the Quality Checking of Radioactive Waste Packages. This network has been established in 1992 on the initiative of the European Commission (EC) with the aim to promote and facilitate collaboration in the development, application and standardization of quality checking of waste packages. The initiative was a direct consequence of the European Commission’s Plan of Action in the Field of Radioactive Waste that recognized the importance of quality checking in the field of nuclear waste management. The EC Plan of Action recommended in 1989 the constitution of an ad-hoc group of experts to advise and investigate the needs, incentives, scope and ways of implementation of a European Network of quality checking laboratories for radioactive waste products. Based on the advice of this expert group, in October 1992 the European Network of Testing Facilities for the Quality Checking of Radioactive Waste Packages was founded by nine laboratories that performed quality checking on behalf of regulatory bodies or authorities. The Network is a non-profit making organization.

The current members of EN-TRAP are laboratories of the European Union Member States that perform examinations of nuclear waste packages. There are also associated members participating in the working groups. Associated members are those institutions or organizations involved in waste management and measurements on wastes, but, are not appointed by the national regulator for independent quality checking of waste, or they are from non-member states. In 2002 EN-TRAP has 12 member laboratories and 5 associated members (the list of participating laboratories is given in the appendix).

EN-TRAP realizes its objectives via an organization consisting of a Steering Committee (SC) and three Working Groups (WGs). The SC decides on the programme of activities, on policy issues and on the membership type of new member institutions. The technical aspects are discussed in the WGs [1,2,3]. Currently 3 WGs exist:
2 Thematic approach and main achievements realised by the working groups

2.1 Achievements by WG A

The aim of the meetings of WG A is to give its participants the possibility to exchange information on their respective NDA methods and their application to certain waste types, and to identify possibilities to improve the quality, efficiency and cost-effectiveness of these NDA methods. So far the main attention was on NDA techniques for low level waste. The meetings of WG A are devoted to certain topics which are of common interest and which are elaborated in a workshop-like manner.

The main topics that have been elaborated so far are:

- Survey of the different gamma and neutron systems used by the members. As a result, a synopsis of respectively the gamma and neutron assay systems, used in the different laboratories participating in the working group, has been set up [4,5];
- A comparison of the differences in national limits for the maximum allowed alpha and nuclide specific activity in waste packages;
- Detection limits and their definition, and how to account for background signals and their variations;
- Calibration methods for NDA systems;
- Discussion and evaluation of data obtained from inter laboratory exercises;
- Uncertainty estimation in gamma assay;
- Possibility and restrictions of tomography in NDA;
- Norms and standards on NDA (ISO standards have been worked out by WG A. Some members of the WG A participate also to the ISO/TC 85/SC 5/WG5 meetings);
- The use of the World Wide Web to present EN-TRAP;

WG A has also been an ideal forum to initiate project proposals for the EC Nuclear Fission Safety Programmes. Since 1995, 5 project proposals have been accepted for funding and were executed. These projects were mainly devoted to improvement of NDA techniques. WG A also organised a workshop on "Past, Present and Future of NDA QA/QC on Radioactive Waste" in June 2000 at NRG Petten.

Accurate assay of waste packages and hence quality checking is an extremely difficult task and ongoing research is devoted to make accurate measurements and sound uncertainty estimations. In the past, uncertainty of NDA on waste packages was more or less overlooked. National regulations more and more now require that measured values be reported together with an extended uncertainty (ISO 17025). Reporting a total uncertainty is not straightforward in NDA of waste packages. Indeed, the uncertainty depends on many unknown parameters of the waste package. Hence the only way to make meaningful uncertainty estimations is to quantify the influence of these parameters which requires sophisticated methods and instruments. An interesting tool, however, is computer simulation, which continues to get more and more attention and also becomes more and more the common practise in many laboratories. Computer simulations are mainly used to optimise systems, predict system response and to estimate measurement uncertainty.
2.2 Achievements by WG B

Destructive techniques are complementary to NDA and are essential to quantify those radio nuclides which cannot be measured, or are difficult to measure, with NDA techniques (e.g. pure \( \alpha \) or \( \beta \) emitting nuclides), but that are important for the evaluation of a safe disposal. Contrary to the relatively limited number of NDA methods many different chemical and radiochemical analytical methods exist and are radionuclide specific. Meetings of WG B revealed that for many analyses each laboratory has its own approaches which are all different and show that no common destructive procedures exist. The aim of WG B is to set up, compare, improve, validate and promote harmonisation of methods applicable to various types of waste streams for treatment and conditioning, for verification and characterisation and for quality control. Fast and selective methods are desirable. In WG B the main attention was devoted to:

- Editing a document compiling the different chemical and radiochemical methods and physical techniques used in the participant's laboratories [6];
- The use of combined radiochemical methods;
- Volatile releases and sampling procedures;
- Sampling of radioactive wastes, representativity and stability of samples;
- Key nuclide measurements;
- Leaching procedures and application;
- The determination of the reliability of methods through the organisation of interlaboratory exercises;
- Possibilities to model the gas release from waste packages.

Until now, 4 project proposals initiated by WG B have been accepted for EC funding. These projects deal with: the evaluation of different sample treatment and analysis steps in fast analytical techniques for the characterisation of nuclear wastes; decontamination techniques for alpha-emitting nuclides in conditioned radioactive waste; analysis automation to speed up characterisation; an interlaboratory test for the analysis of a primary waste flux.

The evaluation of the varying approaches followed by the different laboratories for a more understanding and the ongoing exchange of information undoubtedly benefited the participating laboratories.

2.3 Achievements realised by WG C

This working group deals with the more fundamental aspects of Quality Assurance and Quality Control in nuclear waste management in general. The main objective being the investigation of the requirements of quality checking laboratories to have quality assured procedures and systems of work and accreditation. So far WG C concentrated on:

- Identifying the requirements for quality checking of radioactive waste packages;
- Reviewing procedures, testing and control methods employed by member laboratories and investigating common features;
- Evaluating uncertainties in assessing isotopic inventories of radioactive waste packages.

WG C prepared a document on the relationship between the member laboratories and their respective regulatory authorities, their arrangements for quality control and acceptance of low and intermediate level wastes for disposal or long-term storage [7]. This work was elaborated in the frame work of a concerted action funded under the EC's Nuclear Fission Safety Programme.

3. Conclusions

The Working Groups of EN-TRAP have shown that they are valuable fora for exchange of information between the quality checking laboratories and other parties and they have led to close
scientific and technical collaborations between different members in the field of non-destructive assay and destructive analyses. The information exchange during WGs meetings and the collaboration in EC projects initiated from within the working groups motivated many partners to improve methods based on the ideas of other laboratories.

4. References


5. APPENDIX

Members and associated members of ENTRAP (situation 2002)

- Commissariat à l'Energie Atomique CEA, Centre de Cadarache, France
- NNC Limited, WQCL Winfrith Technology Centre, United Kingdom
- Empresa Nacional de Residuos Radioactivos SA (ENRESA), Madrid, Spain
- Centro de Investigaciones Energeticas, Medio Ambientales y Tecnologicas (CIEMAT), Madrid, Spain
- Belgoprocess, Dessel, Belgium
- Studiecentrum voor Kernenergie/Centre d'Etude de l'Energie Nucléaire, (SCK*CEN), Mol, Belgium
- NRG, Arnhem, The Netherlands
- Ente per le Nuovo Tecnologie, l'Energia e l'Ambiente (ENEA), Saluggia, Italy
- Agenzia Nazionale per la Protezione dell'Ambiente (ANPA), Rome, Italy
- Joint Research Centre, (JRC), Ispra, Italy
- Forschungszentrum Jülich GmbH, (FZJ) (ISR/PKS), Jülich, Germany
- Forschungszentrum Karlsruhe GmbH (FZK), Karlsruhe, Germany
- Technische Universität München (TUM), Institut für Radiochemie (RCM), München, Germany
- Austrian Research Centres (ARC), Seibersdorf, Austria
- Technical Research Centre of Finland (VTT), Espoo, Finland
- Nuclear Research Institute (NRI), Rez, Czech Republic
- COGEMA, France
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USE OF NON-DESTRUCTIVE ASSAY SYSTEMS
IN DISMANTLING PU-CONTAMINATED FACILITIES

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Abstract
For the examination of incoming radioactive waste as well as for product control measurements the Karlsruhe Research Center operates several drum monitors. During the last years a lot of experience could be gained in the examination of Pu-containing wastes arising from research institutes as well as from decommissioning projects. Due to unknown matrix and nuclide composition problems especially occur as far as “historical” waste forms are considered. Major influence factors and their consideration during routine measurement are discussed in this paper.

1 Introduction

The Central Decontamination Department (HDB) of the Karlsruhe Research Center runs various facilities for radioactive waste conditioning. This waste originates mainly from the current dismantling projects of the Karlsruhe Reprocessing Plant (WAK), the decommissioned research reactors of the Research Center, and the European Institute of Transuranium Elements.

Before waste may be transported to HDB for processing, it has to be described completely. For safety purposes and quality assurance the activity declaration is randomly controlled at the waste entrance by non-destructive examination of the waste packages.

2 Measurement Equipment

To perform this examinations three drum measurement facilities are operated by HDB. This facilities allow major parameters to be determined for both waste characterization as well as product control:

1. **Gammascanner for low-level waste drums (LLW scanner):**
   Low-energy and higher-energy gamma emitters can be determined automatically in 200 l and 400 l drums with a surface dose rate of up to 2 mSv/h.

2. **Gammascanner for intermediate level waste drums (ILW scanner):**
   Together with a passive neutron monitor, the ILW scanner is installed in a shielded cell, in which drums of increased dose rate can be subjected to measurement in a remote-controlled manner. Both measurement stations are connected by a chain conveyor for each drum being transported first to the gamma and then to the neutron monitor. The gamma scanner is designed for 200 l and 400 l packages with a surface dose rate of up to 0.5 Sv/h. By varying the scanning parameters, it is possible to determine even smaller, locally restricted activity distributions at a variable shielding thickness.

3. **Passive neutron monitor for determination of the fissile material inventory in LLW and ILW drums:**
   The passive neutron monitor allows measurement of both 200 l and 400 l drums of 1 μSv/h – 1 Sv/h surface dose rate and approx. 10 mg – 100 g plutonium inventory.
So far, measurements have been carried out mainly on solid raw wastes (entry control) and on compacted or solidified waste products. Below, it shall be reported about major influencing factors and their consideration during routine measurement.

3 Influencing Factors

3.1 Matrix Effects
The wastes received for treatment comprise light and heavy metals, glass, insulation material and filters as well as concrete and burnable materials (mainly plastics and cellulose products). In most cases, density and activity distributions in a drum are inhomogeneous. It is envisaged to have the wastes sorted as precisely as possible. Neutron monitor calibration and verification measurements were carried out using special standard drums loaded in accordance with the procedure applied during WAK dismantling:
- Sand (as homogeneous standard drum)
- Metal (iron parts, tubes, sheet metal, filters, etc.)
- Plastics (polyethylene, foil, cellulose, cleaning cloths, etc.)
- Rubble.

In case precise sorting of the waste cannot be guaranteed, the category with the higher neutron absorption behavior is assumed so as to obtain a conservative analysis result.

Within the framework of product control, waste products, i.e. solidified, cast or compacted wastes, are examined. Cast waste products consist of scrap, rubble or mixed wastes embedded in cement stone. Solidified waste products comprise cemented waste waters, concentrates, sludges or ion exchange resins. The latter are assumed to have a homogeneous density and activity distribution due to the conditioning process. In any case, unknown quantities of moderating material, here considerable amounts of water that is chemically and absorptively bound in the concrete, lead to an underestimation of the plutonium content. Compared to atomic nuclei of other matrices, elastic scattering on hydrogen nuclei results in highest energy losses of the neutrons emitted. Consequently, a reduction of the neutrons to be measured and, hence, an underestimation of the measured value is expected with an increasing humidity content. Experimental studies regarding the humidity content (e.g. in cellulose mixes or rubble with residual humidity) revealed the dependence expected, but the results obtained may not just be transferred to calculable water contents of cement stone or concrete. To solve this problem, a special MCNP calibration calculation had to be made for a defined concrete drum (according to HDB cementing requirements) and the package had to be subjected to additional verification measurements. Statistically sound measurement results on real waste packages and above all on cast waste products of inhomogeneous density distribution are still lacking.

3.2 Chemical Composition
For the calculation of plutonium and uranium isotopes, it is generally assumed that they exist in oxidic form in the waste. Fluoride and/or bromide compounds cause the yield of neutrons from the \((a/n)\) nuclear reaction to be increased. As the threshold energy for the separation of a neutron is smaller for several light atomic nuclei than their Coulomb's repulsion energy for \(\alpha\) particles, light nuclei and in particular Li-7, Be-9, B-10, B-11, and F-19 exhibit significant reaction cross-sections for the generation of \((a/n)\) neutrons.
In real waste packages, however, generation of this nuclear reaction strongly depends on the distribution of these elements and the type of chemical bond. Hence, a numerical correction can hardly be made even in case of known contents, which is rather rare anyway.

3.3 Nuclide Vector

To calculate the fissile material contents from the neutron counting rates, the nuclide vector of the uranium and plutonium isotopes as well as of other neutron-emitting nuclides must be known. Incorrect or incomplete nuclide vectors result in an incorrect evaluation of the measurement results and mostly in an overestimation of the plutonium inventory. Nuclides with a high neutron emission rate, above all Cm-244 and Cf-252, cause considerable evaluation errors. Their contents have to be given in the nuclide vector for them to be considered in evaluation and the plutonium and uranium contents not being overestimated.

To verify the nuclide vectors applied and determine lacking vectors, in particular with regard to the above-mentioned neutron emitters, the origin of the neutrons emitted (\(\alpha,\alpha\) reaction or spontaneous fission) is determined by coincidence analysis. As a criterion, an average of more than one neutron is emitted during spontaneous fission. (\(\alpha,\alpha\)) reactions always emit a single neutron only. The number of neutrons emitted per spontaneous fission varies from U-235 (1.7) to Pu-239 (2.24), Cm-242/244 (2.4), and further on to Cf-252 (3.7). In principle, fissile materials may be distinguished in this way. Still, the neutron monitor must have a sufficiently high detection sensitivity (at least 15%). Judging from its design parameters, the HDB monitor is suited for such measurements. Correlation analysis with the HDB monitor is based on the TCA/LCA data processing system and shift register analysis.

Presently, calibration and verification measurements are being performed prior to routine analysis in order to facilitate the interpretation of the measurement results.

4 Quality Assurance

Comprehensive verification measurements on drums of various material categories, i.e. various densities and variable measurement positions, resulted in test instructions being set up. These instructions were controlled by the authorities and the expert during the commissioning measurements. Regular instrument tests, the contents of which are specified in the HDB operations requirements, serve to ensure the functionality of the drum measurement stations. The procedure consists of appropriate radiation sources with known activities being inserted into an inactive reference drum and measured in a routine manner. Reference drums are drums that have been specially manufactured for this purpose. They allow for the following radiation sources being inserted at variable positions:

- Eu-152 volume sources for the LLW scanner (Ge and GeLi detector)
- Cs-137 and Co-60 volume sources for the ILW scanner
- Pu standard with known vector for the neutron monitor

As the ILW scanner and the neutron monitor have been combined in a single unit, the same reference drum is applied for this test. The results of the weekly instrument tests are collected in a file and represented graphically as a function of time in order to visualize deviations and trends in the response behavior of the detectors. By means of this quality assurance program, quality of the measurement results can be maintained.
5 Application of the Results

Results of the drum measurements are used as described below:

1. Control of incoming waste and internal waste products:
   The difference between measured and declared values are calculated and compared with defined acceptance values, that specify the tolerable deviation in activity declarations. The declarations of those drums that exceed the tolerance limits have to be corrected.

2. Substitution or correction of the existing declaration:
   As far as the waste declaration shall be substituted by the measurement results all data measured by the drum monitors, i.e.
   - dose rate on the surface and at 1 m distance,
   - gross and net weight,
   - gamma-emitting nuclides, such as Cs-137, Co-60, etc.,
   - plutonium and uranium contents,
   directly enter the waste declaration. All other nuclides to be declared are determined on the basis of nuclide vectors in correlation to key nuclides.

To verify and optimize the nuclide vectors, the data measured are compared with each other and the values expected. This is the basis of the measurement-supported declaration procedure. Hence, the measurements made by the drum measurement facilities contribute to the updating and optimizing of nuclide vectors of the respective waste producers. Within the framework of WAK dismantling, more than 1300 waste drums have been measured so far.
DEVELOPMENT OF A STATE-OF-THE-ART SOLID WASTE CHARACTERISATION FACILITY AT JRC ISPRA

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Abstract
This paper will review the development and construction of a state-of-the-art waste characterisation facility that represents a fundamental element in the JRC’s strategy to characterise radioactive wastes originating from four decades of nuclear research activities conducted at the Ispra Site. The state-of-the-art NDA measurement solution selected by JRC-Ispra will be presented in the light of special requirements imposed by applicable Italian legislation, norms and the qualities of the waste packages. The intended plant combines the state-of-the-art-measurement solution with comprehensive process automation in an industrial environment and is therefore of significant interest to the radioactive waste management community.

1 Introduction

The Joint Research Centre of the European Commission is distributed in five EU member states. Four of the JRC sites (Geel, Ispra, Karlsruhe and Petten) are host to numerous research facilities in which a variety of scientific and technological activities employing radioactive materials have been undertaken in the framework of the Euratom Treaty. The diversity of such facilities reflects the broad spectrum of nuclear research conducted by JRC since its birth, and includes experimental reactors, hot cells and radiochemical facilities, as well as a variety of radioactive waste management plants and stores.

The changing focus of recent EU Framework Programmes combined with the natural ageing of facilities has resulted, however, in many of the nuclear installations no longer being able or necessary to support the JRC’s present day mission. For this purpose, the JRC has developed a Decommissioning and Waste Management (D&WM) Programme which foresees a safe and economic path for decommissioning the shutdown installations and managing its existing and future radioactive wastes, collectively known as the JRC’s “Historical (nuclear) Liabilities”[1].

Most of the liabilities are related to the largest JRC site at Ispra in northern Italy. Presently, the waste management area of the Ispra Site is host to a variety of unconditioned and conditioned solid wastes with approximately nine thousand waste packages having a volume approaching 4000 m³. To this quantity will be eventually added about 8000 m³ of radioactive wastes originating from decommissioning related activities. To respect applicable legislation, all waste must undergo appropriate characterisation during its management on-site and before its transfer, off-site, to a final national repository. Consequently, waste characterisation equipment has a fundamental role in the site’s waste management strategy.

2 Technical Requirements

The basic technical requirements of the waste characterisation system are primarily determined by Italian regulations that establish radioactivity level thresholds for diverse α- and βγ-radionuclides in different categories of waste. The treatment and conditioning of the waste,
and the on-site interim storage requirements of the resulting waste packages, together with their final repository destination and associated charges, are dictated by the waste’s category.

The activity thresholds and corresponding waste categories are defined in Technical Guide N° 26 of ANPA (ex-ENEA Disp), [2,3], in which the definition of the categories is based on the final disposal route. Cat. I is assigned to short-lived very low level waste (SL-VLLW) not requiring a long-term storage solution in contrast to long-lived very low level waste (LL-VLLW) which needs a disposal site. Cat. II covers wastes that will decay in a few centuries to an activity level below 370 Bq/g, i.e. long-lived low level waste (LL-LLW) and short-lived intermediate level waste (SL-ILW). A detailed breakdown of the specific activity thresholds for Cat. II conditioned wastes is given in Table I. Non-conditioned wastes are also accepted in this category if the specific activity levels are below the thresholds given in Table II. Finally, Cat. III groups those wastes that are not included in Categories I and II, i.e. high level waste (HLW) and/or long-lived intermediate level waste (LL-ILW). The existing and future radioactive wastes at JRC-Ispra fall into all three waste categories.

Table I. Limits for radioactivity concentrations in the Cat. II conditioned waste packages

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Concentration</th>
</tr>
</thead>
<tbody>
<tr>
<td>α-emitters $T_{1/2} &gt; 5$ years</td>
<td>370 Bq/g</td>
</tr>
<tr>
<td>β-emitters $T_{1/2} &gt; 100$ years</td>
<td>370 Bq/g</td>
</tr>
<tr>
<td>β-emitters 100 years in activated metals</td>
<td>3.7 kBq/g</td>
</tr>
<tr>
<td>β-emitters $5 &lt; T_{1/2} \leq 100$ years</td>
<td>37 kBq/g</td>
</tr>
<tr>
<td>$^{137}$Cs and $^{90}$Sr</td>
<td>3.7 MBq/g</td>
</tr>
<tr>
<td>$^{60}$Co</td>
<td>37 MBq/g</td>
</tr>
<tr>
<td>$^{3}$H</td>
<td>1.85 MBq/g</td>
</tr>
<tr>
<td>$^{241}$Pu</td>
<td>13 kBq/g</td>
</tr>
<tr>
<td>$^{242}$Cm</td>
<td>74 kBq/g</td>
</tr>
<tr>
<td>Radionuclides $T_{1/2} \leq 5$ years</td>
<td>37 MBq/g</td>
</tr>
</tbody>
</table>

Table II. Limits for radioactivity concentrations in the Cat. II non-conditioned waste packages

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Concentration</th>
</tr>
</thead>
<tbody>
<tr>
<td>Radionuclides $T_{1/2} &gt; 5$ years</td>
<td>370 Bq/g</td>
</tr>
<tr>
<td>$^{137}$Cs and $^{90}$Sr</td>
<td>740 Bq/g</td>
</tr>
<tr>
<td>Radionuclides $T_{1/2} \leq 5$ years</td>
<td>18.5 kBq/g</td>
</tr>
<tr>
<td>$^{60}$Co</td>
<td>18.5 kBq/g</td>
</tr>
</tbody>
</table>

The cost of final disposal is clearly correlated to the waste category via the necessary storage time and conditions. With regard to final repository costs, JRC-Ispra presently assumes charges will be about 7 Euro/m$^3$ for Cat. II waste packages, a price that includes Italy taking ownership of the waste. In contrast, a factor of fifteen is applied to Cat. III waste packages. Using these values and the likely waste volumes, JRC-Ispra estimates that the charges may exceed 100 MEuro. Consequently, it is extremely important not only for operational but also financial reasons that any waste characterisation system is capable of reliably segregating waste and that waste is not inadvertently over-categorised.

Given the variety of nuclear research studies previously undertaken at JRC-Ispra, it is evident that the wastes contain very diverse radionuclides: from the reactors, all the major activation products and their decay products, while work on the fuel cycle has led to fission product and actinide contaminated wastes. It is worth noting, however, that as the bulk of existing wastes originate
from activities performed in the 1960s and 70s, radionuclides with a very short half-life now make an insignificant contribution to the total remaining activity, although their decay products still need to be considered.

Given this backdrop, a representative set of gamma-emitting isotopes that must be readily quantified by the intended waste characterisation system, was established. The set comprises $^{60}$Co, $^{94}$Nb, $^{125}$Sb, $^{133}$Ba, $^{134}$Cs, $^{137}$Cs, $^{152}$Eu, $^{154}$Eu. Similarly, the principal actinides of interest were defined as $^{235}$U, $^{238}$Pu, $^{240}$Pu and $^{241}$Pu. NDA does not, however, provide all the necessary data for waste categorisation. A number of the radionuclides of interest emit only very low energy gamma rays and/or beta particles that cannot be measured with a NDA system. Fortunately, most of the “soft” contributors, e.g. $^{60}$Ni and $^{63}$Ni in steel and $^{93}$Zr in Zircalloy, can be correlated to the easily measured radionuclides, and this permits their activity contribution to be estimated by applying appropriate nuclide vector correction factors to the measured activity. Likewise, JRC-Ispra recognises that during decommissioning, some waste streams may require sampling and radiochemical analyses due to the expected presence of radionuclides that can neither be quantified by NDA nor correlated to the measurable radionuclides. In particular, given the use of graphite moderators and heavy water in the site’s experimental reactors, it is likely that $^{3}$H, $^{14}$C, $^{35}$Cl and $^{41}$Ca radionuclides are present. Such radionuclides will be calculated using ad-hoc procedures.

Attention must also be given to the heterogeneous activity and matrix attenuation distributions in the existing waste packages. Presently, the major part of the existing waste is conditioned with bitumen and concrete with matrix densities varying from 1000 to 2500 kg/m$^3$. JRC-Ispra also has numerous unconditioned waste packages filled with low-density process waste material and compacted waste or metals with a typical matrix density in the range 100 to 2500 kg/m$^3$. Furthermore, it is envisaged that a supercompactor will be eventually utilised to reduce future waste volumes resulting in waste matrices having densities up to 4000 kg/m$^3$.

To date, the nuclear research facilities at JRC-Ispra have used a variety of drums ranging from 35 to 200 litres. There is a tendency throughout Europe, however, for larger drum sizes to be used in order to reduce the number of waste packages and maximise the radioactive waste to container volume ratio. Indeed, JRC-Ispra will standardise future waste packages on 440 litre drums and it was therefore decided that the proposed waste characterisation system must be able to accommodate such containers as this performance was considered to be still within the bounds of current technology.

Operational requirements arise from regulatory demands, safety requirements and the efficient use of resources. About 30000 waste drums will eventually be characterised and, typically, one hour will be required to handle, measure and analyse each drum. Therefore, the waste characterisation system will be provided with an automatic drum delivery subsystem and will operate unattended overnight to permit the analysis of up to twenty drums per day. This approach will allow a 50% system usage rather than say, <15%, in the case of plant operation being limited to normal working hours, and will help to maximise the return on the capital investment by limiting personnel requirements and by avoiding waste characterisation to become a bottle-neck in the site’s D&WM Programme.

3 Selected NDA Solution

After a thorough analysis of the technical prescriptions and a review of commercial solutions on the market, a NDA measurement system based on tomographic $\gamma$-scanning and active neutron detection for quantifying beta/gamma and alpha activities respectively was chosen. In addition to the NDA measurement system, the contract supply also included a conveyor transport system,
dose and weight measurement stations and a comprehensive measurement and control automation system, see Figure 1. The supply contract was awarded to A.N. Technology Ltd.

![Figure 1: Principal components of the waste characterisation system](image)

The main features of the chosen gamma measurement station (Figure 2) are its transmission and emission tomography capability for drums up to 440 litres having a wide range of matrix densities. The station also supports a conventional SGS-mode, while for very low activity drums or those having low attenuation, it will be possible to make measurements in a total gamma mode. This flexibility will allow JRC-Ispra to select the appropriate operating mode to satisfy a wide range of activities and to measure activity and attenuation distributions in the waste packages. Typically in a tomographic operating mode, a 200 litre drum will be mapped into thirteen segments or vertical sections. In each of these, 150 line averaged transmission and emission spectra will be acquired and analysed resulting in a spatial resolution of approximating a 5 cm cube, although the resolution will be variable by changing the detector collimation and the measurement time.

The gamma system will be able to perform a typical measurement of a complete 200 litre drum containing short-lived LLW comprising $^{137}$Cs and $^{60}$Co (Cat. I threshold of 10 Bq/g) with a matrix density of 2000 kg/m$^3$ in one hour. In general, measurements will be able to determine the category (I, II or III) of the waste drum for all likely matrix and gamma emitter configurations, with the estimated minimum detectable activity levels being ~1 Bq/g for $^{60}$Co and $^{137}$Cs.

The main feature of the neutron measurement station (Figure 3) is the interrogation of the waste with pulses of thermal neutrons that will be produced when generated 14 MeV neutrons from pulsed accelerator are slowed down in a graphite liner surrounding the drum cavity. The thermal neutrons will induce fission in fissile isotopes present in the waste and the ensuing prompt and delayed fission neutrons will be detected in external, fast neutron detector banks. From the detected fission neutrons the amount of fissile material will be deduced. In a passive measurement mode (with the neutron generator switched off), a signal count rate from the spontaneous fission (fertile) isotopes will be obtained in addition to a count rate from $(\alpha, n)$ reactions in the drum. By combining the results of the active and passive neutron measurements, a quantitative measure of the actinides in the waste will be possible.

The neutron measurements will be performed in a specially designed cavity, sufficiently large to accommodate a 440 litre drum, whose main purpose of the cavity is to provide neutron moderation of the interrogating neutron flux and to maximise the measurement efficiency. The neutron moderator surrounding the waste drum will be nuclear grade graphite. The fast neutron detector banks and the neutron moderator will be dimensioned so that the lifetime of the thermal neutrons is of the order of 1000 $\mu$s. The detector banks will be located on the outside of the
moderator, with a number of flux monitors distributed both inside and outside of the cavity. The detector banks will consist of cadmium clad high-density polyethylene blocks with embedded $^3$He proportional counters and will be installed in an approximate 4π configuration surrounding the measurement cavity.

Overall, the neutron station will be capable of performing a typical characterisation measurement of a complete 200 litre drum containing an average long-lived LLW (Cat. II, specific activity <370 Bq/g) with a matrix density of 1500 kg/m$^3$ in one hour. Initial design figures suggest a lower detection level of about 10 Bq/g of $^{239}$Pu for 200 litre drums.

Recognising the degree of plant automation and to minimise additional operational doses, the waste characterisation facility is also envisaged to automatically measure the weight, surface contamination, and dose rates of the waste drums. All the measured and computed data related to each waste package will be stored in a centralised database. It is envisaged that an automatic laser etching system integrated into the facility will be employed to label each drum with the relevant data (e.g. id, date, dose, mass etc.) prior to its dispatch to the site's future interim store.

4 Conclusions

JRC-Ispra believes that the characterisation facility will fulfil all of its standard waste drum NDA requirements and will therefore enable the successful characterisation of existing and future radioactive wastes in a well-qualified, efficient and cost-effective manner. The intended plant is a state-of-the-art characterisation system operating in an industrial environment and therefore of significant interest to the radioactive waste management community. Given that the facility will employ most of the advanced features discussed and promoted by the waste management community, it can serve as a reference plant in Europe and therefore provide European added value to the significant initial capital investment that JRC-Ispra had made. Currently, the facility is in an advanced state of construction. It is envisaged that the facility will be commissioned in 2003 and enter in routine operation in 2004.

5 References

Abstract
An analysis and evaluation system for the characterisation of radioactive waste from nuclear installations for both declaration and free-release measurements is under development. It is intended to allow the determination of the most likely activity of an object and the appropriate uncertainty range. Additionally, it is designed to determine possible compositions of the object (e.g. density and activity distribution). This can be achieved by a consequent implementation of all available information about an object like a-priori information (e.g. origin and age of the waste), knowledge from databases (e.g. nuclide properties like half-lives, gamma energies and emission probabilities) and results from non-destructive measurements (e.g. gamma scanning, transmission measurements). The system is built up of separate modules. There are modules to prepare the input data, to create detector systems and objects, to simulate the interaction between detector and object and to analyse the simulation results. This modular conception requires a precise definition of the different interfaces but provides the possibility of extensibility. Missing or insufficiently known parameters about the object are varied in their supposed range to achieve a simulation result that shows best match with the considered real measurement. For comparison of simulation and measurement results there is a tool which provides mathematical methods for consistency checking.

1 Introduction

Non-destructive assay techniques frequently support the characterisation of waste arising from the decommissioning of nuclear installations. The evaluation of these measurements usually is based on several assumptions on the composition of the object (e.g. its density and activity distributions) since these are not known in detail or at all, resulting in sometimes strongly biased activity values. The quoted uncertainties are often based on simple counting statistics only. Some applications establish at least an empirical “uncertainty factor” to consider in some way the lack-of-knowledge on the real distributions. Nevertheless, these reported activity values and their corresponding uncertainties often show large discrepancies from reality.

To overcome this unsatisfactory situation, a special analysis and evaluation software package is developed allowing the determination of the most likely activity of an object and the appropriate uncertainty range by using all available information. Additionally, possible compositions of the object (e.g. density and activity distributions) being consistent with the measured data can be determined.
2 Concept of the software package

2.1 General

The basic idea to set the data evaluation and analysis on a more reliable basis is to consider all available information about the object under investigation for its characterisation. Based on these data an iterative evaluation process takes place. Its basic concept is shown in Fig. 1. Due to the modular structure of the software package future extensions for including additional data can be adapted easily. In the following the modules are briefly introduced.

2.2 Input Module

Primarily all available information must be collected. For its further manipulation it is classified into three general categories:

- a-priori information,
- knowledge from databases and
- results from measurements.

A-priori information is straight information about an object like its origin, its age or the nuclide vector. Based on this information further consolidated assumptions like waste form (raw, compacted, cemented etc.), activity and density range can be derived.

Databases provide information on container characteristics, attenuation coefficients, nuclide properties etc. and parameters about individual detector configurations.

The third category is composed of all results from measurements. There may be basic measurements like determination of dimensions and mass as well as integral gamma counting and more complex non-destructive techniques like gamma scanning and transmission measurements. The latter may provide information about the inner composition of an object. The results of the subsequent analysis must be tested for conformity within given limits against all available data of this category.

2.3 Preparation Module

All input data must be provided in a form being appropriate for the use within all software modules. Therefore routines exist extracting the information from different analysis protocols and other input sources and converting it in a specific format. All these data are stored in a database. Additionally, consistency checks are performed taking into account the individual uncertainty ranges. Furthermore, a first approximation to describe the composition of the object is performed, resulting in a parameter set necessary for object simulation.
2.4 Simulation Module

This module is responsible for the simulation of the different measurements whose data are available for comparison with measurement results. Two sub-modules are attached:

- detector module and
- object module.

The detector module, which is designed and programmed by ISR/FZJ (Institut für Sicherheitsforschung und Reaktortechnik, Forschungszentrum Jülich GmbH), determines the response $R_{ij}$ of a detector at a defined position in front of the detector called interface matrix. It gives the detector efficiency as a function of position $(i, j)$, energy $E_y$ and direction $(\theta_y, \phi_y)$ of all incident gamma quanta and characteristics of the detector system $(k_1, ..., k_n)$ (see Fig. 2).

The object module provides a detailed description of the object under inspection. This object may consist of individual components having different densities, radionuclides and activities. Furthermore, the module simulates the photon distribution $\phi_{ij}$ of gamma radiation emitted from defined activity volumes by means of Monte Carlo simulation (MCNP).

Using the interface matrix as projection of detector and object characterisation (see Fig. 2), the simulation tool calculates results for a specific measurement procedure (i.e. integral gamma counting, multi-rotational scan in segmented gamma scanning etc.) which are used in the subsequent analysis process.

**Figure 2: Schematic illustration for the definition of an interface matrix which is used as a projection field of emission as well as detection data linking object and detector characteristics.**
2.5 Analysis Module

The simulation results are compared with the corresponding measurement data. Special tools providing mathematical methods for consistency checking are applied. Depending on the level of consistency all appropriate simulation parameters are stored in a database. Then, parameters may be modified within their uncertainty ranges and readmitted in the simulation process to finally obtain different density and/or activity compositions of the object all being consistent with the measured data.

2.6 Result Module

Applying such an iterative analysis procedure the simulation determines the most likely activities of radionuclides within an object. Since the simulation considers possible compositions of the object (e.g. density and activity distributions) appropriate uncertainty ranges can also be deduced. Thus, the aim of this project to improve both accuracy and precision will be achieved.

2.7 Output Module

Since this software package is intended to be applied in routine inspection a protocol is produced, presenting the results of the analysis as well as all information necessary for QA/QC.

3 Conclusions

Experiences and results from a categorisation, which will be done by FZJ, of typical waste forms from decommissioning and of actually applied measuring systems for inspection are considered in this analysis and evaluation software package, thus assuring a close relation to praxis. Therefore the software package will give a valuable help for the improvement of accuracy and precision in declaration and free-release measurements of waste setting the results on a reliable basis. The applicability of the software package will be demonstrated with extensive tests using artificial samples as well as performing in-field measurements.

4 Acknowledgment

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DETERMINATION OF TRITIUM AND CARBON-14 IN ACCELERATOR WASTE

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Abstract
In dismounted parts of the accelerator facilities of Paul Scherrer Institute, tritium and $^{14}$C were determined by low-level counting after chemical separation. In graphite targets used for the production of $\pi$-mesons, tritium amounts from $1.7 \times 10^5$ to $6 \times 10^7$ Bq/g were found; the corresponding $^{14}$C data were 6 and 9 Ci/g, respectively. In the dismantled copper beam dump of Target E, the tritium content extended up to $2.8 \times 10^6$ Bq/g, but no $^{14}$C could be detected. In mechanical parts of the beam dump, consisting of iron and stainless steel, respectively, the tritium amount ranged up to $5.3 \times 10^6$ Bq/g, the $^{14}$C amount from 1 to 800 Bq/g. The separation procedures are described in detail.

1 Introduction
According to the interaction of accelerated particles and target materials, radioactivity is induced. By the primary beam, the target itself is activated, likewise the beam dump. During the setting up of the beam, or by scattered particles, beam lining elements are activated, and by the induced secondary radiation, mostly by neutrons, the surroundings of the target equipment are activated, including the shielding elements. Consequently, when accelerator facilities are decommissioned or when a part of the accelerator system is changed by maintaining, dismounted activated parts must be disposed, and an appropriate concept should be elaborated before.

In Paul Scherrer Institute, the radionuclide inventories of dismounted activated parts have to be determined before disposal. Due to its origin from accelerator facilities, the nuclide vectors should show compositions different from that of well-known nuclear power waste. It is expected to support these data mainly by model calculations, but at least in the first phase, they must be validated by radioanalytical measurements. The measurements include determination of tritium and $^{14}$C contents. The present paper describes the procedures for determination of $^3$H and $^{14}$C in both dismantled graphite targets and the dismounted beam dump of Target E of the cyclotron facilities at Paul Scherrer Institute.

2 Experimental

2.1 Graphite targets
The so-called 'Target E' is shown in Fig. 1. It consists of ultrapure polycrystalline graphite. This target type is used for many years for the production of intense pion and muon beams, by interaction of 590-MeV protons with light nuclei, in our case carbon. The graphite has a density of about 1.78 g/cm$^2$. It rotates with about 1 r.p.s. The proton beam is slightly degraded (by 6 cm graphite, in the new targets by 4 cm graphite); it is heated by the released ionization energy (about 25 MeV/proton) and by secondary particles generated from interaction between the protons and the target nuclei.
(about 5 MeV/protons). The energy produced by both mechanisms is about 45 kW for a proton current of 1.5 mA and results in a operation temperature of 1500 K of the radiatively cooled target cone. By use of high beam currents (presently up to 2 mA), the graphite targets are of limited lifetime. They must be replaced in intervals of some months to a few years. The target investigated in this paper is a typical example: It was dismounted in June of 1994 after 3 years operation with an accumulated integrated beam current of 3.98 Ah.

The crumbled graphite is put into a quartz crucible and placed into a heating furnace. An oxygen stream is allowed to flow for 30 min at room temperature. Then, the system is heated up to 1000°C. The released gases are trapped into three wash flasks containing aqueous sodium hydroxide solution (6.67 g NaOH/g graphite). After 3 hours, the heating is stopped, and the tritium activity of all three flasks is measured by a liquid scintillator counter (range 0 - 18.6 keV, cocktail Hionic Fluor, Packard). Up to 90 % of the tritium activity is collected in the first flask, 8 % is found in the second flask. The tritium content in the third flask is negligible.

To determine the 14C-activity, each solution is evaporated to dryness and carefully dried in vacuum. The residue from the first solution is dissolved in double-distilled water and transferred into a three-necked flask. Under stirring, conc. sulphuric acid is added dropwise at room temperature in nitrogen atmosphere. The released gases are passed through concentrated sulphuric acid and trapped into three wash flasks containing 0.1N sodium hydroxide solution. Then, the 14C-activity is measured in the liquid scintillator counter (range 2 - 256 keV, cocktail Hionic Fluor, Packard). This procedure is repeated with the content of the second and third flask of tritium analyses. As in the tritium determination, more than 90 % of the 14C-activity is detected in the first flask, up to 9 % in the second one, and practically no activity was found in the third flask. Therefore, the distribution of the 14C-activity is roughly proportional to the tritium content. This analytical process requires a time of three days.

2.2 Metallic targets

The metallic samples are the same as described elsewhere in detail [1]. They were taken from the beam dump of Target E and consisted of electrolyte copper, proton activated (No. 6) and activated by secondary particles, respectively (No. 9), shielding cast iron (No. 12), stainless steel (No. 15) and Steel 37 (No. 16); the latter two samples were taken from construction material of the beam dump. As mentioned in [1], the beam dump was decommissioned in beginning of 1990 after 16 years operation and an accumulated integrated beam current of 5.95 Ah. After 10 years cooling time, we may assume that the short-lived nuclides are decayed and the samples are ready for determination of tritium and 14C.

Borings of the metal samples are placed in a three-necked flask. Under stirring at room temperature, they are dissolved in 40 % HNO₃ in a nitrogen atmosphere. The released gases are passing through concentrated sulphuric acid and collected into three wash flasks, each containing 0.1N sodium hydroxide. In this solution, the 14C-activity is measured by liquid scintillation (range 2 - 256 keV, cocktail Hionic Fluor, Packard) as described above. According to the measurement of 14C in graphite, the principal amount of 14C is detected in the first flask.

The acidic solution in the three-necked flask is diluted with double distilled water and the pH is adjusted to 8-9 with 0.1N sodium hydroxide. Then, the tritium activity is measured as described above.

3 Results and Discussion

In the graphite targets, the tritium activity was determined resulting between 1.7 and 6 \cdot 10^8 Bq/g, depending of the location of sample in the target. The corresponding amounts of 14C were determin-
ed resulting between 6 and 9 Bq/g. Obviously, the amounts of both radionuclides are not distributed homogeneously in the target ring. The highest value was found in the entrance region of the proton beam, the lower amount was found in the beam exit position of the ring. The amounts in the spokes are not given here; they are activated by scattered radiation from protons and secondary particles and were determined to 1-2 orders of magnitude less. The tritium is mainly formed by nuclear reactions like $^{12}$C(p,x)$^3$H. From excitation function measurements, conservative estimations suggested a yield in the order of $10^5$-10$^6$ Bq/g [2]. Our results could be interpreted in such a way that most of formed tritium would release during irradiation. Due to the slightly reducing conditions during irradiation, perhaps it is escaped from target in the form of HT. The results of tritium and $^{14}$C determinations in the metallic samples are shown in Table 1.

Table 1. Tritium and $^{14}$C activities (Bq/g) in metallic waste (Target E beam dump)

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Sample 6 (Copper, activated by protons)</th>
<th>Sample 9 (Copper, activated by secondary particles)</th>
<th>Sample 12 (Cast Iron)</th>
<th>Sample 15 (Stainless Steel)</th>
<th>Sample 16 (Steel 37)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^3$H</td>
<td>$2.84 \times 10^6$</td>
<td>$138$</td>
<td>$4.61 \times 10^3$</td>
<td>$5.29 \times 10^3$</td>
<td>$3.20 \times 10^3$</td>
</tr>
<tr>
<td>$^{14}$C</td>
<td>$\leq 1$</td>
<td>$\leq 1$</td>
<td>$801$</td>
<td>$223$</td>
<td>$15$</td>
</tr>
</tbody>
</table>

As mentioned above, tritium is formed mainly by direct reactions with $^3$H as final product. Thus, it seems that in a first approximation, the yield is a measure of the accumulated integrated beam current at the respective position. Even in these samples, the tritium amount is lower than expected from calculations [3]; it seems that tritium is escaped during irradiation by diffusion effects. The low amount of $^{14}$C in the copper samples and the amount of this nuclide in the iron samples nearly proportional to the carbon content indicates that the main source of $^{14}$C is the carbon content in the samples. Thus, $^{14}$C would be formed mainly by interaction of secondary neutrons with carbon. More fine calculations indicates that the measured $^{14}$C amount in cast iron is somewhat lower than expected, while the measured amounts in Steel 37 and in Stainless Steel are higher than expected [3]. These effects are under examination.

4 Conclusions

The tritium and $^{14}$C activities could be determined by low-level counting in graphite targets and in metallic accelerator waste as well. By careful work, the determination of very low amounts of $^{14}$C beside high amounts of tritium is possible. The formation channels of the nuclides are explicable. The statistical uncertainties, obtained by repeated measurements, are distinctly higher than the actual measuring error. This indicates for inhomogeneities in the samples. The assumed statistical error fulfills in any case the requirements of accuracy for final disposal. The graphite targets contain tritium amounts far beyond the exemption limit. This may pose problems for handling. To overcome this, the graphite may be treated chemically (burned) or disposed after some decades of cooling time. For the disposal of the activated metallic waste, on the other hand, tritium and $^{14}$C should pose no problems.
5 Acknowledgments

The authors are grateful to Professor H.W. Gäggeler and Dr. H.F. Beer, both PSI, for their interest and continuous help in this work. They are debtful of gratitude to Dr. H. Maxeiner, NAGRA, Wettingen, for many stimulating discussions.

6 References


7 Figures

Figure 1: Photography of the rotating graphite target, used at PSI for production of pions and muons
Characterisation of the corrosion products of non-irradiated material test reactors fuel elements (MTR-FE)

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Abstract

In a high concentrated Mg-rich brine a non-irradiated MTR-FE corroded. The formed corrosion products consist of an amorphous part and of hydrotalcites, which were identified as Mg-Al-hydrotalcites with chloride anions in the interlayer.

1. Introduction

The final disposal of MTR-FE in a salt dome is under discussion. A water ingress is considered as an accident scenario. Previous work showed, that the formed high concentrated salt brines corroded the fuel container and the fuel and most of the radionuclides were trapped by the formed corrosion products. Because of the high dose rate of burned up MTR-FE, non irradiated fuel elements were used. A non-irradiated material test reactors fuel element (MTR-FE) of the DIDO type was leached in a repository relevant salt brine. The main objective of this study was to characterise the formed corrosion products.

2. Experimental

Two corrosion experiments with non-irradiated MTR-FE pieces (20×28×1.5 mm) were performed in a Mg-rich brine under Ar atmosphere at 90°C with and without iron in glass autoclaves. The experimental details are shown in Table 1. MTR-FE elements of DIDO type contain 20 wt% U/Al alloy, which is covered with metallic Al. The uranium mass in one piece used is about 0.11 g. The Mg-rich brine used represents a repository relevant brine [2].

Table 1. Experimental details for the corrosion experiments with non-irradiated MTR-FE

<table>
<thead>
<tr>
<th></th>
<th>Experiment 1</th>
<th>Experiment 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>solution</td>
<td>Mg-rich brine</td>
<td>Mg-rich brine</td>
</tr>
<tr>
<td>volume of solution, ml</td>
<td>400</td>
<td>400</td>
</tr>
<tr>
<td>weight of MTR-FE plates, g</td>
<td>1.84</td>
<td>2.03</td>
</tr>
<tr>
<td>elemental iron; mass, g</td>
<td>3.22</td>
<td>-</td>
</tr>
<tr>
<td>temperature, °C</td>
<td>90</td>
<td>90</td>
</tr>
<tr>
<td>environment</td>
<td>Ar</td>
<td>Ar</td>
</tr>
<tr>
<td>time, months</td>
<td>17.5</td>
<td>20</td>
</tr>
</tbody>
</table>

The experiments were performed till the plates corroded totally. At the end of the experiments the autoclaves were opened under an Ar atmosphere. The liquid and solid phases were separated. The solution was filtered with a 450 nm filter and then 1 ml of the filtrate was diluted with 0.1M HNO₃ up to 20 mL. The obtained clear solution was analysed by ICP-MS. U was separated with TOPO and the organic phase was measured with an α-spectrometer. Aliquots of the sediment were treated in the following way: one wet aliquot was dried at 90°C for about 24 hours; another aliquot was dialysed. For dialysate a dialysate hose was filled with the substance (2-4 g) and placed in a 2 L vessel containing deionised water. The water was changed two times a day for 2-5 days.
After dialysis, the substance was dried. All dried samples were milled to powder and analysed by XRD and SEM. For the determination of the elemental composition an aliquot of about 0.2 g was dissolved in 5 ml 8 M HNO₃. The U content was measured as described above.

Figure 1. XRD pattern of a) direct corrosion products and b) of dialysed corrosion products. Narrows show the hydrotalcite peaks.
3. Results and Discussion

The duration of the experiments was about 18 months. Non-irradiated MTR-FE plates corroded completely, whereas the iron plate corroded slightly. More than 20 g of the sediment was formed. The corrosion rates, calculated from the weight loss of non-irradiated MTR-FE pieces and from the corrosion time were about 0.1 g/month. The composition of the used Mg-rich salt brine did not change significant. Only the Mg concentration decreased about 10%.

The elemental composition of the corrosion products showed that the main components of the formed corrosion products are Mg and Al. All released Al and U were quantitatively found in the corrosion products.

The XRD investigation of the corrosion products showed that the main crystalline phase is a bischofite, MgCl$_2$·6H$_2$O (Figure 1, a). This phase is present because magnesium chloride is the main component of the used solution. The signals of the other phases were less intensive and suppressed by bischofite. To reveal other crystalline phases a dialysis of the sediment was performed. After the dialyses hydrotalcites were clearly identified. From the XRD data the lattice parameters $a$ and $c$ lattice were calculated. Comparing these results with literature data, the obtained HTlc can be identified as a Mg-Al-hydrotalcite with a Mg/Al ratio close to 3 and with chloride anions in the interlayer space.

Hydrotalcites have a layer structure. Each layer has a brucite, Mg(OH)$_2$, structure. Some Mg$^{2+}$-ions are substituted by a trivalent ion. The positive charge is compensate by anions, which are in the interlayer region between the two brucite-like sheets [3]. The interlayer anion can be easily exchanged. Therefore the second name for HTlc is anionic clays. In literature some reports already exist where the hydrotalcites ability for exchange with radionuclide anions was investigated ([4], [5], [6]). Hydrotalcites are very attractive for radionuclide retardation. Some of our preliminary results [7] showed that hydrotalcites immobilized, for example, I$^-$ as anion and cations like Cs$^+$ and Am$^{3+}$.

4. Conclusion

The final disposal of spent fuel elements is considered to be realised in a salt dome. A potential water ingress will lead to the formation of salt brines, which will corroded the fuel container and the fuel itself. Corrosion products will be formed and the characterisation of these products was the subject of this work. For these purposes two corrosion experiments with non-irradiated MTR-FE in high concentrated Mg-rich brine were performed. The formed corrosion products were characterised. It was found that the sediment contains hydrotalcites besides bischofite and amorphous phases. After the dialysate the XRD pattern clearly indicates that the obtained hydrotalcites belongs to the Mg-Al-type with a Mg/Al ratio close to 3 and with chloride as interlayer anions. All dissolved Al and U were found to be in the corrosion products.

Considering the salt dome as a final repository for spent fuel elements with a metallic Al/U as fuel, it can be surely said that the uranium will not mobilised. In further work the ability of hydrotalcites for radionuclides retardation will be investigated.

5. Acknowledgement

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6. References


Remobilisation of Radionuclides from the Corrosion Products of MTR-Fuel Elements

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Abstract
The retention behaviour of radionuclides in secondary phases formed by corrosion processes of a material-test reactor fuel element in salt brine was analysed as function of the ionic strength of the surrounding aquatic phase. Furthermore the kinetic of the radionuclide remobilization was determined.

1. Introduction
The direct final disposal of spent fuel is being discussed in Germany [1]. A possible repository under consideration is a salt mine. As an accident scenario for long-term safety analysis a water ingress is considered. For final disposal the material-test reactor fuel elements (MTR-FE) 1 will be placed in a cast iron POLLUX container, which is assumed to resist the corrosive attack of the formed high concentrated salt brine for approximately 500 years. After the container failure, the brine will have direct contact with the MTR-FE. Different leaching experiments with the MTR-FE in high concentrated salt brines at 90°C (in a repository the expected temperature caused by the afterheat is 90°C) showed that the radionuclides were mobilised first and then trapped by the corrosion products (secondary phases) of the aluminium matrix and the POLLUX container [2]. In order to quantify the source term for radionuclide release aliquots of the corrosion products were leached in different concentrated salt brines and in water. The kinetic of the radionuclide remobilization was studied especially in dependence on the ionic strength of the surrounding solution.

2. Experimental
Salt brine 2 represents a repository relevant brine. For one litre of brine 2 solution 937.1 g magnesium chloride hexahydrate, 0.126 g magnesium sulfate heptahydrate, 4.13 g sodium chloride, 1.42 g potassium chloride and 39.68 g calcium chloride dihydrate were dissolved in deionized water.

The experiments were performed in a hot cell due to the high dose rate of the secondary phases, which were obtained through corrosion processes of a MTR-BE in salt brine 2. After homogenisation the secondary phases were divided in equal portions. The remaining salt brine 2 was decanted after centrifugation of each aliquot. The wet samples were weighted and 10 ml of solution (brine 2 or half concentrated brine 2 or water) was added to each tube. The tubes were closed, mixed and stored in an oven at 90°C. At different time intervals 1 ml of the solutions were taken (exact 1 ml of the identical solution was added immediately to the aliquots to guarantee an identical sample volume), filtered (450 nm filter) and diluted with 9 ml 0,1 M HNO3.

1 The MTR-FE of the DIDO type contains 20 wt% U-Al alloy with an initial enrichment of 80 % in $^{235}$U. The fuel alloy plates with a thickness of 0.6 mm (average) are covered on both sides with aluminium claddings of 0.38 mm, the total thickness is then 1.36 mm. Three plates are welded to form a tube. A fuel element assembly consists of four concentric tubes of approximately 6, 7, 8 and 9 cm diameter, mounted onto structural parts.
solution. This samples were taken out and the following analytical procedure was performed: After a first gamma measurement, the amount of Cs was determined. Then Cs was separated with AMP (ammoniummolybdatophosphat) followed by a second gamma measurement where $^{60}\text{Co}$, $^{154/155}\text{Eu}^{144}\text{Ce}$ were determined clearly. Then, this solution was used for a subsequent separation and determination of Sr. The determination of the actinoids, Pu and U was done by alpha spectrometry using an aliquot of the original sample solution. Detail description is given in [2]. The determination of the total radionuclide inventory of an aliquot of the secondary phases was achieved by total dissolution with 8 M HNO$_3$. The resulting clear solution was analysed in the way described above.

3. Results and Discussion

In figure 1 the remobilization rate for Cs is shown. The acronym FOI (y-axis) stands for fraction of inventory. This radionuclide inventory was calculated from the total dissolved aliquots of the secondary phases. A quantitative remobilization for Cs was achieved in all samples. Within 7 days equilibrium was reached. Furthermore the release of Cs did not depend on the ionic strength of the used solvents.

![Figure 1: Remobilization of Cs in different solvents.](image)

The same remobilization behaviour was found for the radionuclides $^{60}\text{Co}$ and $^{90}\text{Sr}$ except that Cs was released quantitatively while these radionuclides were remobilized to 70 %. In figure 2 the release rate is shown for the radionuclide $^{154/155}\text{Eu}$. 

Figure 2:  Remobilization of Eu in different solvents.

It can be seen, that the release rate increased in the first 300 days strongly. Especially in water a quantitative remobilization was achieved but then after 300 days, a different process must took place in which Eu was immobilized again. For that immobilization reaction no equilibrium was reached even after 800 days. In half concentrated brine 2 Eu behaved similar but the transfer of remobilization to immobilization reaction occured after 500 days. In brine 2 an equilibrium of remobilization was reached after 300 days and no subsequent immobilisation reaction was observed.

Figure 3:  Remobilization of actinoides in different solvents.
In figure 3 the remobilization rate of the actinoides Pu and U is shown. For these actinoides no release was observed in brine 2 or in half concentrated brine 2 solution. Only in water Pu and U were remobilized first but after 300 days they were immobilised again.

Now, the question did arise: "what did cause the transfer of the remobilization reaction to an immobilisation reaction which was observed for Eu and the actinoides especially in water?". We suggest that due to the change of the ionic strength of the surrounding solvent the secondary phases did change in their compositions. The used secondary phases were formed through corrosion processes of MTR-FE in salt brine 2. Using fresh brine 2 the ionic strength did not change. Therefor in all experiments were brine 2 was used, only a radionuclide remobilization reaction was possible due to dilution effects. A new immobilisation reaction is not possible because the secondary phases did not change in their compositions. In water, which is the medium with the lowest ionic strength used, the compositions of the secondary phases changed significant. The resulting new phases then immobilised Eu and especially the actinoides much better.

4. Conclusion

In this paper secondary phases obtained through corrosion processes of a MTR-FE in salt brine 2 are leached in different concentrated salt brines and in water. The remobilisation behaviour of the fixed radionuclides was studied. It was found that Cs released quantitatively while Sr and Co were remobilized up to 70 % and that the release rates reached equilibrium after 7 days. Furthermore the release rate did not depend on the ionic strength of the used solutions. For Eu and the actinoides Pu and U a different behaviour was observed. A strong release rate was observed in the first 300 days for Eu in all solvents and for Pu and U only in water. Then these radionuclides were immobilised again in the experiments using water or half concentrated brine 2 solutions. No equilibrium was reached during the experimental time period. These immobilisation reactions can be explained by changes in the phase compositions of the secondary phases. In future work the characterisation of the secondary phases formed in different solvents is intended.

5. Acknowledgement

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6. References


Investigation of penetration mechanisms of radionuclides in concrete and their leaching behaviour

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Abstract
Penetration profiles for Cs and Sr in concrete of the strength class B 35 have been determined. Capillary suction experiments with both radionuclides $^{137}$Cs/$^{85}$Sr and natural Cs/Sr have been performed in parallel. Fully characterized cylindrical concrete specimens were in contact with solutions containing the species of interest under well-defined conditions. Different processing techniques have been applied for active and non-active samples followed by gamma spectrometry and atom absorption spectroscopy respectively. Both routes provided satisfactory results, being in good agreement and demonstrating the distinct behaviour of the two elements. Similar work with Co, I and U will be reviewed in near future.

1 Introduction
Decommissioning operations of nuclear plants imply the accumulation of an enormous mass of building materials such as concrete, metals, electric devices, tubes, cables, liquid waste etc. The overwhelming part of this “nuclear waste” is undoubtedly represented by the amount of concrete with different contamination levels. Available data, however, suggest that approximately 92% of these concretes can be categorized as unrestrictedly free and are directly available for other uses. A smaller amount of 6-7% is liable to free release or to unrestricted free release after decontamination treatment i.e. it can also be recycled. Heavily activated concrete fragments from the reactor vicinity (~1-2%) are intended for final storage [1]. Under the aspect of concrete reprocessing, the initiated research project at IfMB offers a more specified direction towards precise evaluation of surface contamination, definition of migration parameters of radionuclides (Cs, Sr, Co, I, U) in the concrete matrix and decontamination possibilities. Previous research work [2] performed at the nineties offered a comprehensive platform for the present project. The experimental work is divided in three main parts:
1. Study of the penetration depth and radionuclide behaviour in concrete surfaces
2. Determination of the radionuclide distribution in various cement phases and between hardened cement paste and aggregate
3. Leaching investigations of radionuclides from concrete fractions according to the German recycling guidelines for building materials
Transport data gained from experiments will serve as the basic input for a realistic modelling approach able to describe radionuclide kinetics in concrete and to calculate released activity. This will be reviewed in a forthcoming paper in combination with diffusion results.

2 Experimental

2.1 Sample preparation
Cylindrical concrete samples of 30 cm length and 10 cm diameter of the strength class B 35 were prepared by mixing Ordinary Portland Cement (OPC) with water (w/c ratio 0.5) and pure quartz
aggregate. Much care was taken for good compaction of the material based on an optimized grading curve. The specimens were demoulded after one day and were stored under water to avoid carbonation. For material characterisation, all relevant properties (compression strength, pore system, gas permeability, water absorption coefficient) were investigated by methods commonly used in building materials technology. In order to estimate the natural background of cesium and strontium in the starting materials, samples of OPC and prepared concrete were analysed by means of atom absorption spectroscopy (AAS) and ICP-MS. The agreement of the values revealed a negligible Cs content in the specimens on the one hand, but a quite high Sr contribution of approximately 0.17 wt.% on the other, which implies a limiting factor for measurements of natural Sr via AAS.

2.2 Capillary suction
These experiments are laboratory methods for testing transport of liquids in porous non-saturated solids due to surface tension existing in thin capillaries. By utilizing same experimental conditions, mobility comparisons between different ions are possible. Experiments of this type were carried out in parallel with $^{137}$Cs and $^{85}$Sr and with inactive cesium and strontium in aqueous solutions. The experimental construction and some details are shown in the Figure 1.

![Figure 1: Experimental setup for capillary suction (T = 20 °C, rel. humidity 65%)](image)

Preliminary experiments to determine the water absorption of B 35 concrete samples (liquid uptake/area unity) confirmed a fast weight increase at the beginning weakening considerably after 10 days reaching an almost steady state after 25 days (Fig 2). The concrete samples were pre-equilibrated by treating them thermally at 40 °C for two weeks. One even face of the specimen

![Figure 2: Water absorption for B 35 (two samples)](image)
Figure 3: Experimentally determined penetration profiles for $^{137}$Cs/$^{85}$Sr and natural Cs/Sr.
was then exposed to a solution containing the investigated ion for the duration of 3-4 weeks. To force quasi one-dimensional transport into the bulk, the cylindrical surface was coated with an epoxy resin impermeable for water. For active experiments, three samples were used for each nuclide. Non-active experiments were carried out double.

2.3 Sample treatment and measurements
The general concept used to find out the profiles of Cs and Sr was a step by step removal of thin layers from the loaded concrete specimens. Different grinding techniques and measurement methods were employed to detect the gamma emitting radionuclides $^{137}$Cs and $^{85}$Sr and natural Cs and Sr. Radioactive specimens were abraded with an especially designed device equipped with a diamond grinding disc, suction apparatus and a collecting unity with filter papers showing a collection efficiency of 75%. Concrete layers between 0.2 and 1.5 mm were ground and the total activity of the filter was directly measured employing a gamma spectrometer with a high purity germanium detector. Concrete layers of the thickness 0.1-0.5 mm were removed in the case of natural Cs and Sr with a Boehme grinder using corundum as abrasive medium. Because of the bigger concentration gradient at surface-near regions, thinner specimen layers (0.1-0.2 mm) were firstly ground. The collected samples, consisting of fine concrete powder mixed with a large amount of corundum, were treated with diluted hydrochloric acid to dissolve the cement paste. After filtration and rinsing of the residue, the solutions were measured by means of AAS.

3 First results and discussion
The achieved depth of an ion as a function of time is based on the perplex interplay of many parameters (adsorption, ion exchange, valency of penetrating ions, precipitation, w/c-value, pore structure, strength class of the concrete, aggregate quality, carbonation status etc.) and each of them can influence the transport mechanism decisively. Although obtained on a completely different way and using strongly different concentrations in the penetrating solution (about $10^{-2}$ mol/l in non-active and from $10^{-7}$ to $10^{-10}$ mol/l in active experiments), the results show a good qualitative agreement (Fig 3a, 3b, 3c, 3d) and clearly reveal the different chemical character of Cs and Sr in the concrete matrix. The first evident conclusion is the long-tailed penetration profile of Cs demonstrating its higher mobility. It is however difficult to relate this behaviour to one or more physicochemical parameters with full certainty. Competition for active places on the hardened cement paste with K and Na present in the pore solution and ion exchange phenomena seem to be of primary importance under these conditions [3]. Some traces of Cs can even be detected in layers deeper than 1 cm. At a distance of 2-3 mm the Cs content has decreased to approximately one tenth when compared to the concentration of the first layer and it amounts only one hundredth and less in layers beyond 5-6 mm. The lower migration potential of Sr, indicated by a more abrupt decrease of the penetration curve, is obviously due to a stronger interaction with the hardened cement paste and could be attributed to its double charge. For the rather pure information from AAS measurements, the high natural background of Sr in cement has to be drawn in to attention.

4 References
Solid-Phase Extraction for the Enrichment of Actinides from Radioactive Waste

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Abstract
For the destructive characterisation of radioactive waste, the actinides to be analysed must be separated from matrix components, if possible, in an element-specific manner. Microwave pressure digestion is a time-saving digestion method for radioactive waste samples. The partitioning of the actinides from the liquid samples are carried out with the aid of solid-phase extraction. For the experiments, on the one hand, commercially available extraction resins such as Actinide, TRU, UTEVA and TEVA Resin® from Eichrom were used. On the other hand, an extraction resin with Cyanex 301 produced in-house was employed specifically for actinide-lanthanide partitioning. It was possible to separate 99.4 % of the lanthanides by a two-stage partitioning process. It is particularly meaningful to perform a prior matrix partitioning with Actinide Resin®. The extractant, which was eluted from the column together with the actinides, was destroyed oxidatively by means of a newly developed technique using microwave pressure digestion.

1. Introduction
The characterisation of actinides in solid radioactive waste samples is only possible if the waste matrix is destroyed before and as result a liquid sample is obtained. With the aid of microwave pressure digestion all kind of radioactive waste matrices can be destroyed effective in a time-saving manner [1]. The enrichment and partitioning of the actinides in these liquid sample was performed by solid-phase extraction. A separation scheme using Eichrom resins developed by Horwitz et al. [2,3,4] was tested with the actinides Thorium, Uranium, Neptunium, Plutonium, Americium, Curium and the lanthanide Europium. These separations had been carried out for aqueous standard solutions first and had to be adapted to different kinds of radioactive waste matrices e.g. concrete, ion exchange resins, filter dusts, slurries and sander grains. With respect to future options for destroying actinides by transmutation it is also essential to carry out an actinide-lanthanide partitioning which was not considered in Horwitz separation scheme. These considerations and experiments with different kinds of radioactive waste matrices led to a successfully tested partitioning scheme. This paper does not go into details on the experiments with the resins impregnated with Cyanex 301 for lanthanide separations which are described in Reference [1].

2. Experimental
A separation scheme based upon Figure 1 was tested. The experiments on the Eichrom resins were carried out both with radioactive standard solutions and with solutions containing radioactive waste matrices. The solutions were transferred to the different columns containing 1g (for Actinide™ Resin) or 0.7g (for TEVA™, UTEVA™ and TRU™) resin. The columns were washed with several mineral acids [1,4] and the actinides were then eluted (see also results and discussion).
The destruction of DIPEX™, which is eluted together with the actinides from Actinide™ Resin, was first realised by the addition of 5 mL 30% H₂O₂ and 0.5 mL 0.2 M NaVO₃ as catalyst [5]. An other possibility to destroy DIPEX™ was to undergo a microwave decomposition with 5 mL HNO₃ and 1.5 mL H₂O₂. The microwave, which was used, was a Mega 1200 of MWS. There was no catalyst necessary for this method.

### 3. Results and discussion

3.1. Results with Actinide™ Resin

All actinides and lanthanides can be separated from matrix components with Actinide™ Resin. The actinides and lanthanides were eluted with 15 mL isopropanol together with the extracting agent DIPEX™. It was possible to prepare 1 mL of the isopropanol fraction directly for α-measurement to have a first look on the actinides which are present in the sample. A quantitative analysis could even be carried out when 100 µL of the fraction was prepared together with 2 mL of a 1% Zapon lacquer solution on a stainless steel target (Ø 200 mm) for α-measurement in a grid chamber. A more time-consuming electrodeposition was not necessary.

For the next separation steps the extracting agent DIPEX™ had to be destroyed. The isopropanol fraction was first evaporated and then heated on a hot plate with H₂O₂ and NaVO₃. This method led to a black residue which had to be treated the described way another three times. This destruction step was very difficult to handle because of the exothermic reaction and was thus also very time consuming. It was replaced by the destruction with HNO₃ and H₂O₂ using a microwave decomposition. This step could be carried out in one hour. After evaporation a residue was obtained which contained mainly amorphous parts and methylen-diphosphonic acid but no DIPEX™. This residue was dissolved in 2 M HNO₃ / 1 M Al(NO₃)₃ and put into the next separation steps without any problems. It was even shown that the recovery rates after the

![Separation scheme with five columns](image-url)
destruction in the microwave were higher than the rates received with the other method (see Table 1).

### Table 1: Recovery rates of the actinides after different destruction methods of DIPEX™

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Recovery rate after destruction with H₂O₂ and NaVO₃</th>
<th>Recovery rate after microwave decomposition</th>
</tr>
</thead>
<tbody>
<tr>
<td>²³⁷Np</td>
<td>90 ± 3 %</td>
<td>85 ± 5 %</td>
</tr>
<tr>
<td>²³¹Am</td>
<td>77 ± 7 %</td>
<td>85 ± 5 %</td>
</tr>
<tr>
<td>²³³U</td>
<td>65 ± 5 %</td>
<td>90 ± 4 %</td>
</tr>
<tr>
<td>²³⁹Pu</td>
<td>65 ± 6 %</td>
<td>90 ± 6 %</td>
</tr>
</tbody>
</table>

3.2. Results with TEVA™ Resin

The actinides Neptunium and Thorium can be separated distinctively on a TEVA™ Resin column. It is important to add ascorbic acid and ferrous-sulfamate to the sample to reduce Neptunium completely to Np(IV) and Plutonium to Pu(III). Experiments were carried out with standard solutions including known activities and real waste matrices e.g. concrete, ion exchange resin. Figure 2 shows the results and the used mineral acids for the extraction and separation. To simplify the graphic the actinides Uranium, Plutonium and Americium are not shown. These nuclides were found quantitatively after the sample was rinsed through the column (first fraction). As expected the recovery rates for Neptunium and Thorium in standard solutions were around 90%. The addition of ascorbic acid had to be increased if waste matrices including Fe(III) are used. This factor was easy to handle, but Thorium caused a problem in real waste matrices. 70% of Thorium was found after the sample was rinsed through the column together with Uranium, Americium and Plutonium. Thorium appears always in its stable oxidation state +4 so there was no need to add another oxidation or reducing agent. That meant that the Thorium was not completely extracted by the resin. We supposed that the waste matrix components influenced the extraction of Thorium on TEVA™ Resin. Cleaning the sample from matrix components with Actinide™ Resin led to the same result. After the destruction of DIPEX™, phosphoric acid residues remained which disturbed the extraction of Thorium on TEVA™ Resin. This effect could not be inhibited by the addition of more Al(NO₃)₃ which normally complexes phosphate ions.

3.3. Results with UTEVA™ and TRU™ Resin

It is possible to separate Uranium and also Thorium on UTEVA™ Resin. The washing fraction of TEVA™ Resin can be used without any further treatment. We made experiments with standard and real waste solutions both for Uranium and Thorium which delivered non satisfying results with TEVA™ Resin. As can be seen in Figure 3 Uranium was eluted with good recovery rates of 95% respectively 90%. Again we found Thorium in the rinsed sample solution. But in this case
it was only 10% and it was possible to analyse the Thorium quantitatively by the addition of a Thorium spike. On TRU™ Resin Plutonium was separated from Americium, Curium and lanthanides. Figure 4 shows that there were the same good recovery rates for standard and real waste solutions. This Resin did not cause any problems.

4. Conclusion

In order to separate actinides (Neptunium, Thorium, Uranium, Americium and Plutonium) and lanthanides from dissolved radioactive waste, we tested a separation scheme presented in Figure 1 successfully. With this partitioning run the actinides and lanthanides were separated from matrix components on Actinide™ Resin. On TEVA, UTEVA and TRU Resin™ the actinides were isolated distinctively. The separation of the lanthanides was successfully carried out with an in house-made resin containing Cyanex 301. 99.4% of the lanthanides were separated by a two stage partitioning process.

The whole separation scheme can be automated with the aid of a laboratory robot system. This is already realised in our laboratories and guarantees a quick partitioning process with a minimum dose rate for the laboratory worker.

5. References

THE USE OF AN HPCIC COLUMN COUPLED TO IE-HPLC FOR A MATRIX INDEPENDENT SEPARATION OF ACTINIDE ELEMENTS

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Research Centre Jülich, Institute for Safety Research and Reactor Technology, Jülich, Germany

Abstract
Within the scope of the destructive waste analysis, there is a need for fast and easy to automate methods for the analysis of the actinide elements. HPLC can provide for the fast multi-element characterisation of radioactive waste. In order to avoid separation problems caused by the sample matrix, an HPLC column has been modified with an actinide selective complexing agent, CMPO. By means of this column coupled to an anion exchange column, the actinides americium, plutonium, thorium and uranium have been separated. The separation method has been tested with simulated salt and cement samples.

1. Introduction

Because of radiation protection reasons and in order to reduce analysis time and costs, there is a need for automated methods in destructive waste analysis. High Performance Liquid Chromatography (HPLC) is such a fast and easy to automate method that can provide for the multi-element characterisation of radioactive waste. Earlier developed Ion Exchange HPLC (IE-HPLC) methods have the big disadvantage of being seriously affected by the sample matrix and thus requiring a matrix preseparation in advance. The goal of our work was to develop a more matrix independent HPLC separation method for the determination of the actinide group elements.

2. High Performance Chelation Ion chromatography (HPCIC)

1.1. Principles of High Performance Chelation Ion Chromatography
In High Performance Chelation Ion Chromatography a reversed phase HPLC column is modified with a chelating agent. The reversed phase column can be either a silica based column or a polystyrene divinylbenzene copolymer based column. The chelating agent loaded on the column provides for selectivity. The aqueous eluents can contain complexing agents like citric acid which can offer an additional tool of selectivity control. HPCIC combines the high efficiency and robustness of Reversed Phase HPLC with the selectivity of the chelating agent. The main advantage of this chromatography method is its relative insensitivity to high ionic strengths. [1]

1.2. An HPCIC column for the separation of actinide elements
Because of the low pH of the Radwaste samples required to prevent hydrolysis and polymerisation of the high valency metal ions, a polystyrene divinylbenzene copolymer based stationary phase has been preferred. A PRP-1 column (5µm, 100 x 4.6 mm) has been modified
with the actinide complexing agent CMPO (octyl(phenyl)-N,N-diisobutylcarbamoylmethylphosphine oxide) well-known from liquid-liquid and solid phase extraction. With this HPCIC column, the actinides could be separated from matrix compounds, but due to the very high retention of the tetra- and hexavalent actinides, their mutual elution from this column was impossible.

3. The HPCIC column coupled to IE-HPLC

Plutonium could be eluted from the CMPO modified column by reducing it to the trivalent state. Uranium (VI) and thorium(IV) could be eluted together from the HPCIC column as anionic carbonate complexes which could be separated on an anion exchange column (DOWEX 1x8, 200-400 mesh, 120x4.6 mm) coupled to the HPCIC column. An overview of the HPLC arrangement with the two column approach can be seen in figure 1. The parameters which were held constant during all experiments are listed in table I. Table II contains the eluents and Table III contains the separation conditions for the separation of the actinide elements. The detection has been performed with an online-LSC detector.

![Figure 1: Experimental arrangement of the HPLC system with online LSC detector](image-url)
Table I: Chromatographic conditions which were held constant during all experiments

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>eluent flow rate</td>
<td>1 mL/min</td>
</tr>
<tr>
<td>sample loop</td>
<td>100 μL</td>
</tr>
<tr>
<td>column temperature</td>
<td>30°C</td>
</tr>
<tr>
<td>volume LSC cell</td>
<td>0.5 mL</td>
</tr>
<tr>
<td>LSC cocktail flow rate</td>
<td>2 mL/min</td>
</tr>
</tbody>
</table>

Table II: Eluents for the actinide elements

<table>
<thead>
<tr>
<th>Eluent</th>
<th>Concentration</th>
</tr>
</thead>
<tbody>
<tr>
<td>Eluent A</td>
<td>0.05M HNO₃ + 0.01M NaN₂O₂</td>
</tr>
<tr>
<td>Eluent B</td>
<td>2M Na₂CO₃</td>
</tr>
<tr>
<td>Eluent C</td>
<td>0.25M Na₂CO₃ + 0.5M NaN₃O₃</td>
</tr>
<tr>
<td>Reduction agent</td>
<td>0.005M Fe(II)SO₄ in 10 mg/L ascorbic acid</td>
</tr>
</tbody>
</table>

Table III: Separation conditions for actinide elements

<table>
<thead>
<tr>
<th>Time [min]</th>
<th>Eluent</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>Eluent A</td>
</tr>
<tr>
<td>5</td>
<td>Injection of reduction agent through the 5mL loop</td>
</tr>
<tr>
<td>10</td>
<td>Closing of the 5mL sample loop</td>
</tr>
<tr>
<td>12</td>
<td>85% eluent A + 15% eluent B</td>
</tr>
<tr>
<td>15</td>
<td>Eluent B</td>
</tr>
<tr>
<td>20-30</td>
<td>Eluent C</td>
</tr>
</tbody>
</table>

4. Results

A chromatogram of the separation of the actinides Am, Pu, Th and U, obtained with the elution program in table III is illustrated in figure 2. The detector efficiency and detection limits (3σ-criterion) for their subsequent quantification with the online LSC-detector are listed in table IV. The detector efficiency differs for each nuclide because of the different alpha-energy and the different sample matrix in the LSC measurement cell. This separation method has been tested successfully with salt and cement samples spiked with known amounts of Am, Th, Pu and U.
Figure 2: Chromatogram of a standard solution containing 19.0 Bq $^{241}$Am, 15.9 Bq $^{238}$Pu, 17.1 Bq $^{229}$Th and 32.9 Bq $^{233}$U

Table IV: Detector efficiency and detection limits (3σ-criterion)

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Injected Bq</th>
<th>Measured CPS</th>
<th>Efficiency ε %</th>
<th>Detection limits</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{241}$Am</td>
<td>201.9</td>
<td>167.7 ± 11.7</td>
<td>83.1</td>
<td>0.50</td>
</tr>
<tr>
<td>$^{238}$Pu</td>
<td>330.5</td>
<td>304.6 ± 19.1</td>
<td>92.2</td>
<td>0.95</td>
</tr>
<tr>
<td>$^{229}$Th</td>
<td>73.0</td>
<td>25.3 ± 2.4</td>
<td>34.7</td>
<td>1.67</td>
</tr>
<tr>
<td>$^{233}$U</td>
<td>168.5</td>
<td>89.4 ± 9.6</td>
<td>53.0</td>
<td>0.82</td>
</tr>
</tbody>
</table>

5. Conclusions

This method can provide for a fast and automated determination of the major actinides. The determination of americium however, can be interfered by the presence of other radionuclides in the sample.

6. References

TECHNICAL ASSESSMENT OF WASTE PACKAGE PRODUCTION

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Abstract
In order to ensure that radioactive wastes are packaged as a quality product with defined characteristics, Nirex performs assessments of quality management systems. To enable future custodians of the package to have confidence in the information defining its characteristics, Technical Assessments are also undertaken. Confirmation of compliance with the waste product specification during production is an important outcome of an assessment. Assessments are also used to demonstrate continuing post-production quality of waste packages.

1 Introduction
The role of Nirex is to provide the United Kingdom with safe, environmentally sound and publicly acceptable options for the long-term management of radioactive materials. These materials include operational and decommissioning wastes from nuclear power plants and other wastes from industrial, defence and medical applications involving radioactivity. Within this role, Nirex provides advice on, and where possible endorsement of, waste packaging proposals with respect to the suitability of the waste package for long-term management. Nirex reviews each proposal under sixteen technical areas, including the control of package production through the implementation of a ISO9001 compliant quality management system (QMS).

It is essential to establish confidence in the package characteristics at the time of production, when there is ready access to operators, plant and equipment. Such checks are necessary before production to avoid incurring doses, costs and the generation of secondary wastes from checking and possibly remediating defective packages. To gain confidence in the completed waste package, Nirex undertakes a continuous assessment process that seeks to demonstrate an appropriate level of quality before, during and after package production.

QMS audit is used to demonstrate quality during the planning phase whilst technical assessment focuses on package production controls, the completed package characteristics, measurement, monitoring and the creation of information and records [1,2].

Technical Assessment is undertaken during the production phase. It provides an opportunity for the Waste Packager to demonstrate to stakeholders the production of fully characterised, quality waste products that meet regulatory and safety requirements and that are consistent with future waste management options. It is also be used to confirm that appropriate records and information, which will enable future generations to make informed decisions on the management of the waste package, are created and preserved.

Therefore the QMS assessment confirms the capability to produce a defined product and the Technical Assessment confirms that the waste product meets expectations.
2 Nirex Advice Process and Waste Package Production

Radioactive waste arises in a wide range of physical and chemical forms. UK Government policy (which is currently under review) requires that waste held in interim storage should be passively safe and, so far as possible, compatible with the requirements for subsequent stages of its management. In order to assist Waste Producers in this activity, Nirex has developed the Phased Disposal Concept, a stepwise and reversible approach to geological disposal. The concept underpins a set of guidance documents and specifications that are used by the Waste Producers for packaging waste in a suitable form.

Prior to packaging a waste, a Waste Producer will submit proposals to Nirex for review. The review will consider the means for ensuring the safe handling and treatment of the waste during each phase of waste management. The phases include waste conditioning, package production, interim storage of waste packages, waste package transport, waste handling, package emplacement and monitoring during the operational phase of a repository and the repository post-closure period.

The endorsement of a packaging proposal, in the form of a Letter of Comfort, will be conditional on the ability of the Waste Producer to demonstrate effective control of package production and interim storage. Hence the importance of employing an appropriate QMS. In order to continue to provide assurance both during and after waste package production, Nirex carries out audits of the production process, the completed waste package, the storage and transport systems and the information management system.

3 Waste Product Specifications

As with any quality product, it is essential that a clear specification of the product is agreed before production begins. Only with this specification in place is it possible to demonstrate 'compliance'. Nirex, in-conjunction with the UK nuclear industry, has developed the concept of the Waste Product Specification (WPrS). In simplified terms, the WPrS draws together data and evidence drawn from research and development activities in nine inter-related areas, as shown in Figure 1.

![Figure 1: The Waste Product Specification](image-url)
The WPrS is developed by the Waste Producer during the planning phase and focuses on the key processes, materials and parameters. The WPrS is used in the control of the production process by identifying and quantifying the key components of the completed package. Production of a package to the WPrS will give confidence that the product will display predictable characteristics over time, thereby making it suitable for future phases of waste management.

A WPrS structured in this way therefore provides the focus for obtaining the objective evidence concerning the activities affecting product quality. Clearly, the WPrS can only provide a summary of the limits and controls necessary during and after production, and therefore it needs to be supported itself by references to the research and development work supporting the project.

4 Technical Assessments

The aim of Technical Assessment is to provide objective evidence demonstrating that the Waste Producer:

- is implementing a robust management system that is capable of producing a quality product;
- has the capability for handling and storing waste packages in such a way that they continue to meet the criteria demanded by the Phased Disposal Concept;
- has the means for creating and preserving accurate and reliable information in a form suitable for use by future generations; and
- is compliant with the packaging proposal that was originally endorsed (through issue by Nirex of a Letter of Comfort).

The focus of the assessment process continues to be the identification of activities likely to affect product quality. These activities will be present at all phases of product realisation and will range from operator training records, through production material control to post-production monitoring.

There are a number of project stages where opportunities exist to assess the Waste Producer's System. With regard to Technical Assessment, it will be important to confirm that the production plant has been adequately commissioned and tested. This should include checking that test batches are compliant with the Waste Product Specification. Having demonstrated compliance, assessments of randomly sampled product might be suitable for providing assurance of the quality and compliance of a large production campaign. The interactions between advice, endorsement and assessment are illustrated in Figure 2.

When a Technical Assessment is undertaken, a defined sequence of activities is followed. This will include a review of the production process control documentation, quality plans and programmes, and the Waste Product Specification. The review is followed by a site-visit where the Waste Producer is invited to present the evidence demonstrating that the operation of the plant, the manufactured product and the interim storage arrangements are consistent with the provisions of the Waste Product Specification.

As Waste Producers increasingly become familiar with the assessment process and its benefits, they have integrated the principles into their own systems of work and this has enabled a joint assessment approach to be adopted.
The assessment findings are recorded in a report that identifies both good practice and areas for improvement. Where evidence shows that waste products fall outside the limits specified in the Waste Product Specification, further evidence will be necessary to demonstrate that the product remains acceptable for future phases of waste management. In these circumstances Nirex will work with the Waste Producer to identify the most appropriate course of action.

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