

PROGRESS REPORT ON NUCLEAR DATA RESEARCH IN THE FEDERAL REPUBLIC OF GERMANY

for the Period April 1, 2001 to March 31, 2002

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Edited by
S.M. Qaim
Forschungszentrum Jülich GmbH
Institut für Nuklearchemie
Jülich, Federal Republic of Germany

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Edited by: S. M. Qaim

Forschungszentrum Jülich GmbH

Institut für Nuklearchemie

Jülich, Federal Republic of Germany

FOREWORD

As in previous years, this report has been prepared to promote exchange of nuclear data research information between the Federal Republic of Germany and other member states of OECD/NEA and IAEA. It covers progress reports from the research centres at Karlsruhe and Jülich, the universities of Dresden, Hannover and Köln, as well as from the PTB Braunschweig. Each contribution is presented under the laboratory heading from where the work is reported. The names of other participating laboratories are also mentioned. The emphasis in the work reported here is on nuclear data for applied science programmes, such as those relevant to reactor technology and safety, transmutation concepts, accelerator shielding and development, astrophysics research, cosmogenic and meteoritic investigations, production of medically important radioisotopes, etc.

The coordination of nuclear data activities at the international level is done by two committees: the NEA-Nuclear Science Committee (NEA-NSC) and the IAEA-International Nuclear Data Committee (INDC). The present Editor has the privilege and the responsibility of representing Germany in both the committees. This report therefore also serves as a background information to some areas of work of those committees.

Jülich, July 2002

S.M. Qaim

This document contains information of a preliminary nature. Its contents should be used with discretion.

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FORSCHUNGSZENTRUM KARLSRUHE INSTITUT FÜR KERNPHYSIK

1. A Measurement of the $^{13}\text{C}(\alpha,\alpha)$ Differential Cross Section and its Application to the $^{13}\text{C}(\alpha,n)$ Reaction*

M. Heil, A. Couture¹, J. Daly¹, R. Detwiler¹, U. Giesen¹, J. Görres¹, G. Hale², F. Käppeler, R. Reifarth, E. Stech¹, P. Tischhauser¹, C. Ugalde¹, M. Wiescher¹

The $^{13}\text{C}(\alpha,n)$ reaction is considered as the main neutron source for the *s* process in AGB-stars of 1-3 solar masses. The reaction takes place at temperatures of about 10^8 K which corresponds to a Gamov peak at 190 keV. At this energy a direct measurement of the $^{13}\text{C}(\alpha,n)$ cross section is extremely difficult and therefore one has to extrapolate the measured data to lower energies. Since the cross section is expected to be influenced by a subthreshold resonance and a resonance just above threshold, R-matrix analyses are especially suited for this extrapolation. An exact measurement of the excitation function and the angular distribution of the $^{13}\text{C}(\alpha,\alpha)$ reaction, which represents another decay channel of the ^{17}O compound nucleus, can provide additional constraints to improve the quality and reliability of the R-matrix extrapolation. New results for the $^{13}\text{C}(\alpha,\alpha)$ differential cross section in the energy range 2.6 - 6.2 MeV are presented. The experiment was performed with the 10 MV tandem accelerator at the University of Notre Dame. Angular distributions for 29 angles between 44 deg and 165 deg could be obtained for 375 energies. Preliminary results for the reaction rate at $kT = 8$ keV are presented.

* Nucl. Phys. A688 (2001) 499c - 501c

¹ University of Notre Dame, Notre Dame, Indiana 46556, USA

² Los Alamos National Laboratory, New Mexico 87545, USA

2. Alpha and Neutron Induced Reactions on Ruthenium*

W. Rapp, H.J. Brede¹, D. Hentschel², F. Käppeler, H. Klein¹, R. Reifarth, T. Rauscher³

The uncertain origin of the proton-rich Mo and Ru isotopes has motivated cross section measurements of α - and neutron-induced reactions. The experiments were performed via the activation technique by irradiating thin layers of natural ruthenium with α -particle beams close to the Gamov window of the *p* process between 7.0 and 10.5 MeV. The cross sections of the reactions $^{96}\text{Ru}(\alpha,\gamma)$, $^{96}\text{Ru}(\alpha,n)$, $^{96}\text{Ru}(\alpha,p)$, and $^{98}\text{Ru}(\alpha,n)$ could be determined with uncertainties of typically 10%. On average, these results are about two to three times smaller than recent statistical model predictions. Additional activations in a quasi-stellar neutron spectrum corresponding to $kT=25$ keV allowed to obtain the complementary stellar (n, γ)-cross sections for ^{96}Ru , ^{102}Ru , and ^{104}Ru . In these cases the agreement with model calculations is considerably better.

*Nucl. Phys. A688 (2001) 427c - 429c

¹ Physikalisch-Technische Bundesanstalt, Bundesallee 100, D-38116 Braunschweig, Germany

² Forschungszentrum Karlsruhe, Institut für Nukleare Entsorgung

³ Departement für Physik und Astronomie, Universität Basel, Klingelbergstrasse 82, CH-4056 Basel, Switzerland

3. The keV Neutron Capture Cross Sections of $^{128,129,130}\text{Xe}$ *

R. Reifarth, F. Käppeler, F. Voss, K. Wisshak.

The (n, γ) cross sections of the important *s*-process nuclei ^{128}Xe , ^{129}Xe , and ^{130}Xe have been measured in the astrophysically relevant neutron energy range from 3 to 225 keV. Neutrons were produced via the $^7\text{Li}(p,n)^7\text{Be}$ reaction by bombarding metallic Li targets with the pulsed proton beam of the Karlsruhe 3.7 MV Van de Graaff accelerator. High pressure gas samples of enriched xenon were used in the experiment, and capture events were registered with the Karlsruhe 4 π Barium Fluoride Detector. The cross sections were determined relative to the gold standard with overall uncertainties of 1.5 - 2.5% over most of the investigated energy range. From these results Maxwellian averaged stellar (n, γ) cross sections with typical uncertainties of 2% were calculated for thermal energies between $kT=8$ keV and 100 keV. In contrast to previous theoretical estimates, which were known to exhibit uncertainties of 30 to 50%, this work provides a reliable basis for quantitative astrophysical analyses.

* Nucl. Phys. A688 (2001) 229c - 232c

FZKA-Report 6694, Forschungszentrum Karlsruhe (2001)

4. The Stellar Neutron Capture Rate of ^{147}Pm for Constraining the *s*-Process Neutron Density *

C. Arlandini, M. Heil, R. Reifarth, F. Käppeler, P.V. Sedyshev¹

The stellar (n, γ) cross section of the unstable isotope ^{147}Pm , which represents an important branch point in the *s*-process reaction path, has been measured by activation at 25 keV thermal energy. The experiment was difficult because the sample mass was limited to only 28 ng due to the relatively short half-life of 2.62 yr. In spite of considerable background from various impurities, partial cross sections for feeding both the 5.37 d ground state and the 41.3 d isomer in ^{148}Pm could be determined, yielding a preliminary value for the total (n, γ) cross section of 685 ± 100 mbarn at a thermal energy of $kT = 25$ keV. This result can be used for improved statistical model calculations of the (n, γ) cross sections of the additional branch point isotopes ^{147}Nd and ^{148}Pm . These data are required as a basis for considerably refined analyses of the *s*-process branchings at $A=147/148$ which are probing the neutron density in the He-burning zone of low-mass AGB stars.

*Nucl. Phys. A688 (2001) 487c - 489c

¹Frank Laboratory of Neutron Physics, JINR, 14980 Dubna, Moscow Region, Russia

5. The *s*-Process Branchings at ^{151}Sm , ^{154}Eu , and ^{163}Dy *

J. Best, H. Stoll, C. Arlandini, S. Jaag, F. Käppeler, K. Wisshak, A. Mengoni¹, G. Reffo¹, T. Rauscher²

The (n, γ) cross sections of the stable europium isotopes ^{151}Eu and ^{153}Eu have been measured by irradiating oxide samples in a quasi-stellar neutron spectrum. From the induced activities, the stellar cross sections of ^{151}Eu and ^{153}Eu at a thermal energy of $kT=30$ keV were found to be $\langle\sigma v\rangle/v_T = 3821 \pm 152$ mb and 2733 ± 110 mb, respectively. These results allowed to resolve severe discrepancies among previous data. Similar activation measurements were also performed on $^{152,154}\text{Sm}$ and $^{164,170}\text{Er}$. Among these results, the stellar cross section of the *s*-only isotope ^{164}Er

$\langle\sigma v\rangle/v_T=1084\pm 51$ mb at $kT=30$ keV is particularly important. Statistical model calculations were performed with emphasis on the effect of excited states as well as on the unstable isotopes ^{151}Sm , ^{152}Eu , and ^{154}Eu . The combined set of cross sections was used for an updated analysis of the branchings at $A = 151, 152, 154$, and 163 . The temperature and density estimates derived via the classical approach are discussed and compared to stellar models for helium shell burning in low mass stars.

***Phys. Rev. C (2001) 64: 015801 (15)**

¹ E.N.E.A., Applied Physics Division, V. Don Fiammelli 2, and Centro Dati Nucleari, V. Martiri di Monte Sole 4, I-40129 Bologna, Italy

² Departement für Physik und Astronomie, Universität Basel, Klingelbergstrasse 82, CH-4056 Basel, Switzerland

6. Neutron Capture on $^{180\text{m}}\text{Ta}$: Clue for an *s*-Process Origin of Nature's Rarest Isotope *

K. Wisshak, F. Voss, C. Arlandini, F. Becvar¹, O. Straniero², R. Gallino³, M. Heil, F. Käppeler, M. Krücka¹, S. Masera³, R. Reifarth, C. Travaglio⁴

The neutron capture cross section of $^{180\text{m}}\text{Ta}$ has been measured in the keV range, yielding a stellar average of 1465 ± 100 mbarn at $kT=30$ keV. Though the sample contained only 6.7 mg $^{180\text{m}}\text{Ta}$ (at an enrichment of 5.5%), the few capture events could be separated from the much larger background by a unique combination of high efficiency, good energy resolution, and high granularity of the Karlsruhe 4π BaF₂ detector. A detailed *s*-process analysis based on this first experimental value indicates that $^{180\text{m}}\text{Ta}$ is predominantly of *s*-process origin.

*** Phys. Rev. Lett. 87 (2001) 251102 (4)**

¹ Faculty of Mathematics and Physics, Charles University, CZ-180 00 Prague, Czech Republic

² Osservatorio Astronomico di Collurania, I-64100 Teramo, Italy

³ Dipartimento di Fisica Generale, Università di Torino and INFN, Sezione di Torino, Via P. Giuria 1, I-10125 Torino, Italy

⁴ Max-Planck Institut für Astronomie, Königstuhl 17, D-69117 Heidelberg, Germany

FORSCHUNGSZENTRUM KARLSRUHE

INSTITUT FÜR REAKTORSICHERHEIT

1. Monte Carlo Calculation of Sensitivities to Secondary Angular Distributions

U. Fischer, I. Kodeli¹, R. Perel²

Based on the differential operator method, computational schemes are being developed for the Monte Carlo calculation of point detector sensitivities to secondary angular distributions (SAD) with the objective to implement them into the MCNP code. By using the Legendre series expansion approximation for the SAD, suitable sensitivity algorithms have been elaborated and implemented into MCSEN, a local update to the MCNP Monte Carlo code. MCSEN is thus capable of calculating point detector sensitivities for the SAD Legendre coefficients. Validation of this approach has been achieved through the benchmarking against deterministic sensitivity calculations for an iron spherical shell assembly with a central 14 MeV neutron source. Comparison calculations for SAD sensitivities were performed with the SUS3D code based on first order perturbation theory using direct and adjoint flux spectra as provided by one-dimensional ANISN transport calculations. Direct comparison calculations using perturbed angular cross-section data were performed both with the ANISN and the MCNP code. Good agreement was achieved for the individual sensitivity profiles as well as for the calculated flux changes as compared to the direct calculations. Table 1 shows the calculated changes due to a 5% decrease of the first elastic scattering moment as predicted by MCSEN and SUS3D sensitivity calculations and as provided by direct MCNP and ANISN calculations with perturbed SAD data. The Monte Carlo technique for calculating point detector sensitivities to secondary angular distributions is thus qualified for application to sensitivity analyses of fusion integral experiments.

Table 1 Relative change [%] of neutron flux integrals due to a 5% decrease of the first elastic scattering moment: Results of MCSEN and SUS3D sensitivity calculations vs. direct MCNP and ANISN calculations using perturbed SAD data.

Upper energy [MeV]	MCSEN	SUS3D	MCNP (perturbed SAD)	ANISN (perturbed SAD)
0.098	0.806	0.801	0.797	0.796
1.00	0.737	0.747	0.700	0.741
4.97	0.699	0.698	0.680	0.695
7.41	0.383	0.447	0.399	0.437
10.0	0.456	0.401	0.422	0.391
13.8	-0.733	-0.464	-0.618	-0.499
19.6	-1.336	-1.460	-1.356	-1.437
Total	0.000	-0.020	-0.015	-0.020

¹ Institut "Jozef Stefan", Jamova 39, 1000 Ljubljana, Slovenia

² Racah Institute of Physics, Hebrew University of Jerusalem, 91904 Jerusalem, Israel

2. Simulation of D-Li Source Neutrons in Monte Carlo Transport Calculations

U. Fischer, A. Konobeyev¹, Yu. Korovin¹, U. v. Möllendorff,
P. Pereslavytsev¹, S. P. Simakov

The International Fusion Material Irradiation Facility (IFMIF) uses the d-Li stripping reaction to produce high-energy neutrons. An appropriate representation of the d-Li neutron source term is required for design calculations of IFMIF. The Monte Carlo code McDeLicious was developed to simulate in the transport calculation the neutron generation on the basis of evaluated double-differential d + ^{6,7}Li cross-section data. These were prepared employing a newly developed method based on diffraction theory, a modified intranuclear cascade model and standard evaluation techniques [1]. The evaluated data include cross-sections for all reaction channels up to 50 MeV deuteron energy as well as energy-angle distributions for the neutrons emitted through the various ^{6,7}Li(d,xn)-reactions. McDeLicious [2] is a further enhancement of McDeLi, based on a built-in semi-empirical d-Li reaction model with the new ability to sample the generation of d-Li source neutrons from the processed d + ^{6,7}Li cross-section data. The McDeLicious approach was tested against available experimental thick lithium target data. It was shown that the resulting total neutron yield as well as the neutron angle-energy spectra can be predicted with considerably better accuracy than with the semi-empirical McDeLi approach and the intra-nuclear cascade model of the high energy particle Monte Carlo code MCNPX, see Figs. 1 and 2 for the comparison of thick lithium target neutron yield data. With the McDeLicious approach the accuracy of the d-Li neutron source term can be steadily improved by updating the d + ^{6,7}Li data evaluations.

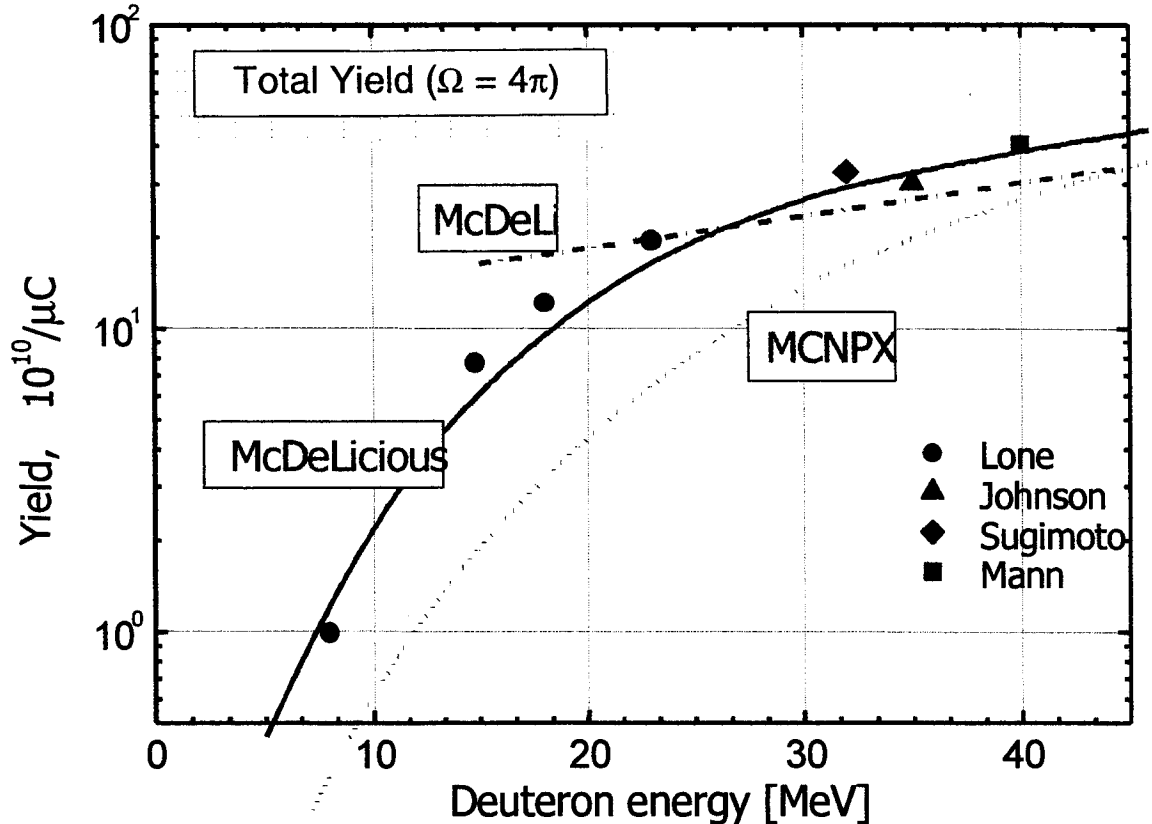


Fig. 1 Thick target d-Li forward neutron yield data.

¹ Institute for Nuclear Power Engineering (INPE), 249020 Obninsk, Kaluga Region, Russia

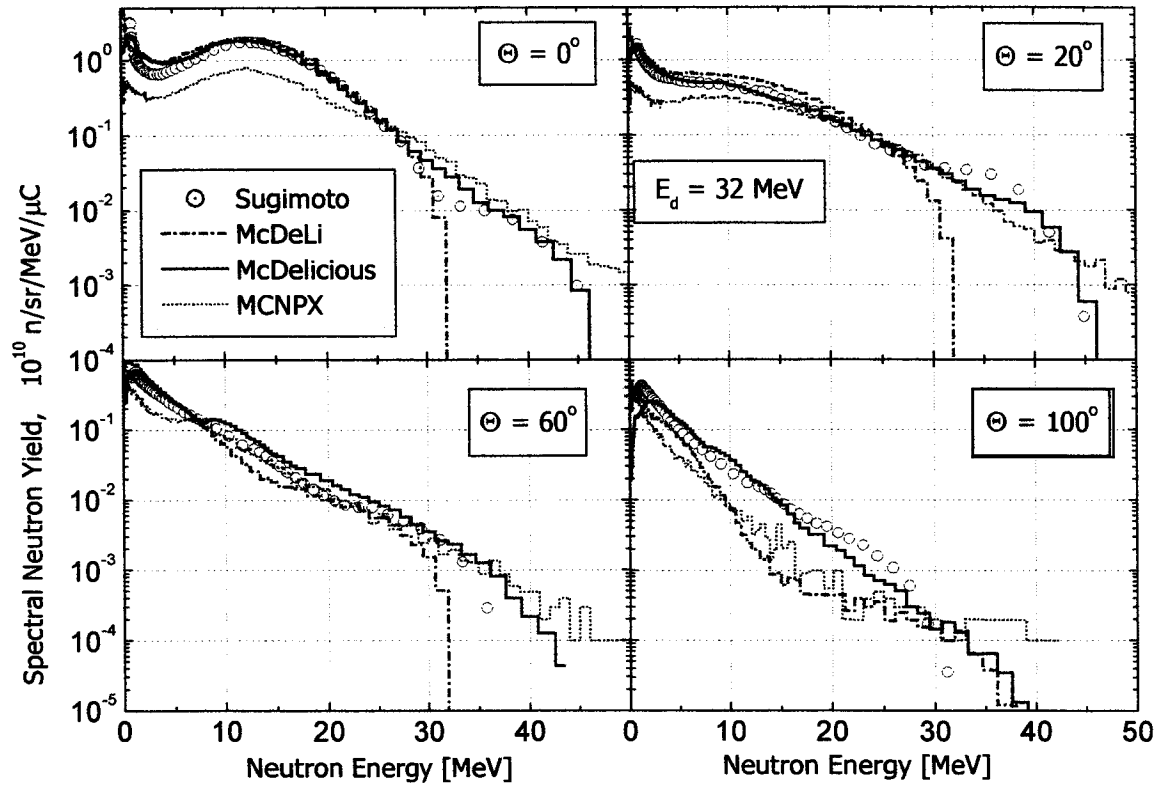


Fig. 2 Thick lithium target neutron yield spectra at 32 MeV incident deuteron energy.

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3. Intermediate Energy Activation File (IEAF-2001)

U. Fischer, A. Konobeyev¹, Yu. Korovin¹, D. Leichtle, U. v. Möllendorff, P. Pereslavl'tsev¹, I. Schmuck, S. P. Simakov

The Intermediate Energy Activation File (IEAF-2001) is a new compilation of neutron-induced activation cross sections in ENDF-6 data format for activation analyses in fusion technology and intermediate energy applications [1]. The IEAF-2001 data library contains neutron-induced activation cross sections in the energy range 10^{-5} eV to 150 MeV for 679 target nuclides from Z=1 (hydrogen) to 84 (polonium). The total number of reaction channels with activation cross-section data is 134,431. The European Activation File (EAF-99) served as a basis for the activation data below 20 MeV neutron energy. Threshold reaction cross-sections were evaluated on the basis of geometry dependent hybrid exciton and evaporation models using a modified version of the ALICE code. The IEAF-2001 data library can be processed with standard ENDF processing tools such as NJOY. A groupwise IEAF-2001 data library has been prepared for 256 energy groups in GENDF data format. Application tests were successfully performed for the activation analysis of the high flux test module of the International Fusion Material Irradiation Facility (IFMIF) using the recent ALARA activation code and the 256 energy group GENDF data library [2,3].

The IEAF-2001 data library is available on CD-ROM from the NEA Data Bank, Paris, both with pointwise ENDF and groupwise GENDF data. The IEAF-2001 library can be used with any activation code capable of handling an arbitrary number of reaction channels. Testing of the IEAF data against integral experiments is underway to qualify and further improve the IEAF library.

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¹ Institute for Nuclear Power Engineering (INPE), 249020 Obninsk, Kaluga Region, Russia

INSTITUT FÜR NUKLEARCHEMIE FORSCHUNGSZENTRUM JÜLICH

1. Neutron Activation Cross Sections

P. Reimer[†], A. Plompen^{††}, F. Cserpák^{*}, S. Sudár^{*}, R. Dóczi^{*}, T. Bastian^{**}, I. Spahn,
S. Spellerberg, J. Csikai^{*}, S.M. Qaim

As mentioned in the last Progress Report, a good triangular cooperation has developed between Jülich, Geel and Debrecen. Measurements in the neutron energy range of 7 to 12 MeV are done using a DD gas target at the compact cyclotron CV 28 in Jülich, for energies around 14 MeV use is made of a DT neutron generator at Debrecen, and the energy range of 14 to 20 MeV is covered by a DT source at the Van de Graaff machine in Geel. The data obtained are of considerable technological importance.

Studies on $^{nat}\text{V}(\text{n},\alpha)^{47}\text{Sc}$ and $^{94}\text{Mo}(\text{n},\text{p})^{94}\text{Nb}$ reactions were completed. Due to high scientific contents, two papers were published in journals dealing with fundamental nuclear physics (one in Phys.Rev.C and the other in Nucl.Phys.A, cf. Publications at the end of this contribution). Work on fast neutron induced reactions on the radioactive target nucleus ^{99}Tc has also been completed. A detailed manuscript is in preparation and shall be soon submitted to a physics journal. Investigations on (n,p) and (n, α) reactions on ^{107}Ag and ^{109}Ag , performed last year up to 14 MeV, have now been extended till 20 MeV. The excitation function of the $^{196}\text{Hg}(\text{n},2\text{n})^{195\text{m,g}}\text{Hg}$ reaction is not known well. Since mercury is a favourite target material for a spallation neutron source, we initiated studies on this reaction, First measurements around 14 MeV have been done. Work in other energy regions is in progress.

For integral testing of differential data some measurements have been started with 14 MeV d(Be) neutrons. Of particular interest are cross section data of a few (n,p) reactions which lead to the formation of low-energy β^- emitting radionuclides of therapeutic importance.

[†] Also at Institute for Reference Materials and Measurements (IRMM), Geel, Belgium

^{††} Institute for Reference Materials and Measurements (IRMM), Geel, Belgium

^{*} Institute of Experimental Physics, Debrecen University, Debrecen, Hungary

^{**} Abteilung Nuklearchemie, Universität zu Köln, Germany

A special case is the $^{89}\text{Y}(\text{n,p})^{89}\text{Sr}$ ($T_{1/2} = 50.5$ d; $E_{\beta^-} = 1.5$ MeV) reaction which is presently under study. Another important case is the $^{67}\text{Zn}(\text{n,p})^{67}\text{Cu}$ ($T_{1/2} = 61.9$ h; $E_{\beta^-} = 0.6$ MeV) reaction which will also be investigated.

2. Charged Particle Induced Reaction Cross Sections

K. Kettern, K. Hilgers, S. Kastleiner, S. Spellerberg, B. Scholten, F. M. Nortier*, T. N. van der Walt*, F. Tárkányi†, Z. Kovács†, S. Sudár**, Y. Skakun††, H. E. Hassan***, H.H. Coenen, S. M. Qaim

As in previous years, systematic studies on charged particle induced reaction cross sections for medical applications were continued. During the period of the present report following investigations were carried out.

a) *Excitation functions of nuclear reactions for production of positron emitters (^{14}O , ^{76}Br , ^{82}Sr (^{82}Rb) and ^{124}I)*

The radionuclide ^{14}O ($T_{1/2} = 70$ s) is formed via the $^{14}\text{N}(\text{p,n})$ -process. It is of interest from three points of view: (a) as an impurity during the production of ^{11}C using the $^{14}\text{N}(\text{p},\alpha)$ -reaction, (b) as a substitute for ^{15}O ($T_{1/2} = 2.0$ min) in PET studies, (c) for producing radioactive beam for astrophysical research. Experimental measurements and data analysis were completed. With the suggested data set, the yields of ^{14}O can now be confidently calculated.

The positron emitting radionuclide ^{76}Br ($T_{1/2} = 16.0$ h) and the γ -emitting radionuclide ^{77}Br ($T_{1/2} = 57.0$ h) are of considerable medical importance. A renewed interest in these two radioisotopes in our laboratory motivated us to measure excitation functions of the following reactions from their thresholds up to about 30 MeV: $^{76}\text{Se}(\text{p,n})^{76}\text{Br}$, $^{77}\text{Se}(\text{p,xn})^{76,77}\text{Br}$ and $^{\text{nat}}\text{Se}(\text{p,xn})^{76,77}\text{Br}$. Preliminary studies have been performed and further work is continuing.

* National Accelerator Centre (NAC), Faure, South Africa

† Institute of Nuclear Research (ATOMKI), Debrecen, Hungary

** Institute of Experimental Physics, Debrecen University, Debrecen, Hungary

†† Kharkov Institute of Physics and Technology (KIPT), Kharkov, Ukraine

*** Cyclotron Project, Atomic Energy Authority, Cairo, Egypt

The radionuclide ^{82}Sr ($T_{1/2} = 25.3$ d) is in great demand for preparing the most commonly used ^{82}Sr (^{82}Rb) generator system for PET studies. It is produced either via spallation or via the $^{85}\text{Rb}(p,4n)^{82}\text{Sr}$ reaction. In the latter case the cross section data base was rather weak and the experimental and theoretical yields showed considerable discrepancies. In connection with our detailed studies on the production of ^{83}Sr via the $^{85}\text{Rb}(p,3n)$ -reaction, reported in the last two Progress Reports, we also measured the excitation function of the $^{85}\text{Rb}(p,4n)^{82}\text{Sr}$ reaction up to 100 MeV using both X-ray and γ -ray spectrometry. From the measured curve the integral yield was calculated and is given in Fig. 1. For comparison the yield calculated from the only other measurement reported in the literature is also given. Our data show that the calculated yield around 70 MeV is about 30 % lower than the hitherto accepted value.

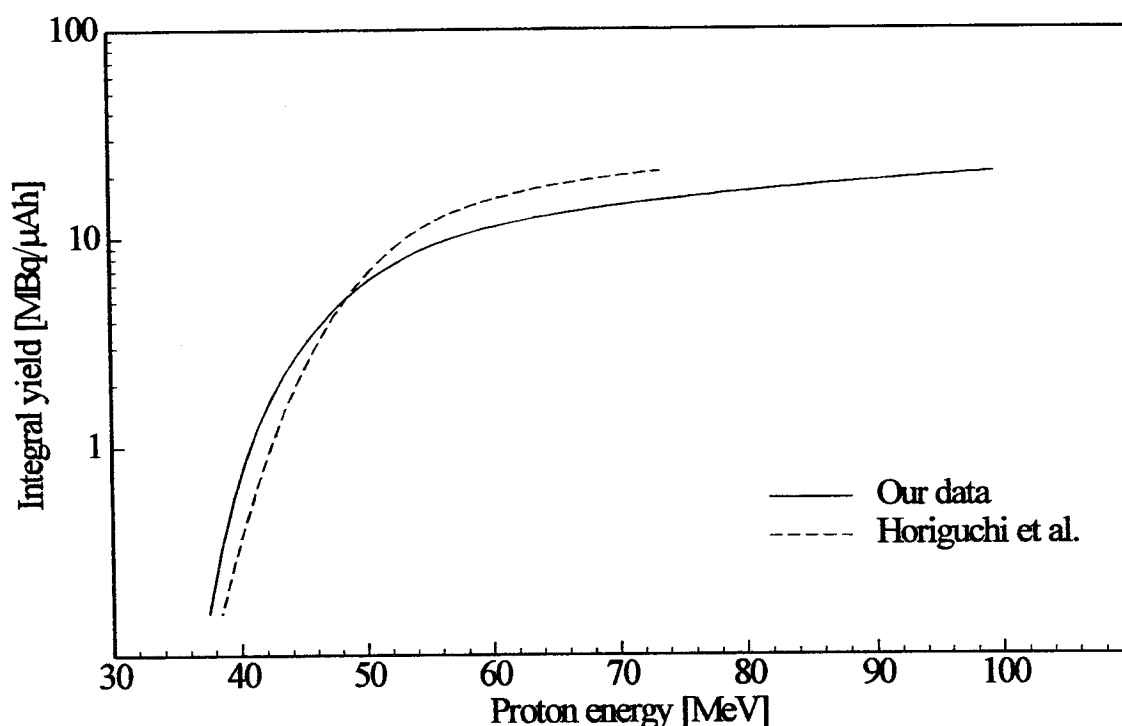


Fig. 1 Integral yield of ^{82}Sr in the $^{85}\text{Rb}(p,4n)$ -reaction as a function of incident proton energy, calculated from the excitation function measured in this work and that reported by Horiguchi et al. (Int.J.Appl.Radiat.Isot. 31, 141(1980)). The present yield value around 70 MeV is about 30 % lower.

The significance of the positron emitting radionuclide ^{124}I ($T_{1/2} = 4.18 \text{ d}$) is increasing both in diagnosis and therapy. As reported in the last two Progress Reports, detailed cross section measurements on the $^{124}\text{Te}(\text{p},\text{n})^{124}\text{I}$, $^{124}\text{Te}(\text{d},2\text{n})^{124}\text{I}$ and $^{125}\text{Te}(\text{p},2\text{n})^{124}\text{I}$ reactions depicted that the (p,n) process leads to the highest purity product whereas the (p,2n) reaction gives the highest yield. We now initiated a study of the $^{126}\text{Te}(\text{p},3\text{n})^{124}\text{I}$ reaction. When completed, it should allow a comparison of the two intermediate energy reactions, viz. $^{125}\text{Te}(\text{p},2\text{n})^{124}\text{I}$ and $^{126}\text{Te}(\text{p},3\text{n})^{124}\text{I}$ processes, with regard to the production of ^{124}I . The criteria for selection of one of the two processes for use at a medium-sized cyclotron would be, as ever, the yield and the radionuclidic purity of ^{124}I .

b) Cross sections and yields of therapy related and other radionuclides (^{64}Cu , ^{88}Y and ^{103}Pd)

The radionuclide ^{64}Cu ($T_{1/2} = 12.7 \text{ h}$) is of great interest in systemic endoradiotherapy. It is produced via two major routes, viz. $^{64}\text{Ni}(\text{p},\text{n})^{64}\text{Cu}$ and $^{64}\text{Zn}(\text{n},\text{p})^{64}\text{Cu}$, the former using a small-sized cyclotron and the latter using a nuclear reactor. Two other processes, namely $^{66}\text{Zn}(\text{d},\alpha)^{64}\text{Cu}$ and $^{68}\text{Zn}(\text{p},\alpha\text{n})^{64}\text{Cu}$, have also been suggested but the cross section data were not known. During the period of the present Progress Report those two reactions were investigated radiochemically in detail, in case of deuterons up to 14 MeV and in case of protons up to 30 MeV. The thick target yields of ^{64}Cu calculated from the measured data are, however, much lower than those from the presently used processes. The only advantage of the reactions investigated here could be that no isotopically enriched target material is needed.

The radionuclide ^{88}Y ($T_{1/2} = 106.6 \text{ d}$) is suitable for studying slow biokinetics of importance in therapy planning prior to the use of the β^- emitting therapy nuclide ^{90}Y ($T_{1/2} = 64.1 \text{ h}$). Detailed cross section measurements were therefore performed on the $^{\text{nat}}\text{Sr}(\text{p},\text{xn})^{87\text{m,g}}\text{Y}$ processes up to 25 MeV and from the data obtained the yield of ^{88}Y (as well as of the $^{87\text{g}}\text{Y}$ impurity) was calculated. The results show that ^{88}Y can be conveniently produced at a small-sized cyclotron.

The X-ray and Auger electron emitting radionuclide ^{103}Pd ($T_{1/2} = 17.0$ d) is gaining increasing importance in brachytherapy. It is produced routinely via the $^{103}\text{Rh}(p,n)$ -reaction but the cross section data were not known well. As mentioned in the last two Progress Reports, we investigated the $^{103}\text{Rh}(p,xn)^{103,101,100}\text{Pd}$ reactions up to 40 MeV in detail, using X-ray and γ -ray spectrometry, and carried out a critical evaluation of the available data for the $^{103}\text{Rh}(p,n)$ -process. Furthermore, statistical model calculation was done using the code STAPRE. The recommended excitation function is given in Fig. 2. It may be useful to all the groups engaged in production of ^{103}Pd . Due to the high importance of this radionuclide we also investigated two alternative routes, namely $^{102}\text{Ru}(^3\text{He},2n)^{103}\text{Pd}$ and $^{100}\text{Ru}(\alpha,n)^{103}\text{Pd}$, which could possibly be used for production of ^{103}Pd . Final analysis of data is under way and the work is reaching completion.

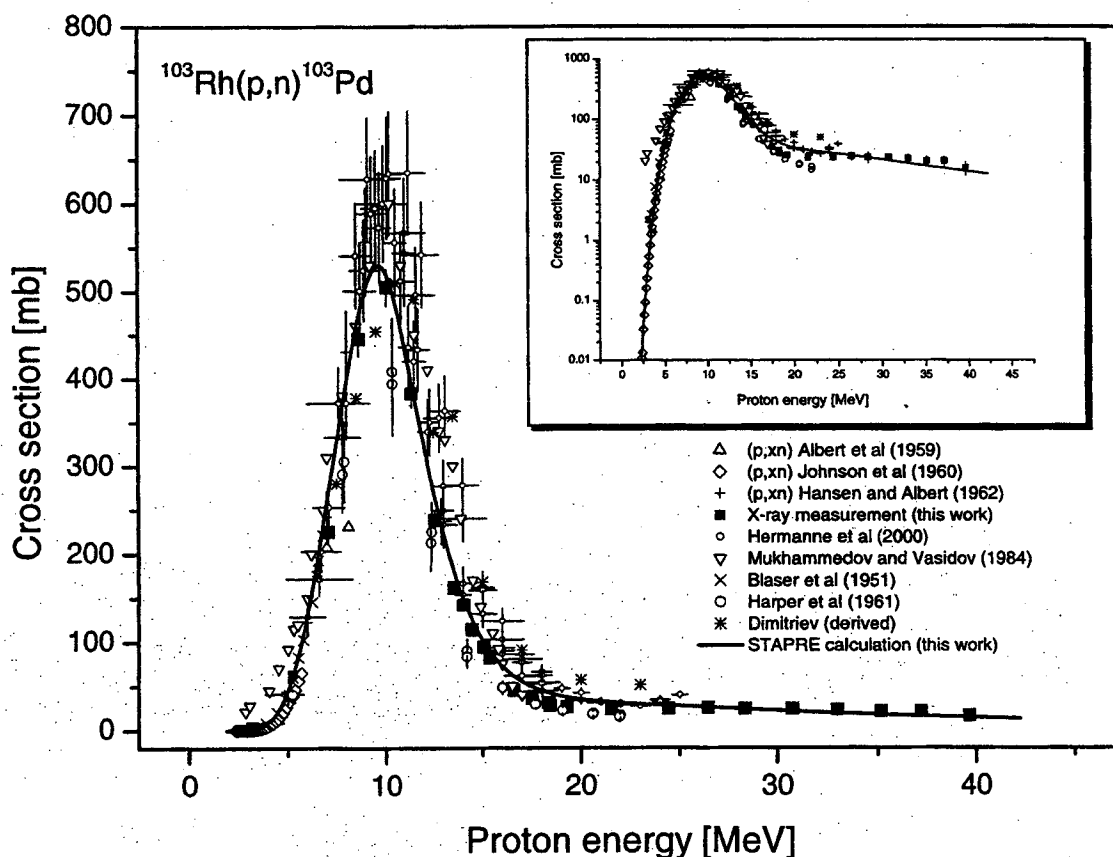


Fig. 2 Excitation function of the $^{103}\text{Rh}(p,n)^{103}\text{Pd}$ reaction with all available data obtained via neutron counting and activation measurement. For comparison the result of nuclear model calculation is also given. In the inset the data are shown on a logarithmic scale to depict the energy dependence in the threshold region.

3. Other Activities

a) Ph.D. Theses

Two Ph.D theses were completed during the period of the Progress Report. They were submitted to the Faculty of Natural Sciences of the University of Cologne and successfully defended. Thereafter they were published as Reports of the Forschungszentrum Jülich. The details are:

Sascha Kastleiner

Kernchemische Untersuchungen zur Produktion einiger medizinisch relevanter Strontium- und Rubidium-Isotope

Report JÜL-3931 (2001).

Peter Reimer

Fast neutron induced reactions leading to activation products: selected cases relevant to development of low activation materials, transmutation and hazard assessment of nuclear wastes

Report JÜL- 3980 (2002)

b) Chair of INDC

Syed M. Qaim was appointed by the IAEA as Chairman of the International Nuclear Data Committee (INDC). The term of office will cover the period January 2002 to December 2005.

Publications (During the Period of the Progress Report)

- [1] S.M. Qaim: Cross section data for production of diagnostic and therapeutic radionuclides. Proc. Advisory Group Meeting on "Long Term Needs for Nuclear Data Development", IAEA, Vienna, 28 November – 1 December 2000, Report INDC(NDS)-428 (2001) 7

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Papers presented at the International Conference on Nuclear Data for Science and Technology, Tsukuba, Japan, October 2001, and to be published in the J. Nucl. Sci. Technology, 2002

- A. J. M. Plompen, P. Reimer, V. Avrigeanu and S. M. Qaim
Vanadium cross section measurements by the activation technique and evaluations from threshold to 20 MeV

- A. J. M. Plompen, D. L. Smith, P. Reimer, S. M. Qaim, V. Semkova, F. Cserpák, A. Avrigeanu and S. Sudár
Recent neutron activation cross section measurements (*Invited*)
- S. M. Qaim, F. T. Tárkányi, P. Oblozinský, K. Gul, A. Hermanne, M. G. Mustafa, F. M. Nortier, B. Scholten, Yu. N. Shubin, S. Takács and Y. Zhuang
Charged-particle cross section data base for medical radioisotope production
- B. Scholten, E. Hess, S. Takács, Z. Kovács, F. Tárkányi, H. H. Coenen and S. M. Qaim
Cross section measurements on gas targets relevant to the production of the positron emitting radionuclides ^{14}O , ^{18}F and ^{76}Br
- S. M. Qaim, S. Kastleiner, A. Hohn, F. M. Nortier, B. Scholten, T. N. van der Walt and H. H. Coenen
Nuclear data for production of the longer-lived positron emitters ^{83}Sr and ^{124}I with intermediate energy protons
- S. M. Qaim
Nuclear data for production of new medical radionuclides (*Invited*)

INSTITUT FÜR KERN- UND TEILCHENPHYSIK TECHNISCHE UNIVERSITÄT DRESDEN

1. Integral Activation Experiment with EUROFER*

R. A. Forrest¹, H. Freiesleben, V. D. Kovalchuk², D. V. Markovskij³, D. V. Maximovich²,
K. Seidel, S. Unholzer

The radioactivity induced by neutrons in the materials of a fusion device represents a central topic of safety-related investigations. Radionuclides of a broad range of half-lives have to be included in the corresponding analyses. The short-term radioactivity (half-life ranging from the order of magnitude of minutes to weeks) is of interest with respect to heat production and shut-down dose rates, whereas the long-term radioactivity (half-life of the order of 10 – 100 years or more) determines the waste management. The radioactivity is mainly produced by neutrons of 14 MeV energy, where the number of open reaction channels is maximum, and at thermal energy, where capture cross sections are large. In the present work, the radioactivity induced by 14 MeV neutrons in EUROFER was investigated. The EUROFER steel was developed in the European Fusion Technology Programme as a structural material with reduced activation [1].

In a calculation with the European Activation System (version EASY-99) [2], EUROFER was taken to be irradiated with 14 MeV neutrons of a flux density corresponding to a power of 1.0 MW/m², for a period of one year. The results obtained for the contact dose rate as a function of the decay time after irradiation are shown in Fig. 1. After about 100 years the dose rate is expected to be below the hands-on limit. 70% of the long-term radioactivity originates from ²⁶Al produced by the ²⁷Al(n,2n) reaction. For shorter decay times, there are two groups of half-lives of the dominant radionuclides, one with T_{1/2} of a few minutes (up to hours) and the other with T_{1/2} of the order of days and month. Therefore, the irradiation experiment was divided into two different parts, directed to the short-term radioactivity and to the longer term one.

* Work supported by the European Fusion Development Activity Programme (Task TTMN-002) and by the German Bundesministerium für Bildung und Forschung (Projekt RUS-564-98)

¹ EURATOM/UKAEA Fusion Association, Culham Science Centre, Abingdon, United Kingdom

² Coordination Centre "Atomsafety", Sergiev Posad, Russia

³ Russian Research Centre "Kurchatov Institute", Moscow, Russia

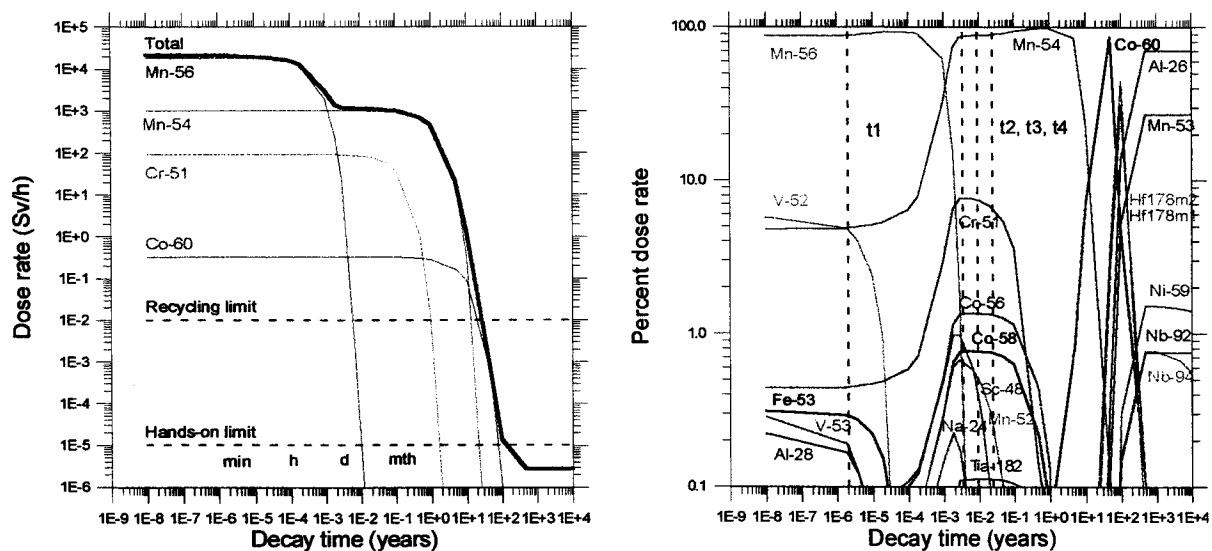


Fig. 1 Contact dose rate (left hand) and contributions of different radionuclides to the total dose rate (right hand) after irradiation of EUROFER with 14 MeV neutrons of 1.0 MW/m² power for one year as a function of decay time.

Small pieces of EUROFER were irradiated at the high-intensity D-T neutron generator SNEG-13 [3] at Sergiev Posad for 3.0 min with a fluence of $3.36 \cdot 10^{11}$ neutrons/cm² and for 13.93 h with a fluence of the order of 10^{14} neutrons/cm² at two different sample positions, resulting in different neutron energies. Gamma-spectra of the samples were measured with a Ge(Li)-spectrometer during decay at times labelled t1 ... t4 in Fig. 1, and nuclide activities were derived. For each of the measured values the activity was calculated with EASY-99, and calculated-to-experimental values (C/E) were determined. The results are presented in Tables 1 and 2. For the activities of radionuclides, which determine the short-term radioactivity (Table 1), deviations from C/E = 1.0, larger than the uncertainty range, were found in three cases only. The sum of the activities measured and of the dose rate, which comprises 97 % of the total one expected, are underestimated by less than 20%.

For the activities of radionuclides, which determine the dose rate up to about 10 y, the C and E values agree within their uncertainties (Table 2), with the exception of ⁵⁴Mn, where a significant underestimation was found. Some energy dependence of the C/E can be stated for ⁵¹Cr as in previous experiments with steels. The sums of the calculated and measured

activities agree within ~ 20 %, and the radioactivity calculated for reactor neutron fluxes can be validated at this level.

^{26}Al and ^{53}Mn are dominant in the range of the long-term radioactivity (Fig.1). The small Al content of the EUROFER sample [1] was confirmed by the C/E obtained for the ^{24}Na activities (Table 2). Cross section and half-life uncertainties for $^{27}\text{Al}(n,2n)^{26}\text{Al}$, the channel with 99.9 % contribution, result in 30 % uncertainty of the calculated value. ^{53}Mn is produced to 98 % by $^{54}\text{Fe}(n,d)$. The uncertainty of the calculated dose rate for this channel was estimated by EASY-99 to be at 60 %.

Table 1 Radionuclides measured after short irradiation, their half-lives, the γ -rays used to measure the activity, the producing nuclear reactions with their contribution to the activity (EASY-99 calculation), and the ratios of experimental-to-calculated activity (C/E). The mean neutron incident energy was $\langle E_n \rangle = 14.93$ MeV.

Radio-nuclide	Half-life	E_γ (keV)	Reaction contribution (%)		C / E
^{28}Al	2.24 min	1778	$^{28}\text{Si}(n,p)$	99.5	1.00 ± 0.32
^{51}Ti	5.8 min	320	$^{51}\text{V}(n,p)$	68.1	0.91 ± 0.20
			$^{54}\text{Cr}(n,\alpha)$	31.8	
^{52}V	3.75 min	1434	$^{52}\text{Cr}(n,p)$	94.5	0.74 ± 0.06
			$^{53}\text{Cr}(n,d)$	3.5	
			$^{55}\text{Mn}(n,\alpha)$	2.0	
^{53}V	1.62 min	1006	$^{53}\text{Cr}(n,p)$	97.8	0.82 ± 0.10
			$^{54}\text{Cr}(n,d)$	2.2	
^{56}Mn	2.58 h	846	$^{56}\text{Fe}(n,p)$	99.2	0.94 ± 0.04
		1810	$^{57}\text{Fe}(n,d)$	0.8	
		2113			
^{57}Mn	1.6 min	122	$^{57}\text{Fe}(n,p)$	97.7	1.11 ± 0.16
			$^{58}\text{Fe}(n,d)$	2.3	
^{53}Fe	8.51 min	378	$^{54}\text{Fe}(n,2n)$	100	1.33 ± 0.56

Table 2 Radionuclides measured after longer irradiation with neutrons of two different energies.

Radio-nuclide	Half-life	E_γ (keV)	Reaction	$\langle E_n \rangle = 14.93$ MeV		$\langle E_n \rangle = 14.37$ MeV	
				contribution (%)	C/E	contribution (%)	C / E
^{24}Na	14.97 h	1369	$^{27}\text{Al}(n,\alpha)$	69.1	0.80 ± 0.21	69.1	1.16 ± 0.29
			$^{27}\text{Al}(n,\alpha)\text{IT}$	30.9		30.9	
^{48}Sc	1.82 d	984	$^{48}\text{Ti}(n,p)$	5.1	1.02 ± 0.11	5.7	1.01 ± 0.12
		1038	$^{51}\text{V}(n,\alpha)$	94.9		94.3	
		1312					
^{51}Cr	27.7 d	320	$^{52}\text{Cr}(n,2n)$	88.7	$1.08 \pm$	86.4	0.83 ± 0.06
			$^{54}\text{Fe}(n,\alpha)$	0.07		13.6	
				11.3			
^{54}Mn	312 d	835	$^{55}\text{Mn}(n,2n)$		0.80 ± 0.05	19.1	0.66 ± 0.04
			$^{54}\text{Fe}(n,p)$	22.5		80.8	
			$^{56}\text{Fe}(n,t)$	76.9			
				0.6			
^{56}Mn	2.58 h	846	$^{56}\text{Fe}(n,p)$		0.92 ± 0.10	99.6	0.96 ± 0.07
		1810	$^{57}\text{Fe}(n,d)$	99.2			
		2113		0.8			

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2. Measurement and Analysis of Neutron and γ -ray Flux Spectra in SiC*

Y. Chen^{1,2}, U. Fischer¹, H. Freiesleben, C. Negoita, K. Seidel, S. Unholzer

Silicon carbide composites are candidates for advanced structural materials in fusion reactors, mainly due to their low decay heat and activation properties. Integral experiments are required to benchmark the nuclear data, involved in design calculations, both with regard to the activation and to the particle transport behaviour. After activation experiments (e. g. Ref. [1]), the transport data were tested in a benchmark experiment. A block of sintered SiC, borrowed from JAERI Tokai -mura, was irradiated at the Frascati Neutron Generator [2] with 14 MeV neutrons and the neutron and γ -ray flux spectra were measured at four positions inside the assembly.

The experimental set-up is shown in Fig. 1. The dimensions of the SiC block were 45.7 cm x 45.7 cm x 71.1 cm of length (x-axis). The neutron source was located at $x = -5.3$ cm. Flux spectra were measured in four positions (P 1,...P 4) at $x = 12.70, 27.94, 43.18$ and 58.42 cm. The SiC material had a density of 3.185 g/cm^3 and an elemental composition (private communication with JAERI) of 30.828 wt-% of C, 68.889 wt-% of Si, 0.19 wt-% of B, 0.079 wt-% of Al and 0.014 wt-% of Fe.

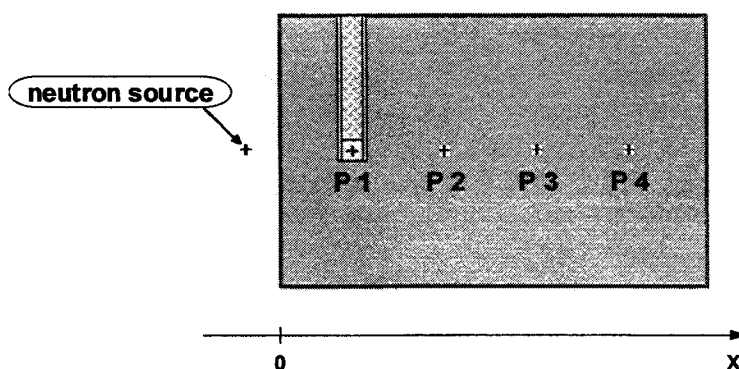


Fig. 1 Geometry of the benchmark assembly with the detector in position 1.

¹ Forschungszentrum Karlsruhe, Institut für Reaktorsicherheit

² Institute of Plasma Physics Hefei, P. R. China

* Work supported by the European Fusion Development Activity Programme (Task TTMN-002)

Neutron and γ -ray flux spectra were measured simultaneously, using a NE 213 liquid-scintillation spectrometer. The dimensions of the cylindrical active volume of the detector were 38 mm in both height and diameter. The material had a mass density of 0.874 g/cm^3 and an elemental composition of 54.8 at-% H and 45.2 at-% C. The scintillator was coupled to a photomultiplier by means of a 500 mm long light guide. When the detector was located at one of the positions (P 1 in Fig. 1), the other ones were filled with bricks of SiC.

The pulse-height response matrix of the detector was determined on the basis of detailed simulations of experimental distributions from mono-energetic neutron and γ -sources with the MCNP-4A code. The DIFBAS code [3] was employed for unfolding the measured pulse-height distributions of the present experiment in order to generate the neutron and γ -flux spectra. They were obtained as absolute fluxes, as the response matrix has been determined on an absolute scale. Results are shown in Fig. 2 together with the calculated flux spectra.

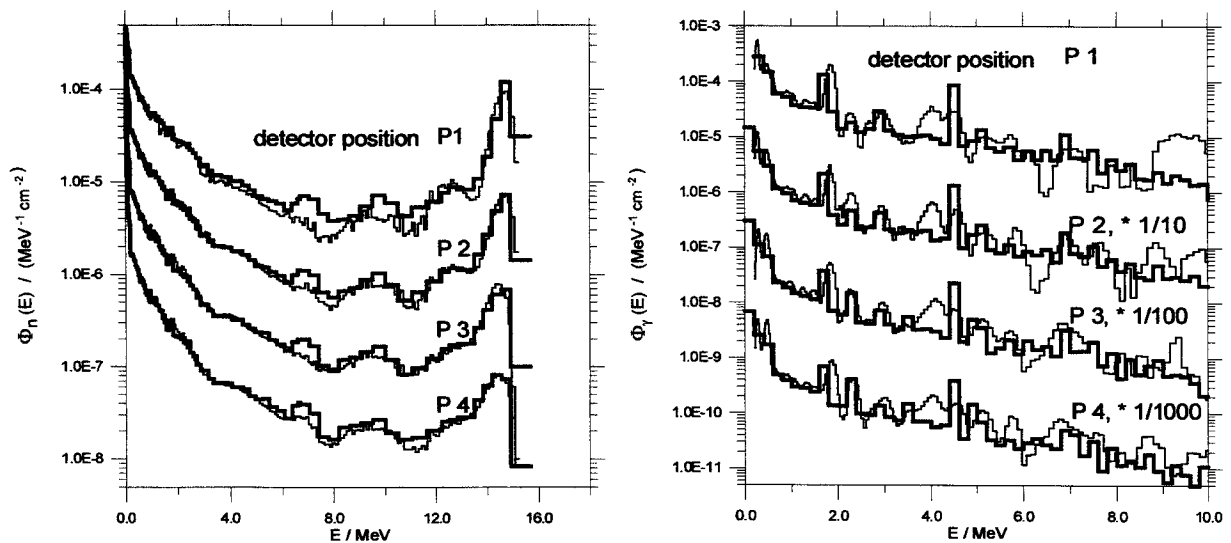


Fig. 2 Measured (thin line) and calculated (thick line) neutron (left hand) and γ -ray (right hand) flux spectra at the four detector positions, normalised to one source neutron.

The computational analysis of the flux spectra was performed with the Monte Carlo code MCNP-4C [4] using a full 3D geometry model of the assembly, the neutron generator and the experimental hall. Nuclear data were taken from the FENDL/MC-2.0 [5] data library except for ^{28}Si , for which the new data evaluation of the European Fusion File EFF-3.0 was used.

A comparison of the measured spectra with the calculated ones shows that the fast neutron flux in a SiC assembly of a thickness, corresponding to about 10 times the mean free path for

14 MeV neutrons, can be reproduced by these calculations within 20%. The underestimation of the neutron flux for the interval $5.0 \text{ MeV} < E < 10. \text{ MeV}$ may be attributed to inelastic scattering cross sections from individual levels. A sensitivity analysis is underway to verify this conjecture.

The γ -ray flux for $E > 0.4 \text{ MeV}$ is underestimated, increasingly with the detector position, by up to 26%. This may be attributed to some γ -peaks that are not sufficiently reproduced and, perhaps, to the uncertainty of the B impurity content of the SiC assembly. Further analysis is required to clarify this problem.

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3. Observation of 3d Metal Ion Charge State Distributions in the Dresden EBIT: Atomic Data for Astrophysics and Plasma Diagnostics

F. Grossmann¹, U. Kentsch, S. Landgraf, V.P. Ovsyannikov¹, F. Ullmann¹, G. Zschornack

Highly charged ions are gaining increasing importance for applications in research fields such as atomic and plasma physics, astrophysics, radiobiology and materials research as well as in technological fields such as nano-structuring, potential sputtering and ion beam lithography. The Dresden EBIT, a room temperature EBIT without any cryogenic techniques, is a long-term stable device for the production of ions of nearly all charged states over a wide range of the Periodic Table of the Elements [1-4]. A 3D-representation and a more detailed explanation of the operation principle can be found in [1,5,7].

¹ LEYBOLD Systems + Service GmbH, Dresden

One of the main activities in 2001 was the observation of charge state distributions of highly charged 3d metal ions by energy and wavelength disperse X-ray spectroscopy. The availability of an easy to handle “table-top” source for spectroscopic investigations on highly charged ions is an excellent possibility to obtain atomic data that are important in astrophysics or plasma diagnostics and modelling.

Highly charged argon, manganese, iron and nickel ions were produced in the electron beam at electron excitation energies of 8.0 and 14.4 keV. The X-ray emissions from direct excitation (DE) and radiative recombination (RR) processes were measured with an energy disperse Si(Li) solid state detector [6,7]. The observed X-ray lines were analysed and compared with results from MCDF atomic structure calculations. In the RR spectrum, the contribution from capture into bare nuclei, hydrogen- and helium-like ions could be identified as shown in Fig.1 for the example of iron. To analyse the ion charge state distribution in the ion trap, the X-ray lines from DE processes were used to determine contributions from helium-, lithium and beryllium-like ions. For helium- and hydrogen-like ions as well as for bare nuclei, the X-ray lines from RR processes were analysed to get information about the contributions of these ions to the charge state distribution. Based on the spectroscopic information the densities of ions from different ionisation stages were calculated. Table 1 shows the results for argon, iron, nickel and manganese.

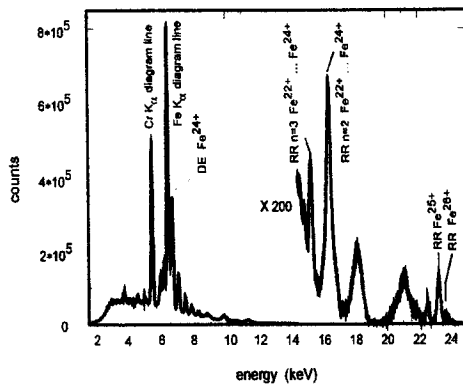


Fig. 1 Iron X-ray spectrum measured at $E_e = 14.4$ keV and $I_e = 43$ mA. The region of direct excitation processes and RR processes into differently ionised iron ions is shown. Note the different scales for DE and RR processes.

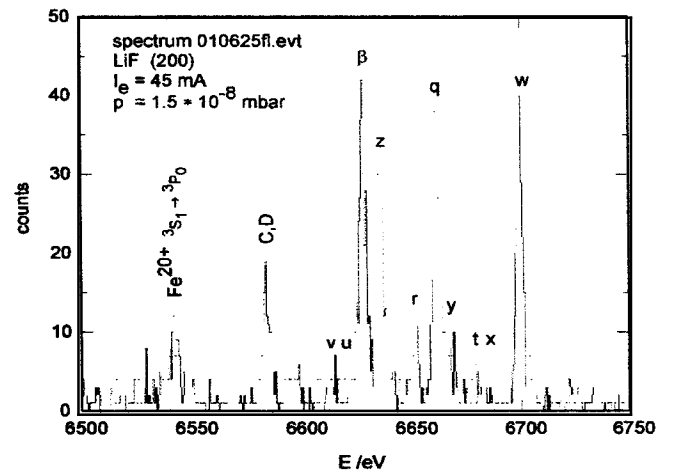


Fig. 2 X-rays from DE processes in highly charged iron ions measured with a LiF (200) crystal at $E_e = 14.4$ keV and $p = 1.5 \cdot 10^{-8}$ mbar.

The ion charge state distributions were studied also by wavelength-disperse X-ray spectroscopy using a crystal diffraction spectrometer equipped with a flat LiF (200) crystal. Contributions from helium- to carbon-like ions were observed with an uncertainty of about 1 eV. Fig. 2 shows an iron spectrum at an electron energy of 14.4 keV and a gas pressure of $1.5 \cdot 10^{-8}$ mbar. Utilising calculated excitation cross-sections, the ion charge state distribution was determined as described in more detail in [6]. The results of the measurements were in good agreement with model calculations using a computer code from Kalagin et al. [8].

The transitions for Fe^{24+} in Fig. 2 are labelled as follows: w: $1s2p \ ^1P_1 \rightarrow 1s^2 \ ^1S_0$, z: $1s2s \ ^3S_1 \rightarrow 1s^2 \ ^1S_0$, x: $1s2p \ ^3P_2 \rightarrow 1s^2 \ ^1S_0$ and y: $1s2p \ ^3P_1 \rightarrow 1s^2 \ ^1S_0$. The notations r, q, v and u characterise transitions in Fe^{23+} , β in Fe^{22+} and C, D in Fe^{21+} , respectively. For the determination of atomic data, it is possible to produce every 3d element at every ionisation stage.

Table 1 Results from the analysis of DE and RR X-ray lines to deduce the ion charge state distribution in the ion trap at different excitation conditions.

Argon $Z = 18$ at $E_e = 8$ keV					
q	16	17	18		
n(RR)	3.1(7)	8.4(6)	1.9(6)		
Manganese $Z = 25$ at $E_e = 14.4$ keV					
q	24	25			
n(RR)	2.1(7)	4.2(6)			
Iron $Z = 26$ at $E_e = 14.4$ keV					
q	22	23	24	25	26
n(DE)	1.7(8)	3.9(8)	5.7(8)		
n(RR)			3.4(8)	3.2(7)	4.1(6)
Nickel $Z = 28$ at $E_e = 14.4$ keV					
q	27	28			
n(RR)	1.0(7)	5.3(5)			

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**ZENTRUM FÜR STRAHLENSCHUTZ UND RADIOÖKOLOGIE,
UNIVERSITÄT HANNOVER
AND
ABTEILUNG NUKLEARCHEMIE, UNIVERSITÄT ZU KÖLN**

**Cross Sections for the Production of Residual Nuclides at Medium-Energies
Relevant for Accelerator-Driven Technologies**

W. Glasser¹, R. Michel¹, S. Neumann², H. Schuhmacher³, V. Dangendorf³, R. Nolte³, U. Herpers⁴,
A.N. Smirnov⁵, I. Ryzhov⁵, A.V. Prokofiev⁶, P. Malmborg⁶, D. Kollár⁷, J.-P. Meulders⁸

Introduction

Integral cross sections for the production of residual nuclides by medium-energy nuclear reactions are of importance for many fields of basic and applied sciences ranging from astrophysics over space and environmental sciences, medicine, accelerator technology, space and aviation technology to *accelerator driven transmutation of waste (ADTW)* and *energy amplification (ADEA)*. The data needs of those applications cover cross sections for proton- and (more importantly) neutron-induced reactions. The particular problem for accelerator driven technologies is that the data needs are extreme with respect to both, target element coverage and types of reaction data. Since it will be practically impossible to measure all relevant data, one will have to rely to a large degree on theoretical estimates.

Given the fact that the predictive power of present day models and codes does not satisfy the requirements, an initiative was taken by a number of European laboratories to improve this situation. To this end, a complete experimental data base was and is being established for a selected number of target elements depending on which newly developed models shall be scrutinized.

¹ Zentrum für Strahlenschutz und Radioökologie, Universität Hannover

² Present address: TÜV Nord, Hamburg

³ Physikalisch Technische Bundesanstalt, Braunschweig

⁴ Abteilung Nuklearchemie, Universität zu Köln

⁵ Khlopin Radium Institute, St. Petersburg, Russia

⁶ The Svedberg Laboratory, University Uppsala, Sweden

⁷ Department of Nuclear Physics, Comenius University, Bratislava, Republic of Slovakia

⁸ Institut de Physique Nucléaire, Université Catholique de Louvain, Louvain La Neuve, Belgium

This initiative started with a European Concerted Action “Lead for Transmutation” [1] and continued by the 5th Framework project HINDAS “High- and Intermediate-Energy Data for Accelerator-Driven Systems” [2, 3]. Here, we report on some aspects of work-package 3 of HINDAS, namely the production of residual nuclides by proton- and neutron-induced reactions.

For proton-induced reactions, these investigations aim to further develop and complete the cross section database which was established by our collaboration in recent years [4, 5, and references therein]. It is now extended to heavy target elements such as Ta [6], W [6 - 8], Pb [9], and Bi [6]. New, but still unpublished data are available for the target element iron. Work on the target element uranium for proton-energies up to 72 MeV is underway. All those data allow for stringent tests of nuclear models and codes when calculating cross sections for residual nuclide production from thresholds up to 2.6 GeV.

For neutron-induced reactions the situation is worse. Contrary to their importance, the availability of neutron cross sections for the production of residual nuclides above 30 MeV is marginal. A solution of this problem is to perform irradiation experiments with quasi-monoenergetic neutrons produced by the ${}^7\text{Li}(p,n){}^7\text{Be}$ reaction.

Production of Residual Nuclides by Neutron-Induced Reactions up to 175 MeV

Within the HINDAS project, activation experiments are performed at TSL/Uppsala and UCL/Louvain La Neuve in order to determine excitation functions for the production of residual radionuclides from a variety of target elements up to 175 MeV.

The neutron beam-line at TSL/Uppsala [10] is equipped with a special irradiation chamber (PARTY facility) allowing for parasitic activation experiments [11]. Irradiations can be performed with peak neutron energies up to 175 MeV. Neutron flux densities of $\sim 0.5 \cdot 10^5 \text{ cm}^{-2} \text{ s}^{-1} \mu\text{A}^{-1} (\text{mm Li})^{-1}$ are obtained in the high energy peak of the neutron spectrum. 99.984 % ${}^7\text{Li}$ -targets with a thickness between 2 and 15 mm can be used. Proton currents of 10 μA are available for energies up to 100 MeV; they are about a factor of ten lower for higher energies.

The UCL cyclotron facility provides neutron beams in the energy range from about 25 to 70 MeV, and well-characterized neutron reference fields have been previously established [12]. Proton currents up to 10 μA are possible; with a 5 mm ${}^{\text{nat}}\text{Li}$ target the resulting neutron flux density in the high-energy peak of the neutron spectrum is $\sim 1 \cdot 10^5 \text{ cm}^{-2} \text{ s}^{-1}$ at a distance of 5m from the target.

A total of 10 activation experiments were performed with proton energies of 36.4, 48.5, and 62.9 MeV at the UCL and of 69.1, 76.4, 98.5, 136.7, 148.4, 162.7 and 178.8 MeV at TSL. The target elements C, N, O, Mg, Al, Si, Fe, Co, Ni, Cu, Ag, Te, Pb, and U were irradiated with the highest beam currents available. Cylindrical target stacks with a diameter of 22 mm and a total length of 66 mm were irradiated in the PARTY facility. Typical beam-on-target times were 40 h at UCL and 80 h at TSL. At UCL, cylindrical target stacks with a diameter of 25 mm containing the different target foils were inserted in the center of a larger stack with a 100 mm $\times$ 100 mm cross section which had the same macroscopic cross section as the target stack. The total length of the target stacks was 81 mm. These stack arrangements were positioned at a distance of $\sim$ 5 m from the neutron-producing target. At this position, measurements of the lateral profile of the collimated neutron beam with a beam profile monitor [13] showed a width of the beam (FWHM) of about 55 mm and confirmed that, on the one hand, the target stacks were irradiated homogeneously and, on the other hand, the entire neutron beam passed through the stack arrangement and was attenuated in the same way.

At UCL, the absolute neutron fluence was determined via 3 methods: a proton recoil telescope, a ^{238}U fission ionization chamber, and an NE213 scintillation detector. The spectral fluence was measured with the TOF method, using the latter two systems. A specially developed multi-wire proportional counter yielded the transverse intensity profile of the collimated neutron beam. All instruments employed for the measurement of the neutron fluence and the energy distributions were described in detail elsewhere [12]. The energy distributions of neutrons entering and leaving the stack were determined by TOF spectrometry at distances of about 11 m from the target without and with the stack in its irradiation position. For the two lower neutron energies, a scintillation detector and for the higher energy a ^{238}U fission ionization chamber was used as a spectrometer. Earlier investigations showed that the results obtained with these two devices are in good agreement [12]. Recent investigations [14] show that the spectral fluence below the threshold of the spectrometer of about 4 MeV is essentially constant down to zero neutron energy. At the position of the stack the neutron fluence in the high-energy peak was determined with a proton recoil telescope [15] (PRT). This device is based on the detection of recoil protons produced by elastic neutron scattering on hydrogen in a high-purity polyethylene (PE) radiator foil of 1.5 mm in thickness. Cross sections for n-p scattering were taken from the phase-shift analysis "VL40" which has been recommended as standard cross section data [16]. The uncertainties of 5% for these data dominate the total uncertainty of the fluence measurements.

At TSL, the fluence of neutrons passing through the activation stack is monitored using $^{238}\text{U}(\text{n},\text{f})$ and $^{209}\text{Bi}(\text{n},\text{f})$ reactions as standards [16]. Thin film breakdown counters [17] (TFBC) are used for fission fragment detection. The principle of TFBC operation is based on the phenomenon of electrical breakdown in a track created by a fragment passing through a thin SiO_2 layer. Typically, four neutron monitors were used. A pair of monitors with ^{238}U (99.999% isotopic purity) and/or ^{209}Bi (>99.995% purity) targets was placed upstream the stack, and a similar pair downstream the stack. Each monitor consists of a fissile target and a fission fragment detector mounted close to each other like a sandwich. The monitors operate in a time-of-flight (TOF) mode. However, the short flight path (about 2 m) and unfavorable time structure of the beam at TSL does not allow an explicit separation of the high-energy peak in the neutron spectrum from the low-energy tail. As a consequence, the use of TOF technique is limited at the PARTY facility to the separation of monitor reaction events from the ones due to an intrinsic detector background as well as due to spontaneous and neutron-induced fission of heavier nuclei contamination in the monitor samples.

To obtain the high-energy peak neutron fluence from the monitor count rate, one needs independent information on the fraction of fission events due to peak neutrons. The latter is provided by additional measurements of TOF fission spectra for the same monitor reactions, with similar detector arrangements, in the same beam line but at a longer flight path in the marble hall (10-13 m). There, a much better separation of the high-energy peak (accuracy ~10 %) in the neutron spectrum from the low-energy tail is possible. Assuming that the neutron spectrum at the PARTY location at an emission angle of about 1° is equal to the neutron spectrum at 0° allows extracting the number of neutrons in the high-energy peak irradiating the target stack.

Residual radionuclides with half-lives between 20 min and 5 years were measured by off-line γ -spectrometry. In spite of the long irradiation times and high beam currents applied, the measurement of the irradiated targets is a low-count-rate problem and requires close-to-detector geometries. This results in particular problems with respect to efficiency determination and to necessary corrections for systematic coincidence. In addition, γ -self-absorption in the targets has to be corrected for. Therefore, a new method for the determination of detector efficiencies was developed which takes into account all these effects simultaneously.

The γ -spectrometry of the activated target foils yields activities proportional to production rates P [$\text{s}^{-1} \text{g}^{-1}$]. A production rate P of a nuclide produced is given by:

$$P = \frac{N_L}{A_T} \int \sigma(E) \cdot J(E) dE \quad \text{with} \quad J(E) = \frac{d^2 \Phi(E)}{dE dt} \quad (1)$$

with N_L being Avogadro's number and A_T the atomic mass of target element. $\sigma(E)$ are the neutron cross sections, $J(E)$ the spectral neutron flux densities and $\Phi(E)$ the neutron fluence. The integral is taken over all neutron energies E .

Cross sections cannot be directly calculated from these response integrals since the neutrons used are just "quasi-monoenergetic" with only about 30 to 50% of the neutrons in the high-energy peak with a width of a few MeV. The neutron cross sections $\sigma(E)$ have to be extracted from production rates P_i ($i = 1, \dots, n$) determined in a series of n irradiation experiments with different neutron energies by unfolding. To derive the final cross sections one requires a "guess" excitation function of the respective nuclear reaction, and the spectral fluences in different experiments are needed. This guess excitation function contains all eventually existing experimental neutron cross sections (mostly for $E < 20$ MeV, if at all). For energies or target elements for which no experimental data are available, theoretical neutron excitation functions are calculated as guess functions up to the highest neutron energies applied in the activation experiments by the ALICE-IPPE code [18]. The calculated excitation functions are normalized to the existing experimental cross sections, if available.

Information on the energy dependence of the neutron spectra in the targets was obtained by modeling the neutron spectra by Monte Carlo techniques using the LAHET/MCNP code system [19, 20]. These transport calculations started either from the experimentally determined neutron spectra (at UCL) or from the systematics of experimentally measured neutron emission spectra of the $^7\text{Li}(p,n)$ -reaction [21] (at TSL). The calculations described the transport of the neutrons into the target stacks and into the individual targets as well the production and transport of secondary particles which inside the massive target stacks cannot be neglected. The normalization of the spectra was done on the basis of either direct measurements of the spectra of the impinging neutrons (at UCL) or the neutron fluences measured by the TFBC-monitors (at TSL). Measurements at UCL with and without the target stacks in the beam allowed to determine the attenuation of the neutrons in the stacks and to validate respective calculations of this attenuation using the LAHET/MCNP [19, 20] codes (Fig. 1).

First reports of the performance of the PARTY facility and some results for short- and medium-lived γ -emitting product nuclides are published [11, 22]. From the target elements activated, excitation functions for a total of about 120 reactions will be obtained.

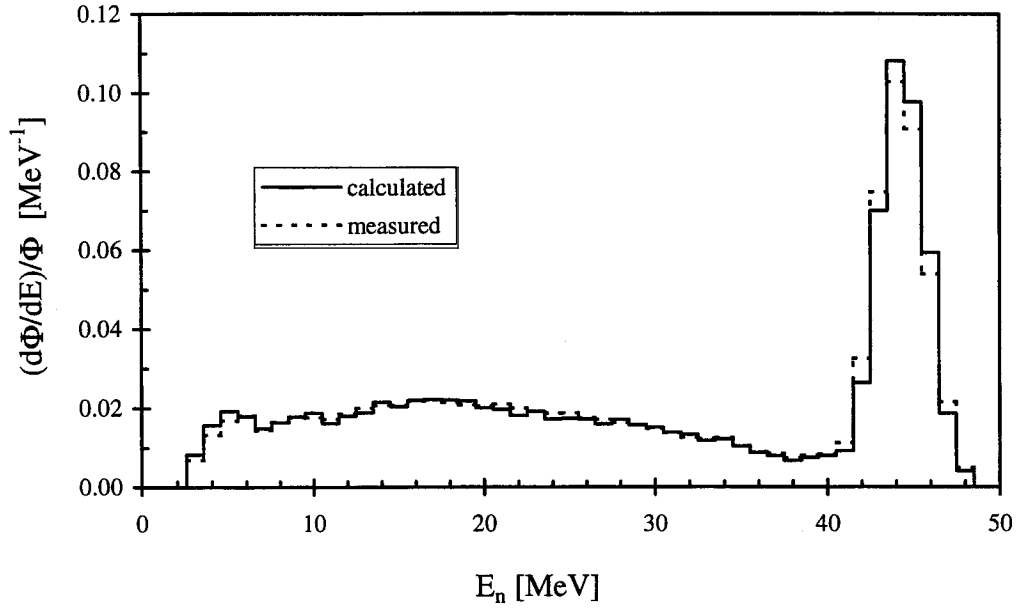


Fig. 1: Comparison of measured (broken line) and calculated (full line) neutron spectra behind the stack of the UCL experiment with $E_p = 36.4$ MeV. The spectra are normalized to a fluence 1 cm^{-2} of neutrons with $E_n > 2.5$ MeV.

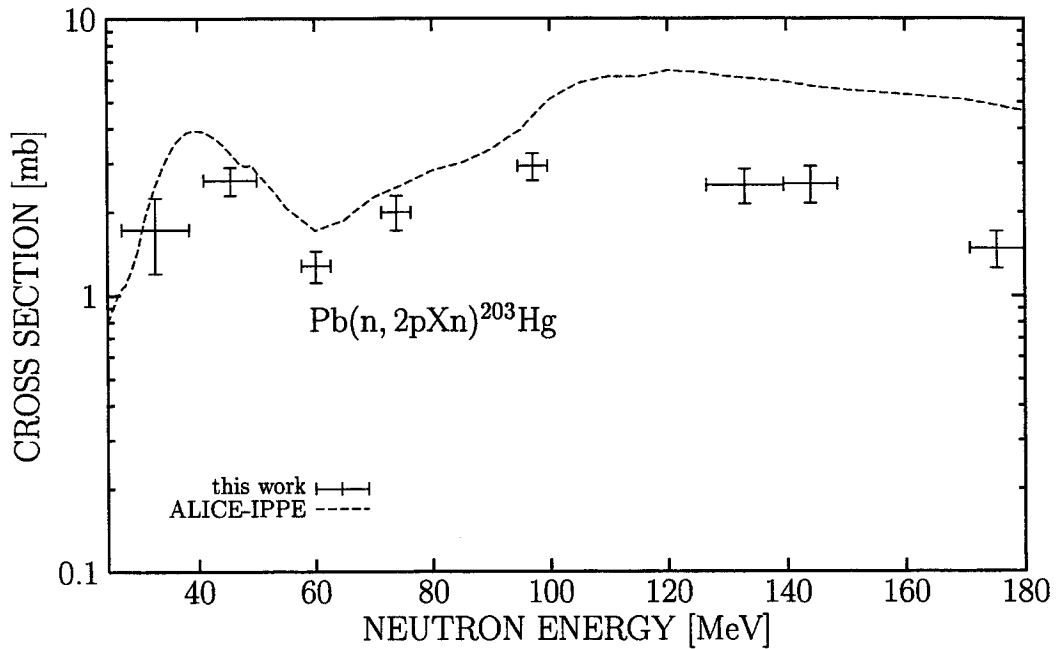


Fig. 2: Guess excitation function and final cross sections for the reaction $^{\text{nat}}\text{Pb}(n,2\text{pxn})^{203}\text{Hg}$. The uncertainties in energy given represent the half-width of the peak in the neutron spectrum.

In Fig. 2, we present as an example the final cross sections and the initial guess excitation function for the $^{nat}\text{Pb}(n,2pxn)^{203}\text{Hg}$ reaction. Presently, work is going on to make use of the total information contained in the experimental production rates for a physically meaningful interpolation of the excitation functions between the data points and to further reduce the uncertainties of the cross sections.

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PHYSIKALISCH-TECHNISCHE BUNDESANSTALT BRAUNSCHWEIG

Measurement of Neutron Scattering Cross Sections on Nitrogen at Energies between 8 and 14 MeV

D. Schmidt, W. Mannhart

Neutron scattering cross sections of nitrogen are needed to calculate the neutron transport, for instance, in ceramic structural materials (fusion technology) as well as in tissue (dosimetry). In the energy region between 8 and 14 MeV, two systematic measurements have been reported [1,2] which differ in many respects. The aim of the present precise measurement was to improve the database for nitrogen in this energy range.

Measurements were carried out at seven incident neutron energies between 7.89 MeV and 13.85 MeV. The measuring setup and the procedure to obtain the differential cross sections was the same as for the previous measurement of iron [3]. The sample consisted of Si_3N_4 with a nitrogen mass content of 38.24%. In order to separate the scattering fractions of silicon, the silicon cross sections were determined in parallel under identical experimental conditions [4]. For excitation energies below 7.5 MeV, eleven partial cross sections of the isotopes ^{28}Si , ^{29}Si and ^{30}Si had been obtained, although the abundances of ^{29}Si and ^{30}Si were below 5%. In this way, all essential scattering contributions of silicon could be determined and correctly separated from the nitrogen scattering part.

Six partial cross sections of ^{14}N were extracted for the J^π/Q levels $1^+/0.0$ (g.s.), $0^+/-2.313$, $1^+/-3.948$, $0^-/-4.915$, $2^-/-5.106$ and the doublet $1^- + 3^-/-5.76$. The 0^+ and 0^- levels are weakly excited, *i.e.* the corresponding cross sections are rather small (in the order of 1 mb/sr). Fig. 1 shows in the upper part an elastic angular distribution as an example. A Legendre polynomial expansion was fitted using least-squares methods, the maximum order was chosen by different criteria [3]. The quality of the measured data is demonstrated in the lower part of Fig. 1 which shows the deviations of measured cross sections from the fitted Legendre polynomial expansion.

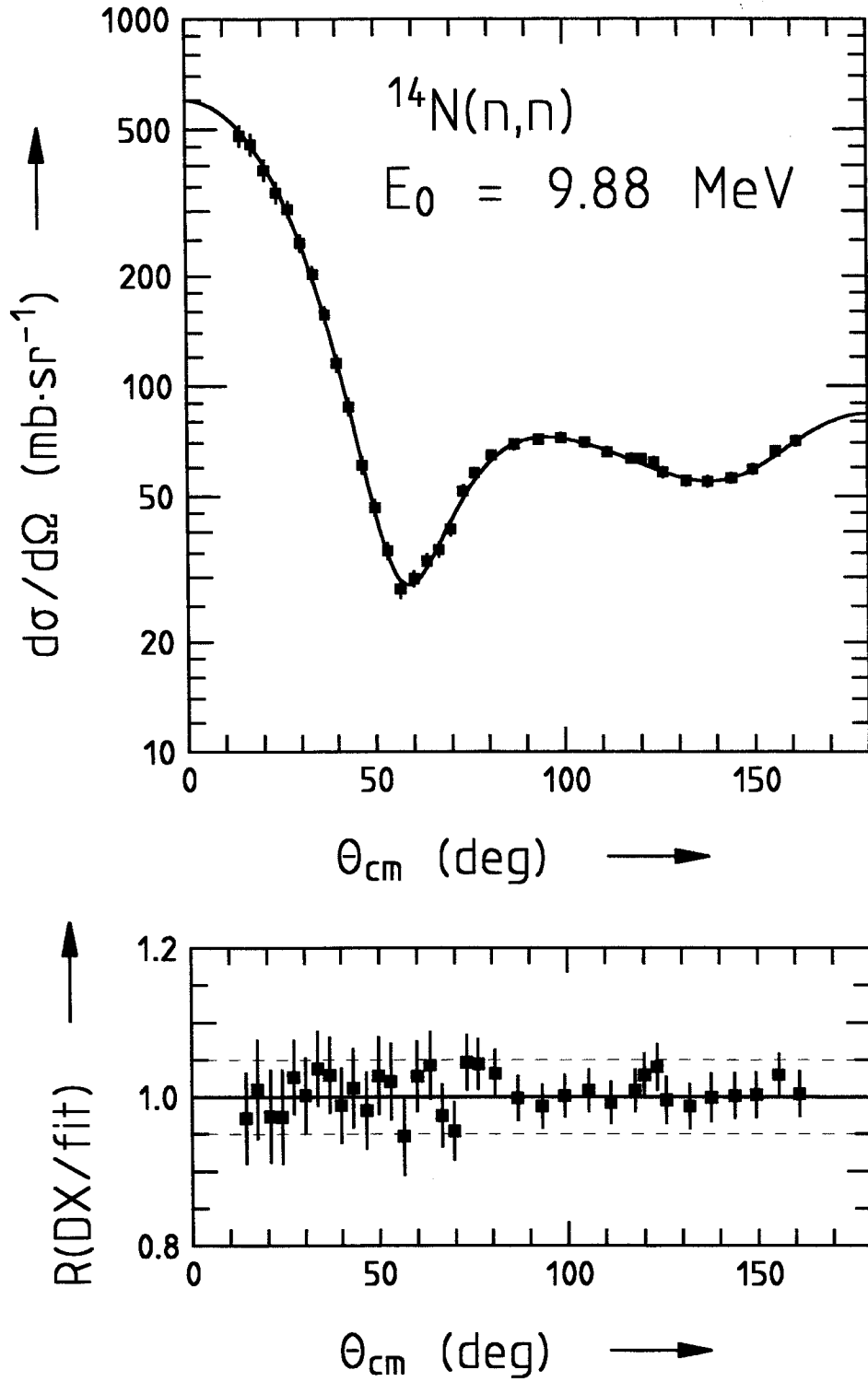


Fig. 1 Upper part: Angular distribution of elastic scattering on ^{14}N ; the points are the measured cross sections, the curve is a fitted Legendre polynomial expansion.

Lower part: Ratio of measured elastic cross sections to the fitted Legendre polynomial expansion; the dashed lines denote $\pm 5\%$.

The angle-integrated elastic cross sections determined are shown in Fig. 2, together with data from the EXFOR database [5]. Our results confirm the measurement of Bruyères-le-Châtel [2], except the energy region between 11 MeV and 13 MeV. Here, our cross sections are about 10% larger, the reason of this discrepancy could not be found.

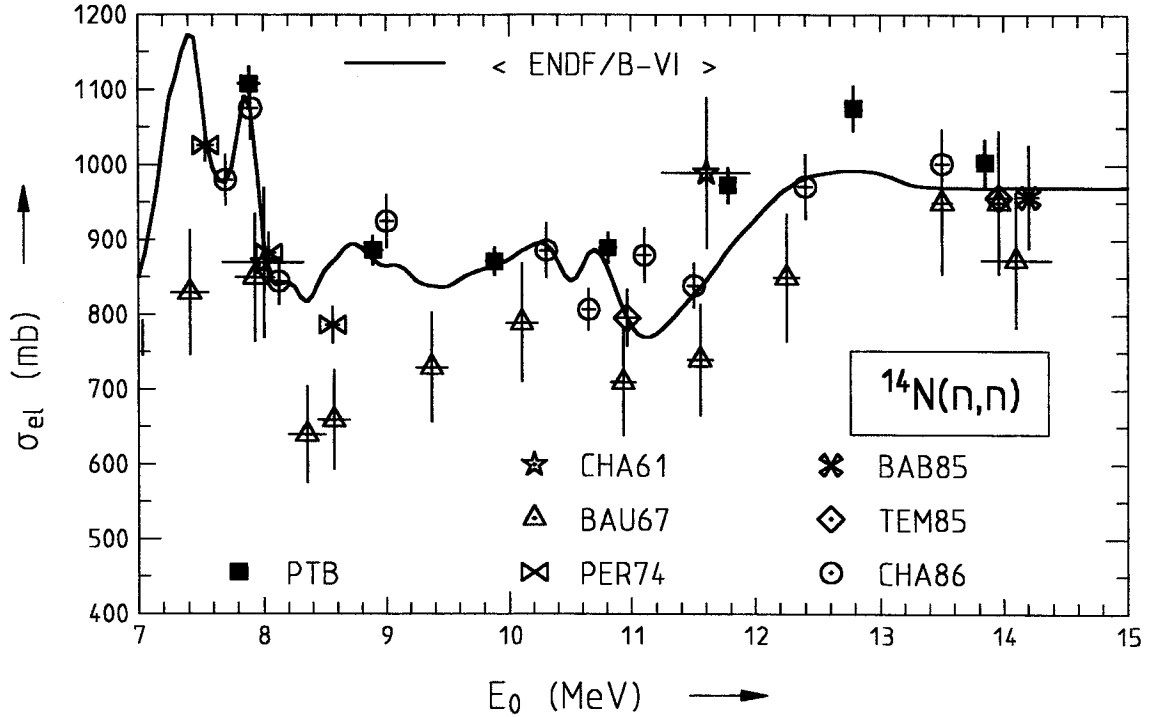


Fig. 2 Angle-integrated cross sections for the elastic scattering of neutron from nitrogen; results obtained by other authors (EXFOR database [5]) are also given; the curve represents the ENDF/B-VI evaluation averaged over 150 keV (energy resolution of the experiment of the present work).

Fig. 3 shows the angle-integrated cross sections of the 0^- -4.915 level. The difference between our results and those from Bruyères-le-Châtel [2] are sometimes by a factor of two. The reason may lie in differences in correction for multiple scattering. We compare the measured TOF spectra with those simulated with an extensive and realistic Monte Carlo code [6]. The multiple scattering fraction which for light nuclei is mostly outside the scattering peaks in the TOF spectra like a "flat background," is taken correctly into account by our method. In a straightforward analysis (raw cross

section determination from peak areas and correction for multiple scattering), this "background" may artificially enhance the peak areas and, hence, the cross sections. This effect plays a major role at small cross sections, as in the case shown in Fig. 3.

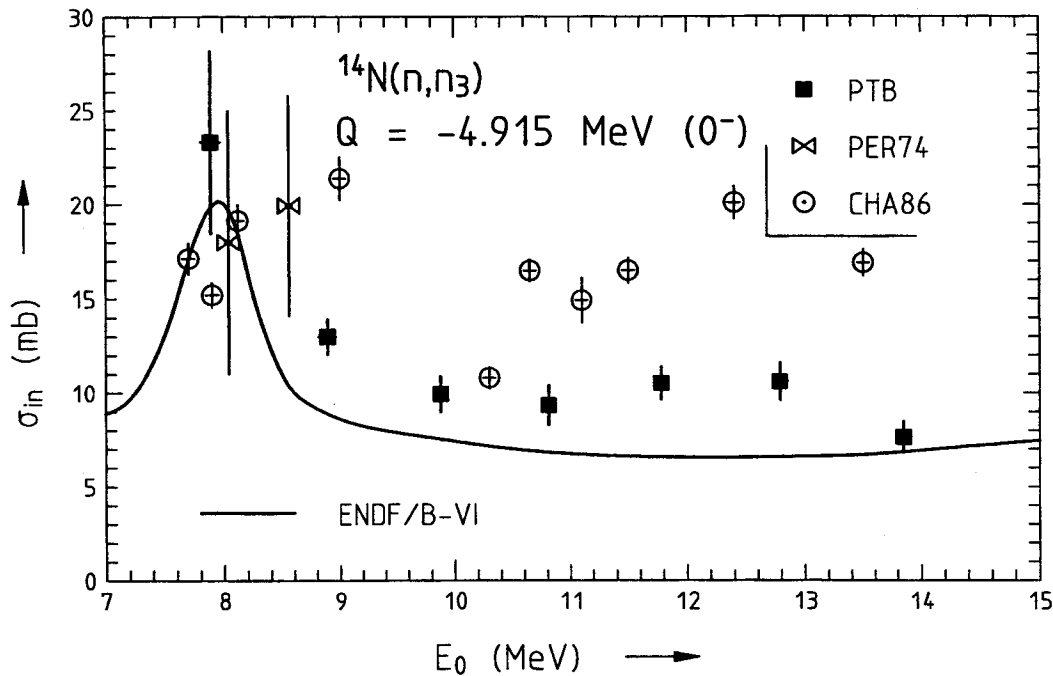


Fig. 3 Same as Fig. 2 but for the $0^-/-4.915$ level.

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A P P E N D I X

Addresses of Contributing Laboratories

Institut für Kernphysik
Director: Prof. Dr. H. Blümer
Reporter: Dr. F. Käppeler
Forschungszentrum Karlsruhe
Postfach 3640
76021 Karlsruhe

Institut für Reaktorsicherheit
Director: Prof. Dr. D. G. Cacuci
Reporter: Dr. U. Fischer
Forschungszentrum Karlsruhe
Postfach 3640
76021 Karlsruhe

Institut für Nuklearchemie
Director: Prof. Dr. H. H. Coenen
Reporter: Prof. Dr. Dr. h. c. S.M. Qaim
Forschungszentrum Jülich GmbH
Postfach 1913
52425 Jülich

Institut für Kern- und Teilchenphysik
Director: Prof. Dr. H. Freiesleben
Reporter: Prof. Dr. K. Seidel
Technische Universität Dresden
Mommsenstr. 13
01062 Dresden

Zentrum für Strahlenschutz und Radioökologie
Head and reporter: Prof. Dr. R. Michel
Universität Hannover
Am Kleinen Felde 30
30167 Hannover

Abteilung Nuklearchemie
Head: Prof. Dr. H. H. Coenen
Reporter: Dr. U. Herpers
Universität zu Köln
Otto-Fischer-Str. 12-14
50674 Köln

Physikalisch-Technische Bundesanstalt
Abteilung Ionisierende Strahlung
Director: Prof. Dr. G. Dietze
Reporter: Dr. W. Mannhart
Bundallee 100
38116 Braunschweig