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Uses of alpha particles, especially in nuclear reaction studies and medical radionuclide production

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Abstract: Alpha particles exhibit three important characteristics: scattering, ionisation and activation. This article briefly discusses those properties and outlines their major applications. Among others, α-particles are used in elemental analysis, investigation and improvement of materials properties, nuclear reaction studies and medical radionuclide production. The latter two topics, dealing with activation of target materials, are treated in some detail in this paper. Measurements of excitation functions of α-particle induced reactions shed some light on their reaction mechanisms, and studies of isomeric cross sections reveal the probability of population of high-spin nuclear levels. Regarding medical radionuclides, an overview is presented of the isotopes commonly produced using α-particle beams. Consideration is also given to some routes which could be potentially useful for production of a few other radionuclides. The significance of α-particle induced reactions to produce a few high-spin isomeric states, decaying by emission of low-energy conversion or Auger electrons, which are of interest in localised internal radiotherapy, is outlined. The α-particle beam, thus broadens the scope of nuclear chemistry research related to development of non-standard positron emitters and therapeutic radionuclides.

Keywords: α-particle beam, elemental analysis, materials research, nuclear reaction studies, medical radionuclide production.

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1 Introduction

1.1 General

An alpha particle is a helium nucleus, i.e. it is a bound entity of two protons and two neutrons. The strong binding of an alpha particle makes it behave more like a nucleon rather than a complex particle, except that it has double the charge of a proton, which leads to a much stronger Coulomb interaction than in the case of a proton. The alpha particle, however, has a spin of 0 in comparison to the value of ½ for a neutron and a proton.

The alpha particle was originally detected in the natural radioactive decay of heavy mass nuclei. Subsequently, a large number of artificially produced heavy mass α-particle emitting radionuclides were characterised. Their half-lives differ considerably but the α-particle energies are usually between 4 and 7 MeV. In general, the α-particle energy increases slowly with the increasing Z of the radioactive nucleus. On the other hand, some synthetically produced neutron-deficient nuclides of medium lifetime in the region of lanthanides emit low-energy α-particles (1–4 MeV), and strongly neutron deficient ones up to 7.4 MeV.

Three important properties of α-particles were established rather early:

a) ionisation (and excitation) of the surrounding medium
b) scattering
c) nuclear interaction

The property of the α-particle to highly ionise and excite atoms (resulting in chemical and biochemical changes) causes rapid loss of its energy in the surrounding medium. Consequently, the ranges of α-particles with $E < 7$ MeV in the matter involved are rather low. This property makes the α-particle very attractive for internal radionuclide therapy. On the other hand, the loss of the intensity of high-energy α-particles is more concentrated towards the end of the path length, resulting in the so-called "Bragg Peak". This characteristic renders the α-particle beam very useful for external radiation therapy, especially for treatment of

deep-lying tumours. Another important use of the highly ionising character of the α-particle is in materials research. Properties of many synthetic and semiconductor materials are improved on irradiation with α-particles.

The scattering of the α-particle, initially observed below the Coulomb barrier, is a very specific property for identifying various elements. It is utilized today for extensive elemental analyses of thin layers of materials using both low and intermediate energy α-particle beams.

The interaction of an α-particle with another nucleus may cause Coulomb excitation or may induce a nuclear reaction. Since the interaction involves transfer of a large amount of angular momentum, often high-lying high-spin nuclear levels are populated. Studies of α-particle induced interactions are thus of considerable interest from the viewpoints of nuclear astrophysics, nuclear reaction mechanisms and formation of radioactive products. In nuclear astrophysics, an understanding of the occurrence of alpha-clustering is of great importance. The formation of ^{8}Be from two α-particles provides an intermediate step in the synthesis of ^{12}C. Similarly, the synthesis of other light mass nuclei is interesting, e.g. of ^{26}Al via the ^{23}Na(α,n)-reaction and of ^{26}Mg via the ^{23}Na(α,p)-process. Furthermore, in the medium and heavy mass regions, there are some stable proton rich isotopes whose nucleosyntheses cannot be satisfactorily explained via neutron-induced reactions: it is postulated that α-particle induced reactions play an important role. As regards nuclear reaction mechanisms, experimental measurement of an excitation function and its comparison with the results of nuclear model calculations provides very useful information. Finally, the formation of radioactive products in α-particle induced reactions is of great practical value since many unique radionuclides are obtained.

1.2 Availability of α-particles

In contrast to isotropically emitted α-particles from radioactive nuclei, the accelerated α-particles move in one direction and are termed as a beam. Tremendous advances have been made in charged particle accelerator technology over the last 100 years. The types of particles accelerated, their energies, beam currents and the beam quality constitute a very broad spectrum of modern engineering. Alpha particle beams of well-defined shapes and high intensities are now commonly available for applications in diversified fields. The main use of low-energy accelerators is in elemental analysis. For radionuclide production high-intensity small and medium-sized cyclotrons are commercially available. Occasionally intermediate energy cyclotrons are used for nuclear reaction studies as well as for radionuclide production.

This review briefly mentions the various applications of alpha particles but deals in more detail with nuclear reaction studies and medical radionuclide production.

2 Applications in materials research

Materials research involves on one hand analysis and study of known materials and, on the other, development of new materials. Ion beams in general, and α-particle beams in particular, are useful in both cases.

2.1 Elemental analysis

Small accelerators of different designs and capabilities, named as Pelletron, Tandem, Tandetron, etc. are available in many parts of the world. They generally accelerate charged particles to energies between a few hundred keV and several MeV; in special cases α-particles of energies up to 25 MeV can be accelerated. Ion-beam analysis is done using the following techniques (for early detailed reviews cf. [1, 2]).

i) Ion back scattering (IBS)
ii) Elastic recoil detection analysis (ERDA)
iii) Particle induced X-ray emission (PIXE)
iv) Particle induced γ-ray emission (PIGE)
v) Charged particle activation analysis (CPAA)

The *ion back scattering* (IBS) at low energies mainly consists of Rutherford back scattering (RBS) which is scattering below the Coulomb barrier. The technique is based on the energy exchange between the elastically colliding particles, the slowing down of the ion in the target material, and the probability of the elastic scattering reaction. The energy and angle of the back-scattered radiation thus give important information on the charge and thickness of the target material. It is probably the most widely adopted single technique in multielement characterisation of materials with α-particle beams of energies between 2 and 4 MeV. The detection sensitivities are high for heavy mass elements, and quantities on the order of a few ng can be easily detected. The light elements, on the other hand, are difficult to detect.

In recent years, intermediate energy α-particles of energy up to about 40 MeV have also been increasingly used in ion back scattering work. However, the back-scattered

spectra then become very complex, and powerful computer programs are needed to unfold them.

The *elastic recoil detection analysis* (ERDA) involves quantitative detection of a recoiling nucleus, released from its environment, subsequent to the interaction of a charged particle projectile with a light nucleus. By determining the energy of the recoiling nucleus, its depth-profile is measured. This technique is particularly suitable for the assay of hydrogen and helium isotopes in surface layers. By a suitable choice of the incident projectile, the technique can be extended also to lithium and beryllium. If the host substrate is thin compared with the range of the projectile, a considerable improvement in detection sensitivity is achieved by the use of the kinematic coincidence technique (KCT), which involves detection of both the recoil light nucleus and the scattered projectile in coincidence.

Analysis by ERDA is generally done using low or intermediate energy projectiles. In the case of α-particles the energies used are either below 5 MeV or around 25 MeV. With 5 MeV α-particles, the limit of detection of concentration of hydrogen and deuterium within a depth of 1 μm of the substrate consisting of light elements is about 0.5% (5000 ppm). By using 25 MeV α-particles, the depth which can be examined is in the range of 100 to 300 μm, depending on the substrate. Thus the detection of hydrogen, helium (and lithium) in a matrix of heavy elements also becomes possible (at a concentration limit of about 0.1%). When KCT is used with 25 MeV α-particles on a thin substrate, the limit of detection of those three light elements, especially in heavy substrates, is considerably improved and lies in the range of 100 ng. Thus, ERDA and its extension KCT provide powerful, accurate and fast methods of determination of very light elements (hydrogen, helium, lithium) in various host matrices, especially polymer surfaces and interfaces. In fact this technique has contributed significantly to polymer science.

The *particle induced X-ray emission* (PIXE) technique involves quantitative determination of X-rays emitted in interactions of an ion-beam with materials. The ionisation or excitation of an inner atomic shell by the projectile leads to a vacancy which is filled by an electron from the outer shell, the process being accompanied by emission of X-rays. As the energy of the X-ray is a signature of the Z of the element, a quantitative determination of the element is possible via characterisation of the intensity of the X-ray. It is an analytical technique of high sensitivity, especially in the medium and heavy mass regions where detection limits lie in the ng range. The technique is, however, more suitable for analysis of trace elements on or near surface layers. In general, the PIXE makes use of protons of energies 1 to 5 MeV. Due to the much lower range of α-particles in matter than that of protons, PIXE-alpha spectrometers are occasionally used in analysis of very thin upper surfaces (cf. [3]).

The *particle-induced gamma ray emission* (PIGE) technique involves quantitative determination of prompt γ-rays emitted in a nuclear reaction or Coulomb excitation of the nucleus. This technique finds only limited specific application in the analysis of light mass elements for which RBS and PIXE are not very successful. Using α-particles of about 5 MeV, for example, the reactions $^{7}\mathrm{Li}(\alpha,\alpha'\gamma)^{7}\mathrm{Li}$ and $^{19}\mathrm{F}(\alpha,\alpha'\gamma)^{19}\mathrm{F}$ allow a rather easy determination of the two target elements lithium and fluorine by on-line determination of the γ-rays (478 keV and 197 keV, respectively).

Charged particle activation analysis (CPAA) involves quantitative determination of a radioactive product formed in the interaction of a nucleus with a charged particle as projectile. Due to the Coulomb barrier effect the technique is applied only in the region of light elements, especially for surface analysis using protons and ^{3}He-particles. The use of α-particles has also been demonstrated in the analysis of several metals like V, Cr, Mn, Fe, etc. (cf. [4]).

In summary, elemental analysis of materials is very successfully carried out using α-particle beams utilizing the RBS and ERDA techniques, the former for heavy elements and the latter for very light elements. A few light elements can also be analysed using PIGE in combination with α-particles. PIXE, on the other hand, commonly utilizes a proton beam, and activation analysis using neutrons has been applied for decades more successfully than charged particle activation.

2.2 Materials properties

In recent years, the ion beam, especially the α-particle beam or heavier mass beam at an intermediate energy up to about 40 MeV, has been finding increasing application in implantation studies related to development of new materials. In general, the main research direction here is the fabrication and characterisation of micro- and nano-structured substances, which are of high interest in materials, environmental and bio-medicinal sciences, micro- and nanotechnology, electronics, optics and laser technology. The doses administered to achieve the desired effects are deduced from the α-particle energy as well as its permissible intensity. This is a very fast developing field and its importance is expected to increase further in the future. Another aspect of α-particle implantation studies is related to the investigation of radiation damage in structural

materials of nuclear technology. As it is well known, radiation damage in nuclear reactors is caused by displacement of atoms from their lattice positions, mainly due to scattering effects of neutrons. In fast reactors, especially in future fusion reactors, the (n,xα) processes will also occur and will lead to the formation of considerable quantities of He gas, whose damage effects are relatively unknown. The implantation and simulation studies using α-particle beams allow investigation of the damage effects, e.g. brittleness, change in elasticity, formation of voids, etc.

A yet another special application of the α-particle beam is to produce a profile of radioactivity in a material whose wear and tear in an industrial process or its erosion and corrosion under various environmental conditions are to be studied. This so called *"thin layer activation technique"* generally makes use of activation with protons or deuterons, but occasionally α-particle activation is also employed (cf. [5–8]), for example, production of ^{58}Co and ^{95}Nb via the ^{56}Fe(α,d)^{58}Co and ^{92}Zr(α,p)^{95}Nb reactions, respectively, in the surface layers. Due to the shorter range of the α-particle in the matter as compared to the proton or the deuteron, the profile of the generated radioactivity in α-particle irradiation is shorter. The study of removal of the radioactivity from the generated profile under simulated conditions leads to an understanding of the phenomenon involved (wear, corrosion, etc.).

3 Nuclear reaction studies

Nuclear reaction studies using α-particle beams deal both with nuclear structure analysis and nuclear reaction cross sections. A brief discussion of the two aspects is given below. The emphasis is, however, on cross section data.

3.1 Nuclear structure data

Studies related to nuclear structure generally involve in-beam γ-ray spectroscopy, i.e. measurement of γ-ray spectra during the irradiation of a sample with α-particles. It may involve Coulomb excitation, if the α-particle energy cannot surmount the Coulomb threshold, or may lead to high-lying levels of an (α,x) reaction product, which subsequently deexcite via γ-ray transitions. Thus, some interesting nuclear structure results may be obtained, especially if direct reactions like (α,d) or (α,t) are involved, because of the possibility of population of high-spin levels which may be difficult to reach via an (n,γ) or a (d,p) process.

3.2 Nuclear reaction data

As regards nuclear reaction studies using α-particles, some on-line measurements on the spectra of emitted neutrons and charged particles have been performed with a view to investigating angular and energy distributions of the emitted particles. The data lead to very useful information on the mechanism of emission of those particles. The technique is, however, very demanding and cumbersome. Most of the work has therefore been done using the activation technique in which the residual radioactive product of a nuclear reaction is determined. This method leads to very little information on the route and mechanism of formation of the product. However, if the result could be compared with nuclear model calculations, developed extensively in recent years, it may be possible to discern the contributions of various nuclear processes involved. Such investigations are of direct relevance to accelerator production of radionuclides as well and involve total and isomeric cross sections. They are therefore discussed below in some detail.

3.2.1 Excitation function

Measurement of the total cross section of a reaction channel as a function of the α-particle energy is a standard approach. A typical case is discussed here. The irradiation of an ^{123}Sb target with α-particles of energies up to 50 MeV leads to ^{123}Sb(α,n)^{126}I, ^{123}Sb(α,2n)^{125}I and ^{123}Sb(α,3n)^{124}I reactions. Many groups reported data for those reactions [9–17] and a critical evaluation [18] provided a set of reliable results. The more consistent set of experimental data [12, 13, 19] are shown in Figure 1, together with the calculated results [19] from a modern nuclear model calculational code TALYS, which is primarily based on the statistical model but takes into account precompound and direct interactions as well as the nuclear structures of the nuclei involved. The agreement between the experiment and theory is excellent, both in terms of shapes and magnitudes of the three excitation functions. From these results it is inferred that multi-neutron emission in the interaction of a medium mass nucleus with α-particles can be described well by the existing theory. Interesting is a comparison of the three curves: the (α,n) cross section has a low value of 380 mb at the maximum of the excitation function, the whole curve is rather narrow, but the tail is long, suggesting a high contribution of the compound nucleus mechanism in the early part but more non-equilibrium contributions at higher energies. The curves for the (α,2n) and (α,3n) reactions, on the other

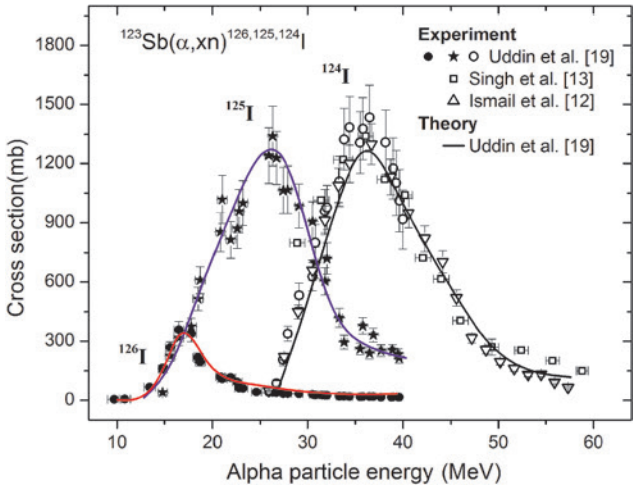

Figure 1: Excitation functions of ^{123}Sb(α,xn)^{126,125,124}I reactions. The experimental data are reproduced well by nuclear model calculations using the code TALYS (diagram adapted from Uddin et al. [19]).

hand, are much broader and have much higher maximum values of about 1400 mb. This suggests occurrence of more precompound interactions. These results show trends similar to those in (p,xn) and (n,xn) processes. The (p,n) excitation function is rather narrow and the (p,xn) curves are broad (cf. [20, 21]). The (n,n′) reaction, on the other hand, is an exception but the (n,xn) excitation functions are rather broad (cf. [22, 23]). From this comparison it may be inferred that (α,xn) reactions are similar to multineutron emission reactions induced by single particles (n or p) which proceed predominantly via compound and precompound processes. The same may not apply to excitation functions of reactions like (α,d), (α,t) etc., where strong contributions from direct interactions may be expected. In order to understand the mechanism of those reactions, spectral measurements dealing with angular and energy distributions are essential.

3.2.2 Isomeric cross section

Whereas the total cross section for the formation of a reaction product (i.e. integral cross section) can be described relatively well by the nuclear model calculation, there are considerable difficulties in the prediction of formation cross section of an isomer (i.e. partial cross section) because it depends on a large number of parameters, such as excitation energy, spin of the level concerned, type of projectile used, reaction channel, etc. (cf. [24–26]). Extensive studies have been performed over the years using neutrons (cf. [27–30]), protons (cf. [31–36]), deuterons (cf. [32–34]), ^{3}He (cf. [35–37]) and α-particles (cf. [32, 33, 35, 37, 38]) to elucidate some important aspects of isomer distribution. In general, it appears that low-spin levels are better formed in low-energy reactions induced by neutrons and protons whereas higher spin levels are preferentially populated in α-particle induced reactions. This is illustrated below through a consideration of three typical isomeric pairs, namely ^{58m,g}Co, ^{94m,g}Tc and ^{194m,g}Ir, which belong to three different mass regions of the Chart of Nuclides.

The radionuclide pair **^{58m,g}Co** consists of a metastable state ($T_{1/2}$ = 9.1 h) having a spin of 5^+ and the ground state ($T_{1/2}$ = 71 d) with a spin of 2^+. The separation energy between the two isomeric levels is only 25 keV and the metastable state decays completely to the ground state. The isomeric cross section ratio ($\frac{\sigma m}{\sigma m+\sigma g}$) was measured in three different charged-particle induced reactions, namely ^{58}Fe(p,n), ^{57}Fe(d,n) and ^{55}Mn(α,n) [32, 33], and the results are shown in Figure 2(a). Evidently, the ratio increases in all reactions with the increasing projectile energy. Conspicuous, however, is the higher formation probability of the high-spin isomer in the α-particle induced reaction.

The radionuclide pair **^{94m,g}Tc** consists of a metastable state ($T_{1/2}$ = 52 min) having a spin of 2^+ and the ground state ($T_{1/2}$ = 4.9 h) with a spin of 7^+. The separation energy between the two states is 75 keV. The two states decay independently to ^{94}Mo. The isomeric cross section ratio ($\frac{\sigma m}{\sigma g}$) was measured in three different reactions, namely ^{94}Mo(p,n), ^{93}Nb(^{3}He,2n) and ^{94}Mo(α,d+pn) [39–42], and the results are depicted in Figure 2(b). Two conspicuous trends are observed: (i) the ratio decreases in the (p,n) reaction with the increasing projectile energy, indicating thereby the increasing population of the higher spin isomer with the increasing projectile energy; (ii) the ratio is low in ^{3}He- and α-particle induced reactions which leads to the conclusion that the higher spin isomer is preferentially populated in the latter two reactions. Since ^{94m}Tc is a low-spin isomer, it is preferably produced via the (p,n) reaction (cf. [39, 40]).

The radionuclide pair **^{194m,g}Ir** consists of a metastable state ($T_{1/2}$ = 171 d) having a high spin of 10 or 11 and the ground state ($T_{1/2}$ = 19.3 h) with a low spin of 1^-. The separation energy between the two isomeric levels is < 440 keV. Both of them decay independently to ^{194}Pt, emitting β^- particles and several γ-rays. The isomeric cross section ratio was reported as ($\frac{\sigma m}{\sigma g}$) for two reactions, namely ^{192}Os(α,d+pn)^{194m,g}Ir and ^{194}Pt(n,p)^{194m,g}Ir [38, 43] and the results are shown in Figure 2(c). Whereas the ratio for the (n,p) reaction is very low, that for the α-particle induced reaction is relatively high.

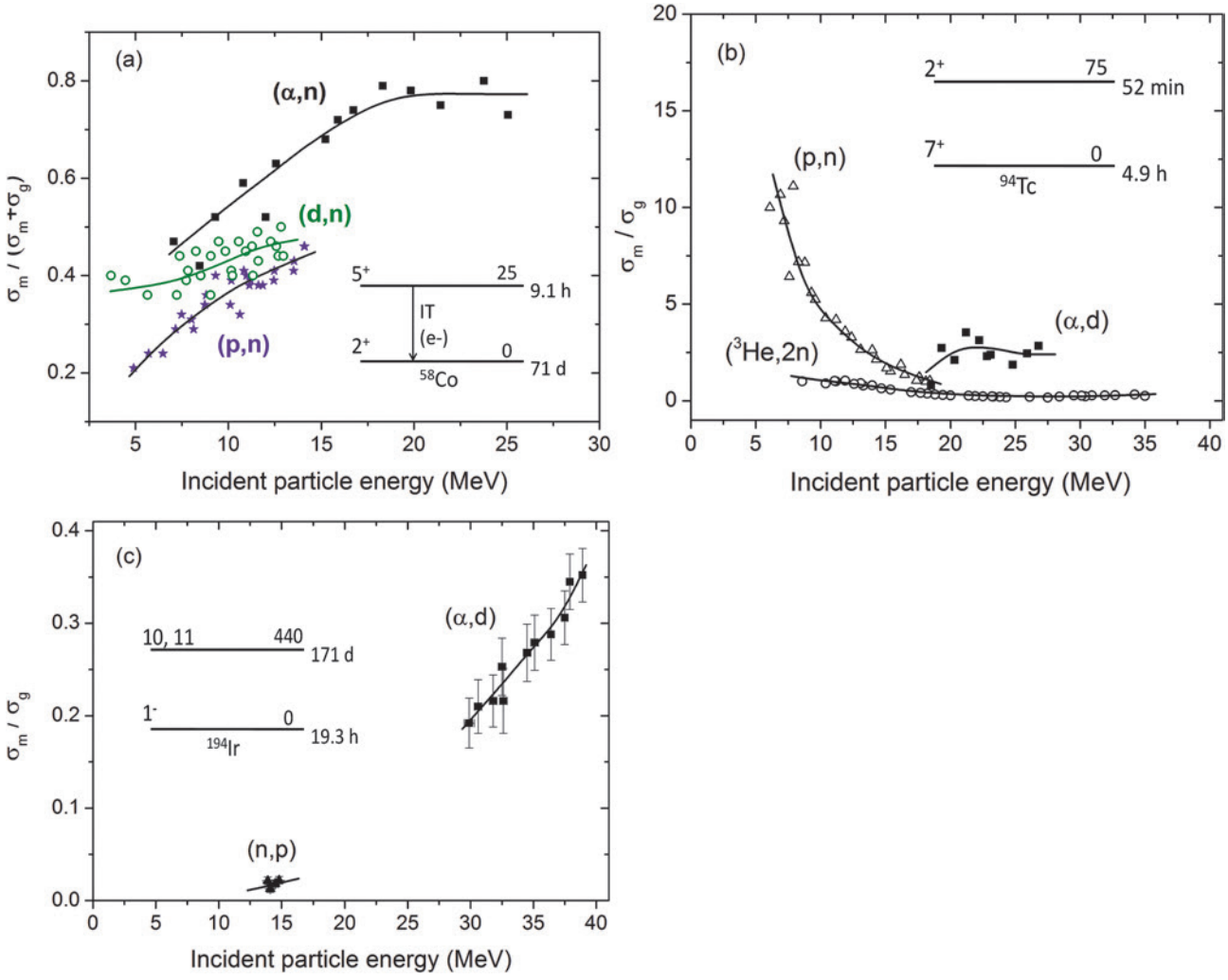

Figure 2: Isomeric cross section ratio as a function of projectile energy for three isomeric pairs: (a) ^{58m,g}Co, (b) ^{94m,g}Tc, (c) ^{194m,g}Ir. In each case experimental data reported in the literature [32, 33, 38–43] are given. The α-particle irradiation leads to a preferential population of the higher spin isomer.

From the above discussed three examples it is qualitatively concluded that, besides other factors, high spin isomers are preferentially populated in α-particle induced reactions, irrespective of the separation energy of the two states concerned. This observation is of considerable significance in the production of a few isomeric states (see below).

4 Medical radionuclide production

4.1 General

Radioactivity is unique in the sense that, in spite of its hazardous nature, it finds application in medicine both in diagnosis and therapy (cf. [44]). Each application, however, demands a special type of radionuclide. For *in vivo* diagnostic investigations involving organ imaging, for example, radionuclides are required that can be efficiently detected from outside of the body. To this end, short-lived γ-ray emitters, like ^{99m}Tc and ^{123}I, and positron emitters, like ^{11}C and ^{18}F, are commonly used, the former finding application in Single Photon Emission Computed Tomography (SPECT) and the latter in Positron Emission Tomography (PET). The underlying principle in diagnostic nuclear medicine is that the radiation dose to the patient is as low as possible, compatible with the required quality of imaging and the diagnostic advantage in comparison to non-radioactive methods. The internal radionuclide therapy, on the other hand, stipulates that a localised, well-defined radiation dose is deposited in a malignant or inflammatory tissue. Therefore, radionuclides emitting

low-range highly-ionising radiation, i.e. α- or β^- particles, conversion and/or Auger electrons, are of great interest.

Many radionuclides are produced using neutron-induced reactions in nuclear reactors. However, in recent years considerable progress has been achieved in production of radionuclides using charged particle induced reactions, and the relevant technology is developing rapidly (cf. [45–47]). In principle, all four light charged particles, namely p, d, ^{3}He and ^{4}He could be utilized. The emphasis is, however, on protons which are more commonly available and can be accelerated to deliver both H^+ and H^- beams. In particular, for production of the commonly used PET radionuclides (^{11}C and ^{18}F), commercially available single or two particle (p and d) small cyclotrons are now routinely used. Many of those cyclotrons are being utilized also for production of several non-standard positron emitters (cf. [46]). On the other hand, for production of some common SPECT-radionuclides, like ^{123}I and ^{201}Tl, or therapeutic radionuclides, like ^{103}Pd and ^{186}Re, a medium-sized cyclotron is required. Furthermore, for production of several other important radionuclides, such as ^{52}Fe, ^{67}Cu, ^{68}Ge (^{68}Ga), ^{82}Sr (^{82}Rb), etc., proton beams of energies up to 100 MeV are desired (cf. [45]). In contrast to protons, the charged particles d, ^{3}He and ^{4}He have found only limited application, though there are cases where the use of one of those particles may be advantageous as compared to the others. Yet such applications constitute special cases. The case of α-particles is interesting. It is discussed below in some detail.

The cross sections of some of the α-particle induced reactions, for example (α,xn) (see above), (α,p), (α,d), (α,t) etc. are fairly high, especially for the light and medium mass target nuclei. As an example, the excitation functions of the four investigated reactions for the production of ^{73}Se, namely ^{75}As(p,3n), ^{75}As(d,4n), ^{72}Ge(^{3}He,2n) and ^{70}Ge(α,n), (cf. [48–53]) are shown in Figure 3. The cross section for the (α,n) reaction is the highest. Thus, from the viewpoint of nuclear data, it is feasible to use the α-particle induced reaction for production purposes. The major disadvantage, however, comes to light while calculating the thick target yield for a certain energy range (E_1 to E_2) which makes use of the following equation (cf. [54]):

$$Y = \frac{N_L \cdot H}{M} I(1 - e^{-\lambda t}) \int_{E_1}^{E_2} \left(\frac{\mathrm{d}E}{\mathrm{d}(\rho\chi)} \right)^{-1} \sigma(E)\mathrm{d}E$$

where N_L is the Avogadro number, H the enrichment (or isotopic abundance) of the target nuclide, M the mass number of the target element, I the projectile current, $(\frac{\mathrm{d}E}{\mathrm{d}(\rho x)})$ the stopping power, $\sigma(E)$ the cross section at energy E, λ the decay constant of the product and t the time

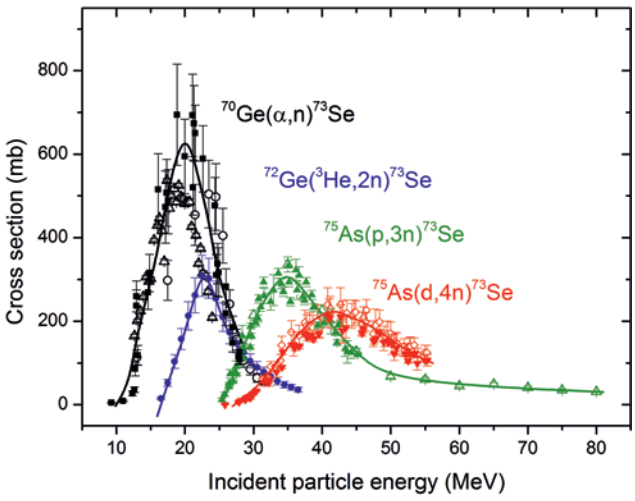

Figure 3: Excitation functions for the reactions ^{75}As(p,3n)^{73}Se, ^{75}As(d,4n)^{73}Se, ^{72}Ge(^{3}He,2n)^{73}Se and ^{70}Ge(α,n)^{73}Se drawn using the data in References [48–53].

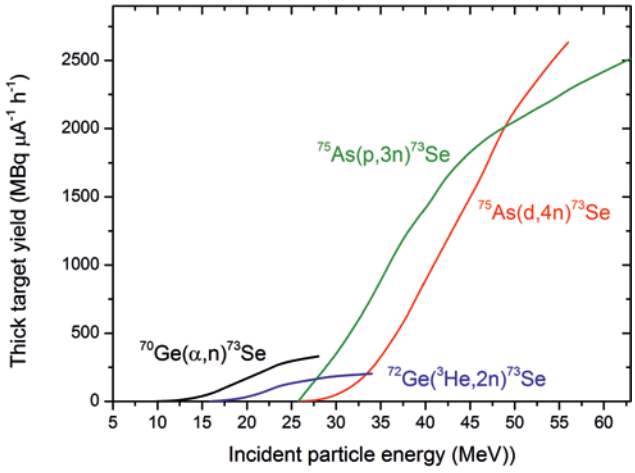

Figure 4: Thick target yields of ^{73}Se calculated from the excitation functions of ^{75}As(p,3n)^{73}As, ^{75}As(d,4n)^{73}Se, ^{72}Ge(^{3}He,2n)^{73}Se and ^{70}Ge(α,n)^{73}Se reactions shown in Figure 3.

of irradiation. The calculated yield (Y) value (in Bq) represents the maximum yield which can be expected from a given nuclear process.

Since the absorption of an α-particle in a target material is much stronger than that of a proton, deuteron or ^{3}He-particle, the total range of the α-particle is much shorter than that of any of the other three particles; correspondingly the number of target nuclei involved in the α-particle induced reaction is the lowest. Thus even if the cross sections of all reactions would be almost equal, the yield of the radioactive product in the α-particle induced reaction would be much lower than in other reactions.

The integral yields of ^{73}Se calculated for the above mentioned four reactions are shown in Figure 4. Evidently,

up to energies of about 45 MeV, the $^{73}As(p,3n)^{73}Se$ reaction leads to the highest yield: yet under certain circumstances (for example the non-availability of a 45 MeV proton beam) the $^{70}Ge(\alpha,n)^{73}Se$ reaction at 25 MeV could also be used for production purposes. This has been practically demonstrated [55] and ^{73}Se was obtained in quantities sufficient for medical applications (see below).

Besides low yields of products in α-particle induced reactions, there is often another disadvantage that the available α-particle beams are of lower intensity than in the case of the proton. On the other hand, there are three major advantages of the α-particle beam: (a) some radionuclides can be produced only via an α-particle induced reaction, (b) high-spin isomeric states are preferentially produced in α-particle induced reactions, (c) the product radionuclide is often two charge units higher than the target nuclide. This makes the chemical separation more specific and the product can be obtained with very high radiochemical and chemical purity. The latter aspect is of considerable significance in the production of metallic therapeutic radionuclides.

4.2 Radionuclides commonly produced using α-particles

The radionuclides commonly produced using α-particles are listed in Table 1, together with their major decay characteristics. In each case the nuclear reaction used, the optimum energy range chosen, the thick target yield calculated from the excitation function, the experimentally achieved level of purity and the major references for its production are given. The last column of Table 1 enlists some additional reactions which were also investigated for the production of the relevant radionuclide. Some of the reasons of non-applicability of those additional reactions for production purposes are discussed below. Some reactions where only cross sections were measured are not given in Table 1; they are mentioned only in the text.

[Correction added after online publication June 08, 2016: "$^{40}Ar(p,3p)^{38}K$" in Table 1 was changed to "$^{40}Ar(p,3n)^{38}K$".]

Table 1: Radionuclides commonly produced using the α-particle beam.

Radionuclide	$T_{1/2}$	Radiation emitted (%)	Nuclear reaction	Energy range (MeV)	Yield[a] (MBq/μAh)	Purity (%)	References to production	Other investigated reactions[e] [Reference]
^{28}Mg	21.1 h	β^- (100)	$^{27}Al(\alpha,3p)^{28}Mg$	140 → 30	1.5	> 99	[56, 57]	$^{26}Mg(t,p)^{28}Mg$ (cf. [56, 57])
^{30}P	2.5 min	β^+ (100)	$^{27}Al(\alpha,n)^{30}P$	24 → 10	ca. 1000[b]	> 99.9	[60, 61]	$^{32}S(n,t)^{30}P$ [61]
^{38}K	7.6 min	β^+ (100)	$^{35}Cl(\alpha,n)^{38}K$	22 → 7	ca. 400[b]	> 99.8	[62–67]	$^{38}Ar(p,n)^{38}K$ [68] $^{40}Ar(p,3n)^{38}K$ [69, 70]
^{43}K	22.2 h	β^- (100)	$^{40}Ar(\alpha,p)^{43}K$	21 → 10	7.0	97.5	[74–76]	$^{44}Ca(\gamma,p)^{43}K$ [77, 78] $^{43}Ca(n,p)^{43}K$ [79]
^{77}Br	57.0 h	EC (99.3) β^+ (0.7)	$^{75}As(\alpha,2n)^{77}Br$	28 → 16	16.6	> 99.9	[84–87]	$^{77}Se(p,n)^{77}Br$ [88, 89] $^{78}Se(p,2n)^{77}Br$ [89, 90] $^{79}Br(p,3n)^{77}Kr \rightarrow {}^{77}Br$ [91, 92] $^{79}Br(d,4n)^{77}Kr \rightarrow {}^{77}Br$ [93]
^{95}Ru	1.65 h	EC (85.0) β^+ (15.0)	$^{92}Mo(\alpha,n)^{95}Ru$	28 → 14	240[c]	> 99	[97, 98]	$^{nat}Mo(^3He,xn)^{95}Ru$ [97, 98]
^{97}Ru	2.9 d	EC (100)	$^{nat}Mo(\alpha,xn)^{97}Ru$	28 → 16	1.8[d]	> 99.8	[97–99]	$^{nat}Mo(^3He,xn)^{97}Ru$ [97, 98]
^{147}Gd	38.1 h	EC (99.7) β^+ (0.3)	$^{144}Sm(\alpha,n)^{147}Gd$	27 → 12	4.8	> 99.8	[100, 101]	$^{147}Sm(^3He,3n)^{147}Gd$ [100, 101]
^{211}At	7.3 h	EC (58) α (42)	$^{209}Bi(\alpha,2n)^{211}At$	28 → 10	17.5	> 99	[81, 104–110]	$^{232}Th,^{238}U(p,spall)^{211}At$ [112] $^{209}Bi(^{6,7}Li,xn)^{211}Rn \rightarrow {}^{211}At$ [113, 114]

[a] Calculated from excitation function.
[b] This is saturation yield.
[c] Value extrapolated to 100% enrichment of ^{92}Mo.
[d] At 15 h after EOB.
[e] For comments on these reactions, see text.

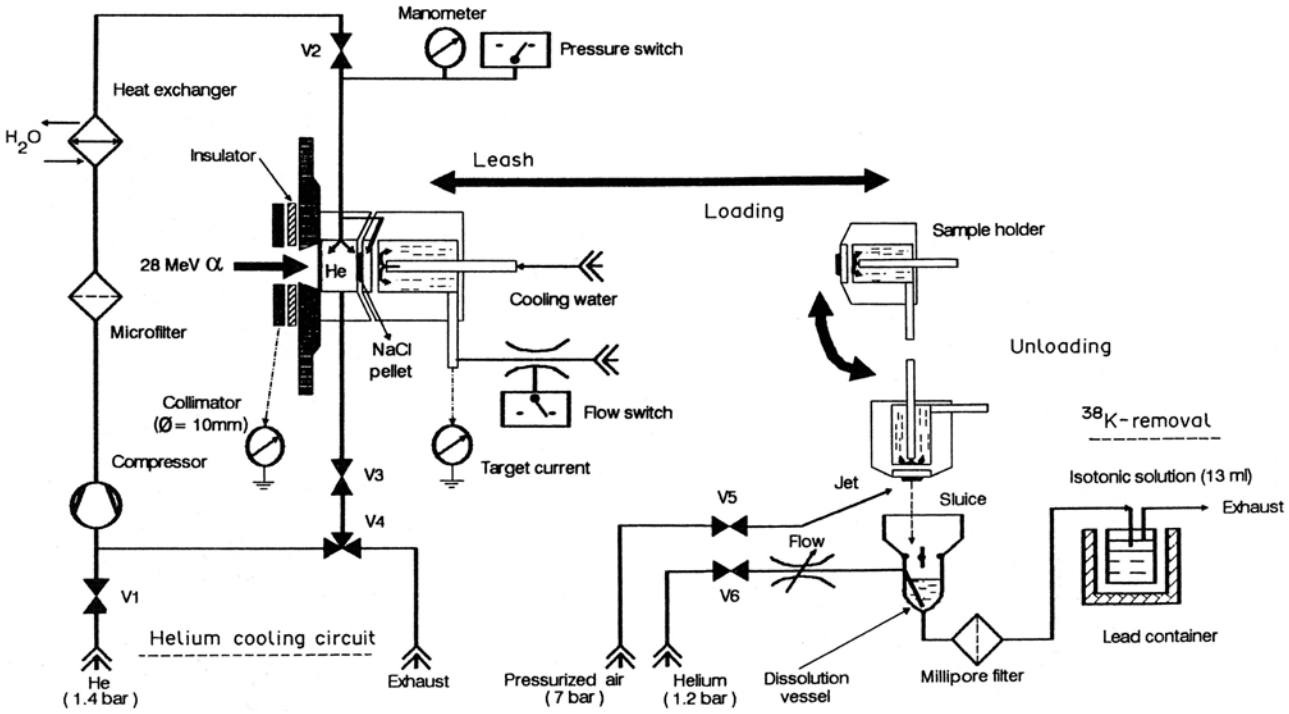

Figure 5: Schematic representation of a remotely controlled target and processing system for the production of ^{38}K via the ^{35}Cl(α,n)-reaction (taken from Blessing and Qaim [66]).

The radionuclide **^{28}Mg** ($T_{1/2}$ = 21.1 h) was produced for about two decades via the ^{27}Al(α,3p)^{28}Mg reaction using intermediate energy α-particles [56, 57]. The yield is rather low but in a 6 h irradiation with 140 MeV α-particles at a beam current of 20 μA, about 100 MBq of ^{28}Mg in no-carrier-added form were obtained for studies on plant physiology as well as γ-scintigraphy of children suffering from magnesium deficiency. The other reaction rather commonly employed was the ^{26}Mg(t,p)^{28}Mg whereby either a tritium beam was used or a sequential reaction in a nuclear reactor was employed (for review cf. [56, 57]). The latter involved irradiating a mixture of Li and Mg whereby tritons released in the interaction of neutrons with Li reacted with ^{26}Mg to yield ^{28}Mg. The product obtained after extensive chemical processing was not carrier-free. The ^{27}Al(α,3p)^{28}Mg reaction therefore became the method of choice for production of ^{28}Mg. Several other reactions with intermediate energy protons, e.g. ^{30}Si(p,3p)^{28}Mg, ^{31}P(p,4p)^{28}Mg, ^{32}S(p,5p)^{28}Mg, ^{nat}Cl(p,6pxn)^{28}Mg, ^{nat}Ar(p,7pxn)^{28}Mg and ^{nat}K(p,8pxn)^{28}Mg, were also investigated [58, 59] but the yields were found to be low [59].

The radionuclide **^{30}P** ($T_{1/2}$ = 2.5 min) was historically the first artificial transmutation product to be identified. It was produced via the nuclear reaction ^{27}Al(α,n)^{30}P which was induced by the low-energy (5.3 MeV) α-particle emitted from a ^{210}Po source. In later years the same nuclear reaction was used to produce large amounts of ^{30}P by employing higher energy α-particles from a cyclotron [60, 61]. This short-lived non-standard positron emitter has found some application in biological studies both in animals and humans using PET. The other investigated reaction, namely ^{32}S(n,t)^{30}P, does not lead to satisfactory result since the yield is low [60]. In three other cases, viz. ^{28}Si(α,d)^{30}P, ^{28}Si(^{3}He,p)^{30}P and ^{32}S(d,α)^{30}P reactions, only cross section measurements were performed [61]. The method of choice for production has therefore been the ^{27}Al(α,n)^{30}P process. Typical batch yields of 740 MBq of $^{30}PH_3$ and 300 MBq of $^{30}PO_4^{3-}$ were achieved [61].

The radionuclide **^{38}K** ($T_{1/2}$ = 7.6 min) is also a short-lived non-standard positron emitter and has been applied in myocardial blood flow studies using PET. It is produced using the ^{35}Cl(α,n)^{38}K reaction [62–67] and the technology is well developed to be able to obtain this radionuclide in a pure form in quantities of about 700 MBq (cf. [66]). A typical automated production system, taken from Reference [66], is shown in Figure 5. A NaCl pellet is irradiated with α-particles. Thereafter it is removed from the target holder and is simply dissolved in water. After filtering through a millipore filter, the solution is ready for application.

The other investigated processes for the production of ^{38}K include the ^{38}Ar(p,n)^{38}K reaction on highly enriched ^{38}Ar [68] and the ^{40}Ar(p,3n)^{38}K reaction with intermediate energy protons [69, 70]. The former reaction is very promising though the cost of the highly enriched target material is rather high and the handling of enriched ^{38}Ar is more difficult than that of an enriched isotope of Kr or Xe. In the second case, i.e. the ^{40}Ar(p,3n)-reaction, a separation of ^{38}K from the accompanying impurity ^{38}Cl ($T_{1/2}$ = 37.2 min) was found to be very cumbersome. A few other investigations report only low yields of ^{38}K (cf. [71–73]). The method of choice to date for producing sufficient quantities of ^{38}K in a radionuclidically pure form is therefore the α-particle induced reaction on ^{35}Cl. The level of the ^{34m}Cl impurity ($T_{1/2}$ = 32.0 min) in ^{38}K is reduced to < 0.2% by keeping the incident α-particle energy on the target below 22 MeV.

[Correction added after online publication June 08, 2016: On page 10, line 2, "^{40}Ar(p,3p)-reaction" was changed to "^{40}Ar(p,3n)-reaction".]

The radionuclide **^{43}K** ($T_{1/2}$ = 22.2 h) is a longer lived isotope of potassium. It is used in plant physiological studies and to some extent in cardiology, though due to the relatively high radiation dose caused by it, the emphasis has shifted over to the β^+ emitter ^{38}K discussed above as well as to the analogue (SPECT)-isotope ^{201}Tl. For the production of ^{43}K the most useful reaction is the ^{40}Ar(α,p)^{43}K process on a gas target (cf. [74–76]) and batch yields of about 300 MBq were achieved [76]. The level of the impurity ^{42}K ($T_{1/2}$ = 12.3 h) at EOB is about 2.5% which can, however, be almost completely eliminated if the incident α-particle energy is kept below 17 MeV [76]. Three other investigated reactions, namely ^{44}Ca(γ,p)^{43}K with 20 MeV bremsstrahlung (cf. [77, 78]), ^{43}Ca(n,p)^{43}K with reactor neutrons (cf. [79]) and ^{51}V(d,x)^{43}K [80] with intermediate energy deuterons need either highly expensive enriched targets or the yield is low.

The radionuclide **^{77}Br** ($T_{1/2}$ = 57.0 h) has been used in labelling of molecules for SPECT studies. It is also potentially useful for Auger electron therapy. For its production a large number of nuclear reactions have been suggested (for early reviews cf. [81, 82] and for a recent evaluation cf. [83]). Most successfully the ^{75}As(α,2n)^{77}Br reaction was utilized [84–87] and the product was achieved in GBq amount. The other investigated reactions include ^{77}Se(p,n)^{77}Br [88, 89], ^{78}Se(p,2n)^{77}Br [89, 90], ^{79}Br(p,3n)^{77}Kr → ^{77}Br [91, 92] and ^{79}Br(d,4n)^{77}Kr → ^{77}Br [93]. The amount of the product obtained in each case, however, was sufficient only for tracer development work. A few other reports dealing with cross section measurements of proton induced reactions on isotopes of selenium, leading to the formation of several bromine radionuclides, have also been published in recent years (cf. [94–96]). For application oriented production of ^{77}Br, however, the α-particle induced reaction has hitherto been most successfully applied.

The radionuclide **^{95}Ru** ($T_{1/2}$ = 1.65 h) is a non-standard positron emitter potentially useful for preparing ruthenocomplexes for diagnostic studies using PET. It has been produced using nuclear reactions induced by both ^{3}He- and α-particles on ^{nat}Mo [97, 98] but due to rare availability of the ^{3}He-particle beam and in view of the radionuclidic purity of the product, the α-particle induced reaction on enriched ^{92}Mo is to be preferred, whereby a batch yield of up to 1 GBq is expected.

The radionuclide **^{97}Ru** ($T_{1/2}$ = 2.9 d) is suitable for preparing ruthenocomplexes for diagnostic studies using SPECT. It has been produced via nuclear reactions induced by both ^{3}He- and α-particles [97–99]. However, in general the α-particle induced production is preferred because it is more economical. The radioruthenium chemically separated from the irradiated ^{nat}Mo target contains three radionuclides, namely ^{94}Ru ($T_{1/2}$ = 51.8 min) ^{95}Ru and ^{97}Ru. Nonetheless, if the product is allowed to decay for about 15 h, radionuclidically pure ^{97}Ru is obtained. Batch yields of up to 200 MBq have been achieved for this radionuclide (cf. [97, 98]).

The radionuclide **^{147}Gd** ($T_{1/2}$ = 38.1 h) is suitable for SPECT studies, especially in combination with Magnetic Resonance Imaging (MRI) because Gd is the most important contrast agent used in MRI. It has been produced with high radionuclidic and chemical purity via the ^{144}Sm(α,n)^{147}Gd reaction on 86.6% enriched $^{144}Sm_2O_3$ target; a batch yield of about 400 MBq was achieved [100, 101]. Another important production reaction investigated was ^{147}Sm(^{3}He,3n)^{147}Gd on 96.5% enriched $^{147}Sm_2O_3$ target, whereby the yield of ^{147}Gd was about 80% higher but the product contained 0.3% ^{149}Gd ($T_{1/2}$ = 9.3 d) as impurity [101]. The enriched ^{144}Sm is more expensive than the enriched ^{147}Sm. Nonetheless, due to the ^{149}Gd impurity and due to the rare availability of the ^{3}He beam, the α-particle induced reaction on ^{144}Sm appears to be more interesting. Some additional reactions like ^{nat}Eu(p,xn)^{146,147,148}Gd at 100 MeV [102] and ^{nat}Eu(d,xn)^{147,149,151,153}Gd at 60 MeV [103] have also been investigated. However, no medium to large scale production of ^{147}Gd has hitherto been reported.

The radionuclide **^{211}At** ($T_{1/2}$ = 7.3 h) is an α-emitting heavy halogen nuclide and thus it is of great potential interest in α-targeted internal radiotherapy. It has been

Table 2: Radioactive high-spin isomeric states advantageously produced using the α-particle beam.

Isomeric state	Spin and parity	$T_{1/2}$	Mode of decay (%)	Preferential production method		Thick target yield [c] (MBq/μAh)	Reference	Other reactions used
				Nuclear reaction	Energy range (MeV)			
^{117m}Sn	$11/2^-$	13.6 d	IT (100) [a]	^{116}Cd(α,3n)^{117m}Sn	60 → 30	8.4	[119–123]	^{117}Sn(n,n′γ)^{117m}Sn [116] ^{121}Sb(p,2p3n)^{117m}Sn [117, 118] ^{nat}In(α,pxn)^{117m}Sn [119] ^{nat}Cd(α,xn)^{117m}Sn [119, 120]
^{193m}Pt	$13/2^+$	4.33 d	IT (100) [b]	^{192}Os(α,3n)^{193m}Pt	40 → 30	10	[124–126]	^{192}Pt(n,γ)^{193m}Pt [d] (cf. [127, 128])
^{195m}Pt	$13/2^+$	4.02 d	IT (100) [b]	^{192}Os(α,n)^{195m}Pt	24 → 18	0.25	[124–126]	^{194}Pt(n,γ)^{195m}Pt [d] (cf. [127]) ^{195}Pt(n,n′γ)^{195m}Pt [116]

[a] Highly converted; energy of conversion electrons ~150 keV.
[b] Highly converted; > 30 Auger electrons emitted per decay.
[c] Calculated from the excitation function for a highly enriched target.
[d] Using enriched target material.

produced for the last 40 years in high radionuclidic purity via the ^{209}Bi(α,2n)^{211}At reaction in GBq quantities (for early works cf. [104–106], and for early review cf. [81]). In recent years more optimisation work has been done (see for example [107, 108]) and the separation methods have been reviewed (cf. [109, 110]). Some new cross section measurements have also been done and the data have been evaluated (for a review cf. [111]), but no substantial change has occurred. The spallation reaction (cf. [112]) gives much lower yield of ^{211}At and the level of impurity is high. Also the reactions ^{209}Bi (^{6,7}Li,xn)^{211}Rn → ^{211}At (cf. [113, 114]) have been investigated. Worth emphasizing is, however, that no reaction other than the (α,2n) process mentioned above has so far delivered ^{211}At with the required quality and in quantities sufficient for medical applications.

4.3 Advantageous production of radioactive isomeric states using α-particles

Radioactive isomeric states of a few nuclides have very suitable decay properties for therapeutic applications. In general, they are low-lying states having high nuclear spins. They decay mostly to their respective ground states by a highly converted internal transition. The low-energy conversion electrons, or an avalanche of emitted Auger electrons, can lead to precise localised internal therapy effect, if the radioactive species is properly attached to an appropriate chemical carrier. Several such isomeric states have been identified. Some of them are advantageously produced using an α-particle beam because they have rather high spins (see Section 3.2.2). They are listed in Table 2. The decay characteristics of each state, its major production method, the suitable energy range and the thick target yield calculated from the excitation function are given. In the last column other investigated reactions are enumerated. Some more production details for each state are given below.

The radioactive **^{117m}Sn** ($T_{1/2}$ = 13.6 d) having a spin of $11/2^-$ lies 315 keV above the ground state ^{117g}Sn which is stable. The emitted conversion electrons of energy about 150 keV are ideally suited for some internal therapeutic studies, e.g. in the form of ^{117m}Sn-labelled porphyrin conjugates (cf. [115]). The radionuclide has been under consideration for more than three decades and its small quantities were hitherto produced via four routes:

(1) ^{117}Sn(n,n′γ)^{117m}Sn, (2) ^{121}Sb(p,αn+2p3n)^{117m}Sn, (3) ^{nat}Cd(α,xn)^{117m}Sn, (4) ^{nat}In(α,pxn)^{117m}Sn (cf. [116–119]). The (α,3n) reaction on a highly enriched ^{116}Cd target (cf. [119–122]) was found to be more interesting and an efficient chemical separation method for ^{117m}Sn was worked out [119]. Very recently, in an effort by Clear Vascular Inc. it has been shown that only this reaction can lead to a product of specific activity and chemical purity high enough to meet the requirement for medical application [123]. So this reaction has now become the method of choice for the production of ^{117m}Sn.

The two radionuclides **^{193m}Pt** ($T_{1/2}$ = 4.33 d) and **^{195m}Pt** ($T_{1/2}$ = 4.02 d) are pure Auger electron emitters with their energies distributed between 10 and 130 keV. Since platinum-complexes (like cis-di-chlorodiaminplatinum)

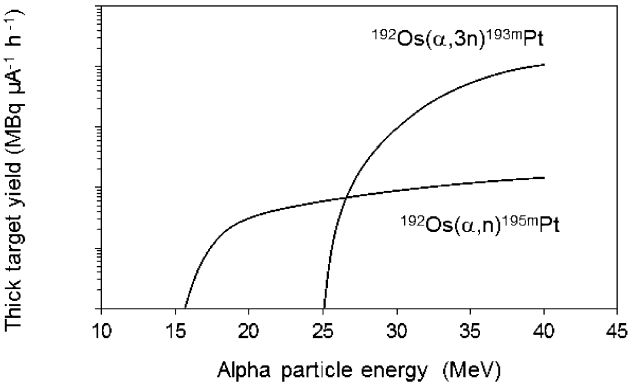

Figure 6: Thick target yields of the radionuclides ^{193m}Pt and ^{195m}Pt, calculated from the measured excitation functions, shown as a function of the α-particle energy (taken from Uddin et al. [125]).

have been in use in chemotherapy as potent anti-tumour agents for a long time, both ^{193m}Pt and ^{195m}Pt have a great potential in Auger electron therapy. So far the major drawback in their widespread use has been their non-availability with a high specific activity. Due to the very high spin ($13/2^+$) of both ^{193m}Pt and ^{195m}Pt, it was considered more fruitful to investigate the α-particle induced reactions on highly enriched ^{192}Os. In recent studies [124–126] the excitation functions of the reactions ^{192}Os(α,n)^{195m}Pt and ^{192}Os(α,3n)^{193m}Pt were measured [124, 125] and thick target yields were calculated which are shown in Figure 6. In small scale production runs it could also be demonstrated that both the radionuclides can be produced with high specific activity; the achieved yield of ^{195m}Pt is rather low but that of ^{193m}Pt over the energy range $E_\alpha = 40 \rightarrow 30$ MeV amounts to about 10 MBq/μA · h [126]. Thus this radionuclide can be produced in quantities sufficient for therapeutic applications.

The other reactions used for the production of the two radionuclides are ^{192}Pt(n,γ)^{193m}Pt (cf. [127]), ^{194}Pt(n,γ)^{195m}Pt (cf. [127, 128]), and ^{195}Pt(n,n′γ)^{195m}Pt [116] but even while using highly enriched isotopes as target materials, the specific activity achieved is not high enough for therapeutic application. The idea to produce ^{195}Ir via double neutron capture by ^{193}Ir and its subsequent decay to ^{195m}Pt (i.e. via the route ^{193}Ir(n,γ)^{194}Ir(n,γ)^{195m,g}Ir $\xrightarrow{\beta^-}$ ^{195m}Pt) is interesting but has not been really attempted. Thus for the production of these two radionuclides, the α-particle induced reactions are most promising.

In addition to the above mentioned three longer lived high-spin isomeric states whose advantageous production using α-particles has been practically demonstrated, there are two other shorter lived isomeric states whose formation through α-particle induced reactions appears to be preferable. The first one is **^{44m}Sc** ($T_{1/2}$ = 2.4 d; spin 6^+) which decays 98.6% by IT to the ground state ^{44g}Sc ($T_{1/2}$ = 3.9 h; spin 2^+) and 1.4% by EC to stable ^{44}Ca. Its formation in the ^{44}Ca(p,n)^{44m}Sc and ^{44}Ca(d,2n)^{44m}Sc reactions has been investigated [129–131] and the latter reaction is more suitable. Cross section measurements on the reaction ^{41}K(α,n)^{44m,g}Sc show a still higher ratio for the formation of ^{44m}Sc [132–136]. Thus this reaction could be more suitable for the production of ^{44m}Sc. Its practical application, however, has not been demonstrated. The isomeric state ^{44m}Sc itself is not very useful for diagnostic or therapeutic studies, but it could serve as an in-vivo generator of ^{44g}Sc (cf. [129]) which is a useful positron emitter, especially in combination with its theranostic counterpart ^{47}Sc (cf. [137]). An alternative way of its production could be via the ^{44}Ti/^{44}Sc generator system (cf. [138, 139]).

The second interesting shorter lived isomeric state is **^{197m}Hg** ($T_{1/2}$ = 23.8 h; spin $13/2^+$) which decays 93.5% by IT to the ground state ^{197g}Hg ($T_{1/2}$ = 64.1 h; spin ½$^-$) and 6.5% by EC to the stable ^{197}Au. The ratio ($\frac{\sigma_m}{\sigma_g}$) was measured in the ^{197}Au(p,n)^{197m,g}Hg and ^{197}Au(d,2n)^{197m,g}Hg processes [140–142] and found to be 0.88 at 19 MeV for protons and 1.35 at 21 MeV for deuterons. The practical use of both the reactions for production has been demonstrated [143, 144]. Very recently the advantageous production of ^{197m}Hg via the proton-induced reaction has been reported and the medical application has also been discussed [145]. On the other hand, the ($\frac{\sigma_m}{\sigma_g}$) ratio increased to about 2.2 in the ^{nat}Pt(α,xn)^{197m,g}Hg process [142] at 27 MeV. We therefore predict that the reaction ^{195}Pt(α,2n)^{197m,g}Hg on highly enriched target material would lead to relatively high yields of the isomeric state.

4.4 Limited scale production of radionuclides using α-particles

Table 3 lists some medically related radionuclides which are either commonly used or are of potential use. They are generally obtained via one of the three production routes: (a) intermediate energy proton (> 30 MeV) induced reaction, (b) low-energy (p,n) reaction on highly enriched target material, (c) (n,γ) process. In all those cases limited scale production could also be achieved through α-particle induced reactions, as discussed below.

The radionuclides **^{52}Fe**, **^{57}Ni**, **^{68}Ge**(**^{68}Ga**), **^{72}Se**(**^{72}As**), **^{72}As**, **^{73}Se**, **^{77}Kr**, **^{82}Sr**(**^{82}Rb**) and **^{83}Sr** are positron emitters

Table 3: Radionuclides produced in limited amounts using the α-particle beam in comparison to their large scale production via other nuclear processes.

Radio-nuclide	$T_{1/2}$	Mode of decay (%)	Production method using α-particles				Major production routes			
			Nuclear reaction	Energy range (MeV)	Thick target yield[a] (MBq/μAh)	Reference	Nuclear reaction	Energy range (MeV)	Thick target yield[a] (MBq/μAh)	Reference
^{18}F	110 min	EC (3) β^+ (97)	^{16}O(α,d)^{18}F	55 → 25	260	[188–192]	^{18}O(p,n)^{18}F[b]	16 → 3	3900	[169, 170]
^{34m}Cl	32.2 min	EC (46) β^+ (54)	^{32}S(α,d)^{34m}Cl ^{31}P(α,n)^{34m}Cl	50 → 20 40 → 10	1608 450	[196, 197] [193]	^{34}S(p,n)^{34m}Cl[b] ^{36}Ar(d,α)^{34m}Cl[b]	20 → 8 8 → 3	1049 252	[194, 195] [198–200]
^{52}Fe	8.3 h	EC (43.5) β^+ (56.5)	^{nat}Cr(α,2n)^{52}Fe	30 → 20	0.11	[164–166]	^{55}Mn(p,4n)^{52}Fe[c]	100 → 60	22	[146, 147]
^{57}Ni	1.5 d	EC (60) β^+ (40)	^{54}Fe(α,n)^{57}Ni[b]				^{59}Co(p,3n)^{57}Ni[c]	40 → 24	15	[158–160]
^{61}Cu	3.4 h	EC (38) β^+ (62)	^{59}Co(α,2n)^{61}Cu ^{58}Ni(α,p)^{61}Cu	33 → 23 18 → 10	185 90	[201–204] [205, 206]	^{61}Ni(p,n)^{61}Cu[b] ^{64}Zn(p,α)^{61}Cu[b]	12 → 9 18 → 11	647 288	[171, 173] [186, 187]
^{66}Ga	9.4 h	EC (43.5) β^+ (56.5)	^{nat}Cu(α,xn)^{66}Ga	20 → 10	33	[168]	^{66}Ge(p,n)^{66}Ga[b]	13 → 8	433	[172]
^{68}Ga	1.13 h	EC (11) β^+ (89)	^{65}Cu(α,n)^{68}Ga[b]	20 → 10	450	[168]	^{nat}Ga(p,xn)^{68}Ge(^{68}Ga)[c]	60 → 15	2.8[d]	[148–153]
^{72}As	26.0 h	EC (12) β^+ (88)	^{69}Ga(α,n)^{72}As[b] ^{69}Ga(α,n)^{72}As[b]				^{75}As(p,4n)^{72}Se(^{72}As)[c] ^{nat}Ge(p,xn)^{72}As	45 → 35 18 → 8	8[d] 93	[49, 50] [154, 155]
^{72}Se	8.5 d	EC (100)	^{70}Ge(α,2n)^{72}Se[b]	37 → 20	115	[49, 207]	^{75}As(p,4n)^{72}Se[c]	45 → 35	8	[49, 50]
^{73}Se	7.1 h	EC (34) β^+ (66)	^{70}Ge(α,n)^{73}Se[b]	28 → 13	126	[49, 51, 55]	^{75}As(p,3n)^{73}Se[c]	40 → 30	1.4×10^3	[48–50]
^{76}Br	16.0 h	EC (42) β^+ (58)	^{75}As(α,3n)^{76}Br				^{76}Se(p,n)^{76}Br[b]	15 → 8	360	[94, 174–176]
^{77}Kr	1.24 h	EC (13) β^+ (87)	^{74}Se(α,n)^{77}Kr[b]				^{79}Br(p,3n)^{77}Kr[c]	40 → 30	1500	[91, 92]
^{81}Rb	4.58 h	EC (69) β^+ (31)	^{79}Br(α,2n)^{81}Rb	27 → 13	40	[208]	^{82}Kr(p,2n)^{81}Rb[b]	27 → 19	1776	[149, 185]

Table 3: Continued.

Radio-nuclide	$T_{1/2}$	Mode of decay (%)	Production method using α-particles				Major production routes			
			Nuclear reaction	Energy range (MeV)	Thick target yield[a] (MBq/μAh)	Reference	Nuclear reaction	Energy range (MeV)	Thick target yield[a] (MBq/μAh)	Reference
^{82m}Rb	6.3 h	EC (77) β^+ (23)	^{79}Br(α,n)^{82m}Rb				^{82}Kr(p,n)^{82m}Rb[b]	14.5 → 10	370	[177]
^{82}Sr	25.5 d	EC (100)	^{nat}Kr(α,xn)^{82}Sr	120 → 20	1.9	[224, 225]	^{nat}Rb(p,xn)^{82}Sr[c]	70 → 30	14[d]	[149, 156]
^{83}Sr	32.2 h	EC (76) β^+ (24)	^{80}Kr(α,n)^{83}Sr[b]				^{85}Rb(p,3n)^{83}Sr[b,c]	37 → 30	160	[157]
^{124}I	4.15 d	EC (78) β^+ (22)	^{121}Sb(α,n)^{124}I[b] ^{nat}Sb(α,xn)	22 → 13 45 → 32	2.1 6.7	[210, 211] [210, 211]	^{124}Te(p,n)^{124}I[b]	12 → 8	16	[178, 184]
^{153}Sm	1.93 d	β^- (100)	^{150}Nd(α,n)^{153}Sm[b]	25 → 13	1.1	[220]	^{152}Sm(n,γ)^{153}Sm[b]	σ_{th}: 206 b	GBq$_{amounts}$[e]	[219]
^{166}Yb	2.3 d	EC (100)	^{164}Er(α,2n)^{166}Yb[b]				^{169}Tm(p,4n)^{166}Yb[c]	45 → 30	~ 100	[161]
^{177}Lu	6.7 d	β^- (100)	^{174}Yb(α,p)^{177m,g}Lu[b]				^{176}Lu(n,γ)^{177m,g}Lu[b] ^{176}Yb(n,γ)^{177}Yb(^{177m,g}Lu)	σ_{th}: 2100 b σ_{th}: 3.1 b	GBq$_{amounts}$[e] GBq$_{amounts}$[e]	[219] [219, 221]
^{178}W	22.0 d	EC (100)	^{176}Hf(α,2n)^{178}W[b,f]				^{181}Ta(p,4n)^{178}W[e]	80 → 30	~ 50	[162, 163]
^{186}Re	3.8 d	EC (7.8) β^- (92.2)	^{184}W(α,d)^{186}Re[b]				^{186}W(p,n)^{186}Re[b]	18 → 5	4.8	[212–217]

[a] Calculated from excitation function.
[b] Using highly enriched target material.
[c] Using intermediate energy protons.
[d] Yield of parent.
[e] Using highly enriched target for irradiation in a medium-flux reactor.
[f] For use as parent of 9.3 min ^{178}Ta.

and useful for PET studies, **^{166}Yb** is a β^- emitter and so of potential therapeutic use, and **^{178}W** has found application in **^{178}W(^{178}Ta)** generator system for SPECT. They are all produced using intermediate energy protons (cf. [48–50, 146–163]). The suitable energy ranges for those reactions and the thick target yields calculated from the respective excitation functions are given in Table 3. However, due to non-availability of intermediate energy protons, in a few laboratories the practical production of ^{52}Fe, ^{68}Ga, ^{72}Se and ^{73}Se using α-particles has been demonstrated (cf. [46, 49, 55, 164–166, 207]). The yields are, however, much lower (cf. Table 3) than via the major production route, but are sufficient for local tracer applications. In order to substantiate the possibility of developing an alternate production route, we considered the reaction ^{70}Ge(α,n)^{73}Se to generate ^{73}Se. A thin layer of electrodeposited $Cu_3^{70}Ge$ alloy on a wedged Cu-backing was irradiated in the internal target system of the compact cyclotron CV28 at an angle of 6.2° with 28 MeV α-particles at a beam current of about 100 μA [55]. After the end of the irradiation the radioselenium was separated by thermochromatography [55]. The profile of ^{73}Se activity deposited in a quartz tube outside the oven is shown in Figure 7. A clean separation of ^{73}Se could be realized and its batch yield amounted to 2 GBq. This quantity of ^{73}Se should be sufficient for many applications. For ^{57}Ni, ^{72}As, ^{77}Kr, ^{83}Sr, ^{166}Yb and ^{178}W, however, the application of the α-particle route for production has so far not been reported, mainly due to the high cost of the required enriched target. The cross sections on natural targets have, however, been measured in relation to the production of ^{57}Ni and ^{178}W [167, 234].

The radionuclides **^{18}F**, **^{34m}Cl**, **^{61}Cu**, **^{66}Ga**, **^{76}Br**, **^{81}Rb**, **^{82m}Rb** and **^{124}I** are also positron emitters and **^{186}Re** is a β^- emitter of interest in therapy. Most of them are produced via the (p,n) or (p,2n) reaction on the respective highly enriched target isotope (cf. [46, 94, 169–185]). For ^{61}Cu production, the low-energy ^{64}Zn(p,α)^{61}Cu reaction has also been utilized [186, 187]. The practical feasibility of the α-particle induced reaction for production has been shown in cases of ^{18}F, ^{34m}Cl, ^{61}Cu, ^{66}Ga, ^{81}Rb and ^{124}I. The α-particle induced reactions to produce those radionuclides make use of either target materials of natural isotopic composition or if enriched material, then its cost is much lower (cf. [16–19, 188–211]). However, as expected, the yield of the product in α-particle induced reaction is lower than in the commonly used reaction. For ^{76}Br only the ^{75}As(α,3n)^{76}Br reaction cross sections have been measured [174, 209]. Similarly for ^{82m}Rb only the ^{79}Br(α,n)^{82m}Rb reaction cross sections have been reported (cf. [237–239]) which are, however, very discrepant. Regarding ^{186}Re, the practical method of production is the

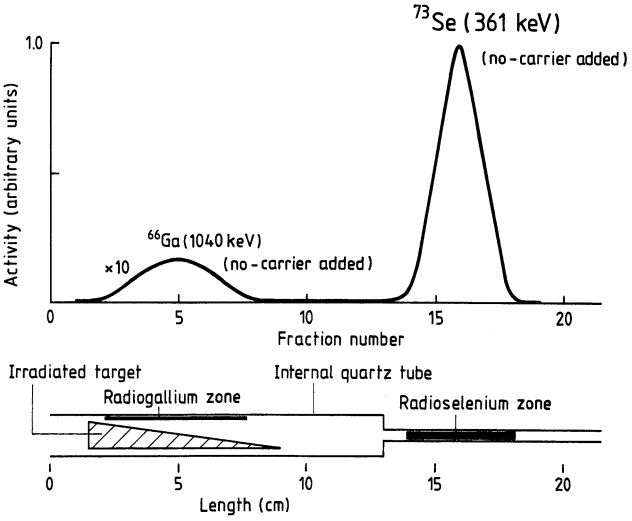

Figure 7: (Lower part) Schematic representation of the thermochromatographic system used for the separation of no-carrier-added ^{73}Se from a wedged $Cu_3^{70}Ge$ target irradiated with 28 MeV α-particles. Radiogallium released from the target was deposited on the inner wall of the quartz tube immediately above the target while radioselenium was carried away to the zone outside the quartz tube where it was collected in a small amount of warm H_2O_2.
(Upper part) Radioactivity profile after separation. The activity of the side-product ^{66}Ga has been increased by a factor of 10 (taken from Blessing et al. [55]).

^{186}W(p,n)-process (cf. [212–217]), and the α-production route has so far not been investigated.

As regards the (n,γ) process, many of the reactor radionuclides are produced via this route. The resulting specific activity, however, is often low. One such case is ^{153}Sm. It is extensively used in radiotherapy [218] but no suitable alternative to (n,γ) reaction has been found (cf. [219]). In our laboratory the excitation function of the ^{140}Nd(α,n)^{153}Sm reaction was investigated [220] and the result is promising. It could be used for production of no-carrier added ^{153}Sm; however, the cost will be rather high. A second case is ^{177}Lu. It is broadly used for tumour treatment: in the form of bisphosphonates for bone metastases, and with peptides and monoclonal antibodies for soft tissue tumours, e.g. in neuroendocrine and prostate diseases. It should, however, be pointed out that besides the 6.7 d ^{177}Lu, there exists a longer lived isomeric state ^{177m}Lu ($T_{1/2}$ = 160 d) which is undesirable. The direct production via the ^{176}Lu(n,γ)-reaction leads to a mixture of the two isomers and since the abundance of the target isotope ^{176}Lu in ^{nat}Lu is only 2.59%, despite its high capture cross section, the specific activity of the product ^{177}Lu achieved is not high. Generally >70% enriched target is used [219]. An alternative indirect production route, namely ^{176}Yb(n,γ)^{177}Yb → ^{177}Lu,

not only gives ^{177}Lu in the n.c.a. form but also the content of ^{177m}Lu is lower [221]. An α-particle induced reaction, namely ^{174}Yb(α,p)^{177m,g}Lu, could also prove to be an interesting alternative production route, but so far it has not been investigated. The formation cross section of ^{177g}Lu using ^{nat}Yb, however, has been measured [228].

In addition to the radionuclides listed in Table 3 the possibility of production of several other radionuclides using α-particle induced reactions has also been investigated, e.g. ^{86}Y, ^{87}Y, ^{88}Y and ^{89}Zr in (α,xn) reactions on ^{nat}Rb and ^{nat}Sr [222, 223] or ^{82}Sr via (α,xn) reactions on ^{nat}Kr and ^{82}Kr [224, 225]. Besides cross section measurements, chemical separations and radionuclidic quality control checks were carried out. The yields of the products, however, were found to be very low. Furthermore, excitation functions of α-particle induced reactions on a large number of target elements have been measured in recent years in several laboratories (cf. [226–236]). Some of those studies are of confirmatory character, dealing with the radionuclides ^{52}Fe, ^{97}Ru, ^{117m}Sn, etc. but a few describe newer routes for ^{99m}Tc, ^{99}Mo, ^{111}In, ^{131}Cs, ^{177}Lu and ^{178}W. Some of the reported data on targets of natural isotopic composition may be potentially useful for developing production routes of a few radionuclides. However, further studies related to practical production using highly-enriched targets are needed.

5 Concluding remarks

The above discussion shows that the α-particle beam has great advantages. It allows on one hand elemental analysis of various materials and, on the other, helps improve their physical properties. The activation of materials in irradiations with α-particles, i.e. the formation of radioactive products, is an interesting topic of study both from fundamental and applied points of views. A comparison of experimental data with results of nuclear model calculations sheds some light on the reaction mechanism. In particular the formation of nuclear isomeric states, i.e. the partial formation cross section, is difficult to reproduce by theory. This topic thus still poses a challenge to both experimentalists and theorists. The cross section data obtained as a function of α-particle energy, on the other hand, are of great practical significance in the production of some medically related radionuclides, though their yields are generally much lower than in proton or deuteron induced reactions. Nonetheless, there are several radionuclides which are exclusively produced through the use of α-particles, and there are a few low-lying high-spin isomeric states which are preferentially populated in α-particle induced reactions. In many other cases, however, production using an α-particle beam is only done when intermediate energy protons are not available or when a commonly used method involving a highly enriched target isotope cannot be employed due to its high cost.

In our institute a new cyclotron (IBA Cyclone 30 XP) has been recently installed and will soon go in operation. Besides protons and deuterons, an α-particle beam of maximum energy 30 MeV will also be available. In addition to routine production of a few standard radionuclides, it is intended to perform fundamental nuclear reaction studies as well as technologically oriented research in support of our radionuclide development program. The α-particle beam is expected to broaden the scope of nuclear chemistry research related to development of both non-standard positron emitters and radionuclides of interest in internal radiotherapy.

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