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Nuclear data for medical radionuclides

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Abstract Radionuclides are used in medicine both for diagnosis and therapy. The radioactive decay data play a key role in the choice of a radionuclide for a certain application. The nuclear reaction data, on the other hand, allow to optimise the production route of a chosen radionuclide. The status of nuclear data of the commonly used diagnostic and therapeutic radionuclides is reviewed and the recent efforts to standardise those data are described. The expected specific activity of the cyclotron produced ^{99m}Tc is briefly discussed. The present efforts are devoted to development of non-standard positron emitters (e.g. ^{64}Cu, ^{86}Y, etc.) and low-range highly-ionising therapeutic radionuclides (e.g. ^{67}Cu, ^{225}Ac, etc.). The need of intermediate-energy multiple-particle accelerating cyclotrons is pointed out.

Keywords Medical radionuclide · Decay and production data · Yield and purity · Specific activity

Introduction

The term nuclear data includes all data which describe either the properties of nuclei or their interactions. In general, all those data can be grouped under three headings, namely, nuclear structure, nuclear decay and nuclear reaction data. Accurate nuclear data are needed for applications in energy-related research (fission, fusion, accelerated driven systems, etc.) as well as non-energy related studies (medical radionuclide development, radiation therapy, astrophysics, etc.). The scope of this article is limited to medical applications, the basic aim of which is to provide a fundamental base for optimum production and application of radionuclides. The activities consist of new experimental measurements, nuclear model calculations, and standardisation and evaluation of existing data [for earlier reviews cf. 1–5].

As regards radioactive decay data, continuous experimental and evaluation efforts are underway [for a recent review cf. 6], and good quality data are available, for example from Evaluated Nuclear Structure and Decay Data File (ENSDF), BNL, USA. Only occasionally in case of some emerging radionuclides small discrepancies are observed, e.g. in the intensities of weak γ-ray lines, β^+ emission branching or Auger electron spectra [cf. 7]. Specific attempts then need to be undertaken to remove those discrepancies. It should be emphasized that the very choice of a radionuclide for medical application depends on its decay data.

With regard to nuclear reaction data for radionuclide production, mainly neutron and charged-particle induced reactions have been of interest. In recent years, however, some attention has also been paid to photon-induced reactions. The processes involved in neutron interactions are (n,γ) (n,f) and (n,p), and extensive data are available, for example from Evaluated Nuclear Data File (ENDF/B-VII.0), BNL, USA. For charged-particle induced reactions also the experimental data base has been considerably strengthened in recent years (cf. EXFOR, IAEA, Vienna). In contrast there is still paucity in photon-induced reaction data [cf. 8]. It should be pointed out that nuclear reaction data play a very important role in the optimisation of a production procedure,

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i.e. maximising the yield of the desired product and minimising the level of impurities [cf. 1, 2].

In this article the status of data of radionuclides used in nuclear medicine is briefly reviewed, with emphasis on data for research-oriented radionuclides.

Commonly used radionuclides in nuclear medicine

Status of nuclear data

Diagnostic radionuclides

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. To this end, in Single Photon Emission Computed Tomography (SPECT), γ-ray emitting radionuclides like ^{99m}Tc ($T_{½}$ = 6.0 h), ^{123}I ($T_{½}$ = 13.2 h) and ^{201}Tl ($T_{½}$ = 3.06 d) are commonly used. In positron emission tomography (PET), on the other hand, short-lived β^+-emitting radionuclides like ^{11}C ($T_{½}$ = 20.4 min), ^{13}N ($T_{½}$ = 10.0 min), ^{15}O ($T_{½}$ = 2.0 min), ^{18}F ($T_{½}$ = 110 min), ^{68}Ga ($T_{½}$ = 68.3 min) and ^{82}Rb ($T_{½}$ = 1.3 min) often find application. The radionuclides ^{99m}Tc, ^{68}Ga and ^{82}Rb are available through generator systems. The parent of ^{99m}Tc (i.e. ^{99}Mo) is produced using in a nuclear reactor but the parents of ^{68}Ga and ^{82}Rb (i.e. ^{68}Ge and ^{82}Sr) are produced using intermediate energy protons from a cyclotron or an accelerator. All of the other radionuclides are produced via direct charged-particle induced reactions at cyclotrons. The decay and production data of all those radionuclides are well known [cf. 2, 9–11].

As an example, the data for the ^{18}O(p,n)^{18}F reaction, which is now commonly used for the production of the most widely used PET radionuclide ^{18}F, are shown in Fig. 1A. The reaction was investigated both by neutron detection and residual activity measurement. The neutron data show better resolution but are difficult to quantify. The resonances are due to population of several high-lying states in the product nucleus. First systematic activation measurements by Ruth and Wolf [12], followed by detailed studies by Hess et al. [13], showed resonances at 5.1, 6.1 and 7.2 MeV which are in agreement with those found in spectral measurements of neutrons. It should be pointed out that due to resonance structure nuclear model calculations cannot reproduce excitation functions of reactions on light nuclei with reliable accuracies, calling for good experimental studies. On the other hand, while calculating the integral yield of the product, the structures disappear and a smooth curve is obtained (see Fig. 1B). The extended data of Hess et al. [13] up to 30 MeV should allow construction of large targets to produce ^{18}F in quantities up to 500 GBq (~14 Ci) or even more.

Therapeutic radionuclides

The spectrum of radionuclides commonly used in internal radionuclide therapy (endotherapy) is very broad. Since in this case a localised, well-defined radiation dose needs to be deposited in a malignant or inflammatory tissue, radionuclides emitting low-range highly-ionising radiation, i.e. α or β^- particles, conversion and/or Auger electrons, are of great interest. Among the β^- emitters, the radionuclides ^{32}P ($T_{½}$ = 14.3 days), ^{89}Sr ($T_{½}$ = 50.5 days), ^{90}Y ($T_{½}$ = 2.7 days), ^{131}I ($T_{½}$ = 8.0 d), ^{153}Sm ($T_{½}$ = 1.9 d), ^{169}Er ($T_{½}$ = 9.4 d), ^{177}Lu ($T_{½}$ = 6.7 days) and ^{188}Re ($T_{½}$ = 17.0 h) are often used. The radionuclides ^{90}Y and ^{188}Re are available through generator systems whereas the others are produced directly or via decay of the respective precursors. Regarding α-emitting radionuclides, hitherto ^{211}At ($T_{½}$ = 7.2 h) and ^{223}Ra ($T_{½}$ = 11.4 days) have found some application but several other radionuclides are in development (see below). As far as Auger electron and X-ray emitters are concerned, so far only ^{125}I ($T_{½}$ = 59.4 days) and ^{103}Pd ($T_{½}$ = 17.0 days), respectively, have found wide application. The radionuclide ^{131}Cs ($T_{½}$ = 9.7 days) is also finding increasing application.

The radionuclides ^{211}At and ^{103}Pd are produced at cyclotrons. All of the other radionuclides are produced via irradiations in a nuclear reactor. Except for some small discrepancies in a few cases, the decay and production data of all those radionuclides are fairly well known [cf. 3, 14].

Standardisation of nuclear data

When enough data have been accumulated for a certain radionuclide, it is recommended to evaluate and standardise the data to assist the user to choose proper conditions for the production of a medical radionuclide with acceptable quality.

As regards commonly used reactor-produced radionuclides, extensive neutron-induced reaction cross section data are available (e.g. in ENDF/B-VII.0), thanks to energy-related reactor programmes. Of special interest are (n,γ) reaction cross sections and fission yields. For some special cases, double neutron capture and (n,p) reaction cross sections are also needed and are available in the data files.

For accelerator-based radionuclides, the quality of available data varies. In case of photon-induced reactions, e.g. (γ,xn) and (γ,xp) reactions, an evaluated data file was prepared [8], but it is mostly based on nuclear model calculations. More work is needed to develop this file further.

As regards charged-particle induced reaction cross section data, the evaluation aspect was partly discussed earlier

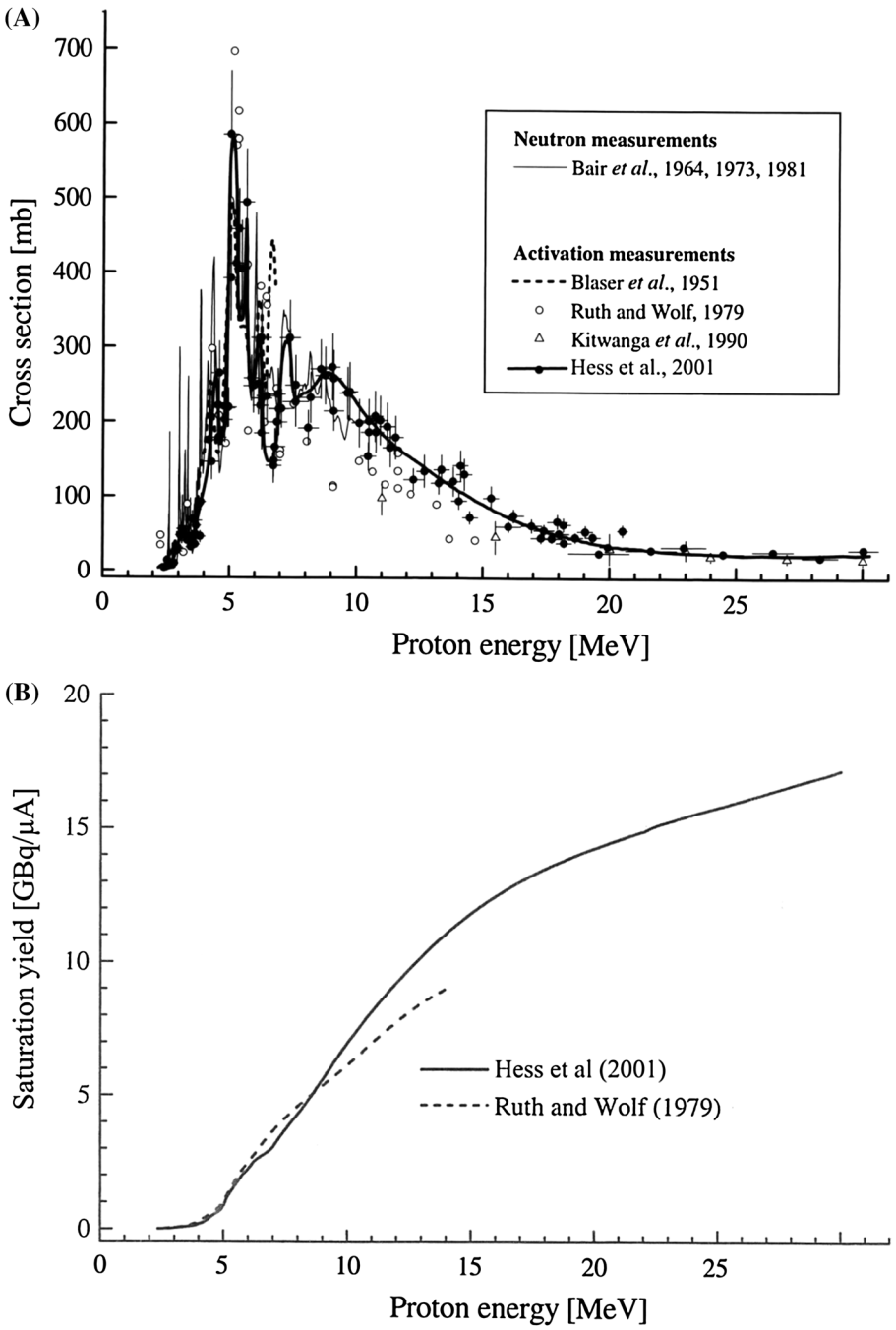

Fig. 1 **A** Excitation function of the $^{18}O(p,n)^{18}F$ reaction. Results of both neutron and activation measurements are shown. The rather *bold curve* is an eye-guide to the activation data (taken from Ref. [13]). **B** Thick target saturation yield of ^{18}F calculated from the data by Ruth and Wolf [12] and those by Hess et al. [13]

[4]. Considerable experimental efforts have been devoted in recent years by many groups. All data are promptly compiled in the EXFOR file. However, no Data Centre or research group was doing evaluation of charged-particle data. Therefore, the IAEA embarked on this mission about 20 years ago and organised two successive coordinated research projects (CRPs) in which about a dozen laboratories participated, under the guidance and chair of this author. Since no evaluation methodology existed at the start of the first project in 1995, the initial work was rather empirical. However, in later years strong application of nuclear models could be built in. All the major available nuclear model codes, namely GNASH, STAPRE, ALICE-IPPE, TALYS and EMPIRE were used.

While developing the evaluation methodology, three points clearly emerged:

(a) the need to adjust the reported formation sections of a radionuclide according to its latest available decay data, especially γ-ray intensities,
(b) the absolute necessity to standardise reactions used for charged-particle flux measurement. This need led

to the development of a large number of monitor reactions for proton, deuteron, ^{3}He- and α-particle beams over a wide range of energies [9]. In fact through the use of those monitor reactions, the accuracy of data reported in recent years from various laboratories has considerably increased.

(c) the nuclear model calculations reproduce the experimental data with varying degrees of success. Their main objective in the evaluation process should be to check the consistency in the reported data.

The final recommended excitation function is generally based on statistical fitting of the selected experimental data. The evaluated and recommended data for the major diagnostic radionuclides are now available in [9] and those for the therapeutic radionuclides in [14]. They should allow a proper selection of the projectile energy range in a target to ensure high radionuclidic purity of the desired radionuclide, and the accompanying radioactive impurities can be accurately calculated. The two files are being continuously updated. The discrepancies in the data for the ^{18}O(p,n)^{18}F reaction found in the file [9] were removed through extensive new measurements [13]. Similarly new experiments were done to remove the discrepancy in the ^{nat}Rb(p,xn)^{82}Sr reaction data [10] as well as in the ^{nat}Ga(p,xn)^{68}Ge reaction data [11]. Recently the IAEA has initiated a third CRP to deal with other emerging radionuclides. A few independent research groups are also engaged in the evaluation work [cf. 15].

Alternative routes of production of ^{99}Mo and ^{99m}Tc

The future supply of ^{99m}Tc (via the fission-produced ^{99}Mo parent) appears to be somewhat jeopardized. On one hand the presently used reactors are ageing and there appears to be no planning for their replacement and, on the other, there is the risk of nuclear weapons proliferation due to the use of highly enriched ^{235}U as the target material. Both these aspects demand a search for alternative methods of production and separation of ^{99}Mo and/or ^{99m}Tc. Presently considerable efforts are underway in this direction.

The suggested routes for the production of ^{99}Mo and ^{99m}Tc which are not dependent on a nuclear reactor are given in Table 1. Although the cross sections of those reactions [16–24] are not small, the non-availability of high fluxes of the respective projectiles leads to the conclusion that presently only the direct production of ^{99m}Tc via the ^{100}Mo(p,2n)^{99m}Tc reaction is promising. The ^{100}Mo(d,3n)^{99m}Tc reaction [20] is also interesting but, due to less availability of high-intensity deuteron beams, the emphasis is presently on the (p,2n) process.

A study of the ^{100}Mo(p,2n)^{99m}Tc reaction done at Jülich at the request of the IAEA had shown that this route using 22 MeV protons on a highly enriched ^{100}Mo target is worth considering for local production and consumption of ^{99m}Tc [21]. Over the last few years a large number of other measurements have been reported but the data show large scatter, especially over the energy range of 10–20 MeV. The values from Prague [22], Edmonton [23] and Debrecen [24], however, appear to be more accurate. Recently a critical evaluation of all available data was carried out [cf. 25] using a rigorous nuclear model analysis and statistical fitting procedure; the recommended excitation function [25] is shown in Fig. 2. This should allow calculation of the yield of ^{99m}Tc with enhanced confidence. Over the suitable energy range of $E_p = 22 \rightarrow 10$ MeV, the calculated yield of ^{99m}Tc amounts to 700 MBq/μAh.

It should be pointed out that the long-lived impurities ^{99g}Tc ($T_{½} = 2.1 \times 10^5$ a) and ^{98}Tc ($T_{½} = 4.2 \times 10^6$ a) associated with the ^{100}Mo(p,2n)^{99m}Tc process for the production of ^{99m}Tc may have strong repercussions on the utilization of ^{99m}Tc for medical applications. The theoretically calculated excitation functions for the formation of both those impurities are shown in Fig. 2 together with the recommended curve for the ^{100}Mo(p,2n)^{99m}Tc reaction. A recent mass spectrometric measurement of ^{99g}Tc [23] confirms the calculation up to 18 MeV. The cross section for the formation of ^{99g}Tc is about four times higher than that for ^{99m}Tc. From about 17 MeV onwards, the formation of ^{98}Tc also becomes significant. Evidently, even while using 100 % enriched ^{100}Mo as target material, the choice of the incident proton energy will have to be carefully controlled

Table 1 Alternative routes for production of ^{99}Mo and ^{99m}Tc

Reaction	Cross section	Status and/or reference
^{nat}U(γ,f)^{99}Mo	160 mb at 15 MeV	Evaluated data available [8]
^{232}Th(p,f)^{99}Mo	34 mb at 22 MeV	[16]
^{100}Mo(γ,n)^{99}Mo	150 mb at 14.5 MeV	[17]
^{100}Mo(n,2n)^{99}Mo	1,500 mb at 14 MeV	[18, 19]
^{100}Mo(p,pn)^{99}Mo	150 mb at 40 MeV	Evaluated data available [25]
^{100}Mo(d,p2n)^{99}Mo	438 mb at 36 MeV	[20]
^{100}Mo(p,2n)^{99m}Tc	284 mb at 17 MeV	Evaluated data available [25]
^{100}Mo(d,3n)^{99m}Tc	325 mb at 24 MeV	[20]

The ^{nat}U(n,f)^{99}Mo process with spallation neutrons also appears interesting, but the cross section is unknown

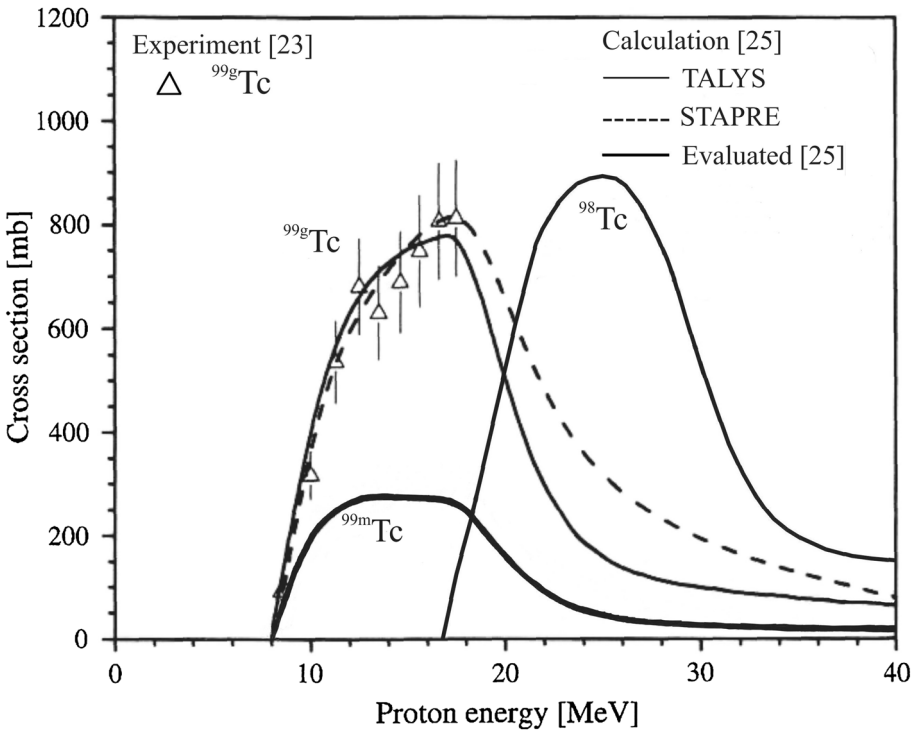

Fig. 2 Excitation functions of the ^{100}Mo(p,2n)^{99m,g}Tc and ^{100}Mo(p,3n)^{98}Tc reactions. The *bold curve* gives the evaluated values for the ^{100}Mo(p,2n)^{99m}Tc reaction [25]. The experimental data for the ^{100}Mo(p,2n)^{99g}Tc reaction were taken from [23] and the calculated data (using the codes TALYS and/or STAPRE) for the formation of ^{99g}Tc and ^{98}Tc are from [25]

to keep the level of the metal technetium impurity low so that ^{99m}Tc of the desired specific activity is obtained. This aspect may turn out to be critical in the acceptance of cyclotron-produced ^{99m}Tc for medical application. Thus, further extensive work is needed regarding the associated radionuclidic and metallic impurities.

Research oriented radionuclides

In recent years, nuclear data research related to medical radionuclides has been carried out mainly in two directions:

- Development of novel positron emitters
- Development of novel highly-ionising radiation emitters for internal radiotherapy

A brief discussion of those two areas is given below.

Novel positron emitters

With the growing significance of PET in diagnostic nuclear medicine, the need for novel positron emitters, also known as non-standard positron emitters, has been increasing, especially for studying slow metabolic processes and for quantification of targeted therapy.

The decay data of the emerging positron emitters are fairly well known; yet some discrepancies exist in a few cases, especially with regard to the β^+ emission intensities. Since in recent years many of the novel positron emitters can be obtained in a radionuclidically pure form, a combination of beta counting and high-resolution X-ray spectrometry can lead to very reliable results. Using this approach, recently β^+ emission intensities were accurately determined for ^{120}I [26], ^{124}I [27], ^{64}Cu and ^{76}Br [28]. In several other cases as well such measurements are recommended [cf. 7].

As regards production data of novel positron emitters, the subject has been treated in detail in two recent reviews [29, 30]. In this article, therefore, only a few salient features are discussed. The development of a suitable production process of a novel positron emitting radionuclide demands investigation of several nuclear reactions. In most cases it is necessary to use a highly enriched isotope as target material. The choice of the production route then depends mainly on the yield of the desired radionuclide and the expected level of the radioactive impurities. Other factors to be taken into account are the cost of the enriched target material, the ease of chemical separation of the product and the recovery of the enriched target isotope. The availability of the charged particles and their energies are also important. All those factors are well illustrated through measurements done in connection with the production of ^{124}I ($T_{½}$ = 4.18 days), mostly at Jülich in collaboration with the groups at Brussels, Cairo, Debrecen and iThemba LABS (Cape Town). The results are summarized in Table 2. The yields were calculated from the single measurements [31–33] on the (p,n), (d,2n) and (p,2n) reactions, but from the evaluated data for the other reactions [34, 35]. Of all the processes investigated, the

Table 2 Routes for production of ^{124}I. The ^{124}Te(p,n)^{124}I reaction gives the purest form of ^{124}I

Nuclear reaction	Energy range (MeV)	Thick target yield of ^{124}I (MBq/μA h)	Impurity (%)		
			^{123}I	^{125}I	^{126}I
^{124}Te(d,2n)	14 → 10	17.5	–	1.7	–
^{124}Te(p,n)	12 → 8	16	1.0	<0.1	–
^{125}Te(p,2n)	21 → 15	81	7.4	0.9	–
^{126}Te(p,3n)	36 → 26	190	148	1.3	1.0
^{121}Sb(α,n)	22 → 13	2.1	895	<0.05	<0.16
^{nat}Sb(α,xn)	45 → 32	6.7	537	1.2	0.3
^{123}Sb(α,3n)	45 → 32	15.5	236	1.2	0.3
^{123}Sb(^{3}He,2n)	19 → 13	0.73	11	0.6	0.6

Table 3 Some novel positron emitters for medical applications

Radionuclide ($T_{½}$)	Major production route	Energy range (MeV)	Application[a]
^{55}Co (17.6 h)	^{58}Ni(p,α)	15 → 7	Tumor imaging; neuronal Ca marker
	^{54}Fe(d,n)	10 → 5	
^{61}Cu (3.3 h)	^{61}Ni(p,n)	15 → 7	Immuno-PET
	^{64}Zn(p,α)	18 → 11	
^{64}Cu (12.7 h)	^{64}Ni(p,n)	14 → 9	Radioimmunotherapy
^{66}Ga (9.4 h)	^{66}Zn(p,n)	13 → 8	Quantification of SPECT-pharmaceuticals
^{72}As (26.0 h)	^{nat}Ge(p,xn)	18 → 8	Tumour imaging; immuno-PET
^{76}Br (16.0 h)	^{76}Se(p,n)	15 → 8	Radioimmunotherapy
^{82m}Rb (6.2 h)	^{82}Kr(p,n)	14 → 10	Cardiology
^{86}Y (14.7 h)	^{86}Sr(p,n)	14 → 10	Therapy planning
^{89}Zr (78.4 h)	^{89}Y(p,n)	14 → 10	Immuno-PET
^{94m}Tc (0.87 h)	^{94}Mo(p,n)	13 → 8	Quantification of SPECT-pharmaceuticals
^{120}I (1.3 h)	^{120}Te(p,n)	13.5 → 12	Iodopharmaceuticals
^{124}I (4.18 d)	^{124}Te(p,n)	12 → 8	Tumour targeting; dosimetry

Produced at a small cyclotron ($E < 20$ MeV) using mostly highly-enriched target material

[a] Each application involves PET imaging

^{124}Te(p,n)^{124}I reaction is the method of choice. Although the yield of ^{124}I is not very high, the product obtained is of the highest radionuclidic purity. It has the added advantage of being used at a small-sized cyclotron. This reaction was originally suggested by Scholten et al. [31] and it is now universally applied for the production of ^{124}I.

Most of the novel positron emitters have been developed using a small-sized cyclotron (with $E < 20$ MeV). A brief overview of the production routes of several emerging positron emitters is given in Table 3. The common route of production of each radionuclide is the low-energy (p,n)-reaction on an enriched target isotope. In a few cases other low-energy reactions, such as (d,n) and (p,α), have also been employed. For a detailed list of references relevant to nuclear data measurements see a recent review article [30]. For all the listed radionuclides some sort of novel medical application has been reported. The radionuclides ^{64}Cu, ^{86}Y, ^{89}Zr and ^{124}I, on the other hand, have already found wide application. The number of relevant reactions studied for their production has therefore also been correspondingly large. However, the low-energy (p,n)-reaction on an enriched target isotope is favoured because it generally leads to a high-purity product.

Figure 3 depicts the excitation function of the ^{64}Ni(p,n)^{64}Cu reaction which may be regarded as a typical (p,n) reaction in the medium mass region. It was used first at Jülich [36] for the production of ^{64}Cu and is now the method of choice at almost all laboratories. A later publication [37] supported those data. The excitation function can be described relatively well by nuclear model calculations [38].

It should be pointed out that three of the novel positron emitters listed in Table 3, namely ^{86}Y, ^{94}Tc and ^{120}I, have isomers. The choice of the nuclear reaction is then critical to obtain one isomer with the least possible amount of the other isomer (for more discussion [cf. 2]). Incidentally, in

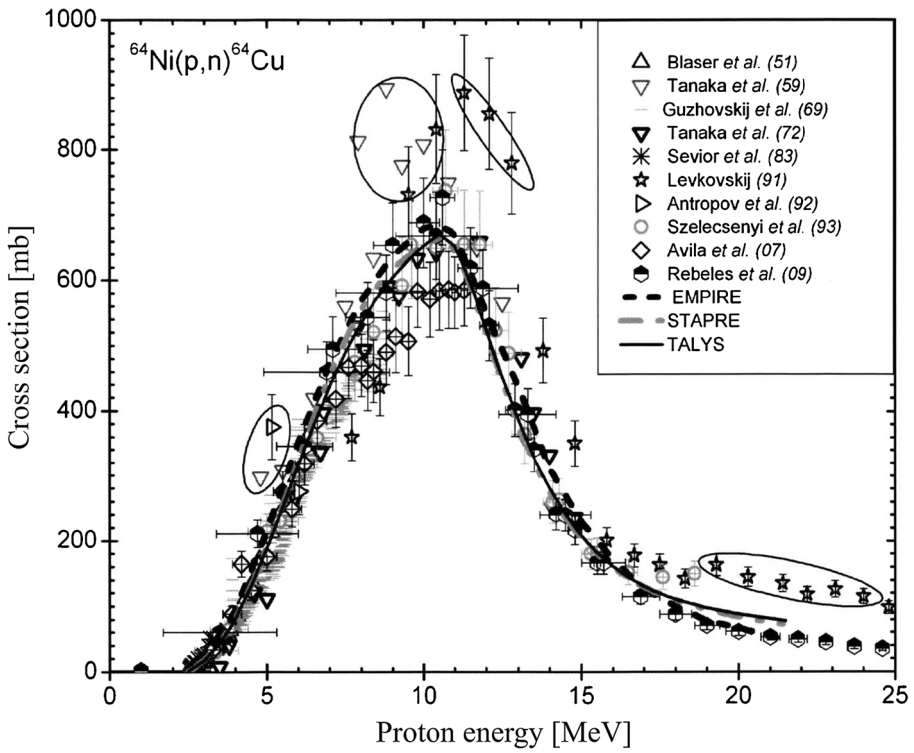

Fig. 3 Experimental data along with the results of nuclear model calculations using the codes EMPIRE, STAPRE and TALYS for the ^{64}Ni(p,n)^{64}Cu reaction. The *encircled data* were deselected in further evaluation (taken from Ref. [38])

all the three cases, the desired radionuclide is a low-spin isomer. So the low-energy (p,n) reaction on the corresponding highly enriched target, viz. ^{86}Sr(p,n)^{86}Y [39], ^{94}Mo(p,n)^{94m}Tc [40] and ^{120}Te(p,n)^{120g}I [41], gave the purest form of the desired product.

In addition to the positron emitters listed in Table 3, nuclear data work on several other potentially useful positron emitting radionuclides, e.g. ^{44}Sc ($T_{½}$ = 3.9 h), ^{45}Ti ($T_{½}$ = 3.1 h), etc. is also going on, especially in the energy region below 20 MeV. However, many useful or potentially useful positron emitters can be produced only using intermediate energy reactions. Extensive nuclear data work has shown that the production of the radionuclides ^{52}Fe ($T_{½}$ = 8.3 h), ^{73}Se ($T_{½}$ = 7.1 h), ^{77}Kr ($T_{½}$ = 1.2 h) and ^{83}Sr ($T_{½}$ = 32.4 h) demands a high intensity cyclotron or accelerator, delivering protons of energies up to about 70 MeV (in the case of ^{52}Fe preferably up to 100 MeV).

Besides protons, heavier charged particles like deuterons, ^{3}He- and α-particles may also induce a few useful reactions. For example, intermediate energy deuterons could be useful in the production of ^{64}Cu via the ^{nat}Zn(d,x)-process, a ^{3}He-particle beam in the production of ^{75}Br by the ^{75}As(^{3}He,3n)-process and an α-particle beam in the production of ^{73}Se via the ^{70}Ge(α,n)^{73}Se reaction (for more details cf. [30]). Several newer studies are in progress in a few laboratories (for example, see the contribution by Ditroi et al., this issue of the journal).

Similar to some commonly used positron emitters made available via generator systems, a few novel and potentially useful positron emitters could also be produced through generators. Some of them are: ^{44}Ti (60.4 a)/^{44}Sc (3.9 h), ^{72}Se (8.5 days)/^{72}As (26.0 h), ^{140}Nd (3.4 days)/^{140}Pr (3.4 min), etc. All of the above mentioned parent radionuclides can be produced only using intermediate energy charged particle beams.

Novel therapeutic radionuclides

The number of potentially interesting therapeutic radionuclides is very large. However, as mentioned above, in internal radionuclide therapy the emphasis has shifted to low-range but highly-ionising radiation emitters. In a recent review [4] many of the novel therapeutic radionuclides were discussed in some detail. The scope of this article is limited to the radionuclides which are presently attracting most attention. Their decay characteristics are summarized in Table 4. The decay and production data of each radionuclide are considered below individually.

^{67}Cu

This radionuclide is gaining increasing significance as a theragnostic pair of the β^{+} emitter ^{64}Cu. Its decay data are well known and the production methods have been recently reviewed [42]. Some of the reaction data have also been evaluated [14]. In general six reactions could possibly be used: (a) ^{70}Zn(p,α)^{67}Cu, (b) ^{70}Zn(d,nα)^{67}Cu, (c) ^{68}Zn(p,2p)^{67}Cu,

(d) ^{64}Ni(α,p)^{67}Cu, (e) ^{68}Zn(γ,p)^{67}Cu, (f) ^{67}Zn(n,p)^{67}Cu. The first reaction has found practical application [cf. 43] but the yield was rather low. The ^{68}Zn(p,2p)^{67}Cu reaction appears to be the most promising. The measured reaction cross sections, however, show some discrepancies [44–46]. Nonetheless, it is a relatively high-yield process and ^{67}Cu could be produced in batches of a few GBq [cf. 47] using a high-current enriched ^{68}Zn target and a proton energy range of $E_p = 80 \rightarrow 30$ MeV. As regards the ^{70}Zn(d,nα)^{67}Cu and ^{64}Ni(α,p)^{67}Cu reactions, so far only cross sections have been measured [48, 49] and the expected respective yield is low. In recent years there has been an increasing interest in the utilization of electron linear accelerators for medical radionuclide production and the reaction ^{68}Zn(γ,p)^{67}Cu has been investigated in some detail [50]. The yield of ^{67}Cu for a ^{nat}Zn target amounting to 1 MBq/(g kW h) is, however, rather low. Furthermore, even the old method used at nuclear reactors, namely the ^{67}Zn(n,p)^{67}Cu reaction, has received some newer attention [51]. While measuring cross sections of the ^{64}Zn(n,p)^{64}Cu and ^{67}Zn(n,p)^{67}Cu reactions in a TRIGA reactor spectrum, Uddin et al. [51] came to the conclusion that for theragnostic studies a good mixture of ^{64}Cu and ^{67}Cu could be produced. The cross sections of the two reactions differ considerably, possibly due to a significant difference in the proton binding energies in the two target nuclei [52]. The calculated yield of ^{67}Cu amounts to 4.4 MBq/(g h for 10^{14} n cm^{-2} s^{-1}). On the other hand, it remains questionable whether one can achieve in reactor production the same chemical and radiochemical purity of ^{67}Cu as in cyclotron production. The emphasis should therefore be on the ^{68}Zn(p,2p)^{67}Cu process using intermediate energy protons.

^{186}Re

Since the chemistry of Re is similar to that of Tc and since a large number of Tc-compounds are already finding applications in nuclear medicine, the radionuclide ^{186}Re is attractive for internal radiotherapy. The γ-ray emission intensities in its decay have been re-measured [53]. The original method of its production, namely the ^{185}Re(n,γ)^{186}Re reaction, has been abandoned due to the resulting low specific activity. The new method of preparation of this radionuclide is the ^{186}W(p,n)^{186}Re reaction, though the ^{186}W(d,2n)^{186}Re reaction has also been investigated and gives a higher yield than the (p,n) reaction. Most of the studies deal with nuclear reaction cross sections [cf. 54–59]; only in a few papers small scale production has been reported. A recent critical analysis [60] showed that, for obtaining a high-purity product, an enriched ^{186}W target is absolutely necessary and the maximum proton energy should not exceed 18 MeV. The formation of the very long-lived isomer, ^{186m}Re ($T_{½} = 2 \times 10^5$ a), also needs special consideration in both (p,n) and (d,2n) reactions. Based on nuclear model calculations the specific activity of ^{186}Re was also predicted [60]. More recent experimental work in this direction [61, 62] appears to support the theoretical considerations. Presently, some practical yield measurements [cf. 63] and other efforts are underway in a few laboratories to produce this radionuclide in larger quantities.

^{225}Ac

There is great interest in this α-particle emitting radionuclide; it is useful in itself as well as for providing ^{213}Bi ($T_{½} = 46$ min; $E_α = 5{,}900$ keV) through a generator system. The decay data involved in the relevant decay chain have been evaluated [cf. 14] and the half-life of ^{225}Ac was recently re-determined [64]. Regarding its production, presently extensive effort is being invested. On one hand its separation from nuclear waste (^{229}Th) is being optimised [65] and, on the other, the ^{226}Ra(p,2n)^{225}Ac reaction, making use of the radioactive target material ^{226}Ra, is being developed [cf. 66]. A third possibility under investigation is its production via irradiation of ^{232}Th with intermediate energy protons [67–69].

Table 4 Decay characteristics of some important emerging therapeutic radionuclides

Radionuclide	$T_{½}$ (days)	Radiation of interest (%)	End point energy (keV)
^{67}Cu	2.58	β$^-$ (100)	577
^{186}Re	3.78	β$^-$ (92.2)	1,070
^{225}Ac	10.00	α (100)	5,830
^{117m}Sn	13.61	IT (100)	161
		Conversion electrons	
^{193m}Pt	4.33	IT (100)	135
		Auger electrons	
^{195m}Pt	4.02	IT (100)	130
		Auger electrons	

The cross sections for the formation of ^{225}Ac in the proton irradiation of ^{232}Th [67, 68] are shown in Fig. 4. The calculated ^{225}Ac yield over the energy range $E_p = 140 \rightarrow 60$ MeV amounts to 4 MBq/μA h. The amount of ^{227}Ac impurity in ^{225}Ac was also estimated by Ermolaev et al. [67] and the result is given in Fig. 4. Over the above mentioned energy range the level of the ^{227}Ac impurity amounts to about 0.2 % of ^{225}Ac. It should be mentioned that the yield of ^{225}Ac via the ^{226}Ra(p,2n)-reaction over the energy range $E_p = 22 \rightarrow 10$ MeV amounts to 7 MBq/μA h. The yield through the ^{232}Th(p,x)^{225}Ac reaction is thus lower and the chemical processing involved is extensive. But it avoids the use of a radioactive target which is quite cumbersome. Anyway, all methods of ^{225}Ac production need further development. In fact extensive efforts are underway with regard to the production of ^{225}Ac via both the latter processes.

^{117m}Sn

This low-lying isomeric state of ^{117}Sn decays by heavily converted isomeric transition to the ground state. The emitted conversion electrons of energies about 160 keV are ideally suited for some internal therapeutic studies and the radionuclide has been under consideration for more than three decades. It has been produced in small quantities via four routes: (1) ^{117}Sn(n,ńγ)^{117m}Sn, (2) ^{121}Sb(p,2p3n)^{117m}Sn, (3) ^{nat}Cd(α,xn)^{117m}Sn, (4) ^{nat}In(α,pxn)^{117m}Sn. Very recently in a joint effort between BNL and Clear Vascular, Inc. it has been realized that the product obtained through the first two processes does not have a specific activity (and also chemical purity) high enough for medical applications. The more promising method for production appears to be the α-particle irradiation of cadmium or indium.

The cross section data for the ^{115}In(α,d + pn)^{117m}Sn reaction were determined by Fukushima et al. [70] up to 40 MeV and by Qaim and Döhler [71] up to 140 MeV. In both the works, a peak at about 38 MeV was observed; the magnitude of the cross section in the latter work was, however, by about a factor of 5 higher. From the data of Qaim and Döhler [71], the optimum energy range for the production of ^{117m}Sn was deduced to be $E_\alpha = 45 \rightarrow 20$ MeV, with the thick target yield of ^{117m}Sn as 370 kBq/μAh and the ^{113}Sn ($T_{½} = 115.9$ days) impurity level of <0.2 %. The yield is rather low but the advantage is that the main target isotope ^{115}In has an abundance of 95.7 % in ^{nat}In so that an enriched target is not necessary.

The cross section data for the ^{116}Cd(α,3n)^{117m}Sn reaction were measured by Montgomery and Porile [72] up to 45 MeV and for the ^{nat}Cd(α,xn)^{117m}Sn process by Qaim and Döhler [71] extensively over the energy range of 15–140 MeV. The excitation curve is composed of two reactions, namely ^{114}Cd(α,n)^{117m}Sn and ^{116}Cd(α,3n)^{117m}Sn. The latter reaction has higher cross section and is more important. The relatively high formation cross section of ^{117m}Sn in α-particle induced reactions is attributed to its high spin ($11/2^-$), which is expected to be preferentially populated [cf. 73–75].

In two more recent measurements under Brussels/Debrecen cooperation the reactions ^{nat}Cd(α,x)^{117m}Sn, ^{114}Cd(α,n)^{117m}Sn and ^{116}Cd(α,3n)^{117m}Sn were investigated up to 39 MeV [76, 77]. The data for the ^{nat}Cd(α,x)^{117m}Sn reaction [76] are slightly higher than those by Qaim and Döhler [71]. However, if the data of Qaim and Döhler [71] for the peak around 45 MeV, which is of prime interest in the

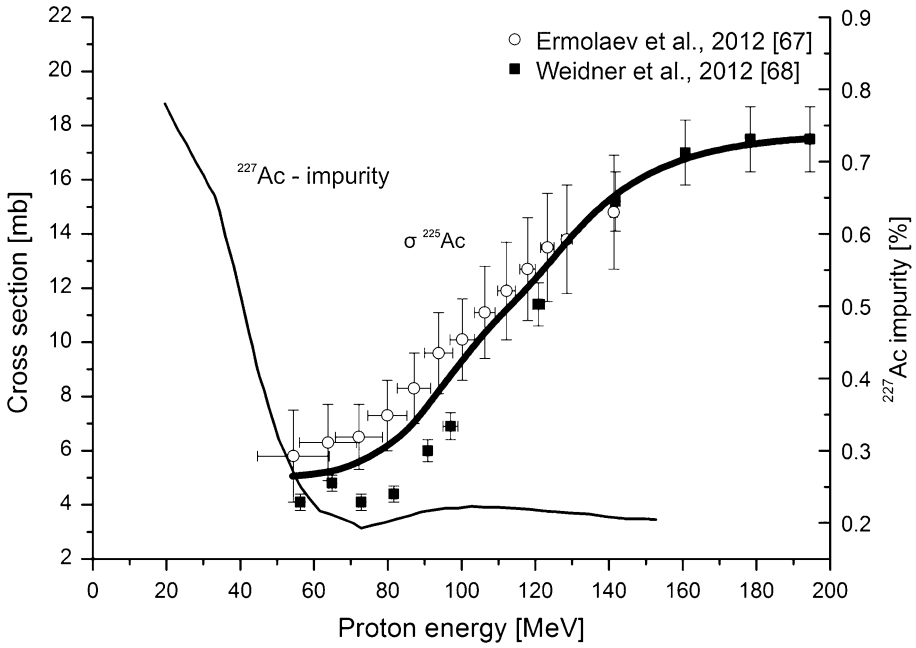

Fig. 4 Cross section data for the formation of ^{225}Ac in proton-induced reactions on ^{232}Th (*left scale*) shown as a function of projectile energy [taken from Refs. 67, 68]. The *thick solid line* is an eye-guide. The level of the ^{227}Ac impurity (*thin solid line, right scale*) is also given [67]

production of ^{117m}Sn, are normalised to 100 % ^{116}Cd enrichment (instead of 7.5 % in ^{nat}Cd), the resulting cross section values of about 1,200 mb are comparable to those reported by Adam Rebeles et al. [77] as well as by Montgomery and Porile [72]. Based on the data by Qaim and Döhler [71], which cover the full energy range of the peak, the energy region for the production of ^{117m}Sn via the ^{116}Cd(α,3n)-reaction is therefore suggested as $E_\alpha = 60 \rightarrow 30$ MeV. The thick yield of ^{117m}Sn for a 100 % ^{116}Cd target would amount to about 8.4 MBq/μAh with almost no isotopic radionuclidic impurity. Due to this reason the Clear Vascular, Inc. has chosen this process for production purposes. In view of the importance of this reaction it is recommended that more cross section measurements be done up to 70 MeV using highly enriched ^{116}Cd as target material.

^{193m}Pt and ^{195m}Pt

These two radionuclides are pure X-ray and Auger electron emitters, each decay leading to more than 30 secondary electrons, with their energies distributed between 10 and 130 keV. Since platinum-complexes (like cis-di-chlorodiaminplatinum) have been in use in chemotherapy as potent anti-tumour agents for a long time, both ^{193m}Pt and ^{195m}Pt have 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. The nuclear reactions employed, namely ^{192}Pt(n,γ)^{193m}Pt and ^{194}Pt(n,γ)^{195m}Pt, even while using highly enriched isotopes as target materials, do not lead to specific activity high enough for therapeutic application. Attempts to produce ^{195}Ir via double neutron capture by ^{193}Ir and its subsequent decay to ^{195m}Pt (i.e. via the route $^{193}\mathrm{Ir}(n,\gamma)^{194}\mathrm{Ir}(n,\gamma)^{195m,g}\mathrm{Ir}\xrightarrow{\beta^-}{}^{195m}\mathrm{Pt}$) have not been very successful. 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 two recent studies involving clean radiochemical separation of radioplatinum followed by X-ray spectrometry the excitation functions of the reactions ^{192}Os(α,n)^{195m}Pt [78, 79] and ^{192}Os(α,3n)^{193m}Pt [78, 79] could be measured. The results are shown in Fig. 5. 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. Thus this radionuclide can be produced in quantities sufficient for therapeutic applications.

Future perspectives and concluding remarks

The foregoing discussion shows that nuclear data play a very important role in the development of radionuclides for applications. The radioactive decay data govern the choice of a radionuclide for medical application and the nuclear reaction data allow optimisation of its production route. Whereas the decay data are well known, except for some small discrepancies in a few cases, the production data need more attention. In the case of neutron-induced reactions, cross sections are needed for radionuclide production in a nuclear reactor, and extensive information is available. However, regarding photon- and charged-particle induced reactions, further research and development work is necessary. The new data needs generally refer to development of novel positron emitters and low-range highly-ionising radiation emitters for internal radiotherapy. Whereas the former are generally produced at a low-energy ($E < 20$ MeV) cyclotron via the low-energy (p,n), (p,α) and (d,n) reactions, etc., for the production of the latter a medium-sized cyclotron is required. On the other hand, for production of several positron emitters, like ^{52}Fe ($T_{½} = 8.3$ h), ^{73}Se ($T_{½} = 7.1$ h), ^{83}Sr ($T_{½} = 32.2$ h), etc. and therapeutic radionuclides such as ^{67}Cu ($T_{½} = 2.58$ days), ^{225}Ac ($T_{½} = 10.0$ days), etc., intermediate energy protons from a cyclotron or an accelerator are needed. Another point worth considering is the availability of multiparticle cyclotrons. The deuteron-induced production of a few radionuclides leads to higher yield than the proton-induced production, e.g. ^{64}Cu ($T_{½} = 12.7$ h) from a ^{64}Ni target, ^{103}Pd ($T_{½} = 17.0$ days) from a ^{103}Rh target, ^{186}Re ($T_{½} = 3.7$ days) from a ^{186}W target, etc. The available deuteron beam currents are, however, generally much lower than for proton beams. On the other hand, a few modern cyclotrons/accelerators can deliver high-intensity deuterons (e.g. ACSI TR30 in Canada, SARAF in Israel) so that the use of deuteron-induced reactions may become advantageous. The α-particle beam is also very useful in populating low-lying high-spin isomeric states [cf. 73–75] which deexcite by IT and emit low-energy electrons (conversion or Auger, or both) that can be used in internal therapy. Examples are ^{117m}Sn, ^{193m}Pt, ^{195m}Pt, etc. The cross section data measurements in those cases are more challenging because often X-ray spectrometry is required. The increasing future demands of the novel radionuclides will decide what type of cyclotrons or accelerators should be favoured and constructed. The emphasis in nuclear data work will also have to be adjusted accordingly.

Over the last few years, two important new developments in the application of radionuclides have been emerging. The first one deals with a combination of PET and internal radiotherapy (as first demonstrated by Herzog et al. [80]), and the second one with a combination of PET and magnetic resonance imaging (MRI). In the latter case a β^+ emitting radionuclide of the contrast agent manganese [e.g. ^{51}Mn ($T_{½} = 46.2$ min) or ^{52}Mn ($T_{½} = 5.6$ days)] would be useful. The production data for those

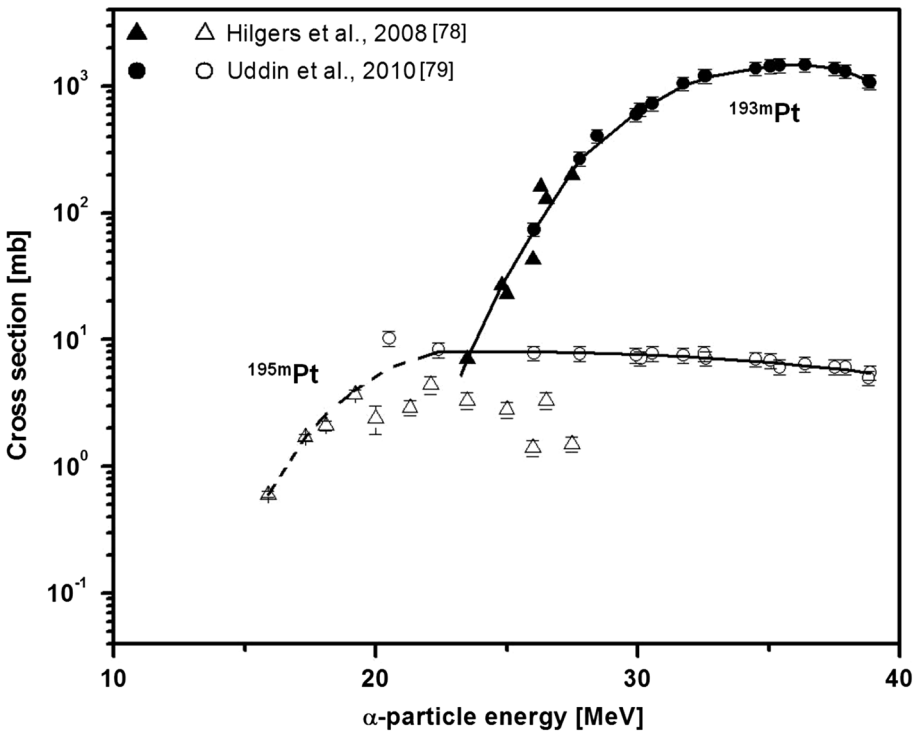

Fig. 5 Excitation functions of the $^{192}Os(\alpha,n)^{195m}Pt$ and $^{192}Os(\alpha,3n)^{193m}Pt$ reactions determined radiochemically using X-ray spectrometry [78, 79]. The *solid lines* are based on nuclear model calculations [cf. 79], and the *dashed line* is an eye-guide

radionuclides have been recently measured [cf. 81, 82]. Evidently, for those two fast expanding applications, further novel radionuclides will have to be developed (for more details cf. [4]). On a longer term basis, the radionuclide research could also involve a combination of radioactivity and nanotechnology, provided toxicity problems are overcome. The future perspectives of radionuclide research for medical applications thus appear to be very bright. In fact it is a flourishing and fast expanding field, even in developing countries. The supporting nuclear data research will therefore also continue to receive further attention.

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