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Recent Advances in Radiochemical Studies of Low and Intermediate Energy Nuclear Reactions Induced by Light Particles

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Radiochemical methods are of considerable significance in studies of low and intermediate energy nuclear reactions induced by light particles, particularly when low-yield and soft-radiation emitting products are involved. A brief description of the pertinent experimental techniques and nuclear model calculational codes is given. The data related to nucleon emission, complex particle emission and formation of isomeric states are reviewed. The nuclear model calculations can reproduce the nucleon emission cross sections fairly well. In the case of isomeric cross sections the calculations demand a careful choice of the input parameters, especially regarding the level structure of the product nucleus. The emission of complex particles is as yet not described well by the theory; the radiochemically determined data serve to develop systematic trends which qualitatively enhance our knowledge on those processes.

1. Introduction

Studies of nuclear reactions in the low and intermediate energy regions are of considerable importance for testing nuclear models and for practical applications. It is known that in the energy region below 10 MeV, compound nuclear processes dominate. Between 10 and 30 MeV the contribution of pre-compound reactions increases and at energies above 30 MeV it becomes appreciable. At energies above 50 MeV till about 150 MeV, a transition occurs from pre-compound to spallation, a region which is relatively less understood. Beyond 200 MeV the spallation process dominates. Thus systematic studies of nuclear reactions from their thresholds up to about 200 MeV, covering emission of various types of radiation and particles (photons, nucleons, complex particles, etc.) should further enhance our understanding of the various nuclear theories involved. Another important fundamental aspect, attracting considerable attention in recent years, has been the formation of isomeric states in product nuclei.[1] The isomeric cross-section ratio for a pair of isomeric states is known to depend on the spins of the isomers concerned, as well as on the spins of the higher lying levels feeding the isomers.[2] A study of the isomeric cross-section ratio as a function of the incident particle energy, especially near the threshold of a reaction, should therefore lead to useful information on the spin-cutoff parameter as well as on the level structure of the product nucleus. As far as practical applications are concerned, the cross section data are needed for calculations on neutronics, activation and heat generation in nuclear technology,[3] as well as for production of high-purity medical radionuclides, especially at cyclotrons.[4]

Nuclear reactions are generally studied by physical techniques involving on-line recording of emitted radiation and particles. Since with those techniques both angular and energy distributions of the emission products can be determined, the results lead to very useful information on the reaction mechanism.[5] The other way of investigating a nuclear reaction is to identify the reaction product by an off-line method. For an unambiguous characterisation of the product often a radiochemical separation is needed.[5] The product itself could be identified via measurement of its radioactivity or, in some special cases, via mass spectrometry (MS)[6] and/or accelerator mass spectrometry (AMS).[7] Obviously the off-line method, usually known as the "radiochemical method", cannot yield the same amount of information on the reaction mechanism as the physical technique which involves recording of the emission spectra. Resort is made here to comparisons of experimental data with the results of nuclear model calculations and, therefrom, to deduce mechanistic information. This paper reviews some of the recent studies performed using the radiochemical technique. This approach is particularly useful when the matrix activity is strong and the desired reaction product is weak. The technique is also very advantageous when the reaction product emits very soft radiation (e.g. low-energy β^--particles or X-rays). Furthermore, for practical applications, the integral data obtained via the radiochemical technique are often more useful than the differential data determined via on-line emission spectrum measurement techniques.

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2. Experimental Methods

2.1. Sample preparation and irradiations. Samples of very high-purity and well-defined chemical composition are needed, particularly when low-yield reactions are under investigation. In work with neutrons, solid, liquid, or gaseous material packed in polyethylene, aluminium, or quartz can be irradiated. In measurements with charged particles, on the other hand, relatively thin samples are needed. They are prepared by methods like sedimentation, vacuum evaporation, electrolytic deposition, etc.

Irradiations and projectile flux determination demand considerable care since in cross section determination quantitative measurements are involved. Monoenergetic neutrons in the region of about 1 MeV are produced via the ^{7}Li(p, n)^{7}Be reaction, those in the range of 3 to 12 MeV via the ^{2}H(d, n)^{3}He reaction, and the ones in the energy range of 13 to 20 MeV via the ^{3}H(d, n)^{4}He reaction. The low-energy deuteron beam (< 500 keV) is produced in a Cockroft-Walton type generator, and the higher energy beam is generally furnished by a Van de Graaff machine or a cyclotron. Irradiations with charged particles are done using beams extracted from cyclotrons or other accelerators. Generally thin samples placed in a row are irradiated together ("stacked-foil technique"). Due to energy degradation in the matter, the effective projectile energy in each sample is different. It is thus possible to determine cross sec-

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tions at several energies in just one experiment. In radiochemical studies, involving both neutrons and charged particles, the projectile flux is measured using suitable monitor reactions.[8,9]

2.2. Radiochemical separations. Specific radiochemical separations facilitate the isolation of transmutation products. A variety of radiochemical methods like precipitation, co-precipitation, solvent extraction, ion-exchange chromatography, thermochromatography, distillation etc. have been employed. Some of the recent examples include the study of (p, ^{7}Be) reaction on several transitional metals via high-performance ion-chromatographic separation of beryllium,[10] the ^{92}Mo(α, 2n)^{94}Ru process via thermochromatography,[11] the short-lived isomeric pairs ^{52m,g}Fe and ^{53m,g}Fe via the solvent extraction technique,[12] and the ^{58}Ni(n, α)^{55}Fe and ^{63}Cu(n, p)^{63}Ni processes via the ion-exchange method.[13,14] For the separation of some reaction products, a combination of several separation methods/techniques may be essential. A good example is the separation of ^{45}Ca from Ti irradiated with protons.[15] In addition to the removal of the matrix activity, the radiochemical separations also facilitate the preparation of thin sources for β^--counting or soft X-ray spectrometry. The excitation functions of the ^{50}Cr(n, np+d)^{49}V and ^{58}Ni(n, α)^{55}Fe reactions, for example, were determined via measurement of the 4.51 and 5.89 keV X-rays of radiochemically separated ^{49}V and ^{55}Fe, respectively[13]. Similarly the ^{48}Ti(n, α)^{45}Ca, ^{45}Sc(n, p)^{45}Ca, ^{nat}Ti(p, x)^{45}Ca and ^{63}Cu(n, p)^{63}Ni reactions were studied by radiochemical separations of the soft radiation emitting ^{45}Ca and ^{63}Ni which were subsequently characterised by β^--counting. In studies on tritium emission, on one hand the residual nuclide, if radioactive, was radiochemically separated and assayed and, on the other, the accumulated tritium was separated and counted.[16]

2.3. Measurement of radioactivity. For characterising relatively strong reaction products, conventional γ-ray spectroscopy or β^--counting was used. In investigations on low-yield reaction products, however, low-background detectors were employed. In the case of low-energy γ-rays, small-sized Ge detectors (with Be-windows) and, for X-rays, Si(Li) detectors have been commonly used. The very soft β^--emitters (like tritium) and the very soft X-ray emitter (like ^{37}Ar) were determined via low-level gas counting.[16]

2.4. Other methods of product assay. In addition to measurement of radioactivity, some of the products could be identified better by mass spectrometry (MS) or accelerator mass spectrometry (AMS). These techniques are especially suitable when the transmutation product is stable or very long-lived. Mass spectrometry has been occasionally applied to the determination of inert gases, like ^{3}He and ^{4}He, formed in the interactions of neutrons with various elements.[6,16] The accelerator mass spectrometry involves mass separation after acceleration of ions to several MeV energies and has been successfully applied to the determination of cosmogenic nuclides like ^{26}Al and ^{36}Cl, environmentally related nuclide ^{14}C, and the high-energy nuclear fragmentation product ^{10}Be.[17,18]

3. Nuclear Model Calculations

Low energy nuclear reactions are commonly treated in terms of the statistical model, generally using the Hauser-Feshbach formalism, which takes into account the angular momentum of the evaporated particle and the level structure of the product nucleus. Later the pre-compound effect was also introduced and the relevant codes GNASH (in USA) and STAPRE (in Europe) in several versions have been very successfully utilized over the last 25 years in evaluating excitation functions, especially of neutron induced reactions up to 20 MeV.

Above 20 MeV the pre-compound effect becomes increasingly important and at energies above 50 MeV, it plays a dominant role. A very commonly used code in the intermediate energy region was ALICE, developed by Blann about 30 years ago. Recently the Obninsk group introduced several modifications and termed it as the code ALICE-IPPE.[19] The incorporated modifications include treatment of the level density in a sophisticated way and consideration of the pre-equilibrium cluster emission (d, t, and ^{4}He). The code has been successfully applied to the calculation of excitation functions of a large number of reactions.

In recent years a further calculational code, namely EMPIRE II, has also been introduced.[20] It combines the general features of the statistical process, the pre-equilibrium exciton model and the various improvements mentioned above. Its use has as yet not been so extensively tested as in the case of the other two calculational codes.

4. Survey of Experimental Data, Comparison with Theoretical Calculations, and Discussion

Extensive experimental and theoretical studies have been performed in the low-energy region up to about 20 MeV, especially in the case of neutron induced reactions. With the increasing incident particle energy, the experimental database becomes weaker. At energies between 100 and 200 MeV, for example, very few data points are available. Nuclear model calculations are fairly successful up to about 100 MeV. The various nuclear processes are considered below separately.

4.1. Emission of nucleons. In the low and intermediate energy regions, the emission of nucleons has been extensively studied, mainly because the reaction cross sections are high. Many of the reaction products have been identified without radiochemical separations, except for cases where β^--emitters or low-energy X-ray emitters are involved. With neutrons, studies are generally limited up to 20 MeV and the reactions of interest are (n, *x*n), (n, pn), etc. For theoretical analysis mostly the codes GNASH and STAPRE have been used. In the case of protons, measurements have been performed often up to 100 MeV, the emphasis being on (p, *x*n) and (p, p*x*n) processes. Besides GNASH and STAPRE, considerable use has been made of the code ALICE-IPPE to calculate the excitation functions. Some recent studies on the target elements Cu, Zn, Rb, Te, Pb, etc. show that both the (p, *x*n) and (p, p*x*n) processes can be described very well by the model calculations,[21-25] provided the input parameters are properly chosen. The same is true for (d, *x*n), (^{3}He, *x*n) and (α, *x*n) processes where, however, more care in parameter choice is necessary.[26-30] In Figure 1 are shown, as typical examples, the experimental results on the (p, *x*n) processes on ^{85}Rb and ^{122}Te. In the former case calculations were done using the code ALICE-IPPE and in the latter the code STAPRE. In both cases generally good agreement was found between the experiment and theory, the latter code having also the capability of calculating the excitation function of the isomeric state ^{120m}I. The code ALICE-IPPE has some problem in reproducing the simple (p, n) reaction at energies > 30 MeV. In the study of neutron and proton induced reactions recently the code EMPIRE II has also been used and the results are more or less similar to those from the other codes. Between 100 and 200 MeV the experimental database is rather weak and the differences between the experimental data and the results of nuclear model calculations become increasingly significant. Similarly when the number of emitted nucleons becomes large, (i.e. several neutrons and several protons) or when an unusual reaction like (p, 2p) occurs, the deviations between the experiment and model calculations may increase.[22,23]

4.2. Emission of complex particles. In contrast to nucleon emission, the emission of complex particles (d, t, ^{3}He, α, ^{7}Li, ^{7}Be, ^{10}Be, etc.) has not been extensively studied, mainly due to both experimental and calculational difficulties (for a detailed review on this topic cf. Ref. 16). This is an area of research demanding extensive use of radiochemical separations. Tritium and ^{3}He emission was studied radiochemically mainly in neutron

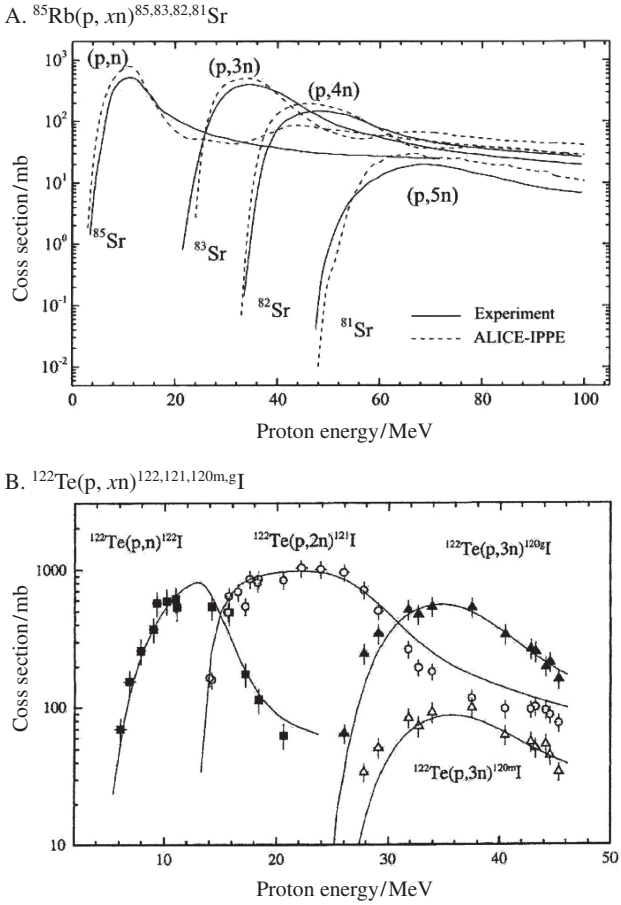

Figure 1. Excitation functions of (p, xn) reactions on (A) ^{85}Rb and (B) ^{122}Te, measured via the activation technique. The experimental data for ^{85}Rb are shown as smooth solid curves and the calculational results from the code ALICE-IPPE as dashed curves. For ^{122}Te, the experimental results are depicted by symbols and the values from the code STAPRE as solid curves (results taken from Refs. 24 and 26).

induced reactions using tritium counting and mass spectrometry (cf. Ref. 16). The α-particle emission data were obtained both via mass spectrometry[17] and measurement of the reaction products. In the case of ^{7}Be and ^{10}Be, radiochemical separations followed by γ-ray spectrometry[31] and accelerator mass spectrometry,[18] respectively, have been applied. As far as nuclear model calculations are concerned, there is as yet no satisfactory method of estimating the complex particle emission cross section, the exceptions being triton and α-particle emissions. In the former case the statistical process has been partly successful and, in the latter, a combination of statistical and pre-compound processes has successfully reproduced the experimental data. In other cases development of systematic trends has proved to be very advantageous in describing some characteristics of those reactions. As an example, some results for the (p, ^{7}Be) reaction[31] are shown in Figure 2. In the light mass region, ^{7}Be is expected to be formed as a product nucleus. For medium and heavy mass target nuclei, however, the emission of the complex particle (^{7}Be) is most probable. The systematic trends of the excitation functions reveal that for proton energies above 40 MeV the emission of ^{7}Be decreases with the increasing mass of the target nucleus. The trend is also valid for the heavy elements (Au and Bi) which undergo fission with intermediate energy protons. In a similar recent radiochemical study[18] the target dependence of ^{7}Be and ^{10}Be emission in neutron and α-particle induced reactions was studied, the former product via γ-ray spectrometry and the latter via AMS. The results for α-particle induced reactions at 400 MeV are shown in Figure 3. Evidently, the heavier targets show a preferential emission of the ^{10}Be fragment over ^{7}Be, probably due to the increasing N/Z ratio with the increasing mass of the target nucleus.

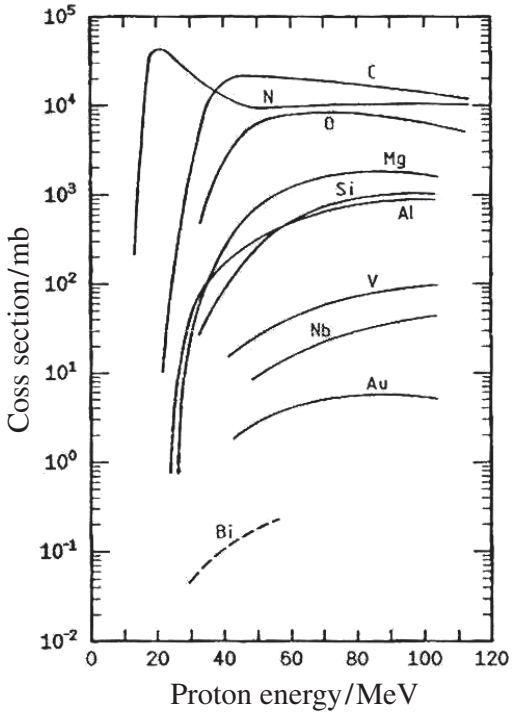

Figure 2. Systematics of (p, ^{7}Be) excitation functions on target elements C to Bi (taken from Ref. 31).

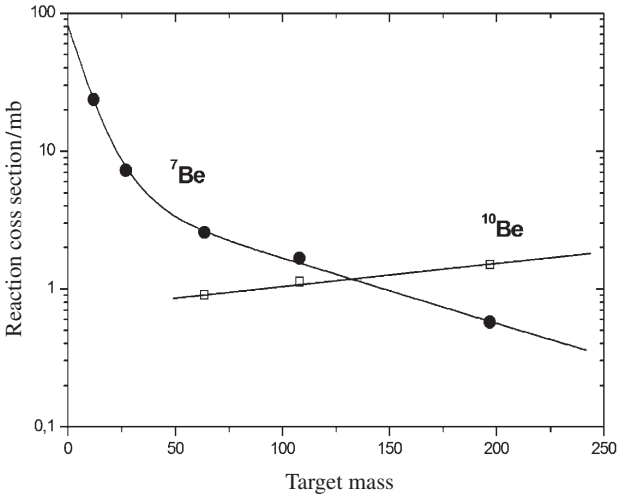

Figure 3. Yields of ^{7}Be and ^{10}Be in 400 MeV α-particle induced reactions shown as a function of the target mass (taken from Ref. 18).

4.3. Formation of isomeric states. In comparison to the total cross section of a reaction channel, the partial cross section describing the population of a particular isomeric state is more difficult to measure and to calculate (for a detailed review on this topic cf. Ref. 1). Due to emission of soft radiation by the low-lying levels involved, radiochemical methods of separation and thin target preparation are ideally suited for the measurement of isomeric cross sections. In recent years extensive investigations have been performed on several isomeric pairs, e.g. ^{52m,g}Mn, ^{58m,g}Co, ^{73m,g}Se, ^{94m,g}Tc, ^{120m,g}I, ^{197m,g}Hg, etc.,[2,26,32-36] involving different combinations of target, projectile, ejectile, and level structure of the product nucleus. Nuclear model calculations have been successfully performed using the code STAPRE and partly EMPIRE II (both incorporating statistical and pre-compound models) in combination with a very careful choice of the input parameters (optical model parameters, level structure of the product nucleus, branching ratios of γ-rays from discrete levels, spin dependence of the level density distribution, etc.). The results for the formation of a typical

isomeric pair ^{58m,g}Co in the ^{61}Ni(p, α)-reaction[32] are shown in Figure 4. The metastable state has a spin value of (5$^+$) and the ground state (2$^+$). The characterisation of the low-energy transition (E=25 keV) was facilitated by the radiochemical method. Evidently, the probability of formation of the high-spin isomer increases rapidly with the increasing kinetic energy of the projectile. The experimental data are reproduced approximately by the STAPRE calculation, especially when the η value (ratio of Θ_{eff} to Θ_{rig}) is properly chosen. In a recent study[35] the formation of the isomeric pair ^{52m,g}Mn was investigated in five nuclear processes and the results are shown in Figure 5. The important role of the spins of the two states involved is confirmed. The cross section of the low-spin isomer in comparison to that of the high-spin ground state initially decreases with the increasing incident projectile energy, but becomes almost constant at high excitation energies. The (p, n) and (^{3}He, t) reactions (curves A and B) occur on the same target nucleus and the product studied is also the same. The same is true for the (d, α), (n, t) and (^{3}He, αp) reactions (curves C, D, and E). The magnitudes of the cross-section ratios, however, differ considerably. It is concluded that the reaction channel affects the isomeric cross-section ratio appreciably, particularly when the channels differ widely, e.g. (p, n) and (^{3}He, t) processes. Very recently the formation of high spin isomers ^{195m}Hg and ^{197m}Hg was studied in several reactions.[36] A description of the isomeric cross-section ratio by the model was possible only with a very low value of η, i.e. the Θ_{eff} to Θ_{rig} ratio. On the basis of those results, a mass dependence of η has been proposed.

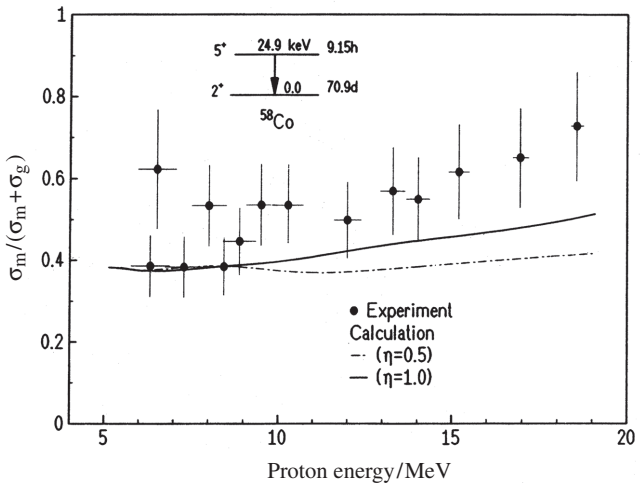

Figure 4. Isomeric cross-section ratio for the ^{61}Ni(p, α)^{58m,g}Co process as a function of proton energy. The nuclear model calculation was done using the code STAPRE for two values of η (ratio of Θ_{eff} to Θ_{rig}) to demonstrate the effect of spin distribution of level density (taken from Ref. 32).

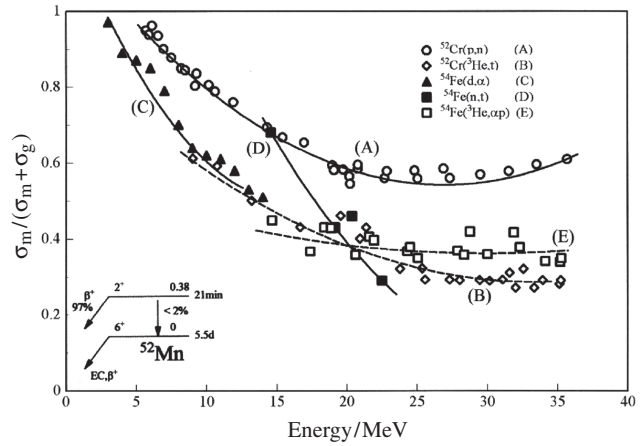

Figure 5. Experimental isomeric cross-section ratios for the formation of ^{52m,g}Mn in several nuclear reactions, plotted as a function of the incident particle energy (taken from Ref. 35).

5. Conclusions

The radiochemical method of nuclear reaction cross section measurements is commonly used in low and intermediate energy regions. It is especially advantageous for studies on complex particle emission and is ideally suited for measurement of isomeric cross sections. In the case of nucleon emission, the experimental total and partial cross sections of a reaction channel are reproduced fairly well by a combination of the statistical and pre-compound models, provided the input parameters are properly chosen. In contrast, the complex particle emission is not described properly by those models, presumably due to strong contributions from direct processes. Further experimental and theoretical work in this direction is necessary.

References

(1) S. M. Qaim, *Proc. Intern. Conf. Nucl. Data for Science and Technology,* Gatlinburg, 1994, Ed. J. K. Dickens (ANS, Inc., LaGrange Park, USA), 1994, p 186.
(2) S. M. Qaim, A. Mushtaq, and M. Uhl, Phys. Rev. C**38,** 645 (1988).
(3) E. T. Cheng, A. B. Pashchenko, and J. Kopecky, Fusion Technology **30**, 1182 (1996).
(4) S. M. Qaim, Radiochimica Acta **89**, 223 (2001).
(5) S. M. Qaim, *Proc. Int. Conf. Nucl. Data for Reactors and other Applications,* Harwell, 1978, OECD-NEA, Paris, 1979, p 1088.
(6) D. W. Kneff, B. M. Oliver, H. Farrar IV, and L. R. Greenwood, Nucl. Sci. Eng. **92**, 491 (1986).
(7) M. Suter, *Proc. Int. Conf. Nucl. Data for Science and Technology,* Jülich, 1991, Ed. S. M. Qaim, Springer Verlag, Berlin, Heidelberg, 1992, p 381.
(8) International Reactor Dosimetry File - IRDF-2002 (International Atomic Energy Agency, Vienna, Austria, 2002).
(9) IAEA-TECDOC-1211, Charged Particle Cross Section Database for Medical Radioisotope Production: Diagnostic Radioisotopes and Monitor Reactions (International Atomic Energy Agency, Vienna, Austria, 2001).
(10) M. Fassbender, S. Spellerberg, and S. M. Qaim, Radiochim. Acta **73**, 39 (1996).
(11) F. O. Denzler, F. Rösch, and S. M. Qaim, Radiochim. Acta **68**, 13 (1995); Erratum **75**, 227 (1996).
(12) A. Fessler and S. M. Qaim, Radiochim. Acta **72**, 121 (1996).
(13) A. Fessler and S. M. Qaim, Radiochim. Acta **84**, 1 (1999).
(14) S. M. Qaim, S. Spellerberg, F. Cserpak, and J. Csikai, Radiochim. Acta **73**, 111 (1996).
(15) K. Kettern, I. Spahn, S. Spellerberg, S. M. Qaim, and H. H. Coenen, *Proc. Int. Conf. Nucl. Data for Science and Technology,* Santa Fe, USA, 2004, Eds. R. C. Haight, M. Chadwick, T. Kawano, and P. Talou (AIP, Melville, New York), Vol. 769, 2005, p 758.
(16) S. M. Qaim, Radiochim. Acta **70/71**, 163 (1995).
(17) R. Bodemann, H. J. Lange, I. Leya, R. Michel, T. Schiekel, R. Rösel, U. Herpers, H. J. Hofmann, B. Dittrich, M. Suter, W. Wölfli, B. Holmqvist, H. Condé, and M. Malmborg, Nucl. Instrum. Methods B**82**, 9 (1993).
(18) H. Matsumura, T. Sanami, K. Masumoto, N. Nakao, A. Toyoda, M. Kawai, T. Aze, H. Nagai, M. Takada, and H. Matsuzaki, Radiochim. Acta **93**, 497 (2005).
(19) I. Dityuk, A. Yu. Konobeyev, V. P. Lunev, and Yu. N. Shubin, *New Version of the Computer Code ALICE-IPPE.* Report INDC(CCP)-410 (International Atomic Energy Agency, Vienna, Austria, 1998).

(20) M. Herman, R. Capote, B. Carlson, P. Oblozinsky, M. Sin, A. Trkov, and V. Zerkin, *EMPIRE-II, Nuclear Reaction Model Code, Version 2.19 (Lodi)* (International Atomic Energy Agency, Vienna, Austria, 2005).
(21) M. Fassbender, Yu. N. Shubin, V. P. Lunev, and S. M. Qaim, Appl. Radiat. Isot. **48**, 1221 (1997).
(22) M. Fassbender, Yu. N. Shubin, and S. M. Qaim, Radiochim. Acta **84**, 59 (1999).
(23) T. Stoll, S. Kastleiner, Yu. N. Shubin, H. H. Coenen, and S. M. Qaim, Radiochim. Acta **90**, 309 (2002).
(24) S. Kastleiner, Yu. N. Shubin, F. M. Nortier, T. N. van der Walt, and S. M. Qaim, Radiochim. Acta **92**, 449 (2004).
(25) A. Hohn, F. M. Nortier, B. Scholten, T. N. van der Walt, H. H. Coenen, and S. M. Qaim, Appl. Radiat. Isot. **55**, 149 (2001).
(26) S. Sudár, A. Hohn, and S. M. Qaim, Appl. Radiat. Isot. **52**, 937 (2000).
(27) A. Hermanne, M. Sonck, S. Takács, F. Tárkányi, and Yu. Shubin, Nucl. Intrum. Methods B**187**, 3 (2002).
(28) F. Tárkányi, A. Hermanne, S. Takács, Yu. N. Shubin, and A. I. Dityuk, Radiochim. Acta **92**, 223 (2004).
(29) K. Hilgers, Yu. N. Shubin, H. H. Coenen, and S. M. Qaim, Radiochim. Acta **93**, 553 (2005).
(30) S. Sudár and S. M. Qaim, Phys. Rev. C**50**, 2408 (1994).
(31) B. Scholten, S. M. Qaim, and G. Stöcklin, Radiochim. Acta **65**, 81 (1994).
(32) S. Sudár, F. Szelecsényi, and S. M. Qaim, Phys. Rev. C**48**, 3115 (1993).
(33) S. Sudár and S. M. Qaim, Phys. Rev. C**53**, 2885 (1996).
(34) B. Strohmaier, M. Fassbender, and S. M. Qaim, Phys. Rev. C**56**, 2654 (1997).
(35) S. M. Qaim, S. Sudár, and A. Fessler, Radiochim. Acta **93**, 503 (2005).
(36) S. Sudár, and S. M. Qaim, Phys. Rev. C**73**, 034613 (2006).