Comparison between Theoretical Calculation and Experimental Results of Excitation Functions for Production of Relevant Biomedical Radionuclides

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Abstract. The radionuclide production for biomedical applications has been brought up in the years, as a special nuclear application, at INFN LASA Laboratory, particularly in cooperation with the JRC-Ispra of EC. Mainly scientific aspects concerning radiation detection and the relevant instruments, the measurements of excitation functions of the involved nuclear reactions, the requested radiochemistry studies and further applications have been investigated. On the side of the nuclear data evaluations, based on nuclear model calculations and critically selected experimental data, the appropriate competence has been developed at ENEA Division for Advanced Physics Technologies. A series of high specific activity accelerator-produced radionuclides in no-carrier-added (NCA) form, for uses in metabolic radiotherapy and for PET radiodiagnostics, are investigated. In this work, last revised measurements and model calculations are reviewed for excitation functions of natZn(d,X)⁶⁴Cu,⁶⁶Ga reactions, referring to irradiation experiments at K=38 variable energy Cyclotron of JRC-Ispra. Concerning the reaction data for producing ¹⁸⁶⁰Re and ²¹¹⁴At/²¹¹⁰Po (including significant emission spectra) and ²¹⁰⁵At, most recent and critically selected experimental results are considered and discussed in comparison with model calculations paying special care to pre-equilibrium effects estimate and to the appropriate overall parameterization. Model calculations are presented for ²²⁹⁰Ra(p,2n)²²⁸Ac reaction, according to the working program of the ongoing IAEA CRP on the matter.

INTRODUCTION

Results are reviewed concerning comparison of experimental values and model calculations by the ongoing activities on accelerator-produced high specific activity radionuclides in no-carrier-added (NCA) form. The following radionuclides, according to Table 1 planning, are reviewed: ⁶⁴Cu produced by natZn(d,αxn) and natZn(d,2p) nuclear reactions, for simultaneous positron/negatron metabolic radiotherapy and PET imaging; ⁶⁶Ga high-energy positron emitter (4.2 MeV), produced by the natZn(d,xn) reaction, for metabolic radiotherapy and PET; ¹⁸⁶⁰Re produced by ¹⁸⁶⁰W(p,n) and ¹⁸⁶⁰W(d,2n) reactions, for negatron (1.1 MeV) metabolic radiotherapy; ²¹¹⁴At/²¹¹⁰Po produced by the ²⁰⁹Bi(α,2n) reaction, internally spiked with the γ emitter ²¹⁰⁵At, produced by ²⁰⁹Bi(α,3n) reaction, and ²²⁵Ac/²¹⁢Bi/²¹³Po for high-LET metabolic radiotherapy.

COOPERATION ASPECTS REFERRING TO INTERNATIONAL INITIATIVES ON NUCLEAR DATA

Recognizing the main relevance of excitation-function determination for the production of medical radionuclides, a special effort has been brought up on the matter over the years, as e.g., from [1-8], and it is going on presently at the INFN-LASA Laboratory, taking advantage of the long-time experience in the experimental nuclear physics and nuclear chemistry fields. Recently, collaboration on the matter with ENEA–Division for Advanced Physics Technologies (FIS) has been intended to compare experimental results of main relevance with predictions from model calculations. Relevant activities have been referred to the international initiatives by the IAEA regarding medical radionuclide production aspects and...
particularly the related nuclear data, by participating with significant results in Specialists’ and Advisory Meetings [9] and the Coordinated Research Projects (CRP), on nuclear data relevant to radioisotopes for SPECT and PET (fulfilled) and for radiotherapy (ongoing). The results presented in this paper are especially referred to the ongoing CRP. More generally, the present matter has been referred to the joint effort devoted in the international context, both by the IAEA and the OECD-NEA, to the nuclear data evaluations, including measurement analyses and model calculations, and to their exchange in computerized files, according to the needs recognized by the NEA-Nuclear Science Committee and by the IAEA-International Nuclear Data Committee [10]. Concerning the experimental values of the excitation functions here considered, they have been deduced from the analyses of irradiation experiments at the MC40 cyclotron of JRC-Ispra for the deuteron-induced reactions on the natZn target [9], or they are taken from the literature [11-17], for the (p,n) and for the (d,2n) reactions on 186W target, respectively, and for the 209Bi(α,2n) production reactions concerning the 211At therapeutic radionuclide. Those experimental data are presented in comparison with the results obtained by model calculations. Moreover, the experimental thick target yield obtained from the first irradiation experiment at Ispra cyclotron for the same production reaction is given for 28.8 ± 0.2 MeV incident alpha energy, in comparison with the integrated value obtained from the calculated cross section data. For the 226Ra(p,2n) reaction cross section, only model calculations have been performed, as proposed at the above ongoing IAEA-CRP, for lack of experimental data. From those theoretical results graphically presented in the following, the considered reaction appears to be confirmed as a promising production route of 225Ac. Then, it is desirable to obtain detailed experimental data in the near future even if the measurements are extremely difficult, due to the irradiation of radioactive 226Ra target and the emission of gaseous 222Rn and its daughters.

NUCLEAR MODELS AND COMPUTING CODES

The high relevance of the nuclear model calculations is well recognized in the framework of nuclear data evaluation activities within the NEA and IAEA context and a number of initiatives on the matter have been undertaken, both for the validation of the computing codes with respect to measured values and for the model parameterization and systematics, as outlined in particular in [9], aimed at reliable data calculations also in case of discrepant, scarce, or lacking measurements. In this work, calculations for the involved nuclear reactions have been carried out at ENEA Division FIS, through internally developed codes and the EMPIRE-II system [18], accounting for the major nuclear reaction mechanisms, including the Optical Model (OM), the Multi-Step Compound and Exciton models and the full featured Hauser-Feshbach model, with a comprehensive parameter library mainly covering nuclear masses, OM data, discrete nuclear levels, and decay schemes. Particularly the Monte Carlo Pre-equilibrium approach has been successful in approximating the experimental values. As the determination of the nuclear levels density is of main impact on the results, the approach previously presented [19] was considered, taking into account the dependence of the crucial “level density parameter” on the nuclear excitation energy.
RESULTS AND RELEVANT COMMENTS

First results have been obtained by the work underway, according to the scheme in Table 1. Significant examples are reviewed in Figs. 1-5, respectively:

- the cumulative excitation functions for the production of \(^{64}\text{Cu}\) and \(^{66}\text{Ga}\) by nuclear reactions on a natural zinc target in the energy range up to 19 MeV, such as \((d,\alpha xn)\) reactions for \(^{64}\text{Cu}\) production, \((d,2p)\) reactions for \(^{64}\text{Cu}\) production, and \((d,xn)\) reactions for \(^{66}\text{Ga}\) production. The experimental values, obtained from the analyses of the above-mentioned irradiation experiments, are consistently compared with the model calculations (full lines), obtained as described above, for \(^{64}\text{Cu}\) and \(^{66}\text{Ga}\) production reactions in the incident energy intervals from thresholds up to 19 MeV;

- the excitation function for the \((p,n)\) and \((d,2n)\) reactions on a \(^{186}\text{W}\) target for the production of \(^{186}\text{gRe}\) in the incident energy interval up to 20 MeV; the experimental values taken from the literature [11-14] are compared with the theoretical ones (full lines), obtained through the EMPIRE-II code, including the Monte Carlo Pre-equilibrium approach for the \((p,n)\) reaction, but not the possible direct-reaction effects. New measurements appear to be highly desirable in order to reduce the present experimental uncertainties. These production methods can provide NCA \(^{186}\text{gRe}\) in comparison with the CA production route through a reactor neutron field by \((n,\gamma)\) reactions on an enriched \(^{185}\text{Re}\) target.

- the experimental cross-section data for \((\alpha,2n)\) reactions on a \(^{209}\text{Bi}\) target for the production of \(^{211}\text{At}\) in the incident energy interval from 20 MeV up to 45 MeV; the experimental values from the literature [15-17] are compared with the theoretical ones (full line) obtained by nuclear model calculations through the EMPIRE-II code. The thick target yield value obtained by integration of the theoretical curve, from 28.8 down to 20 MeV, is 9234 MBq·C\(^{-1}\), in comparison with the experimental value of total energy absorption equal to 8085 ± 176 MBq·C\(^{-1}\) obtained at LASA from a preliminary irradiation: a discrepancy of about 12.4% appears to be reasonable with regard to the present experimental conditions.
FIGURE 4. Experimental cross-section data taken from the literature [15-17] for the production of $^{211}$At via $(\alpha,2n)$ reactions on a $^{209}$Bi target and the corresponding theoretical results by model calculation (full line).

FIGURE 5. Theoretical prediction by nuclear models of cross-section data for the production of $^{225}$Ac via (p,2n) reactions on a $^{226}$Ra target.

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REFERENCES


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