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Investigation of activation cross-sections of alpha-induced nuclear reactions on natural cadmium

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ABSTRACT

We measured production cross-sections of Sn, In, and Cd radionuclides from alpha-induced reactions on ^{nat}Cd from their respective threshold to 45 MeV by using a stacked-foil activation technique at the MC-50 cyclotron of the Korea Institute of Radiological and Medical Sciences. The results were compared with the earlier measurements as well as with the theoretical values obtained from the TENDL-2012 library based on the TALYS 1.4 code. Our measurements for the ^{110,113g,117m}Sn, ^{108m,108g,109g,110m,110g,111g,113m,114m,115m,116m,117m,117g}In, and ^{111m,115g}Cd radionuclides in the energy region from the threshold energy to 45 MeV are in general good agreement with the other experimental data and calculated results. The integral yields for thick target were also deduced using the measured cross-sections and the stopping power of natural cadmium target and found in agreement with the directly measured yields available in the literature. The measured cross-sections find importance in various practical applications including nuclear medicine and improvement of nuclear model calculations.

1. Introduction

Accurate knowledge of light charged particle-induced reaction cross-sections has generated significant interest in the nuclear data community because these reactions are being increasingly used in nuclear medicine, accelerator and nuclear technology, and the testing of nuclear reaction theories. Recently, there are some measurements for the alpha-induced reactions cross-sections on various target elements, because the measured data of the (α,x) processes for different elements are scanty, relative to those of (p,x) processes.

Cadmium (Cd) has a great importance as target material for the production of radionuclides used in medical and industrial applications. Several indium radionuclides are known for potential applications in the diagnostic and therapeutic procedures due to their suitable decay characteristics [1–6]. The relatively short-lived β^+-emitters ^{108m}In ($T_{1/2}$ = 39.6 min), ^{108g}In ($T_{1/2}$ = 58.0 min), and ^{109g}In ($T_{1/2}$ = 4.167 h) are promising candidates for the positron emission tomography (PET). The ^{110m}In ($T_{1/2}$ = 69.1 min, $E^{mean}_{\beta^+} = 1011$ keV, $I^{total}_{\beta^+} = 61.3\%$) could be obtained in pure form via a ^{110}Sn→^{110}In generator system [7,8] and potentially be applied for labeling proteins and peptides in PET imaging procedures [9,10]. The ^{111}In is widely used in diagnostic nuclear medicine and internal radiotherapy due to its favorable half-life ($T_{1/2}$ = 2.8047 d), high intense low energy γ-ray emission (E_γ = 171.28 keV (90.7%), E_γ = 245.35 keV (94.1%)), and abundance of β^- emission (2.72 keV L-Auger line (100.4%); 19.3 keV K-Auger line (15.5%)) [11–13]. It is also used for labeling cellular blood components and monoclonal antibodies, radiolabeled immunoglobulin therapy, and for cancer imaging [3,5,14–16]. Moreover, the ^{111}In and its decay product ^{111m}Cd play a great role in γ–γ time differential perturbed angular correlation (TDPAC) studies which enables to explore properties of material in solid, fluid, and gaseous states [17,18]. The ^{111g}In also shows a great importance as marker in wear and corrosion measurements of indium alloys or implants ^{111}In in plastic for wear measurement by thin layer activation technique [19]. The ^{113m}In ($T_{1/2}$ = 99.476 min) produced via a ^{113}Sn/^{113m}In generator system [20] finds potential applications in diagnostic nuclear medicine [21], in internal radio-therapy [22], and as a tracer radionuclide in industrial activities [23]. The relatively long-lived ^{114m}In ($T_{1/2}$ = 14.51 d) and its decay product ^{114g}In ($T_{1/2}$ = 71.9 s, $E^{mean}_{\beta^-} = 777.9$ keV, $I^{total}_{\beta^-} = 99.5\%$) find importance as a low-energy electron emitter (2.84 keV L-Auger line (65%); 20.1 keV K-Auger line (5.98%)) for radio-immunotherapy [12,24]. There is an increasing interest in studying ^{114m}In to

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determine the long-term stability and bio kinetics of indium-labeled pharmaceuticals [25].

The favorable decay characteristics ($T_{1/2}$ = 14.0 d, low energy Auger electron: Auger-L 2.95 keV (92.8%); Auger-K 21.0 keV (10.8%), and conversion electron: *CE*-K 126.82 keV (65.7%); *CE*-L 151.56 keV (26.57%)) make it possible to use ^{117m}Sn as a high linear energy transfer (LET) radioisotope for the prognosis and palliative treatment of metastatic bone cancer [26–28]. The ^{117m}Sn with a single γ emission of 158.56 keV (86.4%) can be used as an excellent bone imaging agent, enabling accurate quantification of total-body uptake and retention as well as uptake and retention by metastatic lesion and normal bone [29].

A general survey of the literature reveals that only few earlier investigations [30–35] were carried out for the production of aforementioned medically and technologically important radionuclides via alpha particle irradiations on natural and/or enriched cadmium targets but considerable discrepancies are found among them. In general, discrepancies among the different measurements may come from the use of different monitor reactions and/or inappropriate measurement parameters. Therefore, new experimental data are required to reduce the discrepancies and also to complement the data needed to optimize the production of the medically important radionuclides. Recognizing the importance of the measured data in various practical applications we investigated activation cross-sections of the ^{nat}Cd(α,x)^{110,113g,117m}Sn, ^{108m,108g,109g,110m,110g,111g,113m,114m,115m,116m,117m,117g}In and ^{111m,115g}Cd nuclear reactions by using an Azimuthally Field Varying (AVF) cyclotron at the Korea Institute of Radiological and Medical Sciences (KIRAMS). We also deduced the integral yields for thick targets of the investigated radionuclides from their respective threshold up to 45 MeV.

2. Experimental procedures

A well-established activation technique combined with HPGe γ-ray spectrometry was employed to determine production cross-sections of residual radionuclides by irradiating natural cadmium metallic foils with alpha particles. The irradiation technique, activity determination, and data evaluation procedures were similar as described in our previous works [36–43]. Therefore, only the features that are important for an assessment of the procedures relevant to the present investigations are presented here. A high-purity (99.7%) metallic form of cadmium foil (10-μm thickness) having natural isotopic compositions (^{106}Cd 1.25%, ^{108}Cd 0.89%, ^{110}Cd 12.49%, ^{111}Cd 12.80%, ^{112}Cd 24.13%, ^{113}Cd 12.22%, ^{114}Cd 28.73%, and ^{116}Cd 7.49%) [44] was used as a target material. Natural copper (99.9% purity, 10-m thickness) and natural aluminum (99.999% purity, 25-m thickness) foils were also assembled in the stack to be used as a monitor and an energy degrader, respectively. All foils were individually weighed before irradiation for accurate determination and/or cross checking of the foil thickness. The stacked-foils were irradiated by a 45 MeV alpha beam, collimated to 10 mm in diameter, and about 70 nA beam current for 60 min in the external beam line of the MC-50 cyclotron at the KIRAMS. The beam intensity was kept constant during the irradiation.

After irradiations and an appropriate cooling time, the activated foils were removed and measured by using an n-type coaxial HPGe γ-ray spectrometer. The HPGe-detector was coupled to a 4096 multi-channel analyzer with the associated electronics to determine the photo-peak area of the γ-ray spectrum by using the Gamma Vision 5.0 (EG&G Ortec) program. The energy resolution of the detector was 1.90 keV full width at half maximum (FWHM) at the 1332.50 keV peak of ^{60}Co. The photopeak efficiency curve of the γ-ray spectrometer was determined at the different sample measuring distances using a set of standard point sources. The activity measurements of the irradiated samples were started at about 50 min after the end of the bombardment (EOB). The measurements of the activated targets and monitor foils were repeated several times to follow the decay of the radionuclides and thereby to identify the possible interfering nuclides.

The IAEA recommended monitor reactions ^{nat}Cu(,x)^{66}Ga (σ_{max} = 133.84 mb at E_α = 42.1 MeV) and ^{nat}Cu(,x)^{67}Ga (σ_{max} = 52.94 mb at E_α = 42.1 MeV) [45] were used to determine the beam intensity and cross-checking, respectively. The beam intensity was considered as constant to deduce cross-sections for each foil in the stack. The alpha energy degradation along the stacked foils was calculated by using the computer program SRIM-2003 [46].

The cross-sections were determined using the well-known activation formula [40–41]. The decay data relevant for the cross-sections determination were taken from the ENSDF evaluation [47–55] obtained from the NuDat-2.6 library [56], and are summarized in Table 1. The Q-values and threshold energies calculated on the basis of the atomic mass evaluation by Audi and Meng [57] and the Q-tool system [58] are also presented in Table 1. Intense and independent characteristic γ-lines were used to quantify the radionuclides. In some cases, two or more characteristic γ-rays were used to check the obtained results.

The uncertainty of the alpha energy for each representing energy point in the stack depends on the irradiation circumstances and the position of the foil in the stack. These are due to the initial beam energy, the thickness and homogeneity of target foils, and the beam straggling. The estimated uncertainty of a representing point in the excitation function ranges from ±0.7 up to ±2.3 MeV, which is shown in tables and figures.

Total uncertainties of cross-sections were obtained according to the rules of error propagation assuming that all uncertainties are independent. Moreover, some of the sources of uncertainties are common to all data, while others affect each reaction or energy point individually. However, the combined uncertainty in each cross-section was estimated by considering the following uncertainties; statistical uncertainty of the γ-ray counting (0.5–10%), uncertainties in the alpha beam intensity (~6%), in the efficiency calibration of the detector (~4%), due to the sample thickness (~1%), and in gamma intensity (~1%). The overall uncertainties of the measured cross-sections were in the range of 7–12%.

3. Theoretical data from model calculations

Comparison of reliable experimental cross-sections with model calculated ones, without any adjustment of model parameters, may reveal information on accuracies or deficiencies of various reaction mechanisms included in the conventional reaction models. Therefore, we compare our experimental production cross-sections with the TENDL-2012 library [59], which compiles reaction cross-sections based on the nuclear model code TALYS 1.4 [60].

4. Results and discussion

Independent and cumulative cross-sections of the ^{nat}Cd(α,x)^{110,113g,117m}Sn, ^{108m,108g,109g,110m,110g,111g,113m,114m,115m,116m,117m,117g}In and ^{111m,115g}Cd reactions are shown in Figs. 1–17 together with the experimental data available in the EXFOR library [61] and also with theoretical data in the TENDL-2012 library [59]. Since natural cadmium consists of eight stable isotopes, many reaction channels can contribute to the production of a specific radionuclide. The formation of a particular radionuclide via the contribution of these channels only is referred to as 'independent' production. On the other hand, formation of a particular radionuclide via the contribution of direct channels plus decay of isomers and/or precursors is referred to as 'cumulative' production. The measured cross-sections are also tabulated in Tables 2(a) and 2(b) together with their