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Proton induced nuclear reactions on cadmium up 17 MeV

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ABSTRACT

The cross-sections of proton induced reactions on ^{nat}Cd targets was studied in the energy range from threshold up to 17 MeV, using a stacked-foil irradiation technique and classical gamma-spectroscopy. We measured the formation cross-sections of the radioisotopes ^{109g,110m,110,111g,113m,114m,115m,116m}In The obtained excitation functions were compared with the earlier published data and the theoretical model calculations by the codes ALICE-IPPE, EMPIRE and TALYS .

INTRODUCTION

Nuclear reactions have various practical applications in science and technology. Cyclotrons, accelerator and nuclear reactors are commonly employed to produce radionuclides of interest. The physical basis of a radionuclide production is optimized by using data on nuclear cross-sections. Therefore, among the various types of nuclear data, the cross-sections have great significance in the production and the quality control of desired radionuclides. Proton-induced reaction cross-sections for cadmium are important for various practical applications; medical radionuclide production, thin layer activation (TLA) analysis, dosimetry application, nuclear technology, radioactive waste handling and so on Tarkanyi et al (2006).

Cadmium (Cd) is a soft, malleable, ductile, toxic and bluish-white bivalent metal. It is largely used in batteries and pigments, for example in plastic products. It is an ideal target material for the production of medically important radionuclides (for example, ^{109,110,111,114m}In). The radionuclide ^{111}In is widely used in diagnostic nuclear medicine due to its suitable half-life (T1/2 = 2.8 d), abundance of β-emission and high-intense low-energy γ-ray emission. It is also used for labeling cellular blood components and monoclonal antibody, a myocardial damage detection, a localization of an abscess in polyeystic kidney,

radiolabeled immunoglobulin therapy and an imaging for cancer. The short-lived radionuclides ^{110m}In (T1/2 = 69.1 min), and ^{109}In (T1/2 = 4.2 h) have a potential interest in positron emission tomography (PET) studies. It is interesting to note that, ^{114m}In and its daughter radionuclide ^{114}In are usually regarded as undesirable long-lived impurities in ^{111}In-labeled radiopharmaceuticals for a diagnostic use. However, there is increasing interest in studying ^{114m}In to determine the long-term stability and bio kinetics of indium-labeled pharmaceuticals as well as for radionuclide therapy at a low-energy.

EXPERIMENTAL

Cross-sections were measured by the stacked-foil irradiation technique, which involves the simultaneous irradiation of a set of thin foils. The amounts of the produced radionuclides were determined by the activation method, by following the decay of the radioactive products. The details have been described by the authors in publications (Tárkányi et al. 2010, Al-Abyad et al. 2010). Here we give only some salient features relevant to the present measurements.

Samples and irradiations

Sample foils were high-purity thin Cd foils (99.9%, 10 μm) supplied by Goodfellow, England. The stack was irradiated with protons, in the stack Cd foils were placed together with Cu(10 μm) monitor foils. The monitor foils served to degrade the projectile energy as well as to determine the incident particle energy and the beam intensity. the stack was irradiated at 17 MeV primary beam energy for one hour with about 150 nA beam intensity. The irradiations were performed in a Faraday cup like target holder (Tárkányi et al., 2002), having secondary electron suppressor at the external beam of the MGC-20 cyclotron in ATOMKI, Debrecen.

Activity measurement

A HPGe detector was used to measure the activities of the irradiated samples and the monitor foils. The irradiated samples and monitor foils were measured using a HPGe detector. The resulting spectra were analysed using the software GammaVision,version5.10,supplied by EG&G ORTEC (USA). The counting time was adjusted according to the half-life of the product nuclide to get reasonable counting statistics. All the major gamma lines of the resulting radionuclides were identified and the decay was followed. The peak areas were obtained from the spectra that had been taken after suitable decay time.

2.Data analysis

2.1. Calculation of the beam intensity and the energy degradation

The beam intensity was determined from the activities induced in the titanium monitor foils at front of stack and between the target samples. The monitor foils were irradiated simultaneously with the sample foils and measured with the same detector in a comparable geometry. The beam parameters were first estimated from the radiofrequency of the cyclotron (beam energy) and from the measured values of the current integrator in the Faraday cup (beam current), then it was adjusted taking into account a thorough analysis of the excitation function of the monitor reaction natCu(p,x)65Zn measured over the whole energy range and compared with the recommended values from the IAEA-TECDOC-1211, (Tárkányi et al. 2001).

2.1.Calculation of cross-sections and their uncertainties

The count rates at the End of Bombardment (EOB) were converted to activities by introducing corrections for emission probabilities of γ-rays, detector efficiency, coincidence loss, dead time and random pile-up. The decay and spectrometric characteristics of the investigated radionuclides were taken from the NUDAT 2.5 database presented in Table (1). The Q-values refer to the formation of the ground and isomeric states were obtained from Pritychenko et al. 2011. Some of the radionuclides formed are result of cumulative processes, where apart from direct activation decay of either parent nuclides or meta-stable states can contribute. The uncertainties on the cross-sections were estimated by using the error propagation through the formula used for calculation (Guide of uncertainties, 1993). The total uncertainty of the cross-section values was mostly in the range of 11–17%.

Table (1) Decay data of the product nuclides used in the present study.

Nuclei	Half-life $T_{1/2}$	Decay mode (%)	E_γ (keV)	I_γ (%)	Contributing reactions	Q-value (MeV)	Threshold (MeV)
^{109g}In	4.2 h	EC(100)	203.50 426.25 623.70 1149.10	73.5 4.12 5.5 4.3	^{110}Cd(p, 2n)	-12.72	12.83
					IT decay of ^{109m}In		
^{110g}In	4.9 h	EC(100)	641.68 657.76 707.40 884.68 937.50	25.9 98.3 29.5 92.9 68.4	^{110}Cd(p, n) ^{111}Cd(p, 2n)	-4.66 -11.64	4.70 11.74
^{110m}In	69.1 min	β^+ +EC(100)	657.76	98.0	^{110}Cd(p, n) ^{111}Cd(p, 2n)	-4.66 -11.64	4.70 11.74
^{111g}In	2.81 d	EC(100)	171.28 245.39	90.0 94.0	^{111}Cd(p,n) ^{112}Cd(p,2n)	-1.64 -11.04	1.66 11.14
					IT decay of ^{111m}In		
^{113m}In	1.658 h	IT(100)	391.69	64.20	^{113}Cd(p, n) ^{114}Cd(p,2n)	-0.46 -9.5	0.47 9.59
^{114m}In	49.51 d	IT + EC(100)	190.29 558.46	14.74 4.0	^{114}Cd(p,n)	-2.23	2.25
^{115m}In	4.48 h	IT + β^-(100)	336.24	45.83	^{116}Cd(p, 2n)	-8.04	8.11
					β^- decay of ^{115g}Cd		
^{116m1}In	54.29 min	β^-(100)	1097.33 1293.56	56.20 84.40	^{116}Cd(p, n)	-1.25	1.26

4. Nuclear model calculations

With a view to validating the data and testing the predictive power of the nuclear theory, reaction cross-sections were calculated theoretically using the nuclear model codes ALICE-IPPE, EMPIRE and TALYS.

RESULTS AND DISCUSSION

Cross-sections

The experimental reaction cross-sections are plotted as a function of proton energy in Figs. 1-8 in comparison with the results of nuclear model calculations as well as literature experimental data.

^{nat}Cd(p,xn)^{109}In reaction

The nuclide ^{109}In has one long-lived ground state radionuclide ^{109g}In

(T1/2 = 4.20 h) and two short-lived metastable states ^{109m}In (T1/2 = 1.34 min) and ^{109m1}In (T1/2 = 0.21 s). These two metastable radionuclides completely decay to a ground state ^{109g}In by 100% IT process. As the γ-ray counting process was started after a cooling time of about 1.5 h, the measured production cross-sections of the ^{109g}In radionuclide are considered as cumulative ones. Only the 203.5 keV high-intense γ-line was used to identify the ^{109g}In radionuclide. The competing reaction channel for the formation of this radionuclide is: ^{110}Cd (p,2n).

The measured excitation function of the ^{109g}In is shown in Fig. 2 in comparison with the literature data and the theoretical prediction by the theoretical codes. The measured data are in general good agreement with the reported data of Nortier et al. (1990), Kormali et al. (1976) Tárkányi et al. (2006) and Khandaker et al. (2008). The excitation function for $^{109m+g}$In predicted by the ALICE-IPPE code and data from the TALYS and EMPIRE codes are slightly higher than the experimental data in the whole investigated energy region.

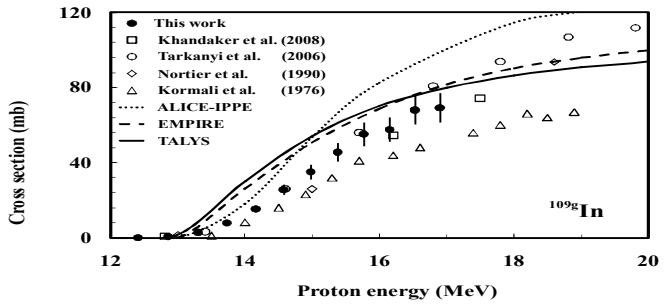

Fig.(1). Excitation function of the ^{nat}Cd(p,xn)^{109g}In reaction (experimental and theoretical results).

^{nat}Cd(p,xn)^{110m,g}In reaction

The ^{110}In radionuclide has two long-lived states, the ground state ^{110g}In (T1/2 = 4.9 h) and its isomeric state ^{110m}In (T1/2 = 69.1 min), decay independently to the stable ^{110}Cd isotope by the EC process. The ^{110g}In radionuclide was produced by the competing direct reactions of ^{110}Cd(p,n), ^{111}Cd(p, 2n) in the investigated proton energy range. The ^{110g}In radionuclide was identified by using its several strong independent γ-lines of 707.4, 884.685 and 937.50 keV. The measured excitation function of ^{110g}In is compared with the available literature data and the calculated values of the theoretical codes as shown in Fig. 3.

The data reported by Khandaker et al. (2008), Al-Saleh et al. (2008) and Tárkányi et al. (2006) showed in general good agreement with the present result. The measured excitation function revealed an excellent agreement with the EMPIRE code but TALYS code is slightly lower in the investigated energy region.

The ^{110m}In radionuclide was identified by using its strong independent γ-line of 657.76 keV. The measured excitation function of ^{110m}In is compared with the available literature data and the calculated values of the theoretical codes as shown in Fig. 4.

The present results of the excitation function of ^{110m}In are in good agreement with the data of Tárkányi et al. (2006) in the whole energy range but the data reported by Nortier et al. (1990) and Kormali et al. (1976) , are too low while Sakakun et al. (1975) agree with the present work up to12 MeV and then fall down.

The present experimental data are in good agreement with results of the nuclear model codes ALICE-IPPE, EMPIRE and TALYS

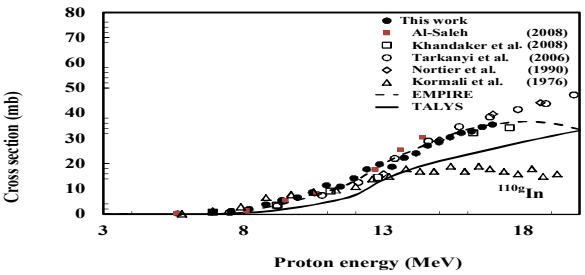

Fig.(2) Excitation function of the ^{nat}Cd(p,xn)^{110g}In reaction (experimental and theoretical results).

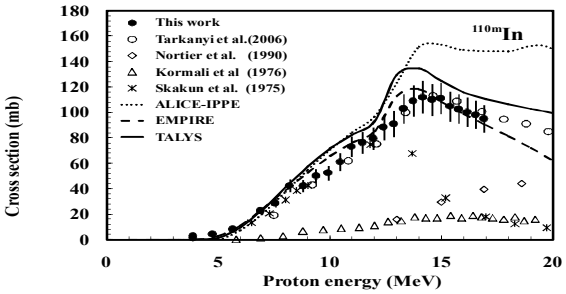

Fig.(3) Excitation function of the ^{nat}Cd(p,xn)^{110m}In reaction (experimental and theoretical results).

$^{nat}Cd(p,xn)^{111g}In$ reaction

The radionuclide ^{111}In has a short half life (7.7 min) isomeric state decaying completely to the long-lived (see Table 1) ground state by IT. Our cumulative values were measured after complete decay of the metastable state. The cross-sections on ^{nat}Cd target were investigated by Nortier etal. [5],in abroad energy range. The contributing reactions in the investegated energy range are ^{111}Cd(p,n) and ^{112}Cd(p,2n). In Fig.5 we present the excitation function in comparison with the earlier data and the theoretical estimation. The measured data are in good agreement with the reported data of Nortier et al. (1990), Tárkányi et al. (2006), Khandaker et al. (2008) and Al-Saleh et al (2008). The excitation function for $^{109m+g}$In predicted by the ALICE-IPPE code and data from TALYS and EMPIRE codes are in good agreement with the experimental data in the whole investigated energy region.

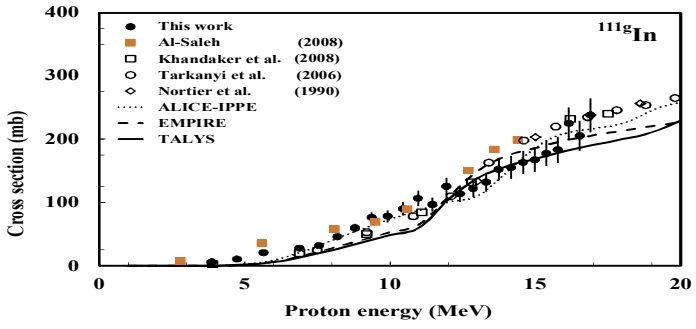

Fig.(4) Excitation function of the ^{nat}Cd(p,xn)^{111g}In reaction (experimental and theoretical results).

$^{nat}Cd(p,xn)^{113}In$ reaction

The metastable radionuclide ^{113m}In (T1/2 = 1.66 h) completely decays to the stable ^{113}In by the IT process. The ^{113m}In is produced mainly by ^{113}Cd(p,n) and ^{114}Cd(p,2n) reactions in our investigated energy region. It can be identified by using a 391.7 keV γ-line. The measured excitation function is shown in Fig. 6 together with the available literature data and the theoretical codes. The present excitation function is in good agreement with the data reported by Tárkányi et al. [1], Kormali et al. [15], Al-Saleh [24] and Khandaker et al. (2008) and also with the TALYS and EMPIRE predictions.

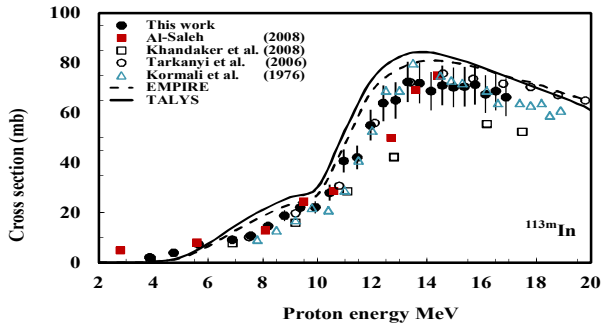

Fig.(5) Excitation function of the ^{nat}Cd(p,xn)^{113m}In reaction (experimental and theoretical results).

^{nat}Cd(p,xn)^{114m}In reaction

Except for the short-lived ground state radionuclide ^{114g}In (T1/2 =1.20 min), the ^{114}In has two isomeric states ^{114m}In (T1/2 = 49.51 d) and ^{114m1}In (T1/2 = 0.0431 s). Under the circumstances of the present experimental conditions, we could only measure the production of the ^{114m}In (T1/2 = 49.51 d) radionuclide. The contributing reactions to the formation of this radionuclide are the ^{114}Cd(p, n) process. The measured data is shown in Fig. 5 together with the available literature data and the theoretical data obtained by the TALYS, EMPIRE and the ALICE-IPPE codes. The present result showed in general good agreement with the previous data obtained by Nortier et al. (1990), Tárkányi et al. (2006) and Khandaker et al. (2008)

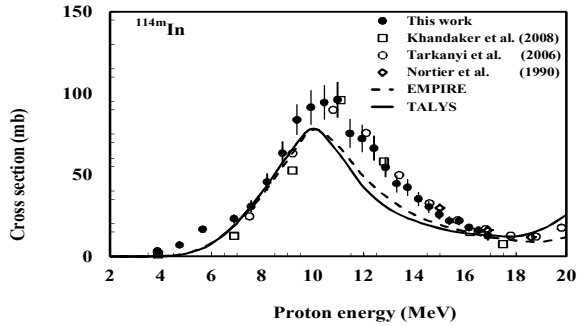

Fig.(6) Excitation function of the ^{nat}Cd(p,xn)^{114m}In reaction (experimental and theoretical results).

^{nat}Cd(p,xn)^{115m}In reaction

The metastable radionuclide ^{115m}In (T1/2 = 4.48 h) decays to the stable ground state of ^{115}In by the IT (95%) and the β^-(5%) processes, and it was

identified by the 336.24 keV γ-line. The production of the ^{115m}In is contributed by not only the direct reaction presented in Table 1 but also the decay of the ^{115g}Cd nuclide. Therefore, the present measured cross-section is a cumulative one. The measured excitation function of this radionuclide is shown in Fig. 8 together with the previous reported one and the calculated one by the TALYS and EMPIRE codes. We found a good agreement with the data reported by Tárkányi et al. (2006) and Khandaker et al. (2008). The TALYS and EMPIRE codes produce a nicely fitted excitation function to the measured one.

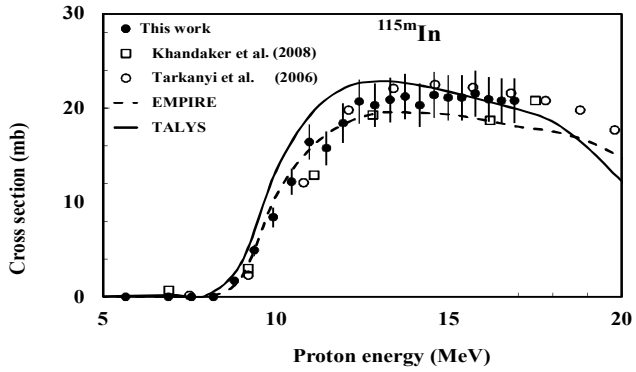

Fig.(7) Excitation function of the ^{nat}Cd(p,xn)^{115m}In reaction (experimental and theoretical results).

^{nat}Cd(p,xn)^{116m}In reaction

The ^{116}In radionuclide has a short-lived ground state ^{116g}In (T1/2 =14.1 s) and two metastable states: ^{116m1}In (T1/2 = 54.29 min) and ^{116m2}In (T1/2 = 2.18 s). The present experimental condition only permits us an identification of the ^{116m1}In radionuclide. The production of this radionuclide is due to the combined contributions from the direct ^{116}Cd(p, n) reaction and the IT decay of the short-lived isomeric state ^{116m2}In (T1/2 = 2.18 s). The measured excitation function is shown in Fig. 8 together with the available literature data and the theoretical data obtained by the TALYS and the ALICE-IPPE codes. The present result agreed with the reported data of Tárkányi et al. (2006) and Khandaker et al. (2008).except for around the 10-MeV region. Al-Saleh (2008) reported scattered data showed larger magnitudes than all other measurements by irradiation on the natural cadmium target. The theoretical data calculated by the TALYS and EMPIRE codes are consistent with the measured data.

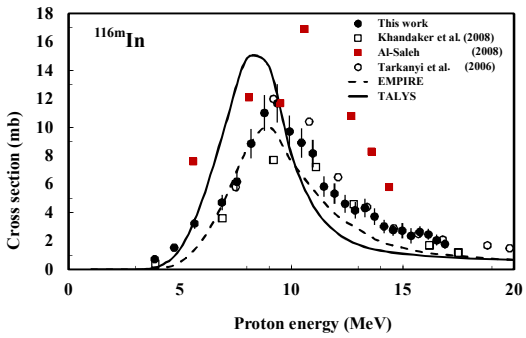

Fig.(8) Excitation function of the ^{nat}Cd(p,xn)^{116m}In reaction (experimental and theoretical results).

REFERENCES

1. Al-Abyad, M., Spahn, I., Qaim, S.M., (2010): Experimental studies and nuclear model calculations on proton induced reactions on manganese up to 45 MeV with reference to production of ^{55}Fe, ^{54}Mn and ^{51}Cr. Appl. Radiat Isot. 68 , 2393–2397.
2. Al-Saleh, F.S. (2008): Cross sections of proton induced nuclear reactions on natural cadmium leading to the formation of radionuclides of indium, Exfor data base
3. EXFOR, 2007. Nuclear reaction data, EXFOR is accessed on line at http://www.nndc.bnl.gov/exfor/exfor.htm.
4. Firestone, R.B., (1998): Table of Isotopes. John Wiley and Sons, Inc., New York, USA.
5. Guide to Expression of Uncertainty in Measurements, ISO Geneva, 1993, ISBN 92-10188-9.
6. Herman, M., Capote, R., Carlson, B.V., Obložinský, P., Sin, M., Trkov, A. Wienke, H. Zerkin, V., (2007): EMPIRE: nuclear reaction model code system for data evaluation. Nuclear Data Sheets, 108, 2655-2715.
8. Khandaker , M.U. K. Kim , M.W. Lee, K.S. Kim , G.N. Kim , Y.S. Cho , Y.O. Lee Production cross-sections for the residual radionuclides from the natCd(p, x) nuclear processes Nuclear Instruments and Methods in Physics Research B 266 (2008) 4877–4887
9. Koning, A.J., Hilaire, S. and Duijvestijn, M.C., (2008): TALYS-1.0", Proceedings of the International Conference on Nuclear Data for Science and Technology, April 22-27, 2007, Nice, France, editors O.Bersillon, F.Gunsing, E.Bauge, R.Jacqmin, and S.Leray, EDP Sciences, 2008. DOI: 10.1051/ndata:07767.
10. Kormali, S.M., D.L. Swindle, E.A. Schweikert, Charged particle

activation of medium Z elements. II. Proton excitation functions, J. Radioanal. Chem. 31 (1976) 437.

11. Lederer, C.M., Shirley, V.S., (1978): Table of Isotopes. John Wiley and Sons, Inc., New York, USA.
12. Nortier, F.M., S.J. Mills, G.F. Steyn. Excitation functions and production rates of relevance to the production of ^{111}In by proton bombardment of ^{nat}Cd and ^{nat}In up to 100 MeV, Appl. Radiat. Isot. 41 (1990) 1201.
13. NUDAT2 Database, <http://www.nndc.bnl.gov/hbin/nudat>.
14. Pritychenko, B., Sonzogni, A. . 2011. Q-value calculator, NNDC, Brookhaven National Laboratory. http://www.nndc.bnl.gov/qcalc.
15. Skakun, E.A. , A.P. Kljucharev, N. Rakivnenko, I.A. Romanij, Excitation functions of (p, n) and (p, 2n) reactions on cadmium isotopes, Izv. Rossiiskoi Akademii Nauk, Ser. Fiz. 39 (1975) 24; Skakun, E.A. A.P. Kljucharev, N. Rakivnenko, I.A. Romanij, English translation, Bull. Russ. Acad. Sci. – Phys. 39 (1) (1975) 18. Exfor data base
16. Tárkányi, F., Hermanne, A., Takács, S. Király, B., Spahn, I., Ignatyuk, A.V., (2010): Experimental study of the excitation functions of proton induced nuclear reactions on ^{167}Er for production of medically relevant ^{167}Tm. Appl. Radiat. Isot., 68 , 250–255.
17. Tárkányi, F., B. Kiraly, F. Ditroi, S. Takács, J. Csikai, A. Hermanne, M.S. Uddin, M. Hagiwara, M. Baba, T. Ido, Yu.N. Shubin, S.F. Kovalev, Activation cross-sections on cadmium: proton induced nuclear reactions up to 80 MeV, Nucl. Instrum. Meth. B 245 (2006) 379.
18. Tárkányi, F., Takács, S., Gul, K., Hermanne, A., Mustafa, M.G., Nortier, M., Obložinský, P., Qaim, S.M., Scholten, B., Shubin, Yu., Zhuang, Y., (2001): Beam monitor reactions: in Charged-Particle Cross-Section Database for Medical Radioisotope Production. IAEA-TECDOC-1211, pp. 49–152.
19. Ziegler, J.F., Ziegler, M.D., Biersack, J.P.,2010. SRIM (2010): Code, Available from <www.srim.org>.