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Activation cross-sections of longer-lived products of proton induced nuclear reactions on dysprosium up to 36 MeV

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ABSTRACT

Activation cross-sections of longer-lived products of proton induced nuclear reactions on dysprosium were measured up to 36 MeV by using stacked foil irradiation technique and γ-spectrometry. We report for the first time experimental cross-sections for the formation of the radionuclides ^{162m}Ho, ^{161}Ho, ^{159}Ho, ^{159}Dy, ^{157}Dy, ^{155}Dy, ^{161}Tb, ^{160}Tb, ^{156}Tb and ^{155}Tb. The experimental data were compared with the results of cross-section calculations of the ALICE and EMPIRE nuclear model codes and of the TALYS nuclear reaction model code as listed in the on-line libraries TENDL 2011 and TENDL 2012.

1. Introduction

A research program is running to study activation cross-sections of proton and deuteron induced reactions mainly for practical applications and to test the presently used theoretical nuclear reaction codes. From a detailed study of the literature it was recognized that data on rare earth elements in most cases are missing. It is well known that many rare earth radionuclides are used in medicine for diagnostic (PET) and, in a larger proportion, for therapeutic (radiopharmaceuticals and brachytherapy) purposes.

In the frame of this systematic study we have investigated the activation cross-sections induced by protons and deuterons on natural dysprosium targets. The part of the study specifically devoted to production of ^{161}Ho, a candidate therapeutic radioisotope, was published separately (Tárkányi et al., 2013a). Here we report on the complete set of activation cross-section data induced by proton irradiation of dysprosium. No earlier experimental data were found in the literature.

2. Experiment and data evaluation

The general characteristics and procedures for irradiation, activity assessment and data evaluation (including estimation of uncertainties) were similar as in many of our earlier works (Takács et al., 2011; Tárkányi et al., 2012).

The main experimental parameters and the methods of data evaluation for the present study are summarized in Table 1 (Andersen and Ziegler, 1977; Bonardi, 1987; Canberra, 2000; Dityuk et al., 1998; Herman et al., 2007; International-Bureau-of-Weights-and-Measures, 1993; Kinsey et al., 1997; Koning and Rochman, 2012; Pritychenko and Sonzogni, 2003; Székely, 1985; Tárkányi et al., 1991, 2001). The used decay data are collected in Table 2.

For beam current and beam energy monitoring and for energy degradation Ti foils were incorporated downstream of each dysprosium foils in the stack. All monitor foil data were considered simultaneously in order to obtain the beam current and beam energy in each target foil by comparison with the IAEA recommended monitor data (Tárkányi et al., 2001). The measured cross-sections of the monitor reaction and the recommended data are shown in Fig. 1.

3. Results and discussion

3.1. Cross-sections

The measured cross-sections for the production of ^{162m}Ho, ^{161}Ho, ^{159}Ho, ^{159}Dy, ^{157}Dy, ^{155}Dy, ^{161}Tb, ^{160}Tb, ^{156}Tb, ^{155}Tb are shown in Tables 3 and 4 and Figs. 2–11. The figures also show the theoretical results calculated with the ALICE-IPPE and the EMPIRE codes and the values available in the on-line libraries TENDL 2011 and TENDL 2012 in comparison with experimental results of this work. We show both TENDL versions, obtained from the

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Table 1
Main parameters of the experiment and the methods of data evaluations.

Experiment		Data evaluation	
Incident particle	Proton	γ-Spectra evaluation	Genie 2000, FORGAMMA (Canberra, 2000; Székely, 1985)
Method	Stacked foil	Determination of beam intensity	Faraday cup (preliminary) Fitted monitor reaction (final) (Tárkányi et al., 1991)
Target stack and thicknesses	Ti–Al–Dy–Al block Repeated 15 times ^{nat}Ti foil, 10.9 μm ^{nat}Al foil, 98 μm ^{nat}Dy foil, 100.59 μm	Decay data	NUDAT 2.6 (Kinsey et al., 1997)
Number of Dy target foils	15	Reaction Q-values	Q-value calculator (Pritychenko and Sonzogni, 2003)
Accelerator	CGR 560 cyclotron Vrije Universiteit Brussels	Determination of beam energy	Andersen (preliminary) Fitted monitor reaction (final) (Andersen and Ziegler, 1977)
Primary energy	36 MeV	Uncertainty of energy	Cumulative effects of possible uncertainties
Irradiation time	71 min	Cross-sections	Isotopic cross-section
Beam current	61 nA	Uncertainty of cross-sections	Sum in quadrature of all individual contribution (International-Bureau-of-Weights-and-Measures, 1993)
Monitor reaction [recommended values]	^{nat}Ti(p,x)^{48}V reaction (Tárkányi et al., 2001)	Yield	Physical yield (Bonardi, 1987)
Monitor target and thickness	^{nat}Ti, 10.9 μm	Theory	ALICE-IPPE (Dityuk et al., 1998), EMPIRE (Herman et al., 2007), TALYS (TENDL 2011, 2012 (Koning and Rochman, 2012)
Detector	HPGe		
γ-Spectra measurements	4 series		
Cooling times	1.5 h, 20 h, 80 h, 330 day		

standard set – not adjusted parameters – to illustrate the difference and the effect of the upgrading.

Due to the experimental circumstances (stacked foil technique, large dose at EOB, limited detector capacity) no cross-section data were obtained for short-lived activation products as ^{164g}Ho(37.5 min), ^{164g}Ho(29 min), ^{162g}Ho(15.0 min), ^{160g}Ho(25.6 min), ^{158m}Ho(28 min), ^{157}Ho(12.6 min), ^{156}Ho (9.5 s, 7.8 min, 56 min) and ^{155}Ho, (48 min).

Also the possibly produced radioisotopes ^{158}Tb (very long half-life, 180 a) and ^{160m}Ho (5.02 h) could not be identified in the measured spectra because of low energy unresolved γ-rays, or small effective cross-section due to the low abundance.

The reactions are discussed separately for each reaction product. Naturally occurring dysprosium is composed of 7 stable isotopes (^{156}Dy – 0.06%, ^{158}Dy – 0.10%, ^{160}Dy – 2.34%, ^{161}Dy – 18.9%, ^{162}Dy – 25.5%, ^{163}Dy – 24.9% and ^{164}Dy – 28.2%). The relevant contributing reactions are collected in Table 2.

The holmium reaction products are produced only through (p,xn) reactions, the dysprosium products directly via (p,pxn) reactions and through the decay of holmium radio-parents, the terbium radioisotopes are produced through directly (p,2pxn) reactions (including complex particle emissions) and decay of simultaneously produced dysprosium radio-products.

3.1.1. ^{nat}Dy(p,xn)^{162m}Ho reaction

Cross-sections for the short-lived ^{162g}Ho ground state ($T_{1/2}$ = 15.0 min) were not measured. Theoretical estimates of ^{162m}Ho ($T_{1/2}$ = 67.0 min) in TENDL 2011 and 2012 are systematically higher by a factor of 1.3 than our experimental results (Fig. 2). In case of ALICE-D and EMPIRE-D the agreement with the experiment is much better. To demonstrate the overestimation of TENDL, 0.7∗TENDL 2012 results were also presented, which gives a good estimation in shape and value of the new experimental data.

3.1.2. ^{nat}Dy(p,xn)^{161}Ho reaction

The experimental and theoretical cross-sections of the reactions producing ^{161}Ho ($T_{1/2}$ = 2.48 h) are shown in Fig. 3. The theoretical overestimation in all cases is significant. Scaling by a factor 0.7 of the TENDL 2011 results shows a rather good agreement.

3.1.3. ^{nat}Dy(p,xn)^{159}Ho reaction

The theory follows both in shape and in magnitude the experimental cross-sections of the ^{159}Ho ($T_{1/2}$ = 33.05 min) in the investigated energy range (Fig. 4), as far as the TENDL calculations regarded. The ALICE-IPPE and EMPIRE overestimate again.

3.1.4. ^{nat}Dy(p,x)^{159}Dy reaction

The cumulative cross-sections of reactions producing ^{159}Dy ($T_{1/2}$ = 144.4 d) contain apart from the direct production, the contribution from the decay of ^{159}Ho ($T_{1/2}$ = 33.05 min) as they were measured after nearly complete decay of the parent isotope. The agreement with the results of the 3 codes is acceptable (Fig. 5). The TENDL results show that the direct production is negligible, especially below 30 MeV.

3.1.5. ^{nat}Dy(p,x)^{157}Dy reaction

The cumulative cross-sections for production of ^{157}Dy ($T_{1/2}$ = 8.14 h) were measured after nearly complete decay of the parent ^{157}Ho ($T_{1/2}$ = 12.6 min). The theoretical data overestimate the experimental results and here also the TENDL results show that the direct production is negligible (Fig. 6).

3.1.6. ^{nat}Dy(p,x)^{155}Dy reaction

The measured ^{155}Dy ($T_{1/2}$ = 9.9 h) was produced directly and through decay of the ^{155}Ho ($T_{1/2}$ = 48 min) parent radioisotope. The comparison with the TENDL results shows good agreement below 30 MeV (Fig. 7), and the agreement with the ALICE-IPPE and the EMPIRE results is also good below 25 MeV.