Activation cross-sections of long lived products of deuteron induced nuclear reactions on dysprosium up to 50 MeV

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Abstract

Activation cross-sections for production of $^{162m,161,155}$Ho, $^{165,159,157,155}$Dy and $^{161,160,156,155}$Tb radionuclides in deuteron induced nuclear reactions on elemental dysprosium were measured up to 50 MeV for practical application and the test of the predictive power of nuclear reaction model codes. A stacked-foil irradiation technique and off-line γ-ray spectrometry were used to determine the activities. No earlier cross-section data were found in the literature. The experimental data are compared with the predictions of the ALICE-D, EMPIRE-D and TALYS codes. Integral production yields were calculated from the fitted experimental data.

Keywords: dysprosium target, deuteron irradiation, holmium, dysprosium and terbium radio-isotopes, physical yield

1. Introduction

In the frame of our systematic study on the excitation functions of light charged particle induced nuclear reactions, the excitation functions for radionuclides production by deuteron irradiation of natDy were measured. Dysprosium is being a high priority and critical strategic metal now used world-wide for high technology, nuclear, clean energy and military applications. The knowledge on the activation cross-sections for light charged particles (ions) and neutron induced reactions are important for various applications. The elemental dysprosium and some of its radionuclides are known to have applications in fission reactor physics and in medical field. The elemental dysprosium has a high thermal neutron absorption cross-section, which makes it interesting for making neutron-absorbing control rods in nuclear reactors (Kannan and Ganesan 2010), and a resonance absorber to control the coolant void reactivity in Advanced Heavy Water Reactor (AHWR) (Kannan and Ganesan 2010). In medical treating damaged joints, irradiation with $^{165}$Dy has proved to be more effective than traditional surgery (Srivastava 2004), and $^{159}$Dy has been advocated for transmission imaging and bone mineral analysis (Rao et al. 1977). An alternative production route for radionuclides of lanthanides using high intensity lasers or light sources was studied recently by a group in Grenoble (Habs and Köster 2012). There were several aims of the study:

- The earlier mentioned global task: the systematic study of the deuteron induced activation data, taking into account that no earlier data were found in the literature.
- Investigation of different production routes of the $^{161}$Ho medical radioisotope
- Investigation of the predictive power of widely used theoretical codes, via systematic comparison between the experimental and theoretical data in connection with the FENDL-3 Project (IAEA).
- To get experimental data in a wide mass region for
(d,p) reactions to prepare systematics to be used in upgrading of nuclear reactions models for codes allowing better description of deuteron induced reactions.

Part of the experimental data for production of $^{161}$Ho was published recently (Tarkanyi et al., 2013). Here we repeat the data related to this radionuclide in graphical form for completeness and for comparison with the theories.

2. Experiment and data evaluation

The general characteristics and procedures for irradiation, activity assessment and data evaluation (including estimation of uncertainties) were similar to those in our earlier works (Takacs et al., 2011; Tarkanyi et al., 2012). The used high purity target foils (<98% purity) were bought from Goodfellow® and were handled according to the producer’s recommendations. The foil thicknesses and homogeneity were accurately re-measured. The main experimental parameters for the present study are summarized in Table 1 (Andersen and Ziegler, 1977; Bonardi, 1987; Canberra, 2000; Kinsey et al., 2010; Of-Weights-and-Measures, 1993; Priyachenko and Sonzogni, 2003; Székely, 1985; Tarkanyi et al., 1991, 2001) together with the methods used in data evaluation. The used decay data and Q values of the contributing reactions are collected in Table 2. To illustrate the reliability of the incident energy and the intensity of the deuteron beam along the stack, the simultaneously measured complete excitation function of the monitor reaction $^{27}$Al(d,x)$^{24}$Na is shown in Fig. 1 in comparison with the recommended data taken from (Tarkanyi et al., 2001).

3. Theory

The ALICE-IPPE-D and EMPIRE-D (Dityuk et al., 1998; Herman et al., 2007) calculations were performed using the recommended values for the input parameters. The results of TALYS (Koning et al., 2007) code were taken from the TENDL 2012 library (Koning et al., 2012). The ALICE-D and EMPIRE-D are the modified version of ALICE-IPPE (Dityuk et al., 1998) and EMPIRE (Herman et al., 2007) codes and were developed at IPPE (Institute of Physics and Power Engineering, Obninsk, Russia) for better description of activation cross-sections of deuteron induced reactions by using energy dependent enhancement factor (Ignatyuk, 2011) for simulation of direct (d,p) and (d,t) transitions. Independent data for isomers with ALICE-D code was obtained by using the isomeric ratios calculated with EMPIRE-D. Separate sets of calculations were performed for all stable Dy isotopes as target nuclei. The excitation functions for natural isotopic composition were obtained by summing all these individual excitation functions with weight of their respective natural abundances.

4. Results

4.1. Cross-sections

Activation cross-sections for production of the $^{162m,161,155}$Ho,$^{165,159,157,155}$Dy and $^{161,160,156,155}$Tb radionuclides were measured. The experimental cross-section data are shown in Figs. 2-11 in comparison with the predictions of theoretical codes. The numerical data are collected in Tables 3 and 4. The cross-section values of holmium production are for direct production via (d,xn) reactions. The dysprosium radio-isotopes are produced directly via (d,pxn) reactions or additionally through the decay of the shorter-lived isobaric parent holmium radioisotope (cumulative cross-section). The terbium radioisotopes are produced directly via (d,p2xn) reaction.

![Figure 1: The simultaneously measured monitor reactions for determination of deuteron beam energy and intensity](image-url)
Table 1: Main experimental parameters and methods of data evaluation

| Parameter                        | Value                                                                 |
|----------------------------------|----------------------------------------------------------------------|
| Incident particle                | Deuteron                                                             |
| Measurement method               | Stacked foil                                                         |
| Track compensation               | Si-200Si, repeated 15 times                                          |
| Target and thickness             | 99.999% Dy foil, 100.59 µm                                           |
| Nuclide of targets               | Tb-Al-Dy, repeated 15 times                                          |
| Accelerator                      | Cyclotron of the Université catholique in Louvain la Neuve (LLN)    |
| Primary energy                   | 50 MeV                                                               |
| Irradiation time                 | 30 min                                                               |
| Beam current                     | 10 µA                                                                |
| Monitor reaction, [recommended value] | 27Al(d,x)24Na reaction (Tarkanyi et al., 2001)                      |
| Monitor target and thickness     | natAl, 26.96 µm                                                      |
| Detector                         | HpGe                                                                |
| g-spectra measurements          | 4 series                                                             |
| Cooling time                     | 4h, 30h, 430h, 2380h                                                |

...and additional comments...

4.1.1. Cross-sections for the natDy(d,x)162mHo reaction

The production cross-sections of 162mHo (T1/2=67.0 min) are shown in Fig. 2. None of the theoretical calculations represents well the experimental data and large disagreements exist among the codes. TENDL-2012 values, which are the results of most recent version of TALYS are a factor of 2 higher and shifted to higher energies compared to the previous results.

4.1.2. Cross-sections for the natDy(d,x)161gDy(m+) reaction

The measured cross-section include the full contribution of the decay of the short-lived isomer (T1/2 = 6.7 s). According to Fig. 3, the shape of the experimental and theoretical excitation functions of 161Ho (T1/2 = 2.48 h) are similar, but the overestimation of all codes is surprisingly high. For the TENDL results (both libraries give about the same values) a scaling factor of 0.55 results in a reasonable representation of the experiment.

4.1.3. Cross-sections for the natDy(d,x)165Dy(m+) reaction

165Dy (T1/2 = 2.334 h) is produced only via the 164Dy(d,p) reaction. The measured cross-section contains the contribution of the complete decay of the short-lived isomeric state (T1/2=1.257 min) that decays with IT
(97.74 %) to the ground state (m+). The experimental data are in good agreement with the systematics of the experimental (d,p) cross-sections above 40 MeV and between the predictions of EMPIRE-D and ALICE-D. The new TENDL 2012 data are closer to the experiment than TENDL 2011 but the underestimation is still significant.

4.1.4. Cross-sections for the \( {}^{nat}\text{Dy}(d,x){}^{159}\text{Dy}(m+) \) reaction

The cumulative cross-sections of \( {}^{159}\text{Dy}(T_{1/2} = 144.4 \text{ d}) \) include the direct production and the contribution from the decay of \( {}^{159}\text{Ho} (T_{1/2} = 33.05 \text{ min}) \), were impossible to measure in our experimental conditions (too long cooling time). The overestimations of the cumulative cross-sections given in both TENDL libraries and by the ALICE-D and EMPIRE-D codes are significant (Fig. 5).

4.1.5. Cross-sections for the \( {}^{nat}\text{Dy}(d,x){}^{157}\text{Dy}(cum) \) reaction

Under our experimental circumstances we can deduce only cumulative cross-sections for production of \( {}^{157}\text{Dy} (T_{1/2} = 8.14 \text{ h}) \) including the contribution of total decay of short-lived parent \( {}^{157}\text{Ho} (T_{1/2} = 12.6 \text{ min}) \). The three codes reproduce well the complex shape of the cumulative experimental excitation function (direct on multiple stable target isotopes at higher energy and parent decay below 15 MeV) (see Fig. 6), but are one order of magnitude lower for the \( {}^{156}\text{Dy}(d,n){}^{157}\text{Ho} \) reaction. At higher energies all codes overestimate the experimental results by a factor of two. According to the theory the direct reaction contributes only by about 20%.

4.1.6. Cross-sections for the \( {}^{nat}\text{Dy}(d,x){}^{155}\text{Dy}(cum) \) reaction

The cumulative cross-section of the \( {}^{155}\text{Dy} (T_{1/2} = 9.9 \text{ h}) \) represents the sum of the direct and indirect production through the decay of \( {}^{155}\text{Ho} (T_{1/2} = 48 \text{ min}) \). The agreement with the results of the three codes is acceptable. (Fig. 7)

4.1.7. Cross-sections for the \( {}^{nat}\text{Dy}(d,x){}^{161}\text{Tb} \) reaction

The radionuclide \( {}^{160}\text{Tb} (T_{1/2} = 72.3 \text{ d}) \) can only be produced directly by \( (d,2pxn) \) reactions as decay contribu-
tions are closed from both sides. The activation cross-sections are shown in Fig. 9. There are significant disagreements with the results of the theoretical codes.

4.1.9. Cross-sections for the $^{nat}$Dy(d,x)$^{159}$Dy(cum) reaction

The production cross-sections of $^{156}$Tb (Fig. 10) represent total activation cross-sections of the ground state: i.e. cumulative cross-section of the ground state after the decay of the two isomeric states ($^{156m_1}$Tb; IT: 100 %, $T_{1/2} = 3$ h and $^{156m_2}$Tb; IT: 100 %, $T_{1/2} = 24.4$ h). Large divergences can be seen between the different codes and the experimental results.

4.1.10. Cross-sections for the $^{nat}$Dy(d,x)$^{155}$Tb(cum) reaction

The cross-sections for $^{155}$Tb ($T_{1/2} = 5.32$ d) production are cumulative. They were determined from the spectra measured after the decay of progenitor $^{155}$Ho ($T_{1/2} = 48$ min) and $^{155}$Dy ($T_{1/2} = 9.9$ h) isotopes. The best description is given by the TENDL calculations, while up to 35 MeV the ALICE-D is also acceptable (Fig. 11).

4.2. Integral yields

We calculated the integral yields for all investigated radionuclides from a spline fit to our experimental data. The yields represent so called physical yields for instantaneous irradiation. No data for directly measured thick target yields were found in the literature. The yields are presented in Figs. 12 and 13. In Fig. 13 the most interesting isotopes are the $^{161}$Ho and $^{162}$mHo, as they have also medical interest (Trknyi et al., 2013). The $^{165}$Dy production yield is with an order of magnitude lower than that of the above two. Another group is formed from the remaining dysprosium isotopes i.e. $^{159}$, $^{157}$, $^{155}$Dy, which have much lower yield. In Fig. 13 the terbium radio-isotopes are presented. From the point of view of yield they rather belong to the latter group of the preceding figure.

5. Summary and conclusions

In the frame of systematic investigation of excitation functions of deuteron induced nuclear reactions, activation cross-sections on dysprosium were investigated up to 50 MeV. Independent or cumulative cross-sections for the formation of the radionuclides $^{162m}$, $^{161}$Ho, $^{165}$, $^{159}$, $^{157}$, $^{155}$Dy and $^{161}$, $^{160}$, $^{156}$, $^{155}$Tb through $^{nat}$Dy(d,x) nuclear reactions were measured for the first time. The experimental data were compared with the theoretical data obtained by the EMPIRE-D and the ALICE-IPPE-D codes and the TALYS data in TENDL library. The prediction of the codes differs significantly from each other, and from the
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Figure 10: Experimental and theoretical excitation functions for $^{nat}$Dy(d,x)$^{156}$Tb (cum)

Figure 11: Experimental and theoretical excitation functions for $^{nat}$Dy(d,x)$^{155}$Tb (cum)

Figure 12: Integral yields as a function of proton energy for the production of $^{162m,161}$Ho, $^{165,159,157,155}$Dy radioisotopes

Figure 13: Integral yields as a function of proton energy for the production of $^{161,160,156,155}$Tb radioisotopes.
Table 2: Decay data and contributing reactions

| Nuclide | Half-life | Eγ (keV) | Iγ (%) | Contributing reaction | Q-value (keV) |
|---------|-----------|----------|--------|-----------------------|---------------|
| 162mHo | 67.0 min  | 57.34    | 4.4    | 162mHo(d,n)           | -5146.61      |
|         |           |          |        | 162mHo(d,2n)          | -11471.62     |
| 162Ho  | 2.48 h    | 77.42    | 103.05 | 162Ho(d,2n)           | -2889.18      |
|         |           | 103.05   | 0.49   | 162Ho(d,3n)           | -12062.2      |
|         |           | 157.26   | 0.43   | 162Ho(d,4n)           | -18333.21     |
|         |           | 175.42   |        | 162Ho(d,4n)           | -25991.35     |
| 161Ho  | ε: 100 %  | 103.89   | 3.80   | 161Ho(d,2n)           | -11280.12     |
|         |           | 256.38   | 0.834  | 161Ho(d,3n)           | -26888.71     |
|         |           | 361.68   | 0.904  | 161Ho(d,4n)           | -35145.3      |
|         |           | 655.413  | 0.913  | 161Ho(d,5n)           | -41340.09     |
|         |           | 715.328  | 0.978  | 161Ho(d,6n)           | -51722.87     |
|         |           | 160Dy(d,n) |      | 161Ho(d,7n)           | -59360.99     |
|         |           | 161Dy(d,2n) |     | 161Ho(d,8n)           | -64020.25     |
| 155Ho  | ε+β−: 100 % | 6.89 d | 74.56669 | 155Ho(d,2p)           | -27500.17     |
|         |           | 105.318  | 0.183  | 155Ho(d,2pn)          | -43099.3      |
|         |           | 148.64   | 0.101  | 155Ho(d,2p2n)         | -49393.5      |
|         |           | 161.29   | 0.078  | 155Ho(d,2p3n)         | -57750.7      |
|         |           | 180.08   |        | 155Ho(d,2p4n)         | -64021.7      |
|         |           | 262.27   | 0.058  | 155Ho(d,2p5n)         | -71785.4      |
| 158Dy  | β−: 100 %  | 9.9 h    | 34.64  | 158Dy(d,2p2n)         | -21322.87     |
|         |           | 184.564  | 68.4   | 158Dy(d,2p4n)         | -30960.99     |
|         |           | 226.918  |        | 158Dy(d,2p5n)         | -42360.19     |
|         |           | 34.64    | 0.183  | 158Dy(d,2p3n)         | -53850.3      |
|         |           | 105.318  | 0.101  | 158Dy(d,2p4n)         | -61144.9      |
| 157Dy  | ε: 100 %   | 5.35 d   | 4.78   | 157Dy(d,2p2n)         | -31300.9      |
|         |           | 74.56699 | 0.183  | 157Dy(d,2p3n)         | -37330.3      |
|         |           | 103.065  | 0.101  | 157Dy(d,2p4n)         | -42360.19     |
|         |           | 106.113  | 0.078  | 157Dy(d,2p5n)         | -48750.5      |
|         |           | 292.401  |        | 157Dy(d,2p6n)         | -50080.9      |
|         |           | 1154.07  | 0.058  | 157Dy(d,2p7n)         | -56380.3      |
|         |           | 1222.44  |        | 157Dy(d,2p8n)         | -62680.7      |
| 155Tb  | ε: 100 %   | 5.32 d   | 32.0   | 155Tb(d,2p2n)         | -24813.9      |
|         |           | 105.318  | 25.1   | 155Tb(d,2p3n)         | -40222.5      |
|         |           | 148.64   | 2.45   | 155Tb(d,2p4n)         | -46678.5      |
|         |           | 161.29   | 2.76   | 155Tb(d,2p5n)         | -54873.9      |
|         |           | 163.28   | 4.44   | 155Tb(d,2p6n)         | -61144.9      |
|         |           | 180.08   | 7.5    | 155Tb(d,2p7n)         | -67414.9      |
|         |           | 262.27   | 5.3    | 155Tb(d,2p8n)         | -73684.9      |

Naturally occurring dysprosium is composed of 7 isotopes (156Dy 0.06 %, 158Dy 0.10 %, 160Dy 2.34 %, 161Dy 18.9 %, 162Dy 25.5 %, 163Dy 24.9 % and 164Dy 28.2 %). When complex particles are emitted instead of individual protons and neutrons the Q-values have to be decreased by the respective binding energies of the compound particles: np-d, +2.2 MeV; 2np-t, +8.48 MeV; n2p-3He, +7.72 MeV; 2n2p-a, +28.30 MeV.
Table 3: Experimental cross-sections of $^{nat}$Dy(d,x)$^{162}$Ho, $^{161}$gHo(m+), $^{165}$gDy(m+), $^{159}$Dy(cum), $^{157}$Dy(cum), $^{155}$Dy(cum) reactions

| $E$ ± $\Delta E$ (MeV) | Cross-section(σ)±$\Delta$σ (mb) |
|------------------------|----------------------------------|
| 3.3 ± 1.1             | $^{162}$mHo                      |
| 11.0 ± 1.0            | $^{161}$gHo                      |
| 20.0 ± 0.0            | $^{165}$gDy                      |
| 25.6 ± 0.7            | $^{159}$Dy                       |
| 26.8 ± 0.7            | $^{157}$Dy                       |
| 29.8 ± 0.6            | $^{155}$Dy                       |
| 32.5 ± 0.6            | $^{161}$Tb                       |
| 35.1 ± 0.5            | $^{160}$Tb                       |
| 37.7 ± 0.5            | $^{156}$Tb                       |
| 40.0 ± 0.5            | $^{155}$Tb                       |
| 42.3 ± 0.4            | $^{156}$Tb                       |
| 44.8 ± 0.3            | $^{155}$Tb                       |
| 46.5 ± 0.2            | $^{161}$Tb                       |
| 48.6 ± 0.1            | $^{160}$Tb                       |
| 50.7 ± 0.1            | $^{156}$Tb                       |
| 52.5 ± 0.1            | $^{155}$Tb                       |
| 54.6 ± 0.1            | $^{156}$Tb                       |
| 56.8 ± 0.1            | $^{155}$Tb                       |

Table 4: Experimental cross-sections of $^{nat}$Dy(d,x)$^{161}$Th, $^{160}$Th, $^{156}$Th(cum), $^{155}$Th(cum) reactions

| $E$ ± $\Delta E$ (MeV) | Cross-section(σ)±$\Delta$σ (mb) |
|------------------------|----------------------------------|
| 3.3 ± 1.1             | $^{161}$Th                       |
| 11.0 ± 1.0            | $^{160}$Th                       |
| 20.0 ± 0.0            | $^{156}$Th                       |
| 25.6 ± 0.7            | $^{155}$Th                       |
| 26.8 ± 0.7            | $^{161}$Th                       |
| 29.8 ± 0.6            | $^{160}$Th                       |
| 32.5 ± 0.6            | $^{156}$Th                       |
| 35.1 ± 0.5            | $^{155}$Th                       |
| 37.7 ± 0.5            | $^{156}$Th                       |
| 40.0 ± 0.5            | $^{155}$Th                       |
| 42.3 ± 0.4            | $^{156}$Th                       |
| 44.8 ± 0.3            | $^{155}$Th                       |
| 46.5 ± 0.2            | $^{161}$Th                       |
| 48.6 ± 0.1            | $^{160}$Th                       |
| 50.7 ± 0.1            | $^{156}$Th                       |
| 52.5 ± 0.1            | $^{155}$Th                       |
| 54.6 ± 0.1            | $^{156}$Th                       |
| 56.8 ± 0.1            | $^{155}$Th                       |

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