Applied Radiation and Isotopes 127 (2017) 7–15

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Activation cross-sections of longer lived radioisotopes of proton induced nuclear reactions on terbium up to 65 MeV

F. Tárkányi[a], A. Hermanne[b], F. Ditrói[a,*], S. Takács[a], A.V. Ignatyuk[c]

[a] Institute for Nuclear Research, Hungarian Academy of Sciences (ATOMKI), Debrecen, Hungary
[b] Cyclotron Laboratory, Vrije Universiteit Brussel (VUB), Brussels, Belgium
[c] Institute of Physics and Power Engineering (IPPE), Obninsk, Russia

ARTICLE INFO

Keywords:
Terbium
Proton irradiation
Theoretical model codes
Cross section
Physical yield

ABSTRACT

Experimental cross sections are presented for the $^{159}Tb(p,xn)^{153,155,157,159}Dy$, $^{152,153,155,156m2,m1,g,158}Tb$ and $^{153,151}Gd$ nuclear reactions up to 65 MeV. The experimental results are compared with the recently reported experimental data and with the results of the nuclear reaction codes ALICE-IPPE, EMPIRE and TALYS as reported in the TENDL-2015 on-line library. Integral thick-target yields are also derived for the reaction products used in practical applications and production routes are discussed.

1. Introduction

A systematic study of proton and deuteron induced reactions up 50 MeV deuteron and 65–70 MeV proton is in progress for testing the prediction possibilities of different theoretical model codes, to complete the activation data file and to investigate production routes of medically relevant isotopes (diagnostic and therapeutic nuclear medicine) of the lanthanide group. Nearly all elements from lanthanum to lutetium were already investigated by our group and discussed in earlier publications.

Activation cross section on Tb are required for production of the medically relevant ^{159}Dy (Nayak and Lahiri, 1999) and ^{157}Dy (Lebowitz and Greene (1971), Yano et al. (1972), Apo (1981). Our results on deuteron induced reactions on terbium were published by Tárkányi et al. (2013). We decided to continue the investigations to compare the production possibilities of the proton and deuteron induced routes.

When our experiment started only a few experimental activation cross-sections of proton induced reaction on terbium were available in the literature, by Lebowitz and Greene (1971), Hassan et al. (2010) and Steyn et al. (2014). Very recently a detailed set of experimental data up to 200 MeV was published by the Los Alamos group (Engle et al., 2012a, 2012b, 2015, 2016).

2. Experimental and data evaluation

The experimental details and the used data evaluation methods were described in many of our earlier reports (Ditrói et al., 2009, 2014). Here we briefly summarize the parameters closely related to this study in Table 1 (experiments), Table 2 (data evaluation) and Table 3 (used decay data), respectively.

3. Model calculations

The new experimental data are shown together with earlier reported experimental data and results of nuclear reaction model calculations. The ALICE-IPPE (Dityuk et al., 1998) and EMPIRE (Herman et al., 2007) codes were used to analyze the present experimental results. The parameters for the optical model, level densities and pre-equilibrium contributions were taken as described in (Belgya et al., 2006). The cross sections for isomers in case of ALICE-IPPE code were obtained by using the isomeric ratios calculated with EMPIRE. The new experimental data are also compared to the data in the TENDL-2015 library (Koning et al., 2015), based on both default and adjusted TALYS (1.6) calculations (Koning and Rochman, 2012).

In a first attempt the EMPIRE calculations were performed using the BNL option (EGSM, Enhanced Generalized Superfluid Model) for level densities, but resulted in large disagreement for excitation functions of gadolinium radio-products. A second series of EMPIRE calculations were performed with the Composite Gilbert-Cameron (GC) description for level densities and gives better agreement for gadolinium isotopes as can be seen on the relevant figures. The differences between the two approaches are rather small for the production of Dy and Tb isotopes and are not represented on the figures. Unfortunately, there are no direct experimental data on the level density parameters for these

* Corresponding author.
E-mail address: ditroi@atomki.hu (F. Ditrói).

http://dx.doi.org/10.1016/j.apradiso.2017.04.030
Received 9 February 2017; Received in revised form 28 March 2017; Accepted 18 April 2017
Available online 02 May 2017

Table 1
Main experimental parameters.

Reaction	^{159}Tb (p,x) (ser. 2)	^{159}Tb (p,x) (ser. 1)
Incident particle	Proton	Proton
Method	Stacked foil	Stacked foil
Number of Tb target foils	15	12
Target composition and thickness (μm)	Tb(22.2), Al(377), La(25), Al(10), CeO(33.1–63.2, sedimented),Al (100) Repeated 15 times	Ti (10.9), Al (102), Tb (22.2). Ti (10.9), Al (102), Al (10) CeO (4.59–34.0, sedimented), Al (100) Repeated 12 times
Accelerator	Cyclone 90 cyclotron of the Université Catholique in Louvain la Neuve (LLN)	CGR-560 cyclotron of the Vrije Universiteit Brussel (VUB) in Brussels
Primary energy (MeV)	65	35
Covered energy range (MeV)	64.9–35.3	33.9–5.8
Irradiation time (min)	59	60
Beam current (nA)	90	100
Monitor reaction, [recommended values]	$^{27}Al(p,x)^{24}Na$ reaction (Tárkányi et al., 2001)	$^{nat}Ti(p,x)^{48}V$ reaction (Tárkányi et al., 2001)
Monitor target and thickness (μm)	^{nat}Al, 377 and (100 + 10)	^{nat}Ti, 10.9
detector	HPGe	HPGe
γ-spectra measurements	4 series	4 series
Cooling times (h)	6.4–8.9 22.2–28.1 127.0–166.1 3120–3241	1.4–3.8 22.1–25.1 292–319 1998–2283

isotopes.

4. Results and discussion

4.1. Excitation functions

The measured cross sections for the production of the investigated radio-products are presented in Tables 4–6 and Figs. 1–13 in comparison with earlier literature values and the results of different model calculations.

4.1.1. Production of ^{159}Dy ($T_{1/2}$ = 144.4 d)

According to Fig. 1. our new data are in acceptable agreement with the recent results of Engle et al. (2016) and with earlier measurement of Hassan et al. (2007) and Steyn et al. (2014). in the overlapping energy range. The values near the maximum at 9 MeV differ however by a factor of 2. The TENDL-2015 underestimates the maximum around 10 MeV and overestimates systematically the high energy tail. The EMPIRE and the ALICE-IPPE describe rather well the shape of the excitation function with a strong underestimation for the ALICE-IPPE results.

4.1.2. Production of ^{157}Dy ($T_{1/2}$ = 8.14 h)

The present and the earlier experimental data of Lebowitz and Greene (1971), Steyn et al. (2014) and Engle et al. (2016) for the ^{159}Tb (p,3 n)^{157}Dy reaction are shown in Fig. 2. The agreement in the common energy range is excellent. The TENDL-2015 underestimates the maximum. The predictions of the EMPIRE and ALICE-IPPE are acceptable.

4.1.3. Production of ^{155}Dy ($T_{1/2}$ = 9.9 h)

Good agreement was found with the earlier experimental data of Steyn et al. (2014) and Engle et al. (2016). The maximum of the excitation function in the TENDL-2015 prediction is shifted to lower energy (Fig. 3). There are significant disagreements in the maximum value for the EMPIRE and ALICE-IPPE codes.

4.1.4. Production of ^{153}Dy ($T_{1/2}$ = 5.35 d)

Our data are consistent with Steyn et al. (2014) and the higher energy data of Engle et al. (2016). There is a significant shift to low energy in the effective threshold of the TENDL-2015 excitation function and a significant underestimation of the maximum in case of EMPIRE results (Fig. 4).

4.1.5. Production of ^{158}Tb ($T_{1/2}$ = 9.9 h)

The experimental and theoretical excitation functions are shown in Fig. 5. In this case our data are more scattered but are not contradictory to Engle et al. (2016). There is a factor of 2 disagreement between the predictions of the maximum values of different model codes but all are confirming the general behavior of the experimental excitation function.

4.1.6. Production of ^{156m2}Tb ($T_{1/2}$ = 24.4 h), ^{156m1}Tb ($T_{1/2}$ = 5.3 h) and ^{156}Tb ($T_{1/2}$ = 5.35 d) (m+)

The radionuclide ^{156}Tb has a long-lived ground state and two shorter-lived isomeric states decaying completely to the ground state.

Table 2
Main parameters of data evaluation (with references).

Gamma spectra evaluation	Genie (2000), Forgamma	(Genie, 2000; Székely, 1985)
Determination of beam intensity	Faraday cup (preliminary) Fitted monitor reaction (final)	(Tárkányi et al., 1991)
Decay data	NUDAT 2.6	(Kinsey et al., 1997)
Reaction Q-values	Q-value calculator	(Pritychenko and Sonzogni)
Determination of beam energy	Anderson (preliminary) Fitted monitor reaction (final)	(Andersen and Ziegler, 1977) (Tárkányi et al., 1991)
Uncertainty of energy	Cumulative effects of possible uncertainties	
Cross sections	Isotopic cross section	
Uncertainty of cross sections	Sum in quadrature of all individual linear contributions	(Guide)
Yield	Physical yield	(Bonardi, 1988; Otuka and Takács, 2015)