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Production cross sections of dysprosium, terbium and gadolinium radioisotopes from the alpha-particle-induced reactions on natural gadolinium up to 50 MeV

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Abstract

The activation cross sections of the alpha-particle-induced reactions on natural gadolinium up to 50 MeV were measured. The stacked foil technique, the activation method and the γ -ray spectrometry were used for the measurement. We determined the experimental production cross sections of $^{155,157,159}\text{Dy}$, $^{153,155,156g,160,161}\text{Tb}$ and ^{153}Gd . No previous experimental data of $^{155,159}\text{Dy}$, and ^{153}Gd were found in the literature. The measured values were compared with the previous experimental data and the TENDL-2019 data.

Keyword

Dysprosium-157; Dysprosium-159; Terbium-155; Alpha irradiation; Gadolinium target; Excitation function; Cross section

1. Introduction

Dysprosium and terbium radioisotopes are of interest for diagnosis and therapy in nuclear medicine. Dysprosium-157 ($T_{1/2} = 8.14$ h), dysprosium-159 ($T_{1/2} = 144.4$ d), and terbium-155 ($T_{1/2} = 5.32$ d) can be used for bone scanning (Skeletal imaging) [1], determination of bone mineral [2] and SPECT imaging [3], respectively. We studied the production cross sections of $^{157,159}\text{Dy}$ in the deuteron-induced reactions on ^{159}Tb [4]. These radionuclides can also be produced by alpha-particle-induced reactions on enriched and natural gadolinium [5–8]. Among them, we focused on the reaction on natural gadolinium, of which isotopic composition is ^{152}Gd (0.20%), ^{154}Gd (2.18%), ^{155}Gd (14.79%), ^{156}Gd (20.47%), ^{157}Gd (15.65%), ^{158}Gd (24.83%), and ^{160}Gd (21.86%). In this work, the activation cross sections of the alpha-particle-induced reactions on natural gadolinium were studied. The production cross sections of $^{155,157,159}\text{Dy}$, $^{153,155,156g,160,161}\text{Tb}$ and ^{153}Gd were determined. The results were

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compared with the experimental data studied earlier [5,6] and the TENDL-2019 data based on calculation using the TALYS code [9].

2. Experimental

The experiment was performed at the RIKEN AVF cyclotron. In the experiment, we used the stacked foil technique, the activation method, and high-resolution γ -ray spectrometry to determine the activation cross sections.

The stacked target consisted of ^{nat}Gd and ^{nat}Ti foils. The thicknesses of the ^{nat}Gd (50×50 mm, 99.9% purity, Nilaco Corp., Japan) and ^{nat}Ti foils (50×100 mm, 99.6% purity, Nilaco Corp., Japan) were 25.4 and 2.25 mg/cm², respectively, which were derived from their measured weight and lateral size. The foils were cut into 8×8 mm to fit a target holder serving as a Faraday cup. Fourteen sets of Gd-Ti-Ti foils were stacked as the target. To handle the recoil effect, for monitoring the alpha-particle beam, using the $^{nat}\text{Ti}(\alpha, x)^{51}\text{Cr}$ monitor reaction, every second Ti foil from the Ti-Ti pairs was used.

The stacked target was irradiated with an alpha-particle beam for 60 min. The beam was accelerated to 51.0 ± 0.1 MeV by the RIKEN AVF cyclotron. The beam energy was measured using the time-of-flight method [10]. Energy degradation in the stacked target was calculated using the SRIM code [11]. The average beam intensity of 258 nA was measured using the Faraday cup.

The γ rays emitted from the irradiated foils were measured by a high-resolution HPGe detector (ORTEC GEM30P4-70). The detector was calibrated by using a standard γ -ray source composed of $^{57,60}\text{Co}$, ^{88}Y , ^{109}Cd , ^{113}Sn , ^{137}Cs , ^{139}Ce and ^{241}Am . The γ -ray spectra were analyzed by using the software Gamma Studio (SEIKO EG&G). The γ -ray spectra of each foil were measured four times after cooling times at different distances (14.3-17.6 h at 2 and 75 cm, 1.4-1.7 d at 2, 10 and 25 cm, 2.4-2.8 d at 2 and 10 cm, 25.4-28.8 d at 2 and 5 cm). The dead time of the measurements was kept below 6.3%. Reaction and decay data for the γ -ray spectrometry were taken from NuDat 2.8 [12], Lund/LBNL Nuclear Data Search [13], LiveChart [14] and QCalc [15] and summarized in Table 1.

Table 1. Reactions and decay data of reaction products [12–15]

Nuclide	Half-life	Decay mode (%)	E_γ (keV)	I_γ (%)	Contributing reactions	Q-value (MeV)
^{155}Dy	9.9 h	$\epsilon + \beta^+$ (100)	184.564	3.39(8)	$^{152}\text{Gd}(\alpha, n)$	-11.2
			226.918	68.7(16)	$^{154}\text{Gd}(\alpha, 3n)$	-26.3
					$^{155}\text{Gd}(\alpha, 4n)$	-32.8
					$^{156}\text{Gd}(\alpha, 5n)$	-41.3
					$^{157}\text{Gd}(\alpha, 6n)$	-47.7
^{157}Dy	8.14 h	$\epsilon + \beta^+$ (100)	182.424	1.33(6)	$^{154}\text{Gd}(\alpha, n)$	-9.9
			326.336	93(3)	$^{155}\text{Gd}(\alpha, 2n)$	-16.4
					$^{156}\text{Gd}(\alpha, 3n)$	-24.9

					$^{157}\text{Gd}(\alpha, 4n)$	-31.3
					$^{158}\text{Gd}(\alpha, 5n)$	-39.2
^{159}Dy	144.4 d	ϵ (100)	58.0	2.27(13)	$^{155}\text{Gd}(\alpha, \gamma)$	-0.5
					$^{156}\text{Gd}(\alpha, n)$	-9.0
					$^{157}\text{Gd}(\alpha, 2n)$	-15.4
					$^{158}\text{Gd}(\alpha, 3n)$	-23.3
					$^{160}\text{Gd}(\alpha, 5n)$	-36.7
^{153}Tb	2.34 d	$\epsilon + \beta^+$ (100)	82.86	5.4(7)	$^{152}\text{Gd}(\alpha, t)$	-15.9
			102.255	5.8(5)	$^{154}\text{Gd}(\alpha, 2nt)$	-31.1
			109.758	6.2(4)	$^{155}\text{Gd}(\alpha, 3nt)$	-37.5
			170.42	5.8(4)	$^{156}\text{Gd}(\alpha, 4nt)$	-46.0
			212.00	31.0(18)	$^{153}\text{Dy}(\epsilon)$	
^{155}Tb	5.32 d	ϵ (100)	86.55	32.0(18)	$^{152}\text{Gd}(\alpha, p)$	-8.3
			105.318	25.1(13)	$^{154}\text{Gd}(\alpha, t)$	-15.0
			148.64	2.65(14)	$^{155}\text{Gd}(\alpha, nt)$	-21.4
			161.29	2.76(15)	$^{156}\text{Gd}(\alpha, 2nt)$	-30.0
			163.28	4.44(23)	$^{157}\text{Gd}(\alpha, 3nt)$	-36.3
			180.08	7.5(4)	$^{158}\text{Gd}(\alpha, 4nt)$	-44.3
			262.27	5.3(3)	$^{155}\text{Dy}(\epsilon)$	
$^{156\text{g}}\text{Tb}$	5.35 d	$\epsilon + \beta^+$ (100)	199.19	41(5)	$^{154}\text{Gd}(\alpha, d)$	-14.3
			262.54	5.8(6)	$^{155}\text{Gd}(\alpha, t)$	-14.5
			296.49	4.5(4)	$^{156}\text{Gd}(\alpha, nt)$	-23.0
			356.38	13.6(13)	$^{157}\text{Gd}(\alpha, 2nt)$	-29.4
			422.34	8.0(8)	$^{158}\text{Gd}(\alpha, 3nt)$	-37.3
			534.29	67(6)		
			1065.11	10.8(10)		
			1154.07	10.4(10)		
			1222.44	31(3)		
^{160}Tb	72.3 d	β^- (100)	86.7877	13.2(3)	$^{157}\text{Gd}(\alpha, p)$	-7.9
			298.5783	26.1(6)	$^{158}\text{Gd}(\alpha, d)$	-13.6
			879.378	30.1(6)	$^{160}\text{Gd}(\alpha, nt)$	-20.7
			966.166	25.1(5)		
			1177.954	14.9(3)		
^{161}Tb	6.89 d	β^- (100)	74.56669	10.2(5)	$^{158}\text{Gd}(\alpha, p)$	-8.1
			87.941	0.183(10)	$^{160}\text{Gd}(\alpha, t)$	-13.0
			103.065	0.101(6)	$^{161}\text{Gd}(\beta^-)$	

¹⁵³ Gd	240.4 d	ε (100)	69.673	2.45(7)	¹⁵² Gd(α, ³ He)	-14.3
			97.431	29.0(8)	¹⁵⁴ Gd(α, nα)	-8.9
			103.1801	22.1(3)	¹⁵⁵ Gd(α, 2nα)	-15.3
					¹⁵⁶ Gd(α, 3nα)	-23.9
					¹⁵⁷ Gd(α, 4nα)	-30.2
					¹⁵⁸ Gd(α, 5nα)	-38.2
					¹⁵³ Tb(ε)	

The self-absorption of low energy γ rays in the ^{nat}Gd foils were corrected using the following formula [16]

$$A(E) = A_0(E) \frac{\rho[\mu(E)/\rho]d}{1 - \exp[-\rho(\mu(E)/\rho)d]}, \quad (1)$$

where $A_0(E)$ is the measured activity for the γ ray at the energy E , $A(E)$ is the corrected activity, d is the thickness of the ^{nat}Gd foil, ρ is the density of the foil, $\mu(E)/\rho$ is the mass attenuation coefficient for the γ ray at the energy E taken from [17].

3. Result and discussion

The cross sections of the ^{nat}Ti(α, x)⁵¹Cr monitor reaction were derived using the γ -ray line at 320.1 keV ($I_\gamma = 9.91\%$). The derived cross sections were compared with the recommended values from IAEA [18]. Based on the comparison, an energy shift toward the lower energy was found. Therefore, the measured target thickness of the ^{nat}Gd foil was corrected within its uncertainty by decreasing 3%. The corrected cross sections of the monitor reaction are shown in Fig. 1. We could obtain a good agreement between our experimental result and the recommended values. The measured thickness of the ^{nat}Ti foil and beam intensity were adopted for the following analysis.

Production cross sections of ^{155,157,159}Dy, ^{153,155,156g,160,161}Tb and ¹⁵³Gd in the alpha-particle-induced reactions on ^{nat}Gd were determined. The numerical data are tabulated in Table 2 and graphically shown in Figs. 2-10 in comparison with the previous experimental data [5,6] and the TENDL-2019 data [9]. The total uncertainties (8.6-29.3%) were estimated from the square root of the quadratic summation of each component; statistical uncertainty (0.02-27.7%), target thickness (3%), target purity (1%), beam intensity (5%), detector efficiency (6%), and γ -ray intensity (3-18%).

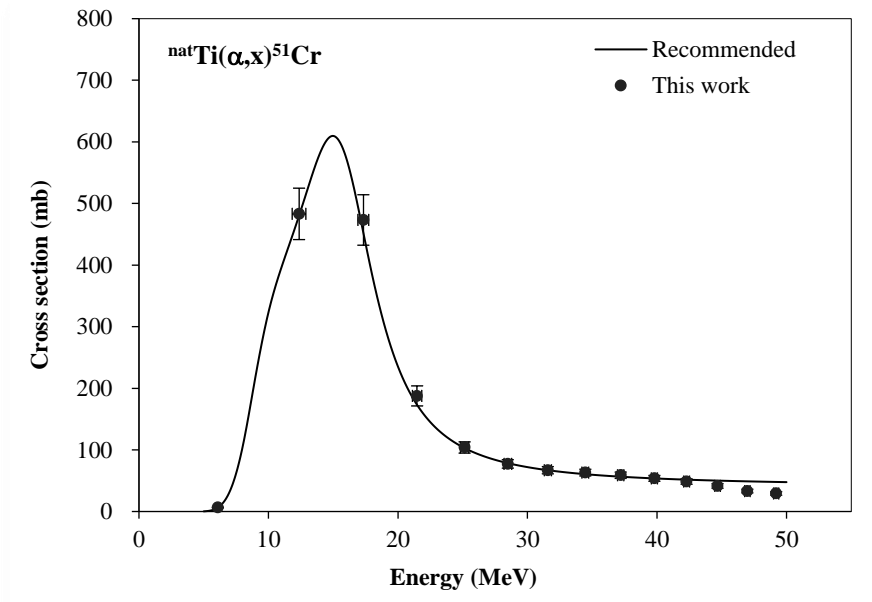


Fig. 1. Excitation function of the ${}^{\text{nat}}\text{Ti}(\alpha, x){}^{51}\text{Cr}$ monitor reaction with the recommended values [18].

Table 2. Production cross sections obtained in the experiment

Energy (MeV)	${}^{155}\text{Dy}$ (mb)	${}^{157}\text{Dy}$ (mb)	${}^{159}\text{Dy}$ (mb)	${}^{153}\text{Tb}$ (mb)	${}^{155}\text{Tb}$ (mb)	${}^{156g}\text{Tb}$ (mb)	${}^{160}\text{Tb}$ (mb)	${}^{161}\text{Tb}$ (mb)	${}^{153}\text{Gd}$ (mb)
50.3±0.9	233±20	367±32	300±32	2.25±0.25	234±23	24.0±3.0	18.9±1.7	18.0±1.9	10.6±1.1
48.1±0.9	205±18	342±30	248±26	2.03±0.22	209±21	19.7±2.5	15.6±1.4	17.8±1.8	9.39±0.94
45.8±0.9	183±16	351±30	212±22	2.61±0.28	189±19	16.2±2.0	13.0±1.2	18.9±1.9	9.75±0.95
43.4±0.9	142±12	371±32	202±22	2.91±0.31	144±14	12.5±1.6	11.5±1.0	16.5±1.7	7.61±0.77
41.0±1.0	80.7±7.0	355±31	203±21	2.53±0.27	75.9±7.6	6.98±0.86	8.77±0.79	12.5±1.2	5.75±0.54
38.5±1.0	42.6±3.7	343±30	279±29	2.21±0.24	39.9±4.0	4.86±0.61	7.67±0.69	10.1±1.0	3.24±0.32
35.8±1.1	25.6±2.3	324±28	350±36	1.54±0.17	24.3±2.5	3.22±0.41	7.47±0.67	7.88±0.79	2.16±0.23
33.0±1.1	13.1±1.2	262±23	346±36	0.494±0.06	12.8±1.3	1.65±0.22	5.69±0.52	5.78±0.58	0.541±0.101
30.0±1.2	3.33±0.31	215±19	312±32	0.0608±0.011	2.91±0.31	0.907±0.129	3.63±0.34	4.16±0.42	
26.8±1.3		162±14	216±22		0.405±0.082	0.255±0.061	1.81±0.18	2.84±0.31	
23.3±1.4		115±10	134±14		0.738±0.108			1.09±0.15	
19.4±1.6		38.2±3.4	78.5±8.1		0.290±0.041			0.190±0.056	
14.8±1.9		0.352±0.031							

3.1 ${}^{\text{nat}}\text{Gd}(\alpha, x){}^{155}\text{Dy}$ reaction

The activation cross sections of the ${}^{\text{nat}}\text{Gd}(\alpha, x){}^{155}\text{Dy}$ reaction were measured for the first time as far as we know. The measurements of the γ -ray line at 226.9 keV ($I_{\gamma} = 68.4\%$) from the ${}^{155}\text{Dy}$ decay ($T_{1/2} =$

9.9 h) after cooling times of 14.3-17.6 h were adopted. We could obtain the cross sections in the energy range from 30 to 50 MeV. Our result is compared with the TENDL-2019 data [9] as shown in Fig. 2. The TENDL-2019 data somewhat overestimates our data between 35 and 45 MeV.

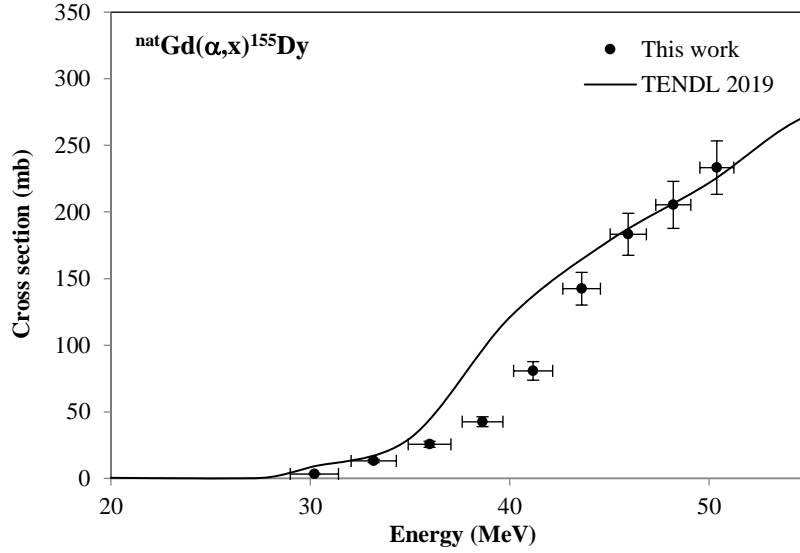


Fig. 2. Excitation function of the ${}^{\text{nat}}\text{Gd}(\alpha,x){}^{155}\text{Dy}$ reaction

3.2 ${}^{\text{nat}}\text{Gd}(\alpha,x){}^{157}\text{Dy}$ reaction

The excitation function of the ${}^{\text{nat}}\text{Gd}(\alpha,x){}^{157}\text{Dy}$ reaction was determined based on the γ -ray line at 326.3 keV ($I_{\gamma} = 93.0\%$) from the decay of ${}^{157}\text{Dy}$ ($T_{1/2} = 8.14$ h). The measurements after cooling times of 14.3-17.6 h were used to derive the cross sections. We could obtain the cross sections in the energy range from 15 to 50 MeV. Our result is shown in Fig. 3 in comparison with the previous experimental study [6] and the TENDL-2019 data [9]. Our result is in good agreement with the experimental data. The TENDL-2019 data show two peaks contributed from the reactions on different Gd isotopes while ours monotonically increases up to 44 MeV. The TENDL-2019 data underestimate most of our experimental data above 25 MeV although they agree with ours below 25 MeV.

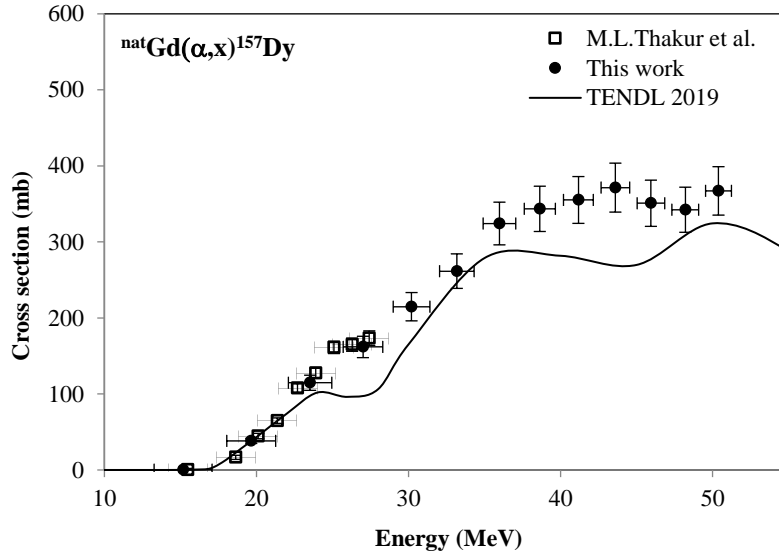


Fig. 3. Excitation function of the $^{nat}\text{Gd}(\alpha,x)^{157}\text{Dy}$ reaction

3.3 The $^{nat}\text{Gd}(\alpha,x)^{159}\text{Dy}$ reaction

The cross sections of the $^{nat}\text{Gd}(\alpha,x)^{159}\text{Dy}$ reaction were determined using the γ -ray line at 58.0 keV ($I_\gamma = 2.27\%$) from the ^{159}Dy decay ($T_{1/2} = 144.4$ d). To reduce γ -ray backgrounds from the short-lived radionuclides, the measurements were executed after cooling times of 25.4-28.8 d. The mass attenuation coefficient adopted for the γ -ray line at 58.0 keV was $12.8 \text{ cm}^2/\text{g}$ [17]. The correction factor calculated from Eq. (1) is 1.17. The cross sections for the ^{159}Dy production derived from the corrected activities are presented with the TENDL-2019 data in Fig. 4. Our experimental data have peaks at around 35 and above 50 MeV. The peak amplitude and positions of the TENDL-2019 data agree with our data. There are no previous experimental data in our literature survey.

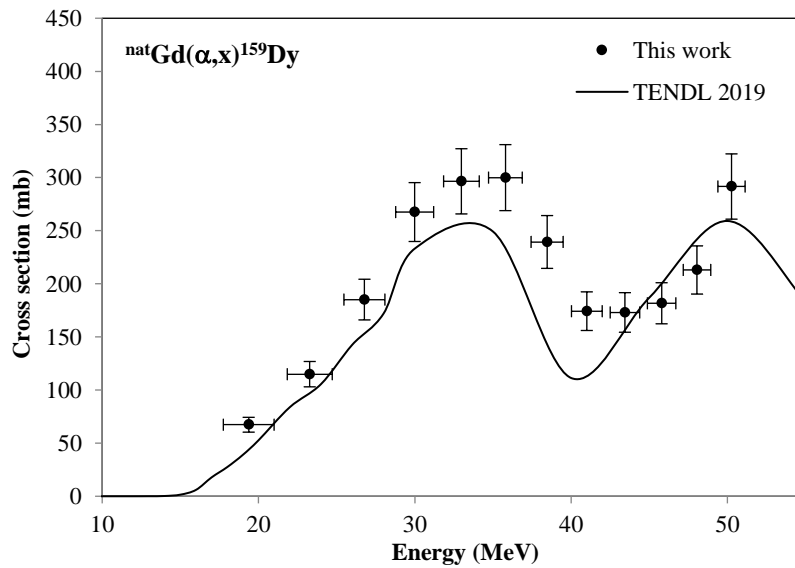


Fig. 4. Excitation function of the $^{nat}\text{Gd}(\alpha,x)^{159}\text{Dy}$ reaction

3.4 $^{nat}\text{Gd}(\alpha, x)^{153}\text{Tb}$ reaction

The radionuclide ^{153}Tb has the half-life of $T_{1/2} = 2.34$ d. The measurements of the 212.048-keV γ -ray line ($I_\gamma = 31\%$) from the ^{153}Tb decay were performed after cooling times of 2.4–2.8 d. The production of its parent ^{153}Dy ($T_{1/2} = 6.4$ h) was energetically possible but not identified because no peaks of the specific γ -lines were found. The cumulative cross sections of the $^{nat}\text{Gd}(\alpha, x)^{153}\text{Tb}$ reaction are shown in Fig. 5 together with the experimental data by Gayoso et al. [5] and the TENDL-2019 data [9]. Our experimental result has a peak around 44 MeV, which is different from the data of Gayoso et al. The TENDL-2019 data are very different from the two experimental data.

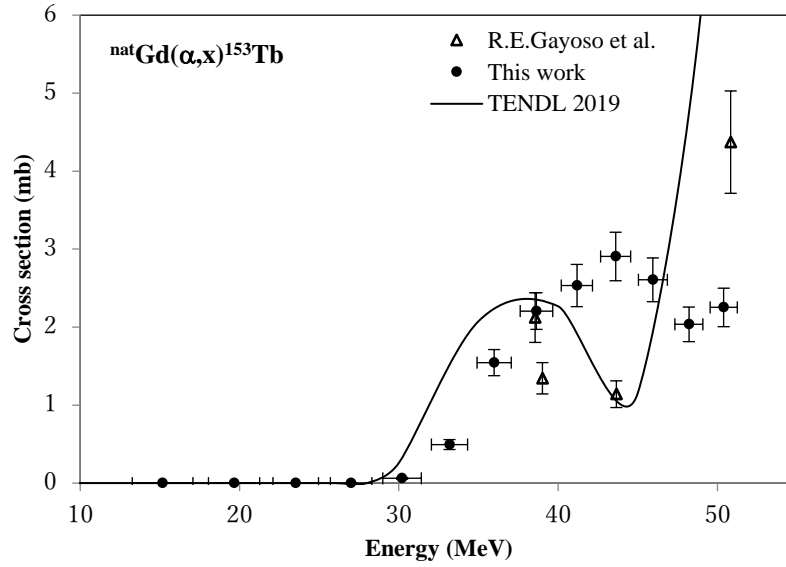


Fig. 5. Excitation function of the $^{nat}\text{Gd}(\alpha, x)^{153}\text{Tb}$ reaction

3.5 $^{nat}\text{Gd}(\alpha, x)^{155}\text{Tb}$ reaction

The γ -ray line at 105.3 keV ($I_\gamma = 25.1\%$) emitted with the ^{155}Tb decay ($T_{1/2} = 5.32$ d) was measured to derive the cross sections of the $^{nat}\text{Gd}(\alpha, x)^{155}\text{Tb}$ reaction after cooling times of 25.4–28.8 d. The cooling time is long enough for the complete decay of the parent nuclide ^{155}Dy ($T_{1/2} = 9.9$ h). The mass attenuation coefficient adopted for the γ -ray line is 2.74 cm^2/g [17] and the calculated correction factor from Eq. (1) is 1.03. The corrected cross sections are presented in Fig. 6 with those in the earlier study [5] and the TENDL-2019 data [9]. Our result shows acceptable agreement with the TENDL-2019 data except the middle energy region of 35–45 MeV. The previous experimental data are smaller than ours above 40 MeV.

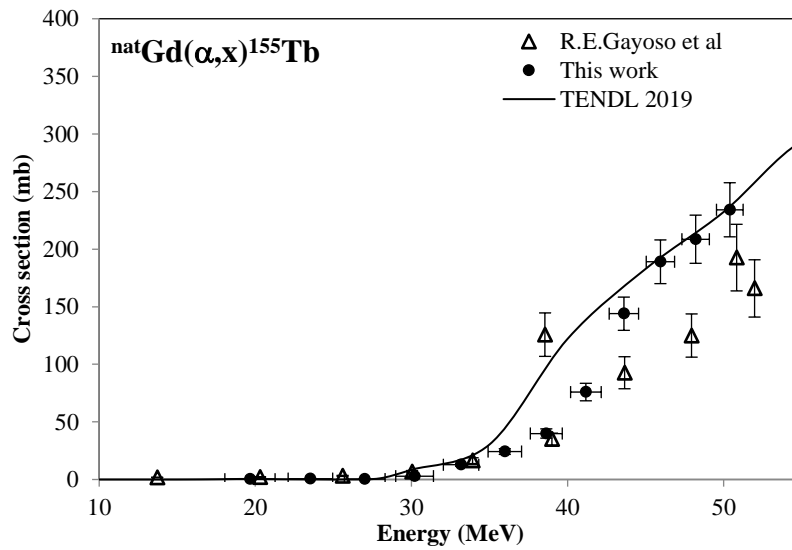


Fig. 6. Excitation function of the $^{nat}\text{Gd}(\alpha, x)^{155}\text{Tb}$ reaction

3.6 $^{nat}\text{Gd}(\alpha, x)^{156g}\text{Tb}$ reaction

The production cross sections of ^{156g}Tb ($T_{1/2} = 5.35$ d) are shown in Fig.7. The measurement of 534.29-keV γ -ray line ($I_{\gamma} = 67\%$) from the ^{156g}Tb decay after cooling times of 25.4-28.8 d was adopted to derive the cross sections. The γ -ray lines from the two isomers at level energy of 0.0496 ($T_{1/2} = 24.4$ h) and 0.0884 MeV ($T_{1/2} = 5.3$ h) could not be identified in the spectra. During the cooling times, the isomers decayed to ^{156g}Tb , and thus the derived cross sections of ^{156g}Tb were cumulative. Our results show a good agreement with the experimental data by Gayoso et al. [5] and the TENDL-2019 data in the energy region.

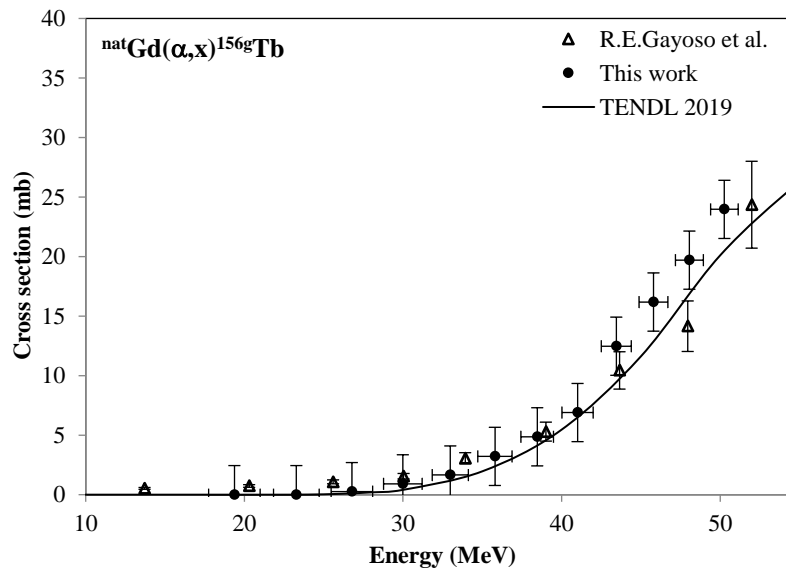


Fig. 7. Excitation function of the $^{nat}\text{Gd}(\alpha, x)^{156g}\text{Tb}$ reaction

3.7 $^{nat}\text{Gd}(\alpha,x)^{160}\text{Tb}$ reaction

The radionuclide ^{160}Tb has the half-life of $T_{1/2} = 72.3$ d. The measurements of the 879.301-keV γ -ray line ($I_\gamma = 30.1\%$) from the ^{160}Tb decay were used to derive the production cross sections after cooling times of 25.4-28.8 d. The derived cross sections are shown in Fig. 8 together with one experimental dataset by Gayoso et al. [5] and the TENDL-2019 data [9]. Both are lower than our results.

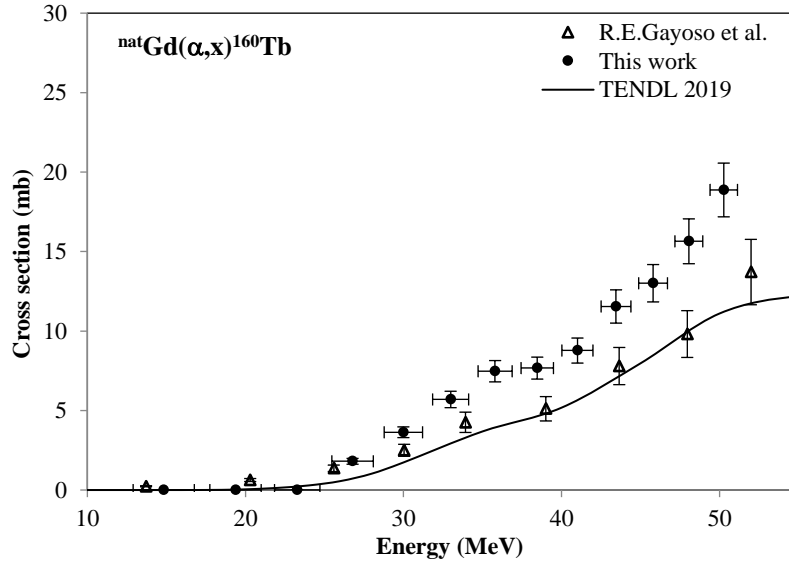


Fig. 8. Excitation function of the $^{nat}\text{Gd}(\alpha,x)^{160}\text{Tb}$ reaction

3.8 $^{nat}\text{Gd}(\alpha,x)^{161}\text{Tb}$ reaction

The excitation function of the $^{nat}\text{Gd}(\alpha,x)^{161}\text{Tb}$ reaction was determined based on the γ -ray line at 74.56 keV ($I_\gamma = 10.2\%$) from the decay of ^{161}Tb ($T_{1/2} = 6.89$ d). The measurements after cooling times of 25.4-28.8 d were used to derive the cross sections. During the cooling time, the parent nuclide ^{161}Gd ($T_{1/2} = 3.66$ min) decayed to ^{161}Tb . The correction factor calculated from Eq. (1) for the γ -ray line is 1.09 using the mass attenuation coefficient of $6.69\text{ cm}^2/\text{g}$ [17]. The cumulative cross sections using the corrected activities are compared with the data of Gayoso et al. [5] and the TENDL-2019 data [9] as shown in Fig. 9. Our data are higher than those of Gayoso et al. and the TENDL-2019 data by about a factor of two.

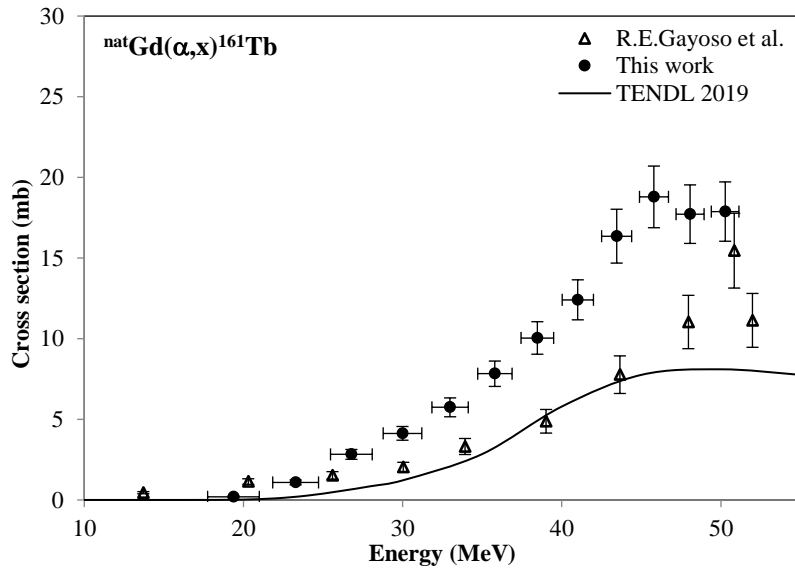


Fig. 9. Excitation function of the ${}^{\text{nat}}\text{Gd}(\alpha, x){}^{161}\text{Tb}$ reaction

3.9 ${}^{\text{nat}}\text{Gd}(\alpha, x){}^{153}\text{Gd}$ reaction

The production cross sections of ${}^{153}\text{Gd}$ ($T_{1/2} = 240.4$ d) using the γ -ray line at 97.431-keV ($I_{\gamma} = 29\%$) are shown in Fig.10. The co-produced ${}^{153}\text{Dy}$ and ${}^{153}\text{Tb}$ decayed to ${}^{153}\text{Gd}$ during cooling times of 25.4-28.8 d. The activities were corrected by the correction factor of 1.04 calculated from Eq. (1) and the mass attenuation coefficient of $3.35 \text{ cm}^2/\text{g}$ [17]. Cumulative cross sections were derived from the corrected activities. We compared our result with the TENDL-2019 data [9]. The TENDL-2019 data are slightly larger than ours. No experimental data studied earlier for the reaction were found in our survey.

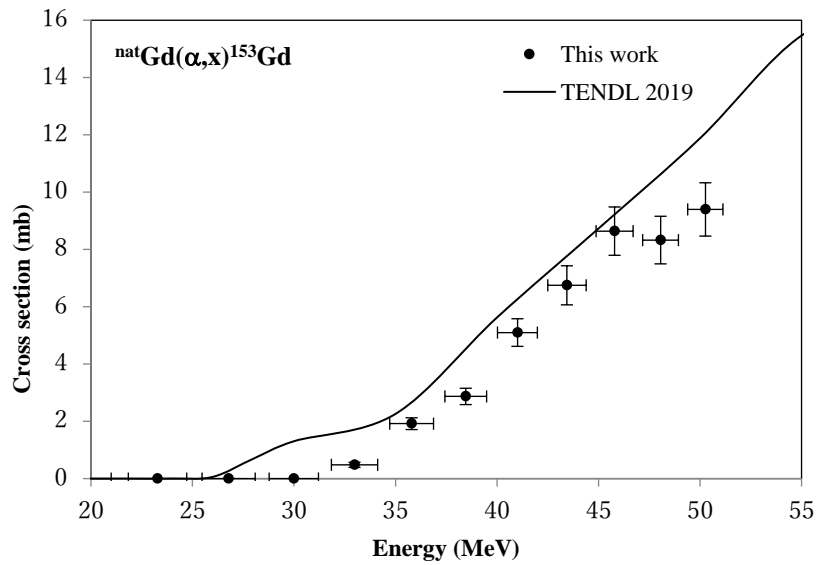


Fig. 10. Excitation function of the ${}^{\text{nat}}\text{Gd}(\alpha, x){}^{153}\text{Gd}$ reaction

4. Summary

We investigated the production cross sections of $^{155,157,159}\text{Dy}$ and $^{153,155,156\text{g},160,161}\text{Tb}$ and ^{153}Gd in the alpha-particle-induced reactions on $^{\text{nat}}\text{Gd}$ up to 50 MeV at the RIKEN AVF cyclotron. The cross sections of $^{155,159}\text{Dy}$, and ^{153}Gd were measured for the first time. The stacked foil technique, the activation method and the high-resolution γ -ray spectrometry were used. The results were compared with the previous experimental data [5,6] and the TENDL-2019 data based on calculation using the theoretical model code TALYS [9]. We found that our data agree with the result of Thakur et al. but data of Gayoso et al. in most of the cases are lower than our results, which systematic deviation can be explained with a systematic error in their experiment. The TALYS calculation provided more or less the same shape for the investigated excitation functions with some differences in the structure and the amplitude of the curves.

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Declarations of interest

None

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