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Measurement of excitation functions of the alpha particle-induced nuclear reactions up to 50 MeV on holmium target

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Abstract ^{165}Er is a pure Auger-electron emitter that shows great potential for therapeutic applications in nuclear medicine. Various production methods have been described in the literature, typically involving the irradiation of erbium (Er) and holmium (Ho) targets with charged particles. This paper reports on the study of the indirect production of ^{165}Er through the ^{165}Ho(α,4n)^{165}Tm reaction. Holmium foils have been irradiated with a 50 MeV alpha beam, and cross sections for producing ^{165,166,167,168}Tm were determined using the activation method and stacked target technique, from the threshold to the maximum bombarding energy.

1 Introduction

Auger electron-emitting radionuclides are of increasing interest as therapeutic agents due to their potentially high levels of cytotoxicity [1]. This is attributed to the high linear energy transfer and low range of the Auger electrons in tissue, which limits their toxicity in areas beyond the targeted cells [2]. These characteristics make Auger-electron-emitting radionuclides particularly well-suited for treatment of small metastatic tumor lesions. ^{165}Er displays promising characteristics due to its decay via electron capture, which results in the emission of Auger electrons at energies of 5.3 keV (65.6%) and 38.4 keV (4.8%). A potential production method involves the reaction ^{165}Ho(α,4n)^{165}Tm, which leads to the formation of ^{165}Er. To choose the optimal alpha beam energy for isotope production, it is crucial to understand the reaction cross-section of all isotopes generated in the different reactions induced by alpha particles in the Ho target. However, the experimental cross-section values reported in the literature for the (α,x) reaction are quite inconsistent.

Natural holmium is composed of one primordial isotope (^{165}Ho). It is the only isotope of holmium believed to be stable, simplifying the selective production of the desired isotope and the evaluation of experimental data. Holmium is a relatively soft and malleable rare-earth element that is corrosion-resistant and chemically stable in dry air at standard temperature and pressure, making it safe to handle in its solid form.

2 Experiment

The experiment was conducted at the RIKEN AVF cyclotron using a nominal alpha beam energy of 50 MeV. The actual beam energy was measured using the time-of-flight (TOF) method [3], which confirmed the bombarding energy to be 50.8 MeV. A stack of holmium, titanium and aluminum foils of different thicknesses (12.6 μm for Ho, 11.0 μm and 5.2 μm for Ti, and 6.7 μm for Al) was irradiated with a beam current of 200 nA for 30 min. To measure the excitation function,

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we employed the well-established stacked foil technique, combined with off-line high-resolution gamma spectrometry. The titanium foils acted as monitor foils, based on the $^{nat}Ti(\alpha,x)^{51}Cr$ monitor reaction. The stack consisted of a total of 25 holmium foils and 25 titanium foils. The measurement process extended for about three months, starting soon after the end of the irradiation. We conducted 10-min measurements to capture short-lived products and extended measurements lasting up to 24 h to observe long-lived products across several series. In total, five series of measurements were performed for the holmium foils and one series for the titanium monitor foils.

3 Results and discussion

Four thulium radioisotopes were identified in the measured spectra: ^{165}Tm, ^{166}Tm, ^{167}Tm, and ^{168}Tm. The final data were compiled from various measurement runs, taking into account their half-lives and cross-sections. The nuclear data for these isotopes were sourced from the NuDat online database [4, 5] and the Q-value calculator [6], as shown in Table 1. This table highlights the lowest threshold energies. The cross-section values for the excitation function were calculated from the measured peak areas and nuclear parameters, then averaged across the strongest measured peaks listed in Table 1 and the different measurement runs where these peaks were detected. The activation cross sections have been calculated using Eq. (1):

$$\sigma = \frac{\lambda N_\gamma}{\varepsilon_d I_\gamma \varepsilon_t F_t N_b (1 - e^{-\lambda t_i}) e^{-\lambda t_c} (1 - e^{-\lambda t_m})} \quad (1)$$

where N_γ is the measured net-peak area (counts), λ is the decay constant (s^{-1}), ε_d is the detector efficiency, I_γ is the gamma-ray intensity, ε_t is the dead-time correction parameter, F_t is the surface density of target atoms (cm^{-2}), N_b is the number of projectiles per unit time (s^{-1}), t_i is the irradiation time (s), t_c is the cooling time measured from EOB (End of Bombardment) to the start of the measurement (s), and t_m is the acquisition time (s) (real-time).

The measured cross sections, along with the TENDL 2023 predictions and experimental literature data, are presented in Figs. 2, 3, 4, 5 and numerically in Table 2.

3.1 Monitor reaction

The reference cross-section for the $^{nat}Ti(\alpha,x)^{51}Cr$ reaction was obtained from the IAEA monitor reaction database [7]. The thickness of the foil was measured and recorded as 5.2 ± 0.1 mm, and this value was used in the data evaluation instead of the nominal thickness. To account for the escape of secondary electrons from the target, the beam intensity was adjusted with a multiplier of 0.935. As a result, our measured monitor excitation function aligns closely with the reference curve from 2017 (Fig. 1).

3.2 $^{165}Ho(\alpha,4n)^{165}Tm$ reaction

The evaluation of the measured data was made based on the 806.312 keV γ-line. The numerical results are presented in Table 2, while the measured excitation function is shown in Fig. 2. Earlier datasets were found in the literature from Martin [8], Sau [9], Rama Rao [10], Mukherjee [11], Singh [12], Gadkari [13], Tárkányi [14], Usman [15] and Kormazeva [16]. Our new data show good agreement with the TENDL 2023 forecast [17], but the previous experimental data systematically underestimate the cross section at its maximum value.

3.3 $^{165}Ho(\alpha,3n)^{166}Tm$ reaction

The evaluation of the measured data was made based on the 705.333 keV, 778.814 keV, 785.904 keV and 1273.54 keV γ-lines. The numerical results are presented in Table 2, while the measured excitation function is shown in Fig. 3. Several earlier measurements were found in the literature [8, 9, 11–16, 18, 19]. In Fig. 3, the TENDL 2023 prediction [17] is also shown. Our new data are in good agreement with those of Tárkányi [14] up to 35 MeV. TENDL 2023 gives a good trend, but around the maximum, it slightly underestimates our results.

3.4 $^{165}Ho(\alpha,2n)^{167}Tm$ reaction

The evaluation of the measured data was made based on the 207.801 keV γ-line. Measured data from Tárkányi [14] and Kormazeva [16] are consistent with our results. All the other experimental data from Martin [8], Sau [9], Rama Rao [10], Mukherjee [11], Singh [12], Gadkari [13], Usman [15], Homma [18], Singh [19], Wilkinson [20] and also the TENDL prediction [17] provide smaller cross-section data (Fig. 4).

3.5 $^{165}Ho(\alpha,n)^{168}Tm$ reaction

The relatively long half-life ^{168}Tm isotope was investigated approximately three months after the irradiation. The evaluation was conducted using the average of two clean (447.512 keV and 815.989 keV) gamma lines (Fig. 5). The results of previous cross-section measurements show a large dispersion for this isotope [8, 9, 11–16, 18–20, 24]. Although the results obtained from the present experiment are approximately in the middle range of previous measurement data, they do not match any previous data set.

Table 1 Parameters of the measured isotopes

Nuclide	Half-life	Decay mode (%)	Eγ (keV)	Iγ (%)	Reaction	E_{th} (MeV)
^{51}Cr	27.701 d	ε (100)	320.0824	9.91	$^{nat}Ti(\alpha,x)^{51}Cr$	2.893
^{165}Tm	30.06 h	ε + β^+ (100)	806.372	9.5	$^{165}Ho(\alpha,4n)$	32.36
^{166}Tm	7.7 h	ε + β^+ (100)	705.333	11.1	$^{165}Ho(\alpha,3n)$	25.21
			778.814	19.1		
			785.904	10		
			1273.55	15		
^{167}Tm	9.28 d	ε (100)	207.801	41.3	$^{165}Ho(\alpha,2n)$	16.34
^{168}Tm	93.1 d	ε + β^+ (99.99)	447.515	23.98	$^{165}Ho(\alpha,n)$	9.39
		β^- (0.01)	815.989	50.95		

Table 2 Measured cross sections of the thulium radioisotopes

Energy (MeV)		$^{165}Ho(\alpha,n)^{168}Tm$ Cross section (mb)		$^{165}Ho(\alpha,2n)^{167}Tm$		$^{165}Ho(\alpha,3n)^{166}Tm$		$^{165}Ho(\alpha,4n)^{165}Tm$	
E	dE	σ	dσ	σ	dσ	σ	dσ	σ	dσ
50.43	0.20	3.11	0.28	40.50	4.12	188.72	14.51	1207.16	91.81
49.69	0.21	3.15	0.28	45.31	4.49	204.66	15.74	1279.21	97.30
48.95	0.21	3.50	0.30	42.94	4.43	226.92	17.43	1340.94	101.97
48.19	0.22	3.50	0.28	48.46	4.76	245.17	18.82	1328.49	101.05
47.43	0.23	3.92	0.32	56.96	8.03	246.55	18.90	1343.19	102.17
45.38	0.27	4.31	0.37	61.26	8.29	414.60	31.57	1186.99	90.29
44.35	0.28	4.78	0.40	76.01	9.67	487.87	37.16	1152.49	87.80
42.78	0.33	5.44	0.46	79.11	9.71	673.21	51.65	956.40	73.14
41.72	0.35	5.83	0.48	91.16	10.52	808.59	61.96	767.05	58.84
40.07	0.41	6.07	0.50	103.99	11.62	1030.34	78.64	448.88	34.84
38.96	0.44	7.06	0.57	70.01	13.28	1166.05	88.93	292.26	23.20
37.23	0.50	7.48	0.61	106.72	17.09	1191.74	90.84	91.38	8.32
36.05	0.54	8.59	0.69	130.12	18.35	1253.90	95.50	34.74	4.50
34.22	0.62	10.29	0.82	264.80	32.70	1212.29	92.32		
32.97	0.67	11.13	0.88	327.93	32.90	1066.86	81.32		
31.01	0.77	12.94	1.00	572.77	48.08	741.43	56.22		
29.67	0.82	13.70	1.05	781.01	61.90	445.91	33.91		
27.55	0.95	16.42	1.27	1103.98	84.37	79.70	6.18		
26.09	1.02	19.27	1.50	1068.31	81.46	3.41	0.44		
23.35	1.21	28.08	2.18	708.95	54.33				
21.71	1.30	42.81	3.28	523.77	40.31				
18.55	1.56	72.09	5.49	98.27	8.29				
16.63	1.67	28.77	2.22	3.69	0.46				
13.36	1.92	0.38	0.10						

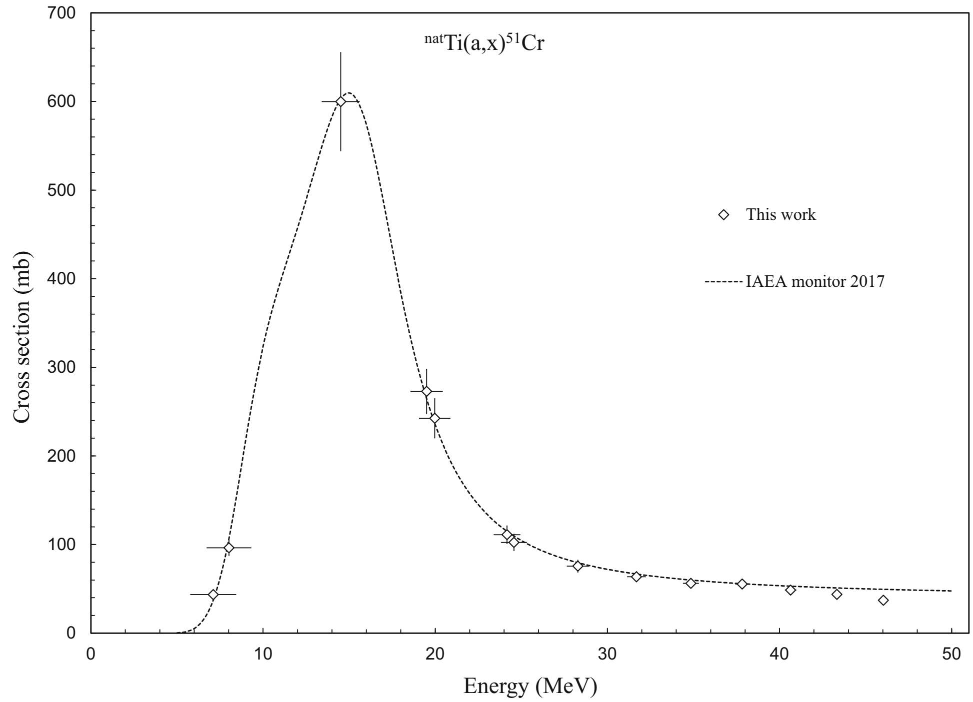

Fig. 1 Comparison of the measured monitor reaction to the reference cross section from the IAEA Monitor Reaction database [7]

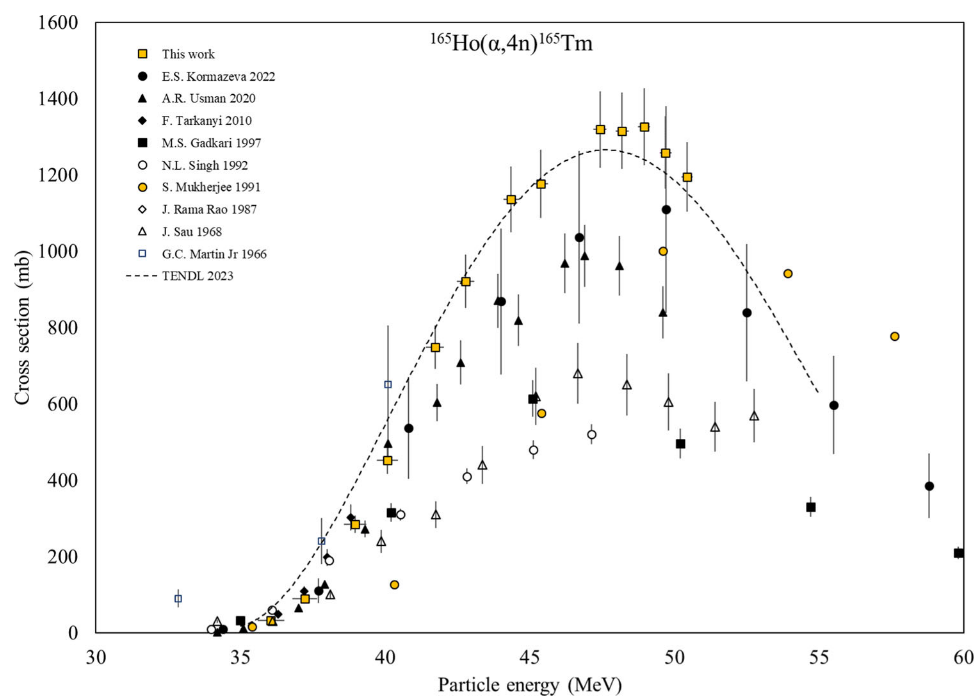

Fig. 2 Measured cross section of the ^{165}Ho(α,4n)^{165}Tm nuclear reaction compared with the TENDL 2023 prediction and the literature data

3.6 Physical yield calculation

The measured cross sections have been used to calculate the integral physical yields for all nuclear reactions, as shown in Fig. 6. The integral thick target yields were determined using the formula provided in Eq. 2 [21, 22].

$$Y_{TT} = \frac{\lambda}{Ze} \int_{0}^{E_b} \frac{\sigma(E)}{S(E)} \quad (2)$$

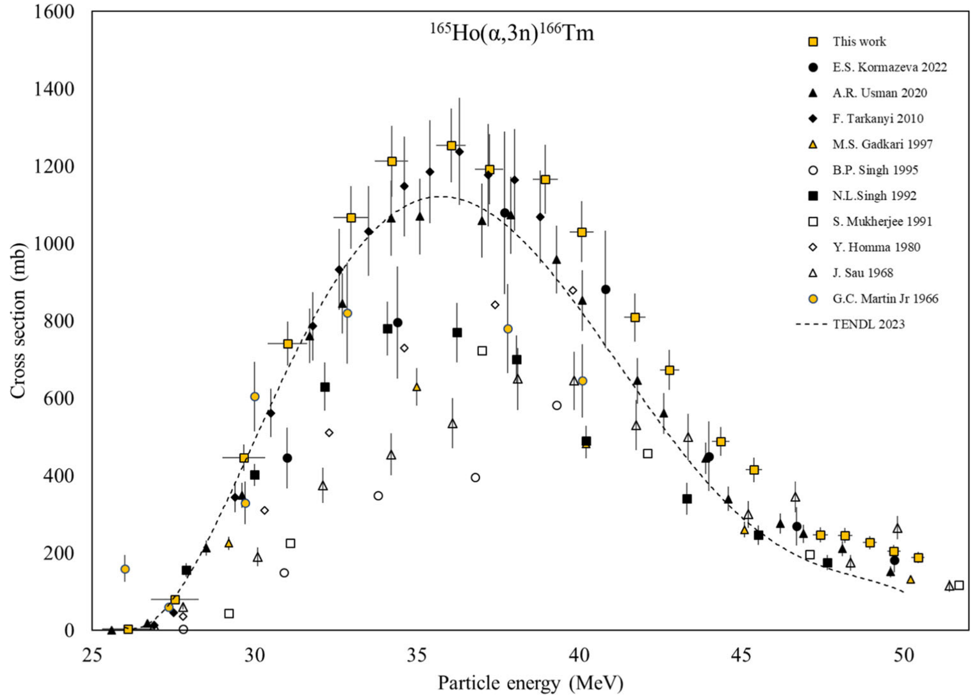

Fig. 3 Measured cross section of the ^{165}Ho(α,3n)^{166}Tm nuclear reaction compared with the TENDL 2023 prediction and the literature data

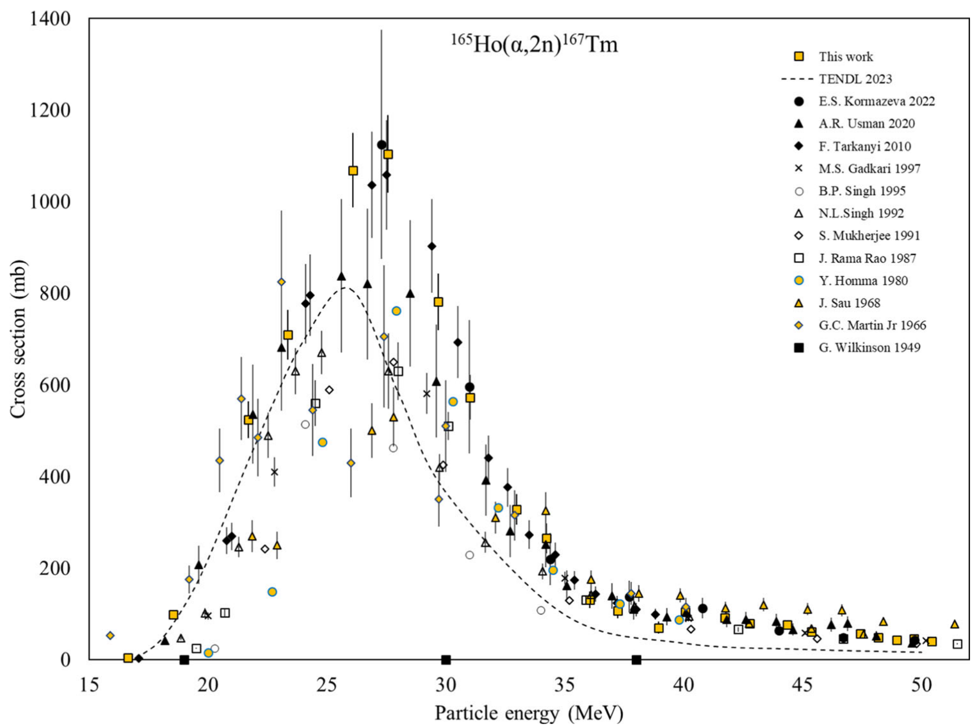

Fig. 4 Measured cross section of the ^{165}Ho(α,2n)^{167}Tm nuclear reaction compared with the TENDL 2023 prediction and the literature data

Dmitriev et al. [23] published experimental yield data for all the investigated reactions, while Kormazeva et al. [16] provided data for ^{167}Tm. Their results are fairly consistent with our calculated curves, as shown in Fig. 6.

4 Conclusion

Our research has demonstrated that within the 50 MeV energy range, the medically significant isotope ^{165}Er can be indirectly produced by alpha particles through the reaction ^{165}Ho(α,4n)^{165}Tm, which shows a reasonable cross-section

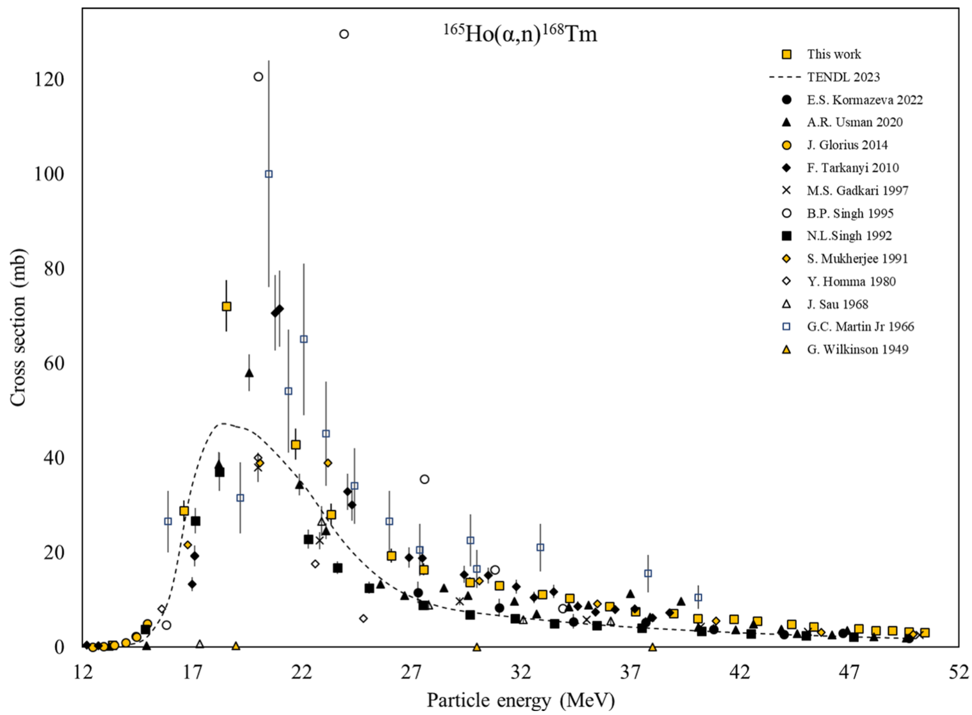

Fig. 5 Measured cross section of the ^{165}Ho(α,n)^{168}Tm nuclear reaction compared with the TENDL 2023 prediction and the earlier literature data

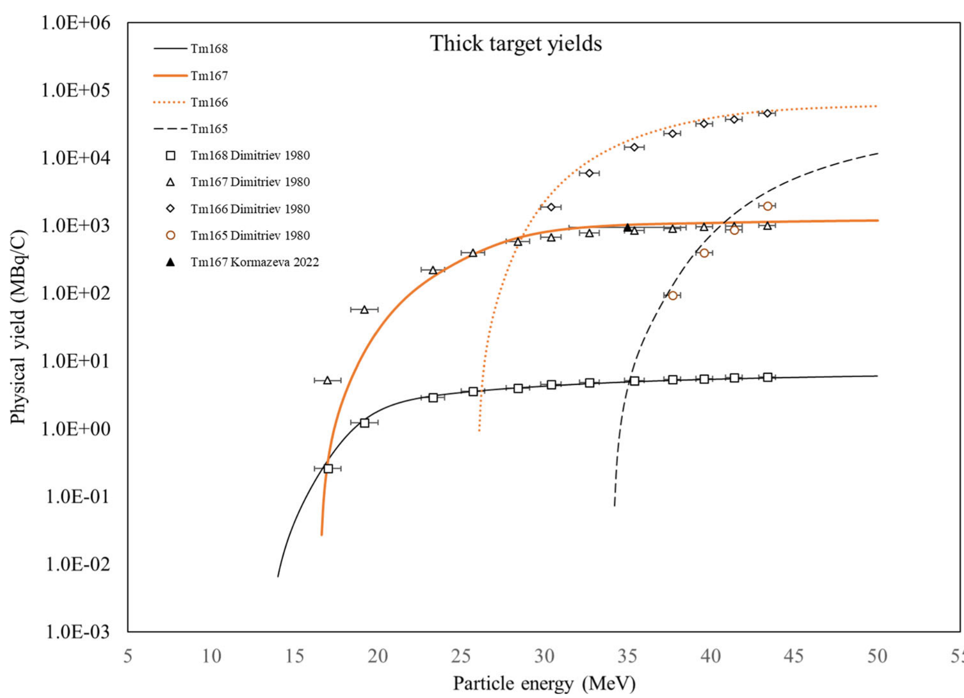

Fig. 6 The calculated physical integral yield from our measured cross sections in comparison with values from the literature.

and production yield. The recent measurements either confirm or refute some earlier experimental results and provide new data in several instances. Additionally, comparing these results with the latest TENDL 2023 predictions will assist the TALYS model developers in refining their algorithms.

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Declarations

Conflict of interest All authors have no conflicts of interest.

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