Production cross sections of ^{169}Yb and Tm isotopes in deuteron-induced reactions on ^{169}Tm

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Abstract

The excitation functions of deuteron-induced reactions on ^{169}Tm were measured using the stacked-foil method and high resolution gamma-ray spectrometry. The production cross sections of a medical radionuclide ^{169}Yb were investigated. The result was compared with the previous experiments and found to be in good agreement. In addition to ^{169}Yb, the production cross sections of Tm isotopes, ^{170}Tm, ^{168}Tm and ^{167}Tm, were measured. These results were compared with the TALYS calculations taken from the TENDL-2015 online data library.

Introduction

Radionuclides are used in many application fields such as engineering and medicine. The best route for production of a certain radionuclide among variety of nuclear reactions can be discussed and selected based on the excitation functions of the different nuclear reactions. The cross sections of the selected nuclear reaction induced by neutrons and/or light charged-particles are therefore fundamental information for applications. However, information of the cross sections of many nuclear reactions is still insufficient, i.e., the amount of experimental cross section data are not enough or the quality of the data is not satisfactory at present even though great efforts of experimentalists.

The ^{169}Yb ($T_{1/2}$ = 32.018 d, EC = 100%) radionuclide is an Auger electron and X-ray emitter and suitable for using in brachytherapy (Deland, 1971; Lymperopoulou, 2006). It can be produced through the neutron capture (n,γ) reaction on ^{168}Yb in reactors (Sims,1970; IAEA-TECDOC-1512,2006). To increase the specific activity of the reactor produced ^{169}Yb, an enriched ^{168}Yb target is required, since the abundance of ^{168}Yb in natural ytterbium is only 0.123%. Charged-particle induced reactions on erbium and

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thulium targets are suitable for production of ^{169}Yb with much higher specific activity than using the (n,γ) reaction. The proton- (Birattari, 1973; Spahn, 2005; Tárkányi, 2012), deuteron- (Tárkányi, 2007; Hermanne, 2009, 2016) and α-induced reactions (Mohan Rao, 1992) on monoisotopic ^{169}Tm target were investigated by several research groups earlier. The α-induced reactions on ^{nat}Er (^{164}Er: 1.601%, ^{166}Er: 33.503%, ^{167}Er: 22.869%, ^{168}Er: 26.978%, ^{170}Er: 14.910%) (Király, 2008) and on enriched ^{166}Er (Glorius, 2014) were also studied before. Based on the experimental cross section data measured earlier the deuteron-induced reaction on ^{169}Tm (Tárkányi, 2007; Hermanne, 2009, 2016) is one of the best candidates for production of high specific activity ^{169}Yb due to the large cross section of the (d,2n) reaction. However, the available experimental cross section data for producing the medically important ^{169}Yb radionuclide by deuteron bombardment have relatively large uncertainties and are scattered, therefore the excitation function is not defined properly. In the experiments reported in Refs. (Tárkányi, 2007; Hermanne, 2009, 2016), the cross sections were measured by using thulium oxide (Tm_2O_3) as a target material. Preparation of a homogeneous thin target from the Tm_2O_3 powder is not an easy task, which could explain the scattering of the reported data. In this paper, we report cross section data of the ^{169}Tm(d,2n)^{169}Yb reaction using the pure, thin metallic thulium foils to reduce the uncertainty of the experimental data.

Method

The excitation functions of the deuteron-induced reactions on ^{169}Tm were measured by the stacked-foil activation method using high resolution gamma-ray spectrometry to assess the activity of the irradiated target foils. ^{169}Tm metallic foils (purity: 99%, Goodfellow Co., Ltd., UK) were stacked with ^{nat}Ti (purity: 99.9%, Goodfellow co., ltd., UK) and ^{27}Al foils (purity: >99.95%, Nilaco Corp., Japan) for monitoring the beam parameters and for degrading the beam energy. The average thicknesses of Tm, Ti and Al foils were determined by measuring the surface area and the weight of a larger piece of the foils and found to be 28.65, 4.95 and 13.44 mg/cm^2, respectively.

The irradiation was performed at the AVF cyclotron of the RIKEN RI Beam Factory. The target was irradiated for 75 minutes with 24.36-MeV deuteron beam with an average intensity of 135.6 nA, which was measured by a Faraday cup. To check the beam stability the beam intensity was recorded in every minute. The incident beam energy was measured by time-of-flight method using plastic scintillator monitors (Watanabe, 2014). The beam energy degradation in the stacked target was calculated

using the SRIM code (SRIM) available online. The gamma-ray spectra of the activated foils were measured by HPGe detectors (ORTEC GEM-25185-P and ORTEC GEM35P4-70) and analyzed by Gamma Studio (SEIKO EG&G). These detectors were calibrated by a multiple standard gamma-ray point source, which consisted of, ^{57,60}Co, ^{88}Y, ^{109}Cd, ^{113}Sn ^{137}Cs, ^{139}Ce and ^{241}Am. The distances between the foils and the detectors were kept 1-30 cm to reduce a dead time. Several series of spectra were recorded to be able to follow the decay of the reaction products. Nuclear decay data in Table 1 are taken from the online NuDat 2.6 database (NuDat 2.6, 2011).

Table 1: The investigated reactions and decay data of reaction products

Reaction product	Half-life (d)	Decay mode (%)	E_γ (keV)	I_γ (%)	Contributing reactions	Q-value (MeV)
^{169}Yb	32.018	EC (100)	177.21	22.28(11)[a]	^{169}Tm(d,2n)	-3.91
^{170}Tm	128.6	EC (0.131)	84.25	2.48[b]	^{169}Tm(d,p)	4.37
		β^- (99.869)				
^{168}Tm	93.1	EC (99.990)	815.99	50.95(16)[c]	^{169}Tm(d,t)	-1.78
		β^- (0.010)			^{169}Tm(d,dn)	-8.03
					^{169}Tm(d,p2n)	-10.26
^{167}Tm	9.25	EC (100)	207.80	42(8)[d]	^{169}Tm(d,tn)	-8.62
					^{169}Tm(d,d2n)	-14.87
					^{169}Tm(d,p3n)	-17.10
					^{169}Tm(d,4n)^{167}Yb(→^{167}Tm)	-19.83

[a]Data are taken from Baglin (2008).
[b]Data are taken from Baglin (2002).
[c]Data are taken from Baglin (2010).
[d]Data are taken from Baglin (2000).

Result and Discussion

To double check the beam energy and intensity, the excitation function of the ^{nat}Ti(d,x)^{48}V monitor reaction was first compared with the recommended values provided by IAEA (IAEA-TECDOC 1211, 2007). The activity of ^{48}V ($T_{1/2}$ = 15.9735 d) of the second foil in each pair of Ti foils to nearly cancel the recoil effect was determined by measuring the 983.53 keV (99.98%) and 1312.11 keV (98.2%) γ-lines from its decay. To decrease the contribution of the ^{48}Sc decay ($T_{1/2}$ = 43.67 h) to the total peak area, the measurements were performed after eight days of cooling time. The measured cross sections are shown in Fig. 1 and are found in

agreement with the recommended values: in Fig. 1, the measured beam intensity was normalized by the cross section at 16.1 MeV to fit the recommended values of the monitor reaction which was 5.7% lower than the measured one. The directly measured beam intensity from the Faraday cup measurement was decreased by 5.7%. The higher beam intensity from the Faraday cup measurement can be explained by the escaping secondary electrons from the surface of the irradiated target. In the data evaluation the beam intensity deduced from monitor reaction was adopted.

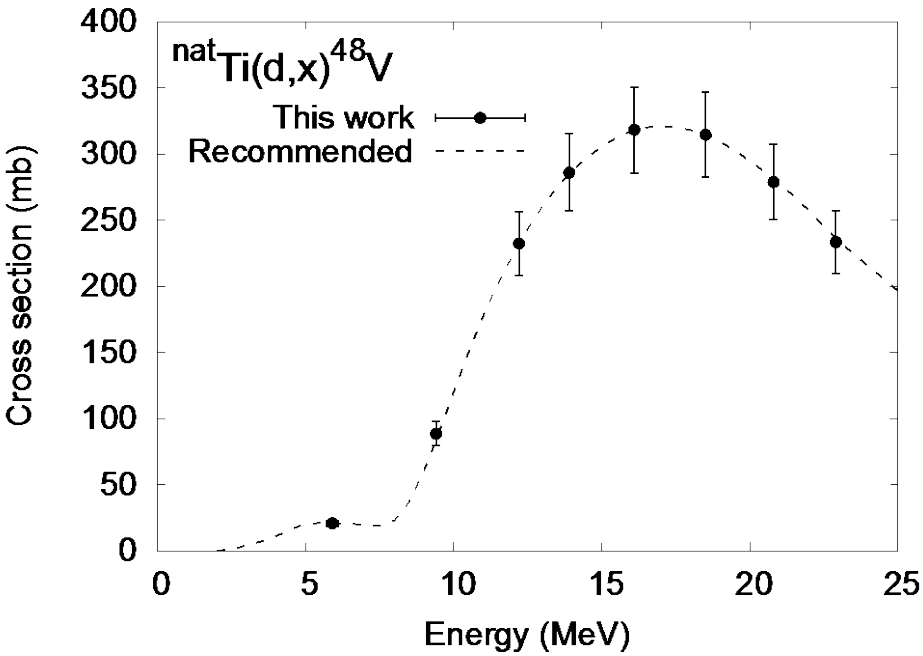

Fig. 1: The excitation function of the monitor reaction ^{nat}Ti(d,x)^{48}V.

From the measured net peak area T_γ, the activation cross sections σ of the deuteron-induced reactions on ^{169}Tm were deduced for ^{169}Yb, ^{170}Tm, ^{168}Tm and ^{167}Tm using the standard activation formula

$$\sigma = \frac{T_\gamma \lambda}{\varepsilon_d \varepsilon_\gamma \varepsilon_t N_t N_b (1-e^{-\lambda t_b}) e^{-\lambda t_c} (1-e^{-\lambda t_m})}, \qquad (1)$$

with the following parameters: surface density of target atoms N_t, number of bombarding particles per unit time N_b, detector efficiency ε_d, gamma-ray abundance ε_γ, measurement dead time ε_t, decay constant λ, bombarding time t_b, cooling time t_c, and acquisition time t_m. The deduced cross section is assigned to the mid energy point of each target foil.

The numerical cross section data as function of bombarding energy for each reaction are presented in Table 2 and graphically in Figs. 2-5. The experimental results

are compared with the previous data and prediction of the TALYS calculations taken from the TENDL-2015 (TENDL-2015, 2015) online data library.

Table 2: Measured cross sections of the ^{169}Tm(d,2n)^{169}Yb and ^{169}Tm(d,x)^{170,168,167}Tm reactions.

Energy (MeV)	^{169}Yb (mb)	^{170}Tm (mb)	^{168}Tm (mb)	^{167}Tm (mb)
24.1 ±0.3	150.2 ±15.4	62.7 ±7.2	97.4 ±10.1	134.6 ±29.1
21.7 ±0.4	192.5 ±19.7	91.1 ±10.1	58.9 ±6.1	5.4 ±1.2
19.5 ±0.4	264.5 ±27.1	110.3 ±12.1	39.4 ±4.2	1.1 ±0.3
17.2 ±0.4	414.7 ±42.4	122.4 ±13.3	32.6 ±3.4	0.3 ±0.09
15.1 ±0.5	615.5 ±62.9	135.2 ±14.8	25.3 ±2.7	0.2 ±0.08
13.5 ±0.5	682.8 ±69.8	154.3 ±16.7	20.5 ±2.3	
11.0 ±0.6	455.6 ±46.6	167.0 ±18.0	7.0 ±0.9	
7.9 ±0.7	52.5 ±5.4	54.0 ±5.9	1.4 ±0.3	
5.1 ±0.9	0.2 ±0.04	2.0 ±0.2	1.7 ±0.2	

The total uncertainty was estimated to be between 10.2 and 44.8% including the statistical uncertainty (1-19%). It was estimated as the square root of the quadratic summation of the propagating components; the beam intensity (6%), target thickness (3%), target purity (1%), detector efficiency (7%) and peak fitting (3%). The initial uncertainty of the bombarding deuteron beam energy was estimated to be ±0.2 MeV which increased through the stack up to 0.6 MeV for the last Tm target foil.

Production of ^{169}Yb

The excitation function of the ^{169}Tm(d,2n)^{169}Yb reaction was derived from the γ-line at 177.21 keV (22.28%) as shown in Fig. 2 together with the earlier measured experimental data (Tárkányi, 2007; Hermanne, 2009) and the result of the TALYS calculation (TENDL-2015, 2015). The first measured cross section point is 0.20 mb at 5.1 MeV, the excitation function increases to 682.8 mb at 13.5 MeV, around the peak of the excitation function. This peak energy is in good agreement with the previous data (Tárkányi, 2007; Hermanne, 2009) although the present cross sections are slightly higher.

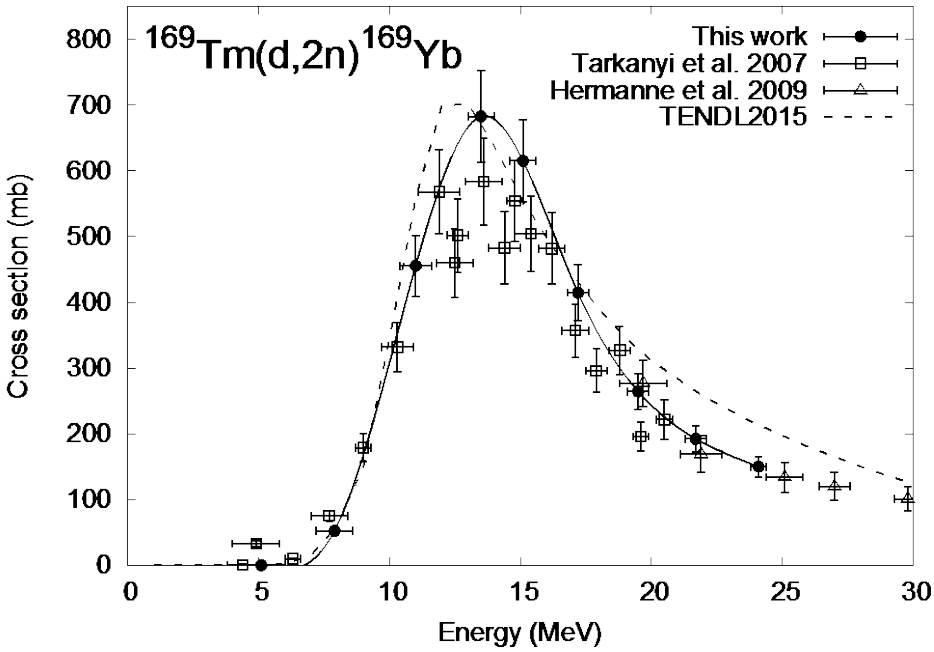

Fig. 2: The excitation function of the ^{169}Tm(d,2n)^{169}Yb reaction is shown with a spline fit over the experimental data. The result is compared with the previous experimental data (Tárkányi, 2007; Hermanne, 2009) and TENDL-2015 (TENDL-2015, 2015).

Production of ^{170}Tm

The radionuclide ^{170}Tm can be produced only in the (d,p) reaction on ^{169}Tm. The reaction product of ^{170}Tm ($T_{1/2}$ = 128.6 d) decays by emission of the 84.25-keV γ ray (2.48%). The contribution of background, X-rays from a lead shield of the HPGe detector to the 84.25-keV γ-line was confirmed to be negligible. Due to the long half-life of ^{170}Tm and its low γ intensity, the activity was assayed after long cooling times, i.e., five months after the end of bombardment. The measured excitation function of the ^{169}Tm(d,p)^{170}Tm reaction is shown in Fig. 3. The peak position is consistent with the result of the TALYS calculation although the amplitude is different from each other. No earlier experimental data are available for this reaction probably due to the long half-life and the low γ intensity of ^{170}Tm.

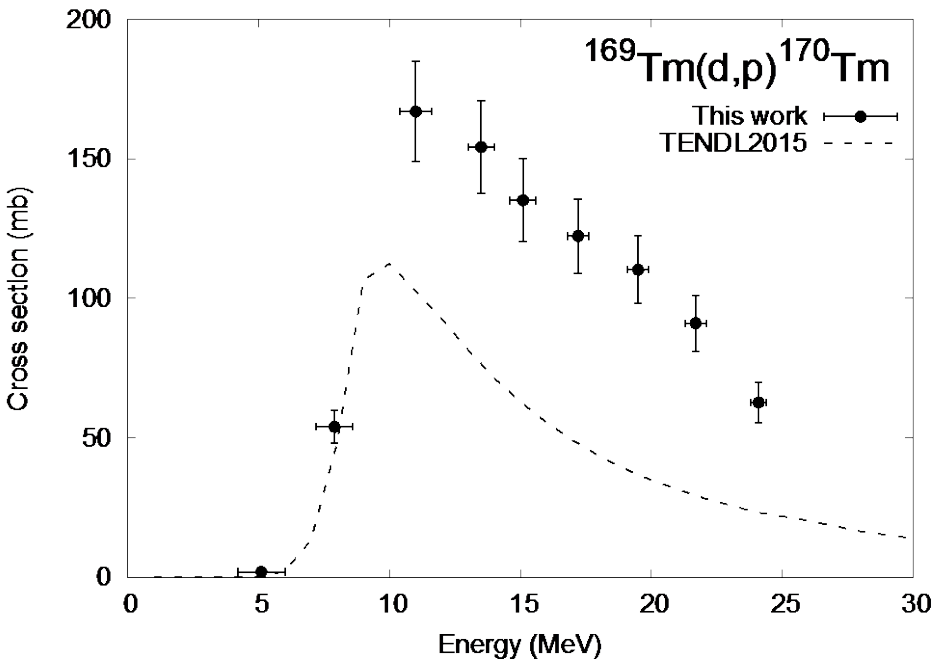

Fig. 3: The excitation function of the ^{169}Tm(d,p)^{170}Tm reaction in comparison with the result of the TALYS calculation.

Production of ^{168}Tm

The reaction product ^{168}Tm ($T_{1/2}$ = 93.1 d) emits the γ ray of 815.99 keV (50.95%). Using this interference free intense γ-line, the excitation function of the ^{169}Tm(d,x)^{168}Tm reaction was derived as shown in Fig. 4. The reaction channel opens above 1.8 MeV and the cross sections slowly increase with the deuteron energy. Our result is compared with the previous data (Tárkányi, 2007; Hermanne, 2009) which show good agreement with each other. The TALYS calculation shows underestimation and overestimation of the experimental data below and above 20 MeV, respectively.

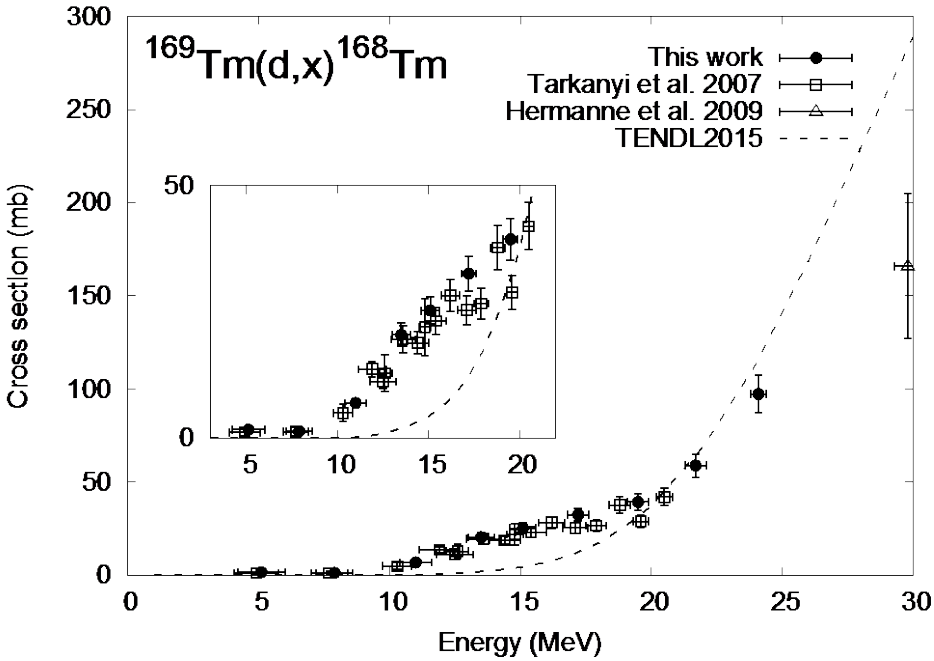

Fig. 4: The excitation function of the ^{169}Tm(d,x)^{168}Tm reaction in comparison with the result of the TALYS calculation and the experimental data measured earlier.

Production of ^{167}Tm

The 207.80-keV γ ray (42%) from the decay of ^{167}Tm ($T_{1/2}$ = 9.25 d) was used to determine the cross section for the ^{169}Tm(d,x)^{167}Tm process. The excitation function of the ^{169}Tm(d,x)^{167}Tm reaction is shown in Fig. 5. The reaction channel opens above 8.72 MeV and the cross sections increase slowly but remain at relatively low level with the incident energy up to about 22 MeV. Above 20 MeV energy the ^{169}Tm(d,4n)^{167}Yb ($T_{1/2}$ = 17.7 min) reaction channel opens and the produced ^{167}Yb decays quickly into ^{167}Tm and the cumulative cross section increases steeply. This behavior is in good agreement with the previously reported data (Hermanne, 2009). The TALYS calculation overestimates the contribution of the ^{169}Tm(d,x)^{167}Yb reaction above 20 MeV.

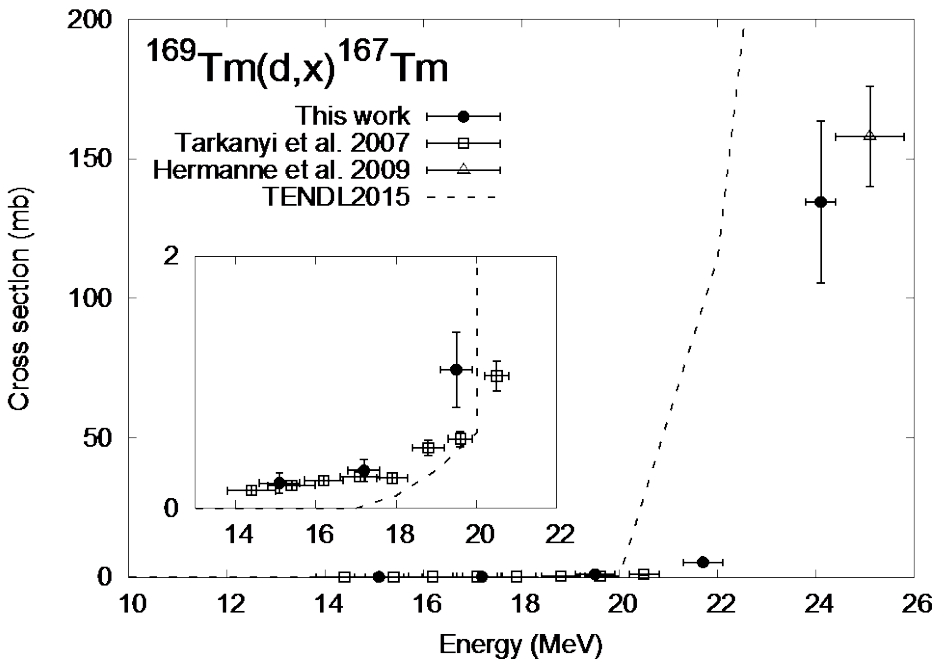

Fig. 5: The excitation function of the ^{169}Tm(d,x)^{167}Tm reaction in comparison with the result of the TALYS calculation and the experimental data measured earlier.

Production yield

Based on our experimental excitation function of the ^{169}Tm(d,x)^{169}Yb reaction, the production yield of ^{169}Yb was shown in Fig. 6 with the previous data (Hermanne, 2009). Our result rises to 5.3 MBq/μAh at 24 MeV, which is higher than previous one due to the higher amplitude of the cross sections.

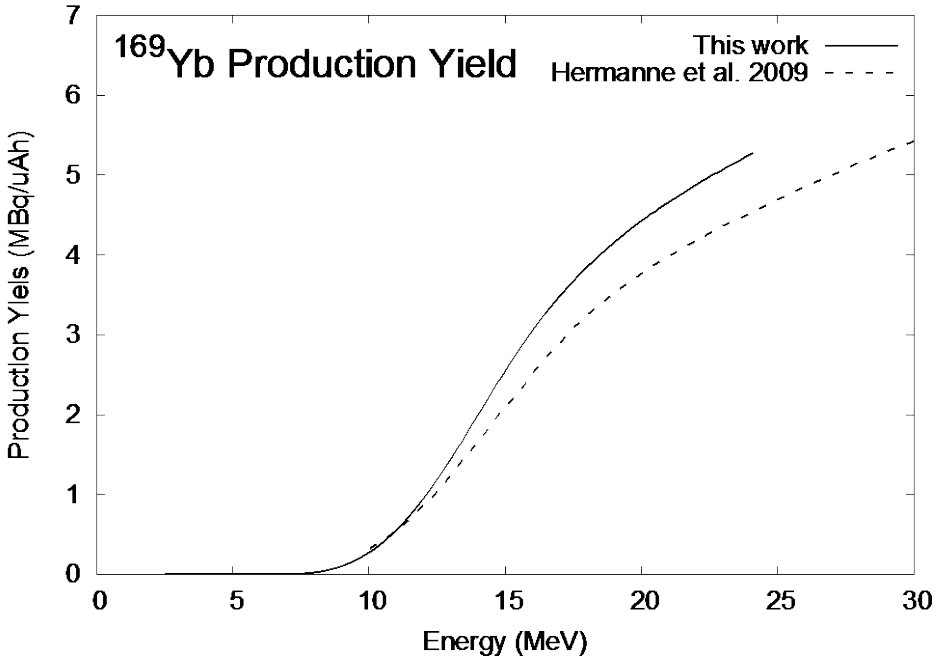

Fig. 6: The production yield of ^{169}Yb in comparison with the result of the experimental data measured earlier.

Summary

We have measured the cross sections of the deuteron-induced reactions on ^{169}Tm to produce ^{169}Yb, ^{170}Tm, ^{168}Tm and ^{167}Tm by using the stacked foil activation method. The thin metallic Tm foils were irradiated by 24.36 MeV deuteron beam and the activity of the produced radionuclides were determined by high resolution gamma spectrometry. The obtained excitation functions were compared with the earlier experimental data (Tárkányi, 2007; Hermanne, 2009) and good agreements in general were found. The excitation function of the ^{169}Tm(d,p)^{170}Tm reaction is reported for the first time.

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