CPC(HEP & NP), 2010, **34**(2): 177–181 **Chinese Physics C** Vol. 34, No. 2, Feb., 2010

Measurement of neutron cross sections and resonance parameters of ^{169}Tm below 100 eV

WANG Wen-Ming(王文明)[1;1)] LI Xia(李霞)[1] ZHAO Zhi-Xiang(赵志祥)[1] ZHOU Zu-Ying(周祖英)[1]
YU Hong-Wei(于洪伟)[1] WU Hai-Cheng(吴海成)[1] WEI Yi-Xiang(魏义祥)[2] T. F. Wang[3]
G. N. Kim[3] M. W. Lee[3] K. S. Kim[3] M. H. Cho[4] I. S. Ko[4] W. Namkung[4]

[1] China Institute of Atomic Energy, Beijing 102413, China
[2] Department of Engineering Physics, Tsinghua University, Beijing 100084, China
[3] Department of Physics, Kyungpook National University, Daegu 702-701, Republic of Korea
[4] Department of Physics, Pohang University of Science and Technology, Pohang 790-784, Republic of Korea

Abstract The neutron total cross-sections of thulium (^{169}Tm) were measured in the neutron energy region from 0.01 eV to 100 eV by using the time-of-flight method at the Pohang Neutron Facility, which consists of an electron linac, a water-cooled tantalum target with a water moderator, and a 12 m time of flight path. Two thulium plates with different thicknesses were used for the neutron transmission measurement. The background level was determined by using a notch-filter of Co, In, and Cd sheets. The present measurement was compared with the previous ones, and a new set of resonance parameters of ^{169}Tm isotope was obtained from the transmission rate by using the SAMMY code, with a comparison with the recommended parameters by Mughabghab.

Key words resonance parameters, thulium, n-TOF method, SAMMY

PACS 24.30.Gd, 25.40.Ny, 28.20.Ka

1 Introduction

The neutron cross-section data are of great importance in the study of the internal structure of atomic nuclei, as well as the safe design of nuclear reactors and the evaluation of both the neutron flux density and the energy spectrum around a reactor. Nowadays, neutron resonance parameters data are playing an increasingly important part in practical applications concerning with computations of reactor temperature coefficients, neutron reaction yields, self-protection effects, and other related matters. Particularly, in reactor design, the individual resonances can be very important.

Thulium, as an important rare-earth element, with ^{169}Tm as the only natural isotope, is widely applied in reactor neutron energy spectrum measurement and weapon design. As an odd-even isotope, ^{169}Tm shows prominent close resonance in low energy range. However, for some reason, in the common nuclear data library, such as ENDF [1] and JENDL [2], there is no data sheet for ^{169}Tm.

The total cross-sections and the resonance parameters of Thulium, ranging from 3 eV to 200 eV, were first measured and analyzed by Harvey et al [3], using the Brookhaven fast chopper in 1955, whose energy resolution was quite poor. The most widely adopted data were from Tellier et al [4] in 1972, ranging from 3.9 eV to 1 MeV, using n-TOF method with a flight path of 103.7 m. The latest reported data which can be found were taken by Danon et al [5] in 1998, with a correction of paramagnetic scattering and more precise thermal cross-sections, ranging from 0.001 eV to 20 eV, which has been adopted by Mughabghab in the latest evaluated resonance parameters [6]. However, in the last decade, there have been rarely such measurements of Thulium reported, while neutron detectors and electronics are developing very fast. Thus, there comes the need to re-measure the cross-sections in the resonance region.

Received 5 March 2009

1) E-mail: eaglewwm@gmail.com

The total cross-sections of ^{169}Tm, ranging from 0.01 eV to 100 eV, were measured with n-TOF method at the Pohang Neutron Facility (PNF), which was constructed at the Pohang Accelerator Laboratory in 1999. The PNF consists of an electron linac, a water-cooled Ta target, and a 12 m- TOF path. The details of the PNF are described elsewhere [7, 8]. The present data are compared with other measurements and analyzed using SAMMY [9] code to generate new resonance parameters, whose results are compared with the resonance parameters recommended by Mughabghab.

2 Experimental arrangement

The experimental setup arrangement of PNF for neutron transmission measurement consists of four parts: the RF gun and Linac, the water-cooled Ta target system to produce neutron, the auto sample changer, and the data acquisition system.

The maximum energy of the electron linac is 100 MeV, and the measured beam currents at the end of linac are 40 mA. The width of the electron beam pulse is 1.0 μs, with a repetition rate of 10 Hz.

Tantalum is selected as the neutron target material in order to produce intense neutrons by means of Bremsstrahlung under high-power electron beams. The neutron guide tubes were constructed with stainless steel, while the neutron collimation system was mainly composed of H_3BO_3, Pb, and Fe collimators, which were symmetrically tapered from 10 cm diameter at the beginning to 5 cm diameter in the middle position where the sample changer was located, and finally to 8 cm diameter at the end of the tube where the neutron detector was placed. In addition, there is a 1.8 m thick concrete slab between the target and the detector room for radiation shielding.

The sample changer, which is controlled remotely by the CAMAC module, is a disc with four holes, of which each is 80 mm in diameter, for mounting samples and set at the midpoint of the TOF path. The exposure time of each sample may be selected individually and precisely controlled by computer, with a 9 s exchange time interval from one position to the next one.

A ^{6}Li-ZnS (Ag) scintillator BC-702 with a diameter of 12.5 cm and a thickness of 1.59 cm was used as the neutron detector. This detector is sensitive for thermal and epithermal neutrons and is on the other hand quite insensitive to gamma radiation; thus, it is suitable for applications where the gamma background is quite high, such as in the transmission measurement at PNF.

3 Data acquisition system and samples

Two different data acquisition systems were used for the neutron TOF spectra measurements: one is a NIM-based system and the other is a CAMAC-based system.

The CAMAC-based system consists of a main data acquisition module and the remote control module of the sample changer. The main purpose of the NIM-based system is for neutron-gamma separation and parallel accumulation of the neutron TOF spectra, if necessary. Parallel data acquisition with both the NIM system and the CAMAC system may be used if one desires to optimize the dwell time for different regions of the TOF spectra.

During this transmission measurement, we used ^{169}Tm templates with a purity of 99.95%, whose physical properties are listed in Table 1. For the energy calibration and background estimation, a notch filter plate which is made of Cd, In and Co, whose physical properties are listed in Table 2, were used. With three of the four sample positions in use, the measurements were performed with two Tm samples with different thicknesses mounted on the sample changer at the same time and another free position was empty to collect the neutron TOF spectra without any sample (open beam). The positions of the samples were chosen in the following sequence: thin sample, open, thick sample, open. The exposure time for each position is set to be 300 s. The interleaving sequence of open positions in the sample changer was chosen to minimize the influence of slow and /or the small variations in the neutron beam intensity. If the beam intensity's variations or its drift was fast and /or large, then the partial measurements were excluded from the total statistics. The total data dating time for the samples was around 80 h.

Table 1. Physical properties of measured sample.

sample	purity (%)	thickness/ mm	diameter/ mm	density/ (g/cm^3)
Tm	99.95	1.41	64	9.32
Tm	99.95	0.41	64	9.32

Table 2. Physical properties of notch filters samples.

sample	purity (%)	size/ cm^2	thickness/ mm	density/ (g/cm^3)
Co	99.9	10×10	0.5	8.90
In	99.99	10×10	0.2	7.31
Cd	99.99	10×10	0.5	8.65

4 Data processing and analysis

4.1 Energy calibration and background estimation

The notch filter templates, were used for energy calibration and background level estimation. Several standard resonance peaks of the notch filter were selected to process non-linear least-square regression in the following formula:

$$E=\left(\frac{72.3\times L}{I\times W-t}\right)^2, \tag{1}$$

where W is the channel width of the time digitizer, with a value of 0.5 μs in this experiment, I is the channel address, L is the flight path and t is the time difference between the start time from the RF trigger and the real time zero when the neutron burst is produced. From the non-linear least square regression, L and t can be determined as (12.06±0.02) and (6.03±0.01), respectively.

The neutron spectrum of notch filters is also used for the background level estimation. In this situation, the black resonances at 132 eV and 1.457 eV together with the starting thermal energy of Cd are adopted to perform fitting in the following formula:

$$B=a\times \mathrm{e}^{-I/b}+c, \tag{2}$$

B represents the estimated background counting of the notch filter spectrum, which will be normalized to reduce the background level of Tm spectrum later. Finally the values of a, b and c are determined as 78.65, 423.05 and 0.5 respectively. The estimation result is shown in Fig. 1.

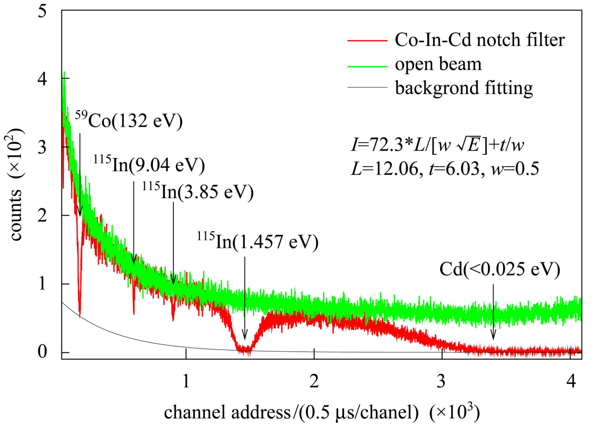

Fig. 1. Background level estimation using notch filters.

4.2 Transmission rate and total cross sections

From the n-TOF spectrum, the transmission rate can be defined as the neutron flux with samples divided by that without samples (open beam). Taking background into consideration, we can get the transmission rate as follows:

$$T(E_i)=\frac{I(E_\mathrm{i})/M_I-B(E_\mathrm{i})/M_B}{O(E_\mathrm{i})/M_O-B(E_\mathrm{i})/M_B}=\frac{kI(E_i)-B(E_i)}{mO(E_i)-B(E_i)}, \tag{3}$$

where I and O represent the neutron flux with and without samples, B is the estimated background level, M_I, M_B and M_O are the normalization factors, which, in this experiment, are generated from the pico-log files of the beam monitor. To make it easier for uncertainty estimation, k and m are introduced to the formula above. The final transmission spectra of samples with different thicknesses in the concerned energy range, from 2 eV to 100 eV, are illustrated in Fig. 2.

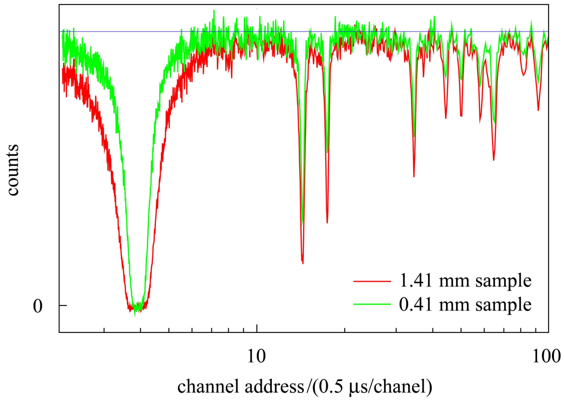

Fig. 2. Tm transmission spectrum after normalization and background correction.

The relationship between transmission rate and total cross sections is as follows:

$$\sigma(E_\mathrm{i})=-\frac{1}{\sum\limits_j N_j}\ln T(E_\mathrm{i}), \tag{4}$$

where N_j is the atomic density per cm^2 of the j-th isotope in the sample. As for different energy groups, the total cross section can be calculated as:

$$\sigma_{\mathrm{eff}}(E)=\frac{\int_{E_\mathrm{i}}^{E_\mathrm{f}}\sigma(E)E\mathrm{d}E}{\int_{E_\mathrm{i}}^{E_\mathrm{f}}E\mathrm{d}E}, \tag{5}$$

Here, the subscript j corresponds to each energy group. The calculated cross section data of this experiment compared with some other experiments and evaluated data are shown in Fig. 3. They are, in a way, in agreement.

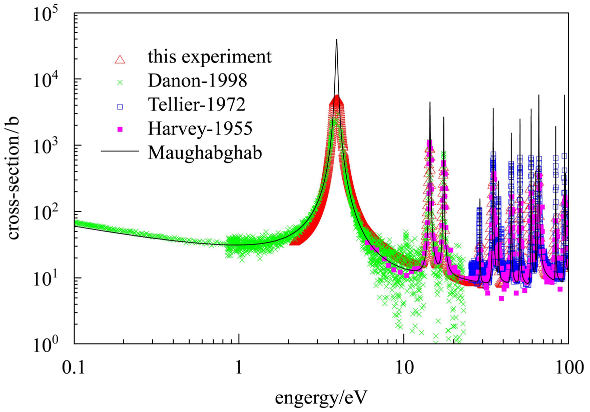

Fig. 3. Cross sections compared with former experiments.

4.3 Energy resolution and uncertainty analysis

From Eq. (1), we can get the energy resolution as

$$\frac{\Delta E}{E}=\frac{2\Delta t}{t}, \tag{6}$$

where, the uncertainty (Δt) of the neutron TOF (t) is composed of uncertainties due to the flight path (0.02 m), the moderator thickness (0.03 m), the pulse width of the electron beam (1 μs), the channel width of the time encoder (0.5 μs), and the time jitter (negligible small) from the neutron detector. The energy resolutions for neutron energies of 1, 10 and 100 eV are 0.65%, 1.01% and 2.63%, respectively.

According to the statistic error of the counting rates of both the background and sample measurements, the statistic error of cross-sections can be generated from Eqs. (3) and (4).

During this experiment, the overall statistic error is less than 5%. The systematic uncertainties came from the uncertainties of flight path and dead time fitting, background estimation and normalization, etc. Taking all above into account, the total systematic error of this experiment is less than 3%.

4.4 Resonance parameters determination

To determine the resonance parameters of each resonance peak in Fig. 2, we fitted the transmission data with the SAMMY code [9], which is a multi-level R-matrix fitting code using Bayes' equations. In the Reich-Moore approximation [10], the total cross sections in the thermal neutron energy region can be described as follows:

$$\sigma_{\mathrm{t}}=\frac{2\pi g}{k^2}\left\{1-\cos 2\varphi\left(1-\frac{\Gamma\Gamma_1}{2d}\right)-\sin 2\varphi\frac{\Gamma_1(E_\lambda-E)}{d}\right\}, \tag{7}$$

here, k represents the wave number associated with the incident channel, ϕ the potential scattering phase shift, g the spin statistical factor, E the neutron energy, and E_λ the resonance energy. Γ and Γ_1 are the sum of the partial widths and the partial width of decay channel 1, respectively. d can be defined as

$$d=(E_\lambda-E)^2+\left(\frac{\Gamma}{2}\right)^2. \tag{8}$$

SAMMY is a code system for multi-level R-matrix fitting to experimental data using Bayes' equations, with corrections for complex experimental conditions. As for this experiment, the Doppler as well as the resolution broadening was taken into consideration, where the MULTI method was applied. The free gas model was applied for Doppler broadening, while the convolution of Gaussian and exponential functions was applied for energy resolution broadening [9], whose mathematical expression is

$$\begin{aligned}R_{\mathrm{GE}}(E,E') = {} & \frac{1}{\varDelta_{\mathrm{E}}\varDelta_{\mathrm{G}}\sqrt{\pi}}\int_{E-\Delta E_{\mathrm{s}}}^{\infty}\mathrm{d}E^0\times\\ & \exp\left\{-\frac{E^0-(E-\Delta E_{\mathrm{s}})}{\varDelta_{\mathrm{E}}}\right\}\times\\ & \exp\left\{-\frac{(E'-E^0)^2}{\varDelta_{\mathrm{G}}^2}\right\},\end{aligned} \tag{9}$$

where the width of Gaussian resolution function $\varDelta_{\mathrm{G}}$ is given by

$$\varDelta_{\mathrm{G}}=E\left[aE+b\right]^{1/2} \tag{10}$$

and the width of exponential resolution function $\varDelta_{\mathrm{E}}$ is given by

$$\varDelta_{\mathrm{E}}=cE^{3/2}, \tag{11}$$

The energy shift ΔE_{s}, which is automatically determined in the SAMMY code, is introduced in order to locate the maximum of the broadening function at $E'=E$. The constant value of a, b and c is 1.21×10^{-6} eV^{-1}, 6.83×10^{-6}, and 8.23×10^{-4} eV$^{-1/2}$, respectively.

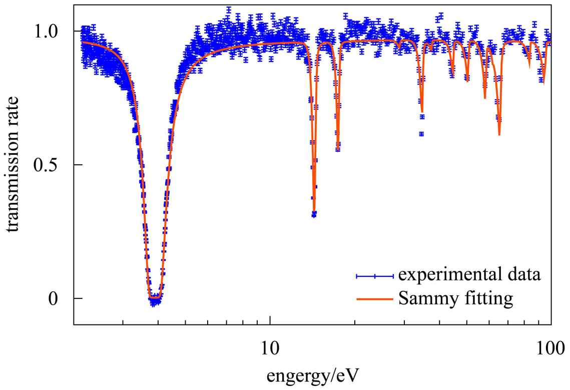

Fig. 4. ^{169}Tm data fitting results.

The fitting results can be seen in Fig. 4, which are in good agreement with present experimental data. Both the thick and thin sample data are processed simultaneously, while only the thin-sample data are shown here.

The new resonance parameters of ^{169}Tm, ranging from 1 eV to 100 eV, compared with the recommended data by Mughabghab, are shown in Table 3. It should be noticed that there are some differences between these two results. For the energy resolution of present experimental data is a little bit poor, the uncertainties of the present work, generated both from experimental errors and Sammy fitting errors, are accordingly larger than the recommended parameters by Mughabghab. The spin group of each resonance is assigned according to the recommended value by Mughabghab at first, which finally shows no big problems.

Table 3. New resonance parameters of ^{169}Tm compared with Mughabghab.

E/eV		$2g\Gamma_n$/meV		Γ_γ/meV	
Mughabghab	present	Mughabghab	present	Mughabghab	present
−20.13	−20.16		358±42	86.37	173±21
3.906±0.001	(3.93)	11.2±0.3	(17)	102.4±1.2	(131)
14.32±0.001	14.41±0.03	4.56±0.06	5.4±0.5	97.1±2.0	160±15
17.42±0.001	17.51±0.02	2.87±0.03	3.8±0.5	81.4±3.0	107±12
28.9±0.1	28.9±0.1	0.31±0.03	0.42±0.04	95±20	93±20
34.79±0.05	34.86±0.06	8.8±0.5	14.1±1.0	86±3	113±20
37.51±0.05	37.47±0.05	0.69±0.07	0.8±0.1		(5.0)
44.79±0.05	44.87±0.06	5.2±0.3	7.0±1.0	93±5	97±8
50.58±0.10	50.7±0.3	8.1±0.8	12.0±2.0	79±5	74±5
59.07±0.10	58.5±0.3	15.0±0.9	25±3.0	84±5	66±6
62.97±0.10	62.9±0.2	1.5±0.2	1.2±0.5	78±15	76±15
65.75±0.10	65.90±0.2	56.5±1.5	71±5	83±4	97±15
83.18±0.10	83.53±0.3	11.3±0.6	15±1	85±5	83±10
94.1±0.1	94.6±0.3	48±5	58±8		(4.9)
95.4±0.1	95.6±0.3	1.9±0.2	3.5±1.0	71±20	71±15

5 Conclusions

The total cross sections of ^{169}Tm, ranging from 0.01 eV to 100 eV, were measured at the Pohang Neutron Facility with n-TOF method and a ^{6}Li-ZnS(Ag) glass scintillator as a neutron detector, whose results are in good agreement with other experimental data. The resonance parameters of ^{169}Tm were determined by fitting the transmission rate with the SAMMY code below 100 eV with a comparison with the recommended parameters by Mughabghab.

The authors would like to express their sincere thanks to the staff of the Pohang Accelerator Laboratory for the excellent operation of the electron linac and their strong support.

References

1 Evaluated Nuclear Data File. ENDF/B-VII.1. Brookhaven National Laboratory, http://www-nds.iaea/org/exfor/endf00.htm, 2007

2 Evaluated Nuclear Data File. JENDL-3.3. Japan Atomic Energy Agency, http://wwwndc.jaea.go.jp/jendl/j33/j33.html, 2002

3 Harvey J A, Hughes D J, Carter R S et al. Phys. Rev., 1955, **99**: 10

4 Tellier H et al. Measurement and Analysis of Total cross-sections of ^{169}Tm and ^{165}Ho. In Proceedings of the International Conference on Nuclear Structure Study with Neutrons, Budapost, 1172. Budapest: Central Research Institute of Physics, 1972. 191–192

5 Danon Y, Werner C J, Youk G et al. Nucl. Sci. Eng., 1998, **128**: 61

6 Mughabghab S F. Atlas of Neutron Resonances. ORNL: Elsevier Science, 2006. 363–367

7 Kim G N et al. J Kor. Phys. Soc., 2003, **43**: 479

8 Kim G N et al. Nucl. Instrum Methods A, 2002, **485**: 458

9 Larson N M. Updated Users's Guide for Sammy: Multilevel R-matrix Fits to Neutron Data Using Bayes's Equations. ORNL: Elsevier Science, 2003

10 Reich C W, Moore M S. Phys. Rev., 1958, **111**: 929