Measurements of fast-neutron capture cross sections for ^{159}Tb and ^{169}Tm*

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Abstract The neutron capture cross sections for ^{159}Tb and ^{169}Tm relative to the ^{197}Au (n, γ) ^{198}Au reaction are measured at neutron energies of 0.57 , 1.10 and 1.60 MeV by using the activation method. The activities of the products are measured with a high resolution HPGe detector gamma-ray spectrometer. The errors of the present work are 5~6% for Tb, 6~7% for Tm. The recommended data in energy region of 0.4~3.0 MeV are given as compared with other data published previously.
Keywords ^{159}Tb, ^{169}Tm, ^{197}Au, Neutron capture cross sections, Activation method

1 Introduction

Fast-neutron capture cross sections are important for the understanding of nuclear reaction mechanisms and for various applications in nuclear technology. Both ^{159}Tb and ^{169}Tm are rare-earth elements. Precise values of the capture cross sections of terbium and thulium also are of major significance for fission and fusion reactor design since they are the fission product poisons.

For the ^{159}Tb (n, γ) ^{160}Tb reaction, capture cross sections in neutron energy range of 0.4~4.0 MeV were determined by directly measuring the prompt gamma-ray of capture events in four laboratories.[1~4] No measurements of ^{159}Tb (n, γ) ^{160}Tb cross sections using activation technique have been reported. The cross sections for ^{169}Tm (n, γ) ^{170}Tm were measured in the neutron energy range of 0.16~3.0 MeV in four laboratories.[5~8] Among them the cross section values measured by Jiang Songsheng[8] using activation method were lower than other ones using the prompt gamma-ray method. Therefore, the measurement of cross sections at 0.5~ 1.6 MeV with higher resolution HPGe detector are necessary in order to check the capture cross sections and obtain the more accurate data.

In this experiment, the neutron capture cross sections for ^{159}Tb and ^{169}Tm relative to the ^{197}Au (n, γ) ^{198}Au reaction were measured by the activation technique at neutron energies of 0.57, 1.10 and 1.60 MeV.

2 Experimental procedure

The experiments were carried out at the 4.5 MV Van de Graaff accelerator in the Institute of Heavy Ion Physics, Peking University. The monoenergetic neutrons with energies of 0.57, 1.10 and 1.60 MeV were produced via the T(p,n) ^{3}He reaction on a solid T-Ti target of 1.42 mg/cm^2 in thickness.

The rare-earth samples were made from natural element oxides powder by pressing into disks of 10 mm in diameter and about 0.5~0.8 mm in thickness (about 353.1, 325.1, 339.8 mg/cm^2 for Tm, 500.5, 544.1, 615.2 mg/cm^2 for Tb, respectively) and sealed in thin polyethylene foils. The mass purities were 86.88% for Tb and 87.56% for Tm. Each sample was sandwiched between two gold disks. The mass purity 99.9% gold disks each of 10 mm in diameter and 0.1 mm in thickness were used to measure the neutron fluence on the sample. The sample groups were wrapped with cadmium foils of 0.5 mm in thickness.

The irradiation was performed at 0° direction relative to the incident proton beam. The distance between the sample and target was 1.5 cm. In order to reduce wall-scattered and floor-scattered neutrons, the target for source

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neutrons was located above a non-scattering environmental underground hollow of 3.0 m in diameter and 1.8 m in depth, which was about 5.5 m away from the wall and at a distance of 1.8 m from the floor. The proton beam currents were generally 10~ 12 μA and the duration of irradiation was 23 to 24 h at each energy. The fluctuation of neutrons fluence rate was monitored with a BF_3 long counter at 0° and at a distance of 315 cm from the neutron source. In order to record the neutron fluence rate as a function of time during the irradiation, the integral count rate of the long counter per 10 min was recorded continuously by microcomputer multiscaler and stored on magnetic disk for calculating the correction of non-uniform irradiation history.

After irradiation, the activities from residual nuclei were measured with a HPGe γ-detector (105 cm^3). The efficiency of the detector was calibrated by using a set of standard gamma ray sources in the energy range of 0.1~1.5 MeV and the efficiency curve was fitted with the least-square method. The γ-detection efficiency of ^{160}Tb was obtained from this efficiency curve. For the decay of ^{170}Tm, the dominant gamma ray is only 84.25 keV. In order to calibrate the efficiency of the detector for ^{170}Tm accurately, a standard gamma-ray source of ^{170}Tm was prepared specially in China Institute of Atomic Energy and its activity was determined by measuring the β-activity. The activities of these samples were also measured comparatively by gamma ray spectroscope using a HPGe-N ORTEC model GX10185 and a HPGe-P Canberra well detector in the Northwest Institute of Nuclear Technology. Their measured results were in good agreement with ours within errors. The decay data used in present work are taken from Ref.[9] and listed in Table 1.

Table 1 Decay data of radioactive products

Resid nucl	$T_{1/2}$/d	E_γ/keV	I_γ/%
^{160}Tb	72.3	1177.95	14.97
^{170}Tm	128.6	84.25	2.48[10]
^{198}Au	2.696	411.8	95.57

Partial corrections were described as follows:

(1) neutron energy: on account of the sample near the target, the incident neutron energy was calculated by

$$\overline{E_n} = \int_0^\alpha E(\theta)A(\theta)2\pi\sin\theta\mathrm{d}\theta \Big/ \int_0^\alpha A(\theta)2\pi\sin\theta\mathrm{d}\theta \tag{1}$$

where $E(\theta)$ is the energy angular distribution of incident neutron, $A(\theta)$ the angular distribution of incident neutron cross sections, α the maximum angle subtended by the sample at the target.

(2) gamma ray self-absorption in the sample: the correction factor of gamma ray self-absorption in the sample is given by

$$f_s = [1 - \exp(-\mu x)]/\mu x \tag{2}$$

where μ is the total-mass absorption coefficient in cm^2/g, x is the density thickness in g/cm^2.

At first, the total-mass absorption coefficient was measured experimentally as gamma-ray passed through a series of samples with different thicknesses. Because the 1173 keV gamma-ray of ^{60}Co almost equals 1177.95 keV gamma-ray of ^{160}Tb, the total-mass absorption coefficient of Tb sample was measured by using 1173 keV γ-ray of ^{60}Co instead of by using 1177.95 keV γ-ray of ^{160}Tb.

It is important for the correction factor of 84.25 keV gamma ray self-absorption in the Tm_2O_3 sample. Its total-mass absorption coefficient was measured experimentally by using 84.25 keV γ-ray of ^{170}Tm source.

The γ-ray peak area measured was analysed using the program H developed for an IBM compatible computer. The counting rates under the concerned full-energy peaks based on the measured γ-spectrum were obtained. After the corrections for the detector efficiency, cascade effect, γ-intensity, fluctuation of neutron fluence rate and γ-ray self absorption in the samples, the activation cross sections of ^{159}Tb and ^{169}Tm were calculated by using well-known activation equation.

3 Results and discussion

The cross sections measured in the present work and the ^{197}Au (n, γ) ^{198}Au cross sections recommended by ENDF/B-6 are listed in Table 2. The principal contributions of errors and their magnitudes are given in Table 3.

Figs.1,2 show the capture cross sections of ^{159}Tb and ^{169}Tm as a function of the incident neutron energy and also include the results of other published measurements, respectively.

Table 2 Measured results of cross sections (in mb)

E_n/MeV	^{159}Tb(n, γ)^{160}Tb	^{169}Tm(n, γ)^{170}Tm	^{197}Au(n, γ)^{198}Au
0.57±0.03	298.8±15.5	160.3±10.1	118.6±4.1
1.10±0.03	179.0±10.7	99.7±6.9	77.2±3.4
1.60±0.04	120.6±7.2	95.5±6.7	66.5±2.9

Table 3 Principal sources of errors

Source of uncertainty	Relative errors/%	
	^{159}Tb(n, γ)^{160}Tb	^{169}Tm(n, γ)^{170}Tm
Reference cross sections	3.5~4.5	3.5~4.5
γ-counting statistics for sample	0.7~1.0	0.6~1.0
γ-counting statistics for ^{198}Au	0.6~0.8	0.5~0.7
γ-detection efficiency for sample	1.5	2.0
γ-detection efficiency for ^{198}Au	1.5	1.5
Correction of self absorption for sample	2.0	4.0
Correction of self absorption for ^{198}Au	1.5	1.5
Sample weight	1.0	1.0
^{197}Au foil weight	0.1	0.1

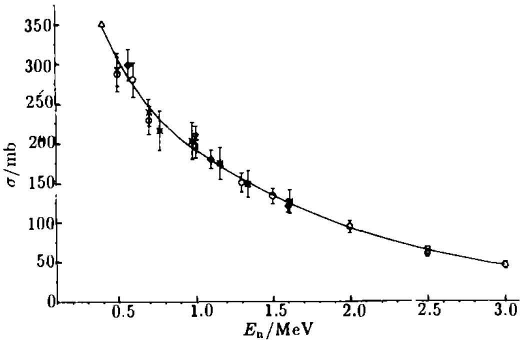

Fig.1 Cross sections for ^{159}Tb(n, γ)^{160}Tb reaction

◇ Present work; o Poenitz's[1]; × Voignier's[2]; * Mu's[3]; △ Brzosko's[4]; — Recommended data

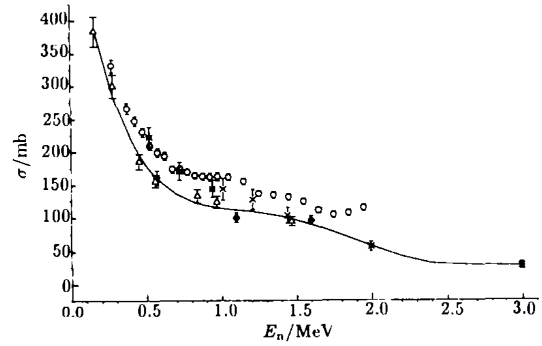

Fig.2 Cross sections for ^{169}Tm(n, γ)^{170}Tm reaction

◇ Present work; o Macklin's[5]; × Xu's[6]; * Joly's[7]; △ Jiang's[8]; — Recommended data

3.1 Tb

The accuracy of our measurement is 5~6%, while Poenitz's 7.5~10%[1], Voignier's 7~8%[2], Mu's 11~12%[3] and Brzosko's no error available[4]. Our results obtained by the activation method are in good agreement with the experimental data of other authors[1~4] obtained by the prompt gamma-ray detection technique in the 0.4~3.0 MeV neutron energy range. From Fig.1, we can find out that all of them have the same decreasing trend with the increasing incident neutron energy. So we consider that the experimental data including our data are reasonable and reliable. We fit these experimental data and get the recommended data in the energy region of 0.4~3.0 MeV as the following

$$\sigma = 630.66 - 1029.0E + 995.07E^2 - 533.52E^3 + 142.55E^4 - 14.795E^5 \quad (3)$$

where σ is the capture cross sections in mb, E is the neutron energy in MeV.

3.2 Tm

The accuracy of the present experimental results is 6~7%, while Macklin's ~5%[5], Xu's 10~12%[6], Joly's 6.7~18%[7] and Jiang's 6~7.5%[8]. For this reaction, we and Jiang[8] measured its capture cross sections by the same activation method. But, we measured the γ-activity of ^{170}Tm while Jiang measured the β-activity of ^{170}Tm. The other authors[5~7] all directly measured the prompt gamma-rays for capture events. From Fig.2, we can find out that our results are in good agreement with Jiang's experimental data but lower than the results of other authors. In the energy range of higher than 1.5 MeV, there is serious discrepancy between the experimental results of Joly and Macklin obtained by the same prompt gamma-ray detection technique. So we can deduce that our and Jiang's experimental results are adequate. The recommended data in energy region of 0.4~3.0 MeV for this reaction are obtained based on our and Jiang's results as well as Joly's experimental data for energy higher than 1.5 MeV. It also can be expressed as

$$\sigma = 582.39 - 1495.0E + 1858.1E^2 - 1105.1E^3 + 305.29E^4 - 31.621E^5 \quad (4)$$

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