Thermal neutron cross section and resonance integral of the ^{159}Tb$(n,\gamma)^{160}$Tb reaction

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The thermal neutron cross section and resonance integral of the reaction ^{159}Tb$(n,\gamma)^{160}$Tb in the thermal and $1/E$ regions, respectively, of a thermal reactor neutron spectrum have been experimentally determined. Literature data on the resonance integral of the reaction show a large scatter ranging from 313 to 780 b. On the basis of such data it becomes extremely difficult to produce a best value for application. In this work we performed a careful experiment in an effort to produce a precise and accurate measured value, which will be a valuable addition to the literature. The ^{159}Tb$(n,\gamma)^{160}$Tb reaction is studied by irradiating spectrographically pure Tb_4O_7 powder samples with thermal and epithermal neutrons in the Pakistan Research Reactor–1 (PARR-1) at PINSTECH. Thermal and epithermal neutron fluence rates were determined with Au and Co activation detectors. The α parameter, which accounts for the deviation of the neutron spectrum shape from the $1/E$ law in the epithermal neutron region, was obtained with good accuracy by using Au, Co, Mn, and Zn activation detectors and adopting the regression and iterative analysis procedures. The induced activities in the target samples and the activation detectors were measured with a high purity germanium detector system. The cadmium ratio method was used to yield the thermal neutron cross section and resonance integral. The measured thermal neutron cross section at 0.0253 eV and resonance integral are 23.6±0.4 b and 443±21 b, respectively. The results are discussed and compared with the literature.

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I. INTRODUCTION

Neutron cross sections are required in both fundamental nuclear research and in a wide range of applied nuclear fields such as shielding calculations, nuclear fuel and waste management, dosimetry, and nuclear medicine. Thermal neutron cross sections and resonance integrals are required for reactor neutron flux parameterization and neutron activation analysis. It has been observed that the resonance integrals of several nuclides described in the literature [1] show considerable discrepancies. A proper evaluation of these data that may be useful for practical purposes requires a large population of experimental data. This puts an emphasis on the need for new measurements. Several nuclides have been studied in the recent past for thermal neutron cross section and resonance integrals [2–7].

In this work we studied the thermal neutron capture cross section (the cross section at 2200 m s^{-1}) and resonance integral (epithermal neutrons in the $1/E$ region) for the rare-earth nuclide Terbium-159. The reported values of the thermal neutron cross section of ^{159}Tb range from 22 to 26 b whereas the resonance integral data of the same nuclide show a large scatter and range from 313 to 780 b, see the Gryntakis and Kim compilation [1]. The present experiment measures the thermal neutron capture cross section and resonance integral of ^{159}Tb$(n,\gamma)^{160}$Tb reaction by irradiating six spectrographically pure terbium powder samples with thermal and epithermal neutrons using the irradiation facilities of the Pakistan Research Reactor (PARR-1) at PINSTECH. The nuclear reactions ^{55}Mn$(n,\gamma)^{56}$Mn, ^{59}Co$(n,\gamma)^{60}$Co, ^{64}Zn$(n,\gamma)^{65}$Zn, and ^{197}Au$(n,\gamma)^{198}$Au were used to determine the α parameter to account for the deviation of the neutron distribution from the $1/E$ law in the epithermal region and the neutron fluxes in the thermal and epithermal regions of the neutron spectrum. The results obtained for the thermal neutron cross section and resonance integral are compared with the literature.

II. THEORY

The flux density in a thermal reactor can be characterized by two components, a temperature dependent Maxwellian thermal neutron component and an epithermal neutron slowing down component with an ideal $1/E$ distribution. A number of formalisms [8–10] have been suggested to describe the reaction rates. A simple convention proposed by Høgdhal [8] for $1/v$ nuclides is employed for the present purpose, which is as accurate as the complex formalisms such as Westcott [9].

The reaction rate per target nuclei, R, of a sample irradiated by reactor neutrons can be described according to the Høgdhal convention as

$$R = R_{th} + R_e = \Phi_{th}\sigma_o + \Phi_e I_o, \tag{1}$$

where $R_{th}(=\Phi_{th}\sigma_o)$ is the reaction rate induced by pure thermal neutrons and $R_e(=\Phi_e I_o)$ is the reaction rate induced by epithermal neutrons. Φ_{th} is the thermal neutron flux and Φ_e is the integrated epithermal neutron flux per unit $\ln(E)$ with E being the neutron energy. σ_o is the thermal neutron capture cross section at 2200 m s^{-1} and I_o is the resonance integral for a $1/E$ spectrum. In practical situations the slowing down neutron spectrum deviates from the ideal $1/E$ shape and requires a correction parameter α. The flux per unit energy is therefore approximately proportional to $1/E^{1+\alpha}$, with α independent of the neutron

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energy [11,12]. The modified reaction rate including the shielding effects can be written as

$$R = R_{th} + R_e = \Phi_{th}\sigma_o G_{th} + \Phi_e I_o(\alpha) G_e, \quad (2)$$

where G_{th} and G_e are the thermal and epithermal self-shielding correction factors. $I_o(\alpha)$ is the irradiation site dependent resonance integral.

The reaction rate of a target irradiated under a cadmium cover is given by

$$R'_e = \Phi_e I_o(\alpha) G_e F_{cd}, \quad (3)$$

where F_{cd} is the cadmium transmission correction factor.

γ Ray spectrometry of the irradiated sample with the use of an efficiency calibrated high-resolution detector enables the determination of the experimentally measured reaction rates R (or R'_e) by employing the following expression:

$$R = \frac{N_p W}{N_a \theta P_\gamma \varepsilon_\gamma m S D C}, \quad (4)$$

where N_p is the net peak counting rate corrected for pulse pileup losses. S, D, and C are the respective correction factors for saturation during irradiation, decay between irradiation and γ counting, and decay during counting. W is the atomic weight, N_a is Avogadro's number, θ is the natural isotopic abundance, P_γ is the γ ray emission probability, ε_γ is the absolute efficiency of the detector for a γ ray photo peak, and m is the mass of the target element.

Experimental measurements of the reaction rates of activation detectors with well-known nuclear data lead to the determination of the neutron flux parameters at the irradiation site and consequently to the measurement of thermal neutron cross section and resonance integral of the element of interest. It is to be noted that the conversion of $I_o(\alpha)$ to I_o is necessary to provide a resonance integral independent of the irradiation position which can then be compared to literature values. This conversion can be made by using the following expression:

$$I_o = E_r^\alpha \left\{ I_o(\alpha) - \frac{0.316\sigma_o}{\sqrt{E_{cd}}} \left[\frac{1}{(2\alpha+1)E_{cd}^\alpha} - E_r^\alpha \right] \right\}, \quad (5)$$

where E_r is the effective resonance energy and E_{cd} (=0.55 eV) is the cadmium cutoff energy.

A. Determination of α parameter

The α parameter can generally be determined by the bare multimonitor method, the cadmium covered multimonitor method and the multimonitor cadmium ratio method [13,14]. In this work the α parameter is determined by using the cadmium ratio method as it utilizes only the ratios, which improves the estimates of the uncertainty. The slope of the following straight line yields the α parameter,

$$Y_i = a + \alpha X_i, \quad (6)$$

where $Y_i = \ln\{E_{r,i}^{-\alpha} G_{th}/(F_{cd,i} R_{cd,i} - 1) Q_{o,i}(\alpha) G_{e,i}\}$ and $X_i = \ln E_{r,i}$, with $Q_{o,i}(\alpha) = I_{0,i}(\alpha)/\sigma_{o,i}$, the subscript i refers to ith nuclide. In this equation the α parameter exists in both sides of the equation. It can therefore initially be set to zero followed by an iterative procedure until it converges.

III. EXPERIMENT

1. Target preparation

The targets of terbium were prepared from spectrographically pure powder Tb_4O_7 from Merck. A small quantity of terbium powder (2–6 mg) was spread onto a Whatman-1 filter paper, covering a relatively large circular area (so as to minimize the self-shielding effect), thereby simulating circular foils of about 8–10 mm in diameter. Four samples were prepared for bare irradiation and another four for irradiation under a cadmium cover. The weighing of the target materials was carefully carried out using a Sartorius semimicro electrical balance.

The irradiations of the activation detectors of Au, Co, Mn, and Zn were carried out to characterize the neutron irradiation facility. The targets of Au weighing 1–4 mg were prepared from thin gold foils of thickness 30 μm. The cobalt samples (6–10 mg) were made from 10-mil cobalt wire. The wires were cut into roughly equal pieces of length 2 cm and rolled spirally. The manganese samples (2–4 mg) were made from manganese metallic pieces. Zinc foils (~2 mg) of 8 mm diameter were used. For the determination of the α deviation parameter, these activation detectors were packaged together in standard polystyrene irradiation capsules of dimension 13.5 mm in diameter and height 11 mm for bare irradiation and another set of detectors was packaged into a cadmium box of dimension 14 mm diameter, height 6 mm, and thickness 1 mm. Each of the activation detectors was also encapsulated individually within polystyrene and cadmium covers for the determination of thermal and epithermal neutron fluxes and cadmium ratios.

2. Neutron irradiation

The Pakistan Research Reactor (PARR-1) at PINSTECH was used for the irradiation of the samples. The reactor core

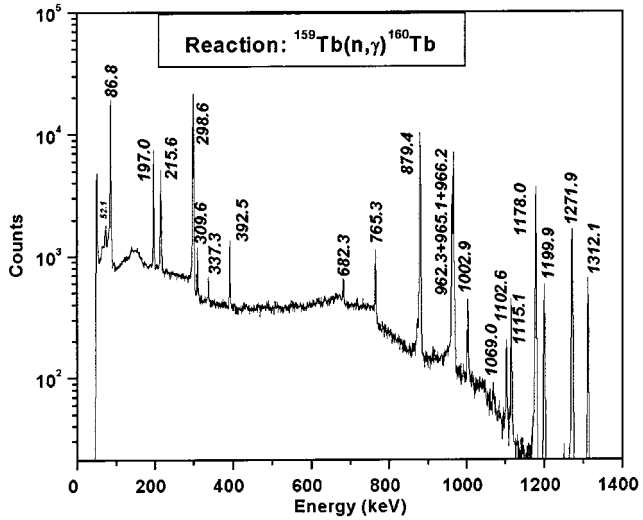

FIG. 1. γ Ray energy spectrum of the irradiated Tb_4O_7 sample.

TABLE I. Nuclear and resonance data for the reactions used in this work. Uncertainties given in parentheses are in the last digits of the principal values.

Nuclear reaction	Atomic weight [17]	Abundance [18] (%)	Effective resonance energy [19] (eV)	Half-life [20] (days)	γ energy [21,22] (keV)	γ emission probability [21,22] (%)	Q_o value [14]	Remarks
$^{55}Mn(n,\gamma)^{56}Mn$	54.938049(9)	100	468(51)	0.107454(4)	846.7715	98.9(3)	1.053(27)	α Monitor
$^{59}Co(n,\gamma)^{60}Co$	58.933200(9)	100	136(7)	1925.1(5)	1173.237 1332.501	99.9736(7) 99.9856(4)	1.993(54)	α and flux
$^{64}Zn(n.\gamma)^{65}Zn$	65.39(2)	48.63(60)	2560(260)	244.26(26)	1115.546	50.60(22)	1.908(94)	α Monitor
$^{197}Au(n,\gamma)^{198}Au$	196.96655(2)	100	5.65(40)	2.69517(21)	411.80203	95.58(12)	15.71(28)	α and flux
$^{159}Tb(n,\gamma)^{160}Tb$	158.92534(2)	100	18.1(9)	72.3(2)	298.58	26.1(6)		
					879.383	30.1(6)		
					1177.962	14.9(3)		

contains 20% LEU fuel. The reactor is of the swimming pool type and normally operates at 10 MW power level. The neutron irradiation of the samples was carried out at the irradiation position RS 3 of the reactor. Suitable irradiation times were chosen (from 2 min to 10 min) so as to induce the optimum activity for the γ ray measurements. The irradiated samples were allowed to decay for an appropriate cooling time before the γ ray spectrometry of the samples.

3. Measurements of radioactivity

The γ ray spectrometry of the irradiated samples was carried out using a high-resolution gamma ray detection system. The system employed a 40% relative efficiency high purity germanium detector with energy resolution of 2.0 keV at the 1332.5 keV peak of ^{60}Co. A computer based 4096 multichannel analyzer carried out the data acquisition. The efficiency calibration of the detector was performed by measuring the radionuclides ^{60}Co, ^{133}Ba, ^{137}Cs, ^{154}Eu, and ^{241}Am. The measured efficiencies were fitted to a suitable function [15], which allowed for the interpolations at the particular energies of interest. Excellent efficiency fitting curves were established for 25- and 15-cm source-detector distances with χ-squared per degree of freedom between 0.9 and 0.95. A detailed study describing the method of the detector efficiency response curves taking into account correlation effects has recently been published elsewhere [16].

The induced activities in the terbium samples were determined by measuring the γ rays at 298, 879, and 1177 keV associated with the decay of ^{160}Tb while counting the samples at source-detector distances of 25 and 15 cm. The long counting distances were selected to avoid true coincidence summing losses. A large number of γ ray spectra were collected for the irradiated terbium samples. A typical experimental γ ray spectrum of the spec pure terbium sample is shown in Fig. 1.

The induced radioactivities in the other irradiated activation detectors were also measured at the same counting distances as above. The necessary nuclear data of the radionuclides used in this work is given in Table I.

TABLE II. Measured flux parameters of the irradiation position RS 3 of Pakistan Research Reactor (PARR-1). (Φ is in n cm^{-2} sec^{-1}).

Deviation parameter α	-0.1623 ± 0.0055
Sub cadmium flux Φ_{th}	$1.98\times10^{13}\pm3.18\times10^{11}$
Epithermal flux Φ_e	$2.53\times10^{11}\pm1.11\times10^{10}$

IV. RESULTS AND DISCUSSION

The specific activities induced in the activation detectors were used to determine the cadmium ratios. Thermal and epithermal correction factors were calculated by using the expression described by Zweifel [23] and Baumann [24]. The experimentally measured cadmium ratios were used in expression (6) to determine the α parameter. The linear least-squares minimization was carried out and the function was iterated to obtain a consistent value of the α parameter (Table II). Figure 2 shows the excellent fitting of a straight line to the experimental data with a goodness-of-fit coefficient of 0.996. The pure thermal and epithermal neutron fluxes (Table II) were determined from the reaction rates measured from

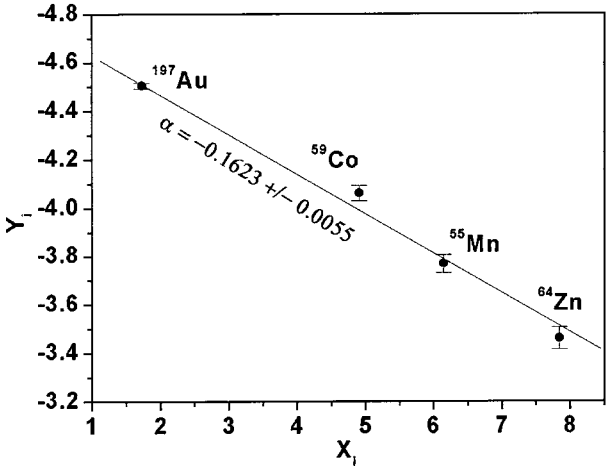

FIG. 2. α-Parameter measurement [X_i and Y_i are defined in Eq. (6)].

TABLE III. Thermal neutron cross section σ_o and resonance integral I_o of ^{159}Tb$(n,\gamma)^{160}$Tb reaction.

	σ_o(barn)	I_o(barn)
Present	23.6±0.4	443±21
Ref. [1]	25.5±1.1(1973)	628±24(1965)/450±50(1967)/780(1967)
	22±2(1967)	402(1967)/780(1969)/352±35(1970)
	23.2±0.5(1974)	429.5(1972)/365(40±1972)/376.4(1972)
	23.2±0.5(1964)	313(1973)/410.3(1973)/430±40(1973)
		434±21(1974)/400±24(1974)/455.45(1975)
		331±25(1976)/375±25(1976)
Ref. [14]	23.8±0.24	426±17(1987)
Ref. [26][a]	23.2±0.5	400±25(1987)
MUGHABGHAB [27][a]	23.4±0.4	418±20(1984)
JENDL-3.3 [28]	26.52	471

[a]Evaluations in Refs. [26,27] include Ref. [1] compilation data.

the induced activities of the radioactive products ^{60}Co and ^{198}Au with respective target samples irradiated under bare and cadmium cover conditions and by using Eqs. (2) and (3). The reaction rates of the ^{159}Tb$(n,\gamma)^{160}$Tb reaction were evaluated [Eq. (4)] from the spectral data of the bare and cadmium irradiated samples. The uncertainties in the overall measurements include (i) statistical errors, (ii) the error in the interpolated efficiencies, (iii) errors in the nuclear data (Table I), and (iv) target weighing errors.

The transmission factor of thermal neutrons through the cadmium, F_{cd}, was taken as 0.995 [25] to take into account the low-lying resonance at 3.339 eV.

The thermal neutron cross section of the reaction ^{159}Tb$(n,\gamma)^{160}$Tb was determined from the ratio of the sub cadmium reaction rates $(R-R'_e)$ to the thermal neutron flux. The same was also deduced from the ratio of sub cadmium reaction rates of terbium to the same ratio of the coirradiated cobalt monitor subsequently multiplied with the thermal cross section of ^{59}Co. The weighted average was taken as the final value of the cross section.

The resonance integral of the reaction was measured by using the cadmium ratios. The weighted mean of the measured cadmium ratios of the chosen γ rays was taken. The resonance integral was determined from the following expression, which is evaluated from Eqs. (2)–(4) followed by its conversion to I_o [Eq. (5)]:

$$Q_o(\alpha) = \frac{I_o(\alpha)}{\sigma_o} = \frac{fG_{th}}{(F_{cd}R_{cd}-1)G_e}, \tag{7}$$

where f is the thermal to epithermal flux ratio.

The measured thermal neutron cross section and resonance integral are presented in Table III along with earlier experimental and evaluated literature data. Comparison of the thermal neutron cross-section results show that the present value of 23.6±0.4 b is in good agreement with six literature values in Table III. However, two of the literature values, 25.5±1.1 b (BNL) and 26.52 b (JENDL 3.3), are somewhat higher. This comparison helps us to conclude that the true mean of the population of the thermal neutron cross sections of the reaction under study lies close to our value.

The resonance integrals in the literature, however, show a large scatter. If we look at the frequency of the literature values (Refs. [1] and [14] in Table III), we may divide them in three groups. We find that in seven cases the integrals are within the range 300–400 b (group 1), in eight cases they lie between 400 and 460 b (group 2) and in the other three cases they are higher than 600 b (group 3). The current measured resonance integral falls within group 2 and is on the higher side of the range. The present result is in agreement, within the uncertainty limit, with the other measurements of the group. The present value of the resonance integral (443±21 b) falls midway between the evaluations of Ref. [26] (400±25 b) and Ref. [28] (471 b) and agrees with the evaluation of Ref. [27] (418±20 b). It is clear that the resonance integral data in Table III merit reevaluation.

V. SUMMARY AND CONCLUSION

The thermal neutron cross section and resonance integral of the reaction ^{159}Tb$(n,\gamma)^{160}$Tb have been measured. The thermal cross section is in excellent agreement with those in the literature and supports the prevailing consistency in the data. The resonance integral measured in this work is in agreement, within the uncertainty limit, with several of the values compiled in Table III. Three ranges of resonance integral data have been identified. The group 2 data are supported by the present measurement and are recommended for the purpose of evaluation. The current results are recommended for use in applications and are expected to help in new evaluations of the thermal cross section and resonance integral.

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