Cross Section of the ^{159}Tb(n,γ)^{160}Tb Reaction at 0.0536 eV Energy

M. S. Uddin[1*] N. Afroze[2], M. A. Islam[1] and M. K. Newaz[3]

[1]*Institute of Nuclear Science and Technology, Atomic Energy Research Establishment Savar, GPO Box No. 3787, Dhaka-1000, Bangladesh*

[2] *Quality Assurance Unit, HEQEP, UGC Bhaban, Agargaon, Dhaka*

[3] *Institute of Computer Science, Atomic Energy Research Establishment, Savar, GPO Box No. 3787, Dhaka-1000, Bangladesh*

Abstract

The cross section of the ^{159}Tb(n,γ)^{160}Tb reaction at energy of 0.0536 eV was measured by activation technique. The activity produced in the irradiated target was measured by HPGe γ-ray detector. The ^{197}Au(n,γ)^{198}Au reaction was used to monitor neutron beam intensity effective in the target. The results amounting to 16.2±1.0 b is in good agreement with the ENDF/B-VII.1 data library. The measured value was extrapolated to 0.253 eV assuming 1/v dependence. Both the results are useful for confirmation of excitation curve as well as available integrally measured values at 0.0253 eV.

Keywords: Cross section, 0.0536 eV neutron energy, HPGe γ-ray detector, research reactor.

1. Introduction

Terbium is a rare earth element. Neutron-induced cross section data for Tb are of great importance both for nuclear technology development and for basic research. Precise values of the neutron capture cross section of terbium are of practical importance in relation to reactor design since this element is the fission product poison. Nuclear reactor design studies call for facility operation at a very low production rate of long-lived radionuclides to minimize the problems associated with reactor maintenance, low-level waste disposal and reactor decommissioning. In this point of view, it is very important to know the cross section of the formation of long-lived nuclei ^{160}Tb produced via the (n,γ) reaction on terbium.

The (n,γ) process has been extensively investigated, but all the measurements have generally been done at 0.0253 eV, i.e. integral measurements over the region of thermal energies. In the thermal region, the cross section follows the well-known 1/v law of neutron capture, but the point of normalization is the value at 0.0253 eV. Therefore, the values in the thermal region derived via the 1/v law would also be not very reliable if some discrepancy on the values at this average energy. It is therefore advisable to obtain experimental data at energies other than 0.0253 eV to be able to check the thermal value.

The aim of the present work was to measure cross sections of the reaction ^{159}Tb(n,γ)^{160}Tb by using monochromatic neutrons at energy of 0.0536 eV at Triple Axis Spectrometer (TAS) installed at TRIGA Mark II reactor, Savar, Dhaka as a part of our systematic studies [1-6]. Thereafter, the measured value was extrapolated to obtain value at 0.0253 eV and compared with the integrally measured values reported in the literature [7-13].

2. Materials and Methods

2.1 *Sample Preparation and Irradiation*

High-purity terbium foil (PFALTZ & BAUER, INC. ID-T00790; Purity: 99.99%; 500 μm thick) was used as the target for the irradiation. Terbium has single stable isotope ^{159}Tb (100%). The Tb foil was cut in circular disc with a diameter of 1.5 cm and then it was sandwiched between two Au foils (Tanaka Kikingzoku Kogyo; Purity: 99.99%; 25 μm thick) for irradiation. The diameter of Au foils was closed to that of Tb foil. The stack of Au-Tb-Au was irradiated for 10 h with monochromatic neutrons having energy of 0.0536 eV at the Triple Axes Spectrometer (TAS). During irradiation the reactor was operated at a power of 1.5 MW. The detail on neutron source has been described in elsewhere [1-6]. The presence of epithermal neutrons were checked by the irradiation of the Au foil covered with Cd.

2.2 *Gamma-ray Measurement and Data Analysis*

High-purity germanium (HPGe) (Canberra, 25% relative efficiency, 1.9 keV resolution at 1332.5 keV of ^{60}Co) gamma-ray detector was used to measure the activities produced in the target and monitor foils. The detector was associated with digital gamma spectrometry system (ORTEC DSPEC jr ^{TM}) and Maestro data acquisition software. The gamma-ray spectrum was analysed by GammaVision software. Due to weak activity and long half-life (72.3 d) of the product radionuclide ^{160}Tb, the irradiated terbium sample was measured for long time and at the surface of the detector to obtain good counting statistics. A gamma-ray spectrum for terbium is shown in Fig. 1. The ^{160}Tb radionuclide was identified by the peak at energy of 298.5 keV (26.1%) in the spectrum. To avoid interference from gamma-lines of undesired unknown sources and to obtain cross section value with adequate precision and accuracy, the sample was recounted 3 times giving enough intervals. From those three activities, the half-life of the radionuclide ^{160}Tb was checked and the result confirmed the γ-ray counts under the peak at 298.5 keV only for the decay of ^{160}Tb and the contribution from natural sources was negligible. As seen in Fig. 1, the peak at 300 keV due to ^{212}Pb is well separated and did not interfere to our desired peak at 298.5 keV.

In gamma-ray spectrum of the irradiated Cd-covered gold and aluminium foils, no peaks for the radionuclides ^{198}Au and ^{24}Na were found. It confirms the negligible amount of

Corresponding author: md.shuzauddin@yahoo.com

epithermal and fast neutrons in the beam used for the irradiation.

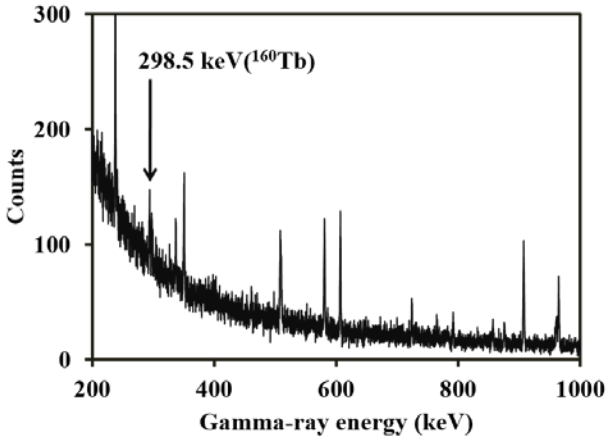

Fig. 1 A typical γ-ray spectrum of the irradiated Tb target: cooling time 3 days and measuring time 15000 sec; peak at 298.5 keV originated in the decay of ^{160}Tb.

The efficiency versus energy curve of the HPGe gamma-ray detector was determined using the standard point sources, ^{57}Co, ^{60}Co, ^{133}Ba, ^{137}Cs and ^{152}Eu. The energies of γ-rays emitted from the radioactive sources were in the range of 81 keV to 1408 keV. The efficiency versus energy curve at 15 cm from the detector surface was determined by measuring the activities of those sources, where both the random and real coincidences were negligible. From the efficiencies at the surface and at 15 cm from the surface obtained using the ^{137}Cs source, a normalization factor was calculated. Thereafter the efficiencies of all the selected energies at 15 cm were normalized to achieve sumcoincidence free efficiencies at the surface. Due to thin samples and the same diameter of both the target and monitor foil the geometry effects on efficiency was negligible.

The gamma-ray count rates of the reaction products were converted to reaction rates (R) by correcting for the gamma-ray intensities and the efficiency of the detector using the following formula:

$$R = \frac{\lambda C}{N \varepsilon I_\gamma \; e^{-\lambda t_c} \; (1-e^{-\lambda t_m}) \cdot (1-e^{-\lambda t_i})} \qquad (1)$$

where,

λ = decay constant, s^{-1}

C = total counts of gamma-ray peak

N = number of target atoms

ε = detector efficiency for the investigated peak

I_γ = gamma-ray intensity

t_c = cooling time, s

t_m = measuring time, s

t_i = irradiation time, s

For the monoenergetic neutrons, the cross section $\sigma(E)$ can simply be obtained as,

$$\sigma(E) = \frac{R}{\phi(E)} \qquad (2)$$

where, ϕ(E) = neutron flux, n cm^{-2} s^{-1}

The combined uncertainty in the experimentally determined cross section was estimated by taking the square root of the quadratic sum of the individual uncertainties: statistical uncertainty of γ-ray counting (0.5-5 %), peak area analysis (0.1- 0.5 %), half-life (0.3 %) sample mass (0.01 %), efficiency (2%), gamma-ray intensity (0.07 %) and neutron flux (3 %). The uncertainty in efficiency calibration amounting to ~4 % was reduced to ~2 %, because the same detector was used to measure the activity both in Tb and Au foils. The overall uncertainty in the cross section is around 6 % (1 sigma). The decay data of the residual radionuclides were taken from the NUDAT (2009) [14] database http://www.nndc.bnl.gov/nudat2.

2.3 *Neutron Flux*

The neutron fluxes both at entrance and exit of the Tb sample were determined from the activity of ^{198}Au formed by the ^{197}Au(n,γ)^{198}Au reaction in the two Au-foils. The standard cross section of the monitor reaction ^{197}Au(n,γ)^{198}Au at 0.0536 eV was evaluated from the experimental data reported by Yamamoto *et al.* [15], Pavlenko and Gnidak [16], Haddad *et al.* [17] and the data reported in ENDF/B-VII.1 [18] data library by a fitting procedure. The standard value of the above monitor reaction at 0.0536 eV amounted to 68.5 ± 1.7 b. The neutron beam intensities obtained at entrance and exit of the target are 9.68 x 10^3 $ncm^{-2}sec^{-1}$and 5.72 x 10^3 $ncm^{-2}sec^{-1}$. No activity was found in the Au-foil covered by Cd and that indicated the absence of epithermal neutrons.

2.4 *Data Correction*

The data were corrected for efficiency, coincidence loss, neutron and gamma attenuation. The uncertainties involved in all parameters needed to convert the count rate into decay rate were considered to deduce uncertainty in cross section. Due to weak activity, the uncertainty due to random coincidences as well as pulse pile up loss was negligible.

The effect of real coincidences was also considered. The correction factor for gamma-ray attenuation in the sample at a given gamma-ray energy at a fixed geometry for the case of a cylinder, coaxially positioned with the detector was calculated by considering the attenuation coefficient for terbium. For terbium the correction factor of gamma-ray attenuation was found 1.01 for 298.5 keV

3. Results and Discussion

The measured cross section of the ^{159}Tb(n,γ)^{160}Tb reaction at the energy of 0.0536 eV amounts to 16.2±1 b. This value was extrapolated to the value at energy of 0.0253 eV by following the 1/v dependence in the thermal region and the result is 23.6±1.4 b, which is comparable with the integrally measured values of the previous works [7-13].

Both the values are shown in Fig. 2 together with literature values. In order to validate the measured value, we constructed the excitation function for the above reaction, filling in the data available at other energies which are shown in Fig.2. Only the evaluated data reported in ENDF/B-VII.1 data library are given in Fig. 2, because the data reported in all the major evaluation libraries are almost the same to ENDF/B-VII.1. The measured values fall on the ENDF/B-VII.1 curve. Malik et al.[7] reported several points in the thermal energy region. Their values are systematically higher than the evaluated curve. Our assumption is that these discrepancies are possibly due to incorrect consideration of perturbations in the neutron irradiation field or errors in the induced activity measurements. The most of the previous integrally measured values are consistent with the normalized value at 0.0253 eV from the measured values at 0.0536 eV. Our measured cross section value at 0.0536 eV should thus serve as new experimental reference points.

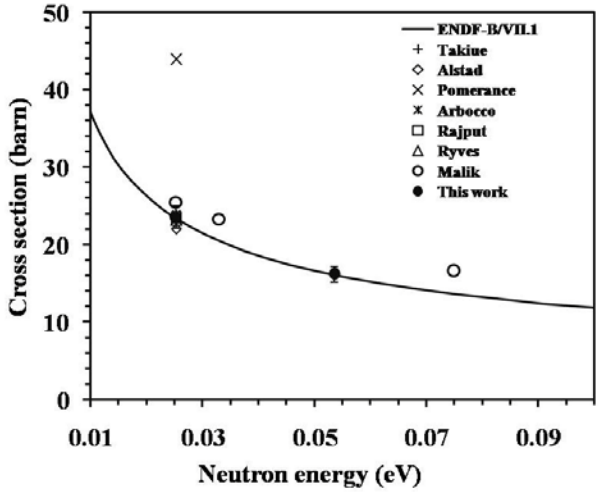

Fig. 2 Neutron capture cross sections for the $^{159}Tb(n,\gamma)^{160}$ Tb reaction. Solid circle-this work; measured at 0.0536 eV and other one extrapolated to 0.0253eV from our measured value.

4. Conclusion

The cross section of the reaction $^{159}Tb(n,\gamma)^{160}Tb$ in the thermal region was measured for the first time using monochromatic neutrons at energy of 0.0536 eV. The cross section at 0.0253 eV was also deduced from the measured values at 0.0536 eV assuming the 1/v dependence in the thermal region. The present value is in good agreement with the ENDF/B-VII.1 evaluation curve. The extrapolated value at 0.0253 eV serve as the confirmatory of integral value over thermal energy.

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