MEASUREMENT OF CROSS-SECTION FOR THE $^{139}La(n,\ )^{140}La$ REACTION USING REFLECTED NEUTRON BEAM AT 0.0334 eV ENERGY

by

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The experimental neutron capture cross-section for the $^{139}La(n, \gamma)^{140}La$ reaction at 0.0334 eV was measured for the first time using activation technique. The obtained cross-section value amounted to 7.86   0.55 b is in agreement with the evaluated data libraries ENDF/B-VII and JENDL-3.3. The present value measured at 0.0334 eV and the previous data at 0.0536 eV were extrapolated to the energy 0.0253 eV assuming 1/v dependence and the results were compared with the integrally measured values reported in the literature. The new data are thus useful to check the evaluated excitation function.

Key words: monochromatic neutron, neutron capture cross-section, lanthanum, activation technique, research reactor

INTRODUCTION

Lanthanum-139 (^{139}La) is essentially the only stable isotope (natural isotopic abundance 99.91 %) of the rare Earth element La. This stable isotope is generally used to produce the ^{140}La radioisotope via the $^{139}La(n, \gamma)^{140}La$ thermal neutron capture reaction. Study of isotopes with closed neutron shells such as ^{139}La (N = 82) are of special importance in nuclear physics. The cross-sections and nuclear structure of these nuclei provide useful information for fundamental nuclear physics studies [1], as well as for applications in nuclear astrophysics and nuclear technology. We chose to study the thermal neutron-induced reaction cross-section of the ^{139}La isotope mainly for three reasons: first, ^{139}La is not only an abundant fission fragment (5 % fission yield [2]) but, it is also widely used for neutron dosimetry in nuclear power plants because of the relatively long half-life ($t_{1/2}$ =1.67855 d) of ^{140}La; second, ^{139}La, known as the neutron magic isotope, is of special importance in the s- and r-process indicator in stellar nucleosynthesis [3, 4]; third, experimental thermal neutron activation cross-sections are very rare at energies other than the average energy 0.0253 eV due to the non-availability of mono-energetic neutron source in this energy region.

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The performances of the existing nuclear data libraries *e. g*., ENDF/B-VII, JENDL-3.3, JEFF, *etc*. depend on the presence of accurate and precise experimental reaction cross-section data. However, in some cases the evaluated data by the existing libraries differ significantly, as the experimental data that have been selected by the evaluators are significantly different. Thus, it is essential to determine experimental cross-section data at new energies other than 0.0253 eV which will help for the evaluators to evaluate accurate and precise data libraries. Moreover, accurate measurements of neutron capture cross-sections for a large number of nuclides, at the thermal energy region, are currently of great importance in neutron transport calculations and in reactor safety assessments. A number of authors have reported thermal neutron capture cross-sections on ^{139}La mostly at average thermal neutron energy of 0.0253 eV [5-20] and there are discrepancies among the data (8.1 to 9.5 b, where 1 b = 1 10^{-28} m^2, differ by about 17 %). In most of these studies, thermal neutron cross-sections were determined experimentally by the activation method using cadmium cut-off energy technique. In this technique special attempt is needed to correct the effect of fast neutrons, which were not absorbed of by Cd-shielding. Moreover, thermal neutrons cannot reach one side of the bare sample due to Cd-filtering. These effects may add a large uncertainty in the measured cross-section [21].

The experimental determination of neutron capture cross sections for various targets using the reflected monochromatized neutron beam (0.0536 eV) from the triple axis spectrometer (TAS) installed at the BAEC TRIGA Research Reactor (BTRR), Dhaka is well established [22-24]. Recently, a high resolution neutron powder diffractometer (locally called SAND) has been installed in the radial beam port-II of the BTRR for neutron scattering experiments. In this diffractometer, neutron beams are monochromized by (115) plane of Si single crystal with wave-length λ = 1.565 10^{-10} m, which corresponds to 0.0334 eV neutron energy. The neutron beam intensity is sufficient to make good activation of lanthanum target. In the present paper, we report on the measurement of the thermal neutron capture cross-section of the ^{139}La(n, γ)^{140}La reaction at the neutron energy of 0.0334 eV using neutron activation technique. To our knowledge, for this reaction there are no experimental neutron capture cross-section data available at this energy,. The measured values are compared with 1/v based evaluated cross-sections reported in the ENDF/B-VII [25] and JENDL-3.3 [26] data libraries.

EXPERIMENTAL

Neutron source

The BTRR is a research reactor having a maximum continuous thermal power output of 3 MW. The reactor installed at the campus of Atomic Energy Research Establishment (AERE), Savar, Dhaka, has several irradiation facilities including four external neutron beam tubes namely tangential, radial piercing, radial-1 and radial-2 beam ports. Neutrons coming out of the reactor are of various wavelengths. For this experiment at radial beam port of BTRR, neutrons are monochromatized before reaching the target for irradiation. In order to obtain appropriate monochromatic beam characteristics, a high performance neutron powder diffractometer namely Savar Neutron Diffractometer (SAND) has been installed at the radial beam port of the BTRR. A detail of the diffractometer as well as neutron monochromator facility has already been reported [27]. Therefore, a brief description is given here. The neutrons of various energies coming from the reactor core were passed through a sapphire crystal to filter fast neutrons before entering the monochromator. Monochromatization can very effectively be done by Bragg reflection from a Si(115) monochromator using a suitable single crystal. The thickness, offset angle and bending radius of silicon slabs have been selected to optimize the intensity and diffractometer resolution for the Si(115) reflection of the monochromator at 97 ° take-off angle and a sample to monochromator distance of 128.62 cm yielding a wavelength of λ =1.565 10^{-10} m, which corresponds to 0.0334 eV neutron energy. The uncertainty in the wavelength as well as energy was derived from refinement of the Ni standard; it amounted to 0.004 %. The diffractometer has been installed at the 334.91 cm from the reactor core, appropriate for the Si(115) reflection. After reflection, the monochromatic neutrons pass through a collimator and then focus on the target.

Sample preparation

The powder sample of high purity (99.99 %) lanthanum nitrate hexahydrate ($La(NO_3)_3.6H_2O$) was used as a natural isotopic target. The $La(NO_3)3.6H_2O$ powder was pressed with a hydraulic press (5 ton/cm^2) to prepare pellet (0.544 g) having diameter of 1.2 cm and thickness 0.2 cm. The pellet was sandwiched between two pure gold foils (having 25 μm of thickness and 12 mm in diameter). The thin gold foils were used to measure the effective neutron flux at 0.0334 eV using the^{197}Au(n, γ)198Au monitor reaction.

Irradiation and activity measurement

The pellet sample sandwiched between two gold foils was irradiated with unidirectional monoenergetic neutrons of 0.0334 eV for 2.5 h. During irradiation the reactor was operated at 2.4 MW power. To follow the effect of epithermal and fast neutrons, the cadmium covered gold foil and bare aluminum foil were also irradiated. The activities of target and monitor foils were measured nondestructively using a high-purity germanium (HPGe) gamma ray detector(Canberra, 25 % relative efficiency, 1.85 keV resolution at 1332 keV of ^{60}Co source) associated with digital gamma-ray spectrometry system (ORTEC DSPEC jr^{TM}) and Maestro data acquisition software. The gamma ray spectrum was analyzed by GammaVision software. The spectrum was also analyzed by the program Hypermet PC 5.12 and the results compared with that of GammaVision. To avoid interference from gamma ray lines of undesired sources and to obtain cross-section value with adequate accuracy and precision, the sample was measured 3 times (5, 10 and 20 h) giving enough intervals. As an example, a gamma ray spectrum of radioactive lanthanum nitrate hexahydrate is shown in fig. 1. In gamma ray spectra of the irradiated Cd-covered gold and bare Al foils, no peaks at energies of 411.8 keV and 1368.6 keV emitted in the decay of ^{198}Au and ^{24}Na, were found. It confirms the negligible amount of epithermal and fast neutrons in the beam used for irradiation of the target.

DATA ANALYSIS

Cross-section determination

The gamma ray count rates of the reaction products were converted to the reaction rates (R) by cor-

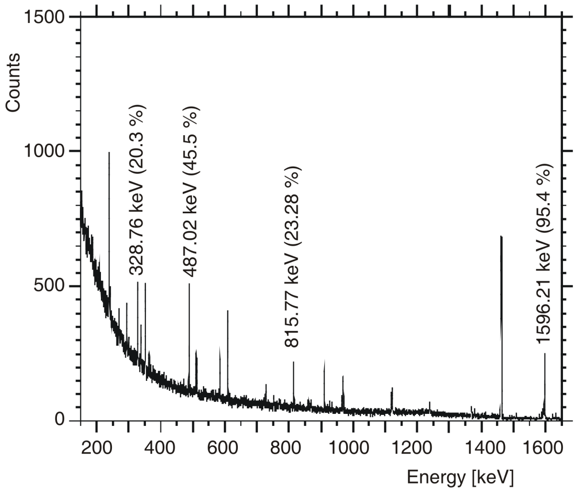

Figure 1. Gamma-ray spectrum of the lanthanum nitrate irradiated with thermal neutrons

recting for the gamma ray intensities and the efficiency of the detector using the following formula

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

where λ is the decay constant (s^{-1}), C – the total counts of gamma ray peak area, N – the number of target atoms, ε – the detector efficiency for the investigated peak, I_γ – the branching ratio of gamma-ray, t_c – the cooling time (s), t_m – the counting time (s), and t_i – the irradiation time (s).

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

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

where $\phi(E)$ [cm^{-2}s^{-1}] is the neutron flux.

The decay data of the radioactive products were taken from the NUDAT database [28]. The decay data are quoted in tab. 1. The efficiency *vs*. energy curve of the HPGe gamma ray detector for the counting distance was determined using the standard point sources, ^{22}Na, ^{60}Co, ^{57}Co, ^{133}Ba, ^{137}Cs, and ^{152}Eu. The combined uncertainty in cross-section was estimated by taking the square root of the quadratic sum of the individual uncertainties as given in tab. 2. The overall uncertainty in the cross-section is around 7 % (1σ).

Table 1. Decay characteristics of the investigated nuclides [28]

Nuclear reaction	Half-life	Gamma-ray energy [keV]	Branching ratio [%]
^{139}La(n, γ)^{140}La	1.678 ± 0.001d	328.76	20.3 ± 0.3
		487.02	45.5 ± 0.6
		815.77	23.28 ± 0.02
		1596.21	95.4 ± 0.1
^{197}Au(n, γ)^{198}Au	2.695 ± 0.002 d	411.8	95.5

Table 2. Sources of uncertainties in the measured cross section

Sources of uncertainty	Uncertainty [%]
Statistical uncertainty of gamma-ray counting	1-2
Peak area analysis	2
Decay branching ratio	0.5
Efficiency calibration	4
Sample mass	0.01
Half-life	0.01
Isotopic abundance	0.016
Self-shielding factor	0.01
Neutron flux	5
Total	~7

Neutron beam intensity and attenuation in the target

The neutron beam intensities at the entrance and exit of the target were determined using the ^{197}Au(n, γ)^{198}Au reaction induced in the Au-monitor foils. The monitor foils were irradiated simultaneously and measured with the same detector and in a comparable geometry as the lanthanum target. The main problem with the monitor reaction lies in the selection of the standard cross-section, because no recommended cross section value has been reported at 0.0334 eV energy. We evaluated the standard cross-section of the ^{197}Au(n, γ)^{198}Au reaction at 0.0334 eV from the experimental data reported in different literatures [29-31] by a fitting procedure. The obtained value is 86.3 ± 2.1 b at 0.0334 eV. The detailed evaluation of the monitor reaction cross-section has been described in our previous study [5]. From the cross-section value and measured activity of ^{198}Au, the neutron beam intensity was calculated using eqs. (1) and (2). The neutron beam intensities $4.75 \times 10^4 \pm 2375$ cm^{-2}s^{-1} and $4.15 \times 10^4 \pm 2075$ cm^{-2}s^{-1} were found at the entrance and exit of the target, respectively. The average value of these two, which was $4.45 \times 10^4 \pm 2225$ cm^{-2}s^{-1}, was used for cross-section determination. The neutron beam intensities at the entrance and exit of the target show that their difference is 13%, which describes the neutron attenuation as they pass through the lanthanum target during irradiation.

Gamma ray attenuation

The correction factor for gamma ray attenuation in the sample at a given energy, with fixed geometry for the case of cylinder, coaxially positioned with the detector, was calculated by considering the attenuation coefficients for lanthanum, nitrogen, hydrogen, and oxygen using the equation

$$F_g = \frac{\mu x}{1 - e^{-\mu x}} \tag{3}$$

where μ [cm^{-1}] is the linear attenuation coefficient and x [cm] – the sample thickness. The attenuation is also dependent on density ρ, which was determined from the measured mass, diameter, and thickness. The mass attenuation coefficient μ/ρ [cm^2g^{-1}] for constituting elements was used from Lide, [32]. The total mass attenuation coefficient for the compound lanthanum nitrate hexahydrate was converted to a linear attenuation coefficient by multiplying with the density of the sample. The correction factors for gamma ray attenuation along the pellet were found to be 1.04, 1.03, 1.02, and 1.01 for the 328.76, 487.02, 815.77, and 1596.21 keV gamma ray energies, respectively.

RESULTS AND DISCUSSION

The radionuclide ^{140}La was identified by its characteristic gamma ray lines of energies 328.76, 487.02, 815.77, and 1596.21 keV as shown in fig. 1. The cross-section values determined individually for the above gamma lines are consistent with each other and the average cross-section value deduced by the gamma ray energies is reported. The coincidence loss at 815.77 keV was considered to be 8 % in cross-section calculation. The necessary corrections for neutron absorption and gamma ray attenuation in target were also taken into account during cross-section calculation.

As far as our knowledge goes, this is the first experimental neutron capture cross-section for the $^{139}La(n, \gamma)^{140}La$ reaction at 0.0334 eV. The measured cross-section with uncertainty at this energy is quoted in tab. 3 together with other literature values at neutron energies of 0.0253 eV and 0.0536 eV. The measured cross-section at 0.0334 eV and our previous data at 0.0536 eV [5] are individually extrapolated to the values at energy of 0.0253 eV by following the 1/v dependence in the thermal region and the results are 9.03   0.63 b and 8.92   0.44 b, respectively. These values are in good agreement with each other. The average of these values at 0.0253 eV is also listed in tab. 3. In order to validate the measured values, we constructed the excitation function for the reaction filling in the data available at other energies which are shown in fig. 2.

A number of authors have reported experimental thermal neutron capture cross-section for the

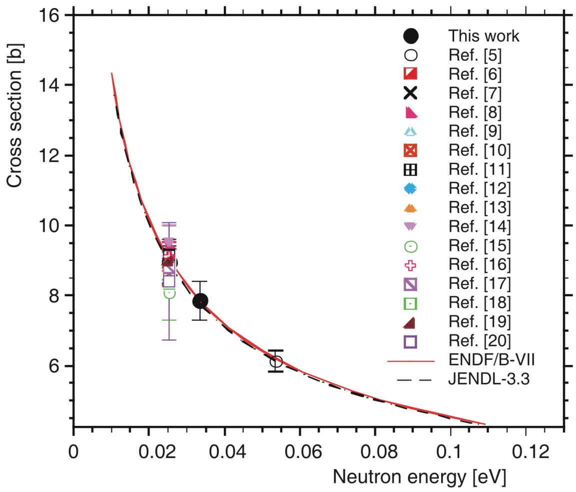

Figure 2. Neutron capture cross-section of the $^{139}La(n, \gamma)^{140}La$ reaction; solid circle-this work at 0.0334 eV and 0.0253 eV, open circle at 0.0536 eV [5]

Table 3. Thermal neutron capture cross-section of the $^{139}La(n, \gamma)^{140}La$ reaction

Year	Reference	Neutron energy [eV]	Cross-section [b]
2015	This work	0.0334	7.86 0.55
2010	Chowdhury *et al.* [5]	0.0536	6.13 0.30
		0.0253	8.98 0.63 (extrapolated*)
2014	Do *et al.* [6]	0.0253	9.16 0.36
1999	Holden [7]	0.0253	9.2 0.2
1988	Corte *et al.* [8]	0.0253	9.43 0.09
1978	Heft [9]	0.0253	9.18 0.05
1978	Takiue and Ishikawa [10]	0.0253	8.63 0.34
1975	Gleason [11]	0.0253	9.15 0.25
1975	Mannhart [12]	0.0253	8.93 0.03
1971	Ryves, Perkins [13]	0.0253	9.03 0.33
1967	O'Brien Jr. *et al.* [14]	0.0253	9.5 0.5
1960	Lyon [15]	0.0253	8.10 0.81
1957	Cummins [16]	0.0253	9.1 0.2
1951	Pomerance [17]	0.0253	8.80 0.44
1951	Benoist *et al.* [18]	0.0253	8.35 0.1
1950	Harris *et al.* [19]	0.0253	9.01 0.05
1947	Seren *et al.* [20]	0.0253	8.40 1.68

* Using the 1/v trend

^{139}La(n, γ)^{140}La reaction and it is interesting that all of those values are at 0.0253 eV neutron energy except our previous data at 0.0536 eV energy [5] (tab. 3). The reported data at 0.0253 eV are in the range of 8.1-9.5 b. To measure cross-section at average thermal neutron energy of 0.0253 eV, they exposed the target in a mixed neutron beam of thermal and epithermal energies for activation. They subtracted the contribution of the epithermal neutrons by the Cd cut-off energy technique. The Cd cut-off energy technique is rather complex. On the other hand, in the present study, a careful measurement at neutron energy 0.0334 eV has been carried out and the obtained cross-section is 7.86   0.55 b. The pure thermal neutron beam of 0.0334 eV energy was used for irradiation of the target, where the complex Cd cut-off energy technique was not involved. Our technique is very simple to obtain data with good precision and accuracy. Data reported both in the ENDF/B-VII and JENDL-3.3 libraries are evaluated and no measurements have been done at 0.0334 eV energy. In these libraries the value at 0.0334 eV is calculated from the thermal cross-section at 0.0253 eV using the $1/v$ relationship. The calculated value of ENDF/B-VII at 0.0334 eV is about 1.4 % higher than that of JENDL-3.3. Assuming a $1/v$ cross-section dependence ENDF/B-VII and JENDL-3.3 give neutron capture cross-sections at 0.0334 eV of 7.88 b and 7.77 b, respectively. Our measured cross-section 7.86   0.55 b is 0.25 % lower than that of ENDF/B-VII but 1.2 % higher than that of JENDL-3.3. If we compare the present normalized value at 0.0253 eV from the measured values at energies 0.0334 eV and 0.0536 eV with the previous integrally measured values, it is observed that the present value is consistent with the previous integrally measured values within associated uncertainties. It is clear that the present cross-section value at 0.0334 eV energy can play an important role to check the energy dependence of neutron capture cross-sections in the thermal region.

CONCLUSIONS

The neutron capture cross-section of the ^{139}La(n, γ)^{140}La reaction at 0.0334 eV neutron energy was measured for the first time using monochromatic unidirectional neutrons coming out of a high resolution neutron powder diffractometer established at the BTRR in Dhaka. The cross-section at 0.0253 eV was also deduced from the measured values at 0.0334 eV and 0.0536 eV energies assuming the $1/v$ dependence in the thermal region. The obtained new results are in good agreement with the evaluated data libraries ENDF/B-VII and JENDL-3.3. The present value at 0.0334 eV and our previous measured value at 0.0536 eV, for this reaction, would be useful to confirm the reliability of the data evaluated by $1/v$ relation in the nuclear data libraries. Our study may encourage cross-section measurements for various reactions, at different energies in the thermal energy region, at many places in the world where neutron diffractometer and spectrometer are installed.

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AUTHORS' CONTRIBUTIONS

The research plan and experimental work were performed by S. M. Hossain, M. A. Islam, and M. S. Uddin. The data analysis and manuscript preparation were performed by M. A. Islam. Tables and figures were prepared by F. Akter, K. Naher, and U. Tamim. All the authors participated in the discussion of the results presented in the paper.

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МЕРЕЊЕ ПРЕСЕКА ЗА СУДАР РЕАКЦИЈЕ ^{139}La(n, γ)^{140}La КОРИШЋЕЊЕМ РЕФЛЕКТОВАНОГ НЕУТРОНСКОГ СНОПА ЕНЕРГИЈЕ 0.0334 eV

Пресек за захват неутрона у реакцији ^{139}La(n, γ)^{140}La на 0.0334 eV експериментално је по први пут одређен коришћењем активационог поступка. Добијена вредност пресека за судар износила је 7.86   0.55 b, у сагласности са провереним библиотекама података ENDF/B-VII и JENDL-3.3. Измерена вредност на 0.0334 eV и ранији податак на 0.0536 eV екстраполисани су до енергије 0.0253 eV претпостављајући 1/v зависности и резултати су упоређени са интегрално измереним вредностима документованим у литератури. Отуда су нови подаци корисни при провери процењене функције побуде.

Кључне речи: монохроматски неутрон, пресек за захват неутрона, лантан, активациона техника, истраживачки реактор