Measurement of Neutron Capture Cross Sections of ^{139}La, ^{152}Sm and ^{191,193}Ir at 55 and 144keV

Vuong Huu Tan
Vietnam Atomic Energy Commission
59-Ly Thuong Kiet, Hanoi, Vietnam

Tran Tuan Anh, Nguyen Canh Hai and Pham Ngoc Son
Dalat Nuclear Research Institute
01-Nguyen Tu Luc, Dalat, Vietnam

Tokio Fukahori
Nuclear Data Center, Nuclear Science and Engineering Directorate
Japan Atomic Energy Agency
Tokai-mura, Naka-gun, Ibaraki-ken, 319-1195, Japan

The neutron capture cross sections of ^{139}La, ^{152}Sm and ^{191,193}Ir at average energies of 55keV and 144keV have been measured relative to the standard capture cross sections of ^{197}Au by means of the activation method. The neutron beams were derived by filtered techniques from the horizontal channel No.4 of the research reactor at the Dalat Nuclear Research Institute, Vietnam. A fast-digital gamma-ray spectroscopy in compacted with a 58% efficient HPGe detector has been used for measurements of gamma-ray spectra from the activated samples. The correction factors for multi-scattering, self-shielding and resonance capture effects of neutron in the irradiating samples were calculated by Monte-Carlo technique. The present results have been compared with the previous measurements, and the evaluated data from JENDL-3.3 and ENDF/B-VI.8 libraries.

Key words: *neutron, capture cross section, nuclear reaction data, filtered neutron beam, resonance neutron capture, correction*

1. Introduction

Accurate measurements of neutron capture cross sections for most of nuclides are currently necessary for the calculations of neutron transport, the assessments of the reactor safety, the investigations of high-burn-up core characteristics, the decay heat power predictions, and for the nuclear transmutation study. In keV energy region, the (n,γ) cross sections of the nuclides at or near magic neutron number, N=50, 82 and 126 are special important for the study on the s-process reaction chain for nucleosynthesis. However, the present status of experimental data for capture cross sections is still inadequate both in quality and in quantity. Therefore, it is important to perform the precisely measurements of capture cross sections for those nuclides, particular in keV energy region [1-6].

In the present experiment, we performed the measurements of capture cross section of ^{139}La, ^{152}Sm and $^{191, 193}$Ir on the filtered neutron beams of 55keV and 144keV, relative to the standard capture cross section of ^{197}Au by the activation method. The neutron beams were derived from the horizontal channel No.4 of the research reactor at the Dalat Nuclear Research Institute (DNRI), by using the filtered compositions of 98cmSi + 35g/cm^{2}S + 0.2g/cm^{2}B^{10} and 98cmSi + 1cmTi + 0.2g/cm^{2}B^{10} for 55keV and 144keV respectively [6,7]. The neutron energy resolution, FWHM, is 8keV at 55keV peak, and 22keV at 144keV peak [7].

Beside determining the corrections for neutron multi-scattering and self-shielding in irradiated samples, it is important to concern that the large resonance capture cross sections of the standards and samples, in the slow neutron background region above the Cd-Cutoff energy, strongly contribute to the uncertainty of experimental results. Therefore, the correction factors for slow neutron resonance capture in the present experiments were also calculated by the general least square method for strong resonance captures at 4.92eV of ^{197}Au, 8.06eV of ^{152}Sm, 1.3eV of ^{193}Ir, 0.67eV, 5,38eV and 6.15eV of ^{191}Ir, and 72.3eV of ^{139}La. Furthermore, a low background and fast-digital spectroscopy with a high efficiency, 58%, HPGe detector has been used for detection of gamma-ray spectrum from the irradiated samples, and the statistical uncertainties are expected to be less than 1%.

2. Experiments

The measurements for neutron capture cross sections of ^{139}La, ^{152}Sm and $^{191, 193}$Ir at the energies of 55keV and 144keV were performed on the filtered neutron beams of DNRI. The neutron beams were collimated to 3cm in diameter by using the usual materials of LiF, Cd, B_4C, Pb and borated paraffin. The physical properties of these beams are given in Table 1. Since the neutron spectra had been experimentally measured before with a recoil-proton counter [6], in this work, the neutron transport and unfolding methods [8,9] were applied to obtain the exact spectra, shown in Fig. 1 and 2, which have been used for calculation of the average quantities and of the correction factors.

The samples were prepared from the natural oxide powders, 99.99% purity, of La_2O_3, Sm_2O_3 and IrO_2. In order to diminish the effect of water on the samples [10], each collected amount of the powders was dried up at about 105°C for several hours before weighting and pressing into pellets. Then the pellets were covered by thin polyethylene foils. The standard gold foils with 2.54cm in diameter and 0.01mm in thickness were used as the neutron flux monitors. Each sample was sandwiched between two gold disks, and the sample groups were wrapped in Cd covers with 0.5mm in thickness with aim to reject most of thermal neutron background. The irradiation time was 70 hours for every sample group.

The specific activities of the samples and the gold disks were measured by using the fast-digital spectroscopy system and with the high efficiency HPGe detector. The system was calibrated by using standard radioisotope sources and a multi-nuclides standard solution, supported by IAEA. Each of irradiated samples, standards and calibrated sources was measured under the same conditions.

Table 1 The properties of the filtered neutron beams [6]

Neutron energy (keV)	Filter combination	Flux density (n/cm^2/s)	FWHM
55	98cmSi + 35g/cm^{2}S + 0.2g/cm^{2}B^{10}	5.61 x 10^5	8 keV
144	98cmSi +1cmTi + 0.2g/cm^{2}B^{10}	2.14 x 10^6	22 keV

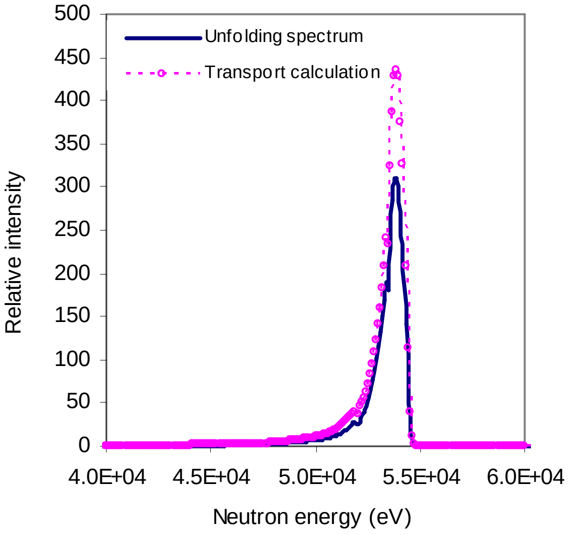

Fig. 1 Neutron spectrum of the 55keV filtered beam

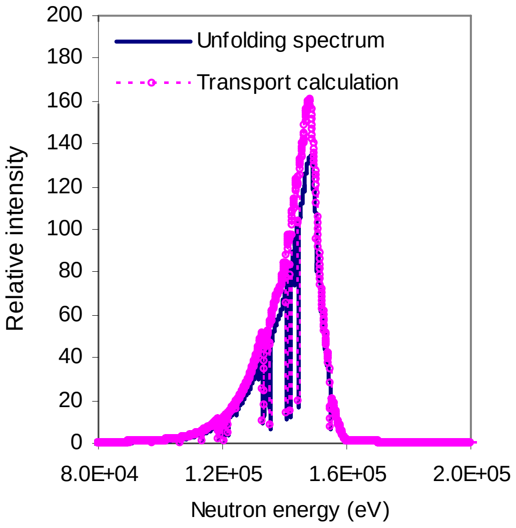

Fig. 2 Neutron spectrum of the 144keV filtered beam

3. Data Analysis

During irradiation in the neutron beam with energy spectrum Φ(E), the reaction rate, R, of samples is defined as follows:

$$R = N\int \Phi(E)\sigma_a(E)dE \;, \tag{1}$$

where N is the number of nuclei in sample, and $\sigma_a(E)$ is the neutron capture cross section at energy E. The average neutron capture cross section, $<\sigma_a>$, and neutron flux, $<\Phi>$, are defined as following:

$$<\sigma_a>= \int \sigma_a(E)\Phi(E)dE \,/ \int \Phi(E)dE \;\; ; \;\; <\Phi>= \int \Phi(E)dE \;.$$

Applying these average quantities, the integrating equation (1) can be rewritten:

$$R = N <\sigma_a><\Phi> \tag{2}$$

The activity, A, of the sample at the end of neutron irradiation is given by expressions:

$$A = R(1-\exp(-\lambda t_1)) \;, \tag{3}$$

$$A = \frac{Cf_c\lambda}{\varepsilon_\gamma I_\gamma \exp(-\lambda t_2)(1-\exp(-\lambda t_3))} \;, \tag{4}$$

where C denotes the net counts of the corresponding gamma peak, and t_1, t_2 and t_3 are irradiating, cooling and measuring times, respectively. λ is the decay constant of the product nucleus, ε_γ the detection efficiency of detector, I_γ the intensity of interesting γ-ray line and f_c is the correction factors. Finally, from equations (2), (3) and (4), the average capture cross sections, $<\sigma_a>^x$, for the samples at average neutron spectrum <Φ> can be obtained relative to that of ^{197}Au standard by the following relations:

$$<\sigma_a>= \frac{C^x f(\lambda,t)^x f_c^x I_\gamma^{Au} \varepsilon_\gamma^{Au} N^{Au} <\sigma_a>^{Au}}{C^{Au} f(\lambda,t)^{Au} f_c^{Au} I_\gamma^x \varepsilon_\gamma^x N^x} \tag{5}$$

$$f(\lambda,t) = \frac{\lambda}{(1-\exp(-\lambda t_1))\exp(-\lambda t_2)(1-\exp(-\lambda t_3))} \;, \tag{6}$$

where the superscript 'x' denotes the nucleus of sample. Calculating from the ENDF/B-VI.8 data library, the average standard capture cross section of ^{197}Au respects to the 55keV beam's spectrum is 414.61mb, and for that to the 144keV beam's spectrum is 277.21mb. The relevant decay data of product nuclei, used in this work, are given in Table 2.

Table 2 Decay properties of the product nuclei [11]

Product nucleus	Half-life	γ-ray energy (keV)	Intensity per decay (%)
^{198}Au	2.6952±0.0002 d	411.8	95.6±0.1
^{140}La	1.6781±0.0003 d	487.02	45.5±0.6
^{153}Sm	46.50±0.21 h	103.2	29.3±0.1
^{192}Ir	73.827±0.013 d	316.5	82.7±0.2
^{194}Ir	19.28±0.13 h	328.45	13.1±1.7

The correction factors for the neutron self-shielding, multi-scattering and the effects of strong resonance capture of samples with slow neutron background were calculated by Monte-Carlo method [9, 12]. In which, the effect of isotopic impurities and oxygen in the samples and the loss energy by scattering of neutron were taken into account. In the resonance capture corrections, the background spectra of neutron beams were measured by unfolding method and resonance thin-foil activation technique. The data used for the correction calculation were taken from JENDL-3.3 [13] and ENDF/B-VI.8 [14]. The calculated correction factors are given in Table 3.

Table 3 Correction factors for multi-scattering, self-shielding and background resonance capture of neutron in the samples

Nuclides	55keV region			144keV region		
	Self-shielding	Multi-scattering	Resonance capture	Self-shielding	Multi-scattering	Resonance capture
Au-197	0.9985	0.9901	0.4269	0.9988	0.9929	0.5338
La-139	0.9962	0.9785	0.6227	0.9986	0.982	0.7531
Sm-152	0.9988	0.9856	0.2816	0.9991	0.9917	0.4890
Ir-191	0.9959	0.9782	0.4937	0.9968	0.9828	0.6593
Ir-193	0.9959	0.9774	0.5214	0.9968	0.9826	0.6944

4. Results and Discussion

In the present work, the new values of average neutron capture cross sections of ^{139}La, ^{152}Sm and $^{191, 193}$Ir at incident neutron energies of 55keV and 144keV are reported with errors about 5-6.5%. The results are given in Table 4. The uncertainties in the present measurements are mainly due to the statistical errors (0.1-2%), the uncertainties of γ-ray detection efficiency (3.5%), the reference cross section (~3%) and the correction factors for neutron resonance capture, self-shielding and multi-scattering effects (~3%). In comparisons with the previous measurements and the evaluated data, The present results are seem to be good agreement with the previous measurements of Musgrove [15], Wisshak [16], Duamet [17], Macklin [18] and with the evaluated data of JENDL-3.3 and ENDF/B-VI.8 within the experimental uncertainties. The comparisons results are shown in Figs.3-6.

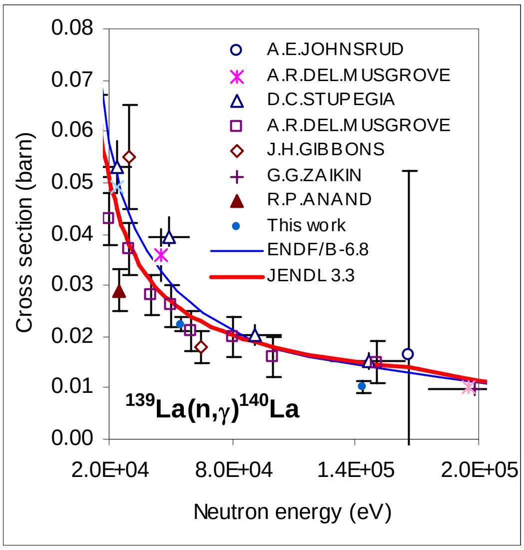

Fig. 3 Neutron capture cross section of ^{139}La in keV region

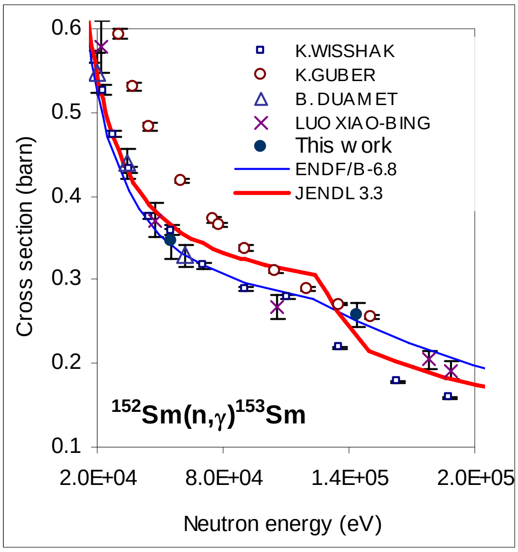

Fig. 4 Neutron capture cross section of ^{152}Sm in keV region

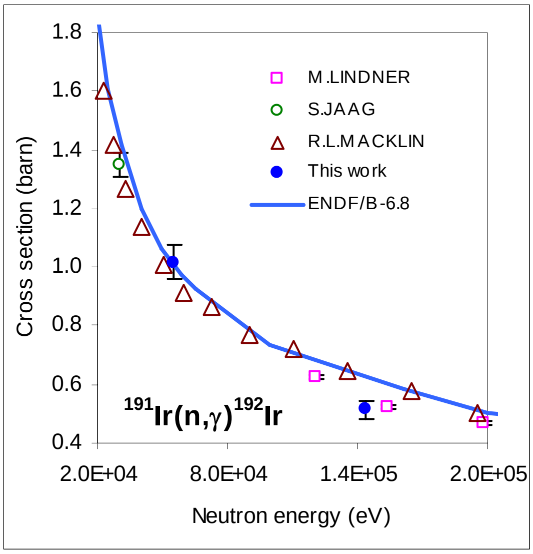

Fig. 5 Neutron capture cross section of ^{191}Ir in keV region

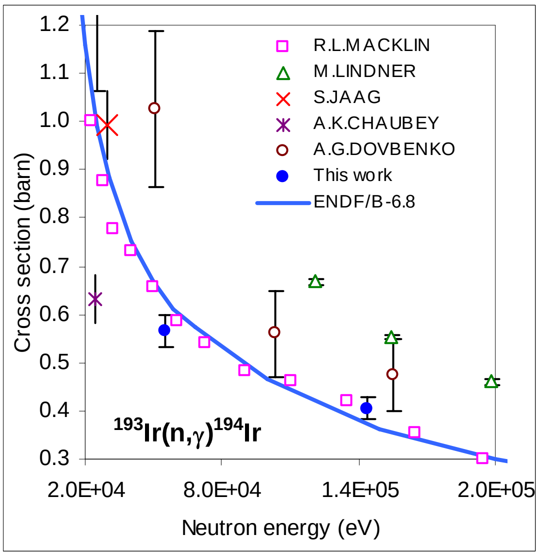

Fig. 6 Neutron capture cross section of ^{193}Ir in keV region

Table 4 The neutron capture cross sections of ^{139}La, ^{152}Sm and $^{191, 193}$Ir obtained in the present study

Average neutron energy [Energy range] (keV)	$<\sigma_a>^{La-139}$ (mb)	$<\sigma_a>^{Sm-152}$ (mb)	$<\sigma_a>^{Ir-191}$ (mb)	$<\sigma_a>^{Ir-193}$ (mb)
55 [51-59]	22.4 ± 1.2	345.5 ± 19.4	1016.5 ± 57.2	566.7 ± 32.6
144 [133-155]	12.01 ± 0.58	258.7 ± 14.5	514 ± 29.4	404.5 ± 22.8

5. Conclusion

The neutron capture cross section of ^{139}La, ^{152}Sm and $^{191, 193}$Ir at average incident neutron energies of 55keV and 144keV have been measured by means of the activation method, using the filtered neutron beams at DNRI. The results, with uncertainties of 5-6.5%, were obtained relative to the standard capture cross sections of ^{197}Au. The energy ranges of the filtered neutron beams are 14 and 15% for 55keV and 144keV beams, respectively. Although the slow neutron background existing in the filtered neutron beam is quite low, the effects of strong resonance capture cross sections of the samples and standards should be taken into account to improve the accuracy of the experimental results.

Acknowledgments

We would like to express our thanks to the Vietnam Atomic Energy Commission (VAEC) and the Dalat Nuclear Research Institute (DNRI) for their great encouragement and helpful in creating an advantage research condition for this work. The present study was sponsored by the Fundamental Research Program of VAEC under the subproject No. 409706. The advanced gamma-ray spectroscopy system used in the experiments was supported by IAEA. We also wish to express our deep gratitude to the members of the Nuclear Data Center of JAEA for their helpful reference data and materials.

References

[1] K. H. Guber, et al., Neutron Cross Section Measurements at the Spallation Neutron Source, Int. Conf. on Nuclear Data for Science and Technology, Tsukuba (JP), 7-12, P. 281, Oct. 2001.

[2] Stefano Marrone, Measurement of the ^{139}La(n,γ) Cross Section at n_TOF, 12th Conference on Capture Gamma-Ray Spectroscopy and Related Topics, CGS12, 4-9 (2005).

[3] S. O'Brien et al., Neutron capture cross section of ^{139}La, Phys. Rev. C68, 035801 (2003).

[4] Z. Y. Bao, H. Beer, F. Kappeler, F. Voss, And K. Wisshak, Neutron Cross Sections for Nucleosynthesis Studies, Atomic Data and Nuclear Data Tables **76,** 70–154 (2000).

[5] Kazuya OHGAMA, et al., Measurement of keV-Neutron Capture Cross Sections and Capture Gamma-Ray Spectra of ^{91,92}Zr, J. Nuclear Science and Technology, Vol. 42, P. 333 (April 2005).

[6] P. Z. Hien, V. H. Tan, N. C. Hai. Radiactive capture cross section measurements for U-238 on the 55 keV and 144 keV filtered neutron beams, KC-09-08, Dalat (1995), P. 117.

[7] V. P. Vertebnyj, et al., Filtered Medium and Thermal Neutron Beams and Their Use, "Properties of Neutro Sources", IAEA-TECDOC-401, (1986).

[8] O. O. Gritzay, et al., Neutron Filters at Kyiv Research Reactor, KIRN-01-6 (2001).

[9] K. Senoo, Y. Nagai, T. Shima, T. Ohsaki, A Monte Carlo code for multiple neutron scattering events in a thick sample for (n, y) experiments, Nucl. Instr. and Meth. A339 (1994) 556-563.

[10] Motoharu Mizumoto and Masayoshi Sugimoto, The Influence of Water Absorption in Samples for Neutron Capture Cross Section Measurements, Nucl. Instr. and Meth., A282 (1989) 324.

[11] Nudat 2.2 Database, http://www.nndc.bnl.gov/nudat2/

[12] Oleg Shcherbakov and Hideo Harada, Resonance Self-Shielding Corrections for Activation Cross Section Measurements, J. Nucl. Sci. Tech., Vol. 39, No. 5, P. 548 (2002).

[13] K. Shibata, T. Kawano, T. Nakagawa, O. Iwamoto, J. Katakura, T. Fukahori, S. Chiba, A. Hasegawa, T. Murata,H. Matsunobu, T. Ohsawa, Y. Nakajima, T. Yoshida, A. Zukeran, M. Kawai, M. Baba, M. Ishikawa, T. Asami, T. Watanabe, Y. Watanabe, M. Igashira, N. Yamamuro, H. Kitazawa, N. Yamano and H. Takano: "Japanese Evaluated Nuclear Data Library Version 3 Revision-3: JENDL-3.3," *J. Nucl. Sci. Technol.* **39**, 1125 (2002).

[14] V. McLane and Members of the Cross Section Evaluation Working Group, "ENDF/B-VI Summary Documentation (ENDF-201)", BNL-NCS-17541, Brookhaven National Laboratory (1996).

[15] A. R. de L. Musgrove, B. J. Allen, R. L. Macklin, Resonance Neutron Capture in ^{139}La, Aust. J. Phys., 30, 599-604 (1977).

[16] K. Wisshak, et al., Neutron Capture in ^{148,150}Sm: A Sensitive Probe of the S-Process Neutron Density, Phys. Rev. C48, 3, (1993).

[17] B. Duamet, et al. Measurement of keV-Neutron Capture Cross Sections and Capture Gamma-Ray Spectra of ^{147,148,149,150,152,154}Sm, J. Nucl. Sci. Tech., Vol. 36, p. 865-876 1999).

[18] R. L. Macklin, D. M. Darke and J. J. Malanify, Fast Neutron Capture Cross Section of ^{169}Tm, ^{191}Ir, ^{193}Ir, ^{175}Lu for En from 3-2000 keV, LA-7479-MS, Aug. 1978.