Resonance Integral of Neptunium(^{237}Np) from Energy Dependent Differential Neutron Capture Cross-Section by Using the Linac TOF Method and C$_6$D$_6$ Scintillation Spectrometer

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Abstract

^{237}Np is very important material in the fission products of nuclear reactors. Resonance integral(RI) tests of this material is necessary to check between the experiments and the evaluated data. Such feedback to the evaluated data is very important to correct data and improve of codes. The RI for the ^{237}Np(n,ɣ)^{238}Np reaction were measured by using the 46−MeV electron linear accelerator (linac) at the Research Reactor Institute, Kyoto University (KURRI). The measurement was performed in the energy region from 0.005 eV and 10 keV. RI obtained as 804.7 barns, compared with those of the evaluated data in JENDL−4.0 and Mughabghab.

Keywords: Resonance integral, ^{237}Np(n,ɣ)^{238}Np, Differential, Neutron, Capture, Cross-section, Neptunium, TOF method

I. Introduction

Neptunium (^{237}Np) is a significant radioisotope in reactors because of a fission product which is produced in light reactor and fast breeders reactors[1]. Therefore, the precise measurement of the Resonance Integral(RI) for the ^{237}Np(n,ɣ)^{238}Np reaction is of great importance in the design of a reactor core because of varying the neutron flux[2]. Furthermore, the efficiency of reactor is greatly influenced by their fission products ratio. However, the previous measurement have large discrepancies among the their experiment data and evaluated data as shown in Table 1, because it has a very big s-wave resonance at 0.491 eV [3-11] . In generally RI data were measured by using activation method with nuclear reactor. However we tried to measure the RI by summing the differential neutron capture data which is from TOF method. We

have obatined the RI for ^{237}Np with the neutron time-of-flight (TOF) method by using the 46-MeV electron linear accelerator (linac) at the Research Reactor Institute, Kyoto University (KURRI)[12]. The RI for ^{237}Np(n,ɣ)^{238}Np reaction was determined from the differential neutron capture cross-sections in the neutron energies from 0.005 eV to 10 keV. The RI was measured by using the detector assembled with C$_6$D$_6$ scintillators at KURRI.

II. Measurement

We measured the neutron capture cross-sections for various samples by using the neutron TOF method for a pulsed neutron source based on a 46-MeV electron. The capture cross-section measurements are described elsewhere[12 - 14]. The previous energy-dependent neutron capture cross section measurement was carried out using a neutron TOF method with the total-energy absorption

detector assembled with BGO scintillators at the 46-MeV electron linac of KURRI. In the present study, we measured it by the C_6D_6 scintillation detectors. The experimental arrangement is almost the same as before [12-14] and is shown in Fig. 1. The parameters of the samples used in the capture cross-section measurement are summarized in Table 2. The [237]Np sample was in the form of neptunium oxide(NpO_2) powder of 1.13g, which was packed in an aluminum disk.
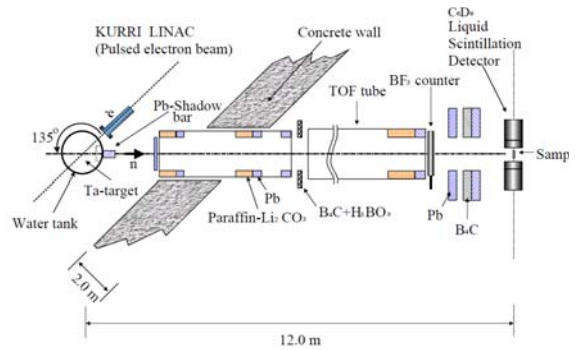

Figure 1. Experimental arrangement

Table 1. Previous experimental RI data and evaluated data

Author	Year	RI(barn)	Facility	Low limit Energy(eV)	Method	Reference
E. Hellstrand et al.	1970	640±50	Graphite-uranium assembly	0.55	Oscillation technique	[3]
K. Kobayashi et al.	1994	652±24	Graphite reactor	0.5	Activation method	[4]
T. Katoh et al.	2003	862±24	KURRI Reactor	0.358	Activation method	[5]
O. Shcherbakov et al.	2005	506±14 807±19	Linac	0.5 0.3	TOF (24.2 m) method	[6]
S. H. Eber le et al.	1971	805	Reactor	0.5	Activation	[7]
L. N. Jurova et al.	1984	730±30 860±40	Reactor	0.5 0.35	Activation	[8]
Yu. E. Titarenko et al.	1999	549±50[*] 576±84[**] 518±47[***]	Heavy Water Reactor	0.5	Activation	[9]
JENDL-4.0	2010	696	Evaluation			[10]
Mughabghab	1981	640±50	Evaluation			[11]

Reference material([*]: Mn, [**]: Cu, [***]: Au)

The sample thickness is important for decreasing the effects of multiple-scattering and self-shielding which depend on the neutron energy, and for obtaining a good signal-to-noise (S/N) ratio. The [10]B sample in the form of powder was packed in a thin aluminum case and was used to measure the energy-dependent neutron flux in the relevant energy region. A graphite sample was also utilized for the experimental investigation of background due to neutrons scattered by the [237]Np sample. Each sample was located at the center of the C_6D_6 scintillation detector assembly. Prompt capture ɣ-rays were detected with the C_6D_6 detection assembly. The C_6D_6 detection assembly consists of two scintillation detectors, each 11 cm in diameter and 5 cm thickness, was used for the measurement of capture ɣ-rays from the sample. The C_6D_6 detection assembly is installed at 12±0.02 m from the neutron source of Ta target. The front of the C_6D_6 detection assembly was covered with [6]LiF tiles, 3 mm in thickness, to absorb neutrons scattered by the sample. However, the tiles were not always enough to shield against higher

energy neutrons above 10 eV. The effect due to scattered neutrons was experimentally investigated by using a graphite sample. A block diagram of the electronics for

the data acquisition system is almost the same as before [12 - 14]. The

Table 2. Physical parameters of the samples used in the experiment

Samples	Physical form	Purity(%)	Size	mass
Np	Powder	99.6	30 mm diameter, 1.4 mm thickness	1.13 g
B	Powder	99.999	1.8 cm x 1.8 cm, 0.5 thickness	3.57 g

KURRI linac was operated with two different modes. One was for the low-energy experiment below 20 eV with a repetition rate of 50 Hz, a pulse width of 3.0 μs, a peak current of 0.5 A, and an electron energy about 30 MeV. The other was for the high-energy experiment above 0.7 eV with a repetition rate of 300 Hz, a pulse width of 0.010 μs, a peak current of 6 A, and an electron energy about 30 MeV. Short experimental runs for 10 to 30 minutes were cyclically made with and without the ^{237}Np sample, which were called the foreground and the background measurement, respectively, and were repeated in order to average out the changes in the measuring conditions, such as the neutron beam intensity, although the linac operation was steady during the experiments. In the case of the ^{10}B sample, a dummy case made of thin aluminum was used for the background measurement. The background corrections were made for the scattered neutrons by considering the graphite thickness equivalent to the scattering cross-section of the capture sample. The BF$_3$ monitor counts were used for the data normalization between the experimental runs. The details of the measuring conditions and method are described elsewhere [12–15].

III. Data Analysis

The capture count rate for the ^{237}Np sample at an energy E, C$_{Np}$(E), can be obtained from the following relation [12–15]:

$$C_{Np}(E) = \varepsilon_{Np} Y_{Np}(E) \phi(E) \quad \cdots\cdots\cdots\cdots\cdots\cdots (1)$$

where the subscript "Np" of ε_{Np} and $Y_{Np}(E)$ means ^{237}Np. Since the C$_6$D$_6$ scintillation detector assembly is not a total-energy absorption detector. Therefore, we couldn't measure neutron capture cross section absolutely in this study. The detection efficiency ε_{Np} may be independent of the neutron energy E, and $\phi(E)$ is the neutron flux as a function of neutron energy E. The capture yield $Y_{Np}(E)$ can be obtained from

$$Y_{Np}(E) = \varepsilon \frac{C_{Np}(E)}{C_B(E)} Y_B(E) \quad \cdots\cdots\cdots\cdots (2)$$

Here, ε is a relative detection efficiency obtained from "ε$_B$/"ε$_{Np}$ and the subscript "B" means ^{10}B. In the case of the ^{10}B sample used in the current measurement, the capture yield $Y_B(E)$ is unity in the relevant energy region. Thus, we can drop the subscript "Np" in the capture yield. The capture yield, Y(E), is the primary value to be determined from the measured number of capture counts in both samples by using Eq. (2). The neutron capture cross-section, $\sigma_c(E)$, is obtained as follows [16]:

$$\sigma_c(E) = f_c(E) \sigma_t(E) \{1 - \exp(-N \sigma_t(E)t)\} Y(E) \quad \cdots (3)$$

where $f_C(E)$ is the correction function due to the neutron scattering and/or the self-shielding in the sample, σt(E) is the neutron total cross-section obtained from the

evaluated data file, N is the atomic density of the sample, and t is the sample thickness. The correction function, $f_C(E)$, was determined by using the MCNP simulation code[17]. The RI is defined by the following relation:

$$I_{Np} = \int_{E_{Cd}}^{\infty} (\sigma(E)/E)dE$$ .... (4)

where (E) is the neutron capture cross-section as a function of the neutron energy E, and E_{Cd} is a Cd cutoff energy, which is usually defined as 0.5 eV [18]. The epi-cadmium dilute RI for a particular reaction in a 1/E spectrum is expressed by using this relationship, where 0.5 eV is determined by using the cadmium cut-off energy, which depends on the cadmium thickness. We assumed that the dE/E neutron spectrum was not perturbed by the absorbing materials. In the present study, the RI for the $^{237}Np(n,\gamma)^{238}Np$ reaction was obtained from an integration of differential data according to the definition of the RI.

IV. Results and Discussion

The neutron capture cross-sections and the RI(804.7 barns) of a $^{237}Np(n,\gamma)^{238}Np$ reaction were measured in the neutron energy region from 0.005 eV and 10 keV by using the 46-MeV electron linear accelerator at KURRI. ^{237}Np isotopes has a very large s-wave resonance at 0.491 eV. Therefore, the measurement of the RI is very difficult because Cd cutoff energy is close to the resonance energy. The neutron capture cross section of $^{237}Np(n,\gamma)^{238}Np$ reaction was shown in Fig. 2 [15]. The integral values of present results depends on the energy were shown in Fig. 3. The present measurement of the neutron capture cross-sections was compared with the previous experimental data and the evaluated data of JENDL-4.0 [10], Mughabghab [11] as shown in Fig. 4. In the case of ^{237}Np sample, it has a very large s-wave resonance at 0.491. Therefore, the discrepancy of the RI value as shown in Fig 3. is more large than the other elements, especially. The present result are seem to be close to the

result of S. H. Eber le et al and more large than the other values. In the calculation of RI with differential neutron capture cross section, we have to choose the data near the 0.5 eV, very carefully because the data value of RI has comparatively large than more higher data in the keV region.
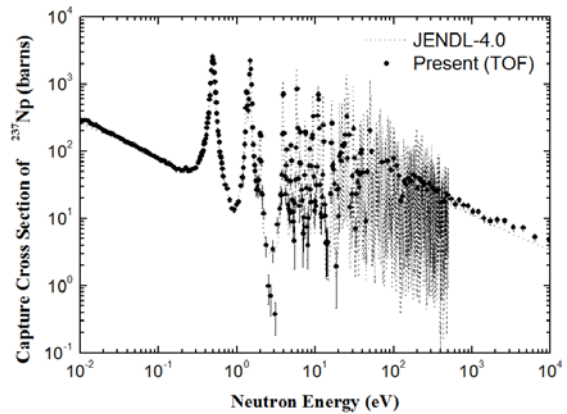

Figure 2. Neutron Capture Cross section for the $^{237}Np(n,\gamma)^{238}Np$ reaction
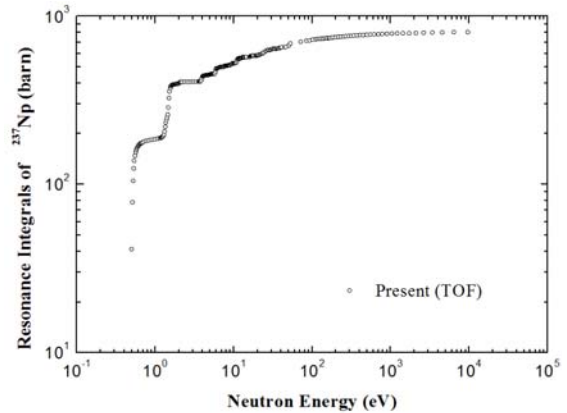

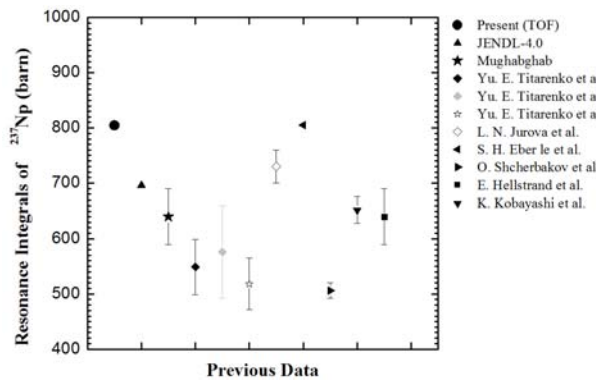Figure 3. Integrated RI value of present result for the $^{237}Np(n,\gamma)^{238}Np$ reaction

Figure 4. Previous and present RI values for the
^{237}Np$(n,\gamma)^{238}$Np reaction

Acknowledgments

References

[1] Mukaiyama, T., et al.: Conceptual study of actinide burner reactors, Proc. of 1988 Int Reactor Phys. Conf., Jackson Hale, Vol. IV, p.369, 1988.

[2] J. Tommasi et al., ibid., ANS, La Grange Park, Illinois, p.1252, 1993.

[3] E. Hellstrand, J. Phelps, C. Sastre, Rep., BNL-50242, (T-577), 1970.

[4] K. Kobayashi, A. Yamanaka, I. Kimura, Journal of Nuclear Science and Technology, Vol. 31, p.1239, 1994.

[5] T. Katoh, S. Nakmura, K. Furutaka, H. Harada, K. Jujiwara, T. Fujii, H. Yamana, Journal of Nuclear Science and Technology, Vol. 40, p.559, 2003.

[6] O. Shcherbakov, K. Furutaka, S. Nakamura, H. Sakane, H. Harada, K. Kobayashi, S. Yamamoto, Jun-ichi Hori, Journal of Nuclear Science and Technology, Vol. 42, p.135, 2005.

[7] S. H. Eber le, W. Robel, W. Jung, I. Bayat, H. J. Bley l, E. Bojarsky, L. Schmit, E. Gantner, J. Reinhardt, C. Krueckeberg, Rep., KFK-1456, p.45, 1971.

[8] L. N. Jurova, A. A. Poljakov, V. P. Rukhlo, Ju. E. Titarenko,

S. F. Komin, Ju. V. Stogov, O. V. Shvedov, B. F. Mjasoedov, A. V. Davydov, S. S. Travnikov, Journal of YK, Vol. 1/55, p.3, 1984.

[9] Yu. E. Titarenko, O. V. Shvedov, M. M. Lgumnov, V. F. Batayev, V. I. Volk, A. Yu. Vakhrushin, S. V. Shepelkov, A. V. Lopatkin, S. G. Mashnik, T. A. Gabriel, Journal of Nuclear Science and Technology, Vol. 131, p.96, 1999.

[10] S. Shibata, T. Narita (Eds.), "Descriptive Data of JENDL-3.2", JAERI-Data/ Code 98-006, part II, 1998.

[11] 11) S. F. Mughabghab, M. Divadeenam and N. E. Holden, Neutron Cross Sections, Neutron Resonance Parameters and Thermal Cross Sections, Vol. 1, part A: Z=1-60(Academic Press, New York, 1981.

[12] S. Yamamoto et al., Journal of Nuclear Science and Technology, Vol 126, p.201, 1997.

[13] K. Kobayashi et al., J. Nucl. Sci. Technol. Vol. 36, p.20 1999.

[14] 14) K. Kobayashi et al., Annu. Rep. Res. Reactor Inst., Kyoto Univ., Vol. 22, p.142, 1987.

[15] 15) K. Kobayashi, Samyol Lee, Shuji Yamamoto, Hyun Je Cho and Yoshiaki Fujita, Journal of Nuclear Science and Technology, Vol, 39, No. 2, p.111, 2002.

[16] H. W. Schmitt, ORNL-2883, 1-28-60, 1960.

[17] J. F. Briesmeister (Ed.), "MCNP－A General Monte Carlo Code for Neutron and Photon Transport, Version 4A", Los Alamos National Laboratory, LA-12625-M, 1997.

[18] K. Shibata, et al., Journal of Nuclear Science and Technology, Vol. 39. p.1125. 2002.