Neutron Capture Cross Section Measurement of Np-237 below 10 keV by Linac Time-of-Flight Method

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The neutron capture cross section of ^{237}Np has been measured in the energy region from 0.01 eV to 10 keV by using the neutron time-of-flight (TOF) method with a 46 MeV electron linear accelerator (linac) at the Research Reactor Institute, Kyoto University (KURRI). A pair of C_6D_6 scintillation detectors, which was placed at a distance of 12.0±0.02 m from the pulsed neutron source, was employed for the prompt capture gamma-ray measurement from the ^{237}Np sample. The measured result has been normalized to the reference value of the ^{237}Np(n,γ)^{238}Np reaction in ENDF/B-VI at 0.0253 eV.

The existing experimental and the evaluated capture cross sections in ENDF/B-VI and JENDL-3.2 have been compared with the present measurement. For the neutron capture cross section of ^{237}Np, the data by Weston et al. and the evaluated data are in good agreement with the present measurement. However, the data by Hoffman et al. are obviously lower in the relevant energy region.

The data, which were measured before using a lead slowing-down spectrometer at KURRI, have been in good agreement with the data obtained by energy-broadening the present TOF measurement.

1. Introduction

The ^{237}Np, which is one of the minor actinides with a long half-life, is abundantly produced in light water reactors. The nuclear data are of great importance for investigating the generation and the burn-up characteristics of ^{237}Np in the reactor. Neutron capture by ^{237}Np produces an intense alpha-emitter of ^{238}Pu through the beta-decay of ^{238}Np. In order to decrease an undesirable inheritance or a risk of these high level radioactive materials, in recent years, a great interest has been taken in the nuclear transmutation using conventional or advanced reactors and accelerator-driven subcritical reactors [1, 2, 3]. Accurate determinations of the fission and the capture cross sections for ^{237}Np are indispensable to research and development of the nuclear transmutation technology.

Although several measurements for the capture cross section of ^{237}Np have been reported at higher energies and thermal energy, the cross section has rarely been measured in the low/resonance energy region [4]. Weston et al. measured the capture cross section between about 0.01 eV and 0.2 MeV by the neutron

TOF method using an Oak Ridge Electron Linear Accelerator [5]. Hoffman et al. measured the neutron capture cross section of ^{237}Np by the neutron time-of-flight (TOF) method using a Moxon-Rae detector [6].

In the present work, the neutron capture cross section of ^{237}Np has been measured in the energy region from 0.01 eV to 10 keV by using the neutron TOF method with a 46 MeV electron linear accelerator (linac) of the Research Reactor Institute, Kyoto University (KURRI). The present result is compared with the previous experimental[7] and the evaluated data in ENDF/B-VI [8] and JENDL-3.2 [9].

The capture cross section was measured before using the lead slowing-down spectrometer (KULS) at KURRI. The result obtained by the KULS is compared with the present TOF measurement, which is energy-broadened by the resolution function of the KULS.

2. Experimental Procedure

2.1. Capture Samples

Neptunium oxide (NpO_2) powder of 1.13 g was purchased from Amersham, which was packed in an aluminum disk container of 20 mm in inner-diameter and 1.4 mm in thickness (outer-diameter: 30 mm and thickness: 2.2mm). The purity of the sample is 99.6% by weight and the major impurities are about 4μg in total weight of Ga, K, P, Rb, and S. The gamma-rays of 86.5 keV from ^{237}Np and 300, 312, 341 keV from ^{233}Pa which was produced through the α-decay of ^{237}Np, were measured with a high-purity germanium detector (HPGe). No peak from the impurities except for ^{233}Pa was found in the pulse height distribution data.

The enriched ^{10}B powder of 90.4% was put into a thin Al (0.2 mm in thickness) plug of 1.8×1.8 cm^2 and 8mm in thickness, and the sample thickness was 1.102 g/cm^2.

2.2. Experimental Arrangement

The neutron capture measurement was performed in the neutron energy region of 0.01 eV to 10 keV using the 46 MeV electron linac at KURRI. The experimental arrangement is shown in **Fig. 1**. Pulsed fast neutrons were produced from a water-cooled Ta target. The flight path, which is 12.0 ± 0.02 m from the neutron source to the sample, is located at an angle of 135° with respect to the linac electron beam direction. The target is consists of twelve sheets of Ta plates of 5 cm in diameter and 2.9 cm long in effective thickness [10]. The target was set at the center of the cylindrical water tank, which was 30 cm in diameter and 1 cm in tank wall thickness, to moderate fast neutrons. A shadow bar made of Pb block (size: 5×5 cm^3 $\times 10$ long) was placed in the neutron flight path in front of the Ta target to reduce the γ-flash generated by the electron burst in the target.

The linac was operated with a pulse width of 3μs, a repetition rate of 30 Hz, electron peak current of 400 *mA* and the electron energy of 22 MeV for the measurement in the lower energy region, and with a pulse width of 33 ns, a repetition rate of 100 Hz, electron peak current of 3 μA and the electron energy of 20 MeV in the higher energy region, respectively.

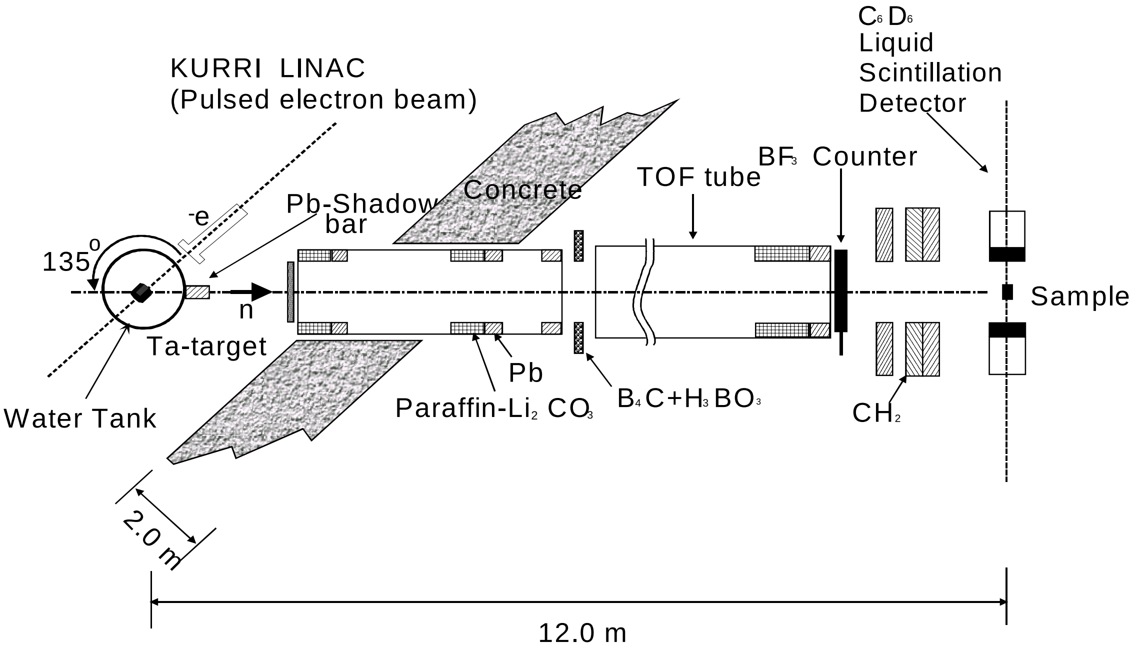

Fig. 1. Experimental arrangement for the capture cross section measurement.

2.3. Detectors and Data Taking

A pair of C_6D_6 liquid scintillation detectors was used for detection of γ-rays from the sample. The ^{237}Np sample was inserted into the neutron TOF beam between the detectors, each 11cm in diameter and 5cm thick. The detectors are adequate to the capture γ-rays measurement because of less sensitive to scattered neutrons. For the measurement of the incident neutron flux/spectrum on the sample, a ^{10}B plug (1.102 g/cm^2) was inserted into the TOF beam instead of the ^{237}Np sample. A background run was carried out with an aluminum disk container without the ^{237}Np sample. In the background measurement, a thick ^{10}B plug (4.54 g/cm^2) was placed in front of the collimator before the C_6D_6 detectors to black-out the neutron beam. The background level was also confirmed by those measured with a 0.5 mm thick Cd sheet and notch-filters of Ag, Co and Mn. The neutron beam intensity during the experiment was monitored with a BF_3 counter, as shown in **Fig. 1**. In order to monitor the neutron intensities during the experimental runs, the BF_3 counter was placed in the neutron beam. Through the amplifiers and the discriminators, signals from a pair of C_6D_6 liquid scintillation detectors or the BF_3 counter for the neutron flux/spectrum monitor were fed into the time digitizer, which was initiated by the linac electron burst. The multi-channel data were stored as two sets of 4096-channel data with a channel width of 4μs.

3. Data Analysis

3.1. Neutron Capture Cross Section

The relative neutron capture yield of the ^{237}Np(n,γ) reaction is given by the following relation:

$$Y_{Np}(E) = \frac{C_{Np}(E)}{C_B(E)} Y_B(E), \tag{1}$$

$$Y_{Np}(E) = (1 - \exp(-N\sigma_t(E)t))\frac{\sigma_c(E)}{\sigma_t(E)} f_c, \qquad (2)$$

where $C_{Np}(E)$ is capture counts of ^{237}Np at energy E, $C_B(E)$ is capture counts of ^{10}B at energy E, N is atomic density of the ^{237}Np sample, f_c is the correction factor for the neutron scattering in the sample, $\sigma_c(E)$ is the neutron capture cross section, $\sigma_t(E)$ is the total cross section, t is thickness of the ^{237}Np sample, and $Y_B(E)$ is the capture yield of ^{10}B. Since the cross section of the ^{10}B(n,α) reaction is a well-known reference one, it has been used to determine the neutron flux/spectrum in the present measurement. When t is thin enough, Eq. (2) can be written as follows:

$$\sigma_c(E) = \frac{Y_{Np}(E)}{Ntf_c} \qquad (3)$$

3.2. Self-shielding Correction

The self-shielding effect of neutrons has to be taken into account in the capture cross section measurements, especially near the large resonance region. We have assumed that the sample is irradiated by the neutron TOF beam. The self-shielding correction in the ^{237}Np sample has been calculated by the Monte Carlo code MCNP [11]. The correction factor has been obtained from the ratio of the effective capture cross section for the ^{237}Np sample to that for the infinite diluted one that is obtained by multiplying the atomic density by 10^{-6}. The result has been applied to the neutron scattering and self-shielding correction for the present cross section measurement of the ^{237}Np(n, γ)^{238}Np reaction.

4. Results and Discussion

The neutron capture cross sections have been measured relative to that of the ^{10}B(n,α) reaction at the energy region from 0.01 eV and 10 keV. The result obtained has been normalized to the reference value of the thermal neutron cross section of 181 *b* of ENDF/B-VI at 0.0253 eV. The experimental uncertainties are in the range of 0.05% to 44%, and the major uncertainties are due to the statistical error and that in the reference cross section for the ^{10}B(n,α) reaction. Since the ^{237}Np sample was almost free from impurities, no correction was made for the impurity effect.

The capture cross sections measured by Weston et al. [5] are in good agreement with the present measurement as seen in **Fig. 2**, but the data measured by Hoffman et at.[6] are remarkably lower than the present values [6]. The evaluated data in ENDF/B-VI [8] and JENDL-3.2 [9] are also in good agreement with the present measurement in the relevant energy region, as shown in **Fig. 3**.

We measured the capture cross section before using the lead slowing-down spectrometer (KULS) at KURRI. The measured data are in good agreement with the data that the present TOF measurement is broadened by the resolution function of the spectrometer, as shown in **Fig.4**.

5. Conclusion

The neutron capture cross section of ^{237}Np has been measured in the energy region from 0.01 eV and

10 keV by using the neutron TOF method and the C_6D_6 liquid scintillation detectors. The data by Weston et al. and the evaluated data in ENDF/B-VI and JENDL-3.2 are in good agreement with the present measurement in the relevant energy region. However, the data by Hoffman et al. are lower obviously. The data measured with the lead slowing-down spectrometer have been in good agreement with the data which were obtained by energy-broadening the present TOF measurement.

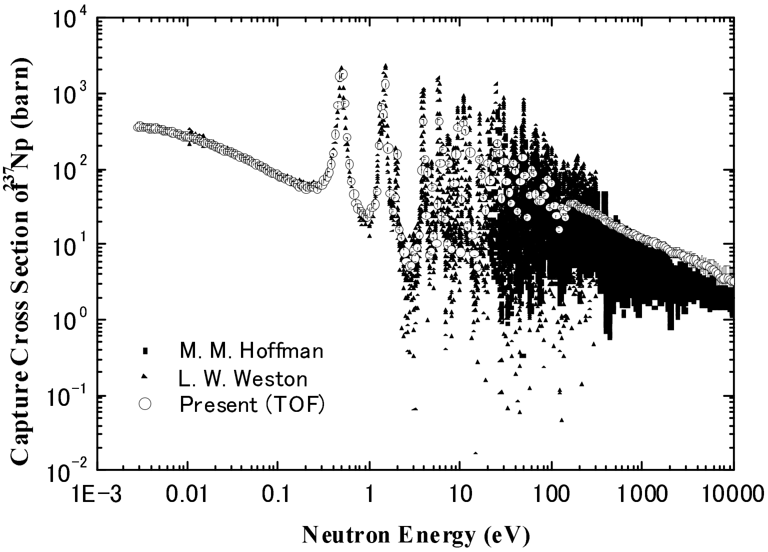

Fig. 2. Comparison of the experimental capture cross sections of ^{237}Np and the present measurement obtained by the linac TOF method.

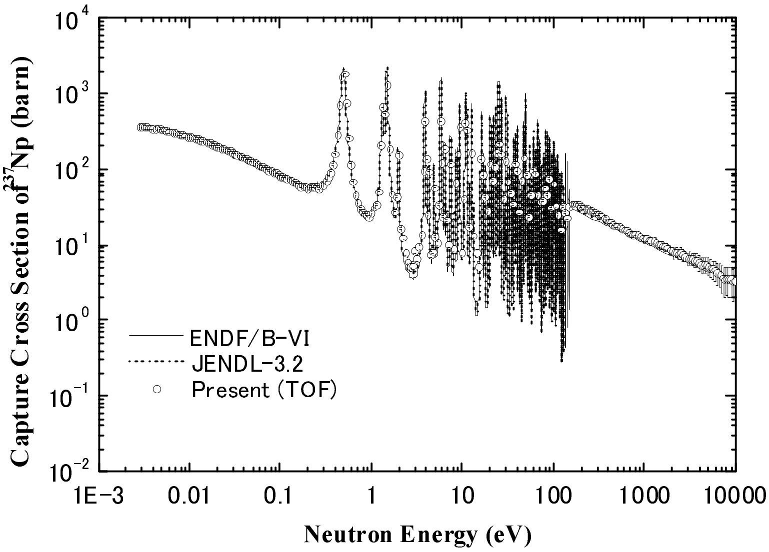

Fig. 3. Comparison of the evaluated capture cross sections of ^{237}Np and the present measurement obtained by the linac TOF method.

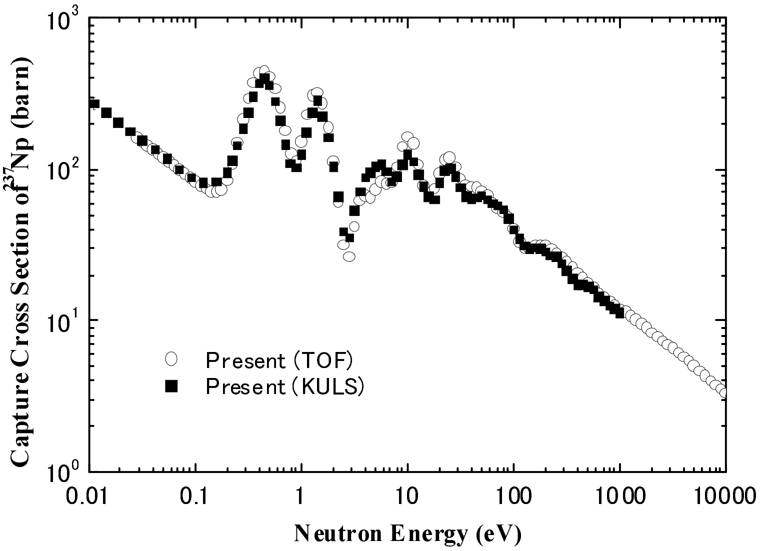

Fig. 4. Comparison of the measurement obtained with the KULS and the linac TOF data broadened by the KULS resolution.

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