Capture Cross Section Measurement of Np-237 below 1 keV with Lead Slowing-down Spectrometer

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Making use of the Kyoto University Lead slowing-down Spectrometer (KULS) driven by a 46 MeV electron linear accelerator (linac) at the Research Reactor Institute, Kyoto University (KURRI), the relative cross section for the ^{237}Np(n,γ) reaction has been measured from 0.01 eV to 1 keV with energy resolution of about 40 % (FWHM).   The neutron flux/spectrum has been measured by a BF$_3$ counter.   The cross section of the ^{10}B(n,α) reaction in ENDF/B-VI was used as a reference one for the cross section measurement.   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, and the measurement has been compared with the experimental and the evaluated data in ENDF/B-VI and JENDL-3.2, whose data were broadened by the energy resolution of the KULS.

1. Introduction

The neptunium (Np)-237, which is abundantly produced in light water reactors, is one of the minor actinides with a long half-life.   In order to make nuclear power more acceptable and practical, much interest has been paid to the disposal of radioactive waste matter [1-3].   One of the waste management methods for ^{237}Np is to adopt the nuclear transmutation using reactor neutrons. The Np-237 has a large capture cross section in the low/resonance energy region.   It has also a relatively large fission cross section in the MeV energy region.   Accurate determination of these cross sections is very important for the evaluation of the transmutation of ^{237}Np which is produced in large quantities in power reactors.

Although several measurements of the ^{237}Np(n,f) cross section have been reported at higher energies, the capture cross section has rarely been measured in the lower/resonance energy region. Hoffman et al. measured the neutron capture cross section of Np-237 by the neutron time-of-flight (TOF) method and Moxon-Rae detector[4].   Weston et al. measured the capture cross section between about 10^{-2} eV and 0.2 MeV by the neutron TOF method using an Oak Ridge Electron Linear Accelerator [5].   The evaluated fission cross section data are appeared in ENDF/B-VI [6] and JENDL-3.2 [7].

In the present study, we measure the neutron capture cross section of Np-237 in the range of

0.01 eV to 1 keV relative to that of the ^{10}B(n, α) reaction by using the lead slowing-down spectrometer [8] coupled to the 46 MeV electron linear accelerator (linac) of the Research Reactor Institute, Kyoto University (KURRI). Below 1 keV, the relative measurement has been carried out with an Ar-gas counter, and the neutron flux/spectrum has been measured with a BF$_3$ counter. The relative cross section obtained has been normalized to the reference value of the thermal cross section at 0.0253 eV. The present result is compared with the existing experimental and evaluated data in ENDF/B-VI and JENDL-3.2.

2. Experimental Methods

2.1. Lead Slowing-down Spectrometer

A lead slowing-down spectrometer has been installed in coupling to the 46 MeV linac at KURRI. This Kyoto University Lead Slowing-down Spectrometer (KULS)[8] is composed of 1600 lead blocks (each size : 10 x 10 x 20 cm^3, purity : 99.9 %) and the blocks are piled up to make a cube of 1.5 x 1.5 x 1.5 m^3 (about 40 tons in weight) without any structural materials. The KULS is covered with Cd sheets of 0.5 mm in thickness to shield it against low energy neutrons scattered from the surroundings. At the center of the KULS, an air-cooled photoneutron target of Ta is set to generate pulsed fast neutrons. One of the experimental holes in the KULS is covered by Bi layers of 10 to 15 cm in thickness to shield from high energy capture γ-rays (6 to 7 MeV) produced by the Pb(n, γ) reaction in the spectrometer.

Characteristics of behavior of neutrons in the KULS have been studied by experiments using the resonance filter method[8]. The slowing-down constant K in the relation of E=K/t^2 was determined to be 190± 2 (keV μs^2) for the Bi hole in the KULS by the least squares method using the measured relation between the neutron slowing-down time t in μ s and the average neutron energy E in keV. The energy resolution for the experimental holes was also deduced from the measured data to be about 40 % at energies between a few electron-volts and about 500 eV and was worse than that below a few electron-volts and above about 500 eV [8]. The relation between the neutron slowing-down time and the energy, and its energy resolution were also verified by Monte Carlo calculations [8].

2.2. The Np-237 sample

Neptunium oxide powder of 1.13 gram was purchased from Amersham, which was packed in an aluminum disk container of 20 mm in diameter and 1.4 mm in thickness. The purity of the sample is 99.6 % by weight and the major impurities are about 4 μg in total weight of Ga, K, P, Rb, and S. The gamma-rays of 86.5 keV to 300, 312, 341 keV from Pa-233, which was produced through the α-decay of Np-237, were measured with a high-purity germanium detector (HPGe). The pulse height distribution of the Np-237 is shown in **Fig. 1.** No peak from the impurities is found in the figure.

2.3. Ar-gas counter

The Ar-gas counter, which is filled with a mixed gas of 97 % Ar and 3 % CO_2 at the pressure of 1 atm., was of a cylindrical type, 12.7 mm in diameter, 6.3 cm in effective length, and high-voltage bias was 1400 V.   The Np-237 sample was put together by the side of the counter as seen in **Fig. 2** and inserted into the Bi hole of the KULS.

A BF_3 counter, 12.7 mm in diameter and 5.8 cm in effective length was applied to measure the neutron flux / spectrum at the Bi hole of the KULS.   In order to monitor the neutron intensities between the experimental runs, another BF_3 counter was placed in an experimental hole of the KULS.
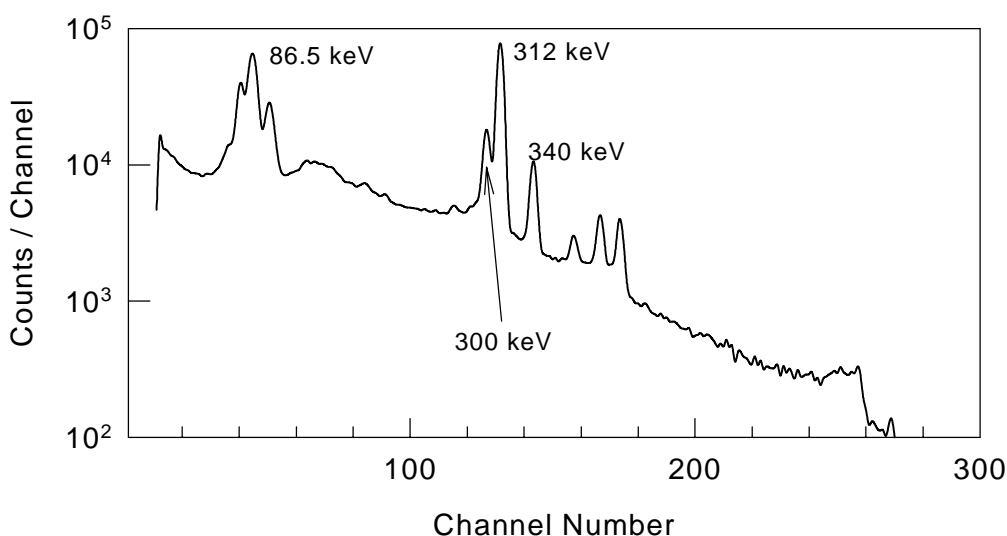

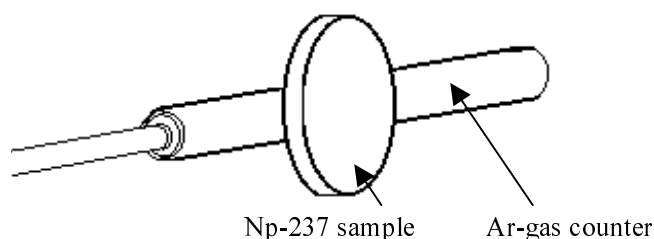Fig. 1. The pulse height spectrum of the Np-237 sample measured with a HPGe detector.

Np-237 sample   Ar-gas counter

Fig. 2. The Ar-gas counter and the Np-237 sample.

3. Measurement and Analysis

3.1. Capture Cross Section Measurement

The relative cross section of the ^{237}Np(n,) reaction is given by the following relation :

$$\sigma_{Np}(E) \quad = \quad \frac{C_{Np}(E)}{C_B(E)}\sigma_B(E) \, , \tag{1}$$

where $C_{Np}(E)$ is capture counts of Np-237 at energy E, $C_B(E)$ is counts from the BF_3 counter at energy E, and $\sigma_B(E)$ is the energy-dependent reference cross section of the $^{10}B(n,\alpha)$ reaction.   The cross section of the $^{10}B(n,\alpha)$ reaction is a well-known reference one and has been used to determine the neutron flux in the current measurement.   The cross section values of $\sigma_B(E)$, whose data were broadened by the resolution function of the KULS, were taken from ENDF/B-VI[6].   The typical operating conditions that the KULS was driven were as follows ; the pulse repetition rate was 200 Hz, the pulse width 6.8 ns, the electron peak current 600 mA, and the electron energy 30 MeV.   A background run was carried out without the NpO_2 powder using the same size of empty aluminum disk container instead of the Np-237 sample.

3.2. Electronics and Data Taking

        Through the amplifiers and the discriminators, signals from the Ar-gas counter 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.   Two sets of 4096 channels with a channel width of 0.5 μ s were allotted to the slowing-down time measurements for the Ar-gas counter or the BF_3 counter and the BF_3 counter for the neutron intensity monitor between the experimental runs.   Pulse height distributions for these counters were also measured together with the slowing-down time measurements.
        For the relative measurement to the $^{10}B(n,)$ cross section, output signals from the BF_3 counter were also fed to the time digitizer through the amplifiers and the discriminators, and were stored in almost the same way as for the measurement with the Ar-gas counter.

3.3. Self-shielding Correction

        As mentioned above, the NpO_2 powder of 1.13 gram was encapsulated in the aluminum disk container of 20 mm in diameter and 1.4 mm in thickness.   Then, the self-shielding effects 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 isotopically irradiated from the surrounding in the Bi hole of the KULS.   The self-shielding correction in the Np-237 sample has been calculated by the MCNP Monte Carlo Code using the following relation.

$$\left\langle \sigma_c(E) \right\rangle = \frac{\int_r \int_E N\sigma_c(E)\varphi(r,E)drdE}{\int_r \int_E N\varphi(r,E)drdE} \tag{2}$$

where, N is atomic density of the Np-237 sample, $\sigma_c(E)$ is capture cross section, ψ (r,E) is neutron energy spectrum at the position r.   The correction function has been obtained from the ratio of the effective capture cross section for the Np-237 sample and that for the infinite diluted sample (the atomic density is multiplied by 10^{-6}).

The correction function broadened by the resolution function of the KULS is illustrated in **Fig. 3**. The broadened function has been applied to the self-shielding correction for the present measurement of the ^{237}Np(n,γ)^{238}Np reaction cross section with the KULS.
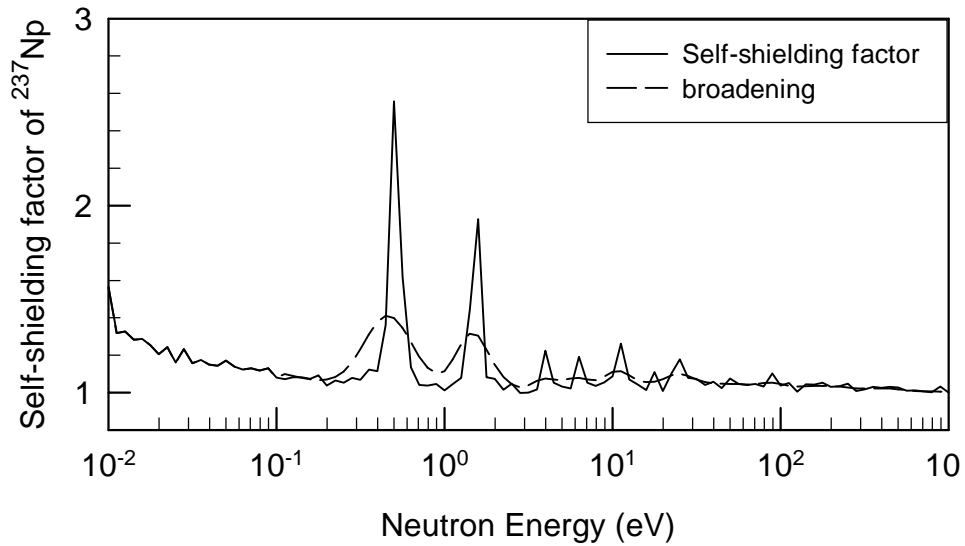

Fig. 3.   The self-shielding factor of ^{237}Np. The solid-line is self-shielding factor of ^{237}Np and dot-line is broadened by the resolution function of the KULS.
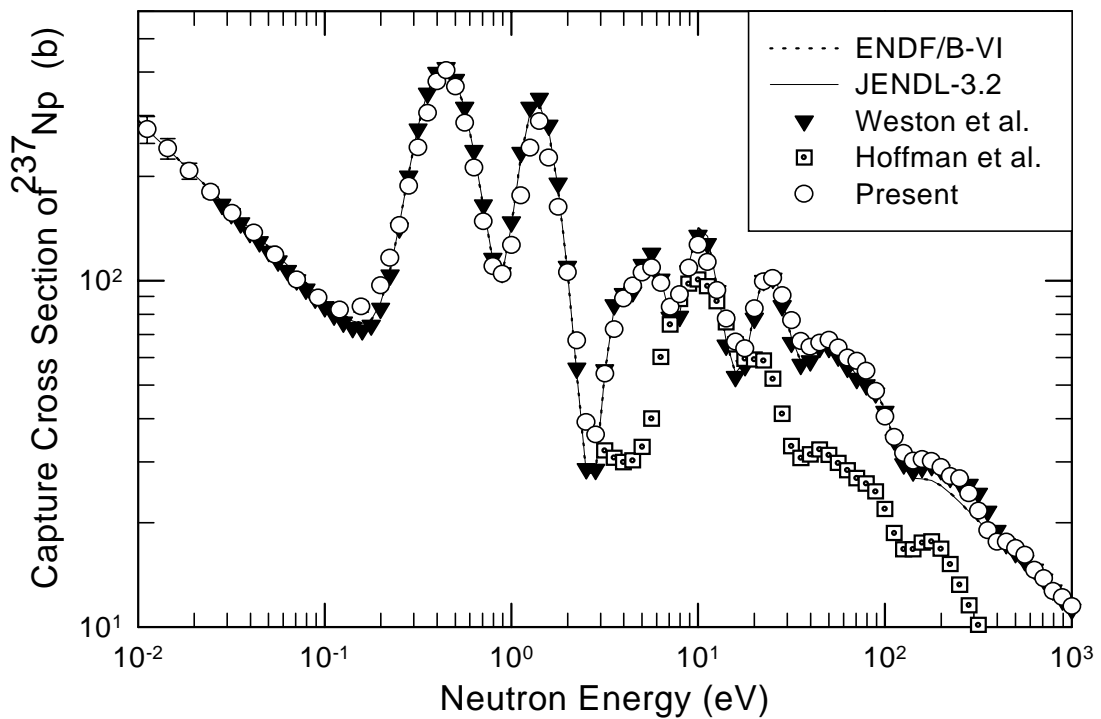

Fig. 4.   Comparison of the measured and the evaluated data for the ^{237}Np(n,γ)^{238}Np reaction, whose values are broadened by the resolution function of the KULS.

4. Results and Discussion

Making use of the KULS, the cross section of the ^{237}Np(n,) reaction has been measured relative to that of the ^{10}B(n,α) reaction at energies below 1 keV. The cross section data were obtained by summing up the slowing-down time data in intervals of about 0.12 lethargy width. The result obtained has been normalized to the reference value of the thermal neutron cross section in ENDF/B-VI at 0.0253 eV. The experimental uncertainties are in the range from 2 % to 10 %, and the major uncertainties are due to the statistical error (2~10 %) and that in the reference cross section (~2 %). Since the Np-237 sample was almost free from impurities, no correction was made for the impurity effect.

The capture cross sections measured by Weston et al. are in good agreement with the present measurement as seen in **Fig. 4**, but the data measured by Hoffman et al. are remarkably lower than the present values. In **Fig. 4**, the experimental and the evaluated data are broadened by the resolution function of the KULS. The evaluated data in ENDF/B-VI and JENDL-3.2 are close to the present measurement in the relevant energy region.

5. Conclusion

The cross section of the ^{237}Np(n,) reaction has been measured from 0.01 eV to 1 keV relative to that of the ^{10}B(n,α) reaction, making use of the lead slowing-down spectrometer KULS at KURRI. 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.

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