Measurements of Neutron Capture Cross Sections for $^{237, 238}Np$

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Recent progress on the measurement of neutron capture cross sections performed by a JNC and Kyoto university group is reviewed focusing on ^{237}Np and ^{238}Np: ① on the measurement of the neutron capture cross section of ^{237}Np for thermal neutrons, the activation method was utilized where a thin cadmium shield was introduced to minimize the errors due to a low energy resonance; ② the effective capture cross section of ^{238}Np for thermal neutrons was measured by utilizing a double-neutron capture reaction; and ③ a total-energy detector and a flash-ADC based data-taking system have been developed and used to measure the capture cross section of ^{237}Np by a TOF method.

1. The neutron capture cross section and resonance integrals of ^{237}Np for thermal neutrons

The thermal neutron capture cross section (σ_0) and the resonance integral (I_0) of ^{237}Np have been measured by an activation method to supply basic data for the study of transmutation of nuclear waste. The neutron irradiation of ^{237}Np samples has been done at the Research Reactor Institute, Kyoto University (KUR). Samples of ^{237}Np were irradiated between two Cd sheets or without a Cd sheet. Since ^{237}Np has a strong resonance at the energy of 0.49 eV, the Cd cut-off energy was adjusted as low as 0.36 eV(thickness of the Cd sheets: 0.125 mm). A high purity Ge detector was employed for the activity measurement of the irradiated sample. The reaction rate to produce ^{238}Np from ^{237}Np was analyzed by the Westcott's convention. The details on the experiments and analyses were described in ref. [1] Results obtained were 141.7±5.4 barns for σ_0 and 862±51 barns for I_0 above 0.358 eV of ^{273}Np. **Table 1** shows the present result and previous one on the σ_0 for ^{237}Np.

Table 1 Comparison of σ_0 for ^{237}Np

Authors (year)	σ_0 (b)	Measurement method
Brown et al. (1956)	172 ± 7	α-ray spectroscopic method
Smith et al. (1957)	170 ± 22	Total cross section
Tattersall et al. (1956)	169 ± 3	Pile oscillation method
Schuman et al. (1969)	185 ± 12	α-ray spectroscopic method
Kobayashi et al. (1994)	158 ± 3	γ-ray spectroscopic method
JNC-Kyoto (2003)	141.7 ± 5.4	γ-ray spectroscopic method
JENDL-3.3 (2002)	161.7	Evaluation

The measurements of the σ_0 by γ-ray spectroscopic method give smaller values than those measured by other methods. The γ-ray spectroscopic method requires absolute γ-ray intensity of ^{238}Np. Recent evaluated nuclear structure data file [2] gave about 10% smaller value for the absolute γ-ray intensity of ^{238}Np; if this value is used for the reduction of the σ_0, it is increased by 10 %. To understand the discrepancy on the σ_0 of ^{237}Np, the absolute γ-ray intensity of ^{238}Np should be re-investigated in detail.

2. The effective capture cross section of ^{238}Np for thermal neutrons

The effective capture cross section ($\hat{\sigma}$) of ^{238}Np for thermal neutrons has been measured as data for the burn-up analysis of irradiated fuels and for the study of nuclear transmutation of minor actinides. A sample of ^{237}Np was irradiated for 10 hours at KUR. Neptunium-238 and –239 were produced simultaneously via the double neutron capture reaction ^{237}Np(n, γ)^{238}Np(n, γ)^{239}Np as are shown in **Fig. 1**.

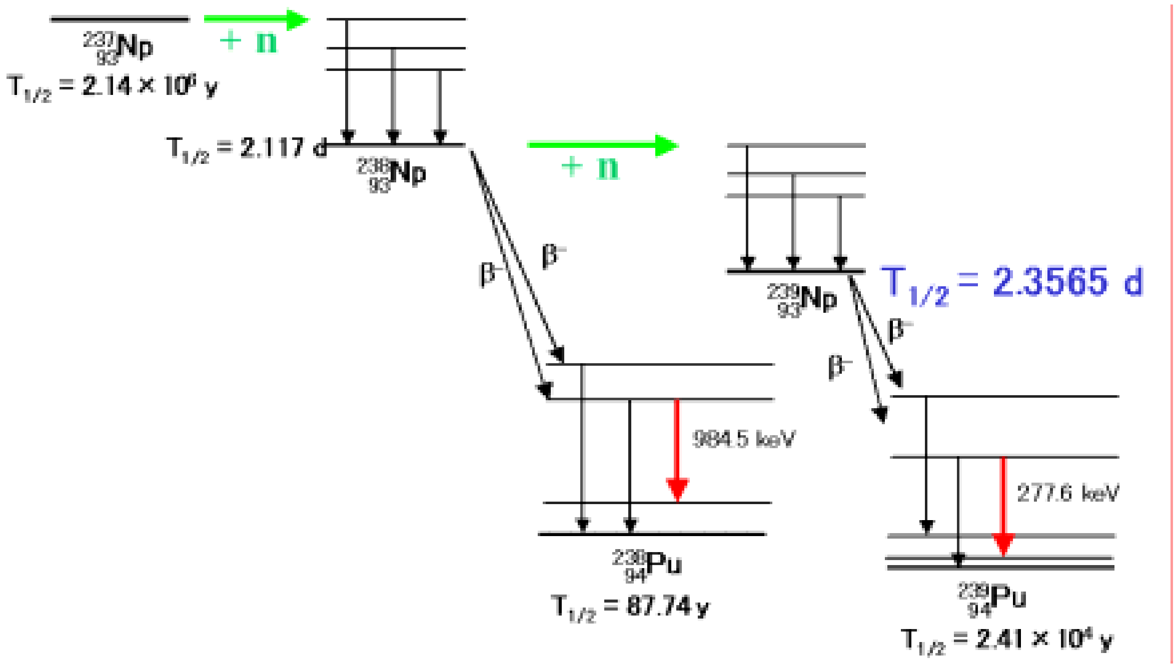

Fig. 1 Reaction scheme of the double neutron capture reaction ^{237}Np(n, γ)^{238}Np(n, γ)^{239}Np

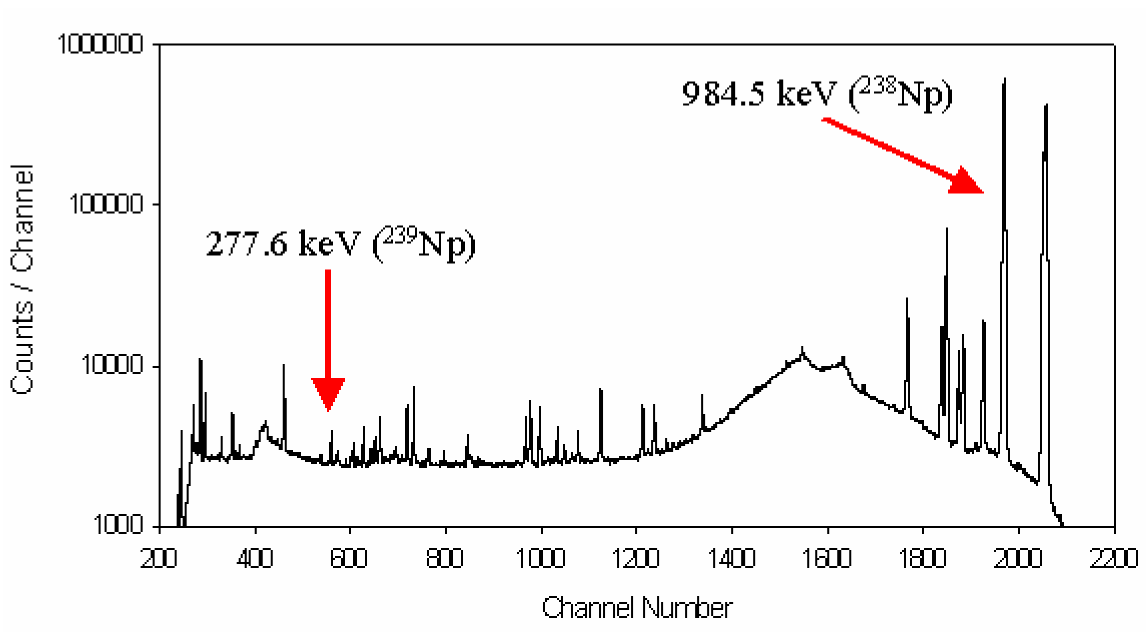

Fig. 2 Gamma-ray spectrum from the ^{237}Np sample irradiated during a period of 10 hours

The neutron flux at the irradiation position was monitored using Au and Co wires. The epithermal index $r\sqrt{T/T_0}$ in Westcott's convention was measured as 0.03. A high-purity Ge detector with a BGO ($Bi_4Ge_3O_{12}$) anti-coincidence shield was used to measure the activities of ^{238}Np and ^{239}Np. **Figure 2** shows an example of the measured γ-ray spectrum. Both of the 277.6-keV γ-ray from the decay of ^{239}Np and the 984.5-keV γ-ray from the decay of ^{238}Np are clearly shown. The $\hat{\sigma}$ of ^{238}Np was deduced from the ratio of these activities.

If ^{238}U is included in the ^{237}Np sample, ^{239}Np is produced via the ^{238}U(n, γ)^{239}U reaction and β decay process. It is difficult to measure ^{238}U in the ^{237}Np sample by measuring the radioactivities since the lifetime of ^{238}U is much longer than that of ^{237}Np. Therefore, the activation method using a neumatic tube of KUR has been utilyzed; the Np sample of about 1 kBq was irradiated for 3 min in the pneumatic transfer tube of KUR and the gamma-rays from the irradiated sample were measured by a HP-Ge detector. The ratio between the number of ^{238}U and that of ^{237}Np nuclei was determined to be less than 5.1×10^{-5}. The contamination of ^{238}U contributes the neutron-capture cross section of ^{238}Np by less than 0.4 %, and is negligible. The result obtained is 479 ± 24 b for the $\hat{\sigma}$ of ^{238}Np [3]. The result is compared with data in the evaluated nuclear data libraries, ENDL-86 [4], ENDF/B-VI [5], and JENDL-3.3 [6]. To explain the present result, the evaluated capture cross sections of ^{238}Np have to be increased about 5 times in the case of ENDL-86 and about 2.4 times in the case of ENDF/B-VI. The value in JENDL-3.3 agrees well with the present result. However, the reduced resonance integral of all libraries are equal to zero. Further fine evaluations should be required on the thermal neutron capture cross section and resonance integral of ^{238}Np.

3. The measurement of neutron capture cross section of ^{237}Np by a TOF method

Fig. 3 The 16-section BGO scintillation detector (back-view in left and side-view in right)

The neutron capture cross section of ^{237}Np has been measured relative to the ^{10}B(n,α)^{7}Li* cross section by the time-of-flight method in the energy range from 0.02 eV to 100 eV. The 46 MeV electron linear accelerator at the Research Reactor Institute, Kyoto University, was used as a pulsed neutron source. The 16-section BGO scintillation detector shown in **Fig. 3** having a total volume of 8.54 l and the associated 40 MHz flash-ADC–based data taking system have been developed for the measurement of the capture cross section. The BGO detector can be used as a total energy gamma-ray detector [7].

Figure 4 shows typical waveforms for γ-rays from a ^{137}Cs source obtained by the flash-ADC system with 100 ns channel width (10 MHz). The input pulse was smoothed and the derived waveform was calculated; this procedure enables to extract pulse-height information even for overlapping pulses.

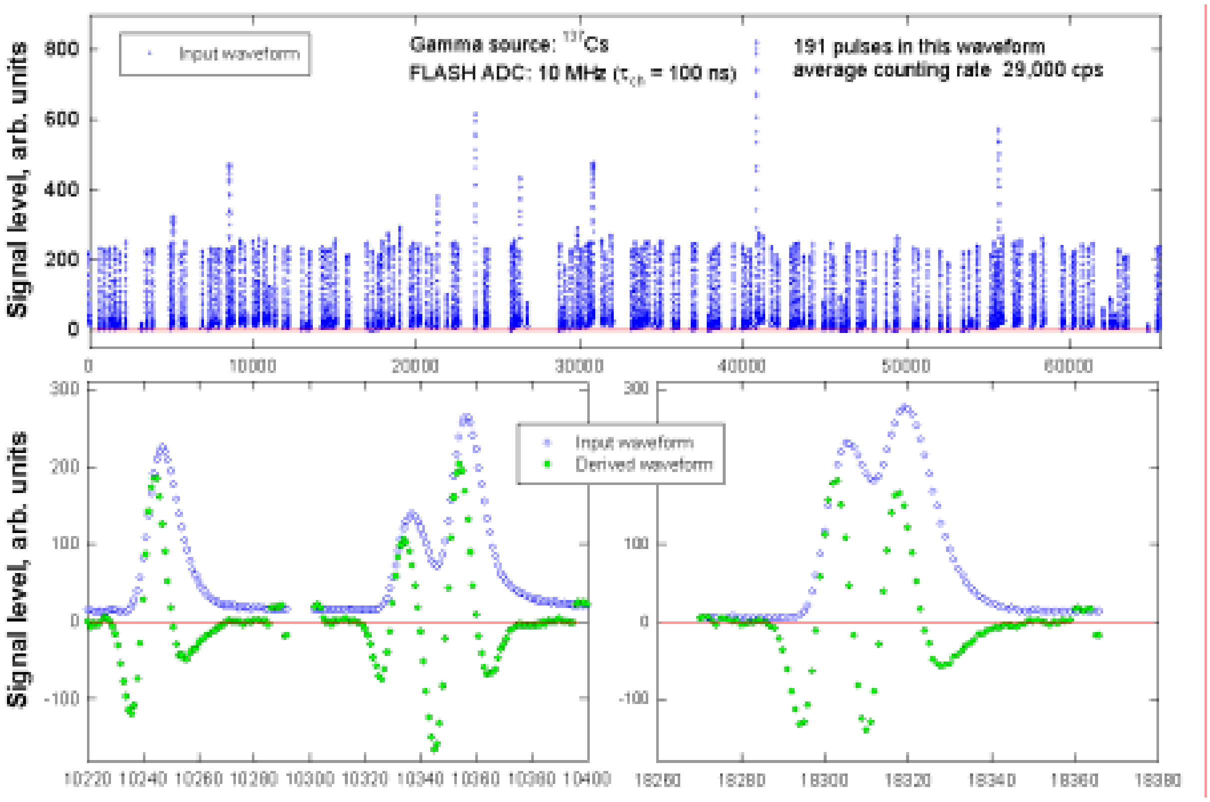

Fig. 4 Waveforms of the BGO detector signal: (top) full-length input waveforms; (down) stretched parts of the input and derived waveforms

The capture cross section of ^{237}Np in resonance energy range was measured using the total energy gamma-ray detector for the first time,. The result of the present measurement has been compared with the evaluated capture cross sections of ENDF/B-VI and JENDL-3.3, as well as with the data measured by other authors. **Figure 5** shows the ratio of the measured capture cross section and the other measurements or evaluated values. To make the comparison easier, the capture cross section was averaged. In the energy range from 1 to 100 eV the energy dependence of the present cross section is close to the experimental data by Weston and Todd [8]. However, there is an obvious discrepancy between the data by Kobayashi *et al.* [9] and the present data. On the otherhand, there is an agreement in the energy range 0.02 –100 eV on the energy dependence of the ^{237}Np capture cross section between the present measurements [10] and the ENDF/B-VI. As far as the JENDL-3.3 is concerned, there is an obvious disagreement with the present data below ~ 0.3 eV, while above this energy an agreement exists, though it is worse than that of ENDF/B-VI. These comparisons show the necessity of the measurements and re-evaluations of the neutron capture cross section at especially theraml energy region.

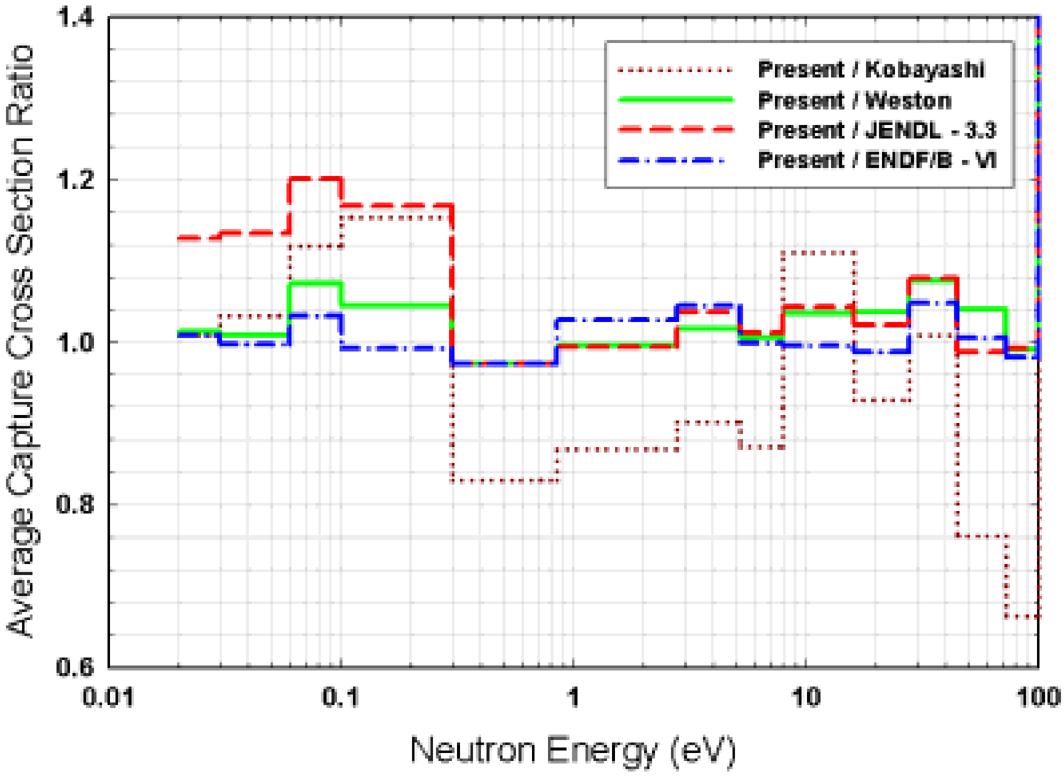

Fig. 5 The ratio of the averaged capture cross section measured by a JNC-Kyoto group and previous measurements or evaluated values

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