Measurement of Fission Cross Section of Pa-231
using Lead Slowing-down Spectrometer

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Making use of a back-to-back type double fission chamber and 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 neutron- induced fission cross section of Pa-231 has been measured from 0.1 eV up to 10 keV with energy resolution (FWHM) of about 40 %. The cross section of the 235U(n,f) reaction in ENDF/B-VI was used as a reference one for the measurement of the fission cross section. The measured result has been compared with the evaluated data in ENDF/B-VI, JENDL-3.2 and JEF-2.2, whose data were broadened by the energy resolution of the KULS.

1. Introduction

Protactinium(Pa)-231 is one of the most interesting nuclei which are related to the production of U-232 in the 232Th-233U fuel cycle(1-3). The fission phenomena, fission energy and mass distributions for the neutron-induced fission of Pa-231 have been investigated(4,5). Although several measurements of the ^{231}Pa(n, f) cross section have been reported at higher energies, the fission cross section has rarely been measured in the lower/resonance energy region(6). One of the reasons may be due to sub-barrier fission which results in a low fission cross section. In addition, Pa-231 is a radioactive actinide element. Then, the pure sample and an intense neutron source are required to overcome the severe experimental conditions for the fission cross section measurement.

Wagemans et al. measured the thermal neutron-induced fission cross section of Pa-231(5,7). Leonard et al. made the cross section measurement at 20 energy points between 0.37 eV and 0.52 eV(8). No experimental data has been obtained in the energy range of thermal neutron to 100 keV except for those measured by Leonard et al. The evaluated fission cross section data are stored in ENDF/B-VI(9), JENDL-3.2(10) and JEF-2.2(11). The ENDF/B-VI and the JEF-2.2 data are markedly discrepant from the JENDL-3.2, especially above about 10 eV.

In the present study, we measure the neutron-induced fission cross section of Pa-231 in the range of 0.1 eV to 10 keV relative to that of the ^{235}U(n, f) reaction by making use of a back-to-back (BTB) type double fission chamber(12) and a lead slowing-down spectrometer(13) coupled to the 46 MeV electron linear accelerator (linac) of the Research Reactor Institute, Kyoto University (KURRI). Below 1 keV, the relative measurement to the ^{10}B(n, α) reaction has been done with a BF3 counter, as before(14,15). The result obtained is compared with the data evaluated in ENDF/B-VI, JENDL-3.2 and JEF-2.2.

2. Samples and Fission Chamber

2.1. Chemical Purification of Pa-231

We have purified the Pa-231 sample by an anion-exchange method from the daughter nuclides, before we proceed to make the electrodeposited layer. The stock solution of Pa-231 in 9M HCl was loaded to a column packed with anion-exchange resin (DOWEX-1 X 8 of 400 mesh), and the column was rinsed with about 10 columns of volume of 9M hydrochloric acid to wash out the daughter nuclides. Then, Pa-231 was quantitatively eluted with 9M hydrochloric acid containing 0.1 M hydrofluoric acid. We have repeated twice the procedures dissolving the Pa-231 sample with nitric acid and making it dry by evaporation. The dried-up sample was dissolved in a few ? of 2-methyl-propanol. By using this solution for non-aqueous electrolysis, Pa-231 was electrodeposited onto an aluminum plate of cathode under 400 V bias for 20 minutes.

2.2. Number of Pa-231 and U-235 Atoms

The amount of Pa-231 deposited was determined by analyzing the α spectrum having energies in the range of 4.74 to 5.03 MeV. Considering the detection efficiency, the α intensities and the half-life, the number of the Pa-231 atoms was found to be (3.32±0.09)x1016. The uncertainties were estimated by taking account of (a) counting statistics of the α measurement, (b) geometrical detection efficiency, and (c) uncertainty in the decay data used.

For highly enriched uranium sample (U-235: 99.91%), we also employed the electrodeposited layer which we used in the previous measurement(15). The number of the U-235 atoms was determined to be (2.81±0.03)x1016 by analyzing the α spectrum at energies of 4.152 to 4.597 MeV.

2.3. Double Fission Chamber

An ionization chamber with two parallel plate electrodes has been employed for the current measurement. Since the back-sides of the Pa-231 and the U-235 deposits face each other, it is called a back-to-back (BTB) type double fission chamber(12). The chamber is made of aluminum and is 40 mm in diameter, 39 mm in length, and the wall thickness of the chamber is ~ 2 mm(14,15). The fission chamber is filled with a mixed gas of 97 % Ar and 3 % N2 at a pressure of 1 atm. The fission chamber collects most of the energy of the fission fragments but not those of the α particles, in order to get good discrimination between pulses of α particle and fission fragment.

3. 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)(13) 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(13). The slowing-down constant K in the relation of E=K/t2 was determined to be 190±2 (keV・μs^2) for the bismuth hole in the KULS(13) by the least squares method using the measured relation between the neutron slowing-down time t in μs 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(13). The relation between the neutron slowing-down time and the energy, and its energy resolution were also verified by Monte Carlo calculations(13).

4. Measurement and Analysis

4.1. Fission Ratio Measurement

The energy-dependant cross section of the 231Pa(n, f) reaction is given by the following relation :

$$\sigma_{Pa}(E) = \frac{C_{Pa}(E)}{C_U(E)} \frac{N_U}{N_{Pa}} \sigma_U(E) \quad , \qquad (1)$$

where, $C_{Pa}(E)$; fission counts of Pa-231 at energy E,
$C_U(E)$; fission counts of U-235 at energy E,
N_U ; number of U-235 atoms in the U-235 deposit,
N_{Pa} ; number of Pa-231 atoms in the Pa-231 deposit,
$\sigma_U(E)$; energy-dependent reference cross section of the ^{235}U(n,f) reaction.

The fission cross section of U-235 is a well-known reference cross section and has been used to determine the neutron flux in the current measurement. The cross section values of $\sigma_U(E)$, whose data were broadened by the resolution function of the KULS, were taken from ENDF/B-VI(9). The typical operating conditions that the KULS was driven were as follows ; the pulse repetition rate was 240 Hz, the pulse width 22 ns, the electron peak current ～1 A, and the electron energy ～31 MeV. After the measurement for more than ～30 hours, the deposited layers of Pa-231 and U-235 in the BTB chamber were interchanged, and another measurement was made for ～45 hours. A background run was carried out without the sample deposits. It was found that the effect of the background-counts was ～0.2 % at most in the cross section values as those in the previous experiments(14,15).

4.2. BF_3 Counter

Below 200 eV, there exist strong neutron resonance peaks in the fission cross sections of Pa-231 and U-235. Therefore, we have employed the ^{10}B(n, α) cross section that shows a smooth and a good 1/v energy dependence in the relevant energy region. The ^{10}B(n, α) reaction is a well-known standard cross section and is often applied to cross section measurements as a reference(16). In order to measure the energy-dependent neutron flux in the resonance energy region, a BF_3 counter was placed in the bismuth hole of the KULS instead of the BTB chamber, as we did before(14,15).

4.3. Electronics and Data Taking

Two identical electronic circuits were employed for the fission-count measurements by the Pa-231 and the U-235 layers in the BTB chamber, as we did before(14,15). Through the amplifiers and the discriminators, signals from the chamber were fed into a time digitizer, which was initiated by the linac electron burst. Two sets of 4096 channels with a channel width of 0.5 μs were allotted to the slowing-down time measurements for the BTB chamber. Pulse height distributions of the fission events were measured together with the slowing-down time measurements.

For the relative measurement to the ^{10}B(n, α) 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 BTB chamber.

5. Results and Discussion

Making use of the BTB chamber and the KULS, the cross section of the ^{231}Pa(n, f) reaction was measured relative to that of the ^{235}U(n, f) reaction at energies from 0.1 eV to 10 keV.

In the resonance energy region below 1 keV, the fission cross section was also measured relative to the $^{10}B(n, \alpha)$ cross section to avoid the resonance interference between Pa-231 and U-235, and the result was normalized to the absolute value of the Pa-231 fission cross section determined relative to the $^{235}U(n, f)$ cross section between 200 eV and 1 keV. The cross section data were obtained by summing up the slowing-down time data in intervals of ～0.12 lethargy width.

Since the Pa-231 sample was chemically purified and was almost free from impurities, no correction was made for the impurity effect. The experimental uncertainties are in the range from 4 % to 40 %, and the major uncertainties are due to the statistical error(2～40%), the error in the reference cross section(～2%) and the number of the Pa-231 and U-235 atoms(2.9%).

The cross sections measured by Leonard et al. are in the range from 30 to 200 mb at 20 energy points between 0.37 eV and 0.52 eV, while the current data show 52 to 64 mb with the uncertainties from 4.7 to 5.4 %. Figure 1 shows the current measurement and the evaluated data in ENDF/B-VI, JENDL-3.2 and JEF-2.2, whose data are broadened by the resolution function of the KULS. The ENDF/B-VI and the JEF-2.2 data almost overlap above 0.1 eV and the later is slightly lower than the former below 0.1 eV. Although the ENDF/B-VI values are in good agreement with the measurement below 4 eV, they are higher near the bump of 5 eV and above about 20 eV obviously. To the contrary, the JENDL-3.2 data are lower than the measured result above about 100 eV as the neutron energy increases.

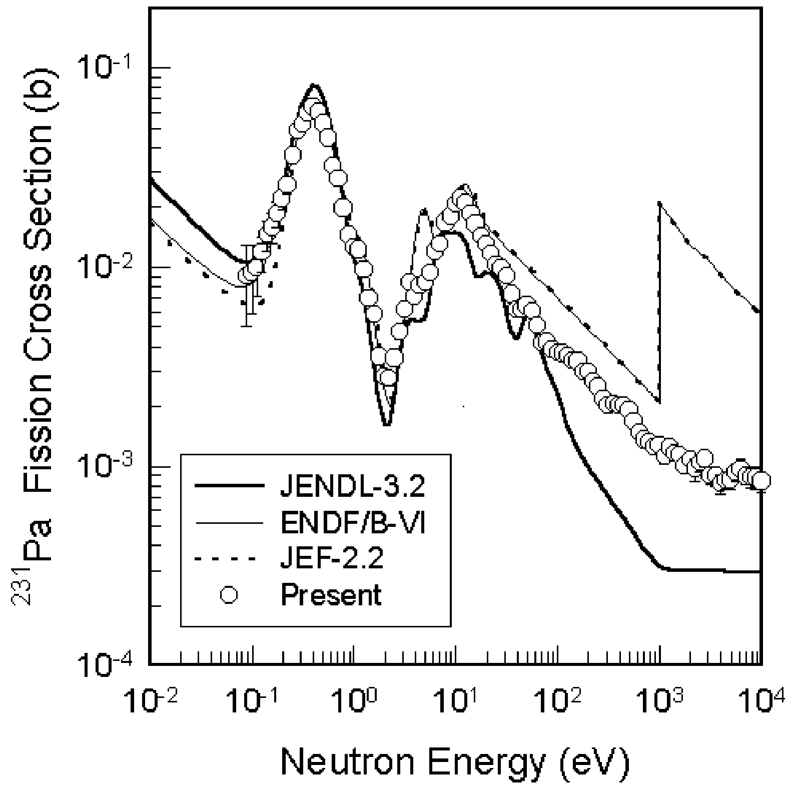

Fig. 1 Comparison of the measured result and the evaluated data in ENDF/B-VI, JENDL-3.2 and JEF-2.2, whose values are broadened by the resolution function of the KULS.

6. Conclusion

The cross section of the ^{231}Pa(n, f) reaction has been measured from 0.1 eV to 10 keV relative to that of the ^{235}U(n, f) reaction, making use of the BTB-type double fission chamber and the lead slowing-down spectrometer KULS at KURRI. No experimental data has been measured before in the energy region from thermal neutron energy to 100 keV except for those by Leonard et al. at energies between 0.37 eV and 0.52 eV. The evaluated data in ENDF/B-VI and JEF-2.2 are in general agreement with the current measurement below 4 eV and are much higher above 20 eV. The JENDL-3.2 data are much lower than the measurement above 100 eV.

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