Measurements of the Neutron Capture Cross Section of Am-243 with the ANNRI beamline, MLF/J-PARC

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Abstract. The neutron capture cross section of ^{243}Am from thermal to 100 keV was measured using the Accurate Neutron-Nucleus Reaction Measurement Instrument (ANNRI) at the Japan Proton Accelerator Research Complex (J-PARC). The time-of-flight (TOF) method using a NaI(Tl) detector was employed in these measurements and the pulse-height weighting technique was used to derive the neutron capture yield. The preliminary cross section was determined by normalizing the results to JENDL-4.0 cross section data at the third resonance of ^{243}Am. In the thermal energy, the preliminary result is in good agreement with the two latest experimental data measured by the TOF and the activation methods but higher than the evaluated values in JENDL-5.

1 Introduction

Precise nuclear data for minor actinides (MAs) are essential for the design of nuclear transmutation systems to reduce the inventory of nuclear waste and for burn-up analysis for actinide composition in fuel cycle scenarios [1–3]. In particular, americium-243 is one of the most abundant MAs in spent fuels and contributes to the long-term radiotoxicity of the spent fuels due to its half-life (7370 y). The neutron capture cross section of ^{243}Am has an influence for the fuel cycle scenarios since it plays a pivotal role to produce ^{244}Cm, an important source of the decay heat in treating radioactive wastes.

Even though the capture cross section of ^{243}Am at thermal energies has been reported in past experiments [4–15] as shown in Figure 1, the data are dispersed between 75 and 85 barns. However, in the kev-energy region, the available experimental data is scare. There are only three data sets measured by using Time-of Flight (TOF) method, those of Wisshak *et al.* [18], Weston *et al.* [19] and Kodama *et al.* [20] and their results have large systematic difference.

In order to improve the accuracy of the neutron capture cross sections of ^{243}Am, we conducted measurements of the neutron capture cross section in the energy range from thermal to 100 keV. In this work, the preliminary results of the neutron capture cross sections are presented and details of the sample, the experimental setup and the data analysis are also described.

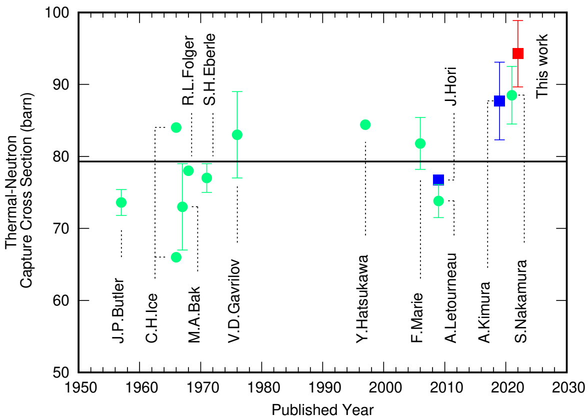

Figure 1. Thermal-neutron capture cross sections of ^{243}Am from past experiments [4–15] and evaluated value from JENDL-4.0 [16] and JENDL-5.0 [17]. Data measured by the activation methods are denoted by filled circles and data measured from TOF experiments are denoted by filled squares.

2 Experimental Setup

The measurements were performed at the Accurate Neutron-Nucleus Reaction Measurement Instrument (ANNRI) of the Materials and Life Science and Technology (MLF) in the Japan Proton Accelerator Research Complex (J-PARC). The pulsed neutron beam was produced through the nuclear spallation reaction at MLF using the 3 GeV proton beam of the J-PARC facility. The proton accelerator was operated in double-bunch mode at a 25 Hz rate and a beam power of about 600 kW.

Prompt gamma-rays emitted from the neutron capture reactions were detected with an NaI(Tl) detector at an-

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Table 1. Isotopic and impurity abundances of ^{243}Am sample.

	Ratio (%)
^{241}Am	2.23
^{242m}Am	0.01
^{243}Am	96.52
^{239}Pu	0.38
^{240}Pu	0.82

gle of 90° with respect to the neutron beam axis. The sample was placed at a neutron flight path of 27.9 m and the Time-of-Flight method was employed to determine the incident neutron energy. In this experiment, a digital data acquisition system (CAEN V1720) was used to analyse anode signals from the NaI(Tl) detector and the pulse height and the TOF of each event were recorded.

3 Samples

An ^{243}Am sample with an activity of 282 MBq (38.14 mg) was used for the measurements. The activity was precisely measured in previous works by γ-ray calorimetry and spectroscopy[21]. In Table 1 the abundances of the isotopes and impurities are listed. The isotopic and impurity abundances were accurately determined by thermal ionization mass spectroscopy (TIMS). The sample in the chemical form of AmO_2 powder was mixed with 39 mg of Y_2O_3 binder. The powders shaped into a disk of a diameter of 10 mm and a thickness of 0.5 mm were packed into a 22-mm Al with 0.1-mm thick walls. A dummy sample, an Al container with the identical shape to the ^{243}Am sample, containing only Y_2O_3, was used to estimate the background induced by the Al case and the yttrium binder. To derive the scattering neutron background by the sample, a ^{nat}C sample with a diameter of 10 mm and a thickness of 0.5 mm was used. In order to measure the energy-dependance of the incident neutron flux, a B_4C sample of 10 mm in diameter and 1 mm in thickness was placed at the sample position and the 478-keV γ-rays from the ^{10}B(n,$\alpha\gamma$)^{7}Li reaction were detected with the NaI(Tl) detector.

4 Data Analysis

The neutron capture yield is calculated from the recorded pulse hight spectrum by applying the pulse-height weighting technique (PHWT)[22]. A weighting function was obtained from the detector response functions which were calculated using the Monte-Carlo simulation code SG.[23]. The ^{243}Am capture yield was obtained by subtracting the backgrounds related to the Al container and the scattered neutrons, respectively. The latter was deduced from the ^{nat}C sample measurement. Corrections for the self-shielding and multiple scattering effects in the sample were applied by using the Monte-Carlo simulation code PHITS[24].

The neutron spectrum was estimated from TOF spectra of the 478-keV γ-ray counts. The 478-keV gamma-ray spectra were divided by the reaction rate of the

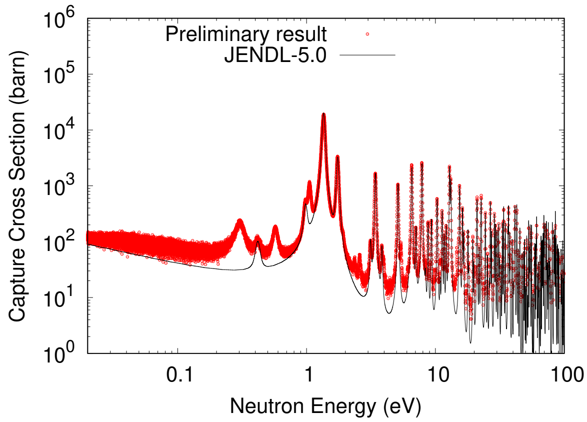

Figure 2. Preliminary neutron capture cross section of ^{243}Am in the energy region from 20 meV to 100 eV and evaluated data of JENLD-5[17].

^{10}B(n,$\alpha\gamma$)^{7}Li reaction. The corrections for the self-shielding and multiple scattering was also applied.

The neutron capture yield obtained by subtracting backgrounds was divided by the neutron energy dependence from the neutron flux in order to derive a relative capture cross section. The derived relative capture cross section was normalized to the evaluated value of the third resonance (1.4 eV) of JENDL-4.0[16].This process is described by the following equation:

$$\sigma_{^{243}Am}(E_n) = N_{3rd}\frac{Y_{^{243}Am}(E_n)C(E_n)}{\phi_n(E_n)} \tag{1}$$

where $\sigma_{^{243}Am}(E_n)$ is the neutron capture cross section, $Y_{^{243}Am}(E_n)$ is the neutron capture yield, $\phi_n(E_n)$ is the incident neutron spectrum, $C(E_n)$ is the correction factor for self-shielding and multiple scattering and N_{3rd} is the normalization factor using the third resonance of JENDL-4.0.

5 Results and Discussion

The preliminary result for the capture cross section of ^{243}Am was derived by nomalizing the relative cross section of ^{243}Am to the evaluated data of JENDL-4.0 for the third resonance of ^{243}Am. Figure 2 shows the preliminary result from 20 meV to 100 eV compared to the evaluated value in JENDL-5[17]. The resonance peaks from the sample impurities of ^{242m}Am and ^{240}Pu are clearly visible. To obtain the final result of the neutron capture cross section, evaluations of the contributions from the capture and fission events of impurities are being conducted.

The preliminary result of the thermal-neutron capture cross section was calculated with the correction for the capture reaction of impurities. The correction value was estimated by using the abundance of impurities and the capture cross section of Am and Pu isotopes from JENDL-4.0. The preliminary thermal-neutron capture cross section of ^{243}Am are shown in Table 2 together with previously reported experimental data and evaluated data from JENDL-5.0. The statistical uncertainty was included in the preliminary result. The preliminary result agrees within

Table 2. Preliminary result of the thermal-neutron capture cross section of ^{243}Am and the past measured and evaluated data.

Author	σ_{ther} (barn)	Method
This work	94.3 ± 4.6	TOF
Nakamura *et al.* [15]	88.5 ± 4.0	Activation
Kimura *et al.* [14]	87.7 ± 5.4	TOF
JENDL-5 (2022)	85.79	Evaluation

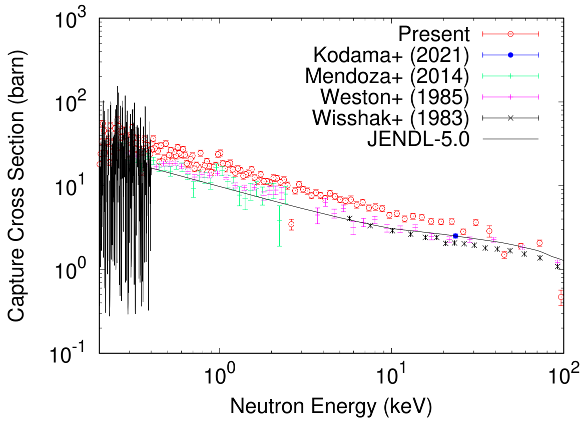

Figure 3. Preliminary result of the neutron capture cross section of ^{243}Am compared with the past data and the evaluated values from JENDL-5.

the uncertainties with the experimental data from Kimura *et al.*[14] and Nakamura *et al.*[15]. However, the present result is 6% larger. This difference probably comes from the contribution of fission events that has not been removed in the present analysis yet. This correction for the fission events will be applied in future work. The evaluated value from JENDL-5.0 is 9% smaller than the present result.

Above 200 eV, the preliminary results of the capture cross section are shown in Figure 3 together with the past data sets and the JENDL-5 evaluation. The present result is systematically higher than the past data since the subtraction of the background of Al case and the scattered neutron is underway. Further analysis will be completed in the future.

6 Conclusions

The neutron capture cross section of ^{243}Am was measured with NaI(Tl) detector at ANNRI beam line in the neutron energy region from thermal to 100 keV. The relative neutron capture cross section was determined using pulse-height weighting technique. The derived relative neutron capture cross section was normalized to the evaluated value of the third resonance value of JENDL-4.0. The preliminary result of thermal-neutron capture cross section is in good agreement with the previous experimental data of Kimura *et al.* and Nakamura *et al.* within uncertainties. The further analysis to obtain the final result of the cross section and the resonance analysis will be completed in the future.

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