

Fast-neutron capture cross section data measurement of minor actinides for development of nuclear transmutation systems

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Abstract. The neutron capture cross sections of ²³⁷Np, ²⁴³Am and ²⁴¹Am in the keV energy region were measured by the time-of-flight method at the Japan Proton Accelerator Research Complex (J-PARC). Characterization of the minor actinide samples were made to reduce the uncertainties of the cross section. A neutron beam filter system was developed and installed in ANNRI to solve the double-bunch mode issue of J-PARC. The pulse-height weighting technique was employed to determine the neutron capture cross sections. The neutron capture cross sections of ²³⁷Np, ²⁴³Am and ²⁴¹Am were determined with higher accuracies than the past experiments.

1 Introduction

Long-lived minor actinides (MAs) in nuclear waste from nuclear power plants are a long-standing issue to continue nuclear energy production. In order to solve the issue, researchers have suggested nuclear transmutation, in which long-lived radionuclides are transmuted into stable or shorter-life nuclides via neutron-induced nuclear reactions [1]. Development of nuclear transmutation systems such as an accelerator-driven system requires accurate neutron nuclear reaction data [2,3]. The present research project entitled “Study on accuracy improvement of fast-neutron capture reaction data of long-lived MAs for development of nuclear transmutation systems” has been conducted as a joint collaboration including Tokyo Tech, Japan Atomic Energy Agency and Kyoto University [4]. This project focuses on the neutron capture reaction of MAs, especially ²³⁷Np, ²⁴¹Am and ²⁴³Am, in the fast neutron energy region. The final goal of this project is to improve the accuracy of the neutron capture cross sections of ²³⁷Np, ²⁴¹Am and ²⁴³Am by employing a high-intensity neutron beam from a spallation source of the Japan Proton Accelerator Research Complex (J-PARC) [5]. To achieve the goal, a neutron beam filter system in J-PARC, sample characteristic assay, and theoretical reaction model were developed. A part of the results of the project are reviewed in this paper.

2 Experiments

The neutron beam filter system was designed to solve the double-pulse beam issue in J-PARC. Neutrons are produced in the spallation target with a cycle of 25 Hz

in the Materials and Life Science Experimental Facility (MLF) of J-PARC. The proton accelerator is operated in a special operational mode such that two 100-ns wide proton beam pulses separated by 600 ns are injected into the spallation target every 40 ms as shown in Fig. 1. The purpose of the double-pulse operation is to increase the thermal neutron flux. The double-pulse beam time structure disappears in the neutron slowing-down process in moderators and thereby does not affect a neutron time-of-flight (TOF) spectrum in the thermal energy region. However, in the higher energy region (> 10 eV), the double-pulse beam is crucially problematic [6]. Resonance peaks on a TOF spectrum become doublet due to the double pulse beam time structure. This makes resonance analysis more complicated, requiring the implementation of a resolution function of the double-pulse beam into resonance analysis.

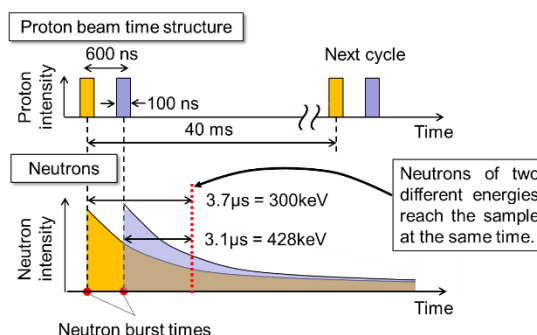


Fig. 1. Time structure of double-pulse mode operation of J-PARC/MLF and neutron generation.

Situation is even worse in the fast neutron energy region that is above the resolved resonance region. The cross section is a smooth function of neutron energy.

Neutrons that have two different energies associated with the double proton beam pulses reach the sample at the same time. It is difficult to deconvolute the measured neutron capture yield into single-pulse components.

To solve the double-pulse beam issue in J-PARC/MLF, a neutron beam filter system has been developed and installed into the Accurate Neutron Nucleus Reaction Measurement Instrument (ANNRI) [7]. The neutron beam filter technique has been often used in nuclear reactors, to make a mono-energetic neutron beam [8,9]. A total cross section minimum caused by an s-wave resonance interference is used as an energy window of neutron energy filtering. Combining optimal filter materials, a neutron energy spectrum can be tailored to be mono-energetic.

In the present project, the neutron beam filter technique was employed to separate two different neutron energy components at the same TOF channels. The sample position of ANNRI was 27.9 m from the neutron source. The neutron beam filter system was placed at a flight length of 18 m as shown in Fig. 2. The selected filter materials were Fe, Bi, Al, Si, Cr and Sc. They have resonances which are suitable for filtering neutrons in the keV energy region. The available resonance energies of the filter materials are summarized in Table 1. The neutron filter materials were fabricated and tested in neutron beam experiments at J-PARC. In the test experiments, transmission neutron spectra were measured by detecting prompt γ -rays from the $^{10}\text{B}(n,\alpha\gamma)^7\text{Li}$ reaction placing a boron sample at the sample position. The 478-keV γ -rays from the $^{10}\text{B}(n,\alpha\gamma)^7\text{Li}$ reaction were detected with a NaI(Tl) detector. Figures 3 and 4 show neutron TOF spectra for 20-cm thick Fe filter and 20-cm thick Si filter, respectively. The neutron spectra show well-filtered peaks at 24, 54 and 146 keV.

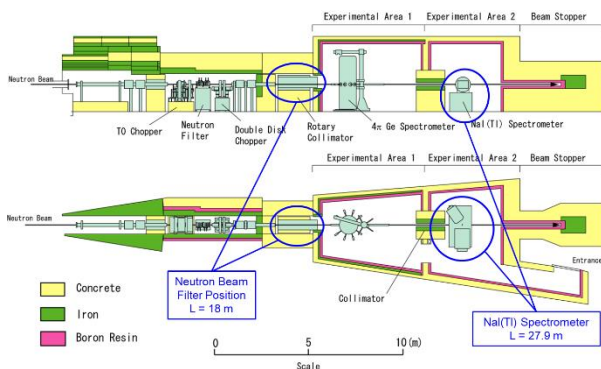


Fig. 2. Accurate Neutron Nucleus Reaction Measurement Instrument (ANNRI), and the positions of neutron beam filter system and NaI(Tl) spectrometer.

This project used NaI(Tl) detectors for measurements of neutron capture cross sections. The ANNRI-NaI(Tl) detectors consist of two differently-sized NaI(Tl) detectors [10]. The larger detector (crystal size: 330-mm diameter, 203-mm long) was placed at a scattering angle of 90° as respect to the beam axis. The smaller detector (203-mm diameter 203-mm long) was at an angle of 125° . The detectors were shielded from background γ -rays and neutrons with various shielding materials such as lead, borated polyethylene and

paraffin, cadmium and ^6Li -enriched lithium compounds. A sample was placed in a carbon-fiber beam duct and set at a flight length of 27.9 m. The beam duct was filled with helium gas to reduce scattering neutron background by air.

Table 1. Available resonance energies of the filter materials

Material	Resonance energies (keV)
Fe	24
Bi	2.2, 5.1, 11.5, 15.3, 32.9
Al	27, 125, 265
Si	54, 146
Cr	56, 82
Sc	7.5, 10.5

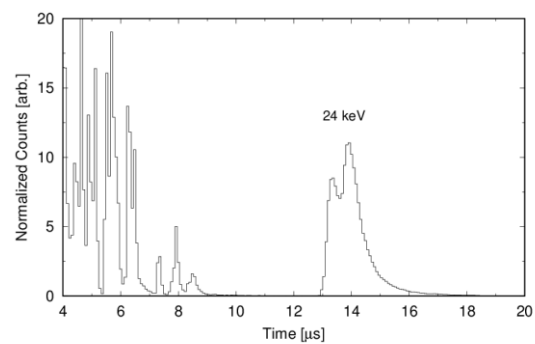


Fig. 3. Neutron TOF spectrum of neutron beam filtered with the 20-cm thick Fe filter.

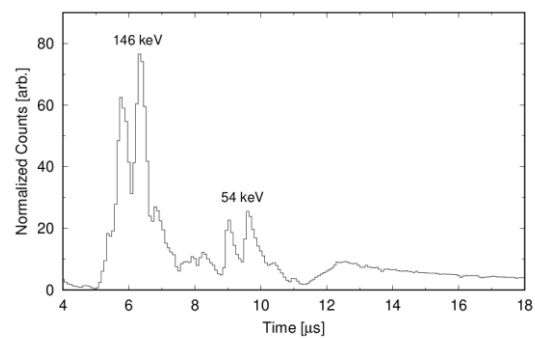


Fig. 4. Neutron TOF spectrum of neutron beam filtered with the 20-cm thick Si filter.

One of the main sources of systematic uncertainties of cross section is sample characteristics. The sample characteristic assay, especially, precise mass spectrometry of isotopes and impurities lowers sample systematic uncertainties. Isotope compositions are sometimes off from the rated values. Radioactive samples often have impurities not listed in a certificate sheet.

In this project, a thermal ionization mass spectrometer (TIMS) of the Institute for Integrated Radiation and Nuclear Science of Kyoto University was used to analyse the isotope compositions and impurities of samples. All MA samples for measurement at J-PARC were sealed in containers. For analysis, unsealed MA solution taken from the same batch as the sealed MA samples were prepared. The results of the impurity

analysis are summarized in Table 2. The impurity ratios were determined very accurately, reducing the uncertainties of the final cross sections.

The radioactivities of ^{237}Np , ^{243}Am and ^{241}Am were 5.18, 278.2 and 957.4 MBq, respectively. The radioactivities were determined from heat measurement by calorimeter and γ -ray measurement by a Ge detector [11].

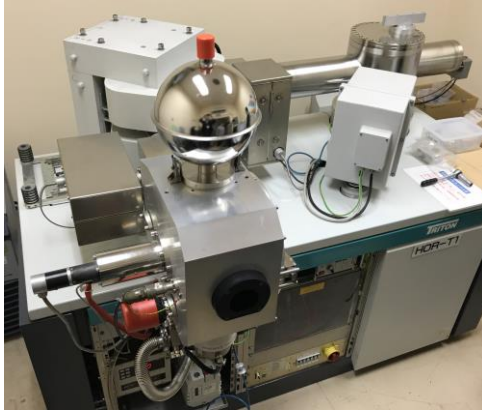


Fig. 5. Thermal ionization mass spectrometer (TIMS).

Table 2. Impurity ratios of the ^{237}Np , ^{241}Am and ^{243}Am samples measured with TIMS.

	^{237}Np sample	^{241}Am sample	^{243}Am sample
^{237}Np	1	-	-
^{241}Am	-	1	$(2.33\pm 0.01)\times 10^{-2}$
$^{242\text{m}}\text{Am}$	-	-	$(0.48\pm 0.05)\times 10^{-3}$
^{243}Am	-	$(1.4\pm 0.7)\times 10^{-6}$	1
^{238}Pu	$(6.1\pm 0.6)\times 10^{-7}$	-	$(1.4\pm 0.2)\times 10^{-5}$
^{239}Pu	$(5.1\pm 0.3)\times 10^{-4}$	$(9.2\pm 2.2)\times 10^{-3}$	$(4.0\pm 0.1)\times 10^{-3}$
^{240}Pu	$(3.9\pm 0.4)\times 10^{-5}$	$(3.1\pm 0.8)\times 10^{-5}$	$(8.6\pm 0.2)\times 10^{-3}$
^{241}Pu	$(2.0\pm 1.4)\times 10^{-6}$	-	-
^{242}Pu	$(2.4\pm 1.6)\times 10^{-6}$	$(5.2\pm 1.3)\times 10^{-5}$	$(9.9\pm 2.7)\times 10^{-6}$

3 Results

The neutron capture cross sections of ^{237}Np , ^{243}Am and ^{241}Am were measured using the neutron beam filter system of ANNRI at J-PARC. The experimental results are shown in Figs. 6-8. Past experimental data [12-17] and evaluated cross sections are also plotted for comparison. Part of the experimental results have been reported in Refs. [18-20]. The details of the experiments and data analysis can be found therein.

Three neutron filters, Fe, Si and Cr, were used for measurement of ^{237}Np . For ^{243}Am and ^{241}Am , two neutron filters, Fe and Si, were used. The pulse-height weighting technique (PHWT) [21, 22] was employed to derive the neutron capture yield. Measurements of gold

samples were also made and the relative ratios of the capture yields of the sample to the gold measurements were obtained. The absolute values of the capture cross sections of the samples were determined from the JENDL-4.0 evaluated cross section of the $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ reaction.

The errors of the present cross sections plotted in Figs. 6-8 include statistical and systematic uncertainties. Uncertainties of total sample mass, impurity abundances, neutron transport coefficients, standard cross section and weighting function of PHTW were considered as the systematic uncertainty. As seen in Figs. 6-8, the capture cross sections obtained in the present experiments have less uncertainties than the past measurements. In particular, the results of ^{243}Am and ^{241}Am achieved much higher accuracies than the past measurements.

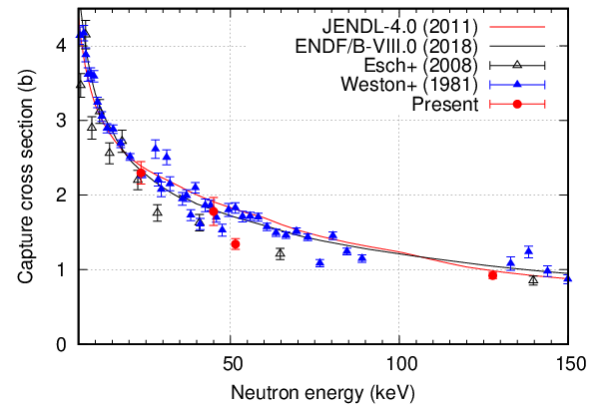


Fig. 6. Neutron capture cross section of ^{237}Np . The present experimental results are compared with past experimental data and evaluated cross sections.

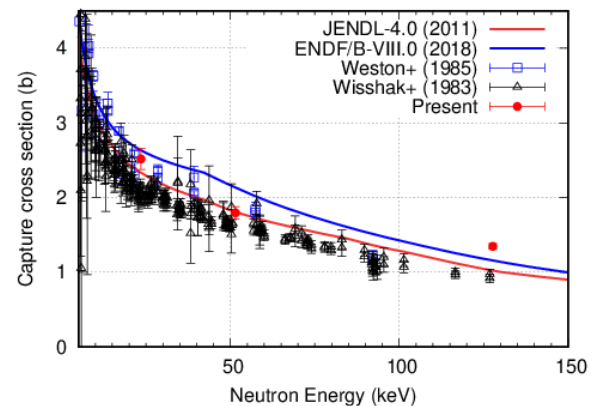


Fig. 7. Neutron capture cross section of ^{243}Am . The present experimental results are compared with past experimental data and evaluated cross sections.

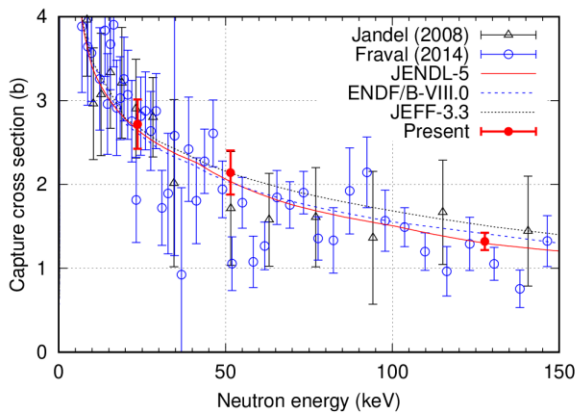


Fig. 8. Neutron capture cross section of ^{241}Am . The present experimental results are compared with past experimental data and evaluated cross sections.

4 Conclusions

The neutron capture cross sections of ^{237}Np , ^{243}Am and ^{241}Am in the keV energy region were measured by the TOF method at J-PARC/ANNRI. Characterization of the MA samples were made to reduce the uncertainties of the cross section measurement. To solve the double-bunch mode issue of J-PARC, a neutron beam filter system was developed and installed in ANNRI. The accuracies of the neutron capture cross sections of ^{237}Np , ^{243}Am and ^{241}Am were improved.

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