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ARTICLE

Measurements of keV-Energy neutron capture cross section of ^{243}Am using neutron filter

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The neutron capture cross section of ^{243}Am were measured in the keV-energy region applying the implemented neutron filtering system of the Accurate Neutron-Nucleus Reaction Measurement Instrument beamline in the Materials and life science facility of the Japan Proton Accelerator Research Complex. A Si beam filter was used to obtain a quasi-monoenergetic neutron beam at averaged neutron energies of 51.5 and 127.7 keV. The neutron capture γ-rays were detected with a NaI(Tl) spectrometer. The pulse height weighting technique was employed to derive the neutron capture yield from pulse height spectra. The cross section was determined relative to the ^{197}Au capture cross section of JENDL-4.0. The results were obtained with higher accuracy than past available experimental data sets.

Keywords: nuclear transmutation; nuclear data; neutron capture cross section; americium-243; ANNRI; MLF; J-PARC

1. Introduction

The study and design of nuclear transmutation systems in nuclear waste management require accurate nuclear data of minor actinides (MAs), including neutron-induced nuclear reactions. The nuclear transmutation systems are expected to reduce the radiotoxicity and the amount of MAs in the nuclear wastes. A promising option among the proposed nuclear transmutation systems is an accelerator-driven system (ADS) that consists of accelerator-based neutron source and a sub-critical core containing MAs. The contribution of uncertainties of the evaluated nuclear data of JENDL-4.0 to reactor physics parameters, which are essential to evaluate the performance of the systems, were quantified in a recent work [1]. It was demonstrated that the neutron capture reaction of ^{243}Am is one of the major contributors among all MAs. In the neutron energy of interest, from 0.454 keV to 1.35 MeV, uncertainties for the capture cross section of ^{243}Am was evaluated to be 10% while the target uncertainty was set to 2% in a sensitivity study of ADS by Subgroup 26 of the Working Party on International Nuclear Data Evaluation Co-operation of OECD/NEA [2]. The large uncertainties stem from the poor quality of the experimental data.

There are only two available data sets for the neutron capture cross section of ^{243}Am in the keV-energy region [3,4]. Both results have discrepancies, although they agree together within the large uncertainties. Therefore, the uncertainty of its neutron capture cross section is not small enough to design nuclear transmutation systems. Currently, to reduce the uncertainties of nuclear data of MAs, measurements of the nuclear data are ongoing at the Accurate Neutron-Nucleus Reaction Measurement Instrument (ANNRI) beamline of the Materials and Life Science Experimental Facility (MLF) in the Japan Proton Accelerator Research Complex (J-PARC). In addition, a neutron filtering system has been installed at the ANNRI beamline for fast neutron cross section measurement [5] and the capture cross section of ^{243}Am was measured by using the neutron filter with ^{nat}Fe as a filter material at 23.5 keV [6].

In order to improve the accuracy of the neutron capture cross section of ^{243}Am, the neutron capture cross section was measured at ANNRI using the neutron filtering technique with another filter material. ^{nat}Si was used as the filter material to obtain a quasi-monoenergetic neutron beam with averaged neutron energies of 51.5 and 127.7 keV. The cross section in the higher energy can be obtain by using ^{nat}Si filter instead of ^{nat}Fe due to the different resonance structures in the cross section of these materials. In this paper, the results of the cross section measurement are presented and the experimental setup, the sample information and the data analysis are also described.

2. Experimental setup

The experiments were performed at the ANNRI beamline of MLF in J-PARC. The pulsed neutron beam was provided

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through the nuclear spallation reaction using the 3 GeV proton beam of J-PARC. The incident neutron energy was determined by the neutron Time-of-Flight (TOF) method. However, the conversion of the neutron TOF to the neutron energy is complicated due to the J-PARC accelerator operative condition. The proton accelerator is operated in the double-bunch mode, where two proton bunches with a separation time of 0.6 μs are injected into the spallation target every 40 ms to generate the neutron beam to achieve a high neutron beam intensity. The time difference between the two proton bunches is not negligible for the neutron TOF measurement in the keV-energy region. Neutrons with two different energies originating the two proton bunches arrive at the sample at the same time. This makes high-energy neutron measurements difficult due to the mixing of the two neutron energies at a TOF. To solve such an issue, the newly implemented neutron beam filtering system was employed in the present measurements. The experiments were conducted with Si neutron filter in order to extract a quasi-monoenergetic neutron beam with the averaged energy of 51.5 and 127.7 keV which were evaluated in the previous study [5]. Alongside the filter, a Cd filter was used to cut off low-energy neutrons. Prompt capture γ-rays were detected using the NaI(Tl) spectrometer located at a flight path of 27.9 m. In the experiments, a digital data acquisition system (CEAN V1720) was used to analyze anode signals from the NaI(Tl) detector. Simultaneously, the pulse height was calculated from the integral area of the anode signals. The details of the neutron filtering system of ANNRI beamline was described in Ref [5].

3. Samples and measurements

A pellet of 282 MBq of ^{243}Am oxide mixed with 39 mg of Y_2O_3 used as a binder was used in the present experiments. The radioactivity of the sample was precisely measured in a previous work by γ-ray spectroscopy and calorimeter measurement [7]. The pellet was 10.0 mm in diameter and 0.1 mm in thickness. The abundances of the isotopes and impurities in the sample are listed in **Table 1**. These isotopic and impurity abundances were determined with a thermal ionization mass spectrometer (TIMS). The sample was enclosed in an Al case having an outer diameter of 22 mm and a wall thickness of 0.1 mm. The sample-dependent background relative to the Al canning was estimated by measuring a replica of the Al dummy case containing only Y_2O_3 without ^{243}Am oxide. In the present analysis, the neutron capture cross section of ^{243}Am was derived relative to the neutron capture cross section of ^{197}Au. Hence, a gold sample having a diameter of 10.0 mm, a thickness of 1.0 mm and a mass of 1452.7 mg was used to derive the ^{197}Au neutron capture yield. Furtherer, the time and energy distribution of the neutron beam was obtained by measuring the 478 keV gamma rays produced via the ^{10}B(n,αγ)^{7}Li reaction in B_4C sample with a 90.4% ^{10}B enrichment. This is a conventional method because the cross section of the ^{10}B(n,$\alpha\gamma$) reaction is well known. In addition to these samples, a ^{nat}C sample with a diameter of 10 mm and a thickness of 0.5 mm was used to estimate the scattering neutron background by the sample, since the elastic scattering is dominant in the neutron-carbon reactions. Lastly, the sample-independent background was derived in a "blank" measurement with no sample.

4. Data analysis

The neutron time distribution of the incident neutron flux was determined using the boron sample measurement by analyzing the TOF spectrum of counts of the emitted γ-rays of 478 keV from the ^{10}B(n,$\alpha\gamma$)^{7}Li. Figure 1 shows the time distribution of the neutron flux for the filtered neutron beam. The upper horizontal axis represents the incident neutron energy corresponding to the TOF. The

Table 1. Impurity information of the Am sample.

Nucleus	^{241}Am	^{242m}Am	^{243}Am	^{239}Pu	^{240}Pu	^{242}Pu	^{244}Cm
Impurities (%)	2.23	0.04	96.5	0.38	0.82	< 0.001	< 0.001

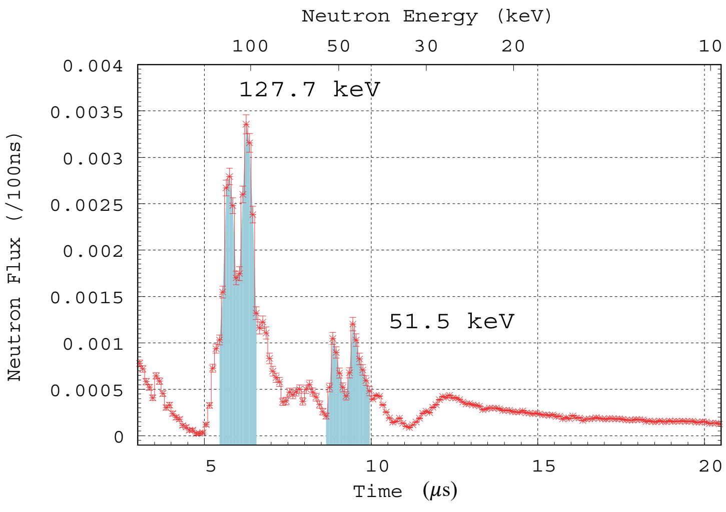

Figure 1. Neutron time distribution of the incident neutron flux.

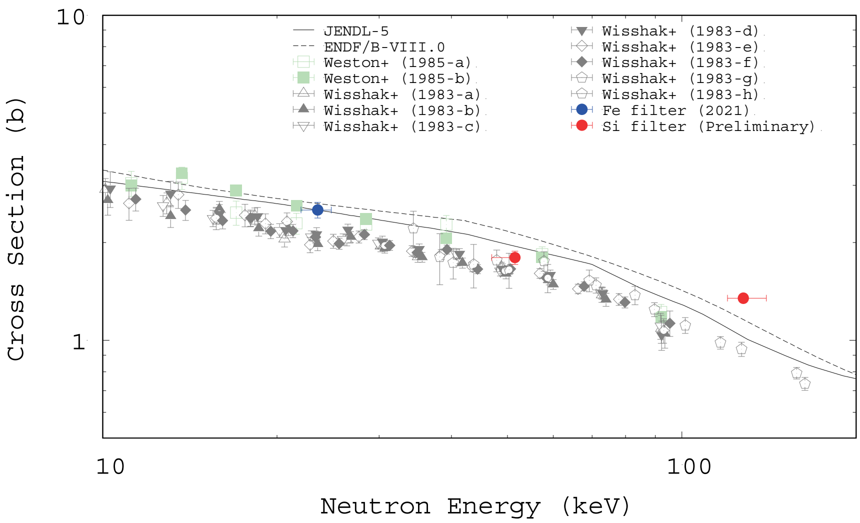

Figure 2. Neutron capture cross section of ^{243}Am compared to the past experimental data and evaluated nuclear data from JENDL-5 and ENDF/B-VBIII.0.

colored region in **Figure 1** is the TOF gate region for PH spectrum analysis because there were sufficient. Each gate region shows doublet peaks to the double bunch operation. Background from the Al container and the scattered neutrons were subtracted to obtain the net capture γ-ray spectrum of ^{243}Am. Then, the neutron capture yields were derived by applying the pulse-height weighting technique [8]. The data from the gold measurement were analyzed in the same manner and the capture yield of ^{197}Au was derived. The neutron capture cross section was determined at averaged neutron energies of 51.5 and 127.7 keV.

5. Results

The preliminary results of the capture cross section of ^{243}Am were determined at averaged neutron energies of 51.5 and 127.7 keV using the neutron filtering system. The cross sections were obtained relative to the neutron capture cross section of ^{197}Au. These results are compared to the TOF experimental results of Weston et al. [3] and Wisshak et al. [4] with the evaluated data from JENDL-5 [9] and ENDF/B-VIII.0[10] in **Figure 2**. The cross section measured in previous experiments using Fe neutron filter is also shown. The horizontal error bars of the present result represent the neutron energy distributions corresponding to the TOF gate in the analysis. The present cross section at 51.5 keV agrees with the experimental data of Weston et al. On the other hand, the present value at 127.7 keV is higher than the evaluated values of JENDL-5 and ENDF/B-VIII.0, and the experimental data of Wisshak et al. Two reported data were measured in TOF experiments with a pulsed neutron beam, while the preliminary results were measured by using quasi-monoenergetic neutron beam. One of the data was normalized to the thermal neutron capture cross section of ^{243}Am by Weston et al. and the other one was determined relative to the capture cross section of ^{197}Au by Wisshak et al in the same manner of this analysis. Since the origin of the discrepancy has not been clear, further investigation is needed. The uncertainties of the preliminary results are smaller than the previous result and the reported data by Weston et al. and Wisshak et al since the statistical error and the systematic error related to the sample mass was reduced.

6. Summery

The neutron capture cross section of ^{243}Am was measured at incident neutron energies of 51.5 and 127.7 keV in the ANNRI beamline of MLF/J-PARC. The neutron filtering system using ^{nat}Si was employed to solve the issue of the double-bunch beam operation of J-PARC. The neutron capture cross section was determined relative to the capture cross section of ^{197}Au from JENDL-4.0. The results achieved higher accuracy than those of the past experimental data.

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References

[1] H. Iwamoto, et al., Sensitivity and uncertainty analysis for an accelerator driven system with JENDL-4.0, *Journal of Nuclear Science and Technology* 50 (2013), pp. 856-862.
[2] M. Salvatore, et al., Uncertainty and target accuracy assessment for innovative systems using recent covariance data evaluations, *Nuclear Science NEA/WPEC-26* (2008).
[3] L.W. Weston and J.H. Tod, Neutron Capture Cross Section of ^{243}Am, *Nuclear Science and Engineering* 91 (1985), pp. 444-450.
[4] K. Wisshak and F. Kappeler, Neutron Capture and Fission Cross Section of ^{243}Am in the Energy Range from 5 to 250 keV, *Nuclear Science and Engineering* 85 (1983), pp. 215-217.
[5] G. Rovira, et al., Neutron beam filter system for fast neutron cross-section measurement at the ANNRI beamline of MLF/J-PARC, *Nuclear Instruments and*

Methods in Physics Section A 1003 (2021), 165318.
[6] Y. Kodama, et al., Measurements of the neutron capture cross section of ^{243}Am around 23.5 keV, *Journal of Nuclear Science and Technology* 53 (2021).
[7] M.L. Macklin and J.H. Gilbbons, Capture-Cross-Section Studies for 30-220 keV Neutrons Using a New Technique, *Physical Review* 159 (1967), pp. 1007-1012.
[8] K. Terada, et al., Technical developments for accurate determination of amount of samples used for TOF measurements, *Web of Conferences* 146 (2017), 03019.
[9] O. Iwamoto, et al., Status of JENDL, *EPJ Web of Conferences* 339 (2020).
[10] D.A. Brown, et al., ENDF/B-VIII.0: The 8th Major Release of the Nuclear Reaction Data Library with CIELO-project Cross Sections, New Standards and Thermal Scattering Data, *Nuclear Data Sheet* 148 (2018), pp. 1-142.