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Neutron capture and neutron-induced fission experiments on americium isotopes with DANCE

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Abstract. Neutron capture cross section data on Am isotopes were measured using the Detector for Advanced Neutron Capture Experiments (DANCE) at Los Alamos National Laboratory. The neutron capture cross section was determined for ²⁴¹Am for neutron energies between thermal and 320 keV. Preliminary results were also obtained for ²⁴³Am for neutron energies between 35 eV and 200 keV. The results on concurrent neutron-induced fission and neutron-capture measurements on ^{242m}Am will be presented, where the fission events were actively triggered during the experiments. In these experiments, the Parallel-Plate Avalanche Counter (PPAC) detector that surrounds the target located in the center of the DANCE array was used as a fission-tagging detector to separate (n,γ) from (n,f) events. The first evidence of neutron capture on ^{242m}Am in the resonance region in between 2 and 9 eV of the neutron energy was obtained.

Keywords: Neutron capture, Neutron-induced fission, Cross sections

INTRODUCTION

High-fidelity neutron capture and neutron-induced fission data on major and minor actinides are desired for many applications, such as the design of the future fast reactors, nuclear forensics/attribution and stockpile stewardship. Much data on neutron capture cross sections exist, however the accuracy of the data is still in question. For fissioning actinides, the uncertainties of the neutron capture-to-neutron-induced fission cross section ratios are still rather large, even for the major actinides such as ²³⁵U and ²³⁹Pu. In case of ^{242m}Am, the neutron capture cross section is unknown and only few experiments determined the thermal cross section using the activation measurement in the reactor neutron flux environment [1, 2, 3, 4]. In this paper, we will present the overview of the measurements carried out recent years with DANCE on americium isotopes ²⁴¹Am, ²⁴³Am and ^{242m}Am at Los Alamos Neutron Scattering Center (LANSCE).

EXPERIMENTS

Details on experimental set-up, data-acquisition and the methods used in the data analysis can be found in [5] and references therein. Here we will briefly summarize the main features. The capture or fission γ-rays are detected by 160 BaF₂ crystals of the DANCE

TABLE 1. Am targets used in measurements with DANCE.

	Year	Backing	Mass [μg]	PPAC used	Prepared
^{241}Am	2005	Ti	~ 50	yes	C-NR, LANL
^{243}Am	2005	Ti	~ 50	yes	C-NR, LANL
^{241}Am	2006	Ti	219	no	C-NR, LANL
^{242m}Am	2006	Be	47	yes	LLNL
^{242m}Am	2007	Ti	154	yes	LLNL
^{243}Am	2007	Ti	267	no	C-NR, LANL

array arranged in 4π geometry around the target. The neutrons impinging on the DANCE target are produced in the 500 MeV proton-induced reactions on tungsten spallation target and moderated by water moderator in the beginning of the 20.25 m long flight path. The neutron time-of-flight is measured using the start signal from the proton beam pick-off and the stop signal from the DANCE array. The stop signal in the DANCE array is obtained from the DANCE crystals that fired in the 20 ns wide coincidence window. The methods of total γ -ray calorimetry are used for the determination of the cross section. Appropriate data reduction is performed by selecting the region of γ -ray multiplicity and total γ -ray energy for the cross section analysis. In case of fissioning actinides, we use an active fission tagging detector (PPAC) to distinguish the neutron-induced fission events from background and neutron capture events [6, 7].

The summary of americium targets used at DANCE is shown in Table 1, together with the year of the preparation, backing material, total mass, PPAC usage, and a place of the preparation. The diameter of the targets were 6.35 mm except the ^{242m}Am targets were 7 mm. In 2005, the PPAC detector was used for the measurements of neutron capture cross sections on $^{241,243}\text{Am}$. The analysis showed however, that the fission component in the neutron capture spectra is negligible and that the backgrounds originating from the neutron scattering off the PPAC components were overwhelming in the neutron energy region $E_n > 1$ keV. The new targets were prepared in 2007 and measurements were repeated without the PPAC with the significantly better signal-to-background ratio.

RESULTS

Neutron capture of $^{241,243}\text{Am}$

The finalized data on the neutron capture cross section of ^{241}Am were published in [5]. The absolute $^{241}\text{Am}(n,\gamma)$ cross sections were obtained in the region of neutron energies from thermal to 320 keV and the results are shown together with the ENDF/B-VII.0 evaluation in Fig. 1. The neutron flux at the target position was normalized using the targets of ^{197}Au with the very well known thickness and diameter. The efficiency of the neutron capture cascades was extracted using the γ -ray energy spectra on the strong resonances. The determination of the neutron flux and detection efficiency enabled the determination of the absolute cross section for the ^{241}Am target. Resonance parameters

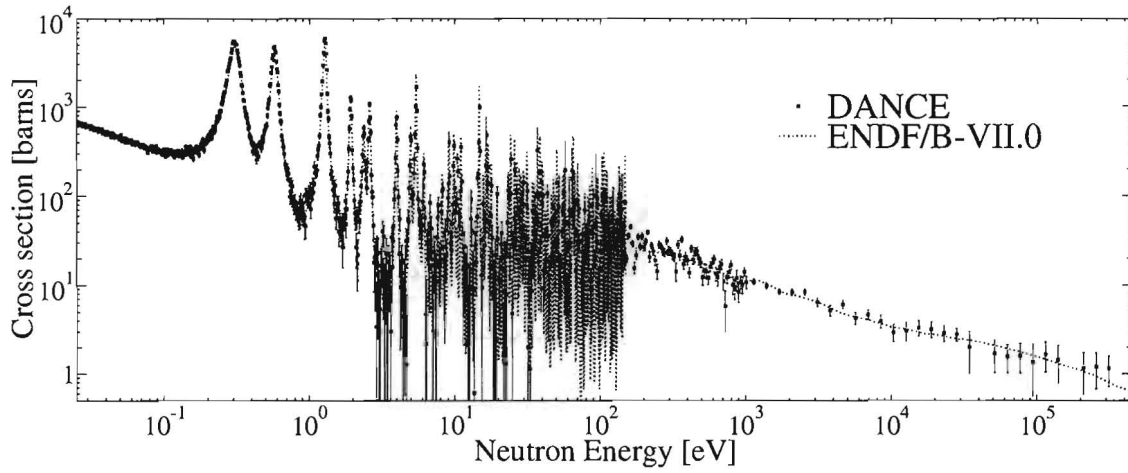


FIGURE 1. $^{241}\text{Am}(n,\gamma)$ cross section measured at DANCE.

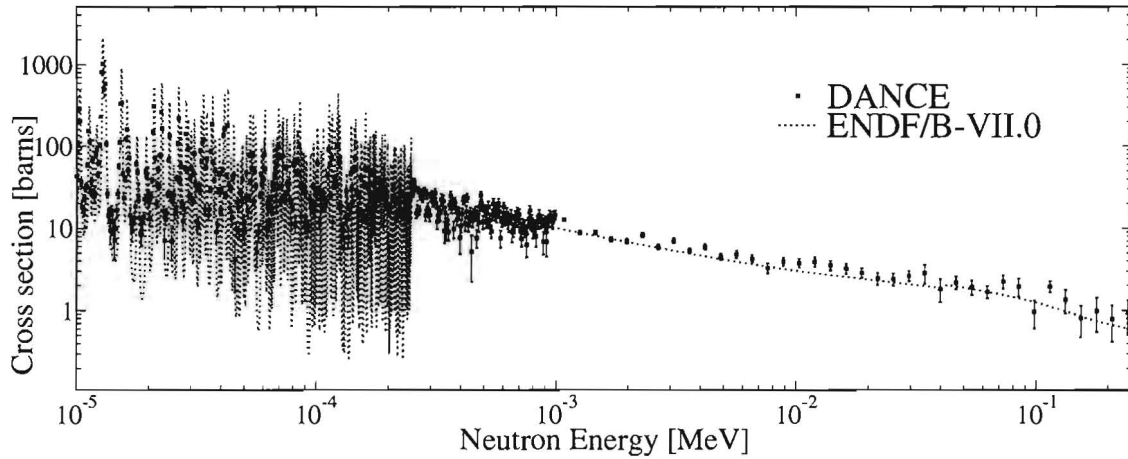


FIGURE 2. Preliminary results on the $^{243}\text{Am}(n,\gamma)$ cross section measured at DANCE.

were obtained also for the resonances below 12 eV and are summarized in Table 1 and 2 of Ref [5].

Preliminary results on the neutron capture cross section of ^{243}Am in the region above 35 eV is shown in Fig 2 together with the ENDF/B-VII.0 evaluation. The DANCE data are normalized to ENDF/B-VII.0 in the region between 100 and 200 eV in neutron energy. High quality of the data is observed. More detailed analysis is underway.

Neutron capture and neutron-induced fission of ^{242m}Am

The first experiment with ^{242m}Am target at DANCE was performed in 2006 [8]. Because of the difficulties during the preparation of very radioactive ^{242m}Am target, the total mass of the sample material was only 47 μg . The experiment was significantly

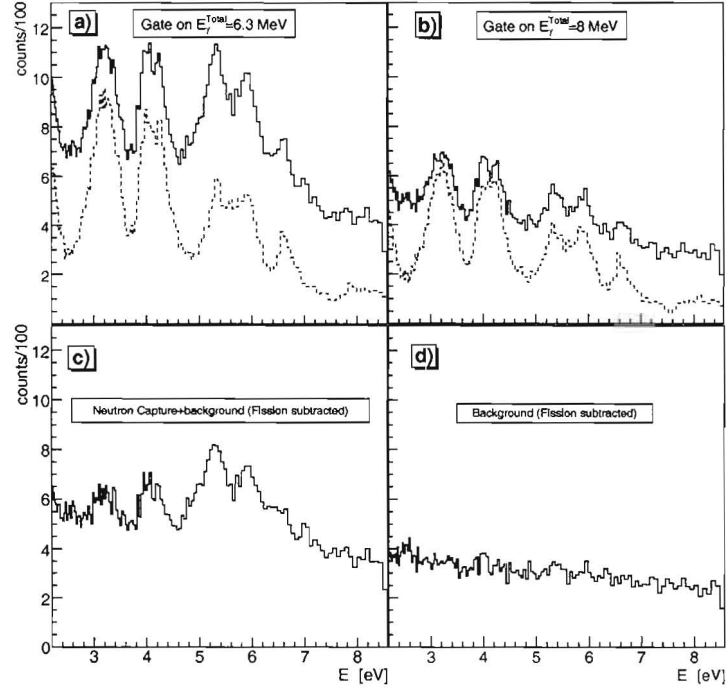


FIGURE 3. Spectra measured at DANCE in the region of neutron energies between 2 and 9 eV for ^{242m}Am target. a) The spectra for $M_\gamma > 2$ are shown for PPAC untaged (solid line) and PPAC tagged events (dashed line), respectively, gated on the total γ -ray energy E_γ^{Total} around the Q value of $^{242m}\text{Am}(n,\gamma)$ reaction 6.3 ± 1 MeV. b) The same as in a) with the gate on E_γ^{Total} around 8.0 ± 1 MeV. c) The PPAC untaged spectrum from a) after the subtraction of the PPAC tagged spectrum from a) normalized to an efficiency of the PPAC. d) The PPAC untaged spectrum obtained in the same manner as in c) for the spectra in b).

complicated by high levels of alpha and γ activity of the target. The further complication for the neutron-capture measurement on ^{242m}Am was the neutron-induced fission. The neutron capture cross section of ^{242m}Am is expected to be two orders of magnitude lower compared to the neutron-induced fission cross section. From experiment on the $47 \mu\text{g}$ sample in 2006, no neutron capture events were identified. However, we were able to extract the neutron-induced fission cross sections and the preliminary results were reported in Ref. [6, 8], where the comparison of our result to other high resolution measurements was shown. In order to obtain the absolute neutron-induced fission cross section of ^{242m}Am , it is required to calibrate the neutron flux and determine the efficiency of neutron-induced fission events. This work is in progress.

The new sample of ^{242m}Am with the total mass of $154 \mu\text{g}$ was prepared in 2007 and the experiment with the PPAC was performed again with the goal to identify the neutron capture events. Fig. 3 describes the procedure of the fission background component subtraction from the PPAC untaged data. Spectra in Fig. 3a and c were gated in the region of the total γ -ray energy of 6.3 ± 1 MeV which corresponds to the Q-value of the (n,γ) reaction. The spectra in Fig. 3b and d were gated on the region of the total γ -ray energy of 8 ± 1 MeV, which is above the Q-value of the (n,γ) reaction where no

capture events should be observed. The PPAC tagged spectra (shown by the dashed lines in Fig. 3a and b) were subtracted from the PPAC untaged spectra (solid lines in Fig. 3a and b) after they were normalized to the PPAC efficiency. The subtracted spectra are shown in Fig. 3c and d. The resonance structure is observed for the spectrum in Fig. 3c which was gated on the Q-value of (n, γ) reaction. The spectrum in Fig. 3d shows no resonance structure, but rather smooth varying character. This fact proves that the fission component from the PPAC untaged spectra were removed properly. Therefore, the resonance structure observed in the spectrum gated on the (n, γ) reaction's Q-value (Fig. 3c) originates from the capture events and provides the first direct observation of neutron capture events in the resonance region between 2 and 9 eV of neutron energy. The work on the subtraction of other smoothly varying background components is in progress, and we expect to obtain the results on neutron capture-to-neutron-induced fission ratio soon.

SUMMARY

The review of the neutron capture and neutron-induced fission measurements with DANCE on americium isotopes was presented. The data on ^{241}Am neutron capture cross section were obtained for the neutron capture cross section for neutron energies from thermal to 320 keV. The preliminary results on ^{243}Am neutron capture cross section were measured for neutron energies from 35 eV to 200 keV. For ^{242m}Am , the first direct evidence of the neutron capture was obtained in the resonance region between 2 and 9 eV of neutron energy. The active neutron induced fission tagging PPAC in the center of the DANCE array was used in this experiment to reduce and characterize the neutron-induced fission component in the neutron capture data.

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