

Neutron capture and (n,2n) measurements on ^{241}Am

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Abstract. We report on a set of neutron-induced reaction measurements on ^{241}Am which are important for nuclear forensics and advanced nuclear reactor design. Neutron capture measurements have been performed on the DANCE detector array at the Los Alamos Neutron Scattering Center (LANSCE). In general, good agreement is found with the most recent data evaluations up to an incident neutron energy of ~ 300 keV where background limits the measurement. Using mono-energetic neutrons produced in the $^2\text{H}(d,n)^3\text{He}$ reaction at the Triangle Universities Nuclear Laboratory (TUNL), we have measured the $^{241}\text{Am}(n,2n)$ excitation function from 7.6 to 14.5 MeV using the activation method. Good agreement is found with previous measurements, with the exception of the three data points reported by Perdikakis et al. around 11 MeV, where we obtain a much lower cross section that is more consistent with theoretical estimates.

1 Introduction

Arising from the β -decay of ^{241}Pu , ^{241}Am can be used as an in-situ nuclear forensic tracer for plutonium-based systems. Neutron capture data on ^{241}Am can provide valuable information about the number of low-energy neutrons that the plutonium has been exposed to, while the (n,2n) reactions can define the number of high energy neutrons. Improved capture measurements on the minor actinides, such as ^{241}Am , are also desired for the Global Nuclear Energy Partnership (GNEP), advanced reactor design, and actinide transmutation. Herein we report on $^{241}\text{Am}(n,\gamma)$ measurements using the DANCE detector array at the Lujan Center/LANSCE and $^{241}\text{Am}(n,2n)$ measurements performed at the TUNL facility.

2 $^{241}\text{Am}(n,\gamma)$ measurements at DANCE

The DANCE (Detector for Advanced Neutron Capture Experiments) is a 160-element BaF_2 4π -array designed to measure neutron capture cross sections on stable and radioactive targets [1]. We have commissioned the detector on a 20.26 m neutron flight path located at the Lujan Center at LANSCE [2, 3]. The neutrons are produced in a moderated tungsten target with a pulsed 800-MeV proton beam that has been extracted from a proton storage ring. For this experiment the ring delivered a 20 Hz beam with a pulse full width of ~ 250 ns at an average current of $100 \mu\text{A}$. The neutron flux at the target position of DANCE is approximately 3×10^5 n/cm²-s-energy decade and falls off with $\sim 1/E_n$ energy dependence above an enhanced thermal bump extending up to 0.3 eV. The neutron flux is

measured with three different neutron monitors that are located ~ 2.4 m downstream of the target. These large-area neutron monitors (^6LiF , BF_3 , and ^{235}U) utilize the $^6\text{Li}(n,t)$, $^{10}\text{B}(n,\alpha)$, and $^{235}\text{U}(n,\text{fission})$ reactions, respectively, and are calibrated against $\text{Au}(n,\gamma)$ resonances measured in DANCE.

Neutron capture in the target of DANCE, induces a cascade of gamma-rays that are detected by the BaF_2 array. Typical gamma-ray multiplicities range from 1–6 with an average around 3.5. The total sum energy for all gamma-rays should equal the Q-value for the capture reaction ($Q = 5.5$ MeV for ^{241}Am) in an ideal detector. With a single gamma-ray efficiency of $\sim 86\%$ and an energy resolution of $\sim 14\%$, DANCE exhibits a total gamma-ray E_{sum} peak that is roughly triangular in shape with the tail extending to lower energies. Gates on the gamma-ray multiplicity ($M_\gamma = 4$), gamma coincidence time (20 ns), and E_{sum} (3.6–6 MeV) have been used to reduce interference from background events and optimize the signal-to-noise ratio. These multiplicity and E_{sum} cuts also reduced interference from fission events whose cross section is small at low energies ($E_n < 300$ keV).

We have modelled the performance of DANCE using GEANT4 with excellent agreement observed between the measured and calculated multiplicity distributions and E_{sum} response [4]. As input to this we have used the DICEBOX code [5] to estimate the gamma-ray cascade for ^{241}Am .

For the actual experiment we prepared a $0.7 \text{ mg/cm}^2 \times 0.7 \text{ cm}$ diameter ^{241}Am target by electrodeposition onto a thin titanium backing. Ti blank target runs were acquired as part of the background subtraction in the data analysis. The fast ($\tau \sim 0.6$ ns) and slow (~ 650 ns) scintillation light produced the BaF_2 crystals by gamma-rays were viewed by photomultiplier tubes and the resulting electronic signals were recorded using 500 MHz, 8-bit flash ADCs which are readout using a MIDAS data acquisition system. More information on the data acquisition and analysis system is given in ref. [6].

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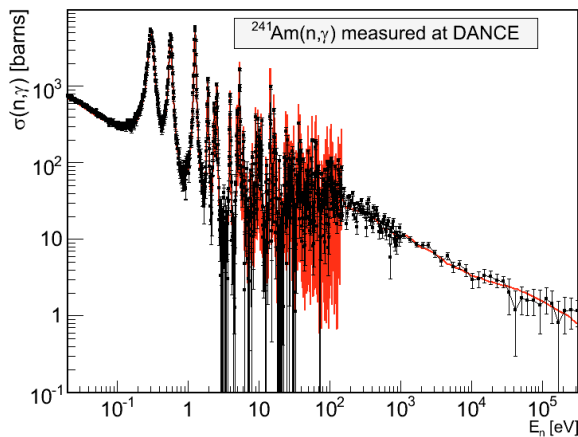


Fig. 1. $^{241}\text{Am}(n,\gamma)$ preliminary data obtained with the DANCE detector array at LANSCE. The ENDF-B/VII evaluation is shown in red.

The preliminary results of a 17-day run on ^{241}Am are shown in figure 1. As we are still working to put these measurements on an absolute scale, these data have been normalized to the resonance at 1.2 eV or by the resonance integral between 17–140 eV, depending on the type of data that was acquired (segmented mode covering a wide energy range or continuous mode covering a narrower energy range – see ref. [6] for details). Good agreement in the overlap region covered by both data collection modes was found. These data have been dead-time corrected.

Excellent agreement is found when these data are compared with the latest ENDF-B/VII, JENDL-3.3, and JEFF-3.1 (n,γ) data evaluations [7] with one exception: the resonance at 10.4 eV is missing from the ENDF/B-VII evaluation (see fig. 2). The good agreement extends up to 300 keV where the measurements become limited by background. In this higher energy region, we have increased the energy bin size over which the cross section is averaged (up to $\Delta E_n/E_n = 20\%$) to maintain reasonable counting statistics. We are presently in the process of finalizing this data to put them on an absolute cross section basis for publication.

3 $^{241}\text{Am}(n,2n)$ measurements at TUNL

The excitation function for the $^{241}\text{Am}(n,2n)$ reaction was measured from 7.6 to 14.5 MeV using the activation technique. Three stippled ^{241}Am targets of $\sim 1 \text{ mg/cm}^2 \times 1 \text{ cm}$ diameter on platinum were prepared at Los Alamos. Six additional targets of the same dimension were prepared at Livermore after further chemical purification of the americium followed by electrodeposition onto platinum. These targets were then sent to the Triangle University Nuclear Laboratory (TUNL) where they were irradiated with monoenergetic neutrons produced by the $^2\text{H}(d,n)^3\text{He}$ reaction using the 10 MV FN Tandem Van de Graff accelerator. A 2- μA deuteron beam on a 3 cm long deuterium gas cell pressurized to 3 atm produced an average neutron flux at the target position (4 cm from the center of the gas cell) of $\sim 3 \times 10^7 \text{ n/cm}^2\text{-s}$. Given the production kinematics

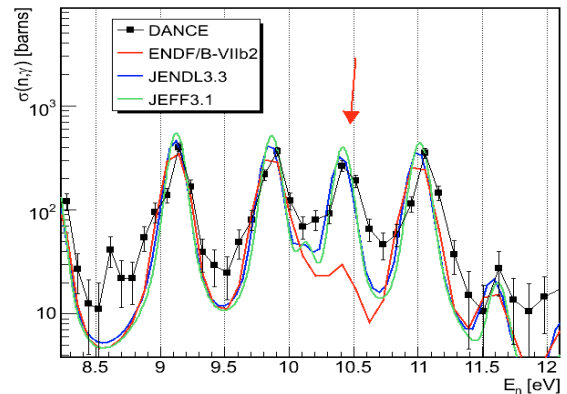


Fig. 2. An enlarged energy region of figure 1 showing the resonance at 10.4 eV that is not included in the ENDF-B/VII evaluation. Preliminary DANCE data are given in black, ENDF/B-VII (red), JENDL-3.3 (blue) and JEFF-3.1 (green).

and this extended cell – target geometry, the energy resolution of the neutron beam is estimated to be 80–280 keV (FWHM) depending on the neutron energy.

To monitor the neutron fluence for each irradiation a packet of 1 cm diameter Al, Ni, and Au foils was placed directly upstream and downstream of the ^{241}Am target. The following monitor reactions were used $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$, $^{58}\text{Ni}(n,p)^{58}\text{Cu}$, $^{58}\text{Ni}(n,2n)^{57}\text{Ni}$, and $^{197}\text{Au}(n,2n)^{196}\text{Au}$. During the activation, the relative neutron flux history was measured using three BF_3 detectors located 4.77 m from the gas cell at angles of 0° (coaxial with the target) and $\pm 10^\circ$. Neutron flux fluctuation, including beam on/off corrections, were applied to all of the activation products measured. Depending on the expected $^{241}\text{Am}(n,2n)$ cross section, the irradiation period ranged from 24 to 72 hours.

After each irradiation the target assembly was dismantled so that the front and back monitor foil stacks could be counted separately from the ^{241}Am target in the same counting geometry. 60% HPGe detectors combined with an automatic Canberra Multiport II multichannel analyzer data acquisition system were used to count each monitor foil pack and the americium target offline. To attenuate the strong 59.5 keV gamma-rays emitted by the ^{241}Am target, 3 mm thick lead was placed directly in front and behind each target before counting. This attenuated the 59.5 keV gamma-rays by $>10^7$ while only reducing the 888.8 keV and 987.8 keV gamma rays associated with the decay of the $(n,2n)$ reaction product ^{240}Am (50.8 h) by $\sim 25\%$. Each Am target and monitor foil pack was counted for several days to follow their decay. The HPGe detectors were calibrated using a set of NIST-traceable gamma-ray (both point-like and 1 cm extended diameter) standards.

In figure 3 we show the high-energy portion of gamma-ray spectra collected after irradiating stippled and electroplated ^{241}Am targets. The stippled targets contained a ^{22}Na contaminant which accounts for the increased background and Compton edge observed at $\sim 1060 \text{ keV}$. The electroplated targets were free of this contaminant. Many of the other gamma-rays observed are due to weak lines fed in the decay

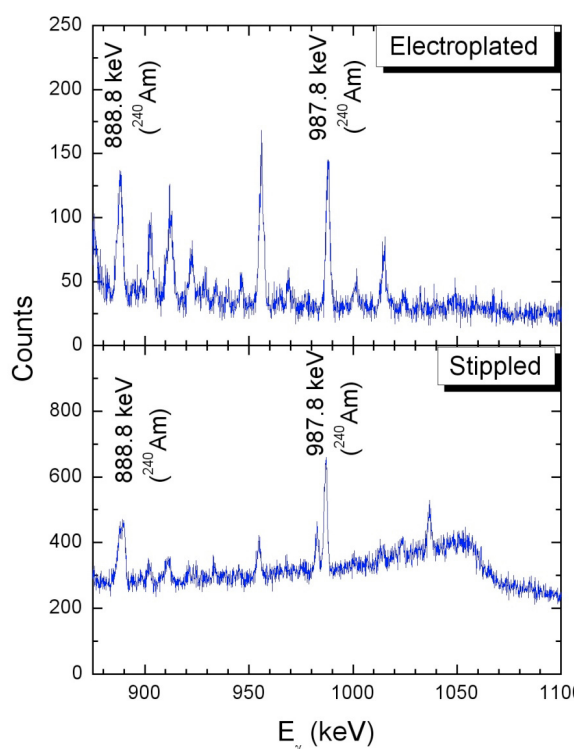


Fig. 3. High-energy portion of gamma-ray spectra collected for a stippled (bottom) and electroplated (top) ^{241}Am targets that were irradiated with 14 MeV neutrons. Gamma lines at 888.8 and 987.8 keV associated with the decay of ^{240}Am are noted.

of ^{241}Am or from activation of the platinum backing. By tracking the decay of the 888.8 keV line, we determined that this line was partial contaminated by a weak unresolved line from ^{241}Am at 887.3 keV, so in this analysis we have used only the 987.8 keV counting results.

Using the 987.8 keV ^{240}Am counting data with the calibrated gamma-ray efficiency, neutron fluence determination obtained from the monitor foil counting results and ENDF/B-VII monitor reaction cross sections, target thickness assay, on/off beam corrections, and decay scheme information (branching ratio and half-lives) given in ref. [8], we have calculated the cross section for each irradiation. The preliminary cross section data are shown in figure 4. Excellent agreement with the Hauser-Feshbach model calculations reported in the ENDF/B-VII and JENDL-3.3 data evaluations [7] is found.

In figure 5 we contrast these preliminary results with previous measurements reported near 14 MeV by Gancarz [9], Filatenkov et al. [10], Loughed et al. [11], and some recent data at lower energies by Perdikakis et al. [12]. The data agree within the reported errors in this energy region. Although we do not expect that deuteron breakup reaction to significantly change our cross sections in the 13-14.5 MeV region, we have collected neutron time-of-flight deuteron breakup data at TUNL to quantify and make a correction for this effect.

For the data at lower neutron energies, we disagree with three data points reported by Perdikakis et al. in the 11-MeV region, but agree with their findings at 8.8 and 9.6 MeV. We have double checked our measurements and

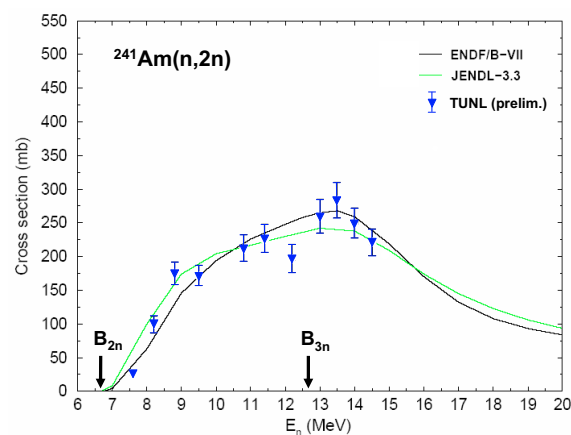


Fig. 4. Preliminary cross sections for the $^{241}\text{Am}(n,2n)$ reaction as measured at TUNL (blue) are shown with the ENDF/B-VII (black) and JENDL-3.3 (green) data evaluations. Given as B_{2n} and B_{3n} are the threshold energies for the (n,2n) and (n,3n) reactions, respectively.

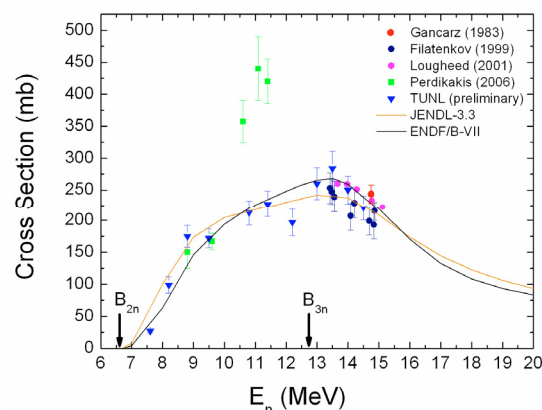


Fig. 5. Cross section measurements for the $^{241}\text{Am}(n,2n)$ reaction by Gancarz (red), Filatenkov (dark blue), Loughed (pink), Perdikakis (green), and our preliminary TUNL results (light blue) compared to JENDL-3.3 (yellow) and ENDF/B-VII (black) data evaluations.

report them here with confidence. As noted earlier, our measurements agree well with the ENDF/B-VII and JENDL-3.3 data evaluations.

4 Summary

We report on the neutron capture cross section of ^{241}Am measured from thermal to ~ 300 keV energies using the DANCE detector array at LANSCE. With the exception of a resonance at 10.4 eV missed in ENDF/B-VII, excellent agreement is found between these preliminary results and current data evaluations.

The $^{241}\text{Am}(n,2n)$ cross section has been measured from $E_n = 7.6$ to 14.5 MeV using the activation technique at TUNL. Good agreement with previous measurements is found with the exception of three data points near $E_n = 11$ MeV that have recently been reported by Perdikakis et al. Outstanding

agreement between our preliminary results and the Hauser-Feshbach model calculations reported in ENDF/B-VII and JENDL-3.3 is highlighted. We are proceeding to finalize these data and submit them for publication.

We acknowledge the outstanding support of the operating staffs at the Los Alamos Neutron Scattering Center and the Triangle Universities Nuclear Laboratory who made these measurements possible. This research is supported in part by Laboratory Directed Research and Development at Los Alamos National Laboratory, the National Nuclear Security Agency (NNSA), Office of Research and Engineering (NA-22), and by the NNSA Stewardship Science Academic Alliance program through research Grant No. DE-FG03-02NA00057.E. Work performed under the auspices of the US Department of Energy at Los Alamos National Laboratory by the Los Alamos National Security, LLC under Contract No. DE-AC52-06NA25396 and at Lawrence Livermore National Laboratory under Contract No. W-7405-ENG-48.

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