UCRL-CONF-232955

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July 23, 2007

International Conference on Nuclear Data for Science and Technology 2007
Nice, France
April 22, 2007 through April 27, 2007

International Conference on Nuclear Data for Science and Technology 2007
DOI:

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}H(d,n)^{3}He reaction at Triangle University Nuclear Laboratory (TUNL), we have measured the ^{241}Am(n,2n) excitation function from threshold (6.7 MeV) 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 the ^{241}Am(n,γ) measurements using the DANCE detector array at the Lujan Center/LANSCE and ^{241}Am(n,2n) measurements performed at the TUNL facility.

2 ^{241}Am(n,γ) 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 width of ~200 ns at an average current of 100 μA. The neutron flux at the target position of DANCE is approximately 3 x 10^5 n/cm^2-s-energy decade and falls off with ~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 (^{6}LiF, BF_3, and ^{235}U) utilize the ^{6}Li(n,t), ^{10}B(n,α), and ^{235}U(n,fission) reactions, respectively, and are calibrated against Au(n,γ) 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 ~86% and an energy resolution of ~14%, DANCE exhibits a total gamma-ray E_{sum} peak that is triangular in shape with the tail extending to lower energies. Gates on the gamma-ray multiplicity (M_γ=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

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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 have prepared a 0.7 mg/cm^2 x 0.7 cm diameter ^{241}Am target by electrodeposition onto a thin titanium backing. Ti blank target runs where also acquired and subtract out in the analysis. The fast (τ~0.6 ns) – slow (600 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].

The preliminary results of a 17-day run on ^{241}Am are shown in fig. 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 the 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.

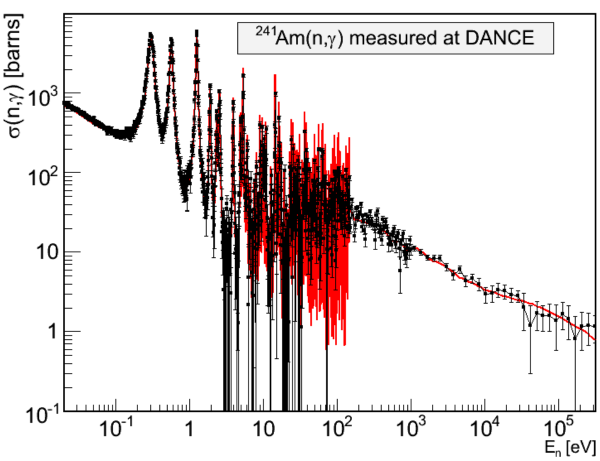

Fig. 1. ^{241}Am(n,γ) preliminary data obtained with the DANCE detector array at LANSCE. The ENDF-B/VII evaluation is shown in red.

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 average (up to $\Delta E_n/E_n$=20%) to maintain reasonable errors. We are presently in the process of finalizing this data to put them on an absolute cross section basis for publication.

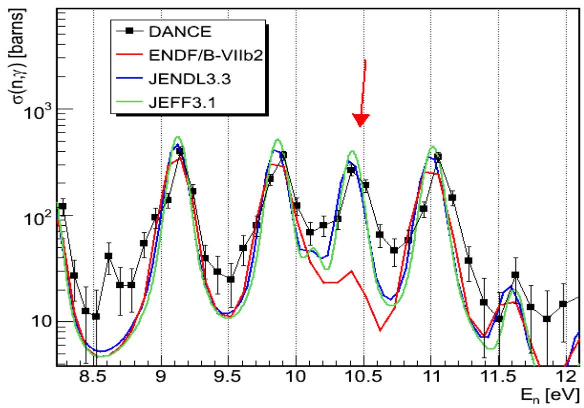

Fig. 2. An enlarged energy region of fig. 1 showing the resonance at 10.4 eV that is not included in the ENDF-B/VII evaluation. Preliminary DANCE data is given in black, ENDF/B-VII (red), JENDL-3.3 (blue) and JEFF-3.1 (green).

3 ^{241}Am(n,2n) Measurements at TUNL

The excitation function for the ^{241}Am(n,2n) reaction was measured from 7.6 to 14.5 MeV using the activation technique. Three stippled ^{241}Am targets of ~1 mg/cm2 x 1 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}H(d,n)^{3}He 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 at an average neutron flux at the target position (4 cm from the center of the gas cell) of ~3 x 10^7 n/cm^2-s. Given the production kinematics and this extended cell – target geometry, the energy resolution of the neutron beam is estimated to be 160-300 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 place directly upstream and downstream of the ^{241}Am target. The following monitor reactions were used ^{27}Al(n,α)^{24}Na, ^{58}Ni(n,p)^{58}Cu, ^{58}Ni(n,2n)^{57}Ni, and ^{197}Au(n,2n)^{196}Au. During the activation, the relative neutron flux history was measured using BF_3 detectors located 4.77 m from the gas cell at angles of 0^o (coaxial with the target) and at ☛10^o. Growth and decay beam on/off corrections were applied to all of the activation products measured. Depending on the expected ^{241}Am(n,2n) cross section, the irradiation period ranged from 24 to 72 hours.

After each irradiation the target assembly was dismounted so that the front and back monitor foil stacks could be counted separately from the ^{241}Am target. 60% HPGe detectors combined with an automatic Canberra Multiport II multichannel analyzer data acquisition system was used to count each monitor foil pack and the americium target offline. To attenuate the strong 59.5 keV gamma-rays emitted by ^{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 ~25%. Each target and monitor foil pack were 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 fig. 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 ~1060 keV. The electroplated targets were clean. Many of the other gamma-rays observed are due to weak lines feed in the decay 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.

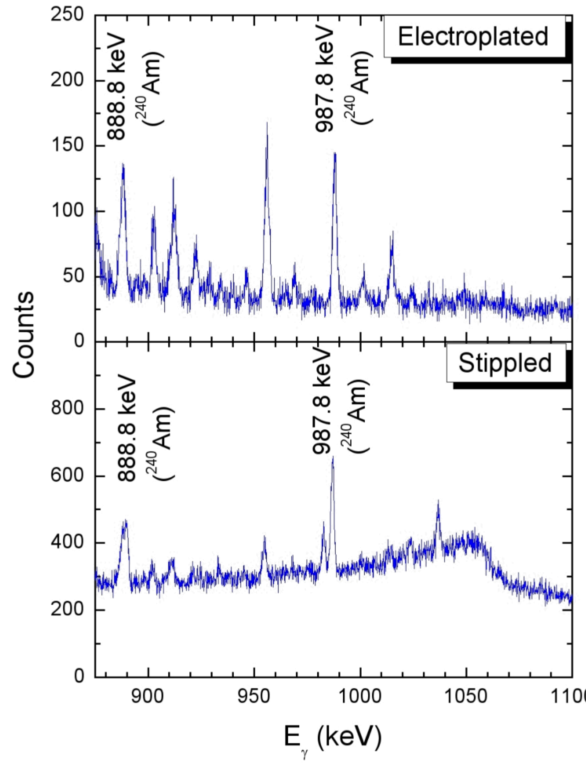

Fig. 3. High-energy portion of gamma-ray spectra collected for a stippled (bottom) or electroplated (top) ^{241}Am targets that have been irradiated with 14 MeV neutrons. Gamma lines at 888.8 and 987.8 keV associated with the decay of ^{240}Am are noted.

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 fig. 4. Excellent agreement with the Hauser-Feshbach model calculations reported in the ENDF/B-VII and JENDL-3.3 data evaluations [7] is found.

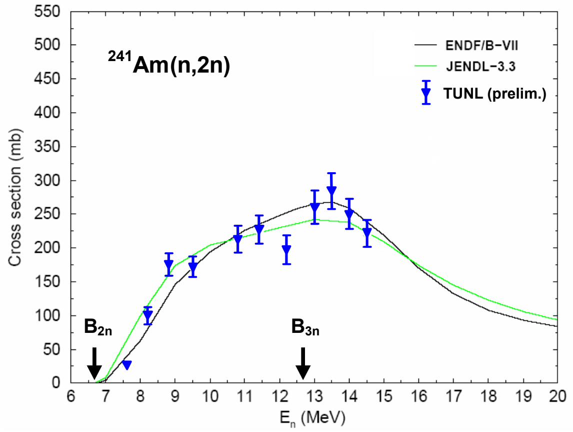

Fig. 4. Preliminary cross sections for the ^{241}Am(n,2n) reaction as measured at TUNL (blue) are shown with the ENDF/B-VII (black) and JENDL-3.3 (green) data evaluations. Given as B_{2n} and B_{3n} are the threshold energies for the (n,2n) and (n,3n) reactions, respectively.

In fig. 5 we contrast these preliminary results with previous measurements reported near 14 MeV by Gancarz [9], Filatenkov *et al.* [10], Lougheed *et al.* [11], and some recent data at lower energies by Perdikakis *et al.* [12]. The data agrees 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.

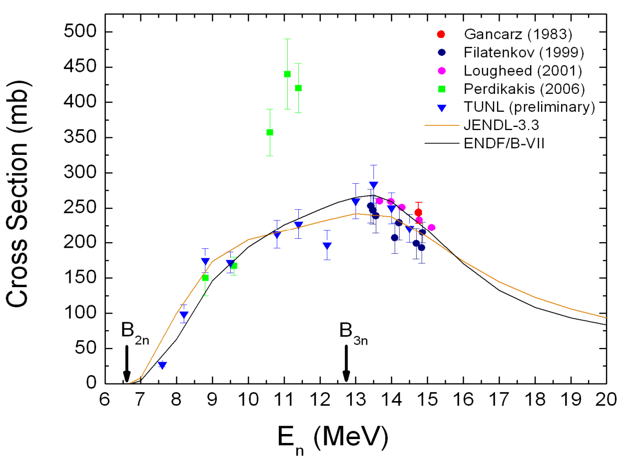

Fig. 5. Cross section measurements for the ^{241}Am(n,2n) reaction by Gancarz (red), Filatenkov (dark blue), Lougheed (pink), Perdikakis (green), and our preliminary TUNL results (light blue) compared to JENDL-3.3 (yellow) and ENDF/B-VII (black) data evaluations.

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 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 on ^{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 the ENDF/B-VII, excellent agreement is found between these preliminary results and current data evaluations.

The ^{241}Am(n,2n) cross section has been measured from 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 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 operating staff at the Los Alamos Neutron Scattering Center and the Triangle University 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. Work performed under the auspices of the U.S. Department of Energy at Los Alamos National Laboratory by the Los Alamos National Security, LLC under Contact No. DE-AC52-06NA25396 and at Lawrence Livermore National Laboratory by the University of California, under Contract No. W-7405-ENG-48.

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