International Conference on Nuclear Data for Science and Technology 2007
DOI: 10.1051/ndata:07592

Recent actinide nuclear data efforts with the DANCE 4π BaF_2 array

T.A. Bredeweg[1,a], U. Agvaanluvsan[2], J.A. Becker[2], E.M. Bond[1], A.J. Couture[1], T. Ethvignot[3], J.R. Fitzpatrick[1], M.M. Fowler[1], T. Granier[3], R.C. Haight[1], T.S. Hill[1], M. Jandel[1], R.A. Macri[2], J.M. O'Donnell[1], W.E. Parker[2], R. Reifarth[1], R.S. Rundberg[1], A.K. Slemmons[1], F.K. Tovesson[1], J.L. Ullmann[1], D.J. Vieira[1], J.B. Wilhelmy[1], P.A. Wilk[2], J.M. Wouters[1], and C.Y. Wu[2]

[1] Los Alamos National Laboratory, Los Alamos, NM 87545, USA
[2] Lawrence Livermore National Laboratory, Livermore, CA 94550, USA
[3] CEA-DAM, BP. 12, 91680 Bruyères-le-Châtel, France

Abstract. Much of the recent work in the DANCE collaboration has focused on nuclides of interest to stockpile stewardship, attribution science and the advanced fuel cycle initiative. As an example, we have recently begun a program to produce high precision measurements of the key production and destruction reactions of important nuclear fuel elements and radiochemical diagnostic isotopes. The neutron capture targets that have been fielded under this program include several isotopes of uranium, plutonium and americium. However, neutron capture measurements on many of the actinides are complicated by the presence of γ-rays arising from low energy neutron-induced fission. To overcome this difficulty we have designed and implemented a dual parallel-plate avalanche counter fission-tagging detector. This design provides a high efficiency for detecting fission fragments and is self-contained to allow loading of pre-assembled target/detector assemblies into the neutron beam line at DANCE. An outline of the recent experimental program will be presented as well as preliminary results from neutron capture measurements on ^{234,235,236}U. Planned measurements on ^{238,239}Pu will also be discussed.

1 Introduction

Much of our current nuclear data efforts have focused on neutron capture cross section measurements on actinides pertinent to the stockpile stewardship and nuclear forensics programs. These same cross sections are of great interest to the Advanced Fuel Cycle Initiative (AFCI) and the new, much broader Global Nuclear Energy Partnership (GNEP). Unfortunately, measurement of the neutron capture cross sections for many of the actinides is complicated by the presence of low energy neutron-induced fission. Efforts to separate prompt γ-rays arising from fission from those following capture, based on γ-ray multiplicity and summed γ-ray energy, have resulted in only partial success, and only by using an existing evaluation of the fission cross section for the isotope of interest. The addition of a fission-tagging detector around the target can be used to characterize the γ-ray spectrum arising from fission. The characterization of the fission component can then be used to perform a "background" subtraction, corrected for the efficiency of the fission-tagging detector, to enable good separation of the capture events.

2 DANCE and fission-tagging

The Detector for Advanced Neutron Capture Experiments (DANCE) is a 160 element γ-ray calorimeter with high granularity and nearly 4π coverage coupled to state-of-the-art data acquisition electronics [1,2]. The primary purpose of the array is to perform neutron capture measurements on small (one milligram or less) and/or radioactive (activity $\leq$ 1 Ci, $t_{1/2} \geq$ 100 days) nuclides. The DANCE array is located on flight-path 14 of the Manuel Lujan Jr. Neutron Scattering Center of the Los Alamos Neutron Science Center (LANSCE). Neutrons are produced through spallation reactions of 800 MeV protons impinging on a tungsten neutron production target. The proton pulse width at the production target is approximately 125 ns full-width at half maximum with a repetition rate of 20 Hz. Flight-path 14 views a partially coupled light water moderator that produces a neutron spectrum that peaks near thermal energies and drops off as $1/E_n$ above approximately 1 eV [3,4]. Neutron energies are determined by time-of-flight relative to the proton pulse arrival at the production target. Calibration of neutron energy from the measured time-of-flight is performed by analyzing neutron capture resonances in a thin (2 mg/cm^2) gold target.

The PPAC fission-tagging detector is a dual parallel-plate avalanche counter which is operated in transmission mode [5]. A photograph of the PPAC fission-tagging detector and removable target/cathode foil is provided in figure 1. The actinide sample is electro-deposited directly on the center cathode foil, which is mounted on a G10 fiberglass frame. Fission fragments are detected in two 3.2 mm low pressure gas regions on either side of the target/cathode foil. Isobutane is used for the fill gas with an operating pressure of 6–10 Torr. Signals are collected independently from the two anode foils. Each signal is amplified through two onboard MAR-6 monolithic microwave IC amplifiers[1] or one onboard MAR-6 amplifier and an external A81-1 amplifier[2] located approximately one meter from the detector. The later configuration was developed to eliminate feedback oscillations

[a] Presenting author, e-mail: toddb@lanl.gov

[1] Mini-circuits RF/IF Microwave Components.
[2] M/A-COM Inc. RF/Microwave Products.

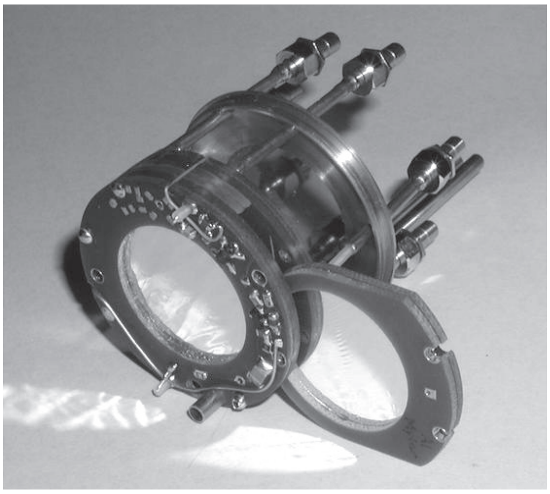

Fig. 1. Picture of a PPAC fission-tagging detector and the removable target/cathode foil partially inserted. The surface mount components for the first amplification stage can be seen on the front ring. The neutron beam passes through a 2.5 cm opening in the center of the G10 frame.

between the cascaded MAR-6 amplifiers when run at high gain. Semi-rigid coax cables are used to carry the detector bias, test inputs and output signals through the beam pipe to a break-out box located just downstream of the DANCE array. Similarly, detector gas supply and return are carried through 0.1 inch I.D. stainless steel tubing running between the detector and break-out box. An external gas handling system, with LabVIEW[3] based monitoring and control capabilities, controls gas flow and pressure. All foils located in the neutron flight path are made of thin metallized Mylar (400–550 μg/cm^2 total thickness) to limit the background from capture on these materials and scattered neutrons that are then captured in the BaF_2 crystals. The removable target/cathode foil allows for reuse of a single detector for several targets and associated background blanks. This design also allows easy recovery of rare or otherwise valuable sample material for use in other experiments. The relatively low cost of this design also allows for dedicated detectors to be used to limit handling requirements for high activity samples such as ^{238}Pu and ^{242m}Am [6]. The decision to use fission-tagging for a particular actinide will depend on the strength of the fission channel relative to capture and the desired level of accuracy for the measurement. For several of the threshold fissioners, such as ^{241}Am and ^{243}Am [7], the fission cross section is two orders of magnitude or more below the the capture cross section in the neutron energy region available for flight-path 14 (thermal to ≈500 keV). In this case fission-tagging might not be necessary for a 5% measurement, but would be required for a 1% measurement. The efficiency of the fission-tagging detector can be empirically determined from the data by comparing 2D histograms of cluster multiplicity[4] versus summed γ-ray energy in the DANCE array for tagged and untagged events.

[3] National Instruments Corp.

[4] A cluster is a group of adjacent DANCE crystals registering simultaneous hits within a small coincidence window, usually ~10–20 ns. Combined data analysis and GEANT4 simulations indicate that the DANCE cluster multiplicity closely tracks the actual γ-ray multiplicity of the γ cascade [8].

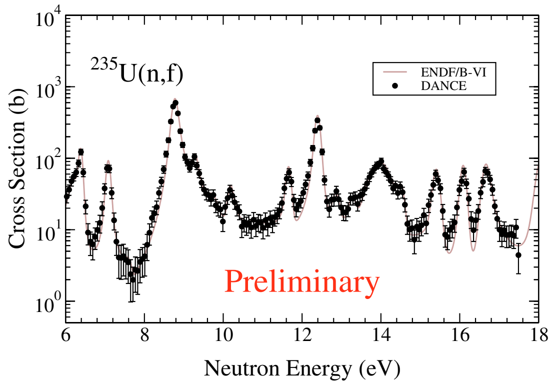

Fig. 2. Preliminary results for tagged events in the ^{235}U measurement. The data are corrected for detection efficiency and normalized to sample size and total neutron fluence. The deduced cross sections are in good agreement with the current ENDF/B-VII [9] evaluation for neutron-induced fission of ^{235}U.

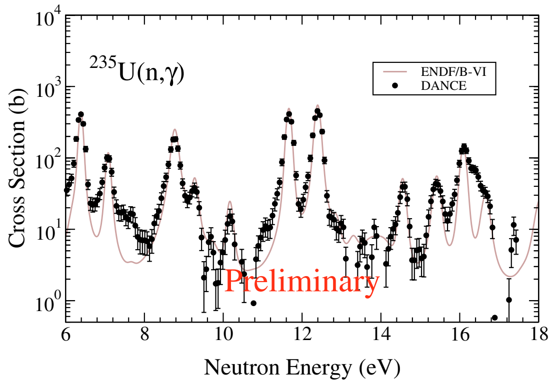

Fig. 3. Preliminary results for untagged events in the ^{235}U measurement. The points represent untagged DANCE data less the remaining fission contribution and background as determined by a blank run and corrected for detection efficiency.

3 Uranium neutron capture cross sections

For the uranium isotopes of most interest ($^{232-238}$U) three have sufficient neutron-induced fission cross sections below $E_n \approx 500$ keV to warrant the use of fission-tagging, ^{232}U, ^{233}U and ^{235}U. To date we have performed measurements on three of the uranium isotopes: ^{234,236}U without fission-tagging and ^{235}U with (and without) fission-tagging.

3.1 ^{235}U(n,γ) cross section and capture-to-fission ratio

A great amount of experimental work that has been done on ^{235}U. This makes this isotope an ideal choice for testing the capabilities of the fission-tagging PPAC. Initial data were collected in 2005 to test the response of the PPAC and to determine optimum running conditions (gas pressure and flow rate, bias on the cathode and anode foils and amplifier configuration) [5]. Production data were taken in 2006 to measure the neutron capture cross section and the capture-to-fission ratio

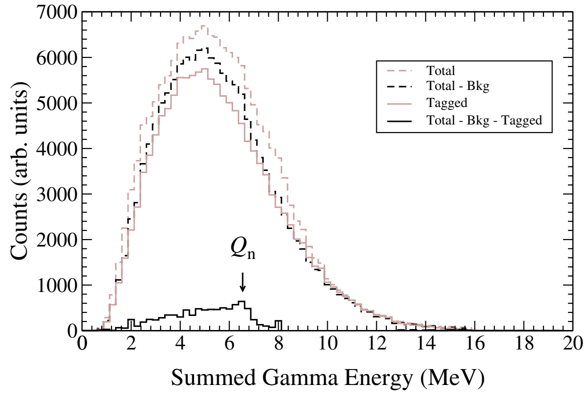

Fig. 4. Summed γ-ray energy in the DANCE array for thermal neutrons on ^{235}U with fission-tagging. The dashed gray line represents the distribution for total events (tagged + untagged + background), the dashed black line is for total events minus background, the solid gray line shows only tagged events and the solid black line shows total events minus tagged events minus background. The arrow indicates the Q-value (6.5 MeV) for the reaction $^{235}U(n,\gamma)^{236}U$.

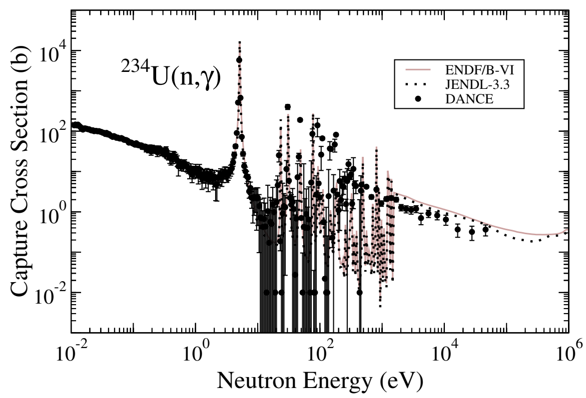

Fig. 5. Preliminary neutron capture cross section for ^{234}U. The solid line is the ENDF/B-VI evaluated cross section, the dotted line is the JENDL-3.3 [11] evaluated cross section and the points are measured cross sections using the DANCE array.

($\alpha = \sigma_\gamma/\sigma_f$). The target consisted of 460 μg of 99.89% ^{235}U electro-deposited directly onto the Mylar cathode foil that was flashed with 2500 Å of titanium on the deposit side and 1000 Å on the opposite side before electro-deposition. The ^{235}U deposit was 0.7 cm diameter (~1.2 mg/cm^2), which is smaller than the 1.0 cm diameter of the neutron beam at the DANCE target position. The uranium deposit was covered by a second titanium flashed Mylar foil to complete the target/cathode assembly. The total thickness of each titanium flashed Mylar foil was approximately 550 μg/cm^2. The isotopic composition of the target material will be determined by mass spectroscopy before the final data are published.

In the off-line analysis the data were segregated into two categories: tagged, in which γ-rays were detected in the DANCE array in coincidence with one or two fission fragments detected in the PPAC, and untagged, in which there was no coincidence with the PPAC. The coincidence window was usaually 10–20 ns. The tagged (fission) events proved to be

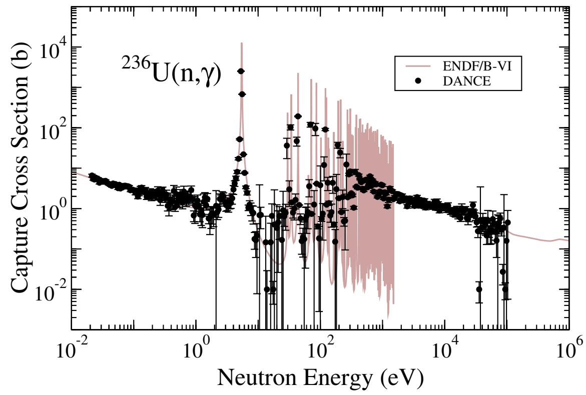

Fig. 6. Preliminary neutron capture cross section for ^{236}U. The solid line is the ENDF/B-VI evaluated cross section and the points are measured cross sections using the DANCE array.

very clean with almost no discernable background component. Correcting for PPAC detector efficiency and normalizing to sample size and total neutron fluence yielded the preliminary result shown in figure 2. The only additional requirement placed on the DANCE data was cluster multiplicities ≥2. The agreement between the DANCE results and the ENDF/B-VI evaluation is quite good. A comparison of total counts for high multiplicity events, as described previously, indicates the PPAC detection efficiency for uranium data ~80%. Our calculations indicate that this is close to the geometry limit expected.

The situation is not so straightforward for the untagged data. This portion of the data contains neutron capture events, the remaining 20% of the neutron-induced fission events that were not tagged and all background events. The situation is exacerbated by the fact that, despite our best efforts the background does increase when the PPAC is installed. The signal-to-noise ratio decreases by a factor of three on average with the PPAC compared to without the PPAC. This can be seen in figure 3 which is the deduced capture cross section starting with all untagged events, subtracting the remaining 20% of the fission contribution and finally subtracting the background as determined by a blank run. The relatively poor results in the valleys between resonances is a clear indication of additional background issues.

Despite the issues with increased background from the PPAC detector it is easy to illustrate the need for fission-tagging. Figure 4 shows the distribution of summed γ-ray energies as determined with the DANCE array for thermal neutrons on the ^{235}U target. The dashed gray line represents the distribution for all events irrespective of fission-tagging. The dashed black line is the distribution after subtraction of background events, while the solid gray line is for tagged events only. The capture events are what remains after subtracting the tagged fission events, the remaining untagged fission events (~20%) and the background, and are represented by the solid black line. The ENDF/B-VII thermal capture cross section is approximately 16.9% of the corresponding fission cross section. Integrating over the fission (solid gray line) and capture (solid black line) distributions in figure 4, and roughly accounting for the different detection efficiencies for capture and fission events, leads to a result of ~15% for the

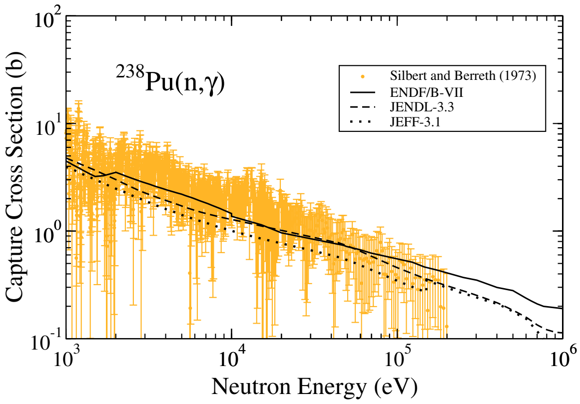

Fig. 7. Neutron capture cross section for ^{238}Pu. The solid and dashed lines are evaluated neutron capture cross sections from the ENDF/B-VII, JENDL-3.3 [11] and JEFF-3.1 [12] libraries. The points are experimental data from Silbert and Berreth [13].

Table 1. Isotopic composition of the ^{234}U and ^{236}U targets.

Isotope	^{234}U isotopics	^{236}U isotopics
^{233}U	0.007%	0.082%
^{234}U	99.39%	–
^{235}U	0.348%	0.205%
^{236}U	0.091%	99.68%
^{238}U	0.166%	0.034%

DANCE data. However, this figure also illustrates that without fission-tagging it would be impossible to extract the capture component from the fission "background". The end result is that any target that requires fission-tagging must be run both with and without fission-tagging. The measurement with fission-tagging is used to characterize the fission spectrum in the DANCE array which is then used to subtract out fission from a second measurement without the PPAC.

3.2 ^{234,236}U(n,γ) cross sections

Neutron capture measurements were made on the two threshold fissioners ^{234}U and ^{236}U soon after the commisioning of the DANCE array [10]. It was at this time that we determined the necessity of developing a fission-tagging capability. Each sample contained small isotopic concentrations of ^{235}U in the amounts shown in table 1. Even this small amount of the highly fissionable isotope introduced a nontrivial fission background component. So, although neither of these measurements required fission-tagging directly, the tagged data for ^{235}U, discussed above, was used to subtract out the contribution from the small amount of ^{235}U present in the two samples. The final data reduction is nearly completed, and preliminary results are presented in figures 5 and 6. The results for ^{236}U are consistent with the ENDF/B-VI evaluation. The results for ^{234}U are as much as 20% lower than the ENDF/B-VI evaluation in the keV region. The DANCE results for ^{234}U(n,γ) have been incorporated into the latest ENDF/B-VII evaluation [9].

4 ^{238,239}Pu(n,γ) cross sections

Both of these isotopes are of great interest to the AFCI and GNEP programs as well as for stockpile stewardship. There are currently discrepancies of 40–50% among the various evaluations for neutron capture on ^{238}Pu in the keV region, as shown in figure 7. Furthermore, the current evaluations for neutron capture on plutonium isotopes in general are based on older experimental data that suffer from various real and/or perceived deficiencies and systematic uncertainties due primarily to technological limitations of the time. Our current program schedule includes a 30 day measurement on ^{239}Pu during the 2007 calendar year. The greater safety considerations associated with ^{238}Pu have forced us to hold off on this measurement until the 2008 calendar year. Because of the relative strength of the neutron-induced fission cross sections these measurements will require the use of fission-tagging.

This work was performed under the auspices of the US Department of Energy by the University of California (under contracts W-7405-ENG-36 [LANL] and W-7405-ENG-48 [LLNL]) and Los Alamos National Security, LLC (under contract DE-AC52-06NA25396 [LANL]). This work benefited from use of the Los Alamos Neutron Science Center.

References

1. J. Wouters et al., *Acquisition-analysis system for the DANCE (Detector for Advanced Neutron Capture Experiments) BaF_2 γ-ray calorimeter*, in *Proc. of the 14th Conference On Real Time (RT2005), Stockholm, Sweden, June 4-10, 2005* (published in IEEE Trans. Nucl. Sci. 53, 2006), pp. 880–885.
2. T. Bredeweg et al. (to be published).
3. G. Muhrer et al., Nucl. Instrum. Meth. A **527**, 531 (2004).
4. G. Muhrer et al., Nucl. Instrum. Meth. A **547**, 555 (2005).
5. T. Bredeweg et al. (to be published).
6. W. Parker et al. (these proceedings).
7. D. Vieira et al. (these proceedings).
8. M. Jandel et al., *GEANT4 simulation of the DANCE array*, in *Proc. of the 19th International Conference on the Application of Accelerators in Research and Industry, Fort Worth, Texas, August 20-25, 2006* (to be published in Nucl. Instrum. Meth. B, 2007).
9. M. Chadwick, P. Obložinský, M. Herman et al., Nucl. Data Sheets **107**(12), 2931 (2006).
10. R. Rundberg et al. (to be published).
11. K. Shibata et al., J. Nucl. Sci. Technol. **39**, 1125 (2002).
12. A. Koning et al., *Status of the JEFF Nuclear Data Library*, in *Proceedings of the International Conference on Nuclear Data for Science and Technology, Santa Fe, 2004* (2004), p. 177.
13. M. Silbert, J. Berreth, Nucl. Sci. Eng. **52**, 187 (1973).