NEUTRON CAPTURE MEASUREMENTS ON PLUTONIUM ISOTOPES FOR GNEP

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A measurement campaign for neutron capture on plutonium isotopes is underway using the Detector for Advanced Neutron Capture Experiments (DANCE) array as part of a data effort for the Advanced Fuel Cycle Initiative and the Global Nuclear Energy Partnership. Measurements have been completed for capture on ^{240}Pu and ^{242}Pu. This represents the first measurement from thermal up to several hundred keV for both isotopes. The cross-section measurements are normalized to the thermal ENDF/B-VI.8 value. The ^{240}Pu cross-section is seen to be in good agreement with the evaluation, while the ^{242}Pu cross-section appears to be slightly higher than the evaluated cross-section.

I. INTRODUCTION

Sensitivity studies performed as part of the Advanced Fuel Cycle Initiative (AFCI) and the Global Nuclear Energy Partnership (GNEP) projects have shown that in certain cases, the uncertainties in nuclear cross-sections contribute significantly to the uncertainties in reactor design parameters.[1] Both fission and capture cross-sections play an important role. Furthermore, the uncertainties are greatest on radioactive samples which have proven the most difficult to measure due to the sample induced background, limited availability of material, and other issues.

The Detector for Advanced Neutron Capture Experiments (DANCE) is a 4-π BaF_2 array which was designed for making time-of-flight (TOF) neutron capture measurements on small, radioactive samples for nuclear energy, threat reduction, and nuclear astrophysics.[2] The instrument is ideally adapted for the types of measurements which are needed for the GNEP program—in particular, differential measurements over a wide energy range on small-mass, often radioactive, isotopes.

II. EXPERIMENTAL PROCEDURE

All of the measurements were performed with the DANCE instrument located at Flight Path 14 at the Lujan Center at LANSCE at Los Alamos National Laboratory.[3]

II.A Neutron Production

The LANSCE facility operates an 800 MeV proton accelerator. Beams are delivered to multiple target stations for a variety of classes of experiments. The Lujan Center neutron production target receives beams of up to 120 μA in 150 ns full-width at half-maximum bursts. Twenty bursts are received on target per second. The neutron production target consists of two tungsten targets.[3] Spallation reactions produce neutrons of a wide range of energies which are then downscattered by moderators which are seen by various beam lines. Flight Path 14 (FP14) views a partially coupled backscatter water moderator. The resulting white spectrum has neutrons from thermal up to 500 keV, though typically the number of neutrons at maximum energy is too small for statistically significant measurements. The neutron energy can be determined by the time-of-flight technique relative to a reference pick-off signal which indicates when a proton burst impinges upon the spallation target.

II.B. The DANCE Instrument

DANCE is a 4-π BaF_2 scintillator array consisting of 160 independent detectors. Four different detector shapes ensure that each crystal observes the same solid angle. The detector face is 18 cm from the center of the array and each crystal is 15 cm deep in the line from the center of the array. A cut-away schematic of the array is shown in figure 1. The geometric design was planned to maximize both granularity and total solid angle.[4] This highly segmented array allows measurements of radioactive samples while minimizing the rate in individual detectors, simplifying pile-up and dead-time corrections. Furthermore, the high efficiency and large solid angle coverage of the BaF_2 crystals offer a total efficiency for a single gamma-ray of $\epsilon_{tot} \approx 90\%$ and a cascade efficiency for a typical 3-4 gamma cascade of $\epsilon_{casc} > 98\%$.[2,5]

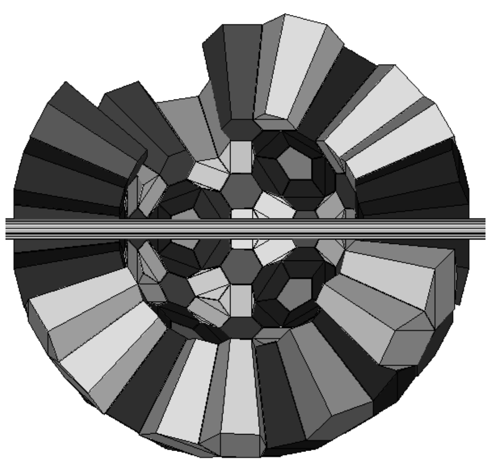

Fig. 1. A cut-away of approximately half of the DANCE array is shown above. The different crystal shapes are shown in different shades of grey.

A ^{6}LiH shell is installed outside of the neutron beam line but inside of the BaF_2 crystals to absorb scattered neutrons. Since the ^{6}Li(n,α)t reaction produces no gamma-rays, it is a very clean way to remove neutrons scattered from the sample before they have the opportunity to interact with the scintillator material.

There are significant advantages of the use of a total energy calorimeter such as the DANCE array over detectors such as C_6D_6 used in past measurements. The greatest advantage is offered by capturing the total cascade energy, which is not possible with other detection techniques. This offers an additional selection which can be placed on reaction Q-value, offering substantial improvement in the signal-to-noise ratio. In addition, it aids in the identification of background and contaminants. Furthermore, BaF_2 is a much higher efficiency material than C_6D_6, so much smaller samples can be used, reducing self-absorption effects and reducing the activity of radioactive samples. Since high-purity radioactive samples are difficult to produce in the first place, the ability to use small (sub-milligram) samples instead of gram size samples opens the door to measurements on a wide range of materials that were previously intractable.

II.C Target Preparation

All of the plutonium targets were prepared at Idaho National Laboratory. The plutonium was electroplated on to thin (2-4 μm) titanium foil and sealed with VYNS, a vinyl chloride and vinyl acetate co-polymer ($C_{22}H_{33}O_2Cl_9$), to ensure that the plutonium did not migrate. The electroplated area was 8 mm in diameter. The targets were prepared as two separate foils which were sandwiched together with the plutonium on the inner surface. Isotopic information about the samples can be found in table 1. Plutonium-239 was known to be a contaminant in the ^{240}Pu sample, so a ^{239}Pu sample was also measured and its mass and composition is listed in table 1 as well. The sample masses were determined by gamma counting each half of the sample before it was assembled. The samples were mounted in a standard DANCE radioactive target holder which consisted of an aluminum cylinder 1.75" in diameter with 0.003" thick KAPTON windows on either side for the beam to pass through. In this way, there were three layers of containment for the plutonium—the VYNS, the titanium, and the KAPTON. A schematic of the target holder is shown in figure 2.

TABLE I. Sample Composition (principal isotopes)

Composition				
Sample	Mass	%(^{239}Pu)	%(^{240}Pu)	%(^{242}Pu)
^{239}Pu	1.0 mg	**99.077**	0.881	0.005
^{240}Pu	0.9 mg	0.729	**98.305**	0.280
^{242}Pu	1.1 mg	0.005	0.022	**99.932**

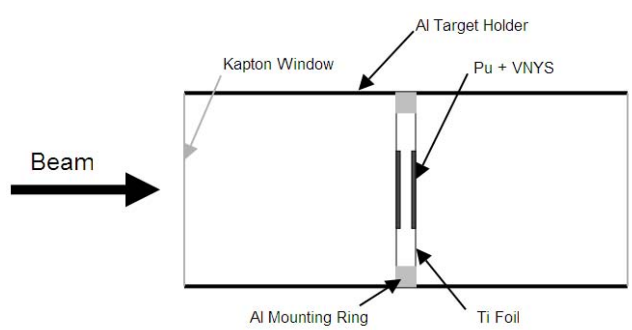

Fig. 2. A schematic of the sample mounted in a radioactive target holder is shown. The schematic is not to scale, but rather intended to show all of the relevant parts.

II.D Sample Irradiations

The measurements on the plutonium samples included measurements on the ^{240}Pu and ^{242}Pu, measurements with a sample prepared in an identical way but without plutonium, measurements with a polyvinylchloride (PVC) sample, which was similar in chemical composition to the VYNS, measurements with a polyethylene filter upstream of the sample cave to determine the gamma-scattering component, and measurements with a carbon sample to estimate neutron scattering into the BaF_2 array. The total beam time spent in the measurement of the ^{242}Pu sample was 21 days, including time for the measurement of contaminants. The time for the measurement of the ^{240}Pu sample was similar.

II.E Data Acquisition

The data was acquired using Acqiris DC265 waveform digitizers. A total of 320 channels of digitization was available for the 160 detectors. The data was sampled at a rate of 500 Msamples/s with 8 bit resolution. For each 24 channels, a single board computer was used to control the digitization and to perform initial data reduction. From the initial waveforms, a peak detection routine extracted a background level, 32 channels of fast waveform, and five slow integrals, reducing the data to be stored more than 15-fold over the event. The MIDAS (Maximum Integration Data Acquisition System) was used to acquire data and manage the experiment.[6] A full discussion of the acquisition system is given by Wouters *et al.*[7]

III. DATA ANALYSIS

The data was analyzed using the ROME (Root based Object oriented Midas Extension) framework.[8] This allowed the data to be replayed in list mode and to dynamically reconstruct physical events. This allowed the time coincidence for discriminating between events to be varied to optimize the signal to noise ratio and provide the cleanest events. A coincidence window of 30 ns was used for this analysis. Studies with highly active samples have shown that coincidence windows of as little as 6-8 ns are achievable, but with some cascade-dependent losses in Q-value detection.

Barium fluoride contains trace amounts of radium due to the difficulties in chemically separating the barium from the radium. The natural decay of radium and its daughters deposits MeV α's in the scintillator which produces a scintillation response. Pulse-shape discrimination was achieved by comparing short (6 ns) versus long (1 μs) integration times, which allowed α-induced events to be removed from the total energy determination. Furthermore, the α spectra were used to generate run-by-run gamma-ray energy calibrations. Due to the nature of scintillator detectors, it is important to make frequent corrections for gain shift. The α calibration routine proved highly reliable and very robust. In addition, it required no additional instrument time as the calibration data were taken while the capture data were being taken.

In the DANCE array, a single gamma-ray frequently interacts with multiple crystals in the process of being detected. As a result, the adjacent crystals which all observe an event have been defined as a "cluster" and the number of clusters which participate in an event is a much better approximation of the number of gammas that participated in the decay than the crystal multiplicity. For the remaining discussion, multiplicity will be assumed to refer to the number of clusters that participated in the event.

The data can then be represented in a three-dimensional matrix of neutron energy versus total γ-ray energy versus multiplicity. Gamma scattering and scattered neutrons predominately populate low multiplicity events, meaning that requiring high multiplicity preferentially selects capture events. Furthermore, gating on total energies near the reaction Q-value further selects capture events.

The cross-sections were extracted by a series of steps. First, the beam-independent background, due to natural radioactive sources, cosmic rays, etc. was subtracted based on the live time of the system. The background was determined from runs when the beam was off. Since the scattered neutron appears as captures in the barium isotopes with Q-values ranging from 4.7-9.1 MeV, by gating on events with total energy from 7.5-9.0 MeV in both backing and plutonium runs, the scatter contribution could be determined for each neutron energy. By scaling the contributions between a backing run and a plutonium run and subtracting the backing, a clean plutonium yield spectrum could be determined. The success of removing the scattering component from the events is illustrated in figures 3 and 4 for ^{242}Pu. Figure 3 shows the total energy deposited as a function of multiplicity for neutron energies of 1-10 eV. The Q-values for ^{240}Pu and ^{242}Pu are 5.24 and 5.03 MeV, respectively.[9] Not surprisingly, the spectrum is dominated by events with a good reaction energy as a large resonance dominates this region. In figure 4, the situation for neutron energies from 1-10 keV is illustrated. In this region, the scattering contamination is much more important and correct accounting is essential.

The main criterion for evaluating the quality of the background subtraction lies in the quality of the regions above and below the reaction Q-value. If the region near 8 MeV, which is dominated by capture on barium, is free of structure and zero, then accounting for scatter is proper. If the low energy region (<2 MeV) is well-behaved, generally tapering to zero and non-negative, then accounting for gamma-scatter is proper. The final yield was determined with a 1 MeV wide total energy gate near the Q-value.

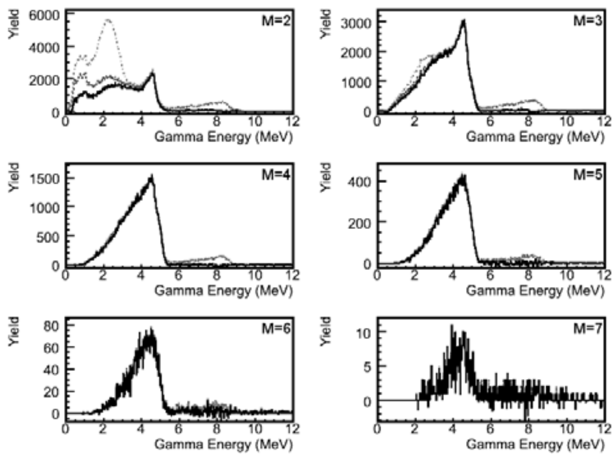

Fig. 3. The total energy deposition in the DANCE array, gated on neutron energies from 1-10 eV for the ^{242}Pu sample is shown above. The multiplicity is indicated in the upper right corner of each plot. The black line is the spectrum after all subtractions and should only show neutron capture on ^{242}Pu. The grey lines show various stages of the background removal.

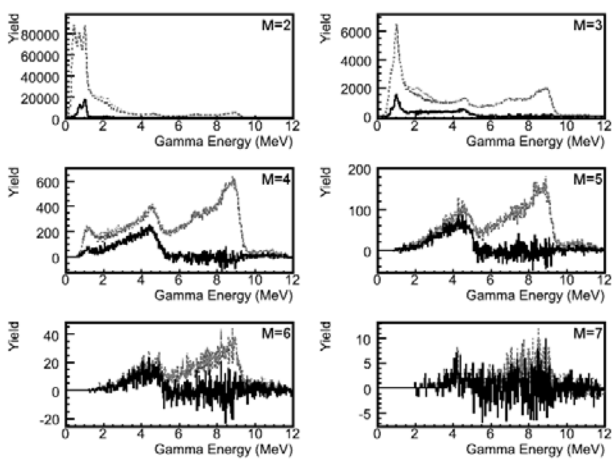

Fig. 4. The total energy deposition in the DANCE array, gated on neutron energies from 1-10 keV for the ^{242}Pu sample is shown above. The multiplicity is indicated in the upper right corner of each plot. The black line is the spectrum after all subtractions and should only show ^{242}Pu. The grey lines show various stages of the background removal.

The neutron flux shape was determined by use of neutron monitors approximately 2 m downstream of the DANCE ball. The flux was simultaneously measured with a ^{235}U fission detector, a $^{10}BF_3$ gas counter, and a ^{6}LiF evaporation layer viewed by a silicon detector. The flux shape determined by the monitors was used to weight the yield spectrum to extract a cross-section. The data were then normalized to the ENDF/B-VI.8 thermal value.[10]

The preliminary extracted cross-sections are compared to the SAMMY-broadened ENDF/B-VI.8 values in figures 5 and 6 for ^{240}Pu and ^{242}Pu.[11] SAMMY is an R-matrix fitting routine for the extraction of resonance parameters. It also allows the broadening of the evaluated cross-section to account for the resolution function of the neutron source, which aids in the comparison between data and evaluated cross-section. It has only been used for this broadening in the figures.

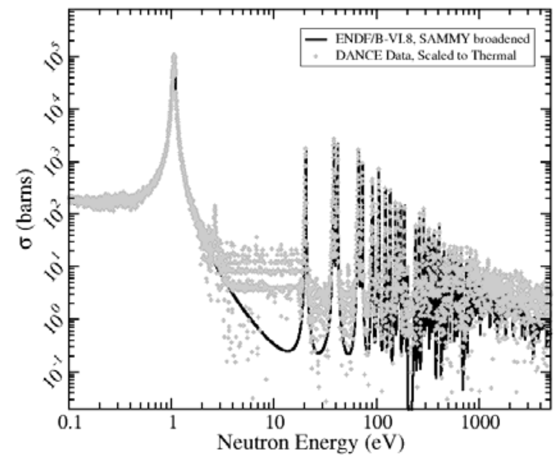

Fig. 5. The preliminary measured ^{240}Pu(n,γ) cross-section is shown above as compared to the ENDF/B-VI.8 value. Contamination from ^{242}Pu known to be in the target can be seen at 2.7 eV. The evaluated data are broadened but do not include contaminants.

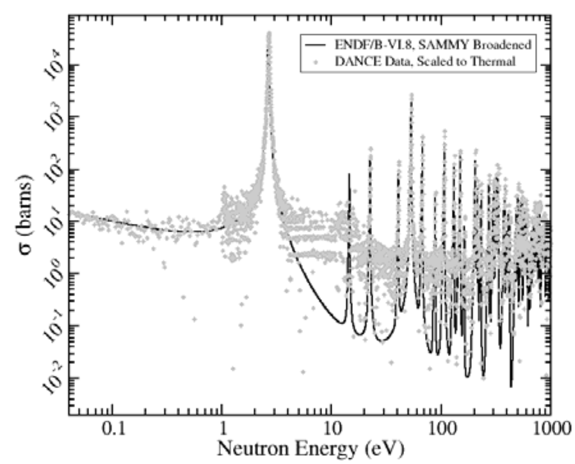

Fig. 6. The preliminary measured ^{242}Pu(n,γ) cross-section is shown above as compared to the ENDF/B-VI.8 value. The weak contamination of ^{240}Pu can be seen at 1 eV. The evaluated data are broadened but do not include any of the contaminants.

IV. RESULTS AND CONCLUSIONS

The measurements are largely in agreement with the previous evaluations, though the ^{242}Pu data appeared to be slightly larger than ENDF both in the resolved resonance region and in the unresolved region. One of the most notable results was that the decreased strength of the first and second resonances in ^{240}Pu(n,γ) reported by Guerrero *et al.* (the n_TOF Collaboration) was not observed.[12] Instead, the agreement between the strength observed in this measurement and the evaluation are in very good agreement. Figures 7 and 8 illustrate the difference in the results seen by the two measurements.

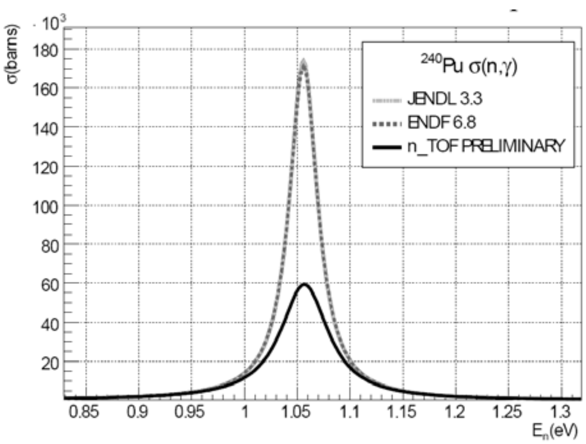

Fig 7. The result reported by Guerrero *et al.* (nTOF) for the ^{240}Pu(n,γ) cross-section show significant discrepancies with the previous evaluations.[12] The JENDL and ENDF data are in good agreement and are difficult to distinguish in the figure.

While the final analysis is still forthcoming, the DANCE measurements are very promising, both from the quality of present measurements and the potential for future measurements. Each of these measurements was made with a sample mass on the order of a milligram, a valuable development for future measurements on small samples. In the case of the ^{240}Pu, the measured values are in very good agreement with the evaluations, in contradiction to the results from another recent measurement. In the case of ^{242}Pu, the observed results are slightly higher and there is at least one resonance previously attributed to ^{242}Pu which was not observed.

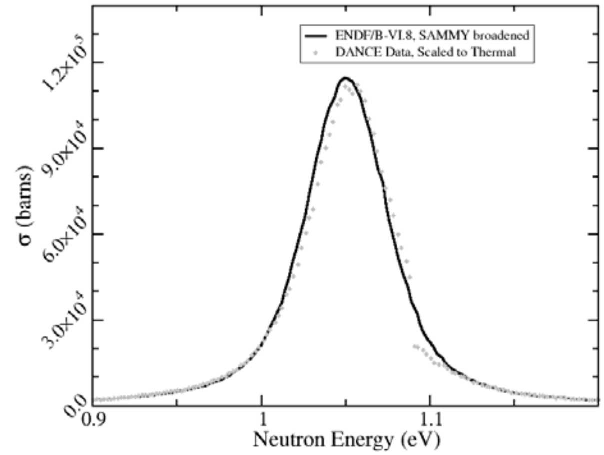

Fig. 8. The preliminary ^{240}Pu(n,γ) DANCE data, scaled to thermal, show excellent agreement with the evaluated strength of the first resonance. The jump in the DANCE data at approximately 1.08 eV is an artifact of dead-time in the data acquisition technique for the energy region from 1.08 up to ~17 eV. Proper dead-time corrections are expected to increase the observed yield and be well-behaved in the final analysis.

V. OUTLOOK

In addition to measurements on non-fissile isotopes such as ^{240}Pu and ^{242}Pu, DANCE has the capability to make measurements on fissile isotopes assuming that the capture cross-section is still at least 1% of the fission cross-section. These measurements are complicated by the high energy and multiplicity gamma background produced by fission events. A specially designed parallel-plate avalanche counter (PPAC) has been designed which fits inside the DANCE array at the target position. The sample mounts inside of the PPAC.[13] By tagging on fission events, the shape and structure of fission events can be determined and the fission background can be subtracted from capture events. This technique has been successfully employed for a determination of the ^{241}Am capture cross-section. Measurements have just been completed on ^{239}Pu. Measurements are planned later in the year on ^{242m}Am and ^{243}Am.

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