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Absolute measurement of the ^{242}Pu neutron-capture cross section

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The absolute neutron-capture cross section of ^{242}Pu was measured at the Los Alamos Neutron Science Center using the Detector for Advanced Neutron-Capture Experiments array along with a compact parallel-plate avalanche counter for fission-fragment detection. The first direct measurement of the ^{242}Pu(n,γ) cross section was made over the incident neutron energy range from thermal to $\approx$ 6 keV, and the absolute scale of the (n,γ) cross section was set according to the known ^{239}Pu(n,f) resonance at $E_{n,R}$ = 7.83 eV. This was accomplished by adding a small quantity of ^{239}Pu to the ^{242}Pu sample. The relative scale of the cross section, with a range of four orders of magnitude, was determined for incident neutron energies from thermal to $\approx$ 40 keV. Our data, in general, are in agreement with previous measurements and those reported in ENDF/B-VII.1; the ^{242}Pu(n,γ) cross section at the $E_{n,R}$ = 2.68 eV resonance is within 2.4% of the evaluated value. However, discrepancies exist at higher energies; our data are $\approx$30% lower than the evaluated data at $E_n \approx$ 1 keV and are approximately 2σ away from the previous measurement at $E_n \approx$ 20 keV.

I. INTRODUCTION

Improving the precision of network calculations of the radiochemical diagnostic chain is one of the priorities for the US/DOE Stockpile Stewardship program. A set of well measured (n,γ), (n,2n), and (n,f) cross sections for the isotopes involved are prerequisites for a precision network calculation. The isotope creation and destruction channels in the Pu-Am diagnostic chain, shown in Fig. 1, are important examples of these reactions. The (n,γ) cross sections for the actinides in Fig. 1 are among the high-value quantities in the network calculations. Our current focus is the ^{242}Pu(n,γ) cross section, and it is a key input to the Pu-Am network calculation. It is particularly relevant for modeling reactor performance and the development of next generation reactors [1, 2] because it has a long half-life of 3.8 $\times$ 10^5 years.

Little experimental data have been published on the ^{242}Pu(n,γ) reaction except for a few isolated incident neutron energy regions such as cross section data at thermal energy [3–10] and the energy range of $\approx$ 6$-$90 keV [11]. However, an extensive set of the ^{242}Pu(n,f) cross sections for incident neutron energies ranging from $\approx$ $10^2$$-$$10^8$ eV were reported [12–26]. In this article, we report a new ^{242}Pu(n,γ) cross section measurement at the Los Alamos Neutron Science Center (LANSCE) using the Detector for Advanced Neutron-Capture Experiments (DANCE) array [27] in combination with a parallel-plate avalanche counter (PPAC) [28]. The absolute neutron-capture cross sections were determined for incident neutron energies ranging from thermal to $\approx$ 40 keV, and this is the first direct measurement of the ^{242}Pu(n,γ) cross section between $E_n \approx$ 0.025$-$6000 eV. The (n,γ) cross sections reported in the evaluation ENDF/B-VII.1 [29] were derived indirectly from total cross section measurements in combination with (n,γ) cross section models and the available (n,γ) data. Details of the experiment, the analysis, and results are described in the sections below.

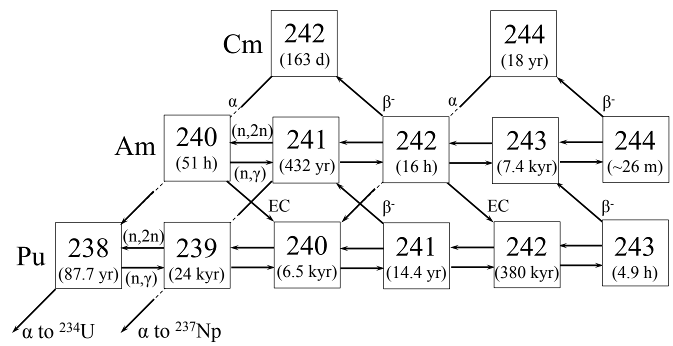

FIG. 1. Production/destruction network of nuclear reactions in plutonium-containing nuclear fuel, which leads to the formation of americium (Am) and curium (Cm) isotopes [1]. The ^{242}Pu(n,γ) reaction is among the key inputs to the series of reactions. Improved ^{242}Pu(n,γ) cross sections would contribute to improved network calculations.

II. EXPERIMENT

The measurement of the ^{242}Pu(n,γ) cross section, as a function of incident neutron energy (E_n), was carried out at the LANSCE Lujan Center [30] using the DANCE array. DANCE is located at a flight path 21.23 m away from the neutron source. Neutrons are produced by bombarding a tungsten target with 800-MeV protons at a

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repetition rate of 20 Hz, and they are then slowed down by a water moderator [31]. The incident neutron energy, ranging from thermal to several hundred keV, is determined from the time-of-flight difference between the beam pulse and event detection (by either the DANCE or the PPAC). The experiment was performed over a period of 17 days with a ^{242}Pu target installed within a PPAC. An additional seven days of beam time on a blank target in a duplicate PPAC assembly were utilized to collect background data in the inclusive mode.

A double-sided, electroplated target composed of 0.642 mg of 99.93% enriched ^{242}Pu with an active area $\approx$ 7.6 mm in diameter, was fabricated at Lawrence Livermore National Laboratory (LLNL) using the electroplating cell described in Ref. [32]. ^{239}Pu, with an atomic ratio of 5.0%, was added to the target to set the absolute scale of the ^{242}Pu neutron-capture cross section using the well-known ^{239}Pu(n,f) cross section. The uncertainty of this atomic ratio was measured to be better than 1% using the mass spectrometer at LLNL. This target was sandwiched between two 1.4 μm thick aluminized mylar foils, acting as the cathode, before it was installed inside the PPAC. Aluminized mylar foils of the same thickness, mounted on either side of the target at a distance of 3 mm, acted as the anodes. Entrance and exit windows were fabricated from 13 μm thick Kapton foils. This PPAC assembly was used successfully in previous experiments including the ^{252}Cf spontaneous fission measurement described in Ref. [33].

The DANCE array is composed of 160 BaF_2 crystals, with equal-volume and equal solid-angle coverage, arranged in a 4π geometry, and it was used to make precision neutron-capture cross section measurements for many actinides including ^{237}Np [34], ^{241}Am [1], ^{235}U [35], and ^{238}Pu [36]. It also was used for measuring the prompt-γ emission in both spontaneous fission and neutron-induced fission [33, 37–39].

This experiment was designed partially to improve the quality of the ^{242}Pu(n,γ) cross section with respect to the previous, unpublished measurement [40] by doubling the beam time to improve the event statistics, and thinning Kapton foils by a factor of $\approx$ 6 for the PPAC entrance and exit windows to reduce beam-induced backgrounds.

III. ANALYSIS

The (n,γ) cross section was determined from the spectrum of total γ-ray energy versus multiplicity measured by DANCE in the inclusive data collection mode. Therefore, it was necessary to impose proper gates on those two quantities to optimize the signal-to-noise ratio and improve the precision of the measured cross section. As a result, gating efficiencies were needed to determine the cross section. As mentioned earlier, the absolute scale for the ^{242}Pu(n,γ) cross section was set by the ^{239}Pu(n,f) cross section, and the PPAC detection efficiency for fission fragments also had to be determined. Detector efficiencies for both DANCE ($\epsilon_{\rm DANCE}$) and PPAC ($\epsilon_{\rm PPAC}$) are addressed below.

A. DANCE Efficiency

The efficiency of DANCE is dependent upon the gates applied to the summed γ-ray energy ($E_{\rm sum}$) and the cluster multiplicity ($M_{\rm cl}$). All γ-ray energies (E_γ) deposited in DANCE were summed over an initial 200 ns coincident time window—during the first stages of data analysis—and then over a narrower, 10 ns [41] coincident time window after "time alignment" was applied to all 160 equal-volume BaF_2 scintillation crystals. To align the crystals, a reference crystal was chosen at the beginning of analysis, and timing for each crystal during each run was corrected based on the time difference with respect to the reference crystal [1]. For the γ-ray energy, the radium (Ra) α-decay inherent to the DANCE BaF_2 crystals was used for alignment along with standard γ-ray calibration sources (^{22}Na and ^{60}Co). The γ-ray cluster multiplicity ($M_{\rm cl}$) was extracted by requiring that any γ rays detected with adjacent BaF_2 crystals triggered, be grouped together within a given time window. The cluster multiplicity was defined this way to minimize over-counting the γ-ray multiplicity because of Compton scattering.

A three-dimensional representation of the summed γ-ray energy versus the cluster multiplicity, with a gate at the neutron resonance energy $E_{\rm n,R}$ = 2.68 eV, is shown in Fig. 2. In the figure, the $E_{\rm sum}$ peak near the ^{242}Pu neutron-capture Q value of 5033.91 $\pm$ 2.63 keV [42] can be observed, particularly for events with the cluster multiplicity ≥ 3. Events with $M_{\rm cl}$ = (3,4) and $E_{\rm sum}$= 3.5$-$4.5 MeV were selected in the determination of (n,γ) cross section to improve the signal-to-noise ratio and thus the precision.

Before calculating the $E_{\rm sum}$ efficiency, the data collected on a blank target in a duplicate PPAC were subtracted from those collected with the active target by normalizing the area $E_{\rm sum}$ = 7$-$9 MeV. This energy range was selected because it corresponds to the summed γ-ray energy produced by randomly scattered neutrons captured by barium isotopes in the BaF_2 crystals [43]. Isotopes ^{137}Ba and ^{135}Ba have (n,γ) Q values of 8611.72 keV and 9107.74 keV, respectively [42]. This background subtraction is imperative in the determination of the cross section because the neutron beam intensity drops quickly as a function of $1/E_{\rm n}$, and there is a sharp deterioration in the signal-to-noise ratio at incident neutron energies exceeding 1 keV.

The background-subtracted, summed γ-ray energy spectrum generated by an incident neutron energy gate on the ^{242}Pu(n,γ) resonance at $E_{\rm n,R}$ = 2.68 eV [29] is shown in Fig. 3. The figure illustrates how the efficiency is calculated from the ratio of $E_{\rm sum}$ areas 3.5$-$4.5 MeV (black) and 0.0$-$5.25 MeV (gray). The same procedure was performed for the less intense ^{242}Pu(n,γ) resonance at $E_{\rm n,R}$ = 67.6 eV [29]. The weighted mean of the $\epsilon_{E_{sum}}$ for these two resonances was 39.58(5)%. The quality of data is illustrated in Fig. 4 by displaying the gated, summed γ-ray energy spectra with incident neutron energies of $E_n = 2.68$ eV (resonance) over the energy range 2.3−3.0 eV (Fig. 4a), $E_n = 1.0$ keV over the energy range 0.75−1.25 keV (Fig. 4b), and $E_n = 10$ keV over the energy range 7.5−12.5 keV (Fig. 4c). The deterioration of the data quality for $E_n > 1$ keV is evident.

To determine the cluster multiplicity efficiency, the E_{sum} spectra as a function of M_{cl} from 1−9 were generated with a gate on the $E_{n,R} = 2.68$ eV resonance. The same background subtraction method mentioned earlier was applied to these E_{sum} spectra for each M_{cl} and then projected onto the M_{cl} axis. However, $M_{cl} = 0$ and 1 could not be extracted from the data and were determined by assuming a Poisson distribution (an analytical approximation of the measured shape) for events with $M_{cl} = 2$–9. A quantitative estimation of the uncertainty due to this assumption was not made since events with $M_{cl} = 0$ and 1 account for no more than 5% of the total events. In Fig. 5, the shaded region represents $M_{cl} = 3$ and 4, and the solid line is the Poisson distribution fit to $M_{cl} = 2$–9. The DANCE multiplicity efficiency was calculated from the ratio between the areas $M_{cl} = (3,4)$ and 0−9. The multiplicity efficiency for the ^{242}Pu(n,γ) resonance at $E_{n,R} = 67.6$ eV was also analyzed in this manner. The weighted mean of the M_{cl} efficiencies for these two resonances was determined to be 59.6(4)%. As a result, the DANCE array efficiency, the product of the

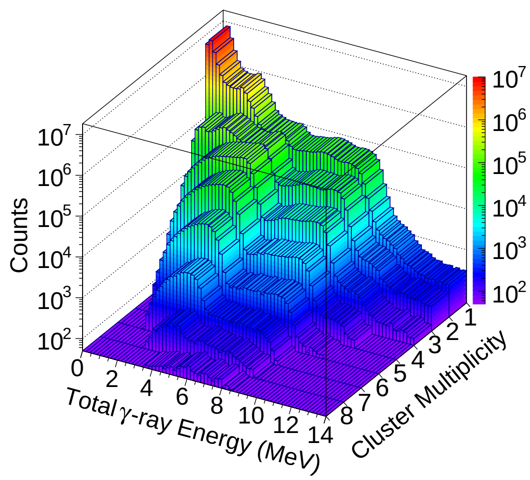

FIG. 2. The DANCE summed γ-ray energy versus cluster multiplicity with a gate at $E_{n,R} = 2.68$ eV. Events with $M_{cl} = 1$–8 and $E_{sum} = 0$–14 MeV are shown.

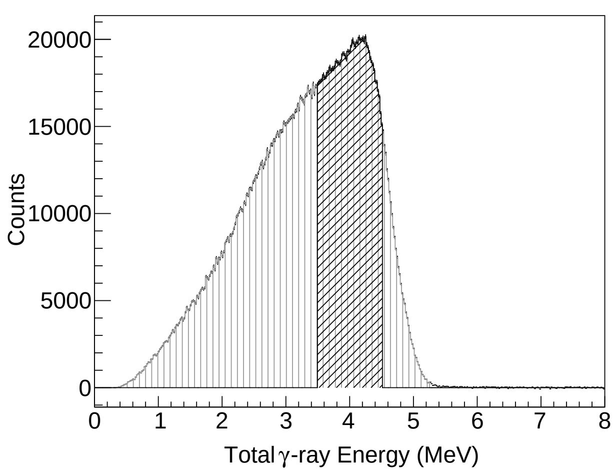

FIG. 3. The background-subtracted, summed γ-ray energy spectrum for the ^{242}Pu(n,γ) resonance at $E_{n,R} = 2.68$ eV. The $E_{sum} = 3.5$–4.5 MeV region is shaded in black and is located on the lower side of the ^{242}Pu(n,γ) Q value of 5.034 MeV. The energy region shaded in gray is $E_{sum} = 0.0$–5.25 MeV. Only cluster multiplicities 3 and 4 are shown.

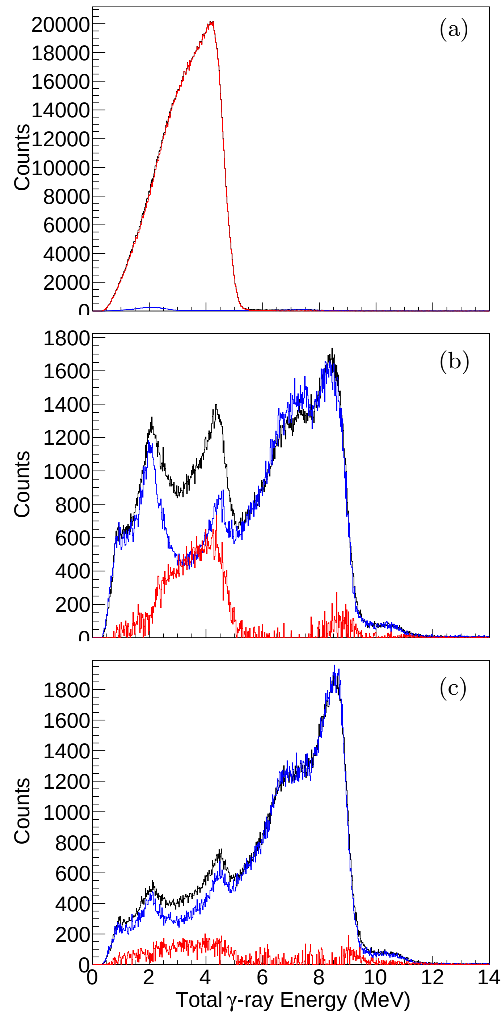

FIG. 4. The summed γ-ray spectra for incident neutron energies at (a) 2.68 eV, (b) 1.0 keV, and (c)10.0 keV with the requirement of $M_{cl} = (3,4)$. The inclusive spectra are black, the backgrounds (data normalized over the area of $E_{sum} = 7$–9 MeV) are blue, and the subtracted spectra are red.

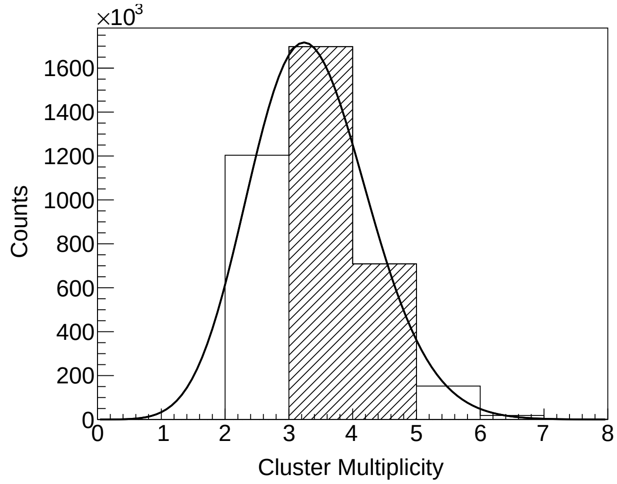

FIG. 5. The cluster multiplicity (M_{cl}) spectrum for the ^{242}Pu(n,γ) resonance at $E_{n,R}$ = 2.68 eV. The highlighted region corresponds to multiplicities M_{cl} = 3 and 4. The solid line is a Poisson distribution fit to M_{cl} = 2−9 to estimate multiplicities M_{cl} = 0 and 1.

M_{cl} and E_{sum} efficiencies, was ϵ_{DANCE} = 23.6(1)%. Note that the quoted uncertainties are statistical only.

B. PPAC Efficiency

In the current work, the absolute scale of the ^{242}Pu(n,γ) cross section was set by the known ^{239}Pu(n,f)

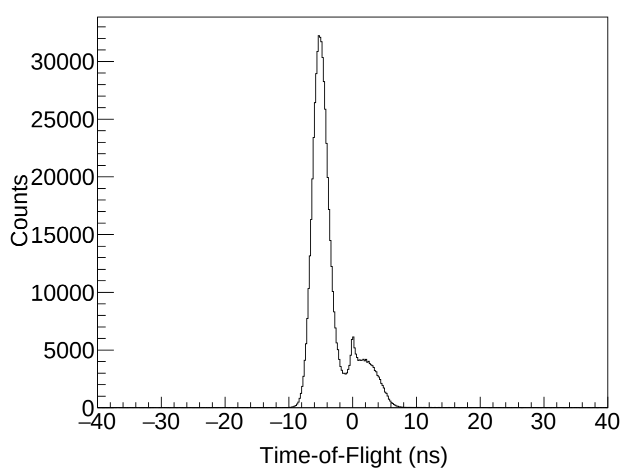

FIG. 6. The time difference between γ rays detected by the DANCE array and charged particles detected in the PPAC for ^{239}Pu(n,f). The time resolution is ≈2.8 ns for the peak at ≈-5 ns. An ≈8 ns gate was placed around this peak. The peak at 0 ns is an artifact of the timing algorithm and is not related to the event time-of-flight.

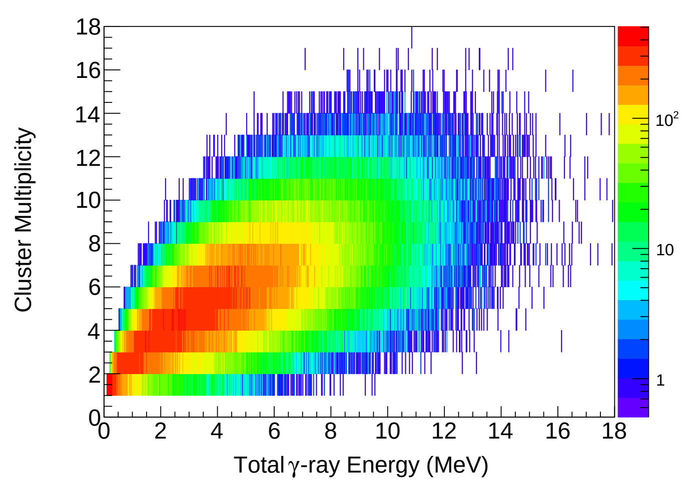

FIG. 7. The measured cluster multiplicity versus total γ-ray energy for the neutron-induced fission of ^{239}Pu.

cross section at E_n = 7.83 eV, measured using the PPAC. Therefore, it was necessary to know the PPAC efficiency, which was determined from the γ rays measured by the DANCE. The γ rays associated with fission fragments were isolated by gating on the PPAC pulse height and the PPAC−DANCE coincident timing spectrum shown in Fig. 6, where the time resolution of ≈ 2.8 ns was obtained. An ≈ 8 ns timing gate was imposed on the PPAC−DANCE coincident timing spectrum. The resulting two-dimensional multiplicity (M_{cl}) versus the summed γ-ray energy (E_{sum}) spectrum is shown in Fig. 7. Comparing this figure to Fig. 2, it is apparent that events with $M_{cl} \geq 8$ are dominated by fission. The efficiency of the PPAC is obtained by comparing E_{sum} spectra for the inclusive measurement and PPAC-coincident measurement where this multiplicity condition was applied. The PPAC efficiency was determined by taking the weighted mean of efficiencies calculated over several different incident neutron energy ranges and was found to be 55.8(12)%. Note that this uncertainty includes the statistics only.

C. Cross section

The ^{242}Pu(n,γ) cross section was determined in two steps. The first step was to extract the relative scale of cross section as a function of incident neutron energy. With gates on cluster multiplicities 3 and 4 as well as E_{sum} = 3.5−4.5 MeV, the cross section spectrum was background subtracted after corrections were made to both the inclusive and background spectra according to the neutron flux as a function of incident neutron energy. The neutron flux was monitored using the ^{6}Li(n,α) reaction rate measured downstream from DANCE. To remove the fission contribution, a DANCE−PPAC coincident cross section spectrum was constructed, using the same M_{cl} and E_{sum} gating condition and scaled by the PPAC efficiency, and then subtracted. Finally, the residual ^{239}Pu(n,γ) events were scaled and subtracted according to the measured (n,γ) spectrum for ^{239}Pu with an appropriate E_{sum} gate.

The absolute scale of the ^{242}Pu(n,γ) cross section was set according to the following equation [36] for the $E_{n,R}$ = 2.68 eV resonance:

$$\sigma_{^{242}\mathrm{Pu}} = \sigma_{^{239}\mathrm{Pu}} \frac{\epsilon_{\mathrm{PPAC}}}{\epsilon_{\mathrm{DANCE}}} \frac{N_{^{242}\mathrm{Pu}}}{N_{^{239}\mathrm{Pu}}} R_{^{239}\mathrm{Pu}/^{242}\mathrm{Pu}}, \quad (1)$$

where $\sigma_{^{242}\mathrm{Pu}}$ is the absolute ^{242}Pu(n,γ) integrated cross section over E_n = 1.5−4.5 eV for the $E_{n,R}$ = 2.68 eV resonance, $\sigma_{^{239}\mathrm{Pu}}$ = 162.9 b eV is the integrated fission cross section over E_n = 6.75−8.5 eV for the $E_{n,R}$ = 7.83 eV resonance [44], ϵ_{PPAC} is the PPAC efficiency, $\epsilon_{\mathrm{DANCE}}$ is the DANCE efficiency, $R_{^{239}\mathrm{Pu}/^{242}\mathrm{Pu}}$ is the atomic ratio of isotopes, $N_{^{242}\mathrm{Pu}}$ is the net counts for ^{242}Pu(n,γ) at the $E_{n,R}$ = 2.68 eV resonance, and $N_{^{239}\mathrm{Pu}}$ is the net counts for ^{239}Pu(n,f) at the $E_{n,R}$ = 7.83 eV resonance.

A first-order correction for the self-shielding effect was made for the ^{242}Pu(n,γ) cross section at $E_{n,R}$ = 2.68 eV because of the non-negligible beam attenuation for neutrons passing through the target. An increase of ≈ 6.5% to the measured cross section was estimated according to the description in Ref. [45].

IV. RESULTS

With all of the corrections mentioned above, the absolute ^{242}Pu(n,γ) cross section was obtained for incident neutron energies from thermal to ≈ 40 keV. The absolute scale was set according to the cross section determined at the $E_{n,R}$ = 2.68 eV resonance, which is 2890 ± 160 b eV integrated over E_n = 1.5−4.5 eV. By comparison, for the evaluated data reported in ENDF/B-VII.1 [29], the cross section integrated over the same E_n range is 2820 b eV which is ≈ 2.4% lower than the current value. Note that the systematic uncertainty for this measurement was not estimated since the statistical uncertainty for the absolute scale of ^{242}Pu(n,γ) cross section was already ≈ 6%. Statistical uncertainty derived from the measured ^{239}Pu(n,f) cross section was the dominant source of this uncertainty in the absolute (n,γ) cross section. Systematic uncertainties associated with the (n,f) measurement and the gating efficiencies were dwarfed by this statistical uncertainty. The absolute ^{242}Pu(n,γ) cross section measured in this work (black) with the evaluated data given by ENDF/B-VII.1 [29] (histogram) are shown in Fig. 8. Data are shown with the incident neutron energy ranges 10^{-2}−20 eV (Fig. 8a), 10−10^3 eV (Fig. 8b), and 10^3−10^6 eV (Fig. 8c). The data were truncated after ≈ 40 keV due to limited statistics. The $E_{n,R}$ = 14.60 ± 0.01 eV [29] resonance was not observed because it probably lies below the experimental sensitivity. The data, in general, are consistent with the evaluated data and the previous measurements. The exception is the cross section above $E_{n,R}$ > 1 keV where the data are systematically lower than the evaluated data listed in ENDF/B-VII.1 [29]. At E_n ≈ 1 keV, our data are ≈30% lower than the evaluated data, and at E_n ≈ 20 keV, the Hockenbury *et al.* [11] measurement is within 2σ of the new data.

In addition to the measured cross section, the

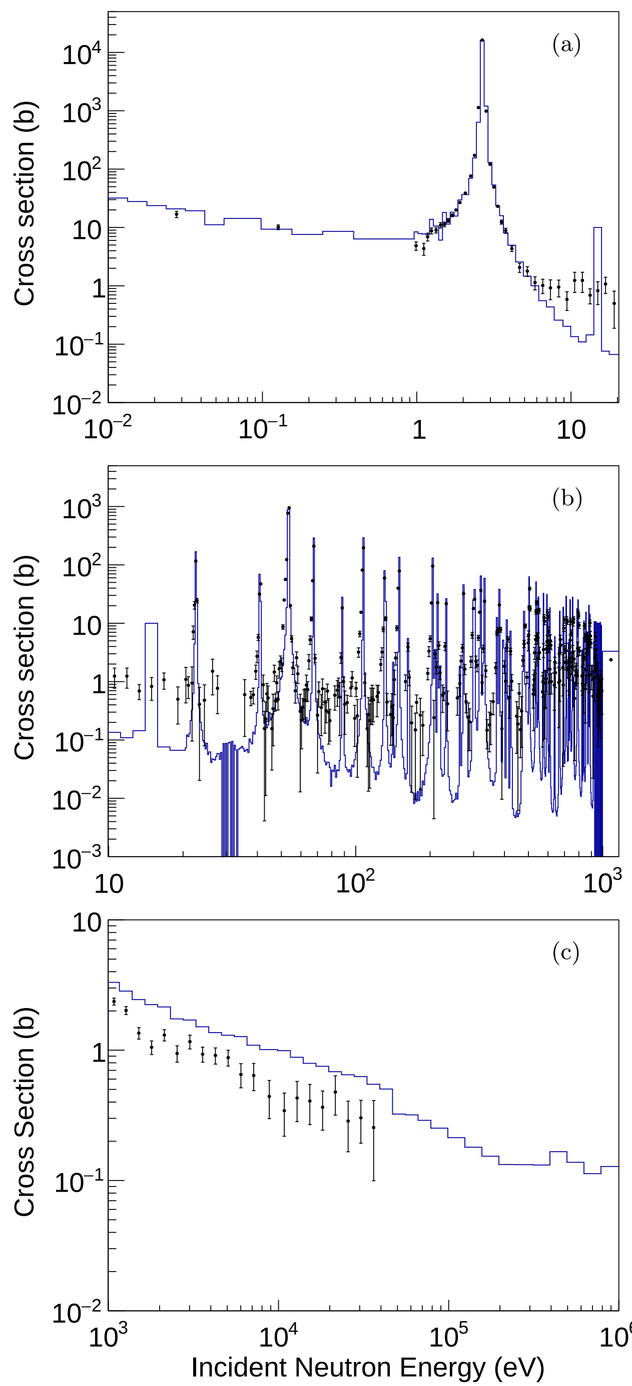

FIG. 8. Comparison of the absolute ^{242}Pu(n,γ) cross section between the current work (black circles) and the evaluated data in ENDF/B-VII.1 [29] (histogram) spanning (a) E_n =10^{-2}−20 eV, (b) E_n = 10−10^3 eV, and (c) E_n = 10^3−10^6 eV.

TABLE I. Comparison between the current ^{242}Pu(n,γ) resonance parameters and the resonance energies and widths reported in ENDF/B-VII.1 [29]. Statistical uncertainties are quoted in the table for the partial widths. An $\approx 0.5\%$ systematic uncertainty, associated with the neutron beam, was assigned for the DANCE (n,γ) resonance energies.

$E_{n,R}$ (eV)		Γ_γ (meV)		Γ_n (meV)	
Present	Ref. [29]	Present	Ref. [29]	Present	Ref. [29]
2.662	2.676(2)	25.1(5)	25	2.02(4)	2.00
–	14.60(1)	–	22	–	0.061
22.50	22.56(1)	22(2)	20	0.261(10)	0.29
40.95	40.95(1)	31(3)	29	0.416(12)	0.45
53.340	53.46(2)	20.6(6)	21.2	66(3)	52.0
67.4	67.6(2)	29(2)	21	4.04(11)	4.6
88.12	88.44	24(2)	26	0.73(6)	0.66
106.84	107.32(3)	24(2)	21	19.5(12)	18
130.4	131.4(1)	27(3)	22	7.4(7)	6.2
141.3	141.4	22(2)	22	0.059(6)	0.06
149.1	149.7	18.1(19)	22	12.6(13)	15
163.4	163.5	22(2)	22	0.53(5)	0.52
204.2	205.0	11.5(12)	22	48(5)	55
209.2	210.0	22(2)	22	0.42(4)	0.42
214.8	215.3	26(2)	22	6.2(6)	5.25
219.6	219.3	22(2)	22	0.126(13)	0.125
232.3	232.7	28(3)	28	4.7(5)	4.7
264.2	264.5	22(2)	22	0.180(18)	0.08
272.3	272.0	22(2)	22	0.080(8)	0.080
273.0	273.6	23(2)	23	16.3(16)	16.0
274.2	274.8	22(2)	22	0.085(9)	0.085
280.7	281.1	22(2)	22	0.066(7)	0.065
298.2	298.7	26(3)	26	8.4(8)	8.4
303.3	303.6	23(2)	22.5	18.5(18)	17.8
318.8	319.9	18.9(7)	22	260(20)	225
327.9	327.6	22(2)	22	0.25(3)	0.25
332.0	332.4	29(3)	25	76(7)	71
370.5	374.3	23(2)	22	6.7(7)	6.4
379.7	379.6	22(2)	22	0.135(14)	0.135
381.8	382.3	23(2)	22.5	51(5)	50
396.5	396.1	22(2)	22	2.5(3)	2.5
399.8	399.7	22(2)	22	1.80(18)	1.8
411.5	410.5	25(2)	22	8.5(8)	7.5
424.0	424.1	22(2)	22	4.1(4)	4.1
425.9	425.2	22(2)	22	0.141(14)	0.14
468.4	468.4	22(2)	22	0.0100(10)	0.01
473.1	473.7	22(2)	22	0.48(5)	0.48
482.0	482.5	24(2)	23.5	21(2)	20.9
495.0	494.8	22(2)	22	0.135(14)	0.135

^{242}Pu(n,γ) resonance energies ($E_{n,R}$), γ widths (Γ_γ), and neutron widths (Γ_n) for 38 resonances with energies between 2.66 and 495 eV were extracted to the first order using the *R*-matrix code SAMMY [45]; the fission widths, Γ_f, were set to values quoted in ENDF/B-VII.1 [29] during the calculation. The comparison between the current resonance energies and parameters and the values reported in ENDF/B-VII.1 [29] is given in Table I. The average Γ_γ for resonance energies within the range 2.66−495 eV is 23.0 meV which is 1.7% higher than the average reported in ENDF/B-VII.1 [29]. The uncertainties quoted in Tab. I are statistical; a systematic uncertainty of $\approx 0.5\%$ was adopted for the DANCE resonance energies due to the LANSCE neutron beam.

V. SUMMARY

The ^{242}Pu absolute neutron-capture cross section was measured successfully for incident neutron energies from thermal up to ≈40 keV using the DANCE array and a PPAC for fission-fragment detection. This was the first direct measurement of the ^{242}Pu(n,γ) cross section for thermal to ≈ 6 keV incident neutrons. The relative scale of the cross section was determined from the summed γ-ray energy and cluster multiplicity, both derived from DANCE and normalized to the measured neutron flux. The proper gates on those two quantities, with a combined efficiency of 23.6%, were necessary to enhance the signal-to-noise ratio and improve the precision of the measured cross section. The absolute scale was determined according to the known ^{239}Pu(n,f) cross section at $E_{n,R} = 7.83$ eV, which was measured using a PPAC for fission-fragment detection. This measurement was possible because a small, fixed amount of ^{239}Pu was added to the ^{242}Pu sample. Our data, in general, are in reasonable agreement with previous measurements and evaluated data; however, the deviation between the current (n,γ) cross section measurement and the ENDF/B-VII.1 [29] evaluation for incident neutron energies greater than 1 keV requires reevaluation. In addition to the cross section, the γ and neutron widths for 38 resonances, with resonance energies up to 495 eV, were evaluated using the code SAMMY. These widths will help improve model calculations of neutron-capture cross sections at higher incident neutron energies beyond the scope of the current work. This absolute measurement of the cross section for the ^{242}Pu(n,γ) reaction will improve the precision of network calculations of the Pu-Am diagnostic chain and benefit the development of next generation nuclear reactors.

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