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^{242}Pu absolute neutron-capture cross section measurement

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Abstract. 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. During target fabrication, a small amount of ^{239}Pu was added to the active target so that the absolute scale of the ^{242}Pu(n,γ) cross section could be set according to the known ^{239}Pu(n,f) resonance at $E_{n,R} = 7.83$ eV. The relative scale of the ^{242}Pu(n,γ) cross section covers four orders of magnitude for incident neutron energies from thermal to ≈ 40 keV. The cross section reported in ENDF/B-VII.1 for the ^{242}Pu(n,γ) $E_{n,R} = 2.68$ eV resonance was found to be 2.4% lower than the new absolute ^{242}Pu(n,γ) cross section.

1. Introduction

The ^{242}Pu(n,γ) reaction is important because precision (n,f), (n,γ), and (n,2n) cross sections are key input to network calculations of the radiochemical diagnostic chain. These calculations are a priority for the United States Department of Energy's Stockpile Stewardship program and related initiatives. New reactor concepts rely on network calculations like the Pu-Am diagnostic chain and catalyzed interest in improved cross section measurements for neutron-induced reactions on actinides [1,2]. In a reactor, plutonium-containing nuclear fuel undergoes reactions that produce long-lived ^{242}Pu ($t_{1/2} = 3.8 \times 10^5$ years). Improved cross sections for neutron capture by ^{242}Pu may impact calculations relevant to the development of next-generation reactors [2].

Prior to this study, little experimental data were published on the ^{242}Pu(n,γ) reaction. References [3–10] report cross sections for thermal neutron capture, and Ref. [11] provides cross sections at incident neutron energies $E_n \approx 6$–90 keV. Meanwhile, the ^{242}Pu(n,f) cross section has been well-studied [12–26], and reported cross sections span an incident neutron energy range from $\approx 10^2 - 10^8$ eV. In this work, a new (n,γ) measurement [27] was performed at the Los Alamos Neutron Science Center (LANSCE) using the Detector for Advanced Neutron-Capture Experiments (DANCE) array [28] in combination with a parallel-plate avalanche counter (PPAC) [29]. The measured absolute neutron-capture cross sections cover an incident neutron energy range from thermal to ≈ 40 keV. 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 [30] 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.

2. 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 [31] 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 repetition rate of 20 Hz, and they are slowed by a water moderator [32]. 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. This experiment was performed over a period of 17 days with a ^{242}Pu target installed within a PPAC. An additional seven days of beam were collected on a blank target in a duplicate PPAC assembly and represent the 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 ≈ 7.6 mm in diameter, was fabricated at Lawrence Livermore National Laboratory (LLNL) with the electroplating cell described in Ref. [33]. With an atomic ratio of 5.0%, ^{239}Pu was added to the target to set the absolute scale of the ^{242}Pu neutron-capture cross section with 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.

3. Analysis

Detector efficiencies for the DANCE array (ϵ_{DANCE}) and the avalanche counter (ϵ_{PPAC}) were key quantities necessary to extract the measured neutron-capture cross sections. Gates

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applied to the event-by-event-built summed γ-ray energy (E_{sum}) versus cluster multiplicity (M_{cl}) spectrum allowed signal-to-noise optimization, and these filters enabled a precision determination of the cross section as a function of incident neutron energy [34].

The E_{sum} efficiency was calculated from the ratio between the 3.5−4.5 MeV area and total area in the γ-ray spectrum associated with the $E_{n,R} = 2.68$ eV resonance. The same procedure was performed for the less intense ^{242}Pu(n,γ) resonance at $E_{n,R} = 67.6$ eV [30]. The weighted mean of these efficiencies was 39.58(5)%. Deterioration of the data quality for $E_n > 1$ keV was evident when this procedure was applied at higher neutron energies.

The cluster multiplicity efficiency was calculated from the ratio between the area of M_{cl} =(3,4) and the total multiplicity from M_{cl} = 0−9. The ^{242}Pu(n,γ) resonance at $E_{n,R}$ = 67.6 eV was also studied 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 M_{cl} and E_{sum} efficiencies, was ϵ_{DANCE} = 23.6(1)%.

The absolute scale of the ^{242}Pu(n,γ) cross section was set by the known ^{239}Pu(n,f) cross section at E_n = 7.83 eV and measured using the PPAC. An $\approx 8\,ns$ timing gate was imposed on the PPAC−DANCE coincident timing spectrum. The efficiency of the PPAC was obtained by comparing E_{sum} spectra for the inclusive measurement and PPAC-coincident measurement with cluster multiplicities restricted to $M_{cl} \geq 8$. 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)%.

The absolute ^{242}Pu(n,γ) cross section was determined from the equation [34]

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

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 $\cdot$ eV is the integrated fission cross section over $E_n = 6.75$–8.5 eV for the $E_{n,R} = 7.83$ eV resonance [35], 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. Self shielding contributed to an increase of $\approx 6.5\%$ in the measured cross section and was estimated according to the description in Ref. [36].

4. Results

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 and was found

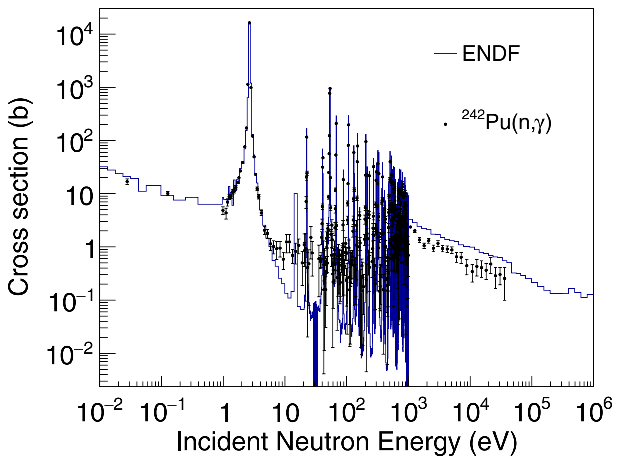

Figure 1. Comparison of the absolute ^{242}Pu(n,γ) cross section between the current work (black circles) and the evaluated data in ENDF/B-VII.1 [30] (histogram).

to be 2890 ± 160 b $\cdot$ eV integrated over E_n = 1.5−4.5 eV. By comparison, for the evaluated data reported in ENDF/B-VII.1 [30], the cross section integrated over the same E_n range is 2820 b $\cdot$ eV and is $\approx 2.4\%$ 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 the ^{242}Pu(n,γ) cross section was already $\approx 6\%$. Statistical uncertainty associated with the ^{239}Pu(n,f) cross section was the dominant source of uncertainty. The absolute ^{242}Pu(n,γ) cross section measured in this work (black) with the evaluated data given by ENDF/B-VII.1 [30] (histogram) are shown in Fig. 1. The data were truncated after ≈ 40 keV due to limited statistics. The $E_{n,R} = 14.60 \pm 0.01$ eV [30] resonance was not observed because it likely 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 evaluation. At $E_n \approx 1$ keV, our data are $\approx 30\%$ lower than the evaluated data, and at $E_n \approx 20$ keV, the measurement by Ref. [11] is within 2σ of the new data.

In addition to the measured cross section, the ^{242}Pu(n,γ) resonance energies, 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 [36]; the fission widths, Γ_f, were set to values quoted in ENDF/B-VII.1 [30]. The average Γ_γ for resonance energies within the range 2.66–495 eV was 23.0 meV and 1.7% higher than the average reported in ENDF/B-VII.1 [30]. These widths may help improve model calculations of neutron-capture cross sections at higher incident neutron energies beyond the scope of the current work.

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