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CONF-850507--5

NEUTRON TOTAL CROSS SECTION OF ^{240}Pu BELOW 6 eV AND THE PARAMETERS OF THE 1.0-eV RESONANCE*

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Abstract Measurements of the low-energy neutron transmission of five different ^{240}Pu samples have been made using the Oak Ridge Electron Linear Accelerator (ORELA) pulsed-neutron source and the time-of-flight technique in an attempt to resolve discrepancies between the evaluated low-energy cross section of this nuclide and the observed production of heavier Pu isotopes in light-water reactors (LWRs). Three "thin" samples consisting of Pu metal foils containing 0.73 at. % ^{240}Pu were measured above 0.4 eV in order to derive the parameters of the 1.056-eV resonance, which is responsible for most of the observed 2200-m/s cross section. Two "thick" samples of 99.97 at. % ^{240}Pu metal were measured from 0.003 eV to about 6 eV to obtain the 2200-m/s cross section and the cross-section shape in the low-energy region. A consistent set of resonance parameters was derived from the above data sets.

INTRODUCTION

At the Knoxville Conference in 1979, it was pointed out[1] that build-up of ^{241}Pu in LWRs was under-predicted in reactor calculations using ENDF/B-IV evaluated parameters for the 1.056-eV resonance of ^{240}Pu. Accordingly, for the ENDF/B-V data set, its capture width was increased by 11%, and the neutron width decreased by 7%. With these adjustments and including a 1-barn contribution for all other positive-energy resonances, the calculated 2200-m/s capture cross section was increased to 287 barns for ^{240}Pu, in accord with a measurement of 289.5 ± 1.4 barn by Lounsbury *et al.*[2] This measured value was obtained from mass-spectrometer analysis of Pu samples after irradiation in a thermal column. Interestingly, these adjustments decreased the capture area of the 1.056-eV resonance. The present measurements were undertaken to confirm the properties of this resonance and to measure the 2200-m/s cross section of ^{240}Pu.

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TRANSMISSION SAMPLES

Three thin samples consisting of 76.2-mm-diam foils of Pu metal containing 0.73% ^{240}Pu were used. These foils had been fabricated in 1963 at LANL. Before use the foils were unpackaged, cleaned, weighed, and resealed in a double layer of 0.005-in. aluminum in an inert atmosphere. Before resealing, three mg-quantity samples were removed from the edge of each foil for mass spectrometer analysis. One set was analyzed at LANL, another at the Y-12 facility in Oak Ridge, and the third was retained. The mean ^{240}Pu analysis of the six samples (0.7255 ± 0.0025%) was used for calculation. Thicknesses of the three foils as determined from their weights and areas were 0.0002914, 0.0006158, and 0.001167 atoms/barn of Pu with an estimated uncertainty of 0.6%. Two thick metal targets of ultra-pure ^{240}Pu were fabricated from oxide material by members of the Isotopes Production and Operations R&D Divisions at ORNL. These two samples were used to measure the thermal neutron cross section. The Pu (+1% aluminum) metal was rolled and then cut in a 0.625-in.-diam die to give disks with areal densities of 0.001676 and 0.004461 Pu atoms/barn. Mass-spectrometer analysis verified the isotopic purity as 99.972% ^{240}Pu.

THE NEUTRON SOURCE

The ORELA was operated at 384 pps and a pulse width of 12 ns for measurements on the three thin samples. The beam was collimated to 22.2 mm at the samples. A 1.6-mm Cd filter was mounted in the beam at 5 m to prevent overlap of slow neutrons from prior bursts. Filters of In, Au, and ^{238}U provided blacking-out resonances at 1.4, 4.7, and 6.6 eV, respectively, for background evaluation. For the thick-sample measurements, a burst width of 26 ns at a repetition rate of 25 pps was used. A Cd filter was cycled into the beam with the sample and the open blank for background determination below 0.3 eV. A fixed ^{238}U filter was placed in the beam to suppress the gamma flash and to provide a blacking-out resonance at 6.6 eV. Normalization of the sample and background rates to the open rate could be calculated either from a "house" neutron monitor or from the total bursts used. The two methods agreed to better than 0.2% for the thin samples and 0.3% for the thick samples.

THE NEUTRON DETECTOR

A 1-mm-thick by 88.9-mm-diameter NE-912 ^{6}Li-loaded glass, viewed from the edge by two RCA 8854 photomultipliers, served as the detector of neutrons. The glass was mounted in an aluminum housing which was lined inside with a highly reflecting thin sheet of aluminum-coated mylar. This detector was placed in a boron-carbide-shielded box at a flight path of 18 m. The photomultipliers were located outside the collimated neutron beam in order to minimize scattered-neutron background.

DATA ANALYSIS

Analysis of the data was carried out with the multi-level, Breit-Wigner shape fitting code SIOB.[3] Both resolution and Doppler broadening were taken into account, the latter according to the assumption of the "weak" binding model. For the 1.056-eV resonance, the resolution width was much smaller than either the Doppler or total widths and contributed negligible uncertainty to the derived parameters. An effective temperature for Doppler broadening of 295°K was assumed, corresponding to the room temperature of 290°K.

RESULTS

The thin samples were run two at a time; the 0.006158-a/b sample was run with each of the other two, but rotated 180° about its normal for the second run. The data and SIOB fits in the region about the 1.056-eV resonance are shown in Fig. 1. The ^{239}Pu "background" transmission was accounted for by fitting the far wings (from 0.4 eV to 2 eV) with SIOB while varying the 0.3-eV ^{239}Pu resonance parameters. The thick samples transmission data resulted in a 2200-m/s total cross section for ^{240}Pu of 284 ± 2 barns. The error includes a 0.4% estimated uncertainty in sample thickness. Due to the large negative correlation in the resonance parameters, a precise value of the product $\Gamma_n \Gamma_\gamma$ was also obtained which is

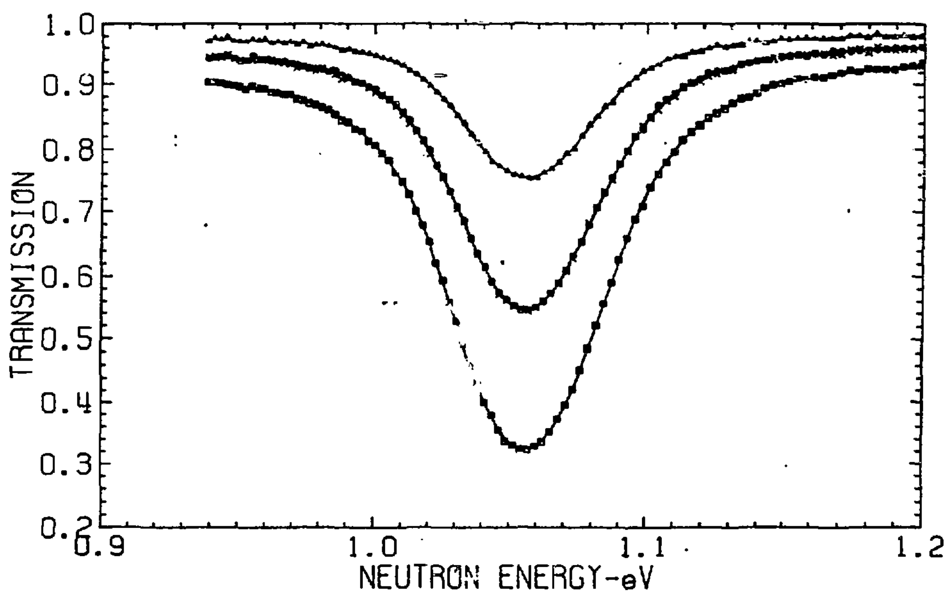

Fig. 1. Thin sample transmission data and the SIOB fits (solid lines) over the 1.056-eV resonance.

independent of resolution and relatively insensitive to Doppler broadening. Table I summarizes the present analysis. The errors in the table are from counting statistics only. Preliminary results of the present experiments are $\Gamma_n = 2.45 \pm 0.02$ meV, $\Gamma_\gamma = 30.3 \pm 0.2$ meV, $\Gamma_n\Gamma_\gamma = 74.13 \pm 0.23$ meV^2, and $E_0 = 1.056 \pm 0.001$ eV, where the errors in widths include sample-thickness uncertainties and the flight-path uncertainty is included in the resonance energy error. The contribution of the 1.056-eV resonance to the 2200-m/s cross section is 282 barns, leaving approximately 1 barn for negative energy resonances. These results agree better with the ENDF/B-IV evaluation than with the ENDF/B-V.

Table I. SIOB Results

A. Thin samples analyzed simultaneously:

$E_o = 1.05641 \pm 0.00002$ eV
$\Gamma_n = 2.449 \pm 0.002$ meV; $\Gamma_\gamma = 30.27 \pm 0.06$ meV
Correlation Coefficient $C(\Gamma_n,\Gamma_\gamma) = 0.65$

B. Thick samples analyzed simultaneously:

$\Gamma_n\Gamma_\gamma = 74.13 \pm 0.10$ meV^2
$C(\Gamma_n,\Gamma_\gamma) = -1.00$

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