Resonance Parameter Measurements and Analysis of Gadolinium

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Abstract

The purpose of the present work is to measure the neutron cross sections of gadolinium accurately. Gd has the highest thermal absorption cross section of any natural element. Therefore it is an important element for thermal reactor applications

Neutron capture and transmission measurements were performed by the time-of-flight technique at the Rensselaer Polytechnic Institute (RPI) LINAC facility using metallic and liquid Gd samples. The liquid samples were isotopically-enriched in either ^{155}Gd or ^{157}Gd. The capture measurements were made at the 25-m flight station with a sodium iodide detector, and the transmission measurements were performed at 15- and 25-m flight stations with ^{6}Li glass scintillation detectors. The multilevel R-matrix Bayesian code SAMMY was used to extract resonance parameters.

The results of the thermal region analysis are significant. Resonance parameters for the low energy doublet, at 0.025 and 0.032 eV, are presented. The thermal (2200 m/s) capture cross section of ^{157}Gd has been measured to be 11% smaller than that calculated from ENDF/B-VI updated through release 8. Thermal capture cross sections and capture resonance integrals for each isotope as well as elemental gadolinium are presented.

In the epithermal region, natural metal samples were measured in capture and transmission. Neutron interaction data up to 300 eV have been analyzed. Substantial improvement to the understanding of gadolinium cross sections is presented, particularly above 180 eV where the ENDF resolved region for ^{155}Gd ends.

Keywords:
gadolinium
resonance parameters
resonance integral
transmission
capture

1. Introduction

The purpose of the current effort was to measure the neutron cross sections of gadolinium. The effort was optimized by the use of isotopically-enriched samples, diluted liquid samples, combined transmission and capture measurements, and Bayesian analysis using the SAMMY [1] computer program. A detailed description of these measurements and analyses is given in References [2,3]. Gadolinium has the highest thermal cross section of any natural element. Its large thermal cross section results from two strong resonances near the thermal energy. One resonance in ^{157}Gd at 0.0314 eV has a capture cross section of 254,000 barns. The other strong thermal resonance, 60,700 barns, occurs in ^{155}Gd at 0.0268 eV. The proximity of these two energies to each other required (partially) separated isotopes. The large cross section of these resonances required exceptionally thin samples. To solve the problem of producing very thin but uniform-thickness samples, diluted liquid samples in thin-walled quartz cylinders were produced. The diluent was heavy water to reduce neutron captures outside of gadolinium.

In the epithermal region only natural elemental metal gadolinium samples were used. Nevertheless, substantial improvement to the ENDF Gd database has been accomplished by careful fitting of thick transmission and capture sample data. Twenty-eight new resonances were clearly identified which are not listed in ENDF. Although the isotope of each resonance was not established, a value of $2ag\Gamma_n$ is quoted for each, where a is the isotopic abundance and g is the spin factor.

2. Historical Review

The seminal measurement of the thermal resonance parameters of gadolinium was made by Mϕller et al.[4]. The resonance parameters for the low energy doublet ($E \approx 0.03$ eV) in ENDF/B-VI [5] updated through release 8 are nearly identical to those of Mϕller et al. [4]

In the epithermal range the key experimenters contributing to the current resonance parameter database are: Mughabghab and Chrien[6], Simpson[7], Fricke et al.[8] , Rahn et al.[9] , as well as Anufriev et al.[10], Macklin[11], Belyaev et al.[12], Karshzhavina et al.[13,14], and Asghar et al.[15].

3. Experimental Conditions

The present experiments employed both transmission and capture measurements. Transmission is the ratio of detector counts with a sample in the beam to the detector counts with the sample out of the beam. Neutron capture experiments measure neutron capture yield. The yield is defined as the number of neutrons captured per neutron incident on the sample.

The RPI LINAC produces a 60-MeV pulsed electron beam. Neutrons are generated from electrons using tantalum photoneutron targets. The neutron energy for a detected event is determined using the time-of-flight technique. The time from the LINAC electron burst, which creates the neutrons, to the time of the detected event defines the flight time of the neutron. Combining this with precise knowledge of the flight path length gives the neutron energy.

The beam conditions during these experiments were 1-3 μs wide pulses at 25 pulses per second for thermal measurements producing an average beam current of 8-19 μA, and 120-160 ns wide pulses at 250 pulses per second for epithermal measurements producing an average beam current of 40-45 μA

The detector used in the neutron capture experiments is a multiplicity-type scintillation gamma detector containing 16 sections of NaI, formed into a 30.5-cm diameter x 30.5-cm length (12-in x 12-in) right circular cylinder with a 8.9-cm (3.5-in) through hole along its axis.[16,17,18] The cylinder is split normal to its axis into two rings, and each ring is divided into 8 equal pie-shaped segments. Each segment is optically isolated in a hermetically sealed aluminum can and is viewed by an RCA 8575 photomultiplier tube. The capture detector was located at approximately 25.5 m from the neutron-producing target.

Neutron transmission measurements were conducted at the 15-meter and 25-meter flight stations. Both transmission detectors use NE 905 ^{6}Li glass scintillation detector (6.6% lithium, enriched to 95% in ^{6}Li).

A variety of gadolinium samples were used in these experiments. Of particular interest was the thermal region. To distinguish between the two thermal resonances, enriched isotopes were measured. Two levels of enriched gadolinium oxide powder were available. One contained 70% ^{157}Gd and 1.4% ^{155}Gd. The other contained 3.8% ^{157}Gd and 74% ^{155}Gd. The remainders were other gadolinium isotopes. The best method for providing a thin, uniform sample was to manufacture liquid samples in quartz cells. Deuterated nitric acid was used to dissolve the Gd_2O_3. It was then diluted with D_2O to produce the desired Gd concentrations. Nine different dilutions of these mixtures were used.

Natural metal gadolinium samples were also measured in the epithermal region. Their thicknesses ranged from 0.025 mm – 1.02 cm.

4. Results

4.1 Data Reduction and Analysis

Data were dead time corrected, run-summed, beam-monitor-normalized, and background-corrected. Final processed data were reduced to transmission or capture yield.

The resonance parameters neutron width, Γ_n, radiation width, Γ_γ, and resonance energy, E_0, were extracted from the capture and transmission data sets using the multi-level R-matrix Bayesian code SAMMY version M6 [1]. This was a combined transmission and capture analysis, which employed the resolution broadening, self-shielding, multiple-scattering, and diluent features of SAMMY.

The thermal capture yields were normalized to the thermal transmission data. The background in thermal capture with liquid samples was estimated by measuring a blank, D2O-filled quartz cell. The further effect of multiple scattering from the quartz cell was estimated using a Monte Carlo simulation. The resulting correction factors, ranging from 1.022 below thermal to 0.97 near 1 eV, were applied to the capture yield data.

The epithermal capture data were normalized to the saturated Gd resonance at 6.3 eV.

4.2 Thermal Results

The SAMMY analysis resulted in resonance parameters for the thermal doublet. Resonance parameters from the present measurement are compared to ENDF in Table 1.

Table 1: Thermal Results: Resonance Parameters

Dataset	Energy, eV	Γ_γ, meV	Γ_n, meV	ISOTOPE	J
ENDF-B/VI	0.0268 $\pm$ 0.0002	108 $\pm$ 1	0.104 $\pm$ 0.002	155	2
	0.0314 $\pm$ 0.0002	106 $\pm$ 1	0.4704 $\pm$ 0.0080	157	2
RPI	0.025 $\pm$ 0.003	104 $\pm$ 3	0.097 $\pm$ 0.003	155	2
	0.032 $\pm$ 0.003	107 $\pm$ 3	0.428 $\pm$ 0.004	157	2

The neutron width of the largest resonance in Gd, at 0.032 eV in ^{157}Gd, has been measured to be (9$\pm$1)% smaller than that given in ENDF/B-VI updated through release 8. The thermal (2200 m/s) capture cross section of ^{157}Gd has been measured to be 11% smaller than that calculated from ENDF. The other major thermal resonance, at 0.025 eV in ^{155}Gd, did not display a significant deviation from the thermal capture cross section given by ENDF.

4.3 Epithermal Results

A set of resonance parameters for gadolinium up to 300 eV as well as a more detailed description of the present measurements is available in References [2,3]. Twenty-eight resonances were introduced that were not included in ENDF. The epithermal results were derived entirely from elemental metal samples, 15 data sets in all. The isotopic assignment of new resonances could not be established. Their widths are assigned in References [2,3] as $2ag\Gamma_n$ where a is the isotopic abundance and g is the spin factor.

Six resonances present in ENDF have been discarded because the literature does not demonstrate their existence nor do the present measurements support their existence.

A new resonance was proposed only when the data provided clear evidence that a resonance was present. Many of the proposed new resonances appear above 180 eV, which was the limit of the resonance region for ^{155}Gd in ENDF/B-VI updated through release 8.

Figure 1 shows a region of the SAMMY fit to the present data and ENDF. The analysis suggested seven new resonances in this region, at 203.4, 209.1, 210.3, 212.3, 213.7, 214.8, and 218.8 eV. Neither the data nor a detailed literature search [6-15] found support for ENDF resonances at 202.7 or 206.9 eV. These two resonances were removed. The literature also only supported one resonance at 201.6 eV. The second resonance, listed at exactly the same energy, is conjectured to be a mistake in the ENDF compilation.

Figure 1 : An overview of transmission and capture data used in the 200-225 eV region and the SAMMY fits using the RPI parameters. The dashed lines represent the ENDF parameters for the thickest samples.

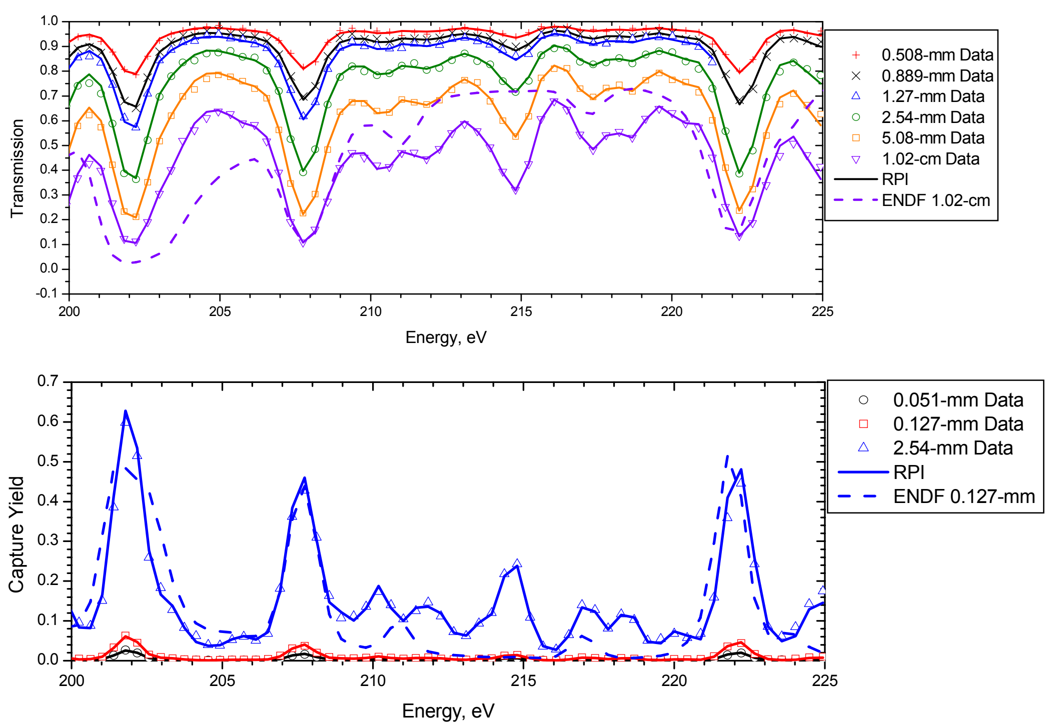
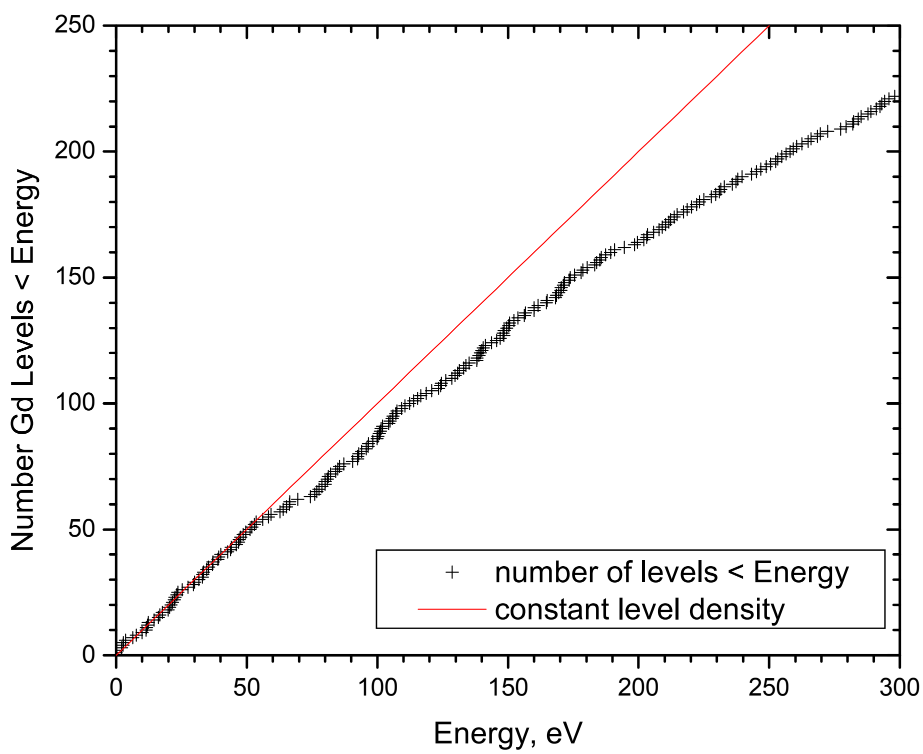


Figure 1 illustrates a typical impact of the present measurement on the understanding of gadolinium resonance parameters. Reference [3] describes the resonance-by-resonance scrutiny that was performed for this analysis.

Figure 2 shows a plot of the gadolinium level density including all new resonances added during the present analysis. The plot of observed levels vs energy shows a good fit to a straight line which agrees with the statistical model of the nucleus up to about 50 eV. All levels are s-wave. Elemental gadolinium is shown because there is no assignment of isotope to the proposed new resonances. Above 50 eV a significant number of levels are missed. Therefore, even with the resonances added in the present analysis, the expectation of constant level density vs energy is not exceeded.

Figure 2: 'Staircase' plot of elemental gadolinium observed levels vs energy shows a good fit to a straight line up to about 50 eV. Above 50 eV, even with the resonances added in the present analysis, the expectation of constant level density vs energy is not exceeded.



4.4 Resonance Integrals and Thermal Cross Sections

Thermal capture cross sections from the present measurement are compared to ENDF in Table 2.

Table 2 : Thermal capture cross sections. A comparison of ENDF/B-VI to RPI results. Cross section units are barns. The units of abundance are percent.

Thermal Capture Cross Sections							
		ENDF			RPI		
Isotope	**Abund**	**Thermal Capture**	**Contribution to Elemental**	**Percent**	**Thermal Capture**	**Contribution to Elemental**	**Percent**
Gd152	0.200	1050.	2.10	0.00430	1050.	2.10	0.00430
Gd154	2.18	85.0	1.85	0.00379	85.8	1.87	0.00422
Gd155	14.80	60700.	8980.	18.4	60200.	8910.	20.1
Gd156	20.47	1.71	0.350	0.000717	1.74	0.356	0.000804
Gd157	15.65	254000.	39800.	81.6	226000.	35400.	79.9
Gd158	24.84	2.01	0.499	0.00102	2.19	0.544	0.00122
Gd160	21.86	0.765	0.167	0.000342	0.755	0.165	0.000372
Gd	100.0		48800.	100.0		44300.	100.0

The most significant departure of the present results from ENDF thermal capture cross sections is in ^{157}Gd. This 11% reduction (from 254000 barns for ENDF to 226000 barns for RPI) is consistent with the ≈9% reduction in neutron width for the thermal ^{157}Gd resonance (see Table 1). An insignificant reduction in thermal capture cross section is seen in ^{155}Gd. This is due to the competing effects of a 7% reduction in neutron width compensated by a 3.7% reduction in total width ($\Gamma_n+\Gamma_\gamma$) and an energy shift toward the thermal energy (0.0253 eV) point. The thermal capture cross section of elemental gadolinium is ≈9% lower than that calculated from ENDF parameters.

Resonance integrals (Table 3) are given for each isotope as well as their contribution to the elemental values. The integrations extend from 0.5 eV to 20 MeV. The low energy cutoff is above the thermal region doublet. The elemental resonance integral for Gd as measured is 2.8% (11 b) larger than that of ENDF. The largest fractional increases in isotopic contributions occur in ^{154}Gd and ^{158}Gd. ^{154}Gd and ^{158}Gd have far fewer resonances than ^{155}Gd or ^{157}Gd. A 14% increase in ^{158}Gd resonance integral compared to ENDF was measured. This is dominated by the 22.3 eV resonance whose neutron width changed by approximately the same amount. The resonance integral of ^{154}Gd is 20% larger than that calculated from ENDF parameters. The ^{154}Gd resonance integral is larger than ENDF due to larger widths for resonances at 22.5, 49.63, 65.21, and 76.00 eV (see Table VII of Reference [3]). ^{155}Gd contributes more than half of the elemental Gd capture resonance integral and its contribution is virtually unchanged when compared to that calculated from ENDF parameters.

Table 3: Infinitely dilute neutron capture resonance integrals. A comparison of ENDF/B-VI to RPI results. Cross section units are barns. The units of abundance are percent.

Capture Resonance Integral							
Isotope	Abund	ENDF			RPI		
		Capture RI	Contribution to Elemental	Percent	Capture RI	Contribution to Elemental	Percent
Gd152	0.200	476.	0.952	0.243	476.	0.952	0.237
Gd154	2.18	217.	4.73	1.21	261.	5.69	1.42
Gd155	14.80	1540.	228.	58.3	1570.	232.	57.7
Gd156	20.47	105.	21.5	5.50	104.	21.3	5.30
Gd157	15.65	755.	118.	30.2	789.	123.	30.6
Gd158	24.84	62.8	15.6	3.99	71.5	17.8	4.43
Gd160	21.86	7.89	1.72	0.440	7.66	1.68	0.418
Gd	100.		391.	100.		402.	100.

5. Discussion of Uncertainties

Counting statistics were propagated through the data reduction and the SAMMY Bayesian analysis. These statistical uncertainties were generally smaller than the systematic differences between data sets. Uncertainties presented in Table 1 reflect the internal inconsistencies between data sets and are of the order of 1σ.

6. Conclusions

Neutron widths and thermal (2200 m/s) capture cross sections of the thermal doublet are smaller than those from ENDF/B-VI updated through release 8. The conclusions of these measurements are as follows:

- The neutron width of the 0.032 eV resonance in ^{157}Gd is 9% smaller than that of ENDF.
- The neutron width of the 0.025 eV resonance in ^{155}Gd is 7% smaller than that of ENDF.
- The radiation width of the 0.032 eV resonance in ^{157}Gd is not significantly different from that of ENDF.
- The radiation width of the 0.025 eV resonance in ^{155}Gd is 4% smaller than that of ENDF.
- The thermal (2200 m/s) cross section of ^{157}Gd is 11% smaller than that of ENDF.
- The thermal (2200 m/s) cross section of ^{155}Gd is not significantly different from that of ENDF.

In the epithermal region, a great deal of improvement has been made to the Gd resonance parameter database. In the energy region near 96 eV, and particularly above 165 eV, significant changes are suggested to ENDF parameters. New resonances have been suggested where comparisons of data to calculations clearly show they are needed.

7. References

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