FY 2016 Progress in Resonance Evaluations of Gadolinium for the NCSP*

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INTRODUCTION

A reevaluation of the resonance regions of the five most abundant isotopes of gadolinium is under way. The task of the reevaluation is being done as a collaboration between the nuclear data group at Oak Ridge National Laboratory (ORNL) and at the Institute for Radiological Protection and Nuclear Safety (IRSN) under a memorandum of understanding between the US Department of Energy (DOE) Nuclear Criticality Safety Program (NCSP) and IRSN.

A new set resonance evaluations for gadolinium is warranted because of the significant role that gadolinium plays in the nuclear industry and because new experimental cross-section measurements made at the RPI Gaerttner LINAC Center [1,2] suggest discrepancies of up to 9% in the thermal cross-section values with respect to the current ENDF/B-VII.1 evaluation. Further, simulations of integral experiments with varying sensitivities to gadolinium, such as in Ref. 3, suggest that the thermal cross section in the ENDF/B-VII.1 evaluation may be over-predicted. The resonance evaluations for gadolinium in ENDF/B-VII.1 will be propagated into ENDF/B-VIII.0. The completed new evaluation will be proposed for inclusion in the next release after ENDF/B-VIII.0.

ORNL has partnered with IRSN to perform an evaluation of a wide body of available experimental data and to seek to resolve discrepancies between several independent experimental measurements to produce a new set of resonance evaluations for gadolinium. The new gadolinium evaluations will benefit the NCSP by significantly improving the predictive power of radiation transport calculations for systems involving gadolinium in the DOE Complex. The new evaluations will also seek to provide evaluated covariance data to support sensitivity/ uncertainty analyses and aspire to evaluate correlations that are known to exist between the resonance evaluations of the different isotopes of gadolinium but have not hitherto been reported.

BACKGROUND

Gadolinium-157 has the largest thermal-neutron cross section of all naturally occurring isotopes. Gadolinium is used as an emergency shutdown measure in some nuclear reactors, as a burnable reactor poison, particularly in nuclear marine propulsion systems, and for tumor treatment in neutron therapy.

The naturally occurring abundance for the isotopes of gadolinium is shown in Table I along with the thermal capture cross section as reported in the *Atlas of Neutron Resonances* [4].

Table I. Naturally occurring abundances for the isotopes of gadolinium and thermal capture cross-section values from Ref. 4.

Isotope	Abundance (%)	Thermal Capture Cross Section (b)
^{152}Gd	0.20	735 +/- 20
^{154}Gd	2.18	85 +/- 12
^{155}Gd	14.80	60 900 +/- 500
^{156}Gd	20.47	1.8 +/- 0.7
^{157}Gd	15.65	254 000 +/- 815
^{158}Gd	24.84	2.2 +/- 0.2
^{160}Gd	21.86	1.4 +/- 0.3

The resolved resonance region evaluations for isotopes of gadolinium currently in the ENDF/B-VII.1 library come from the compilation in Ref. 4. The work done on the gadolinium isotopes was part of the WPEC: NEA Working Party on Evaluation Cooperation Subgroup-23 on the International Library of Fission Product Evaluations in 2004 and 2005 [5].

Table II presents and compares the upper energy limits for the resolved resonance region evaluations for ENDF/B-VII.1, JENDL-4.0, and JEFF-3.2. Of physical significance to the evaluation of the experimental data in the resolved resonance region, the normalized penetrability $\left(P_1(E)/P_0(E)\right)$ is reported at the energy of the end of the resolved resonance region for the ENDF/B-VII.1 evaluations. The physical significance of the normalized penetrability is that, among other factors, it can be used to judge the importance of p-wave resonances to the angle-integrated cross section. Consequently, no p-wave resonances are reported for gadolinium isotopes ^{155}Gd or ^{157}Gd; however, for the other isotopes, p-wave resonances need to be considered and are observed in the experimental data.

Table III further shows the different gadolinium evaluations by presenting the thermal cross-section values

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(at 0.0253 eV) and the resonance capture integrals calculated from the three different nuclear data libraries. Out of the three nuclear data libraries, only ENDF provides uncertainty information. Isotopes, 155, 157, and 160 have resonance parameter covariance matrices reported, and all five isotopes have a multigroup cross-section covariance matrix.

Table II. Gadolinium Isotope Upper Energy Limits for the Resolved Resonance Region Evaluations for ENDF/B-VII.1, JEFF-3.2 and JENDL-4.0

	ENDF-VII.1	JEFF-3.2	JENDL-4.0	P_1/P_0
^{155}Gd	183.3	181.8	181.8	0.00057
^{156}Gd	2227	1580	2214	0.0069
^{157}Gd	306.6	215	303.7	0.00096
^{158}Gd	9980	6037.6	6580	0.030
^{160}Gd	9663	2883.7	4224	0.029

Table III. Thermal Capture Cross-Section Values (Top) and Resonance Capture Integrals (Bottom) Calculated from ENDF-VII.1, JEFF 3.2, and JENDL 4.0*

σ_{cap} (b) RI (b)	ENDF-VII.1	JEFF 3.2	JENDL 4.0
^{155}Gd	60 730 +/- 1858 (349) 1539	60 732 1546	60 736 1560
^{156}Gd	1.8 +/- 0.7 108	1.5 100	1.8 106
^{157}Gd	252 890 +/- 10 119 (151) 759	253 250 763	253 220 784
^{158}Gd	2.2 +/- 0.2 68	2.5 63	2.2 72
^{160}Gd	1.4 +/- 0.4 (0.2) 8.2	0.8 8.5	0.8 11.8

*The reported uncertainty on the thermal values is shown for ENDF-VII.1 with the portion coming from the resonance parameter covariance matrix indicated in parentheses.

METHODS

The final evaluations of the five most abundant isotopes of gadolinium will report the uncertainty in the evaluated resonance parameters through a resonance parameter covariance matrix. The covariance between the resonance parameters of the different isotopes will be reported.

It is intuitive for the resonance parameters of all five isotopes to be correlated through the evaluation process. In this resonance analysis, as is often the case, experimental measurements on natural samples are being evaluated. Therefore, the uncertainty information of the resonance parameters of the individual isotopes is all tied together through the cross-isotope correlations established in the analysis methodology. The joint covariance matrix will be calculated by using the Generalized Linear Least-Squares (GLLS) updating technique implemented in SAMMY:

$$M' = (M^{-1} + G^t V^{-1} G)^{-1} \qquad (1)$$

where M' is the posterior resonance parameter covariance matrix, M is the prior resonance parameter covariance matrix, V is the covariance matrix for the experimental data being analyzed, and G is the sensitivity matrix of the cross section that represents the experimental data being evaluated to the resonance parameters. The prior resonance parameter covariance matrix, M, is a block diagonal in the five isotopes; the posterior resonance parameter covariance matrix, M', is a full matrix.

A full resonance parameter covariance matrix, including the cross-isotope covariances, is necessary to accurately reflect the state of knowledge of the cross section simultaneously, both for the individual isotopes and for their combination in the calculation of the cross section of the natural element of gadolinium. Only through reporting cross-isotope covariances can the evaluator accurately reflect the fact that the cross section for the natural element (the sum of the individual isotopes) can be better known (i.e., it can have a smaller variance) than each of the cross sections of any of the individual isotopes. It follows that an experimental uncertainty on the order of 10% for a measurement of a natural sample does not imply that the cross section for each of the individual isotopes is also known to approximately 10%.

RESULTS

In this section, we provide a demonstration of the consequences of reporting the cross-isotope correlations that are created by the evaluation of experimental data from natural isotopes. We consider the energy region of 30 to 45 eV for an experimental measurement of the capture cross section using a natural gadolinium sample, as reported in Ref. 1 and plotted in Fig. 1. At first, we will only consider the effect of the statistical uncertainty on the experimental data. All of the systematic sources of uncertainty, such as uncertainties in the sample dimensions and experimental resolution function are neglected in this initial analysis but will be rigorously treated in the final covariance evaluation.

In the energy region under consideration, 30 to 45 eV, only the isotopes of ^{155}Gd, ^{156}Gd, and ^{157}Gd have observed resonances. The statistical uncertainty from the experimental data is propagated to uncertainty in determining the resonance parameters for those three isotopes assuming no prior knowledge. The upper triangular portion of the joint resonance parameter correlation matrix is presented in Fig. 2.

As we have argued above, due to resonance overlap between different isotopes, it is natural for correlations to arise between resonance parameters of the three isotopes affecting the cross section of the natural sample in this energy region. An error in the resonance parameters of one isotope will affect the certainty of the resonance parameters of the other isotopes. This is particularly evident in the appearance of the strong correlations in the upper left-hand corner of Fig. 2, where the resonance parameters of ^{155}Gd are correlated to the resonance parameters of ^{157}Gd.

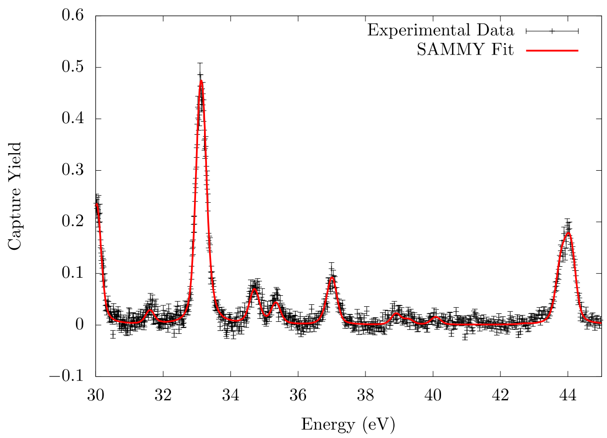

Fig. 1. Experimental capture cross-section measurement based on a natural gadolinium sample plotted with one standard deviation error bars arising solely from statistical uncertainty.

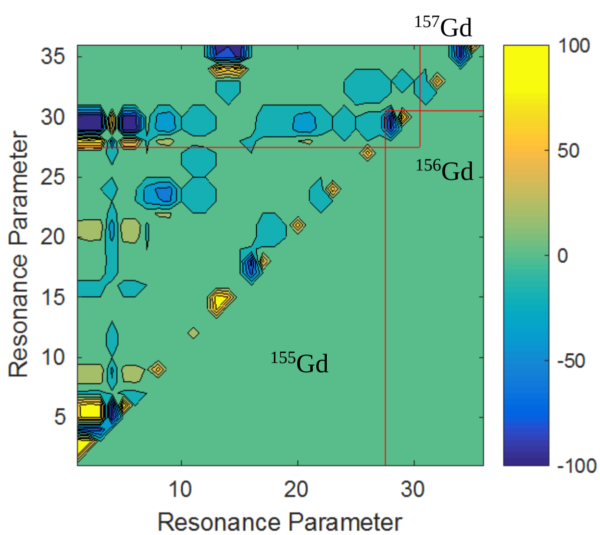

Fig. 2. Upper triangular portion of the joint resonance parameter correlation matrix for resonances of ^{155}Gd, ^{156}Gd, and ^{157}Gd between 30 and 45 eV. Scale is shown in percent.

Previously, however, cross-isotope correlations have not been reported in general-purpose nuclear data libraries. Figure 3 clearly demonstrates one of the consequences of neglecting the cross-isotope correlations. Figure 3 shows, in red, the relative uncertainty on the calculated cross section that corresponds to the experimental measurement shown in Fig. 1 as propagated from the full (with cross-isotope correlations) resonance parameter covariance matrix corresponding to Fig. 2. The green curve in Figure 3 corresponds to the propagated uncertainty from the resonance parameter covariance matrix if the cross-isotope correlations are neglected; only the correlations within the red squares of the individual isotopes are considered. The variance on each of the resonance parameters remains the same.

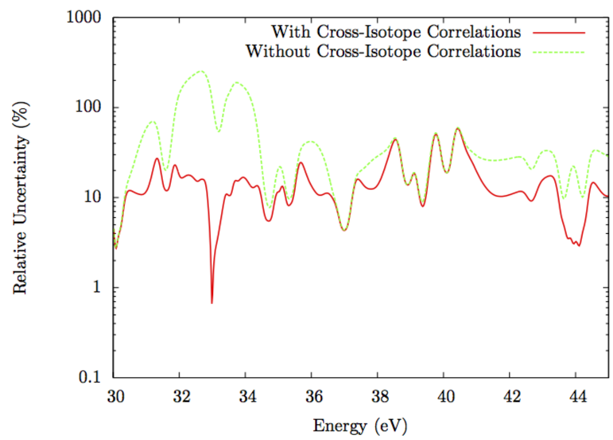

Fig. 3. Relative uncertainty on the cross section corresponding to Fig. 1 propagated from the resonance parameter covariance matrix deduced from the statistical uncertainty on the experimental data presented in Fig. 1.

Figure 3 shows that the systematic methodology of generalized linear least-squares results in a reasonable amount of uncertainty when the cross-isotope correlations are included. However, if the cross-isotope correlations are neglected, the propagated relative uncertainty jumps to rather large values at certain incident neutron energies. In this case, it is evident that the cross-isotope correlations are such that the amount of uncertainty on the cross section of a natural sample is reduced compared to the propagated uncertainty if it is assumed that the individual isotopes are uncorrelated.

Figure 4 shows the capture cross section for the individual isotopes of gadolinium. Gadolinium-156 only has one observed resonance in the energy region of 30 to 45 eV. However, that resonance dominates the cross section of the natural sample around 33 eV, as shown in Fig. 1. It is evident from Fig. 3 that when the correlations between ^{156}Gd and the other isotopes are neglected, the propagated uncertainty in the cross section of the natural sample increases significantly. The same effect is also observed when resonances ^{155}Gd and ^{157}Gd overlap around 44 eV.

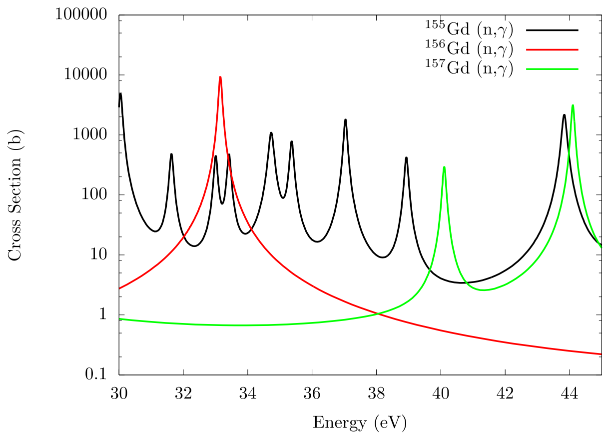

Fig. 4. Capture cross section of the individual isotopes of gadolinium.

CONCLUSIONS

A reevaluation of the resolved and unresolved resonance region of the five most abundant isotopes of gadolinium is ongoing as a collaboration between ORNL and IRSN in support of the NCSP nuclear data request. The new evaluation seeks to resolve some of the discrepancies between the experimentally measured data of different research groups. Through a systematic evaluation of the available experimental data, it is hoped that the new evaluation will also resolve some of the discrepancies noted between computational simulation and experimental measurements of integral experiments with significant sensitivities to the cross section of gadolinium.

The new evaluation will also deliver a covariance matrix with cross-isotope covariances that are a natural by-product of the analysis of natural gadolinium samples in cross-section measurements. A full covariance matrix, a more accurate representation of the confidence in the evaluated cross sections, will enable more reliable propagated uncertainty studies for systems containing gadolinium.

ACKNOWLEDGMENTS

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