Measurement and analysis of $^{155,157}$Gd($n,\gamma$) from thermal energy to 1 keV

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Abstract. We have measured the capture cross section of the $^{155}$Gd and $^{157}$Gd isotopes between 0.025 eV and 1 keV. The capture events were recorded by an array of 4 C$_6$D$_6$ detectors, and the capture yield was deduced exploiting the total energy detection system in combination with the Pulse Height Weighting Techniques. Because of the large cross section around thermal neutron energy, 4 metallic samples of different thickness were used to prevent problems related to self-shielding. The samples were isotopically enriched, with a cross contamination of the other isotope of less than 1.14%. The capture yield was analyzed with an R-Matrix code to describe the cross section in terms of resonance parameters. Near thermal energies, the results are significantly different from evaluations and from previous time-of-flight experiments. The data from the present measurement at n_TOF are publicly available in the experimental nuclear reaction database EXFOR.

1 Introduction

The large capture cross section of gadolinium has considerable influence on applications in nuclear technologies [1, 2] and has significant impact in neutrino physics, hadron therapy and nuclear astrophysics.

The values of the $^{155}$Gd and $^{157}$Gd thermal cross sections retrieved from the experimental nuclear reaction database EXFOR [3] reported in refs. [4–9] are reported together with ENDF/B-VIII.0 [10], CENDL-3.1 [11], JEFF-3.3 [12] and JENDL-4.0 [13] nuclear data libraries and the compilation by Mughabghab [14] in Table 1. Other measurements are not listed because they are not direct measurements of the cross section, whose value cannot be reconstructed from the reported experimental observables. The tabulated cross sections show that large deviations, as high as 11%, with respect to the last evaluation, namely ENDF/B-VIII.0, are present in literature.

The request for high accuracy data for the gadolinium isotopes with the largest capture cross sections [15] motivated a new measurement of these two isotopes at the neutron time-of-flight facility n_TOF [16] at CERN. The details of the experiment, data analysis and results of the n_TOF experiment have been published in ref. [9]. In this conference proceedings we prove the quality of the R-matrix analysis of the experimental data and provide required details to use the n_TOF data available on EXFOR for future evaluations.

2 Measurement

The capture measurement was performed at the n_TOF facility at CERN in 2016 [9]. The longest flight path, at 184 m from the neutron source, was used to take advantage of the better energy resolution. The experimental setup consisted of an array of 4 C$_6$D$_6$ for the measurement of the capture events and of a $^6$Li-based detector for the determination of the neutron flux impinging on the gadolinium samples.

For each isotope, two samples of different thickness were used. The areal density of the $^{155}$Gd samples was $(1.244\pm0.004)\times10^{-4}$ and $(1.236\pm0.012)\times10^{-5}$ atoms/b, whereas for the $^{157}$Gd samples it was $(2.339\pm0.006)\times10^{-4}$ and $(5.74\pm0.12)\times10^{-6}$ atoms/b. The samples were in the form of self-supporting metal discs, highly enriched in the isootope of interest. In addition to the 4 gadolinium samples, $^{197}$Au and lead samples were used for normalisation and for the determination of the background. All the samples were circular in shape with a radius of 1 cm. Particular care was devoted to develop a system able to guarantee an excellent repeatability of sample position and its alignment with respect to the neutron beam. An empty-sample was prepared, as a replica of the Gd samples excluding the metal disc, and it was measured during the campaign to estimate the sample-independent background induced by the neutron beam.

The flux was evaluated with respect to the $^6$Li(n,$\alpha$) reaction standard [17]. The flux detector [18] consisted of a 600 µg/cm$^2$ LiF foil placed in the beam, viewed off-beam by 4 silicon detectors ($5\,\text{cm}\times5\,\text{cm}\times300\,\mu\text{m}$). In the data analysis a correction to take into account the absorption of neutrons passing through the LiF foil was applied.

3 Analysis

The total energy principle was applied by combining the detection system based on C$_6$D$_6$ liquid scintillation detectors with the Pulse Height Weighting Technique (PHWT) [19], which assures the proportionality of the detection efficiency and the corresponding γ-ray energy. As a consequence, this technique makes the efficiency to detect a capture event proportional to the Q-value of the nuclear reaction. A problem arises when the sample contains more than one isotope, since the Q-value is different for each isotope. Therefore, to apply this well-established measurement principle, an approximation was required. In the simultaneous resonance shape analysis of the capture yields with the SAMMY [20] R-Matrix code, the abundance of the contaminant isotopes was scaled accordingly to the Q-value (i.e. the abundance of a contaminant is divided by its Q-value of the neutron capture reaction and is multiplied by that of the isotope of interest). This modification, which introduced a negligible bias on the multi-
ple scattering correction, made the calculated capture yield consistent with the experimental capture yield.

In Table 2 the abundances of the gadolinium isotopes in the $^{155}$Gd and $^{157}$Gd samples are listed together with the corresponding quantity used in the resonance shape analysis. It is worth noticing that the cross contamination of the $^{155}$Gd isotope in the enriched $^{157}$Gd samples and the one of the $^{157}$Gd isotope in the enriched $^{155}$Gd sample is less than 1.14%.

## 4 Discussion on uncertainties

The systematic uncertainties of the experimental capture yield come from: (i) normalisation, (ii) PHWT, (iii) background subtraction, (iv) sample characterisation, (v) neutron flux shape, and, in the energy region below 1 eV, (vi) the uncertainty related to the beam interception factor. More details on the assessment of the different components can be found in ref. [9]. We remind the reader that the individual contributions to the total uncertainty were very similar. It can be argued that at thermal energy the uncertainty component (v) due to neutron flux shape already incorporates the uncertainty component (vi) related to the beam interception factor and therefore the total uncertainty is slightly lower than reported in ref. [9] being reduced to 3% and 3.4%, respectively for the thermal cross section of $^{155}$Gd(n,γ) and $^{157}$Gd(n,γ).

To further decrease the total uncertainty, the samples are now being analyzed in order to reduce the uncertainty related to the sample mass and its distribution. In particular a transmission experiment on the same samples is ongoing at the GELINA facility at JRC Geel (Belgium) and a proton elastic backscattering experiment is also foreseen for the near future.

## 5 Results

From a simultaneous resonance shape analysis of the capture yields, using the R-Matrix code SAMMY, the cross sections were parametrized in terms of resonance parameters in the energy region from thermal to 1 keV neutron energy. An example of the good quality of the fitting procedure is shown in Fig.1 for both gadolinium isotopes.

![Figure 1](image1.png)

Figure 1. (Color online) $^{155,157}$Gd(n,γ) capture yields from the present experiment and results of the resonance shape analysis (RSA) in the neutron energy region from 1 to 40 eV.

The results of the resonance shape analysis were used to reconstruct the cross section and in particular to evaluate the thermal cross section and the Westcott factor. These values are reported in Table 3 for $^{155}$Gd and $^{157}$Gd.

The updated resonance parameters were also used to determine the basic statistical properties of the neutron resonances in $n+^{155}$Gd and $n+^{157}$Gd systems. Neutron strength function and level spacing resulted fully consistent with values in literature, whereas the estimated $\Gamma_{\gamma} =$

\begin{table}[h]
\centering
\begin{tabular}{|l|l|l|l|l|l|l|}
\hline
Reference & Year & Type & $n + ^{155}$Gd & deviation to ENDF/B-VIII.0 & $n + ^{157}$Gd & deviation to ENDF/B-VIII.0 \\
\hline
Møller [4] & 1960 & TOF & 58.9(5)$^a$ & $-3.3\%$ & 254(2)$^a$ & 0.27\% \\
Ohno [5] & 1968 & TOF & 61.9(6)$^a$ & 1.7\% & 248(4)$^a$ & $-2.10\%$ \\
Leinweber [6] & 2006 & TOF & 60.2 & $-1.1\%$ & 226 & $-11\%$ \\
Noguere [7] & 2011 & Pile oscillation & 61.9(15) & 1.7\% & & \\
Choi [8] & 2014 & Spectra averaged & 56.7(21) & $-6.9\%$ & 239(6) & $-5.7\%$ \\
Mughabghab [14] & 2009 & Compilation & 60.9(5) & 0.02\% & 250.0(8) & 0.27\% \\
CENDL-3.1 [11] & 2009 & Evaluation & 60.888 & - & 254.01 & 0.27\% \\
JENDL-4.0 [13] & 2011 & Evaluation & 60.735 & $-0.25\%$ & 253.25 & $-0.03\%$ \\
JEFF-3.3 [12] & 2017 & Evaluation & 60.89 & - & 254.5 & 0.47\% \\
ENDF/B-VIII.0 [10] & 2018 & Evaluation & 60.89 & - & 253.32 & - \\
Mastromarco [9] & 2019 & TOF & 62.2(22) & 2.2\% & 239.8(84) & $-5.3\%$ \\
\hline
\end{tabular}
\caption{$^{155}$Gd and $^{157}$Gd thermal cross sections (in kb) as reported in literature, compilation [14] and evaluations. The results from the present measurement are reported for comparison.}
\end{table}
Table 2. Isotopic composition of the gadolinium isotopes and corresponding scaled quantity used in the resonance shape analysis (the Q-value of the (n,γ) reaction adopted for the calculation of the abundance used in SAMMY is reported).

| Isotope (natural abund.) | Q-value (MeV) | 155Gd samples | Isotopic composition | 157Gd samples | Isotopic composition |
|--------------------------|---------------|---------------|---------------------|---------------|---------------------|
| 155Gd (0.2%)             | 6.246         | 0.03%         | 0.03%               | < 0.01%       | < 0.008%            |
| 155Gd (2.18%)            | 6.435         | 0.63 ± 0.02%  | 0.47 ± 0.015%       | 0.04 ± 0.01%  | 0.032 ± 0.008%      |
| 155Gd (14.80%)           | 8.536         | 91.74 ± 0.18% | 91.74 ± 0.18%       | 0.29 ± 0.01%  | 0.31 ± 0.01%        |
| 155Gd (20.47%)           | 6.36          | 5.12 ± 0.18%  | 3.81 ± 0.13%        | 1.68 ± 0.01%  | 1.346 ± 0.008%      |
| 155Gd (15.65%)           | 7.937         | 1.14%         | 1.06%               | 82.32 ± 0.01% | 88.32 ± 0.01%       |
| 155Gd (24.84%)           | 5.943         | 0.94 ± 0.09%  | 0.65 ± 0.06%        | 9.10 ± 0.01%  | 6.814 ± 0.007%      |
| 156Gd (21.86%)           | 5.635         | 0.40 ± 0.07%  | 0.26 ± 0.05%        | 0.57 ± 0.01%  | 0.405 ± 0.007%      |

106.8(14) and 101.1(20) meV, respectively for 155Gd and 157Gd, are slightly different from literature.

6 Conclusion

The 155Gd(n,γ) and 157Gd(n,γ) cross section measurements were performed at the n_TOF facility. They provided thermal cross sections at E_n = 0.0253 eV of 62.2(20) kb and 239.8(81) kb, respectively. The percentage deviation to ENDF/B-VIII.0 is sizable, in particular for 157Gd, while fair agreement was found for 155Gd. In terms of standard deviations, the present 157Gd(n,γ) cross section is approximately two sigma away from evaluations. To make this result more conclusive, a series of complementary measurements and studies are ongoing to reduce the uncertainty related to the present R-matrix parametrization.

The results of the capture yields obtained at n_TOF are available in the EXFOR database, and they can be used for future evaluations.

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References

[1] F. Rocchi, A. Guglielmelli, D. M. Castelluccio and C. Massimi, Eur. Phys. J. Nuclear Sci. Technol. 3, 21 (2017)
[2] F. Rocchi, A. Guglielmelli, P. Console Camprini, C. Massimi and Luiz Leal, Annals of Nuclear Energy 132, 537 (2019)
[3] N. Otuka, et al., Nucl. Data Sheets 120, 272 (2014)
[4] H. Bjerrum Møller, F. J. Shore, and V. L. Sailor, Nucl. Sci. Eng. 8, 183 (1960)
[5] Y. Ohno, et al., Japanese report to EANDC, Number 10, p. 1 (1968)
[6] G. Leinweber, et al., Nucl. Sci. Eng. 154, 261 (2006)
[7] G. Noguere, P. Archier, A. Gruel, P. Leconte and D. Bernard, Nucl. Instrum. & Methods A 629, 288 (2011)
[8] H.D.Choi, et al., Nucl. Science & Eng. 177, 219 (2014)
[9] M. MastroMarco, et al., Eur. Phys. J. A 55, 9 (2019)
[10] D. A. Brown, et al., Nucl. Data Sheets 148, 1 (2018)
[11] Z. G. Ge, Y. X. Zhuang, J. X. Xie, Z. H. Xie, Journal of the Korean Physical Society 59(2), 1052 (2011)
[12] A. Plompen, et al., The Joint Evaluated Fission and Fusion Nuclear Data Library, JEFF-3.3, Eur. Phys. J. A, accepted for publication
[13] K. Shibata, et al., J. Nucl. Sci. Technol. 48, 1 (2011)
[14] S. F. Mughabghab, Atlas of Neutron Resonances (Elsevier, Amsterdam, 2006)
[15] A. Plompen, et al., The NEA High Priority Nuclear Data Request List for future needs, Int. Conf. on Nuclear Data for Science and Technology, (2007) 765
[16] C. Guerrerro, et al., Eur. Phys. J. A 49, 27 (2013)
[17] A. D. Carlson, et al., Nucl. Data Sheets 110, 3215 (2009)
[18] S. Marrone, et al., Nucl. Instrum. & Methods A 517, 389 (2004)
[19] P. Schillebeeckx, et al., Nucl. Data Sheets 113, 3054 (2012)
[20] N. M. Larson, "Updated Users Guide for SAMMY: Multilevel R-matrix Fits to Neutron Data Using Bayes Equations, SAMMY", Computer Code, Report No. ORNL/TM-9179/R7, Oak Ridge National Laboratory, 2008