Neutron capture and total cross-section measurements of ^{155}Gd and ^{157}Gd at ANNRI in J-PARC

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Abstract. Neutron capture and total cross sections of ^{155}Gd and ^{157}Gd were measured with the neutron time-of-flight method in Accurate Neutron-Nucleus Reaction measurement Instrument at Material and Life Science Experimental Facility of the Japan Proton Accelerator Research Complex using the NaI (Tl) spectrometer and Li-glass detectors. Preliminary cross sections were obtained in the neutron energy region from 4 meV to 100 meV. The derived cross sections agree with evaluated values in JENDL 4.0 and the experimental results by Mastromarco but were not consistent with those by Leinweber.

1 Introduction

Gadolinium (Gd) is used as a burnable poison to compensate for decrease in reactivity under operations of commercial light water reactors, since two nuclides, ^{155}Gd and ^{157}Gd, have large capture cross sections for thermal neutrons. Thus, it is essential for safe and reliable operation of nuclear power plants to know accurate values of neutron capture cross-sections of these nuclides. The capture cross section data of ^{155}Gd and ^{157}Gd at neutron energy of 0.0253 eV available in the literature are summarized in Table 1 with comparison to evaluated values in JENDL 4.0[1]. Between the reported capture cross sections of ^{157}Gd, there is non-negligible inconsistency. Some researchers have pointed out that the current evaluation of the cross section for ^{157}Gd is inconsistent with results of some integral experiments. [2] Therefore, neutron capture and total cross sections of ^{155and157}Gd were measured with a TOF method at Accurate Neutron Nucleus Reaction measurement Instrument (ANNRI) in Japan Proton Accelerator Research Complex (J-PARC).

Table 1. Capture cross section data of ^{155,157}Gd in the literatures.

Reference	year	^{155}Gd (kb)	^{157}Gd (kb)
Tattersall [3]	1960	49.8±0.6	213±2
Møller [4]	1960	60.6±0.5	254±2
Groshev [5]	1962	61±5	240±12
Sun [6]	2003	59.1±4.6	232±12
Leinweber [7]	2006	60.2	226
Choi [8]	2014	56.7±2.1	239±6
Mastromarco [9]	2019	62.2±2.2	239.8±8.4
JENDL 4.0 [1]	2011	60.74	253.2

2 Facilities and samples

Pulsed neutrons are produced using spallation reactions in the mercury target of the MLF [10]. Produced neutrons are slowed down in a supercritical hydrogen moderator and transported to the ANNRI, which is designed for neutron TOF measurements. In the ANNRI, there are two gamma-ray spectrometers and one neutron detector system [11]. More details about the ANNRI beam line are given in Ref.[12]. In this measurement, the NaI(Tl) detectors were used for capture cross-section measurement and the Li-glass detectors were used for total cross-section measurement. J-PARC was operated at a repetition rate of 25 Hz, a power of 210 kW and in the "double-bunch mode", in which each proton pulse consists of two bunches (each with a width of 100 ns) at intervals of 600 ns. The proton beam intensity per shot was stable within 2% in FWHM.

Samples were isotopically enriched Gadolinium nitrate crystalized on pure Al foils with a thickness of 10 μm. Eight sample sheets with effective thickness of 200 nm and twelve sheets with 2 μm thickness were prepared for each ^{155}Gd and ^{157}Gd. To estimate influence due to thickness variation of the samples, thickness of Gd is measured with X-ray Fluorescence Spectrometry at 5 points of all sample sheets. Sample size was 30 mm × 30 mm and it was enough large compared to the neutron beam size at the sample positions. Details of the sample sheets were summarized in Table 2. Isotopic compositions of the ^{155}Gd and ^{157}Gd samples were determined by thermal ioniza-

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tion mass spectrometry (TIMS). The isotopic enrichment for the ^{157}Gd samples was 88.3 % with 9.1 %, 1.7 % and 0.3 % isotopic contaminations of ^{158}Gd, ^{156}Gd and ^{155}Gd, respectively. That for the ^{155}Gd samples was, respectively, 91.7 % with 5.1 % and 1.1 % isotopic contaminations of ^{156}Gd and ^{157}Gd.

Table 2. Characteristics of the sample sheets.

Sheet name	Effective thickness (μm)	Number of sheets	Thickness variation (%)
^{155}Gd 200 nm sheet	0.20	8	1.96
^{155}Gd 2 μm sheet	1.92	12	2.09
^{157}Gd 200 nm sheet	0.20	8	1.80
^{157}Gd 2 μm sheet	2.07	12	2.12

3 Capture cross-section measurement

For the capture cross-section experiment, the NaI(Tl) detectors installed at flight length of 27.9 m were used to measure emitted gamma-rays from neutron capture reactions. The TOF of detected events were measured with a high resolution time digitizer in event-by-event mode. The details of the experimental instruments and method are described in Ref. [13]. In the neutron capture cross-section measurement, two samples with different thickness for each nuclide were used. Thickness and measurement time of the samples were summarized in Table 3. For background estimation, measurements with Al foils and a sample holder without any sample (Blank) were also carried out.

Table 3. Effective thickness and measurement time of samples for capture cross section measurement.

Sample name	Materials	Time (h)
^{155}Gd 600 nmt	^{155}Gd 200 nm sheet × 3	4
^{155}Gd 16 μmt	^{155}Gd 2 μm sheet × 8	4
^{157}Gd 1200 nmt	^{157}Gd 200 nm sheet × 6	15
^{157}Gd 20 μmt	^{157}Gd 2 μm sheet × 10	15
Al Foils	Al Foil (10 μmt) × 5	4
Blank	(Only sample holder)	4

Figure 1 shows the obtained TOF spectrum with a TOF time bin of 25 μs. The dead time correction similar to the one proposed by Katabuchi et al[13] was applied. The frame-overlap backgrounds were estimated and subtracted in almost the same manner as that described in Ref. [14]. The capture yeild for the Gd samples were obtained.

In the analysis, to obtain absolute capture cross section, ratios of capture yields of the thick to those of thin samples were calculated. By taking ratios of capture yields, uncertainties due to neutron intensity and detection efficiency of the spectrometer were canceled out, thus absolute neutron capture cross sections are deduced with high accuracy. Details of this method is written in Ref. [15]. The obtained transmission ratios are shown in Figure 2. By taking the ratios, thermal bumps and many dips due to influence of diffraction (as shown in Figure 1) were disappeared.

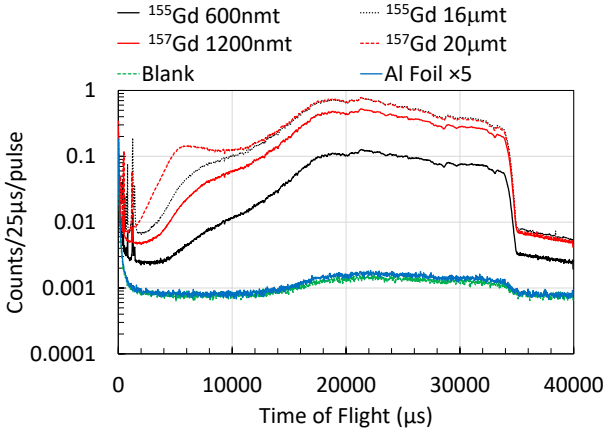

Figure 1. TOF spectra of the ^{155}Gd and ^{157}Gd samples, the Al foils and Blank. The TOF time bin is 25 μs. Each TOF spectrum was normalized with the number of the proton beam pulses.

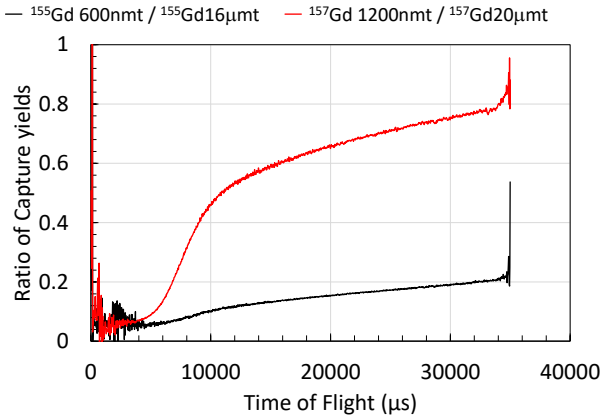

Figure 2. Ratios of capture yields for the ^{155}Gd and ^{157}Gd samples.

Preliminary capture cross sections of ^{155}Gd and ^{157}Gd were derived in the neutron energy range from 4 meV to 100 meV using the ratios of capture yields. The derived capture cross sections and uncertainties are shown in Figure 3. In the analysis, the statistical uncertainty, systematic uncertainty and uncertainty due to neutron intensity were considered. Figure 4 shows the obtained capture cross section of ^{157}Gd at neutron energy of 0.0253 eV with comparison to the values in the literatures and the evaluated value in JENDL 4.0. Comparison to the results of latest two TOF experiments by Mastromarco and Leinweber and the evaluated value in JENDL-4.0, the derived capture cross section of ^{157}Gd agreed within uncertainties to the evaluated value and that by Mastromarco but was not consistent with that by Leinweber.

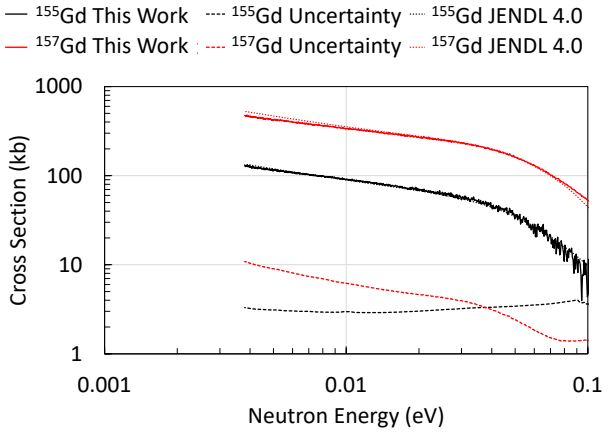

Figure 3. Derived preliminary capture cross sections of ^{155}Gd and ^{157}Gd and uncertainties with comparison to the values of JENDL-4.0 at 300 K.

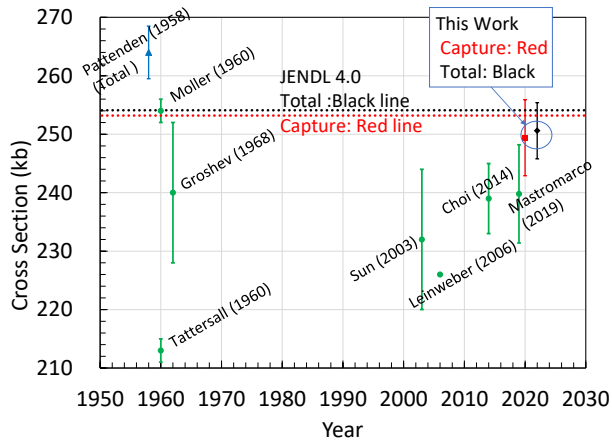

Figure 4. Obtained preliminary capture cross section of ^{157}Gd at neutron energy of 0.0253 eV, and comparisons to capture cross sections (Green) and a total cross section (Blue) in literatures and the evaluated values in JENDL 4.0.

4 Total cross-section measurement

For the neutron total cross section measurement, a ^{6}Li-glass scintillation detector installed at flight length of 28.7 m was used. To determine background, a ^{7}Li-glass detector whose scintillator has same size and chemical component of the ^{6}Li-glass scintillator was set on the upstream side of the ^{6}Li-glass detector. More details about the Li-glass detectors are given in Ref.[16]. In the measurement, a set of four Gd sheets with effective thickness of 200 nm were used for each ^{155}Gd and ^{157}Gd measurement. A measurement of four Al foils with thickness of 10 μm was also carried out to estimate "sample out" conditions. The total measuring time was about 4 hours for the ^{155}Gd sheets, about 6 hours for the ^{157}Gd sheets and about 4 hours for the Al foils.

TOF spectra of the ^{155}Gd and ^{157}Gd samples and the Al foils with the ^{6}Li-glass and ^{7}Li-glass detectors are shown in Figure 5. Because the ^{7}Li-glass detector has slight sensitivity to neutron, thermal bump is also observed in the measured spectra with the ^{7}Li-glass detector.

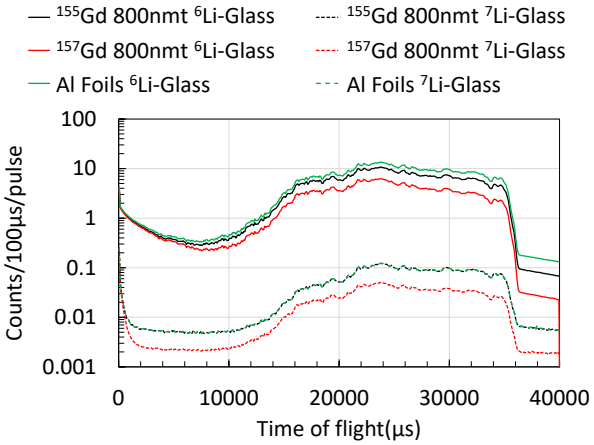

Figure 5. TOF spectra of the ^{155}Gd and ^{157}Gd samples and the Al foils with ^{6}Li-glass and ^{7}Li-glass detectors. Each TOF spectrum was normalized with the number of proton beam pulses.

The analysis procedure was almost the same manner as that described in Ref. [15]. Lower discrimination, dead-time correction and background subtraction were applied. Using the deduced relative neutron beam intensities, transmission ratios the ^{155}Gd and ^{157}Gd samples to the Al foils were obtained. The obtained transmission ratios are shown in Figure 6.

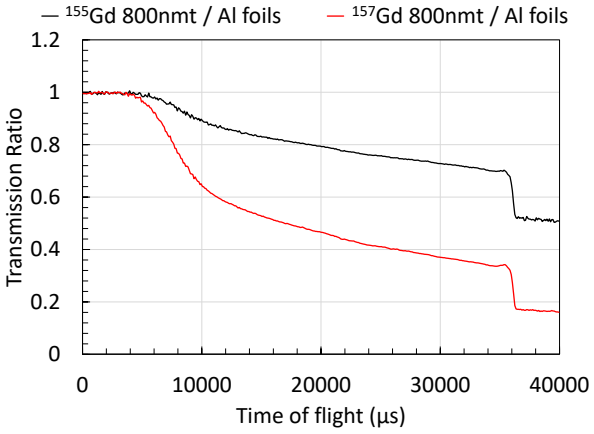

Figure 6. Transmission ratios of the ^{155}Gd and ^{157}Gd samples to the Al foils.

Preliminary total cross sections of ^{155}Gd and ^{157}Gd were derived in the neutron energy range from 4 meV to 100 meV, using the transmission ratio and the areal density of the ^{155}Gd and ^{157}Gd samples. The derived total cross sections of ^{155}Gd and ^{157}Gd are shown in Figure 7. In the analysis, the statistical uncertainty, systematic uncertainty and uncertainty due to neutron intensity were considered. The derived uncertainties are also shown in Figure 7. Obtained total cross-section values of ^{157}Gd at neutron energy of 0.0253 eV was also plotted in Figure 4 with comparison

to the result by Pattenden [17] and the evaluated value in JENDL 4.0. The result is consistent with evaluated values in JENDL 4.0 and the obtained capture-cross section.

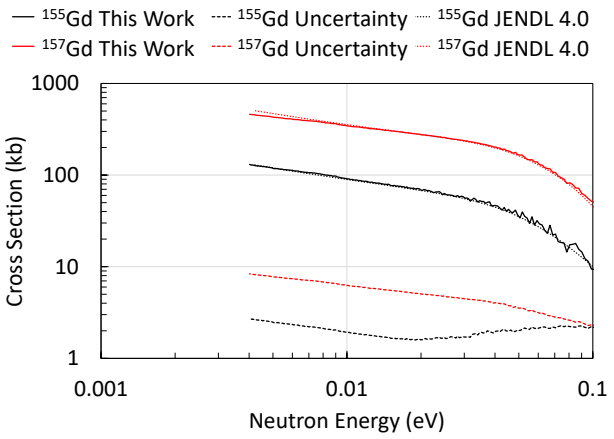

Figure 7. Derived preliminary total cross sections of ^{155}Gd and ^{157}Gd and uncertainties of the cross sections and together with values of JENDL-4.0 at 300 K.

5 Conclusion

Preliminary neutron capture and total cross section of ^{155}Gd and ^{157}Gd were measured in the neutron energy region from 4 meV to 100 meV. The obtained capture and total cross sections are consistent. The derived cross sections were in agreement with evaluated values in JENDL 4.0 and the experimental results by Mastromarco but were not consistent with those by Leinweber. More detailed discussions for systematic uncertainties due to the samples are in progress to obtain final results.

The neutron experiments at the MLF of the J-PARC were performed under the user program (Proposal No. 2016P1001, 2015P1001 and 2012B0093).

Present study includes the result of "Research and Development for accuracy improvement of neutron nuclear data on minor actinides" entrusted to the Japan Atomic Energy Agency by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

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