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Measurements of keV-neutron capture cross sections and capture gamma-ray spectra for Sn and Gd isotopes

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Abstract. The neutron capture cross sections and capture gamma-ray spectra of Sn ($^{116-119}$Sn) and Gd ($^{155-158}$Gd) isotopes have been measured in the incident neutron energy region from 10 to 100 keV and at 550 keV. Pulsed keV neutrons were produced by the ^{7}Li(p,n)^{7}Be reaction. Capture gamma rays were detected with a large anti-Compton NaI(Tl) spectrometer by means of a time-of-flight method. A pulse-height weighting technique was applied to the observed capture gamma-ray pulse-height spectra to obtain capture yields. Using the standard capture cross sections of ^{197}Au, the capture cross sections of the Sn and Gd isotopes were derived. The capture gamma-ray spectra were obtained by unfolding the observed capture gamma-ray pulse-height spectra. The present cross section results were compared with other experimental data and evaluated values.

1 Introduction

Neutron capture cross sections and capture gamma-ray spectra are important quantities in the R&D of innovative nuclear reactors.

Those of long-lived fission products (LLFPs) are significant for the R&D of nuclear transmutation systems, because the performance of system using neutron capture reaction depends directly on these quantities. The nuclide ^{126}Sn is one of LLFPs. However, the present status of experimental data for the neutron cross sections of ^{126}Sn is very poor both in quality and in quantity: only one data was reported at thermal energy [1], because the preparation of high-purity sample is difficult and, moreover, the gamma-ray radiation from a sample causes a serious background in the cross section measurement. On the other hand, keV-neutron capture cross sections and capture gamma-ray spectra of stable Sn isotopes contain important information useful for the theoretical calculation of capture cross sections of ^{126}Sn.

The neutron capture cross sections of Gd isotopes are important in the design of thermal and fast reactors, as well as in nuclear reaction and astrophysics studies. Since a major portion of the natural Gd capture cross sections is due to ^{155}Gd and ^{157}Gd, an accurate knowledge of the cross sections for these isotopes is of importance for calculating the reactor characteristics when Gd is used as a burnable poison in light water reactors. In addition, the data are important for examining the availability of Gd as a control material for fast reactors.

Therefore, we are performing the systematic measurement of keV-neutron capture cross sections and capture gamma-ray spectra for stable Sn and Gd isotopes. In the present paper, we report on the capture cross sections and capture gamma-ray spectra of ^{116,117,118,119}Sn and ^{155,156,157,158}Gd.

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2 Experimental procedure

The experimental procedure has been described in detail elsewhere [2], so it is summarized here.

The capture cross sections and capture gamma-ray spectra of ^{116,117,118,119}Sn and ^{155,156,157,158}Gd were measured in the incident neutron energy region from 10 to 100 keV and at 550 keV, using the 3-MV Pelletron accelerator of the Research Laboratory for Nuclear Reactors at the Tokyo Institute of Technology. An experimental arrangement is shown in figure 1. Pulsed neutrons were produced by the ^{7}Li(p,n)^{7}Be reaction with a pulsed proton beam(1.5 ns width, 4 MHz repetition rate) from the Pelletron accelerator. The incident neutron spectrum on a capture sample was measured by means of a time-of-flight (TOF) method with a ^{6}Li-glass scintillation detector. A 5.0-mm diameter by 5.0-mm ^{6}Li-glass detector located 30 cm from the neutron source was used for the measurements in the 10–100 keV region, and a 102.0-mm diameter by 6.4-mm ^{6}Li-glass detector located 460 cm was used for those at 550 keV.

Each of ^{116,117,118,119}Sn and ^{155,156,157,158}Gd samples was isotopically enriched metal, and the net weight of each sample was 0.2 – 1.0 g. The characteristics of the samples are shown in table 1.

A gold (Au) sample was used as a standard. Each capture sample was located at an angle of 0 degree with respect to the proton beam direction. The distance between the neutron source and the sample was 12 cm for the measurements in the 10–100 keV region and 20 cm for those at 550 keV.

The capture gamma rays emitted from the sample were measured with a large anti-Compton NaI(Tl) spectrometer by means of a TOF method. The main NaI(Tl) detector of the spectrometer had a diameter of 15.2 cm and a length of 20.3 cm or 30.5 cm. The larger NaI(Tl) detector had 30% higher detection efficiency around 5 MeV compared to the smaller detector [3], and was used for the measurements of ^{116,117,118}Sn. One of the main detectors was centered in a

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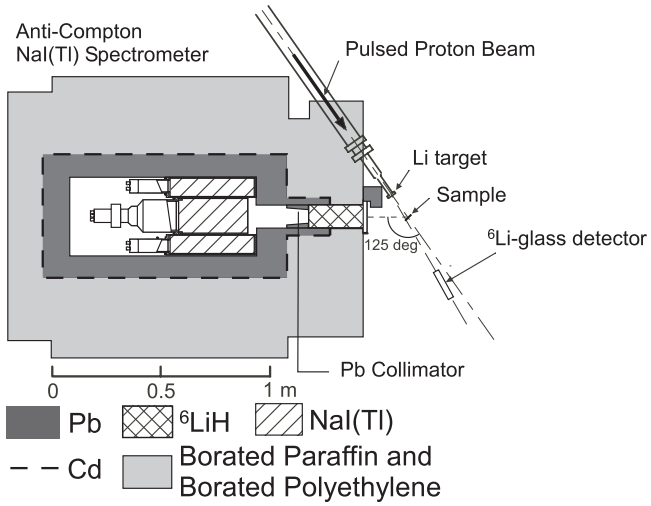

Fig. 1. Experimental arrangement in the 10–100 keV measurement.

Table 1. Characteristics of Sn and Gd samples.

Sample	^{116}Sn	^{117}Sn	^{118}Sn	^{119}Sn
Weight [g]	0.988	1.000	0.999	0.996
Thickness [mm]	1.3	0.55	0.53	0.60
Isotopic composition [%]				
^{112}Sn	<0.01	<0.1	0.07	<0.03
^{114}Sn	0.2	<0.01	0.02	0.02
^{115}Sn	0.48	<0.11	0.02	0.05
^{116}Sn	96.1	4.4	0.48	0.11
^{117}Sn	2.1	95.1	0.62	0.06
^{118}Sn	0.3	0.2	96.2	0.95
^{119}Sn	0.2	<0.05	0.79	93.8
^{120}Sn	0.5	<0.01	0.97	4.89
^{122}Sn	0.1	<0.01	0.71	0.05
^{124}Sn	<0.01	<0.01	0.12	0.04
Sample	^{155}Gd	^{156}Gd	^{157}Gd	^{158}Gd
Weight [g]	0.203	0.198	0.357	0.353
Thickness [mm]	0.1	0.1	0.2	0.2
Isotopic composition [%]				
^{152}Gd	0.04	<0.01	<0.02	<0.1
^{154}Gd	0.64	0.11	0.16	<0.1
^{155}Gd	91.74	1.96	0.81	0.96
^{156}Gd	5.11	93.79	2.21	1.7
^{157}Gd	1.12	2.53	90.96	3.56
^{158}Gd	0.94	1.20	5.08	92.0
^{160}Gd	0.41	0.41	0.80	1.82

hollow Compton-suppression NaI(Tl) detector with an outer diameter of 33.0 cm and a length of 35.6 cm. The spectrometer was set in a heavy shield consisting of borated paraffin, borated polyethylene, Cd, ^{6}LiH and potassium free lead [4]. The capture gamma rays were observed at an angle of 125 degree with respect to the proton beam direction. The signals from the spectrometer were recorded in a personal computer as two-dimensional data of pulse-height (PH) and TOF. The measurements with one of the Sn and Gd samples, the ^{197}Au sample, and no sample (Blank) were performed cyclically to average out changes in experimental conditions such as the incident neutron spectrum.

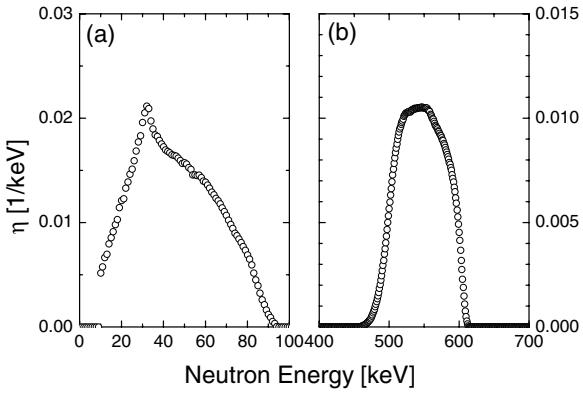

Fig. 2. Typical normalized energy spectra of incident neutrons in the (a) 10–100 keV and (b) 550 keV measurements.

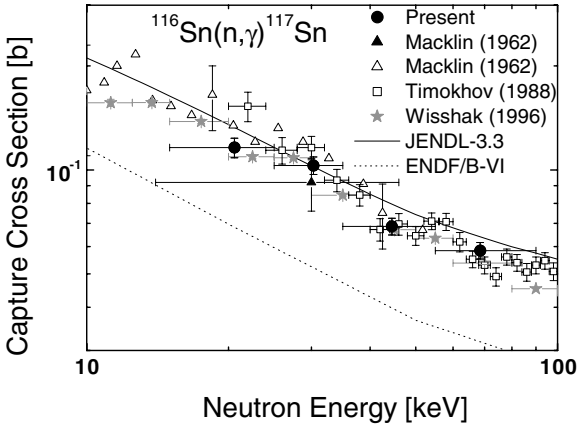

Fig. 3. Neutron capture cross sections of ^{116}Sn.

3 Data processing

The data processing method has been also given in detail elsewhere [2], so it is summarized here. The incident neutron energy spectrum on the sample was derived from the TOF spectra measured with the ^{6}Li-glass detector for the blank run. Typical normalized spectra are shown in figure 2.

In order to obtain the capture yields of individual samples and the ^{197}Au sample, a PH weighting technique [5] was applied to the net capture gamma-ray PH spectra. The number of incident neutrons in the ^{197}Au run was determined by the capture yield of ^{197}Au and the averaged capture cross section of ^{197}Au, which was obtained from the capture cross sections of ENDF/B-VI [6] and the neutron energy spectrum.

The number of incident neutrons in the Sn or Gd isotope run was derived from that in the Au run and the neutron monitor counts of the ^{6}Li-glass detector. The averaged neutron capture cross section of each Sn or Gd isotope was derived from the number of incident neutrons and the capture yield of each sample. Corrections for the neutron self-shielding and multiple-scattering in the sample were made by a Monte Carlo method [7], taking account of impurities in the sample. Moreover, other corrections were made for the gamma-ray scattering and absorption in the sample, for the effect of chemical and isotopic impurities in the sample on the capture yields.

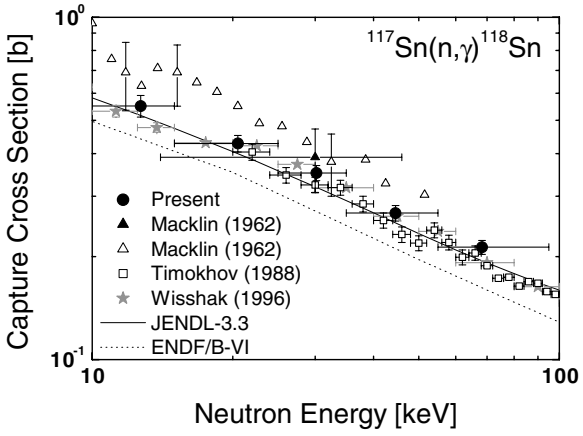

Fig. 4. Neutron capture cross sections of ^{117}Sn.

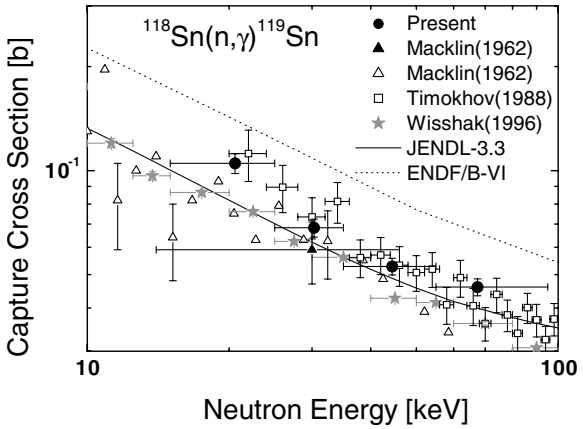

Fig. 5. Neutron capture cross sections of ^{118}Sn.

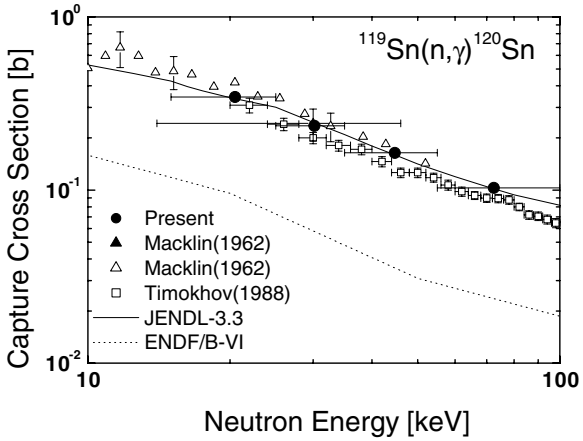

Fig. 6. Neutron capture cross sections of ^{119}Sn.

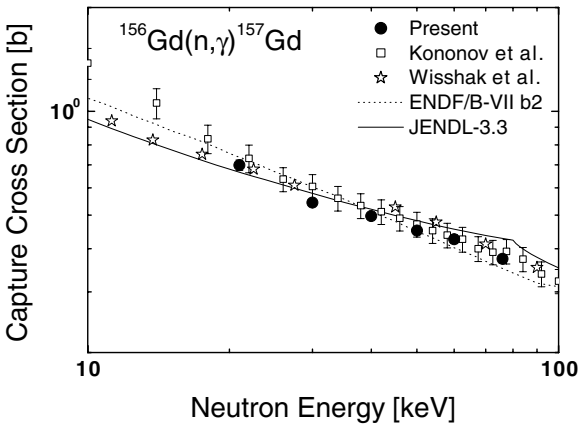

Fig. 7. Neutron capture cross sections of ^{156}Gd.

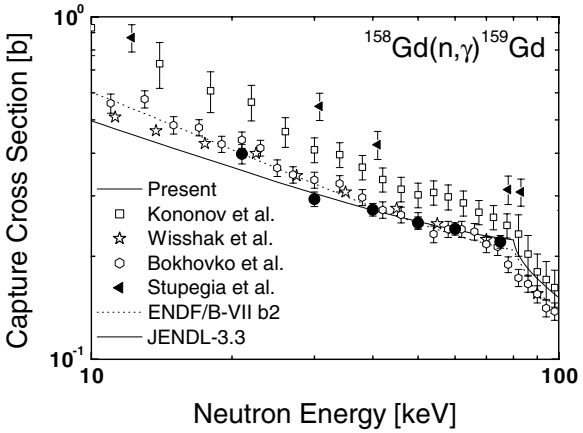

Fig. 8. Neutron capture cross sections of ^{158}Gd.

The capture gamma-ray spectra were derived by unfolding the net capture gamma-ray PH spectra with a computer code, FERDOR [8], and the response matrix of the gamma-ray spectrometer.

4 Results and discussion

The capture cross sections of ^{117,119}Sn and ^{155,157}Gd were derived with the errors from 4 to 5% in the energy region from 10 to 100 keV and at 550 keV. Those of ^{116,118}Sn and ^{156,158}Gd were derived with the errors from 5 to 8% in the energy region from 15 to 100 keV. The obtained cross sections in the energy region from 10 to 100 keV for Sn isotopes are shown in figures 3–6, and compared with previous measurements and the evaluations of JENDL-3.3 and ENDF/B-VI.

As shown in figure 3, the present results of ^{116}Sn are in agreement with those by Macklin et al. [9], Timokhov et al. [10], and Wisshak et al. [11] within the experimental errors. The present results of ^{117,119}Sn in figures 4 and 6 are in agreement with other experimental data within errors. The present results of ^{118}Sn in figure 5 are in good agreement with those by Timokhov et al. [10], but larger than those of Wisshak et al. [11] by about 20%. The results of Macklin et al. [9] are in agreement with the present ones within their large experimental errors of about 25%. The evaluations of Sn isotopes for ENDF/B-VI [12] were theoretically done before the measurements by Timokhov et al. and Wisshak et al., and the evaluated values of ^{116,117,118,119}Sn are very different from the experiments as shown in figures 3–6. On the other hand, the evaluated values of JENDL-3.3 for Sn isotopes [13] are in agreement with the experimental within 20%, because

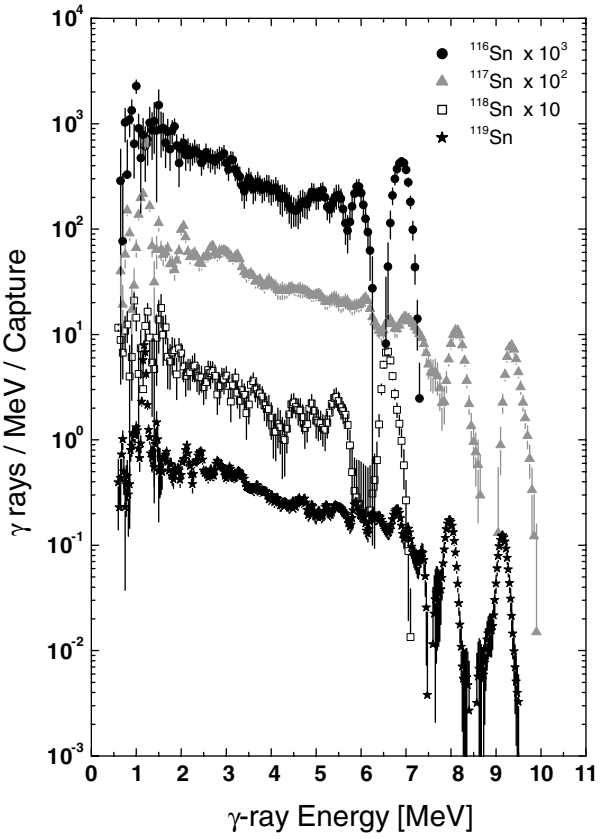

Fig. 9. The neutron capture gamma-ray spectra of ^{116,117,118,119}Sn in the neutron energy region from 15 to 100 keV.

the evaluated values were normalized to reproduce experimental data at several tens of keV. For example, the evaluated values of JENDL-3.3 for ^{116}Sn were adjusted to reproduce the capture cross section of 85 mb at 40 keV measured by Macklin, and are larger than the present ones by 10–15%, except for the values around 30 keV. It means that improvement of theoretical calculation of neutron capture reaction is important for the accurate evaluation of neutron capture cross sections of LLFPs for which measurements are difficult.

The obtained cross sections in the energy region from 10 to 100 keV for ^{156,158}Gd are shown in figures 7 and 8, and compared with previous measurements and the evaluations of JENDL-3.3 and ENDF/B-VII-b2. The results of ^{155,157}Gd have been published elsewhere [14]. As shown in figure 7, the present results for ^{156}Gd are in general agreement with those of Kononov et al. [15] and Wisshak et al. [16] and with the evaluated values in ENDF/B-Z-b2 [17] and JENDL-3.3 [18]. The present results for ^{158}Gd in figure 8 agree with those of Wisshak et al. [16] and Bokhovko et al. [19] and with the evaluated values in both ENDF/B-VII-b2 [17] and JENDL 3.3 [18]. However, the experimental values of Kononov et al. [15] and Stupegia et al. [20] are larger than any other values.

The capture gamma-ray spectra obtained from the measurements for the ^{116,117,118,119}Sn samples are shown in figure 9, each spectrum has strong transitions from the neutron capture state to the ground state, the first excited state and other low-lying states.

5 Conclusion

The neutron capture cross sections and capture gamma-ray spectra of $^{116-119}$Sn and $^{155-158}$Gd have been measured in the incident neutron energy region from 10 to 100 keV and at 550 keV. These data will provide important information on nuclear reaction mechanisms and nuclear excitation modes, and the information will be used for accurate evaluations of the neutron capture cross sections of important nuclide such as ^{126}Sn.

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