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Neutron capture cross section measurements of ^{120}Sn, ^{122}Sn and ^{124}Sn with the array of Ge spectrometer at the J-PARC/MLF/ANNRI

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Abstract. Preliminary neutron capture cross section of ^{120}Sn, ^{122}Sn and ^{124}Sn were obtained in the energy range from 20 meV to 4 keV with the array of germanium detectors in ANNRI at MLF,J-PARC. The results of ^{120}Sn, ^{122}Sn and ^{124}Sn were obtained by normalizing the relative cross sections to the data in JENDL-4.0 at the largest 426.7-, 107.0- and 62.05-eV resonances, respectively. The 67.32- and 150-eV resonances for ^{120}Sn and the 579- and 950-eV resonances for ^{124}Sn which are listed in JENDL-4.0 and/or ENDF/B VII.1 were not observed.

1. Introduction

Accurate neutron capture cross section data for long-lived fission products (LLFPs) are required in the study of transmutation of radioactive waste [1]. One of the most important LLFPs is ^{126}Sn, which is included in spent fuels of light water reactors with relatively large yields and long half-life. However, only one experimental data set is available at the thermal energy [2]. Accurate cross section measurements of ^{126}Sn are strongly required.

It is expected that a ^{126}Sn sample for a cross section measurement is contaminated with a large amount of tin stable isotopes, $^{117-120,122,124}$Sn, because these stable isotopes also have fission yields and the sample is normally prepared only through a chemical process from spent fuels. These isotopes have large influence on neutron capture cross section measurements of ^{126}Sn.

Therefore, to obtain accurate cross section data for ^{126}Sn, the measurements of all tin stable isotopes had been started with Accurate Neutron-Nucleus Reaction measurement Instrument (ANNRI) of Materials and Life science experimental Facility (MLF) in Japan Proton Accelerator Research Complex (J-PARC). The results for ^{112}Sn and ^{118}Sn have been reported in ND2013 [3]. In this paper, results of the neutron capture cross section measurements of ^{120}Sn, ^{122}Sn and ^{124}Sn are reported in the neutron energy region from 20 meV to 4 keV.

2. Experimental procedure

Capture cross section measurements with neutron Time-of-Flight (TOF) method were performed with the array of Ge spectrometer in ANNRI. The array of Ge detectors is installed at the flight length of 21.5 m and is composed of two cluster-type Ge detectors, eight coaxial-type Ge detectors and anti-coincidence shields around each Ge detector described in Refs. [4] and [5]. The neutron intensity at the 21.5-m sample position is described in Ref. [6]. J-PARC is normally operated with “double-bunch mode”, in which each proton pulse consists of two bunches (each with a width of 100 ns) at intervals of 600 ns [7]. The simulated resolution function at the 21.5-m sample position is described in Ref. [8].

In the measurements, two cluster-type Ge detectors were used, but the coaxial-type Ge detectors were not used because they suffered from severe electrical noise. The pulsed neutron beam was collimated to a 7 mm at the sample position. J-PARC was operated with a proton beam power of 270 kW and at a repetition rate of 25 Hz in the “double-bunch mode”.

Samples were isotopically enriched metallic tin with a diameter of 5 mm. The weight of the ^{120}Sn, ^{122}Sn and ^{124}Sn samples was 68.7 mg, 99.7 mg and 88.2 mg, respectively. Isotopic distribution and chemical impurities of each sample are listed in Table 1. The samples were put in fluorinated ethylene propylene (FEP) film bag and attached to a polytetrafluoroethylene (PTFE) sample holder. The total measuring times for the ^{120}Sn, ^{122}Sn and ^{124}Sn samples were about 63, 30 and 32 hours, respectively. To deduce the background, measurements for a ^{208}Pb sample with a diameter of 5 mm, a weight of 159.7 mg, and an isotopic enrichment of 99.60 mole% and a sample holder with an empty FEP film bag (Blank) were also carried out during 16 and 22 hours. For dead-time correction, pulses from a random-timing pulse generator were fed to the pre-amplifier of every Ge crystal [9]. The data acquisition system in ANNRI has a typical dead time of 6 μs.

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Table 1. Isotopic distribution and chemical impurities described on the certification sheets.

	^{120}Sn sample	^{122}Sn sample	^{124}Sn sample
^{116}Sn	0.0012	<0.0001	<0.0001
^{117}Sn	0.001	<0.0001	<0.0001
^{118}Sn	0.003	<0.0001	<0.0001
^{119}Sn	0.004	<0.0001	<0.0001
^{120}Sn	0.988	0.006±0.001	<0.0001
^{122}Sn	0.0015	0.993±0.001	0.001
^{124}Sn	0.001	0.001±0.0005	0.999
Al	0.00025	<0.000004	0.000003
Ca	0.00020	<0.00005	0.00002
Cr	0.000020	<0.000005	<0.000005
Cu	0.000040	0.000025	0.000025
Fe	0.000300	not described	<0.000030
Mg	0.000040	<0.000005	<0.000005
Mn	0.00002	<0.00002	not described
Sb	0.00006	not described	not described
Si	0.00060	<0.00002	<0.00002
Zn	0.000350	<0.000005	<0.000005
In	not described	<0.000002	<0.000002
Te	not described	not described	<0.00005
Ag	<0.00001	<0.0000005	<0.0000005

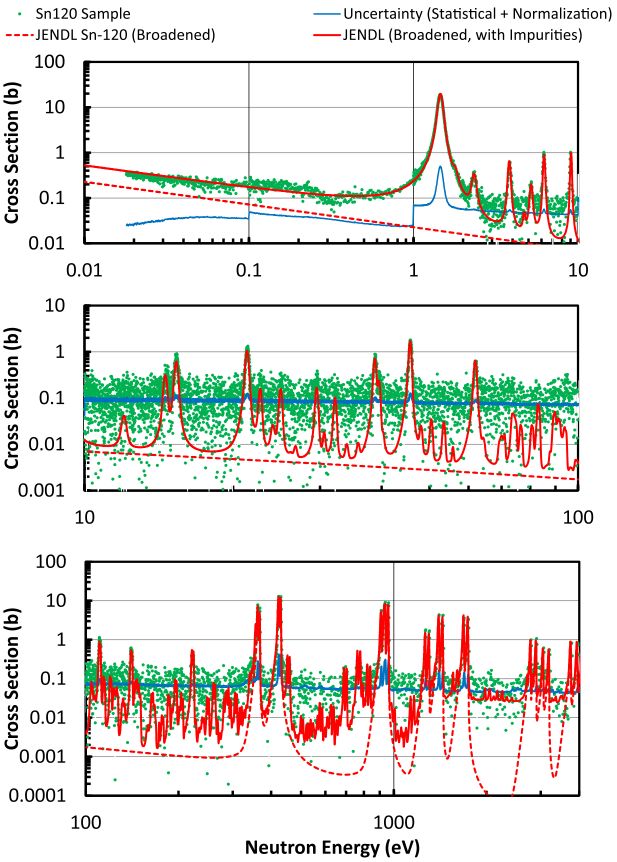

Figure 1. Results of the neutron capture cross sections for the ^{120}Sn sample together with uncertainties due to statistical uncertainty and normalization uncertainty, values of JENDL-4.0 for T = 300 K (broadened with the resolution function) and that with the impurities.

3. Data analysis

The analysis procedure was almost the same manner as that described in Ref. [3].

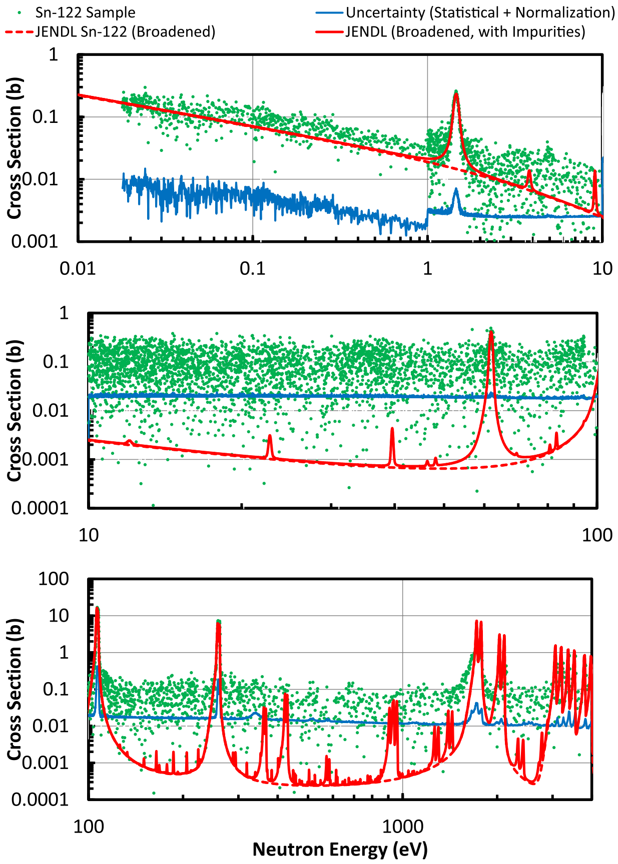

Figure 2. Results of the neutron capture cross sections for the ^{122}Sn sample together with uncertainties due to statistical uncertainty and normalization uncertainty, values of JENDL-4.0 for T = 300 K (broadened with the resolution function) and that with the impurities.

The frame-overlap backgrounds were estimated and subtracted in almost the same manner as that described in Ref. [10]. Because the beam duct and cases of the detectors are made of aluminum, main source of the TOF dependent were γ rays from ^{27}Al(n, γ) reactions due to scattered neutrons. The TOF dependent backgrounds were estimated using the capture γ-ray yields for the empty FEP bag sample and the ^{208}Pb sample. The backgrounds were normalized using the areas of the photo peaks due to the 7724-keV γ rays from ^{27}Al(n, γ) reactions.

Correction factors for neutron self-shielding and multiple scattering were calculated with the Monte Carlo simulation code MCNP [11]. In the calculation, the sample size, shape, mass, isotope abundances, and intensity distribution of neutrons were taken into consideration.

The neutron spectrum was measured by detecting the 478-keV γ rays emitted from the ^{10}B(n,$\alpha\gamma$)^{7}Li reaction. In the measurement, clear photo-peak of the 478-keV γ rays were observed. The photo-peak efficiency for 478-keV γ rays was deduced using ^{60}Co, ^{152}Eu, ^{133}Ba, and ^{137}Cs standard isotopes. The cross section data for ^{10}B(n,$\alpha\gamma$) Li reactions were taken from JENDL-4.0 [12]. Using these data and results, the absolute neutron flux was obtained.

The energy dependences of the relative cross section for ^{120}Sn, ^{122}Sn and ^{124}Sn samples were deduced by using the TOF spectra with the dead time correction, the deduced backgrounds, the self-shielding and multiple scattering correction factors and the neutron flux. Results

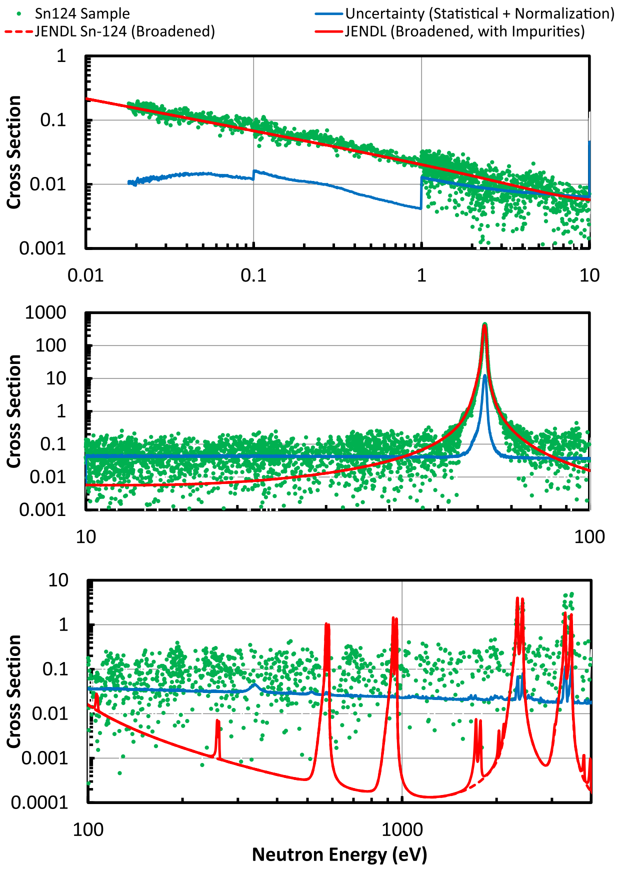

Figure 3. Results of the neutron capture cross sections for the ^{124}Sn sample together with uncertainties due to statistical uncertainty and normalization uncertainty, values of JENDL-4.0 for T = 300 K (broadened with the resolution function) and that with the impurities.

of the neutron capture cross sections for the ^{120}Sn, ^{122}Sn and ^{124}Sn samples were obtained by normalizing the relative cross sections to the data in JENDL-4.0 at the largest 426.7-eV, 107.0-eV and 62.05-eV resonances, respectively.

4. Result

The results of neutron capture cross section for ^{120}Sn, ^{122}Sn and ^{124}Sn samples were obtained in the energy range from 20 meV to 4 keV. Because of "double-bunch mode", the structure appeared on the obtained cross section in the neutron energy range above 100 eV. Figures 1, 2 and 3 show the results for ^{120}Sn, ^{122}Sn and ^{124}Sn samples together with uncertainties due to statistical uncertainty and normalization uncertainty, values of JENDL-4.0 for T = 300 K (broadened with the resolution function [8]) and those with the impurities.

In Fig. 1, the 67.32- and 150-eV resonance were not observed. These resonances were reported by G.V. Muradyan [13] and are listed in ENDF/B VII.1 [14]. This result agreed with the result by P.E. Koehler [15] and evaluation in JENDL-4.0. The 579- and 950-eV resonances for ^{124}Sn were not observed. These resonances were reported by Yu.V. Adamchuk [16] and Fuketa [17], and are listed in both JENDL-4.0 and ENDF/B VII.1.

Pulse-height spectra gated at all resonances of the samples were obtained by subtracting off-resonance spectra from on-resonance spectra [4]. Many prompt γ-rays from ^{120}Sn, ^{122}Sn and ^{124}Sn are observed. The 1114-, 1747- and 2006-keV γ rays observed in the ^{122}Sn (n, γ) reactions were previously unknown γ-rays. The other γ-rays were already reported by R.F. Carlton [18,19] and/or A.I. Egorov [20]. The origin of the resonances were decided using the gated spectra. For example, a photo-peak of 273-keV γ rays was clearly observed at the 1.457-eV resonance of the ^{122}Sn sample. The 273-keV γ rays

Table 2. Resonance energies observed in the measurement with the ^{120}Sn sample along with the evaluated values in JENDL-4.0 and ENDF/B VII.1.

Resonance Energy(eV) This work	JENDL	ENDF	Confirm[a]
^{120}Sn sample			
^{120}Sn			
Not observed	Not Listed	67.32	×
Not observed	Not Listed	150	×
364.2±0.3	364.3	365.2	○
426.7±0.3	426.1	426.1	○
920.7±0.9	919.8	919.8	○
950.0±0.9	948.6	948.6	○
1287±2	1283.1	1287.1	○
1425±2	1420	1426.3	○
1719±3	1713.4	1718.4	○
2847±5	2836	2838, 2852	△[b]
3130±6	3118.4	3123.4	△
^{116}Sn			
111.2±0.1	111.2	111.2	○
^{117}Sn			
38.75±0.04	38.8	38.8	△
^{118}Sn			
45.77±0.03	45.75	45.75	○
359±1	354	359	△
769.9±0.7	771	771	△
^{119}Sn			
141.0±0.1	140.9	140.86	○
222.3±0.2	222.6	222.64	△
454.6±1.0	455.8	455.6	△
696.9±1.4	697	696.1	△
^{124}Sn			
61.99±0.06	62.0	62.0	△
^{115}In			
1.456±0.001	1.46	1.457	○
3.85±0.01	3.82	3.85	○
9.02±0.04	9.07	9.07	○
39.5±0.1	39.56	39.61	△
^{113}In			
14.6±0.1	14.6	14.6	△
32.2±0.2	32.2	32.23	△
^{123}Te			
2.34±0.01	2.334	2.334	○
^{121}Sb			
6.213±0.003	6.217	6.24	○
15.36±0.05	15.37	15.39	△
29.6±0.1	29.58	29.6	△
^{123}Sb			
21.39±0.04	21.37	21.38	○
^{109}Ag			
5.20±0.01	5.19	5.19	○

[a] ○: Photo peaks due to the capture reactions were observed.
△: The number of the events was not enough to observe photo peaks.
×: The resonance was not observed.
[b] Resonances were overlapped.

Table 3. Resonance energies observed in the measurements with the ^{122}Sn and ^{124}Sn samples along with the evaluated values in JENDL-4.0 and ENDF/B VII.1.

Resonance Energy(eV)			Confirm[a]
This work	JENDL	ENDF	
^{122}Sn sample			
^{122}Sn			
107.0±0.1	106.8	106.8	○
260.2±0.2	259.6	259.6	○
1754±4	1751.2	1751.2	○[b]
2076±5	2073	Not Listed	△
3180±5	3138	Not Listed	△
3456±6	3452.9	3452.9	○
3907±7	3896.7	3896.7	△
^{115}In			
1.457±0.001	1.46	1.457	○
^{124}Sn			
61.96±0.06	62.0	62.0	△
^{124}Sn sample			
^{124}Sn			
62.05±0.06	62.0	62.0	○
Not Observed	579	579	×
Not Observed	950	950	×
2381±4	2380	2380	○
3395±6	3390	3390	△

[a] ○: Photo peaks due to the capture reactions were observed.
△: The number of the events was not enough to observe photo peaks.
×: The resonance was not observed.
[b] The prompt γ rays were observed clearly, but some resonances were overlapped.

was not prompt γ-rays from ^{122}Sn (n, γ) reactions but those from ^{115}In (n, γ) reactions. It was validated that the 1.457-eV resonance is one of the ^{115}In resonances. In the same manner, the other observed resonances were confirmed. The results and the observed resonance energies are listed in Tables 2 and 3, and along with the evaluated resonance energies in JENDL-4.0 and ENDF/B VII.1.

In Fig. 1, the ^{120}Sn sample was contaminated by In of 770 ppm, Sb of 450 ppm, Te of 5 ppm and Ag of 8 ppm. The ^{122}Sn sample was contaminated by In of 7.5 ppm as shown in Fig. 2 However, except for Ag in the ^{120}Sn sample, these values do not agree with the certificated values in Table 1. Unexpected contaminations by In, Sb and Te were found in the ^{120}Sn sample and that by In was found in the ^{122}Sn sample.

5. Summary

The preliminary neutron capture cross section of ^{120}Sn, ^{122}Sn and ^{124}Sn were obtained in the energy range from 20 meV to 4 keV with the array of germanium detectors in ANNRI at MLF/J-PARC. The results were obtained by normalizing the relative cross sections to the data in JENDL-4.0 at the largest resonances, respectively. The 67.32- and 150-eV resonances for ^{120}Sn and the 579- and 950-eV resonances for ^{124}Sn which are listed in JENDL-4.0 and/or ENDF/B VII.1 were not observed. Three new prompt γ-ray emissions were observed in the ^{122}Sn (n, γ) reactions.

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