Measurement of neutron total cross sections of Sn-Pb alloys in solid and liquid states

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The use of a Sn-Pb alloy as an emergency in-core heat transport medium is being considered in the design of a small modular reactor proposed by Toshiba Energy Systems & Solutions Corporation. However, there exists no experimental data for the neutron cross sections of Sn-Pb alloy.

In the present study, the neutron total cross sections of Sn-Pb alloy were obtained from neutron transmission measurements by the time-of-flight (TOF) method using the Kyoto University Institute for Integrated Radiation and Nuclear Science – Linear Accelerator (KURNS-LINAC). The sample temperature was changed from room temperature (solid) up to 300 °C (liquid). The total cross sections of the solid and liquid states were compared with the evaluated data and the previous data of Pb and Sn.

Keywords: neutron total cross section, Sn-Pb alloy, time-of-flight, KURNS-LINAC, solid, liquid

1. Introduction

Recently, a small modular reactor (SMR), which has inherent and passive safety, has been receiving attention all over the world. In Japan, a very small modular reactor, namely, MoveluX^{TM} (Mobile-Very-Small reactor for Local Utility in X-mark) [1] is being developed by Toshiba Energy Systems & Solutions Corporation. MoveluX^{TM} is a thermal reactor that uses calcium hydride as a neutron moderator and its heat output is 10 MWth. As for the heat exhaust function in the core, some heat pipes are used, and heat can be transported by using temperature differences. On the other hand, when the heat exhaust function of the heat pipes is lost, an emergency in-core heat transport medium, instead, will carry the heat to the reactor wall by natural convection, and the core wall will be cooled by ambient air circulation. The use of a Sn-Pb alloy as the emergency in-core heat transport medium is being considered.

Since the melting point of the Sn (60 wt%)-Pb (40 wt%) alloy is about 180 ℃, the alloy is solid state when the reactor is just started to work, and it becomes liquid state while the power increases, because the core temperature reaches 660 ℃ in full power operation. Change of the state of the alloy during operation will affect the core characteristics. In particular, the thermal neutron scattering cross section will change between the solid and liquid states, because solid metal has a crystal structure and it causes the coherent scattering effect, and when it melts the effect changes. Alloy also has a crystal structure and its orientation is different from that of single element metal. The wavelength of a thermal neutron is close to the lattice spacing of atoms in the crystal structure and the coherent scattering occurs. Therefore, the neutron cross section in the thermal neutron energy region also differs due to the influence of coherent scattering by the crystal structure and its orientation. However, there are no reports on experimental data for the neutron cross section of Sn-Pb alloy in both solid and liquid states. In the present work, the thermal neutron total cross sections of solid and liquid Sn-Pb alloy were measured and compared with the evaluated nuclear data of the free gas model in JENDL-4.0 [2].

2. Experiment

The neutron total cross section measurements of Sn-Pb alloy were carried out by using the TOF method at KURNS-LINAC. The experimental arrangement is shown in Figure 1. The KURNS-LINAC was operated with a pulse width of 1 μs, a repetition rate of 50 Hz, and an average current of 28.4 μA. The accelerated electrons were incident on a water-cooled Ta-target [3]. Then, fast neutron were produced by the photo nuclear reaction (γ, n). To obtain thermal neutrons, the Ta-target was set at the center of a water moderator tank (20 cm in diameter). The neutron flight tube was used in the direction of 135 degrees with respect to the LINAC beam line [4]. A ^{6}Li-galss scintillator detector, GS20, was used as a neutron detector. The flight length between the Ta-target and the ^{6}Li-glass detector was about 12.1 m.

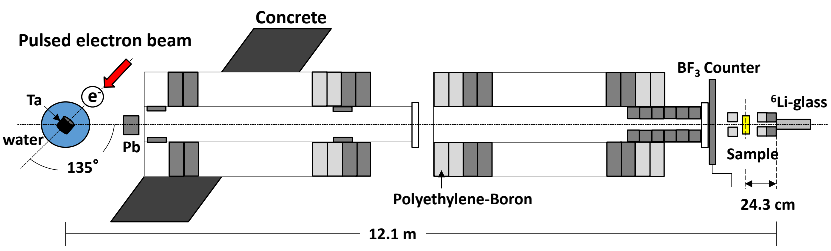

Figure 1: Experimental arrangement for neutron total cross measurement.

The specifications of the Sn-Pn alloy samples and the Al case are listed in Table 1. The Sn-Pb alloys containing 60 wt% natural tin and 40 wt% natural lead were used. We prepared two types of samples in the solid state, one is a re-solidified sample with 10 mm thickness contained in the Al case after melting, and the other is a commercially available rod with 6 mm thickness. To reduce the effects of neutron scattering from the Al case, its thickness was reduced to 1 mm. The measurement times are listed in Table 2.

Table 1: Specifications of Sn-Pb alloys and Al case

	Sn-Pb alloy (Sn : 60 wt%, Pb : 40 wt%)			Al
Sample	liquid (in Al case)	solid (in Al case)	solid (rod sample)	Al case
Weight (g)	413.52	413.52	200.51	193.42
Size (mm^2)	2750	2625	3835	4200
Thickness (mm)	10	10	6	1 (beam line)

Table 2: List of measurement times

	Sample	Measurement time (h)
Sn-Pb alloy	liquid (300 ℃)	12.0
	liquid (250 ℃)	12.0
	liquid (210 ℃)	12.0
	solid (in Al case)	12.0
	solid (rod sample)	9.0
	Al case	12.0
	Blank	9.0
	Al case + res.filter	3.0

In the TOF method, the incident neutron energy was obtained by the following equation:

$$E = \left(\frac{72.3 \times L}{t}\right)^2 \tag{1}$$

where, *E* is the incident neutron energy, *L* is the neutron flight length and *t* is the TOF. The neutron total cross section $\sigma_{tot}(E)$ was obtained by the following equation:

$$\sigma_{tot}(E) = -\frac{1}{n}\ln\frac{(C_s(E) - C_{s,b}(E))/M_s}{(C_o(E) - C_{o,b}(E))/M_o} \tag{2}$$

where, $C_s(E)$ and $C_o(E)$ are the neutron count rates with the sample existed (hereafter, called as "sample-in") and not existed (hereafter, called as "sample-out") measured by the ^{6}Li-glass detector. $C_{s,b}(E)$ and $C_{o,b}(E)$ are the neutron background for the sample-in and sample-out. M_s and M_o are the neutron count rates by the BF_3 detector to monitor neutron intensity fluctuation between measurements. n is the atomic area density [atom/barn] of Sn-Pb alloy samples.

The γ-ray background level was estimated from the pulse height spectrum of each measurement. Figure 2 shows the pulse height spectrum of the ^{6}Li-glass detector. As shown in Figure 2, the counts for 320-420 ch. includes neutron and background γ-ray counts. On the other hand, the counts for 220-319 ch. is only background γ-ray one. Then, the γ-ray background level was evaluated from the TOF spectrum by interpolate the corresponding pulse height normalized by each event. Figure 3 shows the results of evaluated γ-ray background of the TOF spectrum, comparing with the one with resonance filters (Mn, Co, Ag, In and Cd). As shown in Figure 3, the evaluated γ-ray background shows good agreement with the dips of In (1.46 eV), Ag (5.19 eV), Co (132 eV), and Mn (337 eV), which correspond to the large resonance peaks, respectively. Then, it was confirmed that the present background evaluation method is appropriate.

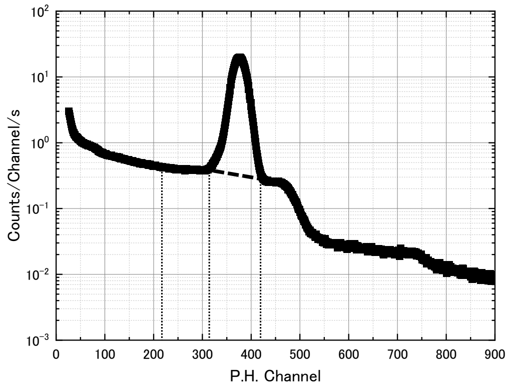

Figure 2: Pulse height spectrum of ^{6}Li-glass detector.

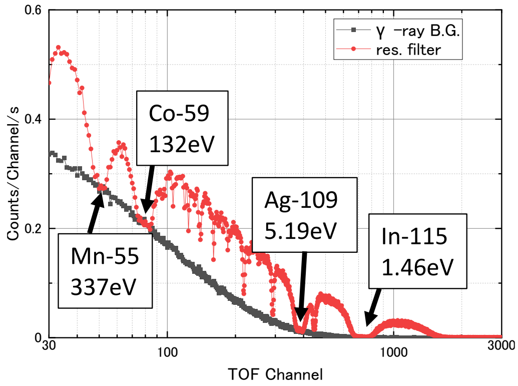

Figure 3: TOF spectra with resonance filters and evaluated γ-ray background.

3. Results and Discussion

We will discuss the results of the measurements from three perspectives. The first perspective is the effect of temperature of the liquid alloy. Figure 4 shows the neutron total cross sections of the liquid Sn-Pb alloys with different temperatures. There are some small differences in the total cross sections between 210 °C and other temperatures from 0.002 eV to 0.01 eV. This seems to be caused by the remaining crystal structure in liquid at 210 °C, which is relatively low temperature. On the other hand, there is a little difference in the cross sections between 250 °C and 300 °C. However, in the present study, it is difficult to say clearly that there is a difference due to sample temperature in the liquid state. Before the experiment, we had considered that the effect of the coherent scattering would disappear in the neutron cross section of the liquid state, and the cross section would be almost close to the one with the free gas model. However, the effect of coherent scattering still exists in the liquid state in the present study.

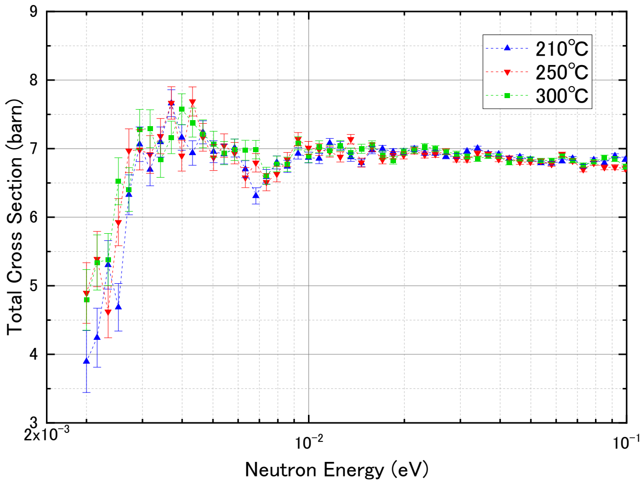

Figure 4: Comparison of neutron total cross sections of liquid Sn-Pb alloy between temperatures (210 °C, 250 °C and 300 °C).

The second perspective is the effect of the solid/liquid sates on the neutron total cross sections. The cross sections of the Sn-Pb alloy in the solid and liquid states, and the JENDL-4.0 data are compared in Figure 5. The JENDL-4.0 data shows the neutron total cross sections of the Sn-Pb alloy calculated using the Sb and Pb cross sections of the JENDL-4.0 with the weights of the atomic ratio of each composition. In the solid state, the Bragg edges due to the crystal structure are observed in the neutron energy below 0.05 eV. However, it turned out that the magnitude (or height) of Bragg edges in the liquid state decreased and widened in the energy region. This would be caused by change of the crystal structure of the solid due to melting. In comparison with the JENDL-4.0, although the measured data shows big difference in the low energy, they show good agreement above 0.05 eV. This is because the influence of coherent scattering due to the crystal structure is little above 0.05 eV.

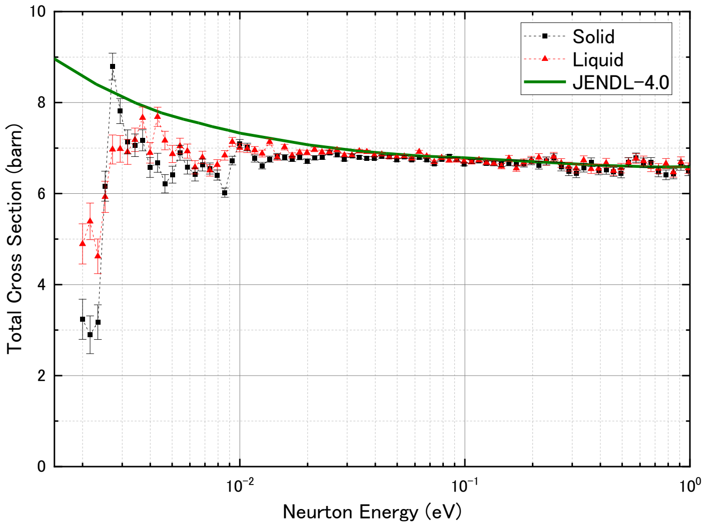

Figure 5: Comparisons between neutron total cross sections of Sn-Pb alloys in the solid, liquid states and JENDL-4.0.

The third perspective is the comparisons in the solid state measurements. In Figure 6, the neutron total cross sections of Sn, Pb measured in the past [5,6] and Sn-Pb alloys in the present measurement are shown. The present Sn-Pb alloy measurements are the results of the two solid samples, the re-solidified sample and the rod one (see Section 2, for detail). As seen in Figure 6, several Bragg edges are in good agreement between the re-solidified and rod samples, Sn, and Pb from 0.002 eV to 0.05 eV. However, there are differences in the Bragg edges between the re-solidified and rod samples. For the re-solidified sample, some Bragg edges of Pb tended to disappear at 3.42 meV, 6.82 meV, and 17.1 meV. The neutron cross section of the material with a crystal structure changes depending on its orientation. Therefore, this difference between the re-solidified and rod samples would be due to the change in crystal orientation caused by melting. However, there is still room for considering the reasons of the difference as further study.

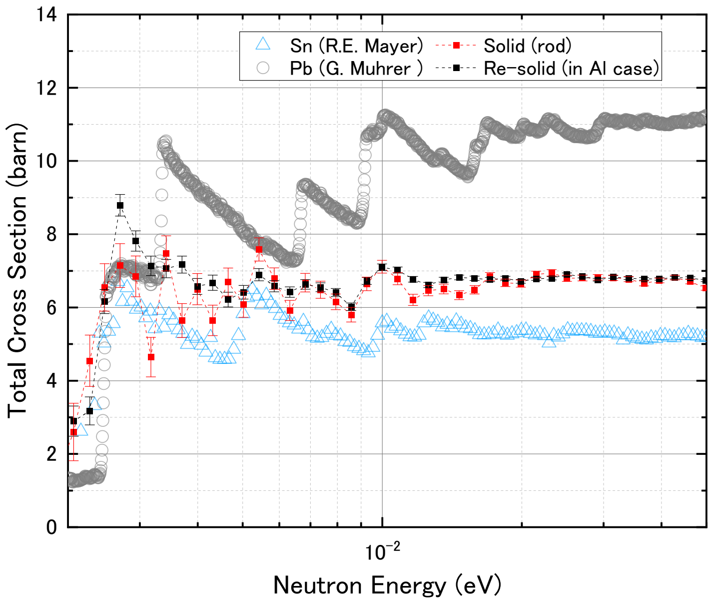

Figure 6: Comparison between the total cross sections of the rod sample (indicated as "solid (rod)"), the re-solidified sample after melting (indicated as "Re-solid (in Al case)") of Sn-Pb alloys and the previously measured data of Pb and Sn.

4. Conclusion

We have performed the neutron total cross section measurements of the Sn-Pb alloy in solid and liquid states using the TOF method at KURNS-LINAC. The measurement results were compared with the evaluated data in JENDL-4.0 and the previous experimental data of Sn and Pb. The total cross sections of the solid and liquid states were compared and changes in Bragg edges were observed in the energy range below 0.05 eV. Comparing the total cross sections of the rod and re-solidified samples, it was confirmed that Bragg edges, which is thought to be due to the crystal structure of Pb, was not observed in the re-solidified sample.

The present study shows important data for using the solid and liquid alloys as heat transport medium in the core of a thermal reactor.

References

[1] R. Kimura et al., Nucl. Sci. Eng., 2020; 194: 213-220p.

[2] K. Shibata et al., J. Nucl. Sci. Technol., 2011; 48(1): 1-30p.

[3] K. Kobayashi et al., Ann. Rep. Res. Reactor Inst. Kyto Univ., 1989: 22; 142p.

[4] Y. Takahashi et al., Phys. B: Condensed Matter, 2018; 551; 488-491p.

[5] G. Muhrer et al., Nucl. Instrum. Methods Phys. Res. Sec. A, 2007; 572: 866-873p.

[6] R. E. Mayer et al., Jour. Atomkernenergie, 1981: 39; 55p.

別紙

日本語タイトル：　固体と液体状態での Sn-Pb 合金の全断面積測定

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