Crystal structure analysis of 143-Zintl phase thermoelectric compounds

K. Ono[A, B], Y. Goto[A], C. H. Lee[A]

[A]*AIST, Japan* [B]*Keio Univ., Japan*

Thermoelectric materials attract interest because they generate electricity from waste heat. Recently, we have reported that the 143-Zintl phase compound $RbZn_4As_3$ shows relatively high thermoelectric performance[1]. Its dimensionless-figure-of-merit (*ZT*) reaches 0.53 at 797 K. In addition, it exhibits low lattice thermal conductivity 0.71 W/mK at 797 K, that contributes to the high *ZT* value. We found that the atomic displacement parameter (B) of Rb is larger than other elements, and this large vibration might be the origin of low lattice thermal conductivity.

For this experiment, we focused on $RbCd_4As_3$. $RbCd_4As_3$ is consisted by electrical conductive layers and spacer layers. The electrical conductive layers are consisted by covalently bonded Cd and As, with doubled tetrahedral structure. Cd_1 and As_1 are located out-plane, Cd_2 and As_2 are located in-plane in the electrical conductive layers. To determine the crystal structure parameters, we conducted powder neutron diffraction measurements of $RbCd_4As_3$ using the HERMES diffractometer. The temperature was varied from 100 K to 700 K. The wave length of incident neutron was 1.34194 Å. All samples were prepared by solid-state reaction. To prevent the absorption of neutrons, the isotope Cd_{114} was used for the raw materials. The sample was enclosed in a vanadium cell (ϕ 10 mm) under Ar filled globe box.

Figure 1 shows neutron powder diffraction patterns of $RbCd_4As_3$ at 300 K with the result of Rietveld analysis by RIETAN-FP, R_{wp} is 11.58 %. The observed and calculated patterns were depicted by red dots and a solid green line, respectively. The vertical bars depict the calculated positions, and the bottom line indicates the difference between the observed and calculated results. Figure 2 shows the temperature dependence of the B parameters. The B parameters increased with heating. In particular, the B parameters of Rb is larger than other elements, despite Rb is heavier than other elements. This could be due to the ionic bonding of Rb ion, in contrast to the covalent bonding of As and Cd. This large B parameter of Rb might suppress lattice thermal conductivity.

In conclusion, the crystal structure parameters of $RbCd_4As_3$ were determined by powder neutron diffraction measurement. We found that B parameter of Rb is larger than other elements, this might suppress lattice thermal conductivity.

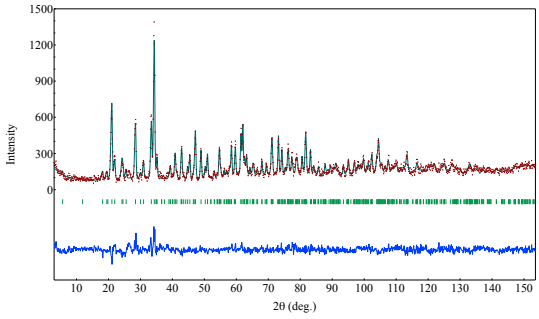

Fig. 1. Neutron diffraction patterns of $RbCd_4As_3$ at 300 K. Vertical bars show the calculated positions of the diffraction peaks.

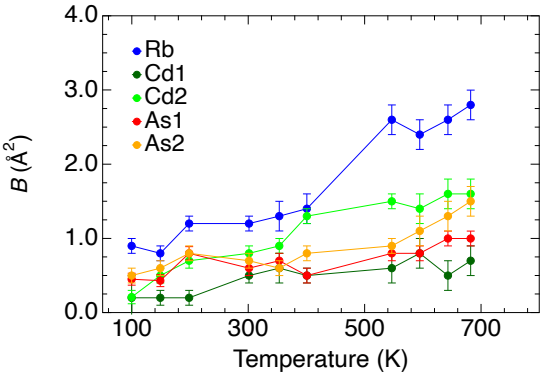

Fig. 2. Temperature dependence of the isotropic atomic displacement parameters of $RbCd_4As_3$.

[1] K. Ono et al., ACS Omega. 8, 42900 (2023).