Thermoelectric Properties of Vacuum Hot-pressed $Ba_8Al_{16}Si_{30}$ Clathlate

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Abstract

Type Ⅰ clathrate $Ba_8Al_{16}Si_{30}$ was produced by arc melting and hot pressing and thermoelectric properties were investigated. Negative Seebeck coefficient at all temperatures measured, which means that the majority carriers are electrons. Electrical conductivity decreased by increasing temperature and thermal conductivity was 0.012 W/cmK at room temperature and dimensionless thermoelectric figure of merit (ZT) was 0.01 at 873K.

Keywords : thermoelectric, $Ba_8Al_{16}Si_{30}$, clathrate, arc melting, hot pressing

1. Introduction

The PGEC (phonon-glass and electron-crystal) concept has been considered to search novel thermoelectric materials. Good thermoelectric materials should possess thermal properties similar to that of a glass and electrical properties similar to that of a perfect single crystal material, *i.e.* a poor thermal conductor and a good electrical conductor. Thermoelectric performance of a give material is described by the dimensionless figure of merit $ZT=\alpha^2\sigma T/\kappa$, where α is Seebeck coefficient, σ is the electrical conductivity, κ is the thermal conductivity and T is temperature [1]. The clathrate that is one of the candidates for good thermoelectric materials has a general formula $A_8^{II}B_{16}^{III}B_{30}^{IV}$, where A^{II}=Ba, Sr, B^{III}=Al, Ga, B^{IV}=Si, Sn. It has been known to have an open framework structure to allow the introduction of atoms that can vibrate inside the cage. The lattice thermal conductivity, κ_L is lowered by scattering of acoustic phonons from the vibrations of these caged atoms, which is well known as the rattling effect. In addition, these materials still maintain considerably high electric conductivity, through the covalent sp^3 hybridized B^{III}-B^{IV} frameworks, which determine the electronic transport properties [2,3]. In this work, the arc melting and hot pressing was attempted to prepare the type I clathrate $Ba_8Al_{16}Si_{30}$ compounds and their thermoelectric properties were investigated.

2. Experimental and Results

High-purity elemental Ba, Al and Si, taken in the stoichiometric atomic ratio 8:16:30 were melted in an Ar atmosphere for 60 seconds with arc current of 300A, and remelted five times to prevent the segregation. The Ti getter was employed to eliminate oxygen in an Ar gas and a vacuum chamber. The ingot was crushed into powders and sieved to 45 ㎛ mesh in a glove box in Ar atmosphere. The powders were hot pressed in a cylindrical Inconel superalloy die with an internal diameter of 10ϕ at 1073K under a pressure of 60 MPa for 1 hour in Ar atmosphere. Phase identification and transformation were analyzed by X-ray diffraction (XRD) with the Cu Kα (40kV, 30mA) radiation within 2θ = 10~80° at 3°/min. Seebeck coefficient, electrical resistivity and thermal conductivity were measured at 300K-600K, and thermoelectric figure of merit was evaluated.

Figure 1 shows the phase identification results analyzed by XRD for the hot-pressed $Ba_8Al_{16}Si_{30}$. Figure 2 represents the Seebeck coefficient with temperature. The $Ba_8Al_{16}Si_{30}$ specimen showed negative Seebeck coefficient at all temperatures measured, which means that the majority carriers are electrons. Temperature dependence of electrical conductivity was shown in Figure 3 Electrical conductivity decreased by increasing temperature, indicating that all specimens showed metallic conduction behavior. Thermal conductivity as shown in Figure 4 was 0.012 W/cmK at room temperature. Figure 5 shows the temperature dependence of dimensionless thermoelectric figure of merit (ZT).

3. Summary

The arc melting and hot pressing were employed to prepare the $Ba_8Al_{16}Si_{30}$ clathlate and their thermoelectric properties were investigated. Negative Seebeck coefficient at all temperatures measured, which means that the majority carriers are electrons. Electrical conductivity decreased by increasing temperature, indicating that all specimens showed metallic conduction behavior. Thermal conductivity was 0.012 W/cmK at room temperature and dimensionless thermoelectric figure of merit (ZT) was 0.01 at 873K.

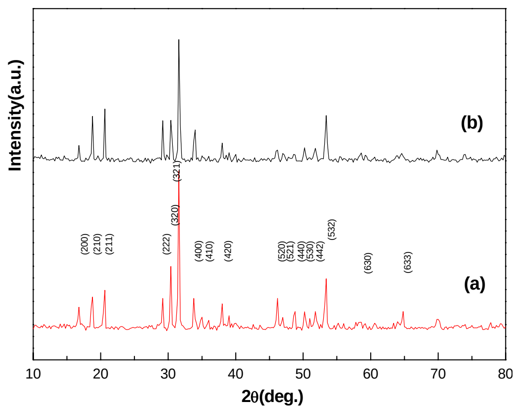

Fig. 1. Phase identifications for hot-pressed $Ba_8Al_{16}Si_{30}$; (a) as powder, (b) hot-pressed at 800℃ for 1hr.

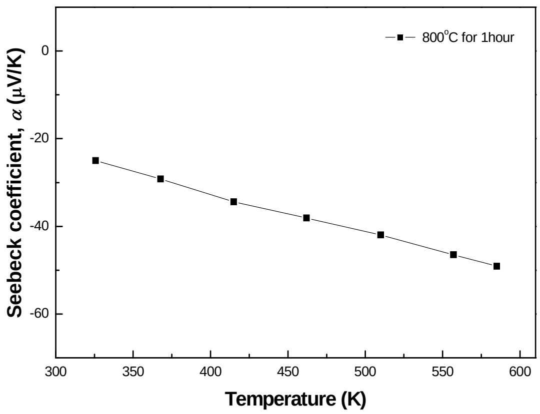

Fig. 2. Seebeck coefficient of hot-pressed $Ba_8Al_{16}Si_{30}$.

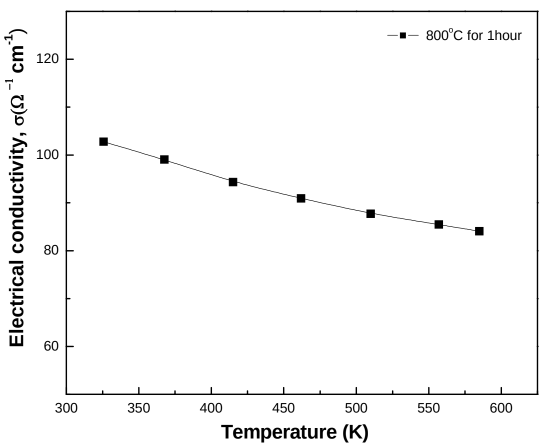

Fig. 3. Electrical conductivity with temperature of hot-pressed $Ba_8Al_{16}Si_{30}$.

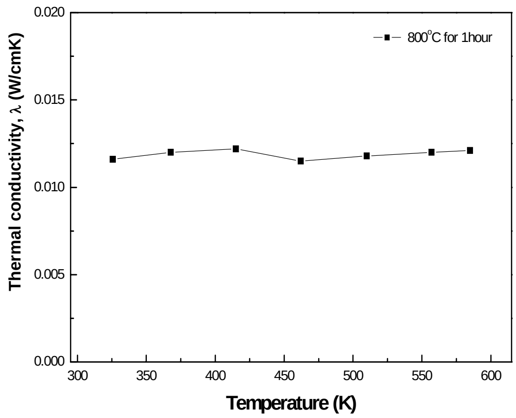

Fig. 4. Thermal conductivity of hot-pressed $Ba_8Al_{16}Si_{30}$.

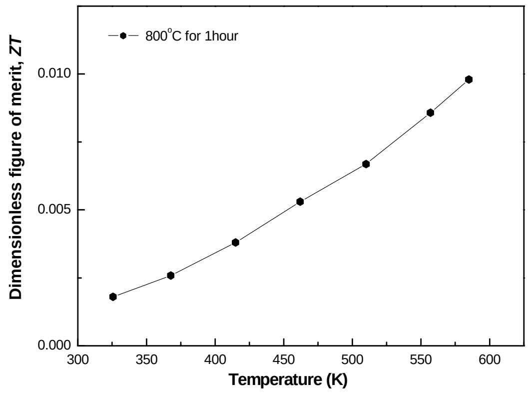

Fig. 5. Dimensionless figure of merit (ZT) of hot-pressed $Ba_8Al_{16}Si_{30}$.

Acknowledgments

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4. References

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