総合論文

ハイエントロピー型熱電材料 $AgBiSe_{2-2x}S_xTe_x$ の熱電特性

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High Thermoelectric Figure-of-merit and Ultra-low Lattice Thermal Conductivity of New High-entropy-type $AgBi(S,Se,Te)_2$

by

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Abstract

Since thermoelectric generators can convert waste heat into electricity, they play an important role in energy harvest. $AgBiSe_2$ metal chalcogenide is one of the high-performance thermoelectric materials with low lattice thermal conductivity. It is well known that $AgBiSe_2$ exhibits temperature-dependent crystal structural transitions from Hexagonal to Rhombohedral, and finally Cubic phase. Previous studies revealed the high figure-of-merit *ZT* in Cubic phase. To decrease phase transition temperature and obtain Cubic phase at lower temperature, we introduced high-entropy alloy (HEA) concept in $AgBiSe_2$ compound. Here, we present successful synthesis of $AgBiSe_{2-2x}S_xTe_x$ and stabilization of Cubic phase utilizing the HEA concept. Through structural analysis using synchrotron X-ray diffraction, decrease of crystal transition temperature was found with increase in *x* amount. Electrical and thermal properties of the obtained samples revealed the ultra-low lattice thermal conductivity and high $ZT \sim 0.9$ at 747 K. The discovery of decreasing the phase transition temperature while keeping or improving thermoelectric properties would offer new way for development of high-entropy-type thermoelectric.

Keywords: Thermoelectric materials, High-entropy-type compounds, Material science

1. 緒言

熱電発電は，ゼーベック効果を用いて熱エネルギーを直接電気エネルギーに変換することができる．熱電モジュールは，排熱や身の回りに存在する熱源から発電が可能であるため，省エネルギー技術として注目されている [1]．熱電材料の性能は，下記の無次元性能指数（*ZT*）で評価される．

$$ZT = \frac{S^2}{\rho\kappa_{\mathrm{tot}}}T \qquad (1)$$

*S*はゼーベック係数，ρは電気抵抗率，κ_{tot}は熱伝導率，*T*は絶対温度に対応しており，高い熱電性能を示す材料には，大きいゼーベック係数，小さい電気抵抗率及び熱伝導率が必要であり，$ZT = 1$ 以上が実用化の目安とされている．

金属カルコゲナイド系熱電材料に分類される $AgBiSe_2$ は，温度上昇に伴い構造相転移を示し（Fig. 1 参照），Cubic 構造では低い格子熱伝導率 κ_{lat} に起因して高い *ZT* を示す [2)-6)]．

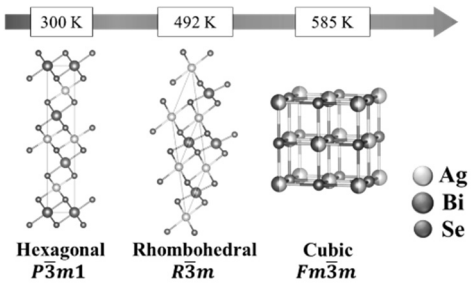

Fig. 1　Crystal structure and space group of $AgBiSe_2$