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Enhanced thermoelectric performance via the solid solution formation: The case of pseudobinary alloy $(Cu_2Te)(Ga_2Te_3)_3$ upon Sb substitution for Cu

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ABSTRACT In this work we have observed the beneficial effect from the solid solution formation on the thermoelectric performance of $(Cu_{2(1-x)}Sb_{2x}Te)(Ga_2Te_3)_3$ upon Sb substitution for Cu. This substitution allows the different occupations of Sb in the crystal lattice, i.e. Sb at Cu sites with $x \leq 0.05$ and at Ga sides with $x \geq 0.05$, which has resulted in the Pisarenko relation does not exactly capture the measured Seebeck coefficient under assumed effective masses m^*. The reduction of the lattice thermal conductivity (κ_L) has been quantified within the temperature range from room temperature to 723 K. Over the entire composition range, the κ_L value is reduced by 33% and 25% at temperature 723 K and 580 K, respectively. This observation is in a good agreement with the theoretical calculation based on the Callaway model used in the solid solutions. Along with the increasing of the mobility and electrical conductivity, the thermoelectric performance has been improved with the highest *ZT* value of 0.58 at 723 K, which is about double the value of intrinsic $(Cu_2Te)(Ga_2Te_3)_3$.

Keywords: Thermoelectric performance; Solid solution formation; Pseudobinary alloy $(Cu_2Te)(Ga_2Te_3)_3$; Hall carrier concentration; Lattice thermal conductivity

1. Introduction

Although the transformation of waste heat into useful electric power is very attractive so far, the conversion effectiveness is still low because of poor thermoelectric (TE) performance of the materials. Therefore, searching for new materials is one of the great challenges facing the members of TE research community. The TE performance is directly dependent on the dimensionless figure of merit (*ZT*),

$$ZT=T\alpha^2\sigma/\kappa \quad (1)$$

where T, α, σ and κ are the absolute temperature, Seebeck coefficient, electrical conductivity, and total thermal conductivity with lattice contribution (κ_L) and electronic part (κ_e), respectively. The correlation among physical parameters above results in a very limited choice of the current state-of-the-art TE materials.

Among the currently developed state-of-the-art TE materials, a few binary chalcogenides, such as SnSe [1,2], In_4Se_3 [3-6] and PbTe [7,8], have been explored extensively; while the ternary chalcogenides, such as $CuGaTe_2$ [9-11] and $CuInTe_2$ [12-15], have also attracted research interests in recent years because of their unique crystal structures.

It was reported that Ga_2Te_3 forms pseudobinary alloys with Cu_2Te in a wide range of compositions $(Cu_2Te)_{1-y}(Ga_2Te_3)_y$ [16]. However, the total cations (Cu^++Ga^{3+}) in this compound is 2, less than anions Te^{2-} ($2y$+1) at $0.5 \leq y < 1.0$, hence ($2y$-1)/($2y$+1) of cation sites are structural vacancies [17]. The structural vacancy is essential to the carrier concentration and lattice thermal conductivity. For example, at y=0.75, i.e. $(Cu_2Te)(Ga_2Te_3)_3$ (denominated as CTGT), one fifth of the cation sites are structural vacancies. The carrier concentration (n_H) in CTGT at room temperature (RT) reaches $0.53 \sim 1.2\times10^{19}$ cm^{-3} [17,18], very close to the

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