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The Effect of Adding Nano-Bi_2Te_3 on Properties of GeTe-Based Thermoelectric Material

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The efficient thermoelectric materials $(GeTe)_{0.85-x}(Mn_{0.6}Sn_{0.4}Te)_{0.15}(Bi_2Te_3)_x$ ($0 \le x \le 0.05$), in which Bi_2Te_3 is nanopowder, were prepared by hot pressing. The effect of adding neutral nano-Bi_2Te_3 content on the thermoelectric properties of germanium telluride was investigated. With increasing x, the thermal conductivity of the prepared samples decreased significantly and the Seebeck coefficient declined slightly, while there was no obvious change in electrical conductivity. In both electrical conductivity and Seebeck coefficient curves at different x values, there are inflection points around 600 K. The maximum dimensionless figure of merit ZT of the prepared materials is 1.54, attained in the temperature range from 700 K to 750 K for $x = 0.03$. The x-ray diffraction (XRD) pattern shows that Bi_2Te_3 has been alloyed into the GeTe-MnTe-SnTe alloy, which is consistent with the high-resolution scanning electron microscopy (HRSEM) images. Adding nano-Bi_2Te_3 to GeTe-based materials could also increase their performance stability at high temperature as a result of decreasing the phase-transition temperature T_c.

Key words: GeTe-based materials, nano-Bi_2Te_3, dimensionless figure of merit, thermal conductivity, phase-transition temperature

INTRODUCTION

Germanium telluride $GeTe_{1-\delta} + Ge_\delta$ ($0.012 \le \delta \le 0.047$)[1] is a promising thermoelectric material deviating from stoichiometry. It is an intermediate-temperature system, and its optimum operating temperature range is 680 K to 780 K. However, using this kind of material is complicated because there is an $\alpha \leftrightarrow \beta$ phase transition between the rhombohedral lattice (space group $R3m$) and cubic lattice (spatial group $Fm3m$) in its working temperature range (about 700 K). The rhombohedral distortion and volume change in the phase-transition process will exert a direct influence on the stability of the thermoelectric performance of such material. Cook et al.[2] proposed that decreasing the molar percentage of GeTe, i.e., adding compounds to GeTe alloy, can reduce the phase-transition temperature T_c and thus widen the existence region of the high-temperature β-phase. As indicated in Ref. 2, GeTe is characterized by a deviation from stoichiometry towards Te. So, GeTe always exhibits p-type conduction, and its main nonstoichiometric defects are vacancies.

Shelimova et al.[3] reported a kind of solid solution with isovalent cation substitution based on GeTe in the GeTe-MnTe-PbTe quasiternary system. Their research results show that, at a specific ratio between MnTe and PbTe, it is possible to move the existence region of β-phase to a lower temperature range. Our laboratory also examined the quasiternary materials $(GeTe)_x(Mn_{0.6}Sn_{0.4}Te)_{1-x}$ ($0.8 \le x \le 1.0$) by substituting lead for tin.[4] The phase-transition temperature T_c of the quasiternary alloy is 572 K, lower than that of GeTe. The maximum dimensionless figure of merit ZT is above 1.5 in the temperature range from 720 K to 750 K for $x = 0.85$.

Mazelsky et al.[5] proposed that Bi_2Te_3 has a donor effect when it is added into GeTe. They proposed a mechanism of doping GeTe by Bi_2Te_3 in which each Bi_2Te_3 molecule is implanted into the GeTe lattice

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