Composition-Dependent Thermoelectric Properties of *n*-Type $Bi_2Te_{2.7}Se_{0.3}$ Doped with In_4Se_3

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We present the effects of In_4Se_3 addition on thermoelectric properties of *n*-type $Bi_2Te_{2.7}Se_{0.3}$. In this study, polycrystalline $(In_4Se_3)_x$ - $(Bi_2Te_{2.7}Se_{0.3})_{1-x}$ pellets were prepared by mechanical alloying followed by spark plasma sintering (SPS). The thermoelectric properties such as Seebeck coefficient and electrical and thermal conductivities were measured in the temperature range of 300 K to 500 K. Addition of In_4Se_3 into $Bi_2Te_{2.7}Se_{0.3}$ resulted in segregation of In_4Se_3 phase within $Bi_2Te_{2.7}Se_{0.3}$ matrix. The Seebeck coefficient of the $(In_4Se_3)_x$ - $(Bi_2Te_{2,7}Se_{0,3})_{1-x}$ samples exhibited lower values compared with that of pure $Bi_2Te_2 Se_{0,3}$ phase. This reduction of Seebeck coefficient in *n*-type $(In_4Se_3)_r$ - $(Bi_2Te_{2.7}Se_{0.3})_{1-x}$ is attributed to the formation of unwanted *p*-type phases by interdiffusion through the interface between $(In_4Se_3)_x$ and $(Bi_2Te_{2.7}Se_{0.3})_{1-x}$ as well as consequently formed Te-deficient matrix. However, the decrease in electrical resistivity and thermal conductivity with addition of In₄Se₃ leads to an enhanced thermoelectric figure of merit (ZT) at a temperature range over 450 K: a maximum ZT of 1.0 is achieved for the *n*-type $(In_4Se_3)_{0.03}$ -(Bi₂₋ $Te_{2.7}Se_{0.3})_{0.97}$ sample at 500 K.

Key words: Bi_2Te_3 , thermoelectric, nanostructure, In_4Se_3

INTRODUCTION

Currently, one of the major research topics in the thermoelectric community is to improve the thermoelectric figure of merit ($ZT = \sigma S^2 T/\kappa$, where S, σ, κ , and T stand for the Seebeck coefficient, electrical conductivity, thermal conductivity, and absolute temperature, respectively) of traditional thermoelectric materials by implementing nanostructures or doping. Since the first report that bismuth telluride, Bi₂Te₃, is a highly effective thermoelectric material, enormous efforts have been dedicated to the discovery of new materials to further improve thermoelectric properties.¹ However, even now, bismuth telluride-based alloys are known as the best thermoelectric materials near room temperature.^{2–4}

Theoretically, elements in group 7 such as Cl, Br, and I can be used to optimize the carrier concentration of *n*-type Bi_2Te_3 - Bi_2Se_3 compounds. However, those elements cannot be used alone as dopants, as they are not stable with very high vapor pressures at high temperature. Instead, stable compounds such as CdCl₂ and SbI₃ can be used as dopants.⁵⁻⁷ However, there are no reports showing that doping with these dopants can improve the thermoelectric properties.

Recently, it was reported that single-crystal In₄Se_{2.35} compound has a large thermoelectric figure of merit of 1.48 at 705 K.^{8–10} The In–Se binary phase diagram contains a variety of intermetallic compounds such as In₂Se₃, InSe, and In₄Se₃ with large and complicated crystal structures.¹¹ In our previous study, we showed that three In-Se compounds (In₂Se₃, InSe, and In₄Se₃) consisting of nanoscaled structures exhibited *n*-type conductivity with Seebeck coefficients ranging from $-380 \ \mu V \ K^{-1}$ to $-570 \ \mu V \ K^{-1}$ at room temperature.¹² Thus, the

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