Journal of ELECTRONIC MATERIALS, Vol. 42, No. 7, 2013
DOI: 10.1007/s11664-013-2606-z

Effect of Cooling Conditions on the Microstructure and Thermoelectric Properties of Zn/Si-Codoped InSb

DONGHUN KIM,[1] KEN KUROSAKI,[1,3] YUJI OHISHI,[1] HIROAKI MUTA,[1] and SHINSUKE YAMANAKA[1,2]

1.—Graduate School of Engineering, Osaka University, 2-1 Yamadaoka, Suita, Osaka 565-0871, Japan. 2.—Research Institute of Nuclear Engineering, University of Fukui, 3-9-1 Bunkyo, Fukui 910-8507, Japan. 3.—e-mail: kurosaki@see.eng.osaka-u.ac.jp

InSb is a good candidate thermoelectric (TE) material owing to its high carrier mobility and narrow band gap around 0.18 eV. However, a high figure of merit (ZT) value has not been achieved with InSb because of its high lattice thermal conductivity (κ_{lat}). To reduce the κ_{lat} of InSb, we prepared a $ZnIn_{18}SiSb_{20}$ alloy by Zn/Si codoping into the In lattice sites of InSb. Polycrystalline samples of $ZnIn_{18}SiSb_{20}$ were prepared by a solid-state reaction method combined with hot pressing. To investigate the microstructures and TE properties resulting from different cooling conditions, samples were prepared by water quenching or slow cooling after an annealing process. The different cooling conditions led to different $ZnIn_{18}SiSb_{20}$ microstructures and TE properties. The electrical transport properties showed that both samples exhibited metal-like behavior and p-type conduction. The thermal conductivity values of the quenched and slow-cooled samples at room temperature were 8.7 W m^{-1} K^{-1} and 11.7 W m^{-1} K^{-1}, respectively. A maximum ZT value of 0.23 was obtained at 723 K for the quenched $ZnIn_{18}SiSb_{20}$ sample.

Key words: Thermoelectric, InSb, zincblende structure, $ZnSiSb_2$, chalcopyrite structure, thermal conductivity

INTRODUCTION

The effectiveness of thermoelectric (TE) materials is determined by the dimensionless figure of merit, $ZT = S^2\sigma T/(\kappa_{el} + \kappa_{lat})$, where S is the Seebeck coefficient, σ is the electrical conductivity, T is the absolute temperature, κ_{el} is the electrical thermal conductivity, and κ_{lat} is the lattice thermal conductivity. The most important issue in research on TE materials is to increase the electrical transport properties ($S^2\sigma$) but decrease κ ($\kappa = \kappa_{el} + \kappa_{lat}$). However, since S, σ, and κ_{el} are interrelated, it is very difficult to control them independently. Thus, reducing κ_{lat} is essential to enhance the ZT value.[1]

Over the past few decades, InSb, which has a narrow band gap with high electron mobility, has been considered to be a good candidate TE material. The carrier mobility of InSb single crystals (10^4 cm^2 V^{-1} s^{-1} to 10^5 cm^2 V^{-1} s^{-1} at 300 K) is the highest among III–V semiconductors. However, a high ZT value has not been achieved with InSb because of its large κ of ~17 W m^{-1} K^{-1}.[2–4] Thus, most research on InSb-based TE materials has focused on reducing its large κ.[5–9]

Substitution of other elements into a crystal lattice is known to be an effective method for enhancing phonon scattering due to the differences in atomic mass and size between the host and impurity atoms.[10–12] Solid solutions with the same crystal structure show good TE performance, e.g., the PbTe-PbSe, Bi_2Te_3-Bi_2Se_3, InSb-GaSb, and Si-Ge[13–16] systems. Recently, Skoug et al.[12] synthesized a copper-based ternary solid solution to reduce κ_{lat} and improve the TE properties. This copper-based system has the same crystal structure as InSb, i.e., the zincblende structure. Moreover, the $AgPb_mSbTe_{m+2}$ (LAST-m, LAST for lead antimony silver tellurium) system has attracted great attention due to its high ZT value of 1.7 at 700 K

(Received July 6, 2012; accepted April 13, 2013; published online May 24, 2013)