Journal of ELECTRONIC MATERIALS, Vol. 42, No. 7, 2013
DOI: 10.1007/s11664-012-2296-y

Solid-State Synthesis and Thermoelectric Properties of $Mg_2Si_{1-x}Sn_x$

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$Mg_2Si_{1-x}Sn_x$ ($0 \le x \le 1$) solid solutions have been successfully prepared by mechanical alloying and hot pressing as a solid-state synthesis route. All specimens were identified as phases with antifluorite structure. The electrical conduction changed from n-type to p-type at room temperature for $x \ge 0.5$ due to the intrinsic properties of Mg_2Sn. The absolute value of the Seebeck coefficient decreased with increasing temperature, and the electrical conductivity increased with increasing temperature; this is indicative of nondegenerate semiconducting behavior. The thermal conductivity was reduced by Mg_2Si-Mg_2Sn solid solution due to phonon scattering by the alloying effect.

Key words: Thermoelectric, magnesium silicide, solid solution, mechanical alloying

INTRODUCTION

The magnesium compounds Mg_2X (X = Si, Ge, Sn) and their solid solutions have attracted increasing attention as promising thermoelectric materials at temperatures ranging from 500 K to 800 K because they are nontoxic, environmentally friendly, and abundantly available.[1–3] Large Seebeck coefficient (α), high electrical conductivity (σ), and low thermal conductivity (κ) are necessary to improve the figure of merit ($ZT = \alpha^2\sigma T/\kappa$), where T is the temperature in Kelvin. Consequently, thermoelectric materials with high ZT value should have low lattice thermal conductivity and high carrier mobility.

In general, the thermal conductivity can be significantly reduced by phonon scattering at point defects such as in solid solutions, which make the low-frequency phonons decrease the thermal conductivity. Among Mg_2Si-Mg_2Sn, Mg_2Si-Mg_2Ge, and Mg_2Sn-Mg_2Ge solid solutions in the Mg_2X system, $Mg_2Si_{1-x}Sn_x$ are expected to exhibit higher ZT because of the greater difference in atomic mass between Si and Sn.[4,5] Zhang et al.[6] reported $ZT = 0.25$ at 400 K for $Mg_2Si_{0.4}Sn_{0.6}$ fabricated by induction melting and spark plasma sintering. Luo et al.[7] reported $ZT = 0.1$ at 420 K for $Mg_2Si_{0.8}Sn_{0.2}$ prepared by solid-state reaction and spark plasma sintering.

In this study, to reduce compositional change due to volatilization and oxidation caused by the Mg element, $Mg_2Si_{1-x}Sn_x$ solid solutions were synthesized by mechanical alloying (MA) and hot pressing (HP) as a solid-state route. Microstructural and thermoelectric property changes with solubility were examined.

EXPERIMENTAL PROCEDURES

$Mg_2Si_{1-x}Sn_x$ ($0 \le x \le 1$) solid solutions were synthesized by MA and HP. High-purity Mg (99.99%, $<149\ \mu$m), Si (99.99%, $<45\ \mu$m), and Sn (99.999%, $<75\ \mu$m) were weighed at stoichiometric ratio and mixed homogeneously. The mixed powders and hardened-steel balls (5 mm diameter) were loaded into a hardened-steel vial in Ar atmosphere at weight ratio of 1:20. The vial was then loaded into a planetary ball mill (Pulverisette 5; Fritsch) and

(Received June 28, 2012; accepted September 25, 2012; published online October 26, 2012)