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The Influence of Grain Boundary Scattering on Thermoelectric Properties of Mg_2Si and $Mg_2Si_{0.8}Sn_{0.2}$

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The temperature dependences of the Seebeck coefficient, and electrical and thermal conductivities of bulk hot-pressed Sb-doped *n*-type Mg_2Si and $Mg_2Si_{0.8}Sn_{0.2}$ samples were measured in the temperature range from 300 K to 850 K together with the Hall coefficients at room temperature. The features of the complex band structure and scattering mechanisms were analyzed based on experimental data within the relaxation-time approximation. Based on the obtained model parameters, the possibility of improvement of the thermoelectric figure of merit due to nanostructuring and grain boundary scattering was theoretically analyzed for both Mg_2Si and the solid solution.

Key words: Thermoelectrics, nanocrystalline materials, electron and phonon transport, grain boundary scattering

INTRODUCTION

In recent years, nanostructuring of bulk materials of different types aimed at increasing the thermoelectric figure of merit has been investigated using different methods.[1–12] Among them there were materials based on Bi_2Te_3–Sb_2Te_3 obtained by ball milling (BM)[1] or melt spinning[2] with subsequent hot pressing (HP), PbTe-based materials with endotaxially grown nanoinclusions,[3] and nanostructured (BM/HP) materials based on Si-Ge solid solutions,[4,5] to name a few. The efficiency of thermoelectric energy conversion is determined by the thermoelectric figure of merit $ZT = S^2\sigma T/\kappa$, where T, S, σ, and κ are the absolute temperature, Seebeck coefficient, and electrical and thermal conductivities, respectively. In these materials, the thermoelectric figure of merit ZT was increased compared with initial solid solutions due to specific scattering mechanisms including scattering of charge carriers and phonons on grain boundaries and nanoinclusions. In the present work, Mg_2Si and $Mg_2Si_{0.8}Sn_{0.2}$ solid solution were investigated. Materials based on $Mg_2Si_{1-x}Sn_x$ solid solutions for $0.4 \le x \le 0.6$ proved to be good thermoelectrics with high $ZT = 1.1$ at 800 K.[7–9] The materials with smaller tin content considered here are also promising due to their sharper increase of ZT with temperature and the possibility of obtaining larger values of specific thermoelectric figure of merit (ZT/D, where D is density). So, it is interesting to investigate the possibility of further improvement of these materials through nanostructuring techniques. Attempts at such investigation were performed recently.[10–12] The mechanical and structural properties were investigated in Refs. 10, 11. In Ref. 12, material preparation and measurements of transport coefficients and the thermoelectric figure of merit of bulk nanostructured Mg_2Si obtained by ball milling and hot pressing were reported. Though the comparison of samples with 30 nm grain size with microsized grain samples showed an increase in ZT of 38%, the absolute value of the thermoelectric figure of merit was too low (0.36 at 823 K). Theoretical analysis of the influence of nanostructuring on materials of different types was performed in Refs. 6, 13–17. In these works it was

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