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Solid-State Synthesis and Thermoelectric Properties of Sb-Doped Mg_2Si Materials

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Sb-doped magnesium silicide compounds have been prepared through ball milling and solid-state reaction. Materials produced were near-stoichiometric. The structural modifications have been studied with powder x-ray diffraction. Highly dense pellets of $Mg_2Si_{1-x}Sb_x$ ($0 \le x \le 0.04$) were fabricated via hot pressing and studied in terms of Seebeck coefficient, electrical and thermal conductivity, and free carrier concentration as a function of Sb concentration. Their thermoelectric performance in the high temperature range is presented, and the maximum value of the dimensionless figure of merit was found to be 0.46 at 810 K, for the $Mg_2Si_{0.915}Sb_{0.015}$ member.

Key words: Solid-state reaction, ball milling, antimony doping homogeneity

INTRODUCTION

Silicide compounds seem to be an advantageous choice for thermoelectrics, not only because of the ample availability of their constituent elements in nature but also because of their nontoxicity, which is consistent with the priority for technology that is safe for both the environment and humans. Mg_2Si-based compounds were found to have a favorable combination of physical and chemical properties and can form a good basis for development of new efficient thermoelectric devices.[1] Mg_2Si compound has already high ZT, of about 1, and its low density encourages the development of economical systems.

Regarding synthesis, it is difficult to prepare Mg_2Si by a melting process due to the large difference in vapor pressures of the constituent elements and their insolubility. Furthermore, it is difficult to control its composition, mainly due to volatilization and oxidation of Mg. In general, solid-state techniques, such as mechanical alloying or solid-state reactions, are preferable because they are relatively easier and better to control, applicable on a large scale, and demand simpler equipment. Mechanical alloying, which is usually preferred for industrial-scale applications, is not an effective technique for this material since severe agglomeration during milling is known to be a major problem. Dry milling is not practical under various conditions.[2] On the other hand, the wet ball-milling process was found to be a useful tool to fabricate nanocrystalline material using Mg_2Si as the starting powder.[3]

Pure Mg_2Si is an indirect-bandgap semiconductor (E_g = 0.77 eV) with less interesting thermoelectric properties, and for this reason, several either p- or n-type dopants have been proposed. Tani and Kido[4,5] first reported that Sb can act as a donor for Mg_2Si. In previous studies, Sb was used as a dopant in either small[6] or larger concentrations.[7] However, Nolas et al.,[7] produced materials with excess Mg, and limited their study to the low temperature range, while Jung et al.[6] produced material with rather low mobility.

In this work, we study high-mobility near-stoichiometric materials, in the high temperature range. A combination of ball-milling, solid-state reaction, and hot-pressing techniques is employed, aiming at completion of the reaction of Mg_2Si with Sb doping and the formation of $Mg_2Si_{1-x}Sb_x$ ($0 \le x \le 0.04$) in the form of highly dense pellets. The thermoelectric properties (Seebeck coefficient, electrical conductivity, thermal conductivity, and figure of merit) are discussed, while the local

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