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# Thermoelectric Properties of the Entire Composition Range in $Mg_2Si_{0.9925-x}Sn_xSb_{0.0075}$

M. SØNDERGAARD,[1] M. CHRISTENSEN,[1,2] K.A. BORUP,[1] H. YIN,[1] and B.B. IVERSEN[1]

1.—Department of Chemistry and iNANO, Centre for Materials Crystallography and Centre for Energy Materials, Aarhus University, 8000 Aarhus C, Denmark. 2.—e-mail: mch@chem.au.dk

Eleven samples of nominal composition $Mg_{2.2}Si_{0.9925-x}Sn_xSb_{0.0075}$ with $x$ = 0, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, 0.9925 were prepared by induction melting, ball milling, and spark plasma sintering. Hall effect, resistivity, Seebeck coefficient, and thermal conductivity measurements were conducted from room temperature to 400°C. Six of the samples were investigated for thermal stability by measuring powder x-ray diffraction while heating to 400°C. All samples were stable in air during the ~12-h-long data collection, except for $Mg_{2.2}Sn_{0.9925}Sb_{0.0075}$, which showed development of an elemental Sn phase after heating. The lattice parameter of each sample was extracted through Rietveld refinement and revealed a linear dependency on nominal composition. Measurement of top and bottom of the pellets exhibited systematic differences in lattice parameter and Seebeck coefficient, indicating that stoichiometry gradients are created during sintering.

**Key words:** Thermoelectric, $Mg_2Si_{1-x}Sn_x$, thermal stability, functionally graded materials

## INTRODUCTION

The efficiency of thermoelectric materials depends on material-specific parameters summarized in the dimensionless figure of merit, $zT$:

$$zT = \frac{S^2}{\kappa\rho}T,$$

where $S$ is the Seebeck coefficient, $\kappa$ is the thermal conductivity, $T$ is the absolute temperature, and $\rho$ is the electrical resistivity. For commercial interest, a $zT$ of unity or above is desired over an extended temperature range.[1–3] In recent years the $Mg_2Si_{1-x}Sn_x$ compounds have been investigated intensively for potential mid-temperature (300°C to 550°C) thermoelectric energy conversion.[4–12] In contrast to state-of-the-art thermoelectrics in this temperature range, the $Mg_2Si_{1-x}Sn_x$ compounds consist of earth-abundant and environmentally benign elements. The best compounds reported have Sn contents between $x$ = 0.3 to 0.7 and are $n$-doped with Sb to reach charge carrier concentrations of $1 \times 10^{20}$ $cm^{-3}$ to $3 \times 10^{20}$ $cm^{-3}$. These compounds reach $zT$ values of 1 and above from 250°C to 530°C.[11] Zaitsev et al.[4,5] reported on the end-members of the solid solution, $Mg_2Si$ and $Mg_2Sn$, having both light and heavy conduction bands with the light being the lowest for $Mg_2Si$ and vice versa for the $Mg_2Sn$, and the possibility for band convergence in the solid solutions. Recently, Liu et al.[11] reported on the convergence of the conduction bands for Sn contents in the range 0.65 to 0.7. This convergence was reflected in the higher numerical Seebeck coefficient of the samples. The method of fabrication can result in large deviations of the performance of the materials; e.g., Liu et al.[13] investigated the importance of excess Mg content. Due to the large difference in melting points between the constituent elements and the high vapor pressure of Mg, the stoichiometry can be challenging to control. Therefore, several synthesis methods have been suggested, including liquid encapsulation,[8] solid-state reaction,[9] direct spark plasma sintering synthesis,[14] and mechanical alloying.[15] The solid solution of $Mg_2Si_{1-x}Sn_x$ has been reported to contain

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