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Synthesis and Characterization of Al-Doped Mg_2Si Thermoelectric Materials

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Magnesium silicide (Mg_2Si)-based alloys are promising candidates for thermoelectric (TE) energy conversion for the middle to high range of temperature. These materials are very attractive for TE research because of the abundance of their constituent elements in the Earth's crust. Mg_2Si could replace lead-based TE materials, due to its low cost, nontoxicity, and low density. In this work, the role of aluminum doping (Mg_2Si:Al = 1:x for x = 0.005, 0.01, 0.02, and 0.04 molar ratio) in dense Mg_2Si materials was investigated. The synthesis process was performed by planetary milling under inert atmosphere starting from commercial Mg_2Si pieces and Al powder. After ball milling, the samples were sintered by means of spark plasma sintering to density >95%. The morphology, composition, and crystal structure of the samples were characterized by field-emission scanning electron microscopy, energy-dispersive spectroscopy, and x-ray diffraction analyses. Moreover, Seebeck coefficient analyses, as well as electrical and thermal conductivity measurements were performed for all samples up to 600°C. The resultant estimated ZT values are comparable to those reported in the literature for these materials. In particular, the maximum ZT achieved was 0.50 for the x = 0.01 Al-doped sample at 600°C.

Key words: Magnesium silicide, aluminum, thermoelectricity

INTRODUCTION

Magnesium silicide (Mg_2Si)-based alloys are promising candidates for thermoelectric (TE) energy conversion for the middle to high temperature range. These materials are very attractive for TE research because of the abundance of their constituent elements in the Earth's crust. Mg_2Si could replace lead-based TE materials, due to its low cost, nontoxicity,[1,2] and low density.

p-Type Mg_2Si can be produced by doping with Ag and Cu, whilst n-type material can be obtained by doping with Sb and Bi. Al doping of Mg_2Si is not completely clear and remains under debate due to the difficulty in predicting the substitutional site of Al in Mg_2Si.[3,4] However, both theoretical[5] and experimental[4,6] studies have shown that Al doping gives rise to n-type behavior, indicating that the Al atoms are mainly located at Mg sites.

The main hindrance of large-scale development of silicide-based devices is the high thermal conductivity value of the material. Several works have reported micron grain sizes of Mg_2Si obtained starting from pure elements (Mg, Si, Al) and employing melting processes or solid-state reaction syntheses.[4,6] The maximum ZT obtained was 0.67 for 3.0 at.% Al-doped Mg_2Si at 855 K.[6]

The aim of this work is to study small-grain-size silicide TE materials synthesized by employing commercial Mg_2Si raw pieces as starting material. This approach was chosen to avoid the difficulties and costs related to the employment of Mg metal powders, with a view to large-scale production of

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