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Microstructures and Thermoelectric Properties of Melt-Spun Zn_xSb_3 Ribbons

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Melt-spun Zn_xSb_3 ribbons were fabricated with weight compositions of x = 3.6, 3.9, and 4.2 through a single-wheel process and were annealed for 2 h at 673 K. The microstructures of the ribbons were investigated using transmission electron microscopy, together with energy-dispersive x-ray analysis. The main structure consisted of β-Zn_4Sb_3 phase, which mainly coexisted with ZnSb phase for $x < 4$ and with Zn phase for $x > 4$. The analyzed composition of the β-Zn_4Sb_3 phase deviated from the stoichiometric composition of 4:3 for all the ribbons. Nanosized voids and zinc inclusions were randomly distributed throughout the β-Zn_4Sb_3 phase. The thermoelectric characteristics of the ribbons were revealed by measuring the Seebeck coefficient, electrical conductivity, power factor, dimensionless figure of merit, and thermal conductivity. The power factor and dimensionless figure of merit increase with increasing x and temperature because either the electrical conductivity or Seebeck coefficient increases.

Key words: Zinc antimonide, thermoelectric materials, transmission electron microscopy, melt-spun ribbon

INTRODUCTION

β-Zn_4Sb_3-structured zinc antimonide is a promising thermoelectric material for generating power by directly converting heat into electricity. The conversion efficiency is characterized by the dimensionless figure of merit defined as $ZT = \alpha^2\sigma T/\kappa$, where α is the Seebeck coefficient, σ is the electrical conductivity, T is temperature, and κ is the thermal conductivity. The preparation process and structure of zinc antimonide have attracted considerable attention since a high ZT was reported for hot-pressed β-Zn_4Sb_3 samples.[1–8]

The structure of a single crystal of β-Zn_4Sb_3 was analyzed, and the β-Zn_4Sb_3 unit cell composed of $Zn_{36-n+i}Sb_{30}$ was found to consist of $36 - n$ zinc atoms at zinc sites, n zinc vacancies, 30 antimony atoms at antimony sites, and i interstitial zinc atoms.[9,10] The interstitial zinc atoms are supposed to act as glass-like contributors to reduce the thermal conductivity and increase the ZT of the crystal.

Transmission electron microscopy (TEM) observations have revealed that the β-Zn_4Sb_3 phase in hot-pressed samples of $Zn_{3.9}Sb_3$ and $Zn_{3.99}Sb_3$ contained randomly distributed voids, while that in a hot-pressed sample of $Zn_{3.9}Sb_3$ contained nanosized zinc inclusions.[11] The zinc inclusions are expected to effectively scatter phonons, thus contributing to phonon-glass behavior, which results in exceptionally low thermal conductivity in order to increase ZT.[11–13]

The aim of the present work is to investigate the structures and thermoelectric properties of isothermally annealed Zn_xSb_3 ribbons with various weight compositions through single-wheel melt-spinning.

EXPERIMENTAL PROCEDURES

High-frequency induction melting was used to prepare Zn_xSb_3 alloy ingots with weight compositions of x = 3.6, 3.9, and 4.2 by melting together

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