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Fabrication and Thermoelectric Power Factor of $CoSb_3$ Prepared using Modified Polyol Process and Evacuated-and-Encapsulated Sintering

AHMAD GHARLEGHI,[1] HON-ZI CHANG,[1] YIN-CHIAO CHEN,[1] YAO-WEI YANG,[1] and CHIA-JYI LIU[1,2,3]

1.—Department of Physics, National Changhua University of Education, Changhua 500, Taiwan. 2.—e-mail: ncuecj@yahoo.com 3.—liucj@cc.ncue.edu.tw

Various reaction temperatures have been investigated to stabilize the $CoSb_3$ phase obtained by a modified polyol process using a refluxing condenser. The resulting powders were cold-pressed and sintered in an evacuated-and-encapsulated ampoule for transport measurements. Single-phase $CoSb_3$ could be produced for the reaction carried out at 448 K for a short duration of 15 min followed by evacuated-and-encapsulated sintering at 848 K for 5 h. In comparison with the sample sintered at 798 K, the sample sintered at 848 K exhibits lower resistivity and thermopower. Due to the sign crossover of the thermopower, the latter sample shows lower power factor for temperatures above 475 K.

Key words: Thermoelectrics, cobalt skutterudites, modified polyol synthesis

INTRODUCTION

Engineering of thermoelectric (TE) materials for use in energy conversion technology has recently attracted huge interest because of the possibility of directly converting both existing heat in nature and waste heat generated in industry into electrical power. The potential of a TE material is evaluated by the dimensionless figure of merit (ZT), which is expressed as $ZT = S^2\sigma T/\kappa$. where S is the Seebeck coefficient, σ is the electrical conductivity, T is the absolute temperature, and κ is the total thermal conductivity. According to this definition, ZT values can be improved by increasing the power factor ($S^2\sigma$), decreasing κ, or optimizing both of them for a given system.[1,2]

Skutterudite compounds with MX_3 composition (where M is a metal atom such as Co, Rh, or Ir and X represents a pnictogen atom such as P, As, or Sb) have promising ZT values for wide usage in TE technology. These compounds have a body-centered cubic structure with 32 atoms in the unit cell and space group $Im\,\bar{3}$; there are also two voids per cell in the structure.[3] Different types of charge carrier have been reported for $CoSb_3$.[4,5] Liu et al. showed that compensating up to 6 at.% Sb when fabricating $CoSb_3$ led to an intrinsic p-type semiconductor in the temperature range from 50°C to 450°C.[6] Moreover, a theoretical study by Koga et al. predicted positive thermopower for undoped $CoSb_3$ compound at low temperature.[7] On the other hand, Zhao et al. demonstrated n-type $CoSb_3$ with reduced thermal conductivity due to the nanostructure size effect.[8,9]

Sign crossover of the thermopower has been reported in $CoSb_3$. Both filling of cages and doping at host ion sites can shift the sign crossover temperature.[3,5,6,10–12] In this work, combinations of reaction temperature and duration were explored to produce fine $CoSb_3$ powders using a modified polyol process. As a result, single-phase $CoSb_3$ could be produced for the reaction carried out at 448 K for a short duration of 15 min followed by evacuated-and-encapsulated sintering at 848 K for 5 h. The resistivity of the as-prepared sample was rather large. The low power factor arises from both its large resistivity and mixed conduction behavior.

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