

Account Paper

Thermoelectric Performance of In Filling in $\text{Co}_{0.33}\text{Fe}_{0.33}\text{Ni}_{0.33}\text{Sb}_3$ Skutterudite

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Abstract

Thermoelectric (TE) performance of CoSb_3 skutterudite materials can be improved by reducing their high thermal conductivity (κ). CoSb_3 has an open structure that can accommodate filler atoms, which enhance phonon scattering and consequently lower the κ . In the present work, indium (In) filling effect on TE properties of $\text{Co}_{0.33}\text{Fe}_{0.33}\text{Ni}_{0.33}\text{Sb}_3$ has been investigated. We synthesized a series of $\text{In}_x\text{Co}_{0.33}\text{Fe}_{0.33}\text{Ni}_{0.33}\text{Sb}_3$ ($x = 0, 0.05, 0.1, 0.15$, and 0.2) polycrystalline samples by solid state reaction method and hot pressed at 600°C to improve their densities. All the samples were crystallized in single skutterudite phase. Negative Seebeck coefficient was observed for all compositions. The introduction of In atoms into the $\text{Co}_{0.33}\text{Fe}_{0.33}\text{Ni}_{0.33}\text{Sb}_3$ skutterudite structure effectively enhanced its TE properties by optimizing its electronic and phonon transport characteristics. The reduction in electrical resistivity and lattice thermal conductivity on filling $x = 0.15$, In amount has resulted in the high figure of merit ZT .

Keywords: In filling, phonon scattering, lattice thermal conductivity, skutterudite, thermal conductivity

1. Introduction

Thermoelectric (TE) technologies have attracted attention as energy-saving technology since they can directly convert the waste heat to electricity. However, current commercial TE materials typically have low conversion efficiencies, which limit their potential for large-scale energy production. The efficiency of TE material depends on ZT , given by: $ZT = (S^2T/\rho\kappa_{\text{total}})$ where S , T , ρ and κ_{total} are Seebeck coefficient, absolute temperature, electrical resistivity, and total thermal conductivity, which is the summation of electronic (κ_e) and lattice (κ_L) components. To achieve the high ZT value, the material should possess high-power factor (S^2/ρ) and low κ_{total} . Meanwhile, the interdependency of these variables creates challenges in enhancing the efficiency of the TE materials [1, 2].

CoSb_3 -based skutterudites are promising materials for TE devices in intermediate temperature range ($500\text{--}900\text{K}$). However, they intrinsically have high κ , which becomes a weakness for achieving the high ZT values. Various approaches such as nano-structuring and alloying have been taken for CoSb_3 to reduce the

κ . Taking advantage of the characteristics of interstitial voids in the body-centered cubic (bcc) structure, filling atoms in the void site has also been developed. Filling these voids with guest atoms induces a rattling effect which enhances phonon scattering and thus reduces the κ_L . Atoms like La, Yb, Eu, and Ce have been extensively studied to achieve high ZT values [3]. Previous studies showed that combination of adding filler atoms such as Ce, and Yb into void site and alloying Co site led to higher ZT [4, 5]. In this regard, we studied indium (In) filling effect, which enables us to handle the materials without inert gas environment dislike the

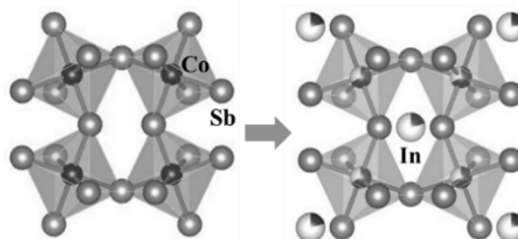


Fig. 1 Crystal structure of CoSb_3 and $\text{In}_x(\text{Co,Fe,Ni})\text{Sb}_3$

previous rare-earth elements, to the alloyed CoSb_3 skutterudites samples i.e. $\text{In}_x\text{Co}_{0.33}\text{Fe}_{0.33}\text{Ni}_{0.33}\text{Sb}_3$ ($x = 0, 0.05, 0.1, 0.15$ and 0.2).

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