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The low and high temperature thermoelectric properties of Sb doped Cu_2SnSe_3

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ABSTRACT

The effect of Sb doping on the thermoelectric properties of Cu_2SnSe_3 was investigated. The $Cu_2Sn_{1-x}Sb_xSe_3$ ($0 \le x \le 0.04$) compounds were prepared by solid state synthesis. The powder X-ray diffraction pattern of the samples showed a cubic structure (space group $F\bar{4}3m$). The electrical resistivity decreased with increase in Sb content up to $x = 0.02$, then it increased with further increase in x. The electrical resistivity data follows variable hopping model at low temperatures. The Seebeck coefficient for all the samples is positive and analysis of data confirms that the variable hopping process is operative at low temperatures. The thermal conductivity is found to decrease with increase in Sb concentration, presumably due to point-defect scattering as a result of Sb substitution. The highest value of figure of merit at 400 K is equal to 0.0137 for the sample $Cu_2Sn_{0.99}Sb_{0.01}Se_3$ which is about eight times greater than that of the pristine sample.

1. Introduction

Thermoelectric materials, which have the ability of converting heat energy directly into electrical energy, are potential candidates to provide source of clean energy. However, the conversion efficiency of most of the thermoelectric materials is too low to be used for large scale applications. The efficiency of thermoelectric materials is given by a dimensionless quantity known as figure of merit (ZT) and is expressed by

$$ZT = S^2T/\kappa r \quad (1)$$

where S is the Seebeck coefficient, ρ is the electrical resistivity, κ is the total thermal conductivity, and T is the absolute temperature. Searching for high efficiency thermoelectric materials is a challenging task because S, ρ and κ are interdependent for any material. In order to attain high ZT values, high Seebeck coefficient, low electrical resistivity and low thermal conductivity must be achieved. It is well-known that materials with high thermoelectric performance occur when the carrier concentrations are in the range 10^{19}–10^{21} cm^{-3}, which is usually observed in semiconductors [1–5].

Thermoelectric materials have two applications viz. thermoelectric generators and coolers. Thermoelectric generators work on the principle of Seebeck effect i.e. a voltage is developed across the junction of two dissimilar materials when subjected to a temperature gradient. Thermoelectric coolers work on the principle of Peltier effect which is just the reverse of Seebeck effect. Most commonly used thermoelectric materials for power generation are PbTe [6–8] and $AgPbTe_2$ [9]. On the other hand, Bi_2Te_3-based compounds are used as Peltier based cooling devices [10]. Unfortunately the elements which are most commonly used in thermoelectric materials are toxic (e.g., Pb, Te), and are not abundantly found in Earth's curst. Thus, using non-toxic element such as Se which is also a naturally abundant element to replace or partially substitute for Te is a common strategy in enhancing/optimizing the ZT values in the Te-contained thermoelectric materials.

Several concepts have been proposed to enhance ZT by reducing thermal conductivity. Among them, one of the most successful achievements in the development of high performance thermoelectric materials is the concept of "Phonon-Glass-Electron-Crystal" (PGEC) as proposed by Slack [11,12]. It was suggested that an ideal thermoelectric material should possess good

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electrical transport properties like crystals and low thermal conductivity like glasses. During last two decades, optimization of thermoelectric properties using PGEC methodology has led to a significant enhancement in *ZT* values of certain thermoelectric materials [4,5,13]. Recently, the concept of PGEC has also been extended to "Phonon-Liquid-Electron-Crystal" (PLEC) which explains the extremely low lattice thermal conductivity observed in superionic materials like $Cu_{2-x}Se$. Thus, high value of *ZT* can be achieved in this class of materials because they have two separate sub-lattices and large Se atoms form a rigid and solid sub-lattice

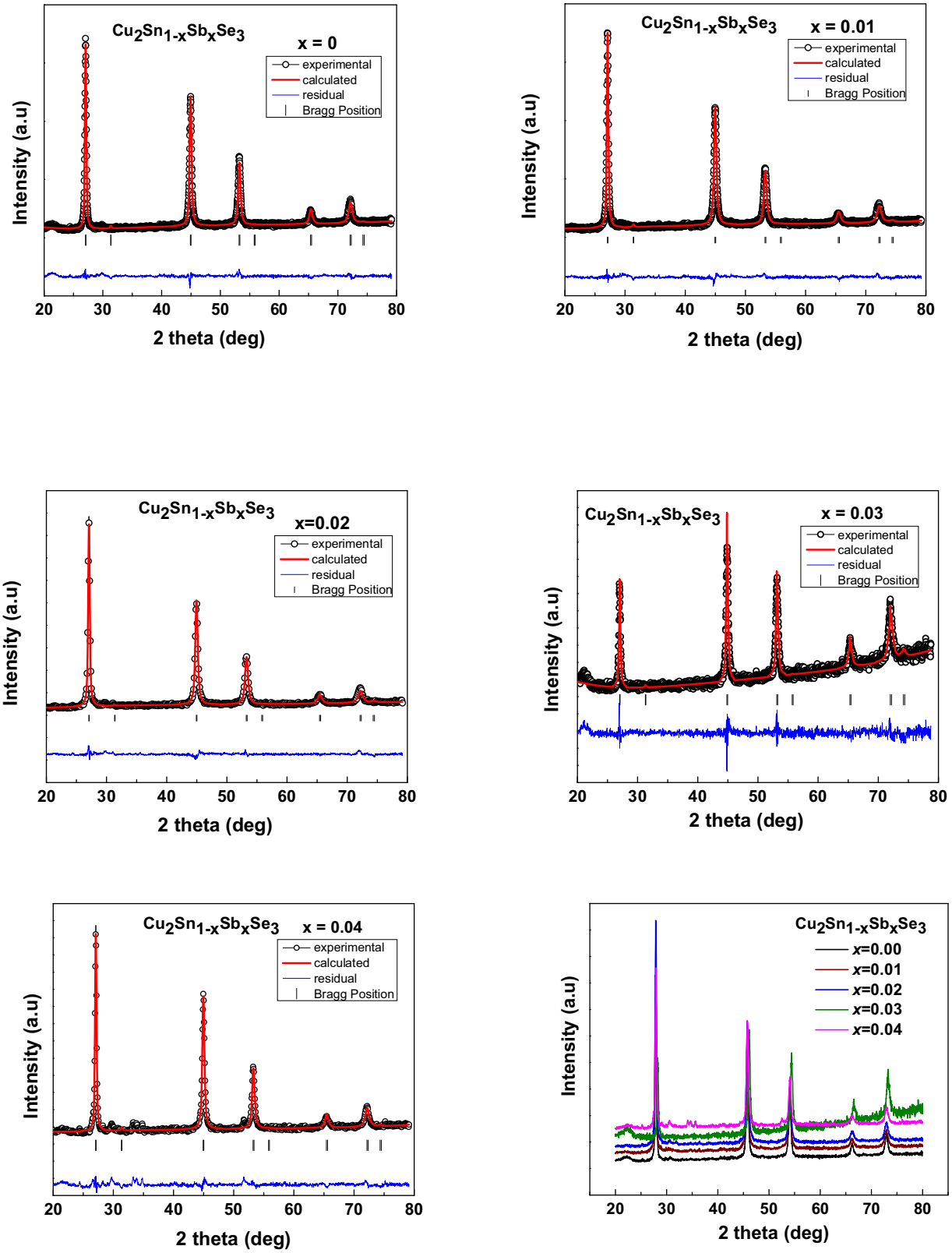

Fig. 1. Rietveld refinement plots of $Cu_2Sn_{1-x}Sb_xSe_3$ ($0 \leq x \leq 0.04$).