Synthesis &Tailoring the Thermal Conductivity of Sr Doped Bi_2Se_3 Thermoelectric Material

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Abstract: We have investigated the thermal transport properties of $Sr_xBi_{2-x}Se_3$ (x=0, 0.05, 0.2). The samples were synthesized by melt route method followed by vacuum hot press. The structural and morphological information of sample has been retrieved using x-ray diffraction (XRD) and scanning electron microscopy (SEM). The thermal transport measurement were performed in the temperature range of 300-550 K. It is found that with increasing Sr content the total thermal conductivity of the material decreases which is attributed to the enhance phonon scattering due to natural grown layered structure and defect induced by Sr doping.

Keywords: Thermoelectric material, Thermal transport properties.
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INTRODUCTION

Growing needs of energy sustainability have given great attention to the field of thermoelectric materials as they directly transforms the waste-heat into useful electrical energy with the advantage of being reliable, lightweight, robust and environment friendly[1]. The efficiency of thermoelectric device is characterized by the dimensionless figure-of-merit, $ZT= (\alpha^2\sigma/\kappa)T$, of the material where α is the Seebeck coefficient, σ is the electrical conductivity and κ is the thermal conductivity. Thus, the ideal thermoelectric material should exhibit high carrier mobility and low thermal conductivity [2]. Bismuth selenide based alloys is one of the conventional thermoelectric materials with narrow band gap of about 0.3 eV and is widely used in development of thermoelectric refrigerator and thermoelectric generator near room temperature applications. [3] Bi_2Se_3 crystallizes in a rhombohedral structure with space group (R-3m). This structure consists of five-atom layers arranged along the c-axis known as quintuple layer as

...Se-Bi-Se-Bi-Se…..Se-Bi-Se-Bi-Se...

The Bi and Se layers are connected by strong covalent/ionic bonds while neighboring Se layer held together by weak Van der Wall type forces.

Thermal conductivity is one of the key parameters in determining the *ZT* of thermoelectric materials. Over the past decade, major progress in the thermoelectric performance of the thermoelectric materials has been made by tailoring their thermal conductivity by adopting various strategies like disorder unit cell, resonant scattering by localized rattling atoms interface scattering etc. while preserving their power factor values [4]. Tailoring of the thermoelectric properties in Bi_2Se_3 systems using various dopants has been successfully evolved since last decade. It has been reported that the Sr doped Bi_2Se_3 is a superconducting topological insulator [5], however no results are reported on the thermal transport behavior of this material above room temperature. In present work, we have tailored the thermal transport properties of $Sr_xBi_{2-x}Se_3$ (x=0, 0.05, 0.2) in the temperature range of 300-550 K.

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EXPERIMENTAL DETAILS

$Sr_xBi_{2-x}Se_3$ (x=0, 0.05, 0.2) samples were prepared by vacuum melt route. The stoichiometric amounts of Sr (99.99%), Bi (99.99%) and Se (99.99%) were sealed in quartz ampoule under a vacuum of 2×10^{-5} mbar. For the uniform mixing of the starting material the vacuum sealed ampoule was kept in a home-made rocking furnace at temperature of 1073 K for 4hr with heating rate of 50^0C/hr. After dwell the furnace was slowly cooled down to room temperature at controlled cooling rate of 10^0C/min. The obtained shiny ingot from the ampoule was finely ground into fine powders and densified at sintering temperature of 873 K for the duration of 60 minutes with an applied load of 2.5 Kg. For the measurement of thermal transport properties disc shaped sample (diameter ~ 10 mm) were hot-pressed and polished. The structural and micro structural characterization of the samples was carried out by PROTO made x-ray diffraction (XRD) using (Cu-Kα) x-ray source and Tescan Vega MV2300T/40 Scanning Electron Microscope (SEM). Thermal transport properties were measured using Linseis make Laser Flash Analysis (LFA-1000) instrument in the temperature range from 300-550K.

RESULTS AND DISCUSSIONS

Structural characterization and morphology

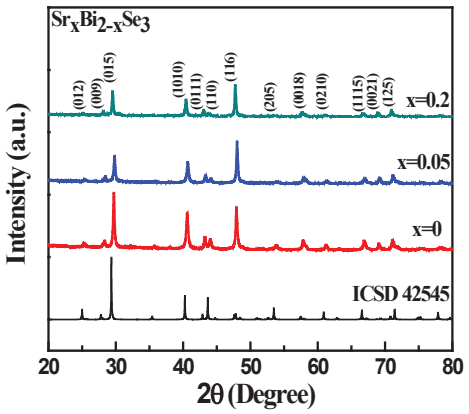

Fig. 1 (a) XRD pattern of $Sr_xBi_{2-x}Se3$

Fig.1 (a) shows the XRD patterns for the synthesised $Sr_xBi_{2-x}Se_3$ (x=0, 0.05, 0.2) hot pressed pellet. The peaks in XRD pattern can be well indexed as rhombohedral phase (ICSD No 42545). It can be seen that there are no impurity peaks suggesting the formation of single phase Bi_2Se_3 compound with lattice parameter a=b=4.137 Å and c=28.630 Å. Intense XRD peaks suggest that the sample are highly crystalline in nature.

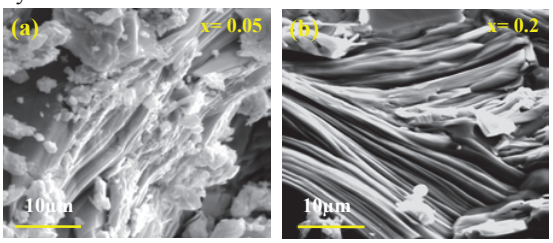

Fig. 2 SEM of fractured hot press pellet with (a) x=0.05 and (b) x=0.2 of $Sr_xBi_{2-x}Se3$.

The morphology of the hot pressed samples were checked by capturing cross section image using SEM. Fig 2 shows SEM image of Sr intercalated Bi_2Se_3 samples which clearly indicates that Sr do not suppress layered growth of the material. Samples show dense and uniform layer structure in the hot press sample. Presence of layer structure provides micron length scale multiple interfaces which can effectively hinder the phonon transport and help in tuning the thermal transport properties.

Thermal Transport Properties

Figure 3(a), 3(b) and 3(c) shows the temperature dependent specific heat, thermal diffusivity and thermal conductivity data, respectively for $Sr_xBi_{2-x}Se_3$ (x=0, 0.05, 0.2) samples.

The specific heat of the samples shown in fig.3 (a) is measured using comparative method using graphite as reference material. The specific heat of sample using comparative method can be estimated by considering approximation that the specific heat of sample is comparable to the reference standard material, according to the following equation,

$$Cp_{sample} = \frac{Cp_{ref} * \Delta T_{ref} * m_{ref}}{\Delta T_{sample} * m_{ref}} \quad \ldots\ldots\ldots\ldots (1)$$

Where ΔT is the raise in temperature and m is the weight.

It is observed that the specific heat of the samples increases systematically with increasing Sr content. The thermal diffusivity of the samples, shown in fig. 3 (b), is measured simultaneously with specific heat. With increasing Sr content thermal diffusivity in the sample decreases which remain almost consistent with increasing temperature. The thermal conductivity (k) of the samples is related to the specific heat (Cp) and thermal diffusivity (D) by following Equation

$$k = D\rho Cp \quad \ldots\ldots\ldots\ldots \quad (2)$$

For calculating thermal conductivity of the samples the physical density of sample is estimated using Archimedes method

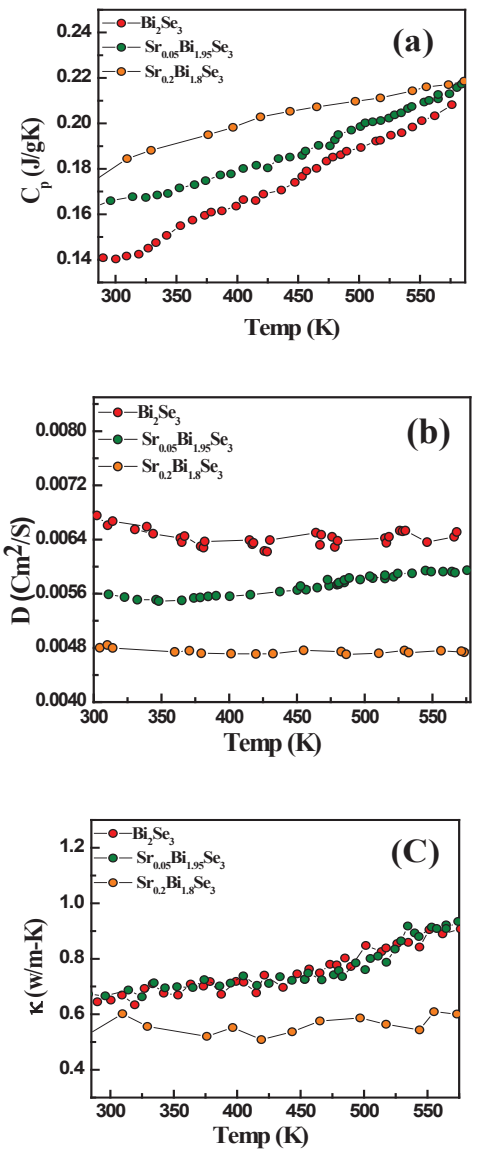

Fig.3 Temperature dependence of (a) specific heat (b) Thermal diffusivity and (c) Thermal conductivity of $Sr_xBi_{2-x}Se_3$.

The temperature dependent thermal conductivity of $Sr_xBi_{2-x}Se_3$ (x=0, 0.05, 0.2) samples estimated using eq. 2, is shown in fig. 3(c) It is observe that with increasing Sr content the thermal conductivity in the samples reduces. Reduction in thermal conductivity with increasing Sr content can be understood in term of enhances phonon scattering due to large number of grain boundaries, defects and micron length scale interfaces. Existence of various length scale scattering centers in the system reduces the mean free path of phonons which suppresses the thermal conductivity of sample

CONCLUSIONS

Thermal transport properties of $Sr_xBi_{2-x}Se_3$ with (x=0, 0.05, 0.2) have been studied. We have seen that with increasing strontium content in Bi_2Se_3 results in reduced thermal conductivity which remains stable with increasing temperature. Reduced thermal conductivity is attributed to the layered structural growth and defect induced due to strontium intercalation in Bi_2Se_3 structures. Reduction in thermal conductivity gives a scope to further explore other thermoelectric parameters in this system for concluding its thermoelectric performance.

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