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Enhancing the Thermoelectric Performance of Bismuth Telluride through Silver Doping

Jai shree Choudhary[a], Karan Grover[a], Monika Tomar[c, d], Anjali Sharma[b, d], & Ranjana Jha[a*]

[a]Research lab for Energy System, Department of physics, Netaji Subhas University of Technology, New Delhi 110 078, India

[b]Department of physics, Atma ram Santan Dharma College, University of Delhi, New Delhi 110 021, India

[c]Department of Physics, Miranda House, University of Delhi, New Delhi 110 007, India

[d]Institute of Eminence, University of Delhi, New Delhi 110 007, India

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In the present work, nanoparticles of pure Bi_2Te_3 and Ag doped Bi_2Te_3 have been prepared using hydrothermal technique. The thermoelectric performance of bismuth telluride has been found to be improving with Ag doping. This may be due to increase in the carrier concentration resulting in enhanced electrical conductivity of the material, and expected to maintain its low thermal conductivity. The morphology, structural properties of the nanoparticles synthesized in this study has been accessed through the utilization of scanning electron microscopy (SEM) and X-ray diffraction (XRD) technique. Thin films ofas prepared powders ofpure Bi_2Te_3 and Ag doped Bi_2Te_3nanoparticles have deposited on to glass substrate using thermal evaporation technique. The thermoelectric measurements of as synthesized materials have studied using an indigenously developed setup. Seebeck coefficient for pure and Ag doped bismuth telluride thin films has found to be 150 $\mu v\,^{\circ}C^{-1}$and 182 $\mu v\,^{\circ}C^{-1}$, respectively.

Keywords: Bismuth chalcogenides, Hydrothermal Technique, Seebeck effect, Thermoelectric generator (TEG)

1 Introduction

In recent years, thermoelectric devices have emerged as highly significant tools for effectively converting waste heat into valuable electrical energy. The dissipation of generated heat leads to a decrease in energy loss, consequently reducing the world's reliance on fossil fuels[1]Meanwhile, thermoelectric materials have become a preferred choice for energy generation over other options due to their advantages, including flexibility, compact size, lack of pollution, and potential for recycling. Bismuth telluride is a widely utilized material in thermoelectric devices operating near room temperature. This is primarily due to its intrinsic properties exhibited by the material. which encompasses including reduced lattice thermal conductivity and enhanced electronicmobility. These properties make it an excellent candidate for efficient thermoelectric energy conversion in this temperature range.[1, 2]

Bismuth telluride-based compounds are widely recognized as the predominant thermoelectric materials in commercial applications, exhibiting a significantly high figure of merit (ZT) at room temperature.[3] Efficiency of the thermoelectric material is quantified using the figure of merit, $ZT = S^2\sigma T/k$, where S represents Seebeck's coefficient or thermoelectric power, σdenotes the electrical conductivity, T signifies the temperature and k represents the thermal conductivity. These parameters are strongly dependent on each other. By decreasing k and increasing σ can lead to the enhancement in the ZT values. The advancement of advanced material processing techniques has led to the development of new nanostructures that exhibit improved ZT value. Although super lattice structures and nano-wires/rods have exhibited promising ZT values, their widespread application in large-scale energy-conversion systems have encountered significant challenges due to constraints associated with heat transfer mechanisms and cost limitations. Enhancement of the ZT value can be attained through three fundamental strategies: augmenting electrical conductivity, minimizing thermal conductivity and enhancing thermoelectric power. In solids, heat conduction occurs through different carriers, including electrons, lattice waves (phonons), magnetic excitations, and, in specific instances, electro magnetic radiation[4,5]. The incorporation of

*Corresponding author (E-mail: drranjanajha@gmail.com)

nanostructures in materials leads to an increased concentration of grain boundaries, which, in turn, enhances phonon scattering at these interfaces, leading to noteworthy decrease in thermal conductivity.

In order to attain optimal thermoelectric performance, it is desirable to have a material characterized by a high Seebeck coefficient, elevatedσ, and minimalk. Nevertheless, the enhancement of thermoelectric performance presents significant challenges owing to the inherent interdependence between electrical conductivity and thermal conductivity.

In this work, to enhance the thermoelectric power and the electrical conductivity of pure bismuth telluride by doping Ag. It is found that by incorporating Ag into the lattice voids, the thermoelectric performance of the Bi_2Te_3 thin filmis effectively controlled. However, the presence of numerous surface defects may hinder the enhancement of the thermoelectric properties. The substrate heating during the thin film deposition process is expected to reduce the surface defects compared to post-annealing[6–8].The present work demonstrates the preparation of both pure and Ag doped Bi_2Te_3 thin films using thermal evaporation technique with *in-situ* annealing. The thermoelectric measurements of the as synthesized$Bi_2Te_3$and Ag doped Bi_2Te_3 thin films were performed using an indigenously developed See beck coefficient measurement setup.

2 Materials and Methods

2.1 Chemical Used

Tellurium powder (Te), Ethylene diaminetetra acetic, Acid Disodium Salt (Na_2-EDTA), Bismuth Trichloride Anhydrous ($BiCl_3$, 99.0%), Sodium tetra hydridoborate ($NaBH_4$), Silver Nitrate Ag NO_3.

All the chemicals used for the synthesis of Bi_2Te_3 and Ag doped Bi_2Te_3 nanoparticles were of high purity.

2.2 Material Synthesis

For the synthesis of Bi_2Te_3 and Ag doped Bi_2Te_3 nanoparticles Firstly, in a beaker6mmol of Te powder was mixed with 60 ml of Distilled water. Subsequently, 4mmol of $BiCl_3$ was added to the same mixture followed with 1 g Na_2-EDTA and then the mixture was stirred for 30 minutes using magnetic stirrer followed by adding $NaBH_4$(Reducing Agent) to obtain the black precipitate. For 1% doping of Ag,3mg of Ag $NO_3$was added in the similarly prepared another mixture as above. Pour the above two mixtures of pure and silver doped solutions in two different 100 ml stainless steel teflon lined autoclave. Further, the sealed autoclaves were kept in the hot air oven for 24hoursmaintained at at 180°C and subsequently the autoclaves were allowed to gradually cool down to room temperature. Finally, theBi_2Te_3 and Ag doped Bi_2Te_3 powders were centrifuged and washed with DI water and ethanol for 3to 4 times.

2.3 Thin film deposition

The deposition of thin films of Bi_2Te_3 and Ag doped Bi_2Te_3onto glass substrates was accomplished using the thermal evaporation technique. The thickness of the as grown films was examined by utilizing a quartz crystal thickness monitor attached in the chamber. Prior to the deposition process, the glass substrates underwent a thorough cleaning procedure with acetone followed by propanol using ultra-sonicator and was dried using N_2 air gun. The pure and Ag-doped thin films were prepared at a substrate temperature of 250°C, in a vacuum o fthe order of 6×10^{-6}Torrwith a deposition rate of 1-3Å/s. The thickness of the both Ag-doped and pure Bi_2Te_3 thin films was kept at 100 nm thick.

2.4 Characterization Techniques

The structural and phase identification analysis of the as synthesized samples were conducted using X-ray diffractometer (make: Rigaku, model: Mini Flex 600) employing Cu-Kα radiation (λ=1.5406A°). Surface morphological characterization was carried out by using scanning electron microscope (make:JEOL, model: JSM6700) operated at 20KV.Hall effect measurements (HMS-5000 series) were carried out in order to measure the electrical conductivity of the thin films and measured of Seebeck coefficients were done through an indigenously developed Seebeck coefficient setup.

3 Results and Discussion

3.1 X-Ray Diffraction Analysis

Figure 1 Represents the X-ray diffraction patterns of both Ag-doped and undoped Bi_2Te_3 thin films. The obtained patterns matched well with the JCPDS file no. 08-0021. The XRD peaks obtained at 27.8° ,40.9°, 46.3°and 68.9°anglescorresponds to (015), (1010), (0015)and (125) planes of Bi_2Te_3, respectively. The XRD peaks indicate that the as synthesized Bi_2Te_3 thin films possess rhombohedral structure with

polycrystalline nature. In Ag-doped Bi_2Te_3 thin films, peaks with hkl plane (0015) and (125) were found to be suppressed as depicted in the XRD pattern.

3.2 Scanning electron microscopy

The surface morphology of as synthesized Bi_2Te_3powder revealed agglomeration of fine needle like grains as depicted in SEM image Fig. 2(a-b)

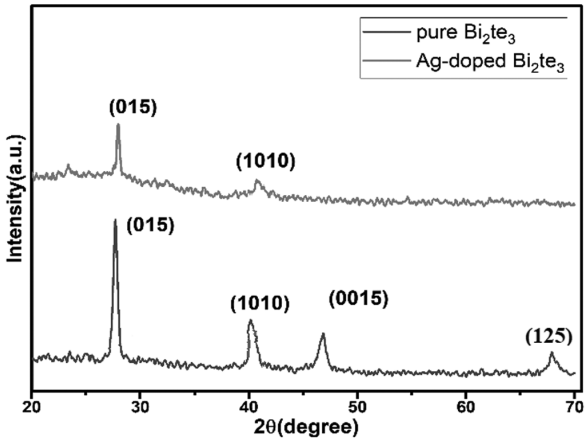

Fig. 1 — X-ray diffraction patternof pure Bi_2Te_3 and Ag-doped Bi_2Te_3thin films.

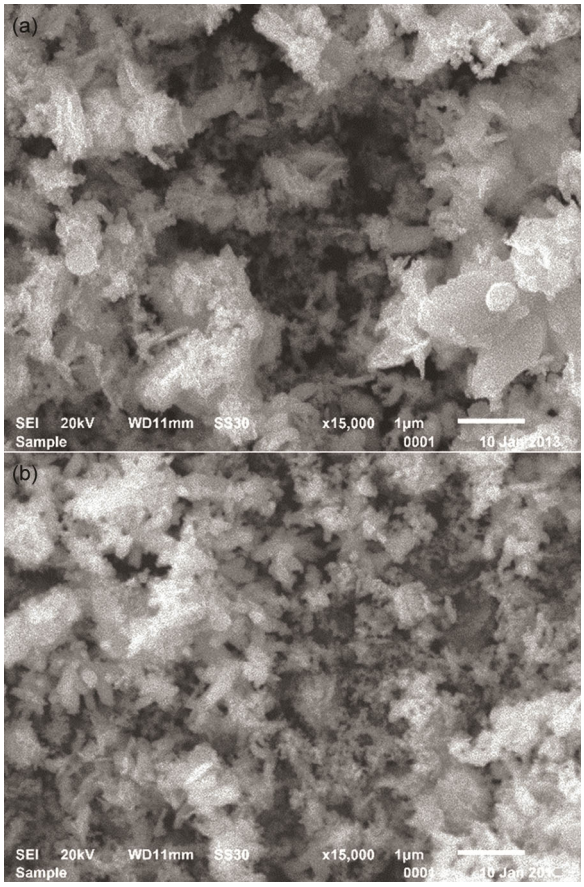

Fig. 2— (a) Scanning electron microscope micrographs of pure Bi_2Te_3, and (b) Ag-doped Bi_2Te_3.

shows the micrograph of Ag-doped Bi_2Te_3 powder shows the uniformly distribution of fine particles with comparatively less agglomeration as compared to un doped Bi_2Te_3 powder.

3.3 Hall Effect measurement

Hall Effect measurements of the pure and doped thin films were studied at 298k temperature with 5mA current. The electrical conductivity and of pure Bi_2Te_3 and Ag-doped Bi_2Te_3 thin films were found to be 9.4075E-01 Sm^{-1} and 2.3630E+00 Sm-1, respectively. The value of average Hall coefficient of pure Bi_2Te_3 and Ag-dopedBi_2Te_3 were observed in data were 5.3957E-03 $m^3A\text{-}S^{-1}$and -5.8453E-03 $m^3A\text{-}S^{-1}$, respectively. in both the films reveals that pure Bi_2Te_3 thin films has p-type carriers whereas Ag doped Bi_2Te_3 has n-type conductivity. Measurements show that there is increase in the electrical conductivity with the Ag doping.

3.4 Thermoelectric measurement

Thermoelectric measurements of the as prepared pure Bi_2Te_3 and Ag-doped Bi_2Te_3 thin films were carried out using an indigenously developed Seebeck measurement set up as shown in Fig. 3. Schematic of indigenously developed setup shows that it consists of glass chamber connected to a rotary pump to attain the vacuum. To achieve the temperature gradient in the sample both the ends are heated using the heater attached with PID controller along with thermocouples. Voltage generated due to temperature gradient is measured using Digital multimeter.

The Fig. 4(a-b) show the induced thermoelectric voltage as a function of material temperature difference for pure Bi_2Te_3 and Ag-doped Bi_2Te_3 thin films respectively. The Seebeck coefficient for pure and Ag doped bismuth telluride thin films were found

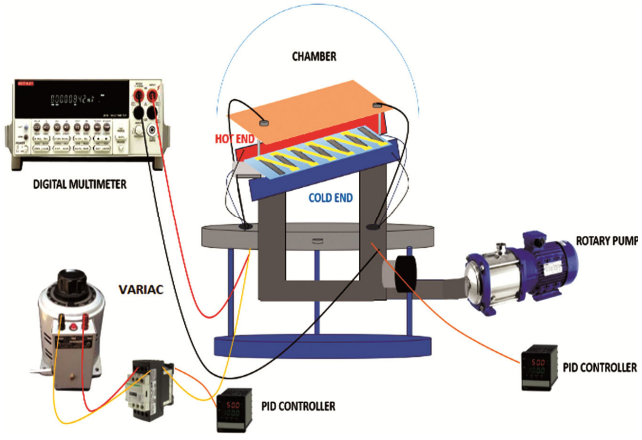

Fig. 3 — Schematic of Indigenously developed setup.

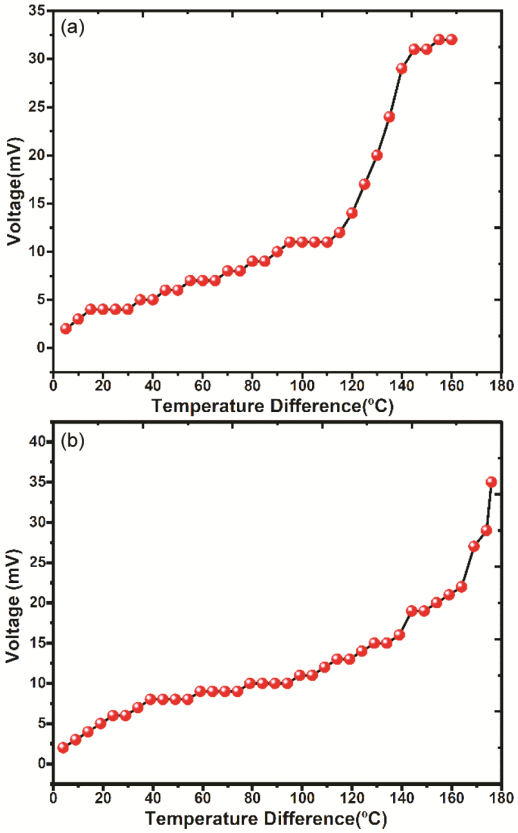

Fig. 4— (a) Seebeck coefficient of pure Bi_2Te_3, & (b) Ag-doped Bi_2Te_3.

to be 150 $\mu v^{\circ}C^{-1}$and 182 $\mu v^{\circ}C^{-1}$, respectively. Thus, with the doping of Ag in Bi_2Te_3, there is an evident enhancement in the electrical conductivity and therefore improved Seebeck coefficient is achieved.

4 Conclusion

A facile synthesis of nanoparticles of pure Bi_2Te_3 and Ag-doped Bi_2Te_3 has been successfully done using hydrothermal route, followed with thin film fabrication using thermal evaporation. On doping of Ag in Bi_2Te_3, the electrical conductivity and Seebeck coefficient has found to be increased significantly which may useful for the overall rise in the ZT value of Ag doped Bi_2Te_3 thin films[9]. This may be due to increase in the carrier concentration resulting in enhanced electrical conductivity of the material, and expected to maintain its low thermal conductivity.

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