Thermal Properties of Bismuth Antimony Telluride with Multi-Wall Carbon Nanotubes using Spark Plasma Sintering

C. K. Nabi, K. Ahmad, and M. A. Al-Eshaikh

***Abstract*— Thermal properties play a vital role in determining the performance of thermoelectric materials. These materials have applications in mainly all fields of life, where there is a considerable temperature difference across a medium. Currently, they are being used, in automobiles by utilizing heat from exhaust, in medical field as implantable medical devices which can utilize body heat, and as cooling devices in commercial transport vehicles.**

Therefore, in this work, we have studied the thermal properties of nanocomposite materials, prepared by uniform dispersion of 2 vol. % of multi-wall carbon nanotubes (MWCNTs) in Bismuth Antimony Telluride (BiSbTe). The MWCNTs were incorporated in pristine BiSbTe powder by uniform dispersion using ultra-sonication, and mild ball milling. The pure BiSbTe and composite powders were consolidated into a disc of thickness 2mm and diameter 12.7mm by Spark Plasma Sintering (SPS) for 3 minutes at temperature 415˚C and pressure 50MPa. Afterwards, the thermal conductivity of the bulk materials was measured using Laser Flash method (LFA-457) from temperature 300 to 500K.

It is observed that the thermal conductivity was reduced by the inclusion of MWCNTs, mainly due to increased phonon scattering. The minimum value of thermal conductivity in 2 vol. % MWCNTs/BiSbTe nanocomposite was found as 0.758 W/mK at 474K. The overall thermal conductivity in 2 vol. % MWCNTs/BiSbTe remains less than pure BiSbTe throughout the measured temperature range.

***Keywords*— BiSbTe, MWCNTs, Thermal Conductivity, Thermoelectric Materials**

I. Introduction

In recent years, advancements in the field of nanotechnology has open new doors in improving the efficiency of TE materials. The rising demand of sustainable energy has urged scientists to develop new materials which can efficiently convert waste heat into electricity. Thermoelectric materials have applications in all fields of life, where there is a considerable temperature difference across a medium. Currently, they have applications in automobile, nuclear, power plants and solar thermoelectric generator (STEG) and promising prospects in the field of medical science and refrigeration applications. [1]-[4].

Considerable research has been conducted during the last decade on low dimensional thermoelectric materials to improve their efficiency as compared to those of bulk thermoelectric materials using quantum confinement effects. Significant improvements in ZT have been achieved in superlattice structures like that of Bi2Te3/Sb2Te3, PbTe/PbSeTe, and Si/Ge [5]-[6]. Such materials are not compatible for large scale applications because of high fabrication cost and too thin to support considerable temperature differences. These constraints of using such materials in large-scale applications are forcing researchers to focus on bulk nanocomposite fabrication to realize nanoscale thermoelectric properties enhancement.

The efficiency of a TE material is closely related to the figure of merit (ZT), where ZT is given by,

$$ZT = \frac{S^2\sigma}{\mathrm{k}}\ T \qquad (1)$$

Where S is the Seebeck coefficient, σ is the electrical conductivity, k is the thermal conductivity and *T* is the absolute temperature. Therefore, the larger value of ZT requires low thermal conductivity (k).

Low dimensional material dispersion into thermoelectric materials has been suggested to reduce thermal conductivity and thus enhance the thermoelectric properties, due to decrease in the lattice thermal conductivity without affecting the electrical resistivity adversely [7]-[8]. In this work, special attention is given to uniform dispersion of MWCNTs into the bulk p-type BiSbTe nanocomposite [13]. We have used ball milling to form BiSbTe nanostructures and incorporated 2 vol. % MWCNTs into it. Spark plasma sintering (SPS) was used to get better consolidation without any grain growths. This technique has several advantages over conventional powder consolidation, such as rapid heating and cooling rates, short sintering time and manageable applied pressure [16]. The effect of nanotubes inclusion on the thermal properties of the bulk composites is analyzed.

II. Experimental Procedures

Pure BiSbTe (American Elements) in the form of lumps and MWCNTs (EMFUTUR, Spain) in powder form were used as

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raw materials. The pure BiSbTe powder was obtained from lump. 2 vol. % MWCNTs powder was dispersed and deagglomerated into BiSbTe powder by ultra-sonication (Q500 USA) and ball milling in an inert environment using a Planetary Micro Mill (PULVERISETTE 7 premium line, FRITSCH Germany). The resulted composite and pure BiSbTe powders were consolidated in graphite mold of 12.7mm diameter using SPS (Type HP D-50, FCT Systeme, Rauenstein, Germany) under 50Mpa pressure. The heating rate was kept at 100˚C/min and held for 3 minutes at 415˚C during SPS. The resulted samples were is the shape of a disc with a thickness of around 2mm and diameter 12.7mm. The densities were measured by Archimedes' principle which are found to be 5.86g/cm^3 for 2 vol. % MWCNTs/BiSbTe sample and 6.67g/cm^3 for pure BiSbTe. The thermal diffusivity was measured along the direction of thickness using laser flash method (NETZSCH Laser Flash Apparatus LFA 457, Germany) in the temperature range of 300 K to 500 K under Argon atmosphere.

III. Results & Discussion

The XRD pattern of pristine BiSbTe and 2 vol. % MWCNTs/BiSbTe bulk samples is shown in fig. 1. The XRD pattern is compared with the pdf reference card for BiSbTe (49-1713). The diffraction peaks match well with the standard peaks of BiSbTe. No peaks of MWCNTs were detected owing to its low content. Thus, the resulted XRD pattern shows that the materials are in single phase and any oxidation or phase change has been prevented. The Scanning Electron Microscope (SEM) image of MWCNTs/BiSbTe nanocomposite is shown in fig. 2.

The thermal conductivity (K) of both samples is calculated after measuring the thermal diffusivity (k), using the relation,

$$K = k\,\rho\,C_p \qquad (2)$$

Where ρ is the density of the given sample, and C_p is the specific heat. The values of C_p is evaluated by using the rule of

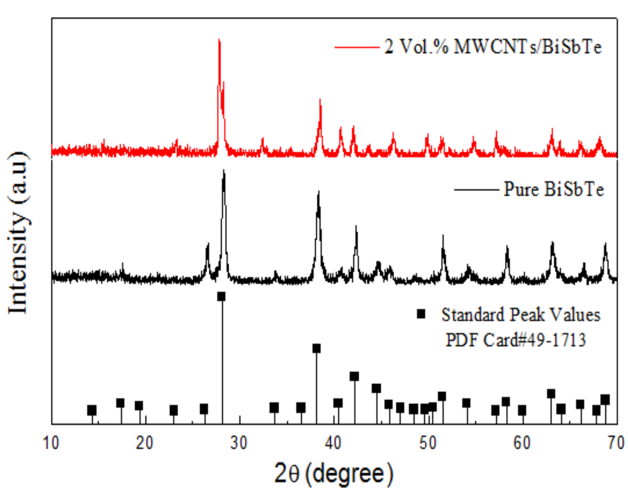

Fig. 1. XRD pattern comparison of MWCNTs/BiSbTe and BiSbTe bulk with standard peaks.

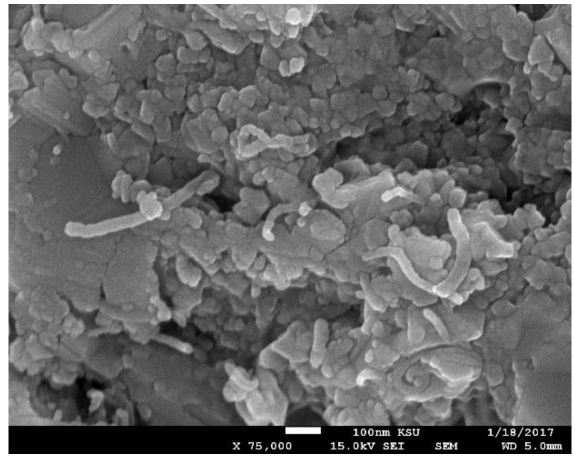

Fig. 2. SEM image of MWCNTs/BiSbTe composite.

mixture from the database [14]-[15]. Laser Flash method (LFA-457) is used to measure the temperature dependent thermal diffusivity, as shown in fig. 3. Although, there is reduction in diffusivity in 2 vol. % MWCNTs/BiSbTe sample as compared to pure BiSbTe, but it is less as compared to reduction in thermal conductivity of the same. The reason is attributed to low density of 2 vol. % MWCNTs/BiSbTe sample (2).

The addition of MWCNTs has reduced the thermal conductivity as compared to the pure BiSbTe sample, as shown in fig. 4. In general, the reduction in the lattice thermal conductivity is due to the adoption of nanostructures which is stemmed from phonon scattering on the boundaries of nanosized grains [9]-[10]. A similar phenomenon has emerged here for the reduction of thermal conductivity, mainly due to phonon scattering at the boundaries of nanotubes and BiSbTe nanostructure. This approach is useful in the development of thermoelectric materials as it does not have much effect on the electrical conductivity. Studies show that nanostructuring usually affect the lattice conductivity rather than the electronic transport properties, which is vital for high electrical conductivity and Seebeck coefficient and thus to improve the thermoelectric properties of the material, which is the later scope of this work [5]-[11].

Another reason for the low thermal conductivity in MWCNTs/BiSbTe sample is due to porosity. The relatively low bulk densities in 2 vol.% MWCNTs/BiSbTe sample may cause porosity [12]. As shown in fig. 4, thermal conductivity at room temperature (300 K) has been reduced from 0.95 W/mK in pure BiSbTe to 0.86 W/mK in 2 vol. % MWCNTs/BiSbTe sample, showing a significant reduction. The overall trend of reduction in thermal conductivity can be seen though out the temperature range from 300 K to 500 K. The minimum thermal conductivity of 0.758 W/mK has been achieved for 2 vol. % BiSbTe/MWCNTs sample at 474 K. Overall, such low thermal conductivity in 2 vol. % BiSbTe/MWCNTs bulk material indicates its potential for improved performance as a thermoelectric material (1).

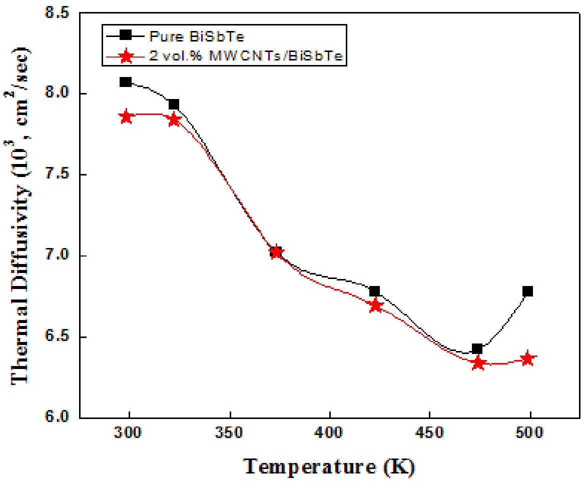

Fig. 3. Temperature dependence thermal diffusivity of pure BiSbTe & 2 vol.% MWCNT/BiSbTe.

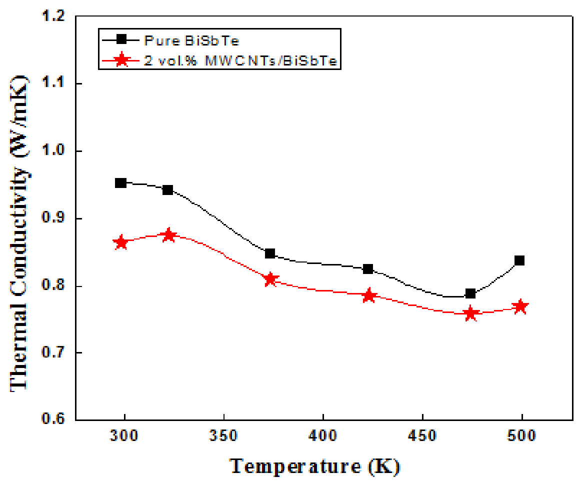

Fig. 4. Temperature dependence thermal conductivity of pure BiSbTe & 2 vol.% MWCNT/BiSbTe.

IV. Conclusion

The temperature dependence thermal conductivity of 2 vol. % MWCNTs/BiSbTe sample and pure BiSbTe has been studied and compared. Special attention is given to uniform dispersion of MWCNTs into pristine BiSbTe. Their consolidation is done using Spark Plasma Sintering (SPS). The maximum reduction in thermal conductivity (almost 10%) is found near room temperature by the addition of 2 vol. % MWCNTs (0.95 W/mK to 0.86 W/mK). This study is important to develop high performance thermoelectric material by using MWCNTs and SPS, as less work is done in past using these nanocomposite materials.

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