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Significant Influences of Selenium on the Electrical Properties of Bi_2Te_3 Compounds Synthesized Using Solid-State Microwave Irradiation

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Abstract. The thermoelectric materials based on p-type $Bi_2Se_{3x}Te_{3\ (1-x)}$ bulk products and dispersed with x compositions of Se (x=0.0, 0.2, 0.4, 0.6, 0.8, 1.0) were fabricated using standard solid-state microwave synthesis procedures. The products were characterized by X-ray diffraction (XRD). The XRD characterizations revealed that these products are pure Bi_2Te_3 and Bi_2Se_3 with uniform structures. The electrical properties of the Bi_2Te_3, Bi_2Se_3 and $Bi_2Se_{3x}Te_{3\ (1-x)}$ samples were measured in the temperature range of 303–523 K. The highest value of the Seebeck coefficient was 176.3 µV/ K for the $Bi_2Se_{0.6}Te_{2.4}$ sample, but only 149.5 and 87.4 µV/K for the Bi_2Te_3 and Bi_2Se_3 samples, respectively.

Introduction

The efficiency of thermoelectric (*TE*) materials is measured by whether it uses the highest *TE* figure of merit (*ZT*) which is possible at the temperature of operation, *T*. *ZT* can be expressed as $ZT=S^2\sigma T/k$, where *S* is the Seebeck coefficient, σ is the electrical conductivity, and *k* is the thermal conductivity. Accordingly, a good *TE* material has to combine both high electrical power factor ($PF = S^2\sigma$) and low thermal conductivity [1]. Compounds based on Bi_2Te_3 and Bi_2Se_3 are widely used as low temperature *TE* materials because their figure of merit at ambient temperature (300 K) is excellent. Their crystal structures are rhombohedral, consisting of atomic layers in the order of (Te, Se)1-Bi-(Te, Se)2-Bi-(Te, Se)1 along the c-axis [2]. Due to their narrow band gap, Bi_2Te_3 and Bi_2Se_3 have attracted wide attention owing to their high *TE* power [3]. The composition of inorganic materials has caught the interest of researchers since these can combine the merits of two materials [4]. Recently, new techniques have been developed in preparing Bi_2Te_3 and Bi_2Se_3, including solid-state microwave synthesis [5]. This method provides a faster, simpler, and more economically feasible method for preparing binary group chalcogenides, such as Bi_2Te_3 and Bi_2Se_3. The compounds prepared using solid-state microwave synthesis may possess improved physical properties compared with compounds synthesized with other techniques [5]. In this work, the successful growth of Bi_2Te_3, Bi_2Se_3, and $Bi_2Se_{3x}Te_{3\ (1-x)}$ bulks was achieved using solid-state microwave synthesis. The room-temperature *TE* and electrical properties of the stoichiometric $Bi_2Se_{3x}Te_{3\ (1-x)}$ (x=0.0-1.0) samples were also measured.

Experimental Details

The bismuth (Bi), tellurium (Te), and selenium (Se) used in this research were of high purity (99.999%). These elements were weighed according to the formula of $Bi_2Se_{3x}Te_{3(1-x)}$ and were mixed and ground for 20 minutes using an agate mortar and pestle. The materials were placed in a quartz ampoule under a vacuum of 10^{-5} mbar. The ampoule was inserted into an alumina crucible, which was located at the center of the microwave cavity. The sample was irradiated in an 800 W microwave oven (MS2147C) for 10 minutes, with periodic shaking. The Bi_2Te_3, Bi_2Se_3, and $Bi_2Se_{3x}Te_{3(1-x)}$ bulk products were then ground before being pressed into disk shapes with a diameter, 13 mm thickness, 0.5 mm using a cold-pressing process at 10 tons. The X-ray diffraction (XRD) patterns of the resulting powders were recorded by an X-ray diffractometer system (PANalytical X'Pert PRO MRD PW3040) using CuKα radiation; *TE* power (the Seebeck coefficient, *S*) and the electrical conductivity σ were measured at 300–523 K. The electrical conductivity measurements as a function of temperature were performed on all samples at constant current (20 mA) using the four probe method under 10^{-3}mbar. The Seebeck coefficient, *S*, was measured by connecting one side of the disk to a heater, while the other side was cooled by chilled water. The temperature of the hot block was raised slowly from room temperature, with regular intervals of 10 K. A thermocouple K- type E© Sun (ECS820C) was used to measure the working temperature.

Results and Discussion

The XRD spectra illustrated in Fig. 1 for the $Bi_2Se_{3x}Te_{3(1-x)}$ powders indicate that these substances are polycrystalline and characterized by a rhombohedral structure (space group $R\bar{3}m$) with a dominant peak representing the plane (0 1 5). They are also in good agreement with the JCPDS values of Bi_2Te_3 and Bi_2Se_3 (Card No.15-0863 and 33-0214), respectively. No remarkable diffractions of other phases can be found, indicating that pure Bi_2Te_3 and Bi_2Se_3 were obtained. In addition, the diffraction peak (0 1 5) shift to lower 2θ angle from 27.79º to 29.35º with increasing Se content (x), suggesting that the lattice parameter decreases with x value. The calculated lattice constant decreases from 4.396 Å for Bi_2Te_3 to 4.177 Å for Bi_2Se_3, which is due to the substitution of Se (~1.15 Å) by larger Te (~1.40 Å).

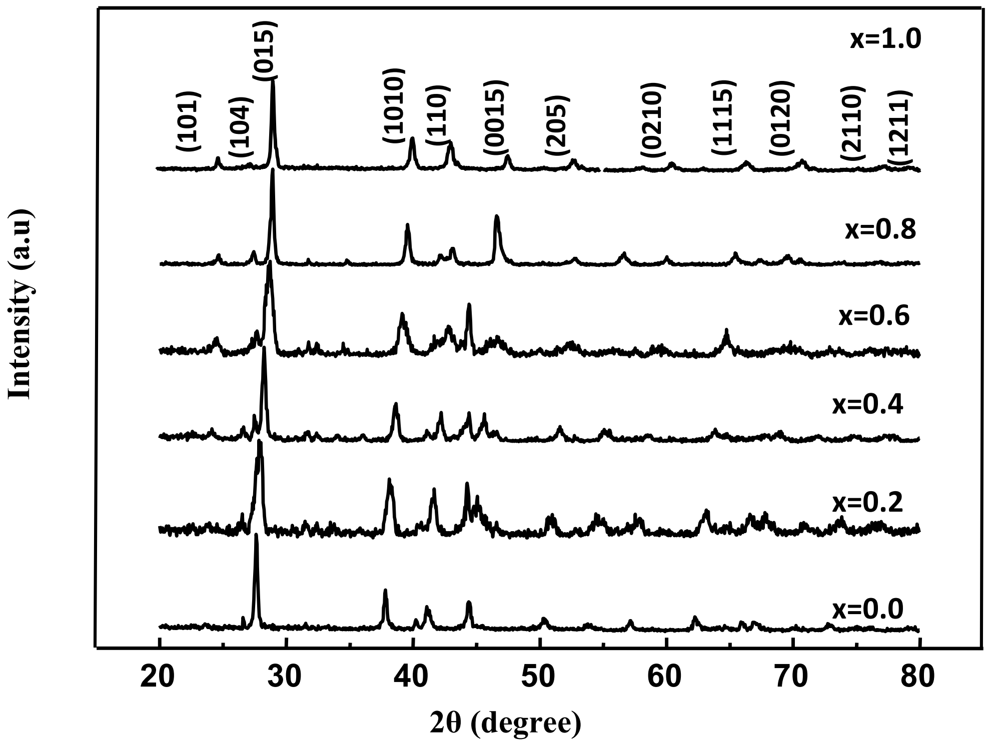

Fig.1. Powder XRD of $Bi_2Se_{3x}Te_{3\,(1-x)}$

Fig. 2 shows the Seebeck coefficient, *S*, as a function of the measured temperature for the $Bi_2Se_{3x}Te_{3\,(1-x)}$ disks. The highest value of the *S* is 176.3 μV/K for the sample with x=0.2. *S* of all the samples are positive indicating that the majority of charge carriers are holes (p-type) in the measuring temperature range. Conversely, it is worthwhile to note that the samples with high selenium content (x=0.4–1.0) exhibit relatively lower *S*. These results are in agreement with that reported by Wang et al. [6]. Fig. 3 shows the electrical conductivity, σ, as a function of temperature for the samples disks. σ of all the samples decrease with increasing temperature, showing a degenerate semiconductor conducting behavior suggesting that the carriers are in an extrinsic state in the whole temperature range. The σ value increases with Se content, its values increase from 1.92×10^5 to 4.04×10^5 S/m at 303 K and from 9.6×10^4 to 1.2×10^5 S/m at 523 K with x increasing from 0.0 to 1.0, respectively. Two competing factors of carrier concentration and mobility, determine the electrical conductivity. Thus, electrical conductivity can be expressed in terms of carrier concentration with the formula $\sigma = pe\mu$. The resultant conductivity that depends on Se content is well explained by the suggestion that the increase in the carrier concentration and hall mobility are due to the Se content. This indicating that σ at a fixed measuring temperature increases with increasing Se content, as mentioned in the previous study [6].

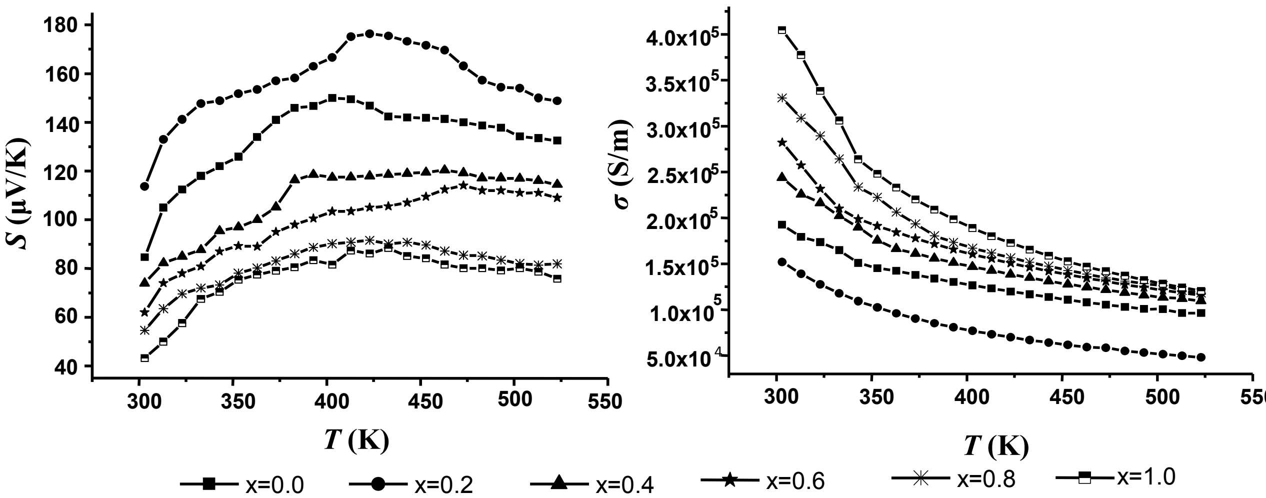

Fig.2. S of $Bi_2Se_{3x}Te_{3(1-x)}$ disks as a function of T Fig.3. σ of $Bi_2Se_{3x}Te_{3(1-x)}$ disks as a function of T

Conclusion

The p-type $Bi_2Se_{3x}Te_{3(1-x)}$ *TE* materials were successfully synthesized using solid-state microwave synthesis. This procedure was more efficient and faster and was a simplified method preparation. A maximum Seebeck coefficient of 176.3 μV/ K at 423 K is achieved for the $Bi_2Sb_{0.6}Te_{2.4}$ sample, which also showed an improved power factor. A maximum power factor value of 2.85 mW/ mK^2 at 403 K was measured for $Bi_2Se_{0.6}Te_{2.4}$ sample, whereas values of 2.37 and 1.34 W/mK^2 were obtained for Bi_2Te_3 and Bi_2Se_3 samples, respectively. This behavior was attributed to the compounds with lower selenium content exhibiting relatively better electrical properties. In addition, the measurements on the mixture Bi_2Te_3 and Bi_2Se_3, such as $Bi_2Se_{3x}Te_{3\,(1-x)}$ prepared using solid-state microwave synthesis showed greater burst in *TE* performance.

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