

ENHANCED THERMOELECTRIC PROPERTIES of n -Bi₂Se₃ AND p -Bi_{0.4}Sb_{1.6}Te₃ SYNTHESIZED BY USING HOT-PRESS METHOD

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Abstract

We present the thermoelectric characteristics of bulk samples of the n -type Bi₂Se₃ and p -type Bi_{0.4}Sb_{1.6}Te₃ that were produced by the vacuum melt technique and then vacuum hot-pressed at 723 and 523 K, respectively. The samples were investigated for their crystalline structure, lattice parameter, bulk density, relative density, and thermoelectric properties by XRD, density kit, ZEM-3, and LFA, respectively. In the same temperature range of 325 to 525 K, the thermoelectric characteristics of bulk n -type Bi₂Se₃ and p -type Bi_{0.4}Sb_{1.6}Te₃ were higher than the literature data. The Bi₂Se₃ and Bi_{0.4}Sb_{1.6}Te₃ bulk samples have maximum zT is 0.84 and 0.88 at 329 K, respectively, and are considered high-performance materials for use in thermoelectric devices.

Keywords: Bi₂Se₃, Bi_{0.4}Sb_{1.6}Te₃, HP method, TE properties, zT

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Introduction

Thermoelectric (TE) materials have been the topic of intensive research due to their unique capability of directly converting heat into electricity and electrical power into cooling or heating. These materials can reduce carbon emissions by converting waste heat into electricity (Yim and Rosi, 1972). Bismuth telluride (Bi₂Te₃) is a well-known bulk TE material for industrial TE cooling and generation applications at low temperatures. TE materials are formed of alloys, oxides, and rare-earth (Goldsmid, 2010). The dimensionless figure of merit is a measure that is connected to the performance of TE materials. The zT is equal to $S^2\sigma/(\kappa_e + \kappa_L)$, where S is the Seebeck coefficient, σ is electrical conductivity, κ_e is the thermal conductivity and κ_L is the lattice thermal conductivity and T is absolute temperature, (Kaibe *et al.*, 1989; Caillat *et al.*, 1992). As a result, the TE materials ought to have high S and σ the same time (Cao *et al.*, 2008). Because of their exceptional TE qualities in those ranges, Bi₂Te₃-based materials are the best TE materials operating at a close low temperature and they are frequently employed for numerous TE applications (Lu *et al.*, 2019). The materials exhibit rhombohedra structure, sometimes knows as rhombohedra, with space group number 166 and lattice parameters $a = b = 4.38$ and $c = 30.48$ Å (Feutelais *et al.*, 1993). However, there are currently very few materials based on n -type Bi₂Te₃. Bi₂Te_{3-x}Se_x (BTS) alloyed with Se is one example (Yan *et al.*, 2010; Hu *et al.*, 2015). The high zT of 0.96 at 370 K in tellurium-free bulk Bi₂Se₃ samples obtained by vacuum melting and regular vacuum HP is reported by Zhang *et al.* (2011). The spark plasma sintered (SPS) technique, Yuho Min *et al.* report achieving scalable synthesis of Bi₂Se₃ nanoflakes and improved TE characteristics of their nanocomposites. At 430 K, the high zT 0.2 is seen in the B Se₃ nanoflakes (Min *et al.*, 2013). Sun *et al.* (2015) use the melting process to report the TE performance of nanostructured topological insulator Bi₂Se₃. At 570 K, the approach for the maximum zT value of Bi₂Se₃ is 0.63 (Sun *et al.*, 2015). The impact of Te nanoparticles on TE performance in p -type BiSbTe is reported by T. Zhang *et al.* They discovered that some Te particles, not graphene, were a second phase in the composite, despite a slight improvement in TE performance. Despite a minor improvement in TE performance, they discovered that certain Te particles were the second stage of composites rather than graphene (Zhang *et al.*, 2013). Suh *et al.* (2015) report the TE performance of Bi_{0.5}Sb_{1.5}Te₃ composites.

At 350 K, the greatest value of zT is 1.35 was obtained.

In this study, we suggested a good synthesis for bulk samples of Bi₂Se₃ and Bi_{0.4}Sb_{1.6}Te₃ using the HP method. The samples were HP at 723 and 523 K under 60 MPa for 2 h. The crystal structure, lattice parameter, bulk density, and TE properties after HP are investigated and discussed for probabilities of TE ink.

Materials and Method

The following reagents were used exactly as supplied. Bismuth (Bi), Selenium (Se), and Tellurium (Te) powder (99.99%, Alfa Aesar) were prepared to have a composition of Bi₂Se₃ and Bi_{0.4}Sb_{1.6}Te₃. The planetary ball mill was used to mix the powders of Bi, Se, and Te for 30 h at 300 rpm in Ar atmosphere. The combined elements were then put into a quartz tube and heated to 1,073 K for 4 h with a heating rate of 50 °C h⁻¹ in Ar atmosphere. The furnace was then gradually brought down to room temperature. The ingots were reground to fine powder by ball milling for 2 h under Ar atmosphere (Lee *et al.*, 2021), and the sample powder was HP at 723 K for Bi₂Se₃ and 523 K for Bi_{0.4}Sb_{1.6}Te₃ under 60 MPa for 2 h step up 5°C min⁻¹ in vacuum (Lee *et al.*, 2014; Bohra *et al.*, 2017). The crystal structures (XRD6100, Shimadzu), patterns were collected using a CuK_{α1} X-ray source (wavelength of 1.5418 Å), operating at 40 kV and 40 mA in the range of 20°-70° with a scan speed of 5° min⁻¹ and step size of 0.01° to determine the crystal structures was evaluated by Scherrer's Equation 1 as follows:

$$D = \frac{K\lambda}{\beta \cos \theta} \quad 1$$

where K is a dimensionless shape factor (0.9), λ is the wavelength of the CuK_{α1} radiation, β is the full width at half maximum (FWHM) of diffraction peak, and θ is the Bragg's angle. The bulk and relative density of Bi₂Se₃ and Bi_{0.4}Sb_{1.6}Te₃ bulk samples were calculated from the Equation 2

$$\rho = \frac{nA}{V_c N_A} \quad 2$$

where n is number of atoms in unit cell, A is the atomic weight, and V_c is unit cell of volume, and N_A is Avogadro's number. The sintered specimens were cut into rectangular pieces (3×3×15 mm²) used

to measure the Seebeck coefficient (S) and electrical conductivity (σ) using a four-probe method (ULVAC- RIKO, ZEM-3) from 325 to 525 K. The thermal diffusivity D was measured by laser flash analysis (LFA Netzsch LFA-457, Germany). The thermal conductivity (κ) was calculated from the Equation 3

$$\kappa = DdC_p \quad 3$$

where the sample density d is determined by the Archimedes method and the specific heat C_p is measured using a differential scanning calorimeter and the three-line method (Radingoana *et al.*, 2023).

Result and Discussion

The XRD patterns of Bi_2Se_3 and $\text{Bi}_{0.4}\text{Sb}_{1.6}\text{Te}_3$ bulk samples, sintered under the vacuum stat at 723 K for Bi_2Se_3 and 523 K for $\text{Bi}_{0.4}\text{Sb}_{1.6}\text{Te}_3$, are shown in Figure 1(a, b). All peaks of XRD patterns showed the single phase that corresponds with the patterns of Bi_2Se_3 (JCPDS#33-0214) and rhombohedral structure (Belec *et al.*, 2019). Since there are no more components in any composition or power, Bi and Se are assumed to be fully reactive, resulting in the formation of the Bi_2Se_3 combination. All of the diffraction peak positions and ($h k l$) values in the XRD patterns of $\text{Bi}_{0.4}\text{Sb}_{1.6}\text{Te}_3$ bulk sample matched the diffraction data of the pure $\text{Bi}_{0.4}\text{Sb}_{1.6}\text{Te}_3$ (JCPDS#049-1713) quite well (Zhang *et al.*, 2022). The prominent peaks are (015), (1 0 10), and (0 0 15), which show that the grains are growing preferentially in these directions. The lattice

parameter, bulk density, and relative density of Bi_2Se_3 and $\text{Bi}_{0.4}\text{Sb}_{1.6}\text{Te}_3$ bulk samples are shown in Table 1.

The temperature dependence of the S , σ , PF , κ , and zT for HP sintered samples of Bi_2Se_3 investigated are presented in Figure 2. The negative value indicated that sample were n -type semiconductors. It can be assumed that the density obtained was higher, which high S and σ as shown in Table 1. The relative density is close to each other and can be excluded because it obtains more than 80% of the relative density of the sample. The S of the sintered sample was observed to increase with increasing temperature from $-145 \mu\text{V K}^{-1}$ at 329 K to $-193 \mu\text{V K}^{-1}$ at 520 K is higher than those obtained with typical Bi_2Te_3 -based bulk ingots as shown in Figure 2(a) (Liu *et al.*, 2017; Byun *et al.*, 2019). The temperature dependence of the σ showed a degenerate semiconductor behavior depending on temperature, as a significant decrease with increasing temperature can be attributed to the increased prevalence of phonon scattering as shown in Figure2 (b) (Sobota *et al.*, 2014). The calculated PF ($PF = S^2\sigma$) was obtained from the experimental value of S and σ . The PF value of the sample decreased with increasing temperature as shown in Figure 2(c). The highest PF value was $2.8 \times 10^{-4} \text{ W m}^{-1} \text{ K}^{-2}$. The Bi_2Se_3 bulk sample is increased with increasing temperature as compared with (Liu *et al.*, 2017; Byun *et al.*, 2019). The κ of bulk sample is $1.1 \text{ W m}^{-1} \text{ K}^{-1}$ at 329 K and it increases to $1.5 \text{ W m}^{-1} \text{ K}^{-1}$ with increasing temperature up to 520 K as shown in Figure 2(d), the maximum zT value was achieved of Bi_2Se_3 bulk marking 0.84 at 329 K, as shown in Figure 2(e).

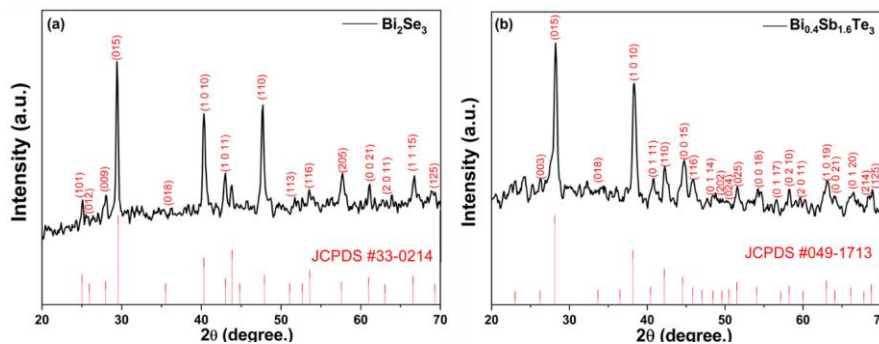


Figure 1. The XRD pattern of (a) Bi_2Se_3 and (b) $\text{Bi}_{0.4}\text{Sb}_{1.6}\text{Te}_3$ bulk samples

Table 1. Lattice parameter, bulk density, and relative density of Bi_2Se_3 and $\text{Bi}_{0.4}\text{Sb}_{1.6}\text{Te}_3$ bulk samples

Sample	Lattice parameter (nm)		Bulk density (g cm^{-3})	Relative density (%)
	a = b	c		
Bi_2Se_3	0.428	3.059	6.20	82.21
$\text{Bi}_{0.4}\text{Sb}_{1.6}\text{Te}_3$	0.448	3.187	6.07	82.92

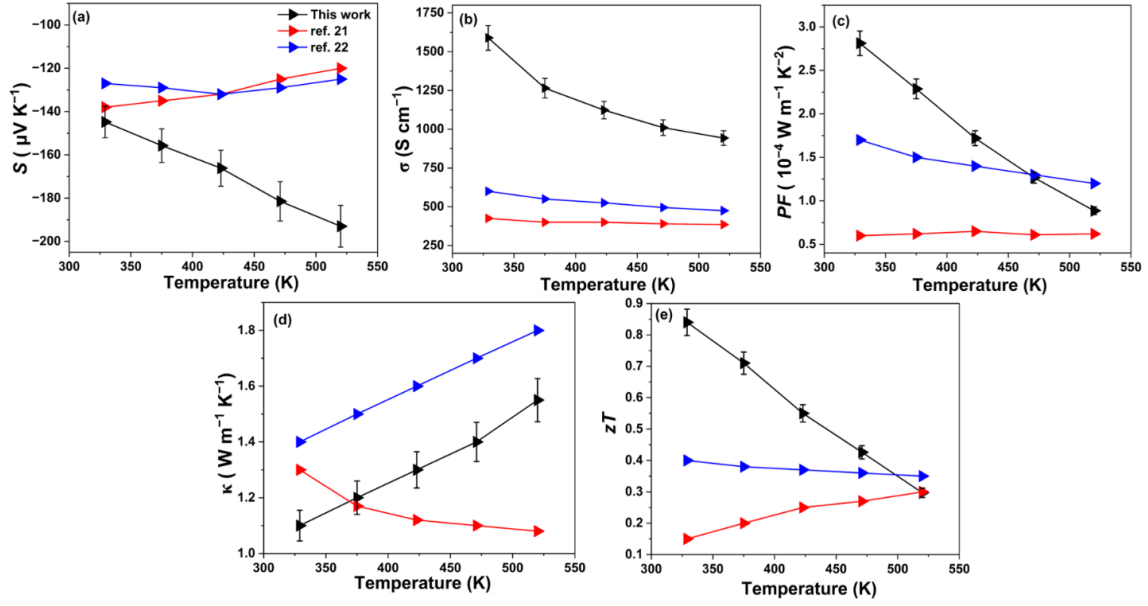


Figure 2. The temperature depends of the (a), S (b), σ (c), PF (d), κ and (e) zT for Bi₂Se₃ bulk sample

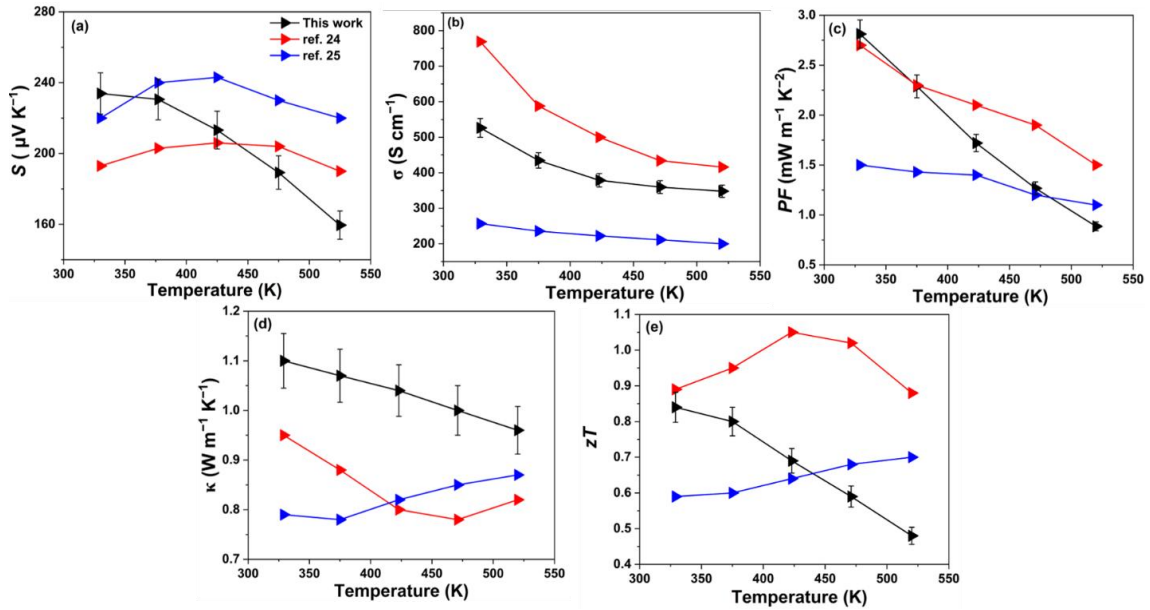


Figure 3. The temperature depends of (a), S (b), σ (c), PF (d), κ and (e) zT of Bi_{0.4}Sb_{1.6}Te₃ bulk sample

The temperature dependence of the S , σ , PF , κ , and zT for HP sintered of Bi_{0.4}Sb_{1.6}Te₃ bulk samples investigated are presented in Figure 3. The S value of Bi_{0.4}Sb_{1.6}Te₃ bulk samples from 234 $\mu\text{V K}^{-1}$ at 330 K to 160 $\mu\text{V K}^{-1}$ at 525 K as shown in Figure 3(a). However, the Bi_{0.4}Sb_{1.6}Te₃ samples show distinct temperature dependence in these samples, these values are higher than those of sample (Zhang *et al.*, 2017), although the Sb₂Te₃

bulk sample had S values that were comparable to those in (Fana *et al.*, 2015). The temperature dependence of the σ showed decreases with increasing temperature due to the scattering of electrons by the intensified lattice vibrations. These vibrations act as obstacles, hindering the smooth flow of electrons and reducing σ as shown in Figure 3(b) (Palenskis, 2022). The PF value of the sample decreased with increasing temperature at high

temperatures as shown in Figure 3(c). The highest PF value was at $2.8 \text{ mW m}^{-1} \text{ K}^{-2}$. The κ of $\text{Bi}_{0.4}\text{Sb}_{1.6}\text{Te}_3$ bulk sample is decreased with increasing temperature as compared with (Fana *et al.*, 2015; Zhang *et al.*, 2017). The lowest value of κ for this sample is $0.96 \text{ W m}^{-1} \text{ K}^{-1}$, which is obtained at 520 K. In Figure 3(d), the higher zT value of $\text{Bi}_{0.4}\text{Sb}_{1.6}\text{Te}_3$ bulk sample can be obtained within the entire temperature range because of the benefit of high PF and low κ ; the zT value is 0.88 at 329 K, as shown in Figure 3(e).

Conclusions

In this work, we investigated the bulk samples of Bi_2Se_3 and $\text{Bi}_{0.4}\text{Sb}_{1.6}\text{Te}_3$, which were synthesized by using the HP sintering method. The crystal structure lattice parameter, bulk density, relative density, and TE properties of Bi_2Se_3 and $\text{Bi}_{0.4}\text{Sb}_{1.6}\text{Te}_3$ were investigated. The maximum S values reached $-193 \mu\text{V K}^{-1}$ at 520 K of Bi_2Se_3 , while $\text{Bi}_{0.4}\text{Sb}_{1.6}\text{Te}_3$ bulk samples obtained and $234 \mu\text{V K}^{-1}$ at 330 K. The temperature dependence of the σ showed decreases with increasing temperature due to the scattering of electrons by the intensified lattice vibrations of the Bi_2Se_3 bulk sample. The highest PF value is $2.8 \times 10^{-4} \text{ W m}^{-1} \text{ K}^{-2}$ at 329 K for Bi_2Se_3 , while $\text{Bi}_{0.4}\text{Sb}_{1.6}\text{Te}_3$ bulk samples was around $2.8 \text{ mW m}^{-1} \text{ K}^{-2}$ along with a low value of κ in both samples. The maximum zT is 0.84 and 0.88 at 329 K of Bi_2Se_3 and $\text{Bi}_{0.4}\text{Sb}_{1.6}\text{Te}_3$ bulk samples, respectively. This indicates that Bi_2Se_3 and $\text{Bi}_{0.4}\text{Sb}_{1.6}\text{Te}_3$ bulk samples can have their TE characteristics improved using the very effective HP process.

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