Synergistic Optimization of Thermoelectric Performance in P-Type Bi0.48Sb1.52Te3/Graphene Composite Postprint

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Abstract

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Full Text

Preamble

Synergistic Optimization of Thermoelectric Performance in P-Type $Bi_{0\cdot48}Sb_{1\cdot52}Te_3$/Graphene Composite

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Abstract

We report the synergistic optimization of thermoelectric properties in p-type $Bi_{0·48}Sb_{1·52}Te_3$ through the addition of graphene. Highly dense $Bi_{0·48}Sb_{1·52}Te_3$ + graphene (x wt%, x = 0, 0.05, 0.1 and 0.15) composites were synthesized by zone melting followed by spark plasma sintering. Scanning electron microscopy clearly revealed graphene at grain edges in the composites. The added graphene improved the power factor to 4.8×10^{-3} $Wm^{-1}K^{-2}$ through modified carrier concentration and suppressed lattice thermal conductivity. Consequently, synergistic optimization of electrical and lattice properties by graphene addition led to a significant improvement in the figure of merit ZT (1.25 at 320 K).

Keywords: $Bi_{0·48}Sb_{1·52}Te_3$; graphene; thermoelectrical materials; microstructure; synergistic optimization

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1. Introduction

Thermoelectric (TE) materials, which enable direct conversion between thermal and electrical energy, have attracted increasing attention [?]. The energy conversion efficiency of TE materials depends on the dimensionless figure of merit, defined as:

$$ZT = \frac{S^2 \sigma T}{\kappa_{tot}}$$

where S is the Seebeck coefficient, σ is the electrical conductivity, κ_{tot} is the total thermal conductivity (including lattice thermal conductivity κ_{lat} and electronic thermal conductivity κ_{ele}), and T is the absolute temperature. These parameters are intercorrelated, making simultaneous optimization challenging.

Bismuth telluride (Bi_2Te_3) based alloys are the best TE materials near room temperature and are widely used for refrigeration and power generation [?]. Researchers have made continuous efforts to improve the TE performance of Bi_2Te_3-based alloys for broader applications [?].

Nanocomposite approaches have proven effective for synergistically tuning electrical and thermal properties by introducing second-phase nanoparticles into the matrix [?]. Various materials, including ZnAlO, WSe_2, and silver, have been employed as second-phase nanoparticles to enhance the TE performance of Bi_2Te_3-based alloys [?]. However, not all composites exhibit synergistic optimization; some show degraded electrical properties [?, ?, ?, ?]. Therefore, proper second-phase selection is crucial for Bi_2Te_3-based alloys.

Graphene possesses very high electrical conductivity and has been explored as an incorporated second phase in conventional thermoelectric materials such as PbTe and $CoSb_3$ to increase electrical conductivity [?]. Despite graphene's high intrinsic κ_{lat}, some composites exhibit lower κ_{lat} than the matrix, likely due to boundary scattering and significant atomic mass differences between the matrix and graphene.

For Bi_2Te_3-based materials, researchers have attempted to optimize TE performance using graphene [?]. A.H. Li et al. reported an enhanced ZT of ~0.45 at room temperature in graphene-doped n-type Bi_2Te_3 [?]. Similar results were reported by B.B. Liang et al. [?] and J.I. Kim et al. [?]. T. Zhang et al. attempted to synthesize p-type graphene-BiSbTe composites by zone melting [?], achieving only modest TE enhancement and observing Te particles rather than graphene as the second phase [?].

Recently, D. Suh et al. reported graphene-BiSbTe composites synthesized from nanoplates via solvothermal methods [?], achieving a promising ZT of 1.24 despite low relative densities of 93-95% that may compromise mechanical properties [?]. Since both Bi_2Te_3-based materials and graphene have layered structures, the microstructure of graphene and matrix significantly influences overall TE performance. In this work, we prepared BiSbTe alloys with added graphene using zone melting combined with spark plasma sintering (SPS). We clearly observed graphene in the composites, located at BiSbTe grain boundaries and edges, demonstrating synergistic effects that enhance both power factor and overall figure of merit ZT.

2.1. Synthesis of BiSbTe/Graphene Powders and Bulk Materials

Elements of bismuth (Bi, 99.999%), antimony (Sb, 99.999%), tellurium (Te, 99.999%), and graphene powders (XF NANO) were used as starting materials without further purification. Stoichiometric amounts of $Bi_{0\cdot48}Sb_{1\cdot52}Te_3$ + graphene (x wt%, x = 0, 0.05, 0.1, and 0.15) were weighed and sealed in quartz tubes under high vacuum ($<10^{-2}$ Pa) using an oxyacetylene flame [?, ?]. The tubes were heated at 1023 K for 2 hours in a rocking furnace to ensure compositional homogeneity. After natural cooling, crystals were grown in a zone melting furnace at 998 K with a growth speed of 25 mm/h. The ingots were ground into fine powders and consolidated by spark plasma sintering (SPS) into cylindrical bulk materials (10 mm diameter, ~10 mm thickness) at 693 K under 60 MPa for 10 min in vacuum.

2.2. Materials Characterizations

Phase structures were characterized by X-ray diffraction (XRD, Bruker AXS, Karlsruhe, Germany) using Cu Kα radiation (λ = 1.5406 Å). Densities were determined by the Archimedes principle, with all samples achieving relative densities above 99%, indicating compact structure after SPS. Microstructure was examined by field emission scanning electron microscopy (FESEM) with energy dispersive spectroscopy (EDS).

2.3. Thermoelectric Properties Measurements

Samples were cut into 3 × 2 × 10 mm^3 bars perpendicular to the pressure direction and $\Phi 6 \times 1.5$ mm^2 plates parallel to the pressure direction for property measurements. Electrical conductivity and Seebeck coefficients were measured simultaneously via the standard four-probe method using a ZEM-3 (ULVAC Technologies, Inc., Chigasaki, Japan) under low-pressure helium from room temperature to 550 K. Thermal conductivity was calculated from specific heat C_p, thermal diffusivity λ, and density using $\kappa_{tot} = C_p \lambda \rho$ [**?**]. Thermal diffusivity was measured by the laser flash method (NETZSCH, LFA-457, Selb, Germany). The Hall coefficient R_H was measured using a physical properties measurement system (Quantum Design, PPMS-9, San Diego, CA, USA) in magnetic fields from 0 to 5 T. Carrier concentration n and mobility μ were calculated via $n = 1/eR_H$ and $\mu = \sigma R_H$, where e is the electron charge.

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3. Results and Discussion

The XRD patterns of BiSbTe + graphene (x wt%, x = 0, 0.05, 0.1 and 0.15) bulk materials are shown in Figure 1a [Figure 1: see original paper]. All diffraction peaks index to the BiSbTe structure with space group R3-m (JCPDS Card No. 49-1713). No obvious graphene impurity peaks are observed, likely because the low graphene content is below the detection limit. To confirm graphene presence, Raman spectroscopy was performed (Figure 1b). Graphene shows three bands at 1354, 1586, and 2893 cm^{-1}, corresponding to the disordered (D) band, graphitic (G) band, and broad (2D) band, respectively [**?**]. The characteristic BiSbTe matrix Raman peaks appear in the 55-215 cm^{-1} range. For the BiSbTe + graphene (x = 0.05) composite, both BiSbTe and graphene characteristic peaks are detected, confirming graphene retention during synthesis.

FESEM was used to further investigate graphene distribution (Figure 2 [Figure 2: see original paper]). The layered BiSbTe structure is evident in all composites (Figure 2a-c). Flocculent material similar to graphene (Figure 2d) appears at grain boundaries (Figure 2b) and grain edges (Figure 2c) in the x = 0.05 sample. EDS results show area A in Figure 2c has higher carbon concentration than area B, confirming the flocculent material as graphene. Similar EDS results for other composites indicate homogeneous graphene distribution.

Temperature-dependent (a) electrical conductivity, (b) Seebeck coefficient, and (c) power factor (PF = $S^2\sigma$) for BiSbTe + graphene (x wt%, x = 0, 0.05, 0.1

and 0.15) are plotted in Figure 3 [Figure 3: see original paper]. Electrical conductivity decreases with temperature, showing typical metallic behavior (Figure 3a). The BiSbTe matrix exhibits conductivity of 1.0 × 10^5 S/m at room temperature, slightly lower than zone-melted samples [**?**, **?**, **?**, **?**] but higher than SPS-prepared samples [**?**]. With added graphene, conductivity slightly enhances for x = 0.05 (1.15 × 10^5 S/m at 300 K, 5 × 10^4 S/m at 550 K), then decreases with further graphene content.

Electrical conductivity depends on carrier concentration and mobility ($\sigma = ne\mu$). As shown in Table 1 , carrier concentration increases after graphene addition while mobility decreases gradually, possibly due to graphene's high specific surface area and potential aggregation. These competing effects cause the complex conductivity changes. Our samples' carrier concentrations (2.66-3.09) × 10^{19} cm^{-3} are slightly higher than previous reports (1.4-2.0) × 10^{19} cm^{-3} [**?**], but with greater mobility, yielding much higher conductivity—likely benefiting from the zone melting process before SPS.

All samples show positive Seebeck coefficients across the temperature range, indicating p-type conduction (Figure 3b). Seebeck coefficients initially increase, peak around 380 K, then decrease due to minority carrier thermal excitation across the band gap. Below 400 K, Seebeck coefficients first decrease then increase with graphene content, opposite to the conductivity trend. At high temperatures, Seebeck coefficients increase with graphene content. The variation can be explained by carrier concentration effects [**?**, **?**]:

$$S = \frac{8\pi^2 k_B^2}{3eh^2} m^* T \left(\frac{\pi}{3n}\right)^{2/3}$$

where k_B is the Boltzmann constant, m^* is the effective carrier mass, and h is Planck's constant. Table 1 shows slight effective mass increase after graphene introduction, consistent with previous studies [**?**].

Power factors decrease with temperature (Figure 3c). Added graphene slightly enhances composite power factors, indicating electrical property optimization. The x = 0.05 sample exhibits higher power factor values than the BiSbTe matrix across the entire temperature range, while x = 0.10 and 0.15 samples show higher values above 370 K. The maximum power factor is 4.8 × 10^{-3} $\text{Wm}^{-1}\text{K}^{-2}$ at 300 K for x = 0.05, substantially higher than ~3.2 × 10^{-3} $\text{Wm}^{-1}\text{K}^{-2}$ in previous work [**?**].

Figure 4 [Figure 4: see original paper] shows temperature-dependent total thermal conductivity κ_{tot} (a) and lattice thermal conductivity κ_{lat} (b). Total thermal conductivity decreases from 300 K to 340 K, reaches a minimum, then rises with temperature—possibly due to ambipolar diffusion. BiSbTe + graphene (x = 0.05, 0.1, and 0.15) composites have lower κ_{tot} than pristine BiSbTe, though it increases with graphene content. The minimum total conductivity decreases from 1.51 $\text{Wm}^{-1}\text{K}^{-1}$ (x = 0) to 1.14 $\text{Wm}^{-1}\text{K}^{-1}$ (x = 0.05). Lattice thermal conductivity is calculated using $\kappa_{lat} = \kappa_{tot} - \kappa_{ele}$, where electronic thermal conductivity κ_{ele} is obtained via the Wiedemann-Franz law ($\kappa_{ele} = L\sigma T$) with Lorenz number L derived from degenerate Fermi energy and scattering parameters for a single band model [**?**]. Lattice thermal conductivity trends match total conductivity, with all composites (x = 0.05, 0.1 and 0.15) showing lower κ_{lat} than the matrix. The minimum lattice conductivity is ~0.6 $\mathrm{Wm^{-1}K^{-1}}$ for x = 0.05. Reduced κ_{lat} in BiSbTe + graphene samples results from extensive effective phase boundary phonon scattering. Graphene's ultrahigh specific surface area creates more interfaces to scatter phonons and reduce lattice thermal conductivity.

Figure 5 [Figure 5: see original paper] presents temperature-dependent ZT values for all bulk samples. Graphene addition enhances composite ZT values through synergistic optimization. Despite much larger power factors than previous studies, higher thermal conductivity yields a peak ZT of 1.25 at 320 K in the BiSbTe composite with 0.05 wt% graphene—nearly identical to the previous study [**?**]. The larger power factors in our samples benefit power generation applications [**?**].

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4. Conclusions

P-type BiSbTe alloys with varying graphene mass ratios were prepared by zone melting combined with spark plasma sintering. Microstructure and TE properties were investigated. Graphene addition slightly improved electrical properties by increasing carrier concentration while simultaneously reducing lattice thermal conductivity through phonon scattering. Both effects significantly enhanced TE performance, achieving a peak ZT of 1.25 at 320 K. These results demonstrate that added graphene can synergistically improve TE performance in the BiSbTe system.

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Conflicts of Interest: The authors declare no conflict of interest.

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