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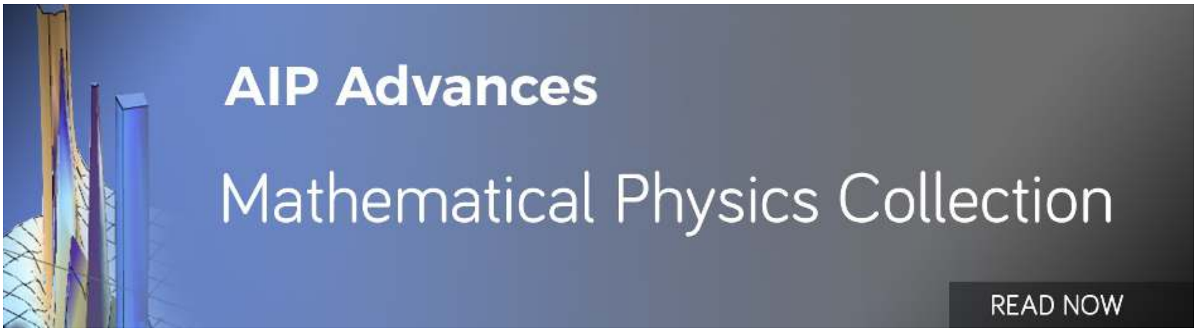

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Thickness dependence of electrical conductivity and thermo-electric power of $Bi_{2.0}Te_{2.7}Se_{0.3}/Bi_{0.4}Te_{3.0}Sb_{1.6}$ thermo-electric devices

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The electrical and thermo-electric (TE) properties of the bismuth telluride (BiTe) -based two-dimensional (2D) thermoelectric (TE) devices with different thin film thicknesses are analyzed systematically. The studied thin film thicknesses are covered from 100 nm to 400 nm. The accurate measured systems for the Seebeck coefficient (S) and electrical conductivity (σ) extractions are also built up in this work. When the thickness of the BiTe-based thin film in the TE device is scaled from 400 nm to 100 nm, the occurred optimized temperature (T) for the highest S value in these devices is found to be shifted from 60°C to 100°C. On the other hand, the best σ is observed in the thinner (100 nm) BiTe-based thin film devices under the higher T (130°C). Based on the understanding of S and σ values, the power factor and the figure of merit (ZT) – i.e., the ability of a TE material to efficiently produce electricity – are also investigated further. Compared with the commercial bulk BiTe TE device, we demonstrate that the ZT value can be improved ~50% with the thinner (100 nm) BiTe-based thin film devices in the higher T (>100°C) region. https://doi.org/10.1063/1.5017252

As we know, in addition to photovoltaic, piezoelectric, electrostatic, and electromagnetic devices, the thermo-electric (TE) device demonstrates one of the highest levels of potential for the application of energy harvesting.[1] Over the last decade, the exploration of high-performance TE materials has attracted much attention from both an academic research perspective and with a view to industrial applications. Bismuth telluride (BiTe) exhibits semiconducting behavior with a narrow band gap of 0.2 eV. Its promise as a usable material for TE applications has stimulated interest in its basic properties.[2] On the other hand, some research groups have also found that nanostructures can provide a chance to disconnect the linkage between thermal and electrical transport by introducing some new scattering mechanisms.[3] These could potentially further improve TE device efficiency. Recently, Dr. Shen et al.[4] demonstrated the micro-level (μm) thickness dependence of the electrical and TE properties of co-evaporated Sb_2Te_3 films. Carrier mobility and the overall TE properties of the Sb_2Te_3 films were affected significantly by changes in the film microstructure; this effect was attributed to the strong anisotropy of Sb_2Te_3 regarding electrical conductivity (σ).

In this work, the dependence of electrical and TE properties for BiTe-based two-dimensional (2D) TE devices with different thin film thicknesses in the nanometer (nm) scale level are analyzed systematically. Compared with the commercial bulk BiTe TE device, we demonstrate that the figure of merit (ZT) value – i.e., the ability of a TE material to efficiently produce electricity – can be improved ~50% with the thinner (100 nm) BiTe-based thin film devices in the higher T (>100°C) region.

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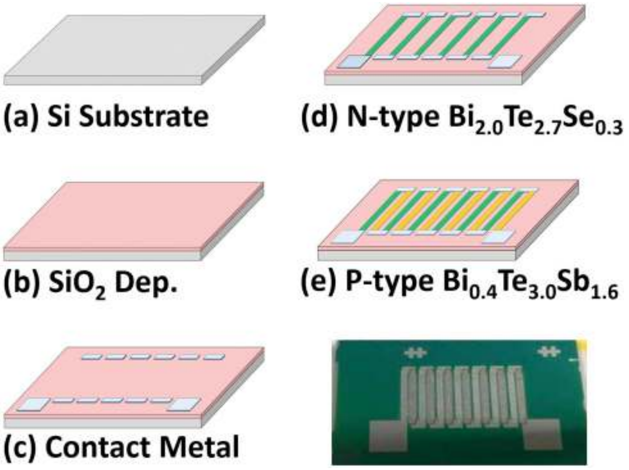
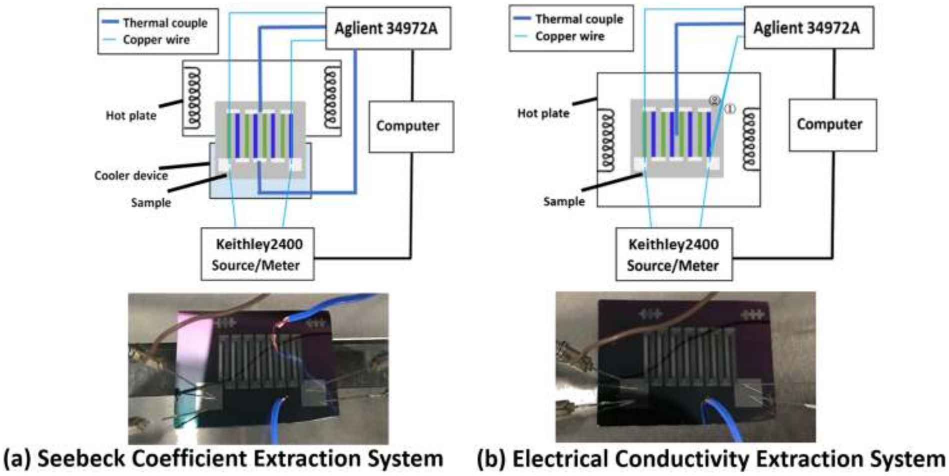


FIG. 1. The process step for the two dimensional (2D) thin film thermo-electric device developed in this work. The $Bi_{2.0}Te_{2.7}Se_{0.3}$ and $Bi_{0.4}Te_{3.0}Sb_{1.6}$ is used for the n-type and p-type materials, respectively.

Fig. 1 shows the step-by-step process flow and the real photo image on our 2D BiTe-based TE device. After the deposition of SiO_2 (~850 nm) for device isolation and Pt (~100 nm) for metal contact, thin $Bi_{2.0}Te_{2.7}Se_{0.3}$ and $Bi_{0.4}Te_{3.0}Sb_{1.6}$ films with different thicknesses from 100 nm to 400 nm are processed as the n-type and p-type materials in the sputter system, respectively. After loading the Si substrate into the sputter system, the deposition chamber was pumped to a base pressure of 4.9 x 10^{-6} torr followed by the introduction of Ar gas. The deposition pressure and the sputtering power were kept at 3.8 x 10^{-3} torr and 20 W, respectively. The detailed process parameters can be referred to Ref. 5. The deposition rate for the n-type $Bi_{2.0}Te_{2.7}Se_{0.3}$ and p-type $Bi_{0.4}Te_{3.0}Sb_{1.6}$ films is found to be ~1.6 A/sec and ~3.15 A/sec, respectively. A JEOL 6500 field-emission scanning electron microscope (FESEM), a JEOL JEM-2100F field-emission transmission electron microscope (FETEM), and X-ray diffractometry (XRD) were utilized for the material analyses.

Fig. 2 shows the measurement systems for the (a) Seebeck coefficient (S) and (b) electrical σ extraction on our 2D thin film TE devices. In order to extract the S value on our 2D TE devices, the equation (1) below is used.

$$\Delta V = IR + S\Delta T \tag{1}$$



(a) Seebeck Coefficient Extraction System (b) Electrical Conductivity Extraction System

FIG. 2. The measurement system for the (a) Seebeck coefficient and (b) electrical conductivity extraction on our two dimensional (2D) thin film thermo-electric device.

where ΔV is the voltage difference between two electrodes in the tested device; I is the constant current of ~0.01 mA input from the Keithley 2400; R is the internal resistance on our TE device; ΔT is the temperature difference on the TE device. The hot plate is used to create the ΔT across the device during measurement, and temperature information is recorded by the thermal couple for the feedback control. On the other hand, the equation (2)–(4) below are used for the extraction of σ.

$$\Delta V = \mathrm{I} \times (R_C + N \times R_{N/P}) \tag{2}$$

where R_C, $R_{N/P}$, and N is the contact resistance, N/P is junction resistance, and the number of N/P junctions in the TE device, respectively.

$$\rho = R_{N/P} \times \frac{A}{L} \tag{3}$$

$$\sigma = \frac{1}{\rho} \tag{4}$$

where ρ, A, and L are resistivity, the cross-section area of materials on the TE device, and the total length across the N/P material, respectively. It is necessary to note that the measured ρ in this work is the real resistivity across the N/P junction material in the TE device system level. Dr. Yamashita separately extracted the individual resistivity of N and P materials, as shown in Ref. 6.

Fig. 3 shows the measured characteristics of (a) S and (b) σ with different operated T and material thicknesses in our 2D thin film TE device. When the thickness of BiTe-based thin films in the TE device is scaled down from 400 nm to 100 nm, the occurred optimized T for the highest S value in these devices is found to be shifted from 60°C to 100°C. This indicates that the thinner film for the TE devices is more suitable for the application of higher T energy harvesting. On the other hand, the best σ is observed in the thinner (100 nm) BiTe-based thin film devices under the higher T (130°C). Our experimental data on the dependency of σ with different BiTe thicknesses and operated T agree well with the theoretical prediction by the Tellier's model.[7,8] It shows that the scaled thickness of BiTe-based thin film in the TE device can indeed lead to the higher σ, which can further contribute to a better power factor (shown in the Fig. 4(a)) and ZT value (shown in the Fig. 4(b)) for efficiency enhancement in the TE device.

In order to further investigate the influence of material thickness and T on the performance of TE devices, the power factor and ZT value are calculated using the equation (5)–(6) as below:

$$\text{power factor} = \mathrm{S}^2 \times \sigma \tag{5}$$

$$\mathrm{ZT} = \mathrm{S}^2 \times \sigma \times \mathrm{T/k} \tag{6}$$

where k is the material thermal conductivity. The value of k used in the calculation is set as 1.5 W/mK.[6] Fig. 4 summarizes the characteristics of (a) power factor and (b) ZT values with different

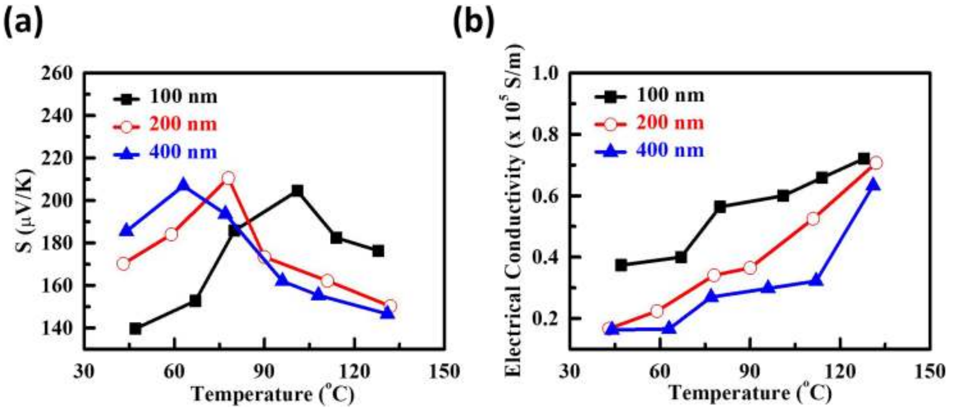


FIG. 3. The characteristics of (a) Seebeck coefficient and (b) electrical conductivity with different operated temperatures and material thicknesses in our two dimensional (2D) thin film thermo-electric device.

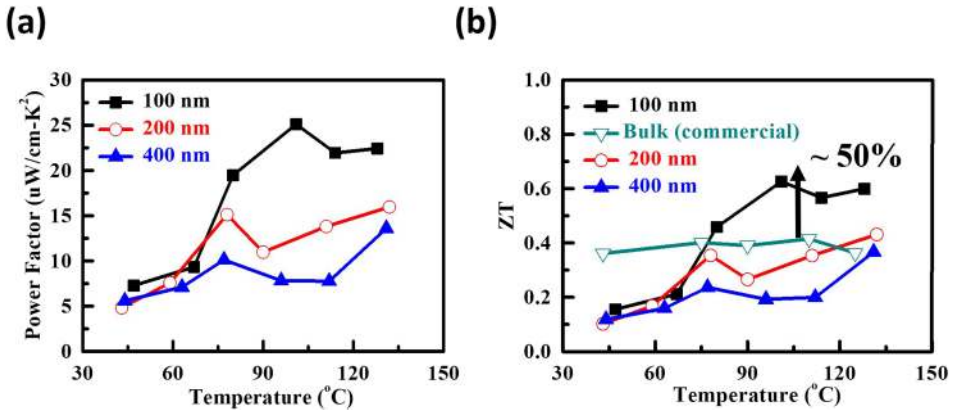


FIG. 4. The characteristics of (a) Power factor and (b) ZT values with different operated temperatures and material thicknesses in our two dimensional (2D) thin film thermo-electric device. The higher ZT value indicates the better efficiency in the developed thermo-electric device.

operated T and material thicknesses in our 2D thin film TE device, together with the bulk commercial device, for reference and comparison. The higher ZT value indicates better efficiency in the developed TE device. We demonstrate that the ZT value in this work can be improved ~50% with the thinner (100 nm) BiTe-based thin film in the higher T (>100°C) region compared to the commercial bulk BiTe-based TE device.

In this work, the dependency of nanometer-level scale BiTe thickness (from 400 nm to 100 nm) and different operated T (from 40°C to 130°C) for TE device efficiency improvement is investigated. The electrical and TE properties, including S, σ, power factor, and ZT value on our developed TE devices, are systemically measured and analyzed. Due to the higher σ value in the thinner (~100 nm) BiTe thickness in the TE device and the theoretical prediction by the Tellier's model, we demonstrate that the ZT value can be improved ~50% with the thinner (100 nm) BiTe-based thin film under the higher operated T (>100°C) region in this work compared to the commercial bulk BiTe-based TE device. The promising and competitive TE device potential demonstrated in this work opens the window for its future application in energy harvesting.

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