Research Article

Influence of Deposition Rate on the Thermoelectric Properties of Sb_2Te_3 Thin Films by Thermal Evaporation Method

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Thermoelectric (TE) materials are crucial because they can be used in power generation and cooling devices. Sb_2Te_3 -based compounds are the most favorable TE materials because of their excellent figure of merit at room temperature. In this study, Sb_2Te_3 thin films were prepared on SiO_2/Si substrates through thermal evaporation. The influence of the evaporation current on the microstructures and TE properties of Sb_2Te_3 thin films were investigated. The crystalline structures and morphologies of the thin films were analyzed using X-ray diffraction and field emission scanning electron microscopy. The Seebeck coefficient, electrical conductivity, and power factor (PF) were measured at room temperature. The experimental results showed that the Seebeck coefficient increased and conductivity decreased with increasing evaporation current. The Seebeck coefficient reached a maximum of $387.58 \mu\text{V/K}$ at an evaporation current of 80 A. Conversely, a PF of $3.57 \mu\text{W/cmK}^2$ was obtained at room temperature with evaporation current of 60 A.

1. Introduction

As energy shortage and carbon reduction are becoming more crucial challenges, green technology is attracting increasing attention. Thermoelectric (TE) materials are advantageous because of their environmentally friendly and renewable nature. TE characteristics facilitate the conversion of thermal energy and electrical energy, which can directly convert thermal energy to electrical energy through the Seebeck effect and electrical energy to thermal energy through the Peltier effect without entailing mechanical moving parts. TE materials have several high-profile applications in such fields as space flight, military, and medicine [1]. The performance of TE materials depends on their Seebeck coefficient, electrical conductivity, and thermal conductivity. The energy conversion efficiency of TE materials is evaluated using the figure of merit ZT: $ZT = (S^2\sigma/\kappa)T$, where S is the Seebeck coefficient (V/K), σ is the electrical conductivity, T is the absolute temperature, κ is the thermal conductivity [1, 2], and $S^2\sigma$ is the power factor (PF). Therefore, an excellent ZT can be obtained by increasing the Seebeck coefficient and electrical conductivity and reducing thermal conductivity. Bismuth telluride- (Bi-Te-) and antimony telluride- (Sb-Te-) based compounds are

currently the state-of-the-art TE materials for operations at near room temperature. Furthermore, Bi-Te- and Sb-Te-based TE materials exhibit the highest ZT and are widely used in commercial TE generators and coolers [3]. Generally, commercial TE devices are manufactured from sintered bulks of these materials. However, bulk materials possess low ZT. To increase ZT, low-dimensional materials are used to improve the TE properties because they increase the density of Fermi-level states and enhance phonon scattering [4] on nanostructured materials. In addition, miniaturizing TE devices using a bulk sample is difficult. Therefore, to miniaturize the device and effectively improve TE properties, several techniques have been reported for growing thin films, such as flash evaporation [5], ion-beam sputtering [6, 7], pulse laser deposition [8, 9], sputtering [10–12], electrochemical deposition [13–15], metal-organic chemical vapor deposition [16, 17], and molecular beam epitaxy [18–20]. However, some processes require long duration and expensive facilities. Therefore, in this study, Sb_2Te_3 -based thin films were fabricated through thermal evaporation, which offers such advantages as low fabricating expenses and short processing time. In addition, we evaluated the effects of the thermal evaporation rate on the TE properties of evaporated thin films on a SiO_2/Si substrate.

2. Experimental

P-type (100) silicon wafers served as the TE thin film substrates. After the RCA cleaning process, a 400-nm SiO_2 layer was thermally grown on a SiO_2/Si substrate through atmospheric pressure chemical vapor deposition. The p-type Sb_2Te_3 thin films were then deposited through thermal evaporation. High-purity (99.99%) Sb_2Te_3 powder with a diameter of 1–10 mm was used as the evaporation source and evaporated from a tungsten boat. The weight of Sb_2Te_3 powders was fixed for each evaporation process. To evaluate the effects of the thermal evaporation rate on the TE properties of thin films, the thermal evaporation current was varied in the range 50–80 A. The thin films were prepared in an evacuated chamber with a working pressure of less than 3.75×10^{-5} Torr at room temperature. The thickness of the evaporated Sb_2Te_3 films was measured from the SEM cross-sectional photographs.

The crystalline phases of TE thin films were examined using X-ray diffraction (XRD; Cu-K α , Bruker D8). The Sb_2Te_3 thin films were revealed by scanning 2θ from 20° to 60° at the speed of 1 second for each step with an angular gap of 0.05 degree. The surface morphologies of the Sb_2Te_3 thin films were analyzed using field emission scanning electron microscopy (FE-SEM, JEOL JSM6700). To evaluate the TE properties, Seebeck coefficient (S) and electrical conductivity (σ) were measured at room temperature. The Seebeck coefficient was obtained by measuring the resulting Seebeck voltage as applying a temperature gradient across the sample, in which the data was acquired by a Keithley 2700 system. As measured temperature was changed in a small range, for example, approximately 5°C , the Seebeck coefficient of the device under test (DUT) could be considered as a fixed value. The Seebeck coefficient obtained in this study is averaged from the values of ten measurements. The electrical conductivity of the specimen was measured using a four-point probe method at room temperature with a Keithly 2400 source meter. Subsequently, PF ($S^2\sigma$) was calculated using S and σ .

3. Results and Discussion

The thickness of the evaporated Sb_2Te_3 films under various evaporation currents was measured to be about 400 nm from the SEM cross-sectional photographs. The results also show that the deposition rate of thin films is approximately linearly increased with the increased evaporation current, as shown in Figure 1.

The X-ray diffraction patterns of the Sb_2Te_3 alloy powder are presented in Figure 2. The peaks agree with the powder diffraction pattern for Sb_2Te_3 material (JCPDS #71-0393). All diffraction peaks of the Sb_2Te_3 powder coincided with those of JCPDS data, indicating the formation of a single phase Sb_2Te_3 . For the Sb_2Te_3 powder, the diffraction angles at 28.24° , 38.29° , and 42.35° corresponded to the (0 1 5), (1 0 10), and (1 1 0) crystalline phases, respectively.

Figure 3 shows the XRD patterns of the Sb_2Te_3 thin films deposited at various evaporation currents. The results show that the diffraction peaks corresponding to the (0 1 5), (1 0 10), and (1 1 0) crystalline phases, respectively.

FIGURE 1: The relationship between deposition rate and evaporation current for the deposition of Sb_2Te_3 thin films.

FIGURE 2: XRD patterns of the Sb_2Te_3 powder.

FIGURE 3: XRD patterns of Sb_2Te_3 thin films deposited on SiO_2/Si substrates at various evaporation currents: (a) JCPDS, (b) 50 A, (c) 60 A, (d) 70 A, and (e) 80 A.

FIGURE 4: FE-SEM top-viewed photographs of the Sb_2Te_3 thin films under various evaporation currents: (a) 50 A, (b) 60 A, (c) 70 A, and (d) 80 A.

and (110) crystalline phases were not obvious. Because these thin films were deposited on an unheated substrate, the atoms did not have sufficient energy to facilitate crystal growth at room temperature. Figure 4 presents the SEM images of the thin films deposited at various evaporation currents at room temperature. The as-deposited thin films had a continuous and smooth surface with a fine grain size and uniformly distributed structure.

The effect of the evaporation current on the Seebeck coefficient is illustrated in Figure 5. The Seebeck coefficient of the as-deposited thin films was positive, confirming that Sb_2Te_3 thin films are p-type semiconductors. The Seebeck coefficient increased with increasing evaporation current. The maximum value of $387.58 \mu\text{V/K}$ was obtained at 80 A. On the other hand, the effect of the evaporation current on electrical conductivity is shown in Figure 6. Electrical conductivity decreased with increasing evaporation current. The sample deposited at 50 A existed as maximum electrical conductivity of $592.70 \text{ S}\cdot\text{cm}^{-1}$. The trend of conductivity may be due to the fact that as the evaporation current increases, the structure of thin film becomes less dense, which results in increased resistivity and decreased conductivity. The trend of Seebeck coefficient is inverse with that of conductivity [21].

Power factor, PF, is a crucial TE parameter and can be calculated from the Seebeck coefficient and electrical conductivity. Figure 7 presents the PFs obtained for the Sb_2Te_3 films deposited at various evaporation currents. The highest

FIGURE 5: Variations of Seebeck coefficient for Sb_2Te_3 thin films deposited at different evaporation current.

PF of $3.57 \mu\text{W}/\text{cmK}^2$ was obtained at an evaporation current of 60 A. The TE properties of the Sb_2Te_3 thin films deposited at various evaporation currents are summarized in Table 1. The maximum Seebeck coefficient and PF of p-type Sb_2Te_3 thin films were approximately $387.58 \mu\text{V/K}$ and $3.59 \mu\text{W}/\text{cmK}^2$, respectively.

TABLE 1: Thermoelectric properties of Sb_2Te_3 films deposited at different evaporation current.

Evaporation current (A)	50	60	70	80
Seebeck coefficient ($\mu\text{V/K}$)	28	129.3	242.4	387.58
Electrical conductivity ($\text{S}\cdot\text{cm}^{-1}$)	592.7	214.75	0.11	0.11
Power factor ($\mu\text{W}/\text{cmK}^2$)	0.46	3.57	0.017	0.028

FIGURE 6: Variations of electrical conductivity for Sb_2Te_3 thin films deposited at different evaporation current.

FIGURE 7: Variations of power factor for Sb_2Te_3 thin films deposited at different evaporation current.

4. Conclusion

In this study, the thermal evaporation method was successfully utilized for the preparation of the Sb_2Te_3 thermoelectric thin films on SiO_2/Si substrates with low cost. The effects of evaporation current on the thermoelectric properties and microstructures of the Sb_2Te_3 thin films have been investigated at the evaporation current of 50–80 A. The electrical conductivity decreased from 592.7 S/cm to 0.11 S/cm, whereas the Seebeck coefficient increased from 28 $\mu\text{V/K}$ to 387.58 $\mu\text{V/K}$ as the evaporation current varied from 50 A

to 80 A. The optimized power factor of 3.57 $\mu\text{W}/\text{cm}\cdot\text{K}^2$ was obtained for Sb_2Te_3 thin film deposited at evaporation current of 60 A.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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