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Full paper

Light-concentrated solar generator and sensor based on flexible thin-film thermoelectric device

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ABSTRACT

In this study, we prepared a multi-functional thin-film thermoelectric device for small-scale energy harvesting and self-powered light sensing. A comprehensive optimization was conducted in terms of thermoelectric material, device, and system integration. The thermoelectric thin films in the device were prepared using the sputtering method together with a post-annealing process. A Fresnel lens was incorporated into the thin-film thermoelectric device for energy concentration. In addition, a simulation procedure was used to facilitate the optimization of the heat sink, which can provide fast heat dissipation and mechanical support for the thin-film thermoelectric device. The electricity generation and light sensing performance was then characterized under solar irradiation, followed by a feasibility study for sensor application. The results indicate that our thin-film thermoelectric device exhibits high voltage output and greatly enhanced responsivity. This work presents a significant progress toward an integrated design of an applicable and multifunctional thin-film thermoelectric sensor for detecting sunlight intensity.

1. Introduction

Recently, extensive research interests have been attracted to develop nanodevices with multifunctionality by harvesting energy to build self-powered sensors. For example, the triboelectric nanogenerator investigated by Wang's group can be applied to harvest mechanical energies and alternatively used as a sensor for actively detecting the processes arising from mechanical agitation [1]. Meanwhile, thermoelectric (TE) devices based on the Seebeck effect have been employed for various applications, including power generation, temperature measurement, and infrared radiation detection [2,3]. The increasing demand of small-scale energy harvesting for power supply or energy sensing provides an opportunity for the investigation of the multi-functional thin-film TE devices, which have the advantages of flexibility, small volume, light weight, high integration, and enhanced compatibility. TE devices are commonly coupled with spectrally selective coatings to harness solar energy and then convert it to electricity [4–8]. The feature of large length to cross-sectional area ratio for thin-film TE legs facilitates a high output voltage and the possibility of a thin-film TE device reasonably serving as detectors or sensors. Thin-film thermopile infrared detectors have become commercially available [9]. The potential use of TE elements based sensors for gas [10] and intravascular flow [11] analysis, or temperature [12] and heat flux [13] measurement are explored. We also reported that our thin-film TE device exhibited a potential light sensing feature, but the responsivity was still low without a specifically optimized design for its sensing application [14].

For thin-film TE devices, low conversion efficiency and fabrication complexity have long been identified as the main obstacles to their development, and efforts should be made in different aspects, including material, device, and system integration. In our previous report, enhanced TE films with hierarchical nanostructures were prepared by increasing the deposition temperature and time [14]. Meanwhile, the research results indicated that a polymer substrate was the best choice in in-plane TE devices to build a large temperature difference due to its intrinsically low thermal conductivity and thin-film feature with flexibility [15]. However, as the limited temperature tolerable of the polymer substrate would suppress a thorough crystallization and growth of thin films, the properties of TE materials still need to be further improved. Recently, significant efforts have been made to enhance the ZT value by introducing nanostructures into TE thin films [16,17], such as superlattice structure [18], high-orientation crystal plane [19,20], and nano arrays [21,22]. Duan et al. [23] also found post-annealing plays a key role in the micro-structures and transport properties of $Bi_2Te_{2.7}Se_{0.3}$ thin films, which would be an efficient way to tune the crystallinity of a nanostructure.

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In solar TE devices, incoming thermal flux rather than the temperature difference is normally fixed. In this case, the temperature distribution varies with several factors including device configuration and external environment. Hence, energy concentration strategies have been combined with bulk TE devices to further enhance the temperature difference. In reports by Hasan et al. [24], a Fresnel lens and bulk TE module were utilized to concentrate the solar beam and generate electrical power. In addition, a high-performance solar bulk TE generator with optical and thermal concentration has been reported by Kraemer et al. with a peak efficiency of 7.4% [25]. There have been few reports, however, on the combination of energy concentration strategy and thin-film TE device owing to the assembly difficulty. The thermal concentrator is typically attached to the TE module, which is not feasible for flexible thin-film TE devices because of the limited contact area and poor mechanical support. Although the separated focusing of solar light was attempted in the literature [26], the fabricated thin-film TE modules still exhibited low power generation.

Solar illumination intensity is mainly measured by an optical power meter by converting light into a corresponding photocurrent. To date, limited research has been reported on solar light detection using TE devices which are commonly used for power generation. In view of these points, a multifunctional thin-film solar TE generator and sensor with ingenious and high integration is studied in this paper. Emphasis is placed on comprehensive optimization in terms of TE material, device and system integration. Post-annealing is adopted to further improve the performance of the TE thin films because the deposition temperature is limited by the substrate. In addition, we also together consider the introduction of an energy concentration strategy with the thin-film TE device and the optimization of a thermal design for the heat sink. The electrical generation and light sensing performance was characterized under solar irradiation, followed by a feasibility study for our thin-film TE sensor in the application of solar illumination detection.

2. Fabrication of thin-film TE device

2.1. Film deposition and annealing

In this work, p-type $Bi_{0.5}Sb_{1.5}Te_3$ and n-type $Bi_{0.5}Te_{2.7}Se_{0.3}$ were deposited by a magnetron sputtering system. The targets were powered by a direct current power supply of 30 W, and the working pressure of the sputtering gas Ar was set at 0.5 Pa. We chose polyimide (PI) with thickness of 0.2 mm and thermal conductivity of 0.35 W/mK as the substrate, which owns excellent characteristics of heat resistance. Because the glass transition temperature of the PI substrate is approximately 390 °C, the deposition temperature for TE films was set at 350 °C for 2 h. Based on the previous report [19], when the film was annealed at approximately 300–350 °C, a synchronous increase of electrical conductivity and Seebeck coefficient was achieved owing to the greatly enhanced carrier mobility and optimized carrier concentration. The post-annealing process was then adopted for the as-deposited TE films with the annealing temperature of 300 °C in N_2 atmosphere for 30 min.

The phase structure was investigated by XRD, as shown in Fig. 1a and b. The three strong peaks of $Bi_{0.5}Sb_{1.5}Te_3$ are (015), (110) and (205), and the peaks of (00 *l*) are very weak in the XRD pattern, indicating a slightly preferential growth along the (015) plane compared with its standard pattern (JCPDS 49–1713). The same situation can be observed for $Bi_2Te_{2.7}Se_{0.3}$ films in Fig. 1b. The intensity of the (015) peak is much higher compared with the standard pattern after annealing, and highly preferential growth along the (015) plane has been achieved. Generally, the intense and sharp peaks on XRD patterns are typical signatures for a high degree of crystallinity.

Fig. 1c and d illustrate the morphology for p-type $Bi_{0.5}Sb_{1.5}Te_3$ film and n-type $Bi_{0.5}Te_{2.7}Se_{0.3}$ film annealed at 300 °C, respectively. The films grow with a thickness of approximate 2 μm and have a relatively uniform and compact structure. The annealing process is beneficial for the diffusion of the atoms in the film, so the grains of $Bi_{0.5}Sb_{1.5}Te_3$ film begin to grow up and "sinter" together (shown in Fig. 1c). Meanwhile, some small precipitates separate from the surface, a phenomenon which was also observed in our previous report [19]. For the n-type $Bi_{0.5}Te_{2.7}Se_{0.3}$ film, the surface morphology (Fig. 1d) reveals a flat film with hexagonal flakes. Many columns can be observed in the cross-sectional structure, which are stacked by many secondary layered nanostructures, resulting in multiscale dimensions.

2.2. Thermoelectric properties

The properties of post-annealing TE films were investigated by using a ZEM3 system, as illustrated in Fig. 2. The electrical conductivity of $Bi_{0.5}Sb_{1.5}Te_3$ film reaches 2.2×10^4 S/m at room temperature and undergoes a small decline to 1.8×10^4 S/m as the temperature increases to 150 °C. Meanwhile, the maximum Seebeck coefficient reaches 220 μV/K at 110 °C and approximately 187 μV/K at room temperature. Regarding the $Bi_{0.5}Te_{2.7}Se_{0.3}$ film, the electrical conductivity changes from 1×10^4 S/m to 1.2×10^4 S/m in the temperature range from room temperature to 150 °C, which is relatively stable. The absolute value of the Seebeck coefficient is approximately 200 μV/K at room temperature and drops to 170 μV/K at 150 °C. Overall, as observed in Fig. 2c, the power factor of $Bi_{0.5}Sb_{1.5}Te_3$ film is nearly twice the value of $Bi_{0.5}Te_{2.7}Se_{0.3}$ film; nevertheless, the power factors for both films exhibit small fluctuations in the test temperature range.

We also summarized the Seebeck coefficient variation at the different test temperatures in Fig. 2d. The average absolute Seebeck coefficients for p-type $Bi_{0.5}Sb_{1.5}Te_3$ and n-type $Bi_{0.5}Te_{2.7}Se_{0.3}$ are calculated to be 200.9 μV/K and 190.6 μV/K, respectively, by integral calculation, which are quite close. In addition, the variation of Seebeck coefficient can be controlled within a small range. Those results make our films especially suitable for the application of a TE generator and TE sensor in a wide-temperature range owing to the relatively stable properties.

2.3. Thin-film TE device

Regarding the thin-film TE device, we have reported a simulation procedure to facilitate the optimization of the thin-film TE device from all aspects including the choice of substrate, the effect of air convection, the dimension of the TE leg, and its configuration [15]. According to the optimized design, the thin-film TE device is prepared as an in-plane device using PI substrate with 12-pair thermoelements in series connection. In the fabrication process, stainless steel masks with designed patterns were used to fabricate devices connected electrically in series. The size and structure of the device can be found in our earlier publication [14]. Initially, n-type (Bi2Te2.7Se0.3) and p-type (Bi0.5Sb1.5Te3) TE films were deposited on the substrate using the aforementioned deposition condition, and then the films were annealed at the annealing temperature of 300 °C in N_2 atmosphere for 30 min. Subsequently, a thin intermediate layer (Ni) was added to improve the contact properties, followed by depositing Cu electrode film on top of the Ni layer to connect the TE legs. Cu and Ni films were deposited at room temperature for 2 h and 15 min, respectively, with a power supply of 30 W and Ar working pressure of 0.5 Pa.

3. Design and fabrication of TE device

3.1. Light-concentrated TE device

In addition to the optimization from the perspective of the TE material and device, the energy concentration strategy and thermal design of the heat sink were also studied with consideration given to system integration. Fig. 3 shows the schematic illustration of the