Fabrication and characterization of roll-type thin-film thermoelectric generators

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Abstract. We propose and demonstrate a facile fabrication method of roll-type thin-film thermoelectric generators to conveniently use a low electric power converted from ambient heat sources. The generators consisted of n-type Bi₂Te₃ and p-type Sb₂Te₃ thin films, which were deposited on a polyimide sheet by radio-frequency magnetron sputtering. Both types of thin films were thermally annealed to increase the thermoelectric properties. The annealed thin films were connected to each other using silver paste, with the p-n junctions connected electrically in series. The completed generator consisted of five p-n pairs was 625 mm long and 26 mm wide. To form the roll-type structure, the generator was bent so that the fold lines were located at the p-n junctions. The performance of the generators, such as the open circuit voltage (Vₜₒ) and maximum output power (Pₘₐₓ), were measured by applying a temperature difference (ΔT) between the ends of the generator. As a result, the generator exhibited a Vₜₒ of 20.1 mV and a Pₘₐₓ of 48.5 nW at a ΔT of 30 K. In addition, we calculated the performances of generators with different film thickness or number of p-n pairs based on the experimental results.

1. Introduction

The use of thermoelectric power generators is one of the key issues to make the internet of things (IoT) widespread. This is because IoT technology uses various sensors such as wireless sensor nodes [1,2], wearable sensors [3,4]. These sensors do not need high electric power, but do need batteries with long duration and high reliability. The thermoelectric generators satisfy these requirements. They directly generate a small amount of electrical energy from the ambient thermal energy in the surrounding space. In addition, the thermoelectric generators do not have moving parts or fluids because they use an all-solid-state technology, leading to an almost maintenance-free throughout the lifespan. Thin-film thermoelectric generators are quite favorable for the application of IoT. The thin-film thermoelectric generators are not able to produce higher electric power, but they are lightweight and can be attached on curved surfaces such as pipes or human bodies by using flexible substrates. In general, thin-film thermoelectric generators have a lateral (in-plane) configuration. A rectangular meander made of many p- and n-type material pairs connected with metal electrodes is formed on a substrate [5-7]. This configuration can increase the output power as the number of pairs and the temperature difference (ΔT) are increased. However, the generation of ΔT in the in-plane direction is limited in application compared to that in the cross-plane direction. To extract the favorable features of the in-plane type of thin-film thermoelectric generators, those with roll-type structures have been
proposed. Shiozaki et al. [8] fabricated flexible thermoelectric generators with a 3D woven shape, which were formed to stretch the cold and hot junction of the thermoelectric legs by mechanical force. This structure can make the $\Delta T$ appear to be in the cross-plane direction. To make the use of roll-type thin-film thermoelectric generators widespread, simple structure and facile fabrication methods, which are not used by MEMS techniques such as photolithography and precision machining, should be considered. In this study, we propose and demonstrate a simple structure of roll-type thin-film thermoelectric generators by a facile fabrication method.

2. Experimental setup

Figure 1 shows the illustration of the fabrication process of a roll-type thermoelectric thin-film generator. The thin films were deposited using RF magnetron sputtering (Tokuda CFS-8EP). The basic experimental setup was described in our previous publications [9,10]. Initially, we prepared ten pieces of polyimide sheets (26 mm × 76 mm × 25 μm). N-type Bi$_2$Te$_3$ thin films with a thickness of 0.8 μm were deposited on the five pieces of polyimide sheets, and p-type Sb$_2$Te$_3$ films with a thickness of 0.8 μm were deposited on the remaining five pieces of polyimide sheets. The polyimide sheet was cut into 10 pieces because a longer polyimide sheet made it difficult to obtain a uniform film thickness due to the limitation of the deposition area in the sputtering equipment, and made it difficult to perform thermal annealing with a uniform temperature due to the limited soaking area in the electric furnace. After depositing the n- and p-type thin films, these films were thermally annealed at the optimized temperature. Then, the sample was assembled by electrically connecting the p-n junctions in series to form the generators. The p-type film was set to adhere to the n-type film, and connected by spraying silver paste. After baking the sample to dry the silver paste, the generator was bent like an accordion so that the fold lines were located at the p-n junctions. To measure the performance of the generators by applying $\Delta T$ between the ends of the generator, the lower half of the generator was dipped in hot water while the upper half was kept in the atmosphere.

3. Results and discussion

Figure 2 shows the performances of the generators, using both types of the thin films annealed at 300°C, as a function of $\Delta T$; the $\Delta T$ at the ends was changed from 10 to 30 K. In Fig. 2(a), a $V_{oc}$ of 11.2 mV was achieved at a $\Delta T$ of 10 K. With increasing the $\Delta T$, $V_{oc}$ linearly increased. A $V_{oc}$ of 20.7 mV
was achieved at a $\Delta T$ of 30 K. This result is in relatively good agreement with that of previous reports with similar device structures and the similar $\Delta T$ [6,11].

The maximum output power ($P_{\text{max}}$) is given by Eq. 1,

$$P_{\text{max}} = V_{oc}^2/4R_{\text{total}},$$  \hspace{1cm} (1)

where $R_{\text{total}}$ is the measured total resistance of the generator. $R_{\text{total}}$ of the generator was measured by a digital multimeter to be 2200 $\Omega$. In Fig. 2(b), $P_{\text{max}}$ was determined to be 14.4 nW at the $\Delta T$ of 10 K. $P_{\text{max}}$ increased as the $\Delta T$ increased, and a $P_{\text{max}}$ of 48.5 nW was achieved at the $\Delta T$ of 30 K. However, $P_{\text{max}}$ is not enough to activate sub-microwatt or microwatt wearable devices such as a temperature sensor [12] or CMOS image sensor [13].

Therefore, to apply the generators using a facile fabrication method into sub-microwatt wearable devices, we calculated the generator performance by changing the film thickness or the number of p-n pairs. Based on the experimental result, the Seebeck coefficient shows no dependence on the film thickness, so that $V_{oc}$ is only proportional to the number of p-n pairs. When the generator is composed of 20 p-n pairs, the $V_{oc}$ is expected to be 82.6 mV.

To calculate $P_{\text{max}}$, $R_{\text{total}}$ should be determined when the film thickness or the number of p-n pairs are changed. $R_{\text{total}}$ is sum of the resistance of the n- and p-type thin films, $R_{\text{film}}$, and the contact resistance, $R_c$. $R_{\text{film}}$ is given by Eq. 2,

$$R_{\text{film}} = n \left[ \frac{1}{(l/w')d} \left( \frac{l}{w' d} \right) + \frac{1}{\sigma_{\text{Bi-Te}} \sigma_{\text{Sb-Te}}} \left( \frac{l}{w' d} \right) \right],$$ \hspace{1cm} (2)

where $n$ is the number of p-n junctions, $\sigma_{\text{Bi-Te}}$ and $\sigma_{\text{Sb-Te}}$ are the measured electrical conductivities of Bi$_2$Te$_3$ and Sb$_2$Te$_3$ thin films annealed at 300°C, respectively. The length (76 mm), width (26 mm), and thickness are represented by $l$, $w'$, and $d$, respectively. On the other hand, $R_c$ is given by Eq. 3,

$$R_c = (2n-1)R_{c0},$$ \hspace{1cm} (3)

where $(2n-1)$ means the number of p-n junctions, and $R_{c0}$ is the contact resistance per p-n junction. Based on the experimental result, $R_c$ for 5 pairs of p-n junctions is calculated by substituting $R_{\text{film}}$ (840 $\Omega$) from $R_{\text{total}}$ (2200 $\Omega$), and was determined to be 1360 $\Omega$. As a result, $R_{c0}$ was determined to be 151

**Figure 2.** The performances of the generators, using both types of the thin films annealed at 300°C, as a function of $\Delta T$; the $\Delta T$ at the ends was changed from 10 to 30 K.

(a) Open circuit voltage, (b) maximum output power.
Ω. We here assumed that the magnitude of $R_{\theta}$ is the same in each p-n junction. Figure 3(b) shows the calculated $P_{\text{max}}$ of the generator at $\Delta T = 30$ K as a function of film thickness for various numbers of p-n pairs with the experimental result. The calculated $P_{\text{max}}$ increased as the number of p-n pairs, regardless of the film thickness. The calculated $P_{\text{max}}$ for all numbers of p-n pairs rapidly increased until the film thickness reached approximately 1.0 μm, because of the decrease in the resistance of the Bi$_2$Te$_3$ and Sb$_2$Te$_3$ thin films. On the other hand, the calculated $P_{\text{max}}$ did not change so much when the film thickness was greater than 3 μm. Therefore, for example, to activate a CMOS image sensor at the consumption power of 226 nW [13] using the roll-type thin-film thermoelectric generator, the structure of the generator is expected to need 20 p-n pairs with 2.0 μm thick-film.

![Figure 3](image_url)

**Figure 3.** The calculated $V_{oc}$ of the generator at $\Delta T = 30$ K as a function of film thickness for various numbers of p-n pairs. (a) Open circuit voltage, (b) maximum output power.

4. Conclusion

The roll-type thin-film thermoelectric generators were prepared by RF sputtering method. By applying a $\Delta T$ between the ends of the generator, we confirmed that the generators produced an electric power.

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