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Flexible thermoelectric generator with polydimethyl siloxane in thermoelectric material and substrate

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ABSTRACT

In this paper, the fabrication and characterization of a flexible thermoelectric generator (TEG) is presented. Bismuth telluride powder (Bi/Te) is utilized as p- and n-type thermoelectric materials, and carbon nanotubes (CNTs) are used to enhance electrical conductivity. Both the CNTs and bismuth telluride (Bi/Te) powder are mixed with a solution of polydimethyl siloxane (PDMS) as a precursor. The same PDMS used for the hybrid thermoelectric materials is also used as the substrate, making the TEG flexible and increasing its stability. The Seebeck coefficients of the fabricated p- and n-type thermoelectric materials are 143 and -174 μV/K, respectively. The output voltage of the fabricated device is 920 mV and the generated power is 570 μW/cm^2 with a temperature difference of 60 °C. The fabricated TEG maintains its performance level during bending reliability tests on a curvature with a radius as small as 5 mm, and after more than 1000 repetitions of bending on a curvature with a radius of 20 mm.

1. Introduction

In recent years, interest in and demand for smart watches such as the Fitbit, Apple Watch, and Samsung Gear S2 have increased significantly. Smart watches measure various types of biometric information, including real-time heart rate, calorie consumption, step posture, and sleep patterns. However, smart watches collect biometric information only when the user is wearing the device. Moreover, all commercially available smart watches are battery operated and cannot operate while charging.

To address these limitations, smart watch manufacturers adopt methods that minimize power consumption, enhance battery performance, and increase the device's operating time between charges. However, none of these improvements allow the continuous use of a smart watch without a charging interval. Therefore, to address the need for batteries that can allow continuous use, research has been conducted on thermoelectric generators (TEGs) that can generate power continuously by using heat in various ways [1–3].

TEGs generate power from the temperature difference between two components, and their power generation efficiency increases when there is less heat loss during the heat transfer into the TEG. Because of this characteristic, it is necessary to consider the curvature and movements of the human body when developing TEGs to be applied in smart watches. Because human bodies have many curves and move, existing TEGs [4,5] using ceramic-based substrates do not have the flexibility to continuously contact body surfaces, thereby increasing heat loss and reducing the power generation efficiency. To resolve this problem, a number of studies have been conducted on flexible TEGs that can operate while attached to an active human body [1,3,6–12].

Flexible TEGs are generally fabricated either from inorganic thermoelectric materials [1,3] or flexible polymer materials [6–13]. Both these methods employ flexible materials for substrates. In the first case, when inorganic thermoelectric materials are used in conjunction with flexible substrate materials, a bending force is concentrated on the substrate when the TEG bends because the thermoelectric materials are rigid. The bending force is focused on interconnects fabricated on the substrate surface, thereby limiting the device's flexibility. On the other hand, in the second case, when flexible polymer thermoelectric materials are used in conjunction with flexible substrate materials, the bending force is uniformly distributed throughout the TEG. This results in more favorable bending characteristics than are exhibited by inorganic thermoelectric materials; however, the flexible polymer thermoelectric material exhibits high contact resistance between the polymer material and interconnects when the interconnects are deposited on the substrate surface [7,8]. A limitation common to both

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methods is the low bonding force between their heterogeneous thermoelectric and substrate materials. This low bonding force indicates that the thermoelectric material and substrate are likely to be separated during use, degrading the performance of the TEG.

To address these limitations, this paper describes a simple method of fabricating a TEG from a hybrid thermoelectric material made by granulating carbon nanotubes (CNTs) into p/n-type bismuth telluride (Bi/Te) and distributing this mixture within the highly flexible material polydimethyl siloxane (PDMS). This hybrid thermoelectric material was processed into pillar shapes, and both the substrate and material base were composed of the same PDMS. Furthermore, the proposed method includes selective etching of the PDMS to reduce contact resistance between interconnects and the fabricated thermoelectric material. The thermoelectric and bending characteristics of the flexible TEG fabricated in this manner were measured to verify that the proposed method produces a new type of TEG that resolves the limitations of previous designs.

2. Design and concept of the proposed TEG

Fig. 1 shows the fabrication process used to manufacture the proposed TEG, which consists of 50 pairs of p- and n-type thermoelectric columns with diameters of 5 mm, heights of 300 μm, and lengths of 12 mm. The p- and n-type thermoelectric materials were connected sequentially via a 300-nm-thick aluminum interconnect at the top and bottom of the columns. At the edge of the device, an electrical pad was positioned to measure performance.

The open circuit Seebeck voltage generated by the TEG can be expressed by

$$V_o = n\alpha_{pn}\Delta T_{TEG} \tag{1}$$

where n represents the number of thermocouples, α_{pn} represents the difference between the Seebeck coefficients of the p- and n-type materials, and ΔT_{TEG} represents the temperature difference between the two ends of the TEG. The maximum output power generated by the TEG can be expressed by

$$P_o = \left(n\alpha_{pn}\Delta T_{TEG}\right)^2 \Big/ \left(4R_{TEG}\right) \tag{2}$$

where R_{TEG} represents the internal resistance of the TEG.

3. Experimental procedure

3.1. Preparation of thermoelectric material

The proposed hybrid thermoelectric material was fabricated from multi-walled CNTs (CM-250, Hanwha Nanotech Corp., Korea), p/n-type Bi/Te (p/n-type, 99.99%, R&D Korea, Korea), and PDMS (Sylgard 184, Dow Corning, USA). To granulate the CNTs into p/n-type Bi/Te, 10 wt% Bi/Te and 2.5 wt% CNTs were added to ethanol (99.9%, Wako Pure Chemical Industries, Ltd., Japan). The solution was stirred with a magnetic stirrer (MS-20D, Witeg, Germany) at 500 rpm for 12 h and heated at 100 °C for 24 h to evaporate the ethanol, thereby fabricating a powder of CNTs granulated on the Bi/Te surface. The 10 wt% of the fabricated powder was added to the PDMS principal material and stirred at 50 rpm for 3 h by using an overhead stirrer (HT-50DX, Witeg, Germany). Subsequently, an even distribution of components within this material was achieved with an ultrasonic machine (WUC-D03H, Witeg, Germany), operated at an input power of 200 W and a frequency of 40 kHz for 3 h. A hardener was then added to the principal material at a ratio of 10:1, followed by stirring at 50 rpm for 30 min, followed by an additional 30 min of ultrasonic processing at the same input power and frequency as the first round. Subsequently, the Bi/Te, CNT, and PDMS mixture was degassed under vacuum conditions.

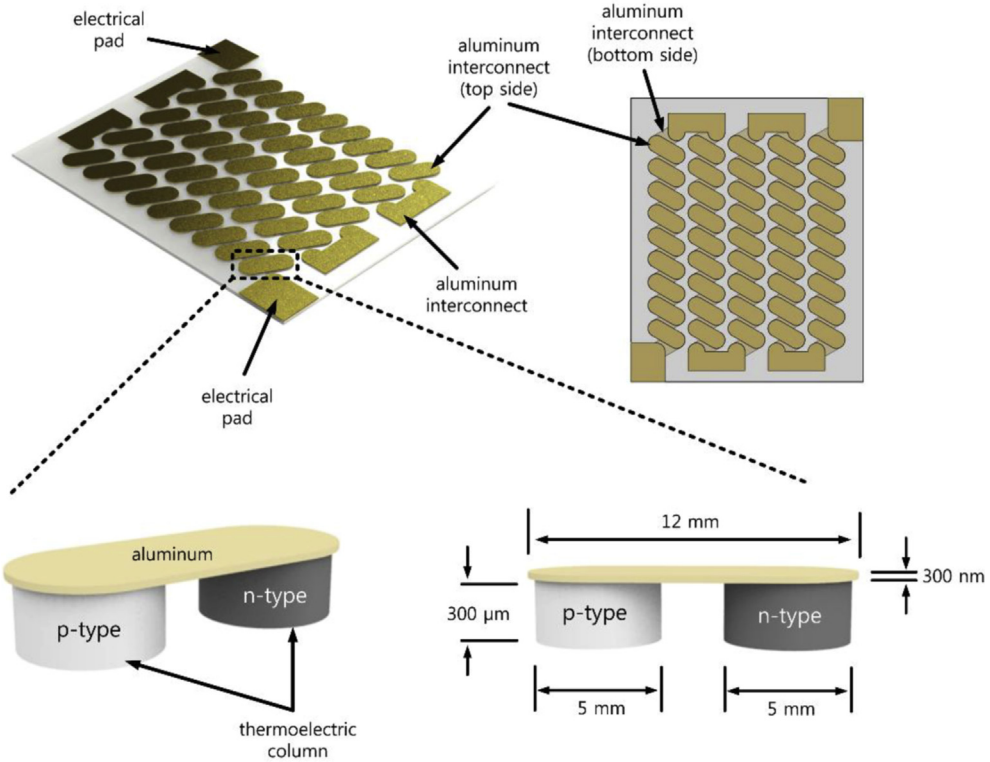

Fig. 1. Configuration of the proposed thermoelectric generator (TEG). The PDMS substrate is not shown in the figure to facilitate visualization of the core components.