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## Thermoelectric generator based on a bismuth-telluride alloy fabricated by addition of ethylene glycol

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#### ABSTRACT

This paper introduces a new method to selectively fabricate n-type and p-type bismuth (Bi)-telluride (Te) thermoelectric materials by the rate of addition of ethylene glycol (EG) in the Bi–Te co-electrodeposition solution. As the amount of added EG is increased, the atomic ratio of Bi in the deposited Bi–Te alloy reached a slope of 0.463 (at% of Bi/vol% of EG), and increased in a linear manner. When the EG content reached approximately 20%v/v, the n-type material changed into a p-type. This change implies that adjusting the EG content in the electrodeposition solution affords simple control of the Bi–Te composition. To demonstrate the applicability of the developed thermoelectric materials, thermoelectric generators (TEGs) were fabricated using electrodeposited n-type (using solution with 30%v/v EG) Bi–Te alloys. The Seebeck voltage of the pair of n-type and p-type thermoelectric materials was 140 mV and the power generated from the pair was 24.36 nW at a 10 °C temperature difference.

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#### 1. Introduction

A thermoelectric generator (TEG) is an energy conversion device that directly converts heat energy into electrical energy. When a heat source exists it is able to produce electricity without other mechanical actuating elements. Continuous generation of electricity is therefore possible and the device has the advantage of not causing vibration or noise. Recently, the development of thermoelectric elements using body heat has been actively pursued and such technology has been commercialized in the form of wristwatches, which consume relatively little energy [1]. Wearable thermoelectric devices are limited by the area of the device. Improvement of the thermoelectric material performance index, along with miniaturization and high integration of the thermocouple, which is composed of a pair of p-type and n-type thermoelectric materials, are therefore necessary in order to increase electricity production.

For the development of thermoelectric materials and high integration of such materials, various methods that are used in nano- and micromachining technologies have recently been actively utilized to fabricate TEGs. Evaporation [2], sputtering [3],

\* Corresponding author. E-mail address: mems@pusan.ac.kr (J.S. Ko). metal organic chemical vapor deposition (MOCVD) [4], and electrodeposition [5] are the main approaches used for the deposition of thermoelectric materials. Among these methods, electrodeposition is relatively inexpensive and provides high deposition speeds. It also offers the advantage of a capability to form large area films. Moreover, electrodeposition can be selectively applied and the p-type and n-type thermoelectric materials can be formed at desired locations [6].

Research on Bismuth (Bi)-Telluride (Te) alloy deposition using electrodeposition has recently increased [5,7–9]. When the Bi atomic percent (at.%) in the coated Bi–Te alloy is lower than 40%, the material becomes n-type, whereas it becomes p-type above this level [6]. In order to fabricate two different types of thermoelectric materials, various methods to control the applied current, voltage, temperature of the electrolyte, stirring speed of the solution, and Bi–Te content have been developed [6,8,10]. However, obtaining thermoelectric materials with the desired Bi–Te composition ratio by controlling the above variables is very complicated and a significant amount of time and effort are required to establish suitable processing conditions.

This study presents a new method to control the Bi and Te composition ratio in the deposited alloy film by controlling only the ethylene glycol (EG) content in the Bi—Te co-electrodeposition solution. TEGs are then fabricated using the p-type and n-type Bi—Te thermoelectric materials developed in this study to demonstrate





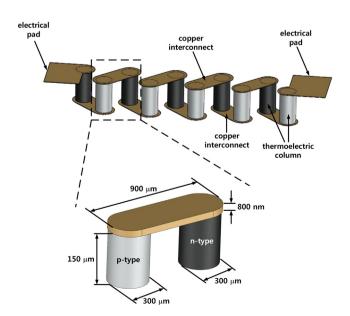
Current Applied Physics Protocological Applied Cological Applied Appli their possible applicability and their performance is evaluated. Ptype and n-type thermoelectric columns are embedded in a polymer (SU-8) substrate and they are electrically connected in series.

#### 2. Experimental

#### 2.1. Preparation of Bi-Te thermoelectric films

To fabricate the electrodeposition solution, the volume ratio of ethylene glycol (107-21-1, Alfa Aesar, USA, 99%) was varied (0, 10, 20, 30, 40%v/v) in the solutions composed of 10 mM Bi (LEES CHEM, Korea, 99.9%), 10 mM TeO<sub>2</sub> (Alfa Aesar, UK, 99.99%), and 1 M HNO<sub>3</sub> (70% electronic grade, OCI Company, Korea). The mole concentrations of Bi, TeO<sub>2</sub>, and HNO<sub>3</sub> were maintained in all electrodeposition solutions, regardless of the EG content. For the solutions, deionized water (OCI Company, Korea) with a specific resistance over 18 MW·cm was used throughout the study. A potentiostat (Versastat2, Princeton Applied Research, USA) was used for the electrodeposition. A  $2.5 \times 2.5 \text{ cm}^2$  platinum mesh (i-Nexus, USA) was used as the counter electrode and Ag/AgCl/KCl +0.197 V vs SHE (Princeton Applied Research, USA) was used as the reference electrode. A scanning electron microscopy (SEM) (S-4800, HITACHI, Japan), energy dispersive spectroscopy (EDS) (7593-H, HORIBA, Japan) and X-ray diffraction (XRD) system (D8 advance, Bruker, USA) were used to analyze the formed structure's shape, composition and lattice structure.

After forming a silicon dioxide film of 300 nm thickness on the silicon wafer, Ti (10 nm)/Au (300 nm) was sequentially deposited via sputtering (ENDURA-5500, AMAT, USA). While the silicon dioxide served as an insulation layer, the Ti/Au layer played the role of a conducting seed layer for electrodeposition. The electrodeposition area was fixed at  $10 \times 10 \text{ mm}^2$ , and the gaps between the working electrode, reference electrode, and counter electrode were maintained at 10 mm. The applied current during electrodeposition was  $-100 \text{ mA/cm}^2$  and of a pulse wave form. One cycle was composed of pulse on-time (0.1 s) and off-time (5 s), and electrodeposition was performed for a total of 3,000 cycles for each specimen. The temperature of the electrodeposition solution was 18 °C and electrodeposition was done with no stirring.



**Fig. 1.** Configuration of a TEG having five thermocouples. The SU-8 substrate is not shown in the figure for better visualization of the core components.

#### 2.2. Design and fabrication of TEG

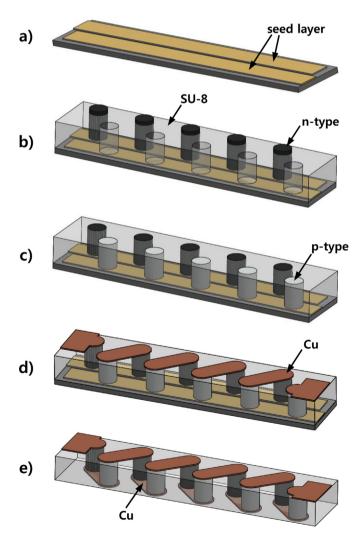
Fig. 1 shows the design of the proposed TEG, wherein five thermocouples are connected in series. Each thermocouple is composed of a pair of p-type and n-type thermoelectric columns. The open circuit Seebeck voltage generated by a TEG can be expressed as [11]

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

where *n* is the number of thermocouples,  $\alpha_{pn}$  the relative Seebeck coefficient of the p-type and n-type thermoelectric materials, and  $\Delta T_{\text{TEG}}$  the temperature difference between the cold and hot surfaces of the TEG. Power is the product of the output voltage and electrical current. Therefore, the maximum output power generated by a TEG can be expressed as

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

where R<sub>TEG</sub> denotes the internal electrical resistance.



**Fig. 2.** Microfabrication process sequence of TEGs: (a) Ti/Au conducting seed layer deposition and patterning, (b) photolithography using SU-8 and electrodeposition of n-type thermoelectric material, (c) electrodeposition of p-type thermoelectric material and CMP of the top surface, (d) topside Cu deposition, and (e) removal of Si substrate and Ti/Au seed layer by CMP process and bottom side Cu deposition.

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