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Silk fabric-based wearable thermoelectric generator for energy harvesting from the human body

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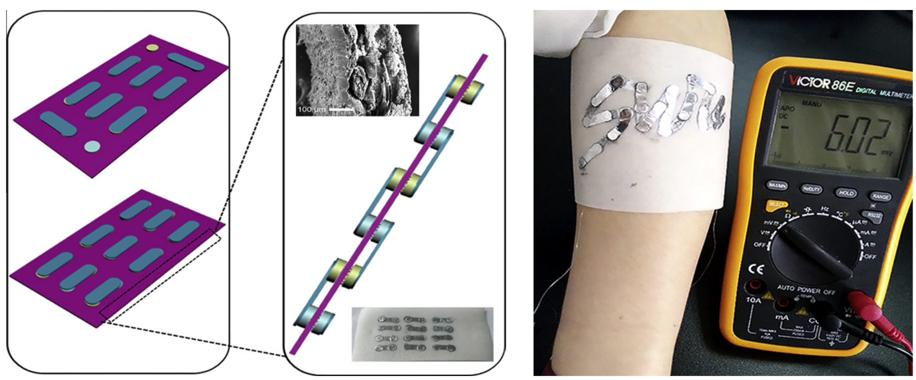
HIGHLIGHTS

- A thermoelectric (TE) generator was prepared on commercially available silk fabric.
- The silk fabric-based TE generator could convert thermal energy into electricity.
- Applications of the TE generator for human body heat harvesting were demonstrated.

GRAPHICAL ABSTRACT



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ABSTRACT

The development of flexible thermoelectric (TE) power generators for harvesting energy from the human body has attracted significant interest in recent years. However, thus far, a wearable TE power generator based on commercially available fabrics has not been realized. In this study, nanostructured Bi_2Te_3 and Sb_2Te_3 were synthesized and deposited on both sides of a silk fabric to form TE columns. These TE columns were connected with silver foils to fabricate a prototype integrating an array of 12 thermocouples. The generator could effectively convert thermal energy into electricity in the temperature difference (ΔT) range of 5–35 K. The maximum voltage and power outputs were ~10 mV and ~15 nW, respectively, with no significant change in both, during 100 cycles of bending and twisting. Different voltage output profiles were collected from an arm-attached generator before and after 30 min of walking, to highlight the immense potential of the silk fabric-based TE generator. This study provides a new approach for developing fabric-based TE power generators for practical applications in wearable electronics.

1. Introduction

Rapid advancements in electronics and miniature technology have facilitated the development of multifunctional wearable devices for real-time health monitoring and fitness tracking in recent years [1–4]. Power-supply systems, which are a critical component, should fulfill the requirements of size, weight, flexibility, and wearability while maintaining their functions [5–7]. Currently, rechargeable lithium-ion (Li-ion) batteries that are compact and lightweight are the most widely utilized energy storage systems for wearable electronics [8–10]. Although they can provide sufficient energy to power the devices, their small size signif-

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icantly restricts their capacity and battery life. This has become the main constraint of wearable technology.

Batteries can be combined with wearable generators to harvest energy from the environment or the human body, creating self-chargeable systems for continuous operation of these devices. Thermoelectric (TE) power generators have been developed to harvest energy from waste heat generated by the human body through physical contact with the human skin [11,12]. According to the Seebeck effect, the power output produced by the TE generator is proportional to the thermal gradient between the hot junction that is in contact with the skin and the environment-exposed cold junction [13,14]. Flexible TE power generators containing several pairs of thermocouples have been fabricated on various substrates for heat collection from the human body [15,16]. Polymer films including polyethylene (PE), polydimethylsiloxane (PDMS), polyimide (PI), and Kapton films are the most popular substrates possessing excellent flexibility for use in TE power systems [17–19]. Because both hot and cold junctions can be fabricated only on the same side of the film, the TE generators can be used as skin or cloth-attached devices, and are not directly wearable. Very recently, thermocouples have been deposited on polyester or glass-based fabrics, which have the feasibility to be fabricated in the form of clothing [20]. However, in these studies, specially designed fabric structures with large holes have been utilized to ensure the penetration of TE materials, which cannot be adapted to well-established weaving techniques. Hence, it is highly desirable to develop an approach that could match the existing weaving techniques for the fabrication of wearable fabric-based TE generators. To the best of our knowledge, there are no previous studies on wearable TE power generators based on commercially available fabrics.

Silk from *Bombyx mori* cocoons has been regarded as "the queen of textiles" owing to its softness, high hygroscopicity, and superior skin affinity [21–25]. This fabric has been used to make clothing for thousands of years, and therefore, it is an ideal substrate for the development of wearable TE generators. Bismuth telluride (Bi_2Te_3) and antimony telluride (Sb_2Te_3) are the most widely used TE materials because of their high TE efficiency near room temperature (25 °C) [26,27]. Herein, we present a wearable TE power generator based on commercially available silk fabrics for energy harvesting from waste heat generated by the human body. Nanostructured Bi_2Te_3 and Sb_2Te_3 were synthesized hydrothermally and deposited on both sides of the silk fabric to form TE columns of *n*-type and *p*-type materials. After connecting the columns with silver foils, a prototype, integrating an array of 12 thermocouples was fabricated and evaluated as a wearable TE generator. The effects of repeated bending or twisting on the performance of the generator were examined. The practical applications of silk fabric-based TE generators with designed patterns for the collection of heat energy from the human body are demonstrated in this study.

2. Experimental section

2.1. Chemicals

TeO_2 (metals basis, 99.99%), $Bi(NO_3)_3{\cdot}5H_2O$ (99.995%), $SbCl_3$ (AR, 99.98%), polyvinyl pyrrolidone (PVP, MW 58000), $N_2H_4{\cdot}H_2O$ (98%), and ethylene glycol (EG, 99%) were purchased from Aladdin (Shanghai, China). Deionized water (resistance over 18 MΩ cm) was generated by a Millipore Q water purification system.

2.2. Synthesis of nanostructured Bi_2Te_3 and Sb_2Te_3

Nanostructured Bi_2Te_3 and Sb_2Te_3 were synthesized hydrothermally as in previous studies [28–30]. 1.2 g PVP and 6 mmol TeO_2 powder were mixed in 100 mL of EG with vigorous stirring at 200 °C. 0.5 g of NaOH was added to the mixture and stirred for 20 min. Then, 0.6 mL of $N_2H_4{\cdot}H_2O$ was injected into the solution, and the solution was stirred for another 60 min to obtain the Te nanotubes. For the preparation of Bi_2Te_3, the Bi precursor was reacted with the Te nanotubes for 60 min at 160 °C. The final product was collected by centrifugation at 8500 rpm, and washed several times with water and absolute ethanol. As to the synthesis of Sb_2Te_3, 0.5 mmol $SbCl_3$, 15 ml hydrazine ($N_2H_4{\cdot}H_2O$), and 0.6 mmol Te nanotubes were added to 20 mL deionized water under ultrasonic irradiation at room temperature for 30 min. The mixed solution was heated in a Teflon-lined stainless steel autoclave (50 mL) at 150 °C for 6 h. After cooling to room temperature, the gray-black product was obtained by centrifugation, and it was washed three times using water.

2.3. Characterization of materials

Morphologies of the materials, silk fabrics, and the TE generator devices were imaged using a JSM-6510LV scanning electron microscope (JEOL, Tokyo, Japan) operating at 20 kV. During SEM measurements, EDS (INCA X-Max 250) data were collected to analyze the chemical elements. XRD spectra were examined using a Cu Ka-ray with tube conditions at 40 kV and 30 mA ranging from 10° to 80° (XRD-7000, Shimadzu, Japan). Bi_2Te_3 and Sb_2Te_3 pellets with diameters of 10 mm and thicknesses of ~2.5 mm were prepared using spark plasma sintering for measuring the Seebeck coefficient. The voltage across the pellet was measured using a Keithley 2400 multimeter. The Seebeck coefficient was extracted from the slope of the voltage versus temperature difference (ΔT) curves.

2.4. Fabrication of silk-based TE power generator

Fig. 1 shows the fabrication process of a silk-based TE power generator. A 4 cm × 8 cm silk fabric coated with a layer of polyvinyl alcohol was pricked with a needle to generate holes at the designated locations, to ensure good contact on both the sides of the material. A paste consisting of TE materials (80 mg Bi_2Te_3 or 120 mg Sb_2Te_3), liquid adhesive binder, and deionized water was repeatedly deposited on both the sides of the silk fabric to create TE material columns with a thickness of ~300 μm and a diameter of 4 mm. The as-prepared silk fabrics were vacuum dried at 120 °C for 10 min. Then, conductive silver foils were pasted onto the TE columns with silver paste to connect the *p*-type and *n*-type material columns. A prototype consisting of 12 thermocouples was fabricated for performance measurements.

2.5. Performance evaluation of the silk-based TE power generator

A homemade system consisting of a heating stage and an aluminum-cooling surface was set up for the generation of ΔT, and the characteristic output of the TE generator (Fig. S1 in the Supplementary Information). ΔT from 5 to 35 K was generated using the system. The electrical performance of the TE device was measured using a Victor 86E digital multimeter.

3. Results and discussion

3.1. Characterization of Bi_2Te_3 and Sb_2Te_3 nanomaterials

Because the performance of the TE power generators significantly depends on the TE materials, it is very important to check the quality of the synthesized products. Morphologies of the *p*-type and *n*-type TE nanomaterials were investigated using SEM.