

Full paper

Fabrication and excellent performances of Bi_{0.5}Sb_{1.5}Te₃/epoxy flexible thermoelectric cooling devices

Weikang Hou¹, Xiaolei Nie¹, Wenyu Zhao*, Hongyu Zhou, Xin Mu, Wanting Zhu, Qingjie Zhang*

State Key Laboratory of Advanced Technology for Materials Synthesis and Processing, Wuhan University of Technology, Wuhan 430070, China

ARTICLE INFO

Keywords:

Flexible thermoelectric device
Bi_{0.5}Sb_{1.5}Te₃/epoxy thick film
Hot-pressing curing
Electrical transport properties
Cooling performance

ABSTRACT

How to fabricate flexible thermoelectric (TE) films with excellent electrical transport properties remains a major bottleneck for high-performance flexible TE cooling devices. Herein, a simple and efficient method is developed to fabricate high-performance flexible TE cooling device by selecting Bi_{0.5}Sb_{1.5}Te₃ as TE component and epoxy resin as adhesive. It is discovered that the (000 l) preferential orientation of Bi_{0.5}Sb_{1.5}Te₃ grains could be successfully realized in Bi_{0.5}Sb_{1.5}Te₃/epoxy thick films by hot-pressing curing process, thus causing the remarkable enhancement in the electrical transport properties. The highest power factor of the (000 l)-oriented Bi_{0.5}Sb_{1.5}Te₃/epoxy thick films reached 0.84 mW m⁻¹ K⁻² at 300 K, increased by 250% and 223% as compared with that of our non-pressure thick film and the highest value by other groups, respectively. A prototype flexible TE cooling device had been fabricated with the Bi_{0.5}Sb_{1.5}Te₃/epoxy thick films. The stable temperature difference of the prototype device reached 6.2 K under a working current of 0.06 A, increased by about 24% as compared with the highest temperature difference of 5 K reported by other groups. Our work demonstrates that the hot-pressing curing process provides an effective approach to fabricate high-performance flexible TE cooling devices with Bi_{0.5}Sb_{1.5}Te₃/epoxy thick films through the (000 l) preferential orientation.

1. Introduction

How to take away the harmful waste heat occurring at nonplanar local areas have posed great challenges in various microelectronic integrated devices as widely used in powerful processors or end-user operation platforms [1–4]. The thermal management technology based on Peltier effect of thermoelectric (TE) materials has attracted increasing attention due to a series of unique advantages such as high reliability, no moving parts, good compactness, and lightweight [5–7]. However, all the currently commercial TE devices are composed of sintered bulk TE legs connected electrically in series through metallic electrodes and then sandwiched between ceramic plates [8,9]. The conventional structure has poor flexibility and thus it is very difficult to realize dimensionally good match between the commercial TE device and the electronic component. The miniaturized flexible TE devices based on TE films are highly promising to solve the bottleneck [3,4,7,10]. However, the preparation of thin films with less than 100 nm in thickness is not only time-consuming but also very inefficient. Alternatively, the thick films prepared by cost-effective printing technologies become a potential candidate for realizing miniaturization and flexibility of TE devices.

The conversion efficiency of a TE cooling device is expressed by the coefficient of performance (COP) [11]

$$COP = \frac{T_c}{T_h - T_c} \cdot \frac{\sqrt{1+ZT} - T_h/T_c}{\sqrt{1+ZT} + 1}$$

where T_h and T_c are the temperatures of the hot side and cold one of the TE device, ZT is the dimensionless figure-of-merit of TE materials $ZT = \alpha^2 \sigma T / \kappa$, where T , α , σ and κ are absolute temperature, Seebeck coefficient, electrical conductivity and thermal conductivity, respectively. Apparently, a TE device with high COP requires high ZT of TE materials.

Great progress has been made in developing high-performance bulk TE materials in the recent two decades and several TE materials have ZT values exceeding 2.0 [12–17]. Unfortunately, when printing the high-performance bulk TE materials on the flexible substrates and transforming into flexible TE thick films with the same chemical compositions, the ZT values were greatly decreased due to the largely deteriorated electrical transport properties originating from low density and various structural defects [18,19]. Several strategies such as cold isostatic pressing, [20,21] selecting additives as sintering aids [22,23] and burning off the insulating binders via high temperature sintering [24]

* Corresponding authors.

E-mail addresses: wyzhao@whut.edu.cn (W. Zhao), zhangqj@whut.edu.cn (Q. Zhang).

¹ These authors contributed equally to this work.

were employed to enhance the electrical transport properties of flexible TE composite thick films. However, so far, the poor electrical transport properties of flexible TE thick films are still a major bottleneck in developing high-performance flexible TE devices.

In this work, a series of $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ /epoxy flexible thick films with (000 l) preferential orientation were prepared on polyimide substrates by the combination of brush-printing and hot-pressing curing processes. The (000 l)-oriented $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ /epoxy flexible thick films exhibited excellent electrical transport properties due to the (000 l) preferential orientation of $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ grains induced by the hot-pressing curing process. The highest power factor reached $0.84 \text{ mW m}^{-1} \text{ K}^{-2}$ at 300 K for $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ /epoxy thick film cured under 623 K and 4 MPa, increased by 180% and 223% as compared with that of our non-pressure thick film and the highest value reported by other groups, respectively. The FTIR spectra show that the main structure of three-dimensional cross-linked network of epoxy resin should keep well in all the thick films cured in the temperature range of 473–623 K. A prototype flexible TE cooling device was designed and fabricated with the $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ /epoxy thick films. The cooling performance of the prototype flexible TE device was evaluated by measuring the temperature difference between the hot side and cold one of TE legs. This work demonstrates that the hot-pressing curing process provides an effective approach to fabricate high-performance $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ /epoxy flexible TE devices through producing the (000 l) preferential orientation of $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ grains in $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ /epoxy thick films.

2. Experimental section

2.1. Preparation of printable slurries

The slurries were made of $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ powder, epoxy resin, hardener, catalyst and reactive diluent [19]. Diglycidyl ether of bisphenol-F (NanYa electronic material Inc) was used as epoxy resin. Methylhexahydrophthalic anhydride (Sinopharm chemical reagent) was used as hardener. 2-ethyl-4-methylimidazole (Sinopharm chemical reagent) was used as catalyst. Butyl glycidyl ether (Sinopharm chemical reagent) was used as reactive diluent. Firstly, $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ ingots were crushed and pulverized with a planetary ball mill apparatus at a speed of 200 rpm for 2 h under the protection of argon. Fig. 1 shows the FESEM image of ball-milled $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ powders. It can be seen that the ball-milled $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ grains have flake-like shape, which must be re-arranged along the (000 l) orientation or the ab -plane during the hot-pressing curing process based on the minimal energy principle. Secondly, epoxy resin, hardener, catalyst and reactive diluent were blended to form the epoxy resin mixture. The weight ratio of epoxy-to-hardener was 1:0.85 while the weight ratio of epoxy-to-catalyst was 100:1. The weight ratio of epoxy-to-diluent was 1:2. Thirdly, $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ powders and epoxy resin mixture were blended for 1 h with a mechanical stirrer and then ultrasonically dispersed for 15 min to obtain printable slurries. The optimized weight ratio of $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ powders and epoxy resin mixture was 3:1.

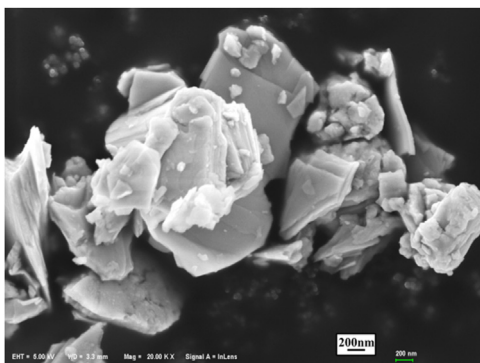


Fig. 1. FESEM image of ball-milled $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ powders.

2.2. Design of prototype flexible TE cooling device based on the (000 l)-oriented $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ /epoxy thick films

Fig. 2 schematically shows the procedure of fabricating prototype flexible TE cooling device based on the (000 l)-oriented $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ /epoxy thick films through the combination of brush-printing and hot-pressing curing processes. Besides the preparation of $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ /epoxy slurries, the whole fabricating procedure still included flexible substrate pretreatment, template process, brush printing, drying, hot-pressing curing, and electrode deposition. The polyimide substrate with about 150 μm in thickness had been first plasma-cleaned and then was compactly covered with template. The $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ /epoxy slurries were brush-printed and formed thick films with about 150 μm in thickness. The thick films were dried at 373 K for 30 min and then transferred into a homemade hot-pressing apparatus to have been cured at different temperatures (473 K, 523 K, 573 K and 623 K) and different hot-pressures (0 MPa, 2 MPa and 4 MPa) to yield compact TE thick films with (000 l) preferential orientation. The TE legs were connected in series by depositing Al/Cu/Ni multilayer thin-film electrodes on the polyimide substrate and TE legs using a vacuum evaporation technique.

2.3. Measurements

Differential scanning calorimeter (DSC) and thermogravimetry (TG) curves of epoxy resin matrix and $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ /epoxy composite were determined by a Netzsch STA449F3 integrative thermal analyzer in N_2 atmosphere in the range of 300–773 K at a heating ratio of 10 K/min. The constituent phases of all the $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ /epoxy thick films were determined by X-ray diffraction (XRD, PANalytical X' Pert PRO) using $\text{Cu K}\alpha$ radiation ($\lambda = 0.15418 \text{ nm}$). The microstructures were examined with a field emission scanning electron microscope (FESEM, Zeiss ULTRA-PLUS-43-13). The surface roughness and thickness of all the thick films were measured on a $500 \mu\text{m} \times 500 \mu\text{m}$ area by employing 3D surface profilometer (NanoMap, 500LS) attached with a scanning probe image processor and analysis software. The density of the thick films was measured with the Archimedes method. Hall coefficient (R_H) and electrical resistivity (ρ) of the thick films were measured with van der Pauw method using an Accent 5500 Hall measurement system at room temperature. The μ_H and n were calculated according to the equations $n = 1/(eR_H)$ and $\mu_H = R_H/\rho$, where e is the charge of an electron. The in-plane electrical conductivity (σ) and Seebeck coefficient (α) were measured with the standard four-probe method (Sinkuriko, ZEM-3) in He atmosphere. The measurement error for σ and α is about $\pm 5\%$. The bending tests of the thick films were performed using a homemade bending test apparatus. The cooling performance of the prototype devices based on $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ /epoxy thick films were measured by a homemade apparatus. Currents generated from a DC power supply (Gwinstek, GPD-2303S) were applied on the prototype device, and the temperatures of hot side and cold one of TE legs were monitored by two high accuracy resistance temperature detectors (RTD).

3. Results and discussion

3.1. Structural evolution of $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ /epoxy thick films during hot-pressing curing process

To determine the temperatures of the curing reaction and decomposition of epoxy resin, the DSC and TG curves of the epoxy resin matrix and $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ /epoxy composites were measured and the results are displayed in Fig. 3. The excess diluent was added into the $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ /epoxy slurries in order to decrease the viscosity. From the DSC curve (solid line) of the epoxy resin matrix, it can be seen that the largest exothermic peaks occur nearby the range of 375–410 K, originating from the curing reaction between the epoxy resin and anhydride hardener catalyzed by imidazole derivative [25]. The endothermic peak

Download English Version:

<https://daneshyari.com/en/article/7952517>

Download Persian Version:

<https://daneshyari.com/article/7952517>

[Daneshyari.com](https://daneshyari.com)