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High-performance and high-stability bismuth selenide core thermoelectric fibers

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1. Introduction

Thermoelectric (TE) materials are considered as critical components for electrical power generation devices and solid state TE cooling [1]. The performance of TE materials is quantified by a dimensionless figure of merit, ZT, which is defined as $S^2 \sigma T/\kappa$, where S is the Seebeck coefficient, σ is the electrical conductivity, κ is the thermal conductivity, and *T* is the absolute temperature [2]. It has been proved that low-dimensional TE materials have large ZT [3,4], which provides a direction of fabricating high-performance TE materials. Low-dimensional Bi-based materials are the most promising TE materials. And many approaches have been used to synthesize low-dimensional Bi-based TE materials, such as solvothermal method [5] and melt spinning method [6]. However, these methods have complicated experimental process, harsh conditions, and low yield, suggesting it is difficult to synthesize lowdimensional TE materials efficiently. Furthermore, the Bi-based TE materials often have poor stability due to the lack of effective protection, which will limit their application [7]. Recently, many

ABSTRACT

Bismuth selenide exhibits high thermoelectric performance, which is a promising candidate for thermalelectrical energy conversion. Here, Bi_2Se_3 core thermoelectric fibers with K9 glass cladding were fabricated by a molten core drawing method. The 50-µm-diameter Bi_2Se_3 core fibers exhibit an ultrahigh Seebeck coefficient of $-150.85 \,\mu$ V/K. In addition, it has a high dimensionless figure of merit of 0.18 (at 300 K) and a long-term stability in air. The results indicate that the drawing approach is an effective way to fabricate high-performance and high-stability thermoelectric fibers, which will have potential application in fiber-integrated thermoelectric devices.

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functional materials have been incorporated into glass fibers by using the molten core drawing approach [8,9]. This approach opens the door to fabricate very long TE materials effectively, featured high environment stability with the protection of a stable glass cladding. In this work, Bi₂Se₃ core fibers were successfully prepared by using the molten core drawing approach. They have enhanced Seebeck coefficient and reduced thermal conductivity. Furthermore, the fibers exhibit outstanding stability in air. This work paves an effective way to fabricate high-performance and high-stability TE fibers.

2. Experiments

 Bi_2Se_3 powder of 99.99% purity was filled into a commercial K9 glass tube. After the powder was filled, the other end of the preform was also closed under N₂ atmosphere. Finally, the preform was drawn into fibers at 840 °C by using a fiber drawing tower. Electro-probe micro-analyzer (EPMA-1600, Shimadzu) equipped with a wavelength-dispersive X-ray spectrometer (WDS) was used to study the distribution of elements. The crystalline phase of the core was identified by X-ray powder diffractometer (XRD, X'Pert PROX, Cu K_{α}). The micro-Raman spectra were collected on the core using a Renishaw RM2000 instrument. The fibers were cut into 4 cm length and the voltage differences between two ends of them were obtained by a digital source meter. Furthermore, some centimeter order Bi_2Se_3 cores were obtained by etching the fiber glass







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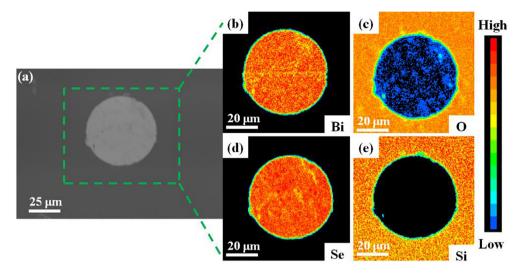


Fig. 1. (a) Electron image of the polished Bi₂Se₃ core fiber. (b)-(e) The WDS mapping images of the marked area in (a).

cladding for the measurement of thermal conductivity. Details of the measuring techniques can be found in the previous study [9,10].

3. Results and discussion

Fig. 1(a) shows the electron image of the Bi_2Se_3 core fiber. It can be seen that the core-clad structure of the fiber is preserved completely. The fiber has an outer diameter of 580 µm and inner diameter of 50 µm. Elemental analysis was performed on the crosssection of the Bi_2Se_3 core fiber to determine the extent of the elemental diffusion. Fig. 1(b)–(e) show the EPMA images of the Bi_2Se_3 core fiber. The distribution of elements O and Si are mainly in the glass-cladding region. Meanwhile, elements Se and Bi are mainly distributed in the core region.

Fig. 2 shows the elemental profiles (Bi, Se, O, Si) across the fiber core. The core possesses a composition of 61.1Bi-38.0Se (wt%). There is a little diffusion of Bi and Se into the cladding region. In addition, the diffusion of oxygen is 0.9 wt% in the Bi₂Se₃ core, which is much lower than that in In₄Se₃ core TE fibers (5 wt%) [11]. It could be attributed to the better thermal stability of Bi₂Se₃ than In₄Se₃.

The XRD patterns of the Bi_2Se_3 core and the Bi_2Se_3 powder are shown in Fig. 3(a). Both of them are crystalline. The patterns of them are well-indexed to the rhombohedral Bi_2Se_3 phase

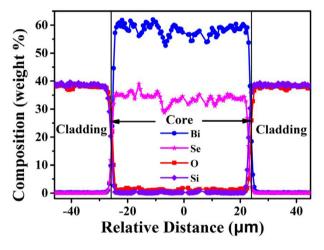


Fig. 2. Elemental profiles for the Bi₂Se₃ core fiber.

(JCPDS#330214), indicating that crystalline Bi_2Se_3 core fibers were obtained. Fig. 3(b) provides the Raman spectra for the Bi_2Se_3 core and the Bi_2Se_3 powder. Three peaks centered at 71, 131, 172 cm⁻¹ are observed from the Bi_2Se_3 powder, which can be attributed to the A_{1g}^1 , E_g^2 , and A_{1g}^2 Raman active modes of Bi_2Se_3 [12], respectively. Similarly, the A_{1g}^1 , E_g^2 , and A_{1g}^2 modes also can

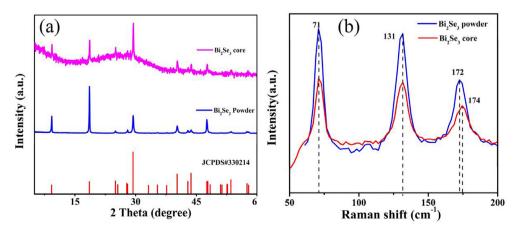


Fig. 3. (a) XRD patterns of Bi₂Se₃ core and the Bi₂Se₃ powder. (b) Raman spectra of Bi₂Se₃ core and the Bi₂Se₃ powder.

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