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Enhancing the thermoelectric property of Bi_2Te_3 through a facile design of interfacial phonon scattering

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ABSTRACT

From the perspective of green development, the pursuit of more facile and effective method toward improving thermoelectric efficiency of Bismuth Telluride (Bi_2Te_3) and its alloys has long been attractive. Recent researches indicate that the phonon scattering effect emerging on the nano-grain boundaries or the interfaces in composites can lead to remarkably improved ZT value. Inspired by such methodology to design effective interfacial phonon scattering, we have succeed in fabricating a novel thermoelectric composites based on conducting bismuth telluride and insulating calcium silicate (Bi_2Te_3-$CaSiO_3$) via constructing multiple interfaces. It is found that a low thermal conductivity is achieved coupling with high electrical conductivity and Seebeck coefficient through adjusting the synergetic effects between Bi_2Te_3 and $CaSiO_3$ interfaces. A high ZT value (0.72) is obtained at relatively high temperature of 373 K for the composites with 85 wt% Bi_2Te_3, which is much higher than that of the bulk Bi_2Te_3 (~0.59). Accordingly, since only materials of industrial grade were adopted, this work would also provide solid evidence for industrial fabrication of Bi_2Te_3 based TE materials toward low cost, high performance and large scale commodities.

1. Introduction

Thermoelectric (TE) material, characterized by the ability to realize direct energy conversion between heat and electricity, has drawn significant attention in recent years due to ever-growing concern over the environment and sustainability issues associated with traditional fossil fuel resources [1–7]. Combined with merits such as low thermal conductivity, relatively high electrical conductivity and tunable Seebeck coefficient, Bismuth Telluride (Bi_2Te_3) has long been regarded as the front runner in TE field [8–13]. Thanks to the tremendous efforts made by researchers in the last few decades, Bi_2Te_3-based TE materials have dominated the small-scale cooling components and power generators for ambient temperature in industry. Unfortunately, the dimensionless figure of merit (ZT) value of commercial Bi_2Te_3 remains to be improved in order to meet the growing demand of high-performance applications. More specifically, the state-of-the-art Bi_2Te_3 materials only have a ZT value around 1 at room temperature, producing great limitation on the fabrication of TE device with superior performance [14–16].

The energy conversion efficiency of TE materials is determined by the ZT value, which is defined as $ZT = S^2\sigma T/\kappa$, where S, σ, and κ represent the Seebeck coefficient, the electrical conductivity and the thermal conductivity (a sum of the electronic κ_{elec} and lattice κ_{lat} vibrations), respectively. The development of TE material with superior property is embarrassed because of the well-known interdependence of S, σ and κ complicated efforts [17]. According to the researches presented in last decade, representative methods such as designing nano-structure or compositing with advanced components have been proved effective in regulating and enhancing the ZT values by decoupling the independence among the TE properties [18–22]. Thus, great importance have been attached to the development of new-type Bi_2Te_3, including Bi_2Te_3 superlattices [23,24], Bi_2Te_3 nano-wires/rods [25–27], Bi_2Te_3 doped with metal elements [28–32], Bi_2Te_3 nanoparticles and nanoflakes [33–35], inorganic and organic hybrid Bi_2Te_3 nanocomposites [36–40]. By virtue of the phonon scattering effect at the nano-grain boundaries or interfaces between different phases, most of these strategies have succeeded in achieving a high-power factor (electrical transport properties) and reduced lattice thermal

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conductivity simultaneously without a severe sacrifice of electrical conductivity. Furthermore, the boundaries in composites are capable of increasing the Seebeck coefficient by means of carrier-filtering or quantum confinement [23,24,26,35,41,42]. However, efficient exploitation of waste heat for power generation requires inexpensive and scalable TE materials from an economic perspective. Most of Bi_2Te_3 forms are difficult for large-scale or industrial applications because of complex fabrication process and high costs [43,44]. Herein, proposing new routes for more facile fabrication of Bi_2Te_3 based TE materials toward industrial application remains a great challenge.

Driven by the fascinating effect originating from nano-boundaries and phase interfaces, doping thermally insulating components into Bi_2Te_3 matrix toward structuring multiple interfaces may be potential solution to developing high-performance TE composites. Moreover, these materials could be endowed with low intrinsic thermal conductivity and moderate electrical conductivity due to the maintenance of conductive pathways. On the other hand, by means of the interfacial scattering effect, namely the preferable scattering of phonons and low energy charge carries compared with their counterparts (energy filtering) at the interfaces, the Seebeck coefficient can be kept at fairly high value from existing literatures [45–49]. Besides, thermal insulating materials possesses other advantages [50] such as abundance, non-toxicity, and easy processability. Given its thermal and electrical insulating characteristic, Calcium Silicate ($CaSiO_3$) can be ideal candidate to compositing Bi_2Te_3 for construction of multiple interfaces [51]. Inspired by such design philosophy, we disclose a facile and feasible route to fabricate a novel thermoelectric material based on Bi_2Te_3 and $CaSiO_3$. Particularly, since only raw materials of industrial grade were employed, this study carries extreme importance for industrial fabrication of TE materials. Compared with pristine Bi_2Te_3, the samples doped with $CaSiO_3$ perform decreased thermal conductivities and improved power factors, which indicates that we can favorably realize the separate adjusting of the thermal conductivity and electrical conductivity. Consequently, the Bi_2Te_3-$CaSiO_3$ composites show a peak ZT value (0.72) when 15 wt% $CaSiO_3$ was incorporated. This study intends to demonstrate the effectiveness of achieving high TE performance in Bi_2Te_3-$CaSiO_3$ composite by virtue of interfacial phonon scattering. Moreover, the easy processability as well as great potential in industrial production would provide an avenue for such Bi_2Te_3-$CaSiO_3$ composite to be applied as power generators.

2. Experimental

2.1. Materials

Commercial bismuth telluride (Bi_2Te_3) of industrial grade was provided by Sichuan Xinglong Company (Chengdu, China), and Calcium Silicate ($CaSiO_3$) was purchased from Chengdu Kelong Reagents (China).

2.2. Preparation of Bi_2Te_3- $CaSiO_3$ composites

Bi_2Te_3 and $CaSiO_3$ powders with different weight ratio were mixed together directly without any further purification, followed by grounding into uniformly fine composite powders in the glove box. The weight ratio of Bi_2Te_3 in the mixture is 67%, 80%, 85%, 91% and 100%. Thereafter, the hybrid powders were compressed into pellet-shaped samples with a size of $\Phi 10\,mm \times 2\,mm$ under 10 MPa at room temperature (as shown in Fig. 1). Due to the existence of some impurity substance in raw materials, the melting point of Bi_2Te_3 employed in this work is about 550 °C. Through orthogonal design of experiments, it is found that the best preparation condition is calcination at 558 °C for 10 min in the vacuum furnace.

2.3. Characterizations

X-ray diffraction (XRD) data was collected on a DX-1000 (Dandong Fangyuan Instrument Co. LTD, China) X-ray diffractometer

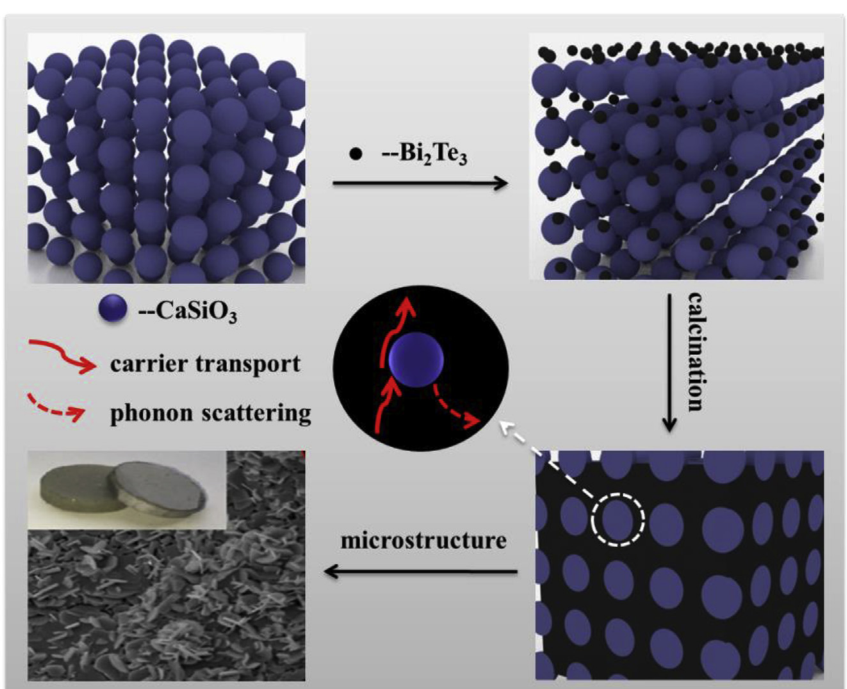

Fig. 1. Illustrations for the preparation process of Bi_2Te_3-$CaSiO_3$ composites.