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Microwave activated hot pressing: A new consolidation technique and its application to fine crystal bismuth telluride based compounds

Zhenzhou Rong, Xi'an Fan *, Fan Yang, Xinzhi Cai, Guangqiang Li

The State Key Laboratory of Refractories and Metallurgy, Wuhan University of Science and Technology, Wuhan 430081, China
Key Laboratory for Ferrous Metallurgy and Resources Utilization of Ministry of Education, Wuhan University of Science and Technology, Wuhan 430081, China

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ABSTRACT

In this work, a new consolidation technique of microwave activated hot pressing (MAHP) was recommended and some fine-grained and highly densified n-type $Bi_2Te_{2.85}Se_{0.15}$ compounds were prepared via mechanical alloying combining with MAHP process for the first time. The phase, microstructure and composition of the obtained samples were evaluated by X-ray diffraction, field emission scanning electron microscopy, and energy dispersive X-ray spectroscopy, respectively. The effect of MAHP processing parameters on the microstructure and thermoelectric (TE) properties of the $Bi_2Te_{2.85}Se_{0.15}$ compounds was investigated detailedly. The $Bi_2Te_{2.85}Se_{0.15}$ powders could be well compacted by the MAHP process and the relative densities of the as-MAHPed bulks were all above 95%. The grain size of the as-MAHPed samples increased gradually with raising sintering temperature and a partially oriented lamellar structure formed at some regions when the sintering temperature was relatively high. The maximum *ZT* value of 0.57 was obtained for the as-MAHPed $Bi_2Te_{2.85}Se_{0.15}$ sample at 373 K, which should be ascribed to the sharply reduced thermal conductivity due to enhanced phonon scattering effect from the contribution of a large number of nanoscale and submicrometer pores dispersed in the matrix. All evidences about electrical and thermal transport properties suggested that the MAHP technique was a potential excellent method to obtain a fine microstructure and improve the TE properties. Different from the conventional hot pressing and microwave pressureless sintering techniques, the MAHP method introduced here was more suitable for practical industrial application due to its cost-saving, energy-saving and high efficiency.

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1. Introduction

Thermoelectric (TE) materials, which can achieve the direct conversion between electrical energy and thermal energy, have received extensive attention recently for their great potential applications in power generation and TE cooling [1–3]. As is well known, bismuth telluride (Bi_2Te_3) based compounds are known as the best commercially used TE materials at ambient temperature from 300 K to 500 K. Commercial Bi_2Te_3-based compounds are commonly prepared by unidirectional crystal growth methods, such as Bridgman and zone melting (ZM) techniques [4,5]. However, their low TE conversion efficiency and poor mechanical performance resulted from the weak Van der Waals bonding between Te(1)–Te(1) layers limit their further applications [6].

The TE conversion efficiency can be characterized by a dimensionless figure of merit, *ZT*, which is defined as $ZT = \alpha^2 T/\rho\kappa$, where α, ρ, T, κ and α^2/ρ are Seebeck coefficient, electrical resistivity, absolute temperature, and thermal conductivity and power factor (*PF*), respectively [1]. One focus of present studies in this field is the applications of nanotechnology and low-dimensional technology. Refining grain has been proved to be a quite feasible method to reduce thermal conductivity due to the increasing phonon scattering effect at the grain boundaries [7–10].

To overcome the poor mechanical performance of Bi_2Te_3-based compounds and improve their TE properties, powder metallurgy technique has been widely investigated and applied. Powder metallurgy, such as hot pressing (HP), hot isostatic pressing (HIP), spark plasma sintering (SPS) and equal channel angular extrusion (ECAE) can achieve simultaneously a fast heating rate and a relatively short dwelling time under high pressure [10–17]. These methods are confirmed to be helpful to control the grain growth and preserve the nanostructure. Poudel et al. broke through the stagnant *ZT* of 1 and achieved a peak *ZT* of 1.4 at 373 K for p-type nanocrystalline Bi–Sb–Te bulk alloys by mechanical alloying (MA) combined with HP process [10]. In our previous works [11–16], the Bi_2Te_3-based TE materials were prepared by MA-HP, MA-SPS or MA-ECAE processes, and maximum *ZT* values of 0.66 and 1.5 for n-type and p-type materials were obtained, respectively. However, expensive equipment, high energy consumption, and low production efficiency during ECAE, HP and SPS processes cannot be ignored.

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* Corresponding author at: P. O. Box 185#, School of Materials and Metallurgy, Wuhan University of Science and Technology, 947 Heping Road, Qingshan District, Wuhan 430081, China. Tel.: +86 27 68862018.
E-mail address: groupfxa@163.com (X. Fan).

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Microwave heating has been widely used for the consolidation of ceramics, cemented carbides, ferrites, etc. Compared with conventional sintering, the advantages of microwave sintering can be concluded as higher energy efficiency, higher sintering density, lower sintering temperature, shorter sintering time, reduced activation energy, etc. [18–20].

Recently, Kim-Hak et al. obtained a p-type Bi_2Te_3 based bulk with a relative density of 85.6% and a maximum *PF* of 2.9×10^{-3} WK^{-2} m^{-1} by microwave pressureless sintering in a specially designed multimode cavity under a nitrogen atmosphere [21]. In the work of Delaizir et al. [22], the TE properties of p-type $Bi_{0.49}Sb_{1.51}Te_3$ compounds prepared by microwave pressureless sintering, HIP and SPS techniques were compared. The maximum compactness was only ~90% for the alloys prepared by microwave pressureless sintering, while the almost full densities (≥97%) were achieved for the samples by SPS and HIP techniques. The *ZT* values of p-type $Bi_{0.49}Sb_{1.51}Te_3$ alloys prepared by microwave pressureless sintering, SPS and HIP were 0.68, 0.7, and 0.53, respectively.

Generally speaking, the relative density of bulk sample prepared by pressureless sintering is much lower than that of bulk prepared by pressure-assisted sintering process under the same sintering conditions. Compared with pressureless sintering, pressure-assisted sintering can significantly achieve lower sintering temperature, shorter holding time, higher compactness, smaller porosity, better mechanical properties, etc. Therefore, on the basis of microwave pressureless sintering technique, a microwave activated hot pressing (MAHP) technique was developed independently in our lab for the first time. A sectional view of the mold is shown in Fig. 1. Similar to the traditional method of hot pressing, a uniaxial pressure is applied during the whole microwave heating process, which presents the combined merits of HP and microwave heating. Because microwave heating and uniaxial pressure are simultaneously applied in the MAHP process, they can improve compacting process such as improving powders contact, powders flow and elements diffusion. The main advantage of this technique is the creation of a dense material with a fine microstructure as a result of shorter holding time at lower sintering temperature. In the present work, the n-type $Bi_2Te_{2.85}Se_{0.15}$ bulks were prepared via MA-MAHP method for the first time. The effect of sintering temperature of the MAHP process on the microstructure and TE properties of $Bi_2Te_{2.85}Se_{0.15}$ alloys was investigated detailedly. The results of the as-HPed $Bi_2Te_{2.85}Se_{0.15}$ bulks in our previous work [12] are compared with the TE properties of the as-MAHPed samples.

2. Materials and methods

According to the nominal composition of $Bi_2Te_{2.85}Se_{0.15}$, high purity powders (99.99 wt.%, 200 mesh) of Bi, Se, and Te elements were weighed and loaded into a planetary ball mill (QM-4F, Nanjing Nanda Instrument) to MA. The weight ratio of ball to powder was 15:1, and the process was conducted at a speed of 400 rpm for 10 h under argon atmosphere. The speed ratio of revolution to rotation is 1:2. Stainless steel pot and balls were used and the inner radius of the milling pot is 80 mm. Subsequently, the as-MAed powders were loaded into a graphite die with an inner diameter of 20 mm, which was surrounded by a circle SiC mold, as shown in Fig. 1. The consolidation process was performed in a MAHP equipment (homemade device). The input microwave frequency was fixed at 2.45 GHz, which is the most commonly used frequency in household purposes and is available worldwide. The

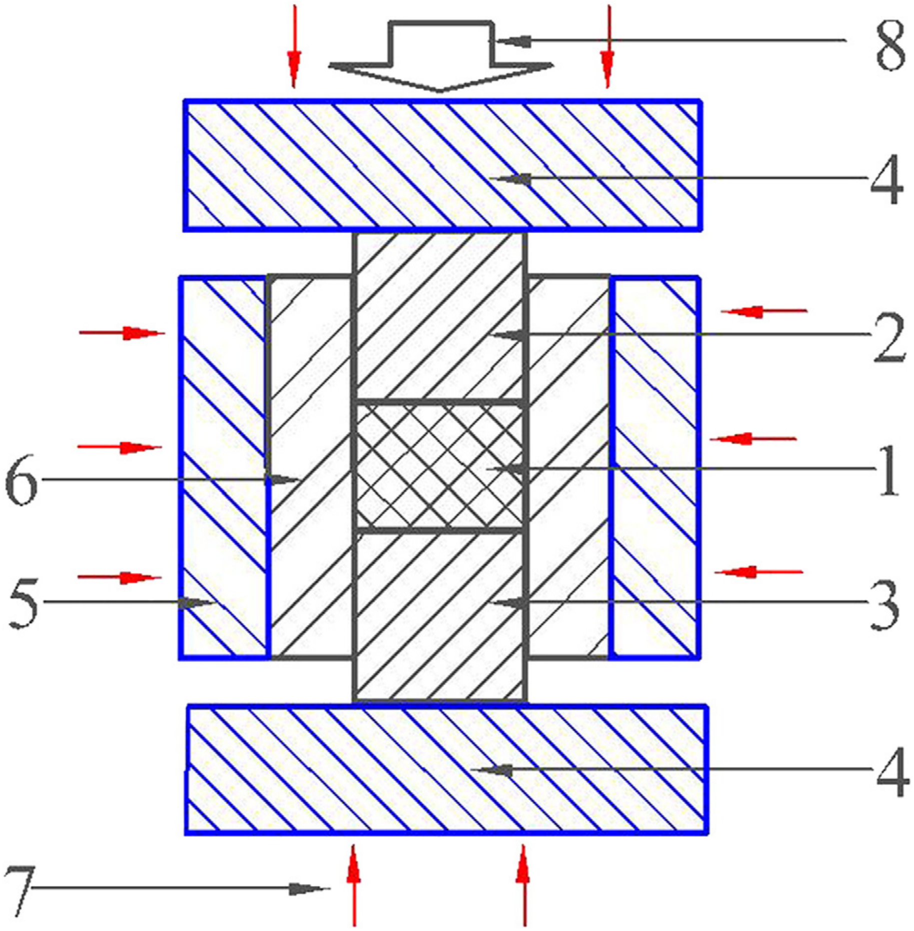

Fig. 1. The sectional view of the mold: 1—pre-sintered powders, 2—top graphite indenter, 3—bottom graphite indenter, 4—SiC gasket, 5—SiC mold, 6—graphite die, 7—input microwave, and 8—uniaxial pressure.

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