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Design and Optimization of Gradient Interface of *p*-Type $Ba_{0.3}In_{0.3}FeCo_3Sb_{12}/Bi_{0.48}Sb_{1.52}Te_3$ Thermoelectric Materials

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A series of *p*-type $Ba_{0.3}In_{0.3}FeCo_3Sb_{12}/Bi_{0.48}Sb_{1.52}Te_3$ (FS/BT) thermoelectric (TE) materials containing one gradient layer (1GL) with FS/BT volume ratio of 5:5, 3GLs-I with 7:3–5:5–3:7, 3GLs-II with 3:7–5:5–7:3, and 3GLs-III with 3:7–5:5–3:7 from FS to BT were fabricated by a two-step spark plasma sintering method. The interface structure and mechanical properties of the *p*-type FS/BT TE materials were investigated in this work. Designing the GLs at the interface of FS and BT bulk TE materials can effectively relax the thermal stress induced by the large difference in thermal expansion coefficient and eliminate the macroscopic cracks that occur in FS/BT TE materials with no GL, hence resulting in a remarkable enhancement in the interface mechanical properties of the FS/BT TE materials with the GLs. The optimized gradient interface of the FS/BT TE materials is 3GLs-II with FS/BT volume ratio of 3:7–5:5–7:3. The highest flexural strength of the 3GLs-II sample reached 13.68 MPa, increased by 116%.

Key words: $Ba_{0.3}In_{0.3}FeCo_3Sb_{12}/Bi_{0.48}Sb_{1.52}Te_3$ thermoelectric materials, gradient interface, microstructure, mechanical properties

INTRODUCTION

Solid-state thermoelectric (TE) devices are new energy devices which can convert heat directly into electricity and vice versa. TE devices have many advantages, such as no moving parts, environmental friendliness, reliability, and so on, and have been successfully used in space power generation and a variety of cooling applications.[1,2] However, there is still a long way to large-scale applications for TE devices because of their low conversion efficiency.[3] Extending the working temperature range of TE devices is considered as an effective way to increase their efficiency. Use of segmented TE modules, composed of different TE materials, has been confirmed to enable achievement of high TE efficiency over a wide temperature range.[4] Among various state-of-the-art TE materials, Bi_2Te_3-based alloys are the most widely used materials for TE coolers and power generation in the low temperature range of 300 K to 500 K,[5,6] whereas filled skutterudites are identified as some of the candidates for industrial power generation applications in the intermediate temperature range of 500 K to 800 K.[7] In recent years, several groups have reported segmented $Bi_2Te_3/CoSb_3$ TE devices with high efficiency.[8–10] However, as a key technology for fabricating segmented TE devices, the joining of two kinds of TE materials is still an obstacle. Müller et al.[11] found delamination cracks in the $FeSi_2$ region near the interface when Ni was selected as a joint interface for $Bi_2Te_3/FeSi_2$. Snowden et al.[12] discovered that the sample separated at the interface when Mo was selected as a joint interface for Zn_4Sb_3 and $(Bi,Sb)_2(Te,Se)_3$, and the same bulk materials exhibited cracks when Fe was selected as the joint interface. The mismatch of the thermal expansion coefficient between two kinds of TE materials might produce large interface thermal stress and result in macroscopic cracks or even interface fracture. According to functional gradient material (FGM) theory, a

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