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Simultaneous enhancement of mechanical and thermoelectric properties of polycrystalline magnesium silicide with conductive glass inclusion

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Abstract

Thermal fatigue and mechanical failure are two challenging aspects of thermoelectric (TE) module design and fabrication. Among the well-known TE materials with high conversion efficiencies are various brittle semiconductors or ceramics. However, formation of micro-cracks during fabrication and mechanical failure due to the high stress response in the course of thermal cycles are commonly observed in such material systems. In this work, we report the results of a novel technique to improve the mechanical and TE properties of magnesium silicide (Mg_2Si) via addition of a small quantity (0.25–1 vol.%) of conductive glass-frit. Mechanically alloyed and hot-pressed Mg_2Si specimens separately doped with 2 at.% Bi and 2 at.% Al were sintered at 1173 and 1123 K, respectively. The TE properties of both compounds were characterized by measurements of electrical resistivity (ρ), Seebeck coefficient (S) and thermal conductivity (κ) in the temperature range 300–970 K. The beneficial effects of addition of a minuscule quantity of Mg–Si–B–R-based (R = rare earth) conductive glass-frit to Al-doped Mg_2Si samples were investigated. Both Al-doped and Bi-doped Mg_2Si specimen were tested for mechanical reliability using diametric compression tests. Power factors times temperature ($S^2\sigma T$) of $>2\ \mathrm{W\,m^{-1}\,K^{-1}}$ were obtained from Al-doped samples containing conductive glass-frit. It was also found that addition of 1% of the conductive glass-frit results in significant improvement of the mechanical properties of Mg_2Si by eliminating microcracks in the brittle Mg_2Si system. Nearly 150% improvement was observed in the mechanical strength of the Al-doped samples reinforced with conductive glass-frit as compared to the samples without glass-frit.

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1. Introduction

Energy harvesting by thermoelectric (TE) devices has gained prominence in the search for alternative energy resources due to the many advantages such devices offer, such as low cost, low maintenance, small size, zero emissions and high reliability. Although refrigeration via TE devices has been widely implemented, the pursuit of high-performance power generation devices has recently attracted much attention. In fact, with the invention of new high-efficiency TE materials, the TE market continues to grow and now includes power generation, cooling, and sensing and imaging applications. Accordingly, new generations of TE devices [1–6] and characterization techniques [7–11] are also being developed. New materials based on nano bulk forms, such as nanocomposites and nanostructured single-component bulk materials, have attracted particular attention due to their ease of fabrication and compatibility with existing forms of TE devices [12–15]. Although bulk nanostructuring methods have been proven to be beneficial for improving TE properties [13,16–18],

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they sometimes result in excessive material defects that can deteriorate the mechanical properties of these materials.

The power generation efficiency of TE materials is based on numerous factors such as high electrical conductivity, high Seebeck coefficient and low thermal conductivity in order to maximize the dimensionless figure of merit, ZT, defined by [19]:

$$ZT = \frac{\sigma S^2}{\kappa_e + \kappa_l} T. \tag{1}$$

In Eq. (1), σ is the electrical conductivity, S is the Seebeck coefficient, κ_e and κ_l are the electronic and lattice contributions, respectively, to the total thermal conductivity, and T is the absolute temperature. TE materials generate voltage due to a thermal gradient applied across the TE material or wire. Therefore, TE devices are continually subjected to high levels of thermal stress during operational cycles. Consequently, in addition to high electrical and low thermal conductivities, the materials are expected to possess high mechanical strength and fracture resistance in order to sustain a large number of thermal cycles over the entire lifetime of the devices. The mechanical reliability is primarily an end result of the material characteristics as well as the fabrication technique. The requirement for low thermal conductivity in TE materials in order to maximize the ZT implies that the thermal resistance (R_T) of the material is expected to be high. However, the thermal resistance is a function of the tensile strength and is directly proportional to the fracture toughness via Eqs. (2) and (3), respectively. In Eqs. (2) and (3), S_T is the tensile strength, v is the Poisson's ratio, κ is the total thermal conductivity, α_T is the thermal expansion coefficient, E is the elastic modulus, K_C is the fracture toughness, Y is the crack shape factor dependent on the sample shape and c is the Griffith flaw size [20]. Therefore, the mechanical strength is directly related to the thermal resistance (R_T) via the Griffith criterion defined in Eq. (3) for brittle materials [21].

$$R_T = \frac{S_T(1-v)\kappa}{\alpha_T E}, \tag{2}$$

$$S_T = \frac{K_C}{Y\sqrt{\pi c}}. \tag{3}$$

For highly brittle materials, the brittleness index (B) relates to the fracture toughness (K_C) and hardness (H) as per Eq. (4) [22,23]. The brittleness index is directly proportional to the hardness of the material, indicating a need for mechanically stable materials with high fracture resistance for better mechanical reliability. A higher K_C in brittle materials implies a higher resistance to growth of microcracks in the specimen.

$$B = \frac{H}{K_C}. \tag{4}$$

Fatigue performance is known to be a result of the presence of residual stresses introduced during the processing of the specimen. However, in the absence of residual stresses, fatigue is caused due to microstructural features and thermal cycling of the material [24]. Formation of microcracks is a known phenomenon in TE samples sintered by means of spark plasma sintering (SPS) technique [25,26]. Microcracking during heating and cooling cycles was reported as a result of thermal fatigue in materials such as lead–antimony–silver–tellurium alloy fabricated via casting and hot-pressing techniques [27]. Similarly, techniques such as inclusion of nanoparticles or compositional inhomogenities aimed at reducing thermal conductivity in TE materials are also known to result in mismatches in thermal expansion coefficients of the inclusions and the host matrix, leading to microcracking [28]. Many such materials systems have been reported to show high stress responses to thermal transients owing to their high thermal expansion coefficients [29–31]. It is known that in microcracked samples both the electrical conductivity and thermal conductivity are dependent on the crack damage parameter, which in turn is a function of the volumetric number of density of microcracks and the crack size distribution moment [29]. Deterioration in electrical properties is also associated with thermomechanical stresses in TE modules and is reported to result in a significant decrease in the figure of merit by 97% after 45,000 thermal cycles [32]. Such degradation in performance and mechanical instability of TE modules was described as being chiefly a consequence of the formation of defects in the materials. It is known that physical properties such as Young's modulus, hardness and fracture toughness are functions of microcracking and porosity [29]. The effects of mechanical deformation on the TE properties of p-type $(Bi_{0.225}Sb_{0.775})_2Te_3$ were also reported by Jung et al. [33].

Several fabrication techniques have been investigated as means of improving the mechanical reliability of TE devices. Qi et al. reported that melt spinning followed by a quick SPS resulted in improved mechanical properties of nanostructured β-Zn_4Sb_3. An improvement of 130% in the compressive strength was demonstrated for the nanostructured sample compared to that of the ingot [34]. Smitchz et al. highlighted that tubular designs of TE modules also help in the distribution of tensile and compressive stresses, resulting in better mechanical properties [30]. TE sample synthesis via quenching was also reported to eliminate microcracks in the two-phase alloy $Cu_{0.2}Ag_{2.8}SbSeTe_2$ [35]. Improved TE and mechanical properties were shown in melt-quenched PbTe and PbTe-$Ge_{1-x}Si_x$ eutectic and hypereutectic composites [36]. Angular extrusion techniques applied to p-type $Bi_{0.4}Sb_{1.6}Te_3$ TE material also revealed improvement in the TE as well as the mechanical properties [37].

Various TE materials have been recognized as appropriate for a wide range of temperatures [19]. Among the prominent medium-temperature (500–800 K) TE material systems, brittle semiconductors such as alloys of Mg and Si are known to possess many beneficial properties such as light weight, non-toxicity of the constituent elements, abundance in nature and suitability for recycling [38–40]. However, they are also characterized by poor mechanical

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