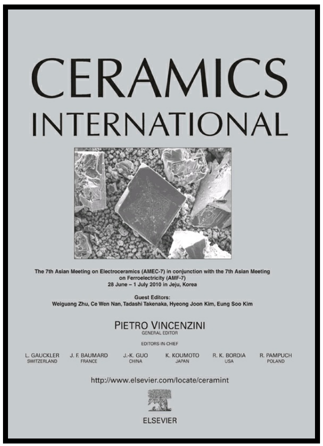

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Effect of Zn migration on the thermoelectric properties of Zn_4Sb_3 material

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Abstract:

β-Zn_4Sb_3 is interesting as thermoelectric material at moderate temperature due to the extreme low thermal conduction. Recent success in energy band engineering or nano-engineering led to a significant improvement in the thermoelectric properties of β-Zn_4Sb_3. In this work, we utilize the direct current to drive the migration of Zn by designing of sintering mould. Obvious Zn migration under the direct current applied in the plasma activated sintering (PAS) process is found in Zn_4Sb_3 compounds, and Zn exhibits significantly heterogeneous gradient composition distribution. At the top of sample, the single-phase Zn_4Sb_3 decomposes into ZnSb phase because of the loss of Zn, while Zn originated from lattice and interstitial sites in Zn_4Sb_3 is abundant in the bottom. The temperature-dependent transport measurements are also carried out at 323–673 K. Zn migration has a huge influence on the thermoelectric properties because of the sensitivity of Zn_4Sb_3. The maximum power factor can reach ~1.44 $mWm^{-1}K^{-2}$ at 673 K due to the high Seebeck coefficient and low resistivity, which is one of the highest values in the reported results. The resulting peak ZT value of ~1.2 at 673 K is obtained. To control the Zn distribution by tuning the current is a feasible approach to improve the thermoelectric properties of Zn_4Sb_3 material.

Keywords：Zn migration; Zinc antimonide; Power factor; Thermoelectric properties

1 Introduction

Thermoelectric (TE) materials, which can directly convert heat into electricity and vice versa, have attracted significant interest for potential applications in power generation and solid-state refrigeration [1–4]. The conversion efficiency of a thermoelectric module is defined by a dimensionless figure of merit, i.e., $zT=S^2T/\rho k_{tot}=S^2T/\rho(k_{ele}+k_{lat})$, where S, T, ρ, k_{tot} are the Seebeck coefficient, absolute temperature, electrical resistivity and total thermal conductivity, respectively; k_{ele} and k_{lat} are the electronic and lattice contributions to the total thermal conductivity k_{tot}, and the k_{lat} is the most critical factor to the k_{tot}. Thus, a relatively high power factor (S^2/ρ) together with a significant reduction in the total thermal conductivity will contribute to the improved properties of thermoelectric devices [5–7]. However, the thermoelectric parameters S, ρ and k_{ele} are closely intertwined and interacted via carrier concentration [8,9].

The binary compound β-Zn_4Sb_3 is a promising thermoelectric material for moderate temperature applications with the peak ZT value of 1.3 at 673 K [10]. In the past years, high-performance Zn_4Sb_3-based compounds were achieved by introducing resonant states [11–13]. For instance, Wang et al. reported that Sm doping caused the resonant distortion of electronic density of states (DOS) of Zn_4Sb_3 compounds, and almost 2-fold increase in effective mass m^*, resulting in a ZT value of 1.1 at 615 K [12]. Numerous investigations shown that nano-structuring could effectively scatter phonons while having less effect on the electronic transport, and thus can improve thermoelectric properties of the materials [14–16]. The "bottom-up" nano-structured