



Oxide formation mechanism and its effect on the microstructure and thermoelectric properties of *p*-type Bi_{0.5}Sb_{1.5}Te₃ alloys

May Likha Lwin^a, Peyala Dharmiah^a, Babu Madavali^a, Chul-Hee Lee^a, Dong-won Shin^a, Gian Song^a, Kap-Ho Lee^b, Soon-Jik Hong^{a,*}

^a Division of Advanced Materials Engineering, Kongju National University, 275, Budae-dong, Cheonan City, Chungcheongnam-do, 330-717, Republic of Korea

^b Department of Materials Science & Engineering, Chungnam National University, Daejeon, Republic of Korea



ARTICLE INFO

Keywords:

Oxygen contamination
p-type Bi_{0.5}Sb_{1.5}Te₃
 Heat treatment
 Thermoelectric properties
 Microstructure
 Gas atomization

ABSTRACT

Bismuth antimony telluride based thermoelectric (TE) materials have been intensively developed and synthesized using different mechanisms, for the room temperature TE applications. In particular, bismuth antimony telluride based TE alloys are very sensitive to deviations in their composition, and to contamination during the materials synthesis. Oxygen contamination during Bi-Sb-Te based materials synthesis is one of the critical factors that alters or diminishes thermoelectric-transport properties. Thus, in this study, how the oxide formation mechanism on the powder surface and bulks of *p*-type Bi_{0.5}Sb_{1.5}Te₃ alloys affected the microstructural features and thermoelectric properties were elucidated quantitatively. While applying heat treatment (HT) to Bi_{0.5}Sb_{1.5}Te₃ powder, the constituent elements interacted with the ambient atmosphere and formed a new oxide phase which acted as a barrier to carrier transport. At the initial stage (300 °C) of heat treatment, only the powder surface was oxidized due to the reaction of outer surface atoms with atmospheric air and moisture. While increasing in temperature during HT, this surface oxygen contamination diffused further inside the powder through the grain boundaries. More diffusion and spreading occurred throughout the matrix at 450 °C. The increment of oxygen content from 0.05 to 0.82 wt% drastically decreased the electrical conductivity by 67%, and thermal conductivity by 7% at the heat treatment temperature of 450 °C. This reduction behavior is mainly due to severe scattering of the carriers/phonons at the new formation of oxide (Sb₂O₃) phase near grain boundaries and within the matrix. At a glance, a small increase in the oxygen content wouldn't significantly influence the thermoelectric properties; however, at a certain level of oxide formation (0.82 wt%), severe effects could occur due to the intensified scattering or trapping of carriers by the oxide barrier formation at the grain boundaries.

1. Introduction

Thermoelectric materials (TE) have become very attractive in relation to their power generation (Seebeck effect) and active refrigeration (Peltier effect) [1]. The efficiency of TE materials can be deduced by the dimensionless figure of merit (ZT), which is defined as the ratio of the power factor ($\sigma\alpha^2$) to the thermal conductivity (κ). Thus, a good TE material should exhibit high electrical conductivity with a high Seebeck coefficient, and low thermal conductivity. It should also minimize the Joule heating effect, to ensure the large potential/thermo-voltage across the junction and create a steep temperature gradient [1,2]. Managing all these parameters within a certain material can be achieved by doping [3], scattering from nanoscale endotaxial precipitation and mesoscale grain boundaries, atomic-scale alloy scattering [4], nanostructuring [5], quantum confinement, superlattices [6], and

creating nanocomposites [7]. As mentioned above, improving the thermoelectric properties of thermoelectric materials using powder metallurgy processes has given promising results.

On the other hand, the thermoelectric materials fabricated by powder metallurgy techniques such as gas-atomization [8,9], mechanical alloying [10], hot extrusion method, and hot-pressing [11,12] are commonly used for the fabrication of TE elements. However, there is a possibility of excessive oxidation of the final powder during fabrication or in the next process. To maintain high purity powder, the particles must be isolated from atmospheric oxidation or any contamination. It is known that reducing the powder using hydrogen can upgrade the powder characteristics because of a lower oxygen concentration. Even so, the reduction of powder using hydrogen or ideal gases does not markedly or perfectly affect the TE properties.

In particular, the control of the oxides in materials, or the oxygen

* Corresponding author.

E-mail address: hongsj@kongju.ac.kr (S.-J. Hong).

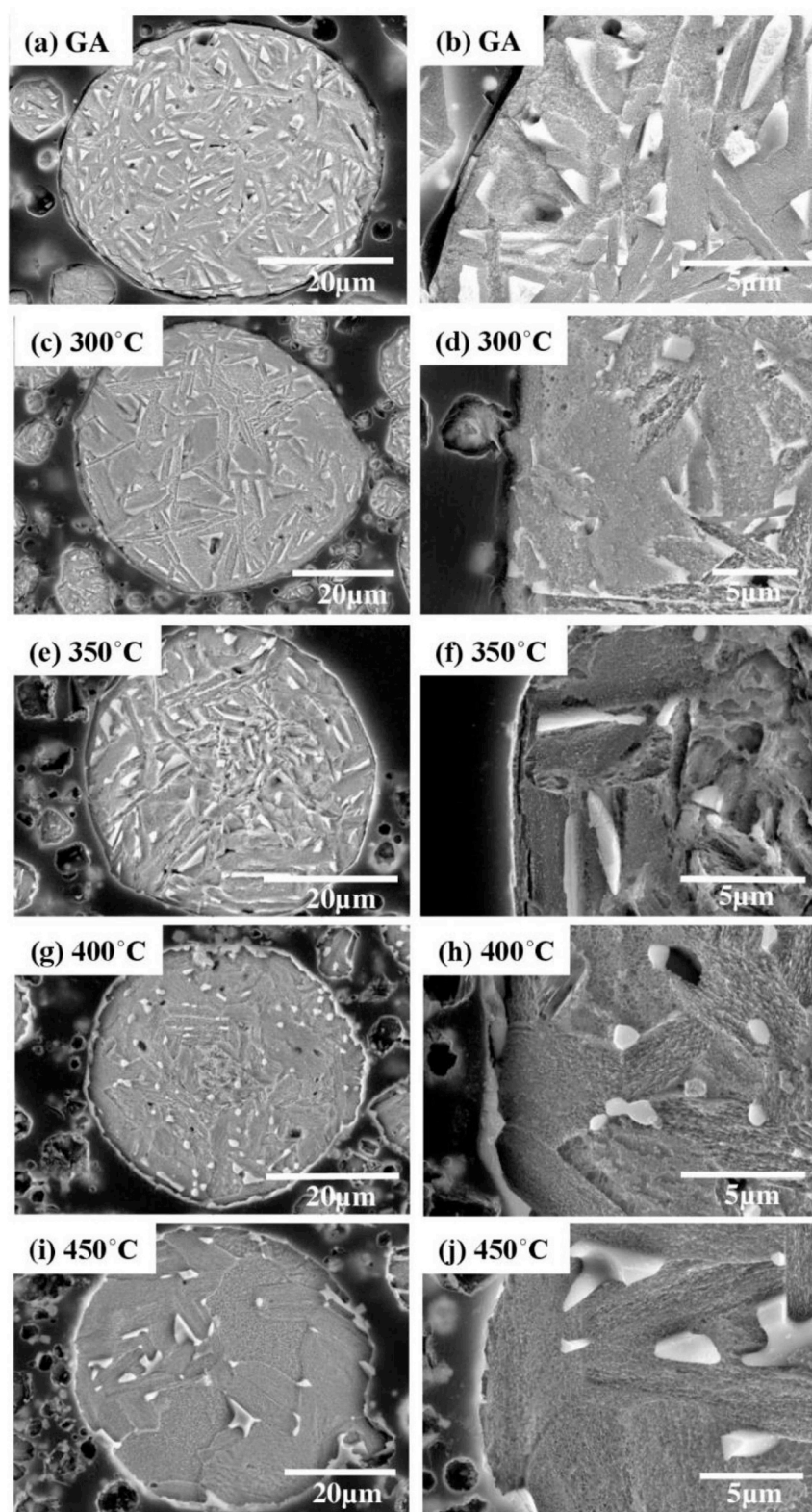


Fig. 1. SEM micrographs of heat-treated GA powder at different temperature held for 1 h: Low and high magnification SEM images of (a, b) GA powder, (c, d) GA powder heat treated at 300 °C, (e, f) GA powder heat treated at 350 °C, (g, h) GA powder heat treated at 400 °C, and (i, j) GA powder heat treated at 450 °C.

atoms present in bismuth telluride alloys, is considered essential because it influences their electrical properties. This is because atomic-scale defects such as vacancies and impurities affect carrier behavior [13]. M.W. Oh et al. reported that dissolved oxygen would affect the thermoelectric properties of $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ because the electrical

conductivity would be decreased by impurity scattering [14]. Lim et al. observed that the oxygen concentration was reduced after hydrogen reduction, but also that the Seebeck coefficient did not vary in spark plasma sintered $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ compounds [15].

In a few reports, it was suggested that the donor-like behavior of

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