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Synthesis and Characterization of Bismuth Telluride ($Bi_{0.48}Sb_{1.52}Te_3$) Single Crystals

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ABSTRACT

The objectives of this research are to study the effect of the crystal growth rate of $Bi_{0.48}Sb_{1.52}Te_3$ prepared by Bridgman method on its thermoelectric properties. Solid solutions with the composition of $Bi_{0.48}Sb_{1.52}Te_3$ have been grown using the crystal growth rates of 3.6, 6.0 and 12.0 mm/hr, respectively. The thermoelectric characterizations of samples was cut along the ingots of these solid solutions are carried out. The crystal growth rate and temperature measurement are responsible for thermoelectric properties. By increasing the temperature measurement, the figure of merit increased. The figure of merit decrease with increasing crystal growth rate. The crystal growth rate of 3.6 mm/hr exhibited a large figure of merit equal to 5.476 x 10^{-3} K^{-1} at 348 K. The best method may be used to fabricate thermoelectric cooling and power generator devices.

Keywords: Thermoelectric properties, Bridgman method, crystal growth rate

INTRODUCTION

The thermoelectric device can convert thermal energy from a temperature gradient into electric energy. This phenomenon was discovered in 1821 and it was called "Seebeck effect". As mentioned above, when a temperature differential is established between the hot and cold ends of the semiconductor materials, a voltage is generated. Based on this Seebeck effect, the thermoelectric devices can also act as power generators. Oppositely, thermoelectric devices can convert electrical energy into a temperature gradient. This phenomenon was discovered by Peltier in 1834. The application of this cooling or heating effect remained minimal until the semiconductor materials was applied. With the advent of semiconductor materials came the capability for a wide variety of practical thermoelectric refrigeration applications (Rowe, 1995).

Bi_2Te_3, Sb_2Te_3 and their solid solutions $(Bi, Sb)_2Te_3$ are of great interest for near-room temperature applications in thermoelectric cooling and thermoelectric generator devices. As a measure of the performance of a thermoelectric device, the figure of merit (Z) of the constituent thermoelectric material is defined by $Z = S^2\sigma/\kappa$, where S, σ and κ are the Seebeck coefficient, electrical conductivity and thermal conductivity, respectively. As conventional fabricating techniques of bismuth antimony telluride bulk compounds, there are single-crystal techniques due to Bridgman (Yamashita and Tomiyoshi, 2003 ; Ebisumori *et al*., 1998), Czochralski

(Zemskov *et al*., 2000) and Zone-melting (Iang *et al*., 2005) methods and powder metallurgy techniques such as hot-pressing (Kim *et al*., 2000; Kitagawa *et al*., 2001) and hot-extrusion (Seo *et al*., 2000) methods. The specimens prepared by Bridgman method were highest figure of merit. The composition of $(Bi_xSb_{1-x})_2Te_3$ had an effect on thermoelectric properties. From the review literature, it was found that x = 0.24 were highest figure of merit (Iang *et al*., 2005).

The classical Bridgman method consists of a slow lowering of a crucible containing the melt from the higher temperature zone, through the temperature gradient, into the lower temperature zone (Figure 1). As the crucible crosses the melting point between the zones, the melt directionally crystallizes and under certain conditions, may result in a single crystal (Rowe, 1995).

In the present study, the crystal growth rate by Bridgman method had an effect on the thermoelectric properties of $Bi_{0.48}Sb_{1.52}Te_3$.

EXPERIMENTAL PROCEDURE

Pure Bismuth (99.9%), Antimony (99.5%) and Telluride(99.8%) powder were weighed for the composition of $Bi_{0.8}Sb_{1.52}Te_3$ (Iang *et al*., 2005) and then mixed in planetary mill for 48 hrs under Ar atmosphere. The mixture was then put in the graphite crucible in vacuum tube to grow crystal growth in Bridgman furnace. The tube were evacuated to ~10^{-2} mbar and sealed off under 23 mbar with Ar gas. As shown in Fig.1, the crystal growth was then performed in Bridgman furnace at temperature gradient of 25°C/cm and the crystal growth rates adjusted are 3.6, 6.0 and 12.0 mm/hr, respectively.

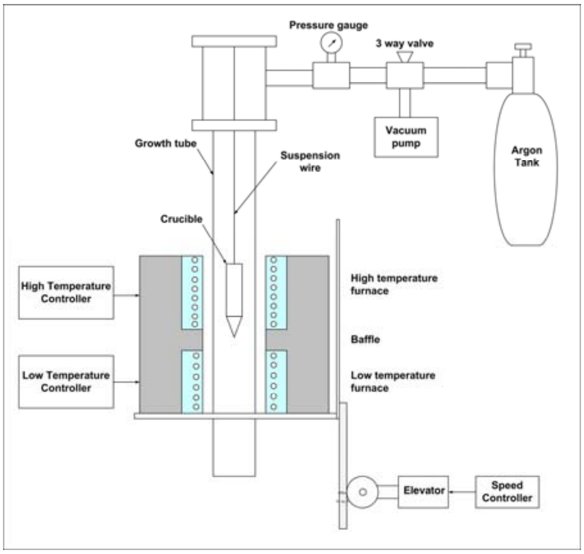

Figure 1 Diagram of the developed vertical Bridgman method.

The ingots were cut into a parallelepiped of 5x5x15 mm^3 and a square plate of 10x10x2 mm^3 from the central part of ingots, As shown in figure 2. The sample was subjected to Seebeck coefficient (S) and electrical resistivity (ρ) measured by ULVAC ZEM-2. Density (d) was measured by the Archimedes method. X-ray diffraction (XRD) was performed with an X'Pert PRO diffractometer (Philips) by using CuK_{α} radiation (λ = 1.5406 A^{o}). The specific heat capacity (C_p) and thermal diffusivity (α) were measured by a laser flash technique(ULVAC TC-7000H). The thermal conductivity (κ) was calculated assuming $\kappa = d*C_p*\alpha$. The figure of merit (Z) was determined according to the equation: $Z = S^2/(\rho\kappa)$

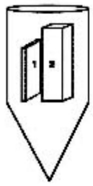

Figure 2 The ingots were cut into a parallel crystal growth direction.

RESULTS AND DISCUSSTION

Crystal orientation

Figure 3 shows the X-ray diffraction (XRD) patterns of ingots for different crystal growth rate by Bridgman method. All diffraction peaks of ingots are assigned to those of the Bi-Sb-Te type structure. The crystal growth rate at 3.6 mm/hr indicated high intensity of (0 1 5) planes, which are parallel to the c-axis. The crystal growth rate at 6.0 mm/hr indicated mainly intensity of (0 0 6) and (0 0 15) planes, which are perpendicular to the c-axis. The crystal growth rate at 12.0 mm/hr indicated patterns of Bi-Sb-Te and Sb, which the mainly patterns of Bi-Sb-Te are parallel to the c-axis. So that, the crystal growth rate had effect on planes of crystal growth.

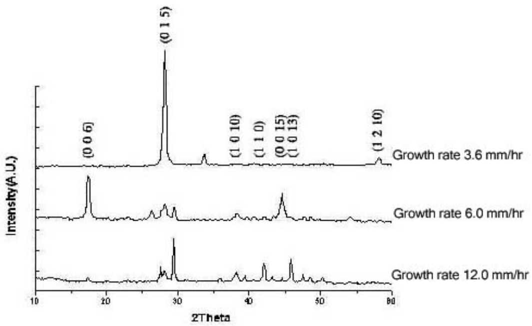

Figure 3 XRD patterns of the crystal growth by Bridgman method for different growth rate.

Thermoelectric properties

Figure 4 shows temperature dependence of Seebeck coefficient of $Bi_{0.48}Sb_{1.52}Te_3$. The values of the Seebeck coefficient are positive along the ingots varying from about 184 μV/K at the tip to 213 μV/K and then increase with increasing temperature measurement.

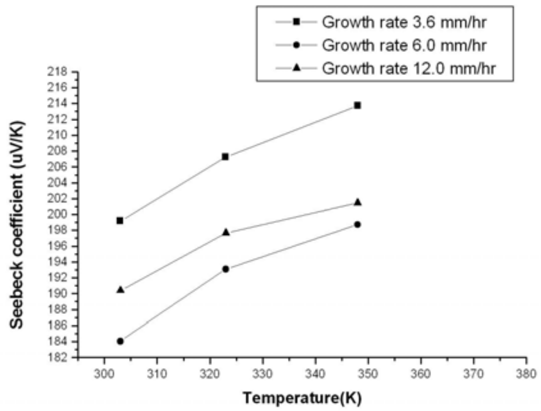

Figure 4 Temperature dependence of Seebeck coefficient of $Bi_{0.48}Sb_{1.52}Te_3$.

Figure 5 shows temperature dependence of electrical resistivity of $Bi_{0.48}Sb_{1.52}Te_3$, where the values increase with increasing crystal growth rate and temperature measurement.

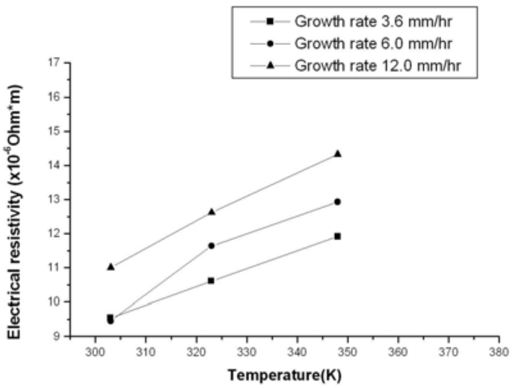

Figure 5 Temperature dependence of electrical resistivity of $Bi_{0.48}Sb_{1.52}Te_3$.

Figure 6 shows temperature dependence of thermal conductivity of $Bi_{0.48}Sb_{1.52}Te_3$. The values decrease with decreasing crystal growth rate and temperature measurement.

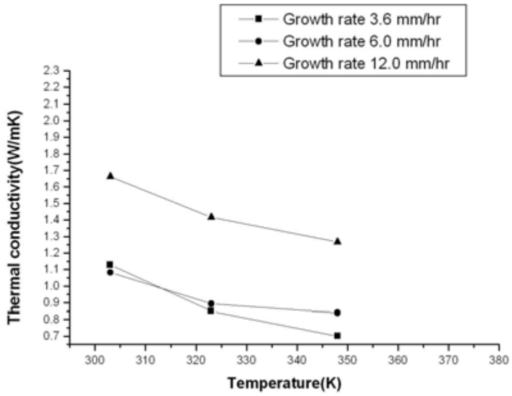

Figure 6 Temperature dependence of thermal conductivity of $Bi_{0.48}Sb_{1.52}Te_3$.

Figure 7 shows temperature dependence of figure of merit of $Bi_{0.48}Sb_{1.52}Te_3$. The values increase with decreasing crystal growth rate and increasing temperature measurement.

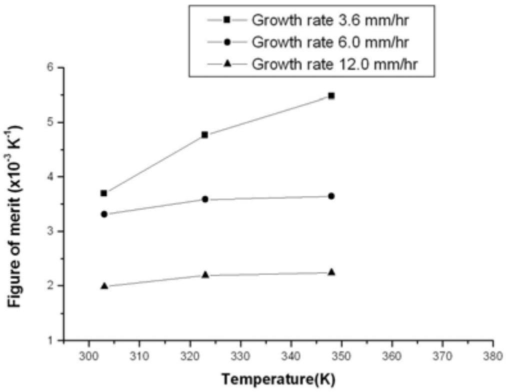

Figure 7 shows temperature dependence of figure of merit of $Bi_{0.48}Sb_{1.52}Te_3$.

The figure of merit depend on the single crystallizes, from XRD pattern. By the crystal growth rate of 3.6 mm/hr exhibited a large intensity of single plane, was highest the figure of merit and decrease with increasing the crystal growth rates. The figure of merit increased when temperature measurement increasing. The maximum the figure of merit values occur at relatively high temperature, suggesting that the materials are suitable for power generation (Ni *et al*., 2005).

CONCLUSION

P-type $Bi_{0.48}Sb_{1.52}Te_3$ crystals of various crystal growth rate (3.6, 6.0 and 12.0 mm/hr) were fabricated by the Bridgman method. The figure of merit increased with decreasing crystal growth rate and increasing temperature measurement. The maximum figure of merit value reached 5.476 x 10^{-3} K^{-1} at about 348 K, corresponding to the crystal growth rate of 3.6 mm/hr.

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