JOURNAL OF MATERIALS SCIENCE **40** (2005) 6439–6444

High-performance bismuth-telluride compounds with highly stable thermoelectric figure of merit

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The *p*-type $(Bi_{0.25}Sb_{0.75})_2Te_3$ ingot doped with 8 wt% excess Te alone and the *n*-type Bi_2 $(Te_{0.94}Se_{0.06})_3$ ingot codoped with 0.068 wt% I and 0.017 wt% Te were grown by the Bridgman method and annealed at 673 K for 5 h in a hydrogen stream. The electrical resistivity ρ, Seebeck coefficient α and thermal conductivity κ before and after annealing were measured at 298 K, so that the annealing degraded significantly *ZT* of the *p*-type specimen but enhanced remarkably that of the *n*-type one. The temperature dependences of ρ, α and κ of the as-grown *p*-type and annealed *n*-type specimens with higher *ZT* were investigated in the temperature range from 200 to 360 K. As a result, *ZT* values of the as-grown *p*-type and annealed *n*-type specimens have a broad peak and reached great values of 1.19 and 1.13 at approximately 320 K, respectively. The present materials were thus found to be far superior to any other bismuth-telluride compound in the thermal stability of energy conversion efficiency in addition to the high performance.

1. Introduction

Bismuth-telluride (Bi_2Te_3)is one of the best thermoelectric materials with the highest figure of merit ($ZT{=}\alpha^2T/\rho\kappa$) or the highest power factor ($P{=}\alpha^2/\rho$), where α is the Seebeck coefficient, ρ the electrical resistivity, κ the thermal conductivity and *T* the absolute temperature [1]. Whereas much effort has been made to raise *Z* of bulk materials based on Bi_2Te_3 by doping or alloying other elements in various fabricating processes, *ZT* was not much more than 1 untill recently. Indeed, so far, the highest *ZT* at room temperature was 1.06 in the *p*-type $(Bi_{0.25}Sb_{0.75})_2Te_3$ alloy doped with 4 wt% excess Te and 0.05 wt% Ge [2] and 1.75 wt% excess Se alone [3] and 0.96 in the *n*-type $(Bi_{0.25}Sb_{0.75})_2(Te_{0.95}Se_{0.05})_3$ alloy doped with 0.16 wt% SbI_3 [4]. Recently, the highest *ZT* of 1.14 at 300 K has been reported by Ettenberg *et al.* [5] for the *p*-type $(Bi_{0.25}Sb_{0.75})_2(Te_{0..97}Se_{0.03})_3$ alloy. However, one of us has most recently reported significantly high *ZT* of 1.19 at 298 K for the *n*-type Bi_2 $(Te_{0.94}Se_{0.06})_3$ doped with 0.068 wt% I and 0.017 wt% Te [6] by annealing the ingots prepared by Bridgman method and 1.41 at 308 K for the as-grown *p*-type $(Bi_{0.25}Sb_{0.75})_2Te_3$ alloy doped with 8 wt% excess Te alone [7], so that both *ZT* were able to exceed 1. This *n*-type specimen has a maximum *ZT* when annealed at 673 K for 5 h in a hydrogen stream. Recently the demand for the thermal stability of *ZT* in addition to the high performance has increased significantly, in order to stabilize the energy conversion efficiency. However, the temperature dependence of *ZT* of these *p*- and *n*-type specimens [6, 7] has not yet measured at temperatures below room temperature, which corresponds to the operating temperature of a Peltier module.

As conventional fabricating techniques of bismuth-telluride bulk compounds, there are single-crystal techniques due to Bridgman [4, 8], Czochralski [9] and zone-melting [8] (i.e., traveling heater [10]) methods and powder metallurgy techniques such as hot-pressing [11, 12] and hot-extrusion [11] methods. The specimens prepared by these methods were often subjected to the annealing in various atmospheres to raise *ZT* as high as possible[4, 6–8, 12]. However, the annealing has had either a favorable effect or an adverse effect on the improvement in *ZT* of bismuth telluride bulk compounds [4, 6–8, 12, 13]. Indeed, the annealing had a favorable effect on the improvement in *ZT* of the *n*-type specimen [6], but it has no effect on the *p*-type specimen [7]. The *p*- and *n*-type specimens used here were cut from the same ingots used for the previous high-performance specimens [6, 7], in order to investigate the temperature dependence

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DOI: 10.1007/s10853-005-1712-6

of ZT in the wide temperature range from 200 to 360 K.

The purpose of this study is to investigate the temperature dependence of ZT of the high-performance p- and n-type bismuth-telluride compounds, particularly in the temperature region important for a Peltier cooling, and to confirm whether the present materials are superior to other bismuth-telluride compounds in the thermal stability of ZT.

2. Experiments

The p-type $(Bi_{0.25}Sb_{0.75})_2Te_3$ ingot doped with 8 wt% excess Te alone and the n-type $Bi_2(Te_{0.94}Se_{0.06})_3$ ingot codoped with 0.068 wt% I and 0.017 wt% Te were grown by the Bridgman method, using purer Bi granules of 99.999% and pure Sb, Te, Se and I granules of 99.99% as starting materials. The materials were weighed out in appropriate atomic ratios, charged into a quartz tube of $\phi 30$ in inner diameter and melted in an evacuated quartz tube by an induction heating to make a homogeneous melt without segregation. The degree of vacuum was then of the order of 10^{-4} Pa. After melting, the compounds were vertically grown by the Bridgman method at a fast rate of 6 cm/h, to produce intentionally scattered second-phase precipitates in the ingot. It is close to one of various growth conditions reported by Yim and Rosi [4]. Naturally, the resulting ingots were not single crystals and consisted of relatively coarse grains with the cleavage planes aligned partially parallel to the freezing direction.

In order to investigate the thermoelectric properties of the as-grown ingots, a parallelepiped of $5\times5\times15$ mm^3 was cut from the central part of ingots with a total length of about 20 cm and a square plate of $10\times10\times3$ mm^3 was cut from the same central part of ingots, where the length of 15 mm and thickness of 3 mm were cut parallel to the freezing direction. Some of the former and latter specimens were annealed at 673 K for 5 h in a hydrogen stream, using purer hydrogen gas of 99.99%, which is an optimum annealing condition for the n-type specimen [6]. The former specimens were subjected to Seebeck coefficient and electrical resistivity measurements (Sinku-Riko, Inc., Model ZEM-1), and the latter ones to thermal conductivity after being cut into a disk of $\phi 10\times3$ mm. The Seebeck coefficient α, electrical resistivity ρ and thermal conductivity κ before and after annealing were measured at 298 K within an accuracy of 1, 2 and 2%, respectively. The resultant accuracy was about 5% as a thermoelectric figure of merit.

Subsequently, the temperature dependence of α and ρ was measured in the range from 200 to 360 K using the same type of apparatus, which makes it possible to observe the thermoelectric properties at temperatures below and above room temperature. In addition, the thermal conductivity κ was measured at intermittent temperatures in the range from 173 to 433 K. The microstructures of these materials before and after annealing were investigated by an electron probe microanalyzer (EPMA) (JEOL, Model JXA-8600MX).

3. Result and discussion

3.1. Characterization of p-type $(Bi_{0.25}Sb_{0.75})_2Te_3$ and n-type $Bi_2(Te_{0.94}Se_{0.06})_3$ specimens

Since the thermoelectric properties of bismuth-telluride compounds depend strongly on the part cut from the ingot, the thermoelectric properties was studied using only the specimens cut from the central part of the ingot.

Fig. 1 shows the micrographs of the p-type $(Bi_{0.25}Sb_{0.75})_2Te_3$ and n-type $Bi_2(Te_{0.94}Se_{0.06})_3$ specimens before and after annealing, which were observed on the section perpendicular to the growth direction, where annealing was done at 673 for 5 h in a hydrogen stream, whose temperature is close to the melting point (686 K) of the second phase composed mainly of Te. As shown in Fig 1a, c and e, the as-grown p-type specimen is a two-phase material consisting of an alloy matrix

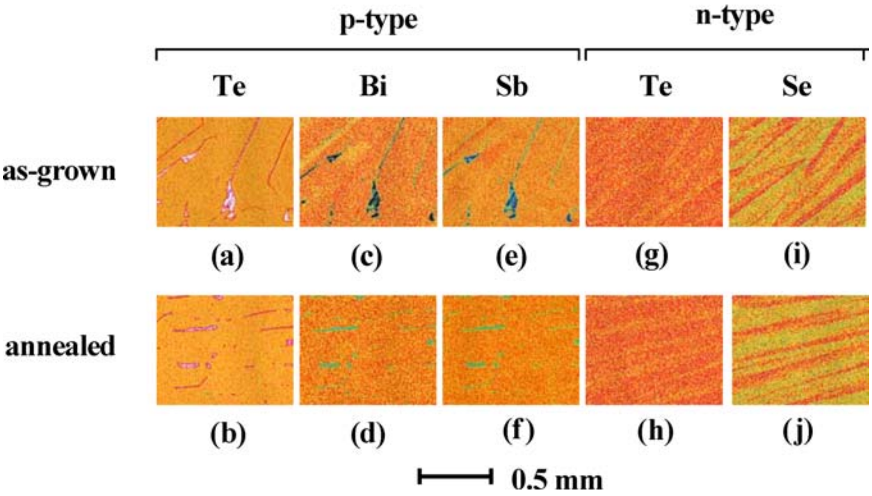

Figure 1 EPMA micrographs of Te ((a) and (b)), Bi ((c) and (d)) and Sb elements ((e) and (f)) in the p-type $(Bi_{0.25}Sb_{0.75})_2Te_3$ doped with 8 wt% excess Te alone and of Te ((g) and (h)) and Se elements ((i) and (j)) in the n-type $Bi_2(Te_{0.94}Se_{0.06})_3$ codoped with 0.017 wt% Te and 0.068 wt% I before and after annealing at 673 K: (a), (c), (e), (g) and (i) before annealing, (b), (d), (f), (h) and (j) after annealing at 673 K for 5 h in a hydrogen stream, where microphotographs are taken on the surface perpendicular to the growth direction. (a), (c) and (e), (g) and (i), (b), (d) and (f), and (h) and (j) micrographs show the same area, respectively.

and a second phase, where the grain has an average size of about 0.5 mm in length and about 0.1 mm in width on the cross-section perpendicular to the growth direction. The second phase appears as lamellae and islands, which are precipitates of insoluble constituents. The EPMA analysis revealed that the second phase was composed of the Te-rich phase of >90 at% Te, <6 at% Sb and <2 at% Bi and precipitated out along grain boundaries and cleavage planes. When it was annealed at 673 K for 5 h in a hydrogen stream, however, the second phase of the excess Te was dispersed to some extent, due to the diffusion of Te into the matrix, so that lamellae became shorter. The hydrogen annealing at a high temperature of 673 K close to the melting point of the Te-rich phase, promoted somewhat the diffusion of the second phase.

On the other hand, the as-grown *n*-type specimen consists mainly of Te- and Se-rich lamellar structures, as shown in Fig. 1g and i. However, no apparent grain boundary was observed as shown in their figures. As shown in Fig. 1g and j, Te- and Se-rich structures remained almost unchanged, even when annealed at 673 K for 5 h in a hydrogen stream, although approximately 1% of an initial Te content was vaporized during annealing. The Se-rich regions just correspond to the Te-poor regions and vice versa, as shown in Fig. 1g andj, but a constitutive element of Bi showed a homogeneous distribution.

3.2. α, ρ and κ of the as-grown *p*-type $(Bi_{0.25}Sb_{0.75})_2Te_3$ and annealed *n*-type $Bi_2(Te_{0.94}Se_{0.06})_3$ specimens

The thermoelectric properties of the *p*- and *n*-type specimens before and after annealing were investigated along the freezing direction. The thermoelectric properties of the as-grown *p*- and *n*-type specimens coincided closely with those of the previous specimens [6, 7], except for κ of the *p*-type one which is 22% higher than that of the previous *p*-type one. The annealing degraded significantly the power factor *P* and thermoelectric figure of merits *ZT* of the annealed *p*-type specimen but enhanced remarkably that of the *n*-type one, as listed in Table I. The annealing has the same effect as that reported previously [6, 7]. As reported previously, when the *p*-type specimen was doped with 3 wt% excess Te, *ZT* was improved after annealing at 673 K for 2 h in a vacuum, while when it was doped with 8 wt% excess Te, *ZT* was degraded after annealing. Thus, whether the annealing has a favorable effect or an advertise effect on *ZT* depends strongly on the additional content of dopant. Of course, the same holds even for the *n*-type specimen. Generally, the optimum annealing condition changes entirely with the type and additional content of dopants, even when the same fabrication process was employed [14].

Fig. 2a and b show the temperature dependences of α and ρ of the as-grown *p*-type and annealed *n*-type specimens. ρ values of both specimens increase almost linearly and abruptly with an increase of temperature. However, α of the *n*-type specimen increases more slowly with an increase of temperature than that of the *p*-type one. As shown in Fig. 2c, the power factor *P* values of the *p*- and *n*-type specimens reached great values of 7.4 and 8.7 mW/K^{2}m at 200 K, respectively, which are much higher than those reported by Abrikosov and Ivanova [15] for the *p*-type $(Bi_{0.325}Sb_{0.675})_2$ $(Te_{0.9}Se_{0.1})_3$ doped with 5 wt% excess Te alone and by Yim and Rosi [4] for the *n*-type $(Bi_{0.9}Sb_{0.1})_2$ $(Te_{0.95}Se_{0.05})_3$ doped with 0.16 wt% SbI_3. The abrupt increase in ρ with temperature is reflected in the temperature dependence of *P*, so that *P* of the present specimens exhibited a tendency to decrease almost linearly with increasing temperature. In contrast, *P* values of the high-performance bismuth telluride compounds prepared by them [4, 15] change only a little with temperature.

As shown in Figs 3a and 4a, κ values of the as-grown *p*-type and annealed *n*-type specimens change parabolically with temperature in the range from 173 to 433 K and have a minimum at approximately 320 K. This temperature dependence of κ is similar to those of the experimental values obtained by Abrikosov and Ivanova [15] for the *p*-type $(Bi_{0.325}Sb_{0.675})_2(Te_{0.9}Se_{0.1})_3$ doped with 5 wt% excess Te alone and by Yim and Rosi [4] for the *n*-type $(Bi_{0.9}Sb_{0.1})_2(Te_{0.95}Se_{0.05})_3$ doped with 0.16 wt% SbI_3, as shown in Figs 3b and 4b. However, κ values at 300 K of the present *p*- and *n*-type specimens were 44 and 21% higher than those of their high-performance bismuth-telluride compounds [4, 15]. This difference in *k* arises predominantly from the difference in the electronic component $\kappa_{el.}$ of κ which was calculated from the Wiedemann-Franz law [1], $\kappa_{el.}=LT/\rho$, where *L* is Lorentz number, because the difference in κ almost equals the difference in $\kappa_{el.}$. The difference in κ is thus found to result from the difference in κ.

$\kappa_{el.}$ of the present *p*- and *n*-type specimens tends to decrease almost linearly with an increase of temperature. In general, the lattice component $\kappa_{lat.}$ of κ is inversely proportional to the temperature at higher temperatures above Debye temperature θ_D (θ_D=155 K

TABLE I Thermoelectric properties measured at 298 K for the *p*-type $(Bi_{0.25}Sb_{0.75})_2Te_3$ and *n*-type $Bi_2(Te_{0.94}Se_{0.06})_3$ specimens before and after annealing at 673 K for 5 h in a hydrogen stream

		p-type		*n*-type	
		As-grown	Annealed	As-grown	Annealed
Electrical resistivity	ρ (μ Ω m)	8.71	11.7	10.7	8.23
Seebeck coefficient	α (μV/K)	+221	+221	−227	−223
Thermal conductivity	κ (W/mK)	1.48	1.36	1.57	1.65
Power factor	*P* (mW/K^{2}m)	5.60	4.17	4.82	6.04
Figure of merit	*ZT*	1.13	0.91	0.91	1.09

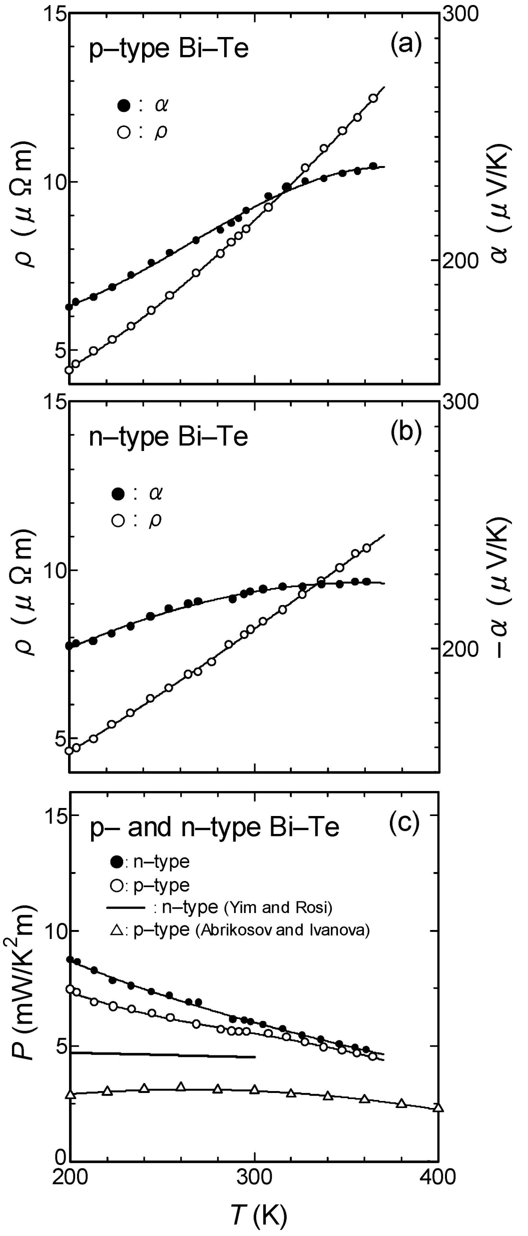

Figure 2 Electrical resisitivity ρ, Seebeck coefficient α and Power factor P as a function of temperature for the as-grown p-type $(Bi_{0.25}Sb_{0.75})_2Te_3$ specimen (a) doped with 8 wt% excess Te alone and the annealed n-type $Bi_2(Te_{0.94}Se_{0.06})_3$ one (b) codoped with 0.068 wt% I and 0.017 wt% Te, where annealing was done at 673 K for 5 h in a hydrogen stream. The open triangles (△) in Fig. 2c denote P obtained by Abrikosov and Ivanova for the p-type $(Bi_{0.325}Sb_{0.675})_2$ $(Te_{0.9}Se_{0.1})_3$ doped with 5 wt% excess Te alone (Ref. [15]). The heavy solid line in Fig. 2c denotes P obtained by Yim and Rosi for the n-type $(Bi_{0.9}Sb_{0.1})_2(Te_{0.95}Se_{0.05})_3$ doped with 0.16 wt% SbI_3 (Ref. [4]).

for Bi_2Te_3) [1]. However, $\kappa_{\text{lat.}}$ estimated by subtracting $\kappa_{\text{el.}}$ from $\kappa_{\text{obs.}}$ tends to change parabolically with temperature and has a minimum at approximately 300 K, as shown in Figs 3a and 4a. $\kappa_{\text{lat.}}$ values at 300 K of the present p- and n-type specimens are very close to those reported by Abrikosov and Ivanova [15] for the p-type specimen and by Yim and Rosi [4] for the n-type one, respectively. The difference between their and our $\kappa_{\text{lat.}}$ at 300 K was approximately only 0.01 W/mK corresponding to approximately 1% of $\kappa_{\text{lat.}}$, independently of the conduction type. Theoretically, both $\kappa_{\text{el.}}$ and $\kappa_{\text{lat.}}$ ought to decrease with increasing temperature, at least in the present temperature range, but it is inconsistent with the practical changes in κ with temperature. The actual change in κ with temperature, therefore, may be associated with whether the contribution of bipolar component $\kappa_{\text{bi.}}$ [13] to κ surpasses or almost equals the decrease in both $\kappa_{\text{el.}}$ and $\kappa_{\text{lat.}}$ with temperature; the rate of increase in $\kappa_{\text{bi.}}$ with temperature becomes large as the band gap is narrowed with increasing temperature, so that the thermal excitation across the band gap is more apt to occur. Particularly, $\kappa_{\text{bi.}}$ should change with the dopant distribution in polycrystalline, because the gap width of the band gap varies with the slight change in the dopant distribution. It is thus considered that the temperature dependence of the resultant κ is determined predominantly by the gap width of the band gap.

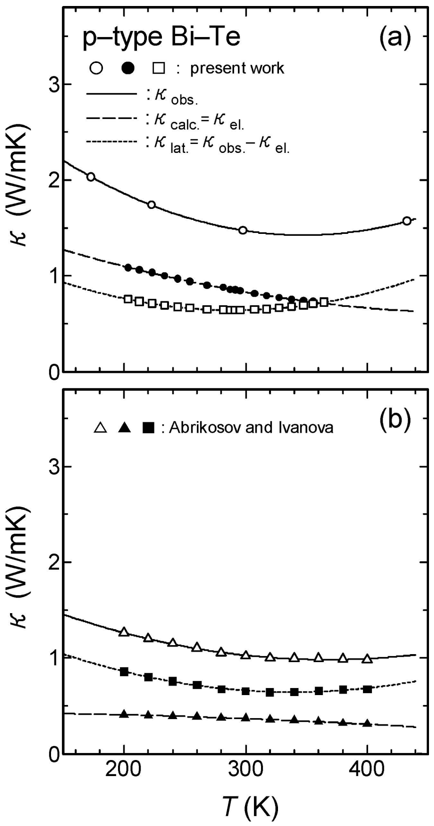

Figure 3 Thermal conductivity κ as a function of temperature for the as-grown p-type $(Bi_{0.25}Sb_{0.75})_2Te_3$ specimen (a) doped with 8 wt% excess Te alone and the p-type $(Bi_{0.325}Sb_{0.675})_2(Te_{0.9}Se_{0.1})_3$ one (b) doped with 5 wt% excess Te alone (Ref. [15]). The solid and dotted curves were drawn by the least square fitting of a quadratic function to $\kappa_{\text{obs.}}$ and $\kappa_{\text{lat.}}$, respectively.

3.3. Temperature dependence of thermoelectric figure of merit

Fig. 5 shows the temperature dependences of Z and ZT for the as-grown p-type and annealed n-type specimens. Z of the present specimens tends to decrease monotonically with an increase of temperature and has no local maximum above 200 K, while Z of the high-performance bismuth-telluride compounds reported by Abrikosov and Ivanova [15] and Yim ans Rosi [4] tends to have a local maximum at approximately 300 K. ZT values of their high-performance p- and n-type compounds have a local maximum at 360 and 300 K, respectively. However, ZT values of the present specimens have a local maximum at

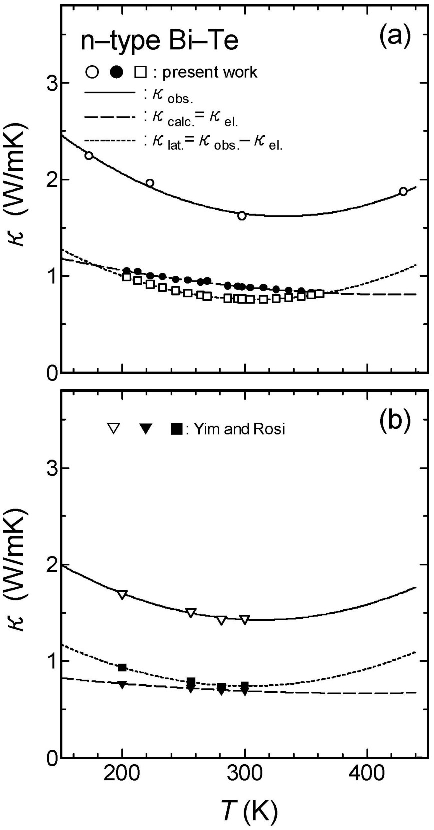

Figure 4 Thermal conductivity κ as a function of temperature for the annealed n-type $Bi_2(Te_{0.94}Se_{0.06})_3$ specimen (a) codoped with 0.068 wt% I and 0.017 wt% Te and the n-type $(Bi_{0.9}Sb_{0.1})_2$ $(Te_{0.95}Se_{0.05})_3$ one (b) doped with 0.16 wt% SbI_3 (Ref. 4), where annealing was done at 673 K for 5 h in a hydrogen stream. The solid and dotted curves were drawn by the least square fitting of a quadratic function to $\kappa_{obs.}$ and $\kappa_{lat.}$, respectively.

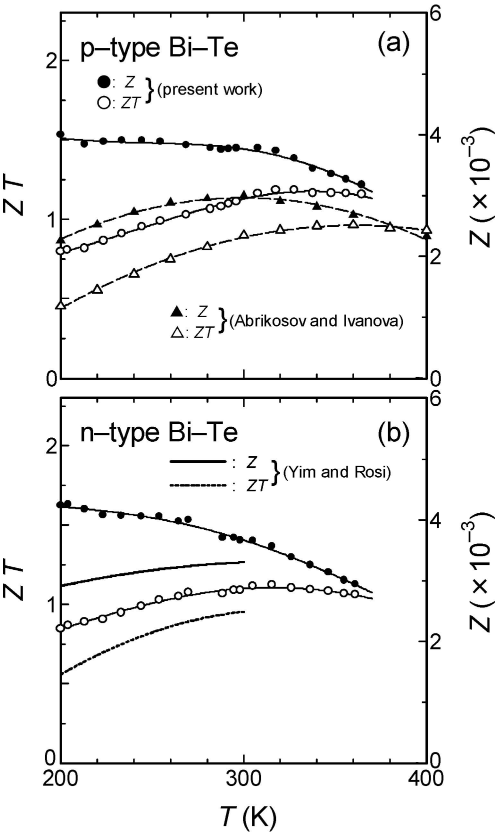

Figure 5 Thermoelectric figure of merits Z and ZT as a function of temperature for the as-grown p-type $(Bi_{0.25}Sb_{0.75})_2Te_3$ specimen (a) doped with 8 wt% excess Te alone and the annealed n-type Bi_2 $(Te_{0.94}Se_{0.06})_3$ one (b) codoped with 0.068 wt% I and 0.017 wt% Te, where annealing was done at 673 K for 5 h in a hydrogen stream. The open and filled triangles (△ and ▲) in Fig. 4a denote Z and ZT, respectively, which were obtained by Abrikosov and Ivanova (Ref. [15]) for the p-type $(Bi_{0.325}Sb_{0.675})_2$ $(Te_{0.9}Se_{0.1})_3$ doped with 5 wt% excess Te alone. The heavy solid and dotted lines in Fig. 4b denote the Z and ZT, respectively, which were obtained by Yim and Rosi (Ref. [4]) for the n-type $(Bi_{0.9}Sb_{0.1})_2$ $(Te_{0.95}Se_{0.05})_3$ doped with 0.16 wt% SbI_3.

approximately 320 K. The maximum ZT reached great values of 1.19 for the as-grown p-type specimen and 1.13 for the annealed n-type one. ZT values at 300 K of the present p- and n-type specimens are 26 and 23% higher than those reported by them [4, 15]. However, the maximum values of the present p- and n-type specimens were approximately 15 and 5% smaller than the experimental values obtained for our previous specimens [6, 7], although they were cut from the same ingots. Probably this difference in the maximum ZT may arise from the slight difference in the dopant distribution, because ρ and κ are highly sensitive to the slight change in the dopant distribution [14].

ZT of the present specimens exhibits a temperature dependence much weaker than those of the high-performance bismuth-telluride compounds in the temperature range from 200 to 360 K; the ratios of maximum ZT to the minimum one for the present p- and n-type specimens are 1.48 and 1.32, respectively, but those for their high-performance p- and n-type compounds [4, 15] are 2.14 and 1.70. It indicates that the present materials are far superior to any other bismuth-telluride compound in the thermal stability of energy conversion efficiency in addition to the high performance. This is attributed to the feature that P of the present specimens tends to decrease with an increase of temperature. When these materials are produced in a large scale, however, it would be required to control precisely the distribution of dopant elements spread in bismuth-telluride bulk in the fabrication process.

4. Summary

The present experimental results can be summarized as follows.

The p-type $(Bi_{0.25}Sb_{0.75})_2Te_3$ ingot doped with 8 wt% excess Te alone and the n-type Bi_2 $(Te_{0.94}Se_{0.06})_3$ ingot codoped with 0.068 wt% I and 0.017 wt% Te were grown by the Bridgman method and annealed at 673 K for 5 h in a hydrogen stream. When the p-type specimen is as-grown and the n-type one was annealed at 673 K for 5 h in a hydrogen stream, the highest ZT was obtained as in the case of the previous specimens. The thermoelectric figure of merits ZT of the as-grown p-type and annealed n-type specimens were measured in the temperature range from 200 to 360 K. The electrical resistivity ρ and Seebeck coefficient α of the present specimens tend to increase with an increase of temperature, but their thermal conductivities κ have a local minimum at approximately 320 K, like κ of other high-performance bismuth-telluride compounds. As a result, ZT values of the as-grown p-type and annealed n-type specimens have a broad peak and reached great values of 1.19 and 1.13 at approximately 320 K, respectively. The present materials were thus found to be far superior to any other bismuth telluride compound in the thermal stability of energy conversion efficiency in addition to the high performance.

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Received 8 January
and accepted 26 April 2005