

## P-type and N-type Bi<sub>2</sub>Te<sub>3</sub>/PbTe Functional Gradient Materials for Thermoelectric Power Generation

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### Abstract

The p-type (Bi<sub>0.2</sub>Sb<sub>0.8</sub>)<sub>2</sub>Te<sub>3</sub>/(Pb<sub>0.7</sub>Sn<sub>0.3</sub>)Te functional gradient material (FGM) was fabricated by hot-pressing the mechanically alloyed (Bi<sub>0.2</sub>Sb<sub>0.8</sub>)<sub>2</sub>Te<sub>3</sub> and the 0.5 at% Na<sub>2</sub>Te-doped (Pb<sub>0.7</sub>Sn<sub>0.3</sub>)Te powders. Also, the n-type Bi<sub>2</sub>(Te<sub>0.9</sub>Se<sub>0.1</sub>)<sub>3</sub>/PbTe FGM was processed by hot-pressing the mechanically alloyed Bi<sub>2</sub>(Te<sub>0.9</sub>Se<sub>0.1</sub>)<sub>3</sub> and the 0.3 wt% Bi-doped PbTe powders. With  $\Delta T$  larger than 300 °C, the p-type (Bi<sub>0.2</sub>Sb<sub>0.8</sub>)<sub>2</sub>Te<sub>3</sub>/(Pb<sub>0.7</sub>Sn<sub>0.3</sub>)Te FGM exhibited larger thermoelectric output power than those of the (Bi<sub>0.2</sub>Sb<sub>0.8</sub>)<sub>2</sub>Te<sub>3</sub> and the 0.5 at% Na<sub>2</sub>Te-doped (Pb<sub>0.7</sub>Sn<sub>0.3</sub>)Te alloys. For the n-type Bi<sub>2</sub>(Te<sub>0.9</sub>Se<sub>0.1</sub>)<sub>3</sub>/PbTe FGM, the thermoelectric output power superior to those of the Bi<sub>2</sub>(Te<sub>0.9</sub>Se<sub>0.1</sub>)<sub>3</sub> and the 0.3 wt% Bi-doped PbTe was predicted at  $\Delta T$  larger than 300 °C.

**Keywords :** Thermoelectric materials, Bismuth telluride, Lead telluride, Functional gradient materials.

### 1. Introduction

Recently, there has been renewed interest in thermoelectric power generation that can recycle the waste heat as one of energy-saving technologies [1]. Thermoelectric power generation depends on the characteristics of thermoelectric materials and temperature difference at both ends of the module. As thermoelectric materials exhibit large temperature dependence on their thermoelectric properties, it is difficult to obtain high energy-conversion efficiency for thermoelectric modules where large temperature difference is essential. To improve the energy-conversion efficiency, functionally graded structures with segment bonding of different materials have been proposed [2,3].

In this study, functional gradient materials (FGMs) of the p-type (Bi<sub>0.2</sub>Sb<sub>0.8</sub>)<sub>2</sub>Te<sub>3</sub>/(Pb<sub>0.7</sub>Sn<sub>0.3</sub>)Te were fabricated by hot-pressing the mechanically alloyed (Bi<sub>0.2</sub>Sb<sub>0.8</sub>)<sub>2</sub>Te<sub>3</sub> and the 0.5 at% Na<sub>2</sub>Te-doped (Pb<sub>0.7</sub>Sn<sub>0.3</sub>)Te powders. Also, the n-type Bi<sub>2</sub>(Te<sub>0.9</sub>Se<sub>0.1</sub>)<sub>3</sub>/PbTe FGM was processed by hot-pressing the mechanically alloyed Bi<sub>2</sub>(Te<sub>0.9</sub>Se<sub>0.1</sub>)<sub>3</sub> and the 0.3 wt% Bi-doped PbTe powders. The thermoelectric properties of the p-type (Bi<sub>0.2</sub>Sb<sub>0.8</sub>)<sub>2</sub>Te<sub>3</sub>/(Pb<sub>0.7</sub>Sn<sub>0.3</sub>)Te and the n-type Bi<sub>2</sub>(Te<sub>0.9</sub>Se<sub>0.1</sub>)<sub>3</sub>/PbTe FGMs were evaluated.

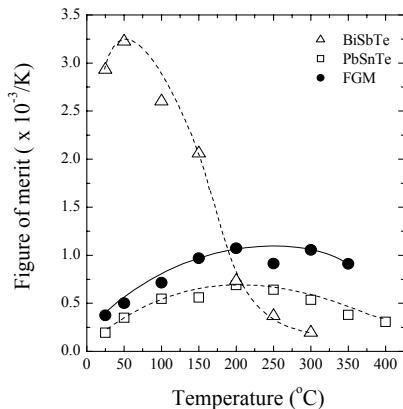
### 2. Experimental and Results

Mechanical alloying of the (Bi<sub>0.2</sub>Sb<sub>0.8</sub>)<sub>2</sub>Te<sub>3</sub> powders was conducted at 1200 rpm using a Spex mill for 5 hours. The 0.5 at% Na<sub>2</sub>Te-doped (Pb<sub>0.7</sub>Sn<sub>0.3</sub>)Te powders were mechanically alloyed at 1200 rpm for 10 hours and reduction-treated at 400 °C for 24 hours in (50% H<sub>2</sub> + 50%

Ar) atmosphere. The (Bi<sub>0.2</sub>Sb<sub>0.8</sub>)<sub>2</sub>Te<sub>3</sub> and the (Pb<sub>0.7</sub>Sn<sub>0.3</sub>)Te powders were then cold-pressed at 475 MPa to form 5 mm×5mm×10mm compacts of (Bi<sub>0.2</sub>Sb<sub>0.8</sub>)<sub>2</sub>Te<sub>3</sub> and (Pb<sub>0.7</sub>Sn<sub>0.3</sub>)Te, respectively. To form the 5 mm×5mm×10 mm compacts of the (Bi<sub>0.2</sub>Sb<sub>0.8</sub>)<sub>2</sub>Te<sub>3</sub>/(Pb<sub>0.7</sub>Sn<sub>0.3</sub>)Te, the (Bi<sub>0.2</sub>Sb<sub>0.8</sub>)<sub>2</sub>Te<sub>3</sub> and the (Pb<sub>0.7</sub>Sn<sub>0.3</sub>)Te powders were charged into the tool-steel mold with a segment ratio of 1:1, and cold-pressed together. The (Bi<sub>0.2</sub>Sb<sub>0.8</sub>)<sub>2</sub>Te<sub>3</sub>, the (Pb<sub>0.7</sub>Sn<sub>0.3</sub>)Te, and the (Bi<sub>0.2</sub>Sb<sub>0.8</sub>)<sub>2</sub>Te<sub>3</sub>/(Pb<sub>0.7</sub>Sn<sub>0.3</sub>)Te FGM compacts were hot-pressed at 500°C for 1 hour in vacuum. The n-type Bi<sub>2</sub>(Te<sub>0.9</sub>Se<sub>0.1</sub>)<sub>3</sub> and the 0.3 wt% Bi-doped PbTe powders were also fabricated by mechanical alloying and were hot-pressed at 500°C for 1 hour in vacuum after cold compaction. The segment ratio of the Bi<sub>2</sub>(Te<sub>0.9</sub>Se<sub>0.1</sub>)<sub>3</sub>/PbTe cold compact was kept 1:1 as like as the p-type FGM.

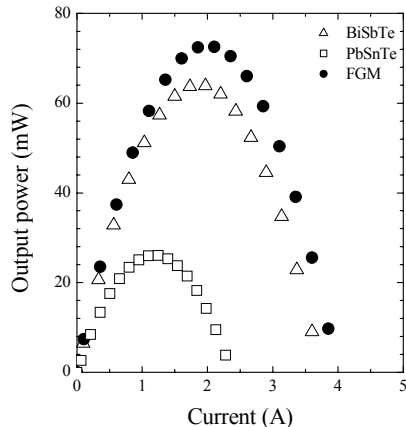
Seebeck coefficients( $\alpha$ ), electrical resistivities( $\rho$ ) and thermal conductivities( $\kappa$ ) of the hot-pressed specimens were measured at 25~400°C. The figure-of-merit was evaluated with the relation  $Z = \alpha^2/(\rho\kappa)$ . Power-generation characteristics were measured with applying a temperature difference of 20~320°C at both ends of a specimen.

Fig. 1 shows the figure-of-merits of the p-type (Bi<sub>0.2</sub>Sb<sub>0.8</sub>)<sub>2</sub>Te<sub>3</sub>/(Pb<sub>0.7</sub>Sn<sub>0.3</sub>)Te FGM as well as those of the (Bi<sub>0.2</sub>Sb<sub>0.8</sub>)<sub>2</sub>Te<sub>3</sub> and the (Pb<sub>0.7</sub>Sn<sub>0.3</sub>)Te. The figure-of-merit of the p-type (Bi<sub>0.2</sub>Sb<sub>0.8</sub>)<sub>2</sub>Te<sub>3</sub>/(Pb<sub>0.7</sub>Sn<sub>0.3</sub>)Te FGM was improved than those of the (Bi<sub>0.2</sub>Sb<sub>0.8</sub>)<sub>2</sub>Te<sub>3</sub> and (Pb<sub>0.7</sub>Sn<sub>0.3</sub>)Te at temperatures above 180 °C, implying that the efficiency of thermoelectric energy conversion can be improved by segment bonding of the different thermoelectric materials [2,3].



**Fig. 1. Temperature dependence of figure-of-merit of the p-type  $(\text{Bi}_{0.2}\text{Sb}_{0.8})_2\text{Te}_3/(\text{Pb}_{0.7}\text{Sn}_{0.3})\text{Te}$  FGM, the  $(\text{Bi}_{0.2}\text{Sb}_{0.8})_2\text{Te}_3$ , and the  $(\text{Pb}_{0.7}\text{Sn}_{0.3})\text{Te}$ .**

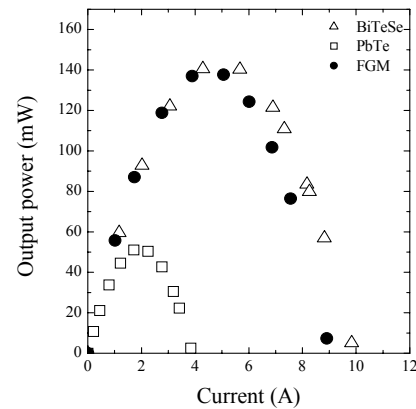
Fig. 2 shows the maximum output power of the p-type  $(\text{Bi}_{0.2}\text{Sb}_{0.8})_2\text{Te}_3/(\text{Pb}_{0.7}\text{Sn}_{0.3})\text{Te}$  FGM, the  $(\text{Bi}_{0.2}\text{Sb}_{0.8})_2\text{Te}_3$  and the  $(\text{Pb}_{0.7}\text{Sn}_{0.3})\text{Te}$  with  $\Delta T$  of 320 °C. The  $(\text{Bi}_{0.2}\text{Sb}_{0.8})_2\text{Te}_3/(\text{Pb}_{0.7}\text{Sn}_{0.3})\text{Te}$  FGM exhibited the maximum output power of 72.6 mW, which was the 13.6% and 179% improved value compared to 63.9 mW of the  $(\text{Bi}_{0.2}\text{Sb}_{0.8})_2\text{Te}_3$  and 26 mW of the  $(\text{Pb}_{0.7}\text{Sn}_{0.3})\text{Te}$ , respectively.



**Fig. 2. Maximum output power of the p-type  $(\text{Bi}_{0.2}\text{Sb}_{0.8})_2\text{Te}_3/(\text{Pb}_{0.7}\text{Sn}_{0.3})\text{Te}$  FGM, the  $(\text{Bi}_{0.2}\text{Sb}_{0.8})_2\text{Te}_3$  and the  $(\text{Pb}_{0.7}\text{Sn}_{0.3})\text{Te}$  with  $\Delta T$  of 320 °C.**

Fig. 3 illustrates the maximum output power of the n-type  $\text{Bi}_2(\text{Te}_{0.9}\text{Se}_{0.1})_3/\text{PbTe}$  FGM, the  $\text{Bi}_2(\text{Te}_{0.9}\text{Se}_{0.1})_3$ , and the 0.3 wt% Bi-doped PbTe with  $\Delta T$  of 300 °C. The n-type  $\text{Bi}_2(\text{Te}_{0.9}\text{Se}_{0.1})_3/\text{PbTe}$  FGM exhibited the maximum output power almost identical to the  $\text{Bi}_2(\text{Te}_{0.9}\text{Se}_{0.1})_3$ . As the maximum output power of the  $\text{Bi}_2(\text{Te}_{0.9}\text{Se}_{0.1})_3/\text{PbTe}$  FGM was improved with increasing  $\Delta T$ , it could be predicted that the n-type  $\text{Bi}_2(\text{Te}_{0.9}\text{Se}_{0.1})_3/\text{PbTe}$  FGM would generate the output powers superior to those of the  $\text{Bi}_2(\text{Te}_{0.9}\text{Se}_{0.1})_3$

and the 0.3 wt% Bi-doped PbTe with  $\Delta T$  larger than 300 °C.



**Fig. 3. Maximum output power of the n-type  $\text{Bi}_2(\text{Te}_{0.9}\text{Se}_{0.1})_3/\text{PbTe}$  FGM, the  $\text{Bi}_2(\text{Te}_{0.9}\text{Se}_{0.1})_3$ , and the  $\text{PbTe}$  with  $\Delta T$  of 300 °C.**

### 3. Summary

At temperatures higher than 180 °C, the p-type  $(\text{Bi}_{0.2}\text{Sb}_{0.8})_2\text{Te}_3/(\text{Pb}_{0.7}\text{Sn}_{0.3})\text{Te}$  FGM exhibited superior figure-of-merits to those of the  $(\text{Bi}_{0.2}\text{Sb}_{0.8})_2\text{Te}_3$  and the 0.5 at%  $\text{Na}_2\text{Te}$ -doped  $(\text{Pb}_{0.7}\text{Sn}_{0.3})\text{Te}$  alloys, implying that the efficiency of thermoelectric energy conversion can be improved by segment bonding of the different thermoelectric materials. With  $\Delta T$  larger than 300 °C at both ends of the specimen, the p-type  $(\text{Bi}_{0.2}\text{Sb}_{0.8})_2\text{Te}_3/(\text{Pb}_{0.7}\text{Sn}_{0.3})\text{Te}$  FGM exhibited the higher output powers than those of the  $(\text{Bi}_{0.2}\text{Sb}_{0.8})_2\text{Te}_3$  and the 0.5 at%  $\text{Na}_2\text{Te}$ -doped  $(\text{Pb}_{0.7}\text{Sn}_{0.3})\text{Te}$  alloys. The output power of the n-type  $\text{Bi}_2(\text{Te}_{0.9}\text{Se}_{0.1})_3/\text{PbTe}$  FGM was predicted to be superior to those of the  $\text{Bi}_2(\text{Te}_{0.9}\text{Se}_{0.1})_3$  and the 0.3 wt% Bi-doped PbTe with  $\Delta T$  larger than 300 °C.

### Acknowledgement

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### 4. References

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