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STUDY ON Bi-Sb-TeALLOY THERMOELECTRIC FOR VEHICLE EXHAUST BY ZONE MELTING METHOD

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Abstract-This study investigated the uses zone melting method to acquire P-type thermoelectric material $Bi_{0.4}Sb_{1.6}Te_3$ and N-type thermoelectric material $Bi_2Te_{2.55}Se_{0.45}$. There are 16 thermoelectric material masses in series connection to form the thermoelectric module. At temperature 381K, the measured Seebeck coefficient of $Bi_{0.4}Sb_{1.6}Te_3$ is 223µV/K, and its thermoelectric figure of merit ZT is 1.36. At temperature 400K, the measured Seebeck coefficient of $Bi_2Te_{2.55}Se_{0.45}$ is 206µV/K, and its thermoelectric figure of merit ZT is 1.03. Using SEM, EDS and Hall effect measurement, the study explores the Seebeck coefficient, electrical conductivity and thermal conductivity of thermoelectric materials. Finally, the powder of thermoelectric material is cold-pressed under 766.4Mpa to make square thermoelectric pairs with size 8.2mm×8.2mm and thicknesses 0.8mm and 1.5mm. Solder paste $Sn_{42}Bi_{58}$ and copper electrode plate are in series connection with 16 pieces of P/N thermoelectric material to form thermoelectric modules.
Keywords -Tribological;benzotriazole; composite; anti-wear.

I. INTRODUCTION

For the petrol engine of internal combustion engine currently used in vehicles, only 30% of it (or around 35% of diesel engine) is used to drive vehicle; around 30% is consumed in engine cooling; and around 40% of waste heat is discharged to the atmosphere through the exhaust. It is hoped that such 70% of energy can be recycled for reuse, and this would be an important research direction. Power generation and energy recycle from waste heat have great prospects, and this study involves both energy autonomy and awareness of environmental protection. Thermoelectric conversion technique is a potential energy technique that can perform fusion between thermoelectric material and multiple industries, such as steel refining, kilning industry, etc. Many advanced countries in the world have been putting in a lot of resources for the related studies. Thermoelectric material is up to now the most effective solution for waste heat recovery.

The principle of thermoelectric material is that Seebeck effect, coming from the temperature gradient at the two ends of material, makes carrier transmission asymmetric. When an electronic hole inside a material is observed, it is found that when temperature difference is zero, the carrier transmission at the two ends is the same, implying that the two ends keep having equal amount of carriers. If there is temperature difference existed, the hot end would provide higher kinetic energy for the carrier at this end. Hence, the amount of carriers transmitted from hot end to cold end is more [1] [2]. In the 20th century studies of compound semiconductor started to have prosperous development. Scientists found that the thermoelectric nature of compound semiconductor is even better than pure gold. Right now, the most commonly used alloy under room temperature is Bi2Te3 alloy series. Poudel et al. [3] used nanostructure to enhance the thermoelectric figure of merit ZT of Bi2Te3 alloy to above 1.2 at temperature 373K. Huleihel et al. [4] design a high gain dc-dc converter for energy harvesting of thermal waste by thermoelectric generators. It can increase the voltage of thermoelectric chip.

II. EXPERIMENTAL DETAIL

The flow chart of zone melting method for thermoelectric (TE) material processes is shown in Figure 1.The commercially powder materialsas starting raw thermoelectric material are used. After high energy ball milling for particle synthesis, the cold pressure method was used to form the thermoelectric generator (TEG) bulk. This material was melted in furnaces that operate about 600 k for 30 minutesforBiSbTealloy’s sintering. When alloy solidify, the boule can grow as a perfect single crystal at the solid/liquid boundary, the impurity atoms will diffuse to the liquid region. Thus, by passing a crystal boule through a thin section of furnace very slowly, such that only a small region of the boule is molten at any time, the impurities will be segregated at the end of the crystal. It is lack of impurities in the leftover regions [6].The crystallization structure and microstructure were investigated by X-ray diffraction, scanning electron microscopy and energy-dispersive.

The thermoelectric material was evaluated by the dimensionless figure of merit ZT = S2бT/K [7], where S, б, T and K are the Seebeck coefficient, electrical conductivity, absolute temperature and thermal conductivity, respectively.

As for measuring method of Seebeck coefficient, T-type thermocouple is used as the probe for measuring temperature difference. T-type thermocouple is made by single copper wire, which is then connected to a multifunctional meter instrument, Model:Agilgent 34970A, for measuring voltage and temperature difference simultaneously. Labview is used

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to draw the curves of voltage and temperature difference to show their relationship.

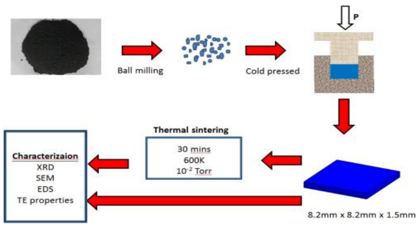


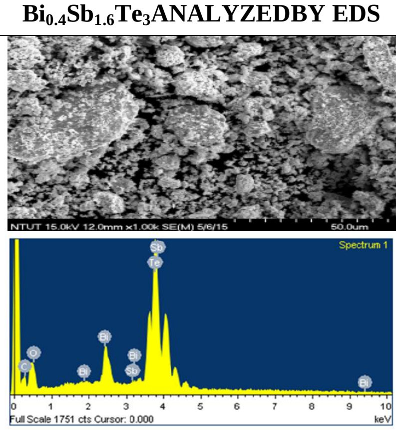
Fig. 1 Processes of BiSbTe thermoelectric alloy

Before measuring Hall Effect, the thermoelectric mass has to be grinded to be below 0.5mm, and its surface also has to be polished to reduce measurement error. The sample is under cold-pressed pressure 766.4Mpa. Meanwhile, the study also makes comparison of items before and after sintering at 327 ^{o}C. The sintering conditions are: temperature rise at 3^{o}C/min.; sintering temperature 327 ^{o}C kept for 30 minutes; and vacuuming 3.2×10^{-2} torr. After that, the sample is cooled in furnace until it reaches room temperature to form the thermoelectric generator (TEG) bulk.

III. RESULTS AND DISCUSSION

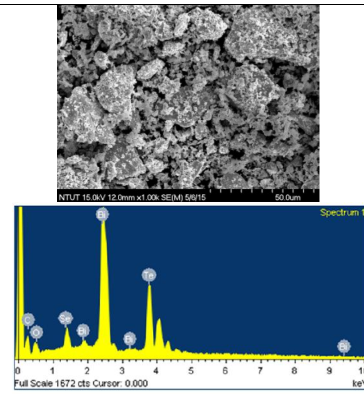
Table 1 shows the bismuth telluride thermoelectric material system $(Bi_2Te_3)_x(Sb_2Te_3)_{1-x}$, and its composition is $Bi_{0.4}Sb_{1.6}Te_3$; and Table 2 shows that its composition is $Bi_2Te_{3-x}Se_x$. As analyzed by EDS, its compound is $Bi_2Te_{2.55}Se_{0.45}$; and as analyzed by SEM, its particle size is 30µm. X-ray diffraction (XRD) analysis is made to analyze the angle and wave crest. Figure 4 shows the XRD diagram of $Bi_{0.4}Sb_{1.6}Te_3$, and the related card number is JCPDS 49-1713. Figure 5 shows the XRD diagram of $Bi_2Te_{2.55}Se_{0.45}$, and the related card number is JCPDS 50-0954. That confirmed the bismuth telluride thermoelectric material. P-type thermoelectric material is $Bi_{0.4}Sb_{1.6}Te_3$ and N-type thermoelectric material is $Bi_2Te_{2.55}Se_{0.45}$.The thermoelectric materialswere made in P/N thermoelectric ingots, with size 8.2mm×8.2mm and thicknesses 0.8mm and 1.5mm.

TABLE1 COMPOSITION OF $Bi_{0.4}Sb_{1.6}Te_3$ANALYZEDBY EDS



Element	Weight%	Atomic%
C K	1.71%	13.92%
O K	2.44%	14.93%
Sb L	28.01%	22.51%
Te L	56.29%	43.38%
Bi M	11.25%	5.26%

TABLE2 COMPOSITION OF $Bi_2Te_{2.55}SE_{0.45}$ANALYZEDBY EDS



Element	Weight%	Atomic%
C K	6.11%	41.53%
O L	2.13%	10.85%
Te L	38.21%	24.43%
Bi M	50.01%	19.53%
Se L	3.54%	3.66%

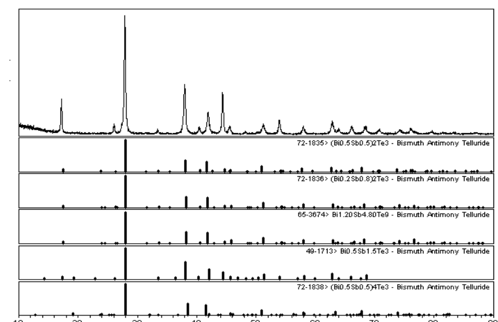


Fig. 2 XRD diagram of $Bi_{0.4}Sb_{1.6}Te_3$

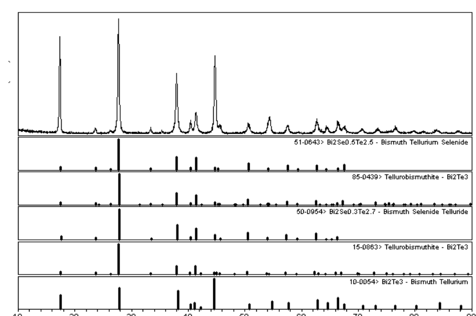


Fig. 3 XRD diagram of $Bi_2Te_{2.55}Se_{0.45}$

After sintering, the bulk concentration of $Bi_{0.4}Sb_{1.6}Te_3$ is decreased, and carrier mobility is increased; but $Bi_2Te_{2.55}Se_{0.45}$ is just the opposite. At temperature 381K, the maximum Seebeck coefficient of $Bi_{0.4}Sb_{1.6}Te_3$ is measured to be 223µV/K; and at temperature 400K, the measured Seebeck coefficient is 206µV/K. The curve of Seebeck coefficient is shown in Figure 4. The maximum electrical conductivities of $Bi_{0.4}Sb_{1.6}Te_3$ and $Bi_2Te_{2.55}Se_{0.45}$ at room temperature are 1736 S/cm and 1391 S/cm respectively. As temperature rises slowly to 400K, the electrical conductivity falls gradually. This is because bismuth telluride is a semiconductor material, and its resistivity increases with temperature rise, thus leading to decline of electrical conductivity, as shown in Figure 5.

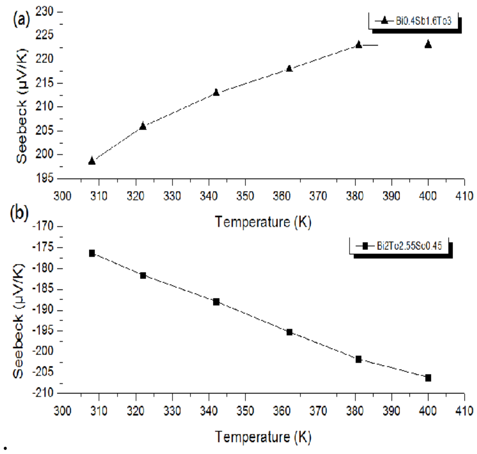


Fig. 4 Seebeck coefficient curves of (a) $Bi_{0.4}Sb_{1.6}Te_3$; and (b) $Bi_2Te_{2.55}Se_{0.45}$

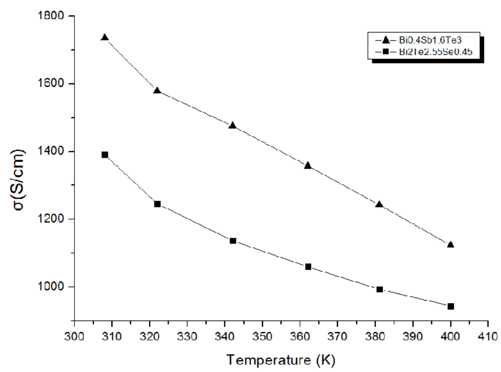


Fig. 5 Electrical conductivity curve of

Thermoelectric figure of merit ZT is performance of thermoelectric material in conversion efficiency. A thermoelectric material with higher thermoelectric figure of merit ZT would turn the medium temperature between high-temperature side and low-temperature side to be energy that pushes carriers. Figure 6 shows the relationship between ZT and electrical conductivity of $Bi_{0.4}Sb_{1.6}Te_3$ and $Bi_2Te_{2.55}Se_{0.45}$. In the figure, the solid line shows thethermoelectric figure of merit ZT, and the dotted line shows the thermal conductivity. It can be seen from the figure that at temperature 381K, the maximum ZT is measured to be 1.36; and at temperature 400K, the ZT is 1.03. Thermoelectric figure of merit is expressed as $ZT = S^2\sigma T/K$. When thermal conductivity is low, ZT will be enhanced. At temperature 381K, the measured thermal conductivity of $Bi_{0.4}Sb_{1.6}Te_3$ is 1.72mW/mK2; and at temperature 400K, the measured thermal conductivity of $Bi_2Te_{2.55}Se_{0.45}$ is 1.56mW/mK2.

The thermoelectric material is cold-pressed at pressure 766.4Mpa to make P/N thermoelectric ingots, with size 8.2mm×8.2mm and thicknesses 0.8mm and 1.5mm. They are connected to form a thermoelectric module with 16 pieces. Aluminum nitride (AlN) serves as the ceramic substrate, and thin copper plate serves as the conductive electrode plate. Solder paste $Sn_{42}Bi_{58}$ is used to weld the thermoelectric masses on a copper plate. The electrical resistance of the 16 thermoelectric material masses in series connection can be measured by multimeter. The electrical resistance of those with thickness 0.8mm is 0.785 ohm; and the electrical resistance of those with thickness 1.5mm is 0.6 ohm, implying that there is no disconnection between welding points, and the resistance at welding points is small.

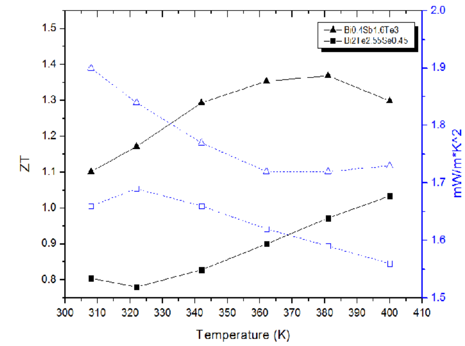


Fig. 6 ZT and thermal conductivity curves of thermoelectric material

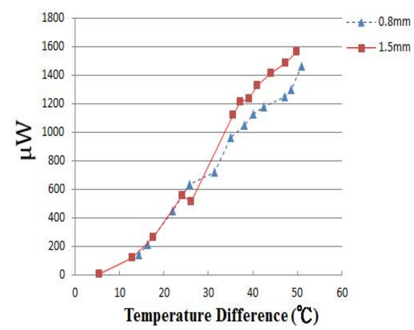


Fig.7 Power and temperature difference curves of thermoelectric modules

In the experiment of thermoelectric modules, those with thickness 0.8mm are compared with those with thickness 1.5mm. Although the thickness 1.5mm of modules is 75% more than the thickness 0.8mm of modules, as shown in Figure 7, the generated power in watts is slightly increased only. Under the fixed temperature difference at the two ends, increasing the leg thickness of thermoelectric module cannot obviously enhance the performance of thermoelectric module. But if it is practically applied to waste heat source, it will be affected by thickness of thermoelectric module, making the low-temperature-side temperatures of those with thickness 0.5mm and those with thickness 1.5mm different. In the experiment of thermoelectric modules, those with thickness 0.8mm are compared with those with thickness 1.5mm. Although the thickness 1.5mm of modules is 75% more than the thickness 0.8mm of modules, the generated power in watts is slightly increased only. Under the fixed temperature difference at the two ends, increasing the leg thickness of thermoelectric module cannot obviously enhance the performance of thermoelectric module. This is one of the reasons for minimizing the thermoelectric material to be nanoscaled.

IV. CONCLUSIONS

This work investigated the thermoelectric generator used zone melting method to acquire P-type thermoelectric material $Bi_{0.4}Sb_{1.6}Te_3$ and N-type thermoelectric material $Bi_2Te_{2.55}Se_{0.45}$. There are 16 thermoelectric material masses in series connection to form the thermoelectric module. From the above experimental data, the following conclusions are drawn:

1. The thermoelectric generatorswere made in P/N thermoelectric ingots, with size 8.2mm×8.2mm and thicknesses 0.8mm and 1.5mm. It can increase the thermoelectric generator voltage and suit for motorcycle exhauster.

2. The thermoelectric generator with higher thermoelectric figure of merit ZT would turn the medium temperature between high-temperature side and low-temperature side has been explored. The relationship between ZT and electrical conductivity of $Bi_{0.4}Sb_{1.6}Te_3$ and $Bi_2Te_{2.55}Se_{0.45}$ can be seen from the figure that at temperature 381K, the maximum ZT is measured to be 1.36; and at temperature 400K, the ZT is 1.03.The performance of ZT is higher than that normal value 1.

3. The aim of acquiring thermoelectric figure of merit ZT ($S^2\sigma T/K$) of thermoelectric material is to enhance Seebeck coefficient and electrical conductivity simultaneously. And reducing thermal conductivity is the most dilemmatic part to thermoelectric material. Since the mutually affecting relationship between thermal conductivity and electrical conductivity is very complicated, it has to rely on the grain boundaries or defects in thermoelectric material to reduce thermal conductivity. When thermoelectric module is actually pasted on the waste heat source, the cold-end temperature can be easily affected by thermal conductivity, also making the cold-end temperature increased and actual temperature difference decreased at the same time. As a result, the voltage properties of thermoelectric module cannot have good function. Additional heat dissipation has to be carried out to enhance the temperature difference between it and waste heat so as to increase the output power.

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