THE EFFECT OF SILVER AND BISMUTH DOPED Mg_2Si ON CRYSTAL STRUCTURE AND THERMOELECTRIC PROPERTIES

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ABSTRACT

Magnesium silicide have been high performance in temperature ranges 500 − 800 K which are medium-temperature and appropriate for applications with waste heat. The Ag and Bi doped Mg2Si were synthesized by hot press method. The crystal structure was analyzed by X-ray diffraction. Thermoelectric properties were measured in temperature rang 323 – 473 K. The XRD pattern of all samples show single crystal and crystallite size slightly increase for doped sample. The doped samples show thermal conductivity lower than un-dope and Ag-doped show maximum decrease about 44.44% at 473 K. The electrical resistivity of doped samples was decreased, and decrease 33% for Bi doped at 473 K. The doped materials slightly decreased Seebeck coefficient about 4% and 16% for Ag and Bi doped, respectively. The highest ZT was found in Ag doped about 0.47×10^{-2} at 473 K.

KEYWORDS: *Alternative Energy; Thermoelectric; Magnesium silicide; Silver; Bismuth*

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INTRODUCTION

Thermoelectric (TE) materials has been investigated in recent years for renewable heat energy into electricity. The performance of TE materials were confirmed by dimensionless figure of merit $ZT=S^2T/\sigma\kappa$, where S is Seebeck coefficient, T is temperature, σ is electrical resistivity and κ is thermal conductivity). Good TE materials needed to high Seebeck coefficient, low electrical resistivity and thermal conductivity. Magnesium silicide have been high performance in temperature ranges 500–800 K which are medium-temperature and appropriate for applications with waste heat. The highest ZT was reported about 1.4 for the $Mg_2(Si_{0.4}Sn_{0.6})Sb_{0.018}$ sample preparing by twice mechanical alloying and spark plasma sintering process [1], $Mg_2Si_{0.53}Sn_{0.4}Ge_{0.05}Bi_{0.02}$ preparing by solid state synthesis and sintering via hot pressing [2], $Mg_{2.16}(Si_{0.4}Sn_{0.6})_{0.97}Bi_{0.03}$ preparing by a two-step solid state reaction followed by spark plasma sintering. [3] and $Mg_2Si_{0.55}Sn_{0.4}Ge_{0.05}$:Bi=1:0.02 preparing by powder methods [4]. This research has objective for improve ZT of Mg_2Si by doped with Ag and Bi.

MATERIALS AND METHODS

The Ag and Bi doped Mg_2Si were prepared by solid state reaction and hot press method. Mg (99%, Aldrich), Si (99.9%, Aldrich), Ag (99.9%, Aldrich) and Bi (99%, Aldrich) were used as starting raw materials. The raw materials were mixed by ball milling for 12 h and calcined at

973 K for 1 h in Ar atmosphere. The powder placed in the graphite mold (ø20 mm) for hot press method at 1053 K under pressure 33 MPa for 1 h in Ar atmosphere.

The crystal structure was characterized by X-ray diffraction (XRD; Shimadzu 6100, Japan) method using CuKα radiation at 40 kV, 30 mA with a scanning speed of 5^{o}/min at 2θ steps of 0.02^{o}. The morphology was observed by scanning electron microscope (SEM; JEOL JSM–5401, Germany). Seebeck coefficient, electrical resistivity and thermal conductivity were measured by steady state method [5] at temperature ranges of 323 – 473 K.

RESULTS AND DISCUSSION

The X-ray diffraction analysis of Mg_2Si, $Mg_{1.98}Bi_{0.02}Si$ and $Mg_{1.98}Ag_{0.02}Si$ after hot press are compare with PDF#2 00-035-0773 as shown in Fig. 1. The crystal structure of all samples are show cubic Fm3m space group as a major phase, and the minor secondary phase MgO and Si are detected in all compositions due to the oxidation of Mg during the solid state reaction [6]. The SEM images magnification ×3500 show distribution, size and shape of particle. The particle size show 2–6 µm and evenly distribution as show in Fig. 2.

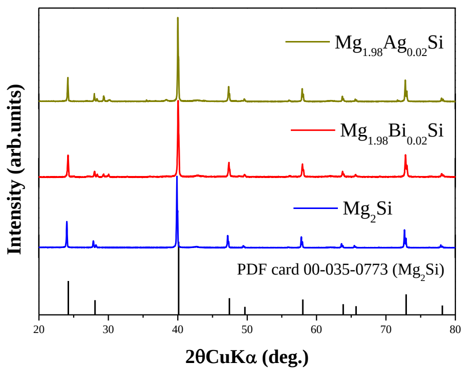

Fig. 1 XRD pattern of Mg_2Si (blue line) $Mg_{1.98}Bi_{0.02}Si$ (red line) and $Mg_{1.98}Ag_{0.02}Si$ (green line).

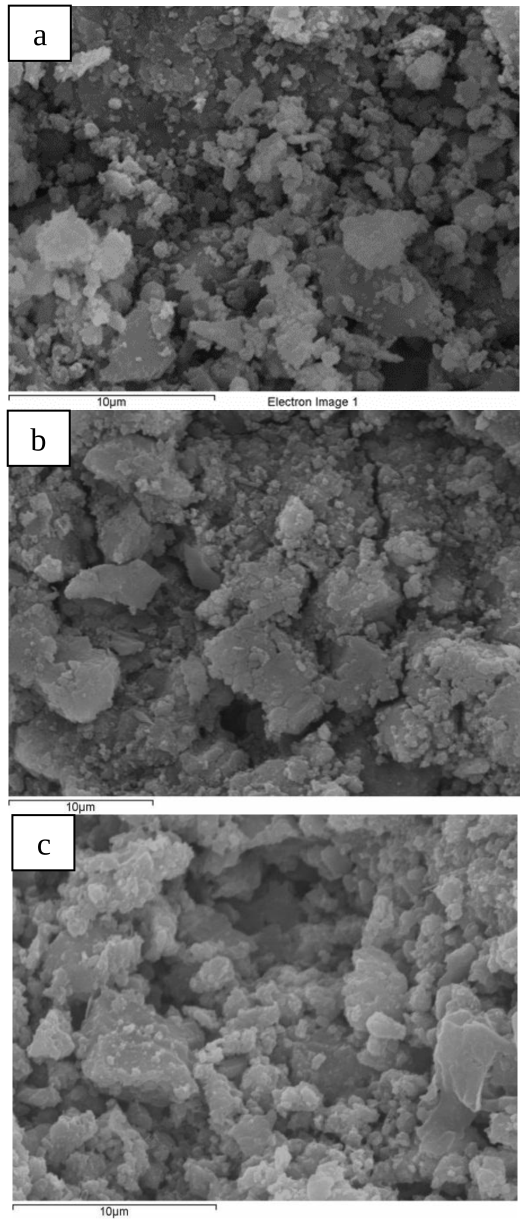

Fig. 2 SEM images of (a) Mg_2Si (b) $Mg_{1.98}Bi_{0.02}Si$ and (c) $Mg_{1.98}Ag_{0.02}Si$.

The Seebeck coefficient increases with temperature increasing but decreased when doping with Ag and Bi. The Mg_2Si shows highest value about –150 µV/K at 473 K as shown in Fig. 3.

Temperature dependence of electrical resistivity for Mg_2Si, $Mg_{1.98}Bi_{0.02}Si$ and $Mg_{1.98}Ag_{0.02}Si$ shows in Fig. 4. The Ag and Bi doped effect to decease electrical resistivity due to increase in carrier concentration and Bi doped shows lowest value about 60 mΩ cm at 473 K.

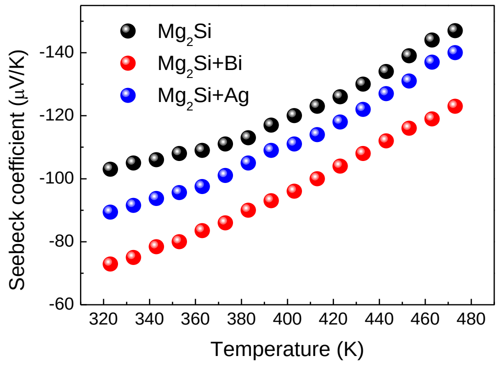

Fig. 3 Seebeck coefficient of Mg_2Si, $Mg_{1.98}Bi_{0.02}Si$ and $Mg_{1.98}Ag_{0.02}Si$ depend on temperature.

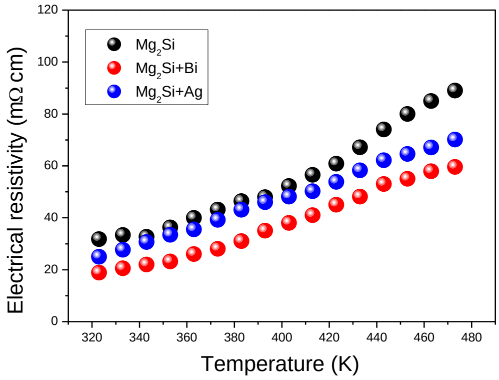

Fig. 4 Electrical resistivity of Mg_2Si, $Mg_{1.98}Bi_{0.02}Si$ and $Mg_{1.98}Ag_{0.02}Si$ depend on temperature.

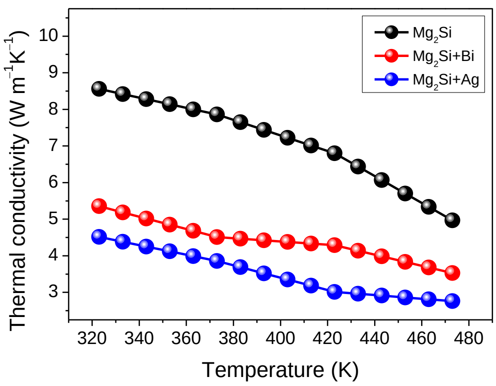

Fig. 5 Thermal conductivity of Mg_2Si, $Mg_{1.98}Bi_{0.02}Si$ and $Mg_{1.98}Ag_{0.02}Si$ depend on temperature.

Temperature dependence of thermal conductivity for Mg_2Si, $Mg_{1.98}Bi_{0.02}Si$ and $Mg_{1.98}Ag_{0.02}Si$ shows in Fig. 5. The doped samples can be decease thermal conductivity and Ag doped shows lowest thermal conductivity about 3 W/m K at 473 K as shown in Fig. 3 (c).

The Fig. 6 shows dimensionless figure of merit of Mg_2Si, $Mg_{1.98}Bi_{0.02}Si$ and $Mg_{1.98}Ag_{0.02}Si$. Doped samples show ZT value higher than un-dope and the Ag doped shows maximum ZT value about 0.47×10^{-2} at 473 K.

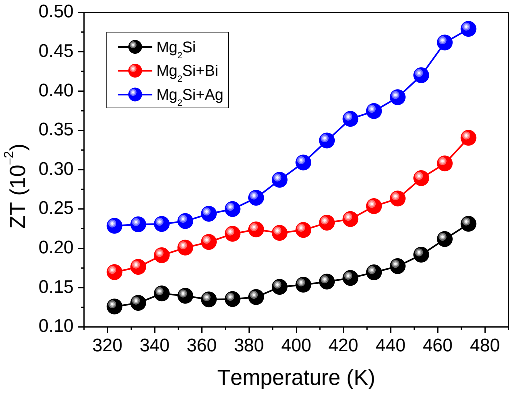

Fig. 6 Dimension less figure of merit of Mg_2Si, $Mg_{1.98}Bi_{0.02}Si$ and $Mg_{1.98}Ag_{0.02}Si$ depend on temperature.

CONCLUSION

The doped samples show thermal conductivity lower than un-dope and Ag doped show maximum decrease about 44.44% at 473 K. The electrical resistivity of doped samples was decreased, and decrease 33% for Bi doped at 473 K. The doped materials slightly decreased Seebeck coefficient about 4% and 16% for Ag and Bi doped, respectively. The highest ZT was found in Ag doped about 0.47×10^{-2} at 473 K.

REFERENCES

[1] L. Zheng, X. Zhang, H. Liu, S. Li, Z. Zhou, Q. Lu, J. Zhang, F. Zhang, Optimized nanostructure and thermoelectric performances of $Mg_2(Si_{0.4}Sn_{0.6})Sb_x$ solid solutions by in situ nanophase generation, J. Alloy. Compd. 671 (2016) 452–457.

[2] A.U. Khan, N.V. Vlachos, E. Hatzikraniotis, G.S. Polymeris, Ch.B. Lioutas, E.C. Stefanaki, K.M. Paraskevopoulos, I. Giapintzakis, Th. Kyratsi, Thermoelectric properties of highly efficient Bi-doped $Mg_2Si_{1-x-y}Sn_xGe_y$ materials, Acta Mater 77 (2014) 43–53.

[3] W. Liu, Q. Zhang, K. Yin, H. Chi, X. Zhou, X. Tang, C. Uher, High figure of merit and thermoelectric properties of Bi-doped $Mg_2Si_{0.4}Sn_{0.6}$ solid solutions, J. Solid. State. Chem. 203 (2013) 333–339.

[4] A.U. Khan, N. Vlachos and Th. Kyratsi, High thermoelectric figure of merit of $Mg_2Si_{0.55}Sn_{0.4}Ge_{0.05}$ materials doped with Bi and Sb, Scripta Mater. 69 (2013) 606–609.
[5] T. Sumphao, C. Thanachayanont, T. Seetawan, Design and Implementation of a Low Cost DAQ System for Thermoelectric Property Measurements, Procedia Eng. 32 (2012) 614 – 620.
[6] G. Kim, J. Kim, H. Lee, S. Cho, I. Lyo, S. Noh, B-W. Kim, S.W. Kim, K.H. Lee, W. Lee, Co-doping of Al and Bi to control the transport properties for improving thermoelectric performance of Mg_2Si, Scripta Mater. 116 (2016) 11–15.