

## The Effect of Sintering Temperature on Thermoelectric Property of Higher Manganese Silicide

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### Abstract

The Higher Manganese Silicide (HMS) was synthesized by solid state reaction and hot pressing. The various sintering temperature was started from 1123 K to 1223 K. The crystal structure and thermoelectric properties of samples were measured. The dimensionless figure of merit (ZT) was calculated from thermoelectric properties. The tetragonal structure of MnSi<sub>1.75</sub> was confirmed by X-ray diffraction (XRD). The Seebeck coefficient and Electrical resistivity are decreases, while the thermal conductivity increase when increasing sintering temperature. The maximum ZT of MnSi<sub>1.75</sub> was found in 1223 K of sintering temperature about 0.23 at 473 K.

**KEYWORDS:** Higher Manganese Silicide; Thermoelectric; Hot press

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### Introduction

Thermoelectric materials have been used for alternative energy wherewith it can convert heat into electricity. However, the efficiency of thermoelectric material is limited for a wide variety of applications. The efficiency of the thermoelectric materials is defined by dimensionless figure of merit,  $ZT = (S^2/\rho\kappa)T$ ,  $S$  is the Seebeck coefficient,  $\rho$  is the electrical resistivity,  $\kappa$  is thermal conductivities, and  $T$  is the absolute temperature. The Higher Manganese Silicide (HMS) was candidate for middle temperature of thermoelectric application due to the HMS showed high ZT in temperature ranges of 673 – 973 K. Recently, the undoped HMS was reported the ZT about 0.4 – 0.7 range by mechanical alloying and pulse discharge sintering [1], spark plasma sintering method [2, 3], hot press [4, 5], etc. In this work, has the objective for prepare the HMS by hot press method with various the sintering temperature from 1123 K to

1223 K to study the impact on thermoelectric properties.

### Materials and Methods

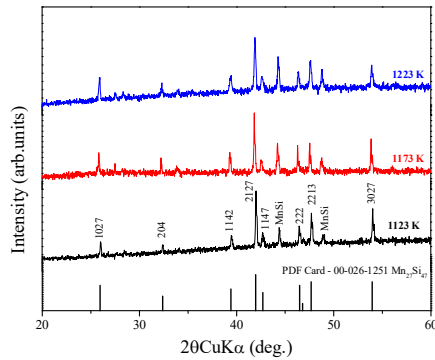
The Higher Manganese Silicide was synthesized by solid state reaction and hot press method. Mn (99%, Aldrich) and Si (99.9%, Aldrich) were used raw materials. The raw materials were weighed in atomic ratio and mixed by ball milling for 2 h. The powder were placed in the alumina crucible and calcined in quartz tube furnace at 1073 K for 1 h in Ar atmosphere. The calcined powder were pressed in graphite mold ( $\phi$ 20 mm) and heated in temperature 1123 – 1223 K under pressure 33 MPa for 1 h in Ar atmosphere. The pellets were cut in size of  $10 \times 10 \times 1$  mm<sup>3</sup> and  $3 \times 3 \times 15$  mm<sup>2</sup> for crystal structure analysis and thermoelectric property measurement, respectively.

The X-ray diffraction (XRD; Shimadzu 6100, Japan) was used for crystal structure characterization. The CuK $\alpha$  radiation at 40 kV, 30

mA and a scanning speed of 5°/min at 2θ steps of 0.02° were setups for XRD condition. The thermoelectric properties include Seebeck coefficient, electrical resistivity and thermal conductivity were measured by steady state method [6] at temperature ranges of 323 – 473 K.

### Results and Discussion

The powder XRD patterns of all samples are given in Fig. 1. The result of XRD show the main crystalline phase is Mn<sub>27</sub>Si<sub>47</sub> with tetragonal structure (space group P - 4n2) and agree with PDF Number 00-026-1251. In addition, the common impurity phase MnSi was found for all sintering temperature. The 3×3×15 mm<sup>3</sup> HMS pellets were set up in the copper probe for Seebeck coefficient and electrical resistivity measurement and show the result in Fig. 2 and 3, respectively.

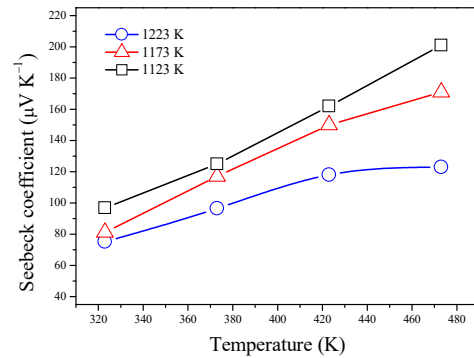


**Fig. 1** XRD pattern of MnSi<sub>1.75</sub> as sintering temperature 1123 K, 1173 K and 1223 K

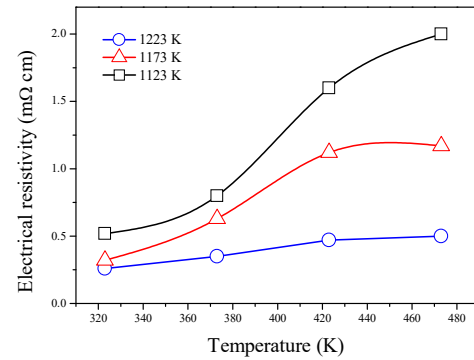
The Seebeck coefficient of all samples are positive value indicate that *p*-type thermoelectric material and decreases when sintering temperature increasing. The increase of *S* value with temperature indicate semiconductor behavior. The maximum of *S* value was found in 1123 K sintering sample about 201 μV K<sup>-1</sup> at 473 K.

The higher sintering temperature can reduce electrical resistivity which the 1223 K sample shows the lowest value about 0.5 mΩ cm at 473 K. The electrical resistivity of all samples increases with temperature increase.

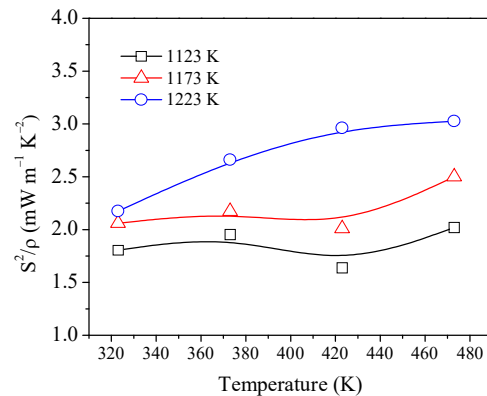
Fig. 4 shows the variation of the power factor (*S*<sup>2</sup>ρ) in function of temperature for sintered samples. The power factor shows evolutions of the electrical conductivity and Seebeck coefficient in function of temperature. The 1123 K and 1173 K of sintering samples are small increases while 1223 K sintering sample shows the large increase with temperature. The maximum *S*<sup>2</sup>ρ = 3.02 mW m<sup>-1</sup> K<sup>-2</sup> is observed for the 1223 K sintering sample.



**Fig. 2** Seebeck coefficient of MnSi<sub>1.75</sub> as sintering temperature 1123 K, 1173 K and 1223 K depend on temperature.



**Fig. 3** Electrical resistivity of MnSi<sub>1.75</sub> as sintering temperature 1123 K, 1173 K and 1223 K depend on temperature

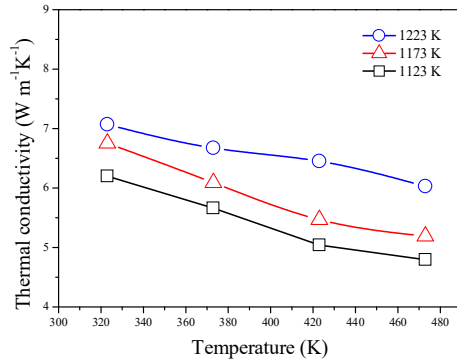


**Fig. 4** The power factor of MnSi<sub>1.75</sub> as sintering temperature 1123 K, 1173 K and 1223 K depend on temperature

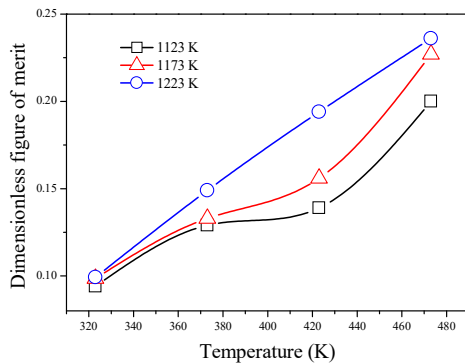
The thermal conductivity of sintered samples as a function of temperature is shown in Fig. 5. The thermal conductivity increases with sintering

temperature. The lowest  $\kappa = 4.79 \text{ W m}^{-1} \text{ K}^{-1}$  is observed for the 1123 K sintering sample.

Fig. 6 shows the variation of the dimensionless figure of merit in function of temperature for sintered samples. The ZT of 1223 K sample shows the highest value about 0.23 at 473 K due to shows high S and low  $\sigma$  although highest  $\kappa$ .



**Fig. 5** Thermal conductivity of MnSi<sub>1.75</sub> as sintering temperature 1123 K, 1173 K and 1223 K depend on temperature



**Fig. 6** Dimension less figure of merit of MnSi<sub>1.75</sub> as sintering temperature 1123 K (black line), 1173 K (red line) and 1223 K (blue line) depend on temperature

## Conclusion

The synthesis of MnSi<sub>1.75</sub> was completed by hot press method. The various sintering temperature was affected to thermoelectric properties by means of high sintering temperature render to decrease S and  $\rho$  but increase  $\kappa$ . The maximum power factor is observed in 1223 K sintering sample about  $3.02 \text{ mW m}^{-1} \text{ K}^{-2}$  at 473 K. The highest sintering temperature shows highest ZT about 0.23 at 473 K.

## References

- [1] T. Itoh, M. Yama, Synthesis of Thermoelectric Manganese Silicide by Mechanical Alloying and Pulse Discharge Sintering, *J. Electron. Mater.* 38(7) (2009) 925 – 929.
- [2] W. Luo, H. Li, Y. Yan, Z. Lin, X. Tang, Q. Zhang, C. Uher, Rapid synthesis of high thermoelectric performance higher manganese silicide with in-situ formed nano-phase of MnSi, *Intermetallics.* 19 (2011) 404 – 408.
- [3] G.B. Granger, M. Soulier, H.I. Mouko, C. Navone, M. Boidot, J. Leforestier, J. Simon, Microstructure investigations and thermoelectrical properties of a P-type polycrystalline higher manganese silicide material sintered from a gas-phase atomized powder, *J. Alloy. Compd.* 618 (2015) 403 – 412.
- [4] D. Shin, K.Jang, S. Ur, I. Kim, Thermoelectric Properties of Higher Manganese Silicides Prepared by Mechanical Alloying and Hot Pressing, *J. Electron. Mater.* 42(7) (2013) 1756 – 1761.
- [5] Y. Sadia, Z. Aminov, D. Mogilyansky, Y. Gelbstein, Texture anisotropy of higher manganese silicide following arc-melting and hot-pressing, *Intermetallics.* 68 (2016) 71 – 77.
- [6] T. Sumpao, C. Thanachayanont, T. Seetawan, Design and Implementation of a Low Cost DAQ System for Thermoelectric Property Measurements, *Procedia Engineering.* 32 (2012) 614 – 620.