

THERMOELECTRIC PROPERTIES OF HOT PRESSED $Mg_2Si_{1-x}Sn_x$ ALLOYS

Samunin A.Yu., Zaitsev V.K., Fedorov M.I., Konstantinov P.P.,
Isachenko G.N., Gurieva E.A.

A.F.Ioffe Physical-Technical Institute, Polytekhnicheskaya ul., 26, St.-Petersburg, 194021,
Russia

Contact author: samunin@yandex.ru,

Abstract

In the present work the samples of solid solutions $Mg_2Si_{1-x}Sn_x$ ($0 \leq x \leq 1$) have been produced by hot pressing in vacuum. Transport properties (electrical and thermal conductivities and Seebeck coefficients) of produced samples have been measured in wide temperature range. It is shown that the properties of the pressed samples are very similar to those of melted homogeneous samples. As the hot pressing does not changes essentially the properties of the material it can be used well in the thermoelectrics and thermoelement production.

Introduction

Mg_2X ($X = Si, Ge, Sn, Pb$) compounds for a long time attract the attention of researchers as perspective thermoelectrics. There are wide areas of solid solutions between these compounds [1].

Complex investigation of electrical properties, thermal conductivities and the features of band structure, which took place previously [1,2], has shown up that the solid solutions in Mg_2Si - Mg_2Sn system are very promising.

Long-time annealing is necessary to prepare homogeneous alloys by melting. Hot-pressing in vacuum or inert atmosphere can be used to decrease this time and get more homogeneous samples. Besides, this method give a chance to improve mechanical properties, to adjust serial issue of materials and considerably simplify the production technology of thermoelements.

In the present work main attention was directed at the investigation of $Mg_2Sn_{0.6}Si_{0.4}$ solid solution, because it is the most

effective material at temperatures up to 850K.

In addition the comparison of thermal conductivity of melted and pressed solid solutions $Mg_2Si_{1-x}Sn_x$ ($0 \leq x \leq 1$) was made.

Experiment

The set-up for hot-pressing in vacuum or in inert atmosphere has been created for the adjustment of method. It consist of modernized press P250 and the vacuum chamber developed by us. High-frequency generator has been used for heating of press-mold with material powder. Synthesized solid solutions of Mg_2Si - Mg_2Sn were crushed in a crusher. Size fraction of 100 – 350 μm was taken from this powder. Required quantity of the powder was put in the mold. The mold is done from steel with molding inserts and punches of graphite (fig.1). Pressing was made in vacuum (2Pa), temperature was higher than temperature of yield point and the effort on punches was 2.5 – 3.5 tons. Process temperature was measured by K-type thermocouple. Sample density was not less than 95-98% of theoretical one. Pressed samples were annealed for several days.

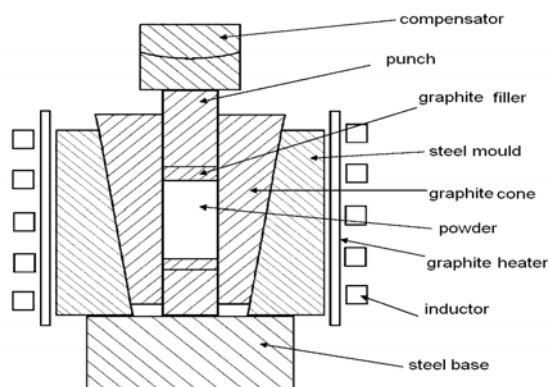


Figure 1: Press cell.

X-ray analysis show, that the solid solution samples are of single-phase and well formed.

The Seebek coefficient, electrical and thermal conductivities have been measured

on prepared samples in the temperature range 300 – 850K. Thermoelectric properties of some pressed and some bulk $Mg_2Sn_{0.6}Si_{0.4}$ solid solutions at room temperature are represented in tab.1. Temperature dependences of samples with marked * are shown at figures 2-4. Therewith figures 2 – 4 show the typical temperature dependences of thermal conductivity (fig.2), electrical conductivity (fig.3) and Seebek coefficient (fig.4) for $Mg_2Sn_{0.6}Si_{0.4}$ solid solution with different carrier concentration, prepared by hot-pressing. On the figures appropriate dependences of similar melted samples are shown for comparison.

Table 1: Thermoelectric properties at room temperature of pressed (a) and bulk (b) doped samples $Mg_2Sn_{0.6}Si_{0.4}$ solid solution.

a					b				
	α	σ	κ	Z		α	σ	κ	Z
	-112	2069	36,3	0,75		-113	2129	36,9	0,73
	-115	2312	33,5	0,98		-116	2065	36,2	0,76
*	-116	2106	33,4	0,84	*	-116	1999	37,1	0,73
*	-122	2000	35,0	0,86	*	-124	1979	35,1	0,86
	-127	1856	34,0	0,89		-129	2058	34,1	0,99
	-131	1830	30,6	1,03		-133	1843	34,7	0,94
*	-145	1360	28,6	0,97	*	-148	1361	29,5	1,01

Fig.5 show the comparison of thermal conductivity of melted and pressed $Mg_2Si_{1-x}Sn_x$ ($0 \leq x \leq 1$) solid solutions with low carrier density.

Fig. 2 – 5 and Tab.1 show that the properties of hot-pressed and melted samples distinguish insignificantly.

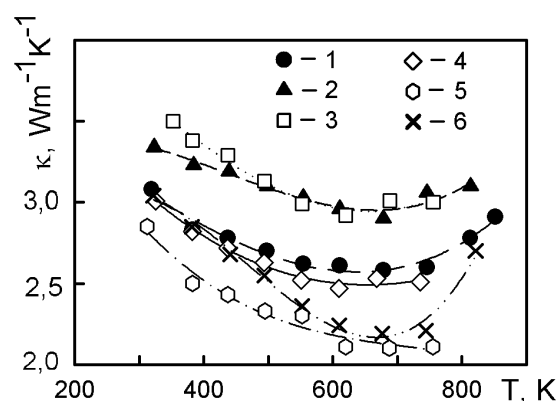


Figure 2: Temperature dependences of thermal conductivity for $Mg_2Sn_{0.6}Si_{0.4}$ solid solution with different carrier concentration, prepared by hot-pressing and melted samples. $n, 10^{20} cm^{-3}$: 1 - 1.9, 2 - 6.2, 3 - 2.1, 4 - 5.8, 5 - 6.8, 6 - 6.5.

Curves 1, 2, 6 – melted samples, curves 3, 4,5 – hot-pressing samples.

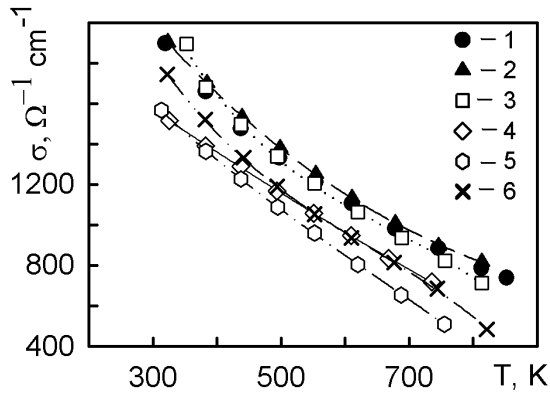


Figure 3: Temperature dependence of electrical conductivity for $Mg_2Sn_{0.6}Si_{0.4}$ solid solution prepared by hot-pressing and melted samples. Notations are the same as on fig.2.

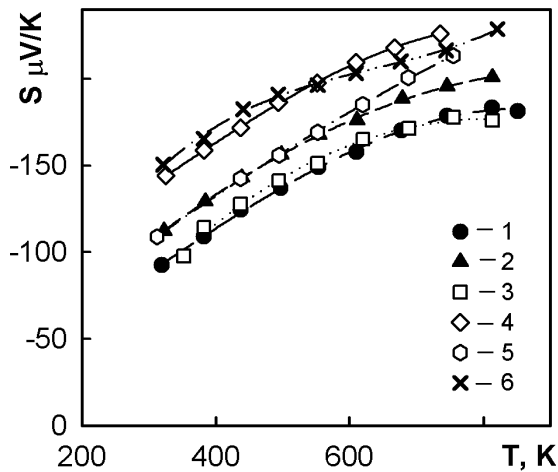


Figure 4: Temperature dependence of Seebeck coefficient for $Mg_2Sn_{0.6}Si_{0.4}$ solid solution prepared by hot-pressing and melted samples. Notations are the same as on fig.3. Curves 3 and 1 are shifted down at $40\mu V/K$, than the measured values and the curves 2 and 5 at $20\mu V/K$ for visualization.

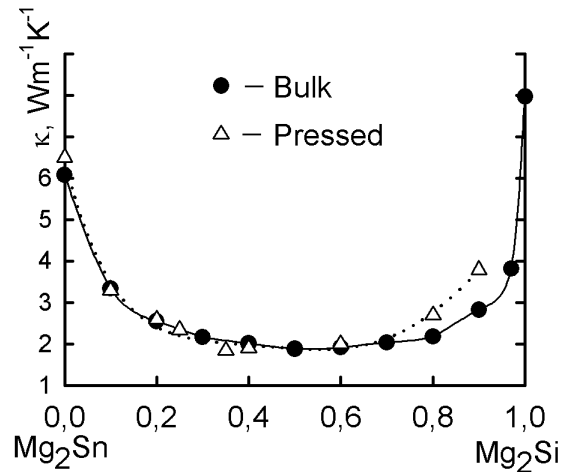


Figure 5: Lattice thermal conductivity of $Mg_2Si_{1-x}Sn_x$ ($0 \leq x \leq 1$) solid solutions at room temperature.

Conclusions

The method of hot-pressing in vacuum of Mg_2Si - Mg_2Sn solid solutions has been developed. This method allows to prepare homogeneous samples with the density, exceeding 95% of theoretical density. Thermoelectric properties of hot-pressed material practically coincide with the properties of melted material of the highest thermoelectric figure of merit. Thermoelectric properties of the pressed samples have been reproduced reliably. The method permits to appreciably decrease the time of material production and simplify fabrication technique of thermoelements based on this material.

Acknowledgements

Authors sincerely thank E.P.Zajats for the help in measuring thermoelectric properties and N.F.Kartenko for X-ray analysis of the samples.

The work was supported by the program of Department of Physical Sciences of the Russian Academy of Sciences "New principles of energy conversion in semiconductor structures", the project: "Thermoelectrics based on silicon compounds - new approach to the increase of efficiency of thermoelectric energy conversion".

References

1. Zaitsev V.K., Fedorov M.I., Gurieva E.A., Eremin I.S, “Thermoelectrics on the Base of Solid Solutions of Mg_2B^{IV} Compounds ($B^{IV}=Si,Ge,Sn$)”, in Thermoelectrics Handbook: macro to nano, edited by D.M.Rowe, CRC Press 2006, chapter 29.
2. Zaitsev V.K., Fedorov M.I., Gurieva E.A., Eremin I.S., Konstantinov P.P., Samunin A.Yu., Vedernikov M.V. “Highly effective $Mg_2Si_{1-x}Sn_x$ thermoelectrics”. Phys. Rev. B, 2006, v.74, N4, p.045207