Investigation of properties of n-type Mg_2Si-Mg_2Sn solid solutions prepared by hot pressing.

A.U.Samunin, V.K.Zaitsev, M.I.Fedorov, P.P.Konstantinov

A.F.Ioffe Physical-Technical Institute
Polytekhnicheskaya ul., 26, Sankt-Petersburg, 194021, Russia
E-mail: samunin@yandex.ru, Phone: +7 812 514 19 97, Fax: +7 812 515 67 47

Abstract

Hot-pressing technology has been developed and the samples based on Mg_2Si-Mg_2Sn solid solutions with the density, exceeding 95% of theoretical density, have been prepared.

The thermoelectric properties (Seebek coefficient, electrical and thermal conductivities) have been measured in the temperature range 300 – 850K.

Introduction

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

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

It is significant, that when preparing Mg_2Si-Mg_2Sn solid solutions by crystallization from liquid phase the following features have been observed:

1. strong segregation because of different specific gravity.
2. strong intragranular and intergranular segregation because of big difference between liquidus and solidus temperatures.
3. high capability for crystallization results in large grains formation (Fig.1.)

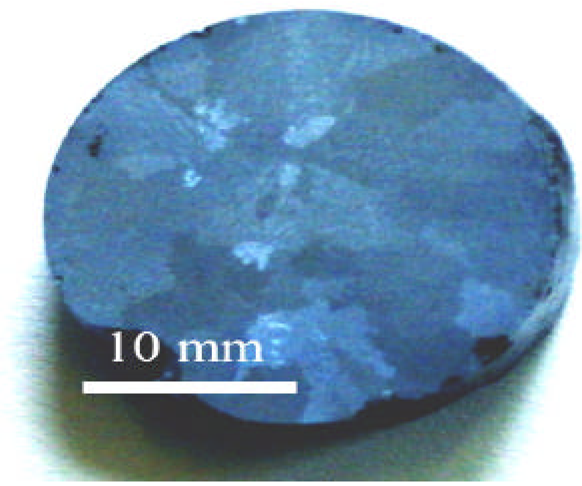

Figure 1: Structure of grains of solid solution $Mg_2Sn_{0.6}Si_{0.4}$

Therefore because of these crystallization features a long time of annealing for homogenization of the alloys is required.

In the present work main attention was directed at investigation of $Mg_2Sn_{0.6}Si_{0.4}$ solid solution, as the most effective at the temperature up to 850K.

Fig.2 shows the dynamics of microstructure changing depending on the annealing time.

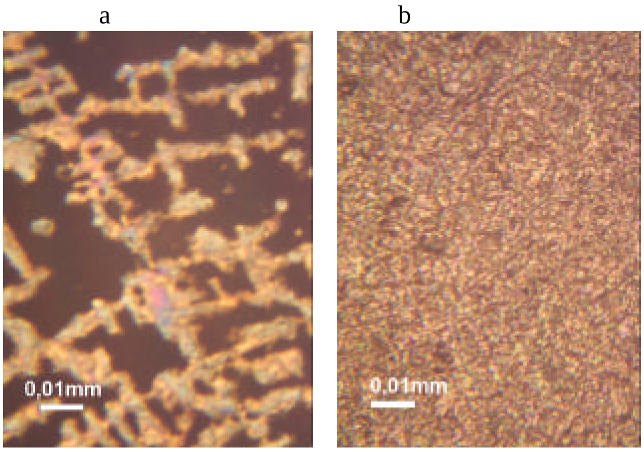

Figure 2: the dynamics of microstructure changing of $Mg_2Sn_{0.6}Si_{0.4}$. a) before annealing; b) after long annealing.

The method of hot-pressing in vacuum or inert atmosphere could be used to decrease the annealing time and to produce more homogeneous samples. Besides, this method gives a chance to improve mechanical properties, to develop industrial methods of the materials manufacturing and considerably simplify thermoelements production.

The set-up for hot-pressing in vacuum or in inert atmosphere was developed and manufactured for the method realization.

Experiment

The set-up consists of modernized hydraulic press P250 and the vacuum chamber developed by us. High-frequency generator has been used for the heating of press-mold with the powder of material. The mold has been done from steel with molding inserts and punches of graphite. Synthesized solid solutions of Mg_2Si-Mg_2Sn were crushed in ball mill or in crusher. Size fraction 100 – 350 μm was taken from this powder. Required quantity of powder was put in the die. The following pressing parameters have been used: vacuum - 2Pa, temperature - higher than temperature of yield point and pressure load on punches - 2.5-3.5 tons. Temperature of the die has been measured by K-type thermocouple. Density of samples was about 95-98% of theoretical. Pressed samples were annealed during several days.

X-ray analysis has shown, that the samples are single-phase and well organized solid solutions. It has confirmed by the lattice distance measurement, that the composition of the solid solution is $Mg_2Sn_{0.6}Si_{0.4}$.

The Seebek coefficient, electrical and thermal conductivities have been measured in the temperature range 300 – 850K. Figures 3 – 6 show typical temperature dependences of thermal conductivity (fig.3), electrical conductivity (fig.4), Seebek coefficient (fig.5) and dimensionless thermoelectric figure of merit (fig.6) for $Mg_2Sn_{0.6}Si_{0.4}$ solid solution with different carrier concentration, prepared by hot-pressing. Corresponding dependencies of similar melted samples are shown for comparison on these figures.

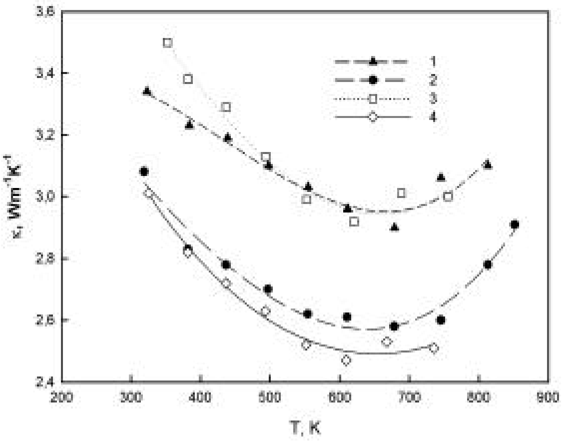

Figure 3: Temperature dependence of thermal conductivity for hot-pressed and melted samples of $Mg_2Sn_{0.6}Si_{0.4}$ solid solution with different carrier concentration, n,$10^{20}cm^{-3}$: 1 -1.9, 2 - 6.2, 3 – 2.1, 4 -5.8. Curves 1, 2 – melted samples, curves 3 and 4 – hot-pressed samples.

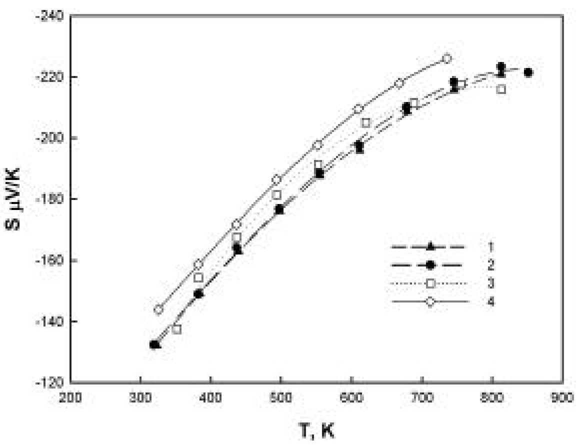

Figure 4: Temperature dependence of electrical conductivity for hot-pressed and melted samples of $Mg_2Sn_{0.6}Si_{0.4}$ solid solution. Notation is the same as on fig.3.

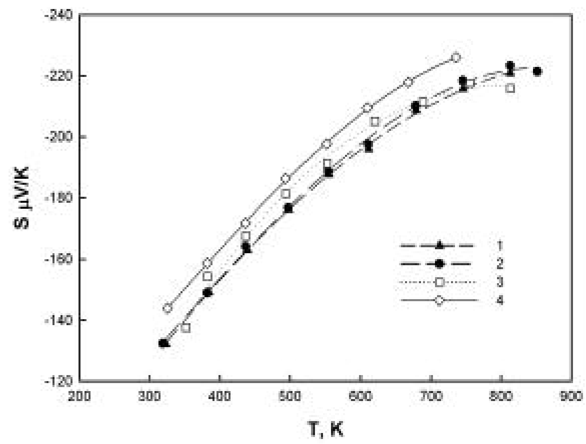

Figure 5: Temperature dependence of Seebek coefficient for for hot-pressed and melted samples of $Mg_2Sn_{0.6}Si_{0.4}$ solid solution. Notation is the same as on fig.3.

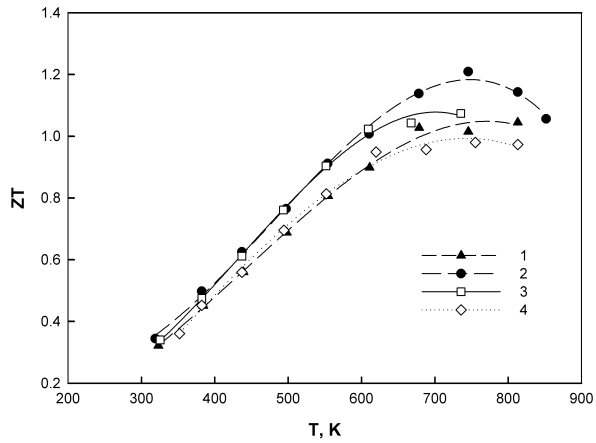

Figure 6: Temperature dependence of dimensionless thermoelectric figure of merit for hot-pressed and melted samples of $Mg_2Sn_{0.6}Si_{0.4}$ solid solution. Notation is the same as on fig.3.

Fig. 3 – 6 show, that the properties of hot-pressed and melted samples have been distinguished insignificantly.

Conclusions

The method of hot-pressing in vacuum for Mg_2Si-Mg_2Sn solid solutions is developed. It allows to prepare homogeneous samples with the density, exceeding 95% of theoretical density. High repeatability of thermoelectrics properties has been achieved. The method permits to appreciably decrease the time of material production. Thermoelectric properties of hot-pressed material practically coincide with those of melted material.

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