

Microstructure and thermoelectric properties of p-type Mg₂Sn prepared by RF induction melting

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Abstract – Silver-doped Mg₂Sn bulk crystals were prepared by RF induction melting in a boron nitride crucible under 1 atm Argon. The microstructure of the ingots was analyzed by X-ray diffraction, scanning electron microscopy and energy-dispersive X-ray spectroscopy. The Hall coefficient, Seebeck coefficient, electrical conductivity, and thermal conductivity were measured in the temperature range of 80–700 K to establish a correlation between the composition, microstructure and thermoelectric properties. The whole process of preparing one ingot needed only 5 hours, and the thermoelectric properties are very similar to those of the near-single-crystal ingots prepared by the Bridgman method requiring 100 hours.

Thermoelectric materials which convert heat directly to electricity or electricity to refrigeration are of interest for applications in power generation and as cooling devices. The efficiency of a thermoelectric material is characterized by its figure of merit $ZT = S^2\sigma/\kappa$, where S is the Seebeck coefficient, σ the electrical conductivity, and κ the thermal conductivity. The compounds Mg₂X ($X = \text{Si, Ge, Sn}$) are promising thermoelectric materials since they have high ZT and the preparation difficulties encountered in the past due to the high vapor pressure and high reactivity of Mg are being overcome. Zaitsev *et al.* [1] has reported a $ZT \approx 1.1$ for Mg₂Si_{1-x}Sn_x solid solutions prepared by direct co-melting followed by lengthy annealing. In this paper we report on the microstructure and thermoelectric properties of Mg₂Sn ingots doped with Ag (0 – 1 at. %) and prepared by RF induction melting, a very simple and rapid method.

High-purity Mg (4N), Sn (6N), and Ag (3N) were mixed with the desired atom ratio, and then loaded into a high-purity boron nitride crucible (BN) with a tight screw-top lid which was then sealed in a graphite container with screwed cover. The graphite container, attached with a thermocouple, was suspended inside a RF induction coil inside a stainless steel chamber, and the charge was melted under 1 atm Ar gas. The system was heated to 1100 K, held there for 10 minutes and then cooled at a rate of about 20 K/min. The entire process of preparing an ingot lasted only 5 hours. The obtained Mg₂Sn ingots are dense and homogenous with perfect large-grain polycrystals. The undoped sample is single-phase Mg₂Sn while the heavily doped samples contain a uniformly dispersed Ag₃Mg phase as shown in Figure 1.

Measurements of the Hall coefficient, thermal conductivity, Seebeck coefficient and electrical conductivity were carried out at 80 – 700 K. The results are comparable to those of the near-single-crystal Mg₂Sn ingots prepared recently by us [2] at very slow cooling rates using the vertical Bridgman method. Representative results of the Seebeck coefficient and electrical conductivity are shown in Figure 2. The undoped sample shows a transition from p -type conduction at low temperatures to n -type conduction above 250 K that is characteristic of lightly doped semiconductors. Samples doped with Ag show p -type behavior in the whole temperature range of measurement, and the room temperature electrical conductivity increases by two orders when silver is added to the Mg₂Sn compound. These results demonstrate that the RF induction and Bridgman methods produce ingots of comparable quality. This is very encouraging since the RF induction melting is a much simpler and rapid method, and thus of lower cost.

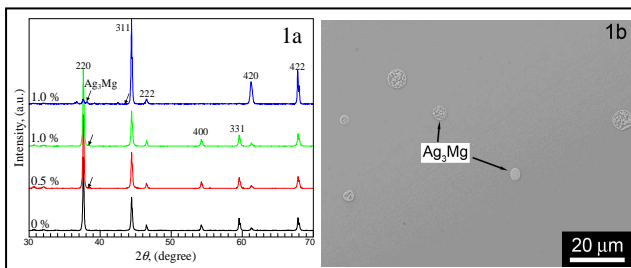


Figure 1: Microstructure of Mg₂Sn prepared by RF induction melting: (a) XRD spectra of the ingots doped with 0–1.0 at. % Ag; (b) SEM of the ingot doped with 0.5 at. % Ag

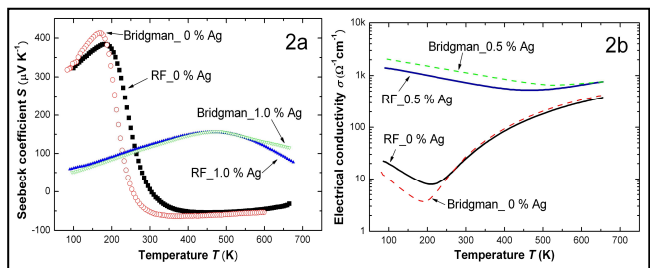


Figure 2: Representative results of thermoelectrical properties of Mg₂Sn prepared by RF induction melting: (a) Seebeck coefficient; (b) electrical conductivity

[1] V.K. Zaitsev, M.I. Fedorov, E.A. Gurieva, I.S. Eremin, P.P. Konstantinov, A.Y. Samunin, and M.V. Vedernikov, Phys. Rev. B. 74 (2006) 045207

[2] H.Y. Chen and N. Savvides, Mater. Res. Soc. Symp. Proc. 2009 (accepted).