THE INFLUENCE OF HIGH GRAVITY IN PbSn EUTECTIC ALLOY

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ABSTRACT

The study of materials processed in centrifuges improves the understanding of the acceleration influence in the convection behavior in materials processing. This work aims to study the influence of high gravity in PbSn eutectic alloy solidification using a small centrifuge designed and built in the Associate Laboratory of Sensors and Materials of the Brazilian Space Research Institute (LAS/INPE). The samples were analyzed by densitometry and scanning electron microscopy (SEM).

Keywords: High Gravity; Centrifuge; Eutectic Alloy; Solidification.

INTRODUCTION

The solidification of metals and alloys are strongly influenced by gravity from the initial stages of nucleation and grain growth (1). The study of non-equilibrium solidification fundamentals is important to understand metastable phases (2). The formation of dendritic structures in PbSn alloy is related to the presence of sedimentation and convective flow during solidification, and the size of these structures is connected to the solidification time (3).

All over the world, relatively large centrifuges are expensive and rarely available for crystal growth experiments. To overcome this difficulty, a small centrifuge was designed and built in the LAS/INPE, which provides an acceleration of up to 5g (4). This centrifuge was upgraded with new technologies, improving its performance (5).

Tin-lead eutectic alloy is a solder for metals widely used in electrical and electronic industries. The alloy have a regular eutectic structure and possess a low cost material that provides convenient physical properties such as low melting point, low vapor pressure, and does not react with the wall of the quartz ampoule, making it an ideal material for solidification studies of eutectic alloys.

The objective of this work is to study the influence of high gravity in PbSn eutectic alloy solidification.

EXPERIMENTAL

The alloy was prepared from pure lead (99.998 %) and tin (99.9999 %). A mass of 20 grams of the alloy was put into a quartz ampoule with 8 mm diameter and 150 mm length. The ampoule is evacuate (10^{-6} Torr) and sealed, one final growth ampoule is shown in Fig. 1.

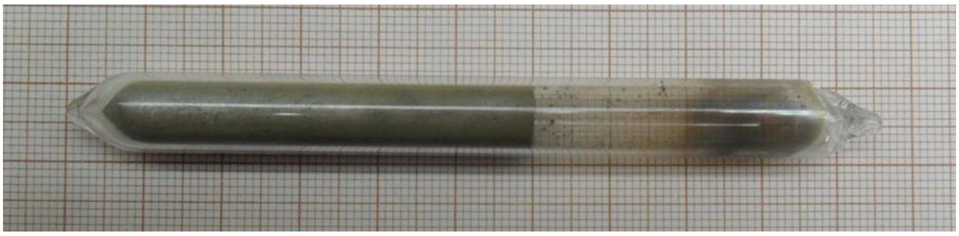

Fig. 1. Ampoule of PbSn eutectic alloy used in the experiments.

The solidification experiments were performed with the centrifuge furnace of the LAS/INPE (Fig. 2) in accordance with the scheme and furnace temperature gradient shown in Fig. 3.

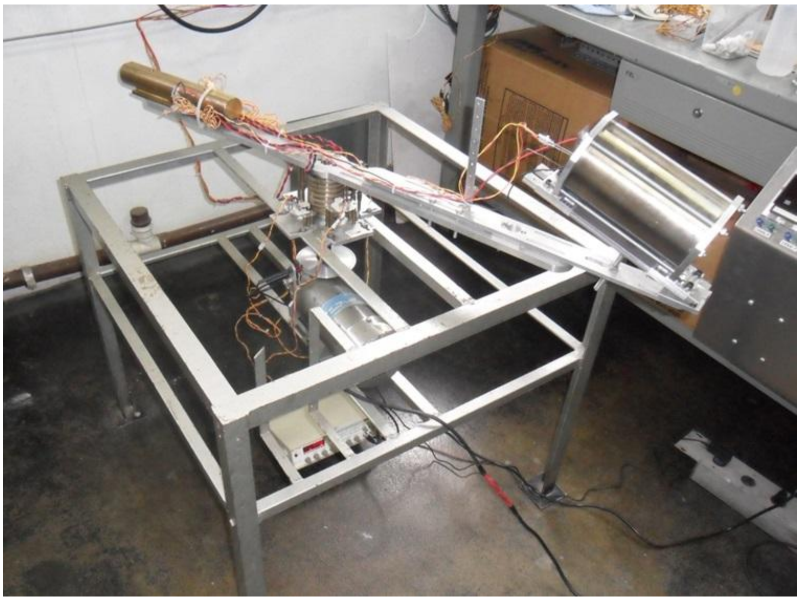

Fig. 2. Photo of LAS/INPE centrifuge

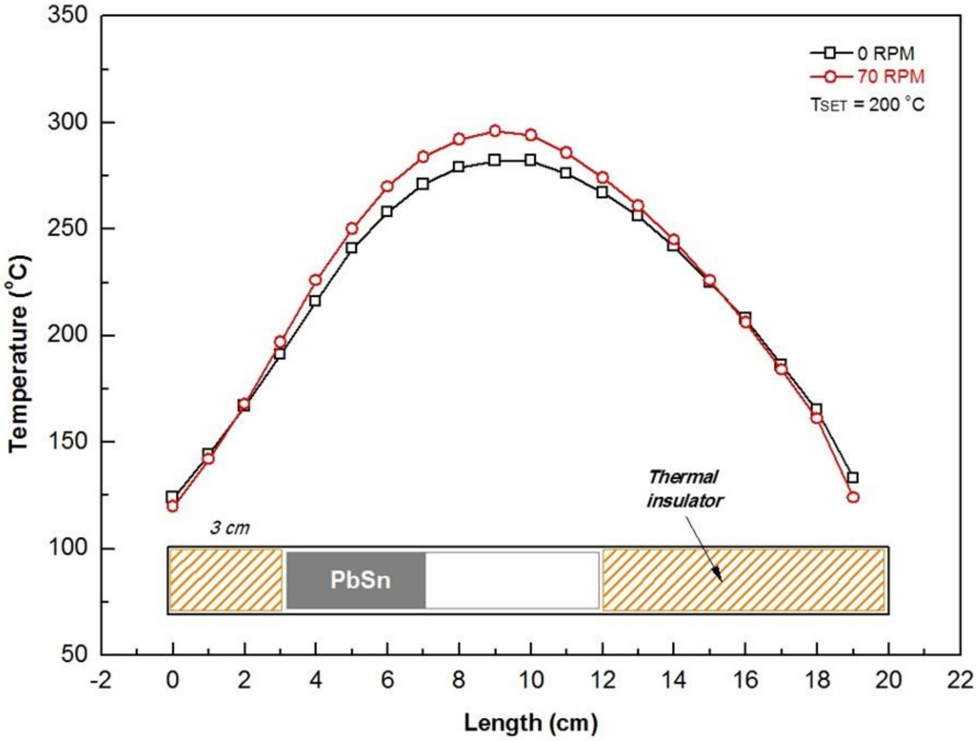

Fig. 3. Temperature gradient of the solidification furnace of centrifuge and schematic diagram of position of ampoule.

Two experiments were realized: with the furnace standing vertically at 1 g and with the furnace rotating at 3.5 g, being 1 g the gravity acceleration on earth (9.8 m/s^2). In the second experiment, the furnace was adjusted with an angle of 74º relative to the Earth gravity vector and heated up to 200 °C for about 2 hours. The motor was turned on at 70 rpm and, after 5 minutes, the furnace temperature was switched off and the motor kept running at constant speed during the solidification process of the sample. The same thermal parameters used with the rotating furnace were used for the solidification made with the standing furnace. Fig. 4 shows the solidification curve of the samples obtained by data-logger.

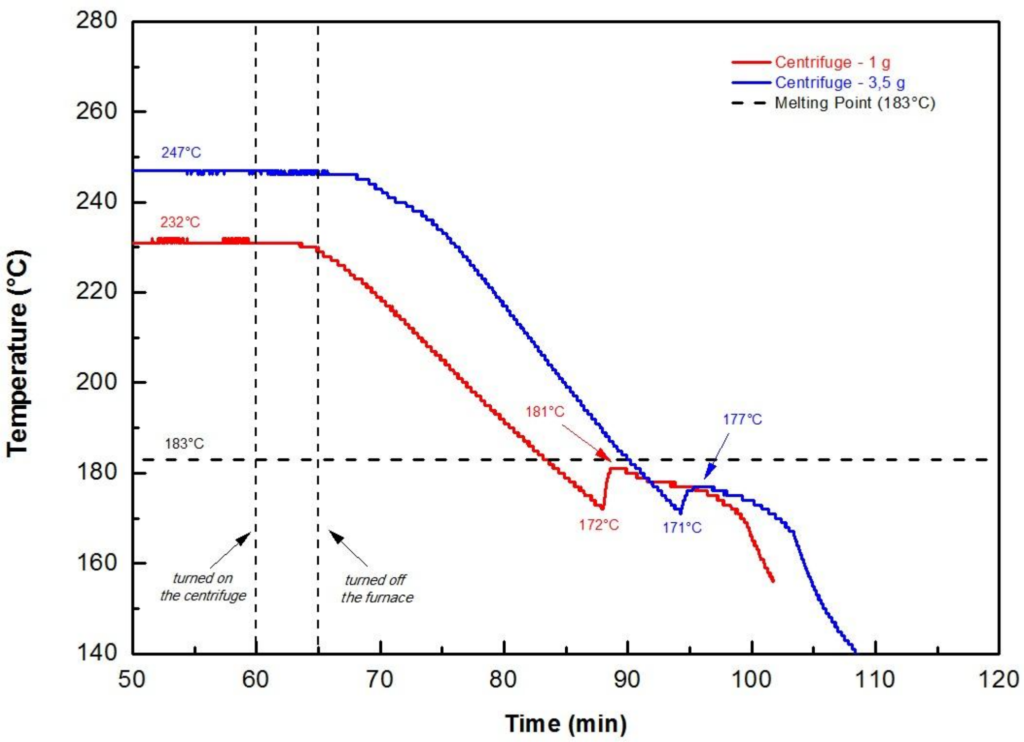

Fig. 4. Solidification curve of PbSn eutectic alloy.

Two samples were made in each growth method and, after solidification, were cut into several discs and its density was determined by Archimedes method to calculate the solute distribution versus the solidified fraction (fs). The other sample was cut in the longitudinal direction. After polishing and etching, the structure morphologies of the samples were examined using an electronic microscope JEOL/SEM model 5310.

RESULTS AND DISCUSSION

In the Pb-Sn system, an aligned lamellar structure was obtained by directional solidification. The directionally solidified Pb-Sn eutectic alloy produces parallel strips alternating the Pb-rich solid solution and the Sn-rich solid solution, respectively [6, 7].

The graph (Fig. 5a) shows that the density profile determined by the Archimedes method in this work and the density value found in the literature. A general expression (Eq. (A)) can be used to determine the composition x ($0 \leq x \leq 1$) of eutectic alloys $M_{1-x}N_x$, in function of the density, where ρ, m and A are respectively the density, mass and atomic weight. The formula is based on the rule of mixture, such as used by An et al. in the deduction of the general equation for the density of isomorphic binary alloys [8]. Based on this equation, Fig. 5b shows the graphics of the solute distribution versus the solidified fraction (fs).

$$x = \frac{(\rho_M - \rho)}{\left(1 - \frac{A_N}{A_M}\right)\rho_M - \left(1 - \frac{A_N \rho_M}{A_M \rho_N}\right)\rho}, \qquad (0 \le x \le 1) \tag{A}$$

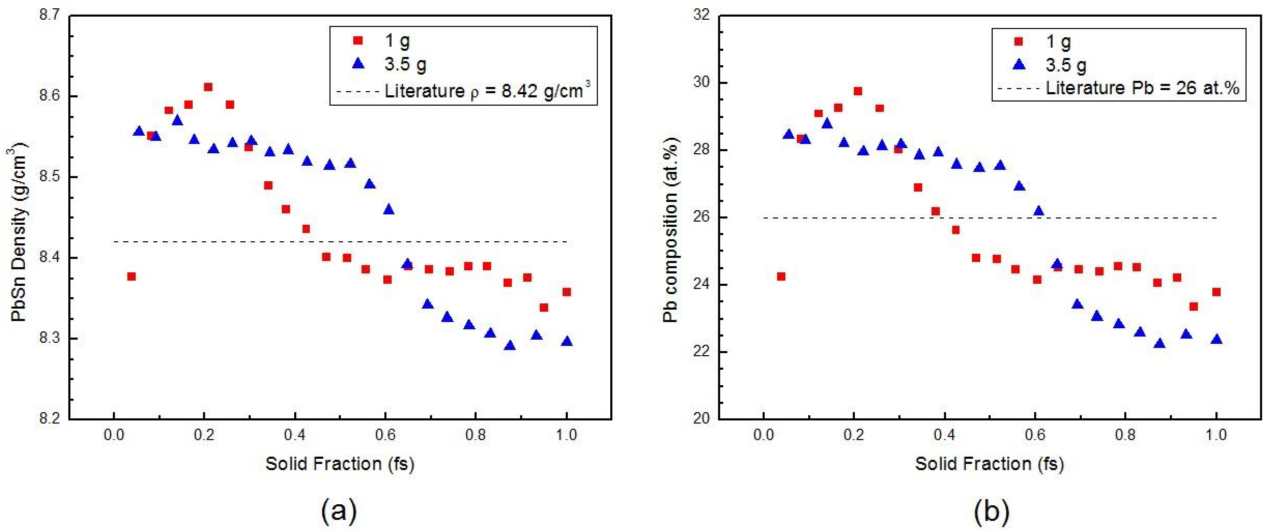

Fig. 5. (a) Average density of PbSn eutectic alloys as function of the solidified fraction; (b) Lead composition profile for 1 g and 3.5 g PbSn eutectic alloy analyzed by densimetric technique.

The graphics shows that, at the beginning of samples, the PbSn density is well above the nominal values for both samples: about 50% of the solidified fraction (fs) for samples grown in 1g and between 10% and 30% of fs for samples grown in 3.5 g. This is due to accumulation of lead caused by natural convection and sedimentation.

Figures 5a and 5b show the variation in density and the variation of the solute (lead) in the eutectic PbSn alloy solidified by 1 g and 3.5 g. It is observed in Fig. 5a for sample grown at 1 g that the PbSn density values rise in the interval 0 < fs < 0.2 due to accumulation of lead (show Fig. 5b) caused by natural convection and sedimentation, decreases in the interval 0.2 < fs < 0.6 and present a constant profile in the interval 0.6 < fs < 1. As for 3.5 g the convection increases and the lead accumulates in a bigger solidified fraction, 0 < fs < 0.6, decreasing strongly in the interval 0.6 < fs < 1.

Fig. 6 and 7 shows the images, obtained by electron backscattering, of the samples solidified by centrifuge furnace. Regions with different eutectic compositions are formed, due to the solute gradient caused by the gravity, resulting in diverse microstructures. It is observed the presence of two eutectic phases: an α phase rich in Pb (light gray color) and a β phase rich in Sn (dark gray color).

In Fig. 6 (radial direction analysis) dendritic structures (in the α phase) over irregular eutectic structures were observed at the beginning of the both samples (fs < 0.50 in 1 g and fs < 0.25 in 3.5 g), and lamellar eutectic structure plus irregular eutectic structures in the remainder of the samples.

In Fig. 7 (longitudinal direction analysis) dendritic structures (in the α phase) over irregular eutectic structures were observed at the beginning of the both samples (fs < 0.50), and lamellar eutectic structure plus irregular eutectic structures in the remainder of the sample grown in 1 g, whereas dendritic structures (in the β phase) over irregular eutectic structures is observed in the remainder of the sample of 3.5 g.

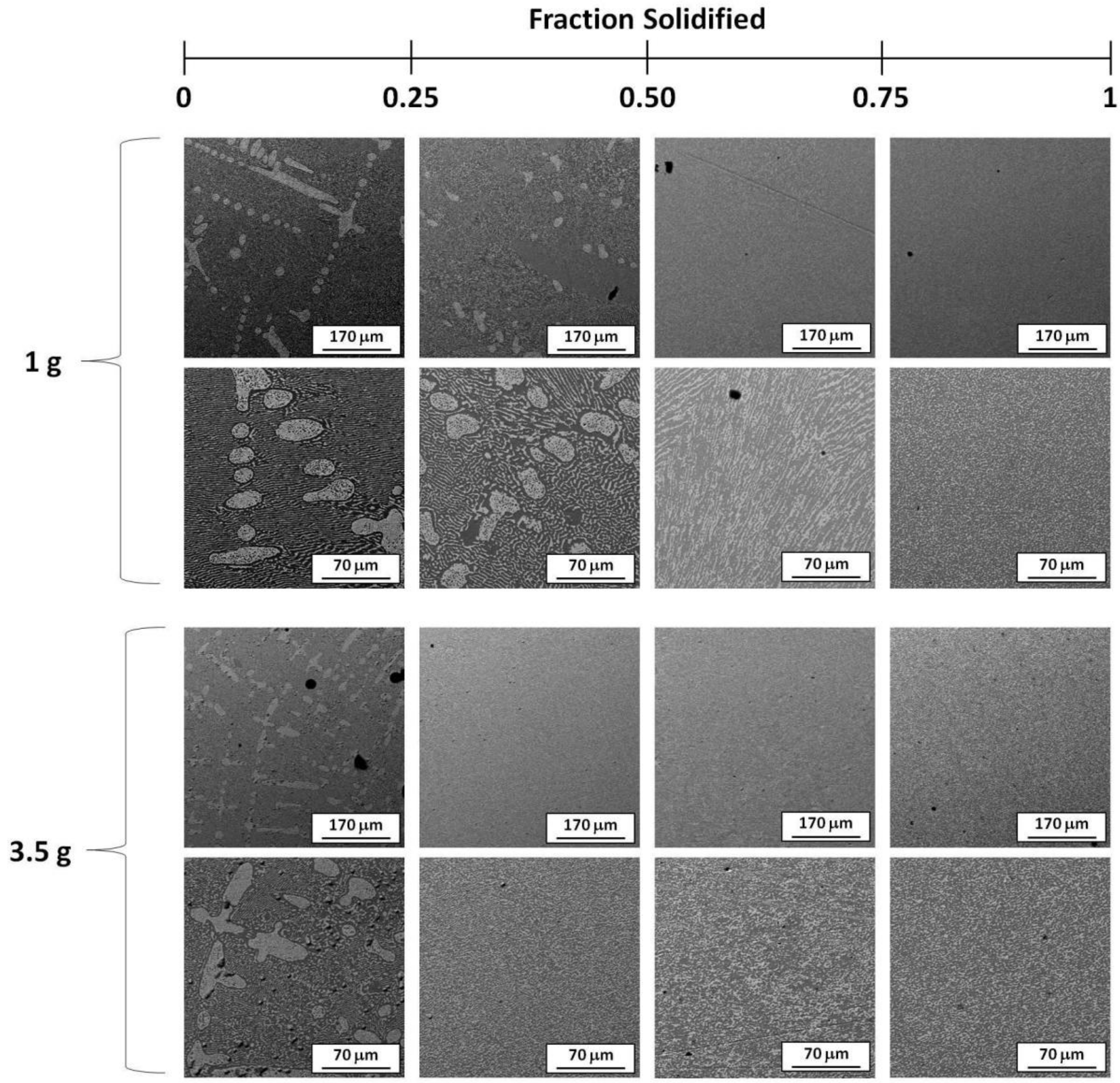

Fig. 6. SEM images of the radial direction analysis the PbSn eutectic obtained by centrifuge.

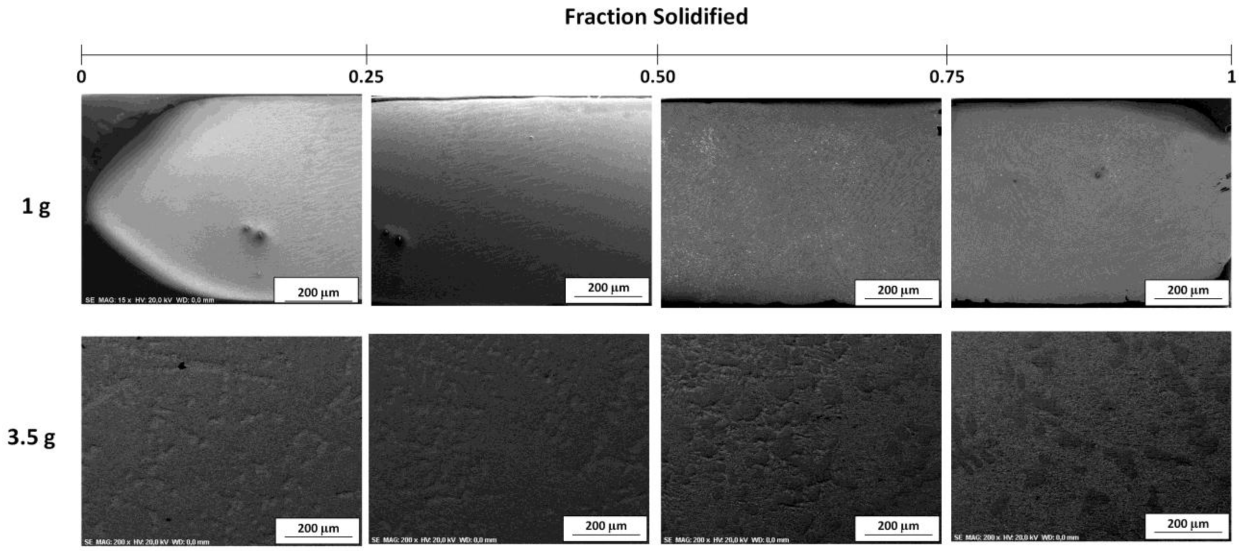

Fig. 7. SEM images of the longitudinal direction analysis the PbSn eutectic obtained by centrifuge.

CONCLUSION

PbSn eutectic alloys were solidified by centrifuge in high gravity. The samples solidified in 3.5 g had density and solute distribution profiles more stable and a lower region with dendritic structure plus irregular eutectic structure (fs < 0.25) as compared with the sample grown in 1 g.

References

1. REGEL, L. L.; WILCOX, W. R. **Materials Processing In High Gravity,** New York: Plenum Press, 1994.
2. TOLEDO, R. C.; FREITAS, F. E.; AN, C. Y.; BANDEIRA, I. N. Containerless Solidification of Eutectic PbSn Alloy Droplets in a Drop Tube. **Materials Science Forum,** v. 727-728, p.1633-1637, 2012.
3. AN, C. Y.; TOLEDO, R. C.; BOSCHETTI, C.; RIBEIRO, M. F.; BANDEIRA, I. N. Solidification of Lead Tin and Lead Telluride Eutectic Alloys in Microgravity. Microgravity Science And Technology, v. 25, n. 5, p.267-273, fev. 2014.
4. AN, C. Y.; RUSSO, L. C.; RIBEIRO, M. F.; BANDEIRA, I. N. A Low Cost Centrifuge for Materials Processing in High Gravity. In: **Centrifugal Materials Processing**. New York: Plenum Press, p.115-119, 1997.
5. FREITAS, F. E. **Use of a low cost centrifuge for study of the influence of high gravity in PbSn eutectic alloy solidification.** 2011. Coursework (Degree in Materials Engineering) - UNIVAP, São José dos Campos, 2011.
6. KARAKAYA, I.; THOMPSON, W. T. The Pb-Sn (Lead-Tin) system. **Journal of Phase Equilibria**, v.9, n.2, p.144-152, 1988.
7. TU, K. N.; ZENG, K. Tin–lead (Sn-Pb) solder reaction in flip chip technology, ***Materials Science and Engineering R***, v.34, n.1, p.1-58, 2001
8. AN, C. Y.; BANDEIRA, I. N.; ROWE, D. M.; MIN, G. An exact density formula for substitutional solid solution alloys. **Journal of Materials Science Letters,** v.13, p. 1051-1053, 1994.