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Solidification of Sn–Pb alloys: Experiments on the influence of the initial concentration

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ABSTRACT

A comparative study among solidification experiments for three selected alloys (Sn–3 wt.%Pb, Sn–6.5 wt.%Pb and Sn–10 wt.%Pb) was conducted on a benchmark experiment model under the same experimental conditions. The goal of this paper is to analyze the effect of variation in concentration on the solidification process with respect to different aspects: thermal, dynamic, structure, and morphology of segregation. Experimental results consist of instantaneous temperature maps provided by a lattice of 50 thermocouples welded on the large crucible side and post-mortem characterizations of the samples, such as X-ray imaging, solute local composition and metallography. Measurement of the instantaneous temperature field and numerical computation of liquid–solid interface evolution allows us to evaluate the effect of variation in concentration on thermosolutal convection behavior. Experimental results show that an increase in concentration greatly enhances the mechanism of the columnar-to-equiaxed transition (CET) and leads to refinement of the equiaxed structure. However, a significant effect of solutal element (lead) stratification is observed, which can slow down thermosolutal convection, in particular for large concentrations. Especially, without any stirring lead segregation which likely occurs during the melting phase may suppress natural convection. Furthermore, lead stratification is significantly reduced when electromagnetic stirring is opposed to natural convection before the solidification phase begins.

1. Introduction

The behavior of the solidification process of metallic binary alloys is different from that of pure metal. First, solidification takes place over a temperature range and not at a fixed point. Second, solid and liquid zones are no longer separated by a distinct front, but by a complex region made up of dendrites and liquid, known as the mushy zone. Third, one component is preferentially rejected into the adjacent liquid zone to form a solute-rich layer at the interface front. A large number of experimental and numerical works have described the effects of such factors, such as control of cooling temperature (directional solidification) and initial concentration [1–3]: the main experimental parameters of controlled solidification are cooling rate (CR), temperature gradient (ΔT) and nominal concentration (C_0). A real solidification problem including heat and mass transfer and phase changes at different stages was described by a variety of authors such as Kurz and Fisher [4]. L.S. Chao et al. [5] studied experimentally the effects of different initial concentrations of Sn–Pb alloys on the final microstructure of the ingot. They observed a non-lamellar eutectic structure when initial concentration was closest to eutectic composition. Moreover, a transition mechanism between the columnar and equiaxed structure was also observed, the presence of which increases with initial concentration. Jin et al. [6] investigated in detail the effect of nominal concentration (C_0) on the size of the mushy zone, and revealed the extent of its influence, especially on solid fraction distribution. Nucleation of solidified grains, followed by growth occur when melt temperature drops below the nucleation temperature T_N which is below the liquidus temperature T_L. For further details the reader is referred to Kurz and Fischer [4]. When undercooling ($T_L - T_N$) exceeds the critical value required for formation of nuclei, the nucleation rate rises. In the case of equiaxed growth, the nucleation stage will continue until recalescence occurs. At the growth stage, grain radius increases due to adherence of liquid atoms to the solid/liquid interface until impingement takes place. In the impingement stage, the liquid between grains

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solidifies, but the grain radii do not change. It is at this level that the final grain morphology is determined.

This present paper is based essentially on a comparative experimental study performed on a benchmark solidification experiment between three different Sn–Pb alloys (3%, 6.5% and 10%). These alloys are characterized by their thermo-physical properties available, as well as low liquidus temperature: 228.14 °C, 223.64 °C and 219.16 °C respectively. The purpose of this paper is primarily to investigate a controlled solidification experiment and to evaluate the effect of nominal concentration (C_0) on the fluid flow induced by thermosolutal convection and its consequence on macrostructures, solutal distribution and segregation morphology.

2. Experimental procedure

A benchmark solidification model is established and validated to study the effect of different concentrations on the solidification process. The experiment device consists in four major parts:

1. Fifty thermocouples were fixed on one of the largest surfaces of the crucible by laser-welding; on the opposite surface, the other sixteen thermocouples are arranged in the relative positions in order to calibrate temperature measurement. The interval distance in either the horizontal or vertical direction is 1.0 cm (see Fig. 1). Temperature fields and the evolutions are recorded during the melting/solidifying process by this thermocouple array. Note that all the thermocouples are in flush with the inner surfaces of the crucible to avoid any friction with the molten in the liquid state. The relative accuracy of the temperature measurement is ±0.1 K and, the response time of the thermocouples is 0.7 s, the recording time step was 1 s.
2. The newly designed heat exchangers yield novelty features: they have the same cross-sectional shape (1 cm × 6 cm) of the sample in the y-direction to ensure the heat flux release through them being more homogenous; the compact three part structure, cooper–stainless steel–cooper, is designed in order to shorten the heat transfer time; and to overcome chemical corrosion between tin and copper, two metal-layers Cr, Ni are deposited on the narrow vertical surfaces of the heat exchangers. The arrangement with nine thermocouples in each heat exchanger (Fig. 1), with a vertical distance of 2 cm and a horizontal distance of 1.5 cm, allows us to measure the heat flux.
3. An enclosure Kirchhoff box assures thermal insulation of the largest surface of the crucible via the heat radiation compensative method. This procedure is realized by a PID regulating system, the details can be found in Refs. [8,9].
4. The entire set-up is installed inside a vacuum chamber in order to limit heat transfer by air convection and protect the sample from oxidation; the degree of vacuum is of the order of 0.01 atm.

All samples consist of a 6 × 10 × 1 cm^3 rectangular ingot solidified by means of lateral cooling [7]. The entire experimental process of all solidification experiments consists of five steps illustrated in Fig. 2: melting, initial thermal stabilization to obtain a homogenous temperature field, application of experimental temperature difference ($\Delta T = 40$ K) between the two heat exchangers, second stabilization of temperature field to obtain a stationary fluid flow, followed by cooling of the sample at a fixed rate (CR = 0.03 K/s). During initial thermal stabilization, electromagnetic stirring can be used to homogenize the liquid sample. Stirring was mandatory to avoid probable lead stratification during the melting step. It was observed that, after melting and application of the temperature differences between the two lateral walls (i.e., step 4 in Fig. 2), without any stirring, temperature field distribution consisted of vertical isotherms as in a solid (see the temperature map presented in Fig. 3). This seemed to indicate that natural convection was practically suppressed by the existence of density stratification due to lead segregation during the melting step. This effect was more pronounced with a higher lead composition; e.g., 6.5% or 10%, as expected.

3. Results and discussion

3.1. Effect of concentration on thermosolutal convection

The temperature contours and gradients for an experimental study of the three compositions selected at different times during cooling are shown in Fig. 4 in the next section. The corresponding numerical values of the instantaneous temperature field are given in Appendix B. The experimental conditions are: $\Delta T = 40$ K, CR = 0.03 K/s. Fig. 4 illustrates the behavior of natural convection during the solidification process. Isotherm deflection provides some indication of the flow pattern generated by natural convection. As shown in Fig. 4(a), the flow pattern consists of clockwise recirculation. The temperature gradient (represented by arrows) on the crucible surface was calculated at each thermocouple position from the temperature difference between two adjacent points by means of a numerically centered difference scheme. The figure shows clearly the necessity of the electromagnetic stirring during the first phase of thermal stabilization (7000 s–11,000 s), before

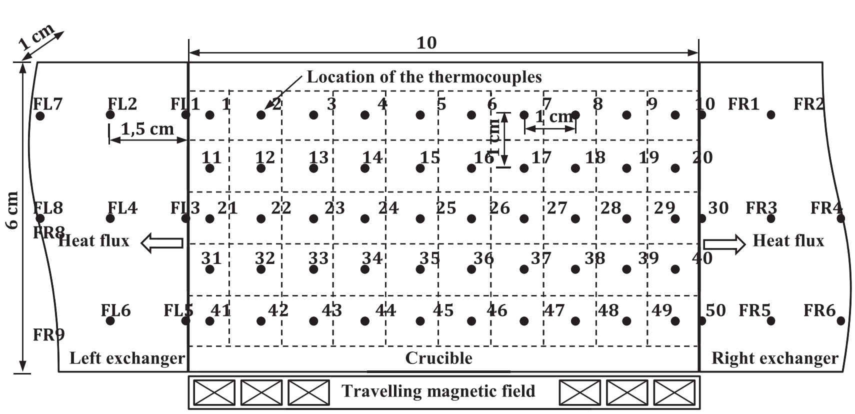

Fig. 1. Sketch of the solidification benchmark experiment: the location of the lateral thermocouples, the heat exchangers, and the traveling magnetic field.