EXPERIMENTAL ANALYSIS OF THE COLUMNAR TO EQUIAXED TRANSITION OF SN-PB HYPOEUTECTIC ALLOYS DIRECTIONALLY SOLIDIFIED UNDER UNSTEADY-STATE CONDITIONS

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Abstract. *Many of the mechanical properties of metallic alloys depend on the size and distribution of grains in the structure. Therefore the structure refinement and the distribution of the refined grains have been the subject of numerous researches in the field of metallurgy. The structure is almost determined in the solidification process, so it is essential to control the solidification structure. In this article the columnar to equiaxed transition (CET) was studied in tin-lead alloys during the horizontal directional solidification as a function of solidification parameters, cooling rates, temperature gradients, growth rates, and composition* C_O. *Specimens were solidified under unsteady state heat flow conditions. A combined theoretical and experimental approach is presented in order to quantitatively determine the solidification thermal variables considered. The macrostructures obtained has indicated that the columnar-to-equiaxed transition occurred on a near vertical plane in the castings and that resulting thermo-solutal convection seems to favor the structural transition which occurs for a critical cooling rate of about 0.027 K/s for the two alloy compositions investigated.*
Keywords*: directional solidification; tin-lead alloys; columnar to equiaxed transition; unsteady-state conditions; solidification parameters.*

1. INTRODUCTION

In spite of the extended history of study of solidification, many aspects of the physics of this phenomenon remain unclear. Aspects of forming of various types of micro and macrostructures in solids obtained, the physical mechanisms of which remain to a large degree unclear, are of particular importance. Solidification is therefore, one of the most important phase transformations in industrial production routes. It plays an important role in casting and foundry industry. In these cases, it takes place under near equilibrium conditions.

On the whole, the macrostructure of cast ingots consists of three different zones, that is, the chill, columnar, and equiaxed zones respectively. The origin of each one has been the subject of intense experimental and theoretical investigation because of the well-known correlation between grain structures and mechanical properties. The three zones may or may not be present in a particular case. The macrostructure of cast ingots have been of interest for many years and despite this effort there is as yet no way that the macrostructure of an ingot can be predicted nor even any clear agreement on how the columnar to equiaxed transformation (CET) actually occurs. The CET has been reported to be dependent on thermal conditions associated with the casting process, including alloy system, alloy composition, melt superheat, mold temperature, mold material, heat-transfer coefficients at the metal-mold interface, cooling rate, casting size, melt convection, transport of solute, and the concentration of nucleating particles, some of which vary with time and position during solidification (Flood and Hunt, 1987; Wang and Beckermann, 1994; Canté et al. 2007).

Lack of a quantitative understanding of the relationship between casting thermal conditions and the resulting structure has limited the development of certain procedures and methods for quality castings. Although, many experiments (Ziv and Weinberg, 1989; Ares and Schvezov, 2000; Siqueira et al. 2003), analytical methods (Hunt, 1984; Flood and Hunt, 1987a; Flood and Hunt, 1987b) and numerical simulations (Wang and Beckermann, 1994; Dong and Lee, 2005; Badillo and Beckermann, 2006) of this process have been carried out in recent years, the mechanism of CET is still not clear.

The gravity effects in relation to the CET, for example, have been investigated with the chill placed in general on the bottom or top of the mold. In the case of vertical upward directional solidification the influence of the convection is minimized when solute is rejected for the interdendritic regions, providing the formation of an interdendritic liquid denser than the global volume of liquid metal. On the other hand, when the process is carried through vertically downward the system provides the melt convection which arises during the process. In the horizontal unidirectional solidification, when the chill is placed on the side of the mold, the convection in function of the composition gradients in the liquid always goes to occur. An interesting feature of the horizontal configuration is the gradient of solute concentration and density in vertical direction, because solute-rich liquid falls down whereas free solvent-crystals rise due buoyancy force. Moreover, there will be also a vertical temperature gradient in the sample as soon as a thermo-solutal convection roll emerge. In spite of this only a few studies have reported these important effects of melt convection to the CET for this particular case.

The objective of this contribution is the presentation of experimental results on the CET in two hypoeutectic Sn-Pb alloys during the horizontal unsteady-state directional solidification in a cooled mold, under different conditions of superheat and heat transfer efficiencies at the metal/mold interface. A combined theoretical and experimental approach was applied to calculate the solidification thermal variables like metal-mold heat transfer coefficients (hi), tip growth rates (V_L), thermal gradients (G_L), and cooling rates (T_R), which affect the structure transition. A comparative study between the results of this article and those from the literature proposed to investigate the CET during upward and downward vertical solidification of Sn-Pb hypoeutectic alloys is also presented.

2. EXPERIMENTAL PROCEDURE

The casting assembly used in solidification experiments is shown schematically in Fig. 1. It was designed in such way that the heat was extracted only through the water-cooled system, placed on the side of the mold, promoting horizontal directional solidification. The carbon steel mold used had a length of 110 mm, a width of 80 mm, a height of 60 mm, and a wall thickness of 3 mm. The lateral inner mold surfaces were covered with a layer of insulating alumina and the upper part of the mold was closed with refractory material to prevent heat losses. The thermal contact condition at the metal/mold interface was also standardized with the heat extracting surface being polished. Previous measurements of the temperature field were carried out confirming that the described experimental set-up fulfills the requirement of an unidirectional heat flow in horizontal direction.

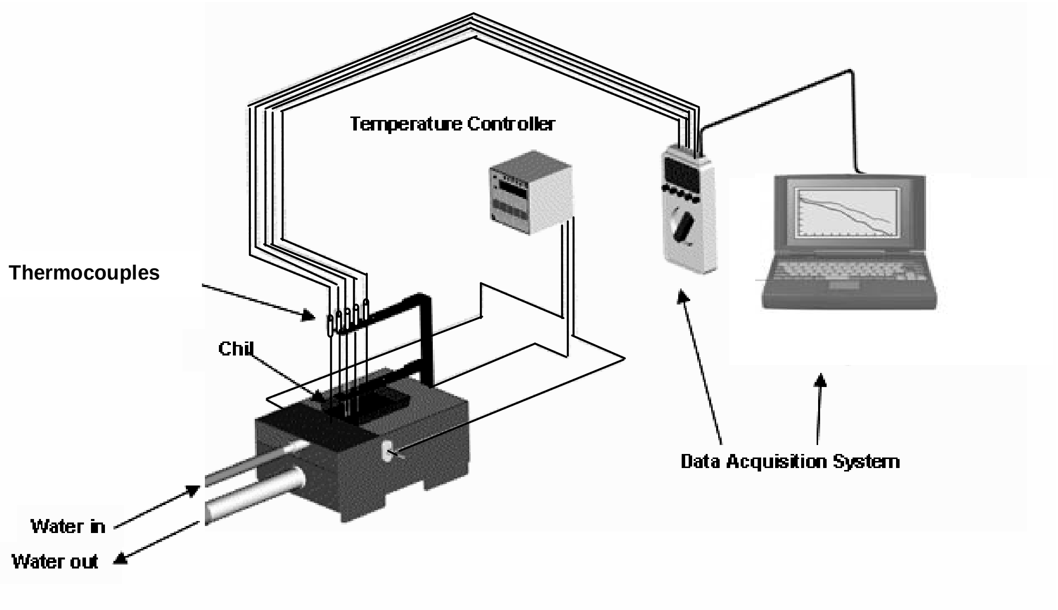


Figure 1. Schematic representation of the experimental setup for the directional solidification.

Experiments were performed with Sn-15wt%Pb and Sn-20wt%Pb hypoeutectic alloys. The chemical compositions of metals that were used to prepare the alloys investigated are summarized in Tab. 1. The corresponding thermophysical properties of these alloys are those reported in Tab. 2.

Table 1. Chemical analyses of metals used to prepare the Sn–Pb alloys.

Metal	Cu	Sn	Pb	Ni	Si	Mg	Cr	Mn	Zn	Fe
Sn	0.101	-	0.03	0.056	0.0496	-	0.042	0.033	0.168	0.067
Pb	0.100	0.0306	-	0.0198	0.070	0.042	0.090	0.080	0.270	0.0162

Table 2. Thermophysical properties of Sn-Pb alloys used for experimentation (Rocha *et al.* 2003).

Properties	Density	Latent Heat of Fusion	Specific Heat	Thermal Conductivity	Solidus Temperature	Liquidus Temperature
Symbol/units	ρ_S [kg/m^3] (solid) ρ_L [kg/m^3] (liquid)	L [J/kg]	c_S [J/kg.K] (solid) c_L [J/kg.K] (liquid)	K_S [W/m.K] (solid) K_L [W/m.K] (liquid)	T_{Sol} [^{o}C]	T_{Liq}[^{o}C]
Sn–15 wt%Pb	7,867 7,552	55,861	208 241	62.5 32.5	183	211
Sn–20 wt%Pb	8,108 7,735.6	53,809	202.8 234.8	60.5 32.3	183	204

The alloys were melted in situ and heated until a superheat of 10% above the liquidus temperature (T_{Liq}) using an electrical furnace. Approaching the superheat temperature, the mold was taken from the heater and set immediately on a water cooled carbon steel chill. Water was circulated through this cooling jacket keeping the carbon steel plate during the solidification at a constant temperature of about 25°C and thus inducing a longitudinal heat transfer from the mold. Solidification occurred dendritically from the lateral chill surface, forming a columnar structure.

Temperatures were measured at different positions in the alloy samples during the solidification process and the data were acquired automatically. For the measurements, a set of five fine type K thermocouples, arranged as shown in Fig. 1, was used. The thermocouples were calibrated at the melting point of tin, exhibiting fluctuations of about 0.4 and 1ºC respectively, and connected by coaxial cables to a data logger interfaced with a computer. The thermocouples were sheathed in 1.6 mm diameter steel tubes, and positioned at 6, 10, 16, 34 and 54 mm from the heat-extracting surface.

The resultant ingots were sectioned in the longitudinal direction, which is parallel to both the sample axis and the direction of solidification. After this, the metallographic specimens were mechanically polished with abrasive papers and subsequently etched with an acid solution composed of 100 ml H_2O, 2 ml HCl, and 10 g $FeCl_3$ to reveal the macrostructures. Etching was performed at a temperature between 30 and 35ºC during approximately 15 min. The position of the CET was located by visual observation and optical microscopy, and the distance from the side of the sample was measured.

The values of experimental thermal analysis in alloy samples were compared with theoretical results given by a finite difference heat flow program (Santos et al. 2001), and an automatic search has selected the best theoretical–experimental fit from a range of transient heat transfer coefficients profiles. In the heat transfer model it is assumed that heat flow is linear parallel to the ingot axis. The heat transfer coefficients at the metal-mold interface has been expressed as a power function of time, given by the general form $hi = C(t)^{-n}$ where hi (W/mK), t (s), C, and n are constants which depend on superheat, mold material and alloy composition respectively. The numerical model (Santos et al. 2001) was selected from the literature as the most appropriate model for representing the solidification conditions assumed in this paper. Furthermore, it has been frequently used by other authors in researching unidirectional solidification in horizontal (Santos et al. 2001) and downward vertical (Spinelli et al. 2004) systems, which produce thermo-solutal convection effects. On the other hand, simulated hi curves were used in another numerical model (Spinelli et al. 2004) , also employed in this paper to calculate theoretical V_L, G_L, and T_R, which are usually associated with CET.

3. RESULTS AND DISCUSSION

For determination of the values of hi was used the referring thermal profile to the thermocouple next to the cooled base (6 mm), position for which eventual heat losses by lateral walls of the mold is more improbable. Typical experimental thermal responses were compared to those simulated numerically by using transient hi profile which provide best curve fitting, as shown in Fig. 2.

The directionally solidified structures of alloys with starting melt temperatures 10% above the liquidus are shown in Fig. 3. The columnar to equiaxed transition occurred at 83 and 72 mm from the metal-cooling chamber interface for Sn–15 wt%Pb and Sn–20 wt%Pb alloys, respectively. The macrostructures show an important influence of the alloy composition with the anticipation of the structural transition with increasing solute content. The basic feature of the CET shown by these macrostructures is that the columnar-to-equiaxed transition essentially occurred on a near vertical plane parallel to the chill wall.

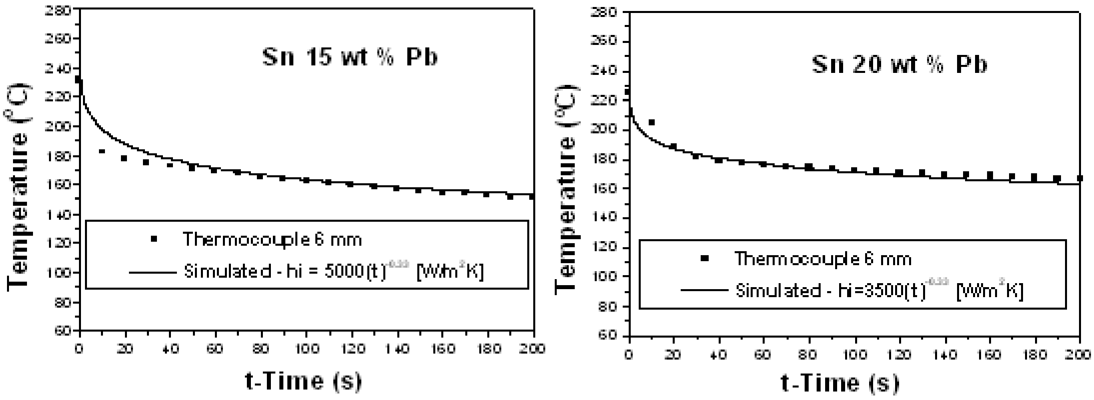
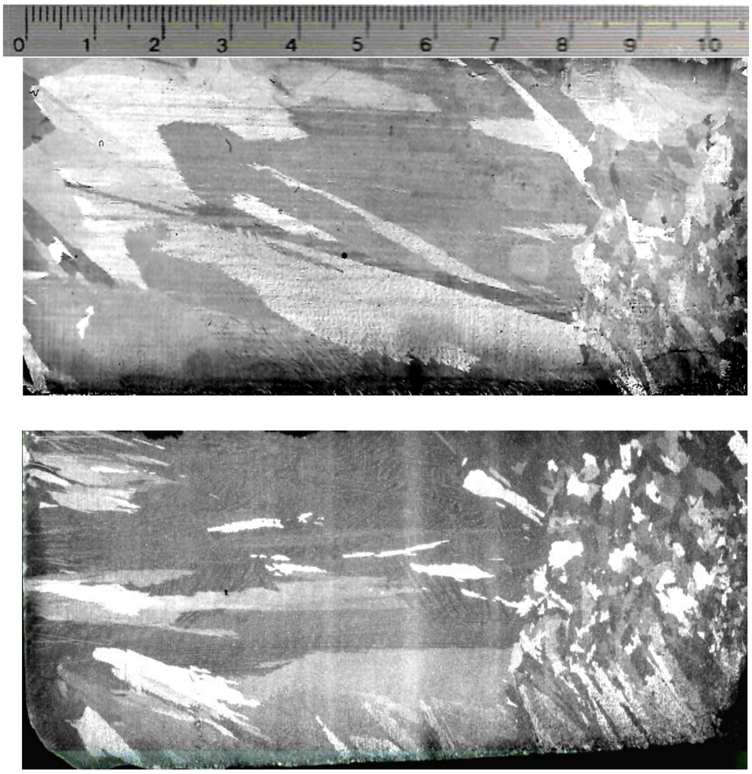


Figure 2. Simulated and typical experimental thermal responses of thermocouple at position 6mm from the heat-extracting surface of alloys investigated.



Sn-15%wtPb

Sn-20%wtPb

Figure 3. Solidification macrostructures of alloys castings.

Figure 4 shows schematically that the solute rejected from the melt during the solidification process elevates the melt density and this difference in density promotes interdendritic convection which can initiate the nucleation of cores in association with dendrite arm detachments in the interdendritic regions, which will behave as equiaxed nuclei, accounting for grain multiplication. As the new cores usually have liquidus temperatures higher than the melt temperature around, the intense presence of solid cores can allow the formation of equiaxed grains and columnar growth can be blocked, stimulating the CET occurrence.

In order to determine more accurate values of V_L, G_L, and T_R all of which vary with time and position during solidification, the results of experimental thermal analysis have been used to determine the displacement of the liquidus isotherm, i.e., a plot of position from the metal-mold interface as a function of time corresponding to the liquidus front passing by each thermocouple. A curve fitting technique on such experimental points has generated power functions of position as a function of time. The corresponding experimental points of alloys examined are presented in Fig. 5.

The derivative of this function with respect to time has yielded values for tip growth rate. Fig. 6 shows typical comparisons between experimental and numerical predictions (Spinelli et al. 2004) of V_L. A good agreement has been observed for the alloys investigated.

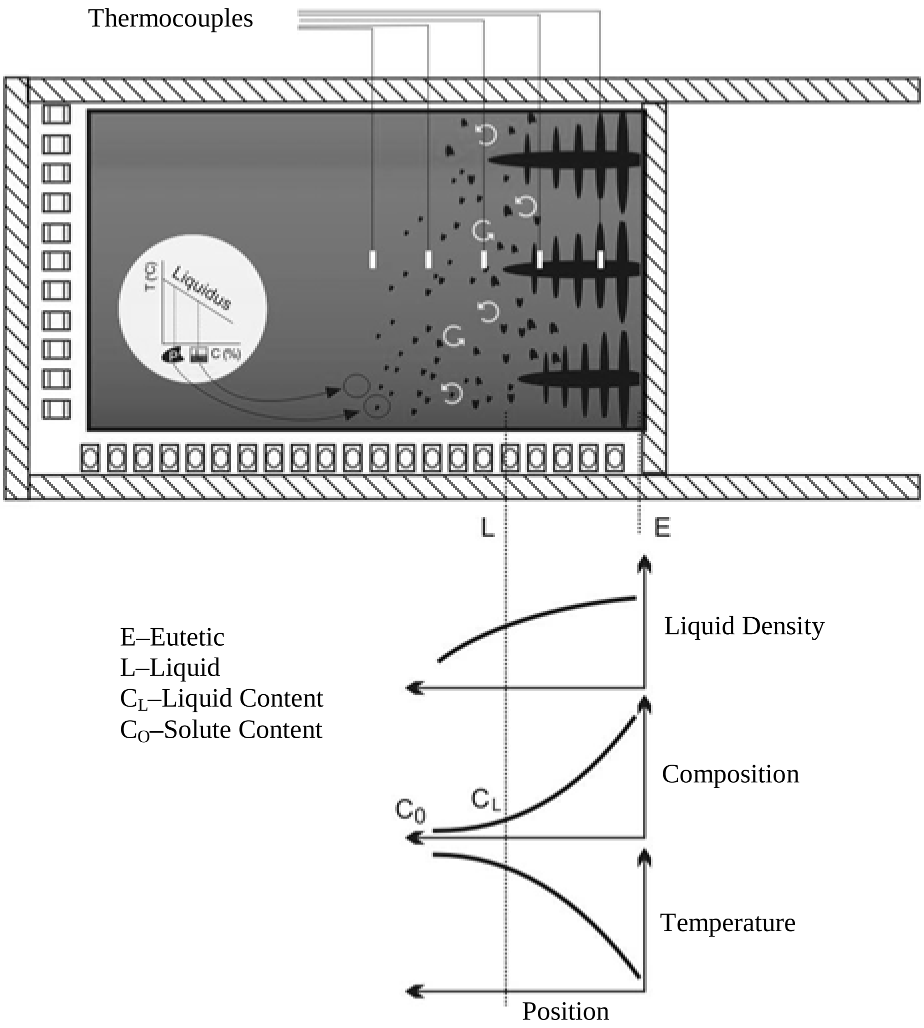


Figure 4. Schematic representation of melt convection effects during horizontal solidification.

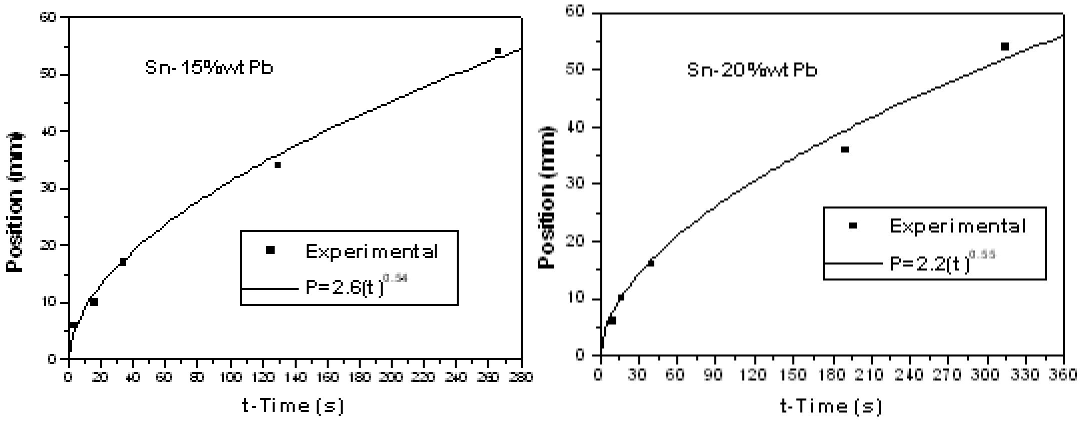


Figure 5. Position of liquidus isotherm from the metal-cooling chamber interface as a function of time.

The cooling rate was calculated by considering the thermal data recorded after the passing of the liquidus front by each thermocouple. These values are compared with theoretical predictions furnished by the numerical model (Spinelli et al. 2004) in Fig. 7. A good agreement has been observed between the experimental values and those numerically simulated for the alloys analyzed. The thermal gradient, G_L, has been obtained from the relationship between the cooling rate and tip growth rate, i.e., $T_R = G_L V_L$ (Spinelli et al. 2004).

Experimental results of CET positions, tip growth rate (V_L), temperature gradient in the liquid ahead of the tip interface (G_L), and cooling rate (T_R) at the transition are listed in Tab. 3.

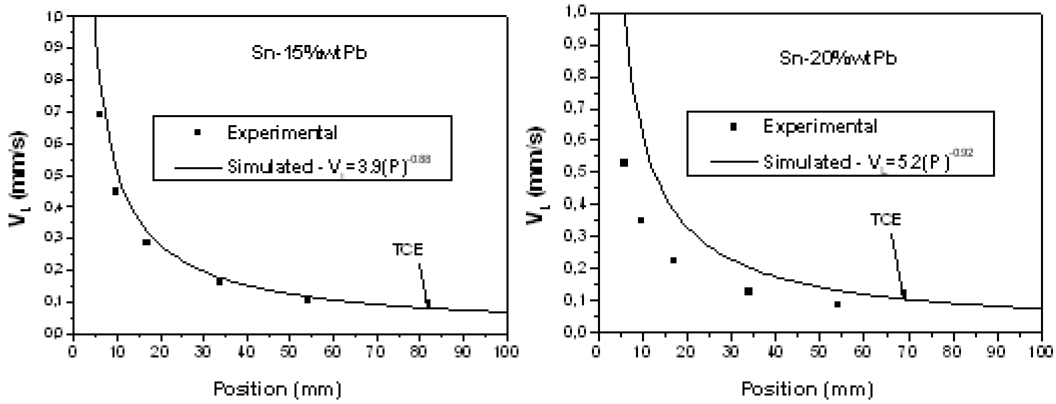


Figure 6. Tip growth rate as a function of position from the metal-cooling chamber interface.

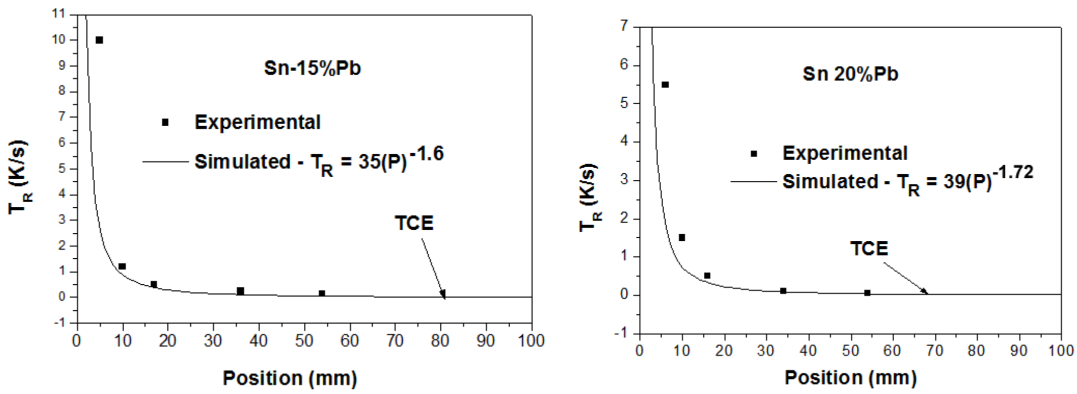


Figure 7. Tip cooling rate as a function of position from the metal-cooling chamber interface.

It can be observed that the structural transition occurred at different positions from metal-mold interface for the experiments carried out in this study and that the solute content in Sn–Pb alloys affected the experimental position of the CET.

Table 3. Solidification thermal parameters associated to the CET position.

Alloys	V_L (mm/s)	G_L K/mm)	T_R (K/s)	CET position (mm)
Sn-15wt%Pb	0.08	0.36	0.029	83
Sn-20wt%Pb	0.10	0.25	0.025	72

Investigations by Siqueira et al. (2003) and Spinelli et al. (2004), on vertically upward and downward directional solidification of Sn–Pb alloys, have proposed a CET criteria based on critical cooling rates. The results obtained in the present work suggest that the CET occurs when a critical cooling rate is reached at the dendrite tips. For the investigated alloys the average critical value is of about 0.027 K/s. When T_R does not reach the critical value no CET should occur. Thus, the same proposed criterion based on a critical cooling rate, which depends only on the alloy system (Siqueira et al. 2003), can be applied to the present experimental results.

Table 4 summarizes the results of the present experimental investigation and those obtained Siqueira et al. (2003) and Spinelli et al. (2004) concerning values of V_L, G_L, and $\dot{T}_R$ at the CET position during solidification of Sn–Pb alloys in systems with different configurations. In all the cases, the columnar length was found to decrease with increasing solute content. The average values obtained during horizontal and downward vertical solidification of Sn–Pb hypoeutectic alloys show that the Spinelli's observations were conducted under conditions of higher values of tip growth rates and lower values of thermal gradients in the melt at the liquidus temperature when compared with those reported in the present study. In all the situations investigated the solidification thermal variables are not very similar at the CET and it is not possible to identify a transition criterion based on a particular variable. In spite of the observed differences, the tip cooling rate at the transition in Spinelli's experiments (mean value of 0.030 K/s) is quite close to the critical value of about 0.027 K/s found in this work.

Table 4. Solidification thermal parameters associated to the CET position for the pesent experiments and those conducted by others researchers for Sn-Pb alloys.

Author	System	Melt Convection	Alloys	V_L (mm/s)	G_L (K/mm)	T_R (K/s)	CET Position (mm)
Siqueira et al. (2003)	upward vertical	No	Sn-10%Pb	0.283	0.052	0.015	60
			Sn-20%Pb	0.323	0.047	0.015	65
Spinelli et al. (2004)	downward vertical	Yes	Sn-15%Pb	0.180	0.155	0.028	83
			Sn-20%Pb	0.227	0.142	0.032	76
This work	horizontal	Yes	Sn-15%Pb	0.080	0.360	0.029	83
			Sn-20%Pb	0.100	0.250	0.025	72

It is very important to observe that the end of the columnar zone during horizontal and downward unidirectional solidification is abbreviated as a result of a two times higher critical cooling rate than that verified during upward unidirectional solidification of Sn–Pb alloys because the rejection of solute into the melt during solidification results in increased melt density and the difference in density will induce interdendritic convection. Melt convection induced by solute segregation may be causing pileup of equiaxed grains formed from fractioned dendritic arms which must stimulate the CET occurrence (Fig. 4) when comparing these results with those reported by Siqueira et al. (2003) which does not take the convection effect into account. The solid cores, denser and less enriched in solute than in liquid form, tend to settle due to the effects of gravity, and survive because they have liquidus temperatures higher than the surrounding liquids. These particles become equiaxed nuclei, agglomerated at the bottom of the cavity, and in sufficient numbers block the advance of the columnar front, which justifies anticipating transition in the lower region of the ingot in the vertical direction of the horizontal solidification system. Thus, the experimental results presented in Tab. 4 suggest that the CET occurs when a critical cooling rate is reached at the dendrite tips, i.e., 0.015 K/s for upward vertical and 0.030 K/s for downward vertical and horizontal unsteady-state directional solidification conditions of Sn–Pb alloys with the columnar growth prevailing throughout the casting for cooling rates higher than these critical values. When T_R does not reach the critical value no CET should occur.

4. CONCLUSIONS

The columnar-to-equiaxed transition has been studied in tin-lead alloys during the horizontal directional solidification under unsteady state conditions. From the results and discussion of the previous sections the main conclusions of this investigation on the correlation between different solidification parameters on the CET are:

- The CET was observed to occur essentially on a near vertical plane parallel to the chill wall.

- The CET occurs when the temperature gradient in the melt ahead of the columnar grains ranges from 0.25 to 0.36 K/m and for values of tip growth rates ranging from 0.08 to 0.10mm/s.

- The columnar length decreases with increasing solute content which is abbreviated as a result of a two times higher critical cooling rate than that verified during upward unidirectional solidification of Sn–Pb alloys investigated.

- The resulting thermo-solutal convection seems to favor the structural transition which occurs when a critical cooling rate is reached at the dendrite tips. For the investigated alloys the average critical value is of about 0.027 K/s. When T_R does not reach the critical value no CET should occur.

- Melt convection induced by solute segregation may be causing pileup of equiaxed grains formed from fractioned dendritic arms which must stimulate the CET occurrence when comparing the results of this work with those obtained during upward directional solidification conditions of tin–lead alloys.

- A comparison of the results obtained in the present work with previous investigations concerning the CET in Sn–Pb hypoeutectic alloys, conducted under conditions of upward and downward unidirectional solidification, has shown that the proposed criterion based on a critical cooling rate can be applied to the present experimental results, except for the value of the critical cooling rate.

- The heat transfer coefficient at the metal/mold interface was found to decrease with increasing lead concentration.

5. ACKNOWLEDGEMENTS

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6. REFERENCES

Badillo, A. and Beckermann, C., 2006, "Phase-Field Simulation of the Columnar-to-Equiaxed Transition in Alloy Solidification", Acta Materialia, Vol. 54, pp. 2015-2026.

Canté, M.V., Cruz, K.S., Spinelli, J.E., Cheung, N. and Garcia, A., 2007, "Experimental Analysis of the Columnar-to-Equiaxed Transition in Directionally Solidified Al–Ni and Al–Sn Alloys", Materials Letters, Vol. 61, pp. 2135-2138.

Dong, H.B. and Lee, P.D., 2005, "Simulation of the Columnar-to-Equiaxed Transition in Directionally Solidified Al–Cu Alloys", Acta Materialia, Vol. 53, pp. 659-668.

Flood, S.C. and Hunt, J.D., 1987, "Columnar and Equiaxed Growth I. A Model of a Columnar Front with a Temperature Dependent Velocity", J. of Crystal Growth, Vol. 82, pp. 543-551.

Flood, S.C. and Hunt, J.D., 1987, "Columnar and Equiaxed Growth II. Equiaxed Growth Ahead of a Columnar Front", J. of Crystal Growth, Vol. 82, pp. 552-560.

Flood, S.C. and Hunt, J.D., 1987, "A Model of a Casting", Applied Science Research, Vol. 44, pp. 27-42.

Hunt, J.D., 1984, "Steady State Columnar and Equiaxed Growth of Dendrites and Eutetic", Materials Science and Engineering, Vol. 65, pp. 75-83.

Rocha, O.L., Siqueira, C.A. and Garcia, A., 2003, "Heat Flow Parameters Affecting Dendrite Spacings During Unsteady-State Solidification of Sn-Pb and Al-Cu Alloys", Metallurgical and Materials Transactions A, Vol. 34A, pp. 995-1006.

Santos, C.A., Quaresma, J.M.V. and Garcia, A., 2001, "Determination of Transient Interfacial Heat Transfer Coefficient in Chill Mold Casting", J. of Alloys and Compounds, Vol. 319, pp. 174-186.

Siqueira, C.A., Cheung, N. and Garcia, A., 2003, "The Columnar to Equiaxed Transition During Solidification of Sn–Pb Alloys", J. of Alloys and Compounds, Vol. 351, pp. 126-134.

Spinelli, J.E., Ferreira, I.L. and Garcia, A., 2004, "Influence of Melt Convection on the Columnar to Equiaxed Transition and Microstructure of Downward Unsteady-State Directionally Solidified Sn–Pb Alloys", J. of Alloys and Compounds, Vol. 384, pp. 217-226.

Wang, C.Y. and Beckermann, C., 1994, "Prediction of Columnar to Equiaxed Transition During Diffusion-Controlled Dendritic Alloy Solidification", Metallurgical and Materials Transactions A, Vol. 25A, pp. 1081-1093.

Ziv, I. and Weinberg, F., 1989, "The Columnar-to-Equiaxed Transition in Al 3Pct Cu", Metallurgical Transactions B, Vol. 20B, pp. 731-734.

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