Lima Rocha, Otávio; Gouvea Gomes, Laércio; Moutinho, Daniel Joaquim Conceição; Leão Ferreira, Ivaldo; Garcia, Amauri

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Universidade Federal de Ouro Preto
Ouro Preto, Brasil

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A transição colunar-equixial na solidificação directional de ligas multicomponentes à base de alumínio

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Otávio Lima Rocha
Professor
Federal Institute of Education,
Science and Technology of Pará – IFPA
Belém – Pará – Brazil
otvrocha@oi.com.br

Laércio Gouvea Gomes
Professor
Federal Institute of Education,
Science and Technology of Pará – IFPA
Belém – Pará – Brazil
laercio.gomes@ifpa.edu.br

Daniel Joaquim Conceição Moutinho
Professor
Federal Institute of Education,
Science and Technology of Pará – IFPA
Belém – Pará – Brazil
djcmoutinho@hotmail.com

Ivaldo Leão Ferreira
Professor
Fluminense Federal University - UFF
Volta Redonda - Rio de Janeiro – Brazil
ileao@puvr.uff.br

Amauri Garcia
Professor
State University of Campinas - UNICAMP
Campinas - São Paulo – Brazil
amaurig@fem.unicamp.br

Abstract

In this work the columnar to equiaxed transition (CET) was experimentally investigated in the unidirectional solidification of three aluminum based multicomponent alloys (Al-nSi-3Cu), with "n" equal to 5.5, 7.5 and 9 wt.%. The main parameters analyzed include the tip temperature gradient (G_L), tip growth rate (V_L), tip cooling rate (T_R) and Si content. A water-cooled solidification experimental setup was developed, and specimens were solidified under unsteady state heat flow conditions. It is shown that for the alloys examined, the solute concentration influences the position of the CET, which occurs for an average cooling rate of about 1.17 ^{0}C/s. A comparative analysis between the results of this work and those from literature proposed to analyze the CET during upward vertical solidification of Al-Si alloys is reported and the results show that the end of the columnar region is abbreviated as a result of seven times higher critical cooling rate than that verified for Al-Si alloys.

Keywords: Directional solidification; Macrostructures; Columnar/equiaxed transition; Al-Si-Cu alloys.

Resumo

Nesse trabalho, a transição colunar/equiaxial (TCE) foi investigada experimentalmente na solidificação unidirecional de três ligas multicomponentes à base de alumínio (Al-nSi-3Cu), com "n" igual a 5,5, 7,5 e 9% em massa. Os principais parâmetros analisados foram o gradiente de temperatura (G_L), a velocidade de deslocamento da isoterma liquidus (V_L), a taxa de resfriamento (T_R) e a composição de Si. Um dispositivo de solidificação direcional refrigerado à agua para fluxo de calor transitório foi utilizado. Para todas as ligas examinadas, os resultados mostram que a concentração de soluto inflencia na posição da TCE, a qual ocorre para uma taxa de resfriamento igual a 1.17 ^{0}C/s. A análise comparativa entre os resultados desse trabalho com os da literatura, propostos para analizar a TCE em ligas Al-Si, é conduzida e os resultados mostram que o final da região colunar acontece para uma taxa de resfriamento sete vezes maior que a encontrada para ligas Al-Si.

Palavras-chave: *solidificação direcional; macroestruturas; transição colunar/equiaxial; ligas Al-Si-Cu.*

1. Introduction

It is well known that most of the mechanical properties of metallic alloys depend on the size and distribution of grains in the structure which is almost determined during the solidification process. However, despite the history of investigation of solidification, many aspects of the physics of this phenomenon remain unclear. Aspects related to the development of various types of microstructures and macrostructures in solids, mainly the physical mechanisms involved, remain to a large degree unclear and are of particular importance. Since solidification is one of the most important phase transformations in industrial production routes, the understanding of the interrelations on operational thermal variables, solidification structures and mechanical properties is essential for the development of improved methods for quality castings. In this context, studies on macrostructural evolution of polycrystalline materials during solidification show two types of grain morphologies: columnar and equiaxed. The origin of each one has been the subject of numerous theoretical and experimental researches in the field of metallurgy for many years. Columnar grains often grow from the mold surface, where the thermal gradients are high, and the growth is preferentially oriented in a direction close to the heat flux. When the gradients are reduced near the center of the casting, equiaxed grains grow in all space directions leading to a material with isotropic mechanical properties and a more homogeneous composition field than that of a columnar structure. Depending on the application, one type of grain is preferred and thus favored, e.g. equiaxed grains in car engines and columnar grains in turbine blades as reported by Reinhart *et al.* (2005) and Liu *et al.* (2006). Thus, equiaxed grains can nucleate and grow ahead of the columnar front causing an abrupt columnar to equiaxed transition (CET). As a consequence, it is critical for industrial applications to understand the physical mechanisms which control the CET in solidification processes.

The CET occurrence, therefore, has been the subject of numerous studies undertaken with a view toward modeling this phenomenon. 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 to improve the materials efficiency and performance. Although, many experiments (Gandin, 2000; Siqueira *et al.*, 2002; 2003; Spinelli *et al.*, 2004; Peres *et al.*, 2004; Canté *et al.*, 2007) and theoretical simulations (Badillo & Beckermann, 2006) of this process that have been carried out in recent years, the mechanism of CET is still not clear. These studies highlight the importance of the relative growth of the equiaxed and columnar grains and develop expressions or numerical procedures to describe a criterion for the columnar-to-equiaxed transition. All these works have been developed for binary alloys.

Aluminum alloys castings had a fundamental role in the growth of the metal-mechanics industry. Nowadays these alloys are supplied in a wide range of chemical compositions. Investigations on vertically upward directional solidification of hypoeutectic Al–Si alloys have proposed a CET criterion based on a critical cooling rate of about 0.17 K/s with the columnar growth prevailing throughout the casting for cooling rates higher than this critical value (Peres, 2004). The CET was observed to occur essentially at the same position from the casting surface and no particular solidification thermal variable could be identified as the responsible for the structural transition since all of them were very similar at the CET.

Studies on the transient solidification of ternary alloys related to macrostructural and microstructural parameters, solidification modeling, solute segregation and porosity formation are very scarce. The main difficulty is related to the determination of the ternary solidification path and the intermediate phases reactions. The microstructural evolution and growth models for ternary alloys cannot be found in literature. Ferreira and coworkers (Ferreira *et al.*, 2004), proposed a numerical scheme to simulate the transient solidification of an Al-Cu-Si alloy for the analysis of inverse segregation, considering the absence of intermediate phases and reactions. Al–Cu–Si ternary alloys, because of particular outstanding properties such as high mechanical strength, low weight and very good fluidity, are a good choice for applications in the automotive and aerospace industry. The potential of such alloys has attracted much attention of researchers with a view to investigating the microstructure evolution, and the formation of macrosegregation and porosity during the solidification process. The mechanical properties of these alloys are related to the grain size, the scale of the dendritic microstructure exhibited by the Al-rich phase, the shape, size and distribution of the Si particles and intermetallics, etc, i.e. the design of mechanical properties of these alloys depends on the strict control of the macro and microstructures during solidification.

The objective of this study is to investigate the influence of thermal parameters on the CET in three hypoeutectic Al-Si-Cu alloys during the upward unsteady-state directional solidification in a cooled mold. Cooling curves for five thermocouples strategically placed along the casting length are used to quantitatively determine the solidification thermal parameters. A comparative study between the results of this study and those obtained by Peres *et al.* (2004) who studied the CET during upward vertical solidification of Al-Si hypoeutectic alloys is also presented. In this context, this study aims to contribute to a better understanding of the solidification thermal variables affecting the CET of Al-Si-Cu alloys.

2. Materials e methods

Experiments were carried out with Al-5.5wt.%Si-3wt.%Cu, Al-7.5wt.%Si-3wt.%Cu and Al-9wt%Si-3wt%Cu alloys, prepared from 99.7% Al, 99,9% Cu and 99.6% Si. The chemical compositions of metals that were used to prepare these alloys and the corresponding thermophysical properties are those reported by Gomes (2012).

The casting assembly used in directional vertical upward solidification experiments has been detailed in previous articles (Siqueira *et al.*, 2002; 2003; Rocha *et al.*, 2003; Spinelli *et al.*, 2004; Peres *et al.*, 2004). The experimental setup consists of a water-cooled mold with heat being extracted from the bottom, promoting a vertical upward directional solidification. The stainless steel mold had an internal diameter of 50 mm, height of 110mm and wall thickness of 3mm. To minimize radial heat losses, a solution of alumina in water has been applied to the inner surface of mold side walls by using a spray gun. After drying, the layer of insulating alumina was about 500 mm thick. The bottom part of the mold was closed with a thin (3 mm thick) carbon steel sheet, which physically separates the metal from the cooling fluid. The alloys were melted in situ and starting melt superheats were standardized in 5% above the liquidus temperature (T_L) using an electrical furnace whose heaters had their power controlled in order to permit a desired superheat to be achieved. Approaching the superheat temperature, the mold was taken from the heater and set immediately on a water cooled stainless steel chill. Water was circulated through this cooling jacket keeping the stainless steel plate during the solidification at a constant temperature of about 25°C and thus inducing a lateral longitudinal heat flow along the vertical direction.

Continuous temperature measurements in the casting were monitored during solidification through the output of a bank of fine type K thermocouples sheathed in 1.6 mm steel tubes and positioned at 5, 10, 15, 30, 50 and 70 mm from the chill. Each ingot was sectioned along its 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 70 ml H_2O, 10 ml HCl, 15 ml HNO_3 and 5 ml HF to reveal the macrostructures. Etching was performed at a temperature between 30°C and 35°C during approximately 30 seconds. The result of the chemical attack is the revelation of the macrostructure, used to confirm the directionality of solidification, the structural morphology and especially the verification of the columnar to equiaxed transition. The position of the columnar to equiaxed transition (CET) was clearly delineated by visual observation and optical microscopy on the etched surface, and the distance from the bottom of the casting was measured.

The thermocouple readings were used to generate 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 these experimental points yielded a power function of position as a function of time. The derivative of this function with respect to time gave values for the liquidus isotherm velocity. Moreover, the data acquisition system employed permits accurate determination of the slope of the experimental cooling curves. Hence, the cooling rate ahead the liquidus front was determined by considering the thermal data recorded immediately after the passage of the liquidus front by each thermocouple (Rocha *et al.*, 2003).

3. Results and discussions

Experimental cooling curves for the six thermocouples inserted into the castings during solidification of the alloys investigated in this study are shown in Figures 1(a), (b) and (c).

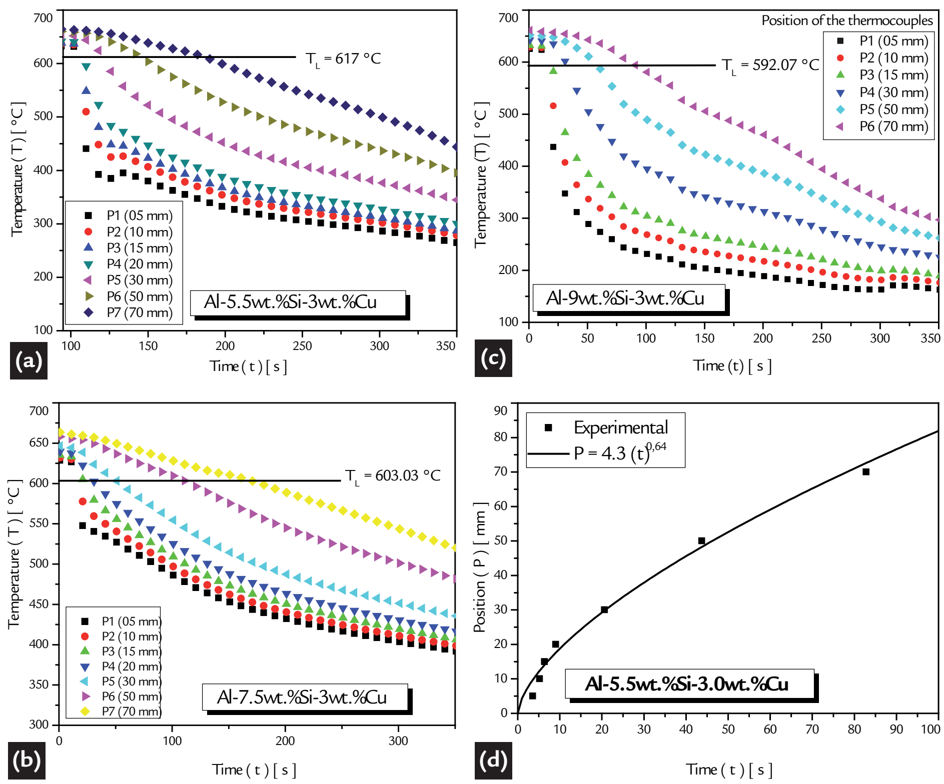

Figure 1
(a), (b) and (c) Typical experimental thermal responses of alloys investigated; (d) Experimental position of liquidus isotherm from the metal-mold interface as function of time for Al-5.5wt.%Si-3wt.%Cu alloy.

The CET is dependent on solidification thermal parameters such as V_L, G_L, and T_R all of which vary with time and position during solidification. It is well known that is very difficult to measure accurately these parameters at the CET, using thermocouples, since the position of the CET is not known before each experimental test. In order to determine more accurate values of these parameters 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.

The experimental points corresponding to one of the alloys examined are presented in Figure 1(d) for the Al-

5.5wt.%Si-3wt.%Cu alloy. The derivative of these function position vs time yielded values for the tip growth rate (V_L), shown in Figure 2(a) for all alloys investigated. The tip cooling rate (T_R) is shown in Figure 2(b).

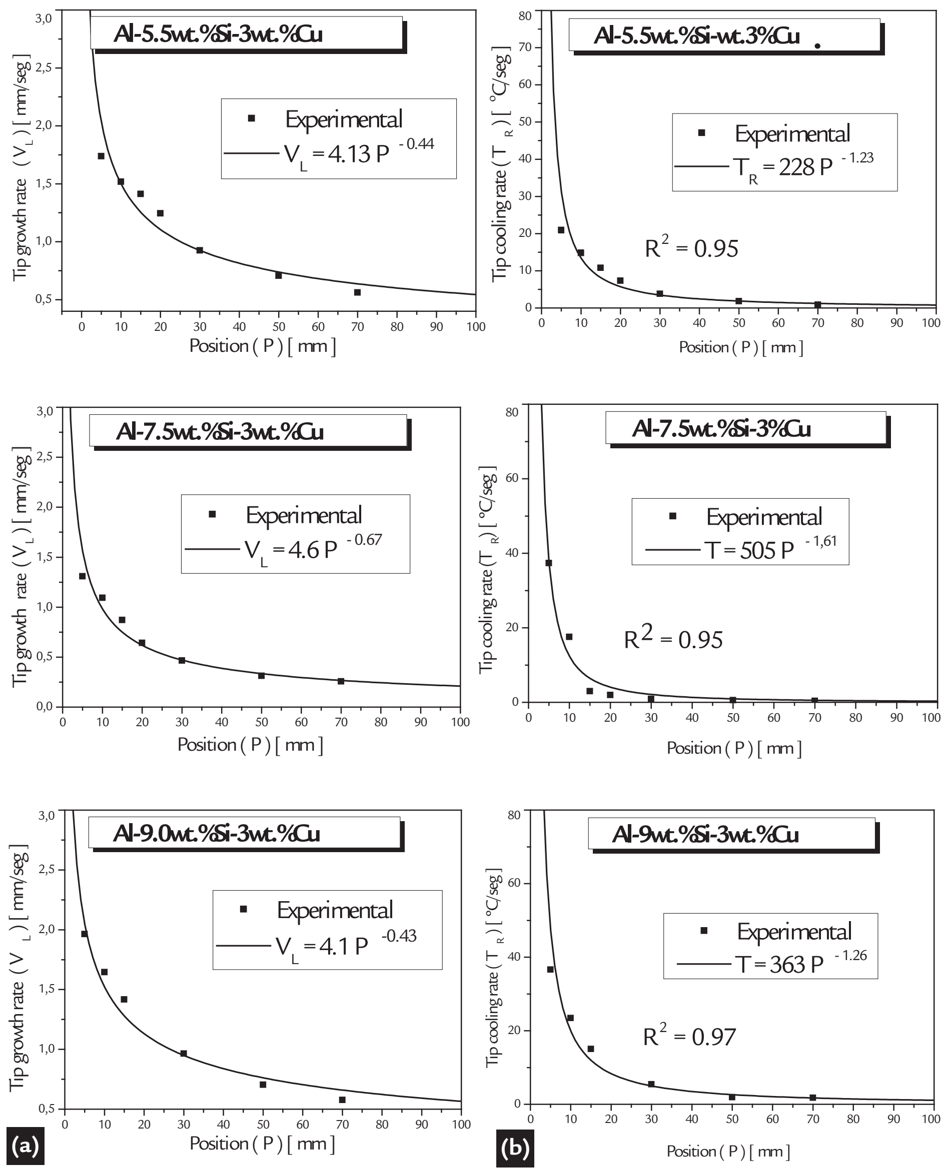

Figure 2
(a) Experimental tip growth rate from the metal/mold interface as a function of position and
(b) Experimental tip cooling rate from the metal/mold interface as a function of position.

The directionally solidified macrostructures of alloys with starting melt temperatures of 648, 633 and 622 ^{0}C above the liquidus temperatures, respectively, are shown in Figure 3. The columnar to equiaxed transition occurred at 70, 78 and 95 mm from the metal/cooling chamber interface for the alloys investigated.

The basic feature of the CET shown by these macrostructures is that the transition is sharp, i.e., there is clear evidence that the columnar-to-equiaxed transition occurred in a plane parallel to the chill wall.

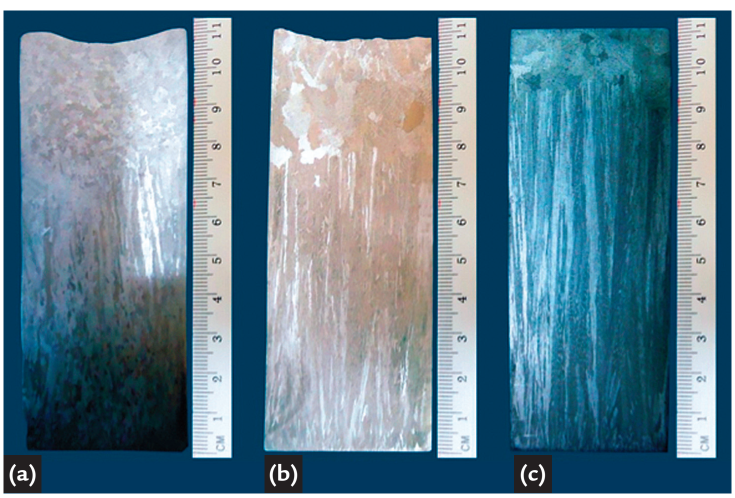

Figure 3
Solidification macrostructures of the Al-Si-Cu alloys investigated:
(a) Al-5.5wt%Si-3wt%Cu,
(b) Al-7.5wt%Si-3wt%Cu,
(c) Al-9wt%Si-3wt%Cu.

Not known are studies in literature on columnar to equiaxed transition for ternary alloys. Investigations on vertically upward and downward directional solidification of Al-Cu, Al-Si and Sn-Pb alloys conducted by Siqueira *et al*. (2002), Peres *et al*. (2004) and Spinelli *et al*. (2004), respectively, proposed a CET criteria based on critical cooling rates. When T_R does not reach the critical value, no CET should occur, with the columnar growth prevailing throughout the casting. The analysis of the thermal parameters of Table 1 allows us to suggest the same proposed criterion based on a critical cooling rate, which depends only on the alloy system, and can be derived from the present experimental results. For the investigated alloys the average critical value is of about 1.17 K/s.

As shown in Figure 3, the CET changes significantly with the increase in the alloy composition, contrarily to the results obtained by Peres *et al*. (2004) with the increase in Si content of hypoeutectic Al-Si alloys, which did not influence the position of the columnar-to-equiaxed transition. 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).

Table 1
Solidification thermal parameters associated with the CET position for the present experiments.

Ternary Alloys	CET Position [mm]	Tip growth rate (V_L) [mm/s]	Tip cooling rate (T_R) [°C/s]	Tip thermal gradient (G_L) [°C/mm]
Al-5.5wt.%Si-3wt.%Cu	70	0.63	1.23	1.95
Al-7.5wt.%Si-3wt.%Cu	78	0.25	1.06	4.24
Al-9wt.%Si-3wt.%Cu	95	0.58	1.23	2.11
Average	-	-	1.17	-

4. Conclusions

Experiments were conducted in order to study the CET occurrence during the upward unsteady-state directional solidification in a cooled mold of three ternary Al-Si-Cu alloys. The following main conclusions are derived from the present investigation:

(a) The basic feature of the CET is that the transition is sharp, i.e., the columnar-to-equiaxed transformation occurs rapidly along a plane parallel to the chill wall, as also observed by Peres *et al*. (2004, 2005) for hypoeutectic Al-Si binary alloys.

(b) The structural transition has been shown not to occur at the same position from the metal/mold interface for the experiments carried out and the increasing Si solute content of the Al-nSi-3Cu alloys investigated in this work, was found to affect the experimental position of the CET, unlike the observations in the study of Peres et al. in which the Si content was shown not to affect the CET position;

(c) The CET has occurred, for ternary Al-Si-Cu alloys, for tip growth rates ranging from 0.25 to 0.63 mm/s and temperature gradients in the melt ahead of the liquidus isotherm ranging from 1.95 to 4.24 ^{0}C/mm, i.e these results do not give support to a CET criterion based only on particular values of such thermal parameters.

(d) It seems that a more realistic CET criterion should encompass both the tip growth rate and the thermal gradient, through the tip cooling rate ($T_R = G_L.V_L$). The columnar growth is observed to prevail throughout the casting for cooling rates higher than a critical value, which depends only on the alloy system, and has been observed to be about 1.17 ^{0}C/s for the ternary alloys investigated in this work.

(e) Finally, the comparative analysis between the results of this work and those from literature has shown that the end of the columnar region is abbreviated as a result of a seven times higher critical cooling rate than that verified for hypoeutectic Al-Si alloys.

5. Acknowledgements

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