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PREDICTION FOR THE COLUMNAR-TO-EQUIAXED TRANSITION SOLIDIFICATION IN BINARY ALLOYS

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ABSTRACT. *Mathematical modeling of solidification of metallic alloys in order to predict the macro grain suffered very important advances in recent decades, but there is a need for current models capable of modeling grain growth during all stages of solidification, especially at the time of occurrence of the transition region columnar-equiaxed. The change grains columnar to equiaxed is common for the solidification of metal alloys, the transition region is called columnar-equiaxed transition (CET) and its study provides the prediction of the mechanical properties that can be applied for example in the development of new products. The present study aims to model and simulate the prediction of CET in solidification problems. The model is based on the conservation equations of mass and chemical species, together with the laws of nucleation and growth of dendritic structures. This model comes from unidirectional solidification and is validated through comparisons with other existing model mentioned in this present paper. Among the results, the model shows the movement of the front of columnar growth, and the position of the transition from columnar to equiaxed during solidification of binary alloys.*

KEYWORDS: *Columnar-equiaxed transition; CET-multiphase solidification; Mathematical model; Simulation.*

1. INTRODUCTION

The increasing development of industries requires the need for new advances in the science and technology of materials and processes. The properties of the products produced by casting depend on the solidification of the liquid metal in the mold and, particularly, of the resulting microstructure during the solidification process. Solidification is a description of scientific and technological processing of a material of the liquid phase to the solid phase (Muller, 2002). It can be estimated that at least 1 billion tons of metal products under go this processing annually, through numerous industrial processes ranging from the production of raw material (cast iron, aluminum, copper, zinc), its processing (steel, cast iron, aluminum electrolytic) and finally finish (casting, welding). Even having attracted the attention of scientists and engineers since the dawn of the Industrial Revolution. The study of solidification grew in importance during the decade of 60, when technology began requiring materials with purity and structures increasingly controlled (Muller, 2002).

The mechanical properties of a piece obtained by casting are closely related to the amount of equiaxed and columnar grain, which depends on the region of the columnar-equiaxed transition (CET - "columnar-to-equiaxed transition") present in the crude solidified structure. It is known that the CET during solidification occurs when the equiaxed grains block the grain growth columnar (Flood, et al., 1988). This study can be accomplished by implementing a mathematical model for the unidirectional solidification to predict the position of the columnar-equiaxed transition, providing a forecast of the macrostructure of grains. Mathematical modeling of solidification of metallic alloys has undergone significant advances in recent years. In the initial numerical models, the existence disregarded grain microstructure and the presence of dendritic growth during solidification. Currently the models developed consider macroscopic aspects, such as heat conduction and flow of liquid metal and microscopic aspects, such as those related to the nucleation and grain growth.

Among the many variables that affect the final structure of grain, we can highlight: cooling rate, alloy composition and the presence of inoculants. The influence of each of these variables in the growth of dendrites is a complex task, since the effects are interdependent and to assess their effects simultaneously is necessary to use mathematical models.

The mathematical model implemented in this work is based on the model developed by Martorano et al., (2003) in order to show the movement of the front of columnar growth and position of the columnar-equiaxed transition in binary alloys, then comparing the results with the theories of solidification in the foremost literatures.

2. METALLURGY OF SOLIDIFICATION

2.1 SOLIDIFICATION: CONCEPTS AND METALLURGICAL ASPECTS AFFECTING GROWTH OF GRAIN:

The final microstructure resulting from solidification directly influence the mechanical properties of metals. And alloys and are directly associated with, those properties depend on grain size, dendrite arm spacing or cellular, fibrous or lamellar spacing, the heterogeneity of chemical composition, size, shape and distribution of inclusions, porosity formed, and further. The pouring temperature of the molten metal emerges as the first variable influence, together with the intensity of convective currents during the mold filling. This will act as the primary means to extract heat from the metal, ensuring the processing liquid / solid, and the rate of heat extraction will depend on the heat absorption capacity of the mold to directly influence the rate of colds (Martorano, et al., 2003) .

2.2 MICROSTRUCTURES AND MACROSTRUCTURES:

The evolution of the shape of the interface between solid and liquid (S / L), during the solidification, is strongly related to the types of microstructures present in an alloy. One can spend the flat shape of the pure metals for cellular and dendritic structures, due to changes in the thermal parameters of the system metal / mold during solidification. The solute or solvent is segregated in this solid-liquid interface, which causes a non-uniform distribution of the liquid across the interface, causing their instability. This accumulation of solute content ahead of the solid / liquid interface promotes the emergence of a phenomenon conducive to nucleation and responsible for its gradual instability, known as Super-cooling constitutional (SRC).

The different morphologies are originated from instabilities caused in the solid / liquid interface depending on the value of the SRC, which in ascending order of this value are labeled by: planar and dendritic cell, according to the scheme shown in Figure 1.

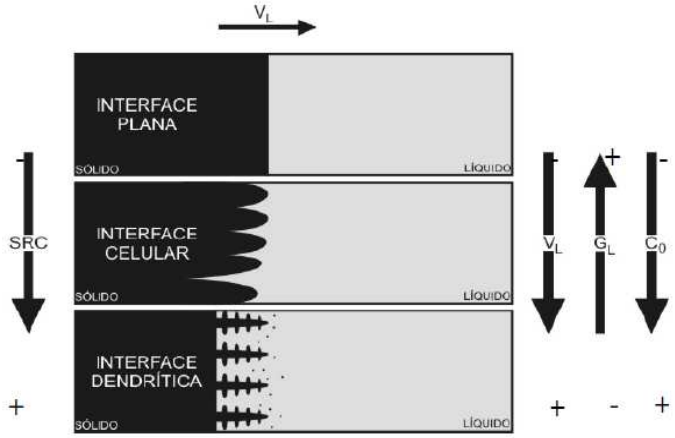

Figure 1. Scheme of the action of factors influence the formation of solidification structures: SRC - Super-cooling constitutional; GL - thermal gradient ahead of the interface; VL – Speed liquidus isotherm, and C0 - solute concentration. Garcia (2007).

Generally, the solidification leads to two characteristic morphological types: equiaxed and columnar (Kurz, et al., 2001). In revealing structural aspects of ingots, commonly are these different structures distributed in a characteristic way and a peripheral region, consisting of equiaxed grains of reduced grain size, followed by an intermediate region of elongated grains in one direction known columnar grains, and finally central equiaxed grains of larger size to that found in the periphery. Such an arrangement can be seen in the schematic drawing of Fig. 2. After nucleation of crystals in a super cooled liquid isothermal usually occurs equiaxed crystal growth.

But the zone columnar growth occurs preferentially in one direction, as well as the heat fluxes, extremely unidirectional, at least locally (Kurz, et al., 2001). The generation of a thin layer of equiaxed grains together the walls of the molds granular zone, the associated high rates of cooling arising from the high temperature difference between the mold and the metal. The region between the tip of the dendrite and where the remaining liquid solidifies region is defined as biphasic or paste and is associated with the temperature gradient and the evolution of the cooling conditions off balance.

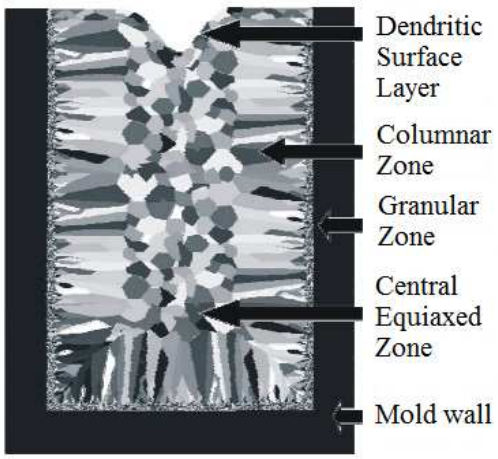

Figure 2. Different macro-regions commonly found in longitudinal sections of ingots cast.

3. MATHEMATICAL MODELING IN BINARY ALLOYS SOLIDIFICATION:

Optimization and process control in various areas has used mathematical modeling as a fundamental tool. The modeling of solidification phenomena has been progress, especially since 1980. Initially efforts were concentrated on the macroscopic aspects involved in the solidification and described by general equations of conservation. Virtually nothing was done to order to model the microstructure phenomena that occur during the solidification and the nucleation and growth of grains, among others, that are important to the properties and quality of the final product.

Then, to obtain a model that represents the solidification with higher property is required interconnection between the processes occurring in the macroscopic and microscopic scale, which has led to many studies on this field.

The mathematical model is based on the conservation equations of mass, energy and chemical species, together with the laws of nucleation and growth of dendritic structures. The solution of these equations is not an easy task in the case of dendritic solidification, due to the complexity of its geometry. This approach was used in early mathematical models seeking to solve the conservation equations for each phase and two models were named areas (liquid and solid).

Therefore, models were developed single domain in which the conservation equations are integrated within each phase present in a representative elementary volume (REV-"Representative elementary volume"). It was later introduced the concept of grain envelope, whereby, is associated with each grain within the mushy zone that surrounds a wrap, as shown in Fig. 3.

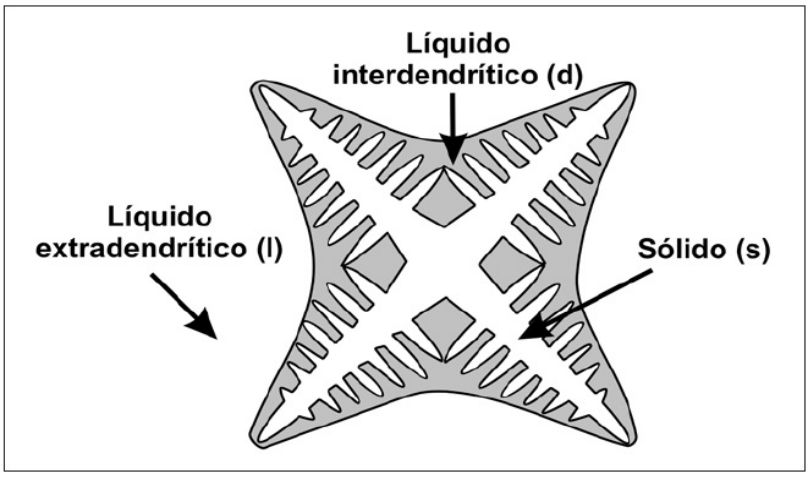

Figure 3. Concept of envelope applied to grain equiaxed grains. Aguiar (2006).

The mathematical model implemented in this work is based on the model developed by Martorano et al. (2003). Therefore, the equations are only briefly described, followed by detailed comments on the new features of the present model.

Martorano et al. (2003) defined three distinct pseudo fases within a representative elementary volume: solid (*s*); interdendritic liquid (*d*) and extradendritic liquid (*l*). The volume fractions of each of three pseudo fases are denoted by εs, εd and εl, respectively ($\varepsilon s + \varepsilon d + \varepsilon l = 1$). The inter and extradendritic liquid are separated by an extradendritic

imaginary envelope which just touches the arms primary and secondary dendrite. In the case of equiaxed grains, the envelopes are spherical and involve each grain equiaxed (Figure 3).

The following assumptions were also assumed: Eqation (1) cooling rate constant; Equation (2) has the interdendritic liquid solute concentration (*Cd*) homogeneous, Equation (3) the solute concentration in liquid follows the phase diagram (thermodynamic equilibrium with the solid), that is, $Cd = (T - Tf) / Ml$, where *T* is the local temperature, *Tf* is the melting temperature of the pure metal *Ml* is the slope of the liquidus line (approximated by a straight line), (diffusion of solute in the solid and the mass and heat transport by convection is negligible, the specific heat (*Cp*) and density (ρ) are constant and equal for all phases. The thermal conductivity (*K*) is given by the average conductivity of the solid phase (*Ks*) and liquid phase (*Kl*) weighted by their volume fraction locations. Then, for the one-dimensional system (in *z*) considered in the present study, the volume averaged conservation differential equations for energy, interdendritic liquid solute, and extradendritic liquid solute can be written, respectively, as (Martorano et al. 2003):

$$T(z,t) = Tl - G\ x\ VT \tag{1}$$

$$(1-K)Cd\ \frac{\partial \varepsilon s}{\partial t} = \varepsilon d\ \frac{\partial Cd}{\partial t} + Se\frac{Dl}{\delta e}\ (Cd - Cl) \tag{2}$$

$$\frac{\partial(\varepsilon l Cl)}{\partial t} = Cd\frac{\partial \varepsilon l}{\partial t} + Se\frac{Dl}{\delta e}\ (Cd - Cl) \tag{3}$$

The simulations are performed for a cooling rate of $G\ x\ VT = 0.005\ K / s$ and by a single control volume, so that all dependent variables are only a function of time.

Where *Tl* is the liquidus temperature corresponding to the initial alloy composition, *L* is the latent heat of fusion per unit mass, *Dl* is the diffusion coefficient of solute in the liquid; *Se* is the interfacial area envelope per unit volume, δe is the thickness of effective diffusion layer around the solute and envelopes and *k* is the partition coefficient of solute. Then, Eq. (2) can be solved for the solid fraction, εs, and Eq. (3) provides the average solute concentration in the extradendritic liquid, *Cl*.

The following equation is used to calculate the temporal evolution of the volume fraction of grain (or extradendritic liquid):

$$\frac{\partial \varepsilon g}{\partial t} = -\frac{\partial \varepsilon l}{\partial t} = SeV \tag{4}$$

knowing that: $\varepsilon g = \varepsilon s + \varepsilon d$ is the fraction of grains and *V* is the speed of radial growth of equiaxed grains and primary arms of columnar grains. This velocity can be obtained from the growth model originally proposed by Lipton et al. (1984):

$$V = \frac{4\sigma * Dl(k-1)Cd}{\Gamma}\ \left(\frac{1}{Iv}\ (\Omega)\right)^2 \tag{5}$$

$$\frac{1}{Iv}\ (\Omega) = 0.4567\ \left(\frac{\Omega}{1-\Omega}\right)^{1.195} \tag{6}$$

where $\sigma * \approx 1 / (4\pi^2)$ is the stability constant, Γ is the coefficient The Gibbs-Thomson, IV^{-1} is the inverse function of Ivantsov and Ω is the dimensionless undercooling solute, calculated as $\Omega = (Cd - Cl) / [Cd\ (1 - k)]$ for *V*, where *Co* is the average concentration of solute in the alloy.

The initial results are presented for two different equiaxed nuclei densities: $n = 2.4 \times 10^5$, $8.8 \times 10^6\ m^{-3}$. These nuclei densities give the following characteristic half-spacings for the equiaxed grains, respectively: $Rf = 10mm$ *and* $3mm$. Assuming these values of *Rf*, it is possible to calculate the instantaneous radius (*Re*) envelope by the following equation: $Re/Rf = (1 - \varepsilon l)^{1/3}$.

In earlier studies by Wang and Beckermann (1993) revealed that the thickness of effective diffusion δe can be calculated by the equation:

$$\frac{\delta e}{Re} = \left(1 - \frac{3}{2}Re\ \frac{(Rf^2 - Re^2)}{(Rf^3 - Re^3)}\right) \tag{7}$$

The system of coupled differential and algebraic equations presented above was solved numerically by the finite difference method using the Taylor series expansion.

4. RESULTS AND DISCUSSION

The current model shows the simulation results in solidification grain alloy Al-3%Cu with heat extraction unidirectional condition in which it is commonly found in the production of parts for casting. Whose properties are listed in Tab. 1:

Table 1. Physical properties and parameters used in the simulation of alloy Al-3 pct CU

Property	Al-3 pct Cu
Dl ($m^2.s^{-1}$)	5×10^{-9}
Ml (K.pct wt^{-1})	-3.37
K (-)	0.17
Γ (m.K)	2.41×10^{-7}
Tl (K)	923
T_{eut} (K)	821

The first results are shown for two different nucleation densities equiaxed grains: $n = 2.4 \times 10^5$ *and* 8.8×10^6 m^{-3}. Solving the equations described in the mathematical model presented above, we obtain a spacing between envelopes for the equiaxed grains, respectively of *Rf = 10 mm* and *3 mm*. Figure 4 shows the time course of the volume fraction of equiaxed grains in both cases, with the temperature variation. Figure 5 shows the corresponding variation in solute concentrations liquid.

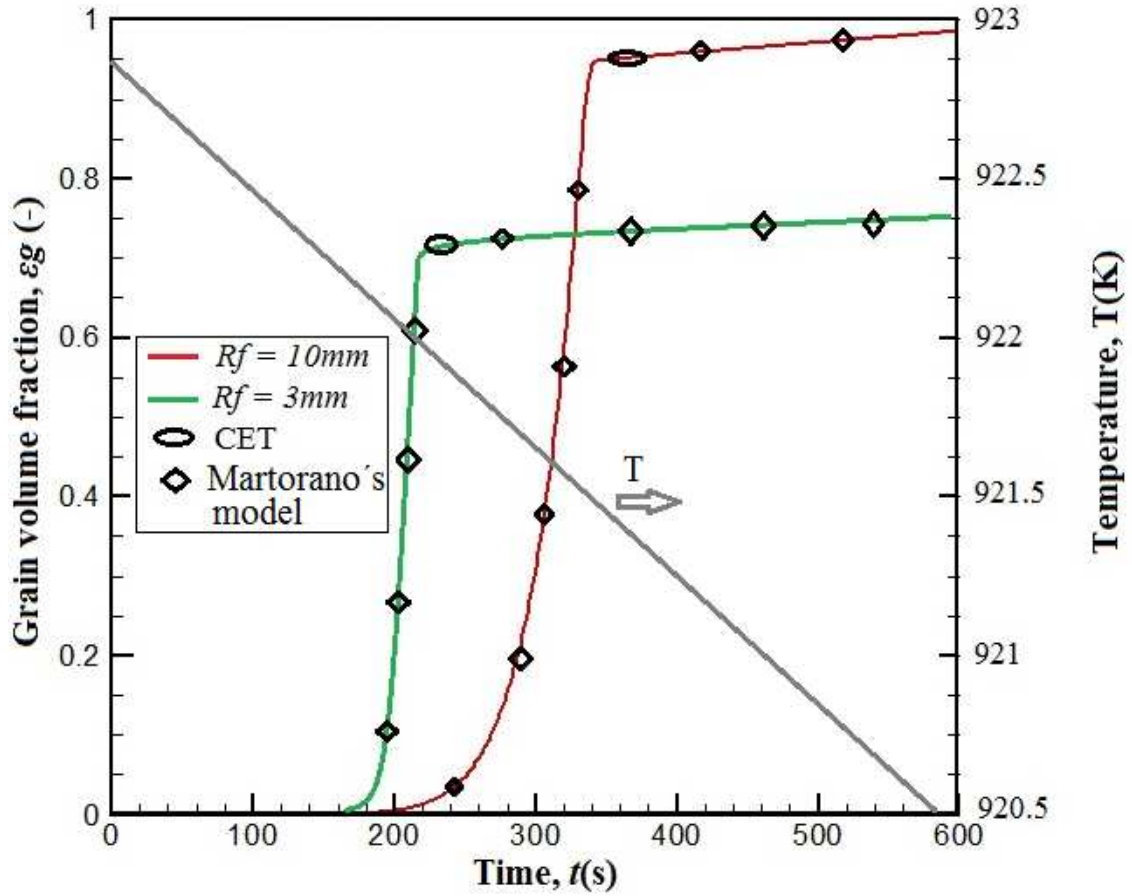

Figure 4. Comparison of grain volume fraction *εg* versus solidification time t(s) of the present paper; with the Martorano´s model.

Analyzing the curve corresponding to the largest equiaxed grain spacing *Rf = 10 mm* from Figure (4), the grain growth is initially very slow (up to 200 sec), which may be attributed to small amounts of concentration area envelope (*Se*) in Eq (4), knowing that: *Se ~ 1/Rf*. Figure 4 shows that, during this period, very little is rejected solute in the liquid

extradendritic (due to small values in the last term of equation (3)), and the concentration of extradendritic liquid, *Cl*, remains *Co* . From 200 seconds grains are subjected to a period of rapid growth until the grain fraction reaches a value of approximately 0.95 to 350 sec (Fig. 4). This rapid increase in εg can be attributed to *Se* in Eq (4) and cooling of solute, the same way by Eq (5) observed that the dendrite tip velocity reaches higher values simultaneously. In response to the decrease of the fraction of liquid extradendritic ($\varepsilon l = 1 - \varepsilon g$), the concentration rises very quickly *Cl* to *Cd* about 350 second (Figure 4), i.e., the sub-cooling is dissipated in a few seconds. The moment the super-cooling the solute reaches a very small value ceases dendritic grain growth is marked by an ellipse in Figures 4 and 5. This ellipse is called the "CET" only to indicate that, if the grains columnar were present, they would stop growing too.

A similar result is obtained for the case with *Rf* = 3 mm, except that cooling dissipates earlier due to higher values of *Se* at the beginning of the simulation (starting at approximately 150 sec). However, the fact *Cl* remains equal to *Co* throughout most of the growing season, combined with the rapid increase in εg near the point labeled CET.

Initially, the results obtained by the present model, *Rf* = 3mm and 10mm were compared with the results obtained by Martorano et al. (2003). The results obtained by Martorano, et al. (2003) identified with the direction of diamonds were almost the same as the current model compared to the two cases, as shown in Fig. 4. In Figure 5, we observe a small difference between the models, the curve of *Rf* = 3mm. But for both the CET occurs almost at the same instant (approximately 290 sec) with minimum variation of the liquid solute concentration, *C*. Soon the Figures 4 and 5 show a good agreement between the two models for both the volume fraction of grain εg, as for the liquid solute concentration *C*.

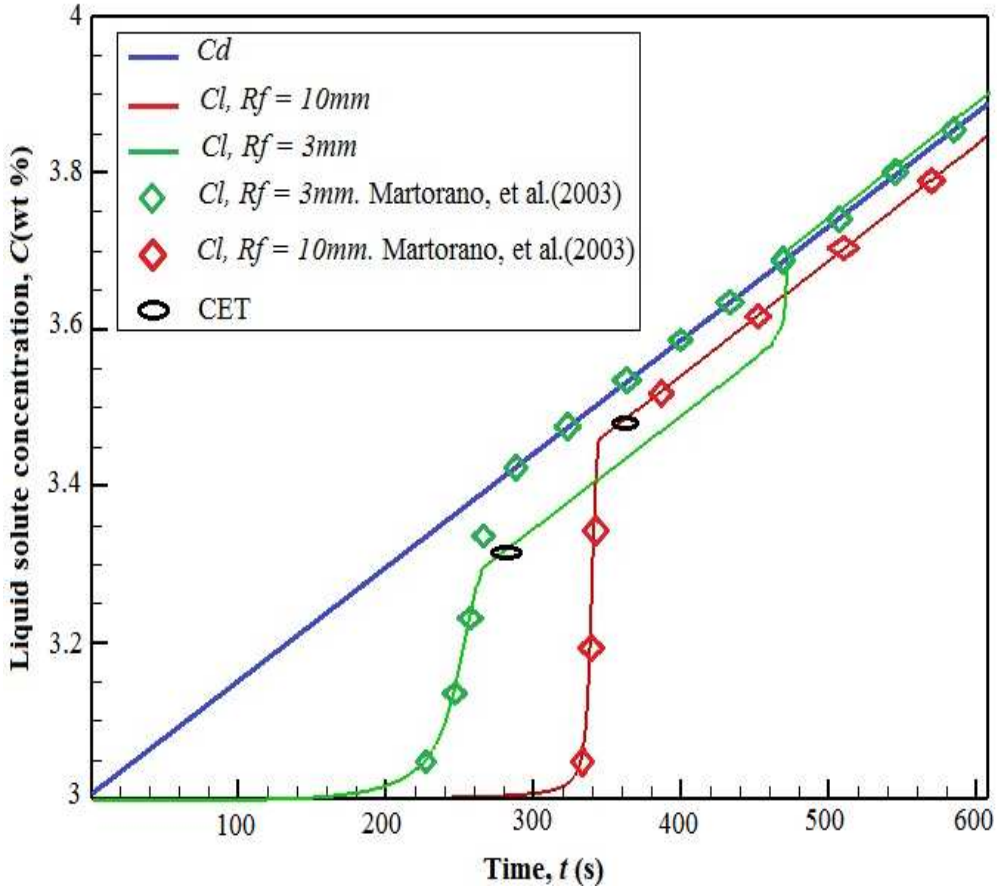

Figure 5. Liquid solute concentration C(%wt) versus solidification time t(s).

After validation of the results through the comparison of this model with the model Martorano, et al (2003) have presented results for three different spacing equiaxed nuclei density: $n = 4.0 \times 10^4$, 1.8×10^5 e 3.4×10^5 m^{-3}. According to the equation: $Rf = (3/4\pi n)^{1/3}$, these nuclei generate a spacing of approximately equiaxed grains characteristic, respectively: *Rf* = 18mm, 11mm and 9mm. The results of the model predicting the evolution of the grain fraction and temperature for three different spacing equiaxed grains, *Rf*, in a single volume control can be seen in Fig. 6. Similarly, the results of the model prediction of the evolution of solute concentration liquid for three different spacing equiaxed grains, *Rf*, in a single control volume can be seen in Fig. 7.

It can be concluded, from current data generated by this work, the density of nuclei equiaxed *n* is inversely proportional to the spacing between the envelopes *Rf*. However, the *Rf* value is directly proportional to the time of occurrence of the CET, as shown in Fig.6: For *Rf* = 18mm (present work occurs approximately at 410 sec), *Rf* = 11mm (present work occurs approximately 360 sec), *Rf* = 9mm (present work occurs at approximately 340 sec). The same can be seen by Fig.7.

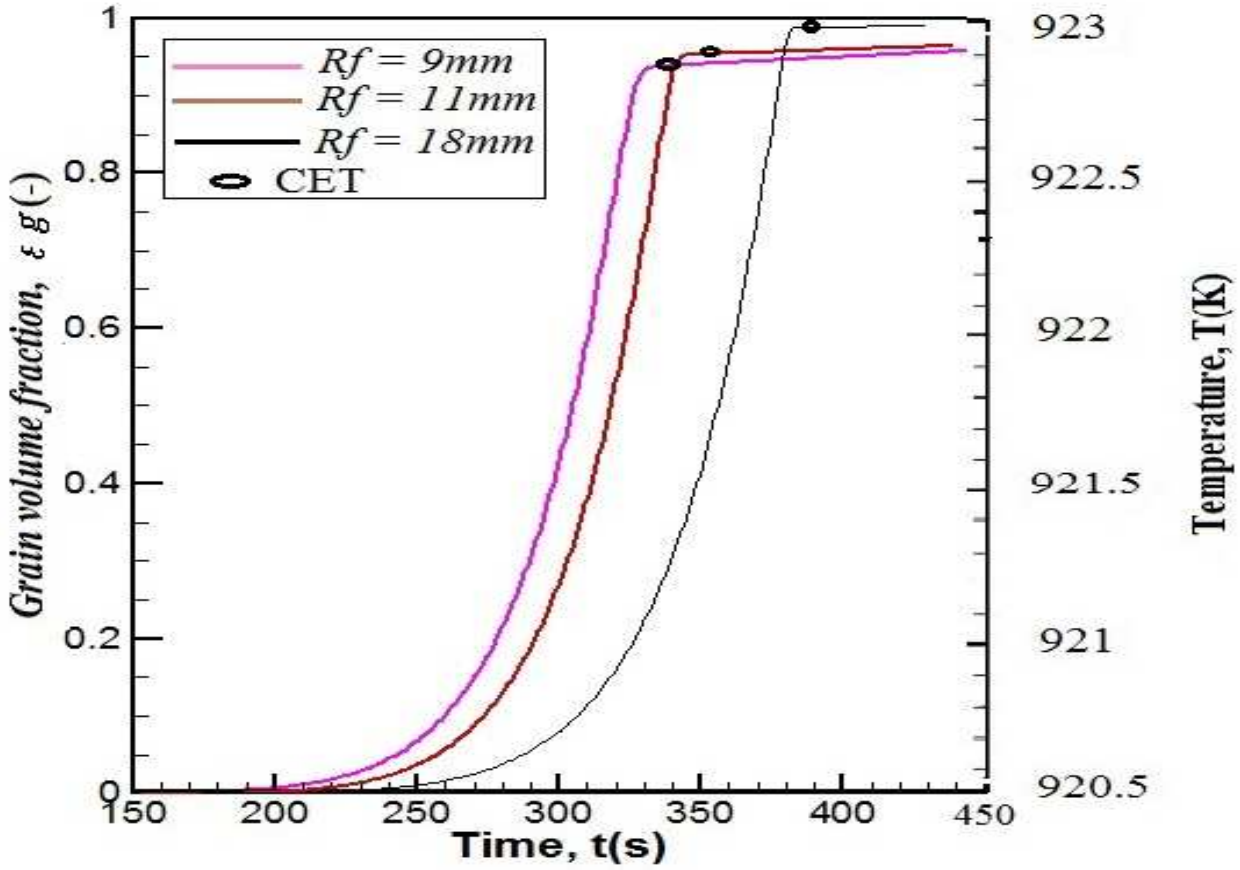

Figure 6. Grain volume fraction εg (-) versus solidification time t(s).

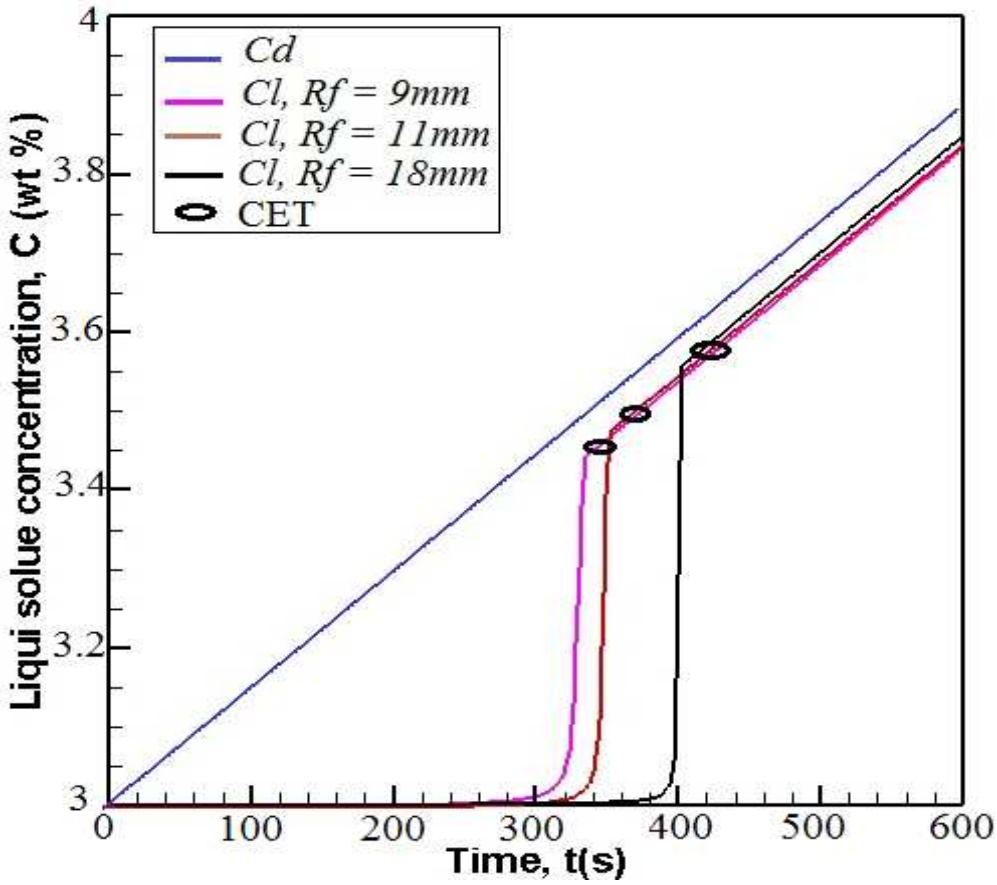

Figure 7. Model predictions for the solute concentration C(%wt) in liquid region versus solidification time t(s).

5. CONCLUSIONS

A mathematical model for multiphase unidirectional solidification of binary alloys has been implemented in this present work. In this paper, the forecasts on the CET were compared to the Martotano´s model, and we can note a good agreement between them. The same growth parameters were considered for both models.

The results were obtained by monitoring grain fraction and the concentration of the solute during the cooling of the alloy Al-3% Cu. Simulations were performed with different densities of nucleation and, therefore, different spacing between envelopes equiaxed grains, allowing the prediction of the size of the zone along the grain equiaxed and columnar size of the area indicated by the position of the columnar equiaxed transition (CET).

The simulation model can be used during the whole period of solidification, after the growth of the individual grains of different sizes being able to predict the time of occurrence of the CET.

6. ACKNOWLEDGEMENTS

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