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On an expression for the growth of secondary dendrite arm spacing during non-equilibrium solidification of multicomponent alloys: Validation against ternary aluminum-based alloys

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ABSTRACT

The technological importance of the microstructure length scale is directly related to the influence exerted on solute redistribution and microporosity formation and on mechanical properties, such as, toughness, ductility, ultimate and yield tensile strengths. There is a huge lack of literature concerning theoretical predictive dendritic growth models for unsteady-state solidification of multicomponent alloys. Most of the existing models have been proposed for steady-state solidification and for binary alloys. One of these models, initially restricted to binary alloys, has been extended for multicomponent alloys; however, it was shown to be valid only for low growth rates and small dendrite tip undercooling, that is, conditions that are very close to those of equilibrium cooling from the melt. In this paper, an extended approach is proposed, encompassing the back diffusion parameter β to allow back diffusion treatment to be included in the analysis. A technique based on Butler's formulation and on thermodynamic databases is used to permit necessary thermophysical parameters, such as the surface energy and the Gibbs-Thomson coefficient to be calculated for Al-Cu-(Si;Mg) alloys. Directional solidification apparatuses are used to provide a wide range of experimental solidification cooling rates and growth rates along the length of the directionally solidified castings. The model predictions are validated against the experimental scatters of secondary dendrite arm spacings of Al-Cu-Si; Mg) alloys castings solidified under transient upward and horizontal heat flow conditions. It is shown that the predictions fit quite well the experimental results.

1. Introduction

The primary advantage of Al-based alloys is their relatively high-strength to weight ratio, which make them commercially attractive to the manufacture of components in the automotive, transportation, and aerospace industries. Most commercial casting Al-alloys are multi-component, multiphase alloys, whose properties are associated with the microstructure that develops during solidification. For multicomponent Al alloys, the microstructural evolution along solidification is less understood than in the case of single phase and multiphase binary alloys. One of the key challenges is to understand the way the microstructures of multicomponent alloys are formed and how they can be controlled during solidification. The design of optimized microstructures for a particular application depends in one hand on the knowledge of microstructural evolution represented by appropriate theoretical and experimental growth models; on the other hand on reliable databases for all the relevant thermophysical properties that will be necessary in other to perform model predictions for any alloy to be examined [1].

The dendrite arm spacing is known to play a significant role in the improvement of mechanical properties. The solidification thermal parameters affect the microstructure length scale, the microsegregation pattern, the size, morphology and distribution of second phases and porosity, which in turn influences toughness, ductility, ultimate and yield tensile strengths and homogenization kinetics of as-solidified alloys [2–5]. The prediction of the microstructural development during transient solidification is possible by the application of phase field models for binary alloys [6–10] and ternary alloys [11–13]. Since the phase field model is applied to microscopic domain [14,15], the macroscopic solidification model has several orders of magnitude in space and time scales [16], making the coupling between both models a very

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hard and computational intensive task. Because of the complexity of dendritic growth, another approach is the application of predictive growth models existing in the literature. The majority of them have been developed for binary alloys, and limited to steady-state solidification, exception being the theoretical models proposed by Hunt and Lu [17] for cellular and dendritic arm spacings and Bouchard and Kirkaldy [18] for primary and secondary dendritic arm spacings. Both models are supposed to encompass also the dendritic growth in unsteady-state heat flow conditions.

Theoretical and experimental models for prediction of dendritic growth in multicomponent alloys are very scarce in the literature because of the difficulty of the task. An important mathematical model was proposed by Kirkwood based on the dissolution of small arms from their tips, providing secondary dendrite arm spacing as a function of time during solidification for both isothermal and constant cooling rate [19]. An expression used for calculation of secondary dendrite arm spacing (λ_2) of binary alloys, which is based on dendrite ripening as the main coarsening mechanism, has been extended by Rappaz and Boettinger to multicomponent systems [20]. These authors proposed a consistent model of equiaxed dendritic solidification for multicomponent alloys, which is able to encompass the coupling of the dendrite growth kinetics and the global solute balance performed at the local scale of the grain. They validated their approach against experimental λ_2 values for various superalloys, and reported that the calculated values fitted quite well the experimental scatter. Easton *et* al. [21] compared experimental λ_2 values of six different multicomponent Al-alloys with calculations performed with the approach suggested by Rappaz and Boettinger, and reported that the general agreement was shown to vary from 20 to 70%. Costa et al. [5] compared experimental results of λ_2 for an Al–6 wt%Cu–4 wt%Si alloy solidified both upward and horizontally under transient heat flow conditions with values calculated by the approach proposed by Rappaz and Boettinger. These authors observed that, in the case of the horizontally solidified alloy casting, the solutal induced convection during growth from the melt does influence the composition gradient in the melt, and hence the growth of λ_2. They state that, since the theoretical approach does not encompass convective effects, the resulting predictions were not expected to fit adequately the corresponding experimental scatter. However, the experimental λ_2 scatter for the upward solidified casting, which was solidified under stable melt conditions, was shown to be located even farther from the calculated values.

By deriving their approach, Rappaz and Boettinger [20] have taken into consideration the following assumptions for the growth of dendrite tips: (i) no thermal gradient at the scale of the grain; (ii) negligible thermal undercooling; (iii) low growth rate; (iv) growth at the marginal stability limit; (v) independent solute fields given by Ivantsov solution; (vi) negligible off-diagonal diffusion term. Regarding these assumptions, added to the fact that they also neglected high solidification rate factors, the behavior is approximately that expected in the condition of phase diagrams global thermodynamics equilibrium, where the undercooling is small, and so is the growth rate. The inaccuracy in calculations observed for case of transient solidified samples reported by Easton *et* al. [21] and Costa *et* al. [5] may be associated with the higher solidification cooling rates and to the corresponding solute redistribution. In this paper, a modified approach is proposed for the prediction of λ_2, which takes into consideration the back diffusion parameter β to allow a back diffusion treatment. A theoretical approach is used to calculate the Gibbs-Thomson coefficient for Al-Cu-(Si;Mg) alloys, a fundamental parameter for the prediction of λ_2. The calculated λ_2 values are validated against experimental results of Al-Cu-(Si;Mg) alloys, for both upward and horizontal transient solidification conditions.

2. Secondary dendrite arm spacing of multicomponent alloys

2.1. Rappaz and Boettinger approach for multicomponent alloys

Rappaz and Boettinger [20] extended a usual expression for secondary dendritic growth of binary alloys for multicomponent systems. The general expression is given by:

$$\lambda_2 = 5.5\,(M \bullet t_{SL})^{\frac{1}{3}} \tag{1}$$

where t_{SL} is the local solidification time, and M is defined as follows

$$M = \frac{-\Gamma}{\sum_{j=1}^{n} m_j(1-k_j)(c_{f,j}-c_{0,j})/D_j} \ln\left[\frac{\sum_{j=1}^{n} m_j(1-k_j)c_{f,j}/D_j}{\sum_{j=1}^{n} m_j(1-k_j)c_{0,j}/D_j}\right] \tag{2}$$

where Γ is the Gibbs–Thomson coefficient, m_j is the liquidus slope, $c_{f,j}$ is the final liquid composition at the dendrite root of the component j (generally assumed to be a eutectic composition), $c_{o,j}$ is the nominal concentration of the alloy, D_j is the diffusion coefficient in the liquid, n is the maximum number of solute elements (j = 1, n) and k_j is the redistribution coefficient. The subscript j represents each alloying element, and the sum encompasses all the solute elements of the multicomponent alloy.

By deriving their approach, the authors assumed that the dendrite is growing at the marginal stability limit, in which the radius is given by:

$$R = 2\pi\sqrt{\frac{\Gamma}{\sum_{j=1}^{n} m_j G_{C,j}}} \tag{3}$$

where $G_{C,j}$ is the solute gradients at the dendrite tip, which is expressed by the following equation:

$$G_{c,j} = -\frac{v}{D_j}(c^*_{l,j}-c^*_{s,j}) = -\frac{v}{D_j}\frac{c_{0,j}(1-k_j)}{1-(1-k_j)\,Iv(Pe_j)} \tag{4}$$

where v is the tip growth rate and $c^*_{l,j}$–$c^*_{s,j}$ are the concentrations at the tip in the liquid and solid phases, respectively.

The difference between the liquid concentrations at the tip and far from the tip, i.e., the supersaturation associated with the solute element j, is given by:

$$\Omega_j = \frac{c^*_{l,j}-c^*_{0,j}}{c^*_{l,j}-c^*_{s,j}} = \frac{c^*_{l,j}-c^*_{0,j}}{c^*_{l,j}(1-k_j)} = Iv(Pe_j) \tag{5}$$

where the Peclet number Pe_j is

$$Pe_j = \frac{Rv}{2D_j} \tag{6}$$

The Ivantsov function [19–22] of species j Peclet number $Iv(Pe_j)$, can be given by

$$Iv(Pe_j) = Pe_j\, e^{Pe_j}\int_{Pe_j}^{\infty}\frac{e^{-u}}{u}du \tag{7}$$

The dendrite radius (R) assumed in this approach is that from Bobadilla *et* al. [23]:

$$R = 2\pi\sqrt{\frac{\Gamma}{\sum_{j=1}^{n} m_j G_{C,j}\xi(Pe_j)-G}} \tag{8}$$

where $\xi_C(Pe_j)$ is a function of the Peclet number, which for low tip cooling rate is approximately 1 [24] and G is the average liquid thermal gradient near the dendrite tip.

The approach proposed by Rappaz and Boettinger assumed the Peclet function $\xi_C(Pe_j) \approx 1$, nevertheless it should be greater than 1 when associated with high solidification rates, in this case neglected by