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A new concept for growth restriction during solidification

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ABSTRACT

Growth restriction refers to the phenomenon of reduced growth velocity due to the solute enrichment/depletion at the solid/liquid interface during alloy solidification. Although significant progress has been made to understand this phenomenon, so far there has been no effective parameter to quantify growth restriction. In this paper, we have derived a new parameter, β , to quantify the growth restriction in multicomponent systems effectively, and which incorporates the nature of solutes, solute concentrations and solidification conditions holistically. Theoretical analysis and phase field simulations have confirmed that growth velocity is a unique function of β regardless of the nature of solutes, solute concentrations and solidification conditions, but it is not a unique function of the widely used growth restriction factor, Q . Our analysis suggests that the overall β for a multicomponent alloy system can be either calculated accurately by the ratio of the liquid fraction to the solid fraction ($\beta = f_l/f_s$) or approximated with great confidence by a linear addition of the β values of the constituent binary systems. In addition, we have shown theoretically that for a given alloy system solidifying under a given undercooling, there is a critical solute concentration, below which solidification becomes partitionless and therefore there is no growth restriction during solidification. Furthermore, our analysis has shown that the physical origin of growth restriction is the blockage of the supply of the critical elements for crystal growth, i.e., solvent atoms in the case of eutectic-forming.

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1. Introduction

During solidification of metallic alloys, solutes partition differentially between the solid and liquid phases, resulting in either accumulation or depletion of solutes at the solid/liquid interface depending on the nature of the solute(s) in a given alloy. It has been long recognized that such accumulation/depletion of solutes leads to a slower growth rate of the solid phase [1], and this phenomenon is usually referred to as growth restriction in the literature. Historically, Cibula [1] was the first person to recognise such a phenomenon by suggesting that there is a concentration gradient in the liquid around a growing solid during solidification. Ivantsov [2] discussed the solute field in the liquid around a growing solid and proposed the concept of constitutional supercooling, which was described more rigorously later by Rutter and Chalmers [3], and formulated quantitatively by Tiller et al. [4]. Soon after, Winegard and Chalmers [5] made the connection between constitutional supercooling and equiaxed grain formation during solidification.

Since then the effect of solute concentration on grain refinement has been demonstrated experimentally by a number of researchers [e.g., 6–10].

Our current knowledge of growth restriction and its effect on solidification is mainly obtained from the research on grain refinement. Due to the desire for homogenous microstructure, improved crack susceptibility and better machinability, grain refinement has been widely investigated both experimentally and theoretically over the past decades, especially in Al- and Mg-alloys (see reviews in Refs. [11–13]). In the beginning, attention was focused mainly on the importance of the nucleant particles on grain refinement [1,14,15]. However, Wallace [16] recognized the role of solute on grain refinement, and Tarshis et al. [17] demonstrated that in a range of Ni- and Al-based alloys the addition of solute led to significant grain refinement. Since then, substantial attentions have also been paid to the solute effect on grain refinement [13,18,19]. Easton and StJohn [13] reviewed the mechanisms of grain refinement and divided the theoretical and experimental work into two categories, the “nucleant paradigm” and the “solute paradigm”. The former emphasises the importance of the nucleant particles on grain refinement, while the latter incorporates the influence of solutes on the grain refinement process. It is now well

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accepted that effective grain refinement requires the presence of both potent nucleant particles with adequate number density and sufficient solute contents [20]. For instance, it has been demonstrated that solidification of alloys without the presence of adequate solute leads to a columnar microstructure even if an exceptionally large number of potent nucleant particles are present in the melt [21,22]. It is generally accepted that increasing solute content results in a later start and slower rate of recalescence, which in turn allows more time for further grain initiations to proceed.

So far, there has been substantial effort dedicated to a quantitative description of growth restriction during solidification (see review in Ref. [23]). Tarshis et al. [17] proposed a parameter, the constitutional supercooling parameter P , to quantify the solute effect, and correlated grain size with P in Ni- and Al-based alloys:

$$P = \frac{mC_0(k-1)}{k}, \quad (1)$$

where m is the liquidus slope in a linear phase diagram, k is the equilibrium solute partition coefficient, and C_0 is the solute content in the alloy melt. Spittle and Sadli [24] studied the effect of Cr, Cu, Fe, Mg, Mn, Si, Zn and Zr on the grain size of high purity Al with and without 0.2 wt.% addition of Al-5Ti-1B grain refiner. Their results showed that there appears to be a good correlation between the grain size and the constitutional supercooling parameter P .

Another more popular parameter for quantifying growth restriction is the growth restriction factor, Q , which was first defined by Maxwell and Hellawell [25] as $1/X$ in their modelling of spherical growth restricted by the partitioning of a single solute. This factor ($1/X$) was considered to be the growth restriction factor, and denoted as Q later by other researchers:

$$Q = mC_0(k-1). \quad (2)$$

From Eqs. (1) and (2), one has $P = Q/k$. Greer et al. [26] replotted the grain size data of Spittle and Sadli [24] as a function of the growth restriction factor Q , and found that Q is a better parameter than P for quantifying the degree of growth restriction. In addition, it has been suggested that Q could be defined as the available undercooling for the formation of the initial solid and therefore Q can be expressed as [27]:

$$Q = \left(\frac{\partial(\Delta T_C)}{\partial f_S} \right)_{f_S \rightarrow 0}, \quad (3)$$

where ΔT_C is the constitutional undercooling, and f_S is the solid fraction. One of the advantages of Eq. (3) is that it offers a thermodynamic approach to predicting the Q value for multicomponent alloys [28,29]. However, the approach to the growth restriction factor of a multicomponent system commonly used in the literature is often a simple linear addition of those for the constituent binary systems [8,10,30]:

$$Q = \sum_{i=1}^n m_i C_{0i} (k_i - 1), \quad (4)$$

where i refers to the individual solute in the multicomponent system.

Hodaj and Durand [31] proposed another parameter, U , to quantify the growth restriction of solutes in multicomponent systems by considering the difference in diffusion coefficients of solutes during solidification under low undercooling,

$$U = D \sum_i \frac{1}{D_i} m_i C_i (k_i - 1), \quad (5)$$

where D is the self-diffusion coefficient of the solvent, and D_i is the diffusion coefficient of the i th solute in the liquid. In Eq. (5) the contributions of the constituent solutes are weighted inversely by their diffusivities D_i . However, it is usually difficult to obtain reliable data for solute diffusivities in liquid alloys, and therefore, it is a common practice to use a constant diffusion coefficient for all the solutes during numerical modelling of solidification processes.

The parameters, P and Q , have been used extensively to account for the experimentally observed effect of solutes on grain size [10,17,24,30,32–35]. Tarshis et al. [17] and Spittle and Sadli [24] explained their experimental results in terms of P and found that their measured grain sizes were closely related to the parameter P . However, more work [10,30,32–35] has focused on the relationship between grain size and the growth restriction factor Q . In addition, a few theoretical models for predicting grain size have also been developed involving Q [25–27], [36–38]. For example, Maxwell and Hellawell [25] developed a simple model for spherical growth during solidification of an isothermal melt, in which the parameter $1/X$ ($1/X = Q$) was identified as the growth restriction parameter. Based on the Maxwell and Hellawell model [25], Greer et al. [26] developed a numerical model for predicting grain size of Al-alloys with grain refiner addition, and found that a grain grows from a refiner particle at an undercooling inversely proportional to the particle diameter, which has been referred to as the free growth model:

$$\Delta T_{fg} = \frac{4\gamma}{\Delta S_v d}, \quad (6)$$

where ΔT_{fg} is free growth undercooling, γ is the solid/liquid interfacial energy, ΔS_v is entropy of fusion per unit volume, d is particle diameter. They found that their numerical predictions [26] agree well with the experimental data of Spittle and Sadli [24] up to 400 μm , but not beyond that. In addition, extensive experimental work has revealed an empirical relationship between the average grain size (\bar{l}) and the growth restriction factor Q [32,37,39–42]:

$$\bar{l} = a + \frac{b}{Q} \quad (7)$$

where a and b are constants. It was believed that a is related to the number density of active nucleant particles and b is related to the potency of the nucleant particles [42,43]. Further analysis has led to the development of the interdependence theory [37]. Furthermore, in the recent years we have seen more theoretical models for grain size prediction [36,44,45], which involve the growth restriction factor Q either directly or indirectly.

However, although experimental results in dilute binary alloys have demonstrated that grain size decreases monotonically with increasing Q , there has been increasing evidence to show that Q does not always work well for quantifying growth restriction during solidification. When examining the effect of solute content on grain size over the entire range of hypoeutectic compositions in binary alloys, Xu et al. [46] found that the relationship between the grain size and P or Q is not monotonic at high alloy concentrations [46]. They suggested that the grain size decreases monotonically with increasing freezing range of aluminium alloys, with the minimum grain size occurring at the maximum freezing range. How-

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