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An analytical approach for predicting as-cast grain size of inoculated aluminum alloys

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ABSTRACT

An analytical model is proposed to predict as-cast grain size of inoculated aluminum alloys during isothermal and non-isothermal solidification. The model is derived from a unified nucleation ceasing criterion which takes into account two stifling effects resulted from recalescence and solute segregation. The theoretical framework adopted by Greer et al. is utilized to account for the phase transformation kinetics during both isothermal and non-isothermal solidification. The proposed analytical model is able to predict maximum nucleation undercooling and hence the as-cast grain size. With some rational assumptions, the model yields a novel relationship between grain size and diffusivity-weighted Grain Restriction Factor, *U*. The model predictions on grain size for a variety of binary alloys has been compared with that from the reported semi-empirical relation, numerical solution and experimental measurements, and good agreements have been achieved. Unlike previous analytical models, all the input parameters of the present model are physically meaningful, including thermo-physical properties, alloy composition, cooling and inoculation conditions. Therefore the model can be applied directly to various alloys systems. It is concluded that the proposed model is valuable in identifying alloy composition and processing parameters to optimize as-cast grain size.

1. Introduction

The prediction of as-cast grain size of aluminum alloys is not only of a great significance in fundamental researches on the solidification microstructures but also of vital practical importance in industries. As-cast grain size prediction has therefore been a very attractive research field and great efforts have been made by numerical approaches [1–5] and analytical models [6–10]. Maxwell and Hellawell [1] made the seminal contribution by developing a numerical approach to predict grain size, and concluded that recalescence was the main factor to stifle nucleation under isothermal solidification condition. Greer et al. [2] has proposed a free growth model to describe the onset free growth of grains on the inoculant particles in the undercooled melt, and enabled a quantitative prediction of grain-refining efficiency. Quested et al. [3] studied the effect of solute field on grain size in the directional solidification where recalescence was absent, indicating that solute effect was the controlling factor to stifle nucleation. Shu et al. [4] also emphasized the importance of solute stifling effect, however, they overestimated this effect due to the violation of the solute conservation law in their model as illustrated by Du et al. [5]. StJohn and his coworkers [10] proposed the interdependence theory to describe the competition between grain growth and nucleation, and concluded that minimizing the nucleation-free zone was crucial to improve inoculant efficacy and promote nucleation and grain refinement.

As well reviewed in the literatures [11–13], there are several measures which could be applied to refine as-cast grain size and have to be included in a predictive as-cast grain size model. The first is the additions of inoculants into the melt of aluminum alloys. The inoculants act as effective substrates for the initiation of grains, and this could be well described by the free growth model proposed by Greer et al. [2]. Secondly, controlling the cooling rate is commonly employed to refine grains in the industrial production. In general, faster cooling gives a finer grain size. Alloying is the third important approach to reducing grain size [14]. Alloying components are proved to have three effects on the grain refinement of aluminum alloy: reducing the growth velocity of the grains (i.e., limiting the release of latent heat) which allows more time to nucleate, generating nucleation free zone via solute enrichment [3], and poisoning effect (i.e., solutes may react with grain refiners, forming some unexpected compounds and exerting side effects on inoculants) [15]. In the present work, we are aiming to

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develop an analytical model to predict as-cast grain size by taking into account inoculation conditions, cooling rate and solute content. The chemical interaction of solute with grain refiners is neglected.

When quantifying the effects of a solute on grain refinement, the parameter Q, referred to as Growth Restriction Factor (GRF), is often used. For binary alloys, Q could be written as

$$Q = mX_0(k-1) \tag{1}$$

where m is the liquidus slope, k is the equilibrium partition coefficient and X_0 is initial solute content in the alloy melt. Some research groups have established analytical approaches to describe the relationship between grain size and Q. StJohn et al. [8,10] have suggested that the average grain size $\bar{l}$ is a linear function of $1/Q$ for a number of alloy systems

$$\bar{l} = a + b/Q \tag{2}$$

where a and b are empirical constants and related to the number density of active nucleating particles and the efficiency of the nucleating particles, respectively. In contrast to this linear relation, Men and Fan [9] have proposed a cubic root law relation for isothermal solidification

$$\bar{l} = K(1/Q)^{1/3} \tag{3}$$

where K is a constant which is independent of alloy composition but strongly dependent on the alloy system, solidification conditions and physical nature of the nucleating particles. The different exponents of $1/Q$ in these two models stem from the different assumptions used in the contributions. The exponent 1 in Eq. (2) is a semi-empirical number [7,8]. However, the plots of grain size against $1/Q$ frequently deviate from the linear approximation at the high Q end, as illustrated by Men et al. [9]. The exponent 1/3 is derived from three fitted polynomial relations for solid fraction, nucleation undercooling and average growth velocity with $1/Q$ at a power of −0.71, −0.71 and 0.14, respectively (see Fig. 6 in Ref. [9]). The two models could be improved as they both need several input tuning parameters. For example, according to the model proposed by StJohn et al. [10], the growth rate and nucleation undercooling are necessitated to be known in advance, while in the model by Men et al. some careful and accurate experiments are needed to determine the coefficient K in Eq. (3) for a given alloy system solidifying under similar experimental conditions. Therefore, dedicated work to derive a parameter-free analytical model which relates the alloy thermo-physical properties and solidification conditions to final grain size is desired. Furthermore, it should be pointed out that using Q as a parameter to measure the effects of solute elements is not sufficient. A simple argument for this conclusion is that different alloy systems with the same Q value usually have different grain sizes as indicated by many experimental measurements [16]. In fact, besides Q, the diffusivity of solute elements also plays a vital important role in determining grain size. Although there are some attempts to address this issue by weighting Q with diffusivity [17,18], a new and general parameter derived in a rigorous manner to replace Q is needed.

Given inoculation condition, cooling condition and melt chemical composition, a predictive as-cast grain size prediction model needs to integrate nucleation termination mechanisms. As summarized in Ref. [5], there are two mechanisms leading to the termination of nucleation events, i.e., recalescence stifling and solute segregation stifling. In the isothermal solidification of a small volume aluminum melt, nucleation events often terminate due to recalescence stifling. The solute segregation stifling is dominant in the case of non-isothermal solidification, for example, directional solidification of a large volume aluminum melt and industrial DC casting [3,5]. In the present paper, the solidification kinetics model adopted in [6] and the unified nucleation ceasing criterion proposed in Ref. [5] are combined to derive an analytical grain size prediction model. The model will be validated using the reported experimental and numerical simulation results. As to be shown in this paper, the proposed model also enables the derivation of a novel relationship between grain size and diffusivity-weighted Growth Restriction Factor.

The paper is organized as follows: Section 2 concerns how the analytical model is constructed; then the model is applied in Section 3 to predict grain size of inoculated as-cast aluminum alloy under the two typical circumstances: isothermal and directional solidification; the model validations are also included in this section. Section 4 illustrates the derivation and validation of a novel relationship between grain size and diffusivity-weighted Q.

2. Model description

The final grain size of an as-cast alloy is a result of the competition between nucleation and solid grain growth. Here we follow the framework proposed by Greer et al. [6] to describe the liquid-to-solid transformation kinetics and apply the unified criterion [5] to treat both isothermal and non-isothermal solidification. The construction of the model is described below.

2.1. Nucleation

The free growth model proposed by Greer et al. [2] is adopted here to calculate nucleation rate, of which the validity has been partially confirmed by Tóth et al. using dynamical density functional theory [19], and has been widely applied by other researchers [4,5,10]. The model considers nucleation as a deterministic phenomenon and directly links the nucleation undercooling ΔT with critical diameter d of a refiner particle that is able to initiate a grain, and it is expressed as

$$d = \frac{4\gamma}{\Delta S_f \Delta T} \tag{4}$$

where γ is the solid–liquid interfacial energy and ΔS_f is the entropy of fusion. The model illustrates that at an undercooling ΔT, the particles with a diameter larger than d are active for nucleation [2].

One of the key input parameters to the free growth model is nucleant particles size distribution (*PSD*). The exponential form, fitted on the experimentally measured particle distribution by Greer et al. [2], is used to describe the nucleant particle size distribution

$$PSD(d) = \frac{N_0}{d_0}\exp\left(-\frac{d}{d_0}\right) \tag{5}$$

where N_0 is the total population of particles, d_0 is the characteristic width of the distribution. Integrating the above equation from d to ∞, the population, N, of the particles whose diameters are larger than d can be obtained

$$N = N_0\exp\left(-\frac{d}{d_0}\right) \tag{6}$$

The interfacial energy and entropy of fusion of aluminum alloys used in following calculations are listed in Table 1, which are taken from [2,5].

2.2. Solid grain growth

As a result of solid grain growth, latent heat is released into the melt which in turn increases its temperature. Meanwhile, rejected solute piles up ahead of the solid–liquid interface, developing a solute diffusion boundary layer and leading to the solute impingement effect [3]. Therefore solid grain growth will impose a significant effect on both temperature field and solute field, and thus affects the final grain size.