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ScienceDirect

Acta Materialia 71 (2014) 380–389

Acta MATERIALIA

www.elsevier.com/locate/actamat

An extension of the Kampmann–Wagner numerical model towards as-cast grain size prediction of multicomponent aluminum alloys

Qiang Du *, Yanjun Li

SINTEF Materials and Chemistry, 7465 Trondheim, Norway

Received 2 August 2013; received in revised form 12 March 2014; accepted 14 March 2014
Available online 14 April 2014

Abstract

The Kampmann–Wagner numerical (KWN) model, which has been widely adopted as a precipitation modeling framework accounting for concurrent nucleation, growth and coarsening kinetics, was extended to predict the as-cast grain size of inoculated multicomponent aluminum alloys. In the model, the heterogeneous nucleation of grains on inoculant particles was modeled based on the free growth criterion, while the influence of the solute on the nucleation behavior, in terms of the solute suppressed nucleation (SSN) effect, was rigorously defined and integrated. In order to fully address the solidification behavior of multicomponent alloys, a coupling of the KWN model to CALPHAD was carried out. These extensions allow the treatment of two different nucleation-ceasing mechanisms induced by grain growth: recalescence stifling and solute segregation stifling. Given melt composition, inoculation and heat extraction rate, the model is able to predict maximum nucleation undercooling, cooling curve and the final as-cast grain size of multicomponent alloys without invoking the binary equivalence assumption used in the existing models. The proposed model was tested with a variety of binary and multicomponent aluminum alloys, and the predictions were compared with the experimental measurement results and previous grain size prediction models. The simulation results show that the SSN effect has a negligible influence on the nucleation behavior and the final grain size during isothermal melt solidification, but a strong influence on the ceasing of grain nucleation during directional solidification. Reasonable agreement was obtained between the model prediction and measurement results on a direct chill casting experiment of an AA5182 alloy. Our work proves that the application of the precipitation modeling framework for the solidification problem is successful.

Keywords: Solidification modeling; Grain size prediction; Aluminum alloys; Precipitation modeling; CALPHAD

1. Introduction

As-cast grain size, resulting from the competition between heterogeneous nucleation and the growth of grains during solidification, is a very important microstructure feature which exerts a strong influence on the casting properties and mechanical properties of castings. In general, it is difficult to predict the grain size as casting involves nucleation. Fortunately, based on the free growth concept proposed by Greer et al. [1], the nucleation in the heavily inoculated melt such as the one encountered in aluminum alloy casting is deterministic and could be well described provided that the size distribution of inoculation particles is known. Therefore the difficulties related to nucleation modeling have been circumvented, and the focus is on the competition between nucleation and growth to predict as-cast grain size.

Most of the recent numerical models for grain size prediction of inoculated aluminum alloys [1–6] have followed the approach pioneered by Maxwell and Hellawell [7]. The approach was developed on the hypothesis that the grain growth is controlled by the diffusion of solute elements in the melt and the nucleation of grains stops due to recalescence. The other interesting alternative approach is the

* Corresponding author.
E-mail addresses: qiang.du@sintef.no, duqiang57@hotmail.com (Q. Du).

http://dx.doi.org/10.1016/j.actamat.2014.03.028
1359-6454/

"interdependence theory" proposed and experimentally validated by StJohn et al. [8]. However, the approach, being semi-empirical in nature, requires the input of the two solidification-condition-dependent parameters: grain growth rate and maximum nucleation undercooling (equal to the liquidus of the bulk melt minus the nucleation-ceasing temperature).

Maxwell and Hellawell's numerical (MHN) approach is only applicable to isothermal solidification where recalescence occurs [7]. While for the spatially non-isothermal melt solidification such as industrial direct chill (DC) casting processes and directional solidification experiments where recalescence is not present [9], the as-cast grain size cannot be predicted by MHN type models as no nucleation-stopping mechanism is available, as pointed out in Ref. [4]. Therefore, an extension to the MHN model by including other nucleation-ceasing mechanisms is necessary. Quested et al. proposed that the solute effect induced by grain growth on nucleation is important when recalescence is absent, and nucleation ceases when the reduced constitutional supercooling zones surrounding growing grains start to impinge [4]. This zone has also been analyzed by Shu et al. [6] (called the "solute suppression nucleation" (SSN) zone), StJohn et al. [8] (called the "nucleation free zone") and by Men and Fan [5]. To avoid computational complexity, Quested and Greer approximated the reduced constitutional supercooling zone size as twice the effective size of the growing grain and developed the dendritic impingement model to treat the recalescence-absent solidification. This simple approximation implies that nucleation ceases when solid fraction reaches 12.5% (or an average grain grows to the half of the final size). In contrast to Quested and Greer's approximation, 4.6 times the radius of the growing grain was used by StJohn et al. [8], which would give a nucleation-ceasing solid fraction of 1.0%. Of course the solid fraction at which nucleation ceases should depend on solidification conditions and a refined treatment of the solute stifling effect is still in demand.

A rigorous treatment of this solute effect on nucleation has been carried out recently by Shu et al., who tried to calculate the thickness of SSN from solidification conditions [6]. Their simulation results showed that the SSN effect has a significant influence on the nucleation behavior and the final grain size for the isothermal melt solidification where recalescence is present. However, they may have overestimated the SSN effect; it is not difficult to find that in their calculation the amount of solute rejected by the solidification was much less than the one received in the bulk melt. In other words the solute conservation law was violated in their model. One of the goals of this paper is to treat the solute effect on the basis of the solute conservation law, and investigate the solute suppression effect induced by grain growth in both isothermal and non-isothermal melts.

In all the existing numerical grain size prediction models [1,4,6,7] a multicomponent aluminum alloy, where more than one alloying component is exerting restriction on growth, was treated as an equivalent binary alloy with the same Q value. Being fully aware of the validity of this binary equivalent treatment requires the alloying components to have identical diffusivities, Quested et al. justified their binary equivalence treatment with the speculation that the solute transportation by convection would cancel out the effect of the variation in diffusivities [10]. It is necessary to examine this speculation before developing a multicomponent model.

As Maxwell and Hellawell pointed out, the convection effect on growth during the initial solidification stage is negligible due to the existence of a stagnant layer adjacent to the growing solid grain whose size exceeds the solute boundary size [7]. A simple indicator for quantifying the dominance of convection is the dimensionless Péclet number, Pe_L, which can be calculated by the following equation:

$$Pe_L = \frac{LV_{rel}}{D} \tag{1}$$

where L is the size of diffusion boundary layer, V_{rel} the relative velocity between the solid grain and the melt and D is diffusivity of an alloying component. Small Pe_L values indicate diffusion dominance while large ones indicate advection dominance. According to Wang and Beckermann [11], V_{rel} tends to vanish because the small nucleus causes such a large interfacial drag that it travels together with its surrounding melt. Taking $1 \times 10^{-5}\ \mathrm{m\ s^{-1}}$ for V_{rel}, 10 μm for L and $5 \times 10^{-9}\ \mathrm{m^2\ s^{-1}}$ for D, one obtains a Péclet number of 0.2. This is a very small number, indicating that convection is not dominant. This estimate is in accordance with the dedicated work on the convection effect carried out by Reddy and Beckermann [12] and Tveito et al. [13]. Solute diffusivities of alloying elements in aluminum melts can differ by about half an order of magnitude [14], and it is necessary to release the binary equivalence assumption and develop a numerical multicomponent model.

In addition to the variation of diffusivities, another motivation for the extension to multicomponent alloys is from a thermodynamic point of view. Summing up Q of different elements in binary systems is not representative for multicomponent alloys, as illustrated in Ref. [10]. The solution to this problem is coupling directly the grain size model with the thermodynamic database developed and validated in the CALPHAD community for multicomponent alloys.

In this paper, we set out to develop a multicomponent grain size prediction model, accounting for both of recalescence stifling and solute segregation stifling. The development is based on the extension of the Kampmann–Wagner numerical (KWN) model, a widely adopted modeling framework for predicting precipitate size distribution and number density [15–18]. The extension of the KWN framework to solidification modeling is straightforward due to many similarities between solid–solid and liquid–solid phase transformation