Available online at www.sciencedirect.com
ScienceDirect
Acta Materialia 59 (2011) 4787–4797
Acta MATERIALIA
www.elsevier.com/locate/actamat

Modeling grain refinement for undercooled single-phase solid-solution alloys

Haifeng Wang [a], Feng Liu [a,*], Yiming Tan [b]

[a] State Key Laboratory of Solidification Processing, Northwestern Polytechnical University, Xi'an, Shaanxi 710072, People's Republic of China
[b] School of Astronautics, Northwestern Polytechnical University, Xi'an, Shaanxi 710072, People's Republic of China

Received 16 August 2010; received in revised form 31 March 2011; accepted 12 April 2011
Available online 10 May 2011

Abstract

A grain-refinement model for rapid solidification of undercooled single-phase solid-solution alloys was developed in which a combination of the model of dendrite fragmentation with the model of overall solidification kinetics allows dendrite fragmentation to occur along with the solidification process (e.g. during mushy zone solidification). Compared with Karma's model, the dendrite break-up time as well as the time for the interdendritic liquid to be solidified completely can be predicted. Thus, the model can predict, in the absence of experimental data (e.g. the plateau duration), the microstructure transition. The model was applied to describe the rapid solidification of undercooled Ni–15 at.% Cu alloy, and a good agreement between the model predictions and the experimental results was obtained. On this basis, the effects of capillarity and back-diffusion on dendrite fragmentation during mushy zone solidification, as well as the effects of back-diffusion and cooling rate on the prediction of the microstructure transition, were found.

Keywords: Rapid solidification; Model; Grain refinement; Dendrite fragmentation; Ni–Cu

1. Introduction

Rapid solidification of bulk undercooled melts is an effective method for preparing metals in metastable states [1], e.g. grain-refined materials. Compared with conventional solidification (magnetic and mechanical stirring, copious nucleation induced by a grain refiner, etc.), such grain refinement occurs spontaneously. To date, a great deal of experimental as well as theoretical work has been undertaken [1–22], though the results have failed to reach a consensus due to controversy over the grain-refinement mechanism. In recent work [23], the evolution of microstructure and microtexture was analyzed for an undercooled Ni–15 at.% Cu alloy, and direct evidence was provided to show that grain refinement at both low and high initial undercoolings should originate from dendrite fragmentation. This agrees with Karma's model [2,3], which not only accounts for such physical origins, but also predicts the critical initial undercoolings for the microstructure transition.

In Karma's model [2,3], the microstructure transition is determined by comparing the dendrite break-up time Δt_{bu} with the plateau duration Δt_{pl}: if $\Delta t_{bu} > \Delta t_{pl}$, a coarse dendritic microstructure forms, whereas, if $\Delta t_{bu} < \Delta t_{pl}$, a refined equiaxed microstructure results. Generally, for a given initial undercooling ΔT, Δt_{bu} can be obtained simply by [2,3]:

$$\Delta t_{bu} \approx \frac{3}{2}\frac{R_{trunk}(\Delta T)^3}{d_0 D^l}\left|\frac{m_l C_0(1-k_e)}{\Delta H_f/C_p}\right| \quad (1)$$

whereas Δt_{pl} is obtained from experimental measurement. Here, $R_{trunk}(\Delta T)$ is the radius of the primary dendrite trunk for a given ΔT, m_l is the slope of the equilibrium liquidus, C_0 is the initial concentration, k_e is the equilibrium partition coefficient, ΔH_f is the latent heat of fusion, C_p is the heat capacity, D^l is the diffusion coefficient of the liquid, and d_0 ($= \Gamma C_p/\Delta H_f$ with Γ the Gibbs–Thompson

* Corresponding author. Tel.: +86 29 88460374.
E-mail address: liufeng@nwpu.edu.cn (F. Liu).

1359-6454/$36.00 © 2011 Acta Materialia Inc. Published by Elsevier Ltd. All rights reserved.
doi:10.1016/j.actamat.2011.04.021

coefficient) is the capillary length. In fact, a number of approximations adopted in Karma's model [2,3] need to be improved as follows:

1. The model limits itself to a single stationary state. Δt_{bu} is obtained from a stability analysis of an idealized cylinder that is embedded in a uniform liquid of composition C_0 and temperature $T_L(C_0)$ (the equilibrium liquidus temperature). However, the composition and temperature in fact vary upon solidification and the maximum temperature of recalescence generally cannot reach $T_L(C_0)$.
2. The interdendritic liquid is believed to be solidified completely at the plateau stage.[1] This is satisfied only if the level rule is fulfilled, e.g. a well-mixed solute in the solid and liquid upon equilibrium solidification. Generally, solidification should be completed at a temperature between the prediction of the level rule and Scheil's equation, e.g. between the equilibrium solidus temperature $T_S(C_0)$ and the melting temperature of pure Cu T_m^{Cu} for Ni–Cu alloy [24,25].
3. The model itself cannot predict Δt_{pl}. In a thorough modeling, the model itself should have the capability to predict the microstructure transition.
4. An exact treatment for the back-diffusion effect is not available. Back-diffusion, as a driving force for dendrite fragmentation, is actually incorporated into the stability analysis. However, only an upper bound for the concentration gradient of the solid at the interface, i.e. $G_c^s = C_0(k_e - 1)/R_{trunk}$, is proposed. That is why Eq. (1) is usually used to predict Δt_{bu}.
5. The effect of a nonlinear liquidus and solidus is not considered. The stability analysis and the calculation of $R_{trunk}(\Delta T)$ are carried out assuming a linear liquidus and solidus. As shown in Refs. [26–31], the effect of a nonlinear liquidus and solidus, particularly for solidification subjected to high ΔT, should not be neglected.

In the present work, a grain-refinement model was developed by combining the model of dendrite fragmentation with the model of overall solidification kinetics; the new model incorporates the effects of back-diffusion and a nonlinear liquidus and solidus. In this way, dendrite fragmentation is allowed to occur along with the solidification process, and the dendrite break-up time and the time required for complete solidification of the interdendritic liquid can be obtained directly (i.e. the model itself can predict the microstructure transition).

This paper is organized as follows. First, the effect of back-diffusion is incorporated into the model of overall solidification kinetics given in Ref. [24] (Section 2.1). Then, Karma's model [2,3] is modified by considering a transient state during mushy zone solidification, in which the effects of back-diffusion and a nonlinear liquidus and solidus are incorporated (Section 2.2). On this basis, a model for grain refinement is established, and the prediction of the microstructure transition, as well as the driving or the frictional forces for dendrite fragmentation, is discussed (Section 3). In Section 4, the model is applied to the rapid solidification of undercooled Ni–15 at.% Cu alloy, in which the effects of capillarity and back-diffusion on dendrite fragmentation during mushy zone solidification, as well as the effects of back-diffusion and cooling rate on the predicted microstructure transition, are discussed in terms of the model calculations. Finally the conclusions are given (Section 5).

2. Model derivation

2.1. Modeling the overall solidification kinetics

Departing from Wang and Beckermann [32,33], an overall solidification kinetic model was developed recently for rapid solidification of undercooled single-phase solid-solution alloys [24]. However, it must be noted no consideration is given to back-diffusion, as this not crucial to obtaining a good agreement between the predicted and the measured cooling curves. In the current work, back-diffusion, as a driving force for dendrite fragmentation [3], is incorporated to obtain the solute gradient of the solid at the interface (see Section 2.2). Because back-diffusion can be easily treated following Ref. [24], only a concise description is given as follows.

2.1.1. Solute and energy balances

A single grain is divided into three different regions or phases: the solid dendrite s, the interdendritic liquid l_i and the extradendritic liquid l_e; see Fig. 1 in Ref. [24]. If the densities and the heat capacities of s, l_i and l_e are equal and constant, the solute and the heat diffusion in the macroscopic scale are omitted, the heat diffusion in s and l_i is not considered, and the solute diffusion in l_i and l_e and the heat diffusion in l_e are under quasi-steady states. Then, following the volume-averaging method (e.g. [34,35]), the solute balances for s, l_i and l_e, and the energy balances for the mushy zone (s plus l_i) and l_e, respectively, can be obtained as:

$$g^s \frac{\partial \langle C^s \rangle^s}{\partial t} = \left(kC_l^* - \langle C^s \rangle^s\right)\left(\frac{\partial g^s}{\partial t} + S^s \frac{D^s}{l_C^{s l_i}}\right) \tag{2}$$

$$g^{l_i} \frac{\partial \langle C^{l_i} \rangle^{l_i}}{\partial t} = \left(\langle C^{l_i} \rangle^{l_i} - kC_l^*\right)\frac{\partial g^s}{\partial t} - \left(C^e - \langle C^{l_i} \rangle^{l_i}\right)\frac{\partial g^{l_e}}{\partial t} - S^s \frac{D^s}{l_C^{s l_i}}\left(kC_l^* - \langle C^s \rangle^s\right) - \left(C^e - \langle C^{l_e} \rangle^{l_e}\right) S^e \frac{D^l \psi^{l_e}}{l_C^{l_e l_i}} \tag{3}$$

$$g^{l_e} \frac{\partial \langle C^{l_e} \rangle^{l_e}}{\partial t} = \left(C^e - \langle C^{l_e} \rangle^{l_e}\right)\left(S^e \frac{D^l \psi^{l_e}}{l_C^{l_e l_i}} + \frac{\partial g^{l_e}}{\partial t}\right) \tag{4}$$

$$\frac{\partial g^s}{\partial t} = \left[\left(1 - g^{l_e}\right)\frac{\partial T_i}{\partial t} + S^e \frac{\alpha^l}{l_T^{l_e l_i}}\left(T_i - \langle T^{l_e} \rangle^{l_e}\right)\right] \Big/ \frac{\Delta H_f}{C_p} \tag{5}$$

[1] The plateau stage is herein defined as the stage between $T_L(C_0)$ and $T_S(C_0)$ in the measured or predicted cooling curve after recalescence.