Diffuse-interface modeling of solute trapping in rapid solidification: Predictions of the hyperbolic phase-field model and parabolic model with finite interface dissipation

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Abstract

Two recently developed phase-field models, a hyperbolic model and a parabolic model with finite interface dissipation, are employed to study the solute trapping in a Si-0.25 at.% As alloy during rapid solidification. The hyperbolic model is applied at the nanometer scale of the interface width δ. The parabolic model is derived by a coarse-graining procedure and is intended to operate with mesoscopic resolution of the interface η. The coarse-graining numerical parameters, namely interface width η and the interface permeability P, are adjusted in the parabolic model to fit the segregation coefficient calculated by the microscopic model on the nanoscale. Based on the optimal sets of η and P selected at small interface velocity, a linear relation between their logarithm values is obtained. This logarithmic relation provides a theoretical basis for choosing the appropriate values of η and P in the numerical phase-field simulation in three spatial dimensions.
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1. Introduction

Considerable theoretical and experimental research has been devoted to the kinetic descriptions of solidification [1]. Most of this research has treated the near-equilibrium or quasi-equilibrium limits of a small growth velocity [2–4]. In these limits, either the local equilibrium condition (i.e. the condition of equal chemical potential) or the condition of equal diffusion potential are employed. By contrast, the understanding of systems far from equilibrium remains less advanced. As a simple but illustrative example of a far-from-equilibrium process, the rapid solidification of a highly undercooled melt has been frequently used for the study of non-equilibrium interface kinetics due to the simplifications resulting from the lack of coherency stress effects, and from the simple interface structure for systems with an atomically rough interface [5]. Rapid solidification occurs in many techniques in contexts such as laser-induced surface melting, spray forming and welding, in which supersaturated solid solutions, metastable compounds and glasses can form [6]. During rapid solidification, the solute may be entrapped by the rapidly moving solid–liquid interface with a quantity in concentrations that differ significantly from those given by the equilibrium phase diagram. This phenomenon is commonly referred to as "solute trapping". Due to its theoretical and technological importance, in addition to experimental investigations, the effect of solute trapping has been also

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extensively studied theoretically using, for instance, analytical models based on a sharp or semi-sharp interface hypothesis [7–13], and phase-field simulations based on diffuse interface models [14–19].

Solute trapping can be characterized by the velocity-dependent solute segregation coefficient $k(V)$, which is defined by the ratio:

$$k(V) = \frac{\text{concentration in solid}}{\text{concentration in liquid}}\bigg|_{\text{interface}} = \frac{c_S}{c_L}\bigg|_{\text{interface}}, \qquad (1)$$

with the interface velocity V. This definition of the solute segregation in Eq. (1) is based on the sharp-interface picture. As for analytical models for describing solute trapping, two important theories are available: one is the continuous growth model (CGM) [9], while the other is the local non-equilibrium model (LNM) [11,12]. The CGM is formulated by assuming a flux balance across a moving solid–liquid interface. The V-dependent solute segregation coefficient $k(V)$ in the CGM takes the form (for dilute alloys) [9]:

$$k(V) = \frac{k_e + V/V_D^I}{1 + V/V_D^I}, \qquad (2)$$

where k_e is the equilibrium partition coefficient given by the equilibrium phase diagram, while V_D^I is the speed of diffusion at the interface defined as $V_D^I = D_I/\lambda$. Here, D_I is the diffusion coefficient at the interface while λ is a characteristic distance, equal to the width of the solid–liquid interface. The quantitative analysis of such a function $k(V)$ in Eq. (2) according to CGM shows a reasonable agreement with the experimental data at small and moderate growth velocities of the solid. However, the experimental results suggest a complete solute trapping regime, i.e. $k(V) = 1$, occurs at a finite interface velocity, which cannot be predicted by CGM. In order to describe the increasing $k(V)$ up to $k(V) = 1$ at a finite interface velocity, both the speed of diffusion at the interface V_D^I and the speed of the atomic diffusion in the bulk V_D^B should be included in the model, as in LNM [11]. The LNM is based on a similar approach as the CGM, but makes use of a generalized Fick's law that accounts for the finite relaxation time of the diffusion flux into its steady state. As a result, the solute segregation coefficient in LNM is described by:

$$k(V) = \frac{\left[1 - (V/V_D^B)^2\right]k_e + V/V_D^I}{1 - (V/V_D^B)^2 + V/V_D^I}, \qquad V < V_D^B,$$

$$k(V) = 1, \qquad V \geqslant V_D^B. \qquad (3)$$

where V_D^B is the propagative speed of the front of solute diffusion profile.

Even though the analytical models may predict the velocity-dependent segregation coefficients, the evolution of the concentration or microstructure can be simulated using the phase-field method. For the past two decades, many attempts have been made to describe non-equilibrium effects in phase transformations [20–22] and, especially, in rapid

solidification [23,24]. In particular, the effects of solute trapping and solute drag have been treated by various phase-field models [14–20,25,26], which fall into two general classes. The first class is based on parabolic governing equations and, therefore, is termed the class of "parabolic phase-field models" [14–17,23,25,26]. The second class is described by hyperbolic-type partial differential equations, and is termed the class of "hyperbolic phase-field models" [18,19,21,24].

The simulated segregation coefficient $k(V)$ in the parabolic phase-field models increases monotonically and gradually as the interface velocity increases, which is consistent with the CGM model [8,9]. However, the complete solute trapping regime, i.e. $k(V) = 1$, cannot be reached by the parabolic phase-field models due to the infinite bulk diffusion speed V_D^B assumed in the model: only the diffusion speed within the interface, V_D^I, is considered in the diffusion equations for the parabolic models. In order to achieve the complete solute trapping regime and the transition to diffusionless solidification (observed in many experiments [6]), the hyperbolic phase-field model [18,19] has been developed by introducing a couple of partial differential equations of hyperbolic type into the original parabolic phase-field models, i.e. the Wheeler–Boettinger–McFadden (WBM) model [14] or the Echebarria–Folch–Karma–Plapp (EFKP) model [27]. By taking both speeds V_D^B and V_D^I into account, the hyperbolic phase-field model [19] predicts the complete solute trapping at the finite interface velocity with the exact value of V_D^B, which agrees well with the experimental data [28] and the atomistic simulations [29]. Considering the lack of experimental data (i.e. segregation coefficient) for most alloys, the simulation results given by the hyperbolic phase-field model can serve as a test as to whether reliable values for speeds V_D^B and V_D^I, and values for other material parameters are being used for the simulation. Note that, so far, the hyperbolic phase-field model can reproduce the complete solute trapping and experimental data with the nanometric width of the diffuse interface.

Very recently, a phase-field model with finite interface dissipation has been developed for the description of non-equilibrium phase transformations [30] on a mesoscopic scale. At this scale, the concentration field, which must be assumed continuous on the atomistic scale, is split into the phase concentrations defined for the individual bulk phases [3]. The solute redistribution at a moving phase boundary is then considered by a local redistribution flux between the phase concentration fields which overlap at the interface. The key feature of this model is that the two concentration fields are linked by a kinetic equation which describes the exchange of the components between the phases, instead of an equilibrium partitioning condition. To adjust the interface dissipation in this exchange, an interface permeability, P, was introduced into the model. For fast exchange (high permeability), the model recovers the phase-field model with equal diffusion potentials in coexisting phases [4,31,32]. In the case of small values of P, the non-equilibrium states of the diffuse interface can be modeled. The model has been applied to simulate the solute trapping in Si-9 at.% As alloy during rapid solid-