

# Phase-field modeling of an abrupt disappearance of solute drag in rapid solidification

Haifeng Wang,<sup>a,\*</sup> P.K. Galenko,<sup>b,c,\*</sup> Xiao Zhang,<sup>a</sup> Wangwang Kuang,<sup>a</sup> Feng Liu<sup>a</sup> and D.M. Herlach<sup>d,e</sup>

<sup>a</sup>State Key Laboratory of Solidification Processing, Northwestern Polytechnical University, Xi'an, Shaanxi 710072, PR China

<sup>b</sup>Friedrich-Schiller-Universität Jena, Physikalisches-Astronomische Fakultät, D-07743 Jena, Germany

<sup>c</sup>Laboratory of Multi-Scale Mathematical Modeling, Ural Federal University, 620002 Ekaterinburg, Russia

<sup>d</sup>Institut für Materialphysik im Weltraum, Deutsches Zentrum für Luft- und Raumfahrt (DLR), 51170 Köln, Germany

<sup>e</sup>Ruhr-Universität Bochum, Fakultät für Physik und Astronomie, 44780 Bochum, Germany

Received 26 January 2015; revised 9 February 2015; accepted 10 February 2015

Available online 16 March 2015

**Abstract**—An effective mobility for non-equilibrium solute diffusion is introduced to develop a hyperbolic phase-field model of rapid solidification in which long-range solute diffusion and short-range solute-redistribution are under local non-equilibrium conditions. At equilibrium, the model provides decoupling of bulk and interface properties. Far from equilibrium, the model predicts a transition from diffusion-limited growth to diffusionless solidification at an interface velocity that is equal to the solute diffusion speed in liquid. At this critical velocity, the solute drag effect disappears abruptly, being consistent with the previous local non-equilibrium model for the sharp interface. A comparison with other phase-field models is made and an agreement between the present model predictions and the experimental results of rapid solidification of Si-9at.%As alloy is obtained. © 2015 Acta Materialia Inc. Published by Elsevier Ltd. All rights reserved.

**Keywords:** Solute drag; Phase-field model; Thermodynamics; Kinetics; Rapid solidification

## 1. Introduction

The interaction between solute atoms and migrating grain boundaries results in a retarding force, i.e. solute drag [1]. Due to its particular importance (e.g. in design and preparation of modern steels [2]), solute drag has been studied extensively by experiments (e.g. Refs. [3,4]) and theories (e.g. the classical solute drag models of Cahn [5] and Hillert–Sundman [6]). It also plays one of the predominant roles in solidification, such as interface kinetics [7,8] and dendritic growth in undercooled melts [9].

For a migrating sharp interface upon rapid solidification, solute drag effect can be either introduced completely or excluded absolutely by the model with or without solute drag [10,11]. As an intermediate case, a partial solute drag model [12] can be proposed to reproduce better the experimental results. By introducing the non-equilibrium solute diffusion effect into the bulk liquid and the interface, the local non-equilibrium models for the sharp interface [13,14] are flexible enough to describe the transition from

the regime with solute drag at low growth velocities to degeneration and abrupt absence of solute drag at high growth velocities. An abrupt transition from diffusion-limited growth to diffusionless solidification is followed by a complete disappearance of solute drag.

By introducing a phase field ( $\phi$ ) that is constant (e.g. 0 or 1) in bulk phases and changes continuously (e.g. from 0 to 1) across the diffuse interface, phase-field modeling avoids the complex front tracking procedure in sharp interface models and becomes quite a powerful simulation tool for the prediction of microstructure formations [15,16]. Assuming that solute diffusion is under local equilibrium conditions for such diffuse interface, the transition from diffusion-limited growth to diffusionless solidification is accompanied by a gradual disappearance of solute drag as the interface velocity increases to infinite [1,17,18].

In order to describe non-equilibrium solute diffusion, a kinetic energy term  $\alpha \bar{J}_B^2/2$  is added to the classical free energy density  $f$  [19]. Here  $\bar{J}_B$  is the overall solute diffusion flux and  $\alpha = (\partial \bar{\mu} / \partial c) v_m^2 / V_D^2$  is a kinetic coefficient independent on  $\bar{J}_B$ ,<sup>1</sup> in which  $\bar{\mu}$  is the overall solute diffusion

\* Corresponding authors at: State Key Laboratory of Solidification Processing, Northwestern Polytechnical University, Xi'an, Shaanxi 710072, PR China. Tel.: +86 29 88460311 (H. Wang). Tel.: +49 3641 947715 (P.K. Galenko); e-mail addresses: [haifengw81@nwpu.edu.cn](mailto:haifengw81@nwpu.edu.cn); [peter.galenko@uni-jena.de](mailto:peter.galenko@uni-jena.de)

<sup>1</sup> It must be pointed out that the kinetic energy term  $\alpha \bar{J}_B^2/2$  is different to the dissipation term of solute diffusion which is also a quadratic function of diffusion flux, e.g. the second term on the right hand side of Eq. (21).

potential,  $c$  is the overall solute molar fraction,<sup>2</sup>  $V_D$  is the solute speed and  $v_m$  the specific atomic volume is assumed be the same for the solvent  $A$  and solute  $B$ . The classical parabolic solute diffusion equation [20] is modified by adding a second derivative of  $c$  with respect to time  $t$  as follows [21],

$$\begin{aligned} \frac{\partial c}{\partial t} + \tau_D \frac{\partial^2 c}{\partial t^2} &= \vec{\nabla} \cdot \left( M_c \vec{\nabla} \frac{\partial f}{\partial c} \right) \\ &= \vec{\nabla} \cdot \left[ M_c \left( \frac{\partial^2 f}{\partial c^2} \vec{\nabla} c + \frac{\partial^2 f}{\partial c \partial \phi} \vec{\nabla} \phi \right) \right] \end{aligned} \quad (1)$$

In the case of one-dimensional steady-state growth, a standard coordinate transformation  $\partial/\partial t \rightarrow -V\partial/\partial z$  with  $V$  as a constant interface velocity reduces the hyperbolic solute diffusion equation Eq. (1) to:

$$-V \frac{\partial c}{\partial z} = \begin{cases} D \left( 1 - \frac{v^2}{V_D^2} \right) \frac{\partial^2 c}{\partial z^2} + \frac{\partial D}{\partial \phi} \frac{\partial \phi}{\partial z} \frac{\partial c}{\partial z} + \frac{\partial}{\partial z} \left[ D \left( \frac{\partial^2 f}{\partial c^2} \right)^{-1} \frac{\partial^2 f}{\partial c \partial \phi} \frac{\partial \phi}{\partial z} \right], & V < V_D \\ \frac{\partial D}{\partial \phi} \frac{\partial \phi}{\partial z} \frac{\partial c}{\partial z} + \frac{\partial}{\partial z} \left[ D \left( \frac{\partial^2 f}{\partial c^2} \right)^{-1} \frac{\partial^2 f}{\partial c \partial \phi} \frac{\partial \phi}{\partial z} \right], & V \geq V_D \end{cases} \quad (2)$$

Here  $\tau_D = D/V_D^2$  is the relaxation time of  $\vec{J}_B$  with  $D$  the solute diffusion coefficient, and  $M_c = D(\partial^2 f/\partial c^2)^{-1}$  is the mobility for  $\vec{J}_B$ . Eq. (2) shows that at  $V < V_D$ , solute diffusion occurs in bulk phases and within the diffuse interface. At  $V \geq V_D$ , the solute profile is homogeneous in bulk phases where  $\partial\phi/\partial z = 0$  and  $\partial c/\partial z = 0$  but is inhomogeneous within the diffuse interface where  $\partial\phi/\partial z \neq 0$  [22,23]. This inhomogeneous solute profile however results in a weak but still existent solute drag effect, being inconsistent with the local non-equilibrium models [13,14] which have already been verified by experiments. In order to show the reason gives rise to this problem, Eq. (1) is reformulated with the conservation law  $\partial c/\partial t = -v_m \vec{\nabla} \cdot \vec{J}_B$  as:

$$\vec{J}_B = -\frac{D}{v_m} \vec{\nabla} c - \tau_D \frac{\partial \vec{J}_B}{\partial t} - \frac{D}{v_m} \left( \frac{\partial^2 f}{\partial c^2} \right)^{-1} \frac{\partial^2 f}{\partial c \partial \phi} \vec{\nabla} \phi \quad (3)$$

where the first and second terms on the right hand side are the long-range solute diffusion flux and, the third term is the flux of short-range solute-redistribution [24,25]. One can see that the non-equilibrium solute diffusion effect is introduced into only long-range solute diffusion (i.e. the second term on the right hand side of Eq. (3)) but not simultaneously long-range solute diffusion and short-range solute redistribution.

In this work, an effective mobility for non-equilibrium solute diffusion is proposed to derive a hyperbolic phase-field model for rapid solidification. Compared with the previous hyperbolic models [22,23] based on the so-called “kinetic energy approach” [19], the present work introduces simultaneously the non-equilibrium solute diffusion effect into long-range solute diffusion and short-range solute redistribution, thus resulting in a sharp transition from inhomogeneous solute profiles at low growth velocities to homogeneous ones in bulk phases and within the diffuse interface at high growth velocities. Noting that solute drag happens in the case of an inhomogeneous solute distribution within the interface but disappears completely in

the case of a homogeneous one, the present model becomes able to predict an abruptly concurrent occurrence of diffusionless solidification and absence of solute drag.

## 2. Effective mobility for non-equilibrium solute diffusion

Let us start from the Maxwell–Cattaneo equation for non-equilibrium solute diffusion [26].

$$\vec{J}_B^i + \tau_D^i \frac{\partial \vec{J}_B^i}{\partial t} = -\frac{D_i}{v_m} \vec{\nabla} c_i \quad (i = S, L), \quad (4)$$

which can be rewritten as

$$\vec{J}_B^i = -\frac{D_i}{v_m} \left( \frac{\partial \tilde{\mu}^i}{\partial c_i} \right)^{-1} \left( \vec{\nabla} \tilde{\mu}^i + \frac{\alpha_i}{v_m} \frac{\partial \vec{J}_B^i}{\partial t} \right) \quad (i = S, L), \quad (5)$$

or

$$\begin{aligned} \vec{J}_B^i &= -\frac{D_i}{v_m} \left( \frac{\partial \tilde{\mu}^i}{\partial c_i} \right)^{-1} \left( 1 + \frac{v_m}{V_D^i{}^2} \frac{1}{\vec{\nabla} c_i} \frac{\partial \vec{J}_B^i}{\partial t} \right) \vec{\nabla} \tilde{\mu}^i \\ &= -\frac{D_i}{v_m} \left( 1 + \frac{v_m}{V_D^i{}^2} \frac{1}{\vec{\nabla} c_i} \frac{\partial \vec{J}_B^i}{\partial t} \right) \vec{\nabla} c_i \quad (i = S, L). \end{aligned} \quad (6)$$

Here  $\tilde{\mu}^i$  is the solute diffusion potential, the superscript or subscript “S” and “L” denote the variable in solid and liquid, respectively. Compared with the classical irreversible thermodynamics, an extra kinetic contribution  $(\partial \vec{J}_B^i/\partial t)$   $\alpha_i/v_m$  is enclosed into Eq. (5). The so-called kinetic energy approach [19] in the case of a prescribed kinetic mobility for solute diffusion changes the thermodynamic state.

On the other hand, the kinetic mobility for solute diffusion may change from its standard definition  $(D_i/v_m)$   $(\partial \tilde{\mu}^i/\partial c_i)^{-1}$  to an effective one  $(D_i/v_m)$   $(\partial \tilde{\mu}^i/\partial c_i)^{-1}$   $\left[ 1 + (v_m/V_D^i{}^2) (1/\vec{\nabla} c_i) (\partial \vec{J}_B^i/\partial t) \right]$  if the thermodynamic driving force  $\vec{\nabla} \tilde{\mu}^i$  is given; see Eq. (6). The kinetic mobility is changed in the case of a prescribed thermodynamic state. If the driving force is taken as the gradient of solute molar fraction  $\vec{\nabla} c_i$ , the non-equilibrium solute diffusion effect changes the diffusion coefficient  $D_i$  to a generalized effective diffusion coefficient for non-steady growth  $D_i [1 + (v_m/V_D^i{}^2) (1/\vec{\nabla} c_i) (\partial \vec{J}_B^i/\partial t)]^3$ .

The “effective mobility approach” provides an alternative choice to describe non-equilibrium solute diffusion. This approach is adopted in the next Section 3 to derive the present hyperbolic phase-field model. For comparison, the previous hyperbolic phase-field model [23] derived from the kinetic energy approach is described concisely in Appendix A.

## 3. The model

### 3.1. Total free energy

Let us consider an isothermal solidification of a binary alloy in a closed system. The overall solute molar fraction  $c$  is:

<sup>2</sup>The definitions of molar fraction, site fraction and concentration are distinguished clearly in recent work of Fischer and Svoboda [20]. Accordingly, the overall solute concentration is  $c/v_m$  (mol m<sup>-3</sup>) and the overall solute molar fraction is  $c$ .

<sup>3</sup>It is straightforward to obtain that the generalized diffusion coefficient  $D_i [1 + (v_m/V_D^i{}^2) (1/\vec{\nabla} c_i) (\partial \vec{J}_B^i/\partial t)]$  reduces to the effective diffusion coefficient [26] in the case of one-dimensional steady-state growth.

Download English Version:

<https://daneshyari.com/en/article/7880387>

Download Persian Version:

<https://daneshyari.com/article/7880387>

[Daneshyari.com](https://daneshyari.com)