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PHASE-FIELD SIMULATION OF SOLIDIFICATION WITH DENSITY CHANGE

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ABSTRACT

Phase-field models of solidification with convection often assume the existence of a single (mixture) velocity at any location inside the diffuse interface, and the phase-field, ϕ , is advected by this mixture velocity. In this paper, the advection of the phase-field is examined for a one-dimensional normal flow to a solidification front induced by a density difference between the solid and liquid. It is found that the results from a phase-field model that assumes a single velocity inside the diffuse interface are generally not in agreement with the sharp interface condition for the kinetic undercooling of the front in the presence of unequal densities, regardless of the interface width. By introducing a two-phase approach, where the solid and liquid are assumed to coexist inside the diffuse interface with different velocities, good agreement with the sharp interface condition is obtained irrespective of the density ratio between the two phases.

NOMENCLATURE

L	latent heat of fusion per unit mass
J	mass flux across the interface
M	interface mobility
S	interfacial area per unit volume
T	temperature
T_m	melting temperature
X_s	existence function
c_p	specific heat
l_a	atomic-scale width of a solid/liquid interface
\mathbf{n}	interface normal vector
r_ρ	density ratio
\mathbf{u}	velocity vector

Greek symbols

β	kinetic coefficient, s/m
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Δ	dimensionless undercooling
δ	diffuse interface width
ϕ	phase-field
Γ	interfacial mass transfer per unit volume due to phase change
μ_k	kinetic coefficient, m/(s·K)
ρ	density
ξ	dimensionless length

Superscripts

–	ensemble or volume average
*	dimensionless variable

Subscripts

i	interface
l	liquid
k	phase
s	solid

INTRODUCTION

Phase-field methods and other diffuse interface approaches have been a popular tool in the direct simulation of solidification [1-3]. By introducing an order parameter, ϕ , to represent the transition between the solid and liquid, a unique set of evolution equations is solved over the entire domain without explicitly tracking the solid/liquid interface. Due to the strong influence of melt motion on the evolution of the microstructure during solidification [4,5], efforts have been made to include convection within phase-field models [3,6-10]. Some phase-field models of solidification with convection treat the solid phase as an extremely viscous fluid and derive the governing equations from basic concepts of irreversible thermodynamics [8,9], or simply make the viscosity a function of the phase-field in the regular Navier-Stokes equations [7]. In

a different approach, Beckermann *et al.* [3] viewed the diffuse interface region as a porous medium where the solid has a zero velocity and the liquid flows through the porous matrix representing the diffuse interface. The diffuse interface version of the governing equations was derived “backwards” from well-established sharp interface equations via a formal ensemble (or volume) averaging method for two-phase flows.

All thermodynamically derived phase-field models assume the existence of a single velocity and pressure at any point inside the diffuse interface between the solid and liquid. Moreover, single thermophysical properties (e.g., density and viscosity) are assumed to exist and their variation across the diffuse interface is postulated in some ad-hoc manner. For large differences in these variables between the phases, thermodynamically derived phase-field models can give results that are very dependent on the choice of the interface width [11] and the way the property variations are specified [10]. The two-phase averaging approach introduced by Beckermann *et al.* [3] offers an alternative by assuming that each phase possesses its own velocity, pressure, and properties, and separate conservation equations are solved for each phase. This avoids the potentially steep variations of the variables across the diffuse interface, and the property variations follow naturally from the derivations. By simply adding the averaged conservation equations for each phase and making use of averaged interfacial balances, a so-called mixture model can be derived from the two-phase model [11]. Assuming furthermore equal velocities of the two phases inside the diffuse interface, a direct connection with thermodynamically derived models can be made [8,9].

The objective of this paper is to examine the advection of the phase-field ϕ in various phase-field models of solidification with flow and to compare the results with those from a corresponding sharp interface model. In the next section, both two-phase and mixture phase-field models are derived using ensemble averaging. The models are then applied to a simple one-dimensional solidification problem where the interface motion is driven by a kinetic undercooling and a normal flow exists that is caused by a density difference between the solid and liquid.

THE MODEL

A detailed derivation of a diffuse interface model for two-phase flows using ensemble averaging of the sharp interface equations is presented in Ref. [11]. In this approach, the phase-field ϕ is defined as the ensemble average of an existence function, X_s , which is unity in the solid and zero otherwise. Here, only the continuity and phase-field equations are presented. The system is assumed to be isothermal and solidification is driven solely by a kinetic undercooling. The material is assumed to be a pure substance with different densities in the solid and liquid states.

Mass conservation

The local mass conservation equation for each phase and the jump condition for a sharp interface are given, respectively, by

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{u}) = 0 \quad \text{and} \quad \|\rho(\mathbf{u} - \mathbf{u}_i) \cdot \mathbf{n}\| = 0 \quad (1)$$

where ρ is the density, which is assumed constant in each phase, \mathbf{u} is the velocity, \mathbf{n} is the local interface normal vector, $\|\cdot\|$ denotes a jump across the interface of a function f as $\|f\| = f_s - f_l$, and \mathbf{u}_i is the velocity of the interface between the solid and liquid. Multiplying the local continuity equation by the existence function, X_k , and averaging yields the following averaged continuity equation for phase k

$$\frac{\partial(\phi_k \rho_k)}{\partial t} + \nabla \cdot (\phi_k \rho_k \bar{\mathbf{u}}_k) = \Gamma_k \quad (2)$$

where the average velocity of phase k is defined as $\bar{\mathbf{u}}_k = \langle X_k \mathbf{u} \rangle / \phi_k$ and the interfacial mass transfer rate per unit volume due to phase-change as $\Gamma_k = \langle [\rho(\mathbf{u} - \mathbf{u}_i)]^k \cdot \nabla X_k \rangle$. Multiplying the jump condition for mass by $|\nabla X_s|$ and averaging, results in the following averaged interfacial mass balance

$$\Gamma_s + \Gamma_l = 0 \quad (3)$$

where the subscripts s and l denote the solid and liquid, respectively.

Defining a mixture density as $\bar{\rho} = \rho_s \phi + \rho_l (1 - \phi)$ and a mass-averaged mixture velocity as $\bar{\mathbf{u}} = [\rho_s \phi \bar{\mathbf{u}}_s + \rho_l (1 - \phi) \bar{\mathbf{u}}_l] / \bar{\rho}$, Eqs. (2) and (3) can be combined to yield the following mixture continuity equation

$$\frac{\partial \bar{\rho}}{\partial t} + \nabla \cdot (\bar{\rho} \bar{\mathbf{u}}) = 0 \quad (4)$$

Equation (4) shows that the mixture is compressible inside the diffuse interface if the densities of the two phases are different. For the mixture approach, where the phase velocities are assumed equal, i.e., $\bar{\mathbf{u}}_s = \bar{\mathbf{u}}_l = \bar{\mathbf{u}}$, Eq. (4) can be rewritten as

$$\nabla \cdot \bar{\mathbf{u}} = \Gamma_s \left(\frac{1}{\rho_s} - \frac{1}{\rho_l} \right) \quad \text{and} \quad \frac{\partial \phi}{\partial t} + \bar{\mathbf{u}} \cdot \nabla \phi = \Gamma_s \left(\frac{1 - \phi}{\rho_s} + \frac{\phi}{\rho_l} \right) \quad \text{for } \bar{\mathbf{u}}_s = \bar{\mathbf{u}}_l = \bar{\mathbf{u}} \quad (5)$$

Phase-field equation

The derivation of the phase-field equation starts with the sharp interface condition for the solid/liquid interface temperature. For the purpose of the present study, solidification is assumed to be controlled solely by atomic attachment kinetics. The effects of pressure, surface tension, and viscous stresses on the interface temperature are neglected.

For a certain choice of the interpolating function for the density variation inside the diffuse interface, Anderson *et al.* [12] derived the following expression for the kinetic undercooling at a solidification front in the limit of a sharp interface

$$T_m - T_i = -\frac{JT_m M}{l_a \rho_s \rho_l L} \quad (6)$$

where T_i is the interface temperature, T_m is the equilibrium melting point, J is the mass flux across the interface defined as $\rho_s(\mathbf{u}_s - \mathbf{u}_i) \cdot \mathbf{n} = \rho_l(\mathbf{u}_l - \mathbf{u}_i) \cdot \mathbf{n} = J$, l_a is the atomic-scale width of a “sharp” solid/liquid interface (which is not necessarily the same as the width of the diffuse interface in the phase-field models below), L is the latent heat of fusion per unit mass, and M is the atomic mobility of the interface. Equation (6) is valid for unequal densities of the solid and liquid as well as in the presence of flow (as long as the pressure and viscous effects are neglected). Also note that it is symmetric with respect to the two phases (*i.e.*, the subscripts s and l could be interchanged). Relating the mobility, M , to the linear kinetic coefficient of the interface, μ_k , via [12]

$$\mu_k = \frac{l_a \rho_l L}{T_m M} \quad (7)$$

Eq. (6) can be rewritten as

$$T_m - T_i = -\frac{J}{\rho_s \mu_k} = \frac{(\mathbf{u}_i - \mathbf{u}_s) \cdot \mathbf{n}}{\mu_k} \quad (8)$$

For $\mathbf{u}_s = 0$, Eq. (8) reduces to the form commonly used in modeling solidification without flow. It is important to note that for a given solid velocity the sharp interface velocity is independent of the density ratio between the solid and liquid, *i.e.*, $r_\rho = \rho_s / \rho_l$.

Equation (8) is now used to derive the phase-field equation. Multiplying both sides of Eq. (8) by $|\nabla X_s|$, making use of the definition for the interface normal vector, $\mathbf{n} = -\nabla X_s / |\nabla X_s|$ [11], and averaging yields

$$\left\langle \frac{(\mathbf{u}_s - \mathbf{u}_i) \cdot \nabla X_s}{\mu_k} \right\rangle = \langle (T_m - T_i) |\nabla X_s| \rangle \quad (9)$$

where the parentheses $\langle \rangle$ denote an ensemble average. The average interfacial temperature is defined as $\bar{T}_i = \langle T_i |\nabla X_s| \rangle / \langle |\nabla X_s| \rangle$, where $\langle |\nabla X_s| \rangle$ is modeled as [11]

$$\langle |\nabla X_s| \rangle = |\nabla \phi| = \frac{\phi(1-\phi)}{\delta} \quad (10)$$

In Eq. (10), δ is a measure of the width of the diffuse interface in the phase-field model. Substituting Eq. (10), making use of the definition for Γ_s , and assuming μ_k to be constant, Eq. (9) becomes

$$\frac{\Gamma_s}{\rho_s \mu_k} = (T_m - \bar{T}_i) \frac{\phi(1-\phi)}{\delta} \quad (11)$$

Substituting Eq. (2) for Γ_s in Eq. (11), yields

$$\frac{\partial \phi}{\partial t} + \nabla \cdot (\phi \bar{\mathbf{u}}_s) = \mu_k (T_m - \bar{T}_i) \frac{\phi(1-\phi)}{\delta} \quad (12)$$

The left-hand side of Eq. (12) describes the evolution of ϕ in a “conserved” form with the solid velocity, $\bar{\mathbf{u}}_s$, inside the divergence operator. Note that no assumption was made with regard to the liquid velocity, and Eq. (12) is valid for arbitrary

$\bar{\mathbf{u}}_l$ as well as for unequal solid and liquid densities. In the case of a rigid and stationary solid, *i.e.*, $\bar{\mathbf{u}}_s = 0$, Eq. (12) reduces to

$$\frac{\partial \phi}{\partial t} = \mu_k (T_m - \bar{T}_i) \frac{\phi(1-\phi)}{\delta} \quad (13)$$

Equation (12), or Eq. (13) for $\bar{\mathbf{u}}_s = 0$, is the phase-field equation for the two-phase approach explained in the Introduction, because the phase velocities inside the diffuse interface are not assumed equal.

The phase-field equation for the mixture approach can be obtained from Eq. (12) by assuming that only a single velocity exists inside the diffuse interface, *i.e.*, $\bar{\mathbf{u}}_s = \bar{\mathbf{u}}_l = \bar{\mathbf{u}}$. Hence,

$$\frac{\partial \phi}{\partial t} + \nabla \cdot (\phi \bar{\mathbf{u}}) = \mu_k (T_m - \bar{T}_i) \frac{\phi(1-\phi)}{\delta} \quad (14)$$

Using Eqs. (5), Eq. (14) can be rewritten as

$$\frac{\partial \phi}{\partial t} + \bar{\mathbf{u}} \cdot \nabla \phi = \frac{\bar{\rho}}{\rho_l} \mu_k (T_m - \bar{T}_i) \frac{\phi(1-\phi)}{\delta} \quad (15)$$

It can be seen that in Eq. (15) the phase-field is advected by a single (mixture) velocity $\bar{\mathbf{u}}$. Equation (15) is equivalent to the thermodynamically derived phase-field equation of Anderson et al. [8,9]. In fact, Eq. (15) can be derived from the phase-field equation in Ref. [8] by choosing appropriate double well and interpolating functions in the free energy functional, and neglecting the effects of pressure, surface tension, and viscous stresses on the interface temperature.

ONE-DIMENSIONAL NORMAL FLOW TO A SOLIDIFICATION FRONT DUE TO A DENSITY DIFFERENCE BETWEEN THE PHASES

Problem description

The two-phase and mixture approaches are compared for the example illustrated in Fig. 1 involving solidification in the presence of a simple one-dimensional flow. The flow normal to the planar solidification front is induced by a density difference between the phases. The interface moves at a constant speed V_i into the positive x -direction. The problem becomes steady if one introduces the moving coordinate $x = x' - V_i t$, where x' is the fixed coordinate normal to the interface. The far-field velocity of the solid ($x \rightarrow -\infty$) is taken to be zero. The system is held isothermally at the interface temperature.

For this example, a *sharp interface analysis* would give a zero velocity in the solid and a constant contraction or expansion flow in the liquid normal to the interface. Hence, with $\mathbf{u}_s = 0$, the sharp interface condition for the interface temperature, Eq. (8), can be written in the following dimensionless form

$$V_i^* = \Delta \quad (16)$$

where Δ is the dimensionless undercooling defined as $\Delta = (T_m - T_i) / (L/c_p)$, and $V_i^* = \beta V_i$ is the dimensionless interface velocity, where $\beta = c_p / (\mu_k L)$. Hence, for a given undercooling, the sharp interface velocity is the same regardless of the density ratio.

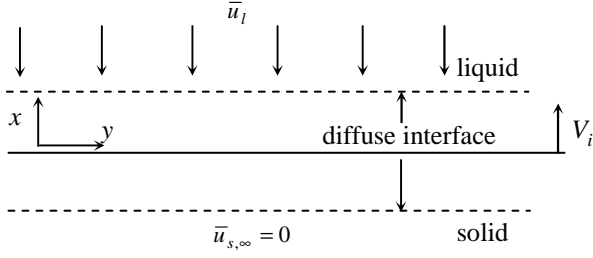


Figure 1. Schematic illustration of the system used in the analysis of normal flow due to phase-change in the presence of a density difference between the phases.

Velocity profiles for a diffuse interface

The velocities for a diffuse interface are solely determined by the mass conservation equations. In the moving coordinate system, the mixture continuity equation, Eq. (4), can be written in a dimensionless form as

$$\frac{d}{d\xi} \left[(r_\rho \phi + 1 - \phi) (\bar{u}^* - V_i^*) \right] = 0 \quad (17)$$

where the dimensionless length is defined as $\xi = x/\delta$ and the dimensionless mixture velocity as $\bar{u}^* = \beta \bar{u}$. Integrating Eq. (17) yields

$$\bar{u}^* = -V_i^* \left(\frac{r_\rho}{r_\rho \phi + 1 - \phi} - 1 \right) \quad (18)$$

Equation (18) provides the velocity profile for the mixture approach. Note that for unequal densities the mixture velocity profile across the interface is not symmetric with respect to $\phi = 0.5$.

The continuity equations for each phase, Eq. (2), become

$$\frac{d}{d\xi} \left[r_\rho \phi (\bar{u}_s^* - V_i^*) \right] = \Gamma_s^* \quad (19)$$

$$\frac{d}{d\xi} \left[(1 - \phi) (\bar{u}_l^* - V_i^*) \right] = \Gamma_l^* \quad (20)$$

where the dimensionless interfacial mass transfer rates are defined as $\Gamma_s^* = \beta \Gamma_s \delta / \rho_l$ and $\Gamma_l^* = \beta \Gamma_l \delta / \rho_l$. Using the definition for Γ_s^* and Eq. (3), Eqs. (19) and (20) can be integrated to yield

$$\bar{u}_s^* = 0 \quad \text{and} \quad \bar{u}_l^* = V_i^* (1 - r_\rho) \quad (21)$$

Hence, for the two-phase approach the individual phase velocities are constant inside the diffuse interface. It can be verified that Eqs. (21) together with the definition of the mixture velocity, \bar{u} , lead to Eq. (18).

Phase-field profiles

The solutions of the phase-field equations for the two-phase ($\bar{u}_s = 0$) and mixture approaches ($\bar{u}_s = \bar{u}_l = \bar{u}$) are discussed separately in the following.

Two-phase model

Equation (13) can be expressed in the moving coordinate system as

$$V_i^* \frac{d\phi}{d\xi} = -\Delta \phi (1 - \phi) \quad (22)$$

Fixing $\phi = 0.5$ at $\xi = 0$, the solution of Eq. (22) is given by Eq. (16) for the interface velocity, i.e., $V_i^* = \Delta$, and the following phase-field profile

$$\phi = \frac{1}{2} \left[1 - \tanh \left(\frac{\xi}{2} \right) \right] \quad (23)$$

Note that the above profile is the same as that encountered in the phase-field model for equilibrium situations. It is emphasized here again that the two-phase model gives an interface velocity that is independent of the density ratio.

Mixture model

Equation (14) can be written in the moving coordinate system as

$$\frac{d}{d\xi} \left[(\bar{u}^* - V_i^*) \phi \right] = \Delta \phi (1 - \phi) \quad (24)$$

Substituting the mixture velocity profile, Eq. (18), into Eq. (24), yields

$$V_i^* \frac{d}{d\xi} \left(\frac{r_\rho}{r_\rho \phi + 1 - \phi} \phi \right) = -\Delta \phi (1 - \phi) \quad (25)$$

which can be rewritten as

$$V_i^* \frac{r_\rho}{(r_\rho \phi + 1 - \phi)^2} \frac{d\phi}{d\xi} = -\Delta \phi (1 - \phi) \quad (26)$$

Fixing $\phi = 0.5$ at $\xi = 0$, and using $\phi = 1$ or $\phi = 0$ far from the interface as a boundary condition, Eq. (26) can be solved numerically to determine the phase-field profile and the interface velocity.

Discussion

Comparing the phase-field equations for this example, i.e., Eqs. (22) for the two-phase model and Eq. (26) for the mixture model, it can be seen that they only differ in the advection term on the left-hand side. In the mixture model, the advection term is modified by a complex factor involving the density ratio, i.e., $r_\rho / (r_\rho \phi + 1 - \phi)^2$, that originates from the variation of the mixture velocity inside the diffuse interface.

It is important to note that the measure of the diffuse interface width, δ , is absent from the dimensionless continuity and phase-field equations for both the two-phase and mixture models. Hence, δ can be chosen as the reference length to scale the problem, and the solution of the dimensionless equations is independent of δ . This independence of the solution on the diffuse interface width is a unique feature of the present problem and can be explained by the fact that the interface velocity is determined solely by the kinetic undercooling and that the system is isothermal. In the presence

of surface tension, however, the solution would generally depend on the interface width [12].

Results

Calculated steady-state phase-field and velocity profiles across the diffuse interface are shown in Figs. 2a and 2b for density ratios of $r_\rho = 2.0$ and $r_\rho = 0.5$, respectively. In each figure, results are given for both the two-phase and mixture models and for an undercooling of $\Delta = 1.0$. The profiles are plotted in the moving coordinate system, such that the interface ($\phi = 0.5$) is fixed at $\xi = 0$. It can be seen from Fig. 2a that for the two-phase model the velocities are constant in each phase and equal to $\bar{u}_s^* = 0$ in the solid and $\bar{u}_l^* = -0.51$ in the liquid, where the negative sign implies that the flow for $r_\rho = 2.0$ is in a direction opposite to the interface velocity (contraction flow). The phase-field profile corresponding to the mixture model is much steeper than the hyperbolic tangent profile corresponding to the two-phase model, which can be explained by the complex form of the advection term in the phase-field equation for the mixture model. For a density ratio larger than unity, such as $r_\rho = 2.0$, the phase-field in the mixture approach is advected towards the solid, which reduces the effective width of the diffuse interface region. For a density ratio less than unity, as shown in Fig. 2b for $r_\rho = 0.5$, the calculated phase-field profile for the mixture model is less steep than the hyperbolic tangent profile corresponding to the two-phase model, since the phase-field is advected away from the solid.

Figure 3 shows a comparison of the variations of the dimensionless interface velocity with undercooling calculated from the two-phase and mixture models for a density ratio of $r_\rho = 0.8$. The two-phase model gives the expected linear dependence of the interface velocity on the undercooling, in agreement with the sharp interface solution given by Eq. (16). The mixture model also gives a quasi-linear variation of the interface velocity, but the interface moves at a speed that is about 20% higher than the sharp interface solution. The results from the mixture model are in agreement with previous observations of Anderson et al. [8] and Conti [9] who solved a similar problem using thermodynamically derived phase-field models. Clearly, if the goal is to simulate a solidification problem that is defined by the present sharp interface condition, the mixture or thermodynamically derived phase-field models will yield incorrect interface velocities. This is especially of concern because the interface velocity is independent of the diffuse interface width.

Figure 4 shows a comparison of the calculated variations of the interface velocity with density ratio between the various models (for $\Delta = 1.0$). Again, the two-phase model agrees with the sharp interface solution and the interface velocity is independent of the density ratio. On the other hand, the interface velocities calculated from the mixture model are a strong function of the density ratio, as expected from Eq. (26). For a density ratio of $r_\rho = 2.0$ the mixture model gives an

interface velocity that is 50.5% lower than the sharp interface value, and for $r_\rho = 0.5$ it is 94.2% higher.

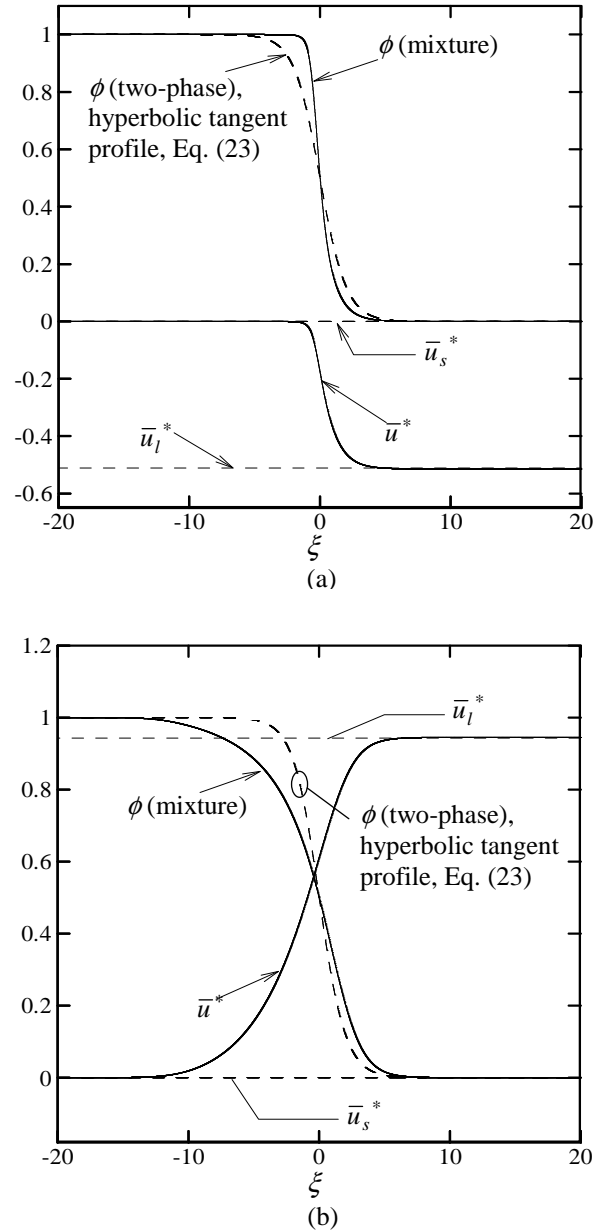


Figure 2. Calculated phase-field and velocity profiles for $\Delta = 1.0$; the dashed lines correspond to the two-phase model and the solid lines to the mixture model: (a) $r_\rho = 2.0$ and (b) $r_\rho = 0.5$.

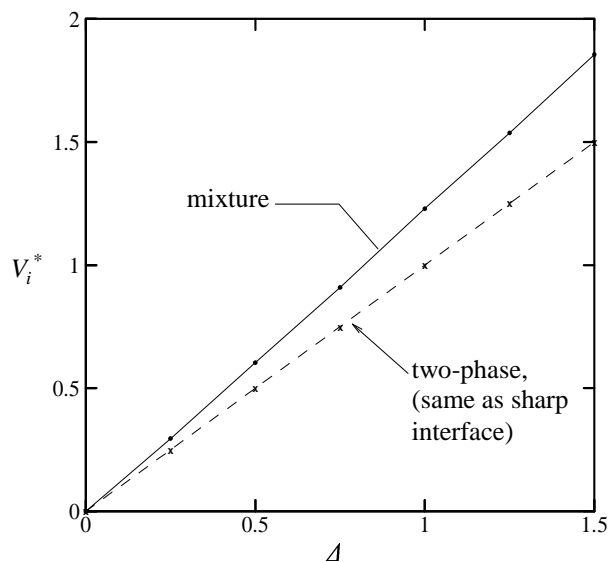


Figure 3. Calculated variations of the interface velocity with undercooling for $r_\rho = 0.8$; the dashed line corresponds to the two-phase model and the solid line to the mixture model.

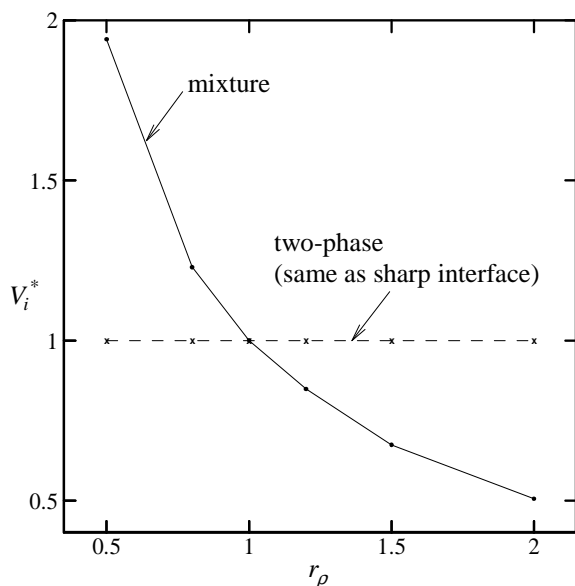


Figure 4. Calculated variations of the interface velocity with density ratio for $\Delta = 1.0$; the dashed line corresponds to the two-phase model and the solid line to the mixture model.

CONCLUSIONS

Phase-field modeling of solidification is examined for the case where there is convection induced by a density difference between the phases. Using an ensemble averaging approach, two different phase-field models are derived from the same set of sharp interface equations: (i) a two-phase model where the velocities of the two phases inside the diffuse interface are not assumed equal and (ii) a mixture model where only a single velocity is assumed to exist inside the diffuse interface. The mixture model is equivalent to thermodynamically derived phase field models previously presented in the literature. The models are applied to a simple one-dimensional solidification problem where solidification is controlled solely by interface attachment kinetics and the flow is driven by a density difference between the phases. It is found that the two-phase model gives results that are in complete agreement with the sharp interface solution of this problem, regardless of the diffuse interface width. On the other hand, the mixture model shows a dependence of the interface velocity on the density ratio that is not in agreement with the sharp interface solution. This can be attributed to the advection of the phase-field by the motion of the mixture inside the diffuse interface. In the two-phase model, the phase-field is not advected because the solid velocity is equal to zero for the present example. In conclusion, it is unclear how a phase-field model that is based on the mixture approach can be used to obtain accurate results for solidification problems involving convection. Future work will include extension of the two-phase model to non-isothermal systems including surface tension.

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