MODELING OF MICROSTRUCTURE EVOLUTION IN THE SOLIDIFICATION OF MULTI-COMPONENT ALLOYS USING LEVEL SET METHODS

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Abstract

A level set method combining features of front tracking methods and fixed domain methods is presented to model microstructure evolution in the solidification of multi-component multiphase alloy systems. Phase boundaries are explicitly tracked by solving the multi-phase level set equations. Diffused interfaces are constructed by extending a small width in both directions from these explicitly tracked phase boundaries. Based on the constructed artificial diffused interfaces, volume-averaging techniques are applied for energy, species and momentum transport. This sacrifice of accuracy by adopting a diffused interface for computational convenience is small considering that the interfaces are still explicitly tracked. By avoiding explicit application of temperature essential boundary conditions on the freezing front, the numerical scheme is energy conserving and the numerical results insensitive to the mesh size. For the numerical analysis of melt flow, a SUPG (streamline-upwind/Petrov-Galerkin), PSPG (pressure stabilizing/Petrov-Galerkin) and DSPG (Darcy stabilizing/Petrov-Galerkin) stabilized velocity-pressure finite element algorithm is adopted. Microstructure evolution in multi-component alloy systems is solved directly using input from phase diagrams. This avoids the difficulty of parameter identification needed in most diffused interface models, and allows easy application to the solidification of various practical alloy systems. Comparable accuracy is observed to front tracking and phase field models in a number of examples available in the literature. Computational techniques including fast marching and narrow band computing are utilized to speed up the level set computations. Adaptive mesh refinement in the rapidly varying interface region makes the method practical for coupling phenomena in meso- and macro-scales during the solidification process.

Introduction

Simulation of solidification and crystal growth processes has been of interest for a number of years not only because of its scientific value in understanding pattern formation in nature but also because of its importance in many technological applications. The main difficulty for this problem lies in tracking the moving boundaries. The sharp freezing interface model is well accepted for modelling microstructure evolution in solidification systems. Front-tracking techniques have been widely used for such sharp front solidification processes [1, 2, 3]. The complexity in implementation of this method restricts current applications to...
solidification systems with only one solid phase. Recent developments of phase field method allow numerical simulation of multi-phase systems. However, without explicitly tracking the interface, the phase field method often requires a very fine grid at a significant computational effort. In this work, a level set method combining features of front tracking methods and fixed domain methods is developed to overcome these difficulties.

Mathematical Model

Let us consider a solidification alloy system with $n$ components (one major component, $n - 1$ minor components) and $N$ phases (one liquid phase, $N - 1$ possible solid phases). Each phase $\alpha$ ($\alpha = 1, 2, ..., N$) at initial temperature $T^\alpha(x, t = 0)$ and at initial concentration $C^\alpha_i(x, t = 0)$ for each component $i$ ($i = 1, 2, ..., n$) is assumed to occupy a region $\Omega^\alpha$ (see Fig. 1). The regions, $\Omega^\alpha$ and $\Omega^\beta$, are separated by the phase $\alpha$-phase $\beta$ interface $\Gamma^\alpha\beta$, which could be $\emptyset$ when the two phases are not neighboring each other. The phase domain $\Omega^\alpha$ is time-dependent and its boundary $\Gamma^\alpha = \bigcup_{\gamma \neq \alpha} \Gamma^{\alpha\gamma}$ is moving with normal velocity $V^\alpha$. The normal $n^\alpha$ is defined as pointing away from the phase region $\Omega^\alpha$. The total domain $\Omega = \bigcup_{\alpha} \Omega^\alpha$ containing all phases and its external boundary $\partial\Omega$ are assumed constant (time-independent). Notice that on the interface of two phases $\Gamma^{\alpha\beta}$, which is both a part of $\Gamma^\alpha$ and a part of $\Gamma^\beta$, we will have $V^\alpha = -V^\beta$ and $n^\alpha = -n^\beta$. In this work, we use $i$ to denote different species with $i = 1$ as the major component and $i = 2, 3, ..., n$ as minor components, $\alpha$, $\beta$ and $\gamma$ to denote different phases with $\alpha, \beta, \gamma \in \{1, 2, ..., N\}$, $s$ to denote different solid phases with $s \in \{2, 3, ..., N\}$ and $\ell$ to denote the unique liquid phase with $\ell = 1$. The signed distance function $\phi^\alpha$ is defined as the minimum distance to $\Gamma^\alpha$ with + sign outside the $\alpha$ phase and − sign inside the $\alpha$ phase.

In the model, we consider constant thermo-physical and transport properties, including densities $\rho^\alpha$, thermal conductivities $k^\alpha$, heat capacities $c^\alpha$, latent heats $L_s^\beta$, partition coefficients $k_{p^\alpha^i}$, diffusion coefficients $D^\ell_i$, and viscosity $\mu^\ell$. Solute diffusion in solid phases and solid-solid phase transformation are neglected $D^\ell_i = 0$ and $V^\alpha = 0$ on $\Gamma^{\alpha\beta}$ $\forall \alpha, \beta \neq \ell$. To avoid directly applying the heat flux jump condition, solute rejection flux condition and no slip condition on the solid/liquid boundaries (as is the case in fixed domain methods) and to simultaneously take advantage of the front-tracking capability of level set methods, we introduce the extended Stefan problem with the following important assumptions [4]:

1. We assume that solidification occurs in a diffused zone of width $2w$ that is symmetric around $\phi^\ell = 0$. The volume averaging technique can be extended to this diffused zone, although it is originally developed for the purpose of macroscopic modeling. In the
volume averaging technique, the phase volume fraction is introduced as follows:

\[ \epsilon^\alpha \equiv \Phi^\alpha / \sum_\beta \Phi^\beta, \quad (1) \]

where the function \( \Phi^\alpha(x,t) \) is defined using the signed distance as follows:

\[ \Phi^\alpha(x,t) \equiv \begin{cases} 0, & \phi^\alpha(x,t) > w, \\ 1, & \phi^\alpha(x,t) < -w, \\ -\phi^\alpha(x,t)/2w + 0.5, & \phi^\alpha(x,t) \in [-w,w]. \end{cases} \quad (2) \]

2. The solid-liquid interface temperature \( T_{s\ell}^I \) is allowed to vary from the equilibrium temperature \( T_s^* \) in such a way that

\[ \frac{dT_{s\ell}^I}{dt} = -k_N(T_{s\ell}^I - T_s^*), \quad (3) \]

where \( k_N \) controls the rate with which \( T_{s\ell}^I \) is designed to approach the desired equilibrium temperature. Moreover, mean temperature in the diffused zone can be approximated as \( T_{s\ell}^I \). Applying energy conservation for the diffusion interface zone, we obtain the following:

\[ V_s = -V_\ell = q^s - q^\ell \rho^s L_s \dot{\epsilon}^s + 2\tilde{c}^s w L_s k_N (T_s^* - T_{s\ell}^I), \quad \text{on } \Gamma^{s\ell}, \quad (4) \]

where \( \tilde{c}^s \equiv 0.5(1 + \rho^\ell c^\ell c^s) c^s \), \( q^s \) and \( q^\ell \) are heat fluxes computed at the boundaries of the diffused interface.

Based on these important assumptions, governing equations for the multi-phase multi-component system can be derived and are shown in the following equations:

\[ \frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{v}) = 0, \quad (5) \]

\[ \frac{\partial (\rho \mathbf{v})}{\partial t} + \nabla \cdot (\rho \mathbf{v} \mathbf{v}) = -\epsilon^\ell \mathbf{v} < \rho^\ell >^\ell + \nabla \cdot \left[ \mu^\ell \left( \nabla \left( \frac{\rho^\ell}{\rho^\ell} \mathbf{v} \right) + \left( \nabla \left( \frac{\rho^\ell}{\rho^\ell} \mathbf{v} \right) \right)^T \right] \right] 
- \epsilon^\ell \mu^\ell \rho^\ell \frac{\mathbf{v}}{\rho^\ell} K(\epsilon^\ell) + \epsilon^\ell \rho^\ell g, \quad (6) \]

\[ (\Sigma_{\alpha=1}^{N} \epsilon^\alpha \rho^\alpha c^\alpha) \frac{\partial T}{\partial t} + c^\ell \nabla \cdot (\rho \mathbf{v} T) = \nabla \cdot (k \nabla T) + \Sigma_{s=1}^{S} \rho^s L^s \dot{\epsilon}^s, \quad (7) \]

\[ \frac{\partial (\rho C_I)}{\partial t} + \nabla \cdot [\rho \mathbf{v} (\epsilon^{\ell} \rho^{\ell} + \Sigma_s \epsilon^s \rho^s k_{pi} C_i)] = \nabla \cdot \left[ \rho^\ell \epsilon^\ell D^\ell \nabla (\epsilon^{\ell} \rho^{\ell} + \Sigma_s \epsilon^s \rho^s k_{pi} C_i) \right]. \quad (8) \]

A number of simplifications is achieved with the above diffused interface model, since applying boundary conditions on the solid-liquid interface for momentum, temperature and species transport is avoided. The sacrifice of accuracy in solving the extended Stefan problem with a diffused interface instead of the classical Stefan problem with a sharp interface is actually small considering that the interfaces are still explicitly tracked by solving the multi-phase level set equations:

\[ \phi^\alpha_t + V^\alpha |\nabla \phi^\alpha| = 0, \quad \alpha = 1, 2, ..., N. \quad (9) \]
Numerical Techniques

There are two types of numerical error in $\phi$, while solving the multi-phase level set equations: (1) $\phi$ does not remain a signed distance, (2) gap or overlap may form between zero level sets for neighboring phases. To address these issues, we use a re-initialization scheme with the following three steps:

1. For each node point $x$, find the two smallest signed distance functions $\phi^\alpha(x), \phi^\beta(x)$ such that $\phi^\alpha(x) \leq \phi^\beta(x) \leq \phi^\gamma(x), \forall \gamma \neq \alpha, \beta$. $\alpha$ will be the phase containing node point $x$ and $\beta$ will be the nearest phase to $x$.

2. Compute $err = \frac{\phi^\alpha(x) + \phi^\beta(x)}{2}$. For all $\gamma$ (including $\alpha, \beta$), update $\phi^\gamma(x)$ as $\phi^\gamma(x) - err$.

3. Use a fast marching technique to re-initialize each signed distance function.

The operation costs for these three steps are only $O(m)$, $O(m)$ and $O(m \log m)$, respectively, where $m$ represents the number of nodes.

A uniform mesh is computationally inefficient since phase boundaries often require finer mesh density. To speed up computation, we implemented the following technique (the 2D case is highlighted here): (1) First, an initial coarse triangle mesh is generated using any suitable mesh generator. (2) If refinement is desired in a region, a triangle element is subdivided into four small triangles with the same size and shape by inserting middle points of the three edges (Fig. 2).

![Figure 2](image-url): Left: Typical sequence of refinement along with the associated data structure. Right: Schematic of generating a conforming grid.

Information about the level of refinement is stored in a tree data structure, which is capable of tracking sons (refined elements) or ancestor (unrefined element) for each element as demonstrated in Fig. 2 (left). In order to make the mesh conforming, two more steps are taken: (1) Elements are further refined so that the refinement level for each two neighboring elements differ by at most 1 as shown in the 1st step of Fig. 2 (right). (2) An element neighboring another element with higher refinement level is subdivided into further connecting elements by connecting its nodes and mid points on edges as shown in the second step of Fig. 2 (right).

Using this adaptive meshing technique, refinement does not change the element shape. Moreover, interpolation of data only needs to be done for newly inserted middle points.

Numerical Examples

Crystal Growth In An Undercooled Binary Alloy

At time zero, we consider an undercooled binary alloy with a small solid seed in the middle of the computational domain. Using same material parameters and initial/boundary condi-
tions as in [5], we obtain comparable segregation pattern and interface position as shown in Fig. 3(b,c).

Figure 3: (a) $\frac{1}{4}$ mesh for solute (b) Solute concentration (c) Interface position.

Planar/cellular/dendritic Transition

Here we present the results of planar/cellular/dendritic transition for Ni-Cu alloy. With $C_0 = 0.4$, only transition from planar to cellular is observed Fig. 4(a). Relation of wavelength and growth velocity is plotted in Fig. 4(b) together with the Mullins-Sekerka loop and comparisons to [6, 7]. By increasing the copper concentration to 0.6, transition to dendritic growth is obtained as shown in Fig. 5.

Figure 4: Left: Planar/cellular transition. Right: Wavelength/growth velocity relation.

Figure 5: Solute concentration and adaptive mesh near tips of dendritic growth ($C_0 = 0.6$).

Eutectic Growth

Here we present some of the results obtained with the prescribed model for an example previously solved using the phase field method in [8]. Consider a binary alloy directional
solidification system with two solid phases $\alpha$ and $\beta$. A temperature gradient $G = 50K/cm$ is maintained at the right side, while a cooling rate of $R = 0.005K/s$ is applied at the left side. Initially, a few $\beta$ seeds are embedded in the $\alpha$ phase. By putting 4 initial $\beta$ seeds, the eutectic growth follows a stable pattern as shown in Fig. 6. If only 2 $\beta$ seeds are inserted in the $\alpha$ phase initially, then the eutectic growth becomes unstable. Valleys with high solute concentration form in the middle of $\alpha - \ell$ phase interface as shown in Fig. 6. This is because, with only 2 $\beta$ seeds, the solute rejected during the transformation of liquid to $\alpha$ cannot be easily absorbed by the $\beta$ phase. The accumulation of solute further slows down the growth rate of $\alpha$ phase and forms valleys. Since the high solute concentration favors the growth of $\beta$ phase, its width increases as shown in Fig. 6.

![Figure 6: Concentration at 80s and evolution of the interface (with 2 and 4 $\beta$ seeds).](image)

Ternary Alloy With Melt Convection Effects

In most studies of crystal growth with convection \[9, 10, 11\], the growth of only one crystal is investigated with a prescribed inlet flow velocity. Fully coupled dendritic growth with heat, solute and momentum transport for an alloy is computationally very difficult due to the various length scales involved in alloy solidification. In order to resolve the dendrite tip or solute boundary layer, a small grid spacing is required. Development of substantial buoyancy driven flow requires larger domain and correspondingly larger grid node/element number. The example in this section is the first study fully coupling dendritic growth with heat, solute and momentum transport for alloy solidification. In this example, we consider a Ni-based alloy with 5.8wt%Al and 15.2 wt% Ta with properties given in \[12\]. Only one solid phase is considered. Initially a cavity of dimensions $0.025m \times 0.025m$ is filled with alloy at its liquidus temperature. A cooling rate of 0.28 K/s is applied at the bottom and sides. The top side is assumed to be adiabatic. Utilizing symmetry, computation is done only in the left half domain. We use adaptive meshing based on $\phi$ with $w = 78\mu m$.

In order to study the effects of convection, we carried two simulation runs: one without convection and one with buoyancy driven flow. With convection, the growth rate is about 20% faster than without convection. The dendrite tip front reaches roughly the same position as shown in Fig. 7 at approximate time of 101 s in the case with convection and 122 s in the case without convection. Without convection, the growth pattern is initially stable when the interface is within 1.0 mm away from the boundary. However, for the case with convection, the interface becomes unstable when the interface is only about 0.5 mm away from the boundary. In the presence of convection, the dendrite tips are more unstable as shown in Fig. 7. In the case without convection, the solute boundary layer developing before dendrite tip hinders phase transformation from liquid to solid. In the presence of fluid flow, the solute accumulated in front of the dendrite tips is taken away by both diffusion and convection. This
results in an increase of growth speed, and a decrease in solute concentration in the solid. Segregation is then more obvious in this case. Without convection, the solute concentration in the solid is slightly less than the initial concentration. Segregation mainly exists between the dendrite arms. A more detailed study of this problem is presented in [13].

Figure 7: Left: Comparison of Al concentration without and with convection. Right: Flow and computational mesh colored with concentration of Ta.

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References


