# A quantitative phase-field model combining with front-tracking method for polycrystalline solidification of alloys

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A B S T R A C T

As the orientation of an individual crystal is constant in solid and diffusive interface layer, a sharp interface model can be coupled to calculate the orientation in phase-field simulation of polycrystalline growth. Here, a two-dimensional phase-field model in combination with the front-tracking method is provided for simulating polycrystalline growth during solidification. In this model, the quantitative phase-field formulations for slow solidification of dilute binary alloys are employed to describe the dynamical evolution of diffusive solid–liquid interface while the front-tracking method is utilized to track the spatial dependent orientation of each grain. Because of the high computing efficiency that is resulted from only one order parameter used to depict the phase transformation during solidification, the model overcomes the disadvantage of reported phase-field approaches for polycrystalline growth. The built model was firstly solved to simulate a free equiaxed dendrite with different orientations growing from undercooled melt for benchmarking. The comparison results indicate that the model is able to compute the orientation exactly. Secondly, the model was extended to simulate the solidification with many equiaxed dendrites. The growth behaviors of the simulated crystals were characterized and analyzed, which demonstrate that the model is feasible to quantitatively and efficiently predict growth dynamics of crystals in a large number and scale during solidification of alloys.

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## 1. Introduction

As polycrystalline materials that are composed of an abundance of randomly oriented crystals in microscopic scale exhibit isotropic properties, polycrystal is the most common grain feature in engineering structural materials. The shape, size and composition distribution of grains determine the mechanical strength, fatigue behavior and some other relevant service properties of materials. Controlling the crystalline growing morphology from melt at the solidification stage is always important to match the required microstructures and composition distribution for engineering alloys. By the aid of computer numerical simulation of crystal growth during solidification, tremendous advancements on the dendritic crystal growth dynamics and the underlying physical mechanism have been achieved up to date. These numerical approaches, including front-tracking [1–3], cellular automaton [4–9] methods based on the sharp interface model and the phase-field method [10–16] based on the diffusive interface model, have been well developed and widely used in simulations on dendritic crystal growth. Due to the neglect of detailed description

of the interface layer between solid and liquid, simulations using the sharp interface methods are usually implemented more efficiently than the diffusive interface approach. Calculating polycrystalline growth with plenty of crystals in a relative large scale of microstructure or even process-scale is thus available in solidification and casting process. However, quantitative description of the interface morphology and moving dynamics is always a tough issue in the simulations of complex dendritic growth for the sharp interface methods. In contrast, it is very convenient for the phase-field model to handle the complicate interfacial physics and geometric topology, which results in the model being particularly prone to capturing the morphological characteristics associated with dendrite growth.

Yet, the simulated number of grains and the scale of dimension are strictly restricted in the diffusive interface scheme owing to the consideration of the finite width of interface. In the phase-field model, an order parameter $\phi$ is introduced to dictate phases, which changes continuously over the thin diffusive interface layer but remains constant within the bulk phases, such as 0 representing liquid and 1 representing solid. The parameter $\phi$ which evolves with time, is governed by a partial differential equation in the entire simulation domain, needless explicitly to distinguish between solid and liquid. Because of this treatment in mathematics

to deal with interface dynamics, the phase-field model has currently emerged as a powerful and quantitative method of choice to simulate dendritic crystal growth during solidification. However, as the width of the interfacial layer is several orders of magnitude smaller than the size of the simulated grain structure or the scale of diffusion field, the computing efficiency of the model is extremely low.

The low computing efficiency of the phase-field model is exaggerated in the situation of coping with polycrystalline growth. To be capable of simulating distribution of either different phases or grains with different orientations, the so-called multi-phase field model has been proposed for solidification and solid-state grain growth [17–23]. Each grain or phase is defined with an order parameter $\phi_i$ to distinct from other grains or phases or both. To model the polycrystalline growth during solidification, it assumes in an individual solid grain $i$, $\phi_i = 1$, and for all other grains at this location the parameter $\phi_j = 0$ $(j \neq i)$. Hence, at any point of the simulation domain with $N$ grains, the spatial distribution of grains or phases can be described by a set of order parameters $(\phi_1, \phi_2, \phi_3, \ldots, \phi_N)$ [21,22,24]. The evolution with time and space of each order parameter is governed by a partial differential equation, which is similar to the model of single phase or crystal growth. The tough problem—low computing efficiency—becomes more stubborn when computing polycrystalline growing process where at least $N$ partial differential equations are solved simultaneously. Consequently, quantitative simulation of large numbers of equiaxed dendritic growth is a great challenge, in particular for solidification crystal growth with various orientations. In order to simulate polycrystalline growth efficiently, another vector-valued phase-field model for crystallization of polycrystalline material has been developed by Kobayashi et al. [11,25,26]. In this model an external variable $\theta$ is used to describe the grain orientation, which allows simulations of solidification, grain growth and grain rotation. Nevertheless, $\theta$ is obtained by solving a time-dependent partial differential equation, which is indeed a step function that has a constant value inside grains and varies sharply at the grain or phase boundaries. Due to such feature of the orientation distribution, the equation specifies a singular diffusivity problem and renders a thorny trouble for numerical calculation. Another difficulty of this model in quantitative simulation of multi-grain growth is the uncertainty of the orientation mobility in the $\theta$ evolution equation.

In the most widely used multi-phase field model [21,22,24], the orientation of a grain is treated as a constant value in the solid crystal and the diffusive interfacial layer, hence it can be described with the sharp interface model. This simplified treatment of orientation is able to lower the calculation cost because the sharp interface model always has a high computing efficiency in tackling polycrystalline growth [27–29]. In this work, the front-tracking procedure is therefore integrated into the quantitative phase-field model [15] to govern the variation of orientation of each grain. A scalar variable $\theta$ whose temporal and spatial evolutions are tracked in advance using the front-tracking algorithm, is introduced to describe the orientations of all grains. The tracking of orientation depends on the local distribution of order parameter, therefore the solid growth dynamics are still controlled by the phase-field formulations. Details of the computational approach used in the tracking of solidification boundaries are available elsewhere [1,3,29–31]. It should be noticed that the orientation contribution to the free energy which represents the excess free energy due to inhomogeneity of crystal orientation in space [32] is neglected because of the simulation just focusing on the solidification stage. This part of free energy is in particular important and necessitated to be considered when two crystals touch each other and start to coarsen by migration of grain boundaries.

In this paper, the proposed model was firstly applied to simulate a single free dendritic crystal with different orientations growing from undercooled melt. The model was solved by the finite element method on an adaptive mesh. The simulated results were compared to the published data to benchmark the accuracy of the suggested model on handling orientation calculation for polycrystalline growth. Secondly, the simulation of a number of crystals with different orientations growing from melt was performed using the built phase-field model. The dynamics involving polycrystalline growth were characterized and then analyzed theoretically.

## 2. Numerical models and implementations

### 2.1. The quantitative phase-field model of polycrystalline growth

For slow solidification of dilute binary alloys, the quantitative phase-field model proposed by Karma [15] is used to describe the motion of solid–liquid interface. The model can be adopted to simulate crystallization with the interfacial width smaller than the thickness of the diffusion boundary layer or the radii of dendrite tips but much larger than its physical width. The solute antitrapping current is included into the model to eliminate the interface stretching and surface diffusion and to guarantee the local chemical equilibrium at the interface. The comparison of the simulated initial transient stage of the directional solidification of Al–Cu alloy with in situ and real-time observation has recently demonstrated that the model could reflect the solidification dynamics quantitatively. To keep consistent with the Karma model [15], in this paper, the order parameter $\psi$ is used to represent the different phases, that is taking $\psi = 1$ and $\psi = -1$ as solid and liquid, respectively. The governing equations are as follows:

$$\tau_0 a^2(\varphi)\frac{\partial \psi}{\partial t} = W_0^2\left\{\boldsymbol{\nabla} \cdot \left[a^2(\varphi)\boldsymbol{\nabla}\psi\right] - \frac{\partial}{\partial x}\left[a(\varphi)a'(\varphi)\frac{\partial\psi}{\partial y}\right]\right.$$
$$\left. + \frac{\partial}{\partial y}\left[a(\varphi)a'(\varphi)\frac{\partial\psi}{\partial x}\right]\right\} + [\psi - \lambda U(1-\psi^2)](1-\psi^2) \quad (1)$$

$$\frac{[(1+k)-(1-k)\psi]}{2}\frac{\partial U}{\partial t} - \frac{1}{2}[1+(1-k)U]\frac{\partial\psi}{\partial t} = \boldsymbol{\nabla} \cdot [Dq(\psi)\boldsymbol{\nabla}U - \mathbf{j}_{\mathrm{at}}] \quad (2)$$

where $x$, $y$ are the horizontal and vertical axes, respectively, $W_0$ the interface width, $\tau_0$ the dimensionless scale of time and $\varphi = \varphi_n - \theta$, $\varphi_n = \arctan(\psi_y/\psi_x)$, is the angle between the normal to the interface pointing to the liquid and $x$ the horizontal direction, $\theta$ is the orientation. $U$ is the dimensionless solute concentration, $U = \frac{2c/c_{l0}-[1+k-(1-k)\psi]}{(1-k)[1+k-(1-k)\psi]}$ with $c_{l0}$ the equilibrium concentration in the liquid at a fixed temperature $T_0$. The parameters $\lambda$, $\xi$ and $q(\psi)$ are set as $\lambda = a_1\xi$, $\xi = \frac{W_0}{d_0}$, $q(\psi) = (1-\psi)/2$. The chemical capillary length is $d_0 = \frac{\gamma T_m}{L_m|m_l|c_{l0}(1-k)}$ with $m_l$ the liquidus slope and $k$ the solute partition coefficient, which are taken as constant values, $\gamma$ the surface tension, $T_m$ the melting point of pure substance and $L_m$ the latent heat. For simplicity, the usual choice of fourfold anisotropy of surface energy is adopted in the phase-field model, $a(\varphi) = 1 + \varepsilon_4\cos4\varphi$, $a'(\varphi) = -4\varepsilon_4\sin4\varphi$. The coefficient $\varepsilon_4$ is the fourfold anisotropy strength of the solid–liquid interfacial energy. As the growth rate of solidification front is not high, the interface kinetic effects are avoided in the model, so that the additional constraint is imposed in the solute conservation equation: $D = a_1 a_2 \xi W_0^2/\tau_0$, $a_1 = 0.8839$ and $a_2 = 0.6267$ as derived by Karma and Rappel [33]. The solute anti-trapping current in Eq. (2) reads [15]

$$\mathbf{j}_{\mathrm{at}} = -\frac{1}{2\sqrt{2}}[1+(1-k)U]\frac{\partial\psi}{\partial t}\frac{\boldsymbol{\nabla}\psi}{|\boldsymbol{\nabla}\psi|} \quad (3)$$