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Microstructure evolution under isothermal and continuous cooling conditions via a combined multiphase field and nucleation approach

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ABSTRACT

We demonstrate the microstructure evolution involving the nucleation and grain growth of a binary alloy during the continuous cooling process via a combined multiphase field model and stochastic nucleation computational model. The nuclei are initiated by adding into the total free energy a term of the nucleation energy related to the variables that represents grains of different orientation. Moelans' interpolation functions are implemented to construct the chemical bulk free energy from coexisting phases/grains for increased stability of multiphase/grain junctions. It was found that the temperature dependent nucleation rate and interface mobility are the main materials properties controlling the features of the resultant microstructures. More even-sized and fine grains can be formed under a high cooling rate given the specified temperature dependent nucleation rate. Solute trapping is the most prominent and solute segregation is the slightest at the highest cooling rate. Equipped with a grain-tracking method, this computational framework provides a viable and computationally efficiently pathway to investigate the large-scale microstructure evolution under various temperature histories that could occur in manufacturing processes.

1. Introduction

Additive manufacturing processes (AMPs) have enabled the development of novel parts due to their increased flexibility in enabling creation of features across multiple scales not possible by traditional manufacturing processes. However, only a handful of metal alloys can be reliably utilized by AMPs among the more than 5000 alloys in use today [1,2]. Pathological microstructures developing during the rapid heating/cooling processes present in AMPs, including large columnar grains and solute segregation, are dominant causes that prevent the wider applicability of AMPs. Since the resulting microstructure has a direct impact on various mechanical properties of manufactured products such as yield and ultimate strengths, fracture toughness and fatigue performance, creating crack-free, fine-grained microstructures is of vital importance. Increasing the nucleation rate by introducing heterogeneous nucleation sites has been used in conventional processing methods such as casting and welding to create microstructure with refined grains. This concept has also been extended to the AMPs. A recent work by Martin et al. [3] introduced nanoparticles of grain refiners to control the solidification microstructure, in order to create a more ideal equiaxed structures that can better accommodate strain and prevent cracking. These lattice-matched nanoparticles are provided in a high density to heterogeneously nucleate the primary equilibrium phases during cooling of the melt pool. This approach can potentially be applied to other advanced manufacturing techniques such as selective laser melting, electron-beam melting to create equiaxed and fine-grained microstructure.

To understand the solidification process during the rapid cooling process that involves nucleation, growth, and coarsening, computational modeling and simulation can play an increasingly crucial role. The development of a computational framework for this reason is also driven by the need to shorten the alloy development and production cycle. The phase-field method has received a tremendous amount of interest in recent years due to its ability to simulate the evolution of realistic microstructures in a thermodynamically consistent manner, and to incorporate the effects of chemical diffusion and convection processes [4,5]. Even though it is largely a phenomenological approach, it has increasingly demonstrated its capability to capture the complex processes and morphologies involved in the microstructure evolution in quantitative manner. Using the phase field approach in a simple two phase (solid-liquid) system, one phase-field variable, which is a function of position and time, is introduced to describe whether solid or liquid exists at a point. The governing equation representing the spatial and temporal evolution of this phase-field variable can be (and often is)

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coupled to equations for heat and solute transport [6,7].

Most of the alloy systems are constituted of multiple phases and/or multiple grains and multiple components. Therefore, the development of the multiphase field approach for multicomponent alloy becomes essential under these conditions. Early phase-field models by Karma [8] and Wheeler et al. [9] for multiphase systems are restricted to a three-phase transformation. The multiphase field model (MPF) by Steinbach et al. [10,11] can be applicable to an arbitrary number of different phases or grains of the same phase yet distinct by their orientation. Similar models were also developed by Fan and Chen [12]. In these models, each phase or grain α distinct from others by its orientation is prescribed by an individual order parameter ϕ_α. A different approach was proposed by Kobayashi and Warren [13] who developed a model that uses the grain orientation as an order parameter to allow for the simulation of solidification, grain growth and rotation.

According to the work of Wheeler et al. [14,15], the so-called Wheeler, Boettinger, and McFadden (WBM) models employ a single continuous composition field c that is defined over the diffusive interface and an equal phase composition $\{c_\alpha\} = c$ is implied [16,17]. Furthermore, Steinbach's approach [10,11] assumes the existence of equal phase diffusion potentials $\{\widetilde{\mu}_\alpha = \partial g_\alpha/\partial c_\alpha = \widetilde{\mu}\}$. It was also shown by Kim et al. [17] -through for a binary alloy system- that Steinbach's approach eliminates the extra chemical potential from the interface in the WBM model. This extra interfacial chemical contribution to the free energy in the WBM model is not only debatable in terms of its physical interpretation, but also problematic in numerical application when the interface width presents a numerical parameter that is usually much larger than its physical value.

Another critical issue resides in the interpolation function used to construct the bulk free energy from the free energies of coexistent phases at a spatial point. The ideal interpolation functions should allow for a thermodynamically consistent interpolation between the free energies of the coexisting phases and keep the local minima of the free energy at their intended positions. However, the interpolation function introduced for two-phase systems, for example, $h(\phi) = \phi^3(10-15\phi + 6\phi^2)$ that is frequently used to interpolate between the bulk free energies of a liquid f_L and solid phase f_S to give $f_b = h(\phi)f_L + (1-h(\phi))f_S$, are not generalizable to multiphase system easily [4,18]. Many models including that of Steinbach et al. uses $h_\alpha(\phi_\alpha) = \phi_\alpha$ to ensure $\sum_\alpha h_\alpha(\phi_\alpha) = 1$ [5], but this function shifts the local minimum of the total free energies and introduces inaccuracies [5]. Moelans [19] introduced an alternate type of interpolation functions to resolve this issue, which satisfies the desirable features including (1) a zero slope at the equilibrium values of the order parameters, (2) $\sum_\alpha h_\alpha(\phi_\alpha) = 1$.

Another issue not frequently discussed is the fact that the phase field methods do not account for the nucleation process naturally. A traditional way to incorporate nucleation is to introduce fluctuations in the field variables via a Gaussian noise term in the evolution function of the field variables [20–22]. Nucleation can occur when these fluctuations are of large enough magnitude to produce a viable nucleus. A common approach to simulate the slow grain growth process while including the nucleation that occurs on a very short time scale is to apply the fluctuation only at the onset of the simulation. A gradual decrease in the fluctuation magnitude with time is enforced so that the nucleation can be neglected as the microstructures further evolve. Simmons et al. [23,24] proposes an alternative Poisson seeding algorithm to introduce viable nuclei in the microstructure at a rate matched to the rate of the formation of nuclei in the real material. The advantage of this approach is that the time scales can be quantitatively incorporated into the nucleation law by adjusting the expectation value of the nucleus formation rate.

In the present paper, it is demonstrated that the microstructural evolution of a binary alloy during the rapid cooling process can be achieved by the multiphase field model and incorporating the stochastic nucleation through the Poisson seeding algorithm by Simmons et al. [23,24]. The employed multiphase field model is based on the work of Kim et al. [17,25] and Steinbach et al. [10]. The nucleation energy of the non-conserved order parameter that represents grains of different orientation is included into the total free energy. The contribution of the chemical bulk free energy to the total free energy is constructed via Moelans' interpolation functions [19]. The microstructure evolution including the nucleation and grain growth process is investigated under different equilibration temperature and various cooling rates. The present work continues with a description of the details associated with the computational model. It follows with the simulation details as well as the description of the relevant material parameters. Results and discussion are presented the next and we close with the conclusions.

2. Computational approach

2.1. The multiphase field model

A system consisting of N_ϕ phases ($n = \alpha, \beta, ..., N_\phi$) and N_C components ($k = 1, 2...N_C$) is considered. For each phase ρ, p_ρ variables are introduced to represent the different crystallographic orientations ($i = 1...p_\rho$). Notations for the phase field variables are similar as those in Noelans' work [19]. The phases and grains are represented as a set of non-conserved order parameters:

$$\boldsymbol{\phi} = (\phi_{\alpha 1}, \phi_{\alpha 2}, ...\phi_{\alpha i}...,\phi_{\alpha p_\alpha}, \phi_{\beta 1}, \phi_{\beta 2}, ...\phi_{\beta i}...,\phi_{\beta p_\beta}, ...,\phi_{\rho 1}, \phi_{\rho 2}, ...\phi_{\rho i}...\phi_{\rho p_\rho}, ...)$$
$$= (\boldsymbol{\phi}_\alpha, \boldsymbol{\phi}_\beta, ...,\boldsymbol{\phi}_\rho, ...) \tag{1}$$

The first symbol in the indices, $\rho = \alpha, \beta, ...$ refers to the different phases and the second symbol $i = 1, ..., p_\rho$ represents the different crystallographic orientations of each phase. Within the grain i of phase ρ, $\phi_{\rho i} = 1$ and $\phi_{\sigma j} = 0$ for $\{\sigma, j\} \neq \{\rho, i\}$. It should be clarified here that the non-conserved order parameters are those which are not required to be conserved over the total volume of the system, i.e., $V_{\phi_{\rho i}} = \int_\Omega \phi_{\rho i} d\Omega$ is not necessarily constant. These non-conserved variables vary smoothly across the grain/phase boundaries.

A conserved concentration field is represented by a set of local molar fractions of N_C-1 independent components since $\sum_{i=1}^{N_c} c_i = 1$:

$$\boldsymbol{c} = (c_1, c_2, ...c_k, ...c_{N_C-1}) \tag{2}$$

The total free energy F of the system is composed of the chemical bulk energy f_b and the interfacial energy f_{int} terms:

$$F = \int_\Omega (f_b + f_{int}) d\Omega \tag{3}$$

The thin-interface approach for alloys as used in Steinbach et al. [10] and Kim et al. [17,25] is adopted in the current study, where the interfaces are treated as a mixture of multiple phases, each having its own phase composition $\boldsymbol{c}_\rho$ and composition-dependent free energy function $f_\rho(\boldsymbol{c}_\rho)$. Thus the bulk free energy f_b is constructed via the interpolation function h_ρ,

$$f_b = \sum_\rho h_\rho f_\rho(\boldsymbol{c}_\rho) \tag{4}$$

where f_ρ is the free energy density of phase ρ and can be calculated as the molar Gibbs free energy divided by the molar volume. The interpolation function h_α for phase α proposed in Moelans' work is in the form [19]

$$h_\alpha = \frac{\sum_i \phi_{\alpha i}^2}{\sum_\alpha \sum_i \phi_{\alpha i}^2} = \frac{|\boldsymbol{\phi}_\alpha|^2}{\sum_\alpha |\boldsymbol{\phi}_\alpha|^2} \tag{5}$$

Here h_α can be understood as phase fraction and satisfies $\sum_\alpha h_\alpha = 1$. It also can be easily verified that h_α has a zero slope at the equilibrium values of non-conserved phase field variables. For example, in a two-phase system, the slope of h_α remains zero at $(\phi_\alpha, \phi_\beta) = (1, 0)$ or $(0, 1)$.