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The evolution of interfacial morphology during coarsening: A comparison between 4D experiments and phase-field simulations

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The evolution of the solid–liquid interface in an Al–Cu dendritic microstructure is predicted using a phase-field model and compared to experimental data. The interfacial velocities are measured during isothermal coarsening using in situ X-ray tomographic microscopy. Good qualitative agreement is found between experimental and simulated velocities. The diffusion coefficient of solute in the liquid is calculated by comparing the magnitude of velocities. The phase-field model is applicable to much larger physical systems than previously tested, increasing its utility for studying coarsening.

Keywords: Three-dimensional tomography; Liquid diffusion; Phase field model; Interface dynamics; Coarsening

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In recent years, the phase-field method has been explored to model the evolution of multi-phase systems [1–9]. It is especially useful for complex microstructures because it avoids the need to explicitly track the boundaries between phases. However, due to the difficulty of observing microstructural evolution in real time and in performing 3D simulations, there have been a limited number of attempts to directly compare phase-field predictions of microstructural evolution with experimental data. Past work has compared the dendrite tip stability parameter σ^* [10], tip velocity during growth in the presence of fluid flow [11], secondary dendrite arm spacing [12], or volume fraction during a phase transformation [13]. Here, we directly compare 3D phase-field predictions of solid–liquid interfacial velocities with the corresponding interfacial velocities calculated from experimental microstructures in an Al–Cu system during isothermal coarsening, captured in situ using X-ray tomographic microscopy. This work thus serves as a validation of the phase-field method and shows how the results of phase-field simulations can be used to determine physical parameters that are difficult to measure independently, in this case the diffusion coefficient of solute in the liquid phase.

The microstructure of a dendritic Al–Cu alloy undergoing coarsening was measured using in situ X-ray tomographic microscopy. Samples of Al–15 wt.% Cu (>99.99% purity, provided by Ames Laboratory) were directionally solidified as in [14]. These samples were then cut into 2 mm diameter specimens. The sample is placed inside a thin-walled boron nitride holder, glued atop an alumina rod, and then mounted on a rotating stage. A custom made X-ray transparent furnace, developed at Risø National Laboratory, is used to coarsen the samples at 836 K, 15 K above the eutectic temperature. The sample is held at constant temperature during data collection.

In situ X-ray tomographic microscopy is conducted on the TOMCAT beamline located at the X02DA port of the Swiss Light Source (SLS) at the Paul Scherrer Institut (Villigen, Switzerland). Further information about the TOMCAT beamline can be found in [15]. X-ray photon energy of 30 keV was used, with an exposure time of 400 ms, and 721 projections were captured over 180° of rotation. A total of 1024 slices, with voxels measuring 1.4 μm per side, were collected every 346 s. Continuous scans were completed for up to 12 h for each sample. The reconstructed, two-dimensional grayscale images were then converted to binary. These images were then combined to create a three-dimensional representation of the

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microstructure as an array of an order parameter ϕ, where $\phi = 0$ is solid and $\phi = 1$ is liquid.

Interfacial velocities in the normal direction, v_n, were calculated from two subsequent experimental microstructures using the level set method [16]:

$$v_n = \frac{1}{|\nabla\phi|}\frac{\partial\phi}{\partial t} \tag{1}$$

The first microstructure was captured after 2.6 h of isothermal coarsening the second 346 s later. The arrays were smoothed using a five-voxel volume average. The velocities were then calculated at each point in the array using Eq. (1), with central finite differences used to approximate spatial derivatives. To determine the velocities on the solid–liquid interface, velocities calculated at each point of the array were linearly interpolated the interface of the first tomographic scan, as determined using a marching cubes algorithm. To check for consistency, velocities were also calculated using experimental data two time steps (692 s) before 2.6 h and three time steps (1038 s) after 2.6 h; velocities were consistent between all three calculations.

The phase-field model is based on the work of Echebarria et al. [8]. It uses a free-energy functional for a dilute binary alloy, and allows for zero solute diffusivity in the solid phase. Spurious effects at solid–liquid interfaces caused by an unrealistically thick solid–liquid interface and the unequal diffusivities are corrected with a phenomenological "anti-trapping current". Although [8] includes a temperature gradient and anisotropic interfacial energy, these features were not implemented, since isothermal coarsening is being simulated, and the degree of anisotropy in the Al–Cu system is insufficient to alter dramatically the shapes of the interfaces, e.g. to induce edges or facets. Physical parameters used in the simulations are shown in Table 1. Central differences were used to approximate spatial derivatives on a regular isotropic grid, and explicit time steps were used. The ratio of grid spacing to interface width was set at $\Delta x/W = 0.4$ as in [8].

To calculate interfacial velocities using the phase-field model, an experimental microstructure, captured after 2.6 h of isothermal coarsening, was resampled onto a finer grid spacing of $\Delta x = 0.7$ μm, and used as the initial conditions for the simulation. Because the composition within each phase is not measured by X-ray tomographic microscopy, the composition field in the system must be chosen before starting the simulation. The order parameter ϕ must vary smoothly across the interface and, consistent with the rapid solute diffusion rate, we assume the concentration field is a quasi-stationary, Laplacian field. Furthermore, any frozen-in concentration gradients remaining from the solidification process are smoothed after 2.6 h of coarsening (more details on the validity of this assumption follow). To equilibrate the fields, ϕ and C were allowed to evolve simultaneously just long enough for ϕ to achieve its equilibrium profile through the interface (during this time, the interface position did not move significantly). Then ϕ was held fixed and C was allowed to evolve until it reached equilibrium. Finally ϕ and C were simultaneously evolved to begin the simulation. Interfacial velocities were calculated using Eq. (1), using the equilibrated order parameter field for the initial microstructure and the evolved order parameter field for the final microstructure.

Table 1. Physical parameters used in phase-field simulations.

Parameter	Value	Reference
D_L (liquid diffusion coefficient)	2.4×10^{-9} m^2/s	[17]
m_L (liquidus slope)	-7.25 K/at.%	[18]
k (partition coefficient)	0.145	[18]
Γ (Gibbs–Thomson coefficient)	2.41×10^{-7} m K	[19]
d_0 (capillary length)	2.27 nm	$\frac{\Gamma}{m_L(C_L^0 - C_S^0)}$ [8]

Experimental results are compared with simulation data in Figure 1. Interfacial velocities are shown for a $104 \times 112 \times 126$ μm subset of the microstructure. This bulb-shaped region (with liquid on the interior of the bulb) was chosen because a smaller subset of the data was needed to make phase-field modeling computationally feasible, and because this region is relatively isolated by the narrow neck of the bulb and the much slower diffusion in the surrounding solid. After the order parameter and composition fields were equilibrated, the system was allowed to evolve for 0.5, 1, 5 s and interfacial velocities were calculated. (Even using 20 processors, the 5 s simulation took over a week to compute, so simulating 346 s between experimental time steps is not currently practical.) The interfacial velocities were approximately the same for all three simulation times. In order to exclude edge effects caused by no-flux boundary conditions, a larger volume than that shown was input to the model, which confirmed that velocities in the vicinity of the bulb were independent of the simulation volume.

Figure 1 shows good qualitative agreement between experimental and simulated velocities. However, as shown by the color bars, the magnitude of the velocities estimated by the phase-field simulations is higher by approximately a factor of three.

To better quantify how much the simulation overestimates the interfacial velocities, the distributions of velocities from experiment and simulation were plotted as normalized histograms. A least-squares fit was used to determine the best-fit scale factor between the normalized histograms (Fig. 2). The simulated interfacial

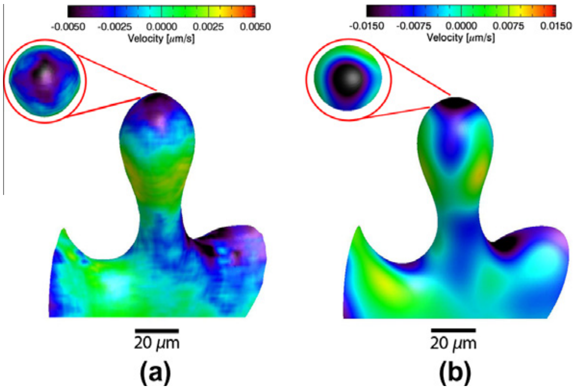

Figure 1. Experimental and simulated velocities. Liquid is inside the bulb shape, and the surrounding solid phase is transparent. The solid–liquid interface is colored by normal velocities, with negative velocities pointing into the liquid. (a) Interfacial velocities calculated from experimental data, 346 s between X-ray scans and (b) interfacial velocities calculated from phase-field simulations, 5 s simulated time elapsed. Note color bars between left and right images differ by a factor of 3.