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Microstructure selection in thin-sample directional solidification of an Al-Cu alloy: *In situ* X-ray imaging and phase-field simulations

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ABSTRACT

We study microstructure selection during directional solidification of a thin metallic sample. We combine *in situ* X-ray radiography of a dilute Al-Cu alloy solidification experiments with three-dimensional phase-field simulations. We explore a range of temperature gradient G and growth velocity V and build a microstructure selection map for this alloy. We investigate the selection of the primary dendritic spacing Λ and tip radius ρ. While ρ shows a good agreement between experimental measurements and dendrite growth theory, with $\rho \sim V^{-1/2}$, Λ is observed to increase with V ($\partial\Lambda/\partial V > 0$), in apparent disagreement with classical scaling laws for primary dendritic spacing, which predict that $\partial\Lambda/\partial V < 0$. We show through simulations that this trend inversion for $\Lambda(V)$ is due to liquid convection in our experiments, despite the thin sample configuration. We use a classical diffusion boundary-layer approximation to semi-quantitatively incorporate the effect of liquid convection into phase-field simulations. This approximation is implemented by assuming complete solute mixing outside a purely diffusive zone of constant thickness that surrounds the solid-liquid interface. This simple method enables us to quantitatively match experimental measurements of the planar morphological instability threshold and primary spacings over an order of magnitude in V. We explain the observed inversion of $\partial\Lambda/\partial V$ by a combination of slow transient dynamics of microstructural homogenization and the influence of the sample thickness.

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1. Introduction

Microstructural characteristics, such as dendritic spacing, determine the properties, performance, and lifetime of metal cast parts. For most industrially relevant metallic alloys, solidification is the first processing step accompanied by microstructure formation. Solidification is also a prime example of nonlinear pattern formation [1,2], where complexity stems from the broad range of length and time scales involved [3,4]. Even though our understanding of crystal growth and solidification as a whole has evolved significantly over the past decades [5,6], microstructure selection mechanisms are still not completely understood.

Many experimental studies of solidification patterns have made use of transparent organic compounds that solidify in a similar fashion as metallic alloys [7]. Such *in situ* solidification observations have led to significant insights into dendrite growth in undercooled melts [8,9] as well as the formation of cellular and dendritic arrays during directional solidification of binary alloys [10–18].

Synchrotron X-ray facilities around the world now provide intense and coherent X-rays, opening the door to similar *in situ* observations of metals and metallic alloys during solidification, with ever increasing spatial and temporal resolutions [19–21]. Aluminum alloys have proven to be particularly good candidates for *in situ* X-ray observations and have been extensively used in metal solidification experiments (e.g. Refs. [19,21–26]).

Both in experiments and simulations, the concept of microstructure selection maps as a function of temperature gradient G

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and growth velocity V, as illustrated schematically in Fig. 1 (see e.g. Ref. [27]), has existed for decades [28]. In a diffusive regime, the critical velocity V_c for the onset of planar morphological instability [29] may be estimated by the constitutional undercooling criterion [30] (see Sec. 4.2). The transition from cellular to dendritic growth occurs over a diffuse range around a transition velocity V_{CD} that may be estimated by the limit of stability of cells as $V_c(1+k)/k$ [31], i.e. close to V_c/k when the solute partition coefficient at the interface k is small [13].

In terms of dendritic tip shape, it is now commonly accepted that for given growth conditions, e.g. a given solute concentration c_∞, a given temperature gradient G, and a given growth velocity V, the selection of dendritic tip radius is unique, and follows microscopic solvability theory [1,32,33]. In contrast, cellular growth leads to a cell tip radius essentially proportional to the primary cellular spacing (see e.g. Ref. [34]).

In contrast to dendrite tip radius selection that occurs over a narrow distribution range, similar conditions can lead to primary cellular or dendritic spacings within a wide range, which has been shown both experimentally and theoretically [11–17,27,35,36]. However, a similar processing history usually results in similar spacings [17,37]. Microstructures may also exhibit oscillatory states close to spacing stability limits during both cellular [38–41] and eutectic growth [39,42,43].

The mechanisms associated with the lower and upper spacing instability are well identified (see e.g. Refs. [11,18,27,35,44,45]), namely cell/dendrite elimination below the lower limit Λ_{min}, and cell tip-splitting or dendritic tertiary branching above the upper limit Λ_{max}. In early modeling studies, Hunt and Lu suggested that the upper spacing stability limit Λ_{max} had to be at least twice the lower limit Λ_{min}, so that the spacing resulting from tip splitting or branching (hence half of the initial spacing) would remain higher than Λ_{min} [35]. However, they pointed out that it was unclear what could be used at the time to model the maximum dendrite spacing, and also suggested that in practice the average spacing was likely to be closer to the lower spacing limit than the upper spacing limit [35]. Since dendritic branching could not be treated with their model, they assumed the maximum dendrite spacing limit to be twice the lower stability limit [27]. This assumption has led to good agreement with experimentally measured distributions [27,35]. It has since then been commonly assumed that $\Lambda_{max}/\Lambda_{min} \approx 2$ (see e.g. Refs. [44,46]) and later theoretical studies have focused mostly on the lower limit of spacing stability. Yet, recent simulations using quantitative phase-field [34,45] and multiscale dendritic needle network modeling [47,48] have shown that the upper spacing stability limit Λ_{max} may actually be larger than Λ_{min} by a factor of 3 to 4 in a dendritic regime. However, most of the dynamically selected spacings occupy the lower half of the stability range, hence remaining between Λ_{min} and about $2\Lambda_{min}$.

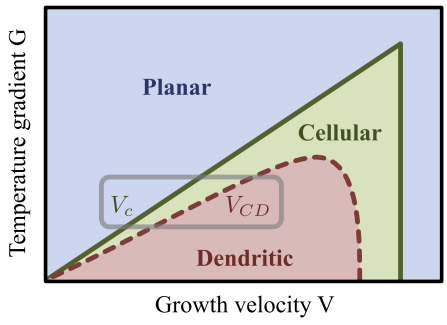

Fig. 1. Schematic microstructure selection map. The gray box shows the typical range of parameters for the Al-1.4 wt%Cu alloy in our experiments (see Fig. 11 in Ref. [27]).

In the immediate vicinity of the critical velocity V_c corresponding to the onset of morphological instability, the average selected primary spacing $\Lambda(V)$ typically shows a sharp decrease with increasing V, followed by an increase of Λ with V in the cellular regime, up to the cell-to-dendrite transition, which occurs over a finite range of velocity around $V \approx V_{CD}$, and finally a decrease of Λ with V in the dendritic regime [49,50] (for such observations in Al-Cu alloys, see Refs. [51,52]). Thus, the $\Lambda(V)$ curve typically exhibits a maximum in the vicinity of the cell-to-dendrite transition at $V \approx V_{CD}$. In the dendritic regime the average primary spacing then decreases as $\Lambda \sim V^{\alpha}$, typically with $\alpha \approx -0.25$ [13,53].

Additionally, the aforementioned studies have focused mostly on purely diffusive transport conditions. Yet, significant convection may take place in liquid alloys, due to both thermal and solutal gradients, combined with the effect of gravity [16,51,54–58]. Convection has been shown to play an important role in microstructure selection in aluminum-copper (Al-Cu) alloys [51,52,59–61], even in thin sample experiments in which confinement is intended to limit the extent of convection [62–64].

In terms of microstructure selection, liquid convective transport is understood to strongly affect heat and mass transport but to have a negligible influence on the dendrite tip operating state [65–71]. On the other hand, gravity-induced liquid convection is known to have a significant impact on dendritic spacing selection. However, most theoretical studies of directional solidification with convection to date have been performed on relatively limited sample sizes and in two dimensions (2D) [61,72].

In the current study, we combine synchrotron X-ray *in situ* observations of a dilute Al-Cu alloy solidification with quantitative phase-field (PF) simulations in 3D to explore microstructure selection mechanisms in thin sample directional solidification. We explore a range of temperature gradients and growth velocities to build a morphological selection map analogous to Fig. 1 and systematically measure cellular and dendritic tip radii and primary spacings. We compare these measurements quantitatively with existing analytical theories and PF simulations that incorporate approximately the effect of convection using a diffusion boundary-layer approximation. Despite its simplicity, this approximation has the advantage that it allows us to simulate the formation of spatially extended cellular/dendritic arrays on experimentally relevant length and time scales. Furthermore, it reproduces well salient features of the experiments including a strong effect of convection on the limit of planar stability and on the spacing-velocity relationship.

2. Experiments

2.1. Sample preparation

We prepared an Al-0.6 at.%Cu alloy (i.e. 1.4 wt%Cu), using high purity (99.999%) Al and Cu by arc-melting. The buttons were flipped and remelted at least five times to ensure homogeneity. Thin samples were rolled to a thickness (z) of about 200 μm. Individual samples with ($x \times y$) dimensions of about 30 × 10 mm^2 were sheared for X-ray imaging experiments.

2.2. Directional solidification

Samples were directionally solidified at a constant velocity within an imposed temperature gradient with a standard *power down* method, directly controlling the temperature gradient and the cooling rate.

Each sample was placed in a boron nitride crucible with a 2 mm diameter observation window that is thinned to 0.1 mm thickness (see additional pictures and schematics in the Supplementary