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Influence of growth velocity variations on the pattern formation during the directional solidification of ternary eutectic Al-Ag-Cu

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ABSTRACT

In order to control the evolving microstructure in complex eutectics and other multi-phase systems, it is important to understand the adjustment mechanisms and the parameters that determine pattern evolution. Here, a combined experimental and simulation approach is taken to investigate the response of a three-phase eutectic system to changes in solidification velocity. Using Al-Ag-Cu as a model system, large scale three-dimensional phase-field simulations are compared to directionally solidified samples containing both, velocity increases and decreases. The experimental results are obtained by synchrotron tomography for detailed consideration of the microstructure directly before and after a targeted velocity change and by traditional SEM analysis of sample cross sections to capture effects over longer length scales. In addition to qualitative analysis of the images, the microstructures are statistically assessed using phase fractions, shape factor and nearest neighbor statistics. Both, simulation and experiment show an immediate change in phase fraction as a result of a change in growth velocity. Adjustment of the microstructural pattern occurs more slowly over a relatively long length scale, due to splitting, merging and overgrowing events. Novel quantification techniques emphasize that ternary eutectic phase arrangements are complex and continuously evolving structures which, even under ideal conditions, do not reach steady state growth as quickly as previously believed.

1. Introduction

It is well known that the scale of eutectic microstructures is dependent on solidification velocity. The relationship was first described by Jackson and Hunt, who observed that the lamellar or rod spacing is equal to the square root of velocity multiplied by some material-specific parameter [1]. These geometrically simple structures are convenient to treat, because a single characteristic length can be used to define the system, controlling both, solid-liquid interfacial curvature and diffusion length. Subsequent works showed the Jackson-Hunt scaling behavior to be consistent for more complex and faceted structures as well [2–4], including various arrangements of three-phase eutectic patterns [5]. In order to accommodate changes in length scale due to velocity adjustments, it is necessary for growing phases to split or overgrow at the solid-liquid interface. This has been extensively studied for binary eutectic growth using both, metallic and transparent organic systems [4,6–8], in which these adjustment mechanisms are generally well understood. However, patterns in higher order eutectics can be far more complex, requiring different adjustment behaviors to respond to velocity changes. Likewise, a variety of patterns have been identified in three-phase eutectics, sometimes with different patterns occurring under the same experimental conditions [9–14]. The way in which such three-phase eutectic systems respond to imposed changes in growth velocity has not previously been studied. Understanding the way in which pattern adjustment occurs is an important step to identify the fundamental parameters that control morphology development in multi-phase, multi-component systems.

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In order to gain a more complete understanding of this complex behavior, a combination of experimental and simulation work was carried out. Recent advances in computer science now allow for large-scale, three-dimensional simulations to be carried out that reasonably approximate the behavior of the physical system [15–19]. Using Al-Ag-Cu as a model system, full 3D phase-field simulations of directionally solidified ternary eutectic growth are carried out. The results are compared to experimental results of the same alloy system. In order to capture the specific spacing adjustment mechanisms in three dimensions, X-ray synchrotron tomography is performed on small regions of the experimental samples where the velocity change have been imposed. Because tomography results are necessarily limited in scale, additional experimental work was done to measure the behavior of the system over longer length scales using individual cross-sectional images taken across the length of the sample.

The paper is structured in the following way: first, details of the experiments and simulations are provided. Then the results obtained by experiment and simulation are discussed and compared. Qualitative analysis is done via comparison of cross-sectional images of the microstructures; quantitative analysis is done via statistical analysis of phase fractions, shape factor and number of nearest neighbors. Finally, the implications and conclusions of these findings are discussed.

2. Setup

Experiments with Bridgman furnaces and simulations with a phase-field model based on the Grand potential approach are conducted to study the influence of growth velocity changes during the directional solidification of the ternary eutectic system Al-Ag-Cu. First, the experimental setup and the procedures to obtain the microstructures are described. Subsequently, the applied phase-field model and the simulation setup is presented.

2.1. Experimental setup

Experimental microstructures are obtained by gradient-stage directional solidification of Al-Ag-Cu alloys at the invariant eutectic composition of 69.1 at-% Al, 18.1 at-% Ag and 12.8 at-% Cu, producing coupled growth of the solid solution with the three solid phases Al, Ag_2Al and Al_2Cu. In this study, two sets of experimental data are studied.

The first set of experimental data, in the following labeled as EXP⇑, is obtained by using a Bridgman-type furnace. The specimen is initially fabricated by casting an alloy, containing elements with a purity of 99.999 *wt%* with the ternary eutectic composition. The melt is cast in a steel mold coated with boron nitride. The initial sample length was 260 *mm* with a rod diameter of 5 *mm*. For directional solidification, this cylindrical specimen is placed into an alumina crucible with inner diameter of 5.5 *mm* and held for 1 h in a stationary gradient of approximately 9 K/mm to create a stable solid-liquid interface. The gradient is then translated past the sample with a rate of 0.7 $\mu m/s$ for a distance of 85 *mm*. The translation velocity is increased to 4.0 $\mu m/s$ at a rate of 1.5 $\mu m/s^2$ and translation continued for an additional height of 50 *mm*. This is close to a stepwise velocity change, with the acceleration taking only 2.2 *s* and corresponding to a solidification distance of less than 5.2 μm. The remainder of the sample is quenched at a rate of 20 mm/s by translating the samples into a water-cooled liquid-metal bath of Ga-In-Sn. After processing, the sample is cut, polished and examined in the transverse plane at different solidification distances, before as well as after the velocity change. For this sample, the microstructure parallel to the solidification front is evaluated at the growth heights 80 *mm*, 85 *mm*, 90 *mm*, 95 *mm*, 115 *mm* and 125 *mm*. Scanning electron microscopy (SEM) using electron-backscatter imaging is carried out to obtain maximum phase contrast. Additional details about the experimental procedure can be found in Ref. [20].

The second set of experimental samples, labeled as EXP⇓, are produced using a somewhat different experimental procedure. Rods of the eutectic alloy with a mold diameter of 10 *mm* and length of 100 *mm* are also cast in boron nitride-coated steel mold and subsequently machined to 8 *mm*. For the cast alloy, metals with a purity of 99.9 *wt%* for Ag and Cu and 99.99 *wt%* for Al are used. Directional solidification is carried out using the ARTEMIS facility [21]. This furnace contains an upper and a lower heater controlled by separate PID controllers with a chilled water heat sink at the bottom. Independent control of each heater allows for translation of the solid-liquid interface without physical movement of the sample or the furnace. The sample is contained inside a transparent aerogel crucible allowing the progress of the solidification front to be determined directly via optical measurement. In the first part, the solidification velocity is measured as 0.32 $\mu m/s$ with a gradient of 2.8 K/mm. After approximately 51 *mm*, the solidification velocity is decreased to 0.08 $\mu m/s$, which resulted a slight decrease in the gradient to 2.2 K/mm. Transverse cross-sections are prepared after the solidification experiment from the ARTEMIS facility and subsequently, SEM images are used to identify different regions of interest, i.e. different grains and morphologies.

For the tomography, ultrasonic drilling is applied on EXP⇓ samples to extract cylindrical samples from these regions at the same solidification height, each 0.5 *mm* in diameter and 8.5 *mm* long, that included material both, before and after the velocity change. The tomography experiments with the extracted needle-shaped samples are performed at the ESRF imaging beamline ID19 and are described in more detail in Ref. [14]. For each sample four overlapping image stacks are obtained with a voxel edge length of 0.325 μm. Smoothing, sharpening and labeling of these single volumes are performed with AVIZO Fire 8.0 software. Images with an interval of about 97.5 μm were evaluated in detail after a manual correction of labeling errors.

2.2. Simulative setups

For the simulations, a thermodynamic consistent multi-component multi-phase-field model based on the Grand potential approach [19,22] of Allen-Cahn type is applied. Beside the four phase-field evolution equations, three coupled chemical potential evolution equations, derived from the diffusion equation of the concentrations, are solved numerically on a discrete regular grid. The equations are spatially discretized using finite differences and, for the temporal evolution, an explicit Euler scheme is applied [23]. Due to the thermal diffusion rate being many times faster than the concentration diffusion in the liquid, a frozen temperature approach [15,24,25] is employed. Therefore an analytic temperature field to control the growth velocity and the slope of the gradient is defined. The multiple times larger concentration diffusion in the liquid than in the solid phases, allows the neglection of the solid diffusion and hence a moving window technique is utilized [26,27]. In the ternary eutectic system Al-Ag-Cu, the solubility of Ag in the Al phase decreases in the order of nine percentage points over 20 *K* below the ternary eutectic temperature T_{eu}, which leads to different phase-fractions of the solid phases at room temperature compared to the thermodynamically predicted phase-fractions at T_{eu} [9,28,29]. To avoid the simulation of this computationally intensive solid diffusion process, the thermodynamic Gibbs energies are adjusted in order to reproduce the phase-fractions from experimental micrographs at room temperature. The simulation parameters are presented as dataset **EXP** in