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Coarsening evolution of dendritic sidearms: From synchrotron experiments to quantitative modeling

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ABSTRACT

The local dynamics of dendritic sidearms during coarsening are studied by combining *in-situ* radiography observations with numerical and analytical models. A flat sample of a Ga-In alloy is partially solidified and then held isothermally in a vertical temperature gradient. The evolving dendritic microstructure is visualized using synchrotron X-ray imaging at the BM20 (ROBL) beamline at ESRF, France. During the coarsening stage, the temporal evolution of the geometrical features of sidebranches is captured by automated image processing. This data is then used to quantify the dynamics of two basic evolution mechanisms for sidebranches: retraction and pinch-off. The universal dynamics of sidearm necks during pinch-off are exploited to determine the product of liquid diffusivity and capillarity length Dd_0, as a parameter that is crucial in the calibration of quantitative models. By employing an idealized phase-field model for the evolution of a single sidebranch, the behavior of selected sidebranches is reproduced from the experiments in a consistent way.

1. Introduction

The solidified microstructure of metal alloys ensues largely from the growth and coarsening of dendrites. During their initial growth into an undercooled melt, dendrites form a characteristic tree-like structure of primary stems and higher-order branches. At a later stage, when the surrounding melt approaches equilibrium, the dendritic structures undergo a slow coarsening process that continuously reduces the number of sidebranches and leads to an increase in the average microstructural length scale. This process is primarily governed by capillarity effects, which cause diffusive material exchange between adjacent structures of different curvature. The coarsening of dendritic structures is characterized by transformation of the side-arm morphology present after growth. It typically proceeds by three mechanisms: (i) retraction of small sidebranches towards their parent stem, (ii) pinch-off or detachment of sidebranches at the narrow neck with the parent stem, and (iii) coalescence of neighboring sidebranches.

The pinch-off of dendrite branches is of particular interest as the resulting dendrite fragments can initiate the growth of equiaxed grains and therefore promote a fine, isotropic microstructure. The underlying detachment mechanism is a capillarity-driven shape instability that leads to a gradual constriction and collapse of the sidearm neck near the junction with the parent stem.

Over decades, the observation of microstructure formation has been limited to post-mortem analysis of sectioned samples or microscopic imaging in transparent alloys; however, new X-ray sources and innovative procedures for image analysis have dramatically advanced the *in-situ* analysis of evolving microstructures in metal alloys starting around the year 2000 [1,2]. Initial studies involved the radiographic observation of thin samples (2D projections). More recently, microscopic tomography is used to obtain time-resolved, volumetric data [3–6]. Samples are often placed within a temperature gradient, which dictates the direction of growth during cooling. The orientation of the growth with respect to gravity has a strong effect on the strength of buoyancy-

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driven convection [7]. The resulting solidification conditions affect the dendrite morphology and the tendency to fragment [7–10].

Flat-sample radiographic solidification studies have addressed the effects of composition, cooling rate, gradient magnitude and orientation, and natural or forced convection. Additional information on strains and crystallographic misorientations have been obtained by means of white-beam X-ray topography [11]. Although radiography-based methods allow a relatively large area of the sample to be captured, the limited sample thickness restricts the growth of the dendrites and the flow of the melt compared to bulk sample conditions [12].

X-ray micro-tomography is able to capture volumetric information on the microstructure in small bulk samples. The restrictions in terms of sample size and time resolution are, however, more severe than in radiography. Therefore, tomography has mainly been useful for observing slow processes in small samples, e.g. during dendrite coarsening on a sub-mm scale [3–6]. Nonetheless, the availability of three-dimensional images has enabled the quantification of local and global features of the morphology and their evolution over time.

For metallic alloys, Aagesen et al. [13] investigated the isothermal pinch-off of rod-like solid structures surrounded by liquid melt. Such configurations are prone to a Rayleigh-Plateau like shape instability, where a small local reduction in the cross-section of the structure becomes amplified. This mechanism is caused by the increasing curvature difference between nearby interface regions, which induces diffusional transport through the bulk liquid. Shortly before the structure breaks up, the local curvature of the neck approaches infinity. Due to the strong localization of the neck dynamics and geometry, this process takes on a universal, self-similar behavior. During this stage the geometry of the neck approaches a double cone with an opening angle of 80° and the neck diameter a_N follows

$$a_N(t) = 1.76\left[Dd_0(t_p - t)\right]^{1/3} \tag{1}$$

where D is the solute diffusivity in the liquid phase, d_0 the chemical capillary length and t_p the time when the pinch-off occurs. These theoretical predictions were confirmed in Ref. [13] by in-situ X-ray tomography of Al-Cu samples during isothermal coarsening.

Recently, Neumann-Heyme et al. [14] performed a computational study of the effects of the initial geometry and cooling rate on the dynamics of sidebranch evolution. The study revealed that sidearms can only pinch off within a limited range of model parameters. Beyond this interval, coalescence or retraction will occur before the sidebranches can detach from the parent stem. The authors confirmed that the pinch-off follows the universal dynamics described in Ref. [13] during a short time interval just before pinch-off. These dynamics were found to be independent of model parameters such as the cooling rate.

In the present work, a radiographic analysis of the growth and coarsening of dendrites in a low-melting-point Ga-In alloy is presented using the ROBL beamline (BM20) at the European Synchrotron Radiation Facility (ESRF, Grenoble). The high spatial and temporal resolution achieved in the experiments enables important local geometric features to be accurately captured during the coarsening stage, and their dynamics to be evaluated quantitatively. It is then demonstrated that the measured dynamics can be well reproduced by means of a numerical simulation model of an axisymmetric sidearm, as first developed in Ref. [14].

A prerequisite for the implementation of the simulation model is the knowledge of the relevant material parameters, since they select the length and time scale of the interface evolution process. On one hand, the use of a Ga-In alloy in solidification experiments enables simple handling due to its low melting point, but on the other hand, the material properties required for quantitative modeling are highly uncertain. In particular, the product of the diffusion coefficient and capillary length is difficult to measure by conventional means, but is of utmost importance in solidification models. The present study exploits the universality of the pinch-off behavior in the form of Eq. (1) as a tool for the direct determination of Dd_0 based on in-situ observations of the neck dynamics coupled with a simple theoretical analysis. This approach is not restricted to Ga-In, but is also applicable to other alloy systems that have not been thoroughly characterized.

2. Experimental methods and model description

2.1. Experimental setup

The visualization experiments were performed at the ROBL beamline (BM20) at ESRF (Grenoble). The experimental setup, cf. Fig. 1a, used here for the solidification experiments was already employed in previous radiographic investigations carried out by means of a microfocus X-ray tube [15,16].

All experiments were conducted using a low-melting-point hypereutectic Ga–25 wt%In alloy that was prepared from gallium and indium of 99.99% purity. The low melting point of the alloy (liquidus temperature 25.7 °C) enables the experiments to be implemented efficiently and flexibly. Furthermore, the Ga–In alloy exhibits a high X-ray contrast between the growing indium dendrites and the interdendritic Ga-rich liquid. A compilation of the material properties is provided in Section 2.4.

The alloy was melted and filled into a Hele-Shaw cell made of Plexiglas with a liquid metal volume of $28 \times 28 \times 0.15\ \mathrm{mm}^3$. The rectangular observation window determined by the width of the X-ray beam was $20 \times 23\ \mathrm{mm}^2$ in size. The Hele-Shaw cell was cooled at the bottom by means of a Peltier cooler, while a second array of Peltier elements was mounted as a heater on the upper part of the solidification cell. The distance between the heater and the cooler was 19 mm. The simultaneous regulation of the power of both Peltier elements by means of a PID controller unit allowed the cooling rate and the temperature gradient to be adjusted flexibly during the process. Three miniaturized K-type thermocouples (<0.1 mm) were attached to the lateral surface of the cell to monitor the temperature. The accuracy of the temperature control is ± 0.2 K. In the present experiments, a cooling rate of 0.01 K/s and a temperature gradient of ~ 1 K/mm were applied. The temperature gradient was calculated from the temperature difference measured between the thermocouples T_1 and T_2.

The solidification cell was exposed to a monochromatic X-ray beam with an energy of 28.5 keV. Conventional transmission radiographs were obtained by means of a scintillator that provides a resolution of 2 μm and was coupled to an optical magnifier and a PCO 2000 CCD camera with 2048×2048 pixels (pixel size of $0.34 \times 0.34\ \mu\mathrm{m}^2$). This equipment leads to a field of view of about $700 \times 700\ \mu\mathrm{m}$. The distance between the detector and sample was 20 cm. In order to change the location of the observation window, the position of the solidification cell was manipulated with respect to the X-ray beam by a motorized positioning system with a minimum translation step of 10 μm. Images were acquired at exposure times ranging from 2 to 20 seconds.

Since mainly X-ray scattering experiments have so far been performed on the ROBL beamline, the initially available hardware was not optimized for imaging. Thus, some initial difficulties with the illumination conditions of the sample had to be improved in order to find a good balance between illumination homogeneity, low image noise level, and reasonably low exposure times. Imperfections of the beam line optics caused by the waviness of the