THE EVOLUTION OF DENDRITE MORPHOLOGY DURING ISOTHERMAL COARSENING

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Abstract

Dendrite coarsening is a common phenomenon in casting processes. From the time dendrites are formed until the inter-dendritic liquid is completely solidified dendrites are changing shape driven by variations in interfacial curvature along the dendrite and resulting in a reduction of total interfacial area. During this process the typical lengthscale of the dendrite can change by orders of magnitude and the final microstructure is in large part determined by the coarsening parameters. Dendrite coarsening is thus crucial in setting the materials parameters of ingots and of great commercial interest.

This coarsening process is being studied in the Pb-Sn system with Sn-dendrites undergoing isothermal coarsening in a Pb-Sn liquid. Results are presented for samples of approximately 60% dendritic phase, which have been coarsened for different lengths of times. Presented are three-dimensional microstructures obtained by serial-sectioning and an analysis of these microstructures with regard to interface orientation and interfacial curvatures. These graphs reflect the evolution of not only the microstructure itself, but also of the underlying driving forces of the coarsening process. As a visualization of the link between the microstructure and the driving forces a three-dimensional microstructure with the interfaces colored according to the local interfacial mean curvature is shown.

Introduction

When a metal alloy is cast, solidification begins with the formation of a thin layer of solid metal at the surface of the mold. As the solidification front progresses into the ingot, however, the solidification front becomes unstable and forms dendrites throughout the remaining liquid. This partially solidified region is termed a mushy zone. The bulk of the ingot remains in the solid-liquid state for a substantial amount of time while heat is extracted and the interdendritic liquid solidifies. During this time the dendrites undergo a coarsening process that determines their ultimate morphology and length scale. The dendrite morphology and lengthscale in turn is crucial in setting the mechanical properties of the final ingot.

Research on dendrite coarsening was, until recently, restricted to examining either individual dendrites in transparent systems, see e.g. [1], or planar sections through a dendritic structure in opaque systems. Thus, the analysis of the evolution of the microstructure in opaque systems is usually reduced to determining the secondary dendrite arm spacing, λ_2, as a function of coarsening time, see e.g. [2,3]. Such experiments show that $\lambda_2 \sim t_f^{1/3}$, where t_f is the time over which the solid and liquid coexisted. It is well known that dendrite coarsening is driven by the variation in mean interfacial curvature, H, along the interface between the solid (dendritic) and liquid (matrix) phase. This variation in curvature with position results in variations in the concentration of solute in the liquid along the interface as described by the Gibbs-Thomson equation,

$$x_A = x^0_A + \ell^\alpha_A H,$$

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where x_A and x^0_A are the concentration of A atoms at a curved and flat interface, respectively, ℓ^α_A is the capillary length in the α-phase and H is the mean curvature.

The variation in concentration with position along the dendrite interface in turn leads to diffusional fluxes between areas of high interface mean curvature, where solid dissolves, and areas of low interface mean curvature, where the solid grows. The overall effect is that the structure becomes coarser.

Several models have been proposed [4,5,6] to explain certain phenomenological aspects of the coarsening process. In general these models employ an oversimplified dendritic morphology and are limited to considering the evolution of localized areas of the dendrites. To formulate more realistic models, information on the complete topology of the dendritic structure and its evolution is required. This is because the solute flux, as discussed below, extends to large length scales. An overview on the coarsening of mushy zones and a discussion of the influence of the local interfacial shape on the coarsening process can be found in [10].

It is now possible to analyze the three-dimensional microstructure of ingots due to the development of a fully automated technique for serial-sectioning of metallic microstructures, which is capable of making 20-30 sections per hour, combined with a method to reconstruct three-dimensional microstructures using these sections [7]. This approach not only characterizes the morphology of the microstructure, but also allows the quantity that drives the coarsening process to be measured: the interfacial curvature as a function of position. For the first time microstructural evolution in dendritic mushes can be linked to its driving forces. The results can thus serve as a test of three-dimensional calculations of dendrite coarsening as well as provide much needed insight into the morphological evolution of topologically complex dendrites during coarsening.

Experimental Procedure

Ingots of Pb-80wt.%Sn have been directionally solidified in a Bridgman-like furnace to give a uniform dendritic microstructure containing ca 60% of Sn-dendrites and 40% of Pb-Sn eutectic phase. Cylindrical samples (6mm length, 12mm diameter) were cut from the rods using a wire cutter in order to preserve the microstructure. The samples were then coarsened at 185°C (2K above the eutectic temperature) for various lengths of time and subsequently quenched and serial-sectioned with a sectioning distance of 4.75μm. The microstructure was then reconstructed in a computer to give three-dimensional images and to conduct curvature analysis of the microstructure.

Analysis

Figure 1 shows the reconstructed volume for two samples that were held for 3 and 210 minutes at 185°C. Clearly, the dendritic microstructure coarsens over time. Figure 2 shows the inverse surface area per unit volume, a shape independent lengthscale measure, as a function of coarsening time.

Despite the large scatter of the data, a linear fit seems to agree reasonably well, which suggests that $\lambda_2 \sim t_f^{1/3}$. The interfacial curvature of the dendritic microstructure, shown in figure 1, was then determined. A full characterization of the curvature at each point of the interface requires either the mean and the Gaussian curvature, H and K, or equivalently the two principal curvatures, κ_1 and κ_2, to be determined. Using mean and Gaussian curvature to characterize the structure is useful since the mean curvature, through the Gibbs-Thomson equation, sets the interfacial composition, and hence the dynamics of the coarsening process. Furthermore, the evolution of mean curvature is a function of the Gaussian curvature and vice versa, see [8]. Characterizing the local interface using the principal curvatures is, however, more intuitive. Figure 3

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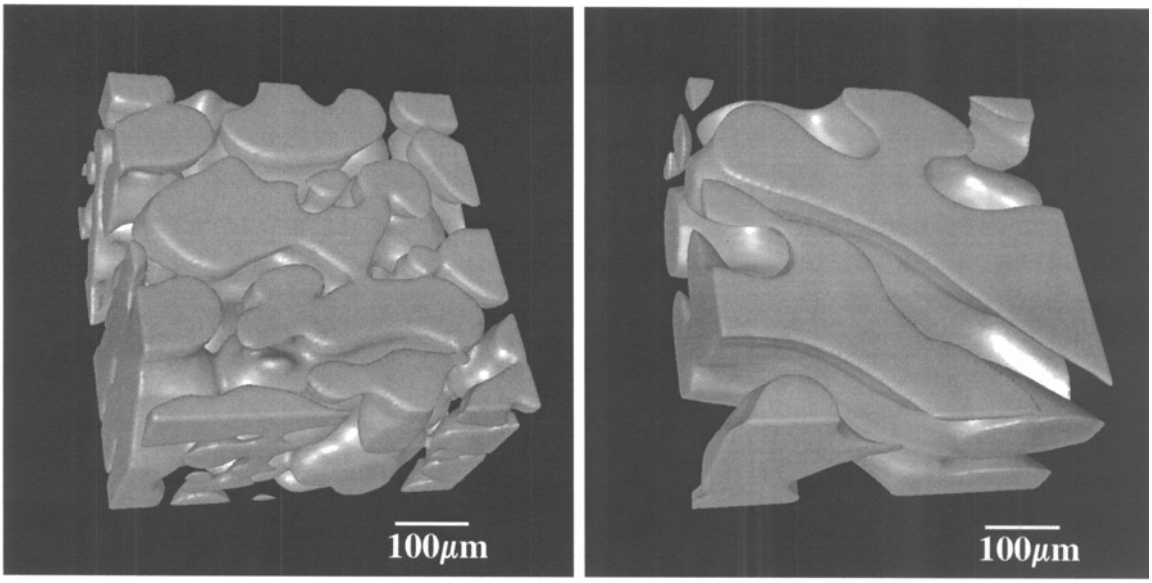

Figure 1. Reconstruction of the microstructure of Pb-80wt.%Sn samples coarsened for (a) 3 minutes and (b) 210 minutes at 185°C. The volume fraction of the dendritic phase, shown as solid, is approximately 60%. The matrix phase is not shown.

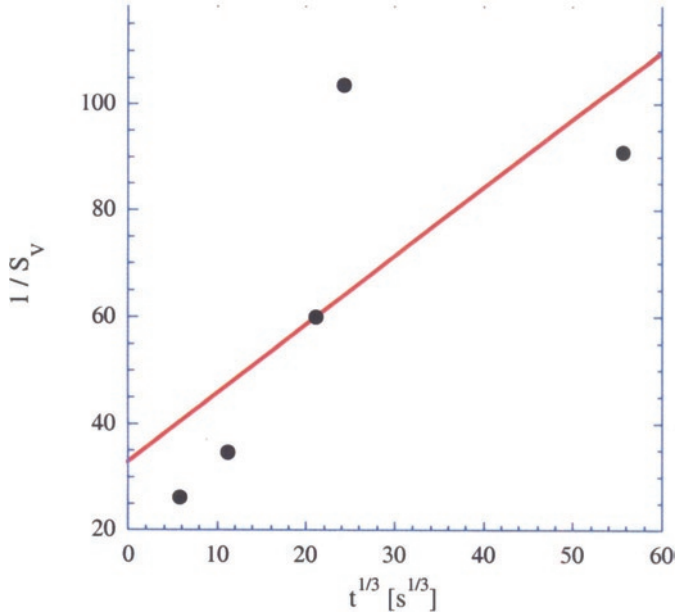

Figure 2. $1 / S_v$ as a function of the cube root of the coarsening time.

shows the solid-liquid interfaces of the 3 minute sample colored based on the mean curvature (here a larger section of the microstructure is shown). Although the details of the microstructure are almost to small to be distinguished, one can clearly see that the sample contains region of mostly negative mean curvature (bluish colors) and regions of mostly positive mean curvature (green/red colors). At this early stage the microstructure is clearly divided into these two kinds of regions. With ongoing coarsening, however, this separation diminishes.

Figure 5 (a) and (b) show the mean and Gaussian curvature as a probability density distribution for the 3 minutes and 2900 minutes coarsened samples. A map of the different regions and the according shapes can

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be found in figure 4. In both cases we find a maximum at near spherical shapes and a distribution which declines from there. The total range of the distribution shrinks as the total lengthscale extends as one would expect from general coarsening theory. The coarsening is, however, not self similar. Using a similar representation in κ_1/κ_2- space this becomes more obvious (see figure 7, figure 6 for a map).

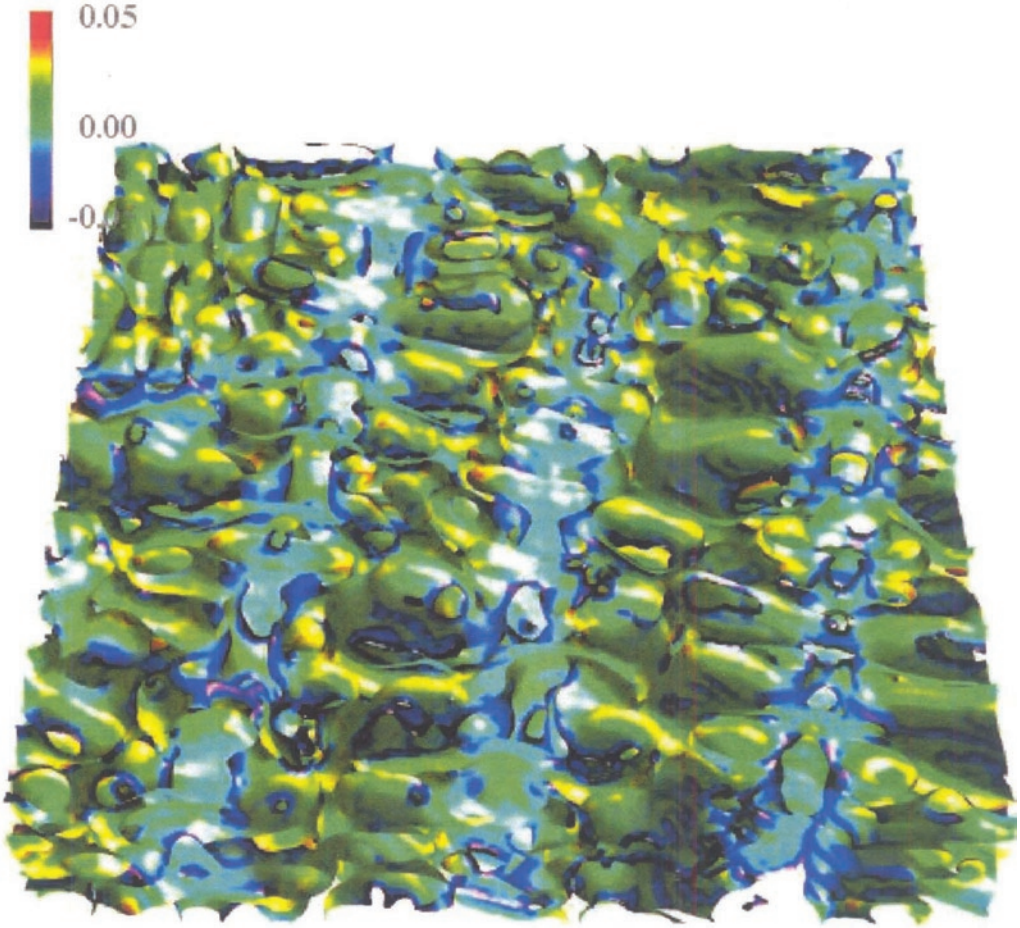

Figure 3. The interface of the 3 minute coarsened sample are shown colored based on the local mean curvature. The legend is given in the upper left corner.

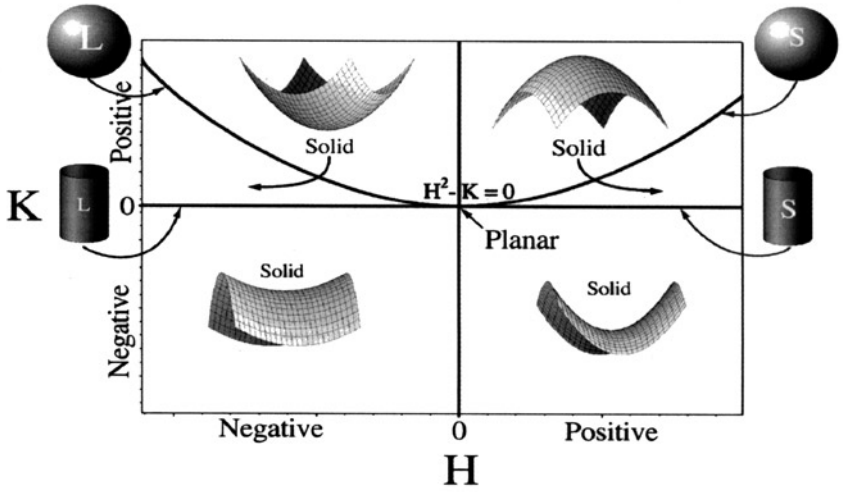

Figure 4. Map of the local interface shapes in the *H-K* space.

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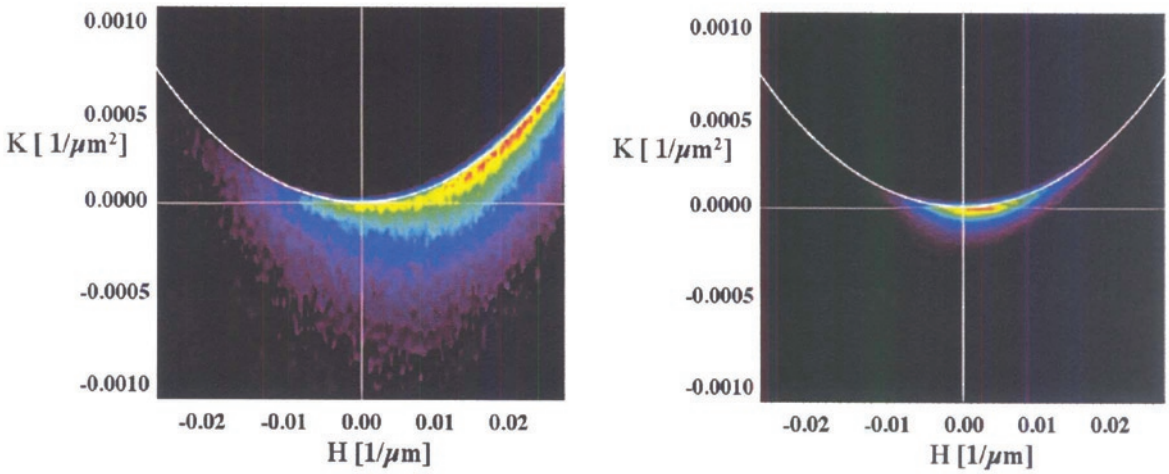

Figure 5: (a) and (b) show the probability density distributions in *H-K*- coordinates for the microstructures of the 3 minutes and 2900 minutes coarsened samples, respectively. The probability density function *P(H,K)* is defined so that *P(H,K)dHdK* is the probability that a randomly chosen interface point has a mean curvature between *H* and *H+dH* and a Gaussian curvature between *K* and K+dK. The colors follow the rainbow with white/red as the highest probability density and black the lowest.

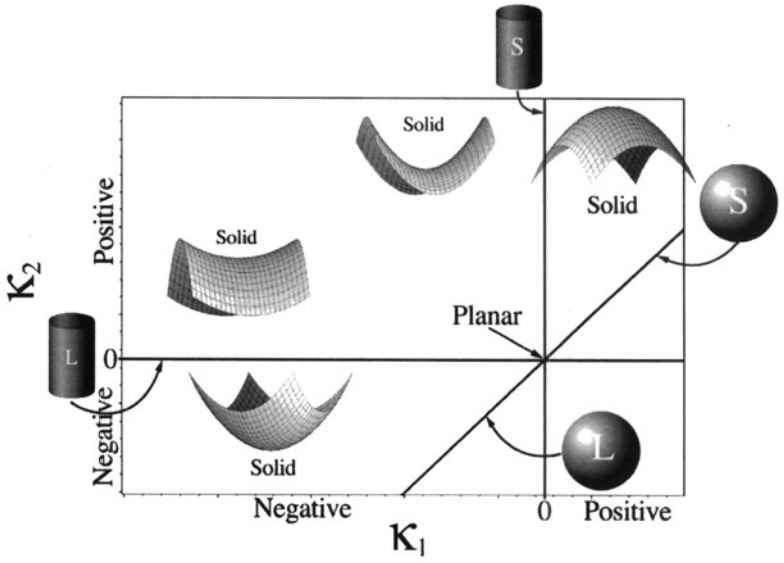

Figure 6: Map of the local interface shapes in the κ_1/κ_2-space.

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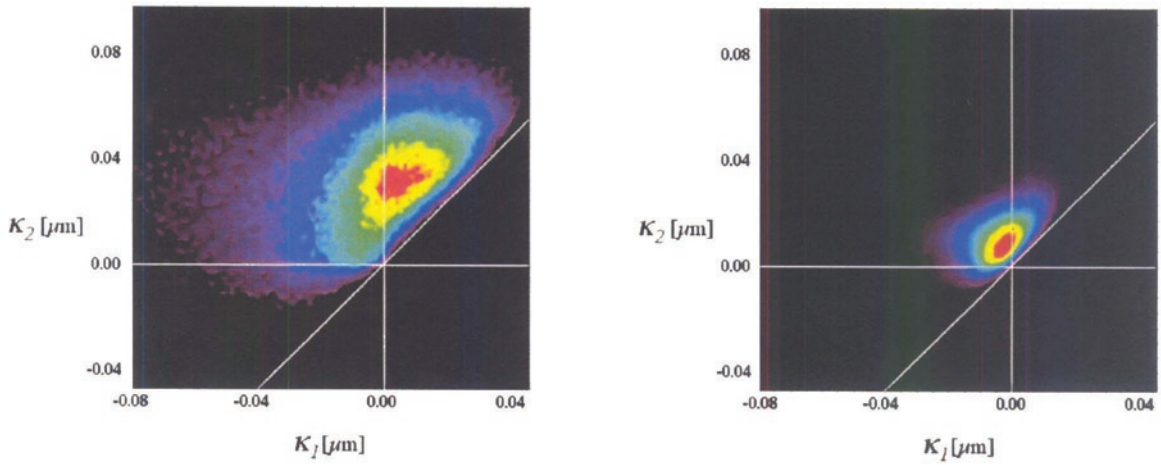

Figure 7: (a) and (b) show the probability density distributions in κ_1 κ_2 - coordinates for the microstructures of the 3 minutes and 2900 minutes coarsened samples, respectively. The probability density function is defined similar to the one used in figure 5.

The maximum for the 3 minute sample lies at positive κ_1 and κ_2. If self similar coarsening were present this maximum would with increasing coarsening time drift towards 0/0. The 2900 minute coarsened sample shows, in contrast, that the maximum has actually moved across the $\kappa_1 = 0$ line.

Conclusions

The evolution of dendritic microstructures during isothermal coarsening for Pb-80wt.%Sn was studied. Using an automated serial-sectioning technique we obtained three-dimensional reconstructions of the dendritic microstructures coarsened for different lengths of time. These reconstructions can also be used to determine plots of the probability density of the mean and Gaussian curvatures. These plots show quantitatively the increase in size scale of a solid-liquid mixture that is expected during coarsening. Although the overall change in lengthscale suggests classic self-similar coarsening, plots of the curvature distribtuions show that the microstructure is undergoing morphological changes as it coarsens. This might eventually lead to a break up of the dendritic structure for very long coarsening times.

Information on the three-dimensional morphology will advance substantially our understanding of the dendrite coarsening process and will provide insights into the coarsening process that can be used to improve theoretical descriptions and simulations of the entire solidification process.

Acknowledgements

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