THREE-DIMENSIONAL CHARACTERIZATION OF DENDRITIC MICROSTRUCTURES

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Abstract—The first complete characterization of a dendritic microstructure is presented. The dendrite morphology is obtained via a novel serial-sectioning process that allows the three-dimensional microstructure of opaque materials to be determined in a routine manner. Using the reconstructed dendrite we determine the spatial distribution of the mean and Gaussian curvature as well as the probability density distributions of these curvatures. These measurements yield many insights into the local processes that shape these topologically complex structures that are vital in setting many material properties.

Keywords: Phase transformations; Diffusion; Coarsening; Dendrite

1. INTRODUCTION

Dendrites are topologically complex structures that are found in systems ranging from metal castings to snow-flakes. These tree-like structures result from a morphological instability of the solid–liquid or solid–vapor interface and often build a connected solid network throughout an entire region undergoing a phase transformation. The morphology of the dendritic structure is of technological importance as dendrites constitute the primary growth morphology during the early stages of virtually all solidification processes and as many important properties of materials are intimately related to the morphology of the dendrite. In nearly all systems dendrites begin coarsening immediately upon formation. During the coarsening process the average length scale of the system increases and the dendrite shape evolves resulting in a microstructure determined largely by the coarsening process.

Although the importance of characterizing and understanding the processes that shape the morphology of dendrites is clear, experiments as well as simulations aimed at measuring or predicting the morphology have proven to be challenging. Simulations of the evolution of dendrite morphology in three dimensions are difficult due to the complex topology of the dendrites and the large size of the computational domain required. Experimentally, only one average microstructural parameter, usually the secondary dendrite arm spacing, is determined for various conditions, e.g. different coarsening times. Using one metric to characterize the topologically complex structure of a dendrite is clearly inadequate. Realistic simulations are even more difficult to undertake as neither the initial dendrite morphology to be employed in such calculations nor the morphology of a coarsened dendrite are known. Thus, the veracity of such simulations cannot be tested. In an attempt to overcome these shortcomings Feijo´ o and Exner measured the mean curvature density distribution of dendritic structures using a deep-etching stereo-imaging technique [1]. However, their results are incomplete as only the secondary dendrite arm spacing is determined for various conditions, e.g. different coarsening times. Using one metric to characterize the topologically complex structure of a dendrite is clearly inadequate. Realistic simulations are even more difficult to undertake as neither the initial dendrite morphology to be employed in such calculations nor the morphology of a coarsened dendrite are known. Thus, the veracity of such simulations cannot be tested. In an attempt to overcome these shortcomings Feijo´ o and Exner measured the mean curvature density distribution of dendritic structures using a deep-etching stereo-imaging technique [1]. However, their results are incomplete as only the mean curvature density distribution was measured and no information was obtained on the local interface curvatures along the dendrite surface. Thus, the morphology of these widespread technologically important structures remains mostly uncharacterized.

The morphology of a dendrite is completely characterized by the curvature tensor along the dendrite surface. Here we consider the two invariants of the curvature tensor, the mean curvature, defined by $H = \frac{1}{r_1 + 1/r_2}$, and the Gaussian curvature given by $K = \frac{1}{r_1} \times \frac{1}{r_2}$, where $r_1$ and $r_2$ are the two principal radii of curvature of the surface. The variation of the mean curvature along the interface results in variations in the chemical potential along the interface, as is described by the Gibbs–Thompson equation. This then gives rise to a diffusion flux which, in turn, leads to coarsening. The importance of the
coupling between the mean curvature, diffusion fluxes, and dendrite evolution during coarsening was recently emphasized in a review article by Marsh and Glicksman [2]. The paper presents an overview of the current understanding of the mechanisms controlling dendrite coarsening. It is clear from this paper that experimental validation of these mechanisms requires an evaluation of the three-dimensional microstructure. The Gaussian curvature provides an essential measure of the morphology since it distinguishes between a saddle-shaped surface and a convex or concave surface. In addition, the evolution of the Gaussian and mean curvatures are coupled [3]

\[
\frac{\partial H}{\partial t} = -(2H^2 - K)v - (\partial_1^2 H + \partial_2^2 H)/2 \tag{1}
\]

\[
\frac{\partial K}{\partial t} = -2HKv - H(\partial_1^2 H + \partial_2^2 H) + \sqrt{H^2 - K(\partial_1^2 H - \partial_2^2 H)} \tag{2}
\]

where \( t \) is the time and \( v \) is the velocity of the surface along the surface normal; 1 and 2 denote the two principal directions along the surface. This coupling can be illustrated by the comparison of the evolution of \( H \) and \( K \) for a spherical surface with \( H \) and \( K \) for a saddle-shaped surface, a surface where the two principal radii of curvature are of opposite signs. From equations (1) and (2) it is clear that both \( \partial_1 H \) and \( \partial_2 K \) for the spherical surface can be quite different from those for a saddle-shaped surface, even if both have the same \( H \) and \( v \). Thus, although only the local mean curvatures influence the transport of material via diffusion, the local mean curvature at any time is not independent of the local Gaussian curvature. Therefore, a complete characterization of a dendritic structure requires that the mean as well as Gaussian curvature be measured with high spatial resolution.

2. RESULTS AND DISCUSSION

To measure the mean and Gaussian curvature of a dendrite surface we employed a newly developed serial-sectioning method [4] that overcomes many of the experimental difficulties associated with the more standard polishing approach: the rate at which sections are prepared is orders of magnitude faster and it is far more accurate. This new approach allows us to report the first complete characterization of a dendritic microstructure. The method, however, has applications beyond the study of dendrites as it can be employed to analyze the morphology of a vast array of microstructures in opaque materials. Since it employs only small-scale equipment and can be done in a lab, the technique has significant advantages over more standard tomographic approaches (X-ray imaging methods). We used a Pb–Sn sample containing ca. 73 wt% Sn that was cooled from the liquid state at a rate of 3.8 K/min. At ca. 197°C tin-dendrites formed, grew, and coarsened for about 220 s. At 183°C the liquid between the dendrites solidified into a eutectic structure. The samples were then reheated to 185°C, held at this temperature for 200 s, and quenched to room temperature. During this second stage only coarsening occurred. This procedure resulted in a fine eutectic between the dendrites. After this treatment the samples were serial sectioned, etched, and then reconstructed using a computer.

Figure 1 shows the reconstructed microstructure based upon 60 sections spaced 2 μm apart each. The reconstructed three-dimensional image was smoothed prior to surface rendering in order to reduce noise on the pixel level. The reconstructed volume consists of secondary and ternary dendrite arms that show some signs of the ongoing coarsening process. Evident is the interconnected nature of the microstructure.

For illustration purposes we have extracted one secondary dendrite arm, its sidebranches and a portion of another secondary arm to which it is contacting; see Fig. 2. The structure shows clear signs of coarsening: ternary arms have coalesced (1–3) and the secondary arm in the front has thinned and then split (4).

Once the interface has been rendered, it is possible to determine the mean and Gaussian curvature at each point on the surface. These curvatures are determined using a method developed by Hashimoto and coworkers [5, 6]. Using their approach we determined the mean and Gaussian curvature at each point along the surface and probability density distributions of the mean and Gaussian curvatures for the entire volume sampled; see Figs 3 and 4, respectively. The probability density for the mean curvature \( P_m(H) \) is defined such that the integral of \( P_m(H) \) from \( H_1 \) to \( H_2 \) gives the probability that a randomly chosen point on the surface has a mean curvature \( H \), with \( H_1 \leq H \leq H_2 \). The probability density for the Gaussian curvature is defined similarly.

The mean curvature distribution shows an asymmetric peak with a maximum at a positive mean curvature. It is qualitatively similar to that measured by Feijoo and Exner [1]. The average mean curvature is clearly positive. One-eighth of the surface area of the dendrite surface is, however, negatively curved. Negative mean curvatures are present when the surface is dimpled or is saddle-shaped with the negative principal curvature dominating. The mean curvature distribution also provides a characteristic length for the microstructure: the inverse mean curvature, \( \langle H \rangle^{-1} \). For the Sn dendrite \( \langle H \rangle^{-1} = 23.9 \) μm. The commonly used characteristic lengths of a dendritic microstructure are the specific surface area, \( S_\Omega \), and the average secondary arm spacing, \( \lambda_2 \). \( S_\Omega \), however, has the disadvantage of being volume fraction dependent, and \( \lambda_2 \) can be difficult to obtain or is no longer defined when the structure undergoes morphological changes, e.g. if the dendrite splits into separate particles. \( \langle H \rangle^{-1} \) can be used as a characteristic length.
regardless of the topology of the microstructure and it can be used to compare structures across a range of volume fractions [2].

The Gaussian curvature distribution (Fig. 4) identifies saddle-shaped surfaces as those having negative Gaussian curvature and cylindrical surfaces as those having a zero Gaussian curvature. The plot reveals that slightly more than half, 51%, of the surface is saddle-shaped. The strong peak close to zero suggests that the dendritic structure consist mostly of cylindrical or near-cylindrical shapes. Some dendrite coarsening models approximate dendrite arms as cylinders, see e.g. [7], and calculate the rate of thinning or shortening of the arms. Although these models seem to be confirmed by our findings, it should be noted that large simplifications were used in the models and that the dendritic structure will most likely undergo dramatic shape changes as coarsening proceeds. For example, cylindrically-shaped arms can coarsen by Rayleigh instability wherein the arm thins and eventually splits into particles [8]. Although signs of this instability seem be present in Fig. 2, where the sidearms show variations in thickness, we will show in the following that these variations are not due to Rayleigh instability. Nevertheless, later in the coarsening process Rayleigh instability as well as other dramatic topological changes should occur.

In order to understand the structure in more detail we have taken the region shown in Fig. 2 and colored the surface based on the local mean curvature; see Fig. 5. We find that most of the negative mean curvatures are at necks where two arms coalesce. There are two dimples on the surface, one in the front (5) and a second (not visible) directly behind (5). Both are undoubtedly a remnant of one earlier coalescence event. Dimpled regions can evolve in a manner that is qualitatively different than in the well understood case of systems composed of separated convex particles. In systems with particles, small particles (high curvature) tend to shrink and large particles (low curvature) tend to grow.
High mean curvatures are present at thin necks or where an arm has just separated (6, 7). In contrast to the assumptions made in many classical models of dendritic coarsening [2], the mean curvatures close to the stem, the secondary arm in this case, are mostly positive (9, 10). One exception is the pendant-drop like arm (11), where the mean curvature at the root is higher than in other portions of the arm, indicating that this arm may be in the process of separating from the stem. It is important to note that the mean curvature on the sidearms is actually lower where the arms are thinner (8). This is due to the saddle shaped nature of the surface. As a result these thinner locations are actually growing at the expense of the thicker areas. This is the opposite of a Rayleigh instability, where the mean curvature is larger where the arms are thinner and the arms tend to split into convex particles. It is likely that although some coarsening has occurred, the thickness variations of the arms are a remnant of the dendrite growth process, i.e. the morphological instability that led to the formation of dendrite arms. As mentioned earlier, however, in later stages of coarsening we still expect Rayleigh instability to occur (see e.g. [10, 11]). The probability distribution for Gaussian curvature, Fig. 4, indicates that half of the surface is saddle shaped or equivalently has negative Gaussian curvature. Using Fig. 6 it is possible to examine the spatial distribution of this negative Gaussian curvature. Most of the very small Gaussian curvatures are found at the necks. As expected, the surface around the hole near (10) has a negative Gaussian curvature as one principle radius of curvature is negative. The strong positive regions curvature) tend to grow. The average mean curvature decreases monotonically with time. This is in general expected to hold true for dendritic systems [2, 9], although just the opposite can happen locally: the mean curvature of a dimpled surface (low curvature) will increase via the deposition of material in the dimple. Thus, dimples will actually shrink with time until they vanish. The direction of the diffusion flux in the particle as well as the dimple case, however, is from areas of high mean curvature to areas of low mean curvature.
exactly match the regions with the largest mean curvatures. In general, it is easier to determine the signs of the principle radii of curvature visually, and hence the sign of the Gaussian curvature, than it is to determine the sign of the mean curvature.

Predictions of the mean curvature distribution exist only for a system of spherical particles; see Fig. 7. It is not surprising, then, that the measured and predicted mean curvature distributions are quite different. There are two asymptotic states to which the system can evolve: a system composed of spherical solid particles in a liquid or spherical droplets of liquid in a solid. The distribution on the left corresponds to a structure where the solid dendrites become the matrix phase and the liquid becomes the particles, as might happen in a system where there are no solid grain boundaries formed upon particle contact in combination with a sufficiently high volume fraction of dendritic phase. The distribution on the right corresponds to the case where the dendrites break up into spherical solid particles and the liquid becomes the matrix, as has been observed many times [2, 7, 12], when grain boundaries can form or the volume fraction of solid was sufficiently low. Although the mean curvature of the dendritic structure is mostly positive and thus significantly closer to the distribution on the right, it is unclear to which distribution the structure may evolve.

3. CONCLUSIONS

With this work we initiate a new era in the study of microstructures: routine measurement of the three-dimensional morphology of opaque materials with sufficient resolution to permit the interfacial curvature to be measured. We have applied this technique to determine the morphology of a dendrite. Our results show that the majority of the dendrite, although the surface is 51% saddle-shaped, can be reasonably well approximated as contiguous near-cylindrical objects. Because of the high spatial resolution of the data we were able to show that the sidearms are not undergoing Rayleigh instabilities and that the thickness variations of the sidearms can thus be ascribed to the preceding growth process rather than to the coarsening process. Finally, the three-dimensional character of the measurements allows a snapshot of the spatial distribution of the mean curvature to be determined and from this the diffusion fluxes that are at the core of the coarsening process for these topologically complex structures.

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