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The evolution of dendrites during coarsening: Fragmentation and morphology

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ABSTRACT

The process of fragmentation of dendrite arms during coarsening remains poorly understood. We perform isothermal coarsening experiments of dendritic solid-liquid mixtures using PbSn alloys aboard the International Space Station (ISS), since arms that fission from the stem do not sediment and thus can be detected. The morphology of the structure and the number of fragments (fissioned arms) were determined using three-dimensional reconstructions. The evolution of the microstructure, change in length scale, interfacial shape distributions, number and distribution of fragments as well as the connectivity of the structures (handles) across coarsening time are discussed. We find that: the inverse of surface area per unit volume S_V^{-1} increases with time as $t^{1/3}$ in a manner that is almost identical to a sample coarsened on earth; the number of fragments per unit volume scaled by S_V^{3} is independent of time. Thus, it is possible to predict the number of fragments during coarsening by a measurement of S_V; the connectivity of the structures as measured by the number of handles in the structure scaled by S_V^{3} is also independent of coarsening time. We find that there is more coalescence of dendrite arms during coarsening than fragmentation.

1. Introduction

The fragmentation of dendrites has important consequences during the solidification of alloys. Fragments can be transported by convection ahead of the tips of columnar dendrites into the undercooled liquid region, and form equiaxed grains (columnar to equiaxed transition, CET). Dendrite fragments can lead also to misoriented grains (freckles) within the columnar region [1], an important casting defect in single crystal turbine blades [2,3]. Further, dendrite fragmentation has been linked to the development of highly refined grain structures in the solidification of undercooled melts, producing desirable characteristics [4,5].

Inspite of the importance of dendrite fragmentation, the mechanisms by which it takes place, the rate at which it occurs, as well as the effect of fragmentation on the connectivity of the structures remain controversial or poorly understood. There is evidence that solute inhomogeneities in the liquid that result from convection during directional solidification can lead to melting at the roots of the secondary arms [6–8]. Fluid flow during solidification may induce stresses that lead to detachment by changing the local solubility of solute at the root [9,10]. Alternatively, the fissioning process can be driven by interfacial curvatures at the root, similar to a Rayleigh instability [11–13]. In this case, mass is transported from the negatively curved roots to the small positive mean curvature regions of the surrounding interfaces due to the Gibbs-Thomson effect.

In addition to the uncertainty regarding the mechanisms, there is very little knowledge regarding the rate at which fragmentation takes place, especially at long coarsening times. A major problem with the empirical analysis of fragmentation is that, on earth, fragments sediment quickly, coalescing with existing dendrites. This precludes their measurement as well as an analysis of the effect of fragmentation on the structure. This paper reports results from experiments conducted aboard the ISS where the microgravity conditions allow a detection of the fragments as well as of effects on the structure from which they detach. The microgravity conditions, absence of convection, and isothermal nature of the experiments allow a focus on capillary driven fragmentation. Below, we provide the background of the research, discuss the method for determining microstructures and their change across coarsening time, as well as the methods for determining fragmentation.

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2. Background

When a material is cast and solidification occurs, coarsening strongly affects the microstructure of the cast material. In the coarsening of a system of spherical particles, a single parameter, the particle radius, sets the interfacial concentration. Lifshitz and Slyozov [14] and Wagner [15] used the direct link between the curvature of a particle and its volume to develop analytical models for the coarsening process. Topologically complex systems such as dendritic solid-liquid mixtures have both positive and negative interfacial curvatures. As there is no relation between the curvature of a dendrite-like object and its size, analytical models of the dendritic coarsening have not been possible. This has limited the understanding of the evolution of dendritic structures as well as the processes leading to fragmentation, and has resulted in empirical approaches to complement the idealized analytical models.

To characterize dendritic structures, an important morphological measure is the curvature of the solid-liquid interfaces [16,17]. The curvature is characterized in terms of the two invariants of the curvature tensor, the mean curvature, H, and the Gaussian curvature, K:

$$H = \frac{1}{2}(\kappa_1 + \kappa_2) \tag{1}$$

$$K = \kappa_1 \kappa_2 \tag{2}$$

where κ_1 and κ_2 are the two principal curvatures of the interface.

The variation in H is the driving force in coarsening. There is an excess free energy associated with the presence of the interface and the system evolves to minimize the free energy. It does this by reducing its interfacial area through a mass diffusion process. The significance of the variation in the interfacial mean curvature in this mass transfer process is expressed by the Gibbs-Thomson equation for a binary alloy:

$$C_L = C_0 + \Gamma H \tag{3}$$

where C_L is the composition in the liquid at the interface, C_0 is the equilibrium liquid composition at a flat solid-liquid interface, and Γ is the capillary length. Since the mean curvature varies with position along the solid-liquid interface of a dendrite, the composition in the liquid changes as well. This gives rise to diffusive transport of solute and an evolution of the structure.

The spacing between secondary arms, λ_2, has long been advocated as an appropriate length scale, and to quantify the coarsening process, modeling has predicted the increase of the time dependence of the secondary arm spacing λ_2 as $t^{1/3}$ [11,18–26]. Using λ_2 as a length scale has two important drawbacks however: its value can be a function of the manner in which it is measured due to the challenge of characterizing 3D structures using planar sections, and it is definable only for structures with a dendritic morphology [16]. Marsh and Glicksman [16] defined a length scale that is independent of the interfacial morphology, the surface area per unit volume S_V of the coarsening phase and found that $S_V^{-1} \sim t^{1/3}$ despite the morphology of the microstructure. This has been confirmed in a number of other studies [27–29]. Thus, even though the microstructure is not self-similar, some average properties can still evolve as $t^{1/3}$. S_V is related to coarsening time as [16]:

$$S_V^{-3}(t) - S_V^{-3}(0) = Kt \tag{4}$$

Certain microstructural measures require a three-dimensional reconstruction of a microstructure such as the number of independent bodies, or fragments, and the genus. The method for isolating possible fragments is described in Section 4.2, once we have discussed how the coarsened microstructures are reconstructed. In addition to this direct measurement of fragmentation, a related measurement is the change in the genus, g, of the structure. The genus is defined as the maximum number of cuts along closed simple curved surfaces that can be made through a body without breaking it up into smaller bodies [30]. It represents the number of handles or holes in a body. The genus is related to the Gaussian curvature of a surface. A closed surface has an integral Gaussian curvature K_{total} equal to 4π with a genus G of 0. Adding a hole or handle to the structure increases the genus by one and decreases K_{total} by 4π. Thus, the genus is given by Ref. [31]

$$g = 1 - \frac{K_{total}}{4\pi} \tag{5}$$

The Euler characteristic (χ) can be used to measure the genus:

$$g = 1 - \frac{\chi}{2} \tag{6}$$

and is given by

$$\chi = n - e + f \tag{7}$$

with n the number of nodes (vertices) of a surface, e the number of edges (links), and f the number of faces. Each void (cavity) within the structure decreases the genus by one, providing a relationship between g, h, and v [31],

$$g = h - v \tag{8}$$

with h the number of handles and v the number of voids within the structure (0 in the case of our samples). When dendritic microstructures coarsen, their morphology changes. While the morphology may change, the connectivity as measured by the genus need not change. However, when there is fragmentation, the genus will necessarily be affected due to the creation of a new closed body. Tracking the change in the genus, the number of bodies in the volume, N_V, and the handles thus provides complementary information to the count data of the fragments for determining the extent of fragmentation during coarsening.

The genus has been measured in experiments [31,32] but was limited to either liquid domains in high volume fraction solid systems or solid domains in mixtures with relatively high volume fractions of solid, in excess of 43%. In low volume fraction solid mixtures, as soon as a secondary arm detaches, it sediments and can coalesce with another piece of solid, resulting in no change in the genus. This makes it difficult to relate the genus to the secondary arm detachment when the experiments are performed on earth.

To obtain the genus g of our reconstructions, the following process was followed. The total number of nodes n, faces f and edges e were obtained using the mesh that defines the interfaces from each reconstruction. Once the three values were obtained, the Euler characteristic was calculated using Eq. (7). The genus was then calculated using Eq. (6).

In their study of the topology of Al-Cu samples, Mendoza et al. [31] used Eq. (8) to calculate the genus. This was necessary since the number of voids in the structure affects on the measurement of the genus. We similarly searched our reconstructions to determine the number of voids. However, none were found in any of the samples. While our reconstructions do not have voids, they have fragments. This requires us to slightly adjust the conversion from genus to handles. Without voids, the genus equals the number of handles. However, since each closed structure adds “2” to the Euler