

The dynamics of interfaces during coarsening in solid–liquid systems

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Abstract

The isothermal coarsening of dendritic Al–Cu microstructures is examined using time-resolved, in situ synchrotron-based X-ray tomographic microscopy. By obtaining the data at the coarsening temperature (in situ) with speeds that are on the order of or faster than the ongoing evolution of the microstructure (time-resolved, 4-D), we examine the dynamic morphological evolution of the solid–liquid interfaces in two solid volume fractions; as such, the relationship between the velocity of the evolving interface and characteristics that define the morphology of the interface is determined. We find that, while there is a correlation between velocity and mean curvature of the interface, there is a significant dispersion in the velocities for a given value of mean curvature. In addition, the Gaussian curvature plays a role in determining the interface velocity even though it has no effect on the chemical potential at the interface. We find that there are many interface patches of various morphologies and size scales that are not evolving during the coarsening process. At higher solid volume fractions and longer coarsening times, more of the structure is active in the coarsening process, and there is an increase in localized diffusional interactions.

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1. Introduction

Coarsening is a naturally occurring phenomenon that decreases the energy of the system by decreasing the overall curvature of the interface between the solid and liquid phases. It occurs by diffusion of mass from regions of high curvature to regions of low curvature since the concentration of the liquid is given by the Gibbs–Thomson equation:

$$C_L = C_\infty + l_c H \quad (1)$$

C_L is the composition of the liquid at a curved interface, C_∞ is the composition of the liquid at a flat interface, l_c is the capillary length, which is a function of the

solid–liquid interfacial energy [1], and H is the mean curvature of the interface, given by:

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

where κ_1 and κ_2 are the eigenvalues of the 2×2 Hessian matrix that define the interfacial curvature tensor; thus, they are minimum and maximum principal curvatures of the interface, respectively. The Hessian matrix has two invariants, with H being one, defined as half the trace of the matrix. The other invariant is the Gaussian curvature, K [2], defined as the determinant of the Hessian:

$$K = \kappa_1 \kappa_2 \quad (3)$$

As these equations show, the concentration at a point on the interface with a certain mean curvature is independent of the Gaussian curvature. The net effect of coarsening is a

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decrease in the total interfacial area over time and an increase in the characteristic length scale of the two-phase system, while the volume fractions of the two phases remain nearly constant.

In the past, a general understanding of the dynamics of the coarsening process was determined by solving a diffusion equation subject to the boundary conditions given by the Gibbs–Thomson equation. Lifshitz and Slyozov [3] and Wagner [4] (LSW) examined the coarsening process in a system of spherical particles and showed that the growth rate of the average particle size follows a $t^{1/3}$ power law:

$$\bar{R}^3(t) - \bar{R}^3(0) = K_{LSW}t \quad (4)$$

where \bar{R} is the average particle radius at a given time, t , \bar{R}_0 is the average particle radius at the start of self-similar coarsening, and K_{LSW} is a constant that depends on the diffusion coefficient, capillary length, interfacial energy and other material parameters. In the zero solid volume fraction limit, where particles interact with a mean field concentration, the growth rate of a particle of radius, R , is [1]:

$$\frac{dR}{dt} = \left[\frac{Dl_c}{C^s - C^l} \right] \frac{1}{R} \left(\frac{1}{R_c} - \frac{1}{R} \right) \quad (5)$$

where t is time, D is the diffusion coefficient, l_c is the capillary length, C^s and C^l are the compositions of the solid and liquid phases respectively, and R_c is the critical radius. The critical radius is the particle radius with zero growth rate and thus represents the particle with an interface composition equal to the mean field composition.

The particle size distribution and kinetics of the average particle size predicted by LSW only hold for spherical domains where the structures are evolving self-similarly. Self-similar evolution of a microstructure is characterized by time independent equivalent morphologies and topologies when the microstructure is scaled by a characteristic length, like the average particle radius, \bar{R} , in spherical systems. When dealing with dendritic microstructures, complex systems that have spatially varying curvatures and tend to interact with long-range diffusion fields, the kinetics that govern the evolution of the microstructure are expected to be more complicated.

Until recently, most coarsening studies have employed ex situ techniques [5–10], and thus the evolution of the structure is not captured at the coarsening temperature. Instead, the analysis occurs after quenching, and multiple samples provide snapshots of the microstructure at discrete times during the coarsening process. This is sufficient for understanding basic coarsening principles, but it is not adequate for extracting the more detailed dynamics of the process. Using techniques such as synchrotron-based X-ray tomographic microscopy provides the opportunity to study such processes at the coarsening temperature on a single sample volume. X-ray tomographic microscopy has become an instrumental tool for capturing the evolution, both in three and four dimensions (3-D and 4-D, respectively), of many materials; 4-D, in this case, means that

the data is collected in a time-resolved manner. This requires that the speed of the 3-D scan(s) is faster than the dynamics of microstructure evolution and the sequential collection of such data sets is thus adequate to resolve these changes. Al–Cu is an ideal material for imaging with X-rays because there is significant absorption contrast due to the density difference between Al and Cu. Further, dendritic features are typically on the micrometer length scale and thus can be spatially resolved using standard setups at X-ray tomographic microscopy beamlines.

It is thus not surprising that Al–Cu alloys have been imaged relatively frequently using X-rays to study solidification and coarsening of dendrites [11–23]. Such studies have focused on, for example, the evolution of the surface area per unit volume, S_v , during solidification [15], the evolution of the mean and Gaussian curvatures of a single dendrite at high temporal resolution [18] and the topological changes that occur when a dendrite arm fissions [21–23]. (For a recent review, see Ref. [24].)

In this study, we describe the 4-D dynamics of coarsening in binary Al–Cu samples by extracting the velocity of the solid–liquid interface using two experimental microstructures captured in a close interval of time during the coarsening process. This is achieved by using a technique first presented by Aagesen et al. [21]. A relationship is established between these velocities and the principal curvatures that characterize the morphology of the solid–liquid interface. Further, the rate of change in area of the interface for a given pair of principal curvatures is examined. This metric provides complementary information to traditional interface shape distributions (ISDs) [5] and to the velocity information, as it quantifies the effect that localized shape changes have on the overall coarsening of the microstructure. Finally, the effects of solid volume fraction on the dynamics of the interface are discussed.

2. Experimental procedure

Samples of Al–15 wt.% Cu and Al–29 wt.% Cu were previously directionally solidified as in Ref. [5]. These two Cu weight fractions correspond to average solid volume fractions of 77% and 35%, respectively. The samples were prepared for X-ray tomographic microscopy by wire-cutting a 1 mm diameter sample out of the larger ingot and then placing it inside a thin-walled boron nitride (BN) sample holder (BN provided by Goodfellow, Inc, UK).

The experiments were conducted on the TOMographic Microscopy and Coherent rAdiology experimenTs (TOM-CAT) beamline of the Swiss Light Source (Paul Scherrer Institut, Switzerland) [25]. The high solid volume fraction data was collected in 2008 using a pco.2000 camera with standard optics at 10× magnification with a monochromatic energy of 20 keV. A total of 721 projections were captured in 180° of rotation, and the data was binned to decrease the total scan time. This resulted in a pixel size of 1.4 μm with a single 3-D data set collected every

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