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Particle tracking during Ostwald ripening using time-resolved laboratory X-ray microtomography

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ABSTRACT

Laboratory X-ray microtomography is investigated as a method for obtaining time-resolved images of microstructural coarsening of the semisolid state of Al–5 wt.% Cu samples during Ostwald ripening. Owing to the 3D imaging capability of tomography, this technique uniquely provides access to the growth rates of individual particles, thereby not only allowing a statistical characterization of coarsening—as has long been possible by conventional metallography—but also enabling quantification of the influence of local environment on particle boundary migration. The latter information is crucial to understanding growth kinetics during Ostwald ripening at high volume fractions of the coarsening phase. Automated image processing and segmentation routines were developed to close gaps in the network of particle boundaries and to track individual particles from one annealing step to the next. The particle tracking success rate places an upper bound of only a few percent on the likelihood of segmentation errors for any given particle. The accuracy of particle size trajectories extracted from the time-resolved tomographic reconstructions is correspondingly high. Statistically averaged coarsening data and individual particle growth rates are in excellent agreement with the results of prior experimental studies and with computer simulations of Ostwald ripening.

1. Introduction

As the resolution of X-ray tomography has improved from the millimeter to the submicrometer range, the scope of applications for this nondestructive 3D imaging technique has broadened from its early use in medical diagnostics to increasingly widespread employment as a tool for characterizing the complex internal microstructures of materials [1,2]. Tomography is particularly well suited to the study of multiphase materials, as differences in local X-ray absorption can be exploited to map the spatial extent of individual phase regions—even when the latter are interconnected in a complex manner in three dimensions [3,4]. Previously, such information could be gleaned only from a destructive technique like serial sectioning [5]; however, with the advent of high-resolution X-ray tomography, such studies no longer require cutting open the sample, thereby making it feasible to observe the evolution of phase boundaries during protracted mechanical and/or thermal processing [6,10].

This *in situ* 3D imaging capability makes X-ray microtomography ideally suited to the investigation of *Ostwald ripening*, a coarsening phenomenon first studied more than a century ago [11] and still of technological relevance today because of its prevalence during the synthesis and processing of modern multiphase materials [12,15]. In the simplest case of particles of one phase dispersed in a matrix of a second phase, Ostwald ripening manifests itself through the growth of larger particles at the expense of smaller ones, leading to an increase in the mean particle size $\langle R \rangle$ as the total number of embedded particles decreases (Fig. 1). The driving force for this process is

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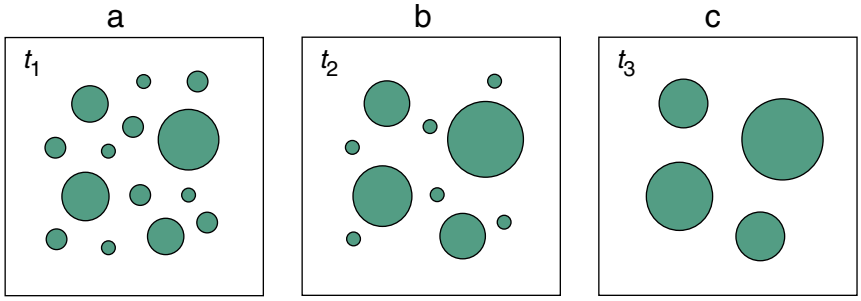

Fig. 1 – Schematic illustration of the Ostwald ripening of particles of one phase embedded in a matrix consisting of a different phase. Larger particles grow at the expense of smaller particles by atomic diffusion from the latter to the former through the matrix. This leads to an increase in the mean particle size $\langle R \rangle$ with time ($t_1 < t_2 < t_3$) without changing the volume fraction V_V of the coarsening phase.

the excess energy of interphase boundaries, the overall area of which is reduced when the volume of the embedded (coarsening) phase is concentrated in fewer particles of larger size.

The first widely accepted analytic model for Ostwald ripening was proposed in 1961 by Lifshitz, Slyozov [16] and Wagner [17]. The so-called LSW model makes three primary predictions regarding the asymptotic kinetics of particle coarsening in a two-phase mixture:

(i) The cubed mean particle radius $\langle R \rangle^3$ of a given phase grows as a linear function of time t.
(ii) The particle size distribution takes on a time-independent shape when normalized by the corresponding mean particle radius $\langle R \rangle$.
(iii) The quantity $R^2 dR/dt$, which is proportional to the instantaneous growth rate of a particle of size R, depends linearly on $R/\langle R \rangle - 1$, entailing particle shrinkage for $R < \langle R \rangle$ and growth when $R > \langle R \rangle$.

Strictly speaking, the LSW results were derived in the limit of a vanishing volume fraction of the coarsening phase (*i.e.* $V_V \rightarrow 0$), which is equivalent to the assumption of no overlap of the concentration depletion zones surrounding the shrinking/growing particles [12,18,19]. This assumption leads to discrepancies between theory and experiment at higher, technologically relevant values for V_V [20,21]. Over the past six decades, numerous attempts have been made to extend the LSW model to higher volume fractions by taking particle–particle interactions into account, at least in a mean-field sense [18,22–26]. For example, Glicksman et al. [24] and Wang et al. [26] developed a "diffusion screening theory" to describe multi-particle systems in the range $0 < V_V < 0.3$, and Marsh and Glicksman [23] established the concept of a "statistical field cell" that is valid for volume fractions up to 0.6. At still higher values for V_V, however, all current approaches to modeling Ostwald ripening begin to break down [27].

In order to guide the development of analytic models for Ostwald ripening in the high-V_V regime, it is necessary to measure the effect of particle–particle interactions on the rate of growth in each particle size class R and to assess their dependence on the overall volume fraction V_V of the coarsening phase. Such information can be obtained only from a nondestructive 3D imaging technique that is able to map out the local environment of each particle in a real coarsening system and to deliver relevant data for quantifying the influence of that environment on the particle's growth kinetics. In this work we demonstrate that absorption-contrast X-ray microtomography meets both of these criteria. Moreover, we show that the necessary characterization capabilities are not exclusive to the specialized instrumentation found at synchrotron beamlines: the latest generation of laboratory X-ray tomographs affords sufficient intensity and resolution for performing such studies, as well.

In Section 2, we describe the sample preparation and tomographic characterization protocol that was followed to investigate Ostwald ripening in a two-phase system with $V_V = 0.74$ during both long-term (LT) and short-term (ST) annealing series. By tracking individual particles over several annealing steps, according to the approach presented in Section 3, we extract particle growth/shrinkage trajectories and compare them to statistical measures for the evolution of the ensemble of particles, discussing the results in the context of prior experimental and theoretical studies of Ostwald ripening. Finally, in Section 4 we assess uncertainties in the microstructural data obtained by this approach, considering not only experimental factors but also the consequences of segmentation artifacts on the accurate tracking of particles from one tomographic reconstruction to the next.

2. Material and Methods

The aggregate state of the phases in a polycrystalline metal undergoing Ostwald ripening can be solid–solid [15] or solid–liquid [21]. Solid–liquid systems offer the advantage of minimizing the interfacial stresses between matrix and coarsening phases [28], thereby eliminating at least one factor having the potential to interfere with conventional coarsening kinetics [20]. Furthermore, ripening occurs faster in solid–liquid systems, owing to the higher rate of atomic diffusion through a liquid than a solid, thereby shortening the annealing time needed to induce a measurable change in the microstructure.

For these reasons, we chose to study a high-purity Al–5 wt.% Cu alloy in the semisolid state at temperatures above the solidus line (~560 °C). Prior to annealing, the alloy was cold-rolled to a thickness reduction of 50% and homogenized