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Stochastic modeling of individual grain behavior during Ostwald ripening at ultra-high volume fractions of the coarsening phase

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ABSTRACT

The evolution of grains during coarsening phenomena like Ostwald ripening is a focus of recent and ongoing research. In the present paper, a new and flexible model is proposed that describes the statistical evolution of the "typical" individual grain size as a function of neighborhood characteristics. The grain size evolution (GSE) model defines a stochastic process based on contemporary mathematical techniques and requires only few (natural) assumptions. It is fitted to time-resolved experimental data of a semisolid Al–Cu alloy, in which the coarsening phase has an ultra-high volume fraction $V_V = 0.93$. Evaluation shows that the model describes the experimental data quite closely. The nature of this modeling approach serves to improve the understanding of coarsening processes at the intermediate level between coarsening mechanisms and global statistical properties. Furthermore, the model enables predictive simulations to be performed, based on an extension of an existing 3D microstructure model (Spettl et al., 2015) to 4D.

1. Introduction

Ostwald ripening is a process that occurs in multiphase systems, where particles (or droplets) of one phase are embedded in a matrix of one or more other phases. In order to minimize the excess energy contributed by interphase boundaries, large particles grow while small particles shrink and, ultimately, disappear. An introduction to Ostwald ripening and its experimental and theoretical investigation can be found in, *e.g.*, [1–6], whereby the most important theoretical treatment of Ostwald ripening is the so-called LSW theory, presented by Lifshitz and Slyozov [7] as well as Wagner [8] in the early sixties. The latter describes the case of a vanishingly small volume fraction V_V of the coarsening phase, for which it is possible to treat every particle as an isolated object – *i.e.*, direct interactions between particles are not considered. The LSW theory predicts a power-law growth of the mean particle size (with exponent 3) and a particle size distribution whose shape does not change over time. In particular, the particle size distribution normalized to expectation unity is always the same distribution (statistical self-similarity [9]). However, the situation is more complicated for volume fractions V_V that are not near zero [10–15], which has become a popular topic of recent research in the field. For example, experimental investigations [16–19], analytical theories [20,21] and large-scale computer simulations – based on, *e.g.*, phase-field and Monte-Carlo methods [22–25] – have focused on volume fractions of technological relevance. Results suggest that the power-law growth and the self-similarity of particle size distributions still hold, although the shape of the particle size distribution changes at higher V_V [22,24], and for $V_V > 0.9$ the power-law exponent may manifest a crossover to that of single-phase grain growth (exponent 2 at $V_V = 1.0$) [22]. However, convergence to steady-state conditions can be slow [26,27].

The approaches mentioned above are all important for an improved understanding of the coarsening process. Yet, other (complementary) approaches have their merits, as well. For example, stochastic modeling of microstructures helps to identify the relevant structural characteristics of a given material, encompassing not only the average values of specific structural parameters but also their local fluctuations and spatial correlations. A stochastic 3D microstructure model for polycrystalline materials has been presented in [28], where the model was fitted to experimental data obtained for an Al-5 wt% Cu alloy heated to 592 °C. However, in [28] the dynamics of microstructural evolution were captured only at the statistical level but not for individual particles (hereafter referred to as *grains*). We aim to remedy this situation in the present paper. Our new treatment has several benefits. A stochastic description of individual grains and their evolution over time is useful to determine simple characteristics that influence (or, at least, are correlated with) the local evolution of grain sizes. The

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stochastic modeling approach proposed in the present paper can identify and account for such dependencies, while subsuming any additional, not-yet-identified factors influencing evolution to the "randomness" of the system. Note that stochastic models describing the evolution of grains have two important advantages. First, it is possible to identify (locally estimated) parameters of the microstructure that have a high impact on the local evolution. Second, predictive simulations are possible. As mentioned in [28], the latter is important for, *e.g.*, the multiscale computational models that are employed in Integrated Computational Materials Engineering (ICME) [29].

In this paper, we present a stochastic model that predicts changes in grain volume based on current grain size and characteristics of its neighborhood. More precisely, the model not only predicts the expected evolution of grain size but also its probability distribution – *i.e.*, the possible new grain sizes and their likelihoods. We call this model the *grain size evolution (GSE) model*. The latter is based on just a few assumptions – power-law growth [7,8], self-similarity [7–9], temporal and spatial Markov property [30,31] – and a further contemporary mathematical technique, the modeling of multivariate distributions with copulas [32,33]. The GSE model describes the dynamical behavior of the "typical"[1] individual grain. In a second step, the 3D model for entire grain systems [28] is extended to 4D by integrating the GSE model.

We show the capability of the model to represent phenomena observed in real materials by fitting it to time-resolved experimental data of a semisolid Al–Cu alloy, in which the coarsening phase occupies an ultra-high volume fraction ($V_V = 0.93$). These data were captured *in situ* with synchrotron X-ray tomography. It turns out that a large number of grains can be tracked, although the complexity of the image data renders it laborious to achieve high tracking efficiencies. Fortunately, even incomplete microstructural information suffices to establish the values of model parameters such that growth of individual grains is predicted quite accurately.

2. Experimental data, imaging and segmentation

In this section, the experimental sample, its time-resolved structural characterization, and image data processing are described.

2.1. Experimental sample and in situ 3D imaging

The experimental data considered here were drawn from the same measurements that were considered in [28]; however, now the time resolution of the data is exploited. Salient facts regarding sample preparation and 3D imaging are reviewed below.

An ingot of the alloy Al-5 wt% Cu was homogenized at 500 °C for 24 h in air and subsequently cold-rolled to a thickness reduction of 50%. Cylindrical specimens – 8.5 mm in length and 4 mm in diameter – were cut from the rolled plate by spark erosion. A furnace was constructed to allow for time-resolved *in situ* tomographic characterization of such samples using X-ray radiation at beamline ID15A of the European Synchrotron Radiation Facility (ESRF). Over the course of 25 h, a tomographic scan was recorded every 10 min while the specimen was held at 592 °C, which placed it in a semisolid state consisting of solid particles surrounded by a liquid matrix. Absorption-contrast tomography is able to distinguish between the particles and the matrix because the liquid absorbs X-rays more strongly than does the solid phase. This is due to the higher concentration of Cu dissolved in the matrix (22.4 wt% vs. only 3.5 wt% in the solid phase) at 592 °C. Application of the lever rule to the Al–Cu phase diagram [35] yields a volume fraction of the solid (coarsening) phase of $V_V = 0.93$ at thermodynamic equilibrium between the particles and the liquid matrix. The nominal resolution of the reconstructed tomographic data sets is given by the voxel side length of 5.36 μm.

We denote the 3D grayscale images obtained in this manner by $I_t = \{I_t(x,y,z) \in \{0,\dots,255\} : (x,y,z) \in W\}$, where $W \subset \mathbb{N}^3$ is the grid of voxel coordinates, the grayscale values are in $\{0,\dots,255\}$, and the annealing times $t \in T_{\text{exp}}$ belong to the set $T_{\text{exp}} = \{200, 210, \dots, 750\}$ (all specified in minutes). Note that the time span between two successive tomograms is always $t_{\text{step}} = 10$ min.

We make the same assumption as in [28] regarding steady-state conditions. We cannot be sure that the sample has actually reached the steady state by 200 min of annealing. Statistical characteristics (*e.g.*, the distributions of grain sizes and coordination numbers) seem to indicate the occurrence of self-similar coarsening [28], but it is still possible that the sample finds itself in a transient regime, the characteristics of which are evolving very slowly. In either event, it makes sense to fit a stochastic model to the experimental data, but in the transient case the stochastic model's predictive ability applies only to a limited extent beyond the time span at hand.

2.2. Identification of grains at fixed annealing times

In order to establish the parameters of the GSE model, we will require information regarding grain-size trajectories [19]. As a result, we must be able to recognize the same grain at subsequent or previous time steps, necessitating a consistent labeling of grains over the course of the measurements. The aim is to obtain a large number of trajectories, each of which extends over numerous time steps. In this section, we discuss the identification of individual grains within 3D data sets. These data form the basis for the tracking of grains across time steps, which we address in the following section.

Image processing was performed in a similar manner as in [28]. Ring artifacts were removed from grayscale data [36] (resulting image denoted by I'_t), a global thresholding step was performed (binary image denoted by B_t), and a smoothing step was applied (resulting image denoted by B'_t). The main difference with respect to the image processing performed in [28] may be found in the identification of grains. We still employ a watershed transformation [37–40], but instead of considering so-called *extended regional minima,* which we introduced in [28] to reduce the occurrence of oversegmentation, we now adopt the following simplified approach, which is computationally faster and therefore better suited to a large number of 3D data sets. For every local minimum, we can interpret the minimal distance to a grain boundary (*i.e.* the matrix phase) as the radius of a sphere centered at the local minimum. We then increase the radius of such a sphere by 10% and remove every other local minimum located within that sphere that has a smaller minimal distance than the local minimum at the sphere center. This thinning of the set of local minima is very simple and reduces oversegmentation in many cases – *i.e.*, when the shape of the grains that are to be detected does not deviate too much from a spherical shape. (The deviations are the reason for the 10% radius increase.) The result of a marker-based watershed transformation [38,41] applied to the thinned set of local minima is a binary image in which any holes in the network of grain boundaries have been filled in. This image is called B''_t. Finally, the same postprocessing was performed as in [28], yielding the

[1] The terminology "typical grain" employed in the present paper conveys the following mathematical concept. Consider a grain ensemble, for which each grain has properties like size, shape, *etc.* For an unboundedly increasing number of such grains, these properties can all be described by probability distributions. Then, the typical grain is a random grain whose properties have these distributions, and, thus, the typical grain is representative of the entire grain system – see [34] for further details.