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Shedding some light on the early grain growth regime: About the effect of the initial microstructure on normal grain growth

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ABSTRACT

In the present paper the effect of the initial microstructure on the coarsening kinetics of normal grain growth is investigated. To that aim, two microstructures characterized by two distinctively different grain size distributions—one following a normal and the other one following a bimodal distribution—are analyzed as they undergo coarsening by means of Monte Carlo Potts model simulations showing different behaviors at early relaxation times but reaching the same quasi-stationary self-similar growth regime for long-time annealing.

1. Introduction

The conventional concept of grain growth is based on the fact that polycrystalline materials are thermodynamically unstable. They undergo continuous grain coarsening, which is driven by a reduction of the total Gibbs free energy of the system. For the case of normal grain growth it is assumed that only the grain boundaries contribute to the reduction of the Gibbs free energy via the decrease of the total grain boundary area. It follows that normal grain growth is characterized by an average growth law of type

$$\langle R\rangle^2 = bt + \langle R\rangle_0^2, \tag{1}$$

where $\langle R\rangle$ is the mean linear grain size (grain radius) of an ensemble of grains, $\langle R\rangle_0$ the corresponding initial value, b is a growth constant and t is the annealing time [1]. In 1986 Mullins [2] showed that the time-dependence of grain and particle size (e.g., average grain size) in normal grain growth, Ostwald ripening, and bubble growth can be deduced from a statistical self-similarity hypothesis, according to which different configurations of the system observed at different annealing times in the self-similar regime are geometrically similar in a statistical sense. Hence, the structures metrical and topological properties can be described by time-independent functions of the scaled grain size:

1. The grain size distribution $F(R,t)$ can be written as a product of a time-dependent factor $g(t)$ and a size-dependent (but time-independent) scaled distribution $f(x)$;

$$F(R,t) = g(t)\cdot f(x) \tag{2}$$

where x is the scaled grain size defined as the grain radius of each grain divided by the average grain radius of the ensemble, $x = R/\langle R\rangle$. The 0th and 1st moment of $f(x)$ are equal to one. Hence standard deviation and skewness of the relative size distribution are constant.
2. The growth law describing the rate of size change for all grains can also be written in a separable form:

$$\dot{R}(R,t) = \langle\dot{R}\rangle(t)\cdot G(x) = \langle\dot{R}\rangle(t)\cdot[U(x)+x] \tag{3}$$

where $G(x)$ and $U(x)$ are time-invariant dimensionless functions depending solely on the scaled grain size x. According to Hillert's classical theory [3] the scaled growth law $U(x)$ takes the form $U(x) = -\Gamma x^{-1} + \Gamma x_c^{-1} - x$, where Γ is a growth parameter. Lifshitz and Slyozov [4] argued that from the requirement of volume conservation follows that the only stable state with a physical meaning must have a tangent to the x-axis resulting in a double-root for $U(x)$. This theory has been generalized by Streitenberger and Zöllner [5] to

$$U(x) = -\frac{Dx_0^2}{a(x_0)}\cdot x^{-1} + \frac{2Dx_0}{a(x_0)} - \frac{D}{a(x_0)}\cdot x \tag{4a}$$

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where x_0 is an upper cut-off, $a(x_0)$ is given by the requirement $\langle x \rangle = 1$, and D is the dimension of the system (here $D = 3$). It follows that the volumetric rate of change cam be written as:

$$R\dot{R}(x) = bx \cdot \left[-\frac{Dx_0^2}{a(x_0)} \cdot x^{-1} + \frac{2Dx_0}{a(x_0)} + \left(1 - \frac{D}{a(x_0)}\right) \cdot x \right] \tag{4b}$$

3. Both, the size distribution and the growth law have to fulfill the continuity equation in size space:

$$\frac{\partial F(R,t)}{\partial t} + \frac{\partial}{\partial R}(\dot{R} \cdot F(R,t)) = 0. \tag{5}$$

Inserting Eqs. (1)–(4) into Eq. (5) results in a one-parameter family of scaled and normalized grain size distribution functions, which all obey the Lifshitz–Slyozov stability conditions [4] yielding [5]:

$$f(x) = \begin{cases} a(x_0) \cdot x_0^{a(x_0)} \cdot e^{a(x_0)} \cdot \frac{x}{(x_0 - x)^{a(x_0)+2}} \cdot \exp\left(\frac{-a(x_0)x_0}{x_0 - x}\right) & \text{if } 0 \leqslant x \leqslant x_0 \\ 0 & \text{otherwise} \end{cases}. \tag{6}$$

4. Additionally, the topology as it can be described for example by the relation between relative grain size x and number of faces and neighboring grains s, respectively, is also time-independent and the structures are called self-similar. It has been shown by theoretical considerations and computer simulations that $s(x)$ can be described by a quadratic function (compare [6] and the references within):

$$s(x) = s_2 x^2 + s_1 x + s_0, \tag{7}$$

where s_0, s_1 and s_2 are geometric parameters.

All in all, scaling and self-similarity are well investigated, in particular, by means of computer simulations (e.g., [7–10]) and analytic theories of normal grain growth (e.g., [5,11–13]) generally under the assumption that grain boundaries are considered as plane surfaces and their migration is directed toward the center of curvature with a speed proportional to the curvature itself. It is especially assumed that all grain boundaries of the polycrystalline microstructure are characterized by a unique value for the surface tension as well as by the same mobility in agreement with the uniform boundary model [14].

Nevertheless, the temporal evolution of a grain microstructure as it happens in experiments as well as in computer simulations from an initial stage to the final quasi-stationary state is still poorly understood, which has been pointed out already more than a decade ago [15]. This initial period of time depends purely on the initial structure and is not self-similar and therewith not characterized by scaling and time-independent size distribution or topology. In the following some main results obtained by Monte Carlo Potts model simulations available in literature are summarized—most of them a by-product:

- In 1998 Miyake [16] reported on lattice-model-independent grain size distributions of normal grain growth in two and three dimensions and showed the temporal evolution of the normalized grain radius variance for different simulation temperatures and simulation lattices, where it can be seen (cf. Fig. 5 in [16]) that there exists an early growth regime, in which the variance is not constant and therewith the associated grain size distribution not time-independent.
- In the same year, Battaile and Holm [17] discussed the evolution of two-dimensional grain microstructures starting from an initial Hillert size distribution [3]. After long-time annealing the scaled grain size distribution deviate significantly from Hillerts classical prediction. Unfortunately Battaile and Holm do not show the evolution of the distribution from initial (Hillert) to the final state.
- A more thorough, albeit short paper on the effect of the initial grain size distribution on grain growth was presented by Wang et al. [18]. They simulated different microstructures characterized by Weibull distributions with different coefficients of variation and observed the temporal development of the latter finding that the difference in the coefficient of variation influences the early growth regime significantly, but not the final self-similar regime.
- The direct evolution of the scaled grain size distribution as well as of the distribution of the number of edges per grain has been shown by Yu and Esche [19] using surface plots of $f(x)[\langle R \rangle(t)]$.
- Thomas et al. [20] showed that coarsening of three-dimensional grains in crystals (or bubbles in dry foams) tends towards a universal, statistically scale-invariant regime. The structures reach a state that is indeed characterized by scaling and self-similarity for three different initial microstructures. As a by-product they presented the temporal development of the surface area as well as of the average number of faces and the associated higher order moments, where one can see clearly the difference between early and self-similar coarsening.

Yet none of these works were specifically aimed at analyzing the initial growth regime of normal grain growth.

The situation is quite different for Ostwald ripening, or particle coarsening, where comparatively many studies on the transient coarsening regime exist. For a recent overview and the relevant literature see Ref. [21], where especially the effect of the initial particle size distribution on the transient dynamics of Ostwald ripening is numerically analyzed using a phase-field model.

However, this problem is of the same importance for grain growth, since all of the aforementioned analytical equations describing grain growth, Eqs. (1)–(7), are strictly valid only for long-time annealing. For practical applications it is assumed that the relaxation time from the initial to the steady state in experiments is rather short and can be neglected. However, this is not true. For example experiments on nanocrystalline palladium [22] have shown that in the first 8–10 h both, average grain size and width of the size distribution, are nearly unchanged marking a rather long and uneventful initial period of annealing, which is followed by a sequence of quick changes from an exponential, over a linear, finally to a parabolic increase of the average grain size within approximately 12 h. On the other hand, the coarsening of solid-Sn particles in a Pb–Sn liquid has been studied [23] showing that while the average particle size changes according to the average growth law ($\langle R \rangle^3 \propto t$) by a factor of three, the scaled particle size distributions were not in agreement with any prediction of a self-similar state.

Hence, the problem to investigate and describe the dynamics of annealing in the initial respectively transient regime is of great importance. In particular, we would like to stress the point that since in this initial, non-equilibrium state, self-similarity does not yet exist, the continuity equation, Eq. (5), cannot be solved analytically and Eqs. (1)–(7) are not fulfilled. The only mathematical model possible is a numerical model namely a simulation. In the following, we will fill this gap by analyzing the early growth regime of normal grain growth by three-dimensional Monte Carlo Potts model simulations using two different initial grain microstructures—one following a normal and the other one following a bimodal distribution.