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GRAIN GROWTH AND MICROSTRUCTURAL EVOLUTION IN A TWO-DIMENSIONAL TWO-PHASE SOLID CONTAINING ONLY QUADRIJUNCTIONS

Danan Fan* and Long-Qing Chen
Department of Materials Science and Engineering, The Pennsylvania State University, University Park, Pennsylvania 16802

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Introduction

Recently, Cahn [1] performed a thermodynamic analysis for the stability of microstructures in a two-dimensional two-phase solid in which the volume fractions are not conserved, i.e., they have the same composition, and hence long-range diffusion and Ostwald ripening are not involved. In his theory, the microstructural stability was analyzed based on the energetic ratios of grain boundaries to the interphase boundary, i.e. $R_\alpha = \sigma_{\alpha\alpha}/\sigma_{\alpha\beta}$ and $R_\beta = \sigma_{\beta\beta}/\sigma_{\alpha\beta}$, where $\sigma_{\alpha\alpha}$ and $\sigma_{\beta\beta}$ are the grain boundary energies of the α and β phases, respectively, and $\sigma_{\alpha\beta}$ is the interphase boundary energy. It is shown that for $0 \le R_\alpha \le \sqrt{3}$, $\alpha\alpha\alpha$ trijunctions are stable; otherwise, they are unstable with respect to the nucleation of β grains. Similarly, $\beta\beta\beta$ trijunctions are stable for $0 \le R_\beta \le \sqrt{3}$ and are unstable with respect to the nucleation of α grains for $R_\beta > \sqrt{3}$. $\alpha\alpha\beta$ and $\alpha\beta\beta$ trijunctions are stable under the conditions of $0 \le R_\alpha < 2$ and $0 \le R_\beta < 2$. More interestingly, he found that the quadrijunctions $\alpha\beta\alpha\beta$ will become stable if the condition $R_\alpha^2 + R_\beta^2 \ge 4$ is satisfied [1].

Following Cahn's work, Holm *et al.* performed Monte Carlo simulations on the same system, i.e., a two-phase solid in which the volume fractions are not conserved [2]. They showed that quadrijunctions can indeed be stable within a certain range of the values for R_α and R_β as predicted by Cahn's thermodynamic analysis. More surprisingly, based on their simulations, they predicted that the grain growth in a system with only quadrijunctions may be frozen [2].

The main objective of this paper is to investigate the stability and evolution kinetics of quadrijunctions in a model two-dimensional two-phase solid in which the volume fractions are CONSERVED, using a continuum diffuse-interface grain growth model, i.e., by numerically solving a set of coupled continuum time-dependent partial differential equations. We have applied this model to grain growth in single-phase systems [3–5] and to coupled grain growth and Ostwald ripening in two-phase solids [6]. It should be emphasized that most two-phase solids in real applications belong to the case of conserved volume fractions.

*Current address: Los Alamos National Lab, T-11, MS B262, Los Alamos, NM 87545

Diffuse-Interface Description of a Two-Phase Microstructure

The details about this diffuse-interface field model have been given in a previous paper [6]. Within the diffuse-interface context, we describe an arbitrary two-phase polycrystalline microstructure using a set of continuous field variables [3–6],

$$\eta_1^{\alpha}(r),\ \eta_2^{\alpha}(r),\ \ldots,\ \eta_p^{\alpha}(r),\ \eta_1^{\beta}(r),\ \eta_2^{\beta}(r),\ \ldots,\ \eta_q^{\beta}(r),\ C(r) \tag{1}$$

where η_i^{α} (i = 1, . . . , p) and η_j^{β} (j = 1, . . . , q) are called orientation field variables with each representing grains of a given crystallographic orientation of a given phase. Those variables assume continuous values ranging from -1.0 to 1.0. $C(r)$ is the composition field which takes the value of C_{α} within an α grain and C_{β} within a β grain.

The total free energy of a two-phase system, F, is written as

$$F = \int \Big[f_o\left(C(r);\ \eta_1^{\alpha}(r),\eta_2^{\alpha}(r),\ldots,\ \eta_p^{\alpha}(r);\ \eta_1^{\beta}(r),\ \eta_2^{\beta}(r),\ \ldots,\ \eta_q^{\beta}(r)\right) + (\kappa_C/2)(\nabla C(r))^2 + (1/2)\sum_{i=1}^{p} \kappa_i^{\alpha}(\nabla \eta_i^{\alpha}(r))^2 + (1/2)\sum_{i=1}^{q} \kappa_i^{\beta}(\nabla \eta_i^{\beta}(r))^2 \Big] d^3r \tag{2}$$

where f_o is local free energy density, κ_C, κ_i^{α} and κ_i^{β} are the gradient energy coefficients, and p and q represent the number of orientation field variables for the α and β phases, respectively.

The kinetics of microstructural evolution is described by the temporal evolution of the field variables by numerically solving the coupled time-dependent Ginzburg-Landau (TDGL) and Cahn-Hilliard (C-H) [7] equations,

$$d\eta_i^{\alpha}(r,t)/dt = -L_i^{\alpha}(\delta F/\delta \eta_i^{\alpha}(r,t)), \qquad i = 1, 2, \ldots, p, \tag{3a}$$

$$d\eta_i^{\beta}(r,t)/dt = -L_i^{\beta}(\delta F/\delta \eta_i^{\beta}(r,t)), \qquad i = 1, 2, \ldots, q, \tag{3b}$$

$$dC(r,t)/dt = \nabla\{L_C \nabla[\delta F/\delta C(r,t)]\} \tag{3c}$$

where L_i^{α}, L_i^{β} and L_C are kinetic coefficients related to grain boundary mobilities and atomic diffusion coefficients, t is time, and F is the total free energy given in equation (2).

Temporal Evolution of a Two-Phase Microstructure With Quadrijunctions

In order to study the stability of a two-phase microstructure with only quadrijunctions, we construct the following free energy density function,

$$f_o = f(C) + \sum_{i=1}^{p} f(C,\ \eta_i^{\alpha}) + \sum_{i=1}^{q} f(C,\ \eta_i^{\beta}) + \sum_{\alpha}\sum_{\beta}\sum_{i=1}^{p}\sum_{j=1}^{q} f(\eta_i^{\alpha},\ \eta_j^{\beta}) \tag{4}$$

where,

$$f(C) = -(A/2)(C - C_m)^2 + (B/4)(C - C_m)^4 + (D_{\alpha}/4)(C - C_{\alpha})^4 + (D_{\beta}/4)(C - C_{\beta})^4$$

$$f(C,\ \eta_j^{\alpha}) = -(\gamma_{\alpha}/2)(C - C_{\beta})^2(\eta_i^{\alpha})^2 + (\delta_{\alpha}/4)(\eta_i^{\alpha})^4$$

$$f(C,\ \eta_j^{\beta}) = -(\gamma_{\beta}/2)(C - C_{\alpha})^2(\eta_i^{\beta})^2 + (\delta_{\beta}/4)(\eta_i^{\beta})^4$$

$$f(\eta_i^{\alpha},\ \eta_j^{\beta}) = (\varepsilon_{ij}^{\alpha\beta}/2)(\eta_i^{\alpha})^2(\eta_j^{\beta})^2$$

where η_i^α, η_i^β are orientation field variables, C is the local composition, C_α and C_β are the equilibrium compositions of α and β phases, $C_m = (C_\alpha + C_\beta)/2$, A, B, D_α, D_β, γ_α, γ_β, δ_α, δ_β and $\varepsilon_{ij}^{\alpha\beta}$ are phenomenological parameters. It can be shown that f_o has p degenerate minima with equal depth located at $\eta_1^\alpha, \eta_2^\alpha, \ldots, \eta_p^\alpha = (1,0,\ldots,0), (0,1,\ldots,0), \ldots, (0,0,\ldots,1)$ in p-dimension orientation space at the equilibrium composition C_α, and has q degenerate minima located at $(\eta_1^\beta, \eta_2^\beta, \ldots, \eta_q^\beta) = (1,0,\ldots,0), (0,1,\ldots,0), \ldots, (0,0,\ldots,1)$ at C_β. This requirement ensures that each point in space can only belong to one crystallographic orientation of a given phase.

The parameters in the free energy density function and the gradient energy coefficients are chosen in such a way that grain boundary and interphase boundary energies satisfy the so-called "double" wetting condition, $R_\alpha^2 + R_\beta^2 \geq 4$, in which no trijunctions are stable and the only stable interfaces are the α/β interphase boundaries [1]. In the following computer simulation, the following numerical values are employed: $C_\alpha = 0.05$, $C_\beta = 0.95$, Cm = 0.5, A = 1.0, B = 4.94, Da = Db = 6.09, $\gamma_\alpha = \gamma_\beta = 2.47$, $\delta_\alpha = \delta_\beta = 1.0$, and $\varepsilon_{ij}^{\alpha\beta} = 7.0$. Using these values, R_α and R_β are found to be $R_\alpha = R_\beta = 2.1$. We assumed that $L_\eta^\alpha = L_\eta^\beta = 1.0$ and $L_C = 0.5$. The coupled TDGL and CH equations are numerically solved using the simple explicit Euler technique in two-dimensions with 256 × 256 or 512 × 512 grid points and with periodic boundary conditions applied along both directions. The time step and grid size are chosen to be 0.1 and 2.0, respectively. The initial condition is generated by assigning small random values to all the orientation field variables and the overall average composition to the composition variable at each discrete lattice point.

The microstructural evolution of a two-phase solid with 50% volume fraction is shown in Fig. 1. It can be seen that, the microstructures are comprised only of quadrijunctions $\alpha\beta\alpha\beta$ and α/β interphase boundaries. The angles of the quadrijunctions vary within a certain range, i.e., there is no thermodynamically fixed angle for quadrijunctions. The above observations seem to be consistent with Cahn's thermodynamic predictions even though his analysis was made on a system in which the volume fractions are nonconserved [1].

Topological Transformations

The topological transformations during grain growth can be directly obtained from the temporal microstructures generated from the computer simulations. It is observed that two-sided grains (grains surrounded by two quadrijunctions) and three-sided grains (grains surrounded by three quadrijunctions) can directly undergo vanishing during grain growth of a two-phase solid with only quadrijunctions. The vanishing of a two-sided grain results in the disappearance of two quadrijunctions and the appearance of a new quadrijunction while the two former grains neighboring to the vanishing grain lose one quadrijunction each. The vanishing of three quadrijunction grains leads to the formation of a hexajunction (a junction at which six grains, with three of each phase, meet). However, a hexajunction is highly unstable in a two-phase solid and it quickly splits into two new quadrijunctions. As a result, each of the two adjacent grains of the other

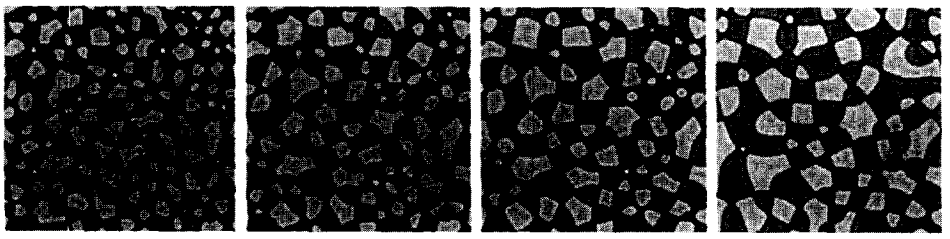

Figure 1. The microstructural evolution in all quadrijunction microstructures ($R_\alpha = R_\beta = 2.1$). The volume fraction of α phase is 50%. (a) t = 5000; (b) t = 10000; (c) t = 30000; (d) t = 50000.

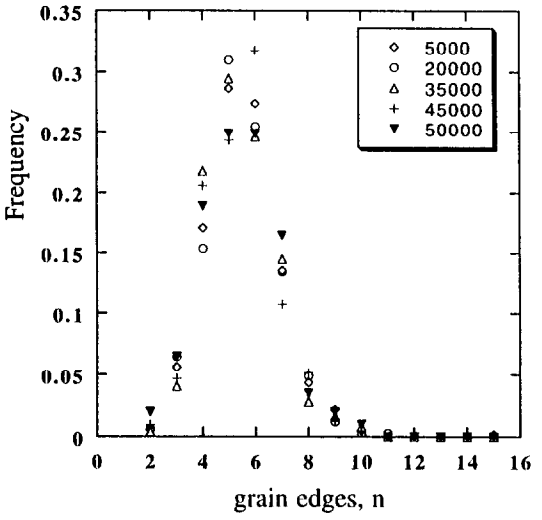

Figure 2. The time dependency of topological distributions of α phase in all quadrijunction microstructures. The volume fraction of α phase is 50%.

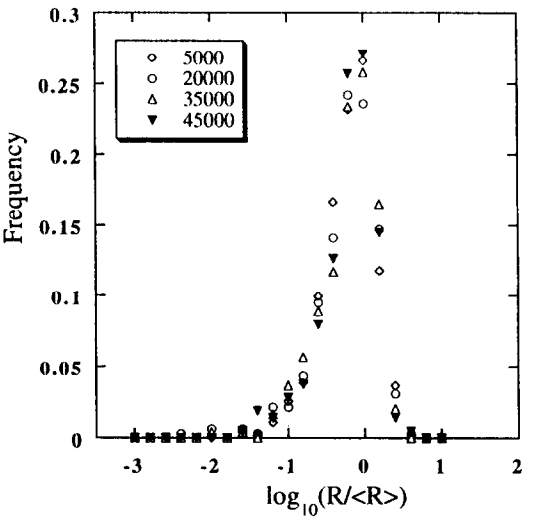

Figure 3. The time dependency of grain size distributions of α phase in all quadrijunction microstructures. The volume fraction of α phase is 50%.

phase loses one interface and one quadrijunction; one of the same phase grains gains one interface and one quadrijunction; and other grains remain unchanged in terms of the topology.

The time dependence of topological distributions is shown in Fig. 2. It can be seen that the shapes of these distributions do not change with time in the steady-state, i.e., the topological distribution is time-

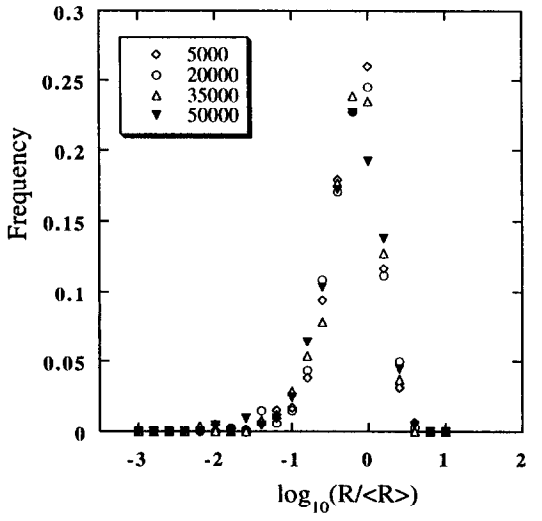

Figure 4. The time dependency of grain size distributions of β phase in all quadrijunction microstructures. The volume fraction of β phase is 50%.

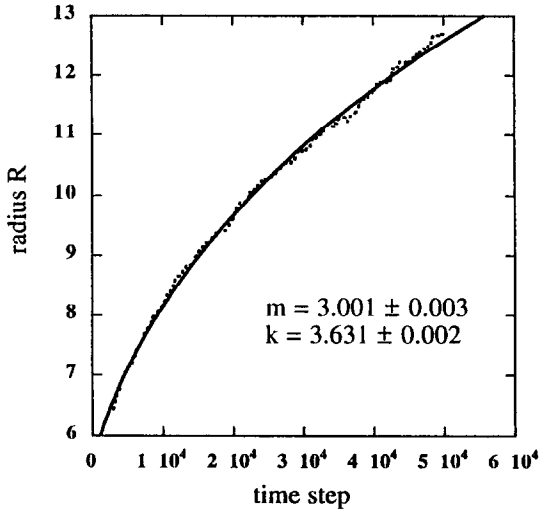

Figure 5. The time dependency of the average grain size of α phase in all quadrijunction microstructures. The volume fraction of α phase is 50%. The dashed line is simulation data and the solid line is a fitting into equation $R^m_t - R^m_0 = kt$.

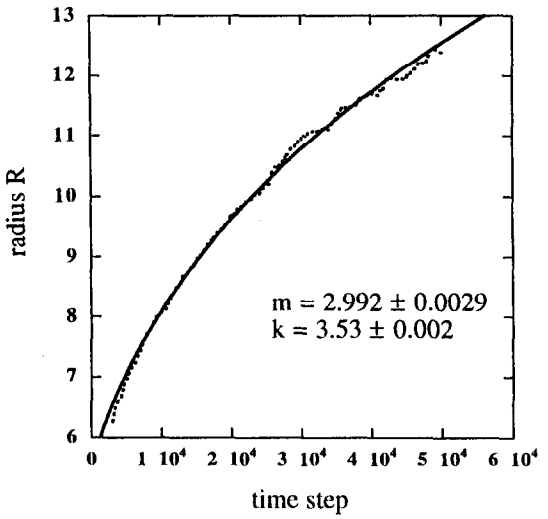

Figure 6. The time dependency of the average grain size of β phase in all quadrijunction microstructures. The volume fraction of β phase is 50%. The dashed line is simulation data and the solid line is a fitting into equation $R^m_t - R^m_0 = kt$.

invariant in a two-phase microstructure with only quadrijunctions, similar to the grain growth in single phase systems [5]. However, in a single-phase system with trijunctions, 2-sided grains never appear whereas in Fig. 2 there are quite a few two-sided grains whose occurrence is due to the fact that grain growth in a two-phase solid with conserved volume fractions is controlled by long-range diffusion, not just by the mean curvature as in single-phase systems. As a result, the avearge number of grain edges in a two-dimensional two-phase solid will not be 6 per grain, which comes from the requirement of space filling and balance of surface tension for a single-phase system. Actually, the average number of grain edges varies with the energetic conditions, i.e., values of R_α and R_β. The average number of grain edges obtained from the microstructures in Fig. 1 is about 5.92 per grain.

Time-Dependence of Grain Size and Size Distributions

The time dependencies of grain size distributions for α and β phases corresponding to the microstructures in Fig. 1 are shown in Figs. 3 and 4 respectively. It can be seen that the distributions for α and β phases are almost identical and the peaks of size distributions for both phases occur at average size ($\log_{10}(R/\langle R\rangle) = 0.0$) position. The shape of size distributions is independent of time, indicating that this system has reached the dynamic steady state after 5000 time steps. The average grain size for the two phases are shown in Figs. 5 and 6, respectively. In contrast to the freezing of grain growth predicted in the Potts model simulations for a two-phase solid with non-conserved volume fractions [2], it is shown that the average grain sizes increase with time and follows the power growth law $R_t^m - R_0^m = kt$ with m = 3, indicating the long-range diffusion controlled coarsening (Figs. 5 and 6).

Conclusions

The stability and evolution of quadrijunctions in a two-dimensional two-phase solid with conserved volume fractions were investigated by computer simulations based on a diffuse-interface field kinetic model.

It is shown that under certain thermodynamic conditions quadrijunctions can be stable in a two-phase solid. The kinetic simulations show that 2-sided grains can be stable in a two-phase solid with conserved volume fractions and the average number of grain edges is less than six. The topological transformations in a two-phase microstructure with only quadrijunctions is dramatically different from those in a single-phase system or a two-phase with only trijunctions. It was shown that in the steady state, both the shape and size distributions of each phase are invariant with time, whereas the average grain radius for each phase with time was found to follow $t^{1/3}$ law, implying the long-range diffusion controlled grain growth in a two-phase solid with stable quadrijunctions and with conserved volume fractions.

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