SAND 96-1149C

CONF-9606237-1

SIMULATION OF ANISOTROPIC GRAIN GROWTH BY OSTWALD RIPENING

Veena Tikare¹ and James D. Cawley²

¹Sandia National Laboratories
Albuquerque, NM 87110-1411

²Case Western Reserve University
Cleveland, OH 44106

ABSTRACT

In this paper, a two-dimensional Potts model that can simulate anisotropic grain growth by Ostwald ripening will be presented. The model defines a Wulff shape for the grains and allows each individual grain to grow in its local environment which is controlled by the solid/liquid interfacial energy, the spatial distribution of neighboring grains, area fraction of grains, wetting by and distribution of the liquid matrix, and the concentration gradients in the liquid. The results of this simulation technique will be presented with emphasis on the kinetics and grain shape evolution and will be compared to those of isotropic grain growth. Finally, the limitations of the Potts model in such microstructural evolution processes will be discussed.

INTRODUCTION

In fully wetting systems, after densification, grain growth occurs by the solution - reprecipitation mechanism and is therefore appropriately considered in the context of Ostwald ripening. Ostwald ripening is the increase in average grain size driven by the reduction in interfacial energy between the grains and surrounding matrix by diffusion through the matrix phase. Modeling anisotropic grain growth by Ostwald ripening is the focus of this investigation.

Predictive modeling of microstructural evolution presents unique challenges because of the large number of thermodynamic, mechanistic and spatial variables that must be considered simultaneously. Analytic models make many simplifying assumptions to make

DISCLAIMER

**Portions of this document may be illegible
in electronic image products. Images are
produced from the best available original
document.**

the problem tractable. In spite of the many assumptions, analytic models have increased the understanding of microstructural evolution processes; however, they have failed to predict the details of the microstructure or to incorporate complexities such as anisotropy^{1,2,3,4}. Motivated by the desire to eliminate these assumptions, many simulation techniques have been developed^{5,6,7}. Among these is the Potts model, a statistical-mechanical simulation technique, which has proven to be robust for modeling a diverse set of microstructural evolution problems, including normal and abnormal grain growth^{8,9}, particle-inhibited grain growth¹⁰, grain growth in multiphase systems¹¹, Ostwald ripening¹² and sintering¹³. In this investigation, the Potts model was modified to simulate isotropic and anisotropic grain growth in liquid-phase-sintered ceramics by Ostwald ripening.

MODEL

The microstructure was represented digitally by discretization on a two-dimensional, square lattice with periodic boundary conditions in both the X- and Y-directions. This lattice was populated with a canonical ensemble of two types of entities, designated A and B. These entities may be thought of as meso-scale domains of a particular phase. Entity A was the primary constituent of the solid phase, and entity B of the liquid phase. The A-entities were allowed to assume one of Q degenerate states, $q_A \in [1, 2, \dots, Q]$ where $Q = 100$ for most simulations. Each state, q_A , is an arbitrary label for the purposes of tracking individual grains, and can be considered to be a particular crystallographic orientation. The B-entities could assume only one state, $q_B = -1$. The negative number has no significance; it simply makes computation easier. The grains are features consisting of contiguous sites of the same state, q_A .

The Potts model used in this investigation incorporated interfacial energy by defining interaction energies between neighboring sites. The Hamiltonian, H , for the simulation and was calculated as the sum of the interaction energy between each site, i , and each of its first and second nearest neighbors, j ,

$$H = \frac{1}{2} \sum_{i=1}^N \sum_{j=1}^8 E(q_i q_j) \quad \text{eq. (1)}$$

where N was the total number of sites, E was the bond energy between neighboring sites in arbitrary units of energy and was a function of the state of the two neighboring sites, and q_i was the phase and orientation of the entity occupying site i . The Hamiltonian had units of energy, the same as that for E , the bond energy. The bond energy, E , was defined so that only neighbors of different states have a bond energy.

The solid-liquid (A-B) interfacial energy, γ_{sl} , and the grain boundary energy between two solid grains (A_i - A_j) of different orientation, γ_{ss} , were defined by their respective bond energies. A fully miscible liquid was considered in the simulation; thus, liquid-liquid interfaces were unnecessary in the simulation. Assigning only one state to the B-sites ensured that no liquid-liquid interfaces could occur. The interfacial energies, γ_{sl} and γ_{ss} , can be calculated as follows:

$$\gamma_{qiqj} = \frac{E_{qiqj} N_{qiqj}}{L_{qiqj}} \quad \text{eq. (2)}$$

where E_{qiqj} , the bond energy, is a function of neighboring phases and states, q_i and q_j , N_{qiqj} is the number of the unlike bonds at the interface, and L_{qiqj} is the length of the interface. The units of interfacial energy are in energy per unit length rather than energy per unit area because two-dimensional space is being used for this simulations. A consequence of eq. (2) is that the ratio of γ_{sl} to γ_{ss} can be varied by varying the ratio of E_{AB} to E_{AA} . The use of a square lattice to digitize the microstructure introduced some anisotropy to the interfacial energies. For isotropic grain growth simulations, both first and second nearest-neighbor interactions were considered to minimize lattice anisotropy effects as shown by Holm et al¹⁴. For anisotropic grain growth, anisotropy was introduced by making the bond energies directionally dependent. Half of the A-entities, even numbered q 's, were given high bond energies in the Y-direction and low bond energies in the X-direction; and vice versa for A-entities with odd numbered q 's. Furthermore, only first nearest neighbors were considered in eq. (1).

Transport was simulated by exchanging neighboring sites with each other. Conserved dynamics were used to maintain constant composition, i.e. fixed number of A- and B-sites. The step-by-step simulation technique used is reviewed below:

1. The two-dimensional, square lattice was populated with the desired composition (fraction of A-sites and B-sites). The lattice was initialized by placing the A- and B-sites at randomly selected lattice sites. The A-sites were assigned random q -values, between $q_A = 1$ and Q , and all B-sites were assigned $q_B = -1$
2. A lattice site was chosen at random, then one of its neighboring sites was chosen at random from its eight first- and second-nearest neighbors.
3. If one site was an A-site and the other a B-site, the simulation continued to step 4; if not, then to step 7. If both were A-sites, the exchange was not permitted because solid state diffusion was not allowed. If both sites were B-sites, the exchange does not change the microstructure; therefore consideration of this exchange was unnecessary.
4. A new orientation for the A-site was chosen at random from Q ($=100$) possible orientations.
5. The new configuration, A- and B-sites exchanging places with the A-site assuming the new orientation, was considered and the change in energy for this configuration was calculated using the Hamiltonian given in eq. (2).
6. The classical Metropolis algorithm¹⁵ was used to determine if the new configuration was accepted or rejected. The Metropolis algorithm uses Boltzmann statistics to determine the exchange probability, ω , as follows:

$$\omega = \exp\left(-\frac{\Delta H}{k_B T}\right) \quad \text{for } \Delta H > 0$$

and

$$\omega = 1 \quad \text{for } \Delta H \leq 0$$

eq. (3)

where k_B is the Boltzmann constant, and T is absolute temperature and has units of bond energy divided by the Boltzmann constant. As is conventional for Monte Carlo simulations, all temperatures are given as $k_B T$ with units of energy rather than just temperature.

7. The number of iterations is incremented by $1/N$, where N is the total number of sites and the program is returned to step 2. The number of iterations is given in units of Monte

Carlo step, MCS. At 1 MCS, the number of attempted exchanges was equal to the total number of sites in the simulation space. It has been shown that MCS scales linearly with time by some constant, τ , for such simulations^{16,12} thus MCS may be considered a measure of time in arbitrary units.

RESULTS AND DISCUSSION

The microstructures of isotropic and anisotropic grain growth simulations are shown in figures 1 and 2. The grain morphologies of the two are very different. In the isotropic simulations, the grains appear regular and equiaxed. In the anisotropic simulations, half the grains are elongated in one direction and rest in the perpendicular direction, since the variation in bond energies is restricted to these two directions. These highly elongated grains are faceted on the edges parallel to the high bond energy direction. The elongated and faceted grains persist throughout the grain growth simulations.

Figure 1. Isotropic grain growth simulation microstructures at time, $t = 4, 35$ and 100 KMCS.

Figure 2. Anisotropic grain growth simulation microstructures at time, $t = 29, 52$ and 100 KMCS.

The data from these simulations are plotted as average grain size versus time on logarithmic scales in figures 3 and 4, where grain size is the average grain radius. The anisotropic grains size is also given as the average radius of circular grains of equivalent area. Although, this seems a non-physical measure, it was chosen as the best one to allow comparison to both isotropic grain growth simulations and predictions by analytical models. The grain growth curves for the anisotropic simulations exhibit two distinct regions; the first is a non-linear region and the second is a linear region indicating power law

behavior. Examination of the simulated microstructures revealed that during the initial, non-linear region, grains are nucleated and solid precipitates onto nuclei from a supersaturated liquid. During precipitation all grain grow, but during the second stage, grains grow by Ostwald ripening, some grains grow at the expense of others. It is this Ostwald ripening regime that is of interest in this investigation.

Figure 3. Isotropic grain growth curves.

Figure 4. Anisotropic grain growth curves.

Ostwald ripening theories predict power law behavior as given by the following equation:

$$R^n - R_0^n = Kt \quad \text{eq. (4)}$$

where R is the average grain radius at time $t > 0$, R_0 is the initial grain size at time $t = 0$, K is the grain growth constant and n is the grain growth exponent. The data do indicate power law behavior with the inverse slope in the Ostwald ripening regime equal to n . The grain growth exponent, n , increased with increasing solid fraction as seen in figure 4. This behavior was in sharp contrast with the isotropic grain growth behavior, which has constant n as shown in figure 3, and with Ostwald ripening theories which also predict constant n , when the grain growth mechanism remains the same.

To understand the origin of varying n , we considered how the grain lengths and widths grew during Ostwald ripening. The grain length was measured as the second moment around the axis going through the center of mass in the direction of low bond energy and grain width was the same except in the direction of high bond energy, as defined by the following relationship:

$$I_L = \frac{\sum_{i=1}^a (X_i - X_c)^2 + \frac{1}{12}}{a} \quad I_W = \frac{\sum_{i=1}^a (Y_i - Y_c)^2 + \frac{1}{12}}{a} \quad \text{eq. (5)}$$

where I_L and I_W are the second moments around the low bond energy direction and high bond energy direction, respectively, and are used as measures of grain length and width, respectively. X_c is the location of the axis of low energy through the center of mass, $X_i - X_c$ is the distance of the site i from that axis, and a is the total number of sites in the grain. Similarly, Y_c is the location of the axis of high energy through the center of mass and $Y_i - Y_c$ is the distance from that axis.

The average length moment and width moment in the Ostwald ripening regime are plotted in figure 5 as a function of time on logarithmic scales for different solid contents. Again, power law behavior is observed for both moments at all three solid contents as indicated by the linear relationship observed in these plots. However, the exponent of the power law was different for

Figure 5. Moments of inertia as a function of time during anisotropic grain growth at different liquid contents.

each moment at the different solid fractions. All three simulations exhibited higher powers for the width moment than the length moment suggesting that the grains grew wider faster than they grew longer. Furthermore, the rate of grain widening increased with decreasing solid fraction. These observations suggested that the shape as characterized by the anisotropy of the grains was changing both with time and solid fraction.

Figure 6. Anisotropy of grains during grain growth.

This was confirmed by determining the anisotropy of the grains; anisotropy, A , was quantified as the ratio of the two moments as given by:

$$A = \frac{I_L}{I_W} \quad \text{eq. (6)}$$

Anisotropy of grains is plotted as a function of logarithmic time in figure 6. Anisotropy decreased with increasing time and decreasing solid fraction. This is consistent with the microstructures shown in figure 2 which show that initially long grains form. These grains

are constrained in the long direction by grains elongated in the perpendicular direction, but almost never by grains elongated in the parallel direction. These initially long grains, which are constrained along their length, widen but remain constrained along their length, thus anisotropy decreases.

The change in grain growth exponent, n , with solid fraction may be due the differing constraints in the two different directions for the various solid contents. In the isotropic grain growth case, the relative ease of growth in any direction is constant, but not so for the anisotropic case. The constraint is changing which in turn changes the rate of growth in the different directions. At high solid fractions the constraint along the width of the grains is high thus growth in this direction lags behind growth in the length direction as seen in figure 7.

Figure 7. Anisotropic grain growth microstructure at time $t = 100$ KMCS for liquid content = 14, 29 and 43%.

The decrease in anisotropy with decreasing grain fraction was unexpected. In silicon nitride ceramics which have anisotropies similar to the one used in this study it was found that at higher liquid contents grains were more elongated than at lower liquid contents. This increase in anisotropy with decreasing grain fraction was attributed to less constraint at lower grain fractions. The contrary behavior in the simulations is a consequence of the limitation of the grain anisotropy to two directions and to two dimensions.

The restriction of anisotropy to two directions is similar to rafting in metals where elastic stress field constrains growth of precipitates in direction aligned with the applied stress. It is also similar to precipitation of a second phase within grains such that the precipitation is aligned in particular crystallographic direction of the parent crystal. These are observed in many martensitic systems such as MgO partially stabilized zirconia.

Conclusions

Anisotropic grain growth by Ostwald ripening can be simulated by the Potts model by assigning different bond energies between neighboring sites along the different lattice directions. The grain growth exhibited power law behavior, but the grain growth exponent was not constant; it increased with decreasing liquid content. Grain anisotropy decreased with increasing grain size and decreasing liquid.

Acknowledgment:

This work was supported by the U.S. Department of Energy under contract number DE-AC04-95AL85000.

- ¹ I.M. Lifshitz and V.V. Slyozov, "The Kinetics of Precipitation From Supersaturated Solid Solutions," *J.Phys. Chem. Solids*, 19 [1-2] 35-50 (1961).
- ² C. Wagner, "Theory of the Ageing of Precipitates by Redissolution," *Zeitschrift fur Elektrochemie*, 65 [7-8] 581-591 (1961).
- ³ P.W. Voorhees and M.E. Glicksman, "Solution to the Multi-Particle Diffusion Problem with Application to Ostwald Ripening - II. Computer Simulations," *Acta Metall.* 32 [11] 2013-2030 (1984).
- ⁴ R.T. DeHoff, "A Geometrically General Theory of Diffusion Controlled Coarsening," *Acta Metall.* 39 [10] 2349-2360 (1991).
- ⁵ K.W. Mahin, K. Hanson and J.W. Morrid Jr., "Comparative Analysis of the Cellular and Johnson-Mehl Microstructures through Computer Simulations," *Acta Metall.* 28 443-453 (1980).
- ⁶ E.A. Ceppi and O.B. Nasello, "Computer Simulation of Bidimensional Grain Growth," *Scripta Metall.* 18 1221-1225 (1984).
- ⁷ A. Soares, A.C. Ferro and M.A. Fortes, "Computer Simulation of Grain Growth in a Bidimensional Polycrystal," *Scripta Met.* 19 1491-1496 (1985).
- ⁸ D.J. Srolovitz, M.P. Anderson, P.S. Sahni and G.S. Grest, "Computer Simulation of Grain Growth - I. Kinetics," *Acta Metall.*, 32 793-802 (1984).
- ⁹ D.J. Srolovitz, G.S. Grest, and M.P. Anderson, "Computer Simulation of Grain Growth - V. Abnormal Grain Growth," *Acta Metall.*, 33 [3] 2233-2247 (1985).
- ¹⁰ E.A. Holm and D.J. Srolovitz, "Effects of Inert Particle Morphology on Grain Growth in Composites," *Proc. of Morris E. Fine Symp.*, edited by P.K. Liaw, J.R. Weertman, H.L. Marcus and J.S. Santner, Minerals, Metals and Materials Soc. 187-192 (1991).
- ¹¹ E.A. Holm, D.J. Srolovitz, J.W. Cahn, "Microstructural Evolution in Two-Dimensional Two-Phase Polycrystals," *Acta Metall.*, 41 [4] 1119-1136 (1993).
- ¹² V. Tikare, Numerical Simulation of Grain Growth in Liquid Phase Sintered Materials, Case Western Reserve Univ., Cleveland, OH (1994).
- ¹³ G.N. Hassold, I-W Chen, and D.J. Srolovitz, "Computer Simulation of Final-Stage Sintering: I. Model, Kinetics, and Microstructure," *J. Am. Ceram. Soc.* 73 [10] 2857-2864 (1990).
- ¹⁴ E.A. Holm, J.A. Glazier, D.J. Srolovitz and G.S. Grest, "Effects of Lattice Anisotropy and Temperature on Domain Growth in the Two-Dimensional Potts Model," *Phys. Rev.*, A 43 2662-2668 (1991).
- ¹⁵ N. Metropolis, A.W. Rosenbluth, M.N. Rosenbluth, A.N. Teller and E. Teller, "Equation of State Calculations by Fast Computing Machines," *J. Chem. Phys.*, 21 1087-92 (1953).
- ¹⁶ Y. Limoge and J.L. Bocquet, "Monte Carlo Simulation in Diffusion Studies: Time Scale Problem," *Acta Metall.*, 36 [7] 1717-1722 (1988).

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.