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INHIBITION OF GRAIN GROWTH BY SECOND PHASE PARTICLES: THREE DIMENSIONAL MONTE CARLO COMPUTER SIMULATIONS

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Introduction

Hillert (1) recently reviewed the analytical models for particle inhibition of grain growth and concluded that the ratio of the limiting mean grain radius, R_L, to particle radius, r, should vary with volume fraction, f, of the particle as:

$$R_L/r = 4/(9f^{0.93}) \quad (1)$$

This result is close to the classic Zener-Smith value (2):

$$R_L/r = 2/(3f) \quad (2)$$

Both these results are derived under the assumption that the density of particles per unit area of grain boundary is that given by random intersections of the grain boundaries with the particles.

Computer simulation studies of two dimensional (2-D) particle inhibited grain growth by Srolovitz, Anderson, Grest and Sahni (3), however, reported a very different result. The limiting grain size was found to be:

$$R_L/r = 1.7/f^{0.5} \quad (3)$$

Not only was the f dependence different from that predicted by the Zener analysis, but a very much higher fraction, ϕ, of particles in contact with the grain boundaries was found than would be expected if the intersections between particles and boundaries were random (4).

It is readily shown (3,4) that the Zener analysis gives essentially the same prediction as Eq. 2 when applied to 2-D grain structures. Srolovitz et al. (3) and Hillert (1), however, recognized that in 2-D particles are especially effective in removing the grain boundary curvature that drives grain growth. Even an isolated 2-D grain would be stabilized against collapse if in contact with only three non co-linear particles - giving a triangular straight sided grain (1). This concept of loss of curvature when applied to the 2-D structures readily predicts a limiting grain size of the form of Eq. 3 (3). The limiting grain size given by Eq. 3 is also that predicted for both 2-D and 3-D grain growth, under the assumption that the fraction ϕ of particles on boundaries remains constant with increasing particle concentration (4). In 2-D the simulation results showed (3) that ϕ was approximately constant at a value near unity ($0.8 < \phi < 0.9$ for $0.005 \leq f \leq 0.05$).

Hillert (1) in his reanalysis of the interaction of particles with grain boundaries in 3-D suggested that such a high correlation between grain boundaries and particles would not be expected for 3-D grain growth. He modified the Zener analysis to give Eq. 1 by more detailed consideration of the interaction of a grain boundary with a single particle, as in

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an earlier paper (5), but left the density of particles on the boundaries at the Zener value. However, he also pointed out that if grains are pinned in 3-D with the majority of particles residing at grain corners then the limiting grain size should be:

$$R_L / r = 1.8/f^{1/3} \tag{4}$$

Hillert suggested that this might apply at high particle concentrations ($f \geq 0.1$).

The computer simulation method allows the details of the complex interaction between particles and grain boundaries to be explored. The present paper describes the extension of a recently developed 3-D computer model of normal grain growth (6,7) to include the effects of a random particle distribution (8). A full presentation of this analysis together with experimental evaluation will be reported in due course. However, the current results given here allow insight into the randomness of the interaction between particles and boundaries and the resulting functional dependence of the limiting grain size on particle volume fraction f.

Simulation Method

The procedure used in the simulation of grain growth in the presence of a particle dispersion is similar to that used in previous studies (3,6,7). A continuum grain structure is superimposed onto a simple 3-D cubic lattice containing N = 100 x 100 x 100 sites. Each lattice site is assigned a number corresponding to a particular grain orientation. A large number, Q, of different grain orientations is used so that grains of the same orientation rarely impinge on each other. A lattice site adjacent to sites with a different Q value are regarded as being part of the grain boundary, while a site surrounded by sites with the same Q value are regarded as being in a grain interior. The grain boundary energy is defined in terms of a lattice site energy:

$$E_i(k) \quad -J \sum_{j=1}^{N(k)} (1 - \delta_{S_i S_j}) \tag{5}$$

Here J is a positive constant, S_i corresponds to the orientation of site i ($1 \leq S_i \leq Q$), δ_{ab} is the Kronecker delta function, and the summation is taken over all j sites within a neighbor shell k of site i. In the present 3-D simulations, Q = 48 and k = 3 were used (7). The kinetics of boundary motion were simulated by a Monte Carlo technique in which a site is selected at random and reoriented to a randomly chosen orientation between 1 and Q. If the change in energy due to the reorientation, ΔE, is less than, or equal to, zero, then the reorientation is accepted. N_p second phase particles of size one lattice site were incorporated by randomly selecting sites at the start of the simulation and assigning them a

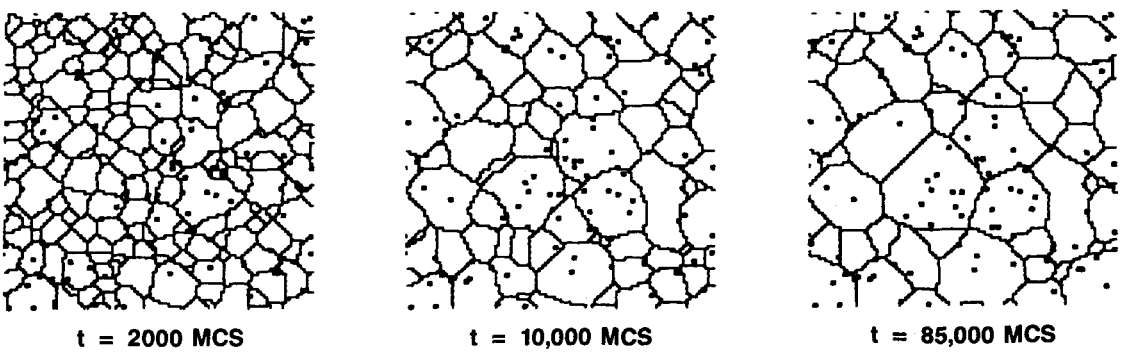

FIG. 1. Cross-section from the 3-D microstructure as a function of time for f = 0.01.

special orientation, Q+1. The particle matrix interfacial energy was set at the same value as the grain boundary. No attempt was made to reorientate the particle sites and they were not allowed to move. As in the grain growth study (7) the simulations were started by randomly assigning one of the Q orientation to each of the remaining ($N-N_p$) sites. That is, the initial grain size was set equal to the particle size and the number of grains is of the same order of magnitude as the number of sites in the system. Time, in the simulations, is proportional to the number of re-orientation attempts. ($N-N_p$) re-orientation attempts was used as the unit of time and referred to as one Monte Carlo step(MCS).

Results

Simulations of grain growth in the presence of a particle dispersion were carried out for particle concentrations, f, of 0.005, 0.01, 0.02, 0.04, 0.08 and 0.16. Owing to the large amount of computer time required, only two simulations were performed at each composition. Example cross-sections for grain growth are shown in Fig. 1 as a function of time for f = 0.01. For all concentrations examined, it was observed that the mean grain volume initially increased with time but eventually reached a limiting value. The mean grain volume, V, as a function of time in MCS is plotted in Fig. 2 for different values of f. The single phase results, f = 0.0, are those previously reported by Anderson et al. (7). Note

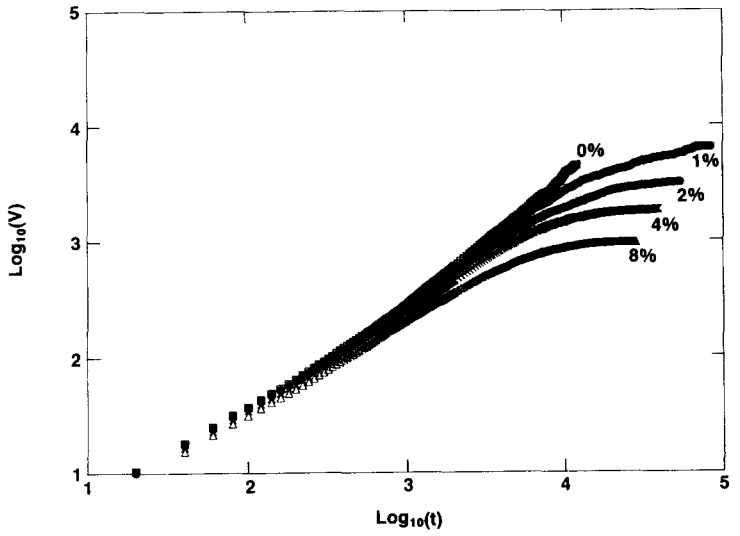

FIG. 2.

Mean grain volume V versus time for different particle concentrations.

that the initial growth rate is almost unaffected by increasing f - not surprisingly since most of the small grains are not in contact with even one particle. As the grain size approaches, and passes the nearest neighbor particle spacing, the growth slows down and becomes fully inhibited at longer times giving the pinned grain structures seen in Fig. 3.

The total fraction, ϕ, of particles on grain boundaries in the pinned structures as well as the fractions on faces between two grains, ϕ_2, on edges between three grains, ϕ_3, and at corners between four grains, ϕ_4, were determined during grain growth and in the pinned structures. Fig. 4 shows the results for the pinned structures. The total fraction, ϕ, and the individual fractions, ϕ_2 to ϕ_4, of particles on boundaries all increased significantly with the volume fraction, f. In particular, it was found that $0.41 \leq \phi \leq 0.64$ and $0.014 \leq \phi_4 \leq 0.075$ as $0.01 \leq f \leq 0.08$. These values are somewhat smaller than those reported for ϕ in the 2-D simulations where ϕ was close to 1. Furthermore, ϕ shows a much stronger dependence on f than was seen in 2-D (3,4).

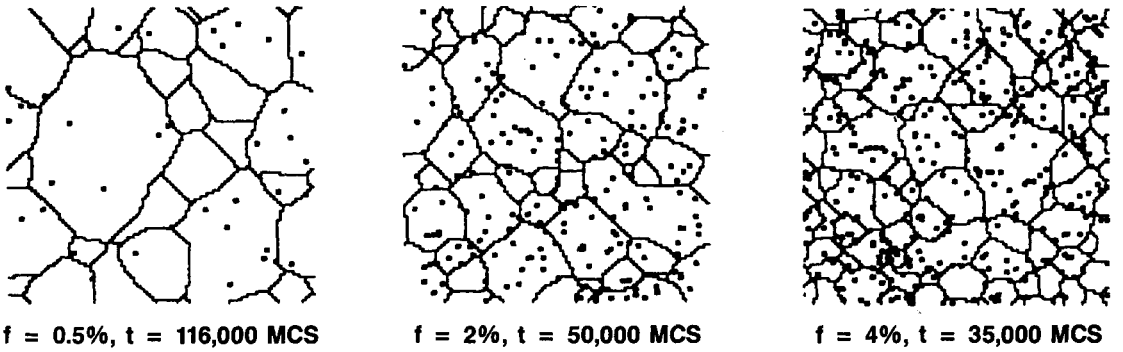

FIG. 3. Pinned microstructures for different particle concentrations, f.

The final and crucial result is given in Fig. 5 where the limiting mean grain volume V_L, in numbers of lattice sites, is plotted against the particle fraction f on a log-log plot. The linear log-log dependence indicates that the following empirical relationship is found in the Monte Carlo simulations:

$$V_L / v = A/f^b \qquad (6)$$

Here v is the particle volume, which is one lattice site, and A and b are constants. A least squares fit to the data gives A = 91.0 ± 16.9 and b = 0.922 ± .045. Assuming $V_L = 4\pi R_L^3/3$ and $v = 4\pi r^3/3$, then (6) reduces to

$$R_L / r = C/f^a = (4.5 \pm 0.8)/f^{(.31 \pm .02)} \qquad (7)$$

where R_L and r are the mean limiting grain radius and particle radius, respectively.

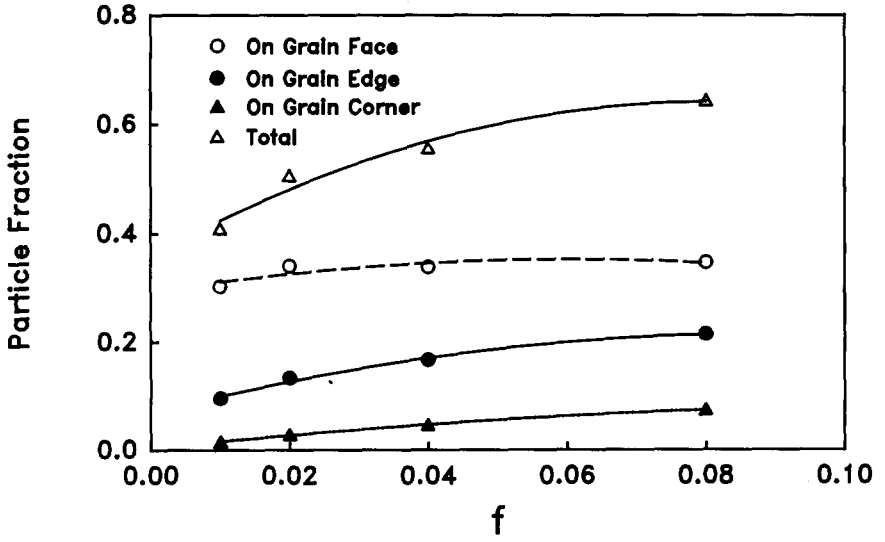

FIG 4.

Fraction of particles on grain faces, edges and corners in the pinned microstructure versus particle concentration, f.

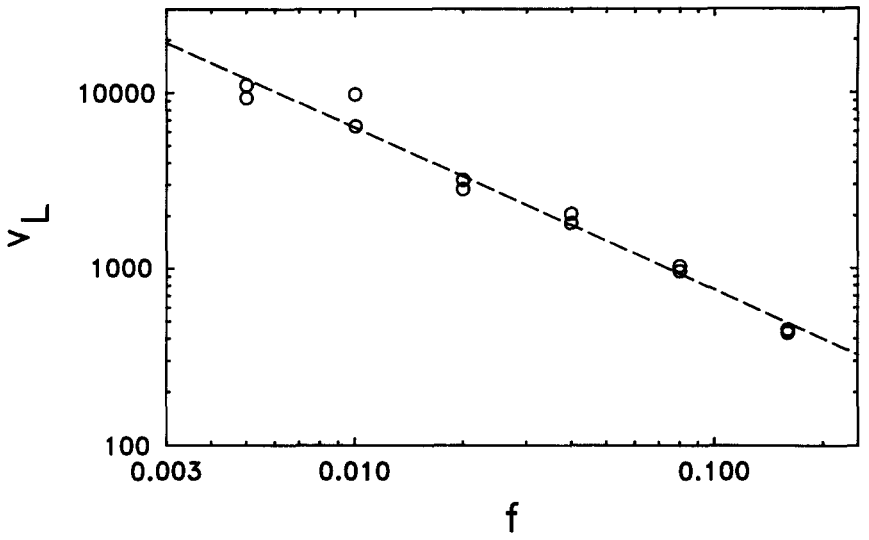

FIG. 5. Mean grain volume of the pinned microstructures versus particle concentration, f.

Discussion

The results of the 3-D computer simulations lead to two important conclusions. The first is that the total fraction of particles observed on the grain boundaries, ϕ, and the fraction of particles observed in grain corners, ϕ_4, of the pinned structures is significantly greater than that expected on the basis of random intersections. This was also found previously in 2-D (3). The expected value for ϕ and ϕ_4 in the limit of random intersections (Zener case) may be estimated employing the method of Doherty et al. (4) as

$$\phi_Z = 3r/R_L \tag{8}$$

and

$$\phi_{Z4} = 6r^3/R_L^3 \tag{9}$$

Eq. 9 may be derived by recognizing that a grain of mean volume $4\pi R_L^3/3$ will have $24/4 = 6$ corners. Each corner has a critical volume $4\pi r^3/3$ in which the center of the particle must lie if it is to be in contact with the corner. The volume of particles in contact with corners is then $f(4\pi r^3/3)N_c = \phi_4 N_p(4\pi r^3/3)$, where N_c is the total number of corners per unit volume and N_p is the total number of particles. By substituting $N_c=6/(4\pi R_L^3/3)$ and $N_p=f/(4\pi r^3/3)$, Eq. 9 results. At f=0.01, using R_L measured in the simulation (Eq. 7), ϕ_Z=0.16 and ϕ_{Z4}=0.0009 are obtained. Both these values are much smaller than the measured values $\phi = 0.41$ and $\phi_4 = 0.014$. Thus, although boundaries can in the simulations escape from particles (e.g., Fig. 1), a strong interaction occurs so that a higher than random correlation of particles with the grain interfaces is found even at low particle concentrations. Hillert predicted such an effect only at high values of f, f≥0.1

The second important result from the simulations is the much stronger inhibition of grain growth, Eq. 7, than expected under the Zener assumption, Eqs. 1-2, at small volume fraction f. This finding is consistent with experimental results compiled by Olgaard and Evans, who showed that the exponent a in the relationship $R_L/r = C/f^a$ varied between 0.3 to 1.0 for a wide range of systems, with most results below 0.5 (9). The simulation results also show the dependence on f expected by Hillert for high volume fractions, Eq. 4. In this case, however, the constant of proportionality generated by the simulation differs from that due to Hillert and there is no evidence for a crossover to a = 0.93 for small f.

More detailed study of the simulations and comparison with experimental results are needed to confirm these ideas and explore the conditions in which abnormal grain growth may occur. The initial results show, however, that the Monte Carlo simulations, in 3-D as in 2-D, are an important new tool for understanding the development of grain structures.

The authors wish to point out that after the work was completed and while this note was in preparation, we received the unpublished results of Oldershaw and Hazzledine (10). who carried out a similar simulation using a 3-D fcc lattice. Their results are in agreement with Eq. 7. These two studies were performed independently and it is nice to see that simulations on different lattices give similar results.

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