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COMPUTER SIMULATION OF RECRYSTALLIZATION IN THE PRESENCE OF SECOND PHASE PARTICLES

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Abstract

Recrystallization in the presence of inert second-phase particles has been simulated with a Monte Carlo technique. The technique is the same one as has been employed successfully to model grain growth, recrystallization and grain growth in the presence of second-phase particles. The results show that at low stored energies, the particles retard the growth of recrystallization nuclei and can prevent the recrystallization process from going to completion. At high enough stored energies, however, growth is not impeded such that the particles do not affect the kinetics of recrystallization. Post-recrystallization is strongly affected by the presence of particles because grain growth is inhibited and the microstructures tend to be less equiaxed than in grain growth.

Introduction

The effect of second phase particles on primary recrystallization has long been known to be profound. This topic has been extensively investigated since the paper of Doherty and Martin[1] and has been frequently reviewed, notably by Köster & Hornbogen[2] Hansen[3] and Humphreys[4]. An interesting feature of the topic is that the presence of particles can either retard or accelerate the kinetics of recrystallization. The reasons for this variable effect have to do with the competition between retardation of grain boundaries by particles and the nucleation of recrystallized grains. The key conclusions to be drawn from the reviews of the topic are as follows.

1) Acceleration of recrystallization occurs when large (>1μm) particles are present. Humphreys[5] has shown that large particles are surrounded by a deformation zone in which there is a higher stored energy and larger misorientations than in the matrix. These factors lead to a significantly higher probability of nucleation at the particle and higher growth rates, at least in the vicinity of the particle. For a sufficiently high particle density, smaller recrystallized grain sizes can be achieved than in the corresponding single-phase material. This phenomenon is known as particle stimulated nucleation (PSN) and is of great importance in commercial materials. It depends, however, on heterogeneities in the deformed microstructure that are not addressed in the simulations described in this paper and so will not be discussed further.

2) Retardation of recrystallization occurs in the presence of fine particles where the mean spacing (Δ_3) is of the order of the sub-grain size, typically <1μm. In this case the fine dispersion retards coarsening of the sub-grain structure which inhibits the nucleation of recrystallized grains.

3) The growth of recrystallized grains is not much affected by the presence of particles on any scale, suggesting that retardation of recrystallization is largely a consequence of the inhibition of nucleation. This is clear from measurements of growth rates by Doherty and Martin[1].

Doherty and Martin[1] showed that a fine, semi-coherent precipitate in an Al-Cu alloy inhibits recrystallization very effectively. In the Al-Al_2O_3 system, where the second phase is incoherent with the matrix, Nobili and Maria[6] showed a similar retardation of recrystallization. They also showed that the recrystallization temperature increased

with oxide content, i.e. that the retardation increased with ... of second phase. The retardation of recrystallization by fine particles is most likely to be the result of pinning of subgrain boundaries by the particles and the consequent prevention of nucleation. This view is supported by the experimental fact that when retardation occurs, but recrystallization goes to completion, the grain size is very coarse (compared with the single phase material), indicating that very few nuclei appeared. Some reviewers of the problem, for example Humphrey[4] and Baker & Martin[7] have used the Zener-Smith analysis to deduce that particle pinning does not produce enough drag to be the sole cause of retardation. The review of particle pinning above, however, shows that this may not be correct. It has been shown experimentally that dispersions of fine particles often produce more uniform, diffuse dislocation substructures, Humphreys[4], and fewer large local misorientations. This lack of large misorientations can clearly contribute to the decrease in nucleation density but is not addressed in this work.

The motivation for this work was previous successful simulations of recrystallization[8,9] and grain growth[10,11,12,13,14] especially the effects of particles on limiting grain growth[15]. The results to be presented here address the issue of retardation of recrystallization and largely confirm the second and third conclusions above. We studied the effect on recrystallization of three parameters, stored energy, nucleation density and particle fraction. Three different stored energies were used with the same numbers of particles and nuclei. Particles were superimposed on microstructures obtained by simulation of grain growth. Nucleation was imposed on the system at the start of recrystallization (site saturated nucleation) with various densities of nuclei. An additional purpose of this study was to study the extent to which the post recrystallization microstructure underwent grain growth.

Simulation Procedure

The simulation procedure employed in the present study is essentially identical to that employed in our previous studies of recrystallization in homogeneous materials[8,9]. In short, a continuum microstructure is mapped onto a two-dimensional triangular lattice containing N=40,000 sites. Each lattice site is assigned a number S_i which corresponds to the orientation of the grain in which it is embedded. Lattice sites which are adjacent to sites having different grain orientations are regarded as as being separated by a grain boundary, whilst a site surrounded by sites with the same orientation is in the grain interior. Each unlike pair of nearest neighbors is assigned an energy J so that the total energy of the system is calculated as

$$E = \frac{J}{2}\sum_{i}^{N}\sum_{j}^{nn}\left(1 - \delta_{S_i S_j}\right) \qquad (1)$$

where the sum on i is over all N sites in the system, the sum on j is over the nearest neighbor sites (nn=6) of site i, and δ_{ij} is the Kronecker delta. Grain growth is simulated by permitting the orientation of a particular site to change, provided that the total energy is reduced or left unchanged as a result of the change in orientation. The sites in the system are sampled in a random order and N such reorientation attempts is defined as one Monte Carlo Step (MCS). The number of Monte Carlo Steps is proportional to time. Particles are introduced into the simulation as sites which have an orientation different from any of the grains and which can not be reoriented during the course of the simulation. This assumption results in the equality of the particle-matrix interfacial energy and the grain boundary energy, which is reasonable for particles that are incoherent with the matrix.

Primary recrystallization in the presence of particles is modeled by incorporating an additional term into the energy expression Eq. (1) which accounts for the energy stored in the matrix, generally in the form of dislocations. This additional energy is present in the unrecrystallized grains but is absent in the recrystallized grains. The total energy E is then calculated as

$$E = \sum_{i}^{N}\left[H f(S_i) + \frac{J}{2}\sum_{j}^{nn}\left(1 - \delta_{S_i S_j}\right)\right] \qquad (2)$$

where the function f(S) is unity for unrecrystallized sites, and zero for recrystallized sites and particles. H is the stored energy per site due to deformation, which is assumed to be uniform over the initially unrecrystallized material. Recrystallization occurs by a nucleation and growth process. Nucleation is simulated by adding small (3 sites) nuclei to the material at random sites at the beginning of the simulation. These nuclei have S values, differing from those of all other grains and for which f=0. If H is too small the nuclei will be sub-critical and shrink away. The value of H required for growth of a nucleus depends on its surroundings. If H/J>2, an isolated nucleus is super-critical and can grow. If H/J<2, the nucleus must be adjacent to an existing boundary in order to survive and its growth tends to occur along the prior boundaries. This discussion shows that a better term would be embryo, as there is no guarantee that the "nucleus" is super-critical in these simulations.

The present simulations are initialized by performing grain growth simulations under the conditions described in Refs. 10 and 11 for a period of 10^3 MCS after which the microstructure contains approximately 1000 grains with

a mean grain area of approximately 40 sites. At this point, single site "particles" are randomly placed within the microstructure with area fractions between 1 and 10 percent. Also, at this time, 3 site nuclei are added to the system such that their area fraction is between 0.75 and 7.5 percent. These nuclei are better described as embryos because they may be sub-critical in the location that they happen to be placed, as discussed above. Simulations were performed with values of the magnitude of the stored energy relative to the grain boundary energy, H/J, of 0.1, 1.01 and 2.01. The simulations were typically run for 30,000 MCS and each data point on the plots below represent averages over 5 simulations, where each simulation had a different initial microstructure.

Results

In order to test the validity of the simulation procedure and to examine one of the simplest cases of primary recrystallization in the presence of second phase particles, a series of simulations were performed in which a relatively large, circular, recrystallized grain was introduced into a matrix of unrecrystallized grains. At low H/J ratios, the recrystallized grain rapidly became pinned and no further recrystallization took place. The only exception to this was for very low fractions of particles, <0.25%, where the single grain did grow without becoming pinned. At higher stored energies, H/J>1, the grain grows without pinning and the growth is unaffected by the particle fraction.

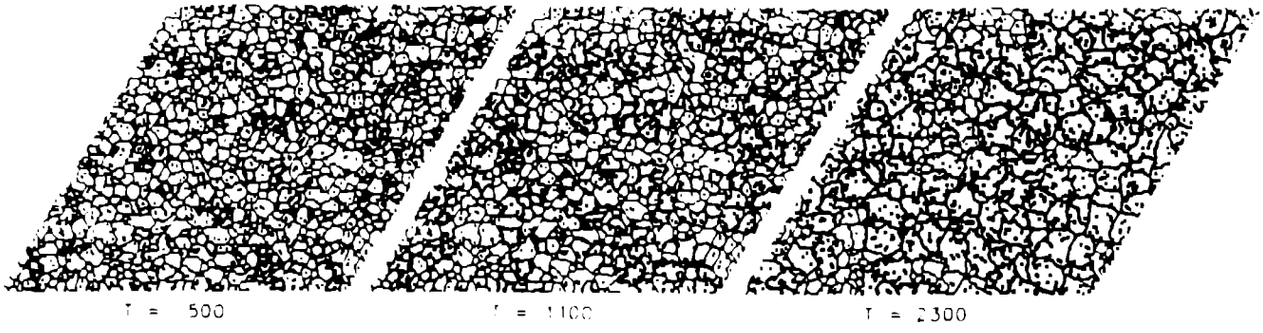

Fig. 1. Temporal evolution of microstructure for H/J=1.01, 200 nuclei at t=0 and 2000 particles (5% area fraction). Recrystallized grains are shaded.

Many of the simple results of the growth of a large, isolated recrystallized grain carry over to the more realistic simulations which include spatial distributions of small recrystallization nuclei. Fig. 1 shows the temporal evolution of such a microstructure for which the particle area fraction is 5%, the initial recrystallized nuclei area fraction is 1.5% and H/J=1. The recrystallized grains grow into the unrecrystallized matrix with a rough interface. The time dependence of the fraction of the sample which has recrystallized, F, is shown as a function of time for three different values of the stored energy, H/J (Fig. 2a). As in the simple test case described above, when H/J≥1 the recrystallized grains grow rapidly and complete recrystallization (F=1) is achieved at relatively early times (t<5000 MCS). On the other hand, for small H/J (=0.1) the recrystallized fraction quickly saturates at a low value and remains unchanged over very long times, i.e. the recrystallized grains are successfully pinned by the particles.

Further analysis of the growth kinetics shows that for both the H/J = 1 and 2 cases, an Kolmogorov-Johnson-Mehl-Avrami (KJMA) exponent of 2 is obtained, fig. 2b. This result is consistent with those obtained in the absence of a second phase dispersion. A plot of the logarithm of the recrystallized grain area <A> versus time (Fig. 2c) shows that the recrystallized grain size saturates at long times. For small H/J this is due to particles restricting the growth of the recrystallized grains into the unrecrystallized matrix. For large stored energy, the saturation of the recrystallized grain size occurs when recrystallization is complete and the dominant growth mode is simply grain boundary curvature driven grain growth. For the cases depicted in Fig. 2c, the grain size at the end of recrystallization is larger than would result if the grain size were determined by grain growth (in the absence of recrystallization) in the presence of a particle dispersion. Therefore, by the time recrystallization is complete, normal grain growth is entirely inhibited. In this way, the final microstructure is simply a picture of the system at the end of recrystallization and further annealing has essentially no effect on the microstructure (assuming no particle coarsening). This feature may be used to help explain why the final grain size for H/J=2 is smaller than that for H/J=1. At larger values of H/J more of the recrystallization nuclei (embryos) added at the beginning of the simulation are supercritical and grow than at lower stored energies. This was verified by examining the number of recrystallized grains as a function of time. Therefore since growth competition between recrystallized grains due to grain boundary curvature effects is negligible, the final recrystallized grain size is a measure of the inverse density of the supercritical nuclei.

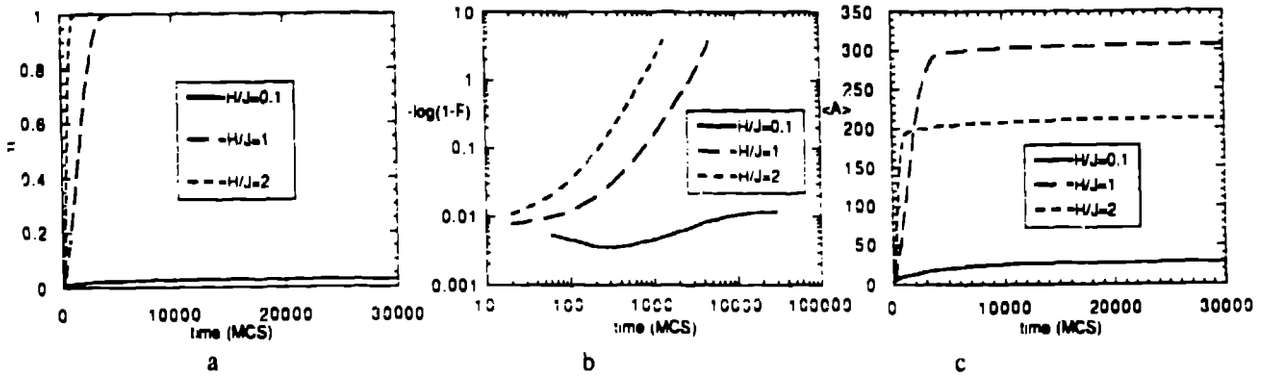

Fig. 2. Kinetics of recrystallization for 200 nuclei, 2000 particles (5% area fraction) and H/J= 0.1, 1.01 and 2.01, as noted on the figure; a) fraction recrystallized (F) versus time, b) JMAK plot and c) mean area of recrystallized grains (<A>) versus time.

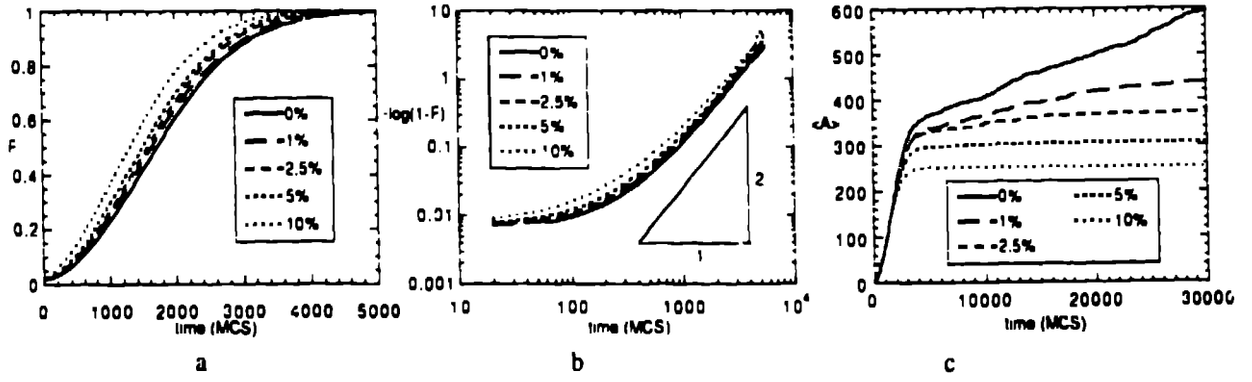

Fig. 3. Kinetics of recrystallization for H/J= 1.01, 200 nuclei and various particle fractions, as noted on figure; a) fraction recrystallized (F) versus time, b) JMAK plot and c) mean area of recrystallized grains (<A>) versus time.

The effects of particle density at fixed stored energy was investigated for particle fractions between 0% and 10%, Fig. 3. The results showed essentially no effect of particle density on the kinetics of recrystallization. The only noticeable effect was that higher particle density was associated with a smaller final recrystallized grain size, Fig. 3c. This slight effect occurs because the particles provide additional heterogeneous nucleation sites. At a stored energy of 1.01, a nucleus can only survive if it nucleates heterogeneously, i.e. on a boundary or particle[9]. It is also worth noting that for the smallest particle density (1%), some grain growth does occur when recrystallization is complete. If grain growth in the presence of a 1% particle dispersion occurred in the absence of primary recrystallization, the pinned grain size at a 1% particle density would be approximately 300 sites[12]. The grain growth at the end of primary recrystallization observed for a particle density of 1% is attributable to the fact that the grain size at that time is of the same order as the pinned grain size for normal grain growth in the presence of a particle dispersion.

The effects of variation in particle density at lower stored energies (where recrystallization does not go to completion) is much more pronounced. Figure 4 shows the temporal evolution of the microstructure for H/J=0.1 and a particle area fraction of 5%. Some growth of the recrystallized grains beyond particles is clearly observed, but unlimited growth as in Fig. 1 where the stored energy is large, does not occur. Figures 5a and 5c show the time dependence of the recrystallized fraction and the mean recrystallized grain area, respectively, for H/J=0.1 and particle area fractions between 0.25% and 10%. For the higher volume fractions, both the recrystallized grain size and the recrystallized fraction saturate at long times at values that increase with decreasing particle concentration. Analysis of the KJMA plot, Fig. 5b shows that the maximum slope is approximately 2 at small particle fractions but decreases with increasing particle density. For the lowest fraction (0.25%) however, recrystallization goes essentially to completion, albeit rather slowly compared to the simulations with higher stored energy. Again this seems reasonable in light of the fact that recrystallization will take place at H/J=0.1 in the absence of particles.

Fig. 4. Temporal evolution of microstructure for H/J=0.1, 200 nuclei at t=0 and 2000 particles (5% area fraction). Recrystallized grains are shaded.

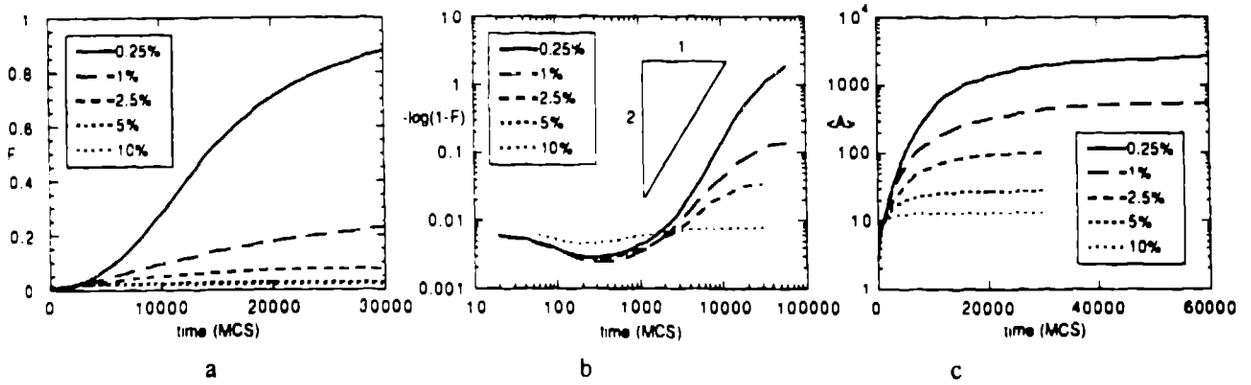

Fig. 5. Kinetics of recrystallization for 200 nuclei, H/J= 0.1 and particle fractions between 0.25% and 10%, as noted on plots; a) fraction recrystallized (F) versus time, b) KJMA plot and c) mean area of recrystallized grains (<A>) versus time, plotted logarithmically.

At large stored energies (H/J=1) variations in the number of nuclei (embryos) added to the system at fixed particle density (5%) have an effect both on the time for complete recrystallization to occur and on the final grain area (see Figs. 6a & 6c). As expected, the time required for complete recrystallization increases as the number of nuclei is decreased. The time dependence of the mean recrystallized grain size is independent of the number of nuclei (>0) at early times before the individual recrystallized grains impinge. At later times, where impingement is important, the curves corresponding to different numbers of nuclei asymptote to different final grain sizes. For comparison, an additional simulation was performed with the identical number of particles but with no nuclei added. Since no nuclei were added in this case, only normal grain growth in the presence of a particle dispersion occurred and the final grain size was approximately 100 sites; this size is similar to that observed in previous simulations of this sort[12]. Since the final recrystallized grain size equals or exceeds this size, no further evolution occurs after recrystallization is complete.

Turning to a comparison of the microstructures in the recrystallized state, Fig. 7 shows the final microstructures for a stored energy of 1.01, various numbers of nuclei at zero time and 5% volume fraction of particles. At this stored energy some interesting features emerge. For a large number of nuclei (small final grain size), the grain shapes are compact and like those observed in grain growth because the final grain size is small enough that grain growth can occur after recrystallization. For a small number of nuclei (large final grain size), however, there are obvious departures from compact, near-spherical shapes, see the lower right corner of the microstructure with the coarsest grain size (100 nuclei initially). These elongated grain shapes reflect the spatially random nature of the nucleation and they are preserved by the pinning action of the particles. When recrystallization occurs in the absence of particles[8] grain growth occurs concurrently with the recrystallization and the non-compact shapes are eliminated. During recrystallization, however, the grain morphology is non-compact as was discussed in Ref. 8.

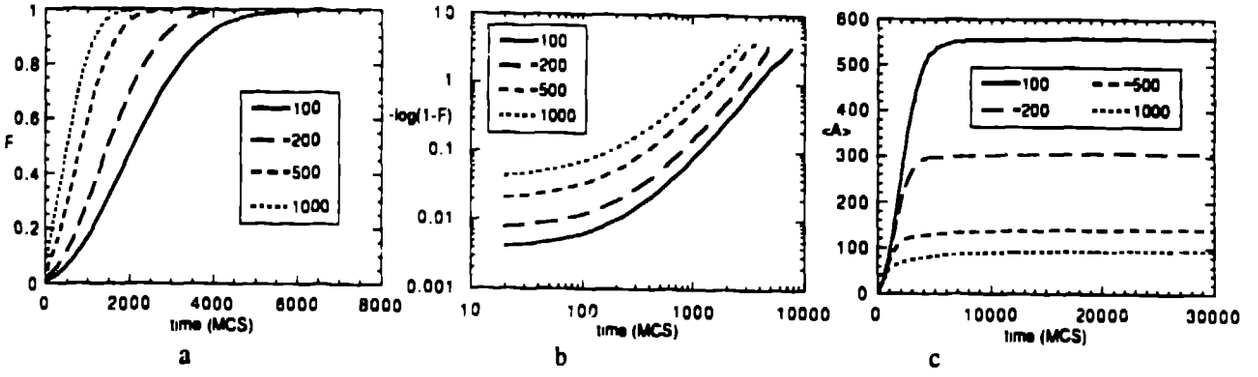

Fig. 6. Kinetics of recrystallization for 2000 particles (5% area fraction), H/J= 1.01 and 100, 200, 500 and 1000 nuclei; a) fraction recrystallized (F) versus time, b) JMAK plot and c) mean area of recrystallized grains (<A>) versus time.

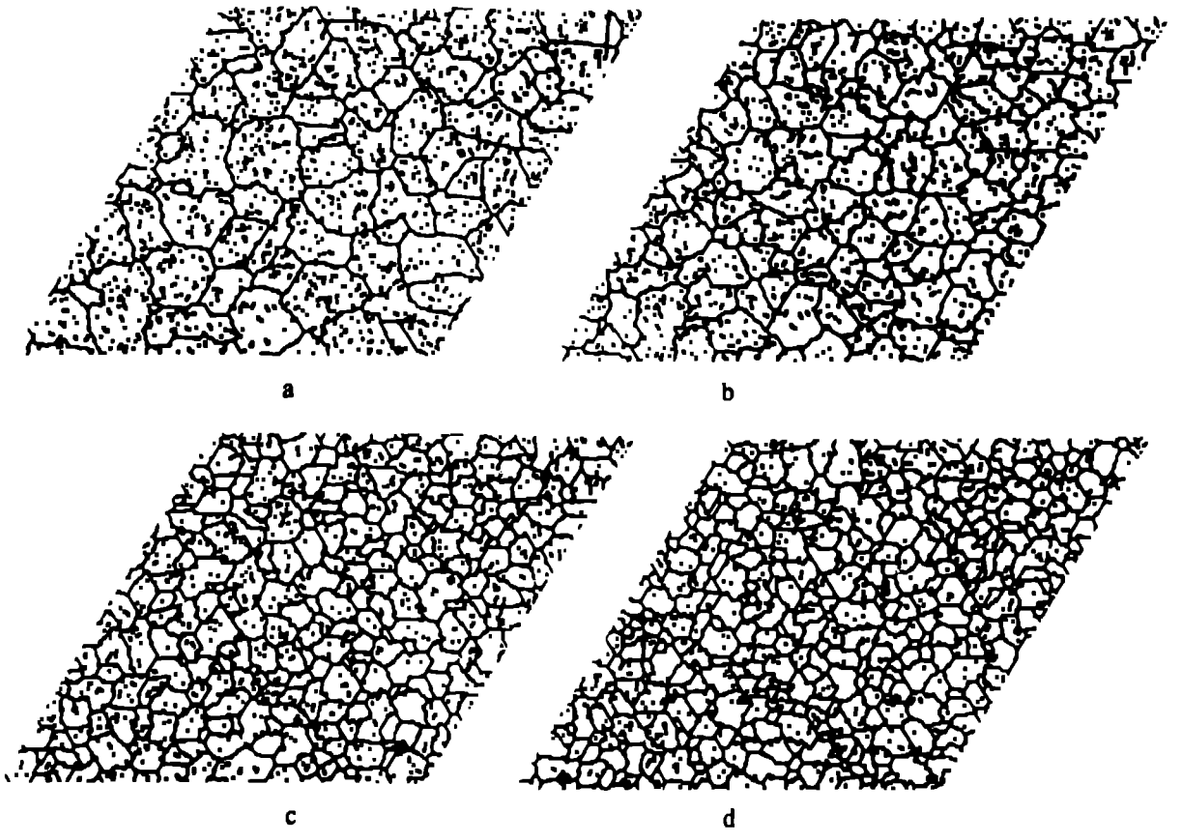

Fig. 7. Final microstructures for H/J=1.01, 2000 particles (5% area fraction) and 100 (a), 200 (b), 500 (c) and 1000 (d) nuclei.

Discussion

The results show that if sufficient driving force is available for recrystallization, the presence of particles does not affect the kinetics of recrystallization. This result has to be qualified by noting that the overall kinetics depend on the nucleation density as well as the growth conditions. Therefore it is more accurate to conclude that, given sufficient driving force, the growth of recrystallized grains is unaffected by the presence of particles. These simulations have not attempted to address the effect of fine dispersions on the nucleation density or rate.

When the driving force is small (H/J<1), on the other hand, the effect of particles is profound. If the particle fraction is large enough, recrystallization is completely prevented and the recrystallized grains introduced at zero time only grow to sizes that are comparable to the pinned matrix grains surrounding them. At small fractions, however, recrystallization does go to completion, as it does in the absence of particles[14]. The presence of a stored energy appears not to be able to affect growth of the recrystallized grains directly: instead, its effect is to bias the

grain growth such that, given sufficient coarsening, the recrystallized grains have an advantage and will dominate the structure. This supposition was tested by repeating the simulations for the lowest stored energy with 200 site saturated nuclei and various particle fractions but instead of setting H/J=0.1, the stored energy was doubled to give H/J=0.2. For particle fractions of 0.25, 1 and 2.5% and H/J=0.2, the evolution of the fraction recrystallized with time was identical to the results for H/J=0.1. This then leads to the conclusion that recrystallization can only go to completion at H/J<1 when the particle fraction is small enough that coarsening can proceed far enough. A crude estimate of this criterion can be made by calculating the particle fraction that corresponds to the pinned grain size (with initial matrix grain area of 40 sites) that corresponds to the nucleation density. For 200 nuclei (which corresponds to a grain area of 200 if all nuclei are supercritical) the minimum particle fraction to pin (see Fig. 13 of Ref. 15) is approximately 0.17. Simulations at H/J=0.1 with 200 nuclei show that recrystallization is complete for f=0.25% but pinned for f=1%. As noted above, the results for H/J=0.2 were identical, suggesting that the criterion is indeed **particle pinning** of grain growth of the **recrystallizing** grains.

Conclusions

- For a small stored energy, the presence of particles strongly retards recrystallization because the nuclei are pinned.
- For low stored energies, H/J<1, particle pinning of grain growth controls the success of recrystallization. The presence of a small stored energy gives the recrystallized grains a grain growth advantage but is insufficient to drive recrystallization fronts past particles.
- At moderate stored energies, 1<H/J<2, recrystallization fronts can pass particles in an essentially unimpeded manner such that the kinetics of recrystallization are unaffected by the presence of particles. Recrystallization itself, however, still depends on the existence of a prior grain structure.
- Even at large stored energies, H/J>1, the presence of particles affects the structure after recrystallization completes because the final structure tends to be pinned. This preserves the non-compact grain shapes that are characteristic of randomly distributed nuclei and affects the grain size distribution.
- When the stored energy is low (H/J<2), giving heterogeneous nucleation conditions, increasing the number of particles also increases the number of nuclei that are super-critical and survive.
- The variation of particle fraction on boundaries, f, shows that Zener analysis for particle drag applies while recrystallization is taking place. Post-recrystallization pinning of the microstructures, however, needs to be thought of in terms of non-random particle-boundary intersections, as is the case for grain growth in the presence of particles.

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