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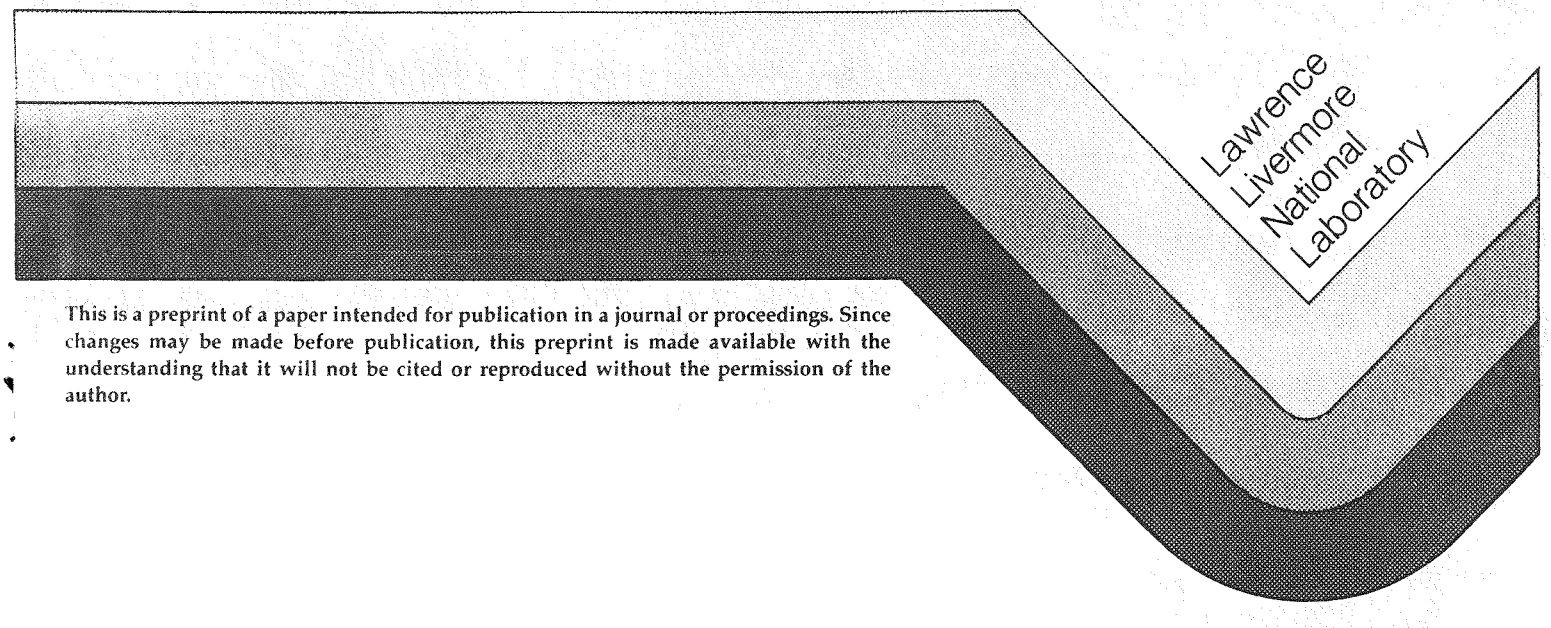

USE OF COMPUTER SIMULATIONS TO ANALYZE LIMITATIONS

OF KINETIC MODELS FOR RECRYSTALLIZATION

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Abstract

Only two kinetic models appear to be appropriate for recrystallization of metals and crystallization of metallic glasses: Kolmogorov-Johnson-Mehl-Avrami (KJMA) and Speich-Fisher (SF). However, much confusion prevails on the proper use of these models because their limitations are not well understood. Fortunately, computer simulations are proving to be effective for simulating kinetic behavior. In particular, a simplified computer model, which simulates a regular geometric grain growing in an impingement cell, provides kinetic simulations that agree with experimental data. The computer model isolates specific concepts that the kinetic models were formulated on, and it also simulates different time-dependent growth relations.

The KJMA model is based on linear interfacial growth and uses an extended-volume concept to compensate for impingement. The computer model isolated the effect of the extended-volume concept, and it also was used to determine why some experimental data conform to KJMA kinetics while others do not. The analyses indicated that essentially linear KJMA behavior can occur in recrystallization of metals that have reached an effectively constant state of recovery or in crystallization of amorphous metals that have reached a constant state of disorder.

The SF relation is based on inverse-time-dependent growth, and it is effective for recrystallization that is retarded from an initial linear rate by factors such as the simultaneous occurrence of recovery. The computer model clearly defines the limitations of the SF model.

This work demonstrates that computer simulations can define the limitations of the existing kinetic models and increase our understanding of them.

Introduction

Much uncertainty exists on the proper use of kinetic relations for recrystallization, primarily because of an incomplete understanding of the limitations of the kinetic relations and the extent to which these limitations affect the predictive capability of the kinetic relations. Many recrystallization studies have relied on the popular model developed independently by Kolmogorov,[1] Johnson and Mehl,[2] and Avrami[3-5] (KJMA). The KJMA kinetic relation has been used extensively to model recrystallization kinetics and is suitable for many materials. A major problem in the use of the KJMA relation, however, is that the later stages of recrystallization in many metals show severe negative deviation from linear KJMA behavior. This deviation has been attributed to factors such as simultaneous recovery and nonuniform deformation.[6-10] Speich and Fisher[7] (SF) developed a kinetic relation that appears to overcome this limitation to some extent and is effective for many materials, but again, confusion seems to prevail on the limitations and the extent to which they restrict the use of the SF relation.

Both kinetic relations are composed of two independent parts: a geometric model and a time-dependent term that models the growth rate. Compensation for grain impingement is

essential in the geometric models; however, impingement geometry is sufficiently complex that impingement compensation can only be approximated. In fact, much of the confusion on the use of the kinetic relations has stemmed from uncertainty about the limitations of the geometric models and the accuracy of their impingement compensation. Fortunately, the limitations of these models and their accuracy can now be analyzed with computer simulations.

Several workers have used computer models to simulate various aspects of recrystallization.[11-13] This paper will concentrate on results generated with a simplified computer simulation (SCS) model[14,15]. The model rigorously compensates for grain impingement and has demonstrated that both impingement models do predict reasonable impingement compensation with respect to the scatter of experimental data. Effective simulations of recrystallization and recrystallization kinetics also have been performed by Srolovitz and co-workers[16,17] using Monte Carlo (MC) simulations, and their work also will be reviewed.

The Kinetic Relations

The geometric model used in the KJMA kinetic relation is the well-known extended-volume concept – an explicit impingement model. This model assumes a random distribution of spherical phases, and the extended volume is defined as the total volume that would be obtained if the spheres grew through each other for the appropriate time, t , with no impingement. The actual volume transformed, V, is related to the extended volume, V_{ex}, by the relation,

$$dV/dV_{ex} = (1 - V) . \tag{1}$$

Integration and normalization to unit volume yields

$$f_{ex} = \ln[1/(1 - f)] \tag{2}$$

where f and f_{ex} are the respective volume fractions. Assuming linear time-dependent growth, which Christian[18] states is appropriate for interface-controlled reactions, Eq. 2 yields the commonly accepted form of the KJMA relation,

$$f = 1 - \exp(- \beta t^n) \tag{3}$$

where f is now the volume fraction and β is an effective reaction-rate constant that includes geometric, nucleation, and growth terms. For three-dimensional growth (which applies to the SCS model), n = 3 for site-saturated (instantaneous) nucleation, and n = 4 for a linear nucleation rate; for two-dimensional growth (which applies to the MC model), n = 2 and 3, respectively.

The geometric model used in the SF kinetic relation – an implicit impingement model – is the empirical parabolic relation,

$$A = K_a f(1 - f) \tag{4}$$

where A is the interfacial area between recrystallized and unrecrystallized material and K_a is a constant. Experimental data obtained from Fe-3.25 Si by SF agreed remarkably well with Eq. (4). The SF kinetic relation uses l/t growth in conjunction with Eq. 4 to obtain

$$f/(1 - f) = K_g t^n \tag{5}$$

where K_g and n are constants. The SF kinetic relation for recrystallization agrees reasonably well with data from materials that show strong negative deviation from KMJA behavior.

The Computer Models

The SCS model is based on the growth of geometric grains distributed on a regular, infinite lattice.[10] This simplified geometry permits simulations with a single grain growing in an impingement cell that conforms to a Wigner-Seitz construct on the distribution lattice. Grain growth is monitored by V and A of the portion of the grain that remains within the impingement cell; V is normalized to f by using a cell size of unit volume. A distinct advantage of this simplified model is that rigorous compensation for grain impingement is executed, and one form of the KJMA extended volume can be defined as the total volume of the SCS grain with no impingement compensation. Disadvantages of the model are that it is highly ordered and it applies rigorously only to site-saturated or instantaneous nucleation, but these conditions appear to correlate better with experimental behavior than random nucleation and growth. The technique

for generating kinetic plots from the SCS data was discussed previously[14] and is demonstrated elsewhere in these proceedings.[15]

The MC technique developed by Doherty and co-workers[9,10,16,17] also yields effective simulations of both recrystallization and various aspects of grain growth. Most of their simulations are two-dimensional and use a finite-element network composed of triangular elements; their typical simulated field is 200 x 200 elements. They define a unit of time as one MC probability sweep through all 40,000 elements (a MC step in their terminology). They assign a stored energy of H to each element in the unrecrystallized fields, and they impose a dimensionless grain-boundary energy, J, between two adjacent elements when the elements are in different grains. The probabilities of both nucleation and growth are proportional to H/J (dimensionless units) in their simulations. An important aspect of the MC technique is that it can model recrystallization in nonuniformly deformed materials by assigning a distribution of energies to H.

Kinetic Simulations

The KJMA Relation

SCS Model. Simulated KJMA kinetic plots are presented in Fig. 1 for a sphere growing in a cubic impingement cell, which conforms to the axial symmetry constraint described elsewhere in these proceedings.[15] The SCS data (indicated by + in Fig. 1) are initially linear. The dotted line through these points has a slope of 3.0, which agrees with three-dimensional KJMA behavior for site-saturated nucleation. Beyond the linear region, which extends to roughly f = 0.1, the upper portion of the SCS kinetic curve shows slight upward curvature. In comparison to the scatter of experimental data, however, the SCS simulations are reasonable and indicate that the extended-volume concept does provide reasonable impingement compensation.

To analyze the contribution of the extended volume to the upward curvature of the SCS data in Fig. 1, extended volumes were calculated as the volume of the spherical grain with no corrections for impingement (termed $f_{1\,ex}$ following notation of Avrami[4]) and from Eq. 3 to obtain f_{ex} from the SCS values of f.[19] As shown in [19], $f_{ex} > f_{1\,ex}$. Equation 3 should apply only to randomly distributed grains and not the ordered array of the SCS model. However, f_{ex} in Eq. 3 formally governs the KJMA kinetic relation and, hence, behavior of the SCS data in KJMA plots, and the difference between f_{ex} and $f_{1\,ex}$ measures the amount of overcompensation of f_{ex} that causes the upward deviation.

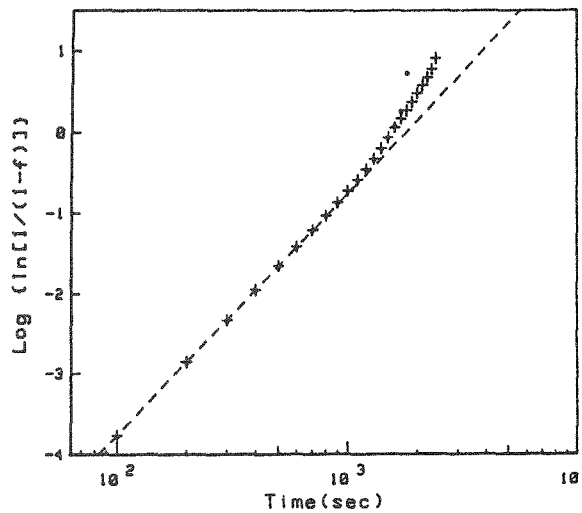

Figure 1 - Simulated KJMA plot of the SCS data for a sphere in a cubic cell.

Figure 1 also includes a plot of $f_{1\,ex}$ (dots) ($f_{1\,ex}$ is limited to f < 1 because of the logarithmic term). When f is regenerated from $f_{1\,ex}$ using equation (2) (as in [14]), the plot falls along the straight line with n = 3 to within plotting accuracy. Similar results also are obtained for a bipyramid with {111} faces as shown in [14] and [15]. Therefore, the difference between f_{ex} and $f_{1\,ex}$ is a measure of the overcompensation of f_{ex} in equation (2) for each SCS model, and this overcompensation correlates directly with the upward deviation of the simulated plots. As discussed in [14] for a sphere growing in a cubic impingement cell, the overcompensation reaches a maximum of about 26% at f = 0.7 but is only about 7% at f = 1. The deviation is somewhat less for the {111} bipyramid. The extent to which the overcompensation is related to the ordered distribution geometry of the SCS model as opposed to the random distribution that is assumed in the KJMA model remains to be isolated.

MC Model. Srolovitz et al.[16,17] simulated KJMA behavior in uniformly deformed metals by using constant values of H/J. KJMA plots of their simulated data had a slope of $n \approx 2$ for site-saturated simulations and $n \approx 3$ for simulations with a constant rate of nucleation; these values agree with two-dimensional KJMA behavior. Their simulations yield homogeneous nucleation for H/J > 2 and heterogeneous nucleation adjacent to grain boundaries for H/J < 2.

Rollett et al.[10] simulated recrystallization in non-uniformly deformed metals by allowing H/J to vary from 0.1 to 5.01. Site-saturated nucleation was modeled by randomly positioning a fixed number of nuclei in the finite-element network prior to the simulation, and continuous nucleation was modeled by adding a fixed number of nuclei at each MC step. Since nuclei positioned in recrystallized regions or low-energy regions were subcritical and decayed away, the nucleation rate decreased with time. An important result from these simulations is that nonuniform deformation can cause negative deviation from KJMA behavior, although the nonuniform energy distributions used in the MC simulations also could correlate with the occurrence of simultaneous recovery.[8]

RSDA simulated $f_{l\,ex}$ by allowing only one grain to grow in the finite-element network. Only site-saturated nucleation was simulated for the extended-volume study; one set of simulations was performed for homogeneous stored energy of H/J = 3 with 500 nuclei and one set for the nonuniform stored-energy distribution with $0.1 < H/J < 5.01$ with 3500 nuclei. Following the extended-volume concept, they assumed the relation

$$f_{ex} = m\, V_{l\,ex} \qquad (6)$$

where m is the number of simulated recrystallized grains at $t = \tau$ adjusted to unit volume. They calculated df/dt and df_{ex}/dt from their MC simulations to obtain df/df_{ex} in equation (1) adjusted to unit volume, and conformance to equation (1) was tested in plots of $\log(df/df_{ex})$ vs. $\log(1 - f)$. Their results for homogeneous stored energy showed reasonably good correlation with Eq. 1. This agreement implies that the extended-volume concept also does not introduce serious errors in the MC simulation for site-saturated recrystallization in a matrix with a homogeneous distribution of stored energy.

For the case of the nonuniform stored energy, serious deviation occurred from the linear plot of Eq. 1. RSDA suggest that this apparently catastrophic decrease results from serious underestimation of the impingement correction in equation (1) and this suggestion presents an intriguing possibility that should not be ignored. However, f_{ex} must be calculated from the sum of all $f_{l\,ex}$ that correspond to the specific time frames and growth rates for each individual grain, and Eq. 6 applies only to the case of a constant growth rate for homogeneous stored energy and not to the case of non-uniform stored energy. Nonetheless, these simulations demonstrate the important possibility that nonuniform energy distributions can cause negative deviation from KJMA behavior and also abnormally low values of n.

The SF Relation

Use of the SCS model to simulate SF kinetics is demonstrated elsewhere[8,14,15] The SCS model shows reasonable agreement with both Eq. 5 and experimental measurements of A and f[8]; simulated SF plots using the SCS model also show excellent agreement with the SF data for Fe-3 1/4 Si[7] and data for vanadium[20].

Limits of f = 0.02 to f = 0.95 were specified by Speich and Fisher for their relation, and SCS simulations indicate that the limits can be extended to f = 0.01 to f = 0.99. These limits are imposed by the breakdown of Eq. 4 at the extremes of f = 0 and f = 1, as first emphasized by Cahn[21]. A and f in Eq. 4 are not geometrically related, and the SCS model has demonstrated that this breakdown occurs because Eq. 4 has finite slopes at the extremes whereas the slopes of geometric growth curves for regular geometric grains become infinite at the extremes.[14] This breakdown causes the SCS data to become asymptotic to the ordinate at each extreme in SF plots.[22]

Discussion

Models for impingement correction can only approximate actual impingement geometry, but they should be relatively simple so that kinetic relations derived from them will be reasonably tractable. Both the KJMA extended-volume concept and the SF parabolic relation conform to this viewpoint. This work demonstrates that computer simulations can define the limitations of the impingement models and increase our understanding of them. Both the SCS and the MC models have isolated the effect of the KJMA extended-volume concept to kinetic behavior, and the SCS model also has isolated the effect of the SF parabolic relation. The simulations reveal that both impingement models provide reasonable impingement compensation. In addition, when the proper time dependence is used in computer simulations, both computer

models yield simulated kinetic data that compare favorably to the KJMA and SF kinetic relations, particularly when compared to the statistical scatter of experimental data on logarithmic plots. Computer simulations also provide information on the applicable ranges of the resulting kinetic relations.

Simulations with the SCS model also reveal the following: (a) conformance to the axial symmetry constraint yields simulated geometric growth curves (plots of A against f[14]) that conform reasonably well to both Eq. 4 and experimental measurements of A and f reported by Speich and Fisher[7]; (b) when the size parameter of a simulated grain is scaled to an appropriate growth rate, simulated kinetic plots conform surprisingly well to either the KJMA or the SF kinetic relations, depending on the modeled growth rate.

The SCS model formally isolates impingement geometry, and it is useful for cursory tests of various time dependences that may control kinetic behavior; however, it effectively is confined to site-saturated nucleation and ordered distribution geometry. In comparison, impingement geometry is only implicitly treated in the MC model (and similar computer simulation models[11-13]), and the MC model suffers some limitations from the minimum finite-element size that is convenient for computer simulations. However, the MC model clearly provides an excellent technique to model the effects of many factors such as incubation periods, nonlinear nucleation rates, non-uniform deformation, and stored-energy decay (simultaneous recovery), and it also models random distributions. It also can simulate the formation of realistic grain structures.

The computer simulations indicate that apparent failures of the kinetic relations can be related to limitations imposed by the time-dependent relations that were used to derive the kinetic relations. This fact often is ignored and leads to improper use of the kinetic relations. In essence, transformations that occur with linear growth should yield linear KJMA plots; examples of such transformations are recrystallization in materials with limited or no recovery, recrystallization in materials with an effectively constant state of recovery (e.g., dynamic recrystallization), and crystallization in glassy metals that have achieved an effectively constant state of disorder[23]. In contrast, when recrystallization in metals is retarded by reactions such as simultaneous recovery, the growth kinetics become nonlinear and do not necessarily yield linear KJMA plots. In this case, the SF relation can yield useful results within the range that is stipulated by the limitations of the impingement model[8,14].

Doherty et al. [9] reviewed evidence of nearly linear KJMA behavior in several metals. A recent review of other evidence of linear KJMA kinetic behavior includes static recrystallization and dynamic recrystallization during hot working of metals and crystallization of metallic glasses.[23] Rationale based on these examples suggests that that conformance to linear KJMA kinetics in metals depends on the kinetics of competitive reactions such as recovery and various recovery-like reactions (including the stored-energy distribution that results from nonuniform deformation), and conformance in metallic glasses depends on the state of disorder in the amorphous matrix. When these reactions approach an essentially steady state, linear KJMA behavior can be approached, and the slopes of KJMA plots should approach the theoretical values of $n = 3$ for site-saturated nucleation or $n = 4$ for a constant nucleation rate. However, if the primary contribution to linear KJMA behavior arises from the constant decay of both nucleation and growth rates, then the KJMA slope will be less than the theoretical value. Many materials exhibit low values of n when they conform to linear KJMA behavior, but Doherty et al.[9] emphasized that few materials can meet the precise conditions that yield linear KJMA behavior.

Variations of n in KJMA plots from the values predicted for linear growth, particularly low values of n for dynamic and static recrystallization in metals, may be misconstrued as a failure of the KJMA model, and nonlinear KJMA behavior does constitute a failure of Eq. 3 to provide an analytical representation of the data. However,Vandermeer[24] emphasizes that these deviations merely signify that the proper growth-rate dependence must be coupled with the KJMA extended-volume model to derive the proper kinetic relation. In fact, the KJMA relation appears to have been misused extensively because the limitations of the relation have either been ignored or not properly understood.

Summary

Computer simulations can be highly effective for analyzing the limitations of kinetic models. The simulations reveal that both the KJMA and the SF geometric models provide

reasonable impingement compensation, but their limitations must be carefully defined. Although failures of the kinetic relations have been attributed to approximations introduced by the impingement models, the computer simulations reveal that major problems arise from limitations imposed by the time dependence that is coupled to the impingement model to derive the kinetic relation. The time dependence often is ignored and can be a major source of confusion.

The KJMA relation does not compensate for retarding effects on recrystallization such as simultaneous recovery or nonuniform deformation, and KJMA plots for recrystallization in cold-worked metals frequently show severe negative curvature. However, reasonably linear KJMA behavior does occur in recrystallization of some metals and in crystallization of metallic glasses, but considerable variation occurs in the slopes of KJMA plots. This evidence suggests that linear KJMA behavior may occur during recrystallization in metals when either a deformed matrix decays to an effectively constant state of recovery, the recovery rate decreases at a constant rate, or nonuniform deformation yields a critical stored-energy distribution. Linear behavior also may occur during crystallization when the amorphous matrix remains in an essentially constant state of disorder or disorder reactions occur at a constant rate. The SF relation appears to be effective for many materials that exhibit nonlinear KJMA behavior.

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