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Grain size distribution in two dimensions in the long time limit

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Abstract

It is shown that the inclusion of a "noise" term in the growth rate of individual grains leads to a stochastic model that provides a more realistic description of grain growth phenomenon. The resulting Fokker–Planck equation for the grain size distribution is solved numerically due to the difficulties in obtaining an analytical solution. The analysis is limited to two dimensions and assumes quasi-stationary distributions in the long time limit. The resulting grain size distribution is shown to be in agreement with that obtained from computer simulations, indicating the validity of the stochastic approach.
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Keywords: Grain size distribution; Stochastic model; Fokker–Planck equation; Numerical solution

1. Introduction

Grains grow during the evolution of grain microstructure after recrystallization results in an increase in average grain size by the motion of grain boundaries and the gradual disappearance of smallest grains. Due to the complexity of the structure of individual grains and the participation of a large number of grain boundaries in the process, developing a realistic model of grain growth is a formidable challenge; many conceptual and mathematical simplifications have to be made. Simplification is possible by considering grain growth in two dimensions (thin films) only. This is due to the von Neumann law [1] whose applicability to grain growth was proved in two dimensions by Mullins [2]. For this reason, the present discussion will be confined to two dimensions. (It should however be noted that an extension of the present work to three-dimensional grain growth is now possible due to the pioneering work of Glicksman [3,4] and MacPherson and Srolovitz [5]. In fact, the resulting equations for grain size distribution are closely related. In the three-dimensional case it is specimen volume that is conserved instead of area, and the resulting equations for grain size distributions are similar though not identical. A modified version of the technique presented below could be used for the three-dimensional case also. However, we find that the equations connecting various constants are much more complicated in the three-dimensional case and require more detailed analysis. We will therefore confine our analysis to the two-dimensional case only.)

Using the concept put forth by von Neumann [1], Mullins [2] showed that the rate of change of the area of an individual grain, $\mathrm{d}A/\mathrm{d}t$, growing under the influence of its curvature-driven motion is given by the expression:

$$\frac{\mathrm{d}A}{\mathrm{d}t} = M(n-6), \tag{1}$$

where n is the number of sides of the grain and M is a constant. This equation appears to be completely deterministic because it specifies the exact areal growth rate of a grain in terms of n. In principle, the areal growth rate can be used to determine the grain size distribution $F(R,t)$ as function of the grain size R at time t. However, Eq. (1) contains two variables, A (or R, with $A \propto R^2$) and n, which are both generally functions of time. If n is uniquely determined by R, a

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deterministic expression for dR/dt can be obtained. Experimentally, and in simulations, this is found to be not true, which has important implications that will be discussed below.

The first attempt to relate R and n was made by Hillert [6] who gave a comprehensive and consistent theoretical treatment of grain growth in both two and three dimensions. Using heuristic arguments he showed that:

$$n = k_1 + k_2 \frac{R}{\bar{R}}, \tag{2}$$

where $\bar{R}$ is the mean grain size, defined in terms of the distribution function as

$$\bar{R}(t) = \frac{\int_0^\infty RF(R,t)\mathrm{d}R}{\int_0^\infty F(R,t)\mathrm{d}R} \tag{3}$$

and k_1 and k_2 are constants. This linear correlation between n and R is discussed further by Rivier and Lissowski [7] and was also first observed in simulations of two-dimensional grain growth by Fradkov et al. [8] using an approach that allowed variation of the unknown rates of topological switching events. It is also supported from some experimental evidence [9].

Eq. (2) leads to an equation for dR/dt as

$$\frac{\mathrm{d}R}{\mathrm{d}t} = -\left(\frac{a}{R} - \frac{b}{\bar{R}}\right) \tag{4a}$$

and for $F(R,t)$ as:

$$\frac{\partial F(R,t)}{\partial t} = \frac{\partial}{\partial R}\left[\left(\frac{a}{R} - \frac{b}{\bar{R}}\right)F(R,t)\right], \tag{4b}$$

where a and b are constants. Eq. (4a) was first obtained by Hillert. Starting from von Neumann's law (Eq. (1)), he assumed that $A \sim R^2$ and $n \sim R$, which leads to Eq. (4a) directly. The relation between n and R was put on firm mathematical grounds by Rios and Glicksman [10] using topological considerations. They show that this relation is indeed true within a few per cent. We have generalized the Hillert equation by using the two constants a and b, instead of the values used by Hillert. Eq. (4a) is thus true for the two-dimensional case only. Hillert, however, assumed that it applies to the three-dimensional case also with somewhat different constants. Eq. (4a) thus has a sound mathematical basis. The physics of this equation is somewhat less clear. From the equation itself one gets that dR/dt is simply proportional to the difference in curvatures of a sphere of radius R and radius $\bar{R}$. We prefer the topological interpretation provided by Rios and Glicksman [10].

Predictions from these types of "mean field" models are in serious disagreement with experiments. There have been many attempts to improve or modify such models to bring them in agreement with experiments. Mullins [11] suggested a cubic relation between n and R instead of Eq. (2), leading to a "mean field" model, which satisfies all necessary constraints, including specimen size conservation. He thus obtained a better agreement with experiments. This presents a dilemma because Abbruzzese et al. [12] have shown quite convincingly that the linear relation is correct, at least approximately, on both experimental and theoretical grounds. Also, Eq. (4) can now be derived on first principles, at least approximately [3,4].

There is another problem of a more fundamental nature with the type of continuity given in Eq. (4). Pande and Rajagopal [13] have shown mathematically that grain growth models based on a mean-field approach provide no mechanism to explain many properties of grain growth that are observed experimentally, no matter what form of the growth rate is assumed. For example, they showed that mean field models do not predict that the long time distribution is independent of the initial distribution. Computationally, Battaile and Holm [14], for example, have shown by using Monte Carlo simulations that the asymptotic self-similar distribution does not depend on the initial distribution. Mean field models also provide no mechanism to explain scaling of the distribution at large times (self-similarity). As far as we are aware these conclusions have not been challenged by anyone. Also, we are not aware of any mathematical technique that can derive, even in principle, the independence of the long time distribution from the initial distribution or the scaling property using "mean field" techniques. In all the derivations of scaling behavior, using "mean field" scaling is assumed as given. We have also assumed scaling in our analysis here, but a stochastic formulation can be used in principle to derive the scaling property [13].

At this point one may be justified in asking if our statement implies that the mathematical technique used in the Lifshitz–Slyozov–Wagner (LSW) derivation is suspect, since Hillert's "mean field" derivation of grain size distribution follows very closely the LSW method. In particle coarsening, the LSW theory and other "mean field" theories predict, in general, that particles with identical size should have the same rate of growth, regardless of their location and environment in the microstructure. Except for vanishingly small volume fraction, experimentally this is found to be not correct. Experiments [15] clearly show the presence of fluctuations, indicating that particles of the same size exhibit different growth rates. Rogers et al. [16] in their experiments found that some relatively large particles shrank, and suggested that growth rates of individual particles depend not only on their size, but also on the details of their local environment.

Voorhees and Glicksman [17] first demonstrated by simulations the presence of fluctuations during phase coarsening. In fact, Glicksman et al. [18] and Wang et al. [19] developed a stochastic analytic model of coarsening that employed a Fokker–Planck equation to estimate the theoretical distributions in coarsening for finite volume fractions. Based on these considerations, these researchers concluded that "mean field" theories predict average behavior of coarsening systems [18,19]. If local details need to be included, then departure from mean field behavior must be considered. Fluctuations occur in the growth rates of particles at any nonzero volume fraction. Technically,

LSW theory is thus valid only for zero volume fraction of particles, and is thus a limit solution. In grain growth there is no physical analog to the zero volume fraction case. However, we will see that our solution approaches the Hillert solution as a limiting case when the fluctuation term approaches zero. In this sense our treatment unifies the LSW treatment with stochastic analysis. LSW results are then obtained from a limit solution of the stochastic model without using the LSW mathematical apparatus. A need for finite cut-off is thus avoided. We thus recover a LSW-type result without using LSW methodology.

These "mean field" models introduce an upper cutoff in the grain size distribution and a mathematical technique that uses "stability conditions" to obtain grain size distribution function in the long time limit. It has been argued that the LSW technique could lead to multiple distributions [20]. However, some of these solutions can be ruled out on physical grounds (see, however, Rios et al. [21]). We would like to state that the LSW technique using cutoff and stability conditions is a perfectly valid mathematical procedure for obtaining grain size distribution in the mean field limit. Below we use an alternate approach, i.e. a stochastic treatment [13,22], which does not require an upper cutoff or "stability conditions". The following description will, we hope, shed some light on the relation between the "mean field" approach and our own approach, which attempts to approximately take into account the problem of local environment.

2. Stochastic effects in grain growth

The continuity equation, Eq. (4), yields a first-order partial differential equation for the size distribution if the growth rate function $\mathrm{d}R/\mathrm{d}t$ is a single-valued function for all grains of size R at time t. This was assumed by Hillert in deriving Eq. (4). The rationale behind the use of this deterministic growth rate expression is that it will describe the statistical grain growth dynamics adequately if $\mathrm{d}R/\mathrm{d}t$ is an appropriate average growth rate for all grains of size R in a given state of the system. However, specific grains having the same R value at a given time will have different shapes and possibly different values of n, so their growth rates may vary. This fluctuation (or "noise") in the $\mathrm{d}R/\mathrm{d}t$ function changes the mathematical character of Eq. (4) significantly.

Pande [22] and Pande and Dantsker [23,24] have attempted to develop a more rigorous theory of isothermal grain growth capable of predicting all of the major attributes of grain growth with minimal assumptions by treating it as a stochastic process. Mathematically, a stochastic process in its simplest form is described by a function of two variables, one of which is time, and involves both a deterministic term and a random term. Specifically in this treatment the relation (2) is retained except that it is regarded as a statistical relation and not an exact one.

The stochastic continuity or Fokker–Planck equation in this case is given by:

$$\frac{\partial F(R,t)}{\partial t} = \frac{\partial}{\partial R}\left\{\left(\frac{a}{R} - \frac{b}{\bar{R}}\right)F(R,t)\right\} + D\frac{\partial F^2(R,t)}{\partial R^2}, \tag{5a}$$

where a, b and D are arbitrary constants yet to be determined. On comparing with the mean field continuity Eq. (4), it is seen that it has an additional term called the diffusion term. The diffusion term cannot be removed by an averaging procedure as suggested by Mullins [11] since the existence of an averaged quantity (first moment) requires that the second moment that corresponds to the diffusion term must also exist. The magnitude of the diffusion term could, however, be small. This will be one of the issues discussed in this paper. The justification for the use of Eq. (5a) is as follows.

Pande and Rajagopal [13] show that in order to describe all the main features of grain growth, the distribution must be described by a Fokker–Planck equation of the form:

$$\frac{\partial F}{\partial t} = \frac{\partial}{\partial R}[A(R,t)F] + \frac{\partial}{\partial R^2}[B(R,t)F] = 0, \tag{5b}$$

where, in general, both A and B are functions of R and t. They have to be determined from the physics of grain growth. We assume that $A(R,t)$, which is $-\mathrm{d}R/\mathrm{d}t$, is given by Eq. (4a). The term $B(R,t)$ takes into account the role of the local environment of a particular grain. Mathematically, Eq. (5b) is equivalent to a Langevin equation:

$$\frac{\mathrm{d}R}{\mathrm{d}t} = A(R,t) + [2B(R,t)]^{1/2}T(t), \tag{5c}$$

where A represents the deterministic or drift term and B is the coefficient of the random or noise term $T(t)$. The fluctuation term $T(t)$ in Eq. (5c) is assumed here to have properties similar to those in classical Langevin equation [25].

In random walk or Brownian motion, $T(t)$ is denoted as a Gaussian random variable with zero mean and having a correlation function that is proportional to a Dirac delta function $\delta_{\mathrm{D}}(t)$. The physical picture embodied by Eq. (5c) is that the random term in this Langevin-type equation creates a tendency for the grain sizes (R values) to spread out over an ever-broadening range of values. Simultaneously, the first (deterministic) term on the right-hand side is akin to a damping (or amplification) term, which here relates the areal growth rate to the instantaneous deviation of the radius from its mean value. The ultimate size distribution $F(R,t)$ achieved during grain growth represents the combined effect of these two tendencies, subject to appropriate global constraints such as constancy of total specimen size (mass conservation.) The function B is, in general, a function of R and t. As a first approximation, we assume that B is a constant and we denote it by D.

It is difficult to solve Eq. (5a) analytically. Our aim in this paper is to solve it numerically in the long time limit, and to compare the results with experiments and computer simulations. For further details regarding the basis for steps leading to Eq. (5c), see Refs. [22–24]. The constant a is a material parameter that can in principle be deter-

mined exactly as shown in Ref. [4]; in our treatment we take it as an unknown parameter. The other two constants, b and D, can be determined exactly in terms of the constant a from boundary conditions as shown below. Hence, there are no adjustable parameters in Eq. (5a) except for the ratio a/D (see Section 6).

3. Solution in the long time limit

The continuity equation, Eq. (5a), can be recast as an ordinary differential equation by imposing the experimental observation of self-similarity of the grain size distribution at long times (see Section 6 for further comments about self-similarity). We assume that all grains sizes are accessible and that the total area (mass) of the polycrystalline system is conserved and finite. Appropriate boundary conditions for the size distribution $F(R,t)$ may thus be written as [22]:

$$F(0,t) = F(\infty,t) = 0. \tag{6}$$

The area (mass) conservation requirement can be expressed in integral form as:

$$A = \int_0^\infty R^2 F(R,t)\mathrm{d}R = \text{constant}. \tag{7}$$

Eq. (5a) admits a family of exact similarity solutions of the general form:

$$F(R,t) = t^{\mu} f\left(\frac{R}{2\lambda t^{1/2}}\right), \tag{8}$$

where the dependence of F on the similarity variable $x = R/2\lambda t^{1/2}$ is commonly found in equations of this type (e.g. in the Boltzmann transformation used in the analysis of nonlinear diffusion problems [26]). The value of the exponent μ of the time-dependent prefactor in Eq. (8) can be determined from the second requirement given in Eq. (7), i.e. specimen area conservation: substituting Eq. (8) in Eq. (7), we find that $\mu = -3/2$. Finally, substituting Eq. (8) with $\mu = -3/2$ in Eq. (5a), we obtain a time-independent ordinary differential equation for $f(x)$ which is given as Eq. (11) below.

Under this transformation the mean grain size $\bar{R}(t)$ is then given by:

$$\bar{R}(t) = 2\lambda t^{1/2}\bar{x}, \tag{9}$$

where

$$\bar{x} = \frac{\int_0^\infty x f(x)\mathrm{d}x}{\int_0^\infty f(x)\mathrm{d}x}. \tag{10}$$

The procedure described above is, in fact, common in both coarsening and grain growth. The time-dependent prefactor is, however, different in coarsening and grain growth. For details see Ref. [13]; see also Ref. [27]. The shape of the renormalized grain size distribution $f(x)$ and its first moment $\bar{x}$ are time invariant, but the scale factor $\bar{R}(t)$ increases as the square root of aging time.

By using Eq. (8), Eq. (5a) can be rewritten as:

$$\frac{D}{\lambda^2} f_{xx} + \left(\frac{a}{\lambda^2 x} - \frac{b}{\lambda^2 \bar{x}}\right) f_x + \left(6 - \frac{a}{\lambda^2 x^2}\right) f = 0. \tag{11}$$

As noted above, the constants λ, D, a and b in the preceding equations are not independent. Relationships among them can be determined so that only two parameters appear in the scaled ordinary differential equation. It is convenient to normalize the grain size distribution $f(x)$ to have zeroth and first moments of unity; we also require a finite second moment (independent of time) for mass conservation. The far-field behavior of a general solution to Eq. (11) can be determined (see Ref. [28, p. 78], and Eqs. (23) and (24) below) to consist of the sum of an exponentially decaying solution and an algebraically decaying solution with $f(x) \sim 1/x^3$. By multiplying Eq. (11) by x^2 and integrating, we find that:

$$\frac{(D + b - a)}{\lambda^2} \int_0^\infty f(x)\mathrm{d}x + \lim_{x\to\infty} x^3 f(x) = 0, \tag{12}$$

so that imposing the condition $(D + b - a) = 0$ eliminates the slowly decaying solution with $f(x) \sim 1/x^3$ and produces a grain size distribution with finite second moment. With this choice and letting $\varepsilon = D/\lambda^2$ and $\alpha = b/\lambda^2$, Eq. (11) has the form:

$$\varepsilon \frac{\mathrm{d}^2 f}{\mathrm{d}x^2} + \left[\frac{\alpha + \varepsilon}{x} - \alpha + 2x\right] \frac{\mathrm{d}f}{\mathrm{d}x} + \left[6 - \frac{\alpha + \varepsilon}{x^2}\right] f = 0, \tag{13}$$

with the added normalization:

$$\frac{\int_0^\infty x f(x)\mathrm{d}x}{\int_0^\infty f(x)\mathrm{d}x} = 1. \tag{14}$$

Eq. (13) can be solved exactly for two limiting cases where the driving force is either due only to the drift velocity (Hillert) or diffusion (Rayleigh).

The Rayleigh grain size distribution [29] occurs when $\alpha = 0$ and $\varepsilon \neq 0$. This means that all curvature effects are ignored, and the grains perform a pure random walk in grain size space. Eq. (13) in this case is reduced to:

$$\varepsilon \frac{\mathrm{d}^2 f_r}{\mathrm{d}x^2} + \left[\frac{\varepsilon}{x} + 2x\right] \frac{\mathrm{d}f_r}{\mathrm{d}x} + \left[6 - \frac{\varepsilon}{x^2}\right] f_r = 0, \tag{15}$$

the only physically acceptable solution of which is

$$f_r(x) = cx \exp\left[\frac{-x^2}{\varepsilon}\right]. \tag{16}$$

Since $f_r(x)$ satisfies the boundary conditions when $x = 0$ and $x \to \infty$, ε and the constant c can be determined by requiring the grain size distribution to be normalized, and its first moment, $\bar{x}$, to be equal to unity. This leads to the normalized Rayleigh grain size distribution:

$$f_r(x) = \frac{\pi}{2} x \exp\left[-\frac{\pi x^2}{4}\right]. \tag{17}$$

The Hillert grain size distribution [6] corresponds to the other limiting case, $\alpha = 8$ and $\varepsilon = 0$, and occurs when the drift velocity due to curvature is the only driving force, i.e. the equation is of the first order (mean field.) Eq. (13) then reduces to a first-order ordinary differential equation:

$$\left[\frac{2(x-2)^2}{x}\right]\frac{df_h(x)}{dx}+\left[6-\frac{8}{x^2}\right]f_h(x)=0, \tag{18}$$

the normalized solution of which is the Hillert grain size distribution:

$$f_h(x)=\frac{8xe^{\frac{-2x}{2-x}}}{(2-x)^4}, \tag{19}$$

for $0<x<2$. This distribution is characterized by an imposed finite cut-off and the assumption that all derivatives of f vanish at $x=2$.

4. Numerical solution for the grain size distribution

The Rayleigh and Hillert distributions thus represent the extreme limits of Eq. (13). When a noise term is present (i.e. both α and ε are finite), an analytical solution is difficult, and therefore we numerically solve Eq. (13) subject to constraint condition of Eq. (14). First, Eq. (13) is reduced to a system of two first-order coupled differential equations:

$$\begin{aligned}\tfrac{df(x)}{dx}&=g(x)\\ \tfrac{dg(x)}{dx}&=-g(x)[\tfrac{\alpha+\varepsilon}{x}-\alpha+2x]+[6-\tfrac{\alpha+\varepsilon}{x^2}]f(x)=0\end{aligned} \tag{20}$$

while the normalization condition $\bar{x}=1$ is rewritten as:

$$h(x)=\int_0^x(1-y)f(y)\,dy\to 0 \quad \text{as } x\to\infty. \tag{21}$$

In differential form, this becomes:

$$\frac{dh(x)}{dx}=(1-x)f(x), \tag{22}$$

with the boundary condition $h(0)=0$. Eqs. (19) and (22) constitute a system of ordinary differential equations that were solved by using an adaptive finite difference package with error control. Due to the singular nature of the coefficients in Eq. (20) at $x=0$, the starting point was taken to be $x_o=0.1$. A series expansion solution for Eq. (13) for small x was developed and used to determine the shifted initial conditions at $x=x_o$.

The numerical solution to the system of equations required a criterion to determine the appropriate pair of (α–ε) that satisfied the constraint Eq. (14). Since $h(x)$ must approach zero as $x\to\infty$, a simple assessment was used, solving the equations over a finite interval $x_o<x<x_l$, and, for a given value ofα, varying ε until $h(x_l)=0$. The upper limit, x_l, where the numerical calculations were terminated, was usually taken to be $x_l=10$, which was large enough to ensure the decay of the grain size distribution $f(x)$ to machine precision. Based upon this scheme, values of (α–ε) were calculated and are shown in Fig. 1.

Normalized grain size distributions were calculated from the results of Fig. 1 along with two limiting cases: Hillert ($\alpha=8$ and $\varepsilon=0$) and Rayleigh ($\alpha=0$ and $\varepsilon=4/\pi$). These results are shown in Fig. 2.

From Fig. 2, we note that the numerical solutions vary smoothly from the Rayleigh to Hillert as ε changes from 0 to $4/\pi$ with the corresponding value of α as required in Fig. 1. The numerical solutions are only valid for $\varepsilon>0$. When $\varepsilon=0$, the ordinary differential equation reduces to first order, and the solution is given by Eq. (18), the Hillert grain size distribution.

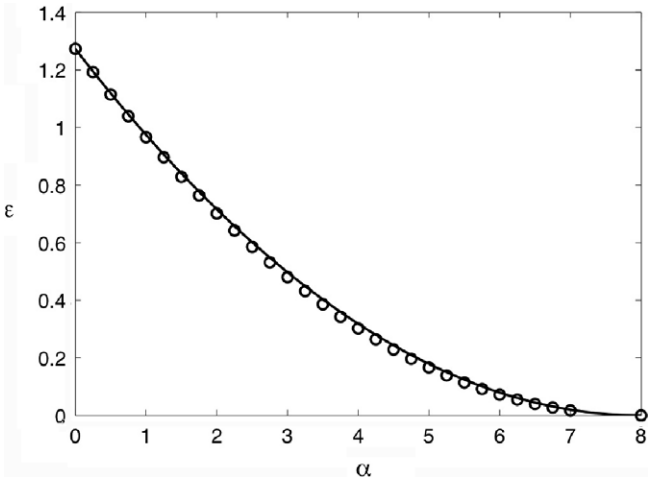

Fig. 1. Numerically determined (α–ε) relationship.

The change in the structure of the ordinary differential equation when $\varepsilon\to 0$ requires a singular perturbation analysis such as the Wenzel–Kramer–Brillouin (WKB) method (see Ref. [28, p. 484]). However, asymptotic solutions will yield forms that are commensurate with the WKB method in the limit as $x\to\infty$. This leads to the asymptotic forms, valid for $x>>1$,

$$f_a\sim e^{-x^2/\varepsilon}e^{\alpha x/\varepsilon}x^{1-\alpha/\varepsilon}(1+b_1/x+b_2/x^2+\cdots), \tag{23}$$

and

$$f_b\sim\frac{1}{x^3}(1+c_1/x+c_2/x^2+\cdots), \tag{24}$$

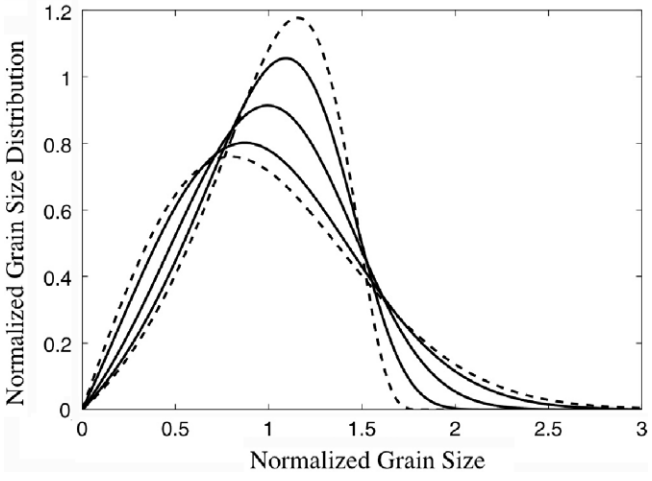

Fig. 2. Normalized grain size distributions. The two limiting cases (non-solid lines) of the Hillert grain size distribution (greatest maximum) and Rayleigh grain size distribution (least maximum) distributions are shown. The solid lines (from top to bottom) are the numerically calculated values for $\alpha=7$ ($\varepsilon=0.017$), $\alpha=5$ ($\varepsilon=0.166$), and $\alpha=2$ ($\varepsilon=0.701$).

where the coefficients b_k and c_k satisfy three-term recurrence relations. The solution f_a decreases faster than x^{-3} and hence will conserve specimen area, while f_b does not. The coefficients b_k and c_k vanish when $\alpha = 0$, in which case Eq. (23) reduces to the Rayleigh grain size distribution.

5. Comparison with numerical simulations

There are several numerical simulations of grain growth by differing techniques. Two examples are examined since the values of α differ greatly with accompanying distinctness in the form of the grain size distribution. Battaile and Holm [14] have utilized a Monte Carlo Potts model to simulate grain growth in two dimensions. Despite starting from two differing morphologies characterized by a Hillert type grain size distribution, the final quasi-static or long-term distributions were the essentially the same. As shown in Fig. 3, the data of Battaile and Holm fits our numerical results when $\alpha = 1$. In this case, the grain size distribution is very similar to the Rayleigh form.

Marthinsen et al. [30] started with a random Voronoi distribution that was allowed to coarsen until a quasi-static distribution was obtained. The comparison with simulated data and numerically calculated results are shown in Fig. 4.

Because of the scatter in the simulation data of Marthinsen et al. [30], two values of α are shown with probably $\alpha = 5$ being a better description. From Fig. 2, for this value of α, the grain size distribution is midway between the Rayleigh and Hillert distributions. From these comparisons, it is quite evident that a noise term is required. From both of these cases, the Hillert grain size distribution is a poor fit of the simulated grain size distribution s. In the latter case, the Rayleigh grain size distribution will also be a poor fit. The effect of the diffusion or noise term is a development of a broader and more left skewed form that is more consistent with calculated and experimental grain size distributions [31].

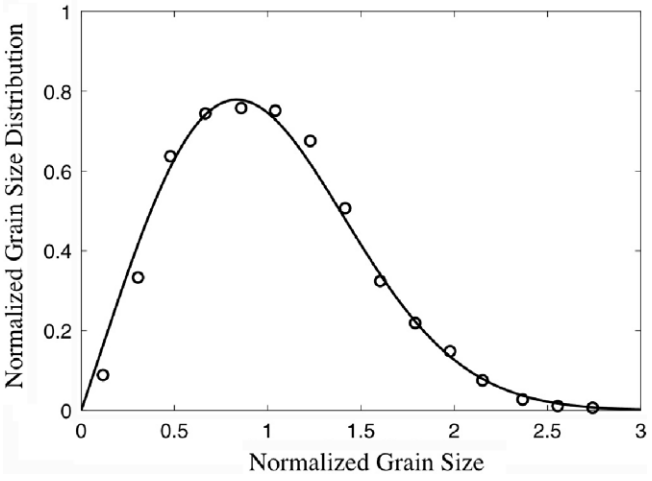

Fig. 3. Comparison of the simulated and numerically calculated grain size distributions. The filled symbols are the simulation data of Battaile and Holm [14]. The solid line is the numerically calculated solution when $\alpha = 1$ and $\varepsilon = 0.967$.

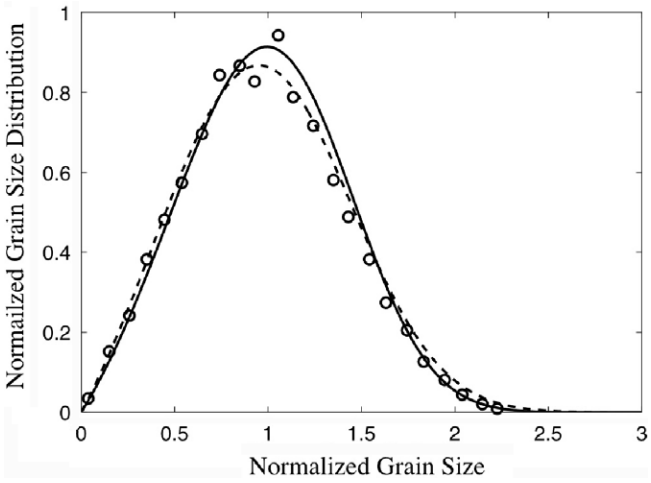

Fig. 4. Comparison with results of Marthinsen et al. [30]. Their data is shown as the open symbols. The numerically calculated results are given for two different values of $\alpha = 4$ and $\alpha = 5$.

At this point we must state that although Hillert's distribution (for both the two-dimensional or three-dimensional case) has never been observed, either experimentally or by computer simulations, several attempts have been made to improve the model. For two-dimensional grain growth, Brandt et al. [32] (cf. also Ref. [33]) and others [34–36] attempted to modify Eq. (4a), while retaining the LSW method, and obtained analytical distributions that were in better agreement with experiments or simulations. An attempt has also been made to modify the LSW procedure while retaining the growth law equation, Eq. (4a) [21]. For three-dimensional grain growth it has been shown that an adequate modification of the effective growth law allows one to obtain an analytical grain size distribution which agrees with simulation data of three-dimensional grain growth fairly well [37].

6. Discussion

The noise term affects the kinetics of grain growth by varying the critical grain size (which in turn determines if a grain will shrink or grow). The drift velocity is usually given as [6]:

$$\frac{dR}{dt} = -a\left(\frac{1}{x} - \frac{1}{x_c}\right), \tag{25}$$

where x_c is the critical size. However, for a more generalized formulation, the drift velocity that is utilized in Eqs. (4a) and (5a) is of the form:

$$\frac{dR}{dt} = -\left(\frac{a}{x} - \frac{b}{\bar{x}}\right), \tag{26}$$

Where $\bar{x}$ is the mean of the grain size distribution and a and b are the constants as in Eq. (4a). Using Eqs. (25) and (26), the critical grain size can be given in terms of $\bar{x}$ as:

$$\frac{x_c}{\bar{x}} = 1 + \frac{\varepsilon}{\alpha}. \tag{27}$$

The critical grain size is equal to one for the Hillert grain size distribution (when $\bar{x} = 1$) and is infinite for the Rayleigh grain size distribution when the values of Fig. 1 are inserted into Eq. (27). For other cases (finite values of both α and ε) the critical size effectively varies between these two limits. This variation of the critical grain size affects the shape of the grain size distribution.

Our formulation for two-dimensional grain growth presented here (see Eq. (11)) employs four constants a, b, λ and D. Out of these constants, a and λ can be determined from first principles [3,4,6]. The constant a involves the mobility of the grain boundary, which is assumed the same for all the grain boundaries in the specimen. The constant λ will depend upon temperature since it controls how rapidly average grain grows with time (see Eq. (9) and Refs. [2,4,6]). The constant b was determined for the constraints of constant specimen size (area) independent of time. That leaves only one unknown constant D. It is interesting to note that the constant D can be obtained from a knowledge of a since a relation exists between α $(=a\lambda^{-2})$, and ε $(=D\lambda^{-2})$ (see Fig. 1). However, in our present treatment we take α as an undetermined independent parameter to be determined by a further analysis of the problem.

This relationship is obtained by constraints imposed on Eq. (13), specifically, that the solutions of this equation must satisfy Eq. (6). In fact, the numerical data obtained in Fig. 1, connecting α and ε, fit the following relationship almost exactly:

$$\varepsilon = \frac{4}{\pi}\left(1 - \frac{\alpha}{8}\right)^2. \tag{28}$$

There has been an attempt in the past [38] to determine D, the strength of the noise, independently. It is interesting and somewhat surprising that D is provided by the formulation itself. It may mean that the source of fluctuations in the growth rate of individual grains arises due to constraints imposed on the system and not due to any special independent source. However, this aspect of the problem needs further experimental and theoretical investigation.

The analysis presented here shows that the distribution function depends only on one adjustable parameter α (it may eventually be possible to obtain α from the physics of the problem). It is true that there are other theories where the distributions have been explained by one adjustable parameter. In that sense our analysis may not appear an advance over other models. Our main object here is to show that the distributions can be obtained by a stochastic formulation, which, in addition, has the potential to explain additional features of grain growth such as scaling, which are not amenable to "mean field" treatments.

It should be noted that, in general, the constant D used in our calculation could be a function of time and grain size. In this work, we have thus taken the strength of the noise as constant. It is remarkable that with this approximation, all the constants, a, b and D, can be exactly determined in terms of one parameter, α.

7. Summary

The inclusion of a noise or diffusive term in the mean field theory leads to grain size distributions that are a better description than the Hillert formulation. This noise term, characterized by the parameter ε, results in a continuous variation of grain size distributions from the Hillert grain size distribution (curvature driven only) to the Rayleigh grain size distribution (diffusive or random walk only). The stochastic model of grain growth falls between these two extremes. We have shown that realistic grain size distributions can be numerically calculated from such a stochastic model of grain growth.

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