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A combined stochastic diffusion and mean-field model for grain growth

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Abstract. A combined stochastic diffusion and mean-field model is developed for a systematic study of the grain growth in a pure single-phase polycrystalline material. A corresponding Fokker-Planck continuity equation is formulated, and the interplay/competition of stochastic and curvature-driven mechanisms is investigated. Finite difference results show that the stochastic diffusion coefficient has a strong effect on the growth of small grains in the early stage in both two-dimensional columnar and three-dimensional grain systems, and the corresponding growth exponents are ~ 0.33 and ~ 0.25, respectively. With the increase in grain size, the deterministic curvature-driven mechanism becomes dominant and the growth exponent is close to 0.5. The transition ranges between these two mechanisms are about 2-26 and 2-15 nm with boundary energy of 0.01-1 J m^{-2} in two- and three-dimensional systems, respectively. The grain size distribution of a three-dimensional system changes dramatically with increasing time, while it changes a little in a two-dimensional system. The grain size distribution from the combined model is consistent with experimental data available.

Keywords: grain growth; stochastic diffusion; curvature-driven process; mean-field model; the finite difference method.

1. Introduction

Most materials, including metals and ceramics, are polycrystalline composed of many small crystalline grains and separated by boundaries or interfaces. The physico-mechanical properties of these materials are closely related to their microstructures, and various structures at different length scales can be formed during grain growth. To improve the physico-mechanical properties and prepare novel materials, many efforts have been made to get a better understanding towards grain growth processes, as reviewed by Tjong and Chen (2004). After Lifshitz and Slyozov's (1961) work, various models have been proposed to describe grain growth, and they can be mainly classified into two categories: deterministic and stochastic models. Both kinds of models have predicated that grain growth follows a power-law kinetics and that their grain size distribution (GSD) approaches a log-normal distribution (Atkinson 1988, Gusak and Tu 2003, Mulheran and

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Harding 1992, Zheng *et al*. 2006a, Zheng *et al*. 2006b). Usually, only one of the deterministic curvature-driven and stochastic diffusion-controlled mechanisms is involved in most models, which makes the models incomplete and introduces discrepancies between experimental observations and theoretical predictions. Recently, an attempt to formulate a combined model involving both mechanisms was made by Helfen *et al.* (2003), but GSDs obtained from the model are still inconsistent with experimental data due to some simple assumptions such as the migration rate of a grain boundary being proportional to its curvature. Here, we propose a special form of continuity equation for grain growth to perform a systematic study of the grain growth in two- and three-dimensional systems, and to explore the competition and interplay between both kinds of mechanisms.

2. Continuity equations

To derive the continuity equations for grain growth, two general assumptions are usually made: (1) Large grains grow by consuming surrounding small grains during grain growth and (2) grains may be classified into groups based on their sizes at a given time. Then the growth process may be described effectively as the change of GSD with time. For a deterministic curvature-driven process, the overall flux $j_d(R, t)$ caused by a driving force is (Atkinson 1988)

$$j_d(R, t) = v(R, t) f(R, t) \tag{1}$$

where $f(R, t)$ is the probability that a grain has a size between R and $R + dR$, and $v(R, t)$ is the drift (or grain boundary) velocity. Here, the change of grain density is due to the difference between incoming and outgoing overall fluxes of grains (Atkinson 1988), namely, $\partial f(R, t) / \partial t = - \partial j(R, t) / \partial R$. Therefore, the continuity equation for the deterministic process is

$$\frac{\partial f(R, t)}{\partial t} = -\frac{\partial}{\partial R}[v(R, t) f(R, t)] \tag{2}$$

The key problem is to determine the exact expression of the drift term, $v(R, t) = dR/dt$. Following the analysis of Gusak and Tu (2003), we consider an m-dimensional system consisting of N grains, in which a "central" grain of size R is embedded in a grain "reservoir" of all others, as depicted in Fig. 1. Assuming that the grain boundary energy is isotropic, then, the total free energy of GBs of the system is

$$F = \frac{1}{2} q \gamma \left(R^{m-1} + \sum_{i=2}^{N} R_i^{m-1} \right) \tag{3}$$

where γ is the GB energy per unit length/area, q a geometrical factor and R_i the size of the i-th grain. Introducing a characteristic size R_c, which is proportional to the average grain size of the system, the total free energy in a normalized size space is

$$\tilde{F} = \frac{1}{2} q \gamma [\tilde{R}^{m-1} + (N-1) < \tilde{R}_i^{m-1} >] \tag{4}$$

where $\tilde{R}_i = R_i / R_c$ is the normalized size of the i-th grain and $<\cdot>$ denotes the average value of a given variable over the size space. Since the ratio of the average grain size to the characteristic size is approximately a constant, the second right-side term in Eq. (4) becomes constant (Gusak and Tu 2003). Then the pressure and the velocity in the normalized size space are

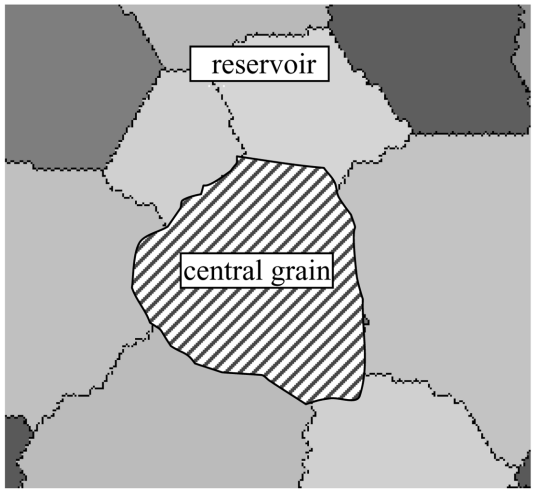

Fig. 1 Sketch map of a single-phase system, in which a "central" grain is embedded in a "reservoir" composed of all others.

$$\tilde{P} = -\frac{\partial \tilde{F}}{\partial \tilde{U}} = -\frac{q\gamma}{4\pi}\frac{1}{\tilde{R}} \tag{5}$$

and

$$\frac{d\tilde{R}}{dt} = \tilde{M}\tilde{P} \tag{6}$$

where $\tilde{U}$ is the normalized area/volume and $\tilde{M}$ the isotropic GB mobility in the normalized space. From a simple dimension analysis, it can be found that the real mobility M and the normalized mobility $\tilde{M}$ follow the relationship: $\tilde{M} = M/R_c^2$ (Gusak and Tu 2003). Thus, Eq. (6) becomes

$$\frac{d\tilde{R}^2}{dt} = -\frac{k}{R_c^2} \tag{7}$$

where $k = q \cdot \gamma \cdot M / 2\pi$. In addition, from the conservation of total volume/area, we have

$$\frac{d\ln R_c^2}{dt} = k\frac{\langle R^{m-2}\rangle}{\langle R^m\rangle} \tag{8}$$

Then the real space velocity can be derived from Eqs. (7) and (8) to take the form of

$$\frac{dR}{dt} = \frac{k}{2}\left(\frac{R\langle R^{m-2}\rangle}{\langle R^m\rangle} - \frac{1}{R}\right) \tag{9}$$

Substituting Eq. (9) into Eq. (2), the continuity equation for the deterministic curvature-driven grain growth process becomes]

$$\frac{df}{dt} = -\frac{\partial}{\partial R}\left[\frac{k}{2} f\left(\frac{R\langle R^{m-2}\rangle}{\langle R^m\rangle} - \frac{1}{R}\right)\right] \tag{10}$$

The corresponding boundary conditions are that, at any time, $f(R, t)$ and $R(\partial f / \partial t)$ vanish in the

limits of $R \to 0$ and $R \to \infty$ (Helfen *et al*. 2003, Mulheran and Harding 1992), and the stationary solution is (Gusak and Tu 2003)

$$f(R,t) = \frac{C}{(k_1 t)^{(m+1)/2}} \frac{R}{(k_1 t)^{1/2}} \exp\left(-\frac{R^2}{k_1 t}\right) \tag{11}$$

where C is a constant, $k_1 = (2/3)^{m-2}k$, and $\langle R\rangle_{GT} = (\pi k_1 t)^{1/2}/2$. In another normalized space, $u = R/\langle R\rangle$, Eq. (11) is a Rayleigh-type distribution function

$$P_{GT}(u) = \frac{\pi}{2} u \exp\left(-\frac{\pi}{4}u^2\right) \tag{12}$$

where the subscript GT represents that the GSD is derived based on the Gusak and Tu's analysis (Gusak and Tu 2003).

On the other hand, grain growth may be considered as a stochastic diffusion-controlled process, namely, all grains undergo purely random walks in the grain size space. The overall flux is assumed to be proportional to the gradient of the probability (Mulheran and Harding 1992) as follows:

$$j_s(R,t) = -B(R,t)\frac{\partial f(R,t)}{\partial R} \tag{13}$$

where $B(R,t)$ is a diffusion term. Louat (Atkinson 1988) assumed that $B(R,t) = D$ (the diffusion coefficient), and obtained a Rayleigh distribution that violates area/volume conservation (Helfen *et al*. 2003). To ensure this conservation condition, a special diffusion term, $B(R,t) = D/R^{m-1}$, which was proposed by Mulheran and Harding (1992), is adopted here to model two- and three-dimensional grain growth. Then the continuity equation corresponding to Eq. (13) is

$$\frac{\partial f}{\partial t} = \frac{\partial}{\partial R}\left(\frac{D}{R^{m-1}}\frac{\partial f}{\partial R}\right) \tag{14}$$

In a normalized space, $\hat{u}$, the stationary solution, P_{MH}, is

$$P_{MH}(\hat{u}) = K\hat{u}^m \exp\left(-\frac{K}{m+1}\hat{u}^{m+1}\right) \tag{15}$$

with

$$K = (m+1)\left(\frac{\Gamma[2m/(m+1)]}{\Gamma[(2m-1)/(m+1)]}\right)^{m+1}$$

where the subscript MH represents that the GSD is derived based on Mulheran and Harding's assumption (Mulheran and Harding 1992), $\Gamma(\cdot)$ is the Gamma function, $\hat{u} = u$ for $m = 2$ and $\hat{u} = u/\{\Gamma(3/2)\cdot[\Gamma(5/4)]^{-2}\}$ for $m = 3$. The corresponding grain growth kinetics is: $\langle R\rangle_{MH} = \Gamma[(m+2)/(m+1)][(m+1)^2 D]^{1/(m+1)} t^{1/(m+1)}$.

Actually, grain growth is a very complex process and many different mechanisms may operate during the process. In the pure curvature-driven process, grains with the same size grow in the same manner. This assumption is obviously too simple since equal-size grains have different local environments, which may introduce flux fluctuations during the course of growth. Thus, both curvature-driven and diffusion-controlled mechanisms should be taken into account simultaneously.

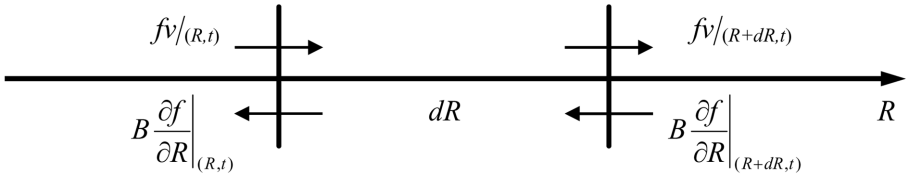

Fig. 2 A sketch of the overall flux of the grain number in the range of R to $R + dR$.

As illustrated in Fig. 2, the overall flux becomes a summation of the two distinct parts,

$$j(R,t) = v(R,t)\,f(R,t) - B(R,t)\frac{\partial f(R,t)}{\partial R} \tag{16}$$

and the corresponding Fokker-Planck continuity equation can be written as,

$$\frac{\partial f}{\partial t} = -\frac{\partial}{\partial R}\left[\frac{k}{2} f\left(\frac{R\langle R^{m-2}\rangle}{\langle R^m\rangle} - \frac{1}{R}\right)\right] + \frac{\partial}{\partial R}\left(\frac{D}{R^{m-1}}\frac{\partial f}{\partial R}\right) \tag{17}$$

3. Finite difference scheme

To solve Eq. (17), we will resort to a finite difference scheme since it is difficult to derive an analytical solution. Noting that the grain size is in the range of 0 to $+\infty$, the form of the continuity equation should be modified to overcome the difficulties from the integration among an infinite range. This can be done by transforming Eq. (17) to a reduced space, $y = R/(R+1)$, then region $R \in (0, \infty)$ becomes $y \in (0, 1)$. Thus, we obtain

$$\frac{\partial g(y,t)}{\partial t} = b \cdot g(y,t) + c \cdot \frac{\partial g(y,t)}{\partial y} + d \cdot \frac{\partial^2 g(y,t)}{\partial y^2} \tag{18}$$

with

$$y_m = \left[1 + \frac{\int_0^1 (1-y)^{-2} g(y,t)\,dy}{\int_0^1 y^m (1-y)^{-m-2} g(y,t)\,dy}\right]^{-1}$$

$$a = \frac{y_{m-2}}{1-y_{m-2}} \cdot \frac{1-y_m}{y_m}$$

$$b = -\frac{k}{2}(1-y)^2\left(\frac{a}{(1-y)^2} + \frac{1}{y^2}\right)$$

$$c = -\frac{k}{2}(1-y)^2\left(a \cdot \frac{y}{1-y} - \frac{1-y}{y}\right) - (m+2y-1)D\frac{(1-y)^{m+2}}{y^m}$$

$$d = D\frac{(1-y)^{m+2}}{y^m}(1-y)y$$

The boundary conditions in the reduced space are $g(0, t) = g(1, t) = 0$.

Here, a backward Euler difference scheme in temporal domain and a second-order difference scheme in spatial domain are adopted to solve the equation (Press *et al.* 1992). The corresponding discrete equations are

$$\frac{g_j^{l+1} - g_j^l}{\Delta t} = b \cdot g_j^{n+1} + c \cdot \frac{g_{j+1}^{l+1} - g_{j-1}^{l+1}}{2\Delta y} + d \cdot \frac{g_{j+1}^{l+1} - 2g_j^{l+1} + g_{j-1}^{l+1}}{(\Delta y)^2} \tag{19}$$

where Δt is the time interval and Δy is the grid spacing. The superscript l denotes time step and subscript j denotes the grid node index in the reduced size space. According to von Neumann's stability analysis (Press *et al.* 1992), the amplification factor for Eq. (19) is

$$\zeta = \frac{1}{1 - b\Delta t + \dfrac{4d\Delta t}{(\Delta y)^2}\sin^2\dfrac{k\Delta y}{2} - i\dfrac{c\Delta t}{\Delta y}\sin(k\Delta y)} \tag{20}$$

Here i is the imaginary unit. It can be easily found that $b < 0$ and $d > 0$, then $|\xi| < 1$ for any time interval Δt, which implies that the implicit difference scheme is unconditionally stable. In the following calculations, we choose time interval $\Delta t = 10^{-8}$ and grid spacing $\Delta y = 10^{-6}$.

4. Results and discussion

Generally, the growth kinetics can be described by a power law, $\langle R\rangle^{1/n} - \langle R_0\rangle^{1/n} = H^{1/n} \cdot t$, where n is referred to as the growth exponent, $\langle R_0\rangle$ the initial average grain size, $\langle R\rangle$ the average grain size at time t and H a constant dependent on temperature (Atkinson 1988). In the case of $\langle R\rangle \gg \langle R_0\rangle$, the kinetics becomes $\langle R\rangle = H \cdot t^n$.

To investigate the effect of the stochastic diffusion-controlled and the deterministic curvature-driven mechanisms on the growth kinetics, we vary one of the two parameters, D and k, keeping another one unchanged. For two-dimensional systems, e.g., columnar polycrystalline materials, the growth kinetics are shown in Fig. 3 for various D and k values. From Fig. 3(a), it can be seen that the diffusion coefficient has a strong effect on the growth of small-size grains in the early stage. With the decrease in diffusion coefficient, the kinetics curve approaches the analytical curve of a deterministic growth process with $k = 2$, which is denoted as the solid line in Fig. 3(a). Fig. 3(b) shows the effect of the curvature-driven mechanism on the grain growth in two-dimensional systems with a constant diffusion coefficient of 10^{-2}. From these curves, it can be seen that the curvature-driven mechanism has an obvious effect on the growth of large-size grains in the later stage. For all studied two-dimensional cases, the growth kinetics curves have a bilinear characteristic in the double-logarithm plots and the corresponding growth exponents in the two distinct stages are ~ 0.33 and ~ 0.5, respectively.

Fig. 4(a) shows the effect of the diffusion-controlled mechanism on the grain growth kinetics of three-dimensional systems with $k = 2$. Square, circle and triangular symbols are the kinetics curves corresponding to $D = 10^{-4}$, $D = 10^{-2}$ and $D = 1$. It can be found that, at the large size region, these curves approach the solid line, which is the kinetics ($\langle R\rangle_{GT} \sim t$) of a pure deterministic three-dimensional growth process. These curves are separated in the small size region while they are superposed in the large one. This indicates that the diffusion-controlled mechanism is dominant for small grains. Fig. 4(b) shows the effect of the curvature-driven mechanism on the grain growth

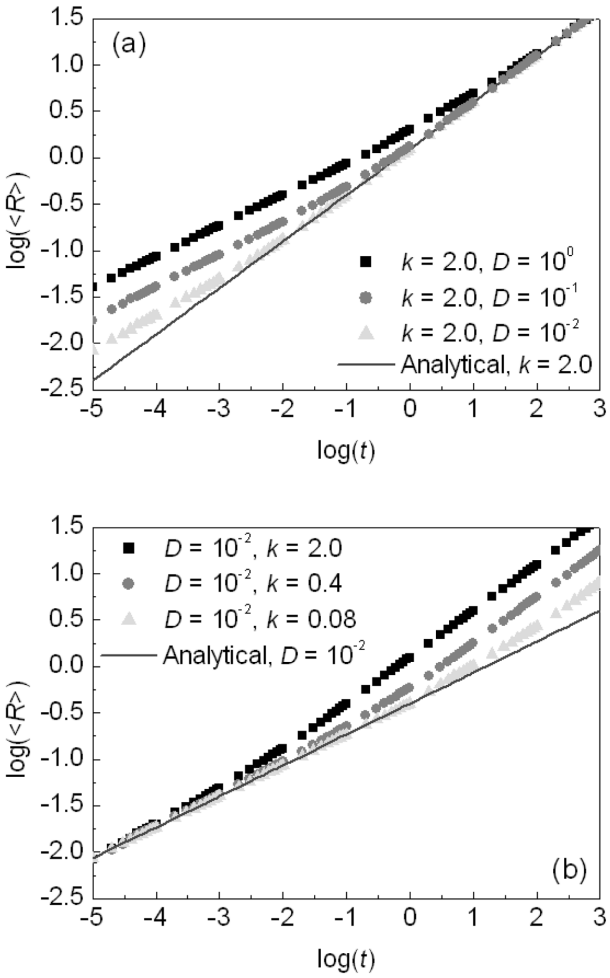

Fig. 3 Effect of (a) the diffusion-controlled and (b) the curvature-driven mechanisms on the grain growth kinetics in two-dimensional systems. The solid lines in (a) and (b) are analytical solutions for the deterministic curvature-driven process with $k = 2$ and the stochastic diffusion-controlled process with $D = 10^{-2}$, respectively.

kinetics of three-dimensional systems with $D = 10^{-4}$. Contrarily, these kinetics curves plotted in Fig. 4(b) are separated in the large size region while they are superposed in the small one, which implies that the curvature-driven mechanism is dominant for large grains. In these three-dimensional cases, it can be also seen that the kinetics curves have a bilinear characteristic in the double-logarithm plots and the corresponding slops are ~ 0.25 and ~ 0.5 for small and large grain size regions, respectively.

From the above analysis, it can be found that small grain growth is dominated by the stochastic mechanism and the exponents approach 1/3 and 1/4, exact values for two- and three-dimensional systems, respectively (Mulheran and Harding 1992); and the curvature-driven mechanism becomes dominant for large grains and the exponent closes to 1/2, a theoretical result for both two- and three-dimensional systems obtained by Gusak and Tu (2003). These different dominative

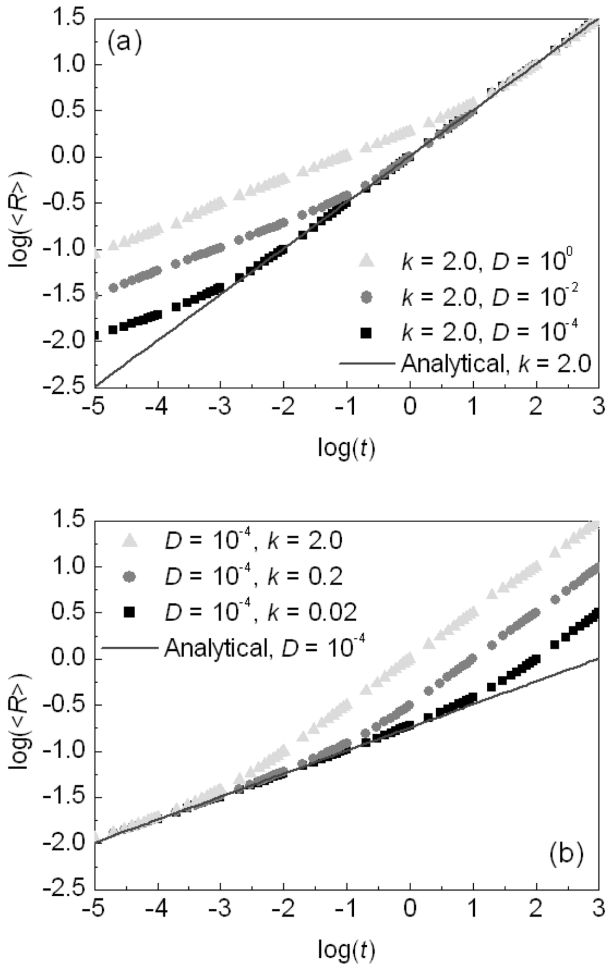

Fig. 4 Effect of (a) the diffusion-controlled and (b) the curvature-driven mechanisms on the grain growth kinetics in three-dimensional systems. The solid lines in (a) and (b) are analytical solutions for the deterministic curvature-driven process with $k = 2$ and the stochastic diffusion-controlled process with $D = 10^{-4}$, respectively.

mechanisms for different stages are mainly due to different effects of atomic jump on the grain growth (Zheng *et al*. 2006b). For a single-phase system, various experimental results show that the growth exponents vary from 0.25 to 0.5, that is, they were obtained at different stages of grain growth (Atkinson 1988).

As shown in Figs. 3(a) and 4(a), the average grain size decreases as decreasing the diffusion coefficient at a given growth time for various systems, which indicates that diffusion motions of atoms can enhance the growth of grains. From Figs. 3(b) and 4(b), the curvature-driven boundary migration has a similar enhancement effect on the grain growth as the diffusion motion. It is well known that impurities have very strong effects on grain growth, they can reduce the mobility of grain boundaries (i.e. the curvature-driven coefficient k) (Atkinson 1988). From Figs. 3(b) and 4(b), we can see that it will take a longer time for a system to grow to a specified average grain size from the same initial configuration with decreasing the curvature-driven coefficient, which reveals

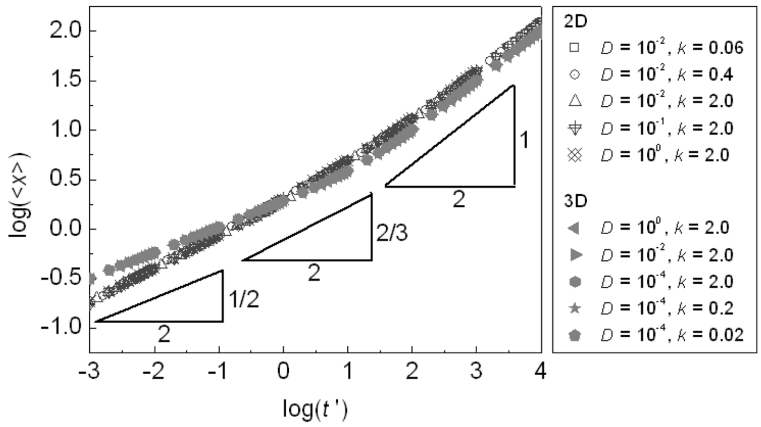

Fig. 5 The scaling laws in two- and three-dimensional systems: the average grain sizes <*R*> are scaled as $x = (k / 2D)^{1/(m-1)}R$ and plotted as functions of scaled time $t' = (k^{m+1} / 2^{m+1}D^2)^{1/(m-1)}t$.

that the grain growth is inhibited due to the presence of impurities and is consistent with experimental observations (Atkinson 1988).

In Fig. 5, the average grain sizes of different systems are scaled as $x = (k / 2D)^{1/(m-1)}R$ and plotted as functions of scaled time $t' = (k^{m+1} / 2^{m+1}D^2)^{1/(m-1)}t$. This figure shows the collapse on a single curve of five data sets corresponding to various D and k parameters for both two- and three-dimensional growth processes. This indicates that there are scaling laws in general grain growth processes and the results are independent of curvature-driven and diffusion coefficients in the scaled size-time space, which is similar with the results of Helfen *et al.* (2003) obtained from another combined model and may be used to determine the grain growth stage in experiments and to extrapolate the material parameters. The bilinear curves in Fig. 5 show two transition regions from the stochastic to the deterministic kinetics and the corresponding bounds are 3.0-6.5 and 3.5-4.3 in the scaled size space for two- and three-dimensional systems, respectively. Based on the Einstein-Nernst relationship, the real space transition grain size, R_{tr}, relates to the scaled one, x_{tr}, according to $R_{tr} = 2x_{tr}(\pi K_B T / q\gamma)^{0.5}$, where K_B is the Boltzmann constant (Helfen *et al.* 2003, Zheng *et al.* 2006b). Then the real space transition regions can be estimated to be 2-26 and 2-15 nm for two- and three-dimensional systems with GB energy of 0.01-1 J m^{-2}, respectively. However, a much larger transition size may be obtained in experiments due to fast grain growth induced by high stress and large plastic deformation during cutting and rolling processes (Zheng *et al.* 2006b).

Fig. 6 shows the GSD evolution at various times in two- and three-dimensional systems with $k = 2$, $D = 10^{-2}$ and $k = 2$, $D = 10^{-4}$, respectively. In the early growth stage, the GSDs in both systems are narrow and approach the theoretical solutions P_{MH} ($m = 2$) and P_{MH} ($m = 3$). With the increase in time, the GSDs become broader and broader, and are close to the GSD of a pure curvature-driven system finally. However, it can be seen obviously that the GSD has a dramatic change in a three-dimensional system, while it changes smoothly in a two-dimensional one, which may be attributed to the additive geometrical constraints in a lower dimensional system.

Fig. 7 shows a comparison of the GSD from present work with those from experimental data and Helfen *et al.*'s results in a three-dimensional grain system. It can be seen that the result obtained from the combined model is in good agreement with experiment data on grain growth in an iron system (Zhang *et al.* 2004), which validates the proposed model.

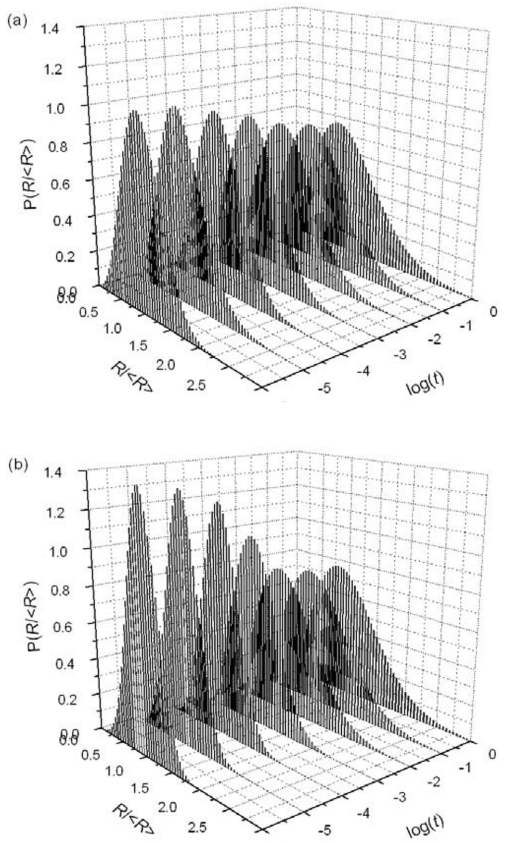

Fig. 6 Evolution of the grain size distribution at various times in (a) two- and (b) three-dimensional systems with $k = 2$, $D = 10^{-2}$ and $k = 2$, $D = 10^{-4}$, respectively.

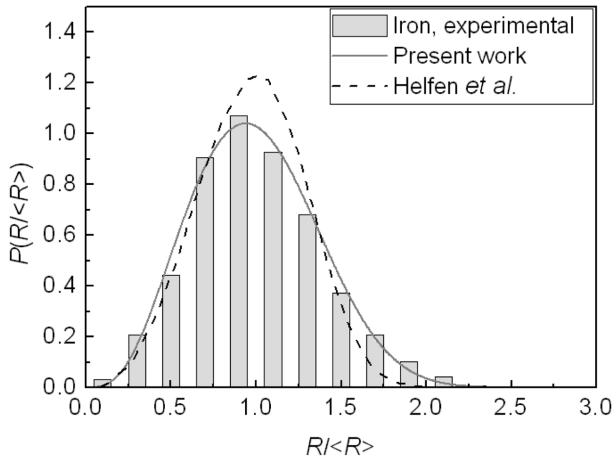

Fig. 7 Grain size distributions obtained from theoretical models and experiments, where bars are experimental data (Zhang *et al*. 2004), dashed and solid curves are the three-dimensional results obtained from Helfen *et al*. (2003) and combined model, respectively.

5. Conclusions

Based on the diffusion-controlled and curvature-driven mechanisms, a combined model for grain growth in a single-phase system has been developed. An implicit finite difference scheme has been used to integrate the corresponding continuity equation that is discretized in spatial domain with a second-order finite difference scheme. The growth kinetics obtained from this model reveals that there exist scaling laws in general growth processes. It has been found that the stochastic diffusion-controlled and deterministic curvature-driven mechanisms are dominant for small and large grains, respectively. The transition sizes between these two mechanisms are 2-26 and 2-15 nm in two- and three-dimensional systems, respectively, for most metals. Good agreement between predicted GSD and experimental data available verifies the proposed combined model. From our knowledge, however, it should be noted that there isn't available experimental data in the opening literatures that can be used to validate these scaling laws, and an integrated experimental, analytical and computational effort is required to improve the proposed model-based simulation procedure.

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