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The kinetics of normal grain growth

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The relationship between the quasistationary distribution functions in normal grain growth and the corresponding grain-growth velocities is investigated. The restrictions imposed by volume conservation lead to a simple differential equation which describes quasistationary grain growth. This equation allows us to express the reduced growth velocity $(dR/dt)/(dR^*/dt)$, where R^* is the scaling grain size, in terms of the corresponding distribution function and to express the distribution function by means of the reduced growth velocity. General conclusions about the shape of distribution functions can be drawn from these expressions.

1. Introduction

During normal grain growth, large grains increase while small grains decrease in size. Since the volume of the system is constant, this results in an increase in the mean grain size and a decrease in the number of grains in the system. Formally, normal grain growth may thus be looked upon as the motion of individual grains in grain size–time–space. The flux j of grains in this space is caused by a diffusion-like process or by a driving force, thus:

$$j = -D\frac{\partial f^0}{\partial R} + f^0 v^0 \tag{1}$$

where f^0 is the distribution function which is a function of size (R) and time (t), v^0 is the drift velocity due to a driving force F, and D is formally identical to a diffusion coefficient, which only depends upon the specific grain-boundary mobility. The continuity of the grain flux in size–time–space gives:

$$\frac{\partial f^0}{\partial t} = \frac{\partial}{\partial R}(-j) = \frac{\partial}{\partial R}\left(D\frac{\partial f^0}{\partial R}\right) - \frac{\partial}{\partial R}(f^0 v^0). \tag{2}$$

This expresses that a change in the number of grains in a size-class (R) is caused by a flux from neighbouring size-classes.

Louat [1] stressed the diffusional character of the process, and put v^0 equal to zero. He also argued that D is virtually independent of f^0, R and t and arrived at the diffusion equation:

$$\frac{\partial f^0}{\partial t} = D\frac{\partial^2 f^0}{\partial R^2}, \tag{3}$$

with the boundary condition $f^0(0, t) = 0$, $f^0(\infty, t) = 0$. The volume conserving solution to this diffusion problem is:

$$f^0(R, t) = A \cdot R\left[\exp -\left(\frac{R}{k}\right)^2\right], \tag{4}$$

where A is a normalization constant, which varies with time, and k is another time-dependent parameter.

Beck [2], Smith [3], Feltham [4], Hillert [5] and others, assumed more or less implicitly that the drift of grains in grain size–space is the dominating factor during normal grain growth, and that the driving force F is in some way related to the elimination of grain-boundary area. Hillert's analysis takes as the starting point:

$$\frac{\partial f^0}{\partial t} + \frac{\partial}{\partial R}(f^0 v^0) = 0, \tag{5}$$

which, of course, is obtained by neglecting the contribution from diffusion to the flux ($D = 0$). Two different approaches are now possible for further analysis. One can start with a particular expression for the drift velocity v^0, and then solve for f^0. This analysis also results in expressions for the variation with time of the mean grain size, which is an important parameter. This is Hillert's approach [5]. He argued for the following relation-

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ship between the drift velocity and the size of the grains:

$$v^0 = \gamma\left(\frac{1}{R_{cr}} - \frac{1}{R}\right), \tag{6}$$

where γ is a factor which depends on the energy and mobility of the grain boundary, both of which were assumed to be independent of R. R_{cr} is a critical grain size, and varies with time. The kinetics of normal grain growth now becomes identical with the kinetics of Oswald ripening with the interphase reactions as the rate-controlling reactions. This process has been analysed in detail by Lifshitz and Slyozov [6] and Wagner [7], and Hillert followed their analysis closely.

The assumed relationship between drift velocity and grain size (Equation 6), which is quite essential in this analysis can hardly be checked experimentally, and only values of v^0 at some limiting values of R can be discussed with reference to experiments. This is a serious limitation. The alternative approach is to take f^0 as the fundamental input in the analysis, as this can be determined experimentally. By means of Equation 5 one can determine v^0 and also the mean growth rate. From a knowledge of v^0 at large values of R, interesting conclusions can be drawn as to the shape of f. An approach along these lines was first tried by Feltham [4], and will be further developed in the present work.

2. Theory and results

In this section we develop a formalism which allows us to calculate v^0 from a given grain-size distribution f^0. A necessary condition is that the distribution is quasistationary, which means that it can be made time-invariant when appropriately scaled. If the following scaling is used

$$\rho = \frac{R}{R^*(t)}$$

$$f(\rho) = R^{*4}(t) f^0(R, t)$$

and

$$v(\rho) = v^0(R,t)/(\mathrm{d}R^*/\mathrm{d}t) = (\mathrm{d}R/\mathrm{d}t)/(\mathrm{d}R^*/\mathrm{d}t),$$

it is shown in Appendix 1 that $f(\rho)$ obeys the following equation:

$$4f(\rho) + \rho\frac{\mathrm{d}f(\rho)}{\mathrm{d}\rho} - \frac{\mathrm{d}}{\mathrm{d}\rho}[f(\rho)\cdot v(\rho)] = 0. \tag{7}$$

The scaling grain size $R^*(t)$ can be chosen in a number of ways but, as shown later, it is convenient to chose $R^* = R_{cr}$, where R_{cr} is the grain size at which v^0 is zero. Equation 7 can now be solved for either v or f (Appendix 2):

$$v(\rho) = \rho - 3\frac{\int_\rho^\infty f(x)\,\mathrm{d}x}{f(\rho)} \tag{8a}$$

$$f(\rho) = \frac{v_0 f_0}{v-\rho}\exp\left(-3\int_0^\rho \frac{\mathrm{d}x}{x-v}\right), \tag{8b}$$

where $v_0 f_0$ is the rate of loss of grains from the system. If the expression for v_0 given by Equation 6 is appropriately scaled and used in Equation 8b, one obtains the Hillert distribution. If, instead, an experimentally determined distribution function is put into Equation 8a, v can be determined as a function of ρ. In Fig. 1 the variation of v with ρ, as calculated from Equation 8a is shown for several proposed grain-size distribution functions.

The log normal distribution function

$$f^0 = k_1\frac{1}{R}\exp\left\{-\left|\frac{1}{k_2}\ln\frac{R}{\bar{R}}\right|^2\right\} \tag{9}$$

where k_1 is the normalization constant, $k_2 = \sqrt{2}\ln\sigma$ where σ is the geometric width of the distribution, and $\bar{R}$ is the geometric mean grain size has been claimed by several investigators [4, 8] to be the distribution function obtained experimentally. After applying Equation A4 to find R_{cr}, Equation 8a is used to find v. The integration can be done analytically and gives:

$$v(\rho) = \rho - \frac{3k_2\frac{\sqrt{\pi}}{2}\left[1-\mathrm{erf}\left(\frac{1}{k_2}\ln\frac{\rho}{\bar{\rho}}\right)\right]}{\frac{1}{\rho}\exp\left|-\frac{1}{k_2}\ln\frac{\rho}{\bar{\rho}}\right|^2} \tag{10}$$

where $\bar{\rho} = \bar{R}/R_{cr}$.

Aboav and Langdon [9] found a better fit of experimental data to the function

$$f^0 = A_1\exp\{-A_2|\sqrt{R} - \sqrt{A_3}|^2\}. \tag{11}$$

A procedure similar to the one carried out for the log normal distribution function gives:

$$v(\rho) = \rho - \frac{3}{A_2^*} - 3\frac{\sqrt{\bar{\rho}}\sqrt{\pi}}{\sqrt{A_2^*}},$$

$$\frac{1-\mathrm{erf}\,(\sqrt{A_2^*}(\sqrt{\rho}-\sqrt{\bar{\rho}}))}{\exp\,(-A_2^*(\sqrt{\rho}-\sqrt{\bar{\rho}})^2}. \tag{12}$$

Here $A_2^* = A_2 R_{cr}$.

In both cases $\bar{\rho}$ will depend upon the width of the distribution function. The graphs in Fig. 1 are

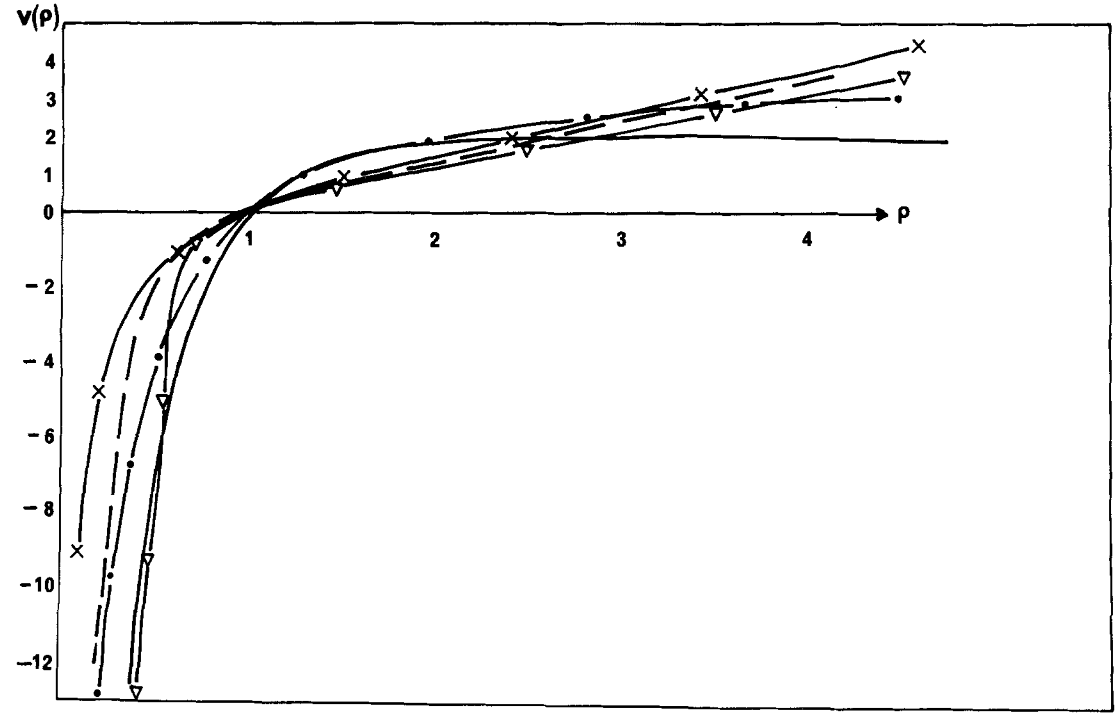

Figure 1 Reduced drift velocity as a function of $\rho = R/R_{cr}$ for several distribution functions. (1) The Hillert-distribution: –•–•–; (2) Log normal distribution, width $\sigma = 1.4$: –▿–▿–; (3) Louat-distribution: –X –X –; (4) Aboav distribution: – – – –; (5) Feltham-distribution: ———.

meant to represent typical experimentally observed widths.

Also included in Fig. 1 is the distribution function found by Louat by considering the grain-growth process as a diffusional process (Equation 4). In this case Equation 8a yields:

$$v(\rho) = \left(\rho - \frac{3k^{*2}}{2}\frac{1}{\rho}\right), \tag{13}$$

where $k^* = K/R_{cr}$ (see Equation 4).

3. Discussion and conclusions

Feltham [4] first tried to derive a drift velocity from a distribution function. Unfortunately, the drift velocity arrived at by him does not give a log normal distribution function as claimed. This can be demostrated by inserting his expression for v into Equation 8b. In fact, his solution is only the first term in a series expansion which gives the complete solution to a Fredholm integral equation. Using the procedure outlined by Hillert to determine the unknown constant in the reduced growth rate, one finds:

$$v(\rho) = 2e\frac{1}{\rho}\ln\rho, \tag{14}$$

and the corresponding distribution function

$$f(\rho) = \frac{f_0 v_0 \rho}{\rho^2 - 2e\ln\rho}\exp\left\{-3\int_0^{\rho^2}\frac{dx}{x - e\ln x}\right\}. \tag{15}$$

This distribution function is shown in Fig. 2. In contrast to the log normal distribution function which has a tail extending to infinity, the correct solution to Feltham's drift velocity shows a cut-off at $\rho = \sqrt{e}$ and the nature of this cut-off is quite similar to the cut-off in the Hillert distribution. We also find that the square of the critical radius increases linearly with time.

As can be seen from Equations 10, 12 and 13 and [4], all the distributions functions except the one given by Hillert give drift velocities that go to infinity or zero for large grain sizes. No driving forces can be imagined that give such drift velocities for large grain sizes. In fact, all distribution functions corresponding to drift velocities limited for large grain must have a cut-off. This can be demonstrated as follows: assume that the drift velocity is limited for large values of ρ and approaches some constant value. If v is put equal to a constant K in Equations 7 or 8b for ρ greater than a, one

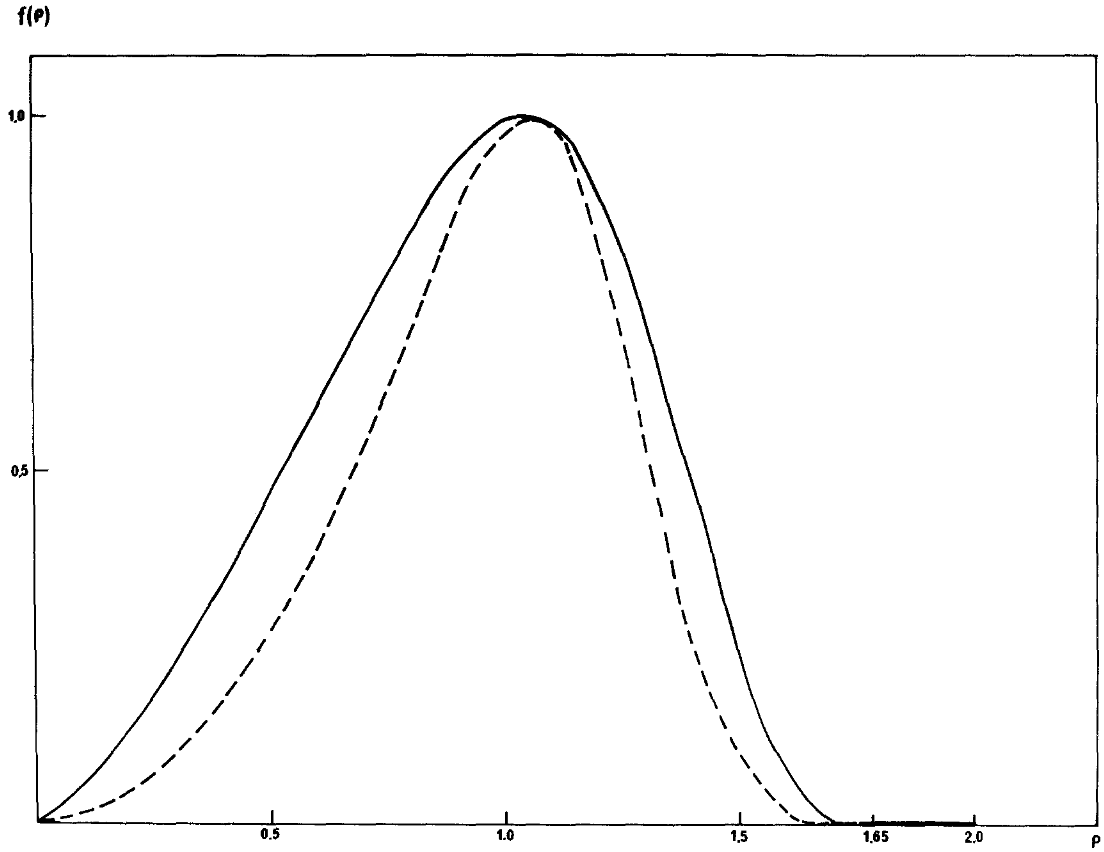

Figure 2 Quasistationary distribution functions as a function of $\rho = R/R_c$: – – – – – Feltham's distribution; ——— Hillert's distribution.

obtains

$$f(\rho) = \frac{v_a f_a}{(\rho - K)^4}. \quad (16)$$

It thus appears as if a distribution with a tail falling off as $1/\rho^4$ is an acceptable solution. However, as shown in Appendix 3, volume conservation places the following restrictions on the distribution functions:

$$\lim_{\rho \to \infty} \rho^3(\rho - v)f(\rho) = 0. \quad (17)$$

Equation 16 does not satisfy this requirement. Hence we conclude that all distribution functions corresponding to a limited drift velocity for large values of ρ have a cut-off. Alternatively, this means that all distribution functions with a tail stretching to infinity cannot have been developed by an Ostwald-ripening process. By a procedure similar to the one used by Lifshitz and Slyozov to determine the growth-rate constant, it can be shown that the v-curves in Fig. 2 must touch the line $v = \rho$ and the cut-off occurs at the grain size of the touching point. As is seen in Fig. 1, only the Hillert and Feltham drift velocities give rise to a cut-off.

The Hillert distribution is much more peaked than the distribution found experimentally. This means that the driving force suggested by him levels off too rapidly with increasing ρ. If the diffusional character of grain growth is a contributing factor, this would bring about a levelling off of the most peaked part of the distribution because of the great gradient in the distribution function in that region. This approach is not followed further here.

We have recently derived quasistationary distribution functions by computer simulation of grain growth [10]. This also resulted in a rather peaked distribution with a cut-off. In this first modelling the contact area between grains was modelled in a rather crude way, and it is hoped that when the contact area is better accounted for, and when the model grains are packed in a real three-dimensional fashion, the resulting quasistationary distribution will be in better agreement with experiment than at present.

Appendix 1

The continuity equation reads

$$\frac{\partial f^0}{\partial t} = \frac{\partial}{\partial R}(f^0 \cdot v^0). \tag{A1}$$

We introduce the following scaling

$$f(\rho) = R^*(t)^4 \cdot f^0(R, t)$$

$$v(\rho) = v^0(R, t)/(\mathrm{d}R^*/\mathrm{d}t) = \frac{\mathrm{d}R/\mathrm{d}t}{\mathrm{d}R^*/\mathrm{d}t} \tag{A2}$$

$$\rho = R/R^*(t).$$

The first of these follows directly from volume conservation of a quasistationary distribution function. When introducing these variables into Equation A1 it transforms to

$$4f(\rho) + \rho \frac{\mathrm{d}f(\rho)}{\mathrm{d}\rho} - \frac{\mathrm{d}}{\mathrm{d}\rho}[f(\rho) \cdot v(\rho)] = 0. \tag{A3}$$

Given an experimentally determined grain-size distribution function $f(R)$, the critical grain size R_{cr} and thus ρ is found from Equation 8a:

$$v(\rho = \rho_{cr} = 1) = 1 - 3\frac{\int_1^\infty f(x)\,\mathrm{d}x}{f(1)} = 0$$

thus

$$R_{cr} = 3\frac{\int_{R_{cr}}^\infty f(R)\,\mathrm{d}R}{f(R_{cr})}. \tag{A4}$$

Equation A4 can be solved numerically when $f(R)$ is known.

Appendix 2

Equation 7 can be rewritten in the following way:

$$3f + \frac{\mathrm{d}}{\mathrm{d}\rho}(\rho \cdot f - f \cdot v) = 0 \tag{A5}$$

This equation can now be integrated directly to give Equation 8a. Equation 5 can also be rewritten in the following way:

$$\frac{\mathrm{d}f/\mathrm{d}\rho}{f} = -\frac{(\mathrm{d}/\mathrm{d}\rho)(\rho - v)}{\rho - v} - \frac{3}{\rho - v}.$$

Multiplying by $\mathrm{d}\rho$ and integrating from $f(\rho = 0) = f_0$ to $f(\rho)$ gives

$$\ln\frac{f}{f_0} = -\ln\frac{v - \rho}{v_0} - \int_0^\rho \frac{3\,\mathrm{d}\rho}{\rho - v} \tag{A6}$$

where $v_0 = v(\rho = 0)$. This is equivalent to Equation 8b.

Appendix 3

Take the differential Equation 7 and multiply by ρ^3

$$4f\rho^3 + \rho^4 \frac{\mathrm{d}f}{\mathrm{d}\rho} = \rho^3 \frac{\mathrm{d}}{\mathrm{d}\rho}(f \cdot v)$$

which can be transformed into

$$\frac{\mathrm{d}}{\mathrm{d}\rho}(\rho^4 f - \rho^3 f \cdot v) = -3\rho^2 f \cdot v.$$

Integrate this equation from zero to ρ

$$-\int_0^\rho 3x^2 f(x)v(x)\,\mathrm{d}x$$

$$= \int_0^\rho \frac{\mathrm{d}}{\mathrm{d}x}[x^4 f(x) - x^3 f(x)v(x)]\,\mathrm{d}x.$$

The integral on the left-hand side is proportional to the change in volume for the range 0 to ρ and is thus zero if all particles are included, hence

$$\lim_{\rho \to \infty} \rho^3(\rho - v)f(\rho) \to 0.$$

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