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# Theory of the size distribution in three-dimensional grain growth

P. A. Mulheran

*Department of Physics, University of Reading, Whiteknights, P.O. Box 220, Reading, RG6 6AF, United Kingdom*

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The evolution of a three-dimensional grain boundary network is calculated in terms of a modified version of the classic theory of Hillert [M. Hillert, Acta Metall. **13**, 227 (1965)]. This has been extended beyond the mean field approximation to account for the size dependence of the incoming flux to grains embedded in the network. The prediction for the scaling distribution of grain sizes shows good agreement with that observed in Potts model simulations. At small volumes the distribution follows a power-law behavior, whereas at larger volumes it falls exponentially.

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Grain growth in polycrystalline materials has inspired several theoretical attempts to describe the network evolution [1]. The fundamental interest has arisen from the observed scaling behavior of the grain size distribution during normal grain growth. Progress towards an understanding of this phenomenon has resulted from the study of Potts model simulations in both two and three dimensions [2,3]. The two-dimensional case has traditionally been more accessible because of the topological rules that govern the network [4,5]. Von Neumann's law states that the growth of a grain in two dimensions depends only on the number of sides [6], and Lewis's law relates the average number of sides of a grain to its scaled area [7]. The combination of these leads to a good description of the scaling distribution of grain areas in the network [8]. However, no such laws exist for the three-dimensional grain boundary network, although some have been conjectured [9–11]. This is one of the reasons why a fundamental description of the grain growth process in three dimensions is lacking. It is the purpose of this communication to provide such a description.

One of the classic theories on the statistics of grain growth is that of Hillert [12]. He derived the grain size distribution from considerations of the average growth rate of a grain within a network whose evolution is driven by capillarity. However, his predicted scaling distribution of grain sizes does not compare well with that observed experimentally or in the Potts simulations. This is particularly true for the two-dimensional simulation [2]. Recent analysis of this Potts simulation has shown that this is due to the use of a mean field approximation and that this can be corrected by employing what I call the randomly connected bubble model [8]. In this current work the same model is applied to the three-dimensional system. It will be shown that this leads to a grain volume distribution that has good agreement with the simulation data and, by implication, with experimental observations. In particular, it is found that in the small volume limit the distribution follows a power law, but it then decreases exponentially at larger scaled volumes.

Before introducing any modifications I will first review Hillert's theory [12]. His expression for the statistically averaged growth rate of a grain with radius $r$ embedded in the grain boundary network is

$$\frac{dr}{dt}=\alpha M\sigma\left[\frac{1}{r_{cr}(t)}-\frac{1}{r}\right], \tag{1}$$

where $M$ is the grain boundary mobility and $\sigma$ is the free energy per unit area. The dimensionless parameter $\alpha\sim 1$ is included because the grains are not perfect spheres but instead are polyhedra. One may assume that $\alpha$ is the same for all grains regardless of size because of the scaling behavior observed in the network evolution. Note how the complicated issue of the network topology does not arise in this formulation. This is a distinct advantage when one wishes to look at the three-dimensional network as mentioned above.

In Eq. (1) the critical radius $r_{cr}(t)$, above which grains grow rather than shrink, is calculated to be $\frac{9}{8}$ times the average radius $\bar{r}(t)$ for a three-dimensional network. Thus (1) has the following mean field interpretation. The grain is shrinking under the action of surface energy minimization, and its radius of curvature is proportional to $r$. In addition, the grain receives an inward flux from its shrinking neighbors. Assuming that the grains are distributed randomly throughout the network one might expect that, on average, this flux is the same for all grains independent of size. The value of the critical radius $r_{cr}$ is determined by the condition that the total volume of the system is conserved. However, this mean field approximation is wrong, because of two neighbors it is the smaller that will shrink, so remaining convex while the larger is concave [8]. Hence the statistically averaged inward flux is not the same for all grains and does depend on the grain size. Grains much smaller than the average $\bar{r}$ obviously will have very few neighbors smaller than themselves, and so receive little inward flux. On the other hand, for large grains, nearly all their neighbors will shrink to contribute more inward flux than the mean field predicts. The growth rate (1) thus underestimates the rate at which small grains shrink and the rate at which large ones grow [13]. These observations will now be used to modify Hillert's theory and create the randomly connected bubble model for a three-dimensional system in the following way.

Consider first the scaling volume distribution $F(z)$ where $z=v/\bar{v}(t)$, $\bar{v}(t)$ being the average grain volume at time $t$. The distribution of grain radii, scaled to the average radius, is $G(x)dx=F(z)dz$ where

$$x=r/\bar{r}=Bz^{1/3}.$$

Since by definition $\bar{x}=1$ the constant $B$ is given by

 

$$\frac{1}{B}=\int_0^\infty z^{1/3}F(z)dz. \tag{2}$$

Now consider a grain of radius $r$ embedded in the three-dimensional network. Statistically its inward flux $J_{in}(r)$ is found from the sum of fluxes leaving smaller grains, $\alpha M\sigma/R$ $(R<r)$, times the probability of being in contact with grains of this size somewhere in the network. Assuming the grains are connected at random across their surfaces, this latter probability is given by the proportion of the total surface area available $[R^2/\int_0^\infty \bar{r}^2x^2G(x)dx]$ times the number of these grains in the system $[G(R/\bar{r})]$. Hence

$$J_{in}(r)=\int_0^{r/\bar{r}}\frac{\alpha M\sigma}{R}\frac{R^2}{\bar{r}^2\int_0^\infty x^2G(x)dx}G(R/\bar{r})\frac{dR}{\bar{r}}$$

$$=\frac{\alpha M\sigma}{\bar{r}C}\int_0^{r/\bar{r}}xG(x)dx,$$

with

$$C=\int_0^\infty x^2G(x)dx. \tag{3}$$

Similarly $J_{out}(r)$ is found from the flux leaving grains of size $r$, which is $\alpha M\sigma/r$, into larger neighbors. The probability of neighboring a larger grain of radius $R$ is as above, so that

$$J_{out}(r)=\int_{r/\bar{r}}^\infty\frac{\alpha M\sigma}{r}\frac{x^2}{C}G(x)dx$$

$$=\frac{\alpha M\sigma}{r}\left[1-\int_0^{r/\bar{r}}\frac{x^2}{C}G(x)dx\right].$$

Thus the statistically averaged rate of growth of the grain radius $r$ in the network is

$$\frac{dr}{dt}=J_{in}(r)-J_{out}(r)=\frac{\alpha M\sigma}{r}\left[\int_0^{r/\bar{r}}\left(\frac{r}{\bar{r}}+x\right)\frac{x}{C}G(x)dx-1\right]. \tag{4}$$

This equation is to be contrasted with Hillert's growth equation (1). The difference between the equations is in the inward flux term, which is size dependent in the current model. Recalling the role played by the geometric factor $\alpha$ we multiply by $4\pi r^2$ to find the equation of motion for grain volume $v$;

$$\frac{dv}{dt}=D\bar{v}^{1/3}(t)w(z),$$

where $D=(48\pi^2)^{1/3}\alpha M\sigma$ and

$$w(z)=z^{1/3}\left[\frac{B^2}{C}\int_0^z(z^{1/3}+y^{1/3})y^{1/3}F(y)dy-1\right]. \tag{5}$$

The continuity equation must be solved with this statistical growth rate to find the predicted distribution of grain volumes. This equation is

$$\frac{\partial f(v,t)}{\partial t}+\frac{\partial}{\partial v}\left[f(v,t)\frac{dv}{dt}\right]=0, \tag{6}$$

where $f(v,t)dv$ is the number of grains between volumes $v$ and $v+dv$ at time $t$. In the scaling state,

$$f(v,t)=\frac{\Omega}{\bar{v}^2(t)}F(z),$$

where $\Omega$ is the total volume of the system and $F(z)$ is of course the scaling distribution function introduced earlier. Since the growth is driven by surface tension the average radius obeys the parabolic growth law in the scaling state. Then $\bar{v}(t)=kt^{3/2}$ and the continuity equation gives

$$\frac{d}{dz}[w(z)F(z)]=\frac{3k^{2/3}}{2D}\left[2F(z)+z\frac{dF(z)}{dz}\right].$$

Integration then reveals

$$F(z)=\frac{\int_0^z F(z')dz'-1}{Aw(z)-z}, \tag{7}$$

with the numerical factor

$$A=\frac{2D}{3k^{2/3}}. \tag{8}$$

Equation (7) clearly requires self-consistent solution, since $w(z)$ is itself a functional of $F(z)$. From the behavior of the Potts model simulations, a first estimate is $F(z)\sim\exp(-z)$. This function fits reasonably well over most of the range, but underestimates the frequency of very small grains. In fact, it shall be shown later that $F(z)\sim z^{-1/3}$ for small $z$. Now with this first estimate the integrals on the right of Eq. (7) may be calculated, using Eqs. (2)–(5), leading to a better approximation for the function $F(z)$. This process is then iterated until the exact solution is found. In practice only three iterations are necessary, and in fact just one iteration yields an estimate for $F(z)$ that is almost indistinguishable from the exact solution. Now the factor $A$ in Eq. (7) is obtained numerically by the condition that

$$\bar{z}=\int_0^\infty zF(z)dz=1.$$

This is equivalent to requiring that the total volume of the system is conserved. In the level of approximation used here it transpires that $A\sim1.4$, so that Eq. (8) gives the growth rate for the average grain volume as

$$\bar{v}(t)\simeq(3.7\alpha M\sigma t)^{3/2}.$$

This can be compared to Hillert's mean field growth rate [12], $\bar{v}(t)\simeq(1.03\alpha M\sigma t)^{3/2}$. Therefore changing the growth law from the mean field equation (1) to Eq. (4) makes the average volume growth rate nearly seven times larger. It may be possible to test this prediction experimentally.

Before the result for $F(z)$ is shown, we consider the term $w(z)$ in the denominator of Eq. (7). If $Aw(z)=z-1$, then $F(z)=\exp(-z)$ exactly. The difference between this and the numerical values calculated from Eq. (5) thus indicates how

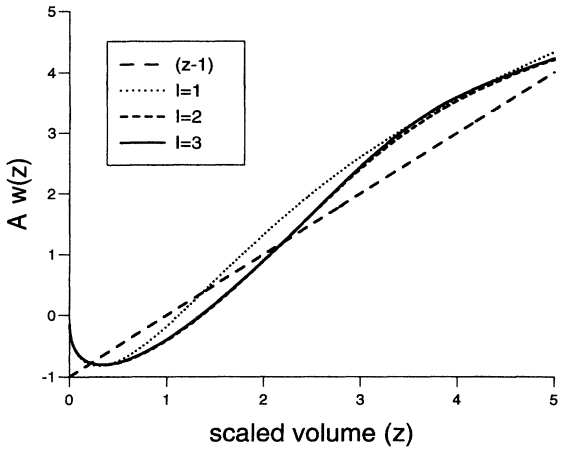


FIG. 1. $Aw(z)$ as calculated from Eq. (5). The variation with iteration number $I$ shows the convergence to the exact solution. The straight line $(z-1)$ is also shown for comparison.

near the new approximation for $F(z)$ is to the initial estimate. The comparison is made in Fig. 1 where the variations upon iteration are also given. It is clear that the numerical values follow the trend of the straight line, implying that the scaled distribution behaves in a similar manner to the exponential decay. The most dramatic deviation occurs in the region of small $z$. This region will be discussed below.

The final distribution function calculated from Eq. (7) is shown in Fig. 2, where it is compared to Hillert's original analytical solution of the mean field model [12] as well as the inverse exponential function. Also shown in Fig. 2 are the data from my own Potts model simulations. These simulations used the so-called extended algorithm [14] whereby spin-flip trials are performed by selecting nearest-neighbor orientations. This differs from the standard procedure where trials are performed by random selections from some $Q_{max}$

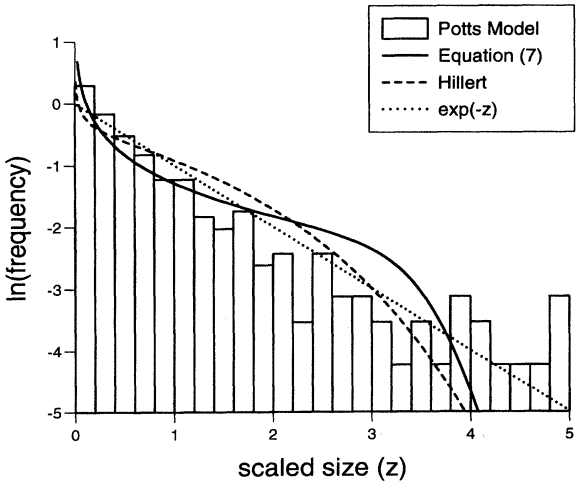


FIG. 2. The scaled volume distribution function as calculated from Eq. (7), plotted on a logarithmic second axis. Hillert's solution [12], the inverse exponential, and the results from simulation are shown for comparison.

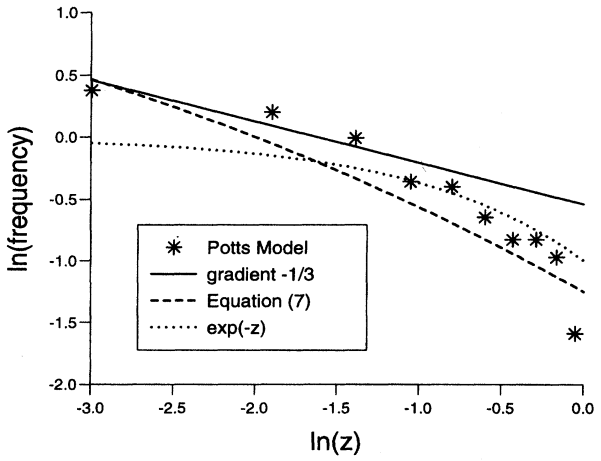


FIG. 3. Log-log plots of the results from simulation and from Eq. (7), to verify the power-law behavior of the distribution at small volumes.

orientations [2,3]. The advantage of the former is that each grain can be assigned a unique orientation so precluding grain coalescence. However, both simulations lead to the same scaling distribution and the reader is referred to [2,3] for detailed analysis of the scaling state. It is clear that the distribution calculated from Eq. (7) compares favorably with the simulation data, particularly in the range $0<z<2$ where about 88% of the grains are found. Our calculation shows more marked deviation from the inverse exponential than Hillert's result giving an improved comparison with the data.

It is apparent from Fig. 2 that the results from Eq. (7) display a high $z$ cutoff in the manner of Hillert's distribution, whereas the Potts model simulations reveal a few grains with scaled sizes greater than $z\simeq 4$. The inverse exponential does not suffer from this cutoff and so would appear to be the best description of the data over the whole range of grain sizes. It is interesting to note that this $\exp(-z)$ function is the maximum entropy solution for the distribution with the constraints of space filling and normalization [15]. However, this solution does not take into account the nature of the grain growth mechanism, i.e., the surface energy minimization that drives the evolution. For this reason this maximum entropy result does not yield the correct form of the distribution at very small grain sizes. It has been emphasized before [3] that the Potts model data are not exponentially distributed at small volumes in three dimensions. In contrast, the calculations presented here do give a good account of the data in this important size range. I shall now demonstrate this by focusing on the low $z$ regime.

Equation (7) has a small volume divergence arising from the fact that $w(z)$ tends to zero with $z$. Is this behavior acceptable? To answer this we must consider how the network coarsens in time. The total number of grains in the system at time $t$ is

$$n(t)=\int_0^\infty f(v,t)dv.$$

Integration of the continuity equation (6) then shows that

$$\frac{dn(t)}{dt} \propto F(0)w(0).$$

But we know that $w(z) \sim -z^{1/3}$ for small $z$ since the grains are shrinking by surface tension. This behavior is evident in Hillert's work [12] as well as in Eq. (5). Thus if the network is coarsening steadily in a scaling state, $F(z) \sim z^{-1/3}$ for small $z$. From Eq. (7) it can be seen that the solution evaluated here satisfies this condition. In Fig. 3 it is confirmed that the Potts model data also show this behavior, verifying that it is coarsening by surface tension effects. It is necessary that the distribution obeys a power law for small grain volumes. Therefore the maximum entropy solution $\exp(-z)$ cannot provide a good explanation for the low $z$ distribution and it cannot be regarded as a satisfactory description of the grain growth data.

In summary, the randomly connected bubble model has been applied to a three-dimensional network. Hillert's theory of grain growth has been modified to account for the shrinkage of grains when neighbored by larger grains and their growth when next to smaller ones. The grains have been assumed to be positioned at random throughout the network. I believe that this point is crucial to understanding why the system displays scaling [8]. The random connectivity of the grains is maintained during the network evolution since new grains are brought into contact by the shrinkage of intermediate ones. The statistical growth laws are thus self-perpetuating. The scaling distribution of grain sizes predicted with this model shows good agreement with that found in Potts model simulations, particularly in the range $0<z<2$ covering $\sim 88\%$ of the grains. This success suggests that we are starting to achieve a good understanding of the microstructural evolution observed in polycrystalline materials.

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