

ON THE THEORY OF NORMAL AND ABNORMAL GRAIN GROWTH*

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A growth equation for individual grains in single-phase materials is suggested. It is used to calculate a rate equation for normal grain growth and the size distribution in the material. It predicts a maximum size of twice the average size. The theory is modified to take into account the effect of second-phase particles.

In an alternative treatment the array of grains is described in terms of a kind of defects introduced into a perfect array. The defects move through the array during grain growth. The rate of grain growth is calculated from the number of defects and their mobility. The defect concentration is predicted by comparing the two treatments. The defect-model predicts two grain size limits due to second-phase particles. Normal grain growth takes place below the lower limit. Abnormal grain growth can take place between the two limits if the material contains at least one very large grain. No grain growth can take place above the higher limit.

Several possible mechanisms for the development of abnormal grain growth are examined. An explanation is offered for the observation that most of the well-known cases occur as the second-phase is dissolving.

SUR LA THEORIE DES CROISSANCES GRANULAIRES NORMALE ET ANORMALE

L'auteur propose une équation rendant compte de la croissance des grains individuels dans un solide monophasé. Cette équation est utilisée pour calculer la vitesse de croissance normale et la distribution des tailles des grains dans la matière. Elle prédit une taille maximum correspondant à 2 fois la taille moyenne. La théorie a été modifiée pour tenir compte de la présence de particules d'une seconde phase.

Par ailleurs, la morphologie des grains est décrite en fonction d'un type de défauts introduits dans un réseau parfait. Ces défauts se déplacent dans le réseau pendant la croissance granulaire. La vitesse de croissance est calculée à partir du nombre de défauts et de leur mobilité. En comparant les deux modes de calcul, on peut prévoir la concentration en défauts: un modèle basé sur la distribution des défauts conduit à prévoir deux tailles de grains limites lorsqu'il reste des particules d'une seconde phase. La croissance granulaire normale se produit donc sous la taille limite inférieure. La croissance anormale se développe lorsque la taille est située entre les deux limites précitées à condition que le solide contienne au moins un grain très grand.

Plusieurs mécanismes possibles pour la croissance granulaire anormale sont examinés. Une explication pourrait être trouvée dans le fait que la plupart des cas de croissance anormale se produisent lorsque la seconde phase est en voie de dissolution.

ZUR THEORIE DES NORMALEN UND DES ANOMALEN KORNWACHSTUMS

Es wird eine Wachstumsgleichung für individuelle Körner in einphasigen Stoffen vorgeschlagen. Sie wird benutzt zur Berechnung einer Beziehung für normales Kornwachstum und für die Größenverteilung in dem Material. Sie sagt eine Maximalgröße von zweimal der durchschnittlichen Größe voraus. Die Theorie wird modifiziert, um dem Einfluß von Teilchen einer zweiten Phase Rechnung zu tragen.

In einer zweiten Behandlung wird die Anordnung von Körnern als eine Art von eingefügten Fehlern in einer idealen Anordnung beschrieben. Während des Kornwachstums wandern die Fehler durch die Anordnung. Die Geschwindigkeit des Kornwachstums wird berechnet aus der Zahl der Fehler und ihrer Beweglichkeit. Die Fehlerkonzentration wird durch einen Vergleich der beiden Verfahren vorausgesagt. Das Fehler-Modell sagt zwei Korngrößengrenzen auf Grund von Teilchen einer zweiten Phase voraus. Normales Kornwachstum findet unterhalb der unteren Grenze statt. Anomales Kornwachstum kann zwischen den beiden Grenzen stattfinden, wenn das Material wenigstens ein sehr großes Korn enthält. Oberhalb der oberen Grenze können die Körner nicht wachsen.

Für die Entstehung des anomalen Kornwachstums werden verschiedene in Frage kommende Mechanismen untersucht. Es wird eine Erklärung für die Beobachtung gegeben, daß die meisten wohlbekannten Fälle bei der Auflösung der zweiten Phase auftreten.

1. INTRODUCTION

By grain growth one usually understands the increase of the grain size in a single-phase material or of the matrix grain size in a material with second-phase particles. The sum of the individual grain sizes is constant and the increase in average grain size is thus connected with a disappearance of some of the grains, usually the smaller ones. In practice, one distinguishes between "normal" or "continuous" grain growth and

"abnormal" or "discontinuous" grain growth. During normal grain growth, the size of the individual grains are relatively uniform. During abnormal grain growth, on the other hand, the differences in individual sizes increase by some of the grains growing rapidly. When they have consumed all the other grains, the remaining grains may again be of a relatively uniform size.

The theory of normal grain growth is based on the grain boundary interfacial free energy being the driving force. Several authors^(1,2,3) have been able to deduce a parabolic growth law for the average grain

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size, $\bar{R}^2 - R_0^2 = A\sigma t$, using slightly different arguments, a common feature being that a fixed distribution of shapes and relative sizes is assumed. C. S. Smith⁽⁴⁾ suggested that there is a natural tendency toward such a fixed distribution. Apart from his discussion of the general topological rules governing the shapes of grains, there seems to be no theoretical discussion of how this distribution might look or whether there is a tendency toward such a distribution. Experimental data have often been interpreted in terms of a lognormal distribution. Accepting this distribution function, Feltham⁽⁵⁾ was able to estimate the size of the rate constant A . The main interest of the experimenters has been focussed on the temperature dependence of the rate constant and on the time exponent in the rate law when written as R proportional $t^{1/2}$. Experimentally one has usually found exponent values appreciably less than 1/2.

The particle size of second-phase particles in a matrix may also increase by the larger particles growing at the expense of the smaller ones. This kind of grain growth is often called coalescence and is closely related to the first kind of grain growth, the main driving force being interfacial free energy in both cases. In the case of coalescence, Lifshitz and Slyozov⁽⁶⁾ have recently been able to prove that there actually is a tendency toward a fixed distribution of relative sizes. They have calculated the distribution function in detail and also the rate constant in the growth law for coalescence, $r^3 = Kt$.

In the present paper it will be shown how the method of Lifshitz and Slyozov can be applied to the first kind of grain growth. The treatment will not be quite rigorous, however. The high complexity of the geometric shapes of the grains in a single-phase material as compared to the perfect spherical shapes of the second-phase particles in the case of coalescence makes some approximations necessary when formulating mathematically the growth law for individual grains. In an attempt to recover some of the finer details that may get lost by these approximations, an alternative model of grain growth will be discussed. In this model, the complicated geometric pattern of the grains in a single-phase material is described simply by a number of defects introduced into a perfect array of grains. This model is particularly interesting in connection with the retarding effect on the grain boundary movements due to second-phase particles, as treated by Zener.⁽⁷⁾

Grain growth in two-dimensional as well as three-dimensional systems will be considered. The three-dimensional case is the most interesting one from the practical viewpoint but the two-dimensional case

seems to lend itself more readily to theoretical treatment.

2. CHOICE OF BASIC EQUATION

For coalescence, Greenwood⁽⁸⁾ derived an equation for the change in size of an individual particle:

$$\frac{dr}{dt} = \frac{k}{r^2} \left(\frac{r}{r_{cr}} - 1 \right) \quad (1)$$

The critical size r_{cr} is related to the average particle ones dissolve. Using equation (1) and assuming a constant total volume of the particles Lifshitz and Slyozov were able to show that the growth of the average particle size \bar{r} , will asymptotically approach the following expression:

$$\bar{r}^3 = \frac{4}{9} kt \quad (2)$$

They were also able to show that $r_{cr} = \bar{r}$ and to calculate the asymptotic distribution function for the particle sizes, finding that the radius of the largest particle should be 3/2 times as large as the average \bar{r} when the asymptotic distribution has been attained.

In order to apply the method of Lifshitz and Slyozov to the case of grain growth in single-phase materials we need an expression similar to equation (1) for the change in size of an individual grain. We shall start with the generally accepted assumption that the velocity of a grain boundary is proportional to the pressure difference caused by its curvature:

$$v = M \cdot \Delta P = M\sigma \cdot \left(\frac{1}{\rho_1} + \frac{1}{\rho_2} \right) \quad (3)$$

The proportionality constant M may be regarded as the mobility of the grain boundary. ρ_1 and ρ_2 are the principal radii of curvature.

The size of each grain will be expressed by the radius R of an equivalent circle or sphere having the same area or volume, respectively. The net increase in size of a grain can, in principle, be calculated by integrating v around the grain. The net increase described by dR/dt is thus intimately related to an average value of v around the grain. We can put $dR/dt = g \cdot v_{\text{average}}$ where the value of the factor g depends upon the shape of the grain. For a circular grain it is unity, for ordinary grains it might be somewhat larger. Equation (3) can now be written

$$\frac{dR}{dt} = M\sigma \cdot g \cdot \left(\frac{1}{\rho_1} + \frac{1}{\rho_2} \right)_{\text{average}} \quad (4)$$

The curvatures will vary from grain to grain as well as around the periphery of each grain, due to the complex shapes of the grains in a single-phase material.

However, for our analysis we are only interested in a value of $g \cdot \left(\frac{1}{\rho_1} + \frac{1}{\rho_2} \right)$ averaged over all the grains of each size R . A central problem in the theory of grain growth is to find this average. In the present paper we shall not try to solve this complex geometric problem in a rigorous way but rather to make the simplest possible choice. Then we shall use it to show how the characteristics of grain growth can be calculated. Some justification for the choice will be given but the final justification may have to await an experimental test of the predicted characteristics of grain growth.

We want an expression of the correct dimension and with the characteristic feature that it is positive for large R but negative for small R . The critical size where the value of the expression goes through zero will be denoted by R_{cr} . The simplest choice seems to be

$$g \cdot \left(\frac{1}{\rho_1} + \frac{1}{\rho_2} \right)_{\text{average}} = \alpha \cdot \left(\frac{1}{R_{cr}} - \frac{1}{R} \right) \quad (5)$$

where α is a dimensionless constant. Of course, the critical size R_{cr} might be related to the average grain size in some way but the exact relationship will not be discussed until later. Introducing (5) into equation (4) yields the following expression for the average growth rate of all the particles of size R .

$$\frac{dR}{dt} = \alpha M \sigma \cdot \left(\frac{1}{R_{cr}} - \frac{1}{R} \right) \quad (6)$$

This is the basic equation on which our calculations will be based.

Assuming a lognormal distribution of the individual grain sizes, Feltham⁽⁵⁾ obtained a similar equation, which we can write

$$\frac{dR}{dt} = \frac{K}{8R} \ln \frac{R}{R_{cr}} \quad (7)$$

With $K = 8\alpha M \sigma$ the two equations are identical in the neighborhood of $R = R_{cr}$ but they are quite different at the extreme values of R . For large grains our equation (6) predicts that the growth rate approaches a constant value of $\alpha M \sigma / R_{cr}$ which is satisfactory, whereas Feltham's equation (7) predicts unreasonably low values. For small grains our equation (6) predicts that dR^2/dt approaches a constant negative value which is quite correct for a two-dimensional system.^(9,10) Feltham's equation (7), on the other hand, predicts indefinitely high negative values. As a consequence, our equation (6) seems to be a much more attractive choice than equation (7).

Comparison of our equation (6) with equation (1) shows a remarkable similarity, thus allowing us to follow the procedure of Lifshitz and Slyozov with very

little modification, as will be demonstrated in the next section.

In the case of two dimensions we can use the result of von Neumann⁽⁹⁾ and Mullins⁽¹⁰⁾ in order to determine the value of α . They were able to show that equation (3) can be integrated around any grain in a two-dimensional system, yielding a very simple result which we can write in the following way by using the radius R instead of the area,

$$\frac{dR}{dt} = \frac{M \sigma}{R} \cdot \left(\frac{n}{6} - 1 \right) \quad (8)$$

Comparison with equation (6) reveals that our choice of equation (5) implies that the following relation holds between the size R and the average number of neighbors per grain of that size in a two-dimensional system:

$$n = 6 + 6\alpha \left(\frac{R}{R_{cr}} - 1 \right) \quad (9)$$

The average number of neighbors per grain in the total system would then be

$$\bar{n} = \frac{1}{N} \sum_N 6 \left[1 + \alpha \left(\frac{R}{R_{cr}} - 1 \right) \right] = 6 + 6\alpha \left(\frac{\bar{R}}{R_{cr}} - 1 \right) \quad (10)$$

We can thus conclude that $R_{cr} = \bar{R}$ because there is a topological rule telling that $\bar{n} = 6$ in a two-dimensional array of grains.⁽⁴⁾ The same conclusion can be drawn directly from equation (6) because the sum of RdR/dt over all the grains must be zero in order to conserve the total size of the two-dimensional system. We can now evaluate α . According to the detailed picture of grain growth given by Smith,⁽⁴⁾ a shrinking grain never gets less than three neighbors. With the value $n = 3$ at $R = 0$ equation (9) gives $\alpha = \frac{1}{2}$. Another estimate can be carried out for an abnormally large grain, $R \gg R_{cr}$. Its periphery may cut through most of the surrounding grains sufficiently close to their centers to justify the following estimate:

$$2\pi R \cong \sum_n 2R = 2n\bar{R} \quad (11)$$

Inserting this in equation (9) and remembering that

$\bar{R} = R_{cr}$ we find $\alpha = \frac{\pi}{6} \cong \frac{1}{2}$. It may thus appear that

equation (5) can properly describe the geometry of the grains over the whole range of sizes in a two-dimensional system if the value $\alpha = \frac{1}{2}$ is chosen.

In three dimensions it is not possible to integrate equation (3) in the general case, obtaining a result like equation (8) for an individual grain. The integration can be carried out in special cases, however, thus allowing us to estimate α . Again we shall examine two

cases, $R = 0$ and $R \gg R_{cr}$. A shrinking grain in a three-dimensional array has 4 neighbors just before it disappears, resembling a tetrahedron. Assuming that the four faces of such a grain have spherical shape and meet each other with an angle of 120° along the grain edges a numerical calculation can be carried out yielding $dR/dt = 1.0 \cdot M\sigma/R$ and comparison with equation (6) at $R \ll R_{cr}$ thus gives the value $\alpha = 1.0$ for a three-dimensional system. For a large grain we can compare with the two-dimensional case which gave $\alpha = \frac{1}{2}$ but in view of the fact that $\rho_2 = 0$ in two dimensions but $\rho_2 = \rho_1$ in three, we estimate that α should be about twice as large in three dimensions, $\alpha \cong 1$.

3. CALCULATION OF NORMAL GRAIN GROWTH

Introducing the relative size $u = R/R_{cr}$ we can transform our basic equation (6)

$$\frac{dR^2}{dt} = 2\alpha M\sigma \cdot (u - 1) \quad (12)$$

and form an expression for the growth of the relative size of a particle

$$\frac{du^2}{dt} = \frac{1}{R_{cr}^2} \left[\frac{dR^2}{dt} - \left(\frac{R}{R_{cr}} \right)^2 \cdot \frac{dR_{cr}^2}{dt} \right] \quad (13)$$

$$\frac{du^2}{dt} = \frac{1}{R_{cr}^2} \left[2\alpha M\sigma \cdot (u - 1) - u^2 \cdot \frac{dR_{cr}^2}{dt} \right] \quad (14)$$

After dividing through by dR_{cr}^2/dt we get

$$\frac{du^2}{dR_{cr}^2} = \frac{1}{R_{cr}^2} \left[2\alpha M\sigma \cdot \frac{dt}{dR_{cr}^2} \cdot (u - 1) - u^2 \right] \quad (15)$$

This equation can be written in an extremely simple form by introducing two new variables

$$\frac{du^2}{d\tau} = \gamma \cdot (u - 1) - u^2 \quad (16)$$

where

$$\tau = \ln R_{cr}^2 \quad (17)$$

$$\gamma = 2\alpha M\sigma \cdot dt/dR_{cr}^2 \quad (18)$$

The new variable τ represents the time because R_{cr} increases monotonically with time. The same equation with the exponent 3 instead of 2 was found by Lifshitz and Slyozov. They were able to show that their equation predicts that the coalescence process will asymptotically approach a steady state where the variable γ has a constant value, no matter what the initial size distribution is. The same arguments can be applied to our case but will not here be repeated in detail. In order to find the asymptotic value of γ , we shall discuss Fig. 1 where the function $du^2/d\tau$ has been plotted for three constant values of γ . It is evident

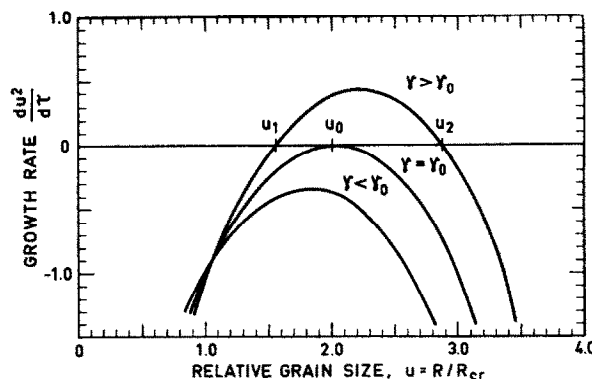


FIG. 1

that a value of $\gamma < \gamma_0$ cannot be maintained during the process because all the grains would then shrink with a nonvanishing rate and after a certain time all the grains would have disappeared. A value $\gamma > \gamma_0$ is also not possible during a steady state because all the grains larger than u_1 would then approach the size u_2 which would mean that they increase their absolute size R indefinitely. On the other hand, all grains smaller than u_1 would disappear within a certain time. As a consequence, the only possible γ value for the steady state is γ_0 which can easily be calculated from equation (16). Let us first calculate u_1 and u_2 :

$$u = \frac{1}{2}\gamma \pm \sqrt{\frac{1}{4}\gamma^2 - \gamma} \quad (19)$$

For $\gamma = \gamma_0$ the two roots must coincide, thus yielding $\gamma_0 = 4$ and $u_0 = 2$. With the value $\gamma = \gamma_0 = 4$, equation (18) gives the growth rate equation for normal grain growth:

$$\frac{dR_{cr}^2}{dt} = \frac{1}{2}\alpha M\sigma \quad (20)$$

This value is quite close to the value obtained by Feltham, a natural consequence of the similarity between Feltham's growth equation (7) and our growth equation (6).

From the shape of the curve for $\gamma = \gamma_0$ in Fig. 1 we can conclude that all grains larger than u_0 would decrease their relative size toward u_0 but they would never be able to pass through this point. Thus, they would increase their absolute size R indefinitely which is physically impossible. This argument allows the important conclusion to be drawn that there must not be any grains larger than $2R_{cr}$ after the steady state condition of grain growth has been reached. If the initial size distribution is too wide, a fraction of large grains will grow in an abnormal manner until all the other grains have been consumed. When completed, this process has resulted in a more narrow size distribution and, at longer times, the steady state may be

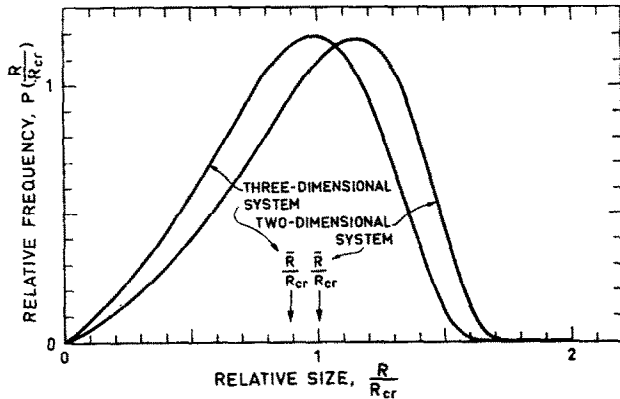


FIG. 2

approached asymptotically. It appears convenient to define this steady state as normal grain growth. Abnormal grain growth may thus sometimes be a necessary initial stage during the development toward normal grain growth.

Following the procedure of Lifshitz and Slyozov, we may calculate the whole distribution of the individual grain sizes. These calculations are presented in the Appendix and they result in the following distribution function:

$$P(u) = (2e)^\beta \cdot \frac{\beta u}{(2-u)^{2+\beta}} \cdot \exp \frac{-2\beta}{2-u} \quad (21)$$

where $\beta = 2$ in two dimensions and $\beta = 3$ in three dimensions.

Figure 2 shows the shape of the function for these two cases. The position of the mean value \bar{R} is indicated. As shown in the Appendix, $\bar{R} = R_{cr}$ in the two-dimensional case, in agreement with the result in Section 2. In a three-dimensional system $\bar{R} = 8/9 R_{cr}$ for this particular grain size distribution.

Experimentally, it is very difficult to determine the true size distribution in a three-dimensional system. It is much easier to make observations on a two-dimensional section and in order to allow comparison with such measurements the size distribution in a section was calculated numerically from the true size distribution assuming that all the grains are spherical. In Fig. 3 the two distributions are compared. The quantity s denotes the apparent radius of a grain as observed in the section. As expected, the distribution curve for a section shows a more pronounced tail on the upper side.

All the distribution curves have a tail reaching up to the theoretical value of 2 but showing negligibly small values long before that limit. For practical purposes one might say that the maximum size is about $1.6 R_{cr}$ in a three-dimensional system and $1.7 R_{cr}$ in a

two-dimensional system. If we compare with the average size we find that the maximum size is about $1.8 \bar{R}$ in a three-dimensional system and $1.7 \bar{R}$ in a two-dimensional system. In a section through a three-dimensional system it is about $2\bar{s}$.

4. EFFECT OF SECOND-PHASE PARTICLES

When a grain boundary is moving through a matrix with second-phase particles, there is a tendency for the grain boundary to stick at the particles. As a consequence, the geometry will be considerably more complex and equation (5) will no longer hold. However, Zener⁽⁷⁾ has shown that one can take into account the effect of second-phase particles by assuming that the geometry is unaffected and instead introducing a back stress by the particles. For particles of uniform size r and a total volume fraction f , Zener found that the hypothetical back stress would be approximately $S = 3f\sigma/4r$. For the general case we shall write $S = \sigma \cdot z$ where z depends upon the number and sizes of the second-phase particles. In agreement with Zener's calculation we shall assume that z is independent of the grain boundary curvature although this may not be strictly true.

Taking the second-phase particles into account we shall change equation (3):

$$v = M \cdot \Delta P_{true} = M \cdot (\Delta P \pm S) \\ = M\sigma \cdot \left[\left(\frac{1}{\rho_1} + \frac{1}{\rho_2} \right) \pm z \right] \quad (22)$$

where ΔP and $\left(\frac{1}{\rho_1} + \frac{1}{\rho_2} \right)$ should be estimated without taking the effect of the second-phase particles into account. As a consequence of equation (22), equation (6) will also change:

$$\frac{dR}{dt} = M\sigma \left[\alpha \left(\frac{1}{R_{cr}} - \frac{1}{R} \right) \pm gz \right] \\ \cong \alpha M\sigma \cdot \left(\frac{1}{R_{cr}} - \frac{1}{R} \pm \frac{z}{\alpha} \right) \quad (23)$$

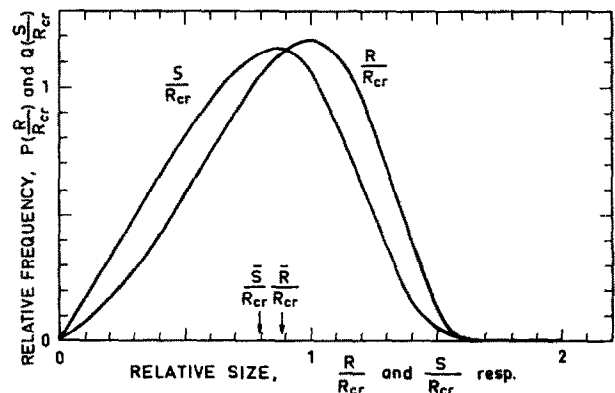


FIG. 3

The sign must be chosen in each case such that the back stress S is acting against the movement of the grain boundary. The negative sign holds when $\frac{1}{R} < \frac{1}{R_{cr}} - \frac{z}{\alpha}$ and the positive sign when $\frac{1}{R} > \frac{1}{R_{cr}} + \frac{z}{\alpha}$. Between these two limits, the back stress would exceed the driving force which is not physically possible. Thus we obtain $dR/dt = 0$ in this range. Considering the fact that the evaluation of z is very approximate, we have neglected the small effect of g being slightly larger than unity due to the non-circular shape of the grains.

Applying the same procedure as in Section 3, we can transform equation (23) into

$$\frac{du^2}{d\tau} = \gamma \cdot \left[u \cdot \left(1 \pm \frac{zR_{cr}}{\alpha} \right) - 1 \right] - u^2 \quad (24)$$

It should immediately be realized that we cannot expect to find any steady state solution in this case, because the second-phase particles grow more and more important as the grain size R_{cr} increases. If we, in spite of this fact, would try to find a steady state solution in the same way as before, we should now obtain

$$\gamma_0 = 4/(1 - zR_{cr}/\alpha)^2 \quad (25)$$

$$u_0 = 2/(1 - zR_{cr}/\alpha) \quad (26)$$

$$dR_{cr}^2/dt = \frac{1}{2}\alpha M\sigma \cdot (1 - zR_{cr}/\alpha)^2 \quad (27)$$

It is evident from equation (26) that the size distribution cannot remain constant when the grain size has grown to such values that the Zener effect becomes important. Instead, the maximum size is predicted to increase from twice the average and to approach infinity as the grain size R_{cr} reaches a limiting value of $R_l = \alpha/z$. According to equation (27) the grain growth will cease at this limiting grain size.

It has long been realized that there should be such a limit and Burke⁽³⁾ has attempted to describe the decrease of the growth rate on approaching the limit, by introducing the factor $(1 - R_{cr}/R_l)$ into the parabolic growth law, obtaining

$$dR_{cr}^2/dt = A\sigma \cdot (1 - R_{cr}/R_l) \quad (28)$$

In view of equation (27) it appears that the square of Burke's factor would have been a better choice. Due to the square, our equation (27) predicts a more gradual retardation of the growth rate.

In view of the fact that equations (25), (26) and (27) do not represent a true steady state, equation (27)

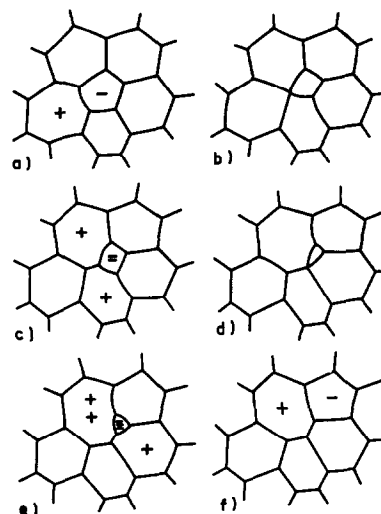


FIG. 4

should only be regarded as a first approximation. It should be noticed, however, that the value of the limit $R_l = \alpha/z$, is not subject to the same uncertainty. It is a direct consequence of equation (23), from which it is evident that a grain cannot grow, however large it is, when the average size is $R_{cr} \geq \alpha/z$.

In order for equations (25), (26) and (27) to represent a true steady state, it is necessary that z vary simultaneously with R_{cr} in such a way that the product zR_{cr} remains constant. It is not quite inconceivable that this condition may be satisfied under special experimental conditions. Such possibilities will be discussed in Section 7.

5. DEFECT-MODEL OF GRAIN GROWTH IN TWO-DIMENSIONAL SYSTEMS

In two dimensions we can attack the grain growth problem by another method which, in some respects, is closer to the true mechanism of grain growth.

The topological rule that the average number of neighbors per grain is $\bar{n} = 6$ in a two-dimensional array of grains, has an important consequence. Let us consider an ideal array of hexagonal grains, the so-called bee-hive structure. If we introduce an imperfection by giving a certain grain five neighbors only, then we must give another grain seven neighbors at the same time. This "5-7 pair" may thus be regarded as one defect, introduced into the perfect array. The important properties of such a defect are demonstrated by Fig. 4. In view of equation (8) the 5-grain will shrink and the 7-grain will grow. During the process, the role of being 7-grain is taken over by other neighbors. The area of the shrinking grain will thus be divided among several of its neighbors. The fundamental process of grain growth would thus seem to be the shrinking of a grain with less than 6 neighbors,

rather than the growth of a grain with more than 6 neighbors. Another important property is demonstrated by the final arrangement of the grains after the shrinking grain has completely disappeared. The defect is still present. It now affects another grain, causing its number of neighbors to decrease to five. The process will thus be repeated.

The defect will move stepwise through the array of grains and for each step it takes, the number of grains will have decreased by one. As a consequence, the rate of grain growth may be regarded as due to the number of defects per grain, c , and the time a a defect will need to make a grain shrink from normal size to zero.

$$-\frac{dN}{dt} = \frac{cN}{a} \quad (29)$$

The number of grains in the system, N , is related to the size of the grains

$$N \cdot R^2 = \text{constant} \quad (30)$$

where R is the root mean square in the general case. However, assuming a constant distribution function we can choose any mean value for R , by changing the size of the constant in equation (30) appropriately. By choosing R_{cr} , we obtain from equation (30):

$$\frac{2dR_{cr}}{R_{cr}} + \frac{dN}{N} = 0 \quad (31)$$

and by combining with equation (29)

$$\frac{dR_{cr}}{dt} = -\frac{R_{cr}}{2N} \cdot \frac{dN}{dt} = \frac{1}{2} R_{cr} \cdot \frac{c}{a} \quad (32)$$

$$\frac{dR_{cr}^2}{dt} = R_{cr}^2 \cdot \frac{c}{a} \quad (33)$$

When there are so many defects that there is a high probability that two or more will cooperate, equation (33) should be slightly modified:

$$\frac{dR_{cr}^2}{dt} = R_{cr}^2 \cdot \sum_p \frac{c_p}{a_p} \quad (34)$$

where c_p is the concentration of grains with 6- p neighbors and a_p is the time it takes such a grain to disappear.

In order to estimate a_p , we shall use the exact equation (8) with $n = 6-p$:

$$\frac{dR}{dt} = -\frac{pM\sigma}{6R} \quad (35)$$

Integration from the normal size R_{cr} to zero gives

$$a_p = -\frac{6}{pM\sigma} \int_{R_{cr}}^0 R dR = \frac{3R_{cr}^2}{pM\sigma} \quad (36)$$

The actual length of a may be somewhat less because the shrinkage rate of a grain will increase when it has grown so small that the number of neighbors decreases further. However, this effect is not very important since it seems to affect a comparatively late stage only.

By combination of (36) with (34) we obtain

$$\frac{dR_{cr}^2}{dt} = \frac{1}{3} M\sigma \sum_p p \cdot c_p \quad (37)$$

which gives a parabolic growth law as long as the number of defects remains constant. This requirement is analogous to the common assumption of a constant size distribution during normal grain growth. By comparing equation (37) with equation (20) we can evaluate the number of defects during normal grain growth and obtain, remembering that $\alpha = \frac{1}{2}$ in a two-dimensional system:

$$\sum_p p \cdot c_p = \frac{3}{2} \alpha = \frac{3}{4} \quad (38)$$

The quantity $\sum p \cdot c_p$ may be regarded as the total concentration of defects. The concentration necessary for normal grain growth is very high according to equation (38) and we must expect a considerable number of them to be situated close to each other and thus to be forced to cooperate.

We must now ask how the concentration of defects in a system can approach the value required for normal grain growth. Let us consider a system with very few defects initially. At the beginning of this section it was demonstrated that the defects have a remarkable stability. One may thus be tempted to expect the number of defects to remain fairly constant during grain growth, and thus the concentration of defects to increase. However, there is at least one very important mechanism by which the number of defects will decrease. As a grain has disappeared the active defect must move on and may then encounter another defect in such a way that they will annihilate each other. A qualitative argument seems to suggest that this mechanism to a first approximation should be effective enough to make the concentration of defects remain constant during grain growth. The mechanism by which the concentration is adjusted toward the value required for normal grain growth may thus be of a quite different nature. Even though we have concluded that the fundamental process of grain growth is the shrinking of the grains with less than 6 neighbors and that the role of being 7-grain is passed around among the neighbors, we should now realize that a grain of sufficient size will have a fairly constant number of neighbors and much higher than 6. Such a grain will grow with a fairly constant rate, and it will even

be able to increase its relative size if the growth rate of the average size is too low. This occurs if there are too few defects according to equation (33) and will then result in a less uniform size distribution, i.e. in a higher concentration of defects. If this line of argument is correct, the shrinking of an ordinary grain is the fundamental process of normal grain growth only, whereas the growth of a large grain is the fundamental process of abnormal grain growth, the concept of "abnormal grain growth" here taken in a very broad sense.

6. THE EFFECT OF SECOND-PHASE PARTICLES TREATED BY THE DEFECT-MODEL

In Section 4 a mathematical analysis of the effect of second-phase particles on grain growth was carried out using the method of Lifshitz and Slyozov. At the end of the section it was concluded that the analysis could be regarded as a first approximation only, in view of the fact that the mathematical solution did not represent a true steady state. It is thus of considerable interest to examine what predictions the defect-model will lead to in the presence of second-phase particles. Again, we shall consider a two-dimensional system.

Accepting equation (9) we can transform equation (23) into:

$$\frac{dR}{dt} = \frac{M\sigma}{R} \left(\frac{n}{6} - 1 \pm zR \right) \quad (39)$$

which reduces to equation (8) when $z = 0$. With $n = 6 - p$ we obtain

$$\frac{dR}{dt} = -\frac{pM\sigma}{6R} \left(1 - \frac{6z}{p} \cdot R \right) \quad (40)$$

Integration from a normal grain size of R_{cr} to zero yields

$$a_p = \frac{6}{pM\sigma} \cdot \left(\frac{p}{6z} \right)^2 \cdot \left[-\ln \left(1 - \frac{6z}{p} \cdot R_{cr} \right) - \frac{6z}{p} \cdot R_{cr} \right] \quad (41)$$

or, approximately

$$\frac{1}{a_p} = \frac{pM\sigma}{3R_{cr}^2} \cdot \left(1 - \frac{6z}{p} \cdot R_{cr} \right)^{2/3} \quad (42)$$

This particular approximation was chosen because it is good for small R_{cr} and also predicts that a_p approaches infinity at $R_{cr} = p/6z$ in agreement with the basic equation (40). By introducing (42) into (34) we get

$$\frac{dR_{cr}^2}{dt} = \frac{1}{3} M\sigma \cdot \sum_p p \cdot \left(1 - \frac{6z}{p} \cdot R_{cr} \right)^{2/3} \cdot c_p \quad (43)$$

where the summation is carried out over all p values

yielding positive terms. By a series expansion it can be shown that equation (43) becomes identical to equation (27) for low values of zR_{cr} if the effect of cooperating defects is neglected and the value $c_1 = 3\alpha/2$ is chosen in agreement with equation (38). This result may be taken as an indication that equation (27) is approximately correct. When comparing the two equations at larger values of zR_{cr} , it is realized that equation (43) predicts a series of limiting values for R_{cr} . At the first limit, $R_{l_1} = 1/6z$, most of the single defects are pinned by the second-phase particles because a 5-grain of the average size, $R = R_{cr} = 1/6z$, will not shrink, according to equation (39). Grain growth can continue only by the action of cooperating defects. At the next limit, $R_{l_2} = 1/3z$, most of the groups of two defects are also pinned because a 4-grain of the size $1/3z$ cannot shrink. In view of this effect one should expect the growth rate to decrease gradually as the final limiting grain size is approached, in qualitative agreement with equation (27). One may guess that the final limit corresponds to the largest p value that is physically possible. This occurs for a grain with three neighbors, $p = 6 - n = 3$, and gives $R_{l_3} = 1/2z$. This is in excellent agreement with the grain size limit predicted by equation (27). On the other hand, in view of the defect-model one might expect that the grain growth will stop earlier if the size distribution is fairly uniform because the number of groups with three cooperating defects might then be negligible. As a consequence, the defect-model seems to suggest that normal grain growth should stop already when the groups of two defects are pinned, i.e. when $R_{cr} = 1/3z$. There should thus exist two grain size limits as indicated in Fig. 5. The lower limit is situated at about $1/3z$ and normal grain growth cannot occur above this limit. The upper limit is situated at $1/2z$ and no grain growth at all can take place in a material that has a larger grain size. Between the two limits abnormal grain growth can take place if there already are some grains much larger than the average. According to equation (23) such grains will grow with a rate of

$$\frac{dR}{dt} = \alpha M\sigma \cdot \left(\frac{1}{R_{cr}} - \frac{z}{\alpha} \right) \quad (45)$$

and this expression yields positive values as long as $R_{cr} < 1/2z$ for a two-dimensional system where $\alpha = \frac{1}{2}$. If the normal grain growth has stopped close to $1/3z$, an abnormally large grain can still grow with a rate of

$$\frac{dR}{dt} = \frac{M\sigma}{6R_{cr}} \quad (46)$$

According to this estimate, the second-phase particles

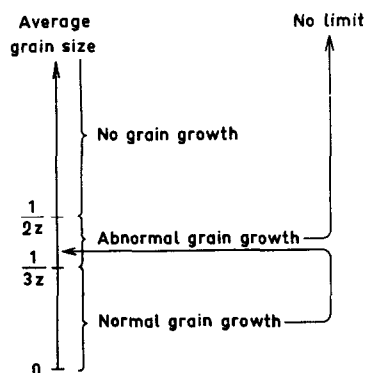


FIG. 5

will decrease the growth rate of an abnormally large grain by a factor of $\frac{1}{3}$ only, when the normal grain growth is completely controlled by them. Of course, the final grain size obtained by abnormal grain growth may be much higher than the upper limit $1/2z$. It is limited only by the number of grains that were large enough to grow and absorb the matrix of grains with normal sizes.

7. THE INITIATION OF ABNORMAL GRAIN GROWTH

As demonstrated in Fig. 5 the defect-model seems to predict that abnormal grain growth can develop in a material if three conditions are simultaneously fulfilled.

- (1) Normal grain growth cannot take place due to the presence of second-phase particles.
- (2) The average grain size has a value below the limit $1/2z$.
- (3) There is at least one grain much larger than the average.

Whether these conditions are automatically fulfilled in a material where the normal grain growth has stopped due to the presence of second-phase particles, is a question of considerable practical importance. We shall therefore examine the process in more detail. From Section 3, we know that there are grains as large as $2R_{cr}$ during normal grain growth. From Section 4, we know that this maximum size should increase further as the limit is approached. In fact, equation (26) even predicts that the maximum size approaches infinity as R_{cr} approaches the limit $1/2z$. However, equation (26) is approximately valid and it does not seem justified to use it for predicting that a few grains will actually have time enough to grow to an abnormally large size before all grain growth has stopped. In view of the fact that the Zener effect becomes appreciable only close to the limit, it would seem more probable that the fairly uniform size distribution attained during normal grain growth far below the limit, will

not tend to change much until close to the limit. Many individual grains may thus reach a size larger than the limit $1/3z$ and may then be stabilized by the second-phase particles. The smaller grains are not stabilized and may shrink and disappear before grain growth stops completely. As a result, we may tentatively suggest that the final grain size may very well be somewhat higher than the limit $1/3z$, as indicated in Fig. 5, or even higher than the upper limit $1/2z$, but not very much higher. As a consequence, it seems reasonable to expect that normal grain growth will not automatically develop into abnormal growth of a few grains. The lower part of Fig. 6 demonstrates how the normal grain growth proceeds when the effect of the second-phase particles is negligible. All the lines are here calculated from equation (16) with $\gamma = 4$. The upper part of Fig. 6 demonstrates what would happen as the grain growth is affected by second-phase particles. In this part of the figure the lines are tentatively drawn, in accordance with the suggested mechanism.

Next, we should ask whether a very large grain, formed by some extraordinary process, could grow abnormally in a material where the normal grain growth has stopped. The answer depends upon the value of the final grain size reached by the normal grain growth. The defect-model does not give any clear-cut answer to this question because of the uncertainty regarding the exact value of the final grain size. The mathematical analysis, on the other hand, predicts that normal grain growth should proceed up to the limit $1/2z$, thus making abnormal grain growth impossible except for a limited time period when the normal grain growth

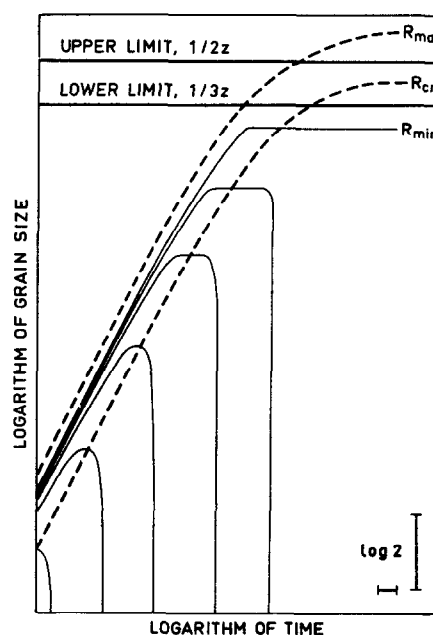


FIG. 6

has started to slow down but has not yet reached the limit.

Finally, we shall consider the possibility of initiating abnormal grain growth by a continuous decrease of the z value. This effect can be accomplished by increasing the particle size r through coalescence or by decreasing the volume fraction f by dissolving the second phase.

During prolonged grain growth experiments one could expect coalescence of the second-phase particles and we could then expect the quantity z to vary as $1/\bar{r}$ and the grain size limits will thus vary as \bar{r} , i.e. proportionally to $t^{1/3}$ in view of equation (2). After sufficiently long times the actual grain size will thus grow slowly as

$$R_{cr} = K \cdot t^{1/3} \quad (44)$$

where the value of K mainly depends upon the constant k in equation (2). If there are two "second-phases" in the material, the back stress will be $S = \sigma \cdot z = \sigma \cdot (z_1 + z_2)$ where z_1 and z_2 are evaluated for each phase separately. If z_1 and z_2 are of different orders of magnitude, one can neglect the effect of the least effective phase. On the other hand, if the most effective phase coalesces faster than the other one, the grain size will gradually change over, as demonstrated by Fig. 7. In view of this picture it does not seem surprising that the experimental values of the time exponent are usually appreciably less than the "theoretical" value of $\frac{1}{3}$.

In Fig. 7 we have assumed that the grain growth proceeds in the normal fashion during the coalescence. Figure 8 demonstrates two other possibilities. This figure is intended as a continuation of Fig. 6. First, it shows how the normal grain growth does not start until the lower limit has reached the value of R_{cr} . From then on, R_{cr} is suggested to follow the limit closely. As a consequence, an unusually large grain can now start to grow abnormally even though it might previously have been inhibited because the final grain size, reached by the normal grain growth in Fig. 6, was too high.

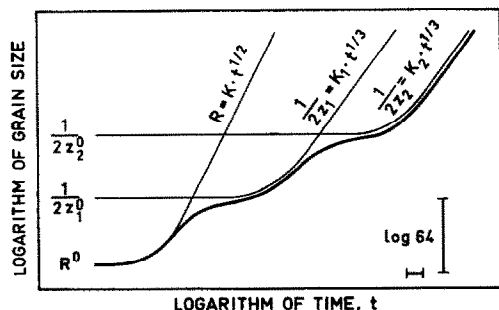


FIG. 7

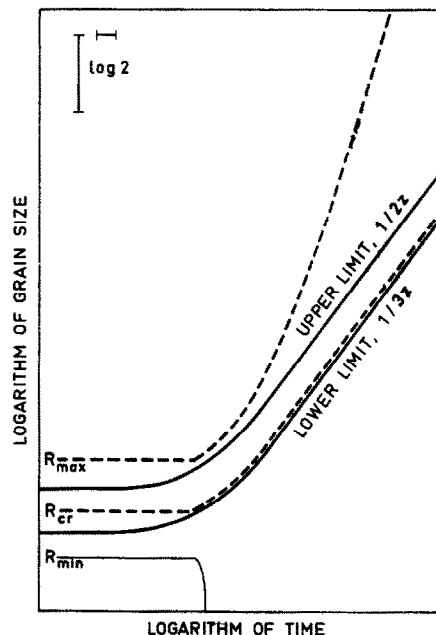


FIG. 8

Secondly, Fig. 8 demonstrates how the grain size distribution grows wider during coalescence by the maximum grain size increasing somewhat faster than the average. This process might develop into abnormal grain growth but it seems impossible at the present to predict how long a time would be needed.

Of course, any process that leads to a slow increase of the grain size limit may initiate the development of abnormal grain growth by the same mechanisms. Most cases of abnormal grain growth, met with in practice, seem to be connected with the dissolving of a second-phase rather than the coalescence. The defect-model does not seem to suggest any immediate explanation of this observation. In terms of the mathematical analysis, on the other hand, there is a slight difference that may prove essential.

As already pointed out, equation (26) seems to suggest that normal grain growth should always develop into abnormal grain growth as the limit $1/2z$ is approached. This conclusion could not be accepted before, in view of the fact that γ_0 is not a constant as the limit is approached. However, if z decreases by coalescence as R_{cr} increases, it seems possible that zR_{cr} could remain constant, thus making the solution described by equations (25), (26) and (27) a true steady state solution. Let us examine the case where R_{cr} follows the limit $1/3z$ closely, thus making $zR_{cr} = \frac{1}{3}$. Equation (26) will then predict that there is a new asymptotic size distribution with a maximum size equal to $6R_{cr}$, instead of $2R_{cr}$. According to the argument by Lifshitz and Slyozov, we should thus expect

that a fraction of grains at the upper end of the size distribution curve, established during normal grain growth, would grow rapidly during coalescence.

Even though we have assumed that zR_{cr} remains constant during coalescence, the solution described by equations (25), (26) and (27) still is not a true steady state because equation (27) predicts a parabolic growth of R_{cr} whereas coalescence only permits R_{cr} to increase proportional to $t^{1/3}$. A true steady state can only be obtained if R_{cr} is allowed to grow faster than during coalescence. This might happen in a limited time period during the dissolution of the second-phase, thus yielding a possible explanation why most of the well-known cases of abnormal grain growth seem to take place when a second-phase is dissolving.

8. ABNORMAL GRAIN GROWTH IN THREE-DIMENSIONAL SYSTEMS

The defect model is strictly applicable only to a two-dimensional array of grains. In a three-dimensional system there is no rule about a constant number of neighbors per grain. In spite of this, it seems reasonable to assume that the general line of argument in Sections 5, 6 and 7 can be applied to a three-dimensional system as well as to a two-dimensional. We may thus suggest that there are always two grain size limits. Normal grain growth can only occur below the lower limit. Between the two limits a large grain would be able to grow abnormally but the risk of developing a grain large enough to grow may be slight under ordinary conditions. The risk of developing a large grain is much higher if the limits slowly move to larger sizes by coalescence or dissolution of the second-phase particles. A safe method of avoiding abnormal grain growth would be to form an average grain size so much larger than the upper limit, that it will remain larger even when the limit moves to larger values during the heat treatment of the material. Such a large size can be obtained by phase transformation or recrystallisation.

Another way to avoid abnormal grain growth would be to decrease the distance between the two size limits. An obvious method to achieve this would be to choose a material with a larger volume fraction of the second phase. Let us consider how the conditions will change if we keep the number of particles constant but increase the volume fraction, f . Both limits will decrease. However, it does not seem possible ever to stop normal grain growth at a grain size smaller than the distance between the particles. After the limit for normal grain growth has reached this value it will stay there but the upper limit will probably decrease further and thus move closer to the lower limit. As a consequence,

we may conclude that abnormal grain growth is not very likely to occur in a material where most of the particles are observed to be situated in the grain boundaries.

APPENDIX

Calculation of the Size Distribution During the Steady State

At the steady state $\gamma = \gamma_0 = 4$ and equation (16) can be written

$$\frac{du^2}{d\tau} = \gamma_0(u - 1) - u^2 = -(2 - u)^2 \quad (\text{A1})$$

$$\frac{du}{d\tau} = -\frac{(2 - u)^2}{2u} \quad (\text{A2})$$

The number of grains between u and $u + du$ at a certain time τ will be denoted by $\varphi(u, \tau) \cdot du$ and the total number of grains at the same time by $N(\tau)$. The distribution function φ must satisfy the continuity equation in the grain-size space

$$\frac{\partial \varphi}{\partial \tau} + \frac{\partial}{\partial u} \left(\varphi \cdot \frac{du}{d\tau} \right) = 0 \quad (\text{A3})$$

By trying the solution

$$\varphi = \chi(\tau + \psi) \frac{du}{d\tau} \quad (\text{A4})$$

where we know that $\frac{du}{d\tau}$ is a function of u only, we find

$$\frac{d\psi}{du} = -1 \frac{du}{d\tau} \quad (\text{A5})$$

and on integration we obtain

$$\begin{aligned} \psi &= \int_0^u \frac{du}{\frac{du}{d\tau}} = \int_0^u \frac{2u du}{(2 - u)^2} \\ &= 2 \left[\ln(2 - u) + \frac{2}{2 - u} - \ln 2e \right] \end{aligned} \quad (\text{A6})$$

The function χ can now be calculated from the information that the size K of the whole system must be constant:

$$\begin{aligned} K &= \int_0^2 R^\beta \cdot \varphi \cdot du = \int_0^2 u^\beta \cdot R_{cr}^\beta \cdot \varphi \cdot du \\ &= \int_0^2 \exp \frac{1}{2} \beta \tau \cdot u^\beta \cdot \varphi \cdot du \\ &= \int_0^2 \exp \frac{1}{2} \beta \tau \cdot u^\beta \cdot \frac{\chi}{\frac{du}{d\tau}} \cdot du \end{aligned} \quad (\text{A7})$$

The constant β is 2 in two dimensions and 3 in three dimensions. The variable τ was introduced using equation (17). The integration is only carried out up to the value $u = 2$ because larger grains are not allowed in the steady state. The value of K must be independent of τ which is possible only if $\chi \cdot \exp \frac{1}{2}\beta\tau$ is independent of τ . It is thus necessary that χ can be expressed as

$$\chi(\tau + \psi) = B \cdot \exp[-\frac{1}{2}\beta \cdot (\tau + \psi)] \quad (\text{A8})$$

The value of the constant B could be calculated by numerical integration, using (A6) and inserting (A8) and (A2) in equation (A7). By inserting (A8) in (A4), we obtain

$$\varphi = \chi \frac{du}{d\tau} = B \cdot \exp -\frac{1}{2}\beta\tau \cdot \exp -\frac{1}{2}\beta\psi \frac{du}{d\tau} \quad (\text{A9})$$

and we can calculate the total number of grains in the system, using (A5):

$$\begin{aligned} N(\tau) &= \int_0^2 \varphi du \\ &= -B \cdot \exp -\frac{1}{2}\beta\tau \cdot \int_0^\infty \exp -\frac{1}{2}\beta\psi \cdot d\psi \\ &= \frac{2}{\beta} \cdot B \cdot \exp -\frac{1}{2}\beta\tau \cdot \left| \exp -\frac{1}{2}\beta\psi \right|_0^\infty \\ &= -\frac{2}{\beta} \cdot B \cdot \exp -\frac{1}{2}\beta\tau \end{aligned} \quad (\text{A10})$$

Let $P(u) \cdot du$ be the probability that the size of a grain is between u and $u + du$. Then

$$P(u) = \frac{\varphi(u, \tau)}{N(\tau)} = -\frac{1}{2}\beta \exp -\frac{1}{2}\beta\psi \frac{du}{d\tau} \quad (\text{A11})$$

Inserting (A2) and (A6) yields

$$P(u) = \frac{\beta u}{(2-u)^{2+\beta}} \cdot (2e)^\beta \cdot \exp \frac{-2\beta}{2-u} \quad (\text{A12})$$

This is the steady state distribution during normal grain growth.

From the experimental point of view, the mean value \bar{R} of the individual sizes is of considerable interest. This value can be calculated using the expression for $P(u)$:

$$\begin{aligned} \bar{u} &= \int_0^2 u \cdot P(u) \cdot du = \\ &= \int_0^2 \frac{\beta \cdot (2e)^\beta \cdot u^2}{(2-u)^{2+\beta}} \cdot \exp \frac{-2\beta}{2-u} \cdot du \end{aligned} \quad (\text{A13})$$

Using the new variable $x = 1/(2-u)$ we obtain

$$\bar{u} = \int_{\frac{1}{2}}^\infty \beta \cdot (2e)^\beta \cdot (4x^\beta - 4x^{\beta-1} + x^{\beta-2}) \cdot \exp -2\beta x \cdot dx \quad (\text{A14})$$

With $\beta = 2$ we find $\bar{u} = 1$ and thus $\bar{R} = R_{cr}$. With $\beta = 3$ we find $\bar{u} = \frac{8}{9}$ and thus $\bar{R} = \frac{8}{9}R_{cr}$. The latter result is formally identical to the case of interface-controlled coalescence which has been treated by Wagner.⁽¹¹⁾

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