GRAIN GROWTH IN METALS[†]

P. FELTHAM[‡]

The sizes and shapes of grains in annealed metals, characterised respectively by the grain diameters and the interfacial angles, are shown to be lognormally distributed in planar sections as well as in space. The similarity of the size and shape distributions facilitates the treatment of grain growth as a univariant statistical problem in which the mean rate of growth of the grains is obtained as the resultant of the surface tension-controlled rate of growth of the individual grains in the distribution. The most probable initial and instantaneous grain diameters D_0^* and D^*, which have approximately the same respective values whether referred to planar or spatial distributions, are then found to be related to the time of isothermal growth t by the equation $(D^*)^2 - (D_0^*)^2 = (\lambda V a\sigma/h)t \exp(-H/kT)$, where λ is a numerical constant of order unity, V the volume per atom, a the lattice spacing, σ the specific grain-boundary energy, h Plank's constant, and H the activation energy for grain-boundary self-diffusion. Agreement with experimental results is good.

CROISSANCE GRANULAIRE DANS LES MÉTAUX

L'auteur montre que les formes et tailles des grains de métaux recuits, caractérisées respectivement par les angles à l'interface et les diamètres des grains, sont distribuées de façon simplement logarithmique aussi bien dans des sections planes que dans l'espace. Cette similitude de la distribution des formes et des tailles facilite le traitement de la croissance comme un problème statistique univariant où la vitesse moyenne de croissance est la résultante de la vitesse de croissance individuelle, contrôlée par mesure de la tension superficielle des grains dans la distribution.

Les diamètres des grains les plus probables (D_0^*) et instantané (D^*), qui ont d'ailleurs approximativement les mêmes valeurs respectives pour les distributions planes ou spatiales, sont liés au temps de croissance isotherme t par l'équation $(D^*)^2 - (D_0^*)^2 = (\lambda V a\sigma/h)^t \exp(-H/kT)$, où λ est la constante numérique d'unité d'ordre, V le volume atomique, a la constante du réseau, σ l'énergie spécifique de la frontière, h la constante de Planck et H l'énergie d'activation pour l'auto-diffusion de la frontière granulaire.

L'accord entre cette formule et les résultats expérimentaux est bon.

KORNWACHSTUM IN METALLEN

Es wird gezeigt, dass Grösse und Form der Körner in ausgeglühten Metallen, charakterisiert durch die Korndurchmesser und die Korngrenzwinkel, sowohl in ebenen Schnitten als auch bezüglich ihrer räumlichen Verteilung eine Normalverteilung mit logarithmischem Argument aufweisen. Die Ähnlichkeit der Grössen- und Form-Verteilung erleichtert die Behandlung des Kornwachstums als statistisches Problem mit nur einer Variablen, bei dem man die mittlere Wachstumsgeschwindigkeit der Körner als Resultierende aus der durch die Oberflächenspannung bestimmten Wachstumsgeschwindigkeit der einzelnen Körner innerhalb der Verteilung erhält. Man findet dann, dass die wahrscheinlichsten Werte für die Korndurchmesser D_0^* und D^* (D_0^* Anfangswert, D^* laufender Wert), die näherungsweise unabhängig davon sein dürften, ob man sie auf eine ebene oder auf eine räumliche Verteilung bezieht, mit der Zeit t durch die Gleichung $(D^*)^2 - (D_0^*)^2 = (\lambda V a\sigma/h)t \exp(-H/kT)$ verknüpft sind; dabei bedeuten λ eine numerische Konstante von der Grössenordnung Eins, V das Volumen pro Atom, a die Gitterkonstante, σ die spezifische Korngrenzenergie, h die Plancksche Konstante und H die Aktivierungsenergie der Korngrenzen-Selbstdiffusion. Die Übereinstimmung mit experimentellen Ergebnissen ist gut.

1. INTRODUCTION

The driving forces leading to grain-boundary migration in crystalline materials are determined not only by the physical constants characteristic of the substances in bulk, but also by the shapes and dimensions of the individual grains. Parameters characterizing the dimensions and shapes of the distribution of grains of an aggregate must therefore appear as variables in the time-laws of grain growth.

Up to now the appropriate statistical treatment has not been given, presumably because of the comparative dearth of suitable experimental material, and the rather complex nature of the problem. Several aspects of grain growth have, however, received considerable attention. In particular, Burke[1] established that the driving force of grain growth in annealed metals was essentially the surface energy (surface tension) of the grain boundaries, which impelled the boundaries to migrate towards their respective centres of curvature. Smith[2,3,4] considered grain shapes and other relevant metallurgical applications of topology, and Beck[5] reviewed some of the more recent experimental work on grain growth. Much earlier an attempt was made by Tammann and Crone[6] to account for the shape of the grain-size distribution curves in cast and annealed aluminium

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and cadmium: the work is unfortunately invalidated by a number of errors.

The problem of grain growth as a whole was approached recently by Cole, Feltham, and Gillam.(7) They found for isothermal growth the relation $(D^*)^2 - (D_0^*)^2 = K_0 t \exp(-H/kT)$, where D^* and D_0^* are the most probable instantaneous and initial grain diameters respectively, t the time, H the activation energy for grain-boundary self-diffusion, and K_0 a constant proportional to the volume per atom, the lattice spacing, and the specific surface energy at grain boundaries. The magnitude of K_0 to be expected for annealed metals should, according to their theory, be of the order of 0·1 cm^2/sec. The above relation gave a very satisfactory description of their experimental results, but in deriving it they by-passed the statistical treatment by making the crucial assumption that the mean growth-rate of the grain area was an invariant fraction of the growth-rate of a grain of diameter equal to the most probable diameter, the boundaries of which were concave parts of spherical surfaces with radii of curvature equal to $\frac{1}{2}D^*$.

The principal object of the present work is to develop a more rigorous theory of isothermal grain growth in metals by using the grain diameters and grain-boundary curvatures as statistical variables, and by making allowance for the restrictive conditions imposed by surface-tension and space-filling requirements.

2. THE OBSERVED DISTRIBUTION OF GRAIN SIZES AND SHAPES IN ANNEALED POLYCRYSTALLINE METALS

The experimental determination of the true spatial grain sizes, although it has been undertaken in a few cases (Roll,(8) Scheil and Wurst,(9) see also Smith(3)) is generally very laborious. In general, therefore, measurements are made on planar sections which are prepared by cutting the specimen in which grain growth has taken place.† It is therefore desirable to study first of all the size and shape distributions in such planar sections, and then to attempt to relate these to the corresponding true spatial distributions.

In relation to distributions as seen in random planar sections, Beck(5) has shown, on the basis of planimetric measurements of grain areas in the central zones of strips of aluminium of high purity containing 500–1000 grains, that at three consecutive periods of isothermal growth at 500°C the grain size-distributions were skew, but that all three curves could be superimposed by making proportional changes in the grain-size scale. The latter observation is of particular interest, because it implies that the functional form of the normalized grain-size distribution should remain invariant if the grain-size scale is put on a logarithmic base.

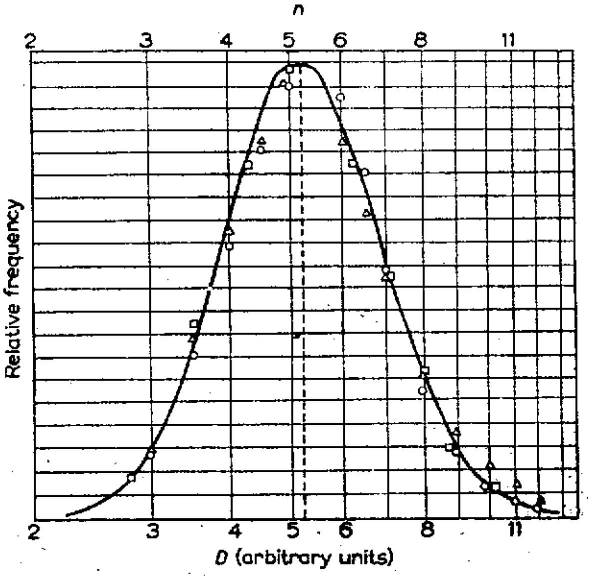

FIG. 1. The relative frequency of grains in planar random sections of annealed aluminium of high purity as functions of the number of sides per planar grain (circles) and the grain diameters (triangles), using data from Beck.(5) The points denoted by squares represent true spatial distribution of grain diameters; the full line is the theoretical lognormal curve.

In fact, if Beck's smoothed composite curve is plotted in this manner, it is found to be very nearly lognormal. In Fig. 1 the lognormal distribution is drawn as a full line; the triangles represent points from Beck's curve. He also found that the relative frequency of occurrence of grains with a given number of sides (or corners) in a planar section remained invariant during isothermal growth. Again, if this distribution is plotted on a logarithmic base, it is found to approximate closely to the lognormal curve. The points denoted by circles in Fig. 1 are taken from the smoothed curve drawn by Beck through the superimposed distributions.

The essential identity of the form of the size and shape distributions apparent from Fig. 1 suggests that a statistical one-one relation might exist between them, i.e. the smaller grains would, in general, be those with few sides, and the larger grains those with many sides. As the actual micrographs used by Beck were inaccessible, and as it was in any case desirable to confirm the generality of the lognormality of the distributions in other metals, similar size and shape determinations were carried out on a micrograph of a

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† As will be seen later, the growth of grains in the surface is not representative of the growth within the specimen; a new section must therefore be made for observation at later stages of isothermal growth if the same specimen is used.

planar section cut through a lump of annealed Chempur tin. A circular zone lying in the centre of the sample (cylinder of 3 cm diameter and 1·5 cm height) containing approximately 1000 grains was utilized. The distribution of shapes, shown in Fig. 2, is again lognormal. The approximate distribution of the sizes of grains with from three to seven sides is shown in Fig. 3. The scatter of the points can be seen to be considerable; an impracticably high count would have to be made to obtain significantly improved accuracy for values of n below 4 or above 6. For the case $n = 7$, measured points are not shown, as this would have rendered the diagram rather confusing. However, the most probable diameters of grains with $n > 7$ were taken to correspond to the centres of the observed respective grain-diameter ranges, the latter being plotted on a logarithmic base. The results are shown in Fig. 4. As was to be expected, the scatter of the points for $n > 7$ is rather greater than for lower values of n. The anticipated linear relation between grain size and grain shape is, however, confirmed over the entire range of n values observed in annealed metals (i.e. from 3 to about 15). By means of this relation it is possible to simplify the statistical treatment of grain growth, as developed in subsequent paragraphs, to a univariate problem with its attendant boundary conditions.

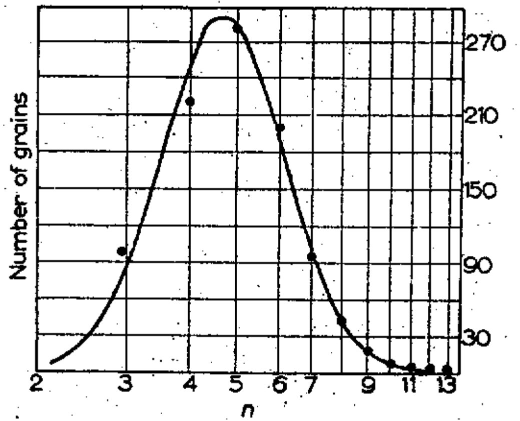

FIG. 2. The distribution of planar grains in a random section of an annealed sample of Chempur tin as a function of the number of sides per grain. The full line corresponds to a lognormal distribution.

The spatial distribution of grain diameters and interfacial angles

A theoretical treatment of the relation between the distributions of grain sizes in planar random sections and the corresponding true spatial distributions has been given by Scheil[10] and Scheil and Wurst.[9] Their method was used, together with tables suitable for computation prepared on the basis of this theory by Johnson,[11] for the determination of the spatial grain-size distribution in the sample of aluminium used by Beck.† The result, denoted by squares in Fig. 1, shows that the spatial grain-size distribution is lognormal, fitting the theoretical curve (full line) rather better than the planar distribution (denoted by triangles). In the particular case of the lognormal distribution the planar grain-size distribution thus

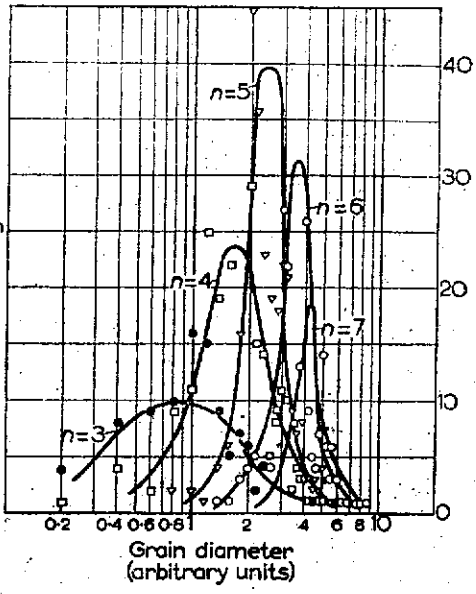

FIG. 3. The approximate distribution of grain diameters as a function of the number of sides per grain, for grains with three to seven sides, obtained from a region containing about 1000 grains in a planar random section of an annealed sample of Chempur tin. (● $n = 3$; □ $n = 4$; △ $n = 5$; ○ $n = 6$. Points for $n = 7$ are not shown.)

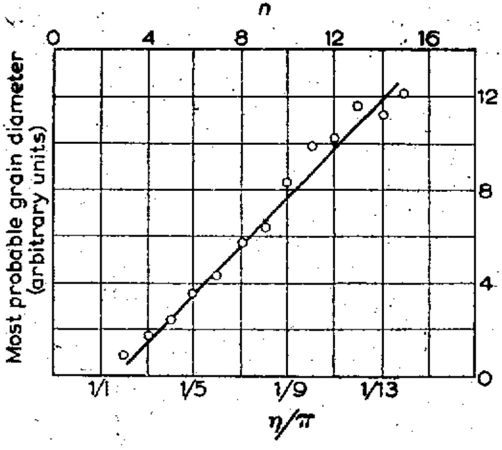

FIG. 4. The relation between the most probable diameters (Fig. 3) and the number of sides (or angle between sides). Section as in Figs. 2 and 3.

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† The regression analysis employed was based upon a nine-step histogram fitted to the curve of planar grain-size distribution.

represents also the spatial distribution of grain diameters closely. The similarity extends not only to the functional form of the distribution curves, but to the actual values of the grain sizes in the two distributions. This is also confirmed in the recent analysis of the relation between planar and spatial grain dimensions given by Spektor.(12) He gives as the ratio of the most probable grain diameters in planar section and in space

$$(D^*)_{\text{sect}}/(D^*)_{\text{space}} = \tfrac{1}{4}\pi(1 + \delta^2), \qquad (1)$$

where δ is the dispersion of the spatial distribution. In a number of cases quoted by him (e.g. armco iron) the ratio is within 10% of unity. We thus have a near-identity of the *size* distributions of grains in planar random sections and in space. If we regard the straight lines joining adjacent corners of grains in a planar section to form regular polygons, then the polygon angle θ and the complementary angle η are related to n, the number of corners (or sides) of the grain, by

$$1/\eta = 1/(\pi - \theta) = n/2\pi. \qquad (2)$$

We then use $1/\eta$ (or η) as an index of the shape of the grain in planar section. We can use the same criterion in the case of spatial grains if θ, in that case, is taken to be equal to the dihedral angle characteristic of the polyhedron. The use of $1/\eta$ is preferable to the use of $n/2\pi$, as the latter is inapplicable to spatial grains. We can now summarize the experimental evidence so far considered in the postulates:

(a) The normalized distributions of the diameters and shapes of grains in planar random sections[†] of annealed pure metals, as well as the normalized distribution of the true grain diameters, are of the same form.

(b) The distributions are lognormal.

As far as the distribution of *shapes* is concerned, Smith(2) has shown that the most probable angle between adjacent sides of grains in planar random sections is equal to the most probable dihedral angle between intersecting grain faces (dihedral angles) in space. Now, as the magnitude of the dihedral angle determines the grain-boundary curvature in the same way as the polygon angle that of a grain in planar section (vide para. 3.2), this observation is particularly useful, as it seems to indicate

(c) That the relations between diameters and shapes and (in so far as the boundary curvatures are assumed to be related to the shapes between) diameters and grain-boundary curvatures are of the same form (vide Fig. 4) whether they refer to planar random sections or to the corresponding spatial distributions.

Although (c) cannot be regarded as rigorously established, the foregoing results render it highly plausible. It will therefore be taken as the third, principal, working hypothesis in developing the theory of grain growth.

3. THEORY

3.1. *The rate-determining mechanism*

We consider the growth of the diameter D_i of the ith grain of the aggregate. This will take place in steps of one interatomic spacing as atoms transfer to it from adjacent regions in the boundary, changing their allegiance from the neighbouring grains to grain i. The frequency of occurrence of translations of sufficient magnitude to effect a translation of an atom is $\nu \exp(-\overrightarrow{Q}/kT)$, where $\nu = kT/h$(13), and $\overrightarrow{Q}$ the activation energy for a jump from the adjacent grain to the end of the diameter of the ith grain. Similarly, the frequency of a return jump, i.e. return of the atom, is $\nu \exp(-\overrightarrow{Q}/kT)$, so that, allowing for the possibility of jumps to both ends of D_i, the rate of addition of atoms to D_i is $2\nu[\exp(-\overrightarrow{Q}/kT) - \exp(-\overleftarrow{Q}/kT)]$. However, only displacements of the atom with amplitude vectors directed towards the grain D_i can be effective. These will comprise only about one-third of the total, so that we have, finally,

$$dD_i/dt = \tfrac{2}{3}a(kT/h)[\exp(-\overrightarrow{Q}/kT) - \exp(-\overleftarrow{Q}/kT)], \qquad (3)$$

where a is the jump distance, i.e. it is equal to the lattice spacing.

We obtain the activation energies from the following considerations. If the diameter terminates in boundaries which have the shape of spherical surfaces with their radii of curvature outside the grain, then if an element of this surface dA advances a distance $d\rho_i$ normal to itself, the accompanying change in area is $(2/\rho_i)dA\, d\rho_i$, and the volume swept out is $dA\, d\rho_i$, where ρ_i is the radius of curvature of the surface. The surface energy per atom of volume V, involved in this advance, is then

$$W_s = 2V\sigma/\rho_i,$$

where σ is the specific surface energy. If the boundaries are not spherical caps, but elements of circular cylinders, the energy is

$$W_c = V\sigma/\rho_i.$$

[†] It is important to differentiate between a planar random section, obtained by sectioning a three-dimensional aggregate, and a true planar distribution (i.e. two-dimensional), such as may be found in coarse-grained thin sheet. The latter type is not, in general, lognormal(6); our present considerations do not apply to it.

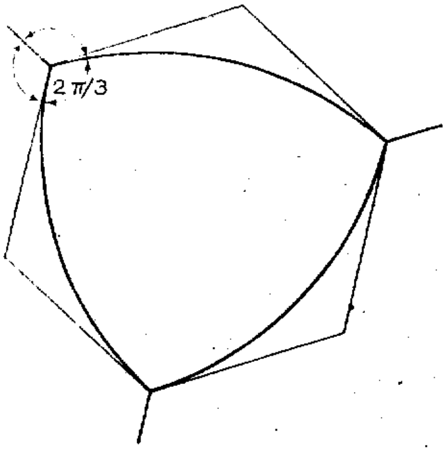

FIG. 5. Triangular idealized grain with curved sides meeting at an angle of 120°. (The sides of the hexagon are tangents at the vertices of the triangle.)

An examination of metal crystals separated from an aggregate[3] shows that a good approximation to the specific energy per volume of atom should be given by the mean of W_s and W_c, i.e.

$$W = \tfrac{3}{2} V\sigma/\rho_i, \tag{4}$$

and this will be used in the present work. In any case, as will be seen, the exact value of the numerical constant in equation (4) is of little consequence.

Thus, if H is the activation energy for grain-boundary self-diffusion, the activation energies in equation (3) will be

$$\overrightarrow{Q} = H - W, \quad \text{and} \quad \overleftarrow{Q} = H + W,$$

and as $W \ll kT$, this gives with equation (3) for the linear growth rate of the grain

$$dD_i/dt = (2Va\sigma/\rho_i h)\exp(-H/kT) \tag{5}$$

on substituting for W from equation (4). Now, σ will be taken as constant, because in randomly oriented aggregates of grains, such as are annealed or recrystallized polycrystals, there is only a small probability that two grains should meet at angles which would give a boundary of low energy.[4] Under isothermal conditions the growth-rate is therefore determined essentially by the magnitude and sign of the local grain-boundary curvature ρ_i.

3.2. *The relation between grain shape and grain-boundary curvature*

If, as was assumed, the specific surface energy σ is independent of the mutual orientation of adjacent grains and the metal is well annealed, the boundaries will tend to meet at the equilibrium angle of 120°, both in spatial and true two-dimensional distributions. This geometric requirement necessitates the introduction of curvature into the grain boundaries. Thus, for example, in a true two-dimensional distribution polygons (idealized grains) with polygon angles less than 120° will develop convex sides, as shown for the triangular grain in Fig. 5, while grains with polygon angles greater than 120° (i.e. number of sides greater than six) will tend to have concave boundaries. Both magnitude and sign of the curvature of the idealized grain will therefore depend upon its "shape" (equation 2).

We shall first consider the relation between shape and grain-boundary curvature in the case of truly planar grains, or, what is equivalent, in the case of a three-dimensional aggregate of columnar grains with their long axes perpendicular to the plane of section. We shall regard the result as a first approximation to the corresponding relation in the case of random planar sections, and subsequently modify it somewhat by semi-empirical means.

Now, curve 1 in Fig. 6 is drawn through the points ($n = 3, 4, \ldots$), representing the ratio

$$f(n/n^*) = r_i/\rho_i = D_i/2\rho_i \tag{6}$$

where n is the number of sides of the regular polygon to which the grain is related, and n^* (=6) the most probable number of sides to be expected in a truly planar distribution. (The adoption of a special symbol for this numerical constant is intended to help in the discussion at a later stage.)

With the convention that the radius of curvature is positive in the case of convex boundaries, e.g. in the case of the three-cornered grain in Fig. 5, it is easily shown by simple geometry that

$$f(n/n^*) = \sin\left[\frac{\pi}{6}\left(1 - \frac{n^*}{n}\right)\right] \Big/ \sin\left[\frac{\pi}{6}\left(\frac{n^*}{n}\right)\right]. \tag{7}$$

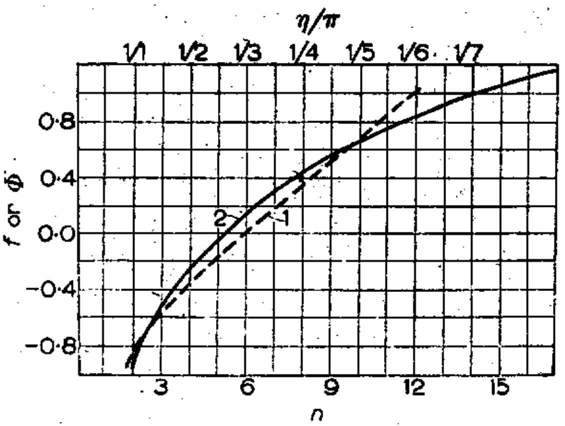

FIG. 6. The functions $f(n/n^*)$ (equations 7 and 8) (curve 1) and

$$\varphi\left[\left(\frac{\eta^*}{\eta} - \alpha\right)\Big/(1-\alpha)\right]$$

(equation 17 and 18) (curve 2).

The grain diameter in equation (6) is taken to be the diameter of the circumscribing circle. As can be seen, $f(n/n^*)$ is negative for n less than 6 and positive for all greater n. As we can write

$$f(n/n^*) = f(\eta^*/\eta) \tag{8}$$

(with $\eta^* = 60°$ in the present case (equation 2), we have as our first approximation, from equation (5) and (6):

$$dD_i/dt = (4Va\sigma/hD_i)f(\eta^*/\eta)\exp(-H/kT) \tag{9}$$

or

$$dD_i^2/dt = Kf(\eta^*/\eta) \tag{10}$$

where

$$K = K_0\exp(-H/kT) = (8Va\sigma/h)\exp(-H/kT) \tag{11}$$

is a constant at any given temperature.

It is apparent from equation (10) that, in a distribution of grains, the grain size D_i and the "shape function" $f(\eta^*/\eta)$ are the two variables to be considered further and that, in view of the functional form of $f(\eta^*/\eta)$, grains with fewer than six sides will tend to diminish in size, while those for which $n > 6$ will tend to grow. The overall increase in the mean grain size on isothermal annealing is then the resultant of the simultaneous operation of both processes taken over all grains.

3.3. *Limiting conditions*

In the preceding paragraph we have dealt with the individual grain. We shall now consider the growth process apparent from planar random sections. As the function $f(\eta^*/\eta)$ relates angles (polygon or dihedral) to grain-boundary curvature as conditioned by surface tension requirements, we would expect it to apply with reasonable accuracy also to grains in a distribution. However, we expect slight deviations for the following reasons, among others:

(a) Grains will not have idealized geometrical shapes, such as are shown for planar six- and three-sided grains in Fig. 5. In fact, it would not in general be possible to fill space with idealized grains without some voids between them. Consequently only the average curvature of, say an n-sided planar grain can be expected to be given by ρ_i if the mean grain diameter is D_i. Equation (6) will therefore not hold exactly.

(b) Although we can reasonably expect η^* to be close to 60°, it may not be exactly equal to that value.

(c) The requirement that, for example, the total area of a planar section under observation must remain invariant during grain growth, as the grains grow at the expense of one another, also imposes limiting conditions on $f(\eta^*/\eta)$.

Now, in relation to (b) we observe that the most probable number of sides per polygon in planar sections is 5·2 (Fig. 1) (and approximately 4·8 in Fig. 2). That the most probable number of sides per polygon in planar sections of annealed metals lies closer to 5 than to 6—in fact, that it is generally equal to about 5·2—has already been observed by Smith.⁽³⁾ As yet, no complete explanation of this fact is available, and, as a first step towards obtaining an improvement on $f(\eta^*/\eta)$, we put $\eta^* = 69°$, (corresponding to $n = 5{\cdot}2$; equation 2), which gives for the most probable polygon (or dihedral) angle $\theta^* = 111°$, i.e. 7·5 % below the ideal value.

The continuity requirement (c) can be stated in a mathematically more convenient form, i.e. the rate of change of the total area must remain zero. Now, if $N(D_p)\,d(\ln D_p)$ is the number of planar grains in the size range $d(\ln D_p)$, and D_p^2 is taken as the grain area, as a first approximation we must have (using the integral rather than the summation):

$$\int_{D_p} N(D_p)(dD_p^2/dt)\,d(\ln D_p) = 0, \tag{12}$$

the integration extending over all grains in the area under observation.

Since by postulate (a) (para. 2.2) we can write

$$N(D_p)\,d(\ln D_p) = A\,N(D)\,d(\ln D)$$

(omitting subscripts i) where A is a constant and D refers to the spatial grain size, and since also (para. 2.2)

$$dD_p^2/dt = dD^2/dt \quad \text{if} \quad D_p = D,$$

the subscript p in equation (12) can be omitted; the latter then applies to the spatial grain-size distribution. To evaluate equation (12) we must now express dD^2/dt in suitable form.

Now, from Fig. 4 we have

$$n/n^* \equiv \eta^*/\eta = \alpha + (1-\alpha)(D/D^*) \tag{13}$$

where $\alpha = \text{const.} = n_0/n^*$, and n_0 the value of n corresponding to $D = 0$. We can therefore define a new function

$$\phi(\eta^*/\eta) = \psi(D/D^*) \tag{14}$$

and write equation (10) (omitting subscript) in the form

$$dD^2/dt = K\phi(\eta^*/\eta) = K\psi(D/D^*) \tag{15}$$

where $\psi(D/D^*)$ is to be determined from the conditions that $\phi(\eta^*/\eta)$ should approximate closely to $f(\eta^*/\eta)$ over the range of η encountered in practice, and that equation (12) be obeyed on substituting $\psi(D/D^*)$ for dD^2/dt (equation 15). Also, of course, we take $\theta^* = 111°$, i.e. $\eta^* = 69°$.

Now, on substituting for the lognormal distribution

$$N(D) \propto \exp\{-b^2[\ln(D/D^*)]^2\}, \quad b = \text{const.}$$

(where D^* is the most probable spatial grain diameter), equation (12) becomes

$$\int_D \exp\{-b^2[\ln(D/D^*)]^2\}\psi(D/D^*)\,d(\ln D) = 0, \qquad (16)$$

the integration extending from $D_{\min}$ to $D_{\max}$. As $\psi(D/D^*)$ is required to approximate to $f(\eta^*/\eta)$, it must be a monotone slowly increasing function over the range of D found in practice. The integral (equation 16) will therefore converge rapidly on both sides of D^*, i.e. for larger and smaller values of D, so that it is permissible to write $D_{\max} = \infty$ and $D_{\min} = 0$. Solutions of the resulting homogeneous Fredholm equation are given by

$$\psi(D/D^*) = \sum_{m=0}^{M} C_m[\ln(D/D^*)]^{2m+1}, \qquad (17)$$

where the C_m's are arbitrary constants. A particular solution which is monotone for all finite values of the variable, which is a good approximation to $f(\eta^*/\eta)$ over the range of dihedral (or polygon) angles found in practice in annealed metals, and which has its zero at the point corresponding to $\theta = 111°$, is given by

$$\psi(D/D^*) = \ln(D/D^*) = \ln\{[(\eta^*/\eta) - \alpha]/(1-\alpha)\}, \qquad (18)$$

where the last equation is obtained from equation (13). The curve obtained with $\eta^* = 69°$ and the appropriate value of α obtained from Fig. 4 is shown as a full line in Fig. 6.

We can therefore now write equation (15) in the form

$$dD_i^2/dt = K \ln(D_i/D^*). \qquad (19)$$

This equation cannot be integrated without a knowledge of the time-dependence of D^*; obviously D^* must increase in the process of grain growth, i.e. with time. In deriving equation (19), our knowledge of the functional *form* of the distribution curve of grain diameters has been utilized (equation 16). In order to determine the time-dependence of D^*, it now becomes necessary to utilize also the *time-invariance* of the shape of the distribution curve.†

3.4. *The mean growth-rate*

A diagrammatic representation of the growth process is shown in Fig. 7. The arrows indicate the respective growth-trajectories of individual grains in their transitions from the lognormal distribution at time t to the distribution at time $t + \Delta t$. Grains which have diameters smaller than D_1 at time t will be absorbed in the interval Δt, and will not appear in the distribution subsequently. Grains with diameters in the range D_1–$D_1{}^*$ will diminish during isothermal annealing, while those with diameters greater than D_1^* will tend to grow. The trajectors will not, in general, intersect.

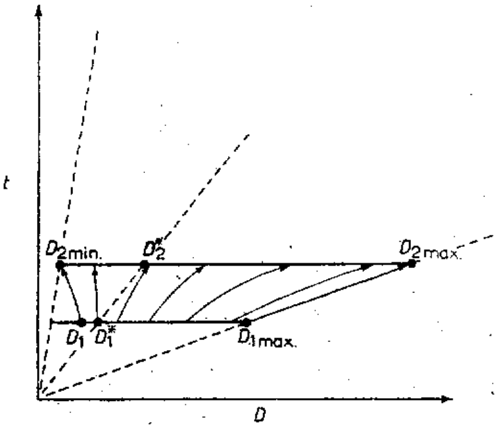

FIG. 7. Diagramatic representation of grain growth for the normalized lognormal (time-invariant) grain-size distribution.

It is important to note that the largest grains in the distribution at time t will also individually be the largest grains at time $t + \Delta t$, since the growth trajectory $D_{1\max} - D_{2\max}$ must, by necessity, be essentially linear. Since the distribution remains lognormal at all times, the ratio

$$D_{\max}/D^* \simeq 2{\cdot}5 \qquad (20)$$

(i.e. 13/5·2 in Fig. 1) is independent of the time, and we can obtain the true, observable, growth-rate of the largest grains from equations (19) and (20):

$$dD^2_{\max}/dt = K \ln 2{\cdot}5,$$

which yields on integration

$$D^2_{\max} - D^2_{0\max} = Kt \ln 2{\cdot}5,$$

or, on substituting for $D_{\max}$ from equations (20), and for K from equation (11),

$$(D^*)^2 - (D_0{}^*)^2 = (\lambda V a\sigma/h)t \exp(-H/kT), \qquad (21)$$

where λ is a numerical constant of order unity.‡ Equation (21), relating the observable variable D^* to the time of isothermal growth, is therefore the required result.

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† It is of interest to observe that the lognormal distribution function should be obtainable as a solution of equation 16 if it were regarded as the unknown function, and $\psi(D/D^*)$ as given by equation (18) were substituted into equation (16) as kernel on account of its close approximation to the "surface tension function" $f(\eta^*/\eta)$. This indicates how surface tension controls the grain-size distribution.

‡ The actual magnitude of D here obtained is 1·3, which compares well with the value of 0·5 given by Cole, Gillam, and Feltham.(7) In cases where σ is not known, it can be estimated from the relation $\sigma \simeq Ga/8$, where G is the shear modulus of the metal.(14)

4. COMPARISON WITH EXPERIMENT AND DISCUSSION

Some factors, apart from those already discussed, which may affect the rate of grain growth in metals were considered by Burke.(1) The most important of these are effects due to nonmetallic inclusions. Zener (vide ref. 1) examined their growth-inhibiting function, and showed that if the growth was surface-tension controlled it would be arrested by inclusions at a stage when

$$\rho_{\mathrm{av}}/\rho_{\mathrm{incl}} = 1/f,$$

where ρ_{av} and ρ_{incl} are the mean radii of curvature of the grain boundaries and inclusions respectively, and f the volume fraction of the inclusion in the metal. The relation provides a means for estimating the magnitude of the effect; no attempt will here be made to incorporate it into equation (21).

The growth-rate may also be influenced by residual internal stresses due to cold work; the energy due to cold work (per atom) may exceed W (equation 4) by an order of magnitude or more if the metal has been heavily cold-worked.(15)

An important effect is observed if the dimensions of the specimen (e.g. thickness of the strip or radius of the wire) are comparable to the mean grain size. Beck(5) and Beck and co-workers(16) first demonstrated that to avoid the slowing-down of grain growth due to "sheet-thickness effect," the minimum dimensions of the specimen should preferably be larger than ten times the mean grain size. The effect is probably due to a number of causes, of which the principal ones are to be sought in the fact that

(a) Owing to the flatness of the surface, the surface grains have much larger average radii of curvature than would correspond to grains of similar respective sizes in the interior of the metal;

(b) In very thin samples, the distribution of grain sizes will not be lognormal (vide footnote in para. 2.2), and the dispersion of grain diameters will be rather narrow,(6) i.e. the grains will be of nearly uniform size with n^* equal or very close to 6. The value of $f(\eta^*/\eta)$ for most grains will thus be low.

This pronounced effect does not appear to have been recognized by a number of workers, with the unfortunate consequence of invalidating much of the work, e.g. studies of the grain growth in nickel,(17) armco iron,(18) in silver, lead, and tin,(6) etc. "Activation energies" derived from such data are invariably of a very high, improbable magnitude, sometimes close to the activation energy of sublimation.

A further precaution which is but rarely observed (but was taken into account, for example, by Wensch and Walker(17)) is the elimination of nucleation such as is likely to persist for considerable periods during growth at comparatively low temperatures if the specimens have not been fully annealed. Again a diminution of the observed growth-rate may result on account of the continued appearance of new, small grains. In work on grain growth in aluminium of high purity, Beck and co-workers(16) found that their result could be represented very well by the empirical relation $(D^*)^m - (D_0^*)^m = K(T)t$, in which m approached the value 2 at temperatures close to the melting-point, but decreased with decreasing temperature. It is possible that but for the variable incidence of nucleation, equation (21) would have proved suitable for the interpretation of their results.

Of the remaining work known to the author, only that of Burke(1) on pure cartridge-brass and that of Cole, Gillam, and Feltham(7) on austenite, lend themselves to comparisons with the theoretical result (equation 21). The latter-named workers found good agreement with equation (21) over a wide range of grain diameters; with very large grains, deviations due to the "sheet-thickness effect" were eventually found. Burke, who actually investigated this effect in annealed cartridge-brass, found the empirical relation

$$dD^*/dt = K(T)[(1/D^*) - (1/D_f^*)]$$

appropriate for the representation of his results, where D_f^* is the most probable limiting diameter attained upon prolonged heating at the temperature T. Its magnitude is determined by the thickness of the strip used as specimen. If $1/D_f^*$ is negligible, integration of Burke's equation leads to the same functional relationship as given by equation (21). Burke obtained an activation energy $H = 40$ kcal/gram atom, but considered his method of obtaining it open to criticism. If, however, H is taken to be 0·7 times the activation energy for volume self-diffusion,(7) and the latter(19) as 43 kcal/gram atom, one obtains $H = 30$ kcal/gram atom. With this value, Burke's data require a value of K_0 of the right order of magnitude to comply with equation (21).

Dean and Hudson,(20) using a high-purity 1% Sb alloy of lead in the form of large cylindrical samples, also found a linear relation between $(D^*)^2$ and t, thus confirming the functional form of equation (21). Although their experiments were carried out at three different temperatures, neither K_0 nor H can be evaluated from their results, as the system was no longer a homogeneous solid solution at the lowest temperature used (150°C).

The experimental work surveyed above, in so far as it lends itself to comparison with the theory, tends to confirm the result given by equation (21). Further data, obtained with the precautions outlined, would nevertheless be valuable.

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