Mechanism of Steady-State Grain Growth in Aluminum

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WITH APPENDIX BY ROBERT T. DeHOFF

Grain growth, defined as the increase in volume of the average grain, is found, in its steady-state, to be directly proportional to the time of isothermal annealing. During steady-state grain growth the grain corners are found all to be quadruple points, the grain edges all triple lines and the ratio of corners to faces to edges to be 6:7:12. The rate constant for steady-state grain growth is shown to be calculable from first principles and from properties that can be measured independently of the growth observation. It is the product of four individual constants, namely: 1) a dimensionless topological constant Θ that is characteristic of steady-state grain growth in any material, 2) the mobility μ of the average grain boundary in the specific material, 3) the surface tension γ of the average grain boundary in the specific material and 4) a dimensionless structural constant σ which expresses the curvature of surface of the grain boundary in the array of grain forms obtaining in the specific specimen of the material and which can be determined metallographically. The topological changes that constitute steady-state growth are shown to exist as a logical sequence of simple events.

GRAIN growth in a polycrystalline aggregate differs fundamentally from the growth of a mass of separate particles, in that the polycrystalline body itself undergoes no growth or shape change. In accord with this restriction, no change can take place in the size or shape of any grain without a compensating adjustment in the system at large. For the size of the average grain to increase, moreover, it is necessary that some grains should disappear from the system. Thus, grain growth in a polycrystalline aggregate amounts to a systematic elimination of grains, coordinated with such grain size and shape changes as are needed to maintain perfect space filling at all times.

A geometry that is suitable for describing the growth of individual particles will be inadequate for the defining of polycrystalline grain growth, unless it can deal also with the connective property that is implied in the coordination of grain sizes and shapes. The familiar Euclidean geometry is deficient in this respect. Models that have been constructed upon a purely Euclidean base have failed to provide an exact, or even realistic, description of grain growth and its kinetics. Such a description can be had, however, by resort to some simple concepts of descriptive topology. All that is required is to express the structure in terms of the numbers of its parts, as connected by Euler's rule for the space network. The numbers that will be used are: the number of grains (N), the number of grain faces (F), the number of grain edges (E) and the number of grain corners (C), all in a specific specimen of the material.

These parameters can be measured directly by serial section analysis, the techniques of which have been reported elsewhere.[1] Such a study has been carried out[2] upon a series of specimens of 99.99+ pct aluminum that had been cold worked at − 195°C and subjected to isothermal grain growth at 635°C for times ranging from 1 min to 1 h. This treatment provided a range of grain size, by volume, of almost three orders of magnitude. In order to avoid the intrusion of surface effects upon the measurements, that portion of each specimen which was analyzed was taken at a distance of not less than ten grain diameters from any external surface. The exact volume of material analyzed in each case was measured and the topological parameters were expressed in terms of number per cubic centimeter, thus: N_V (grains per cm^3), F_V (faces per cm^3), E_V (edges per cm^3), and C_V (corners per cm^3). The results of this study provide the experimental basis for the development that follows.

The *average grain volume*, obtained by taking the reciprocal of the number of grains per cubic centimeter $1/N_V$, provides a measure of grain size* that is independent of grain shape, grain size distribution and shape assortment. It will be shown in the following that the average grain volume is a lineal function of the time, in steady-state grain growth at constant temperature ($1/N_V = kt$).

*The conventional expression for grain size is the mean intercept, obtained by counting the number of grains crossed by unit length of line laid upon a two-dimensional section through the grain structure. This parameter is sensitive to shape. It is a function of the total surface area of the grain boundary, *i.e.*, it is not a grain diameter. Neither this nor any other measurement that can be made upon a two-dimensional section through the material can be used to determine grain volume. N_V must be measured in three-dimensional space.

TOPOLOGICAL NATURE OF THE GRAIN STRUCTURE

The geometric form of the grain structure is defined by the system of grain boundary, which constitutes a topological network. If the grain corners be taken as nodes and the grain edges as branches of a point-line network, Fig. 1, its connectivity in one cubic centimeter of material will be, by network law:[3]

$$\text{Connectivity} = E_V - C_V + 1 \qquad [1]$$

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If, further, the grains themselves be represented as individual points connected by lines representing the grain faces, an interpenetrating network is formed, Fig. 2, and its connectivity is:

$$\text{Connectivity} = F_V - N_V + 1 \qquad [2]$$

The connectivities of the two networks are identical, because both networks correspond to opposite retracts* of the same imaginary surface, Fig. 3, wherefore:

$$N_V + E_V = F_V + C_V \qquad [3]$$

This is a form of Euler's rule for semi-infinite networks. It is valid irrespective of the number of branches connected at each node.

The grain boundary network, being an interface between pairs of crystals of different orientation, has an energy which is felt as a surface tension* [Quincke[5] and Desch[6]). This energy provides the driving force for grain growth. There is always a decrease in the grain boundary area, representing an expenditure of surface energy in accomplishing grain shape change and grain growth, Fig. 4. Surface tension has the immediate effect upon the grain boundary network of causing all junctions of grain faces to occur upon "triple lines"

*The corner-edge network and the grain-face network can be conceived as being separated by a continuous multiply-connected surface, Fig. 3. If this surface be shrunk toward the grain edges, its "retract"[4] becomes the corner-edge network; if it be shrunk upon the grain centers, its "retract" becomes the grain-face network. The connectivity of the surface and of the two networks remains unaltered and, therefore, identical in this operation.

*While it is true that the magnitude of the surface tension of the grain boundary varies with the local orientation difference between pairs of grains, the effect of such orientation difference upon the average behavior of grain growth seems to be minor. This is indicated, for example, by the regularity with which grain faces are found to meet at dihedral angles near 120 deg. It is consistent also with the fact that the grain boundary energy varies only moderately with orientation difference, except in a narrow range of small angle grain boundaries and close to the orientation of twin boundaries (Gjostein and Rhines).[7] In the case of aluminum, few twin boundaries are to be expected.

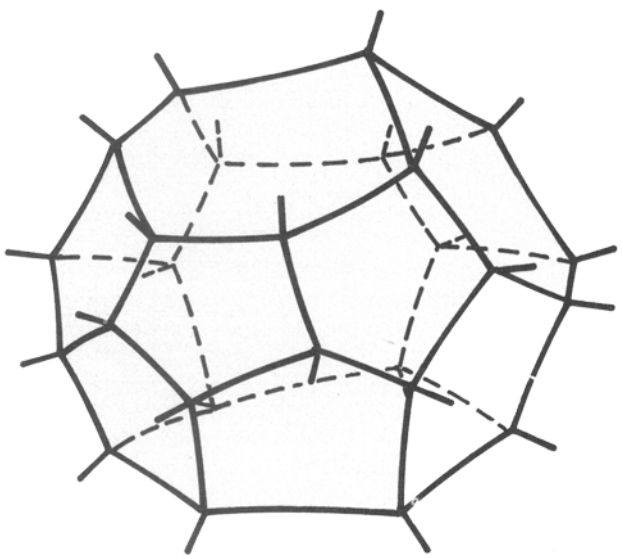


Fig. 1—Edge-corner network surrounding an average grain.

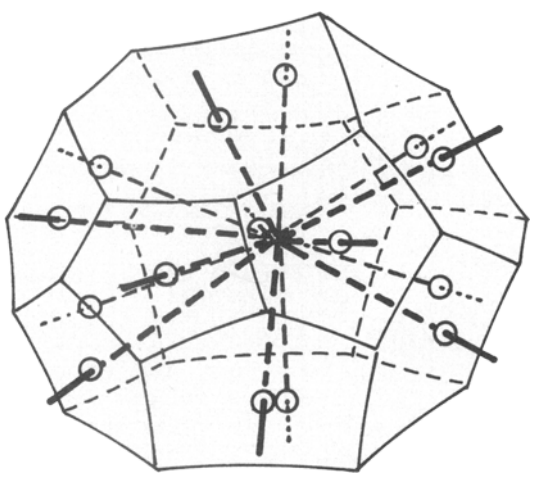


Fig. 2—Interpenetrating grain-face network represented by lines drawn from the center of the grain through each face to the center of each neighboring grain.

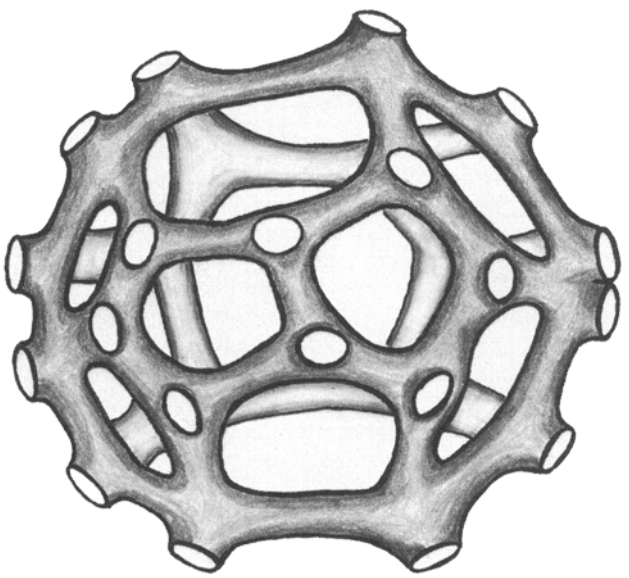


Fig. 3—Imaginary surface separating the edge-corner and the grain-face networks for one grain. Compare with Figs. 1 and 2.

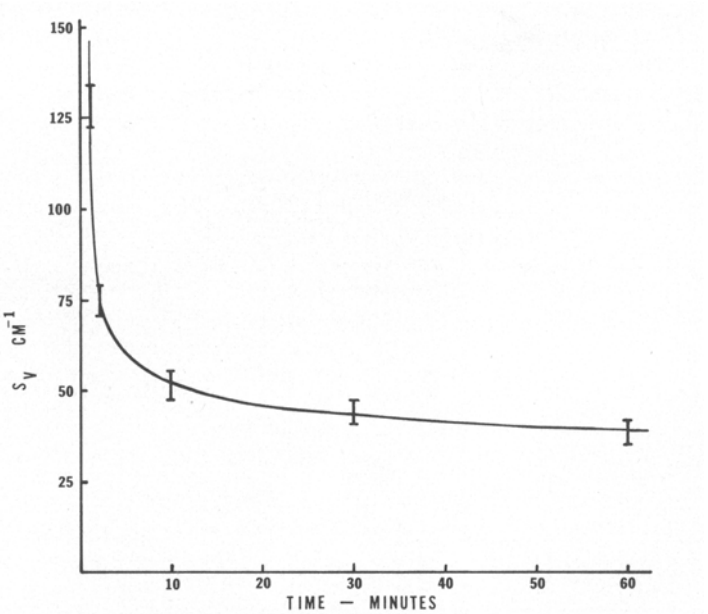


Fig. 4—Decrease in the grain boundary area S_V as a function of annealing time in minutes at 635°C.

where three faces tend to meet at dihedral angles of 120 deg (Harker and Parker)[8] and of causing all triple lines to meet at "quadruple points" where four edges tend to meet at mutually included angles of 109 deg 28 min 16 s, Fig. 5. In the present experimental study, more than 3000 grain corners were analyzed without finding one that was other than a quadruple point. This finding serves to substantiate the belief that surface tension is a major force in shaping the grain boundary network and in motivating grain growth. It also demonstrates that all configurations of the edge-corner network, other than those composed of triple lines and quadruple points, are of such inferior stability as to have extremely short periods of existence.*

J. W. Gibbs[9] has defined the angular relationships obtaining among the interfaces formed by four intersecting bodies, where the respective energies of the several interfaces differ among themselves.

Euler's rule becomes simplified in the triple-line-quadruple-point network by virtue of the fact that there must be exactly twice as many grain edges as grain corners (C. S. Smith):[10]

$$E_V = 2C_V \quad [4]$$

whence, Eq. [3] becomes:

$$N_V + C_V = F_V \quad [5]$$

The grain boundary network is described completely, therefore, by the experimental determination of any two of these three parameters. In practice, it turns out to be advantageous to measure all of the parameters, in order to prove the self-consistency of the measurements by means of Eqs. [4] and [5].*

*An evident source of experimental error resides in the uncertainty of counts where a feature of the network intersects the boundary of the volume being analyzed. The uncertainty in the case of the number of grain corners is negligibly small, because corners occupy no volume and their position with respect to the sampling boundary is rarely ambiguous. Edges, faces and grains can also be identified and counted in terms of the first corner at which each appears. By so counting only those features which originate in the serial section sequence within the test volume, the counting error is made small. A more troublesome source of experimental error exists in the choice of sample size, where it is important to analyze a broad enough area through a sufficient number of serial sections to include a representative sampling of the largest, as well as of the smaller grains. Errors arising from a too small sample produce data that fail to satisfy Eqs. [4] and [5].

Topological quantities are always independent of all dimensional (Euclidean) properties. Thus, the number of grains (N) in a specimen is independent of the shape or size of any grain in the system. The same is true of the number of faces (F), of edges (E) and of corners (C). It follows that a dimensional property, such as the average grain volume, cannot be expressed in purely topological terms. The quantity $1/N_V$ is a hybrid "topological-Euclidean" expression. The introduction of the volume concept (*i.e.*, number *per unit volume*) confers the power to measure and to express average volume. Because the volume of the grain is also independent of its shape, surface area and length of edge, $1/N_V$ remains independent of all dimensions except volume. This expression for the average volume is valid, therefore, for all simple polycrystalline bodies in all stages of grain growth, as long as each grain can be identified as a discrete entity.

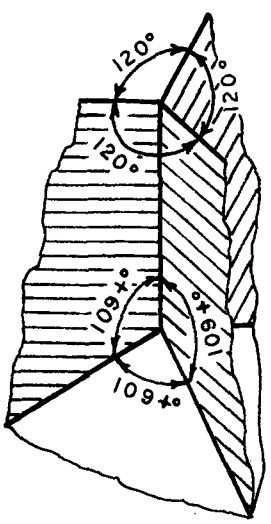


Fig. 5—An equilibrium grain corner where six faces and four edges meet at minimum energy angles, as indicated.

Topological quantities also have the property of existing as whole numbers only. The number of grains (N), in a piece of metal, must be a whole number. There may be grains present in a variety of sizes, but none can be identified as other than a whole grain. The same is true with respect to faces, edges and corners. This situation is not altered by the artifice of counting topological quantities in a limited volume, as is done experimentally. Grain growth occurs in a sequence of discrete steps in which one grain at a time disappears from the system.

This does not mean that an average sized grain vanishes abruptly. On the contrary, the transfer of volume from a shrinking grain to its neighbors and onward to their neighbors is a process that is going on at many sites over extended periods. Ultimately, each grain that is destined to vanish merely arrives at zero size, instantaneously extinguishing all of its associated topological numbers, but causing no discontinuous dimensional change in the system. Its volume and some of its surface have already been contributed to the enlargement of other grains. When it is no longer counted among the grains of the system, the volume assignable to the remaining grains of the system will have increased by a total equal exactly to the *volume of one average sized grain.*

NATURE OF STEADY-STATE GRAIN GROWTH

Steady-state grain growth in metals is characterized by a progressive magnification of the size of the grains without seeming to change their geometric form. This kind of grain growth does not usually occur immediately upon the creation of a new polycrystalline aggregate, but follows after a growth period during which the system of grains has been changing its topological state toward a constant average ratio of corners to faces to edges. At the same time the grain shapes tend toward "equiaxed" forms. Thereafter, steady-state grain growth proceeds in the interior of the metal, where the constraints of external surface pinning are insignificant.* Under the latter circum-

*The two-dimensional external surface network of grain boundary differs from the three-dimensional internal network in that the former can become stable with straight edges outlining hexagons with perfect 120 deg angles. In the absence of curvature there is, then, no further tendency for the two-dimensional network to undergo growth. When this happens, the three-dimensional network, which is anchored to the surface network, is restrained in its growth.

stance, the progress of grain growth is susceptible to rigorous analysis and its kinetics can be derived and expressed exactly.

Serial section studies upon aluminum have shown that, in the steady-state stage, the average number of corners per grain approaches 6, the average number of faces per grain approaches 7, and the average number of edges per grain approaches 12, Fig. 6. Topologically, these numbers describe the tetrakaidecahedron,* Fig. 7, which was identified by C. S. Smith[10] as the geometric figure most likely to fill space with its edges and corners most nearly complying with the angular requirements imposed by surface tension. Thus, *during steady-state grain growth the change that takes place can be described as the elimination, one at a time, of grains of average volume and having the average topological parameters of a tetrakaidecahedron.*

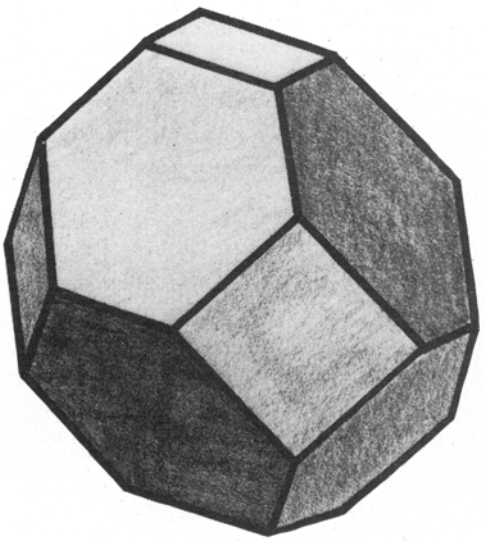
* These are the "sharing numbers" for the tetrakaidecahedron, see Table I, Column *D*.

The change toward equiaxed form, with equilibrium angles at triple lines and quadruple points, constitutes a *local response* to the force of surface tension, tending to minimize surface area. Its immediate goal is an array of grains of assorted sizes, approximating equiaxed form and achieving nearly equilibrium angles by so curving the faces as to permit them to meet at minimum energy angles. It has been demonstrated by C. S. Smith[10] that there exists no grain shape that can permit simultaneous space filling and equilibrium angles without curvature of face, although this condition is approached by regular tetrakaidecahedra.

Grain growth without further significant shape change, is a *long range response* to the force of surface tension, resulting in continued reduction in the grain boundary area. It is characterized by a sweeping migration of the grain boundaries, edges and corners. The driving force for this kind of process arises from the grain boundary surface tension (γ) acting upon curved surface to produce a pressure (P), normal to the grain boundary, Fig. 8, according to the well known relationship:

$$P = \gamma \tfrac{1}{2}\left(\frac{1}{r_1} + \frac{1}{r_2}\right) \text{(dynes/cm}^2\text{)} \qquad [6]$$

where r_1 and r_2 are the principal radii of curvature at the point of application of the pressure.



	C	F	E
On the cell	24	14	36
Shared features	6	7	12

Fig. 7—A 14-sided figure, a tetrakaidecahedron, representing the average topological form of grains in the polycrystalline aggregate.

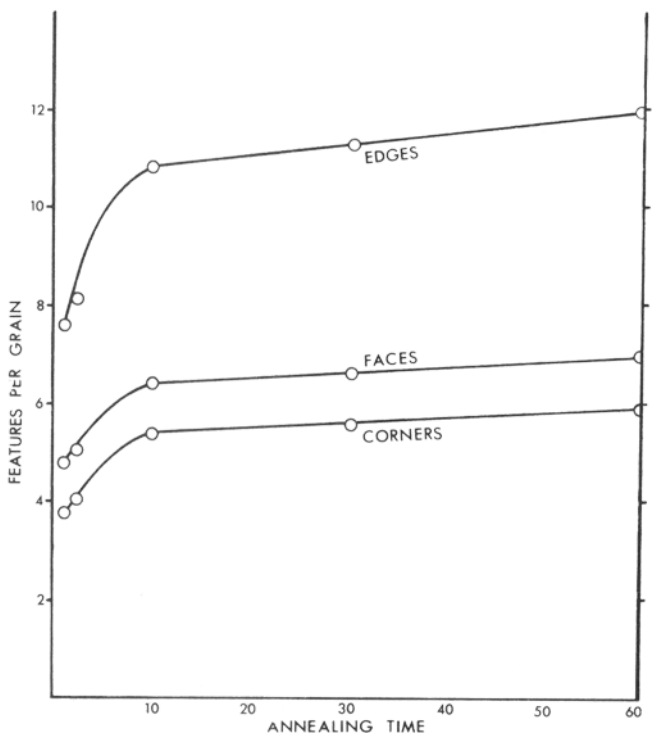


Fig. 6—Numbers of edges, faces and corners per grain, as a function of the annealing time in minutes at 635°C.

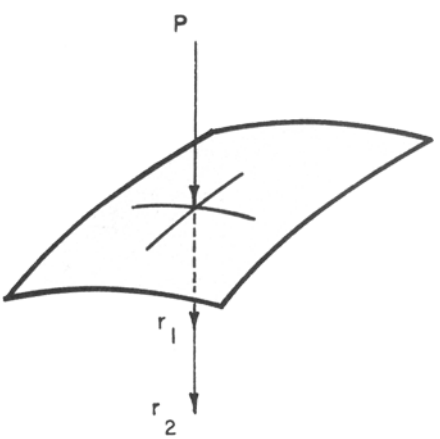


Fig. 8—Illustrating the method of defining the pressure at a point upon a surface, in terms of the surface tension γ and the surface mean curvature at the point.

Simple grains, having small numbers of faces, tend to be convex all over, while complex grains, with large numbers of faces, tend to be concave upon most of their faces, Fig. 9. This happens because grains with fewer than fourteen faces must have a preponderance of convex faces in order to maintain equilibrium angles along their edges and at corners, while the opposite is true of grains having more than fourteen faces. Accordingly, the pressure of surface tension acts upon most faces of simple grains so as to collapse the grain and upon most faces of complex grains so as to enlarge the grain. Under the driving force of surface tension there is, then, a tendency for the larger, more complex, grains to grow at the expense of the smaller, simpler, grains.

Both the total surface area (S_V) and the total surface curvature (M_V) of the grain boundary in one cubic centimeter of specimen can be measured by standard methods of quantitative microscopy.[11]* The total surface curvature is the sum of the mean curvature at every point in the total area of grain boundary in one cubic centimeter of specimen:

$$M_V = \int_{S_V} \tfrac{1}{2}\left(\frac{1}{r_1} + \frac{1}{r_2}\right) dS_V \ (\text{cm}^{-2}) \qquad [7]$$

Like the total surface area (S_V), the total curvature (M_V) diminishes progressively with grain growth, Fig. 10.

In steady-state grain growth the product of the surface area and the surface curvature per grain is found experimentally, Fig. 11, to be a constant (σ):

$$\frac{M_V \cdot S_V}{N_V} = \sigma \quad \text{(unitless)} \qquad [8]$$

This is a necessary consequence of the condition that the grain shapes remain constant during steady-state grain growth. The value of σ is the slope of the graph $M_V \cdot S_V$ vs N_V, Fig. 11. It varies with the distribution of grain forms in the system. In the present aluminum series the spread in grain forms was from 3 to more than 50 faces per grain, Fig. 12, and this distribution was maintained after the beginning of steady-state grain growth. The value of σ was 1.33. If the grains had all been tetrakaidecahedra, with their faces nearly flat, σ would have approached zero, because M_V would have been very small. The value of σ is, thus, a measure of a "structural gradient" in the system.* It will be shown presently that the rate of grain growth is a lineal function of the magnitude of σ.

*$S_V = 2 N_L$, where N_L is the number of intercepts between one cm of test line and grain boundary. $M_V = \pi T_A$, where T_A is the number of tangencies produced between grain boundary and one cm of test line in a sweep through one square cm of two-dimensional section. Where T_A is a total, without reference to the sign of the curvature at each point of tangency, as is the case with the grain boundary measurement, the resulting curvature is absolute, *i.e.*, $\frac{1}{2}(|1/r_1| + |1/r_2|)$. Here the components of curvature $1/r_1$ and $1/r_2$ are added without reference to sign. The pressure acting upon grain boundary is, however, absolute only in the sense: $\frac{1}{2}|(1/r_1 + 1/r_2)|$, where the net curvature at each point on the surface is added without reference to sign. These two kinds of absolute curvature are indistinguishable for either concave or convex surface, but differ for saddle surface. In an equiaxed grain structure the grain faces seem to be largely devoid of saddle curvature, wherefore, the measurement of M_V without reference to sign is acceptable. The same is not necessarily true of non-equiaxed grain types.

*It may be recalled that Zay Jeffries[12] believed that grain growth can occur only in the presence of some kind of gradient, *i.e.*, a temperature gradient, composition gradient, grain size gradient.

TOPOLOGICAL PATH OF GRAIN GROWTH

For the number of grains to decrease, while the relative distribution of grain forms remains unchanged, it is necessary that grains be lost from forms with all numbers of faces in proportion to their frequency in the distribution. If, further, such loss is to occur by the collapse of only simple grains, the process must consist of a progressive loss of faces, culminating in the collapse of any grain that reaches its form of ultimate simplicity. The topological states through which

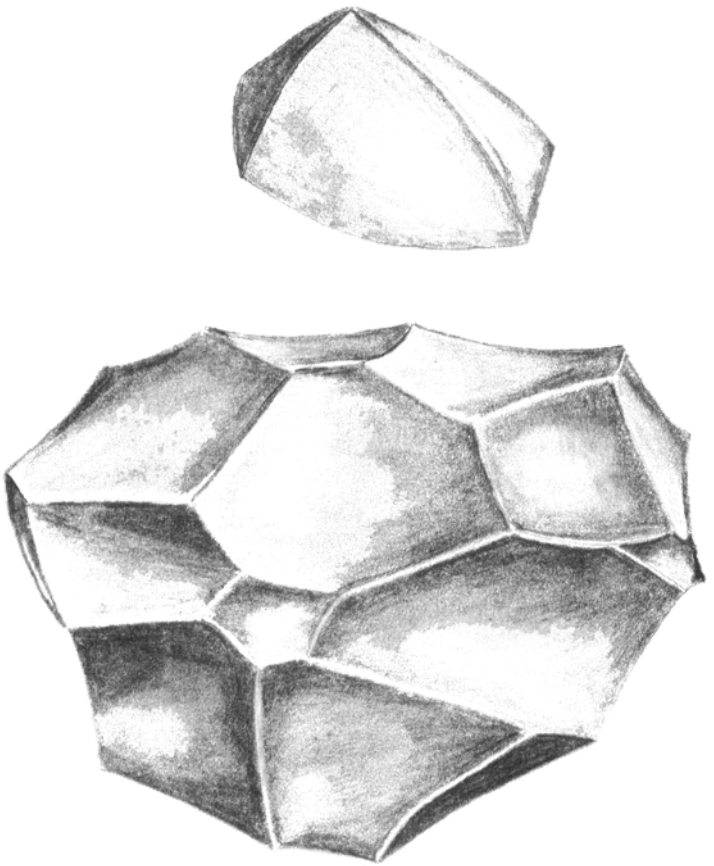


Fig. 9—Typical shapes of simple and complex types of grains. The faces of the simple grains are mostly convex; the faces of the complex grains are mostly concave.

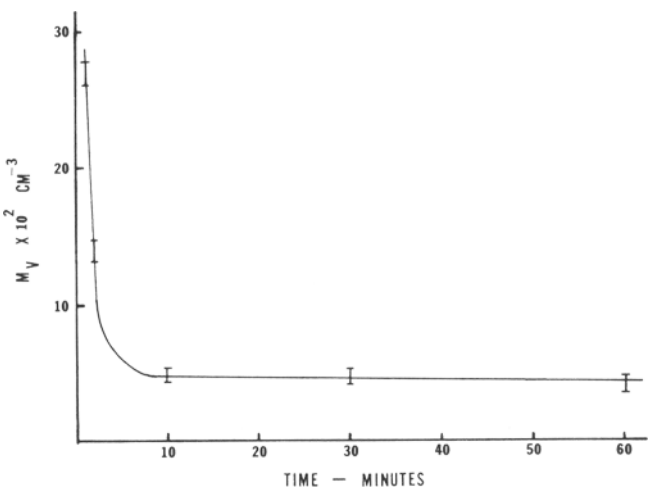


Fig. 10—Total curvature of grain boundary surface M_V as a function of annealing time in minutes at 635°C.

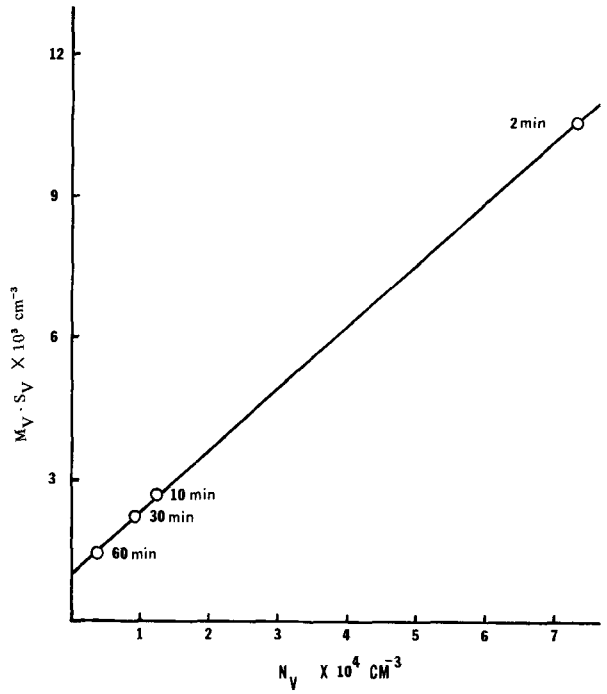


Fig. 11—In steady-state grain growth the product of the total curvature and the total surface per grain ($M_V \cdot S_V/N_V$) is constant with time. Its value is σ.

each grain must pass in such a sequence are itemized, up to 25 faces, in Table I. These states are derived from various forms of Euler's rule, Column *C* corresponding to Eqs. [3], [4], and [5]. It is apparent at once that there exists, under conditions of surface tension equilibrium, only one topological state for any grain with a specified number of faces.

This is not to say that all grains with the same number of faces look alike. The topological features can, in fact, be arranged in various ways to produce different polyhedra with the same number of faces. As an example, the hexahedron may exist in cube-like form with 6 quadrilateral faces and also as a modified prism with 2 triangular faces, 2 quadrilateral faces and 2 pentagonal faces, Fig. 13. If two-edged faces were included, additional modifications of the hexahedron could be constructed. The number of possible variations becomes much greater with the more complex polyhedra. Yet, this diversity has no meaning for grain growth, because all grains having a common number of faces are topologically equivalent.

In reducing a grain of the network to one having one less face, the topological change is always a loss of 1 corner, 1 face and 2 edges, see Table I, Column *C*. This is true regardless of the complexity of the grain, *i.e.*, whether the change is from 2 faces to 1, or from 25 faces to 24. A topological change of this specifica-

Table I. Topology of Grains*

A Grain Form	*B* On the Separate Polyhedron ($C+F-E=2$)			*C* For One Grain in the Net ($C+F-E=1$)			*D* Shared Features per Grain $\left(\frac{C}{4}+\frac{F}{2}-\frac{E}{3}=1\right)$		
	C	*F*	*E*	*C*	*F*	*E*	*C*	*F*	*E*
Monohedron				0	1	0			
Dyohedron				1	2	2			
Triahedron	2	3	3	2	3	4	½	1½	1
Tetrahedron	4	4	6	3	4	6	1	2	2
Pentahedron	6	5	9	4	5	8	1½	2½	3
Hexahedron	8	6	12	5	6	10	2	3	4
Heptahedron	10	7	15	6	7	12	2½	3½	5
Octahedron	12	8	18	7	8	14	3	4	6
Enneahedron	14	9	21	8	9	16	3½	4½	7
Decahedron	16	10	24	9	10	18	4	5	8
Hendecahedron	18	11	27	10	11	20	4½	5½	9
Dodecahedron	20	12	30	11	12	22	5	6	10
Tria-kai-decahedron	22	13	33	12	13	24	5½	6½	11
Tetra-kai-decahedron	24	14	36	13	14	26	6	7	12
Penta-kai-decahedron	26	15	39	14	15	28	6½	7½	13
Hekka-kai-decahedron	26	16	42	15	16	30	7	8	14
Hepta-kai-decahedron	30	17	45	16	17	32	7½	8½	15
Octo-kai-decahedron	32	18	48	17	18	34	8	9	16
Ennea-kai-decahedron	34	19	51	18	19	36	8½	9½	17
Icosihedron	36	20	54	19	20	38	9	10	18
Hen-kai-icosihedron	38	21	57	20	21	40	9½	10½	19
Dyo-kai-icosihedron	40	22	60	21	22	42	10	11	20
Tria-kai-icosihedron	42	23	63	22	23	44	10½	11½	21
Tetra-kai-icosihedron	44	24	66	23	24	46	11	12	22
Penta-kai-icosihedron	46	25	69	24	25	48	11½	12½	23

*Column *B* is derived from the usual form of Euler's rule connecting the numbers of corners, faces and edges of an isolated polyhedron. An additional constraint has been imposed to fulfill the condition that the polyhedron must be constructed exclusively of edges and corners that would appear in the network as triple lines and quadruple points, *i.e.*, $C/2 = E/3$.

Column *C* is derived directly from Eq. [3], setting the number of grains (N) equal to unity and with the added constraint of Eq. [4].

Column *D* is derived from Column *B*, by dividing each feature by the number of neighboring grains among which it is shared in the network.

The monohedron and the dyohedron both are capable of existence in a space network, although neither can be classified among the isolated polyhedra.

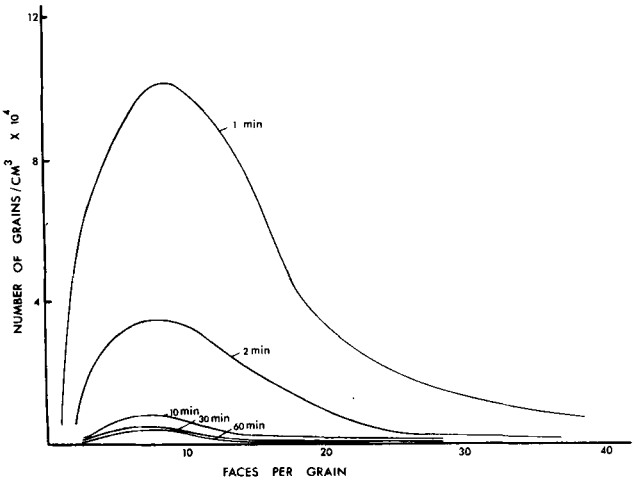
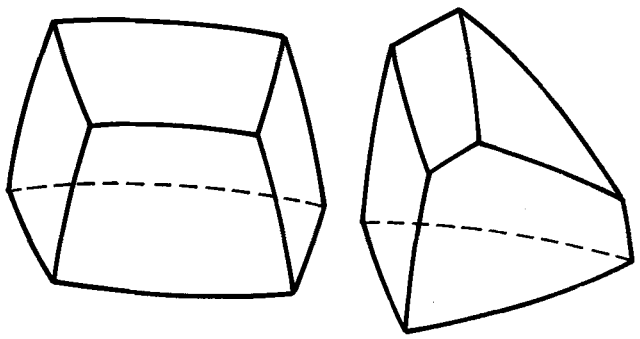


Fig. 12—Distribution of grain forms according to number of faces is unaffected by the annealing time in the course of steady-state grain growth.

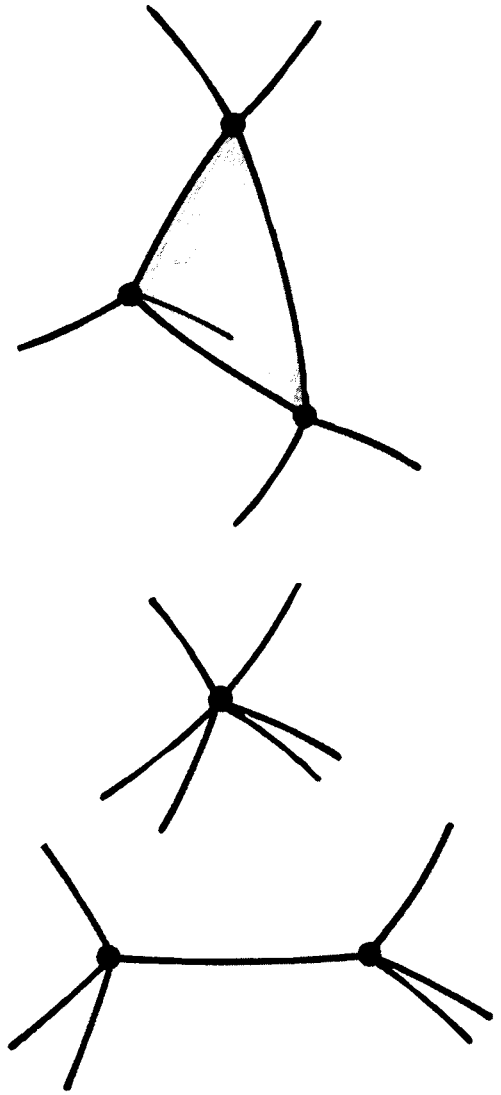


Parameters for both	C	F	E
The polyhedron	8	6	12
In the network	5	6	10
Shared features	2	3	4

Fig. 13—Two kinds of hexahedra, but the topological parameters are identical.

tion represents the loss of *one triangular face*, as is illustrated in Fig. 14. Represented in this sketch is a triangular face between two grains and sharing edges with three other grains. As the three corners of the triangle come together, there exists, momentarily, a five-grain junction with six triple lines meeting at a point. Immediately, the two original neighbors separate, permitting the other three grains to meet along a new triple line that joins two new quadruple points. Destroyed in this exchange are the triangular face, its three edges and its three corners. Created are one new edge and two new corners, resulting in a net loss of: 1 corner, 1 face and 2 edges. The same result can be obtained through other sequences of structural transformation, wherein a triangular face alone is lost. Thus the degrading of complex to simple grains is seen to occur by the loss, one at a time, of triangular faces.

At the end of the degradation sequence lies one of the simple grain forms, the monohedron, the dyohedron, the triahedron, or the tetrahedron, Fig. 15, any one of which might be capable of direct collapse to zero volume. The simplest grain that has been found experimentally in the aluminum grain growth speci-



In the network	C	F	E
Before	3	1	3
After	2	0	1
Lost	1	1	2

Fig. 14—As a pair of neighboring grains moves apart, their mutual triangular face (top sketch) closes (center sketch) to produce a new triple line joining two new corners (bottom sketch). This occurs with a net loss of one corner, one face and two edges.

mens is the triahedron. This polyhedron could collapse by its two corners coming together, whereupon the system loses: 1 grain, 2 corners, 3 faces and 4 edges.* Fig. 16 and Column *C* of Table I. If it be argued that collapse of the triahedron occurs in steps, by first forming a dyohedron and then a monohedron, which finally shrinks to zero volume, the net topological change is found to be the same.

*The loss of the fourth edge is occasioned by the fact that the two triple lines, that had attached the ends of the triahedron to the network, are joined to become one triple line when the triahedral grain is gone.

Since the average number of corners, faces and edges per grain remains in the ratio 6:7:12, Fig. 6, it follows that the elimination of grains, in grain

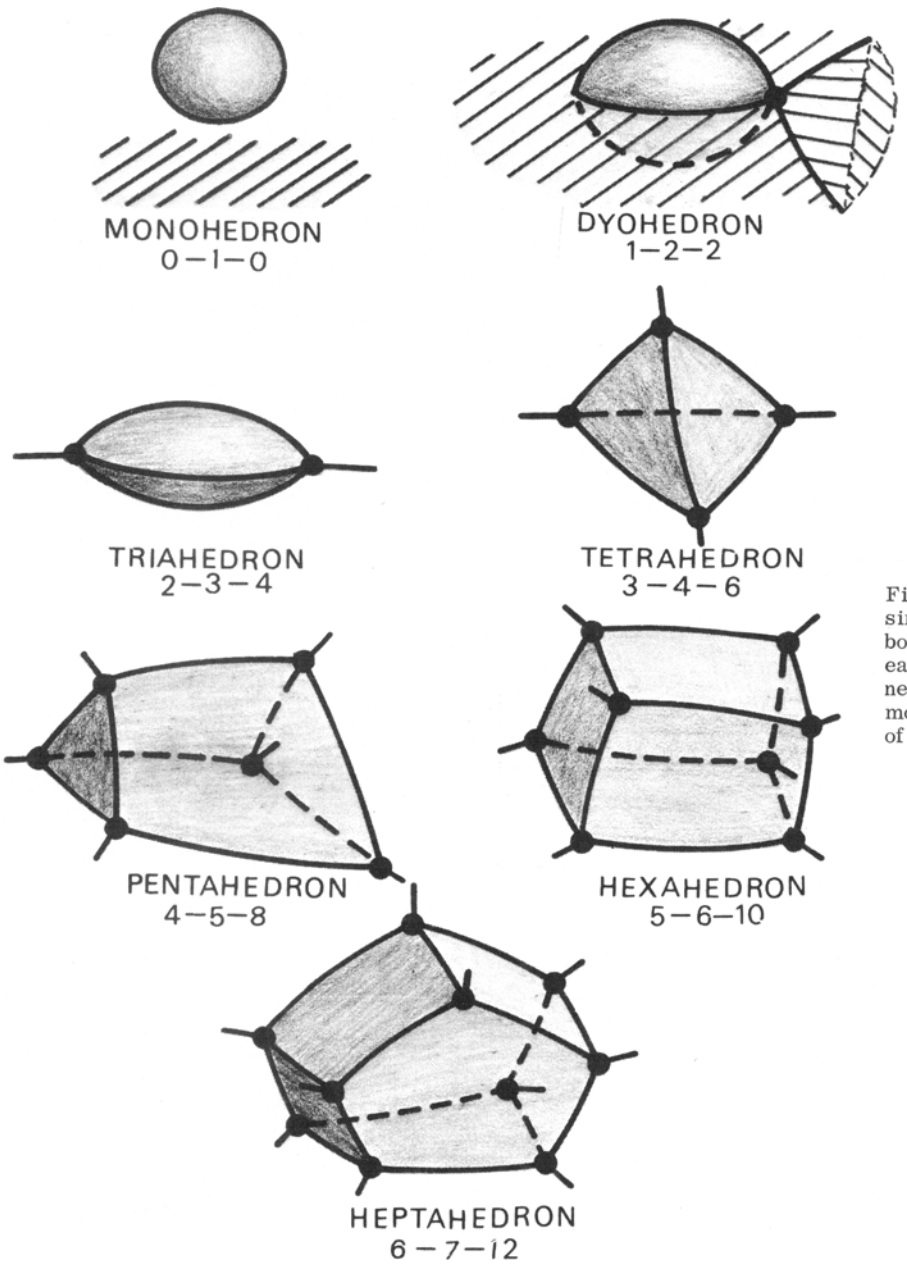


Fig. 15—Topological forms of the seven simplest grains, as they occur in the grain boundary network. The numbers below each sketch denote respectively the corners, faces and edges that would be removed from the network by the collapse of the grain.

growth, must occur by the removal of corners, faces and edges in exactly this ratio. This would be accomplished by collapsing 6 triangular faces and 1 monohedron, or 5 triangular faces and 1 dyohedron, or 4 triangular faces and 1 triahedron, or 3 triangular faces and 1 tetrahedron and so on up to 1 heptahedron, for each grain lost. In terms of shared features per grain, Column *D* of Table I, any of these processes is equivalent to removing a tetrakaidecahedral grain without changing the average topology of its neighbors.

It may appear remarkable that the system could go on losing simple grains and triangular faces without running out of them. The reason lies in an inexhaustible replenishment of these topological units. The removal of one monohedral grain and six triangular faces takes a total of 13 faces away from grains that remain in the system.* This is precisely the number required to reduce one average tetrakaidecahedral grain to a monohedron. The same argument applies to grains of increasing numbers of faces, up to the heptahedron. Thus, the collapse of a simple grain serves, on the average, to provide a new grain of the same kind.

The removal of one triangular face carries away one edge from each of six faces that remain in the system and gives new edge to three other faces, taking away a difference of three edges. The collapse of six triangular faces with one monohedron (a total of 7

*Each triangular face of the network belongs to two grains and thus removes faces from two grains when it collapses. The surface of the monohedron also belongs to two grains, but only one of these grains remains in the system, after its collapse. Thus: 6 X 2 + 1 = 13.

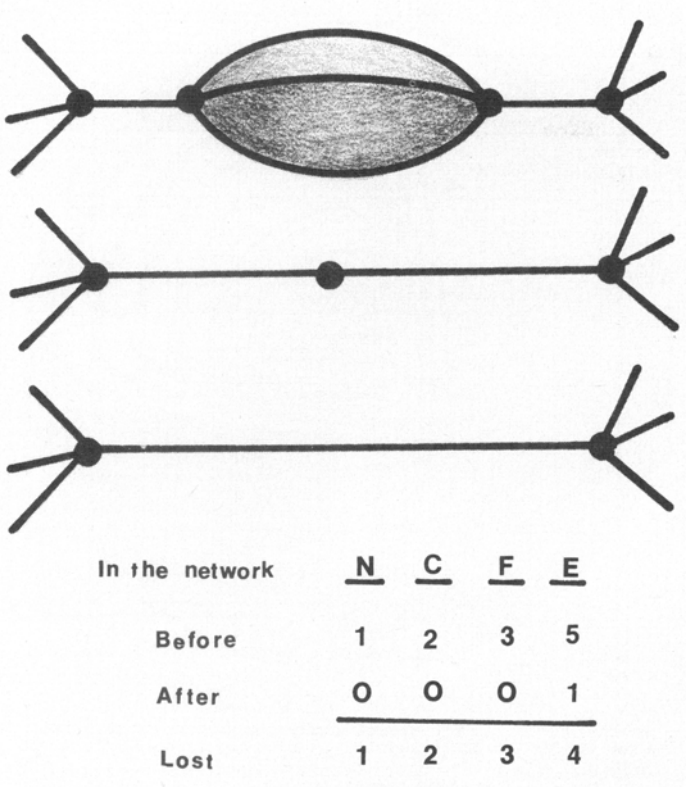


Fig. 16—A triahedral grain (top sketch) collapses (center sketch) leaving one triple line (bottom sketch). This occurs with a loss of one grain, two corners, three faces and four edges.

faces) removes 18 edges from faces that remain in the system. Since the average face of an isolated tetrakaidecahedral grain has 36/7 edges,* it is apparent that the removal of 18 edges from the system is just enough to reduce seven average faces to six triangular faces and one edgeless face, thus replacing the six collapsed triangular faces and the edgeless face of the monohedron.

*Each edge is shared by two faces of the tetrakaidecahedron. Hence, the 36 edges of the separate tetrakaidecahedron, Column *B* of Table I, are divided by 7, that is half the number of faces of the tetrakaidecahedron, in order to obtain the average number of edges per face.

The sharing of a triangular face between two grains occurs mostly when the centers of the grain pair are nearing maximum spacing without parting. Continued separation of the grain centers, with the loss of the triangular face, is a natural consequence of grain growth. The grains that constitute the separating pair are most likely to consist of one grain with fewer than 14 faces and one with more than 14 faces (or both with 14 faces), because the convexity of face of the simpler grains must be mated to the concavity of face of the more complex grains. For this reason, grain complexity does not enter into the probability of the loss of a triangular face. This probability is equal for all grains. As a result, faces are lost from each topological form of grain in proportion to its frequency in the distribution. This maintains a constant distribution of topological grain forms, fulfilling the requirement of steady-state grain growth and accounting for the constant value of σ.

From the foregoing it is amply apparent that extensive migration of the grain boundary is required for grain growth to proceed. The great vigor and frequency with which grain boundaries sweep back and forth during grain growth has been documented in motion picture studies upon grain growth in γ-iron by Grube and Rouze.[13]

THE TOPOLOGICAL TRANSFORMATION

The topological changes that constitute grain growth are immune to being driven by any direct force, because they are independent of any Euclidean dimension of the structure. Physical force can act directly to change a volume, an area, or a length, but it cannot change a number. Topological transformations simply happen when the Euclidean dimensions of some part of the system chance to pass through zero. This occurs when a grain goes to zero volume and when a triangular face goes to zero area, or is created. Such events can occur only as grain boundary sweeps through the system.

It can be deduced that the volume swept by the grain boundary is always the same for the disappearance of one average grain and the readjustment of the remaining grains to equiaxedness, irrespective of the size of the average grain. This principle is most readily understood by reference to a one-dimensional analog. Consider a thin rod divided along its length into "grains" of equal length, Fig. 17. Let one grain be removed by sweeping its "boundary" to the end of the rod and readjust the remaining grains to equal size. In this process, it is evident that boundary has swept through half the length of the rod. The same is true whether the rod goes from three grains to two, from four to three, or from any number to one less. A three-dimensional model can be constructed by placing grains in concentric shells, Fig. 18. When the center grain is collapsed, the adjustment of the remaining shells to equal spacing (approximately equivalent to equiaxing the grains) causes the shell boundaries to sweep a total of one quarter of the total volume of the sphere. The next such event centers upon a collapsing grain located randomly elsewhere in the system, thus maintaining anisotropy of grain shape. This result can be obtained graphically, or it can be derived analytically, as has been done by R. T. DeHoff in an Appendix to this paper.

Although models of this kind possess a degree of artificiality, it is clear from them that a fixed relationship obtains between the number of grains eliminated and the volume swept by grain boundaries, in maintaining a surface tension equilibrium structural state. This geometric constant provides a link between the Euclidean dimensional change and the topological change in number. In so doing, it opens the way for a direct analysis of the kinetics of the topological transformation that constitutes steady-state grain growth. The "sweep-constant Θ" shall be defined as the number of grains lost when the grain boundary sweeps through one cubic centimeter of the material. Its units are number, which is to say that the sweep constant is dimensionless. Upon the basis of the spherical model it will be estimated that the value of Θ is 4.

RATE OF GRAIN GROWTH

The grain size having been defined as $1/N_V$, grain growth becomes the time rate of increase in $1/N_V$.

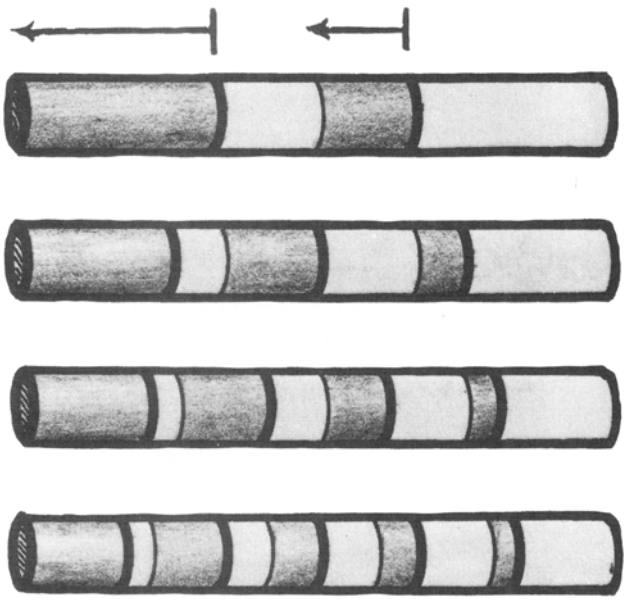


Fig. 17—A one-dimensional model of grain growth, showing that boundary sweeps through half the length (gray zones) when a "grain" is collapsed and the remaining "grains" are adjusted to equal volume. The boundaries before growth are represented as heavy lines, after growth as light lines.

To determine the rate of grain growth it is necessary only to know how many grains are lost in unit time. Making use of the sweep constant Θ, this information can be derived from the rate of sweep of the grain boundary under the force of surface tension.

The average pressure (P) exerted upon a point on the grain boundary is the product of the surface tension (γ) and the average mean curvature (M_V/S_V), which latter is the total curvature (M_V) in one cubic centimeter of material divided by the total area of grain boundary (S_V) in one cubic centimeter. Thus:

$$P = \gamma M_V/S_V \quad (\text{dynes/cm}^2) \qquad [9]$$

The total force (F) is the product of the pressure (P) and the surface area (S_V) in one cubic centimeter of material:

$$F = \gamma M_V \quad (\text{dynes/cm}^3) \qquad [10]$$

If the mobility of the boundary be defined as μ, the number of centimeters of migration of the boundary in one second, under a force of one dyne, the number of sweeps made by all of the grain boundary through one cubic centimeter of the material in one second will be the product of the mobility (μ), the force (F) and the surface area (S_V):

$$\mu\gamma M_V S_V (\text{cm}^{-3}\ \text{sec}^{-1}) \qquad [11]$$

The number of grains lost per cubic centimeter in one second then becomes:

$$\Theta\mu\gamma M_V S_V \quad (\text{cm}^{-3}\ \text{sec}^{-1}) \qquad [12]$$

The total volume transferred from the lost grains to the remaining grains is the product of the number of grains lost per cubic centimeter and the volume of the average grain ($1/N_V$):

$$\Theta\mu\gamma M_V S_V/N_V \quad (\text{sec}^{-1}) \qquad [13]$$

The average growth per grain is obtained by dividing the total volume transferred by the number of grains in one cubic centimeter of the material:

$$\Theta\mu\gamma M_V S_V/N_V^2 \quad (\text{cm}^3/\text{s}) \qquad [14]$$

Substituting the structural gradient (σ) from Eq. [8]:

$$\Theta\mu\gamma\sigma/N_V \quad (\text{cm}^3/\text{s}) \qquad [15]$$

This is the volume increase per grain per second. In time (t) the volume increase per grain is:

$$\Theta\mu\gamma\sigma t/N_V \quad (\text{cm}^3) \qquad [16]$$

The average grain volume at time (t) is the average grain volume at time zero plus the volume increase of the initial average grain in time (t):

$$1/(N_V)_t = 1/(N_V)_0\{1 + \Theta\mu\gamma\sigma t\} \quad (\text{cm}^3) \qquad [17]$$

Steady-state grain growth, expressed as the increase in the volume of the average grain, is thus expected to be proportional directly with the time, as is found experimentally, Fig. 19.

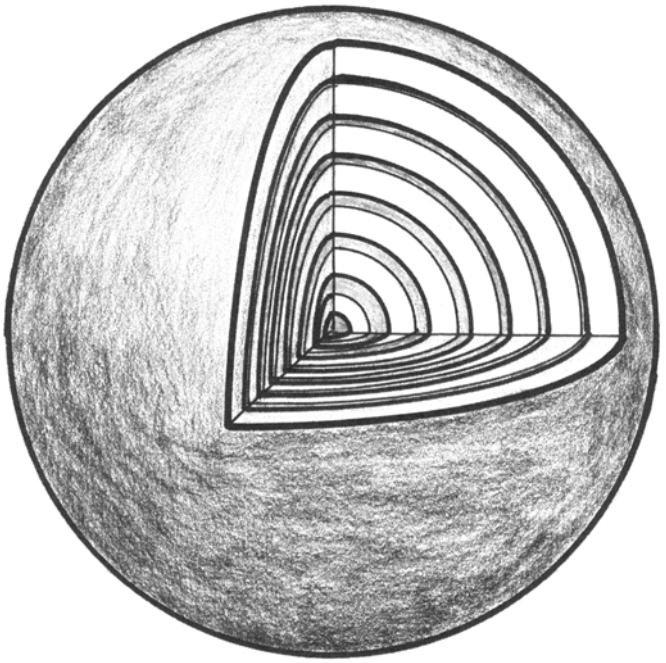


Fig. 18—A three-dimensional model of grain growth, showing concentric shells of boundary sweeping through one quarter of the volume, when the central grain is collapsed and the shells are adjusted to equal spacing. The total sweep fraction is evidently independent of the number of shells. The next collapse occurs randomly elsewhere in the system, thus, maintaining equiaxedness of the grain structure.

DISCUSSION

It has not been required, in developing the foregoing rate law, to introduce any arbitrarily adjustable parameter, as has been done in the usual expression[14] of grain growth kinetics. Each factor in the expression is fully defined and growth is described uniquely, using only fundamental properties of the material. This has resulted from applying a topological analysis to a topological problem, instead of using the usual Euclidean approach, in which adjustment factors are introduced in an effort to compensate for the omission of topological considerations.

The grain growth observations that exist presently

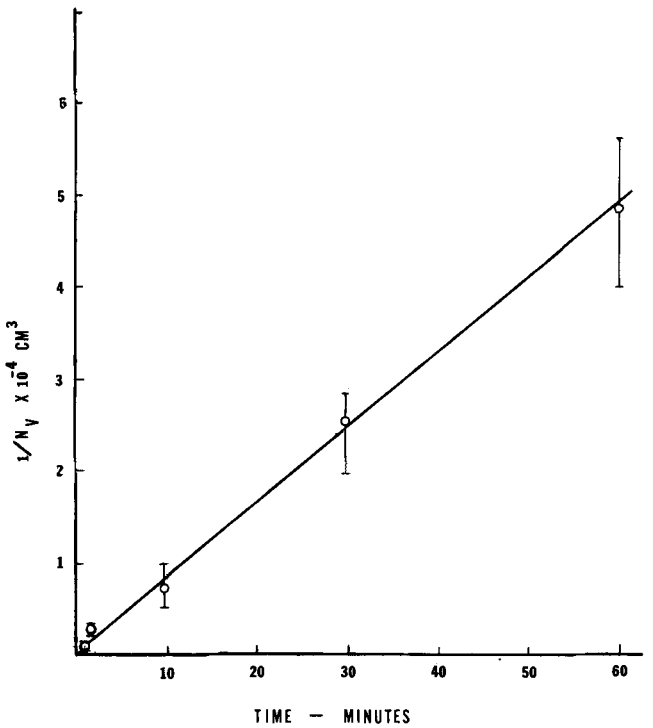


Fig. 19—Grain volume $1/N_V$ is found to be a linear function of the time of isothermal annealing of the aluminum. The larger range of uncertainty at long time results from the greater difficulty in sampling the coarser grained material.

in the literature are unavailable for comparison with the present result, because none can be translated into grain volume growth. A reverse comparison is possible, however, because mean intercept measurements have been made upon the specimens subjected to serial section analysis. The mean intercept readings give a value for the exponent (n) in the expression $D = kt^n$ of 0.43, which is almost identical with the exponent reported by Beck *et al.*[14] for pure aluminum. This correspondence is understood to mean that the material that has been evaluated topologically, in the present study, is typical of materials that have provided the basis for prior expressions of grain growth kinetics.

If a true average grain diameter is computed from the serial section measurements and is used to evaluate (n) in the expression $D = kt^n$, a value close to 0.33 is obtained. This is consistent with proportionality of grain volume to the time. The difference between this and the mean intercept result is believed to arise from the different sensitivity of the two measurements to surface area. The mean intercept, being the inverse of grain boundary area, is highly sensitive to changes in surface area. The grain volume, being independent of surface area, notices only changes in topology, *i.e.*, the number of grains, corners, faces and edges. The establishment of a steady-state with respect to the topological parameters appears to occur long before a really steady state with respect to grain shape has been achieved. The larger value of (n) resulting from the mean intercept measurements probably includes some area decrease resulting from a continued approach to equiaxedness, as well as area decrease associated with the enlargement of the grains *per se*. It has also been noticed, in the present experimental studies, that the mean intercept values display much greater sensitivity to proximity to external surface than do the topological measurements. A distortion of the surface and near surface grains away from equiaxed shape is, of course, to be anticipated.

The discovery of the structural gradient (σ) and of means for its evaluation opens a promising avenue for future investigation and development. This factor, which is so important in establishing the growth potential of the grain structure, doubtless derives its magnitude from the manner in which the initial grain boundary network is created. If all grain centers were to occur upon a pattern of three-dimensional equally spaced points, the system would be made up largely of tetrakaidecahedral grains, of nearly uniform size, with nearly flat faces and with σ approaching zero, because the absolute curvature (M_V) would be very small. Such a dispersion might result from the growth of a system of grains from simple polygonization. The small value of σ would be consistent with the commonly observed stability of sub-grain structures. Cold work, followed by recrystallization, is likely to give an inhomogeneous distribution of grain centers, resulting in a higher value of σ. It may be conjectured that σ would maximize for light deformations, where the inhomogeneity is expected to be greatest and the rate of grain coarsening is commonly large. Thus, the evaluation of σ provides a tool by which the grain form distribution can be studied.

The product $\mu\gamma$ can also be evaluated from the serial section and quantitative microscopy measurements. In the present case, assuming $\Theta = 4$, this product is found to be approximately 2.6×10^{-3} sec^{-1}. Where γ is known, the value of μ is immediately accessible through the metallographic readings. In the case of aluminum, the grain boundary energy has not been measured, but there are determinations of the external surface tension, ranging from 500 to 900 dynes per cm. Upon this basis, it appears that the grain boundary surface tension should be of the order of a few hundred dynes, probably not greater than 500 dynes per cm. If the figure 500 be accepted, the value of the mobility (μ) becomes 5×10^{-6} cm/dyne s.

There are again no values of the mobility in the literature that can be compared with this estimate, because all recorded measurements involve boundaries in direct contact with external surface, as well as other complications. Mobility in the semi-infinite network may be regarded as a fundamental property of the material, because it is relatively uninfluenced by external conditions. Its determination is, therefore, desirable. This may be accomplished, in cases where γ is not already known, by newly measuring γ through the use of the sintering technique,[15] combined with the method of grain boundary grooving.[7]

All of the foregoing has dealt with the behavior of a semi-infinite network of a single kind of interface. Where, through the presence of another phase, a second kind of interface is present, it becomes a part of the network and the properties of the network are modified accordingly. In most cases the added interface is relatively immobile. Topological processes involving such interface are then limited to the traverse of triple lines across the stationary surface, the extinction of grains at the fixed surface and the attachement and detachment of grain boundary from contact with the second phase. All such processes involve increased energies and are likely to be relatively slow. This de-

ters the progress of grain growth and constitutes what has been called "pinning." Obviously, this subject could be approached by the topological route with definite advantage. It is, however, a much larger subject than the one presently undertaken.

Finally, it should be observed that many of the features of microstructure, in general, have the characteristics of a topological continuum. Matters dealing with their measurement and with the kinetics of their change should be made both easier and more exact by analyzing them through their topological properties.

SUMMARY

1) Recognizing grain growth as a topological process, grain size is defined as the average grain volume, obtained by taking the reciprocal of the number of grains in unit volume $1/N_V$.

2) It has been found experimentally that all grain corners are quadruple points, that all grain edges are triple lines and that, during steady-state grain growth, the average grain is topologically a tetrakaidecahedron, with corners, faces and edges in the ratio 6:7:12.

3) The susceptibility of a specific material to undergo steady-state grain growth depends upon a structural gradient (σ) which is experimentally constant and which is equal to the product of the total curvature and the total surface area per grain: $\sigma = M_V \cdot S_V / N_V$.

4) The number of grains eliminated when the grain boundary sweeps through unit volume of the material is a constant Θ, independent of the size of the grains, and the identity of the material.

5) It has been demonstrated that the structural changes required to implement steady-state grain growth, *i.e.*, magnification without net shape change, can occur as a logical sequence of topological transformations in the grain boundary network.

6) The average grain volume ($1/N_V$) increases linearly with time. The rate of increase in the average grain volume is proportional directly to the mobility (μ), the grain boundary surface tension (γ) and the structural gradient (σ) of the grain boundary network.

ACKNOWLEDGMENTS

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APPENDIX

Volume Swept Out By Grain Boundaries Due To The Annihilation Of The Average Grain; Spherical Shell Model

ROBERT T. DeHOFF

I. For the i-th shell, before the grain annihilates,

$$r_i \text{ (before)} = i\left(\frac{R_0}{n}\right) \qquad [1]$$

where R_0 is the radius of the sample.

A) The location of this same shell after annihilation is

$$r_i \text{ (after)} = (i-1)\left(\frac{R_0}{n-1}\right) \qquad [2]$$

because it is now the $(i-1)$-st shell of $(n-1)$.

II. The volume it sweeps in going from r_i (before) to r_i (after) is

$$\begin{aligned}\Delta V_i &= \tfrac{4}{3}\pi [r_i \text{ (before)}]^3 - \tfrac{4}{3}\pi [r_i \text{ (after)}]^3 \\ &= \tfrac{4}{3}\pi \left[i\,\frac{R_0}{n}\right]^3 - \tfrac{4}{3}\pi \left[\frac{(i-1)R_0}{(n-1)}\right]^3 \\ &= \tfrac{4}{3}\pi R_0^3 \left[\left(\frac{i}{n}\right)^3 - \left(\frac{i-1}{n-1}\right)^3\right] \end{aligned} \qquad [3]$$

A) The fraction of the total volume swept by the i-th shell is

$$\Delta V_{V_i} = \frac{\Delta V_i}{\frac{4}{3}\pi R_0^3} = \left\{\left(\frac{i}{n}\right)^3 - \left[\frac{(i-1)}{(n-1)}\right]^3\right\} \qquad [4]$$

III. The total volume fraction swept by the motion of all $(n-1)$ spheres (the n-th, or outside, sphere does not move) is

$$\Delta V_i = \sum_{i=1}^{n-1} \Delta V_{V_i} = \sum_{i=1}^{n-1} \left[\left(\frac{i}{n}\right)^3 - \left(\frac{i-1}{n-1}\right)^3\right] \qquad [5]$$

A) The rest is simply the evaluation of these sums.

IV. Introduce a new index:

$$j = (i-1) \qquad [6]$$

or

$$i = j + 1$$

A) Then the second term in Eq. [5] can be written:

$$\sum_{i=1}^{n-1} \left(\frac{i-1}{n-1}\right)^3 = \sum_{j=0}^{n-2} \frac{j^3}{(n-1)^3} \qquad [7]$$

B) To make the sums run over the same limits, the first term may be written:

$$\sum_{i=1}^{n-1} \frac{i^3}{n^3} = \sum_{i=1}^{n-2} \frac{i^3}{n^3} + \frac{(n-1)^3}{n^3} \qquad [8]$$

(Simply writing the last term explicitly).

C) Since, for $j = 0$, the quantity $j^3/(n-1)^3 = 0$, Eq. [7] may be written:

$$\sum_{i=1}^{n-1} \left(\frac{i-1}{n-1}\right)^3 = \sum_{j=1}^{n-2} \frac{j^3}{(n-1)^3} \qquad [9]$$

D) Put Eqs. [8] and [9] back into [5] to get:

$$\Delta V_V = \frac{(n-1)^3}{n^3} - \sum_{i=1}^{n-2} \frac{i^3}{n^3} - \sum_{j=1}^{n-2} \frac{j^3}{(n-1)^3}$$

1) Since $\sum_{i=1}^{n-2} i^3 \equiv \sum_{j=1}^{n-2} j^3$ [what the index symbol is does not affect the sum.]

$$\Delta V_V = \frac{(n-1)^3}{n^3} + \sum_{i=1}^{n=2} i^3 \left[\frac{1}{n^3} - \frac{1}{(n-1)^3}\right] \qquad [10]$$

2) Since it can be shown that:

$$\sum_{i=1}^{m} i^3 = \left[\tfrac{1}{2}m(m+1)\right]^2 \qquad [11]$$

a) In the present example, the upper index on the sum is:

$$m = n - 2$$

(compare [11] to [10]).

3) Thus:

$$\Delta V_V = \frac{(n-1)^3}{n^3} + \left\{\tfrac{1}{4}(n-2)^2(n-1)^2\right\}\left\{\frac{1}{n^3} - \frac{1}{(n-1)^3}\right\} \quad [12]$$

Now everything is in terms of n, the number of spheres in the system before annihilation.

E) Eq. [12] can be simplified by putting everything over a least common denominator, $4n^3(n-1)$. Then:

$$\Delta V_V = \frac{1}{4n^3(n-1)}\left\{4(n-1)^4 + (n-2)^2[(n-1)^3 - n^3]\right\} \quad [13]$$

1) Expand everything to see what cancels:

$$\Delta V_V = \frac{1}{4n^3(n-1)}\left\{4[n^4 - 4n^3 + 6n^2 - 4n + 1] + [n^2 - 4n + 4][n^3 - 3n^2 + 3n - 1 - n^3]\right\}$$

$$\Delta V_V = \frac{1}{4}\left[1 - \frac{1}{(n^2 - n)}\right] \quad [14]$$

V. Tabulated Values:

n	ΔV_V	n	ΔV_V	n	ΔV_V
2	0.1250	8	0.2455	18	0.2492
3	0.2082	9	0.2465	20	0.2493
4	0.2291	10	0.2472	25	0.2496
5	0.2375	12	0.2481	30	0.2497
6	0.2416	14	0.2486	40	0.2498
7	0.2446	16	0.2489	50	0.2499

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