# RANDOM SUBDIVISIONS OF SPACE INTO CRYSTALS

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**1. Introduction.** This paper considers random subdivisions of a $D$-dimensional Euclidean space into disjoint regions. The regions will be called *crystals* because they represent individual crystal grains in a metal or mineral ($D = 3$) in one application. However, other interpretations are possible. The "crystals" may also be cells in living tissue, bubbles in a froth, and (when $D$ is large) detection regions of a code (see Smith [16], [19], Shannon [15]).

Section 3 describes random processes which have been used to subdivide space into crystals. Given such a process statistical properties of the crystals present interesting problems in geometric probability. For crystals in two different mineral models Meijering [13] derived the mean surface area, mean number of faces, mean total edge length and many other related mean values. Continuing with the same two models, the present paper concerns itself with the distribution of crystal volumes. Section 4 finds the variance of the volume of a crystal and also finds some variances associated with plane or line sections through crystals. Curiously these two models have similar values for many statistical parameters but have very different volume variances. Section 5 gives bounds on the distribution functions for crystal volumes. Section 2 and 3 review related probabilistic results from the literature on mineralogy and metallurgy.

**2. Geometrical formulas.** Basic geometrical facts imply some properties of crystals even before the random process which subdivides space into crystals is specified in detail. For solids composed of several different kinds of crystals, Delesse [6] in 1848 and Rosiwal [14] in 1898 produced unbiased estimates of the volume fraction occupied by the $i$th kind of crystal. These estimates are simply the fractions of area or length which such crystals intersect on a plane or line through the solid. Chayes [3] gives statistical details about these estimates. Even for polycrystalline solids containing a single crystalline phase (the only solids to be considered here), plane or line intersections are informative. A line segment through the solid is a kind of Buffon needle. The expected number of times that a unit length of this "needle" intersects crystal interfaces is related to the mean total interface area per unit volume in the solid (see Smith and Guttman [17], Corrsin [4]:

$$E(\text{intersections in unit length} = \tfrac{1}{2}E(\text{interface area in unit volume}). \tag{1}$$

Another one of several relationships like (1), given by Smith and Guttman, concerns edges where three crystals meet. On a plane section through a solid, edges appear as corners of the plane section through individual crystals. The expected

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number of corners per unit area of the plane is related to the mean total length of edges per unit volume of solid by

$$(2)\qquad E(\text{corners in unit area}) = \tfrac{1}{2}E(\text{edge length in unit volume}).$$

Cauchy's formulas for convex bodies (see Section 30 of Blaschke [2]) provide some other results about crystals. One of Cauchy's formulas relates the perimeter of a plane convex body $C$ to the length $D(\theta)$ of the normal projection of $C$ onto a line of inclination $\theta$:

$$\text{perimeter} = \int_0^\pi D(\theta)\, d\theta.$$

Applied to crystals in a two-dimensional subdivision (e.g., crystal sections in a plane through a three dimensional solid) the formula yields

$$(3)\qquad E(\text{perimeter}) \geqq \pi E(\text{length of projection}).$$

Equality holds in (3) if crystals are convex. A three-dimensional Cauchy formula relates the surface area of a convex body $C$ to an integral of the area of the projection of $C$ onto a variable plane. A result analogous to (3) is

$$(4)\qquad E(\text{surface}) \geqq 4E(\text{area of projection})$$

with equality if crystals are convex. Formulas (3) and (4) may be of interest in the theory of small particles (i.e., powder, sand, etc.) since projections are commonly used in estimating particle sizes (see Herden [8]).

**3. Models.** To estimate other parameters it seems necessary to postulate special models. A simple kind of model uses congruent crystals fitted together to fill space. A popular 3-dimensional shape is Lord Kelvin's tetrakaidecahedron (a truncated octahedron with 6 square faces, 8 hexagonal faces, and 24 vertices; see Thompson [20]). Among space-filling solids of given volume, the tetrakaidecahedron has a small surface area. It was once supposed to approximate the shape of soap bubbles in a froth and other kinds of crystals which minimize surface area. However, tetrakaidecahedra almost never appear as bubbles in froth, cells in living tissue, or crystal grains in a solid (see Lewis [11], Smith [16]). A related approach has been to use formulas which apply strictly only to regular tessellations and to assume suitable additional relations (for example, four crystals meet at each vertex and three crystals meet at each edge). Although the corresponding regular tessellations do not exist, Smith [18] obtains the figures 13.40 faces, 22.80 vertices, and Coxeter [5] obtains 13.56 faces, 23.13 vertices by different assumptions.

Other models introduce some randomness. One, more suitable for experimentation than for analysis, uses deformable spheres randomly packed together and squeezed into polyhedral shapes. Marvin [12] measured an average of 14.16 faces on compressed lead shot. Bernal [1] used balls of modelling clay and obtained 13.3 faces. Both authors observed a large number of pentagonal faces.

The next two models are the ones of main interest here. They follow from simple assumptions about the origin and growth of crystals in solids. In particular, the volumes of the crystals are not assumed to be identical. The crystal volume is a random variable to be studied in Sections 4 and 5.

The following random construction produces what Meijering calls the cell model. First pick points $P_1, P_2, \cdots$ in space at random by a Poisson process of density $\rho$ points per unit volume. Next subdivide space into cells (crystals) $C_1, C_2, \cdots$ by the rule: $C_i$ contains all points in space closer to $P_i$ than to any $P_j$ $(j \neq i)$. In the cell model $C_i$ is a convex polyhedron because it is the intersection of several half-spaces (points closer to $P_i$ than to $P_j$ form a half-space). $P_i$ will be called the *center* of $C_i$. Figure 1 shows a cell model with $D = 2$.

In mineralogy $P_i$ represents the location of the original nucleus or seed crystal from which $C_i$ grew. One assumes

(i) the seeds for all crystals start growing at the same instant;

(ii) seeds grow at the same rate in all directions (i.e., as spheres); and

(iii) seeds stay fixed in space without pushing apart as they grow into contact.

Shannon [15] constructs random codes in a similar way. The centers $P_i$ now form an alphabet of possible transmitted signals chosen at random from a suitable function space. Cell $C_i$ contains all received signals which will be decoded as $P_i$.

Johnson and Mehl [10] gave a more complicated random subdivision which may be a better model of a mineral since it replaces assumption (i) by

(i′) Starting at an initial time $t = 0$, seeds appear at a constant rate, $\alpha$ seeds per second per unit volume.

The Johnson-Mehl model uses a Poisson process of density $\alpha$ in a $D + 1$ dimensional space of *arrivals*. Each arrival point $A_i$ has the form $(P_i, t_i)$ where $P_i$ is the position and $t_i$ the (positive) arrival time of a seed. As in (ii), $A_i$ would grow a crystallized sphere of radius $v(t - t_i)$ at time $t$ if other crystals were absent. However, other crystals interfere with the one from $A_i$. $A_i$ ultimately produces a cell $C_i$ containing all points $P$ such that the sphere growing from $A_i$ is the first one to reach $P$; i.e.,

$$t_i + (r_i/v) < t_j + (r_j/v), \qquad j \neq i, \tag{5}$$

where $r_i$ and $r_j$ are distances from $P$ to $P_i$ and $P_j$. If, for some $j$, the distance between $P_i$ and $P_j$ is less than $v(t_i - t_j)$, then (5) holds for no point $P$. Thus a cell may be empty. Only the nonempty cells are to be considered crystals.

Figure 2 shows a Johnson-Mehl model with $D = 2$. Figures 1 and 2 resemble some actual metal crystallizations (for examples see the photographs in Wells [21], plate II). Evans [7] also interprets the two-dimensional Johnson-Mehl model in terms of circular waves spreading from raindrops which fall at random into a puddle.

In the Johnson-Mehl model, crystals do not have plane faces and are not necessarily convex. However, they are star-shaped; i.e., if $P$ belongs to a crystal $C_i$ with arrival $A_i = (P_i, t_i)$, then the entire line segment $PP_i$ lies in $C_i$. Crystal faces need not be simply connected. Indeed, the common boundary between two crystals may even be disconnected (in that case the different connected parts

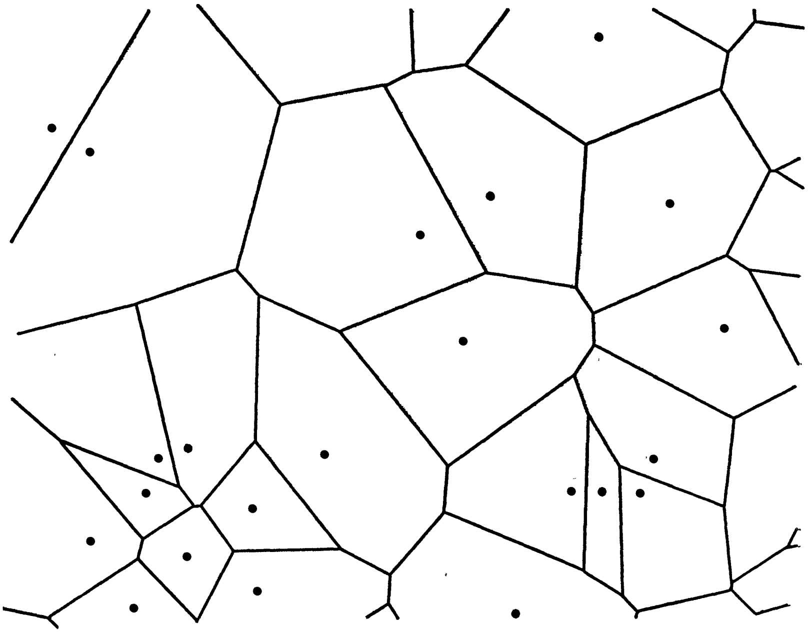

FIG. 1. Cell model

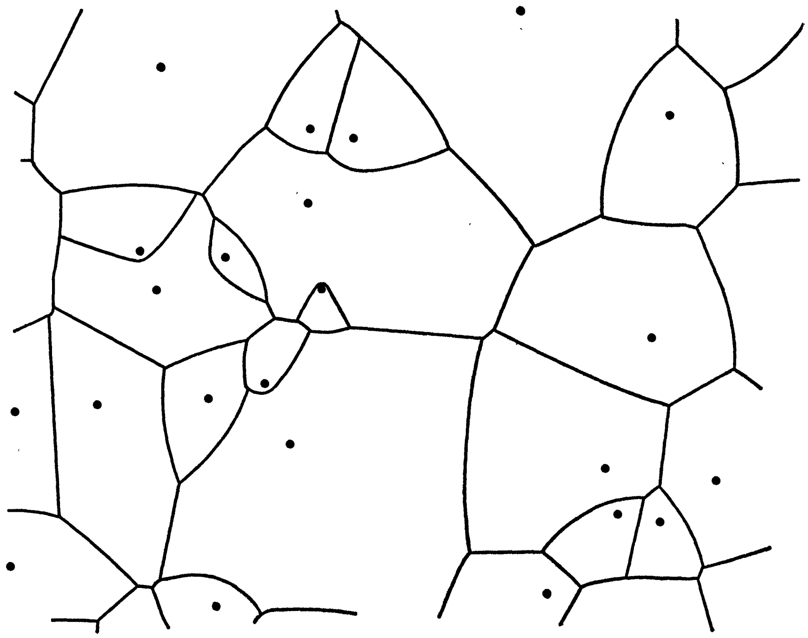

FIG. 2. Johnson-Mehl model

TABLE I

*Meijering's Mean Values for Crystals*

| | Cell Model | JM Model |
|---|---|---|
| 1. Surface area | 5.821 $\rho^{-\frac{2}{3}}$ | 5.143 $\rho^{-\frac{2}{3}}$ |
| 2. Edge length | 17.50 $\rho^{-\frac{1}{3}}$ | 14.71 $\rho^{-\frac{1}{3}}$ |
| 3. Number of vertices | 27.07 | 22.56 |
| 4. Number of edges | 40.61 | >33.84 |
| 5. Number of faces | 15.54 | >13.28 |
| 6. Number of crystal sections per unit area cut by a plane | 1.458 $\rho^{\frac{2}{3}}$ | >1.225 $\rho^{\frac{2}{3}}$ |

of the boundary are considered separate faces). These odd configurations occur when one or more small crystals grow from late arrivals located near the interface between two large crystals.

In arrival space, the set of possible arrivals $A = (P, t)$ which might crystallize the point $P_i$ before time $t_i$ is a cone. It has $(D + 1)$-dimensional volume

$$V = S(D)v^D t_i^{D+1}/(D + 1)$$

where

$$S(D) = \pi^{\frac{1}{2}D}/\Gamma(\tfrac{1}{2}D + 1)$$

is the volume of a unit $D$ dimensional sphere. Hence, as noted for $D \leqq 3$ by Evans and by Johnson and Mehl, $\exp(-\alpha V)$ is the probability that an arrival at time $t_i$ produces a crystal (nonempty cell).

Following Evans, $\exp(-\alpha V)\alpha\, dt_i$ is the expected number of arrivals $A_i$ in a unit volume during a time interval $dt_i$ which produce crystals. Integrate over $0 \leqq t_i < \infty$ to get the ultimate density $\rho$ of crystals per unit volume:

$$\rho = (\alpha/v)^{D/(D+1)}\{(D + 1)/S(D)\}^{1/(D+1)}\Gamma([D + 2]/[D + 1]). \tag{6}$$

Numerically, $\rho = .8794(\alpha/v)^{\frac{2}{3}}$ when $D = 2$ and $\rho = .8960(\alpha/v)^{\frac{3}{4}}$ when $D = 3$.

Table I lists some of the mean values which Meijering derived for crystals in the cell model and Johnson-Mehl (JM) model with $D = 3$. For ease of comparison, the results for the Johnson-Mehl model are expressed (using (6)) in terms of the density $\rho$ instead of $\alpha/v$.

In Table I, items 1, 2, 3 are fundamental parameters from which items 4, 5, 6 and many other mean values follow directly. For instance, $E$ (corners in unit area) in (2) is related to item 2 because

$$3E \text{ (edge length in unit volume)} = \rho E \text{ (edge length of a crystal)}.$$

Next, since a plane through the cell model contains $\frac{3}{2}$ as many boundary lines as corners per unit area, item 6 follows by using Euler's formula for polyhedra [9]. Similarly for the cell model, items 4 and 5 come from item 3 and Euler's formula. For the Johnson-Mehl model, similar arguments give only bounds. In the cell model the mean number of vertices of a face is $3 \times 27.07/15.54 = 5.23$, in agreement with Bernal's observation about pentagonal faces. According to item 1, the interface area per unit volume in a cell model solid is 2.910 $\rho^{\frac{1}{3}}$. Then, by (1), a line intersects 1.455 $\rho^{\frac{1}{3}}$ crystals per unit length on the average. Similarly a Johnson-Mehl solid cuts a line into an expected number 1.286 $\rho^{\frac{1}{3}}$ of sections per unit length. However since Johnson-Mehl crystals need not be convex different line sections may belong to the same crystal.

Since at least one high-dimensional instance of the cell model has been mentioned, formulas which hold for arbitrary dimension $D$ may be of interest. The expected $(D - 1)$-dimensional volume of the surface of a crystal in the cell model is

$$E(\text{surface}) = 2^D\Gamma(2-(1/D))\Gamma(\tfrac{1}{2}D)\{\Gamma(\tfrac{1}{2}D - 1)/\rho\}^{1-(1/D)}/\Gamma(D - \tfrac{1}{2}). \tag{7}$$

The proof of (7) will be omitted; it can be given in the manner of Meijering's derivation for the case $D = 3$. When a line is passed through a solid the expected number of times per unit length that the line intersects a crystal interface can be found by a Buffon needle argument; (1) generalizes to

$$\frac{E(\text{intersections in unit length})}{E(\text{surface})} = \frac{\rho S(D-1)}{DS(D)}. \tag{8}$$

Then for a line through the cell model,

$$E(\text{intersections in unit length}) = \rho^{1/D}\frac{2\Gamma(2-1/D)\{\Gamma(\tfrac{1}{2}D+1)\}^{(2D-1)/D}\Gamma(D)}{D\{\Gamma(\tfrac{1}{2}D+\tfrac{1}{2})\}^2\Gamma(D-\tfrac{1}{2})}. \tag{9}$$

In deriving (9) from (7) and (8), Legendre's duplication formula [22] was used.

**4. Variances.** Section 3 quoted some numbers of crystal sections per unit length or area on a line or plane through a cell model solid. The reciprocals of these numbers are mean lengths or areas of crystal sections. The variances of these intersection lengths and areas and of the crystal volumes will now be considered. In the Johnson-Mehl model different crystal sections may belong to the same (nonconvex) crystal. Some results will be derived for the intersection between a Johnson-Mehl crystal and a line or a plane but such an intersection may contain several crystal sections.

Consider the intersection between a $K$-dimensional hyperplane $H$ and a $D$-dimensional solid. Values of $K$ satisfying $1 \leqq K \leqq D$ will be allowed; if $K = D$ the intersection is the solid itself. Pick a crystal, say $C_1$, which $H$ intersects and let $F(V)$ be the probability density function for the $K$-dimensional volume of the intersection $HC_1$. If one first picks a point on the hyperplane (say the origin) and then examines the $K$-dimensional volume of the intersection of $H$ with the crystal $C^*$ containing that point, this intersection volume has a different density function $\{E(\text{volume})\}^{-1}VF(V)$. In this expression the volume in the expected value is that of $HC_1$; the factor $V$ appears because a large intersection has a proportionally large chance of containing the origin. The expected $K$-dimensional volume of $HC^*$ is

$$E(\text{volume}^*) = E\{(\text{volume})\}^{-1}\{E(\text{volume})^2\}.$$

Then the volume of $HC_1$ has variance equal to

$$\text{(10)} \qquad \text{Var}(\text{volume}) = E(\text{volume})E(\text{volume}^*) - \{E(\text{volume})\}^2.$$

Since $E$(volume) in (10) is known for the cell model, it suffices to find $E$(volume*). When $K = D$, $E(\text{volume}) = \rho^{-1}$ in both models.

Let $P(b)$ be the probability that a point $Q$, at distance $b$ from the origin $O$, belongs to the same crystal $C^*$ as $O$. Then the expected volume of the intersection $HC^*$ is

$$\text{(11)} \qquad E(\text{volume}^*) = E\left\{\int_{HC^*} dm(Q)\right\} = \int_0^\infty P(b)\, d\{S(K)b^K\}.$$

The term $P(b)$ in (11) requires separate derivations for the cell and Johnson-Mehl models.

In the cell model, $Q$ and the origin $O$ belong to the same cell $C^* = C_i$ if and only if the two spheres $K_1$, $K_2$, centered on $O$, $Q$ and with $P_i$ on their boundaries, both contain no other $P_j$. Given the location of $P_i$, the probability that $O$ and $Q$ both belong to $C_i$ is then $\exp\{-\rho \text{ Vol}(K_1 \vee K_2)\}$ where $K_1 \vee K_2$ is the union of $K_1$ and $K_2$. Thus

$$\text{(12)} \qquad P(b) = \rho \int \exp\{-\rho \text{ Vol}(K_1 \vee K_2)\}\, dm(P_i),$$

the integral taken over all $D$-dimensional space. To compute the integral, let $Rb$ denote the distance from $O$ to the integration point $P_i$ and let $u$ be the angle $P_iOQ$. Then the distance from $P_i$ to $Q$ is $b(R^2 + 1 - 2R \cos u)^{\frac12}$ and the angle $P_iQO$ is a function $U(R, u)$ defined by

$$\cos U(R, u) = \frac{1 - R \cos u}{(R^2 + 1 - 2R \cos u)^{\frac12}}.$$

In terms of these variables,

$$\begin{aligned}\text{Vol}(K_1 \vee K_2) &= S(D-1)b^D\left\{R^D \int_u^\pi \sin^D t\, dt \right.\\ &\qquad\qquad \left. + (R^2 + 1 - 2R\cos u)^{D/2} \int_{U(R,u)}^\pi \sin^D t\, dt\right\}\\ &= b^D V(R, u), \quad \text{say.}\end{aligned}$$

This formula follows because $K_1 \vee K_2$ is a union of two disjoint truncated spheres. For any given value of $D$ the integrals defining $V(R, u)$ may be evaluated in terms of elementary functions. Now (12) becomes

$$\text{(13)} \quad P(b) = \rho \int_0^\pi \int_0^\infty e^{-\rho V(R,u) b^D}(D-1)S(D-1)\sin^{D-2} u\, b^D R^{D-1}\, dR\, du.$$

After substituting (13) into (11) and integrating first on $b$ a double integral remains

$$E(\text{volume}^*) = \rho^{-K/D} \frac{(D-1)S(D-1)KS(K)\Gamma((D+K)/D)}{D} \cdot \int_0^{\pi} \int_0^{\infty} \frac{\sin^{D-2} u R^{D-1}\, dR\, du}{\{V(R, u)\}^{(D+K)/D}}. \tag{14}$$

The double integral appears analytically intractable but can be evaluated numerically. The items for the cell model in Table II were computed on an IBM 7090 using (14).

A similar analysis applies to the Johnson-Mehl model. Only cases with $D = 3$ will be considered. In (11) $P(b)$ is now the probability that points $O$ and $Q$, distant $b$ apart, are both crystallized by the same arrival say $A_o = (y_o, y_o, z_o, t_o)$. Thus

$$P(b) = \alpha \iiiint \text{Prob}(A_o \text{ crystallizes both } O \text{ and } Q)\, dx_o dy_o\, dz_o\, dt_o. \tag{15}$$

The integrand in (15) must be of the form

$$\text{Prob}(A_o \text{ crystallizes both } O \text{ and } Q) = \exp(-E), \tag{16}$$

where $E$ is the expected number of arrivals which could crystallize either $O$ or $Q$ or both before $A_o$. $E/\alpha$ is the volume, in arrival space, of the points $(x, y, z, t)$ which satisfy one or both of the inequalities

$$Rb + vt < R_o b + vt_o, \qquad rb + vt < r_o b + vt_o,$$

where $Rb$ is the distance from $(x, y, z)$ to $O$, $R_o b$ is the distance from $(x_o, y_o, z_o)$ to $O$ and $rb$, $r_o b$ are similar distances from $(x, y, z)$, $(x\ , y_o, z_o)$ to $Q$. Introduce new variables $\xi_o, \eta_o, \zeta_o, \tau_o$ by means of

$$x_o = b\xi_o, \qquad y_o = b\eta_o, \qquad z_o = b\zeta_o, \qquad t_o = b\tau_o/v.$$

Then $E = (\alpha/v)b^4 W$ where

$$W = \iiint\!\!\int_{I \vee J} d\xi\, d\eta\, d\zeta\, d\tau. \tag{17}$$

In (17) the region of integration is a union of two four-dimensional cones $I$ and $J$ defined by

$$I\colon R + \tau < R_o + \tau_o, \qquad \tau \geqq 0$$

$$J\colon r + \tau < r_o + \tau_o, \qquad \tau \geqq 0$$

where

$$R^2 = \xi^2 + \eta^2 + \zeta^2, \qquad r^2 = (\xi - 1)^2 + \eta^2 + \zeta^2.$$

Note that $W$ is a function of only two variables $R_o + \tau_o$ and $r_o + \tau_o$.

A quintuple integral for $E(\text{volume}^*)$ now follows from (11), (15), (16), and (17):

$$E(\text{volume}^*) = (\alpha/v)KS(K)\int_0^\infty b^{K+3}\,db\int\int\int\int \exp\{-(\alpha/v)\,b^4W\}\,d\xi_o\,d\eta_o d\,d\zeta_o\,d\tau_o\,.$$

It only remains to simplify this result.

Since $W$ does not depend on $b$, integrate first with respect to $b$ to get

$$E(\text{volume}^*) = C\int\int\int\int W^{-(K+4)/4}\,d\xi_o\,d\eta_o\,d\zeta_o\,d\tau_o \tag{18}$$

where $C$ is the constant $(v/\alpha)^{K/4}\Gamma((K+4)/4)KS(K)/4$.

Next change coordinates from $\tau_o$, $\xi_o$, $\eta_o$, $\zeta_o$ to $\tau_o$, $R_o$, $r_o$ and an angle $\Phi$ of rotation about the $\xi$ axis. The volume element in these coordinates is $r_oR_odr_o\cdot dR_od\Phi d\tau_o$. Since $W$ does not depend on $\Phi$, (18) simplifies further to

$$E(\text{volume}^*) = 2\pi C\int_0^\infty dt_o\int\int W^{-(K+4)/4}\,r_o\,R_o\,dr_o\,dR_o\,.$$

The integration extends over values $r_o$, $R_o$ satisfying both $r_o + R_o > 1$, and $|r_o - R_o| < 1$.

A final reduction follows by introducing new variables $q = r_o - R_o$, $s = r_o + R_o + 2\tau_o$ to replace $r_o$ and $R_o$. Since $R_o + \tau_o = (s - q)/2$ and $r_o + \tau_o = (s + q)/2$, $W$ can be expressed entirely in terms of $s$ and $q$. Now $r_oR_odr_odR_o$ becomes

$$\{(s - 2\tau_o)^2 - q^2)\,ds\,dq/8$$

and the range of integration is $1 < 1 + 2\tau_o < s$, $-1 < q < 1$. Integrate with respect to $\tau_o$ to obtain

$$E(\text{volume}^*) = C'\int_1^\infty ds\int_{-1}^1 \{s^3 - 1 - 3q^2(s - 1)\}\,W^{-(K+4)/4}\,dq \tag{19}$$

where $C' = \pi(v/\alpha)^{K/4}\Gamma((K + 4)/4)KS(K)/96$.

(19) is suitable for numerical integration once $W$ is expressed as a function of $s$ and $q$. The cones $I$ and $J$ of (17) have heights

$$R_o + \tau_o = (s - q)/2 \quad\text{and}\quad r_o + \tau_o = (s + q)/2.$$

Their volumes are

$$\pi(s - q)^4/48 \quad\text{and}\quad \pi(s + q)^4/48.$$

Thus

$$W = \pi(s^4 + 6s^2q^2 + q^4)/24 - \text{Vol}\,(IJ).$$

Since the axes of the cones $I$ and $J$ are unit distance apart, the values of $\tau$ in the intersection $IJ$ range over

$$0 \leqq \tau \leqq \tfrac{1}{2}(R_o + r_o + 2\tau_o - 1) = \tfrac{1}{2}(s - 1).$$

The points of $IJ$, having a fixed value of $\tau$ in this range, lie in the intersection of two three-dimensional spheres. Their radii are

$$(s - 2\tau - q)/2 \quad \text{and} \quad (s - 2\tau + q)/2$$

and their centers are unit distance apart. The intersection has three-dimensional volume

$$\pi\{(1 - 3q^2) + 6q^2(s - 2\tau) - 3(1 + q^2)(s - 2\tau)^2 + 2(s - 2\tau)^3\}/12.$$

Vol $(IJ)$ is the integral of this function of $\tau$ from $\tau = 0$ to $\tau = (s - 1)/2$. The final result for $W$ in (19) is

$$W = \pi\{1 - 2q^2 + 2q^4 + (6q^2 - 2)s + 6q^2s^2 + (2 + 2q^2)s^3 + s^4\}/48.$$

Using (19), Mrs. C. M. Kimme computed the values shown in Table II for the Johnson-Mehl model. The volume variances for the two models are very different. Meijering found both distribution functions for crystal volumes when $D = 1$ and noted that the cell model distribution had a wider spread than the Johnson-Mehl distribution. The methods of this section give the variances as integrals which turn out to be elementary. The variances are $1/(2\rho^2)$ for the cell model and

$$\rho^{-2}\{(\pi/4)(1 + \log_e(\tfrac{3}{2})) - 1\} = .1029\,\rho^{-2}$$

for the Johnson-Mehl model. Meijering expected that the cell model might continue to have the wider spread in crystal size when $D > 1$ but Table II shows the opposite when $D = 3$. Judging from Figures 1 and 2, the opposite may also hold when $D = 2$.

TABLE II

| $D$ | $K$ | $E$(volume*) | | Var(volume) | |
|---|---|---|---|---|---|
| | | Cell | JM | Cell | JM |
| 2 | 1 | $1.027\ \rho^{-\frac{1}{2}}$ | | $.188\ \rho^{-1}$ | |
| | 2 | $1.280\ \rho^{-1}$ | | $.280\ \rho^{-2}$ | |
| 3 | 1 | $.919\ \rho^{-\frac{1}{3}}$ | $1.093\ \rho^{-\frac{1}{3}}$ | $.159\ \rho^{-\frac{2}{3}}$ | |
| | 2 | $1.018\ \rho^{-\frac{2}{3}}$ | $1.482\ \rho^{-\frac{2}{3}}$ | $.228\ \rho^{-\frac{4}{3}}$ | |
| | 3 | $1.180\ \rho^{-1}$ | $2.136\ \rho^{-1}$ | $.180\ \rho^{-2}$ | $1.136\ \rho^{-2}$ |

**5. Large crystals.** This section will examine, for both models, the distribution function for the crystal volume $V$ as $V$ becomes large. Let $Q(V)$ denote the probability that a crystal has volume $V$ or greater. Upper and lower bounds on on $Q(V)$ will be given. These bounds ((20, (22), (24), and (28)), although not very accurate, are sharp enough to show that large crystals are rarer in the Johnson-Mehl model than in the cell model. Specifically, $-\log Q(V)$ is $O(V)$ for the cell model and is $O(V^{(D+1)/D}$ for the Johnson-Mehl model (the exponents 1 and $(D + 1)/D$ are smallest possible.

For a lower bound on $Q(V)$ it suffices to find the probability $g(R)$ that a crystal $C_i$ contains a sphere of radius $R$ (where $V = S(D)R^D$) centered on $P_i$. In the cell model, $g(R)$ is just the probability that no other center $P_j$ lies within $2R$ of $P_i$. Thus

$$g(r) = \exp\{-\rho 2^D V\} \leqq Q(V) \tag{20}$$

for the cell model.

To find $g(R)$ for the Johnson-Mehl model, consider all the arrivals $A_i$ having $P_i$ in a certain unit volume of space. Let $N(R)$ be the expected number of such arrivals for which the cell $C_i$ contains a sphere of radius $R$ and center $P_i$. Since any such cell is certainly a crystal it follows that

$$N(R) = \rho g(R). \tag{21}$$

Also, an argument like the one which derived (6) will give a formula for $N(R)$. An arrival $A = (P, t)$ with $P$ at distance $r$ from $P_i$ will interfere with the sphere about $P_i$ if

$$| r - R | + vt \leqq R + vt_i .$$

In arrival space this inequality describes part of a cone

$$r \leqq 2R + v(t_i - t), \qquad 0 \leqq t \leqq t_i + R/v$$

from which a conical piece

$$r \leqq v(t - t_i), \qquad t_i \leqq t \leqq t_i + R/v$$

has been removed. The expected number of arrivals in this region is

$$m(t_i) = (\alpha/v)\,[S(D)/(D+1)]\{(2R + vt_i)^{D+1} - 2R^{D+1}\}.$$

Then

$$N(R) = \alpha \int_0^\infty \exp\{-m(t_i)\}\, dt_i .$$

After using (21) and simplifying, the result for $g(R)$ is

$$g(R) = \frac{\alpha}{v\rho} \exp\left\{\frac{2\alpha S(D) R^{D+1}}{v(D+1)}\right\} \int_{2R}^\infty \exp\left\{\frac{-\alpha S(D) x^{D+1}}{v(D+1)}\right\} dx. \tag{22}$$

For large $R$, $g(R)$ has an asymptotic expansion which can be obtained by repeatedly integrating (22) by parts. The leading term is

$$g(R) \sim \frac{\exp\{-(2^{D+1} - 2)(\alpha/v)S(D)R^{D+1}/(D+1)\}}{\rho S(D)(2R)^D}.$$

Thus, for the Johnson-Mehl model, the lower bound $g(R)$ on $Q(V)$ has the asymptotic form $\exp\{\text{const. } V^{(D+1)/D}\}/\{\rho^{2D} V\}$.

The proofs of the upper bounds use expected values of certain integrals where the crystal $C_i$ is the region of integration. For the cell model the appropriate formula is

$$(23)\qquad E\left\{\int_{C_i} \exp\,\{s\rho S(D)r^D\}\,dx_1 \cdots dx_D\right\} = \{\rho(1 - s)\}^{-1},$$

which holds for all $s < 1$. In (23) $r$ is the distance from $P_i$ to $(x_1, \cdots, x_D)$. To prove (23) note that a point $(x_1, \cdots, x_D)$ at distance $r$ from $P_i$ has probability $\exp\,\{-\rho S(D)r^D\}$ of belonging to $C_i$. Then the expectation is

$$\int_0^\infty \exp\,\{\rho S(D)(s - 1)r^D\}\,d(S(D)r^D)$$

which has the value (23).

Note, for $0 \leqq s < 1$, that the integral in (23) is greater than a similar integral taken over a sphere of the same volume as $C_i$. Thus

$$s^{-1}E\{\exp(\rho s\,\mathrm{Vol}\,(C_i)) - 1\} \leqq (1 - s)^{-1}$$

$$s^{-1}(\exp(\rho s\,V) - 1)Q(V) \leqq (1 - s)^{-1}$$

when $0 \leqq s < 1$. The choice $s = 1 - (\rho V)^{-1}$ gives a simple bound for the cell model

$$(24)\qquad Q(V) < \{(\rho V - 1)/[\exp(\rho V - 1) - 1]\}$$

which holds for $V > \rho^{-1}$.

For the Johnson-Mehl model, the expected integral to consider is

$$(25)\qquad J(s) = E\left\{\int_{C_i} \exp\,\{s(\alpha/v)S(D)r^{D+1}/(D + 1)\}\,dx_1 \cdots dx_D\right\}.$$

To evaluate (25), consider first an expected value of a sum of integrals

$$(26)\qquad E\left\{\sum_{A_i} \int_{C_i} \cdots\right\},$$

where the integral over $C_i$ is as in (25) and the sum extends over arrivals $A_i$ which appear in some unit volume of space. In (26) it is immaterial whether all arrivals $A_i$ are summed or only the arrivals which produce crystals; if an arrival $A_i$ does not produce a crystal, the cell $C_i$ is empty and the contribution of $A_i$ to the sum (26) is zero. By summing over crystals only, one finds for (26) the value $\rho J(s)$. By summing next over all arrivals one concludes

$$(27)\qquad \rho J(s) = \alpha \int_0^\infty E\left\{\int_{C_i} \cdots\right\} dt,$$

where the integral over $C_i$ is again the one in (25) and where now $C_i$ is a cell (not necessarily a crystal) produced by an arrival at time $t$.

In the integral over $C_i$, a point $P$ at distance $r$ from $P_i$ will belong to $C_i$ only if it remains uncrystallized up to time $t + (r/v)$. Thus (see discussion preceding (6)) $P$ has probability

$$\exp\{-(\alpha/v)S(D)(vt + r)^{D+1}/(D + 1\}$$

of belonging to $C_i$. Now (27) becomes

$$\rho J(s) = \alpha\int_0^\infty \int_0^\infty \exp\ \{(\alpha/v)S(D)\ [sr^{D+1} - (vt + r)^{D+1}]/(D+1\}DS(D)r^{D-1}\,dr\,dt.$$

Make the substitution $t = ry/v$ and integrate on $r$ to get

$$\rho J(s) = D\int_0^\infty \{dy/[(y + 1)^{D+1} - s]\}.$$

Although $J(s)$ can now be evaluated in elementary terms for each value of $D$, it will suffice to note that $J(s)$ exists for $s < 1$.

Again, for $0 \leqq s < 1$, the integral over $C_i$ in (25) exceeds a similar integral taken over a sphere of the same volume as $C_i$. Let $R$ be the radius of this sphere. Then

$$\begin{aligned} J(s) &\geqq E\left\{\int_0^R \exp\ \{s(\alpha/v)S(D)r^{D+1}/(D + 1)\}\ d(S(D)\ r^D)\right\} \\ &> D/(D + 1)E\left\{R^{-1}\int_0^R \exp\ \{s(\alpha/v)S(D)\ r^{D+1}/(D + 1)\}\ d(S(D)\ r^{D+1})\right\} \\ &> D(v/\alpha)s^{-1}\,E\{R^{-1}(\exp\ \{s(\alpha/v)S(D)R^{D+1}/(D + 1)\} - 1\}. \end{aligned}$$

Since the expression in the last expectation is an increasing function of $R$, the expectation is greater than $Q(S(D)R^D)$ times the same function of $R$. In terms of the volume $V = S(D)R^D$ of $C_i$ the inequality is

$$Q(V) \leqq \frac{sJ(s)(\alpha/v)V^{1/D}D^{-1}(S(D))^{-1/D}}{\exp\ \{s(\alpha/v)V^{(D+1)/D}(D + 1)^{-1}(S(D))^{-1/D}\} - 1}. \tag{28}$$

Although one could study this bound further, picking $s$ to make it as small as possible, any value of $s$ in $0 < s < 1$ completes the proof that $-\log Q(V) = O(V^{(D+1)/D})$ for the Johnson-Mehl model.

**6. Generalizations.** The assumption (ii), which both models make, is unrealistic for many minerals. Single crystals, growing without interference, may be as unspherical as flat plates or thin needles. One may alter either the cell or Johnson-Mehl model by assuming that crystals grow as star-shaped figures of some given shape and that they are randomly oriented. A crystal then has a velocity of growth $v(U)$ defined for each direction $U$. This velocity is to be measured along the straight line ray of direction $U$ from the center $P_i$ to the point where the ray intersects the surface of the crystal.

Evans considered a generalized Johnson-Mehl model for $D = 2$ and assigned each point $P$ to the crystal which, if growing free of interference from other crystals, would reach $P$ first. This rule for defining cells $C_i$ seems a natural generalization of (5) and is convenient in analysis. For example, the expected number of arrivals per unit volume which appear at uncrystallized locations is now

$$\alpha\{\alpha S(D)E(v^D)/(D+1)\}^{-1/(D+1)}\Gamma\{(D+2)/(D+1)\}. \tag{29}$$

Here $E(v^D)$ denotes the expected value of $\{v(U)\}^D$ when $U$ is chosen at random with constant density for all directions. The case $D = 2$ of (29) was given by Evans. However crystals in this model appear to grow through one another and in fact may even be disconnected. For example, in the case $D = 2$ with crystals which grow as squares, consider two isolated seeds differing in orientation by 45°. Since the corners are the fastest-growing points each of the two crystals is sure to contain all points sufficiently far out on the rays through its corners; thus both crystals cannot be connected. The same argument applies regardless of the arrival times of the seeds. In particular one seed may arrive after its location is already crystallized. Then (29) is not the density of crystals.

Retaining assumption (ii), similar difficulties arise in constructing a model of a polycrystalline solid composed of two substances with arrival rates $\alpha_1$, $\alpha_2$, and (constant) growth rates $v_1$, $v_2$. A fast-growing crystal may penetrate through an array of slow-growing crystals and become disconnected. A fast-growing arrival may appear inside an existing slow-growing crystal, overtake it, and surround it. The rule for defining crystals in such cases should be changed to stop growth permanently along a ray once it meets another crystal. Such a rule seems to entail analytical difficulties.

Mr. Brian Bayly (University of Chicago) has suggested to the author that in (ii) the rate of crystallization $v$ might be made a decreasing function $v(r)$ of the distance $r$ from the nucleus to the crystal face. In this way one might try to account for the retarding effect of the heat liberated during crystallization. The larger crystals (which have liberated more heat and so are warmer) are made to grow more slowly. This modification is easy to include in the cell model. Although crystal faces now grow at a variable rate, all crystals still have the same growth rate at the same time. Consequently the final crystals are the same cells $C_i$ defined in Section 3. A similar modification of the Johnson-Mehl model presents the following difficulty. Consider a large, and hence slow-growing, crystal $C$. A new nucleus $A$ may arrive inside $C$ but near the surface of $C$. $A$ ought to be discarded. However, the analytically convenient way of generalizing the rule (5) permits $A$ to grow until it overtakes the surface of $C$ and forms a crystal $C'$ which does not contain its own nucleus.

## REFERENCES

[1] Bernal, J. D. (1959). A geometrical approach to the structure of liquids. *Nature* **183** 141–147.

[2] Blaschke, W. (1955). *Vorlesungen über Integralgeometrie*. Deutsche Verlag der Wiss., Berlin.

[3] Chayes, Felix (1956). *Petrographic Modal Analysis.* Wiley, New York.

[4] Corrsin, S. (1955). A measure of the area of a homogeneous random surface in space. *Quart. Appl. Math.* **12** 404–408.

[5] Coxeter, H. S. M. (1958). Close packing and froth. *Ill. J. Math.* **2** 746–758.

[6] Delesse, A. (1848). Procédé méchanique pour déterminer la composition des roches. *Ann. des Mines* (4th Ser.) **13** 379–388.

[7] Evans, U. R. (1945). The laws of expanding circles and spheres in relation to the lateral growth of surface films and the grain size of metals. *Trans. Faraday Soc.* **41** 365–374.

[8] Herden, G. (1960). *Small Particle Statistics*, 2nd Ed. Academic Press, New York.

[9] Hilbert, D. and Cohn-Vossen, S. (1952). *Geometry and the Imagination.* Chelsea, New York.

[10] Johnson, W. A. and Mehl, R. F. (1939). Reaction kinetics in processes of nucleation and growth, *Trans. A.I.M.M.E.*, **135** 416–458.

[11] Lewis, F. T. (1946). The shape of cells as a mathematical problem. *American Scientist* **34** 359–369.

[12] Marvin, J. W. (1939). The shape of compressed lead-shot and its relation to cell shape. *Amer. J. Botany* **26** 280–288.

[13] Meijering, J. L. (1953). Interface area, edge length, and number of vertices in crystal aggregates with random nucleation. *Philips Res. Rep.* **8** 270–290.

[14] Rosiwal, A. (1898). Ueber geometrische gesteinsanalysen. Ein einfacher weg zur ziffermaessigen feststellung des quantitaetsverhaeltnisses der mineralbestandteile gemengter gesteine. *Verhandlungen der K.K. Geologischen Reichanstalt* **5–6** 143–175.

[15] Shannon, C. E. (1949). Communication in the presence of noise. *Proc. I.R.E.* **37** 10–21.

[16] Smith, Cyril Stanley (1952). Grain shapes and other metallurgical applications of topology. *Metal Interfaces*, Amer. Soc. for Metals, Cleveland, 65–113.

[17] Smith, Cyril Stanley and Guttman, Lester (1953). Measurement of internal boundaries in three-dimensional structure by random sectioning. *J. Metals* **5** 81–87.

[18] Smith, C. S. (1953). Further notes on the shape of metal grains: space-filling polyhedra with unlimited sharing of corners and faces. *Acta Metallurgica* **1** 295–300.

[19] Smith, Cyril Stanley (1954). The shape of things. *Scientific Amer.* **190** No. 1, 58–64.

[20] Thompson, D'Arcy W. (1942). *On Growth and Form*, 2nd Ed. **2** Cambridge Univ. Press.

[21] Wells, A. F. (1956). *The Third Dimension in Chemistry.* Oxford Univ. Press.

[22] Whittaker, E. T. and Watson, G. N. (1944). *Modern Analysis.* Cambridge Univ. Press.