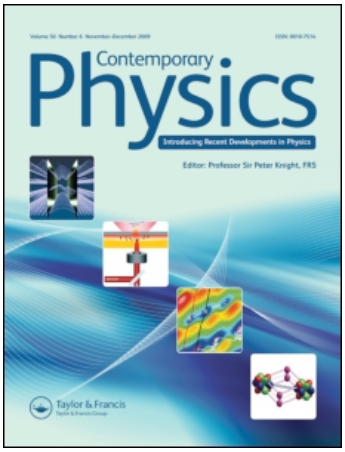

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Soap, cells and statistics—random patterns in two dimensions

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Soap, Cells and Statistics—Random Patterns in Two Dimensions

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ABSTRACT. Random two-dimensional patterns crop up in a wide variety of scientific contexts. What do they have in common? How can they be classified or analysed? These questions are underlined, and partly answered, by a survey of such patterns, paying particular attention to soap cell networks, metallurgical grain structures and the Giant's Causeway.

1. Introduction

All natural structures (and many artificial ones) represent some compromise between order and chaos. When order predominates, the definition and classification of structures can proceed straightforwardly. Consider the case of a solid in the form of a single crystal. A precise space group can be assigned to it, from a finite list of possibilities. A specification of the structure is completed by adding a few numbers for the lattice constants and local arrangement of atoms. Disorder may be incorporated in the conceptual framework by identifying local defects, such as dislocations, or considering the effect of the thermal excitation of lattice vibrations. Such is the programme of crystallography and the foundation of conventional solid-state physics.

But what if *disorder* predominates? In the context of solids, this is the case for a *glass* or amorphous solid, in which the atoms are arranged in an irregular, non-periodic structure. Although this has been a popular academic subject in recent times, we are still not sure where to start in specifying the structure in such a case (Zallen 1979). The term *amorphography* has been coined for the equivalent of crystallography, signifying a systematic approach to the definition of amorphous structures from diffraction data (Wright *et al.* 1980, Wright 1983). For the time being it is only a primitive and tentative procedure.

In some cases of disorder in condensed matter (such as the theory of simple liquids (March 1968, Collins 1967)), headway can be made by appeal to statistical thermodynamics, but often we are dealing with a structure which is far from thermal equilibrium. Metallurgical grains are an important example. Their arrangement is a metastable structure which forms in an accidental manner and will evolve (given a sufficiently high temperature and no factors which impede grain growth) towards a single crystal.

If one looks beyond physics, which even physicists should sometimes do, one sees that such non-ideal structures are the rule rather than the exception. Biological cells, geographical or ecological territories and other natural structures on scales much greater than molecular dimensions are *usually* disordered. Occasional counter-

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examples displaying periodic structures, such as the bee's honeycomb or the insect's compound eye, have always excited the enthusiasm of scientists. Here at last, they would say, is something we can get our teeth into with a bit of rigorous theory. At times, this prejudice was taken a stage further by imagining order when there was none, as in the frequent references to the Giant's Causeway as being 'hexagonal' and its eventual 'explanation' in such terms (Section 3.6). Similarly, plant cells were for long visualized as 'pretty regular dodecahedra' which they are certainly not. Even the bee's honeycomb is not as perfect as has often been supposed (Wyman 1865). Of course, idealization is a valid procedure in science, provided it is done consciously. But it is difficult to sustain a picture of many natural structures as imperfect realizations of some ordered ideal, from which they are distantly removed.

If the need for a proper theory of random structures is so obvious, how have the adherents of the various scientific disciplines responded to it?

Condensed-matter physicists have tended to ignore questions which lie far outside the range of equilibrium statistical thermodynamics, and to study disorder only in that context. Recently, however, there has been a growth of interest in disorder over a wider field—percolation theory, chaos, fractals—as well as the work of the amorphous solid-state community to which we have already referred.

Metallurgists, ceramicists and other materials scientists have always recognized the ubiquity of disorder and have made progress with empirical and analytical techniques for adequate data acquisition. This is no easy matter in the case of a three-dimensional structure, whatever its scale may be, and at best a tedious chore in two-dimensional cases.

Geographers and ecologists, who deal exclusively in two-dimensional structures in in which data acquisition is less of a problem, have been more ambitious in data analysis and in testing idealized mathematical models in terms of statistical criteria.

In recent years, the biological community has not shown much enthusiasm for questions relating to the random arrangement of cells. If one looks at this in the wider context that is offered in our later sections, it is difficult to see how such an attitude can be justified.

This article may offer some tentative insights to all these separate communities and may perhaps help to bring them together. It concentrates mainly on two-dimensional structures of the kind that may be loosely classified as 'cellular', typified by fig. 1. It is enough to take us into many fields—metallurgy, geology, hydrodynamics, ecology, geography, astronomy—so perhaps we may be excused this specialization.

Some of the most important general sources from which we have drawn information and inspiration are:

Thompson (1942): the classic work *On Growth and Form*, best read in this edition, as much amusing detail is suppressed in the later abridged edition.

Smith (1954, and other references below) complements Thompson's work with ideas concerning metallurgical structures, structure in art and aesthetics.

Dormer (1980): an account of the topological and geometrical principles relevant to biological tissue geometry.

Getis and Boots (1979): a survey of work on random patterns in two dimensions, mainly with a geographical perspective.

Stevens (1974) contains several beautiful photographs and a few elementary mathematical principles.

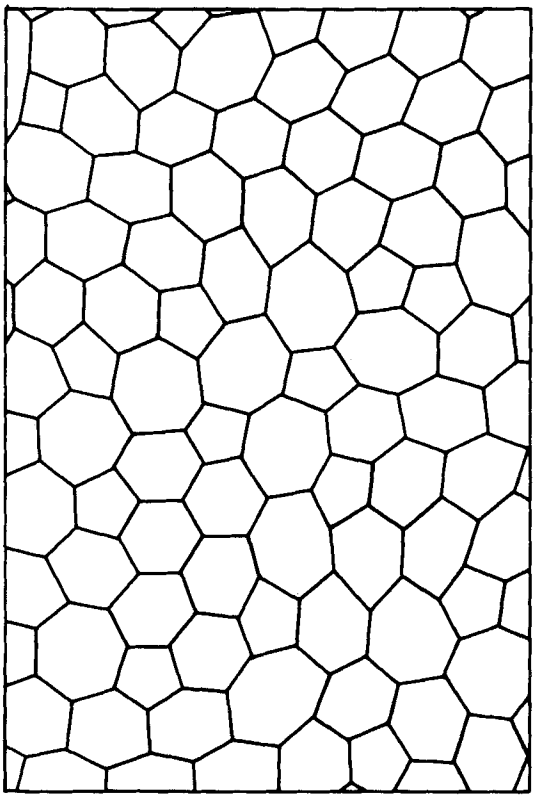


Fig. 1. Typical cellular structure in two dimensions.

Section 2 explains the various topological/geometrical definitions which are essential to the subject and states some of the few general theorems which are helpful. Particular mathematical constructions, leading to creation or modification of idealized random two-dimensional models, will be described in Section 2 with some indication of their motivation.

In Section 3, random cellular structures are taken from many different contexts, mostly in two dimensions (2D). They are compared with each other and with the idealized models of Section 2. Attempts are made to extract significant correlations between various structural parameters. *Much of this material can be read without recourse to the details of Section 2.*

Finally, in Section 4, we review some more speculative ideas on random patterns, arising out of statistical mechanics, and summarize the present state of our understanding.

2. Models and mathematics

2.1. *Geometry and topology*

All the geometrical and topological theorems which we shall use are really very elementary but the topological ones may be unfamiliar so they are reviewed here. It seems natural to include three-dimensional cellular structures at this point, even though they are of little relevance to our later sections. In both cases, the structure includes *vertices*, joined by *edges*, which surround *faces*, and in the three dimensional

case the faces surround *cells*. (When we restrict attention to two dimensions, we shall use the word *cells* for what are here called *faces*, for instance the polygons of fig. 1.)

The number of edges joined to a given vertex is its *coordination number*, z. Usually we shall be interested in *topologically stable structures*, which means that their topological properties are unchanged by small deformations—in practice, this implies $z=3$ (2D) and $z=4$ (3D), everywhere. We shall sometimes call a cellular structure with this coordination number a *froth*.

The cells, faces, edges and vertices of any cellular structure obey the conservation law (Euler's equation)

$$F-E+V=\chi \quad \text{(2D)} \tag{1}$$

$$-C+F-E+V=\xi \quad \text{(3D)} \tag{2}$$

Here C is the number of cells, E of edges, F of faces, and V of vertices. The quantity in the right hand side, χ or ξ, is an integer of order 1, and is a topological invariant of the space in which the tissue is put. For example, $\chi=2$ for a sphere or a rugby ball, and $\chi=0$ for a torus, doughnut or tea cup. $\chi=1$ for a plane and $\xi=1$ for 3D Euclidean space, when the face or cell at infinity is not counted.

An immediate consequence of Euler's equation in two dimensions is that the average number of edges surrounding a cell is six

$$\langle n\rangle=6 \quad \text{(2D)} \tag{3}$$

in the limit of a large system, provided all vertices have coordination $z=3$ as in fig. 1. Indeed one has the valence relations, $3V=2E$ and $\sum nF_n=2E$, where F_n numbers the n-sided cells, because 3 edges meet at every vertex and an edge links 2 vertices, whereas it separates 2 faces. Euler's relation can therefore be written as $(6-\sum nF_n/F)\ F=$a number of order 1, so that the bracketed expression equals zero in the limit of a tissue containing a large number of cells. The three-dimensional equivalent (with $z=4$) is less drastic,

$$\langle f\rangle=12/(6-\langle n\rangle) \quad \text{(3D)} \tag{4}$$

where $\langle f\rangle$ is the average number of faces of three-dimensional cells. Incidentally, equation (4), valid for the froth as a whole, also holds for each individual cell. It relates the number of faces f to $\langle n\rangle$, the average number of sides per face in the cell. It is a direct consequence of Euler's theorem (for 2D) for $\chi=2$ and $z=3$. Most random structures have $\langle f\rangle\simeq 14$, but it is, emphatically, *not* an exact result or even a limit, despite repeated statements in the literature to the contrary (Smith 1952, Laves 1967, Ogawa 1983, ...). A statistical froth with isotropic cells of equal volume has $\langle f\rangle=13{\cdot}40$ (corresponding to equal regular tetrahedra in the dual graph (cf. Section 3)). One 'can' pack 5·1 tetrahedra about a common edge; thus, by duality, $\langle n\rangle=5{\cdot}1$, and $\langle f\rangle=13{\cdot}40$. Fluctuations in the volumes of the cells reduce $\langle f\rangle$, whereas fluctuations in angles (anisotropy of the cells) increase $\langle f\rangle$ (Rivier 1982), as observed. (See Matzke and Nestler (1946), Hulbary (1948), Meijering (1953), Marvin (1939), who obtained a high $\langle f\rangle$ by compressing lead shot *uniaxially*, and Williams and Walker (1983).) Finally, Meijering (1953), by a remarkable feat of geometry, has obtained $f=(48/35)\pi^2+2(\simeq 15{\cdot}54)$ exactly for a Voronoi froth with randomly distributed centres. This corresponds to the territorial division of an ideal gas, whose cells are, accordingly, highly anisotropic.

Interesting but mistaken arguments leading to $\langle f\rangle=14$ rely on an arbitrary, planar cut of a three-dimensional tissue. Concentrate on one particular cell. The cut

divides it into two new cells. One states that (1) the new face (apparent on the cut) is hexagonal on average, as befits a two-dimensional mosaic decorating the planar cut; (2) this new face is typical of the froth. Statements (1) and (2) are not compatible. Either the cut is planar and the face created is not typical of the froth (the dihedral angle between two neighbouring faces in the cut is 180°), or a particular cell is divided, but this division does not produce an extended planar cut, but something with a positive curvature overall, leading to $\langle n \rangle < 6$. If one assumed that statements (1) and (2) were compatible, then one cut would produce a new froth with $C \to C+1$, and $F \to F+1+6$. After m random cuts, $C \to C+m$ and $F \to F+7m$. Given that $2F = \sum f C_f = \langle f \rangle C$, $\langle f \rangle = 2F/C \to 2(F+7m)/(C+m)$, which tends to 14 as $m \to \infty$. This argument is due to Laves (1967) who gives priority to van der Waerden and Meuse. But absolute priority should go to Duchartre (1867), who, in p. 141 of his *Eléments de Botanique*, formulates the correct result and gives fully and precisely the restrictive assumptions for its validity: "Les cellules dont la coupe est hexagonale forment donc chacune, du moins quand elles sont régulières, un solide à 14 faces (tetradecahèdre), et non à douze, comme on le dit souvent." Actually, the new face cannot be typical of the froth since $\langle n \rangle = 6$ implies $\langle f \rangle = \infty$ from equation (4).

There is another conservation law for three-dimensional tissues (Rivier 1979). Faces containing an odd number of edges cannot be found in isolation, but form lines which are either closed or terminate on the surface of the material. These lines thread only odd faces. The proof is similar to that of the Maxwell equation, div $\mathbf{B} = 0$, in electromagnetism, which implies that magnetic induction lines are closed and that magnetic monopoles are absent in classical electromagnetism. Odd lines may play an important part in the physical properties of glasses, both at high and low temperatures.

Returning to the two-dimensional systems which are our main interest, we shall identify three elementary processes by which a hexagonal network might be progressively modified to produce a disordered structure, or by which such a structure might itself change with time; they all maintain $z = 3$, except at the point of transition in the first case.

Firstly, there is the neighbour-switching process shown in fig. 2, which we shall call a T1 process. It can be pictured as taking place as an edge shrinks to zero, to be replaced by another one in such a way that the connections to vertices are rearranged. This, incidentally, justifies the statement that only vertices with $z = 3$ (in 2D) are topologically stable.

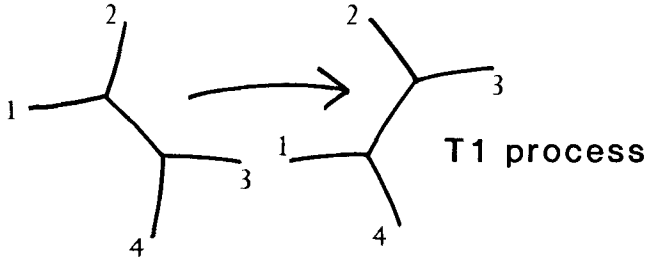


Fig. 2. Elementary local rearrangement of cells.

Secondly, faces (2D cells) may vanish—the T2 process of fig. 3. We confine this to the vanishing of a three-sided cell as shown. (A cell with more than three sides can vanish through a series of T1 processes to make it three-sided, followed by a T2 process.)

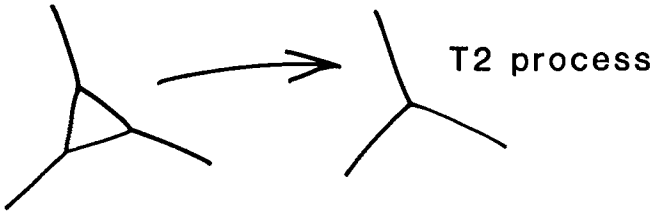


Fig. 3. Vanishing of a cell.

Thirdly, cells may be divided—the process shown in fig. 4. This could be visualized as a continuous process (combining the inverse of T2 with T1s) but it is suggested by mitosis in biology, which is a discontinuous change. In metallurgy, we must sometimes consider the inverse of the process of fig. 4 (coalescence of subgrains).

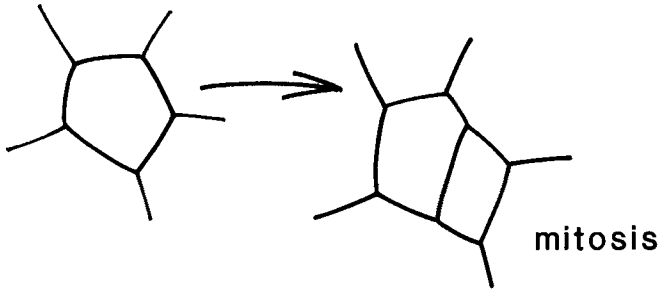


Fig. 4. Cell division.

The reader may like to check that the Euler relation (3) is preserved under such changes. Indeed, this is one approach to proving it. (Another simply follows from the fact that the average angle at a vertex is $2\pi/3$ and hence the average turning angle of polygonal cells must be $\pi - 2\pi/3 = 2\pi/6$).

Note also that a pentagon/heptagon construction in an otherwise hexagonal structure, may be regarded as a topological *dislocation* (fig. 5). For example, Lewis

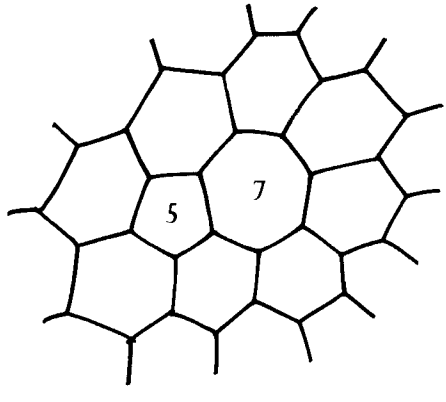


Fig. 5. A 5-7 pair of cells, making up a topological dislocation in an otherwise hexagonal structure.

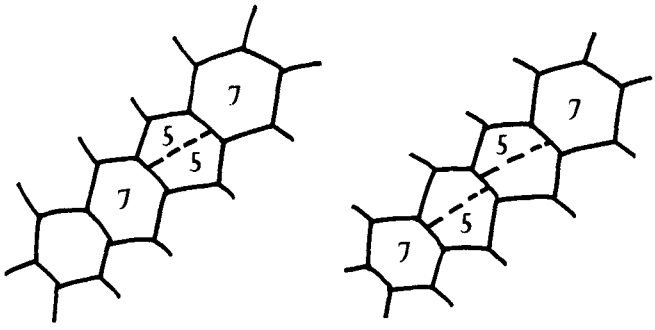


Fig. 6. Creation and dissociation of a dislocation pair by successive cell divisions.

(1943 b) and Pyshnov (1980) have noticed that successive mitoses of neighbouring cells (fig. 6) correspond, in two dimensions, to the creation of a dislocation dipole (a quadrupole of pentagons/heptagons), followed by glide of the two dislocations away from each other, leaving an additional cellular layer in between. This yields a 'topological solution for cell proliferation in intestinal crypt', and, as far as elastic energy is concerned, dislocations are a very efficient way to solve the problem of coping with additional material. The T1 process defined above also creates two such dipoles, which can then be dissociated (by further T1s) to form isolated pentagon/heptagon pairs. The intestinal crypt is a highly corrugated surface: its curvature is locally very high, even if it vanishes on average, as befits a cylindrical object. The dislocation glide mechanism of Pyshnov (1980) is stopped by pentagonal cells present in the regions of positive curvature of the intestine, and cell proliferation is arrested at these points.

Finally we should note that, while our main interest is in *planar* structures, there are plenty of examples of cellular structures on curved surfaces, such as a sphere. An excess of pentagons or other cells having $n<6$ (as in a football or in radiolaria) betrays positive overall curvature of the underlying space, whereas an excess of cells with $n>6$ betrays negative curvature (saddle-like surfaces). It is tempting to think of random tissue, with its non-hexagonal cells, as having been put on a substrate with fluctuating curvature (Gaspard *et al.* 1983, Nelson 1983).

2.2. *Statistical definitions*

It is natural to characterize a random structure in terms of the distribution functions of some of its local geometrical/topological properties, such as the number of edges or the area of a cell. These are indeed two popular choices, which we shall denote by $p(n)$ and $\phi(A)$. These obey

$$\sum_n p(n)=1 \tag{5}$$

$$\sum_n np(n)=6 \quad (z=3) \tag{6}$$

$$\int_0^\infty \phi(A)\,\mathrm{d}A=1 \tag{7}$$

$$\int_0^\infty A\phi(A)\,\mathrm{d}A=A_{\mathrm{mean}} \tag{8}$$

Equations (5) and (7) express the normalization of p and ϕ, (6) is Euler's relation (3), and (8) is just the definition of the mean area per cell. In general, one does not have a well established analytical form for p or ϕ, although various forms have been conjectured. One may therefore resort to consideration of their moments in characterizing them. These are

$$\mu_k = \sum_n (n-6)^k p(n) \tag{9}$$

$$M_k = \int_0^\infty (A - A_{\text{mean}})^k \phi(A)\, dA \tag{10}$$

By definition, $\mu_0 = M_0 = 1$, $\mu_1 = M_1 = 0$. The width of distributions is indicated by the second moment and its asymmetry by the third moment. Two other quantities which are sometimes used are the mean number of sides of neighbours of n-sided cells, which we shall call $m(n)$, and the mean area of n-sided cells, $\bar{A}(n)$.

Finally, it is often more convenient to measure the average linear intercept d, rather than A_{mean}, as a measure of the average size of cells. This is done by finding the average length of the segments into which straight lines (whose directions are random) are broken by their intersections with the cells. Loosely speaking, d is the typical cell diameter.

Examples of the use of all these quantities may be found in Section 3. Typical distributions can be seen in Pieri (1981) (geological crackings or jointings) and Smolyaninov (1980) (biological mosaics).

2.3. *Model structures*

At the risk of putting the cart before the horse, we shall now consider some idealized or artificial random cellular structures, before looking at naturally occurring ones.

2.3.1. *Voronoi polygons*

By far the most commonly studied ideal structure is the Voronoi construction, of which an example is shown in fig. 7. In this an array of points or centres is first defined, which may be completely random or correlated in some way. Then each of these centres is assigned a cell containing all points which are nearest to it. Specifically, one draws the

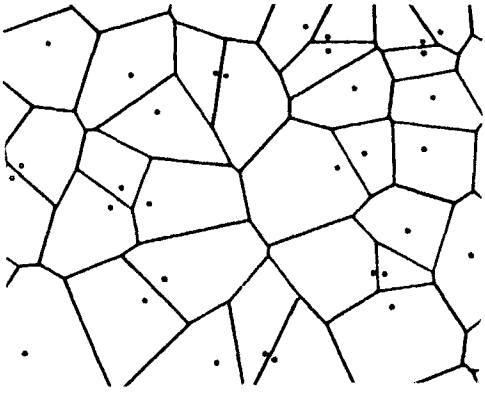


Fig. 7. Voronoi construction for random points, after Kiang (1966).

perpendicular bisecting planes (in 3D) or lines (in 2D) to the lines joining any two centres. The smallest convex polyhedron (polygon) surrounding a given centre contains all points in space closest to this centre. The construction is clearly unique. That it fills space can be seen in two dimensions from the result of elementary geometry that the three mediatrices of a triangle are concurrent. (The vertices of the triangle represent any three centres, and the mediatrices, the perpendicular, bisecting lines making up the faces of the Voronoi polygons. Their common point is a vertex of the Voronoi polygonal froth.)

The method described is not really practical for making the Voronoi construction (other than graphically), and several different computer algorithms have been proposed to do so (Crain 1978, Ocelli 1983, Brostow *et al.* 1978). Some idea of the popularity of this construction over a wide range of specific fields may be gained by considering the variety of its names—*Voronoi polygons, Wigner–Seitz cells, Dirichlet regions or tesselations, the cell model, the S-mosaic, Thiessen polygons, plant polygons, Wirkungsbereich* ... For a recent application in materials science, see Wray *et al.* (1983).

The Voronoi construction also imposes on to the original centres the structure of a graph, by defining unambigously which centres are nearest neighbours and linked by an edge in the graph. Two centres are linked by an edge in the graph if their Voronoi polyhedra have a face in common. Thus, the Voronoi partition of space, a froth of C cells, F faces, E edges, and V vertices, is the *dual* structure to the graph of atomic centres, with V^* centres, E^* edges, F^* faces and C^* cells, in that there is a one-to-one correspondence between elements of the froth and the dual graph

$$V=C^*,\ E=F^*,\ F=E^*,\ C=V^* \tag{11}$$

It also follows that coordination numbers of froth and graph are related by duality. The vertex coordination of the original graph is equal to the number of faces per cell of the froth. Usually, the dual graph has high vertex coordination on average; $z^* \simeq 14$ for three-dimensional packings, and $\langle z^* \rangle = 6$ exactly in two dimensions, which is just Euler's relation.

Apart from its essential simplicity the attractive feature about the Voronoi construction, when applied to a random array of centres (which statisticians call a Poisson point process), is the availability of some *exact* analytical results for some of its statistical properties (Getis and Boots 1979). Specifically, Gilbert (1962) has shown, *inter alia*, that the second moment of the area distribution, as defined in the previous section, is given by

$$M_2 = 0{\cdot}280 A_{\text{mean}}^2 \tag{12}$$

and other results are given by Gilbert (1962) and Meijering (1953) for the distribution of edge lengths.

The form of the distribution function $\phi(A)$ was conjectured by Kiang (1966) to have the form of the gamma distribution,

$$\phi(A) \propto A^{\nu-1} \exp\{-\nu(A/A_{\text{mean}})\} \tag{13}$$

and he found that computer-generated distributions were fitted quite well with $\nu = 4$. Note that for the corresponding *one*-dimensional problem, equation (13) gives the *exact* distribution function, with $\nu = 2$, as can be proved by elementary methods. There is no rigorous justification for it in higher dimensions. In later work (Kiang, personal communication), the value of ν for two dimensions was revised to be approximately 3·5.

Note that the second moment of this distribution is given by

$$M_2 = \nu^{-1} A_{\text{mean}}^2 \tag{14}$$

which is equal to $0{\cdot}25A_{\text{mean}}^2$, for $\nu = 4$ and $0{\cdot}286A_{\text{mean}}^2$, for $\nu = 3{\cdot}50$.

Note also that the empirical data of Crain (1972, 1978) shows that the mean area of n-sided cells is proportional to n for this pattern. (For further discussion, see Section 4.1.)

Sometimes we may wish to assign space to chosen centres in a less democratic manner than this. For example, this might be done in analysing the structure of a multicomponent metallic glass, a herd of different predators, or the work of a crooked electoral boundary commission. There are many ways of doing so. A particular example which is often cited is that of Cox and Agnew (1974) who drew up a 'theoretical partition' of Ireland into ideal counties (which they miscounted as thirty-one instead of thirty-two!) according to 'Reilly's Law of Retail Gravitation'. To the physicist, this seems a bizarre procedure and even the mathematics appears inconsistent. For better-defined procedures, see Boots (1980 b), who discussed boundaries such that (*a*) the boundary points have distances from the centres in proportion to their assigned weights (and form straight lines), and (*b*) the boundary points are equidistant from circles of unequal radii surrounding the centres. These boundaries are arcs of hyperbolas, not straight lines. Another possibility is provided by the 'radical axes' of such circles. The radical axis of two circles is the locus of points from which the *tangents* to the two circles have equal length (Gaultier 1813, Coxeter 1961). It is obvious that radical axes will meet in threefold vertices, but not so obvious that they are straight lines (Coxeter 1961, Ch. 6.5). Coxeter's proof strongly suggests that radical axes are the *only* possible straight-line partition of space between circles. It reduces to a Voronoi partition when the circles have equal radii. Radical partition of space has recently been used by Fisher and Koch (1979) and Gellatly and Finney (1982). Yet another procedure is that of the Johnson–Mehl model, in which the territories of points placed randomly in space *and time* grow radially at uniform rates until they impinge at boundaries (Johnson and Mehl (1939); see Getis and Boots (1979) for further discussion).

In some cases a Voronoi construction is made without any such refinements but the centres are then progressively readjusted. For example, we may wish to make each centre coincide with the centroid of its cell. This procedure has been used in ecology (see Section 3.7), and while iterative computer algorithms designed to implement it do converge, no relevant uniqueness theorems seem to have been proved for the resulting structure.

Yet another model can be generated by making the original points less random. An obvious way of doing this is to surround each point with a 'hard disc' and demand that these do not overlap. Only a certain density of discs can be deposited and the resulting pattern of points will be separated by a minimum distance (the diameter of the hard disc), giving a more regular Voronoi pattern than that of random points (Smalley 1966). This is shown in fig. 8. For a more recent reference, see Lotwick (1982).

2.3.2. *Minimal-surface models*

The Voronoi model is motivated by the idea of a 'territory' which belongs to a given point and is most obviously reasonable in applications in which such centres as a city or a den exist. A very different model can be constructed on the assumption that each cell is of some constant predetermined area (in 2D) and that the system is in equilibrium when the total 'surface energy' (really the total *length* of all cell edges) is a minimum,

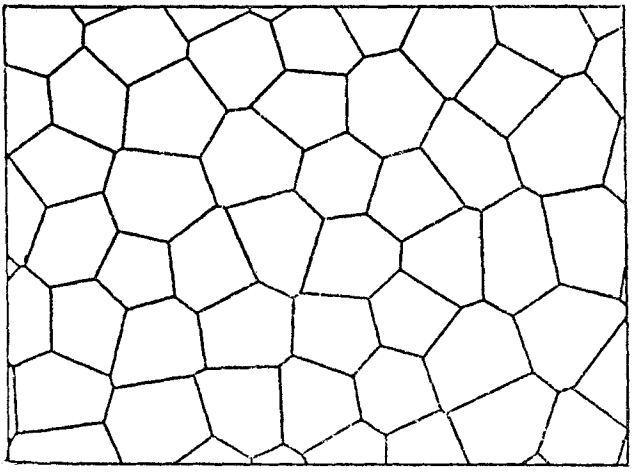


Fig. 8. Voronoi construction for centres of hard discs, at maximum density, after Smalley (1966).

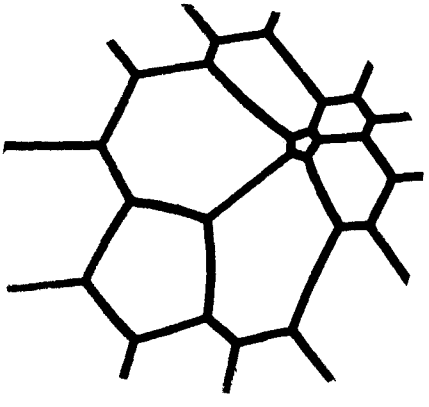


Fig. 9. Typical local configuration of cells in the minimum surface model (taken from the soap cell simulations of Section 3.2).

with respect to any small distortion. A two-dimensional *soap froth* (fig. 9) is just such a system in reality. We shall use the terms appropriate to the soap froth, but these really stand for quantities with more general meanings. Hence we write the total energy (really the energy per unit thickness) as

$$U = 2T \sum_{\substack{\text{edges} \\ i}} l_i \tag{15}$$

where T denotes surface tension and l_i is the length of cell edge i. For equilibrium, (*a*) all cell edges must meet at 120° at vertices, (*b*) each cell edge must be an arc of a circle, with a curvature related to the difference of pressures in the two neighbouring cells (see fig. 10), that is

$$\frac{2T}{r} = \Delta p \tag{16}$$

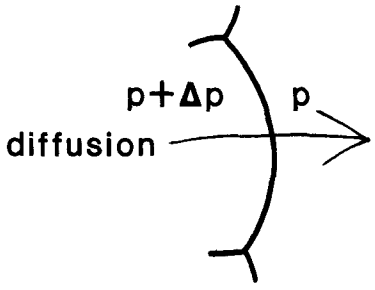


Fig. 10 Intercellular diffusion.

In addition the gas in the cells is treated as incompressible. Note that there is no claim that a structure which is found to obey these conditions for prescribed cell areas is *unique*—indeed it clearly is not. Other minima of surface energy can be found, with different arrangements of cells.

This model has always had a strong aesthetic appeal (Smith 1954, Pearce 1978) but apart from basic topological results, such as Euler's theorem, has not been the subject of much computational or mathematical work until quite recently. It is particularly interesting, because it can be given an internal dynamics, if we allow cells to transfer area to each other at a (slow) rate proportional to the product of the length of their common boundary and the pressure difference. This is what happens in practice with soap cells, or so it is claimed. From this may be derived (by simple arguments of geometry) Von Neumann's growth law which states: *the rate of growth of a cell is proportional to its number of sides minus six* (Von Neumann 1952). Thus many-sided cells grow, few-sided cells diminish and six-sided cells remain constant in area. The result is a trend towards a more and more inhomogeneous structure. We shall discuss all this in detail in the context of actual soap froths (in Section 3.2), because our understanding is still largely based on empirical results, but will note here the surprising conclusion that such a system may tend towards a *fractal* structure (Mandelbrot 1977) in the limit of infinite time! At this point we wish to stress the relevance of the model to other fields, of which metallurgy (grain growth) and ceramics (sintering) are the most obvious.

2.3.3. *Fragmentation*

The idea that a cellular structure is formed by some process of progressive fragmentation or cracking (as in the pattern formed by cracks or glaze—see Section 3.6) might motivate another area for exploration of the definition of ideal patterns, in terms of a hierarchy of cracks. However, this does not seem to have been investigated (but see further remarks in Section 4).

2.3.4. *Other* ad hoc *models*

Two other idealized models are worthy of mention here. Firstly, Thorpe (1983) has generated a number of patterns simply by imposing T1 processes (see Section 2.1) *randomly* on a hexagonal structure. A typical pattern is shown in fig. 11, used for model calculations of properties of disordered solids. Secondly, Shackelford (1982) has generated random two-dimensional networks by attaching units sequentially to a growing cluster, according to a simple computer algorithm. Figure 12 shows one such structure. This work was inspired by the original paper of Zachariasen (1932), advocating the random network model of glasses. Zachariasen's ideas were illustrated by means of two-dimensional analogous structures of just this type. Shackelford claims

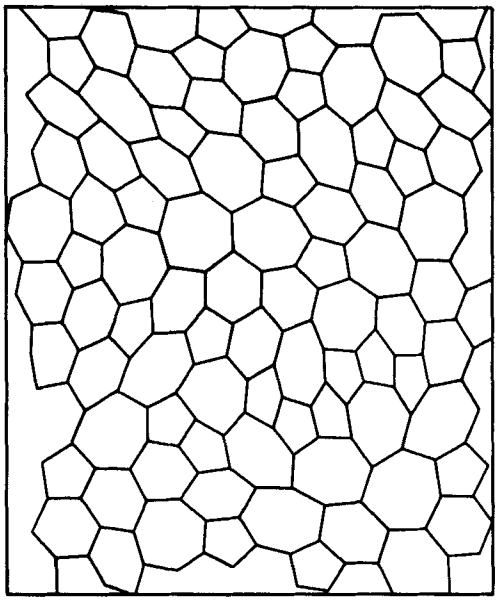


Fig. 11. Pattern produced by random T1 processes (Thorpe 1983).

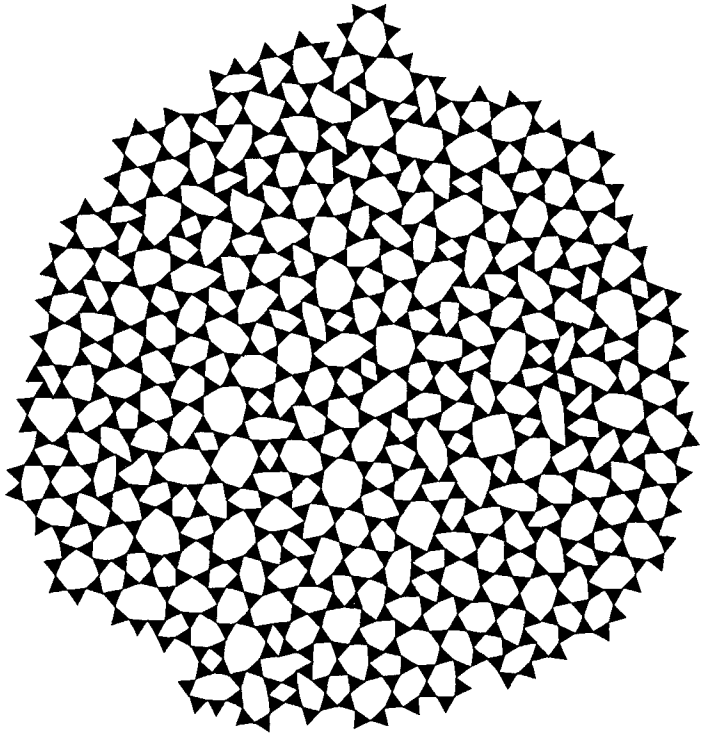


Fig. 12. Pattern produced by adding triangular units sequentially to a cluster (Shackelford 1982).

that the distribution function $p(n)$ for his patterns is best fitted by a *log-normal* distribution and he cites evidence for the widespread occurrence of such distributions in amorphous solids.

3. Examples of cellular structures

3.1. *Introduction*

"The time has come," the Walrus said,
"To talk of many things:
Of shoes—ships—and sealing wax—
Of cabbages—and kings—..."

In doing so, we shall begin with those systems with which a physicist would be most comfortable, and progress to the social and biological ones in which physical principles cannot be so straightforwardly applied.

3.2. *Soap froths in two dimensions*

A two-dimensional soap froth may be formed by squeezing an ordinary soap froth between two plates, or injecting bubles between them (Smith 1952, Aboav 1980). Our knowledge of the resulting system is largely based on the experiments of Smith (1952), who was originally motivated by its relevance to metallurgy (Section 3.3).

We shall assume, following Smith, that it corresponds to the 'minimal surface' model of Section 2.3, which seems reasonable, although one may be uneasy regarding the growth law that is mentioned there. Individual vertical films drain and thin progressively in quite a complicated way, presenting a problem interesting enough to attract Hooke, Newton and Gibbs (Mysels *et al.* 1959). Smith assumed that his soap films could be treated as being of equal and constant thickness so that Von Neumann's growth law (Section 2.3) applied. To our knowledge, this assumption has not been specifically checked, experimentally or otherwise, but we shall provisionally accept it.

Sketches made from some of Smith's photographs (Aboav 1980) are reproduced in fig. 13, showing the interesting progression with time to which we referred in Section 2.3. Smith's own conclusions, based on earlier photographs, were mainly qualitative. If we start from a structure which has relatively little disorder (small $\mu_2 = \sum(n-6)^2 p(n)$), it becomes more disordered (μ_2 increases). This is fairly obvious since larger cells will tend to have more sides and smaller ones less. As they change size, the process by which they lose or acquire sides is just that of fig. 2. Eventually, small cells reach the point of disappearance and so the overall scale, given by A_{mean} for example, begins to increase. Smith asserted that beyond a certain point this change of scale continued, but the increase of μ_2 did *not*. This suggests that the asymptotic behaviour of the system is (in an average sense) just a change of *scale*, with no change of the distribution of cell *shapes*. The conclusions was, however, based on very limited early data and, as we shall see, did not stand up to further scrutiny. This is an important point, since Smith's picture of the asymptotic behaviour is widely accepted and often quoted.

The further analysis of Smith's pictures was undertaken at a much later date by Aboav (1980). Aboav's main interest was in correlation of cell shapes and areas, as described below, so he did not stress the results which bear on the asymptotic behaviour. What he found was a continuing increase of μ_2 up to values ($\simeq 3$) far beyond that to which Smith thought it was converging (about $\mu_2 = 1{\cdot}5$). Moreover, after the initial period in which no cells are lost, there is a simple linear dependence of μ_2 and the average linear intercept d on time t. Also μ_2 and d are proportional to each other! Some of Aboav's results are shown in figs. 14–16.

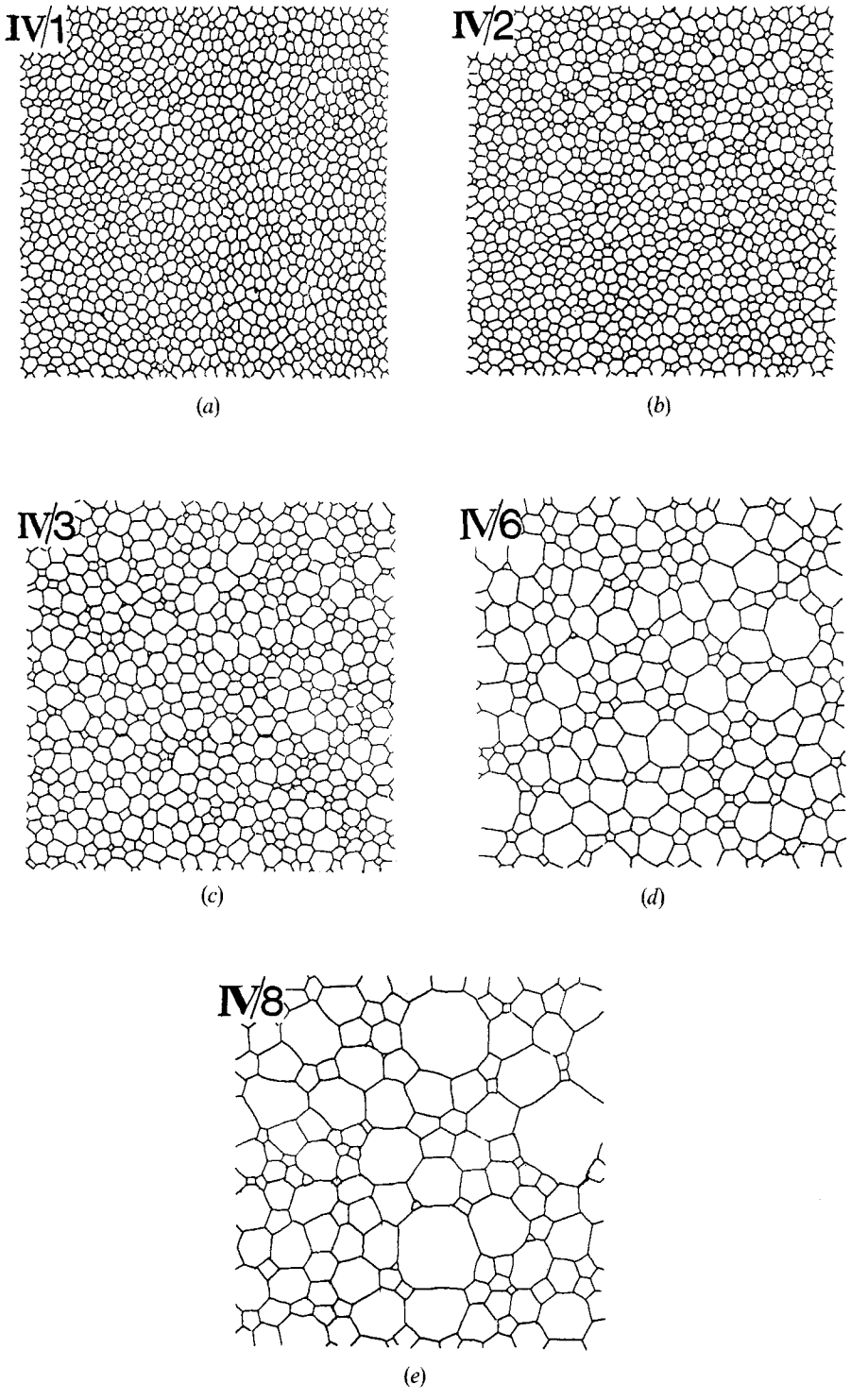


Fig. 13. Evolving cellular structure of soap cells at roughly 15-hour intervals, as observed by Smith (Aboav 1980).

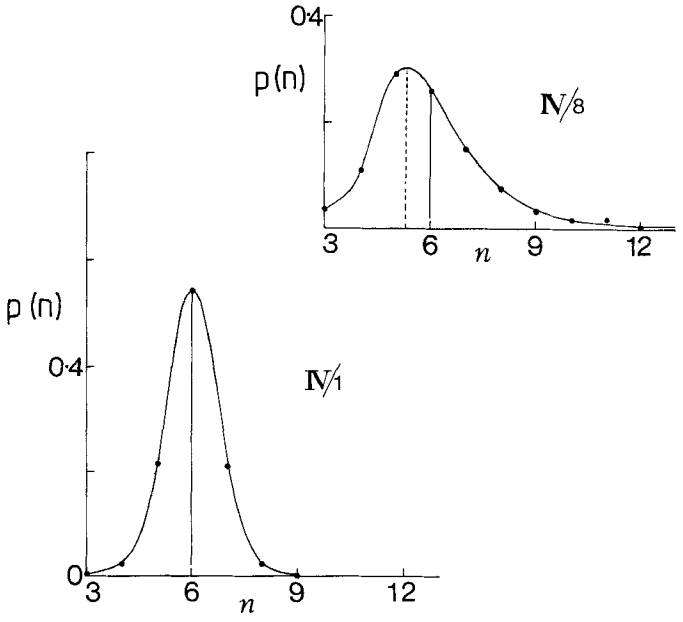


Fig. 14. Distribution function $p(n)$ for structures (a) and (e) of fig. 13.

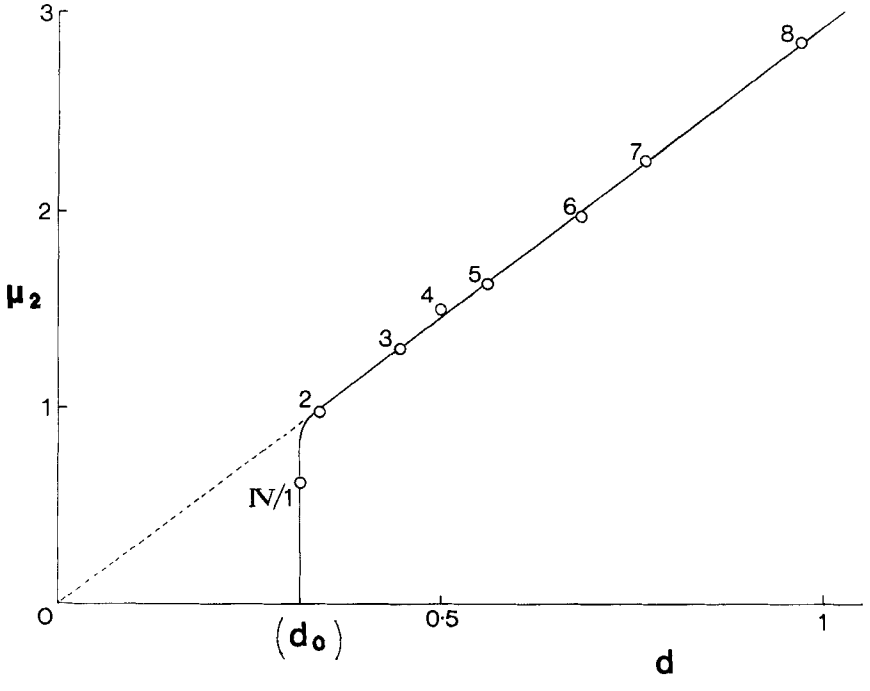


Fig. 15. Variation of μ_2, the second moment of $p(n)$ about its mean, with average linear intercept (cell diameter) d.

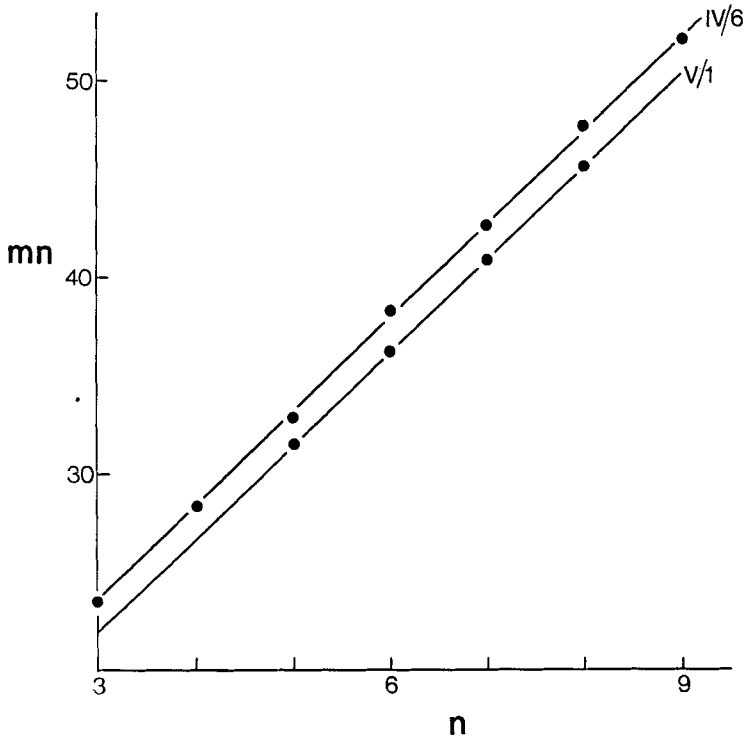


Fig. 16. Aboav's law is shown as a plot of mn against n (n is the number of sides of a cell, m the average number of sides of neighbours) for two different samples.

The linear dependence of μ_2 on t, on which we shall focus, is not clearly stated in Aboav's paper. Weaire and Kermode (1983 a) pointed out that it was a radical departure from the accepted picture. Aboav (private communication) confirms that further photographs show no sign of a change in this behaviour at later times, so it seems reasonable to conjecture that $\mu_2 \sim t$ is the correct *asymptotic* behaviour of μ_2, even though the published data go only some way towards its demonstration.

It should be remembered that a limiting structure with an *infinite* μ_2 is not an absurdity—all that is required is a distribution function $p(n)$ which falls off as $n^{-\alpha}$ where $2 < \alpha \leqslant 3$. In practice such a distribution could develop by the growth of the tail on the distribution toward higher values of n as time proceeds—indeed one can see such behaviour in fig. 14.

At the risk of piling conjecture upon conjecture, Weaire and Kermode (1983 a) suggested that the structure at large times might have a fractal nature (Mandelbrot 1977). What they had in mind was a structure in which cells could have patches of smaller cells at their intersections, and these could in turn have smaller cells, etc. Figure 17 conveys the general idea. Such a structure would be quite unlike anything else that we shall meet in this article, and well worth pursuing experimentally, even though this would not be easy.

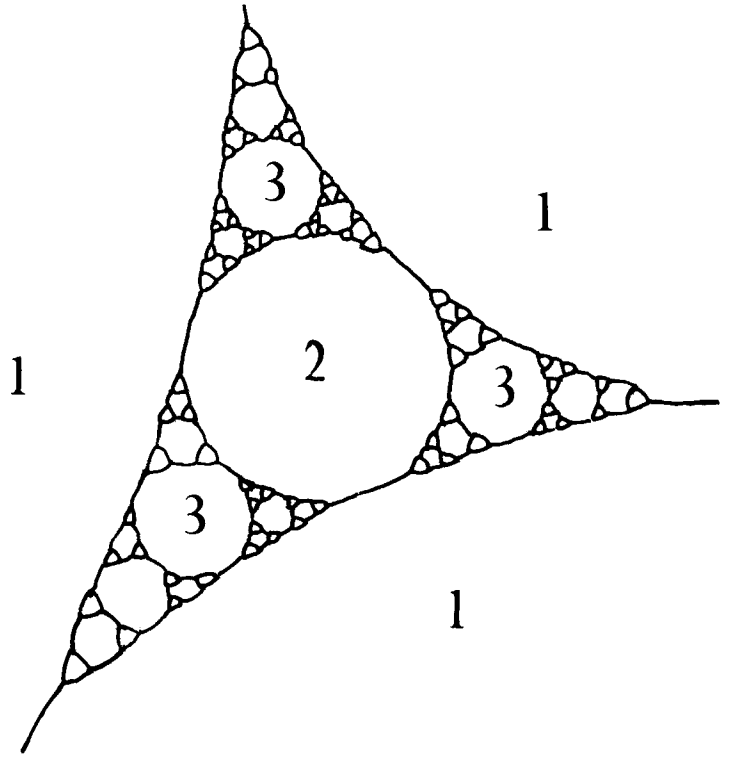


Fig. 17. Illustration of a fractal structure for the soap cell system.

As we have said, Aboav's main concern was with the correlation of neighbouring cells, for which he found the relation

$$m = 6 - a + \frac{6a + \mu_2}{n} \tag{17}$$

for the mean number of sides of cells surrounding n-sided cells, with $a \simeq 1{\cdot}2$ (see fig. 16).

Equation (9) has antecedents in Aboav's earlier work (Aboav 1970). An analysis of the grain structure (in section) of a polycrystalline MgO ceramic led him to the relation

$$m = 5 + 8/n \tag{18}$$

which he interpreted as implying some special arrangement of grains. Weaire (1974) criticized this notion, showing that, if one assumed that the angles at neighbouring vertices were uncorrelated, one could derive by a heuristic argument the relation

$$m = 5 + 6/n \tag{19}$$

which is close to (18). The argument is very simple. Firstly, we replace each cell edge by a straight line joining the vertices at its ends. If a cell has n sides, its average internal angle is $\pi - 2\pi/n$. The other angles associated with the cell vertices must have an average value $(\pi + 2\pi/n)/2$. These belong to neighbouring cells, for which we attribute the average value $2\pi/3$ to their *other* internal angles. Equation (19) then follows easily from the rule that the sum of the turning angles of a polygon equals 2π, applied to the neighbouring cells.

Alternatively, Euler's theorem $\langle n \rangle = 6$ may be applied to a local cluster of cells, consisting of an n-sided cell and its neighbours, to give the same result. The theorem is, of course, valid only in the limit of an infinite cluster, so this procedure is somewhat obscure, although pleasingly 'topological' in character. It was further pointed out by Weaire that there is an exact sum rule which $m(n)$ must obey, namely

$$\sum nm(n)p(n) = 36 + \mu_2. \tag{20}$$

This suggested the relation

$$m = 5 + \frac{6 + \mu_2}{n} \tag{21}$$

as a patched-up version of (19), compatible with (18) (which was for a sample with $\mu_2 \simeq 2$). Finally, Aboav's analysis of Smith's soap froth data, for which a linear relation also works, confirmed that μ_2 should be included as in (21), but the constants were modified, in a way consistent with (20), to give (17).

There thus emerges an interesting structural constant a, whose precise significance has so far escaped explanation. For example, Lambert and Weaire (1981) tried to derive (17) from consideration of a model based on random T1 processes.

Another derivation of Aboav's relation (21) has been recently given by Blanc and Mocellin (1979). They assume, as usual, that there is no correlation in cell shapes beyond nearest neighbours, and obtain the same recursion relation for $m(n)$ for all elementary transformations T1, T2, mitosis and their inverses. (This is no longer the case in 3D.) All these transformations can therefore occur independently of each other, at random in space or time, without affecting the statistical equilibrium of the structure. Aboav's relation is an equation of (the equilibrium) state.

Aboav's law can also be regarded as evidence of the correlation of angles at neighbouring vertices (see above argument). Perhaps this only restates the question, but it does suggest that $a > 1$ is an indication of the tendency to minimize distortion of the polygonal cells, to lower surface energy.

This seems a nice example of the digestion of a mass of statistical data to produce a single significant number characterizing any structure which obeys Aboav's law. We suspect the law to be rather general (recall that it was originally found for a ceramic, and also see Section 3.3). However, Boots (1981) has pointed out that it does *not* hold for the Voronoi polygon model. For further discussion see Lambert and Weaire (1983).

In addition to the experimental results, some further information is available from computer simulations (Weaire and Kermode 1983 c) and more may soon be forthcoming. A computer program has been developed which (*a*) relaxes a given starting configuration to equilibrium, with fixed cell areas, and (*b*) transfers small increments of area between cells, to simulate the growth behaviour, according to Von Neumann's law. This kind of work would be practically impossible without the help of modern graphical output facilities.

Note that, in addition to adjusting vertex coordinates and pressures (and hence curvatures), the program must cope with the T1 and T2 process defined in Section 2.1, i.e. cell rearrangements and disappearences. The T1 process is particularly interesting, because the local configuration makes a sudden change to a quite different one as soon as a side vanishes, with a corresponding decrease in energy.

Figure 18 shows a sequence of structures at roughly equally spaced times, generated in this way, using periodic boundary conditions (Weaire and Kermode 1983 c). It shows

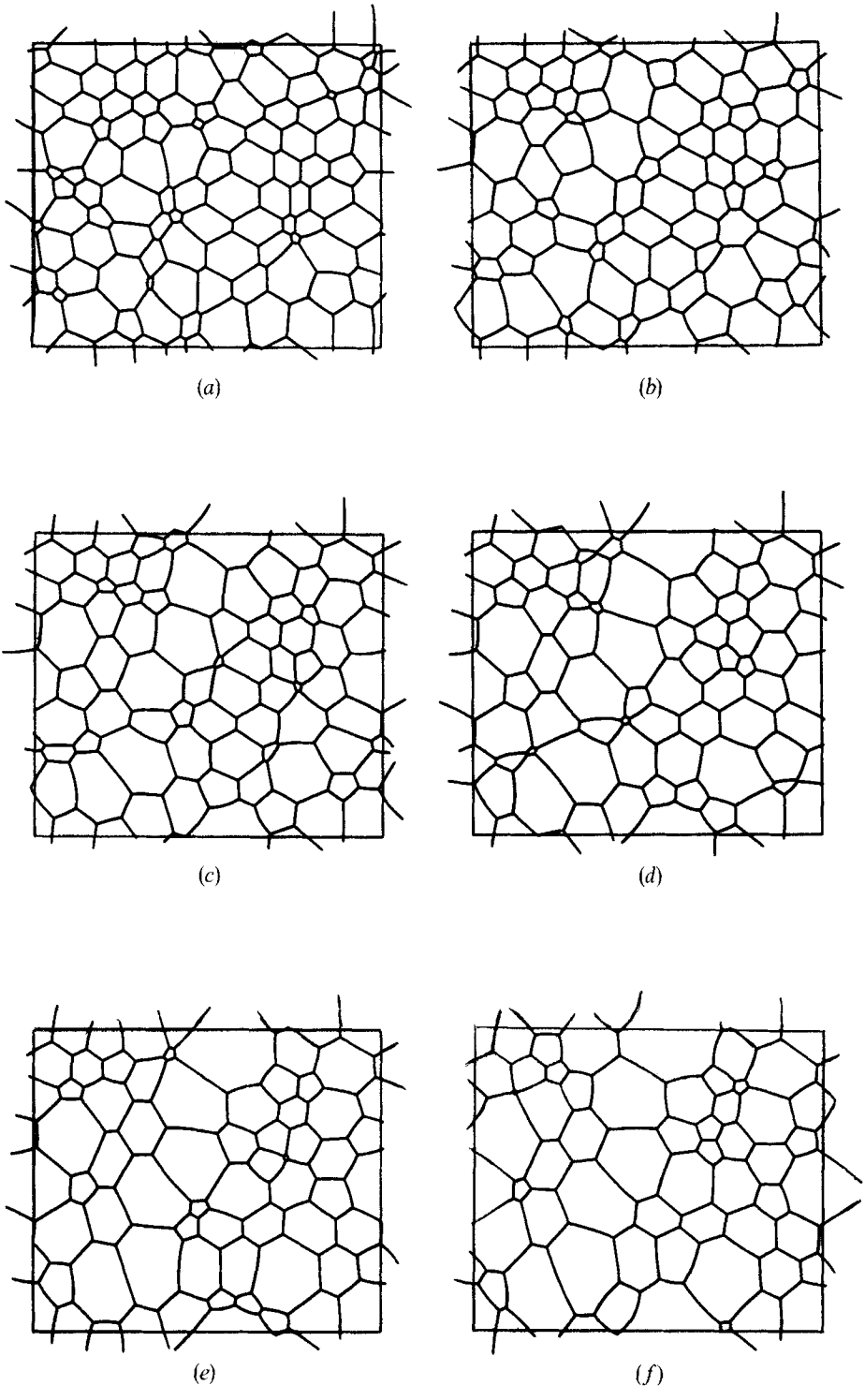
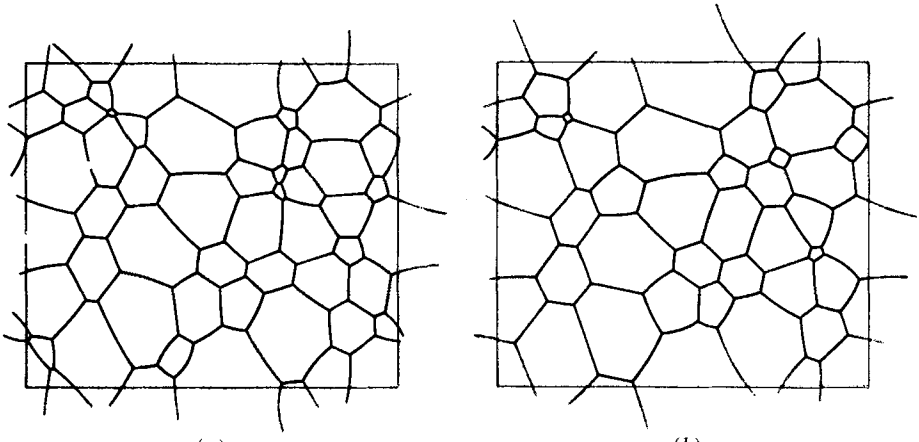


Fig. 18. Sequence of simulated soap cell structures at roughly equal time intervals (continued on facing page).



(*g*) (*h*)

Fig. 18 (continued)

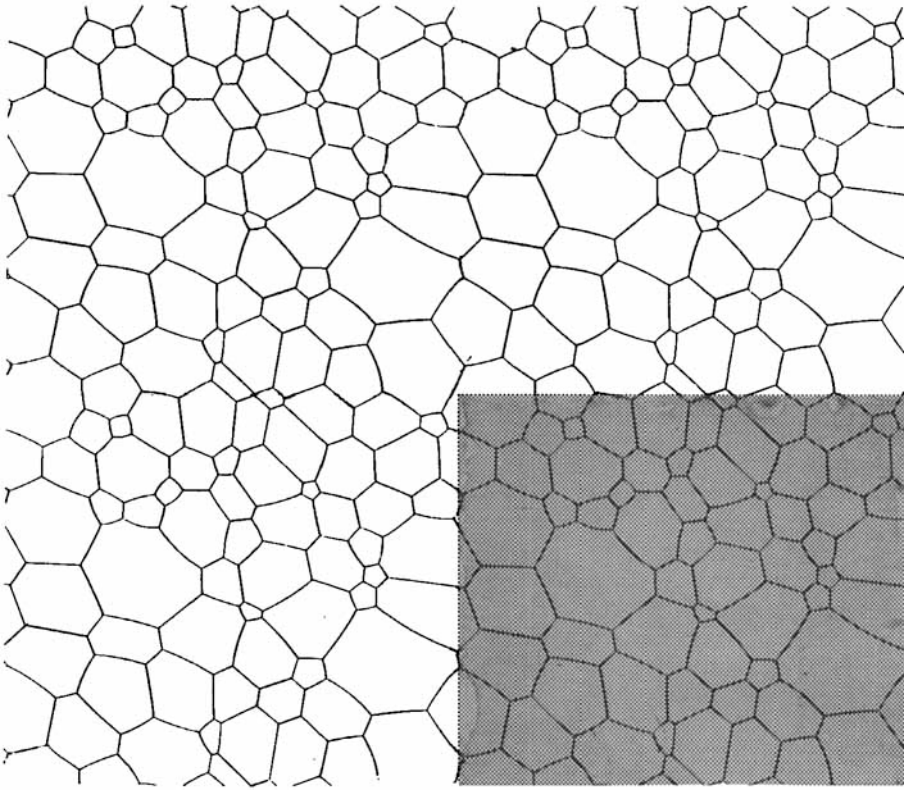


Fig. 19. Periodic boundary conditions are used in these simulations, so that each side of the sample matches the opposite one as shown.

the same steady progression towards higher μ_2 as in the experimental data (although it only gets as far as $\mu_2 \simeq 1{\cdot}5$ before the number of cells is unacceptably low). In this and all other respects, including Aboav's law, the simulated system resembles the real soap froth system (Weaire and Kermode 1983 c).

In all of this work, *periodic* boundary conditions are used, as illustrated by fig. 19. The choice of an initial structure is an awkward one and further work is in progress to test alternatives and to find out how quickly the memory of a given starting configuration is lost in the chaotic development which ensues (Kermode and Weaire 1984).

Ashby and Verrall (1973) have made two-dimensional emulsions which are closely analogous to this system, and have studied their response to stress. The T1 processes, which are increasingly provoked at large strains, play a large role in the plastic behaviour of the system. This behaviour too can be simulated (Kermode and Weaire 1984).

Princen (1983) has recently given an analysis of the rheology of foams and emulsions, including the effects of finite film thickness.

Before leaving soap films, we should note that *three*-dimensional soap froths have long been a subject of interest, being of direct importance in chemical engineering. Analogous principles are certainly at work in this case (small cells shrink and disappear, etc.) but available data is limited. Experimental work does not seem to have progressed beyond that of Matzke (1946), who determined distribution functions for cells. There is a suspicion that the bursting of cell faces contributes significantly to the coarsening of the structure, and that gravity plays a significant role in its evolution.

Simulation of the three-dimensional system presents some intimidating problems, but should be possible.

3.3. *Grain structure in metals and ceramics*

The grain structure of a metal or ceramic is another example of a space-filling cellular structure (fig. 20). In thin films, this has a two-dimensional character. In both the three-dimensional and two-dimensional cases, the structure which is observed in relatively pure single-phase metals is similar to the soap froth. This similarity was

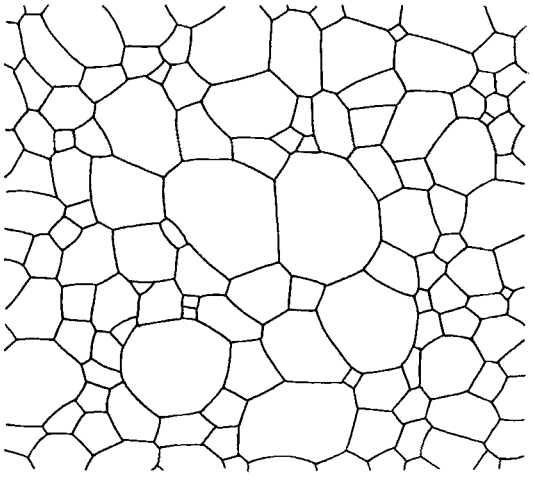


Fig. 20. Section of polycrystalline MgO (Aboav 1980, Aboav and Langdon 1969).

especially emphasized in the work of Smith (1952, 1964) and was the original motivation for his work on the two-dimensional soap froth, discussed in the previous section. It is, of course, no coincidence, since there will be a *surface energy* associated with grain boundaries, and the grains will tend to readjust to lower this energy. The transfer of atoms between grains, in order further to reduce the surface energy (which is precisely what drives the growth process in the previous section) will lead to *grain growth*, an important metallurgical process (Martin and Doherty 1976). For a nice example of grain growth in a two-dimensional (thin film) system, see Guy (1971).

However, on closer inspection, it is difficult to see the precise relationship between the two problems, even though it is very commonly invoked. First, there is the question of crystalline anisotropy, which will make surface energies dependent on the orientation of the boundary. This is generally neglected in theories of grain growth (Martin and Doherty 1976), as is sub-grain coalescence (Byrne 1964). However, there are further questions regarding the various mechanisms of atomic diffusion which contribute to grain rearrangements and grain growth—diffusion within grains, diffusion along grain boundaries and the process of transfer across a grain boundary. A little thought will convince the reader that the situation is really quite different from that of the soap froth in which a *fixed volume* of *gas* is enclosed by each cell. The fact that this has zero shear modulus means that the soap froth is always in equilibrium, to within a very good approximation, and growth is controlled entirely by diffusion through cell walls. In the case of the metallurgical grains, it is not clear that the problem really separates so neatly into equilibrium and growth. To be fair, Smith's original work really offers only a loose analogy. He goes no farther than to say that, "Normal grain growth results from the interaction between the topological requirements (of space filling polyhedra) and the geometrical needs of surface tension equilibrium" (Smith 1952).

There is a substantial body of literature in theoretical metallurgy devoted to grain growth, including the two-dimensional case. The facts to which it is addressed appear to be as follows (Cahn 1974, Aboav 1971). At constant temperature, the mean grain diameter varies with time according to $D \sim t^n$ where n lies between $\frac{1}{3}$ and $\frac{1}{2}$. However, at a given temperature T the grain growth process stops when this diameter reaches a value given by

$$D^{1/2} = C(T - T_0) \tag{22}$$

where C and T_0 are constants. The latter effect is generally attributed to the effect of inclusions (analogous to the pinning of dislocations).

A series of papers addresses the question of the growth law. These include Hillert (1965), Louat (1974), Rhines and Craig (1974) and Kurtz and Carpay (1981). The last two papers are especially interesting in that an attempt is made to use topological properties. Kurtz and Carpay's work is particularly thorough and based on a number of clearly stated premises regarding statistical quantities. It is too detailed to review here but we shall comment on some aspects. Firstly, *log-normal* distributions are assumed for various distributions $p(n)$ in two dimensions. Some experimental evidence for this is given, but the theoretical justification which is offered seems somewhat circular. Shackelford (see Section 2.3) has also suggested that this distribution is the appropriate one for $p(n)$, in another context. Kurtz and Carpay's work also contains interesting ideas on the relationship between three-dimensional cellular structures and the two-dimensional structures obtained by sectioning them. This has long been a key problem in metallography and elsewhere.

All of these theories are based on questionable assumptions and are inadequately tested by the available data. In the circumstances, recent work by the Exxon group (Sahni *et al.* 1983, Srolovitz *et al.* 1983 a, Anderson *et al.* 1983, Srolovitz *et al.* 1983 b) should be the best guide to further progress. This uses Monte Carlo simulation techniques to obtain a number of statistical results. The classic growth law $D \sim t^{1/2}$ favoured by most of the earlier work is *not* found. Instead, the index is approximately 0·38, which is just as consistent with experiment, according to these authors. The discrepancy may seem small but it poses fascinating questions regarding scaling arguments for this problem (Weaire and Kermode 1984).

Another topic which bears on the soap froth model is *superplasticity*. Superplastic alloys can be plastically deformed to very large strains without fracture. In the process, grains are rearranged locally in the manner of the T1 process of fig. 2 (Ashby and Verrall 1973). This also occurs when a 2D soap froth is strained, as remarked in the last section, but here the analogy must be even looser than in the grain growth problem (Kermode and Weaire 1984).

Finally, it should be noted that grain growth in sintered ceramics is thought to be essentially the same process of that in metals, and is well described by Coble and Burke (1963).

3.4. *Other domain structures in materials science*

Other types of two-dimensional cellular patterns are observed in a variety of materials. For example, fig. 21 shows domain structure in a film of As_2Se_3 glass (Chen *et al.* (1981).

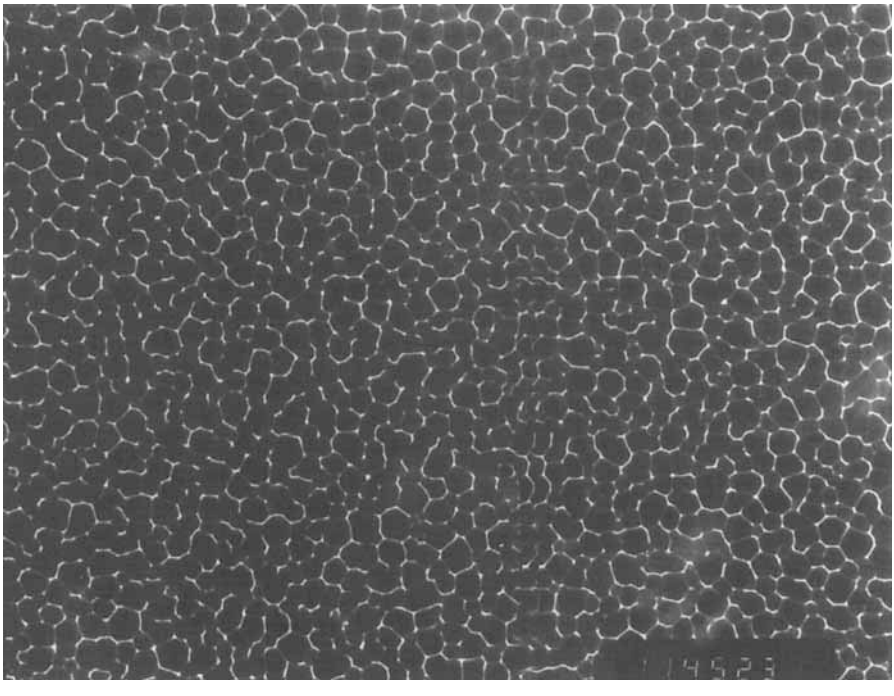


Fig. 21. Domain structure in a thin film of As_2Se_3 glass (Chen *et al.* 1981).

Such patterns, which are testimonials to the refinement of electron microscopy, are usually not sufficiently well defined to invite the kind of statistical analysis which might help in classifying them and explaining the underlying mechanisms. However, in the case of the As_2Se_3 glass, the domains are very well resolved and have very recently been analysed as shown in figs. 22 and 23 (Chen *et al.* 1983).

Other examples, similar but less well resolved, are to be found in the pattern of segregated In in In-implanted single crystal Si (Cullis and Joy 1981), (thin film) amorphous Ge (Donovan and Heinemann 1974), hydrogenated amorphous silicon

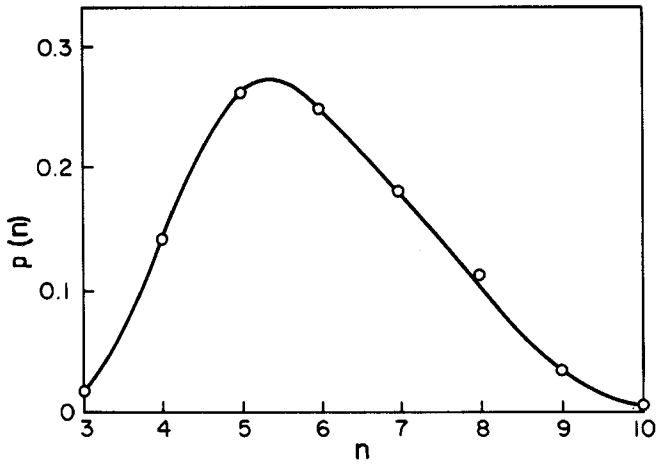


Fig. 22. Distribution function $p(n)$ for the structure of fig. 21. Cf. fig. 14.

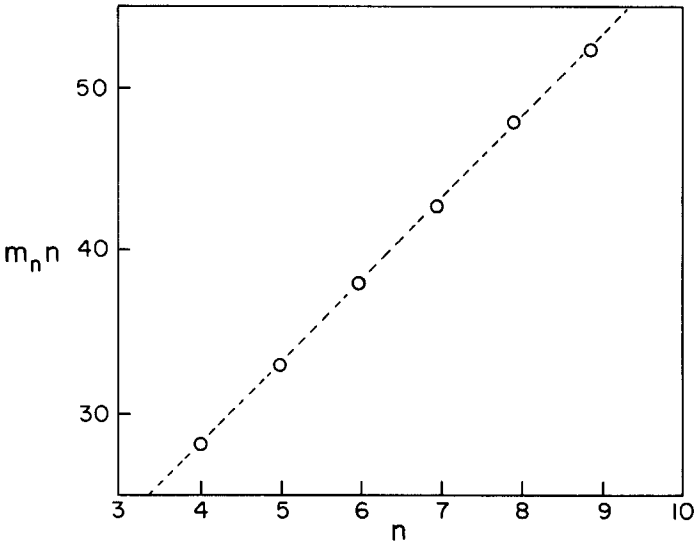


Fig. 23. Test of Aboav's Law for the structure of fig. 21. The straight line is that defined by (17), for $\mu_2 = 2$ and $a = 1{\cdot}1$. Cf. fig. 16.

(Messier and Ross 1982), and various cases of crystal growth instability, analogous to Bénard patterns (Kirkaldy 1966, Langer 1982). We should also mention, in passing, the formation of cellular arrangements of dislocations under plastic strain (Konig and Blum 1980), and the classic experiments of Leduc, described by Thompson (1942), on cellular structures formed by diffusion.

3.5. *Convection cells*

Bénard–Marangoni convection cells, which form under certain conditions when a liquid is heated uniformly from below, usually form a structure close to a perfect hexagonal arrangement. While Bénard (1901) may have been the first to study them systematically, they were reported by Thompson in the nineteenth century (Thomson 1881). They might well be left out of our list of interesting *random* cellular structures, were it not that we shall want to make reference to them elsewhere. In any case, random patterns are sometimes formed. In particular, if the lower surface is heated well above the threshold for convective instability, the hexagonal structure 'melts' (Occelli *et al.* 1983). For a modern discussion of the mechanism of formation of such a structure, see Whitehead (1975).

3.6. *Geological structures*

Geology is concerned with many random structures, formed by faults, phase separation in minerals, etc. Of these, one particular example closely resembles some of our other cellular patterns. This is the structure of *columnar basalt*, a familiar feature in volcanic areas all over the world but usually exemplified by the Giant's Causeway in Northern Ireland for historical reasons. Certainly, fig. 24 must stand as a very early example of scientific interest in a random network.

Concerning the Giant's Causway. Prolixity in a Philosophical Description I'm sure you'l pardon. Thus wrote Sir Richard Bulkley (1693) in an early volume of the *Philosophical Transactions of the Royal Society*, and there has been much prolixity on the subject ever since. It continues to be an inevitable topic to be lightly touched upon in textbooks of structural geology; but the critical reader is likely to be more intrigued than edified by much that is offered by way of explanation. Part of the problem is that many writers have not actually examined the structure and fall easily into the habit of referring to it as 'hexagonal', which it is not—even Bulkley noted that there were 'pentagones'. In fact there are columns of as few as four and as many as eight sides, in an irregular but remarkably perfect network (fig. 25). Had more emphasis been placed on this disorder, the the early idea that the structure had something to do with crystallinity would not have persisted, as it did in some quarters for nearly two hundred years. Other suggested mechanisms included the compaction of cylindrical bodies and, in more modern times, convection cells (Section 3.5). Most of these are reviewed by Spry (1962). In fact, the now accepted basic mechanism was stated by Desmarest (1771) at the time of the French Enlightenment, namely, that the columns were formed by the cracking of deep lava flows upon solidification and cooling. However, credit for this idea is sometimes ascribed to Mallet (1875) who put an attractive mathematical gloss upon it. Mallet's article was rejected by the Royal Society after five months and four referees, but the *Philosophical Magazine* did well to accept it—it has been cited continually ever since. We need not really concern ourselves with it here, because Mallet idealized the structure as hexagonal, and moreover, his calculations are very difficult to interpret or accept.

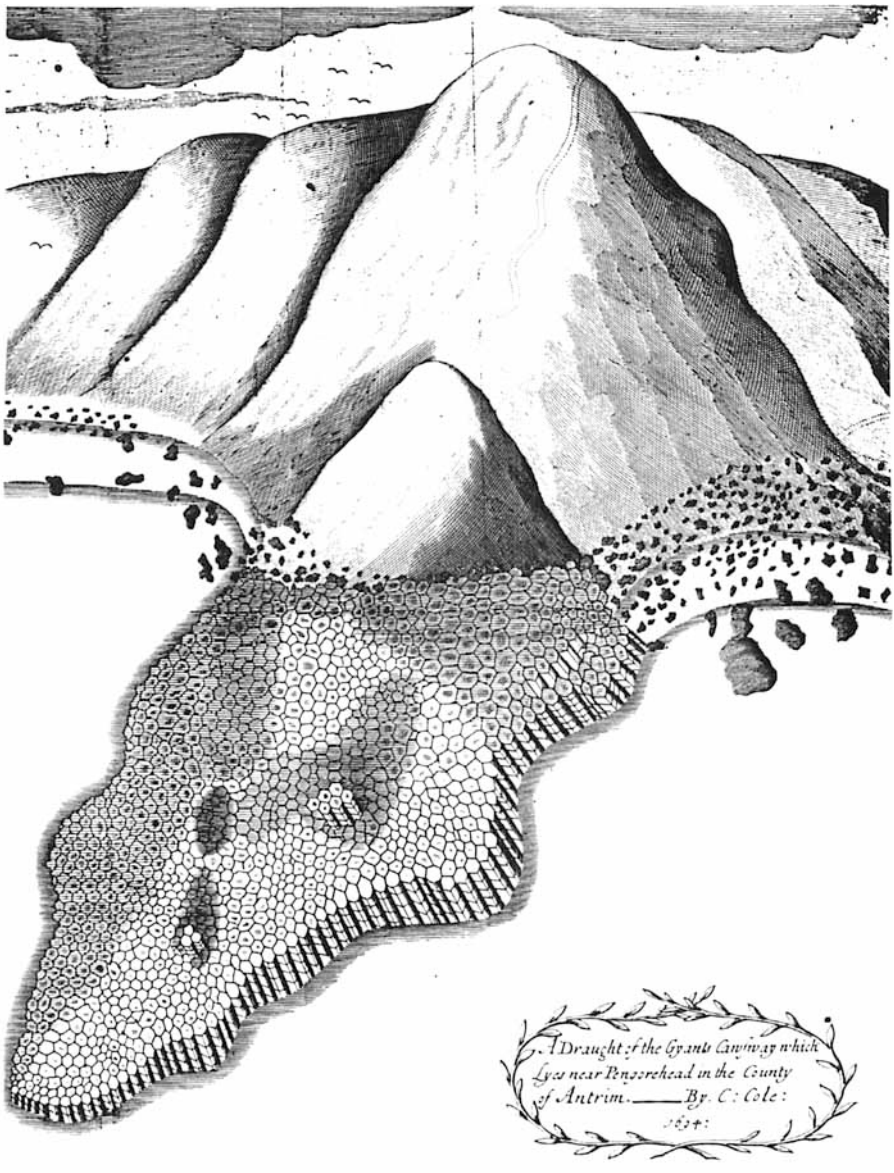


Fig. 24. Sketch of the Giant's Causeway from an early volume of the *Transactions of the Royal Society* (Foley 1694).

In more recent times the notion that cracks must *propagate*, rather than appearing simultaneously, has become familiar, so that the network of cracks is pictured as forming by some process of propagation and bifurcation (Thompson 1942, Lachenbruch 1962). However, it is difficult to see how horizontally propagating/bifurcating cracks could organize themselves to this extent. Even if they did bifurcate regularly at roughly 120° angles, the accidental confluences of the randomly propagating cracks would surely produce a much less coherent structure than is observed. It would have much in common with the kind of structure shown in fig. 26, which is found in glazes and dried mud. Mud or 'gumbo' patterns do sometimes contain a proportion of 120°

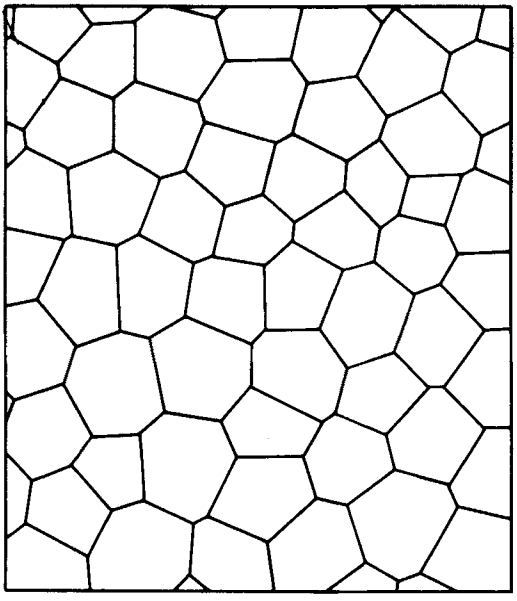


Fig. 25. Accurate map of a small part of the Causeway, taken from O'Reilly (1879).

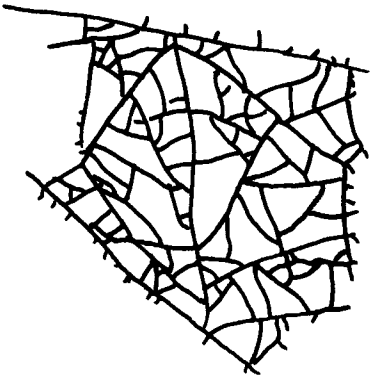


Fig. 26. Typical cracking pattern (mainly T-junctions) in a ceramic glaze.

vertices, but nothing like the overwhelming proportion of these in the basalt columnar structure.

Some idea of the primitive state of our approach to two-dimensional random patterns may be conveyed by the fact that when interesting patterns were observed by a space probe on Europa, rough comparisons were made with $p(n)$ for the Giant's Causeway, in the search for an interpretation (Pieri 1981).

The only work which attempts to model the details of the structure is that of Smalley (1966). His model is in fact just the Voronoi construction for hard discs, described in Section 2.3. Only a vague comparison of the two patterns is made, in terms of $p(n)$, which contains very little information, being quite narrow ($\mu_2 \simeq 0{\cdot}7$). In any case the motivation which is offered for the model, in terms of randomly placed 'stress

centres', seems difficult to justify. While the comparison is interesting, it seems to offer nothing to our understanding.

Weaire and O'Carroll (1983) have suggested that the solution to this dilemma must lie in the *vertical propagation* of the cracks, as the interior of the lava flow slowly cools. The cracks will tend towards a dynamically stable arrangement, and it is suggested that it is this which is on view in locations such as the Giant's Causeway. They also examined a detailed survey and map of part of the Causeway made by O'Reilly in the 1870s (O'Reilly 1879). This was subsequently forgotten or disregarded, so that several others made partial recounts in later years. It can be used to carry the analysis of the structure beyond a mere tabulation of $p(n)$. O'Reilly's compendious data is most valuable, despite his misguided interpretations (he clung to the tradition of crystallinity). A surprising correlation shows up between lengths of sides and the neighbouring angles. This is

$$\theta = 90 + 30LL_{\text{mean}}^{-1} \quad \text{(degrees)} \tag{23}$$

which is the result of a regression analysis of O'Reilly's data (Weaire and O'Carroll 1983). Its significance is unknown but we suspect it to be related to the condition for dynamical stability mentioned above.

Thus we still lack a complete answer to the questions raised by Bulkley in 1693. Fracture mechanics is still a rather sketchy subject so it may be a long time before this beautiful pattern is the object of an equally beautiful theory.

Finally, we should mention that somewhat similar but rather less perfect patterns are formed by 'ice wedge polygons' in permafrost (Lachenbruch 1962).

3.7. *Territories in ecology, geography and economics*

Ecologists, when they turn their minds to the spatial distribution of species or competing groups, are soon concerned with random cellular patterns. The relatively new subject Locational Geography often deals with similar problems. Even when they study patterns of points (such as cities or nesting birds) ecologists and geographers find the idea of cells of dominance or influence attractive. The Voronoi construction (Section 2.3) is generally invoked, in one form or another. While such territories are usually somewhat notional, in at least one case they are visible (fig. 27) (Hasegawa and Tanemura 1976, Barlow 1974). In fig. 28 we show the results of the procedure mentioned in Section 2.3.1. by which a completely random Voronoi structure was

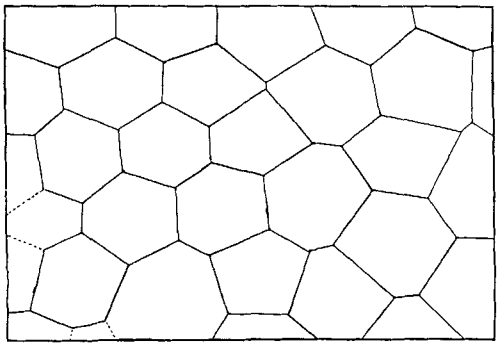


Fig. 27. Territories of mouth-breeder fish (Barlow 1974, Hasegawa and Tanemura 1980).

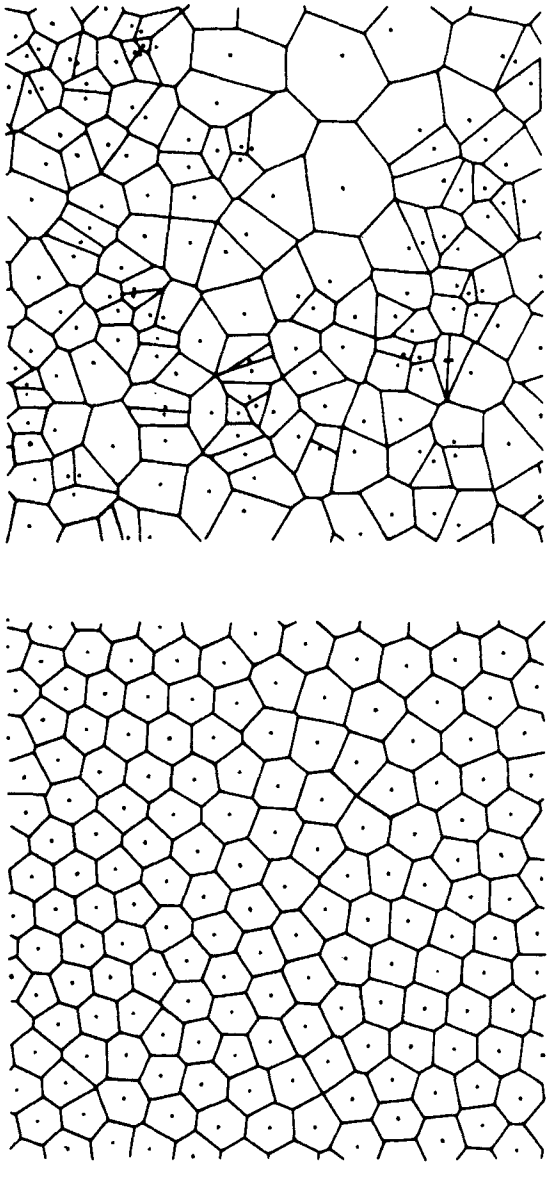


Fig. 28. Upper figure: ——, Random Voronoi structure. Lower: ——, same structure, after relaxation to place each point at the centroid of its cell.

relaxed to make each point the centroid of its Voronoi cell (Tanemura and Hasegawa 1980). Further consideration of mouth-breeder fish, nesting eagles, pectoral sandpipers, etc., might take us too far from the territory of this journal. Equally, we shall not dwell on the geographers' work on county and parish boundaries, although it is worthy of note that a modification of Aboav's law (Section 3.2) has found its way into such areas (Boots 1980 a, c).

For further details and many references, the book by Getis and Boots (1979) may be consulted.

3.8. *Biological cells*

It has been memorably asserted that "Order breeds habit, disorder breeds life"—although we have forgotten by whom! Here at last, in the consideration of biological cells, we reach the most intriguing of our cellular structures but, as we shall explain, we face an anticlimax.

Again, we shall find it convenient to stay in two dimensions, confining our immediate attention to cells in surfaces or stems, which have an essentially two-dimensional structure, for example, see figs. 29 and 30. In many biological specimens, the pattern is greatly complicated by differentiation, just as metallurgical specimens generally exhibit more than one phase. However, it is not difficult to find examples

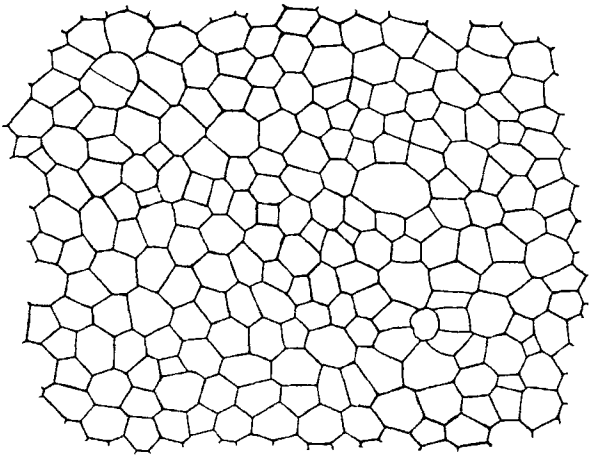


Fig. 29. Cell structure in the skin of a cucumber (Lewis 1928). $\mu_2 = 0{\cdot}8$.

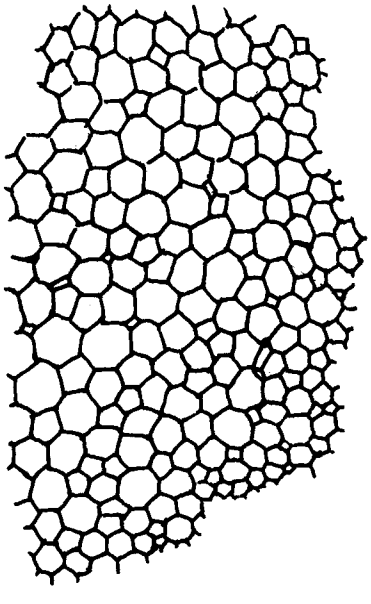


Fig. 30. Cells in the stem of a plant (after Krommenhoek *et al.* 1979).

where there are large areas of undifferentiated cells. Such cells have long been vaguely compared with the other random cellular structures which we have noted.

What principles dictate the arrangement of these cells? This question was asked by the American botanist Lewis from the 1920s (Lewis 1925, 1928, 1931, 1933, 1943 a, 1943 b, 1944, 1949). For example, he compared the structure of biological cells in two dimensions with that of an emulsion (fig. 31). However, Lewis did not really take his mathematical or statistical analysis very far, and it was extended only to a modest extent by Marvin (1939) and Matzke (1950). The questions which he left unanswered seem to be almost entirely ignored today. Given the scope and sophistication of modern biology, this is somewhat surprising. What little has been done on this subject is reviewed by Dormer (1980) but it is mainly concerned with the eternal verities of the relevant mathematics, such as Euler's Theorem. "So long as a system is mathematically determinate there is no room for any biological phenomenon to show itself. Biology begins at the point where there are two or more mathematically admissible results, between which the organism must choose upon some basis other than geometrical necessity. We have to recognize and eliminate from further discussion aspects of tissue structure deriving from pure mathematical necessity" (Dormer 1980, pp. 5–6).

Lewis's early work did, in fact, turn up a surprising relation which we can place beside Aboav's, as possible general features of wide classes of cellular structures. He found that, for various two-dimensional cellular patterns, the mean area $\bar{A}(n)$ of n-sided cells varies *linearly* with n. This relation does not follow from any elementary

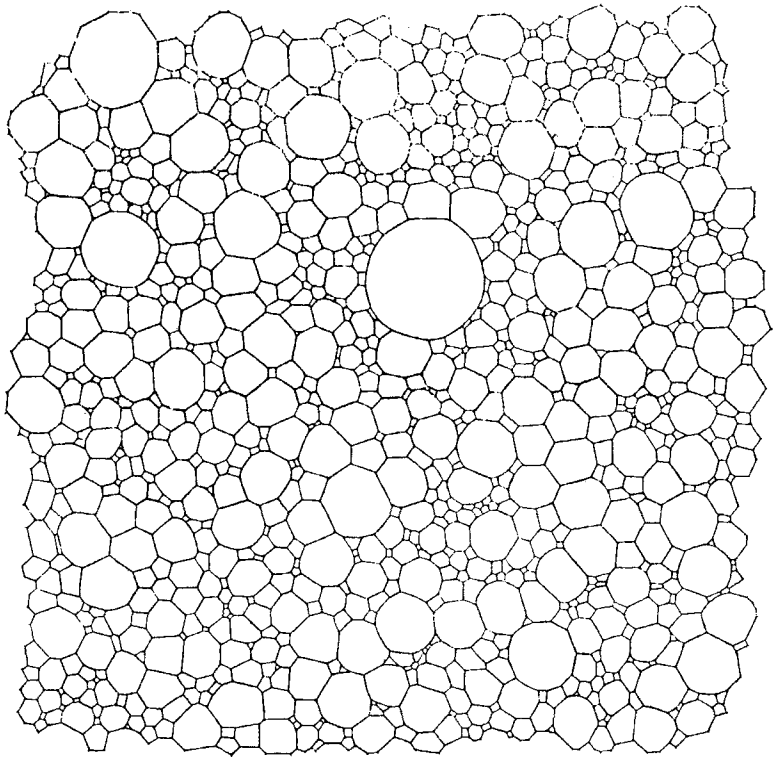


Fig. 31. Cells formed by droplets of photographic emulsion, studied by Lewis as a model for biological cells.

consideration of geometry. Rivier and Lissowski (1982) have suggested an explanation, which we shall discuss in Section 4. For the moment, it is sufficient to note that the Voronoi construction for random points (Section 2.3) also obeys this rule. For the biological systems, the intercept $\bar{A}(n)=0$ is in the range $n=1$–3, while for the Voronoi construction it is approximately $n=0$ (Smolyaninov 1980, Rivier and Lissowski 1982).

A continuation of Lewis's work would surely be most valuable. Perhaps in the end the conclusion may be that the detailed statistical properties of the cell structure have no particular biological importance (apart from disorder itself). This would seem to be the assumption of cell biologists at present. Experience with other, better understood, systems gives one cause to wonder if this can be so. As D'Arcy Thompson said, quoting Aristotle, "Nature does nothing in vain".

Recall what happens in the case of the soap film system. A structure made up of only six-sided cells is *stable*, but any departures from this topology create an instability which feeds the continual evolution of the soap froth. Order breeds habit, disorder breeds life ... Of course, the growth mechanism is quite different for biological cells, involving a combination of mitosis and T1 processes rather than T1 and T2 processes (Section 2.1). Incidentally, Bénard convection cells sometimes undergo a process similar to mitosis (Whitehead 1975).

Specifically, we offer the following comment on Dormer's methodology quoted above. A random cellular structure in statistical equilibrium (the most probable distribution of Section 4) is mathematically determinate, yet its cells can be distributed in an overwhelmingly large number of configurations all consistent with statistical equilibrium. Cells can even undergo transformations (T1, T2 and mitosis) which, in two dimensions, do not affect statistical equilibrium even locally. Such a structure constitutes a fertile soil for any biological phenomenon to manifest itself. The diagnostic potential of statistical crystallography is yet to be exploited.

There is obvious biological value in disorder in a growing tissue—it smoothes out what would otherwise by a clumsy, stepwise growth process, located on selected, dividing cells. At the very least, dislocations (pentagon–heptagon pairs) screen the strain imposed by isolated pentagonal or heptagonal cells, and their glide enables the tissue to grow by successive mitoses under little elastic distortion, and from several cells at the same time.

Rather different considerations, having to do with optics, may favour disordered structures in the retina (Yellott 1983) and it is notable that the hard disc pattern (Section 2.3.1) recurs in attempts to simulate the pattern of receptors.

With these few tentative remarks we leave the problem of biological cell statistics for future study. In our opinion, the botanist and the metallurgist, like the farmer and the cowboy, should be friends. Recently developed techniques of data acquisition, developed for the analysis of grain structures, might well also find application to cell structure.

4. Finding order in chaos

4.1. *Maximum entropy?*

All of the work that we have touched upon in Section 2 suffers from the lack of an established methodology—it is a story of a blind search for significant correlations, without any guiding principle.

There is one school of thought that claims to have found such a principle. Loosely speaking, it is claimed that the methods of statistical mechanics, rephrased in terms of

information theory, can be carried over into a much more general field than equilibrium thermodynamics. The guiding principle is that of Maximum Entropy. In this section we shall comment briefly on this and certain other arguments which are of a similar nature. They are likely to remain controversial for some time, simply because "the weakness of the Maximum Entropy Formalism (MEF) is that it does not contain within itself any criteria for its valid application" (Oppenheim 1980, Landauer 1981). However, we shall argue that our subject of random space-filling structures is as good as, if not a better, field of application for the MEF than equilibrium thermodynamics, and show that empirical laws like Lewis's can be derived by using it. There are certainly no other proofs on the market at the moment.

The Maximum Entropy Formalism (Jaynes 1957 a, b, 1979) is first and foremost the solution of the inverse problem of probability theory: given some prior knowledge of the system, for example, frequencies of a few outputs or some averages, find the probabilities of further outputs. Apart from this specific knowledge, included as 'constraint', the MEF solution is maximally non-commital. It is the solution with minimal prejudice, and it is this variational requirement which grants uniqueness to an otherwise under-constrained problem. Most of the controversy which has been voiced in the literature has to do with the definition of probability as frequency or as the mathematical description of a particular state of knowledge. It is a 200-year-old battlefield, summarized delightfully by Jaynes (1979, p. 56): "One of the major unsolved riddles of probability theory is: how to explain to another person exactly *what is the problem being solved?*" It does not affect the status of MEF as the solution of a well-defined problem, but only expresses doubts of its validity for the description of real, physical or biological situations.

Surely, the reader may interject, *experiment* should be the final judge. But experiment does not measure probabilities, only frequencies (of n-sided cells for example). In a homogeneous random structure, it turns out that the expectation value of the frequency *is* numerically equal to the probability, regardless of any correlation between cells (see, for example, Jaynes 1979, p. 50). Above all, experiment is there to verify or disprove the predicted relations between averaged quantities, the equations of state in thermodynamics, or Lewis's law in random structures. There lies the predictive or diagnostic power of the MEF: "If it can be shown that the class of phenomena predictable by maximum-entropy inference differs in any way from the class of experimentally reproducible phenomena, that fact would *demonstrate the existence of new laws of physics*, not presently known." (Jaynes 1957 c, p. 172.)

Examples of such arguments are to be found in the work of Brostow and Rogers (1983) on fragmentation, Kikuchi (1956) and Tanemura (1979) on soap froth, and Rivier and Lissowski (1982) on Lewis's law. The following is the gist of the latter authors' justification of Lewis's law.

We begin with the remark that all the structures described in Section 3 are roughly similar. They are therefore unlikely to depend on the particular physical, biological or chemical forces which govern their respective constituting materials, apart from their single length scales. Moreover, were short-ranged physical forces dominant in their architecture, they would give rise to an ordered structure in two dimensions; triangular or hexagonal packing, interfaces at 120°, all this is conducive to regular arrangements, like the bees' honeycomb, possibly with a few, well-separated defects like dislocations. The very fact that *random* structures do occur in two dimensions, suggests that specific short-ranged forces are less important in framing the structure than the inescapable, mathematical and universal constraint of space-filling. Indistinguishability of struc-

tures, originating from utterly different areas of Natural Philosophy, strongly supports our prejudice.

The question now is: do these inevitable, mathematical constraints frame in any specific, identifiable way the structures generated under their sole or overriding influence? The answer is yes, and it is provided by the word 'random'. In statistical mechanics, it is well known, although a source of surprise to every undergraduate, that, for large systems, the most probable distribution is overwhelmingly more probable than any other. This is associated with the problem of the 10000 monkeys at their typewriters and the chance of any one of them typing some Shakespeare. Everyone has a definition of 'never', or a price against it, and, less excitingly, every one knows the meaning of 'almost always', or a most probable realization. Consider a large random, space-filling mosaic. Is there a most probable distribution of cells, a most probable structure? If so, is there a criterion for such ideal random structure, analogous to the ideal gas law in thermodynamics? The answer is yes in both cases.

To summarize: because all the structures are, to a first approximation indistinguishable, because they are random, and because their respective, single length scales are so widely different, possible relevant constraints are pruned down to the few (2) mathematical identities pertaining to the space which the cells are filling. These two identities (equations 26 and 27 below) are the constraints of the MEF. Their generality and inevitability in turn grant universality to the (set of most probable) structures. If the argument is circular, it is certainly self-consistent!

One looks for an ideal, random space-filling structure determined solely by the mathematical constraints and the fact that the structure is the most probable one. The structure is in statistical equilibrium, in that any topological rearrangement of the cells leaves its 'arbitrariness', invariant. The arbitrariness is measured precisely by the entropy or information contained in the structure (Shannon 1948, Pierce 1961). If such an ideal structure exists, it is the solution of a statistical problem and can only be the representative archetype of an ensemble of structures. It is not unique. Accordingly, criteria for ideality will be relations between average properties of the structure, like the ideal gas law in thermodynamics, rather than geometrical data like Bragg diffraction spots. One is looking for the statistical analogue of the simple cubic structure in crystallography.

At the lowest order, the statistical problem is set as follows. The random variable is the topological shape of a cell, the number n of its sides in two dimensions. There is indeed, a variety of cell shapes in a random mosaic. A two-dimensional random mosaic is described by the probabilities p_n of finding an n-sided cell. The statistics or correlations between shapes of neighbouring cells is also important, but represent a more detailed and sophisticated description of the structure, neglected at this level. A random mosaic with many cells (thermodynamic limit) will take up the most probable distribution of p_n that maximizes the entropy, arbitrariness or information

$$S = -\sum p_n \ln p_n \tag{24}$$

(Shannon 1948, Pierce 1961, Jaynes 1957), subject to known constraints.

In complete generality, the constraints are

$$\sum p_n = 1 \qquad \text{(normalization)} \tag{25}$$

$$\sum p_n \bar{A}_n = A_{\text{mean}} \qquad \text{(space-filling)} \tag{26}$$

$$\sum p_n n = 6 \qquad \text{(Euler, topology)} \tag{27}$$

Here $\bar{A}_n$ is the average area of an n-sided cell, FA_{mean} the total area available to the F cells in the mosaic. The topological constraint is an immediate consequence of Euler's relation and of the structural stability of the mosaic (all vertices have coordination $z=3$). The corresponding restriction in three dimensions is much weaker. The averaged number of faces per cell is not fixed, but only related to the average number of edges per face, n, by

$$\langle f \rangle = 12/(6-\langle n \rangle) \tag{28}$$

This is all. The problem, as formulated, is entirely mathematical: the concepts of statistical equilibrium (most probable distribution) and entropy or information, and the constraints themselves, are all represented by mathematical expressions. Physics, and biology, are absent at this level, and the resulting structures are expected to be universal. The only subjective step is in the *coding* of the structure by the sole parameter n, and the requirement that it is in statistical equilibrium. (See also Delessert 1972.)

It is unnecessary to evaluate the entropy S. For our purposes, the following argument suffices. The constraints are a linear system of equations between p_n, so that the smaller the dimensionality of the space of constraints, the larger the space of possible solutions p_n, and the more probable one such solution will be. To find the most probable distribution, we must first reduce as much as possible the space of constraints, that is, make them linearly dependent.

$$\bar{A}_n = A_{\text{mean}}\lambda[n-(6-\lambda^{-1})] = A_{\text{mean}}\lambda(n-6)+A_{\text{mean}} \tag{29}$$

The average area of an n-sided cell, $\bar{A}_n$, is linearly related to the number n of its sides. This relation was actually suggested empirically by Lewis (Section 3.8) and is obeyed by Voronoi froths generated from randomly distributed centres (Crain 1978).

The arbitrary parameter λ in (29) is the undetermined multiplier involved in the linear combination between constraints. (The other multiplier is eliminated because the system of equations 25–27 is inhomogeneous.) It is clearly related to the slope and intercept of Lewis's law, and is therefore an important descriptive parameter of the structure. But Lagrange undetermined multipliers have a habit in thermodynamics and in mechanics of possessing a physical meaning of their own. They are not merely arbitrary mathematical factors. What, therefore, is the meaning of λ? It has been further suggested that it is related to the ageing of the structure (Rivier 1983 a).

A full calculation using MEF confirms the above. It is interesting in that it takes the formalism one step further than Jaynes (1957 a, b), by allowing the constraints themselves (the function $\bar{A}_n$, whose average constitutes the space-filling constraint (26)) to be adjusted to increase further the maximal entropy. It also yields the most probable distribution (Rivier 1983 b).

$$\left.\begin{aligned} p_n &= N(n-n_0)^3 \exp(-\gamma n), \quad n \geqslant a \\ &= 0, \qquad\qquad\qquad\quad n < a. \end{aligned}\right\} \tag{30}$$

Here N is the normalization factor, and γ is adjusted so that the topological constraint (27) is satisfied, while $a(>n_0)$, the smallest number of sides per cell, is an arbitrary parameter in the distribution, although experimentally $a \geqslant 3$. Alternatively, one can entertain all n, with the additional constraint that $p_n=0$ for $n<a$, a situation already discussed by Shannon (1948), which enters naturally into the general formalism of

Information Theory. Distribution (30)† is in general agreement with experiment (Crain 1978, Lantuejoul 1978, Blanc and Mocellin 1979, Smolyaninov 1980).

By analogy with the successful application of MEF to equilibrium thermodynamics (Jaynes 1957 a, b), its present application implies that random cellular structures are also in a state of statistical equilibrium, with Lewis's law as equation of (equilibrium) state. MEF yields a more general definition of such statistical equilibrium than does microreversibility under elementary transformations. Statistical equilibrium and Lewis's law are consequences of the balance between entropy (most probable distribution) and lowest level of organization (space-filling territorial partition, encoded by the constraints).

4.2. *Conclusion*

In 1973 there took place the world's first Symmetry Festival (Senechal and Fleck 1977). Nobody has yet suggested that there should be an Asymmetry Festival—it does not have the same immediate appeal, although perhaps Science still lags behind Art in that respect ...

The field lacks coherence, as this article, despite our best efforts, may have demonstrated. Yet it continues to fascinate those who delve into it, and their numbers are growing. The sheer tedium of data-gathering has in the past been an inhibiting factor. Today's instrumentation and computational techniques for pattern recognition

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† The reader may require an outline of the derivation (Rivier 1983 b):

Cells are characterized by shapes (n) and sizes (area A) through the joint distribution $p(n, A)$. Correlation between shape and size,

$$\int \mathrm{d}A p(n, A) A = \bar{A}_n \int \mathrm{d}A p(n, A) \tag{31}$$

is included as an additional constraint, secured by Lagrange multipliers $\lambda(n)$. Absence of correlation corresponds to $\lambda(n) = 0$. The other constraints are (25)–(27), with $p_n = \int \mathrm{d}A\, p(n, A)$.

The entropy must include a geometrical, *a priori* probability $p_0(A) \propto A^{\alpha}$, to find a cell of area A, thus

$$S = -\sum_n \int \mathrm{d}A p(n, A) \ln [p(n, A)/p_0(A)] \tag{32}$$

This is manifest in the case of Voronoi polygons, where $\alpha = 2$ ($\alpha = D$ in D dimensions), because a cell of area A requires at least 3 points within $\sim 2A$ of its seed, for which the probability goes as $A^2 \exp(-\rho A)$ (Poisson distribution). The same geometrical prefactor ($\alpha = 2$) should hold regardless of the type of mosaic, unless triangular cells are forbidden ($\alpha = 3$) or two-sided cells allowed ($\alpha = 1$), for some specific (biological, etc.) reason.

Maximizing the entropy (32) subject to the constraints yields the most probable distribution

$$p_n \propto \bar{A}_n^{\alpha+1} \exp(-\lambda_1 n - \lambda_2 \bar{A}_n), \text{ with } \bar{A}_n \propto \lambda(n)^{-1}$$

given by equation (31). λ_1 and λ_2 are the Lagrange multipliers securing topological (27) and space-filling (26) constraints, respectively.

If the size–shape relationship $\bar{A}_n$ can be adjusted to increase further the entropy, there are two alternatives: either Lewis's law (29) and distribution (30), or $\bar{A}_n = A_{\text{mean}}$, independently of n (no correlation between sizes and shapes of cells) and exponential distribution p_n. The entropy has two disjoint maxima in configuration space $(A_{\text{mean}}, \langle n \rangle, \bar{A}_n)$.

The exact Voronoi distribution in 1D (13) is recovered from this formalism. In general, the exponential decay of $\phi(A) = \sum_n p(n, A)$ is controlled by the space-filling constraint (26).

(Serra 1982) could soon change that, just as graphical output has made computer simulation more feasible. Given more data and more extensive simulations, together with some interdisciplinary interaction, we could see emerging theories of random structures of which we have only fragmentary clues and tentative ideas today.

Even if progress is made in the area which we have mapped out in this article, there will remain the complexities of three-dimensional structures to be sorted out. Perhaps our rapidly developing computer graphics facilities will also make this much easier in the near future.

Theoretically, statistical techniques based on Information Theory or the Maximum Entropy principle have only recently begun to be applied to random structures, whether, as here, to obtain the distribution of random variables and the equation of state for statistical equilibrium, or as the best strategy in direct pattern recognition (Bricogne 1983). However, the growing confidence in the universality of random cellular structures, described by a few simple laws (such as those of Lewis and Aboav) must be credited to the careful and painstaking work of early experimentalists like Lewis, Marvin, Matzke and Smith. Should we regard the publication of a paper on gauge theory on a random lattice (Christ *et al.* 1982) as a certificate of respectability for the Walrus? (cf. 3.1).

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