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Structure of Random Cellular Networks

N. Rivier

Blackett Laboratory, Imperial College, London SW7 2BZ

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The methods of statistical mechanics are applied to the structure of random, space-filling, cellular structures (foams, metallurgical grain aggregates, biological tissues). Microreversibility of the structural properties under elementary transformations is demonstrated. Maximum entropy inference under a few constraints yields structural equations of state, relating the size of cells to their topological shape. These equations of state classify the structures, and are criteria for their randomness. They serve also as reference, from which deviations can be associated with specific constraints.

INTRODUCTION

This paper investigates the form, shape and the relative size of the cells of random, undifferentiated biological tissues, of the grains of metallurgical aggregates, or of the bubbles of a soap froth, in short, of random space-filling cellular networks. These structures are, at first glance or lowest level of discrimination, indistinguishable even though they originate from local building forces which are very different. These specific forces are therefore less relevant in determining the structure than the inescapable mathematical constraints of filling Euclidean space, in the least biaised, most probable fashion.

Here are some examples of identical structures (see also Weaire & Rivier 1984):

Compare two undifferentiated biological tissues, like those (the wing of a fly and a crab apple) adorning the frontispiece of Dormer's (1980) book. Apart from a scale factor (the average cell size), they are indistinguishable, meaning that they can be interchanged without anyone noticing the substitution, even though the two tissues are not exactly superposable. Identity of the two tissues is a topological, rather than geometrical equivalence.

Similarly, the polycrystalline sample of aluminium, shown in the last figure of Cyril Stanley Smith's (1954) essay, is, in all aspects, indistinguishable from a soap bubble froth, and very nearly so from biological tissues.

These structures all belong to a class, or ensemble of random, space-filling cellular tissues. I shall explain here why they appear identical, and which observable criterion on the form of their cells labels them as member of that class of random aggregates.

The most accessible random cellular network is a soap bubble froth. One notices immediately that the size of interfaces between bubbles and of the bubbles themselves is not fixed. It fluctuates in space and in time as if the medium partitioned by the bubbles was made of deformable rubber. Moreover, small bubbles have a small number of (usually large) neighbours, and large bubbles, small neighbours, a spatial correlation which is topological, in that it refers to the shape of the objects making up the structure, rather than their size (which is a fluctuating quantity, only relevant on average). The whole froth is then homogeneous, not in the sense of exact superposition of a pattern with its translated counterpart (as in crystals or in elementary geometry), but because any local difference is not objectively (without using words like "I", "here", "this", or "now") relevant or observable. The biological paradigm is undifferentiated tissue. In material science, glasses and amorphous materials are indeed homogeneous in this objective, but non-metric fashion. And so is a forest in which one has got lost. (An entirely different experience altogether from getting lost in an orange grove - the crystalline or ordered counterpart, with metric generative homogeneity and exact repetition of a pattern ad infinitum).

Botanists, forty years ago, also discovered that undifferentiated tissues had similar architechture to soap bubble froths, and to compressed lead-shot (Matzke 1950, Lewis, 1943), even though the local physical force shaping these two physical models (surface tension versus hard core repulsion) is very different. The situation has been neatly summarised by F.T. Lewis, (1943) as "random avoidence of the niceties of adjustment".

Randomness of the structure, the presence of continuous transformations, and the fact that two different structures can be deemed identical without being superposable, imply that the relevant geometry is topology (rather than metric geometry of unit cells or Bragg peaks). Hence the actual size of edges, cells or angles is of no relevance, since they can expand or shrink as space is continuously deformed. Each structure is not an exact copy of a unique original, but only a member of a statistical ensemble of most probable structures under a few constraints (constant volume and topology of the space which they are filling). Physical or biological forces are irrelevant, to lowest order, in framing that identical and random architecture, which has an equation of state - (Eq. 5,6,9). Randomness also implies that "defects", i.e. shape fluctuations (e.g. topological dislocations - pentagon-heptagon pairs in 2D tissues -, disclin-

ations, or odd lines in 3D (Rivier 1979)), are essential constituents of the structure. It is their presence and their motion under topological transformations, which grant froths, foams, ceramics or tissues, their relevant mechanical, geometrical or growth properties. (Weaire & Rivier 1984).

STATISTICAL MECHANICS OF RANDOM TISSUES

The methodology of statistical mechanics follows a succession of steps:
(1) Definition of microscopic states or configurations, and elementary transformations between different configurations.
(2) Detailed balance or microreversibility of the macroscopic structure under elementary transformations. This is a prerequisite for statistical equilibrium.
(3) Statistical equilibrium is characterized by a probability distribution for the occurrence of microscopic configurations, and by an equation of state, which enables us to classify the various macroscopic structures.

Random tissues are as suitable subjects for the application of statistical mechanics as, for example, physical gases. First of all, the tissue contains a large number of elements (cells) with several possible microscopic configurations (shape, and, possibly, size of the cell). Different microscopic configurations are related directly by elementary transformations (Fig 1). The macroscopic configuration of the structure is characterized by the probability distribution for various microscopic configurations, and by averages of, and spatial correlation between microscopic parameters. The macroscopic configuration of the structure is invariant under elementary transformations of Fig 1, thereby establishing detailed balance or microreversibility (Aboav's law).

Microreversibility is a prerequisite for statistical equilibrium, which can be obtained unambiguously by maximizing the arbitrariness, or entropy of the structure, subject to a few obvious constraints (fixed volume, topology, and, possibly, constant energy). This expresses simply the fact that the system is so large and so loose that the macroscopic configuration with maximal arbitrariness can be realised by many more microscopic configurations than any other, and is therefore overwhelmingly the most probable. But this maximum entropy formalism can be derived rigourously from probability theory (Jaynes, 1957, 1979). It is the least biaised solution of the gambler's (underdetermined) problem: from a few given data, find the distribution of probabilities.

Statistical equilibrium leaves an observable signature, the equation of state, which is a relationship between average microscopic parameters. (Boyle- Mariotte law in ideal gases, for example). In ideal random tissues, the equation of state is Lewis' (1928) relation between average shape and size of a cell (Eq. 5). The equation of state classifies random tissues, which belong to classes or ensembles, each with its own equation of state or its own set of constraints under which entropy is

maximized. For example, metallurgical grains, whose interfaces carry energy, obey the perimeter law (Eq. 9), rather than the area law of Lewis (Eq. 5) which is characteristic of ideal tissues without relevant energy constraint.

Statistical equilibrium yields also the probability distribution for microscopic configurations. This constitutes more detailed information on the structure than the mere equation of state (Maxwell-Boltzmann distribution of velocities in ideal gases, for example). Distributions of sizes and shapes of cells are obtained in Rivier (1985). Experimental distributions can be found in Weaire & Rivier (1984), or in Smoljaninov (1980), and those obtained by numerical simulations, in Srolovitz et al. (1984).

This is a summary of the programme and of some results. The rest of this paper will fill in some details.

GEOMETRY OF TISSUES, ELEMENTARY TRANSFORMATIONS

Random tissues are cellular structures filling a topological space, where edges, faces and cells can shrink or expand continuously as if they were filling rubbery space. This implies a grammar:

1. Elementary Structural Transformations. In 2D, neighbour switching (T1) and cell disappearance (T2) and its inverse (mitosis, which may consist of iterated inverse T2 and T1) (Fig 1). In 3D, switching between neighbouring faces (seen by blowing gently on a soap film formed on a cubic frame) (T1), face disappearance (change of cell neighbourhood), and cell disappearance (T2) and their inverses. If interfaces are planar, and in the absence of symmetry, neighbour switching always requires a face to disappear (Fortes 1985).
2. Structural Stability: Only vertices with coordination z = 4 (in 3D) are structurally stable. Vertices of higher z can be transformed into these by infinitesimal deformations. A tissue or froth with only structurally stable constituents is called maximally random. This will be assumed from now on.
3. Conservation Laws: (Tissue with C cells, F faces, E edges and V vertices, occupying a volume Ω). Euler theorem: $F - E + V = 1$ (2D); $-C + F - E + V = 0$ (3D); continuity of odd lines (lines threading through faces with odd number of edges (3D) (Rivier 1979). (These identities are clearly invariant under elementary structural transformations).

Consequences: The topological random variables are n, the number of edges on a face, and f, the number of faces on a cell. Their expectation values

$$\langle n \rangle = 6 \quad (2D) \ , \ \langle f \rangle = 12/(6 - \langle n \rangle) \quad (3D) \qquad (1)$$

are consequences of the above. They are topological restrictions or constraints on the distribution of shapes of cells. The 3D relation is valid for every cell and for the froth as a whole. For maximally isotropic and isochorous (equal volume) cells,

<f> = 13.4 (exhibited in tetrahedrally close-packed, crystalline structures like A15 or Laves phases (Sadoc 1984)), corresponding to <n> = 5.1, the number of regular tetrahedra sharing one edge in Euclidean space. <f> increases if the cells become more anisotropic; it decreases if fluctuations in their volumes increase (Rivier 1982).

TOPOLOGICAL CORRELATIONS AND DETAILED BALANCE

Denote by m(n) the average number of edges on faces neighbour to a n-sided face in the 2D tissue, and by $m_f(n)$, the same quantity in the 3D tissue, with all faces belonging to the same, f-faceted cell. m (n) represents correlation between shapes of neighbouring cells. m(n) and $m_f(n)$ obey the same recursion relation (Eq. 2-3) if the tissue undergoes any one of the elementary structural transformations or their inverses (Blanc & Mocellin 1979, Rivier 1985). This is a statement of microreversibility or detailed balance: Elementary transformations can occur independently in space or time, without affecting statistical equilibrium and the properties (correlations, etc.) of the structure.

The solution of the recursion relation is Aboav's law (1970),

$$m(n) = 5 + (6 + \mu_2)/n \qquad (2D) \qquad (2)$$

(first justified by Weaire), where $\mu_2 = \langle (n - \langle n \rangle)^2 \rangle$, and

$$n\, m_f(n) = 5f - 11 - K(f - 1 - n) \qquad (3D) \qquad (3)$$

(Rivier 1985), where K is a constant of the froth. The limit f = ∞ is the correct procedure for topological stereology (deduct the properties of a random planar section from those of a 3D froth). Then $K = 5-(12+\mu_2)/f+...$ One can check Eq. 2 in the case of an enormous bubble in 2D. Its neighbours are indeed 5-sided on average (m(∞) = 5). Aboav's law (2) is well obeyed by all types of random structures (Weaire & Rivier 1984).

STRUCTURAL EQUATIONS OF STATE

Microreversibility implies statistical equilibrium. Its properties (probably distributions p(n) or p(f), and of cell sizes, equation of state) are obtained by maximum entropy (arbitrariness) formalism, (MEF) (Jaynes 1957, 1978).

Statistical equilibrium corresponds to the most probable or most arbitrary distribution of cells, which obeys a few constraints. Most probable is equivalent to least amount of implicit, additional biais, apart from that enforced by the constraints. It is this extremal requirement which grants uniqueness to an otherwise underdetermined problem.

Two constraints are unavoidable and mathematical: Topology (Eq. 1.), and filling a fixed volume Ω

$$\langle \overline{A}_n \rangle = \Omega/F \quad (2D), \quad \langle \overline{V}_f \rangle = \Omega/C \quad (3D) \qquad (4)$$

where $\overline{A}_n$, $\overline{V}_f$ are the average area of n-sided faces, volume of f-faceted cells, respectively.

The less restrictive the constraints (1) and (4), the larger the range of solutions. Accordingly, the most probable distribution will also be the least limited, and corresponds to redundant constraints, thus $\overline{A}_n \propto n$ or $\overline{V}_f \propto f$ (Rivier & Lissowski 1982, Rivier 1982)

$$\overline{A}_n = (\Omega/F)\beta \quad [n - (6 - 1/\beta)] \qquad (2D) \qquad (5)$$

$$\overline{V}_f = (\Omega/C)\beta \quad [f - (\langle f \rangle - 1/\beta)] \qquad (3D \qquad (6)$$

Maximizing the arbitrariness has therefore, among others, related the size of cells to their shape. Equation of state (5) has been discovered empirically by Lewis (1928) in undifferentiated, biological tissues. Here β is an undetermined multiplier enforcing the constraints. If the statistical equilibrium is maintained at all times, β is simply proportionnal to the time itself

$$\beta = F/\Omega \int \delta dt \qquad (7)$$

$$d\overline{A}_n/dt = \delta(n-6) \qquad (8)$$

and similarly, ($F \rightarrow C$, $n \rightarrow f$, $6 \rightarrow \langle f \rangle$), in 3D (Rivier 1983). Eq. 8 is called von Neumann's (1952) law. It governs the slow evolution of the tissue.

Equations of state (5) and (6) correspond to an ideal tissue, governed by the minimal number of unavoidable constraints, which are only mathematical and have nothing to do with biology, physics, etc. thereby justifying the universality of the resulting structures. Metallurgical aggregates are non-ideal: Lewis' law is not obeyed by metallurgical grains, where it is the average perimeter or radius $\overline{R}_n$ of n-sided faces, rather than the area, which is proportional to n, as observed experimentally (Desch 1919) or in simulations (Srolovitz et al. 1984). This fact, according to MEF methodology, betrays the presence of (at least) another constraint, which is, obviously, the energy carried by interfaces between grains, i.e. their perimeter. The system increases its entropy by selecting a size-shape relation which could be either Lewis' or the perimeter law:

$$\overline{R}_n = \alpha'(n - n_o') \qquad (9)$$

The maximum entropy associated with the perimeter law is larger than for Lewis' (Rivier 1985), so that the former alternative is selected whenever interfacial energy is relevant.

The final state of a soap bubble froth or a sintering aggregate is also of interest. Evolution by Eq. 8 leads to a more differentiated tissue or froth. In fact, shape fluctuations do saturate, $\mu_2 (t = \infty) = (6 - a)(7 - a)$, where a is the number of sides of the smallest cells (Rivier 1985). If one allows for structural mixtures of two different types of cells (cells and pores), $\mu_2(t = \infty)$ is increased (de Almeida & Rivier 1986).

CONCLUSIONS

Statistical mechanics of structures, and the fact that random tissues are in statistical equilibrium, has brought in important results:

(i) Classification of structures by their equations of state, and uncovering of the specific (physical, biological) constraint responsible for their difference. This constitutes an elementary structural pathology, with the equation of state as a quantitative diagnostic method.

(ii) The structural equations of state (Eq. 5,6,9) are precise, necessary and sufficient criteria for the structure to be random. If there is a science of form, they describe the form of random, space-filling, cellular structures.

(iii) Randomness, through entropy or arbitrariness, is at the essence of all these structures, which are the least biaised partitions of a topological space between cells, grains or bubbles. But biais itself can only be measured against a background of reversible, local elementary structural transformations.

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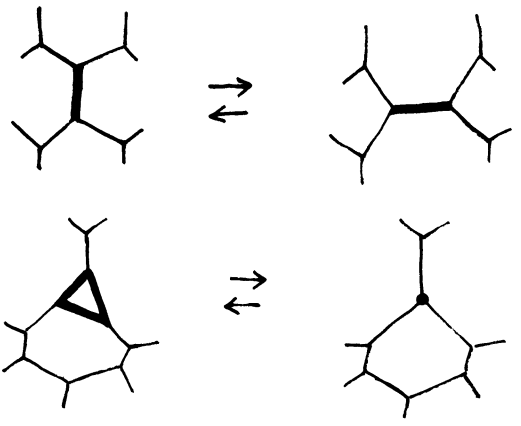

Fig 1: Elementary transformations in 2D: neighbour switching and mitosis.

REFERENCES

Aboav, D.A. (1970): The arrangement of grains in a polycrystal. Metallogr. 3: 383-390

de Almeida, R.M.C. and Rivier, N. (1986): in preparation.

Blanc. M. and Mocellin A. (1979): Grain coordination in plane sections of polycrystals. Acta Metall. 27: 1231-1237

Desch, C.H. (1919): The solidification of metals from the liquid state. J. Inst. Metals 22: 241-276

Dormer, K.J. (1980): Fundamental tissue geometry for biologists. [Cambridge Univ. Press]

Fortes, M.A. and Ferro, A.C. (1985): Topology and transformations in cellular structures. Acta Metall. 33: 1697-1708

Jaynes, E.T. (1957): Information theory and statistical mechanics. Phys. Rev. 106: 620-630; 108· 171-190
(1978): Where do we stand on Maximum Entropy? The Maximum Entropy Formalism: 15-118 [Ed. R.D. Levine and M. Tribus, Boston, MIT press]

Lewis, F.T. (1928): The correlation between cell division and the shapes and sizes of prismatic cells. Anat. Records 38: 341-376

Lewis, F.T. (1943): A geometric accounting for diverse shapes of 14-hedral cells. Amer. J. Botany 30; 74-81

Matzke, E.B. (1950): In the twinkling of an eye. Bull. Torrey Botan. Club 77: 222-227

Rivier, N. (1979): Disclination lines in glasses. Phil. Mag. A40; 859-68
(1982): Recent results on the ideal structure of glasses. J. Physique Colloques 43: C9-91
(1983): On the structure of random tissues or froths, and their evolution. Phil. Mag. B47: L45-49
(1985): Statistical crystallography: structure of random cellular networks. Phil. Mag. B52: 795-819

Rivier, N. and Lissowski, A. (1982): On the correlation between sizes and shapes of cells in epithelial mosaics. J. Phys. A15: 143-8

Sadoc, J.F. (1983): Periodic networks of disclination lines: Application to metal structures. J. Physique Lettres 44: L-707-715

Smith, C.S. (1954): The shape of things. Scient. Amer. 190(1): 58-64

Smoljaninov, V.V.: Mathematical Models of Biological Tissues. [Moscow, Nauka]

Srolovitz, D.J., Anderson, M.P., Sahni, P.S., and Grest, G.S. (1984): Computer simulation of grain growth - II. Acta Metall. 32: 793-802

von Neumann, J. (1952): Discussion-Shape of metal grains. Metal Interfaces [Cleveland Amer. Soc. Metals]: 108-110

Weaire, D. and Rivier, N. (184): Soap, cells and statistics - Random patterns in two dimensions Contemp. Physics. 25: 59-99