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The Arrangement of Cells in a Net. II

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The arrangement of the differently shaped domains into which a thin film of arsenic–selenium glass is found to divide is compared with that of the cells of a foam and the grains of a polycrystal. The significance of the parameters of an empirical equation describing the arrangement is briefly discussed.

Introduction

We return briefly to the question, recently discussed in this journal [1–3], of the shape and relative disposition of cells in a two-dimensional net. In a previous investigation [1] the arrangement of cells in a foam consisting of a single layer of soap bubbles held between two plates of glass was expressed in terms of m_n, the average number of sides of cells adjoining n-sided cells. This quantity was found to be governed, with an average error of 2% of the observed range, by the simple rule

$$m_n = 6 - a + (6a + \mu_2)/n, \tag{1}$$

where a is a numerical parameter equal to 1.2, and μ_2, the other parameter of the equation, is given by

$$\mu_2 = \sum_n (n - 6)^2 f_n \equiv \langle (n - 6)^2 \rangle, \tag{2}$$

f_n being the proportion of cells with n sides. The same rule, expressed in a slightly different form, had already been found to apply to a quite different system of cells, namely, that formed by the grains in a planar section of polycrystalline magnesium oxide [4]. This suggests that the rule may be of wide generality.

It is, however, clear that the parameter a, which experiment shows to be constant for a given system, does not have the same value for all systems. Boots [3], for example, has found that in an artificial net, made

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up of the Voronoi polygons generated by a random set of points in a plane, the observed values of m_n are represented by Eq. (1) but with a equal to less than half that given value.

Since the known facts may not suffice to determine why the dependence of m_n on n should be of this form, it was decided as part of this series of investigations to examine yet another physical system of cells, with the ultimate object of gathering enough data on which to base a general conclusion concerning the rule's validity. The material chosen for this purpose was a thin film of arsenic–selenium glass, which under the electron microscope appears divided into a network of cells, or *domains*. This material is well suited for comparison with a foam and a polycrystal, since it differs from them in its physical properties and in the way its cells are formed. The photographs of the films used in this work were provided by Dr. J. C. Phillips of Bell Laboratories.

Experimental Method and Results

Films of arsenic–selenium (As_2Se_3) glass, 500 Å thick, were prepared in commercial thermal evaporators at room temperature, with a deposition rate of 35–60 Å/min, on silicon wafer substrates coated with a thin

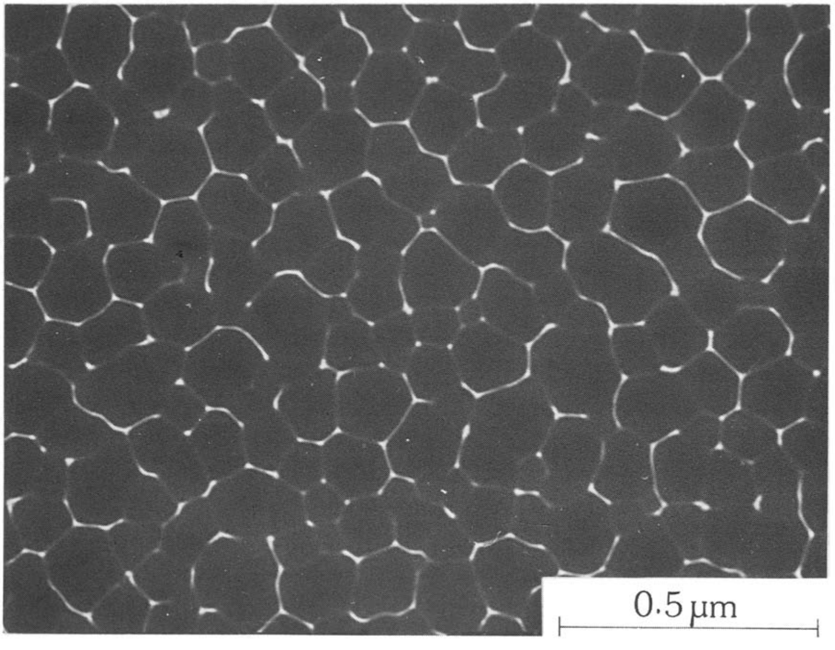

FIG. 1. Electron photomicrograph of a thin film of arsenic–selenium glass.

TABLE 1

Photograph	z	μ_2
114523	1535	2.0
116306	1029	1.8
07	1007	1.9
08	1114	1.9
09	971	1.5
10	919	1.3
11	1010	1.6
12	948	1.7

layer of polymer. The films were removed from the substrate by dissolving the polymer in acetone and were examined in a JEM 200B electron microscope using a low beam ($<10^{-3}$ A/cm^2) obtained by defocusing the second condenser lens. Under these conditions the resulting glass films exhibit a structure of microscopic domains, of the order of 1000 Å in diameter, that appear to be intrinsic to the films rather than induced by the radiation. Figure 1 is a photograph of this structure as it appears under the microscope.

From several such photographs, the number n of sides of each domain and the average number m_n of sides of domains adjoining n-sided ones were determined for a total of 6260 domains. The number of domains z measured in each photograph and the corresponding values of μ_2 are shown in Table 1. In Table 2 are given the values of z_n (the total number of n-sided domains) and of m_n for each value of n. Figure 2 shows the product $m_n n$ versus n.

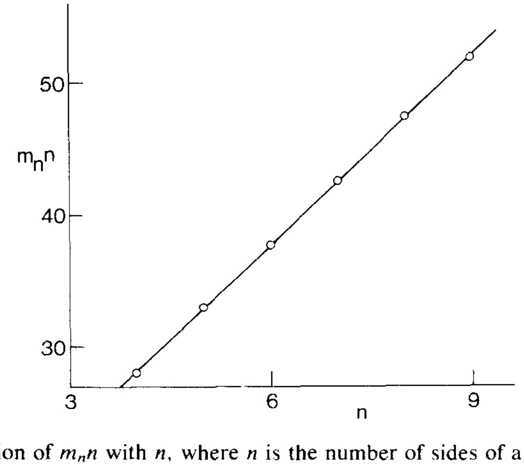

FIG. 2. Variation of $m_n n$ with n, where n is the number of sides of a domain and m_n is the average number of sides of the domains adjoining it.

TABLE 2

n	z_n	m_n	m_n'	$m_n - m_n'$
3	67	—	—	—
4	1079	7.00	7.03_5	-0.03_5
5	2327	6.60	6.59	0.01
6	1638	6.30	6.29	0.01
7	823	6.09	6.07	0.02
8	266	5.92	5.91	0.01
9	59	5.77	5.79	−0.02
10	1	—	—	—

Discussion

An irregular net, or tessellation, of threefold vertices such as we are considering may be looked on as a departure from a regular tessellation of hexagons. For such a regular net, $\langle n \rangle$, the average value of n, is equal to 6, and $\langle n^2 \rangle = 36$. In the irregular net, on the other hand, $\langle n \rangle$ is (by Euler's theorem) still equal to 6, but $\langle n^2 \rangle > 36$. As can be seen from Eq. (2), it is simply related to the quantity μ_2:

$$\langle n^2 \rangle = 36 + \mu_2. \tag{3}$$

μ_2, the amount by which $\langle n^2 \rangle$ exceeds 36, may thus conveniently be taken as a measure of the net's departure from regularity.

Because $\langle n \rangle = 6$ always, it is independent of f_n, the fraction of cells having n sides. $\langle n^2 \rangle$, on the other hand, enjoys no such independence, since for an irregular net it exceeds 36 by an amount depending on the values of f_n [Eqs. (1) and (3)]. Furthermore, since experiment shows that the shape of different cells of an irregular net depends on their relative size [5–7], the distributions of f_n and of the cellular size are not independent. Unlike $\langle n \rangle$, therefore, $\langle n^2 \rangle$—and hence the parameter μ_2 of Eq. (1)—although ostensibly topological quantities, depend on metrical considerations.

Because $\langle n^2 \rangle$ is an average quantity, it can have the same value for different distributions of f_n and hence for different size distributions, which, as we have seen, depend upon the spatial arrangement of the cells. The arrangement of the different kinds of cell in a net is not, therefore, determined by $\langle n^2 \rangle$ but requires for its description a quantity that depends on how the cells associate, such as m_n, whose definition involves pairs of adjoining cells. It is easily shown [8] that $\langle m_n n \rangle$ is also equal to 36 + μ_2 and hence that

$$\langle m_n n \rangle = \langle n^2 \rangle; \tag{4}$$

yet it must be noted that this, too, is a relation between average quantities only, and thus the dependence of m_n on n cannot be deduced from it.

It is, however, found experimentally that in irregular nets, within a limited range of n, individual values of m_n are approximately given by Eq. (1), which may be written

$$(m_n n - \langle m_n n \rangle)/(n - \langle n \rangle) = 6 - a. \tag{5}$$

For a given value of n, that is, the deviations of the quantities $m_n n$ and n from their mean values are proportional. This approximate expression of the dependence of m_n on n is conjectured to be generally valid; it is stated here, without proof, as an empirical rule.

Equation (1), which thus expresses an experimental finding, is based on the hypothesis that two parameters, a and μ_2, suffice to give a quantitative description of the association of cells of different shape in an irregular net. Since this association depends on the relative size of the cells as well as on their shape, and since μ_2 can have the same value for different distributions of cellular size, the parameter a, too, must be assumed to depend on the metrical properties of the net.

The average quantities $\langle m_n n \rangle$ and $\langle n^2 \rangle$, which in theory are equal, in practice differ by an amount depending on the number of cells involved in their computation. They become more nearly equal as that number increases. Similarly, $\langle n \rangle$, which in theory is equal to 6, in practice has this value only approximately, the extent of its departure from 6 again depending on the number of cells used to compute it. These differences, which are caused by fluctuations in the occurrence and distribution of the different kinds of cell, in practice limit the accuracy with which μ_2 can be determined; however, this limitation may, in cases such as those being considered here, be circumvented by assuming Eq. (1) to be valid, as the following example, taken from the work of Boots [3], illustrates.

Table 3 shows, for a tessellation of 1377 polygons, z_n, the number of polygons having n sides, and the corresponding values of f_n and m_n. (Approximate extrapolated values of m_n have been inserted for the solitary hendeca- and dodecagon present in the tessellation, as their contribution to $\langle m_n n \rangle$ is too big to be left out of account.) The values of $n^2 f_n$, $m_n n f_n$, and $(n - 6)^2 f_n$ show that $\langle n^2 \rangle = 37.8$ and $\langle m_n n \rangle = 37.4$. Hence the values of μ_2 obtained from these and from $\langle (n - 6)^2 \rangle$—namely, 1.8, 1.4, and 1.8—differ within fairly wide limits. These will here be referred to as the *observed values* of μ_2. The differences among them stem from fluctuations in the values of f_n and m_n, which can be diminished only by increasing the total number of polygons.

The foregoing computation of $\langle m_n n \rangle$ involves no assumption regarding the dependence of m_n on n, but if this relation is assumed to be given by

Eq. (1), the observed values of m_n can be used to evaluate μ_2, which, because it does not entail a measurement of f_n, can be expressed within closer limits. We shall call the value of μ_2 determined in this way its *effective value,* although strictly speaking this name is misleading; the effective value, it must be emphasized, is not the same as the μ_2 calculated from f_n. It must be kept in mind that, unlike the observed values, the effective value of μ_2 is an *indirectly* determined quantity, and its calculation rests on an assumption.

From the observed values of m_n in the range between $n = 4$ and $n = 8$ of the above tesselation, (the effective values of) the parameters a and μ_2 are found by the method of least squares to be 0.61 and 1.54, respectively. These values of m_n are compared in Table 4 with m'_n, the value of m_n calculated from Eq. (1) using the above-determined values for the parameters therein. It will be seen that the average difference $m_n - m'_n$ over this range of n is about 2% of the corresponding range of m_n. The effective value of μ_2 thus enables m_n in Boots's tessellation to be represented over a limited range of n with the same degree of accuracy as in the case of the cells of a foam [1] or the grains of a polycrystal [4], a result that could not be obtained with an observed value of μ_2 unless far more than 1377 polygons are taken into account.

The accuracy with which μ_2 can be determined in practice has here been discussed at some length because of the obstacle it places in the way of establishing a quantitative relation between n and an average quantity like m_n. With an artificial net like that of Boots, the difficulty can be surmounted by taking into account a sufficiently large number of cells,

TABLE 3

n	z_n	f_n	m_n	n^2f_n	m_nnf_n	$(n-6)^2f_n$
3	16	0.0116	6.96	0.104	0.242	0.1044
4	144	0.1046	6.68	1.674	2.795	0.4184
5	363	0.2636	6.44	6.590	8.488	0.2636
6	406	0.2948	6.26	10.613	11.073	0
7	271	0.1968	6.10	9.643	8.403	0.1968
8	121	0.0879	6.05	5.626	4.254	0.3516
9	43	0.0312	5.81	2.527	1.631	0.2808
10	11	0.0080	5.74	0.800	0.459	0.1280
11	1	0.0007	$(m_nn \sim 63)$	0.085	0.044	0.0175
12	1	0.0007	$(m_nn \sim 68)$	0.101	0.048	0.0252
$\langle n^2\rangle$	—	—	—	37.8	—	—
$\langle m_nn\rangle$	—	—	—	—	37.4	—
μ_2	—	—	—	1.8	1.4	1.8

TABLE 4

n	m_n	m_n'	$m_n - m_n'$
4	6.68	6.69	−0.01
5	6.44	6.43	0.01
6	6.26	6.26	0
7	6.10	6.13	−0.03
8	6.05	6.04	0.01

but that remedy may not always be available in the case of a naturally occurring net. It is, for example, a common experience that even a carefully prepared sample of metal may show slight differences in the general configuration of its grains in neighboring regions; in that case, measuring a larger number of grains does not necessarily lead to a more accurate value of μ_2. Thus, in establishing a relation like Eq. (1) for a naturally occurring net, there is in general a certain accuracy that it is not practical to try to exceed.

We now consider the results obtained with the thin films of arsenic–selenium glass. Figure 2 shows that in this case $m_n n$ varies linearly with n. The film of glass thus resembles the foam and the polycrystal in that the dependence of m_n on n can be expressed

$$m_n = A + B/n, \tag{6}$$

which can be written

$$m_n = 6 - a' + (6a' + \mu_2')/n, \tag{7}$$

where $a' = 6 - A$, and $\mu_2' = B - 6a'$. By the method of least squares and using the observed values of m_n shown in Table 2, $a' = 1.21$ and μ_2' 1.72; in Table 2 these values of m_n are compared with m_n', the value calculated from the equation

$$m_n = 4.79 + 8.98/n, \tag{8}$$

which is obtained on substituting the preceding values of a' and μ_2' in Eq. (7). The comparison shows that the average difference $m_n - m_n'$ is about 1.5% of the observed range of m_n.

Since a' is practically equal to the parameter a in Eq. (1), and μ_2' to the average value of μ_2 computed from the observed values of f_n shown in Table 2, Eqs. (1) and (7) are for practical purposes identical. Subject, then, to the inherent uncertainty in the value of μ_2, m_n for the domains of a thin film of arsenic–selenium glass is governed by the same rule as is found to hold for the cell sections of a foam [1] and a polycrystal [4];

in particular, the parameter a has the same value, 1.2, for each of the three systems. Compared with m_n' the observed values of m_n in Table 2 show a small but systematic departure from linearity in their variation with $1/n$; however, in this case, as in the others, the departure is small enough to allow Eq. (1) to serve as a practical rule.

Despite their similar appearance, the nets thus far considered differ physically in several respects. The way the cells of a typical foam are formed resembles that of the grains of a polycrystal in that each involves the progressive collapse of the smallest cells of an assembly, but the mechanism of that collapse—depending in one case on the surface tension of a liquid and in the other on a property peculiar to the surface, or interface, of a crystal—is far from being the same in each case. Also, the formation of characteristic domains in a thin film of a chalcogenide glass depends on a process that does not, as far as can be seen, involve their collapse. It is remarkable, then, that for systems so differing in their physical attributes the parameter a of Eq. (1) should nevertheless have the same value.

Since we are, as we have seen, obliged on general grounds to assume that this parameter depends on the metrical properties of the net, there is a presumption that if it has the same value for such differing systems as the foam, the polycrystal, and the film of glass, then this is owing not to a physical property of the systems or to their mode of generation but to a geometrical condition to which they are subject—possibly the condition, characterizing many such naturally occurring systems, that the cell edges are constrained to meet at a certain angle. This conjecture receives support from Boots's finding that the parameter a has a different value for a tessellation subject to a different geometrical condition—namely, that the straight edges of its polygons should be perpendicular bisectors of lines joining pairs of neighboring points from a random planar set, with consequently no restriction imposed on the angles at which they meet.

It may therefore be concluded that Eq. (1) with $a = 1.2$ expresses a general property of naturally occurring nets, possibly depending on a restriction imposed on the angle at which the sides of its cells meet. It is further surmised that if the parameter has a different value for nets like those of Boots, it is because their cells are subject to a geometrical restriction of a kind not commonly encountered in nature.

Conclusions

The arrangement of the microscopic domains of a thin film of a chalcogenide (As_2Se_3) glass is found to obey the same rule as applies to the cells of a foam and the grains of a polycrystal;

$$m_n = 6 - a + (6a + \mu_2)/n,$$

where m_n is the average number of sides of domains adjoining n-sided ones, a is a numerical parameter, and $\mu_2 = \langle (n - 6)^2 \rangle$.

The parameter a is found in this case to have the same value, 1.2, as it has for the foam and the polycrystal. It is conjectured that a is a metrical parameter whose value for the systems we have here considered is determined by a geometrical condition governing the angle at which the cell sides meet.

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