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Surface area of coarsening cellular structures

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Abstract

Three dimensional cellular structures serve as models of foams and annealed metals. Such structures coarsen over time. Assuming that cell growth is driven by face curvature, a general formula for the rate of surface area change is obtained. Based on it, a new von Neumann–type relationship linking the rate to topology of individual regular cells with spherical–cap faces is derived. Moreover, the general formula is applied to structures with arbitrary cells. In this case, the rate of total area change is related to structure topology and one geometric parameter – the average difference between principal curvatures of faces. Assuming further that the structure is self–similar, we obtain expressions for the dependence of structure parameters on time. This opens the opportunity to examine conditions, for which the growth of self–similar structures follows the "parabolic law". Also the effect of topological transformations on the coarsening process is considered.

Introduction

Ideal cellular structures are archetypes of dry foams and annealed polycrystalline metals. An important aspect of understanding such structures is figuring out how they evolve over time. Some of the cells grow while other shrink and eventually disappear. The disappearance of cells causes an increase of the average cell size. Therefore, the process is referred to as coarsening.

A cellular structure reduces its total surface area due to surface tension. In foams, long time mechanisms involve diffusion of gas through cell walls. Assuming constant surface tension and constant diffusion coefficient, the pressure difference in two neighboring cells is proportional to the curvature of their common boundary. The resulting boundaries have constant curvatures and move towards their concave sides. For a quantitative description of the movement and structure evolution, boundary velocities are needed. The commonly used (von Neumann's) kinetic postulate is that the velocities are proportional to boundary curvatures. The postulate underpins one of the most frequent motifs in analysis of two dimensional (2D) cellular structures — the von Neumann's law [1, 2]. Let κ denote local curvature of cell sides. For a given cell, the rate of area change $\dot{A} \equiv \mathrm{d}A/\mathrm{d}t$ is proportional to the integral of the interface velocity v over the cell perimeter $\mathcal{L}$ and $\dot{A} \propto \int_{\mathcal{L}} v \propto \int_{edges} \kappa$. With local equilibrium, the angles at triple points are $2\pi/3$, and for an n sided cell $\int_{edges} \kappa = 2\pi - n(\pi/3)$. Hence, one gets $\dot{A} \propto n - 6$. The relationship is remarkable because the rate of area change does not depend on the cell size, shape or environment but only on the number of its sides: an n–sided cell is stationary if $n = 6$, it grows (shrinks) at a constant rate if the number of sides is larger (smaller) than 6.

The von Neumann's law is not entirely strict. When writing $\dot{A} = \int_{\mathcal{L}} v$, a tacit assumption is that the velocity is continuous along the boundary. For 'polygonal' cells, the principle does not take into account the change of the area which occurs at vertices. More importantly, the assumption of interface velocity to be proportional to the curvature does not conform to the requirement that the junction angles are $2\pi/3$ [3, 4]. Nevertheless, the law is a very good approximation, and it is at the core of investigations on 2D structures.

In three dimensions (3D), assuming the von Neumann's kinetics means that boundaries

of cells move with velocities proportional to their local *mean* curvatures; surfaces having zero mean curvature – which are known to minimize surface area – are immobile. Analogously to the 2D case, for a 3D particle having a surface moving with such velocity, the rate of volume change is proportional to $\int_{faces} H$, where H is the mean curvature. However, the analogy stops here because there is no simple relationship between $\int_{faces} H$ and topology of the cell. As a matter of fact, it is possible to create examples showing that an absolute 3D equivalent of 2D von Neumann's law cannot be derived; two cells of a valid structure may have the same number of faces but different $\int_{faces} H$ and, consequently, different rates of volume change. Therefore, considerable efforts have been made to formulate more sophisticated 3D analogues of the von Neumann's law. There is much interest in the relationship proposed by Glazier [5]. Based on numerical simulations, he suggested that a normalized average volume of cells having fixed number of faces is determined by that number. For discussions on that suggestion see [6, 7, 8, 9].

Previous attempts to generalize the von Neumann's law to 3D have been focused on the volume of cells and the rate of its change, whereas we provide a formula for the rate of change of *surface area*. Based on the kinetic postulate of curvature driven motion, we derive a relationship governing the area change rate. The relationship can be seen as a basis for getting a 3D analogue of the von Neumann's law. It is applied to coarsening of individual regular cells having spherical faces; their rate of area change is expressed through the number of cell faces. The general formula for the area change rate is also used to explore the evolution of self–similar structures; this leads to expressions for time–dependence of the total surface area, the number of cells, and the average radius of cells. Moreover, the influence of topological rearrangements on the rate of area change is evaluated.

Rate of area change for an individual cell

An ideal 3D cellular structure ('polyederschaum') consists of space–filling 'polyhedral' cells satisfying Plateau's rules. (Smooth faces separate neighboring cells, each vertex is common to four cells and four edges, and each edge is shared by three faces meeting at the angle of $2\pi/3$.) Let the symbols V, E and F denote the numbers of cell vertices, edges and faces, respectively. Since three edges of a cell meet at one vertex, and each edge has two ends,

$E = 3V/2$. From Euler theorem, the number of faces equals $F = E - V + 2$. Hence, one of the three cell parameters V, E and F determines the other two, and the parameters are used here interchangeably.

The sign of the mean curvature is determined by surface orientation. The latter also determines the direction of the vector $\mathbf{n}$ normal to the surface. Thus, $H\mathbf{n}$ does not depend on the orientation. With the curvature driven motion, the velocity $\mathbf{v}$ of a boundary segment is assumed to be given by $\mathbf{v} = \mu H\mathbf{n}$ with $\mu > 0$.

For a 3D particle having 'smooth' surface $\mathcal{S}$, the rate of change of surface area is related to the interface velocity $\mathbf{v}$ by $\dot{S} = -2\int_{\mathcal{S}}(\mathbf{v}\cdot\mathbf{n})H$; see, e.g., [10]. Similar relationship can be applied to a polyhedral cell by adding the area change χ caused by moving edges and vertices

$$\dot{S} = -2\int_{\mathcal{F}}(\mathbf{v}\cdot\mathbf{n})H \ + \chi \ ,$$

where the integration is performed over the set $\mathcal{F}$ of all faces of the cell. By neglecting the effects caused by the curvature of the edges and these occurring at vertices, the last term can be expressed as

$$\chi = 3^{-1/2}\sum_{i=1}^{F}\int_{\mathcal{P}_i}\mathbf{v}\cdot\mathbf{n}_i \ , \tag{1}$$

where $\mathcal{P}_i$ is the perimeter of the i-th face, and $\mathbf{n}_i$ is an outward directed normal to the i-th face (Fig. 1). With surface velocity proportional to the mean curvature, one gets the key expression for the rate of area change

Fig. 1

$$\dot{S} = -2\mu\int_{\mathcal{F}}H^2 + 3^{-1/2}\mu\sum_{i=1}^{F}\int_{\mathcal{P}_i}H\mathbf{n}\cdot\mathbf{n}_i \ . \tag{2}$$

The squared mean curvature is related to the Gauss curvature K via $H^2 = K + \Delta^2$, where Δ is half of the difference between principal curvatures; H^2 equals K if the surface is a spherical–cap. From the Gauss–Bonnet theorem, for a given cell,

$$\int_{\mathcal{F}}K = 4\pi - V\kappa^V - \sum_{j=1}^{E}\kappa_j^E \ , \tag{3}$$

where κ_j^E is the Gauss curvature 'contained' in the j-th edge, and $\kappa^V = 2\pi - 3\,\mathrm{arcsec}(-3) \approx 0.5512856$ denotes the curvature 'contained' in an equilibrated vertex.

The last terms of eqs (2) and (3) are difficult to estimate for cells of arbitrary shapes but they can be expressed analytically for simple regular 'polyhedra' having spherical faces. Moreover, the complicated terms cancel out in the expression for the rate of total surface area. These two cases are considered below.

The rate of area change for regular cells

One of the approaches to foam evolution is to assume that structures consist of cells satisfying Plateau's rules and having spherical faces [6]. In particular, *regular* cells of this type are considered [11, 12, 13]. For a regular cell, F determines the solid angle corresponding to one face. Since the polyhedra are trivalent, the number of sides of each face n_s equals $2E/F$. The curvature of the faces is calculated from the condition that the spherical faces meet at the angle of $2\pi/3$. Ultimately, the complete shape of the cell is determined by F. Although, regular cells are constructable only for $F = 3, 4, 6$ and 12, formulas are assumed to hold for all $F \geq 3$.

We use results from [12, 13] to determine the last terms of eqs (2) and (3). With $\Delta = 0$ and constant curvature $H = H_F$, one has $\int_{\mathcal{F}} H^2 = \int_{\mathcal{F}} K$, and the last term of eq.(3) is given by $\sum_{j=1}^{E} \kappa_j^E = E\kappa^E$, where $\kappa^E = 2\arcsin\left(3^{-1/2}H_F\right)$, $3H_F/2 = \left(2(3-4c^2)\right)^{1/2} - c$ and $c = \cos(\pi/n_s)$. Moreover, since H_F is positive for convex faces, one has $\mathbf{n}\cdot\mathbf{n}_i = -1$ and $\sum_{i=1}^{F} \int_{\mathcal{P}_i} H\,\mathbf{n}\cdot\mathbf{n}_i = -H_F \sum_{i=1}^{F} \int_{\mathcal{P}_i} 1 = -2\,EH_F l_E = -3^{1/2}E\kappa^E$, where l_E is the length of a single edge.

Now, taking into account the above components, eq.(2) can be expressed as

$$\dot{S} = -\mu\Gamma \quad \text{with} \quad \Gamma \equiv 8\pi - E\kappa^E - 2V\kappa^V \ . \tag{4}$$

Since Γ depends on one independent topological parameter (Fig. 2), this relationship can be seen as an analogue of von Neumann law applicable to regular cells. *Fig. 2*

A different formula of this kind was proposed by Glicksman [12]. In his derivation, the effect corresponding to χ is neglected, and the formula contains only the contribution related to the curvature of faces. Consequently, the resulting $\dot{S}$ is never positive, i.e. a regular cell would loose area no matter how many faces it had.

Also Hilgenfeldt *et al.* [13] considered an analogue of von Neumann's law in relation

to regular cells. From their result, which is given as the rate of volume change, one can calculate the rate of change of area versus number of cell faces. Predictably, it turns out to be different than the rate given by eq.(4); see (Fig. 2). The reason is that the underlying 'empirical' Glazier's formula used in [13] does not fully conform to the conditions of curvature driven coarsening.

The rate of change of total surface area

Considerations of previous section concern individual cells of particular shape; as for arbitrary structures, eq.(2) allows for estimating the rate of change of their *total* surface area. Let C be the number of all cells. We will add the index $m = 1, \ldots, C$ to already defined symbols to numerate cells. With S_m denoting the surface area of the m-th cell, the total surface area S_T is given by $S_T = \sum_{m=1}^{C} S_m/2$. For three neighboring cells, the sum of three Gaussian curvatures 'contained' in their common edge is zero; therefore, the sum over all cells of the last terms of eq.(3) $\sum_{j=1}^{E} \kappa_j^E$ also vanishes [14, 15]. Moreover, within our approximation, arguments analogous to those given in [16] are applicable: at a triple junction, a local change of area of a given face due to a displacement of the junction is compensated by the area changes of the other two faces; see Fig. 3. Hence, $\sum_{m=1}^{C} \chi_m = 0$. *Fig. 3*
Therefore, the rate of change of the total area is governed by $\dot{S}_T = -2\mu \int_{\mathcal{F}_T} H^2$ or

$$\dot{S}_T = -\mu C \left(4\pi - \kappa_V \overline{V}\right) - 2\mu \int_{\mathcal{F}_T} \Delta^2 \ , \tag{5}$$

where $\mathcal{F}_T$ stands for all faces of the structure, and $\overline{V} = \sum_{m=1}^{C} V_m/C$ is the mean number of cell vertices. The analogy of eq.(5) to the von Neumann relationship is spoiled by $\int_{\mathcal{F}_T} \Delta^2$; however, it is unlikely that a better analogue can be obtained without taking additional assumptions.

Let V_T be the number of all vertices in the structure; $V_T = C\overline{V}/4$. Topological transformations (Fig. 4) change the value of $-\mu C \left(4\pi - \kappa_V \overline{V}\right) = -\mu \left(4\pi C - 4\kappa_V V_T\right)$ and consequently the rate $\dot{S}_T$. (The change of $\int_{\mathcal{F}_T} \Delta^2$ is negligible at the instant of transformation.) For a 3D neighbor switching or face elimination (referred to as $+T_1$, and causing $C \to C$, $V_T \to V_T - 1$) the change is $-4\mu\kappa_V$, and for the disappearance of a tetragonal cell by shrinking (T_2 causing $C \to C - 1$, $V_T \to V_T - 3$), it is $+\mu(4\pi - 12\kappa_V)$. Thus, the former *Fig. 4*

transformation is associated with acceleration of the surface loss, and the latter – with deceleration of the process.

Topology puts a lower limit on the integral Δ^2. Since surface tension acts towards reduction of S_T, the rate $\dot{S}_T$ is non–positive. Hence, (5) leads to $\int_{\mathcal{F}_T} \Delta^2 \geq C\left(\kappa_V \overline{V} - 4\pi\right)/2$. It is easier to interpret this inequality using the average value of the integral $\int_{\mathcal{F}_m} \Delta^2$ per cell; let it be denoted by $\overline{\Delta^2}$. The average is given by $\overline{\Delta^2} = (2/C)\int_{\mathcal{F}_T} \Delta^2$, and it is bounded by

$$\overline{\Delta^2} \geq \kappa_V \overline{V} - 4\pi \ .$$

This inequality exposes a formal limit on the applicability of models with cells having spherical faces: for such cells $\overline{\Delta^2} = 0$, and the models are applicable only to structures with $\overline{V} \leq V^* \equiv 4\pi/\kappa_V \approx 22.795$.

Kinetics of self–similar structures

The conventional definition of self–similar structures is based on the cell size distribution $f = f(R,t)$, where R is the 'volume equivalent' radius of cells: an evolving cellular structure is self–similar if the distribution of cell sizes scaled by average radius $\overline{R} \equiv \int_0^\infty Rf\,\mathrm{d}R$, is constant in time. Formally, this means that the product $f(R,t)\overline{R}(t)$ does not depend on t. Let $x = R/\overline{R}$ and $P(x) \equiv f(x\overline{R},t)\overline{R}(t)$. Since $\overline{R^n} \equiv \int_0^\infty R^n f(R,t)\,\mathrm{d}R = \overline{R}^n \int_0^\infty x^n P(x)\,\mathrm{d}x$, the ratios $p_n \equiv \overline{R^n}/\overline{R}^n$ do not depend on time either.

The total volume of all cells is constant. Without loosing generality, this volume is taken to be unity. From the time–independence of p_2 and p_3: total volume $= 1 = Cq_3\overline{R}^3$, and $S_T = Cq_2\overline{R}^2/2$, where $q_3 = 4\pi p_3/3$, and q_2 is constant for a given self–similar structure. Hence, $S_T\overline{R} = q_2 q_3^{-1}/2 = \text{const}$ and $S_T^3 C^{-1} = q_2^3 q_3^{-2}/8 \equiv \alpha = \text{const}$. These relations combined with (5) lead to the following formulas for time–dependence of S_T, C and $\overline{R}$:

$$\begin{aligned} S_T^{-2} - S_{T0}^{-2} &= \alpha^{-1}\ \Psi(t)\ , \\ C^{-2/3} - C_0^{-2/3} &= \alpha^{-1/3}\ \Psi(t)\ , \\ \overline{R}^2 - \overline{R}_0^2 &= 2q_2^{-1}\ \Psi(t)\ , \end{aligned}$$

where

$$\Psi(t) \equiv 2\mu\left(4\pi t - \int_0^t (\kappa_V \overline{V} - \overline{\Delta^2})\,\mathrm{d}t\right)\ ,$$

and S_{T0}, C_0 and $\overline{R}_0$ denote initial values of S_T, C and $\overline{R}$, respectively. It is noteworthy that since the average surface area of cells $\overline{S} = 2S_T/C$ satisfies $\overline{S} - \overline{S}_0 = 2\Psi(t)$, the function $\Psi(t)$ has a simple interpretation: it is half of the total gain in the average surface area of cells. The relationship known in literature as the 'parabolic law of grain growth' (proportionality of $\overline{R}^2 - \overline{R}_0^2$ to t) follows if $\kappa_V \overline{V} - \overline{\Delta^2}$ is constant in time.[1]

If $\overline{V}$ is assumed to be constant in time, due to the non–negativity of $\overline{\Delta^2}$, one gets the lower bound on $\Psi(t)$

$$\Psi(t) \geq \mu\beta t \ ,$$

where $\beta \equiv 2(4\pi - \kappa_V \overline{V})$. Structures involving $\overline{\Delta^2}$ coarsen faster than those corresponding to the bounding case $\Psi(t) = \mu\beta t$. However, the bound is meaningful only if $\overline{V} < V^*$. It is also interesting to note that the constraint $\overline{V} = \text{const}$ allows for estimating frequencies of topological events: Let us assume that the structure changes its topology *only* through the elementary processes T_1 and T_2. The rearrangements $\pm T_1$ cause $\overline{V} = 4V_T/C \rightarrow 4(V_T \mp 1)/C$, while disappearance of a tetragonal cell leads to $4V_T/C \rightarrow 4(V_T - 3)/(C - 1)$. If the number of $-T_1$ exceeds that of $+T_1$, $\overline{V}$ cannot be constant. For $\overline{V}$ to be constant, the frequency f_1 of *net* face elimination events and the frequency f_2 of cell disappearance events must be such that the ratio of $f_2\left(4(V_T - 3)/(C - 1) - \overline{V}\right)$ and $f_1\left(\overline{V} - 4(V_T - 1)/C\right)$ is 1. For large C, this means that the ratio of the frequencies is $f_1/f_2 = \overline{V}/4 - 3$. Of the two processes T_1 and T_2, only the latter causes disappearance of cells, i.e., $f_2 = -\dot{C}$. Knowing the ratio f_1/f_2, the dependence of f_1 on time can be determined.

Summary

Application of the postulate of curvature driven motion to coarsening of ideal cellular structures leads to formula (2) expressing the rate of cell area change through integrals involving the mean curvature of faces. The analysis gets simpler in specific cases. For an individual regular cell with spherical–cap faces, the rate depends only on the number of faces. Moreover, the change rate of structure *total* area is expressed through topological quantities and the (non–topological) average of difference between principal curvatures of

[1] With a more restrictive definition of self–similarity demanding all dimensionless characteristics to be time independent, the parabolic law follows directly (because $\kappa_V \overline{V}$ and $\overline{\Delta^2}$ are dimensionless).

surfaces; that expression is further simplified, if the structure is self–similar. Although the resulting eqs (4) and (5) are not true 3D forms of the von Neumann's law (the first one is based on restrictive assumptions, and the second one involves non–topological parameter), they seem to be its closest analogues currently available via purely theoretical considerations based on the (von Neumann's) kinetic postulate. Since the ideal cellular structure is an important benchmark for numerical treatment of foam and polycrystalline growth, these theoretical results can provide a support for testing numerical models of physically realistic 3D structures.

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Captions

Figure 1: Schematic illustration of the derivation of χ. The shift of the two lines by δ_1 and δ_2 leads to the increase of their length by $s_1 + s_2 = (\delta_1 + \delta_2)/\sqrt{3}$. There are other cases with slightly different geometry (e.g., if $\delta_1 > 2\delta_2$) but all are taken into account in eq.(1).

Figure 2: The dimensionless factor $-\Gamma = \dot{S}/\mu$ versus the number of faces F for regular cells (stars). For comparison, the von Neumann–type relationship of Glazier applied to regular cells [13] gives results marked by diamonds. (All diamonds are below stars.)

Figure 3: Schematic 2D illustration of the relationship $\sum_m \chi_m = 0$ for a triple junction. The junction A of gray boundaries 1, 2 and 3 meeting at the angles $2\pi/3$ is displaced to B; the boundaries at the new location are marked in black. Since $|BC| + |BE| = |AD|$, the loss $|AD|$ in length of boundary 1 is compensated by the gains $|BC|$ and $|BE|$ for boundaries 2 and 3, respectively.

Figure 4: Topological transformations T_1 and T_2.

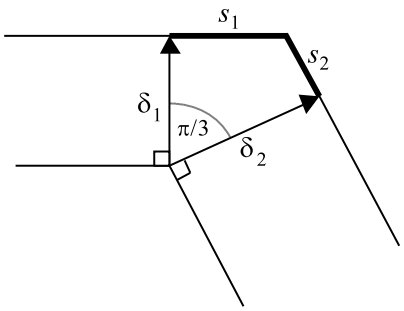

Figure 1: *A.Morawiec ...*

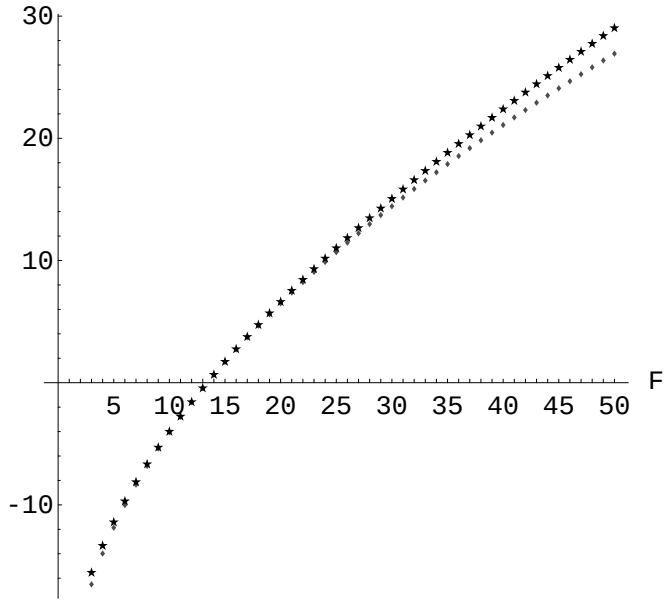

Figure 2: *A.Morawiec ...*

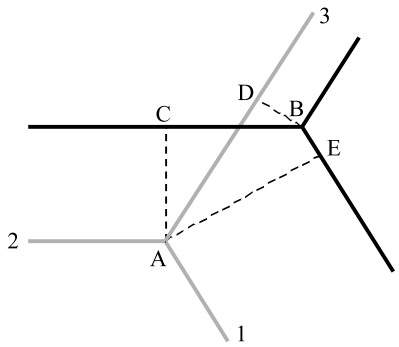

Figure 3: *A.Morawiec ...*

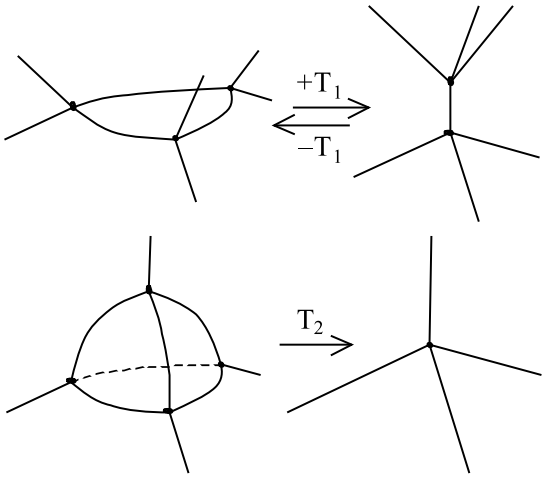

Figure 4: *A.Morawiec ...*