

Does competition between growth elements eventually eliminate self-affinity?

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*"See how various the forms and how unvarying the principles."
Owen Jones*

Abstract Several disparate natural structures have been identified as fractals, but these identifications are usually limited to certain length scales; macroscopic objects, on the other hand, are generally homogeneous and Euclidean. From the observations on several different growth processes, we have arrived at the understanding that competition between growth elements, masses as well as mechanisms, eventually eliminates self-affinity. The concepts of de-sticking probabilities and reversibility are central to this conclusion.

Introduction

Ever since the exposition and the development of fractal concepts,¹ researchers from diverse laboratories and universities have devoted considerable time and efforts in identifying naturally occurring fractals as well as in understanding fractal growth processes.² From cauliflowers and broccoli to clouds and coastlines, from electrode interfaces to thin films, fractal structures have been observed and reported. Elaborate models of diffusion-limited aggregation, ballistic aggregation, as well as of other growth processes have been simulated on computers.³ Various attempts have been made to develop mathematical formalisms which supposedly describe the various attributes of fractal structures.^{4,5} Yet the basic question – Fractals: Where's the Physics? – has largely remained unanswered.⁶ And, very intimately tied to that query, is the dilemma: Is Physics fractal? Or, are fractals merely the outwardly manifestations of physical processes whose true characteristics still remain elusive?

Generally, answers to questions of such import painstakingly conceal themselves, which makes it all the more necessary that some general principles be searched for, some common denominators between apparently different, but self-affine, processes and structures be identified. In what follows, we shall try to convey our perspectives

on natural growth processes to exemplify our understanding that *competition between growth elements, masses as well as mechanisms, eventually eliminates self-affinity*. A recent computer simulation study of aggregation from lattice gas also hints towards a fractal-to-compact transition.⁷ The floor is, however, open for debate that, we hope, will arise as a result of this communication. It is to be emphasised that novelty is not claimed for all the individual parts of this paper; indeed, the works of other researchers, as well as of ours, have been borrowed to support the premise of this paper.

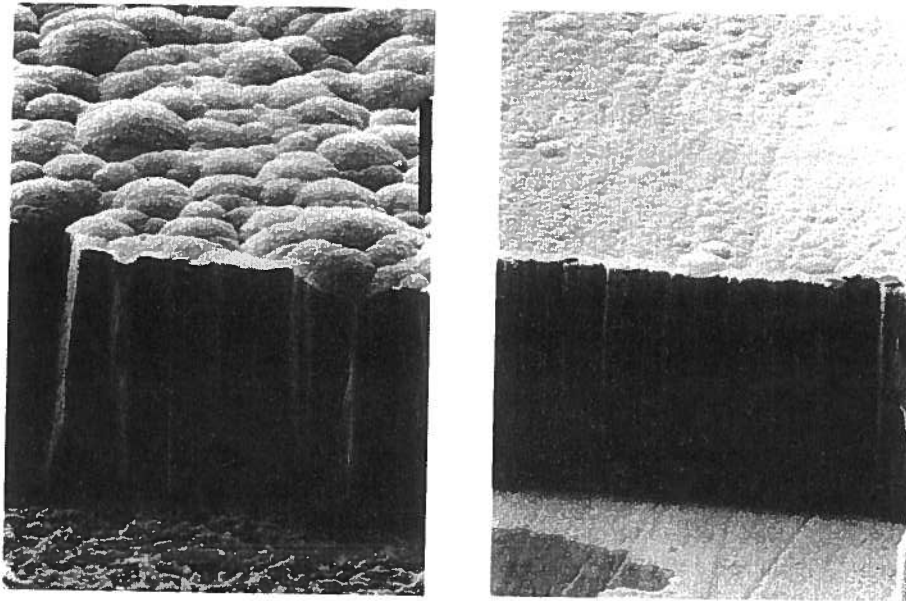
Gaskets and cellular automata

In an earlier paper,⁸ we have described families of fractal gaskets which are derivable from the generalisations⁹ of the Pascal triangle. These structures, called the generalised Pascal–Sierpinski gaskets (GPSGs), are classified by a bi-variate index (K, L) in which $K \geq 2$ as well as $L \geq 2$. It turns out that almost all such gaskets are self-affine; except when $K = 2$ and L is a prime, and in which case the similarity dimension works out to be equal to $\log[1+2+3+\dots+L]/\log[L]$, with L serving as the scale.^{10,11} But even when $K > 2$, it appears from visual examination of the gaskets, that for prime L , the designation of L as the scale is still appropriate. This is because the row $n = L^p$, $p \geq 1$ contains a total of K seeds (occupied sites) and $(K - 1)(L^p - 1)$ nulls (voids), the nulls being grouped together in $(K - 1)$ groups, each group consisting of $(L^p - 1)$ nulls and bounded on either side by a seed; needless to add, since this grouping keeps on recurring every L^p -th row, the proper fractal scale must be logarithmic with base L , regardless of the specific value of K .

The L^p -th row can be regarded, in the manner of Huygens's principle, as a re-seeding row for these GPSGs. Each seed on this row evolves out into a triangular gasketlet made up of $L^p(L - 1)$ rows. Down its L^p -th row, however, each gasketlet contains $[1 + (L^p - 1)(K - 1)]$ sites which is larger than $[1 + (L^p - 1)]$ except when $K = 2$. Consequently, these gasketlets *interfere* with each other. As a result, the number of seeds does not scale with L , unless $K = 2$. It is to be emphasised here that this interference, or the *competition due to re-seeding*, destroys self-similarity even in the presence of a well-established scale.

Typically, well-known gaskets like those of Sierpinski, or the Menger sponge,¹ turn out to be self-similar because, as these structures evolve, increasingly larger voids are created. Consider, however, cellular automata with initially disordered conditions,¹¹ which can be regarded as deterministically grown gaskets. As can be observed from cellular automata, such a gasket's voids do not increase in size indiscriminately with increasing gasket size. Parenthetically, we observe here that total void size, if not growing at the same rate as the total structure, is responsible for the structure exhibiting a fractal nature; this argument augments upon the one given for river basins by Kondoh *et al.*¹² Furthermore, many of the voids have tails and some inter-void connectivities also exist. These features suggest a similarity to materials with highly-interlocked grains, or even to DLA simulations with low sticking probabilities.^{13,14}

These structures have relevance to the cone-growth model of thin films.¹⁵ If the cones grow in a highly competitive environment, the cone diameters do not grow indiscriminately large, the result being that such films are dense as well as fine-grained, and are characterised by large 'fractal' dimensions. Incidentally, the



A

B

Figure 1 SEM view of the top surface and fracture cross-section of 25 μm thick hydrogenated amorphous Si films deposited on: (A) tin oxide sprayed coating on a stainless steel substrate, and (B) polished stainless steel substrate. The bar marker is 10 μm . The substrate surface, upon which the films grew, can be seen in the lower portion of each micrograph. Note that the scratch marks on the relatively smoother substrate (B) are still detectable on the top surface of the thick film, indicating thereby that the growth competition was not extensive enough to 'cover its tracks.' (Micrographs courtesy of R.C. Ross; see ref.18.)

cone-growth model also has relevance to the phenomena of fingering in fluid dynamics.¹⁶

Thin to thick film transition

The observations which have led to the development of a fractal-like model of thin film growth^{15,17} have been mainly based upon visual inspection and limited quantitative data. These initial papers, perhaps, overgeneralised the concept and the geometry of competition for cone growth. Recent work^{18,19} has shown that, although many films exhibit a similar degree of competition over the ranges of film thickness examined and lead to cauliflower-like top surfaces (see Figure 1A), there are distinct conditions when this competition actually ceases. This cessation of competition occurs after about 1 μm of growth evolution; the individual columns, which dictate the top surface's roughness, grow as nearly vertical-sided cones thereafter²⁰ (see Figure 1B). Such a final state has a morphology which is clearly non-fractal in the precise sense; yet there was an evolution to that particular state from a competition between adjacent cones for growth starting from the 10–30 Å thickness to the final

cylindrical column (*i.e.*, cylinder) width of $\sim 2,000\text{--}5,000$ Å. If the initial growth is indeed fractal-like, then the following question can be posed: because the film is microscopically non-uniform and still macroscopically homogeneous, what processes can lead to a continued change in the fractal character?

In a related sense, examination of spectroscopic ellipsometric modelling data^{21,22} has shown that for amorphous germanium (a-Ge) films subject to a high degree of renucleation due to ion-bombardment during growth, the density remains essentially constant regardless of the film thickness. For a-Ge films with a considerable lower degree of bombardment-induced renucleation, the density increases by 14% over the first 7,400 Å of growth, and remains constant thereafter.²³ In fact, such non-uniformity of the film density during the early stages of growth is appearing to be the rule, rather than the exception.²⁴ One tempting explanation for this phenomenon is related to the GPSGs⁸ of orders ($K > 2$, L prime): interference, or the competition due to re-seeding, destroys self-similarity even in the presence of a well-established scale. A related conclusion, drawn from studying cellular automata, is that maybe the films become 'Euclidean' as they evolve since the void distribution turns out to be largely homogeneous. This is because voids of large sizes appear in small numbers and are sparsely distributed, the structure being dominated by the more numerous voids of smaller sizes.¹⁷

Surface smoothening in columnar growth

Thin film growth morphology has many aspects ranging from the atomic scale clustering due to ballistic aggregation to single crystal films. For conditions where the atoms have very low adatom mobility, clustering at the 10–30 Å level is a natural consequence of the random ballistic aggregation process; this has been shown both by computer models^{25–27} as well as by direct observations of real films.^{28,29} Since a thin film has an essentially constant surface area, it means that the clusters which nucleate on the substrate must either grow independently, or they must compete for growth. The latter condition, of course, is what is observed in a variety of films.

This clustering and growth-competition model of the reality of low-mobility thin film growth has seen its main utility in guiding our initial attempts at both experimental¹⁷ and theoretical⁸ quantification of film morphology. We are now at a stage where we are beginning to extend this simple model to address such questions as: are atomic aggregation processes random; what are the consequences of the distribution of cluster sizes and of the rules of competition – albeit, what are the subtle consequences of competition; and, in what ways does re-nucleation, in all of its varied forms, affect final growth forms, such as the internal void network and the related top surface roughness.

In the thin film literature, it is generally found that top surface morphology increases in roughness with increasing film thickness.^{30,31} This is intuitively satisfactory since most studies start with nearly atomically smooth substrates (*e.g.*, polished silicon wafers, glass slides) and result in cauliflower-like (at low mobility) or highly-faceted (at high mobility) top surfaces. The reality of surface smoothening is not usually considered,³² but see Rikvold³³ and Nittmann and Stanley.³⁴ There are two circumstances to consider. Starting from either an initially rough substrate surface or with an initially rough thin film, is it possible that a smoother top surface results due to subsequent growth? From experimental observations, the answer to this

question is in the affirmative. For example, brightening agents are commonly used in the preparation of electro-deposited films.³⁵ These agents are so named because they do just that to a coated surface, turning it from a diffuse-scattering rough surface to a considerably smoother, specular surface.³⁶ It is known that the brightening agents increase the renucleation density and rate during film growth³⁷ and lead to mechanically harder films.

Similar smoothening has also been seen in vapour-deposited multilayer thin films^{32,39} where the contrast between the successive layers unwittingly records the roughness decimation and allows the observer to see subtle, but significant, changes (see Figure 10 of ref.39). In addition, it should be noted that as the layers alternate, there may be changes in the renucleation, especially at the interfaces where the importance of the changes cannot be gainsaid. Yet another example can be seen in the cross-sectional profiles of agates, where banding with growth thickness is common; agates, also referred to as fibrous quartz, grow in a manner analogous to that of thin films.^{40,41} Although the literature is not clear on this matter, concurrent energetic particle bombardment during film growth appears to lead to increased surface smoothening,³² possibly due to control of the nucleation as well as the renucleation densities. Recent attempts to simulate the two-dimensional cross-sectional growth by computerised geometric constructions,⁴² which include nucleation clusters, competition for cluster growth, and renucleation, demonstrate the possibility of initial roughening, and subsequently eventual smoothening, during growth. In particular, such smoothening occurs only at very high, but realistic, values of renucleation.

Cantor-like dusts

After the preceding discussion on the structure of films, we turn our attention to the dynamics of the growth processes. First, some mathematical preliminaries. Let a bi-variate function be defined as a collection of disparate Dirac delta functions through the usual fractal notions of the *initiator* and the *generator*. The initiator is the ensemble:

$$f(\mathbf{x};\mathbf{a}) = \sum_i \delta(\mathbf{x} - \mathbf{a} \cdot \mathbf{x}_{fi}), \quad i \in \{1,2,\dots,M\}$$

in the two-dimensional space $\mathfrak{R}^2: \{\mathbf{x} \mid \mathbf{x} = [x,y]; x,y \in \mathfrak{R}^1\}$; \mathbf{a} is some scaling vector while \mathbf{x}_{fi} is the i -th location on or inside the unit circle. The generator is also similarly defined:

$$g(\mathbf{x};\mathbf{a}) = \sum_i \delta(\mathbf{x} - \mathbf{a} \cdot \mathbf{x}_{gi}), \quad i \in \{1,2,\dots,N\}$$

with \mathbf{x}_{gi} also lying on or inside a unit circle. Then, a cluster (Cantor-like dust) of evolutionary level E , $E \geq 1$, is defined recursively as:

$$p_E(\mathbf{x}) = p_{E-1}(\mathbf{x}) * g(\mathbf{x};\mathbf{a}_E); \quad p_0(\mathbf{x}) = f(\mathbf{x};\mathbf{a}_0),$$

in which $*$ denotes the spatial convolution.⁴³ With specific conditions on the scaling vectors \mathbf{a}_E , and on the locations \mathbf{x}_{fi} and \mathbf{x}_{gi} , these clusters can be made self-affine, or

even self-similar;⁴⁴ randomisation can be implemented to a certain extent by choosing different x_{gi} at different levels.

The point to be made here is as follows: Suppose at a given level of evolution, the cluster contains a particle at some site x_0 . Another particle floats down, as a result of the growth process, and attaches itself to some site x' which is not appreciably far from x_0 , i.e. $|x_0 - x'|$ is much smaller than any other dimension of the cluster. The cluster then could appear to have an *extended* particle which stretches from x_0 to x' . If such occurrences are allowed to take place several times in several locales, the cluster would then appear to be piecewise continuous over several dense sets of points. In other words then, the tendency towards macroscopic homogeneity would be apparent, even though the cluster would be generated from a *fractal* process. In fact, this tendency would come in at some level E , if the scaling vectors $a_E, a_{E+1}, a_{E+2}, \text{etc.}$, are all equal to or less than unity in magnitude.

These clusters have some relevance to the computer simulations of diffusion-limited aggregation as well as ballistic aggregation.²⁷ Suppose there exists a volume, which has internal voids, and which is receptive to further growth. Should not the further acquisition of an incoming particle be influenced by the existing volume as a whole, and not simply by the individual particles to one of which the new particle would adhere to? One envisions, therefore, that these aggregation processes are not as simple as their computer models, because the evolution of these aggregates would certainly have a non-local flavour which has not been simulated on the computers. Perhaps, one of the consequences of non-locality of the cluster-particle interactions would be the creation of a smoother (i.e., less discontinuous) 'continuum'.

More accurately, perhaps, after some initial growth, the cluster-cluster aggregation process would be more important than particle-particle aggregation in order to explain dendritic growth; for example, morphogenetic studies on collagen fibrils suggest that the cornea may have a fractal structure.⁴⁵ In computer simulations, the cluster-cluster aggregation process is usually ignored (but see ref.27). For example, in the snowflake algorithm of Miyazima and Tanaka,⁴⁶ dendritic growth appears on a dense (Euclidean) hexagonal nucleus. This dendritic growth can, with some stretching of the imagination, explain the snowflake in Figure 4a of Miyazima and Tanaka, but not the snowflake depicted in their Figure 4b. On the other hand, the interaction of small clusters could explain very well either of the two natural snowflakes. That conclusion would be considerably assisted by the concept of reversibility, i.e. if the inter-particle bonds have some large, but finite, lifetimes, which would tend to smear out ultrafine detail. In the evolution of large systems, therefore, the history of the growth process cannot be easily ignored without dire consequences.

Reversibility *versus* irreversibility

Most of the physical systems identified as having an underlying fractal geometry have been simulated on computers using the requisite of irreversibility (but see refs 47-49 for recent incorporations of reversibility in computer simulations). Irreversible growth processes occur far from equilibrium at high values of the driving force for the phase transition, as shown in Figure 2. This figure shows the four main morphological responses of a transition to solid state as a function of $\Delta\mu$, which is

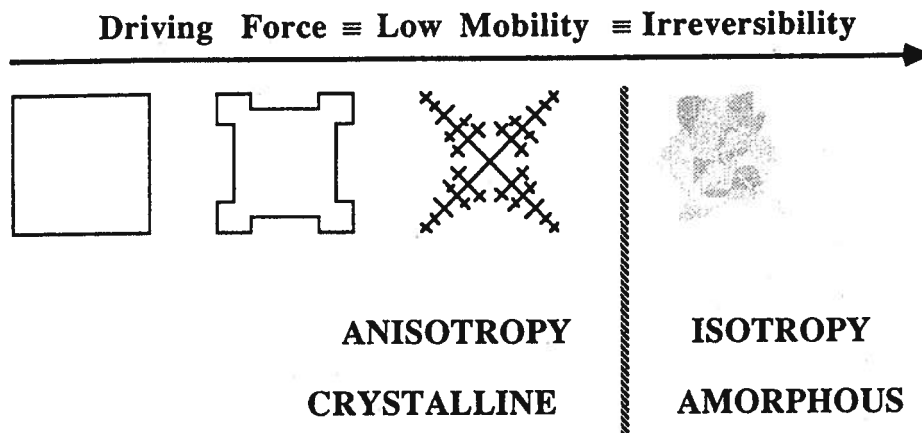


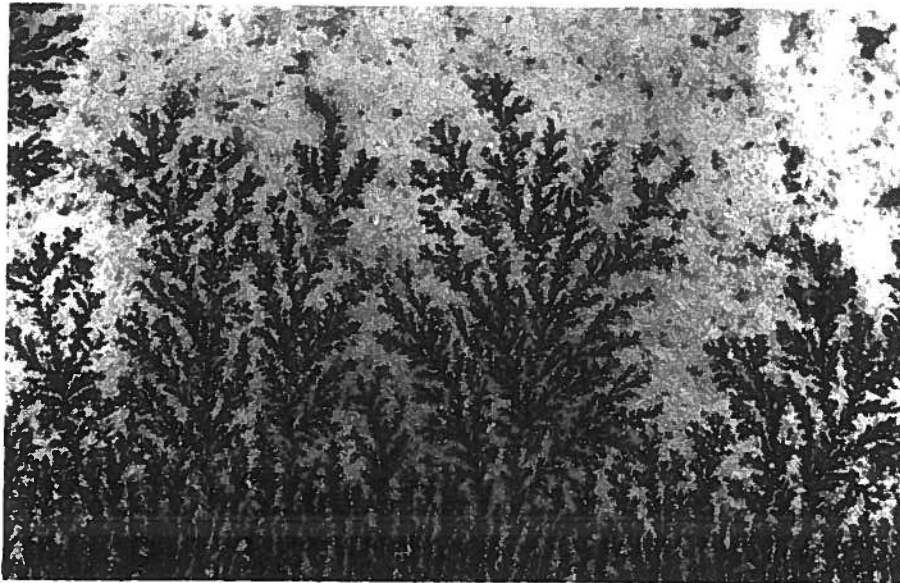
Figure 2 Schematic illustration of the road to equilibrium for single clusters.

the difference between the chemical potentials of the non-aggregated and the aggregated states. To the left of the dashed line, the morphology is controlled by a three-dimensional periodic lattice; therefore, its symmetry belongs to one of the 32 crystallographic classes of point symmetry. On the other hand, to the right of the dashed line, the morphology does not possess any basic rational or non-rational periodic lattice. The transition across the dashed line is controlled by the compromise between the rate of aggregation of particles and the system's ability to relocate them to the low-energy sites, *i.e.* the particle mobility. The transition between the three distinct crystalline categories of the morphology is regulated by the competition between the rates of three types of growth mechanisms: (a) dislocation growth (low $\Delta\mu$), (b) two-dimensional nucleation growth (mid-range $\Delta\mu$), and (c) continuous growth (high $\Delta\mu$). Experimental evidence of such transitions is available elsewhere.⁵⁰

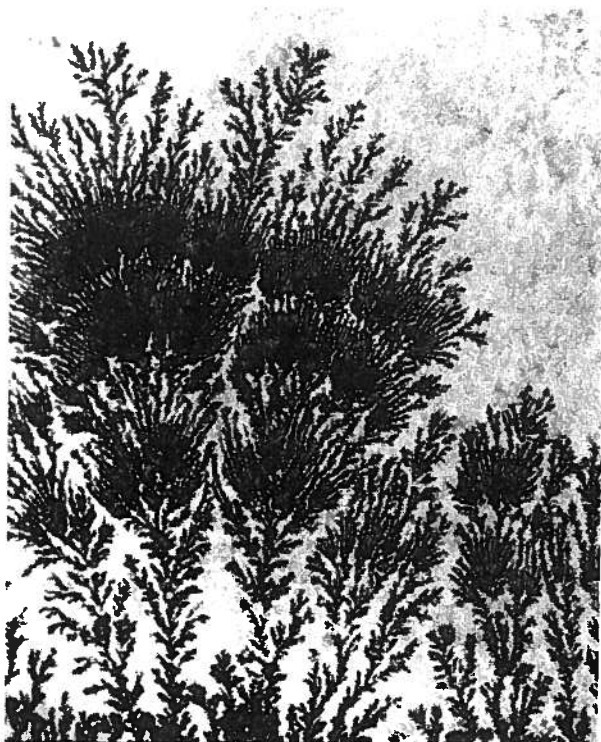
There is also a third factor influencing the competition. It appears when, unlike in the previous cases, we consider the behaviour of multi-macrocluster systems. Then, the whole morphology is controlled by geometrical selection, *i.e.* the competition for available growth spaces between the neighbouring macroclusters, as well as between the neighbouring microclusters constituting the macroclusters. As can be easily envisaged, an unlimited number of possible configurations are realisable because the growth is influenced by several factors:

- (a) the density of seeds, or of the nucleation sites,
- (b) the geometric arrangement of these seeds,
- (c) the surface roughness of the substrate (in particular, the relative position of the nucleation sites in relation to the roughness),
- (d) the flow direction of the incoming particles, *e.g.*, the flow lines of the diffusion field,
- (e) the structural interaction (epitaxy) between the seeds and the substrate, and
- (f) the crystal structure, if any, of the seeds.

The growth of thin films¹⁵ illustrates some of these considerations rather well. An additional example is the geometric selection in the growth of malachite (copper carbonate) spherulites, in which, again, the cone sizes do not grow indefinitely (see Figure 193 of ref.51).



(A)



(B)

Figure 3 Dendritic patterns naturally formed by (MnO_2) in limestone. In (A), note the similarity of the pyrolusite dendrites with the concept of fractal branching. The competition due to successive re-nucleation steps (Liesegang's banding) tends to eliminate self-affinity in (B).

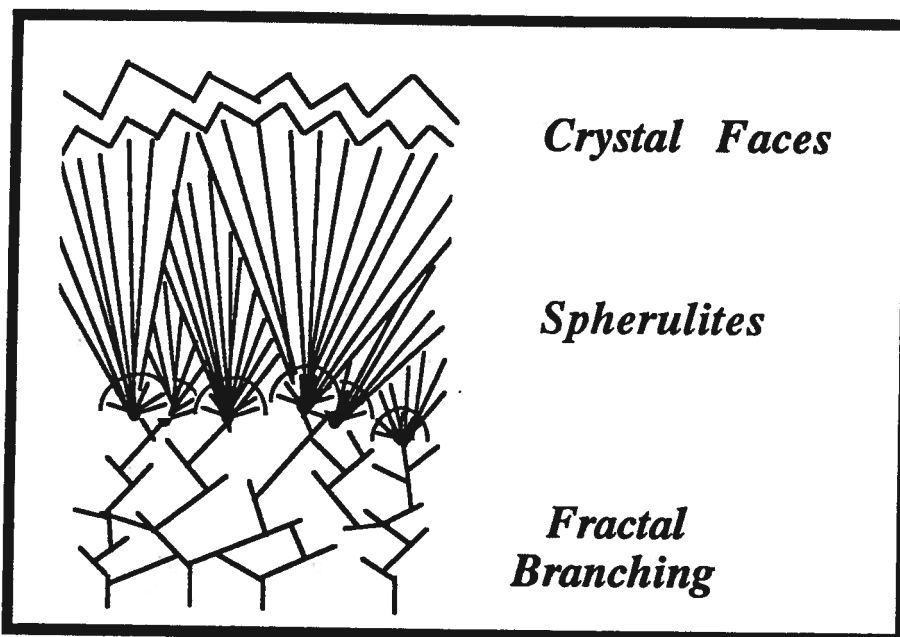


Figure 4 Schematic illustration of the road to equilibrium for multi-cluster growth. Competition for growth is conspicuous at all three stages.

When the purpose of the computer studies is to simulate natural structures with high fidelity, it is important to consider two more points:

(1) Most natural structures, and even laboratory ones, evolve to equilibrium. Therefore, reversibility, mobility, and desticking increase during the growth history; we do not consider here the classical Lemlein–Kliya experiment^{52,53} in which evolution to equilibrium morphology is a result of the surface reorganisation aimed to minimise surface tension. Thus, the mechanisms controlling the morphology are subject to changes. This occurs in growth characterised by either or both, single- as well as multi-clusters. Examples for the single-cluster growth include the case of PbS growth;⁵⁰ and that of the snowflakes.⁴⁶ In particular, Figure 4b of the latter reference shows a clear change from a continuum (DLA mode) to a two-dimensional nucleation-controlled growth. To illustrate the growth of multiclusters, we will choose the very interesting case of pyrolusite ‘fractals’ of Figure 3. A property of the diffusing-reacting systems is the production of hysteresis cycles of $\Delta\mu$ leading to an oscillatory behaviour of the precipitation rate; and therefore, to a patterned structure.⁵⁴ Recently, such an oscillatory pattern has also been found to affect the growth rates of single crystals under diffusion transport.⁵⁵ Clearly, such a structure is observable in Figure 3 where the banding destroys self-similarity. In Figure 4, the morphologies of different evolutionary systems are schematically illustrated.

(2) As pointed out earlier, the assumption of irreversibility is an intrinsic characteristic of most computer simulations devoted to explain the formation of aggregates. Typically these algorithms begin with a seed in two-dimensional space, which may or may not be latticed; then, particles aggregate ‘around’ the seed by a diffusion-limited process.⁵⁶ It turns out that such algorithms are pliable to fractal

analysis. It is, however, important to note that irreversibility derives from high $\Delta\mu$ values. This implies that most of the particles form clusters whose sizes are lower than that of the critical nucleus; a critical nucleus is stable and can grow independently.⁵⁷ To the best of our knowledge, unfortunately there is no formal treatment available in literature that deals with the distribution statistics of clusters of different sizes as a function of $\Delta\mu$, other than the classical nucleation theory. This theory relies on the assumption that the total number of clusters is much smaller than the number of unclustered particles in the system; it must be noted that this assumption is invalid for rigorous studies of systems far from equilibrium, *e.g.* those simulated by diffusion-limited aggregation. However, it is plausible to assume in computer simulations that isolated particles would not be available for seed growth. Thus, Meakin's cluster-cluster aggregation model²⁷ introduces physical meaning to the earlier simulations of diffusion-limited aggregation. The situation remains unchanged when one deals with the snowflake problem,⁴⁶ but now it would be necessary to introduce sticking probabilities related to symmetry on both seeds and micro-clusters.

Surfaces of growing crystals

The transition from fractal structures with rough surfaces to crystal aggregates, and even to single crystals with smooth surfaces, is a ubiquitous morphological response of natural systems when they go from irreversibility to equilibrium. A good example is furnished by the growth of quartz druses from chalcedony spherulites, or from randomly oriented seeds. As observed by Grigor'ev,⁵¹ the number of crystals in a druse is maximum at its base, while it rapidly decreases towards the upper region. In fact, on replotting the graph in Figure 170 on page 193 of Grigor'ev's book on the log-log scale, it can be observed that a linear relationship exists between the log of the distance from the overgrowth surface and the log of the average distance between the crystals. Therefore, as a result of the competition, the surface gets smoother.

Another good example is the naturally occurring growth of pyrolusite (MnO_2) dendrites in limestone. As shown in Figure 3, the morphological pattern of such mineralogical systems is a typical example of fractals (see, for example, plate 56 of ref.1). However, in areas where the amount of pyrolusite is smaller, the morphology is very convoluted; conversely, in locales where the dendritic pattern becomes denser, the surface gets smoother because of the increased competition between the dendrites.

It is worth noting that the rate at which the above-mentioned systems evolved – low rates for quartz druses, and high ones for the MnO_2 dendrites, – is not significant in the evolution processes; and competition between the growth elements turned out to be the predominant factor.

Concluding remarks

We conclude with the following remarks on the evolution of natural aggregation processes: In the beginning, there is extremely short-range order and the seeding may be considered 'random'. Further growth is only piecewise self-similar, *i.e.* it is globally self-affine.^{58,59} particles form small clusters. Then, long-range order begins to assert itself as small clusters interact with each other and influence the accretion

of more particles in a materially significant manner; the tendency towards macroscopic homogeneity starts to manifest itself; the topological (fractal) measures begin to tend towards the Euclidean dimensions. Fractals, thus, are band-limited, and the competition for further growth among the elements eventually brings in homogeneity and destroys the self-affinity present at the microscopic scales.

We end with Alexander Pope:

"Where order in variety we see,
and where, though all things differ, all agree."

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References

- 1 Mandelbrot, B.B. 1983. *The Fractal Geometry of Nature*. Freeman, New York.
- 2 La Breque, M. 1987. *Mosaic*, 18, 22–41.
- 3 Stanley, H.E. and Ostrowsky, N. (eds) 1986. *On Growth and Form*. Martinus Nijhoff, Dordrecht.
- 4 Douady, A. 1986. In: Peitgen, H.-O. and Richter, P.M. (eds), *The Beauty of Fractals*. Springer, Berlin.
- 5 Pietronero, L. 1987. *Physica*, 144A, 257–284.
- 6 Kadanoff, L.P. 1986. *Physics Today* (February issue), 6–7.
- 7 Uwaha, M. and Saito, Y. 1988. *J. Phys. Soc. Jpn.*, 10, 3285–3288.
- 8 Lakhtakia, A., Messier, R., Varadan, V.K. and Varadan, V.V. 1987. *J. Phys. A.*, 20, L735–738.
- 9 Philippou, A.N., Georghiou, C. and Philippou, G.N. 1985. *Fibonacci Q.*, 23, 100–105.
- 10 Holter, N.S., Lakhtakia, A., Varadan, V.K., Varadan, V.V. and Messier, R. 1986. *J. Phys. A*, 19, 1753–1759.
- 11 Wolfram, S. 1983. *Rev. Mod. Phys.*, 55, 600–631.
- 12 Kondoh, H., Matsushita, M. and Fukuda, Y. 1987. *J. Phys. Soc. Jpn.*, 56, 1913–1915.
- 13 Meakin, P. 1983. *Phys. Rev. B.*, 28, 6718–6732.
- 14 Turkevich, L.A. and Scher, H. 1986. In: Pietronero, L. and Tosatti, E. (eds), *Fractals in Physics*, pp.223–229. North-Holland, Amsterdam.
- 15 Messier, R. and Yehoda, J.E. 1985. *J. Appl. Phys.*, 58, 3739–3746.
- 16 Yamada, T. 1986. *Fluid Dynamics Res.*, 1, 163–164.
- 17 Messier, R. 1986. *J. Vac. Sci. Tech. A*, 4, 490–495.
- 18 Ross, R.C., Johncock, A.G., and Chan, A.R. 1984. *J. Non-Cryst. Solids*, 66, 81–86.
- 19 Roy, R.A. 1985. Evolution of Morphology in Amorphous and Crystalline SiC Sputtered Films. Ph.D. Thesis, Pennsylvania State University.
- 20 Wade, R.H. and Silcox, J. 1966. *Phys. Stat. Sol.*, 19, 57–62; 1967. *Appl. Phys. Lett.*, 8, 7–10.
- 21 Pilione, L.J., Vedam, K., Yehoda, J.E., Messier, R. and McMarr, P.J. 1987. *Phys. Rev. B.*, 35, 9368–9371.
- 22 Yang, B., Pilione, L.J., Yehoda, J.E., Vedam, K. and Messier, R. 1987. *Phys. Rev. B.*, 36, 6206–6208.
- 23 McMarr, P.J., Blanco, J.R., Vedam, K., Messier, R. and Pilione, L.J. 1986. *Appl. Phys. Lett.*, 49, 328–330.
- 24 Kim, S.Y. and Vedam, K. 1989. *Thin Sol. Films* 166, 325–344.
- 25 Henderson, D., Brodsky, M.H. and Chaudhari, P. 1974. *Appl. Phys. Lett.*, 25, 641–643.
- 27 Meakin, P. 1987. *CRC Crit. Rev. Sol. State Mater. Sci.*, 13, 143–189.
- 26 Dirks, A.G. and Leamy, H.J. 1977. *Thin Sol. Films*, 47, 219–233.
- 28 Krishnaswamy, S.V., Messier, R., Ng, Y.S., Tsong, T.T. and McLane, S.B. 1980. *J. Non-Cryst. Sol.*, 35/36, 531–536.

- 29 Krishnaswamy, S.V., Messier, R., McLane, S.B., Ng, Y.S. and Tsong, T.T. 1981. *Thin Sol. Films*, **79**, 21–26.
- 30 Messier, R. and Ross, R.C. 1982. *J. Appl. Phys.*, **53**, 6220–6225.
- 31 Roy, R.A. and Messier, R. 1984. *J. Vac. Sci. Technol. A*, **2**, 312–315.
- 32 McNeil, J.R., Wei, L.J., Al-Jumaily, G.A., Shakir, S. and McIver, J.K. 1985. *Appl. Opt.*, **24**, 480–485.
- 33 Rikvold, P.A. 1982. *Phys. Rev. A*, **26**, 647–650.
- 34 Nittmann, J. and Stanley, H.E. 1986. *Nature*, **321**, 663–668.
- 35 Berry, R.M., Hall, P.M. and Harris, M.T. 1968. *Thin Film Technology*, Van Nostrand, Princeton. Chap.5.
- 36 Owen, L.W. 1967. *Trans. J. Plastics Inst.*, **35**, 437–441.
- 37 Nakahara, S. 1977. *Thin Sol. Films*, **45**, 421–432.
- 38 Lepetre, Y., Schuller, I.K., Rasigni, G., Rivoira, R., Philip, R. and Dhez, P. 1985. *Proc. SPIE*, **563**, 258–263.
- 39 Gartner, G., Janiel, P. and Lydtin, H. 1987. *6th Intl. Conf. on Ion and Plasma Assisted Technologies*, pp.17–22. Brighton, UK.
- 40 Jones, F.T. 1952. *Am. Min.*, **37**, 578–587.
- 41 Frondel, C. 1978. *Am. Min.*, **63**, 17–27.
- 42 Yang, B., Walden, B., Messier, R. and White, W.B. 1987. *Proc. SPIE*, **821**, 68–76.
- 43 Goodman, J.W. 1968. *Introduction to Fourier Optics*, Ch. 2. McGraw-Hill, New York.
- 44 Lakhtakia, A., Messier, R., Varadan, V.V. and Varadan, V.K. 1986. *Phys. Lett. A*, **118**, 54–58.
- 45 Trelstad, R.L., Hayashi, M., Hayashi, K. and Birk, D.E. 1987. In: Sheffield, J.B. and Hilfer, S.R. *The Microenvironment and Vision*, pp.1–27. Springer, New York.
- 46 Miyazima, S. and Tanaka, T. 1987. *J. Phys. Soc. Jpn.*, **56**, 441–443.
- 47 Meakin, P. 1985. *J. Chem. Phys.*, **83**, 3645–3649.
- 48 Meakin, P. and Deutch, J.M. 1985. *J. Chem. Phys.*, **83**, 4086–4092.
- 49 Kolb, M. 1986. *J. Phys. A: Math. Gen.*, **19**, L263–L268.
- 50 García-Ruiz, J.M. 1986. *J. Crystal Growth*, **75**, 441–453.
- 51 Grigor'ev, D.P. 1961. *Ontogeny of Minerals*, Daniel Davey, New York.
- 52 Lemlein, G.G. 1954. *Dokl. Akad. Nauk. SSSR*, **98**, 873–877.
- 53 Kliya, M.O. 1956. *Kristallografiya*, **1**, 557–560.
- 54 Henisch, H.K. and García-Ruiz, 1986. *J. Crystal Growth*, **75**, 203–211.
- 55 García-Ruiz, J.M., Santos, A. and Alfaro, E. 1987. *J. Crystal Growth*, **84**, 555–558.
- 56 Witten, T.A. and Sander, L.M. 1983. *Phys. Rev. B*, **27**, 5686–5688.
- 57 Zettlemoyer, A.C. (ed.). 1969. *Nucleation*. Dekker, New York.
- 58 Mandelbrot, B.B. 1986. In: Pietronero, L. and Tosatti, E. (eds), *Fractals in Physics*, pp.3–15. North-Holland, Amsterdam.
- 59 Lakhtakia, A., Messier, R., Varadan, V.K. and Varadan, V.V. 1986. *J. Phys. A*, **19**, L985–L989.