

Diffusion-Reorganized Aggregates: Attractors in Diffusion Processes?

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A process based on particle evaporation, diffusion, and redeposition is applied iteratively to a two-dimensional object of arbitrary shape. The evolution spontaneously transforms the object morphology, converging to branched structures. Independently of initial geometry, the structures found after a long time present fractal geometry with a fractal dimension around 1.75. The final morphology, which constantly evolves in time, can be considered as the dynamic attractor of this evaporation-diffusion-redeposition operator. The ensemble of these fractal shapes can be considered to be the *dynamical equilibrium* geometry of a diffusion-controlled self-transformation process.

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This Letter reports the discovery of a diffusion-mediated process which spontaneously builds a dynamic fractal *equilibrium* structure, in contrast with fractal morphologies linked to *far-from-equilibrium* processes [1–3]. The process is a surface to surface evaporation-diffusion-condensation process which conserves the total mass of the system. During the time evolution, one observes a progressive transformation of the surface through bulk diffusion. After a long relaxation period, the system reaches a dynamical fractal structure. This structure appears as the final equilibrium state towards which *any* initial morphology of M particles will converge after sufficient evaporation-diffusion-condensation iterations. It can then be considered as a general statistical attractor for that specific dynamic process.

The underlying ideas that have suggested this study come from our knowledge of the basic mathematical objects which govern the exchange of Laplacian driven currents across irregular (or fractal) interfaces (as, for example, in the study of irregular or fractal electrodes). This problem can be mapped onto the study of the transfer of Brownian particles across irregular membranes with finite permeability [4]. In this process, Brownian particles strike an irregular surface where they are absorbed with finite probability. When reflected, the particles undergo successive random paths, hitting and hitting again the static surface, until they are finally absorbed. Halsey first indicated that the response of such systems depends on the probability that a particle starting on the interface comes back to it [5]. Generalizing these ideas, it has been recently shown that the Laplacian transfer across irregular interfaces is controlled by a single linear operator \tilde{Q} which maps the *static* surface onto itself through effective “Brownian bridges” [6]. In this context, each surface has an operator \tilde{Q} , which is symmetric and positive.

The question arises naturally of the link between the Brownian bridges and a real diffusion process. This led us to study a dynamical process in which the notion of a \tilde{Q} operator, which maps the surface onto itself through average diffusion, is transformed into a real operation which transforms the surface through discretized diffusion [7]. The

evolution mechanism then proceeds in the following steps (Fig. 1): (1) A particle is chosen at random on the surface of an initial structure and is dissolved, occupying an empty site next to its initial position. (2) The dissolved particle may jump back to its original site or start a Brownian random walk on a square lattice which represents the outer medium. The time step for each jump is $\tau = 1$. This motion is stopped when the particle hits the structure again as it sticks on first hit [8]. (3) The process, which conserves the total mass, is then iterated. For practical reasons, the system is enclosed in a large square window: whenever a diffusing particle strikes the window boundary, this particle is reinitiated on another site of this external box, following a probability law that simulates the random walk in the infinite outer medium. As the probability to return to a starting site is equal to 1 in $d = 2$, this does not modify the morphologies which are generated by the process. The existence of the external box only perturbs the time scale so that the number of computer steps is not proportional to a real time. For this reason, the number of steps will be called pseudotime t .

The dissolution-recondensation process rapidly creates several branches. If particles of these branches were allowed to dissolve, it would lead to a progressive splitting of the structure into two, then many, disconnected parts. In order to avoid this progressive disconnection, one imposes

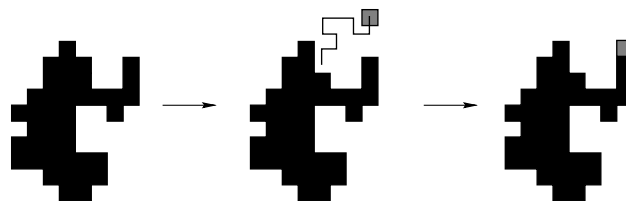


FIG. 1. Schematic of the self-diffusion reorganization process. Step (1): a particle on the boundary of the structure is randomly chosen to be dissolved, according to the rule that it does not break the connectivity of the structure. Step (2): it can go back to its original position or starts a random walk on a square lattice. If the walk brings the particle to a surface site, the particle sticks to the structure under the condition that the new position conserves the simple connectivity of the structure.

the structure to remain connected throughout the process. The only particles allowed to evaporate are then the particles pertaining to the surface of the structure and that are not “red particles”: the red particles are defined as the particles which, if eliminated, would disconnect the structure in two or more distinct parts. The particles allowed to evaporate are therefore called “blue particles.” We also impose that an evaporated particle cannot stick on a site where it would disconnect the outer medium. In fact, one could release this last constraint since it does not modify the long term evolution of the structure. It simply avoids creating temporary “lakes” in which particles evaporating and redeposited would greatly reduce the speed of evolution of the structure. Particles on the internal surface of these lakes would spend most of the time going back and forth until the lake opens spontaneously.

The results of the spontaneous evolution of structures obtained with different initial morphologies are shown in Fig. 2. The left column shows the initial shape and the corresponding final morphologies are shown on the right. In case (a), the initial morphology is a compact dense struc-

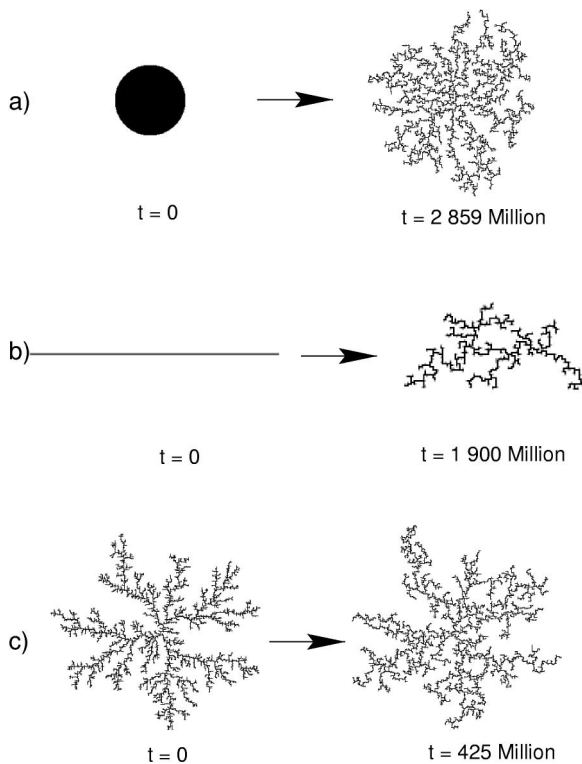


FIG. 2. Three examples of the spontaneous evolution of morphologies towards statistical equilibrium shapes under the diffusion limited reorganization process. Case (a): the initial morphology is dense with dimensionality $D_f = 2$, and contains 7800 particles. Case (b): initial morphology with dimensionality $D_f = 1$, 983 particles. Case (c): initial DLA morphology with dimensionality $D_f = 1.65$, 8000 particles. The final morphologies look all the same and keep the same statistical characteristics after being formed. The time for evolution strongly depends on the initial shape.

ture described by a dimensionality equal to 2. In case (b), the starting shape is a line with dimensionality 1. Finally, in case (c), the initial morphology is an ordinary diffusion-limited aggregate (DLA), first introduced by Witten and Sander [9].

In all three cases (a), (b), and (c), the final morphologies present branched structures, like DLA but less open. The complete statistical study of the morphologies is difficult because the process is very time consuming in the computer. For instance, the time necessary to reach the morphology shown in Fig. 2(a) is of the order of several weeks of CPU time on a Hewlett-Packard C160 workstation. The similarity in the final shapes, independent of initial geometry, suggests that this class of morphologies is an attractor for such self-reorganization processes. We call these structures “diffusion-reorganized aggregates” (DRA).

The fact that morphologies are “attracted” by the fractal final form is also indicated in Fig. 3. The figure gives the determination of the fractal dimension of the final morphologies starting, respectively, with initial dimension $D_f = 2$, $D_f = 1$, and $D_f = 1.65$ [Figs. 2(a), 2(b), and 2(c)]. Although the initial dimensions are very different, the final plots are within reasonable uncertainty

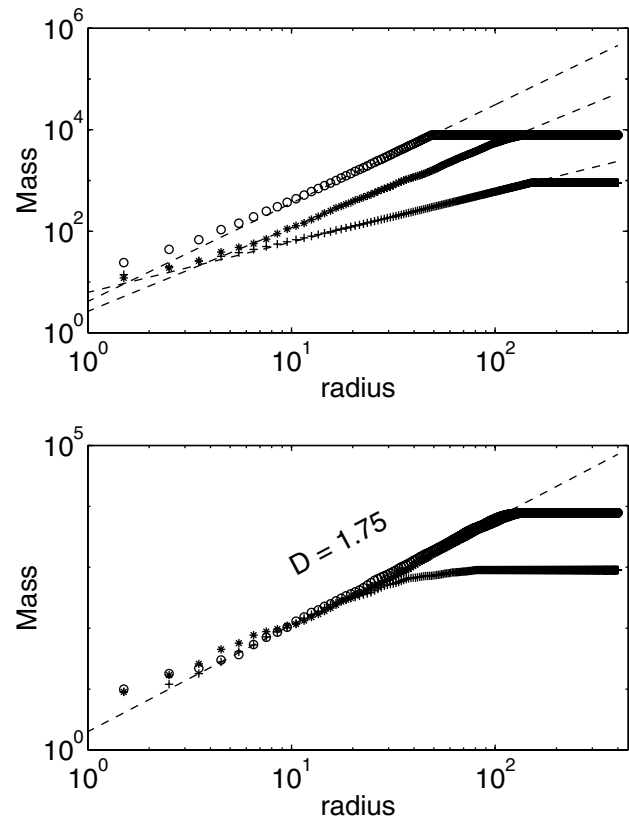


FIG. 3. Evolution of the fractality. Top: mass to radius relation for initial morphologies (a), (b), and (c) defined in Fig. 2 [(a) $\rightarrow \circ$, (b) $\rightarrow +$, (c) $\rightarrow *$]. Bottom: mass to radius relation for the corresponding stationary states. This plot indicates that, independent of the initial dimension, the stationary dimensions are very close.

concentrated and compatible with $D_f = 1.740 \pm 0.02$, a value which is significantly different from the fractal dimension of lattice animals [10] and compatible with that of the branched structures which are the results of a far-from-equilibrium process [11].

The dynamics of the restructuring process has several specific properties. To follow the dynamics, one can compute two memory functions. First is a density-density memory function. For this purpose, the structure is characterized by a number s_i on each site i of the lattice, with $s_i = 1$ if the site is occupied by the structure and 0 if not. Calling M the mass of the initial structure, which is equal to the number of occupied sites, this memory function can be defined as

$$C(t) = \frac{1}{M} \sum_{\text{lattice sites } i} s_i(0)s_i(t). \quad (1)$$

This first function does not discriminate between particles as $s_i(0)s_i(t)$ is equal to 1 if a lattice site i is occupied both at pseudotimes 0 and t (by any particle) and 0 otherwise. The pseudotime evolution of $C(t)$ is shown in Fig. 4. One observes a memory loss which does not go to zero. There can be two reasons for incomplete decorrelation. First, it could happen that a part of the object is not restructured. Second, it could be that the density-density memory does not vanish due to an average statistical overlap, even if the structure has completely forgotten its initial geometry. To clarify this point, one can compute a particle-particle memory function which characterizes the speed at which the particles of a given

structure finally move from their initial positions. This second function is

$$P(t) = \frac{1}{M} \sum_{\text{particles } j} \delta(\vec{r}_j(0) - \vec{r}_j(t)), \quad (2)$$

where $\vec{r}_j(t)$ is the position of the particle j at time t .

The evolution of $P(t)$ is also given in Fig. 4. One observes that the particle-particle memory is lost to zero very rapidly. When it has reached very small values, one can say that the structure has completely lost the memory of its microscopic configuration. Apart from these two rapid processes, one observes in Fig. 4 the very slow global evolution. Note that the DRA geometry loses constantly its memory whereas the DLA growth keeps the total memory of its previous structure.

A more detailed characterization of the rapid regimes is displayed in Fig. 5, where the decorrelations are found to be exponential, an unusual time behavior in this context.

Those results, together with the fractality evolution of Fig. 3, indicate that the final DRA morphologies behave as stable fixed points. The constantly changing DRA can then be viewed as a statistical equilibrium state. In that sense, it is analogous to the homogeneous state of an ideal gas, where the microscopic interactions between particles lead to a statistical macroscopic equilibrium, although it is constantly changing its microscopical structure.

One should also note that the transformation of the \tilde{Q} operator, which maps the surface onto itself through average diffusion, into a real operation is analogous to the dielectric breakdown model (DBM) of DLA [12]. In the DBM model, the operator, that maps the source at infinity onto the growing morphology through average Brownian paths, is transformed into a real stochastic operation. In the model presented herein, the source is the object itself.

It is also interesting to return to the exact concept of the self-transport operator attached to a given morphology [6]. Extending the notion of diffusive self-transport in

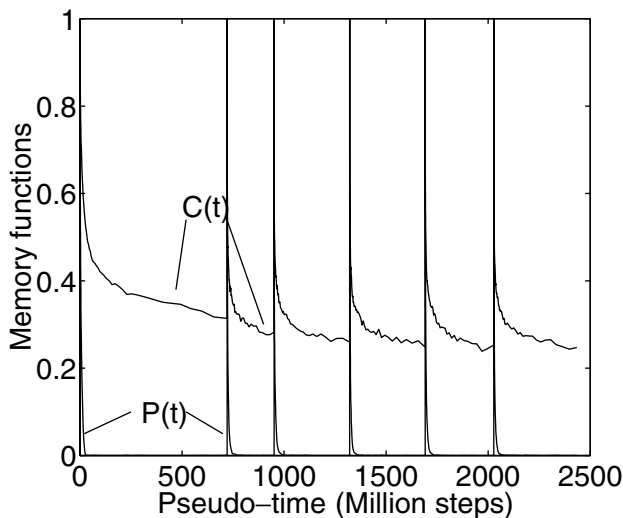


FIG. 4. Time evolution of the memory functions $C(t)$ and $P(t)$ during the evolution of the disk towards DRA structure [Fig. 2a]. These functions are defined by Eqs. (1) and (2). Both functions are reset $5\times$ during the evolution (positions of the peaks). After 1×10^6 time steps, the correlation $C(t)$ is less than $1/2$, indicating that more than half of the initial points have moved and no more belong to the initial structure.

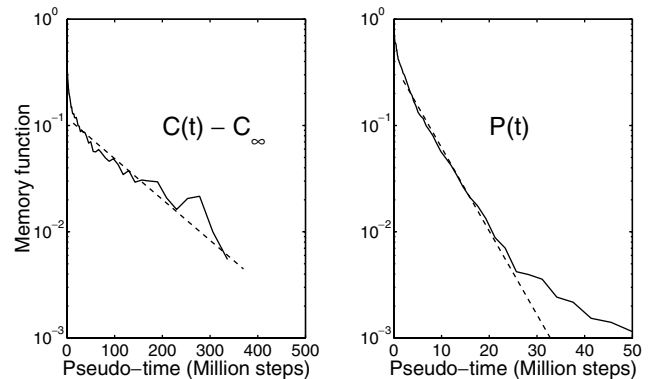


FIG. 5. Detailed behavior of the memory functions decays $C(t) - C(\infty)$ and $P(t)$. In both cases, the final decay is found to be exponential, with characteristic times of $\tau_C \approx 1.1 \cdot 10^8$ and $\tau_P \approx 5 \cdot 10^6$. This suggests a classical equilibrium situation.

this context, it might be that DRA shapes could be considered as “eigenshapes” of these operators. By this, it is suggested here that the self-transport operators attached to these structures transform those structures into themselves, of course in a statistical sense.

All these results should be confirmed in the future through extensive numerical simulations and extension to diffusive self-transport in $d = 3$. Some future extensions of this work can be envisaged. First, one should verify the universal character of this result. Second, if surface energy was included, it would be interesting to study how the equilibrium morphology would depend on its value. Last, one should study whether the self-transport operators \tilde{Q} of these geometries possess some extremal properties.

In summary, it has been shown that diffusive self-transport statistically modifies an object morphology in an irreversible manner. This irreversible process leads to a class of branched structures, called DRA, with fractal dimension around 1.75. This class of structures plays the role of a statistical attractor. Moreover, its attraction basin seems to be the whole configuration space in the sense that, independent of the initial geometry of the system, the dissolution-redeposition process always tends to create the same category of geometries. The fractal DRA can be seen as a statistical equilibrium for this process, which would maximize a “geometrical entropy” still to be defined.

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