Fractal Dimension of Generalized Diffusion-Limited Aggregates

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A Flory-type argument based on a branched-polymer topology is presented for the fractal dimension $D(d, d_w)$ of diffusion-limited aggregates formed by a random walk with fractal dimension d_w.

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Randomly branched aggregates occur in many fields of physics such as branched-polymer physics,[1-4] the sol-gel transition,[5] percolation,[6] turbulence,[7] nucleation,[8] the formation of smoke particles,[9] and electric breakdown.[10] What is remarkable about these objects is that despite their random appearance they all show a strong measure of self-similarity. This in part can be categorized for isotropic aggregates by the scaling behavior of the number of particles in the aggregate N with its radius of gyration R as $N \sim R^{D(d)}$, where D is its fractal dimension[11] and d the Euclidean dimension of the space in which the cluster is grown.

The question then arises as to what are the similarities and differences between differently formed random aggregates. In order to answer this question Witten and Sander[12] introduced a diffusion-limited model. In this model a seed particle is placed at a lattice site and a random walk is allowed to approach it isotropically from a long distance off. If it reaches a nearest-neighbor site, it becomes part of the growing cluster; if not, it is killed off. In this way the cluster grows in a kinetically irreversible manner which may be a good model for instance for smoke particle formation. They carried out computer simulations in $d = 2$, which were extended to higher dimensions ($d = 2$ to 6) by Meakin.[13] Random dendritelike structures were found with a fractal dimension $D \approx 5d/6$. This diffusion-limited model can be generalized[14] by allowing, instead of a normal random walk of fractal dimension $d_w = 2$, a walk to have a general d_w. Thus the fractal dimension is now $D(d, d_w)$, and it is this case we consider here.

In order to get a relation of the type $N \sim R^{D(d, d_w)}$, it is natural to try and find a Flory-type argument which has proved so successful in the case of linear and branched polymers. This would not only give us $D(d, d_w)$ but also the upper critical dimensionality $d_c(d_w)$ if it exists. Also a comparison of the result with the theory for branched polymers should then show whether they belong to the same universality class.[15] When we try to apply such an argument to diffusion-limited aggregates we are faced by two problems.

First there does not appear to exist a free energy in the conventional meaning of the term —there exist no attractive or repulsive interactions between particles forming part of the cluster. In fact the aggregate grows nonergodically —any single aggregate of radius R at radii $r < R$ has essentailly a frozen-in structure, and therefore does not sample phase space as a branched polymer in a solvent would. This is because (and this can clearly be seen in computer simulations) all additional particles added to the growing cluster penetrate no further than a small interfacial region. Nevertheless this is not a real problem as there does exist a well-defined probability for formation for any aggregate of any given configuration which can be found by many repetitions of the growth process, and so long as the asymptotic $N \to \infty$ properties of a single growing aggregate are the same as this ensemble average, the use of a Flory argument should still be valid. The free energy $F(R, N)$ which we shall estimate is in fact to within a constant term simply the negative of the logarithm of the total probability $P(R, N)$ for the formation of any cluster of N particles with radius of gyration R, or $P(R, N) \sim \exp[-F(R, N)]$.

Secondly, and more importantly for the actual calculation of $D(d, d_w)$ is that the generalized diffusion-limited cluster is only defined through the algorithm for its method of formation. However, the essential properties of the structure of the aggregate itself are required for a Flory-type argument. Before suggesting what these appear to be, let us consider the physics which causes the open fractal structure observed—this is screening. Particles performing random walks of dimension d_w stick next to regions of the aggregate where particles already exist. If the outer parts of the aggregate did not prevent penetration

of these particles a compact blob would result with $D=d$. But a screening length l, which by definition is essentially the distance a particle can penetrate into the growing aggregate, does exist and this prevents compact regions from growing indefinitely. This length scale can be estimated in a mean-field manner by smearing out all N particles of the aggregate over a volume R^d. The probability that a lattice site is occupied inside the cluster is $\rho \sim N/R^d$ (here and in the future all lengths are measured in units of the lattice spacing). Thus, if on the average a walk lasts N_w steps inside the cluster before being caught, we have $N_w \rho \sim 1$. But as by definition a walk of dimension d_w obeys $N_w \sim l^{d_w}$, we can estimate the screening length to be

$$l \sim (N/R^d)^{-1/d_w} \sim R^{(d-D)/d_w}. \quad (1)$$

I now argue how a typical cluster of size R has been grown, and what is its resulting structure. Initially the cluster is very small; the screening length is $l \sim 1$, and the random nature of the aggregate formation ensures an essentially branched polymer structure. The cluster grows, and by the time it has reached a size $r < R$ the screening length $l(r) \sim r^{(d-D)/d_w}$ has increased thus hindering to a smaller extent particles penetrating inside the cluster and allowing larger compact regions of particles to form. How large can these compact regions grow before screening hinders further growth? The assumption in this paper is that l is the relevant length scale and that therefore the compact regions are of size l^d. When the blob reaches a size $\sim l^d$ growth stops, and new compact regions appear near the surface, the blobs forming a contiguous randomly branched structure until the cluster has radius of gyration R and the blobs near the surface are of size $l(R)^d \sim R^{d(d-D)/d_w} \sim N^{d(d-D)/d_w D}$. Note that I have made the implicit assumption that $D < d$. If $D = d$, then the aggregate is compact.

Before putting forward the Flory-type argument in this paper, I note that such an argument already exists for the case $d_w = 2$ by Tokuyama and Kawasaki.[16] This paper points out that the relevant length is $l \sim \rho^{-1/2}$, which is the same as Eq. (1) with $d_w = 2$, but argues that the aggregate behaves as a linear chain of blobs of size l^d. This difference in the assumed topological structure of the blobs will lead to divergence with the results given in this paper. They find $D(d, d_w = 2) = (d^2+1)/(d+1)$ for all d which is in good agreement with computer simulations. This same value for $D(d, d_w = 2)$ was found by Muthukumar,[17] though his argument, which treats the growth in a dynamic fashion and assumes a relevant length scale $l' \sim \rho^{-1}$, is very different.

As I have argued previously I would expect the blob length scale to be a very slow function of the radius r inside the aggregate $l(r) \sim r^{(d-D)/d_w}$. For the purposes of calculating the fractal dimension by means of a Flory argument, however, it is sufficient to treat the aggregate as a randomly branched structure consisting of $\mathfrak{N}$ blobs of length scale $l \sim (N/R^d)^{-1/d_w}$, the average blob length scale; clearly

$$\mathfrak{N} l^d = N. \quad (2)$$

With the physical model above in mind, I therefore write down for the "elastic free energy" of the cluster

$$F_{el}(R, N) \sim R^2/\mathfrak{N}^{1/2} l^2 \quad (3)$$

which is the correct term for an intersecting branched polymer[1,2,18,19] consisting of $\mathfrak{N}$ blobs of length scale l. This term, however, which favors small R, does not take into account the self-avoiding nature of the aggregate—the algorithm of formation ensures that no lattice site may be occupied twice. What do we take for the "repulsive contribution to the free energy" $F_{rep}(R, N)$? One estimate would be simply to smear all N particles over a volume R^d. Then $F_{rep}(R, N)$, which here is proportional to the number of particle-particle contacts, would be $F_{rep}(R, N) \sim N^2/R^d$. This estimate for $F_{rep}(R, N)$ would grossly overestimate the excluded volume, as we have completely neglected screening and the resulting clustering of particles into blobs of size l^d. Suppose instead that we treat the blobs as hard spheres of size l^d; then all blob-blob pairs within a distance of $\sim l$ of each other would contribute a constant factor and $F_{rep} \sim \mathfrak{N}^2 (l/R)^d$. In fact, even if we made this assumption, we would be greatly overestimating the excluded volume. The blobs are not hard spheres. They are easily distortable and can in fact approach each other to within a finite number of lattice sites before the real hard spheres—the individual particles of which the blobs are composed—exclude further approach. Thus $F_{rep}(R)$ is proportional to all blob-blob pairs within some finite distance of each other, or

$$F_{rep}(R, N) \sim \mathfrak{N}^2/R^d. \quad (4)$$

We can now combine Eqs. (1)–(4) and find $D(d,$

d_w):

$$F(R,N) \sim R^{2+d(d-4)/2d_w} N^{-(d_w+d-4)/2d_w} + R^{-d(1+2d/d_w)} N^{2+2d/d_w}. \quad (5)$$

On minimizing $F(R,N)$ and using $N \sim R^{D(d,d_w)}$ we find

$$D(d,d_w) = \frac{4d_w + d(2d_w-4) + 5d^2}{5d_w - 4 + 5d}. \quad (6)$$

For the usual diffusion-limited aggregate with $d_w = 2$, we find

$$D(d) = (8+5d^2)/(6+5d). \quad (7)$$

The argument leading to Eqs. (6) and (7) is only correct for dimensions below some upper critical dimensionality $d_c(d_w)$. For we can see by substitution that for $d = d_c(d_w)$ given by

$$d_c^2 - (4-d_w)d_c - 8d_w = 0, \quad (8)$$

$F_{\rm rep}(R \sim N^{1/D}) \sim O(1)$, and is negligible for $d > d_c$. Thus for $d > d_c(d_w)$, the fractal dimension should solely be given by the entropic term in Eq. (5); that is the aggregate behaves like a noninteracting branched chain of blobs with

$$D(d,d_w) = \frac{4d_w - 4d + d^2}{d_w - 4 + d}, \quad d > d_c(d_w). \quad (9)$$

For the usual random walk $D(d) = (8-4d+d^2)/(d-2)$ for $d > d_c(2) = 1+\sqrt{17} \approx 5.1$. From Eq. (9) we see that as $d \to \infty$, $D(d,d_w) \to (d-d_w)$ from above. Now $D(d,d_w) > (d-d_w)$ is a lower bound for the fractal dimension, as first pointed out by Meakin[13] from the fact that a walk of dimension d_w cannot create an object to which it is transparent. Therefore, this model obeys this bound, and suggests that the aggregate becomes "asymptotically transparent" as $d \to \infty$.

Though not stated explicitly, all the previous results are only valid for $1 < d_w < d$. The cases $d_w = 1$ and $d_w = d$ are special and have to be treated separately.

Let us first consider the case $d_w = 1$. For this case $D(d, d_w = 1) = d$, which is due to the infinite "persistence" of such a walk. Once started in some direction, it will carry on along the same path. This is not the case for $d_w = 1+\epsilon$. For instance, scaling arguments suggest that if $\hat{r}_1$ is the direction of the first step of a walk $\hat{l}$ inside an aggregate then

$$\langle \hat{r}_1 \cdot \hat{l} \rangle / l \sim N^{-(d_w-1)(d-D)/Dd_w}.$$

It is clear that the limits $d_w \to 1$, $N \to \infty$ cannot be interchanged and therefore

$$D(d, d_w \to 1) \neq D(d, d_w = 1) = d. \quad (10)$$

In fact we would expect a crossover behavior for an aggregate of N particles formed by a walk of dimension d_w close to 1 between $1 \ll N \ll \exp[k/(d_w-1)]$ when $D = d$ and $N \gg \exp[k/(d_w-1)]$ when D is given by Eq. (6). This crossover effect is distinct from that pointed out by Bensimon, Domany, and Aharony,[20] which is due to the introduction of an additional length scale l_0, the mean free path of the random walk, though related in that both walks give $D = d$ on small scales as the walk will appear to be ballistic under these conditions. Equation (6) will give the limit $D(d, d_w \to 1) = (4-2d+5d^2)/(1+5) < d$, and not $D = d$, as I specifically assumed $D < d$ in the derivation of Eq. (6).

Let us now consider the case $d_w = d$. In deriving Eq. (1) I assumed $N_w \rho \sim 1$. This is only true if the number of times a walk visits a given lattice site, $c_w \sim N_w/l^d \sim l^{d_w - d}$, does not diverge. This is only the case for $d_w < d$. For $d_w \geq d$ the assumption breaks down.

In Table I I compare the results with computer simulations.[13,21] The random walks have been generated experimentally by picking at random the length of a step (x) from a distribution which satisfies $P(x \geq u) = u^{-f}$, $P(x < 1) = 0$, where $P(x \geq u)$ is the probability that the length of the step is greater than or equal to u. Then these walks will have well defined fractal dimension $d_w = f$ for $1 < f < 2$.[11] For $f \leq 1$, $d_w = 1$; and for $f \geq 2$, $d_w = 2$. I note that $f = 0$ is the sober walker $d_w = 1$, while $f = \infty$ generates the Witten-Sander model. The results are satisfactory though several points de-

TABLE I. A comparison of the fractal dimension $D(d,d_w)$ given by theory with the results of computer simulations.

d	f	d_w	$D_{\rm exp}$	$D(d,d_w)$
2	0	=1	≈ 1.95 [a]	2.00
2	4/3	4/3	≈ 1.86 [a]	1.79
2	5/3	5/3	≈ 1.83 [a]	1.77
2	2	$\to 2$	≈ 1.75 [a]	1.75
2	2.5	=2	≈ 1.71 [a]	
2	∞	=2	1.67 ± 0.05 [b]	
3	∞	2	2.49 ± 0.06 [b]	2.52
4	∞	2	3.34 ± 0.10 [b]	3.38
5	∞	2	4.20 ± 0.10 [b]	4.29
6	∞	2	4.90 ± 0.6 [b]	5.00

[a]Ref. 21. [b]Ref. 13.

serve emphasis. The first entry is the result for $d_w = 1$. The limiting value $d_w \to 1$ is very difficult to simulate because of the long-range correlations in the walks. Secondly, Brownian motion in two dimensions is space filling and there appears to be no unique fractal dimension generated with $d_w = 2$. The limiting value $D_{\exp}(2, d_w \to 2) = 1.75$ agrees well with theoretical expectation $D(2, d_w \to 2) = 1.75$; from then on for $2 < f < \infty$, $d_w = 2$, the fractal dimension slowly decreases to the value $D \sim 1.67$ for the Witten-Sander model. The theory makes no prediction for these cases, though I note that the results[16,17] $D(d) = (d^2+1)/(d+1)$ give $D(2) \approx 1.67$ which agrees well with the Witten-Sander model in two dimensions. For the final entries which obey $d > d_w = 2$ the agreement with Eq. (7) is good.

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