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LETTER TO THE EDITOR

Topological properties of diffusion limited aggregation and cluster–cluster aggregation

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Abstract. The detailed topological or 'connectivity' properties of the clusters formed in diffusion limited aggregation (DLA) and cluster-cluster aggregation (CCA) are considered for spatial dimensions $d = 2, 3$ and 4. Specifically, for both aggregation phenomena we calculate the fractal dimension $d_{\min} = \tilde{\nu}^{-1}$ defined by $\ell \sim R^{d_{\min}}$ where ℓ is the shortest path between two points separated by a Pythagorean distance R. For CCA, we find that $d_{\min}$ increases monotonically with d, presumably tending toward a limiting value $d_{\min} = 2$ at the upper critical dimensionality d_c as found previously for lattice animals and percolation. For DLA, on the other hand, we find that $d_{\min} = 1$ within the accuracy of our calculations for $d = 2$, 3 and 4; suggesting the absence of an upper critical dimension. We also discuss some of the subtle features encountered in calculating $d_{\min}$ for DLA.

Considerable recent attention has focussed on models of aggregation, in large part due to their potential promise in providing tractable models for a range of flocculation phenomena. The diffusion limited aggregation (DLA) model of Witten and Sander (1981) is the prototype of modern models of aggregation: a seed particle is placed at the origin and a random walker is released from a large circle encompassing the origin. This particle is assumed to undergo a random walk until it sticks to the seed particle. Another random walker is then released, and this process is continued until typically a large aggregate containing $N = O(10^4)$ particles has been formed. DLA describes a range of natural phenomena in which identifiable 'seed sites' exist, but for the flocculation of particles ranging from soot to colloids no such stationary seed exists. Accordingly, the cluster-cluster aggregation (CCA) model (Meakin 1983d, Kolb *et al* 1983) assumes that a large number of particles randomly diffuse at the same time; when they touch one another they stick, forming clusters, which themselves diffuse randomly touching other particles or clusters until at a large time a ramified fractal aggregate has been formed.

Both DLA and CCA clusters qualitatively resemble aggregates found in nature, and have been the subject of intensive recent study. However, there is thus far only a single quantitative parameter that has been used to characterise these aggregates. This is the fractal dimension d_f that is a direct measure of how the density approaches zero as

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the length scale over which it is measured increases. If $M(R)$ is the cluster mass within a Pythagorean distance R of a cluster point, and $\rho(R) = M(R)/R^d$ is the density, then one writes

$$M(R) \sim R^{d_f} \qquad \rho(R) \sim R^{d_f - d}. \tag{1}$$

The fractal dimension concept has permitted extensive comparisons between large-scale computer simulations (e.g. Meakin 1983a, b, c, e) and several mean-field type theories (e.g. Muthukumar 1983, Tokuyama and Kawasaki 1984, Hentschel 1984, Hentschel and Deutch 1984). More recently, it has become possible to actually *measure* d_f for naturally-occurring aggregates and to compare the experimental values with results from simulations and from theory (see e.g. Forrest and Witten 1979, Nittmann *et al* 1984, Weitz and Oliveira 1984, Niemeyer *et al* 1984, Schaefer *et al* 1984, Schaefer and Keefer 1984, Bale and Schmidt 1984, Laibowitz and Gefen 1984, Matsushita *et al* 1984).

Although d_f has proved extremely useful as a quantitative parameter with which to characterise clusters, it is by no means sufficient. For example, in $d = 3$ both DLA and percolation clusters have $d_f \cong 2.5$, yet even the most casual visual inspection reveals that they are quire different (see e.g. figure 4 of Stanley *et al* (1984a)). Accordingly, one motivation for the present study is to investigate the utility of a second parameter in the quantitative characterisation of DLA and CCA. This is the exponent d_{min} that governs the dependence of the minimum path length between two points, ℓ, on the Pythagorean distance R between them,

$$\ell \sim R^{d_{min}} \tag{2a}$$

(see Middlemiss *et al* 1980, Pike and Stanley 1981, Hong and Stanley 1983a, b, Herrmann *et al* 1984). Equivalently, one may write

$$R \sim \ell^{\tilde{\nu}} \tag{2b}$$

with $\tilde{\nu} = 1/d_{min}$ (Havlin and Nossal 1984, Vannismenus *et al* 1984). Since the minimum path in the cluster should not be *shorter* than the Pythagorean distance nor *longer* than a completely random walk between the two points, we expect

$$1 \leqslant d_{min} \leqslant 2 \qquad (\tfrac{1}{2} \leqslant \tilde{\nu} \leqslant 1). \tag{3}$$

Here $d_{min} = \tilde{\nu}^{-1} = 2$ (as for Gaussian chains) for d above the critical dimension d_c.

The dimension d_{min} is *extrinsic*, in the terminology of Toulouse (see the discussion in Vannimenus (1984) and Stanley (1984)): it measures the dependence of a 'mass' (the number of sites in the minimum path) on a Pythagorean distance (R). One can equivalently study an *intrinsic* dimension d_ℓ, which measures the dependence on ℓ of the mass of sites that lie within a path length ℓ of the origin,

$$M \sim \ell^{d_\ell}. \tag{4}$$

This was first introduced (with the symbol ψ_{13}) by Pike and Stanley (1981); a much more complete discussion is given by Havlin and Nossal (1984). Since $M \sim R^{d_f}$, we can combine (4) and (2) to obtain (Havlin and Nossal 1984)

$$d_\ell = d_f / d_{min} = \tilde{\nu} d_f. \tag{5}$$

Since (4) relates two masses, (5) takes the form of the ratio of two extrinsic dimensions and d_ℓ is termed an intrinsic dimension. Although d_ℓ and d_{min} contain similar information, the actual calculations of these two quantities are quite different and hence study of both provides a useful check on accuracy and systematic errors.

In this work we calculate d_{min} and d_ℓ for both CCA and DLA in $d=2, 3$ and 4 (table 1). We find that the d-dependence of these quantities is quite different for the two sorts of aggregation models. For CCA, we find the same general trends as noticed already for lattice animals and percolation (see e.g. figure 4 of Havlin *et al* 1984a): as d increases, d_{min} increases toward a limiting value of $d_{min}=2$ as $d \rightarrow d_c$. Now for CCA the numerical value of d_c is not known, but our results for $d=2, 3, 4$ lie consistently and significantly *below* the corresponding lattice animal (LA) points, which tends to suggest the possibility that d_c may be larger than the LA value $d_c=8$. For DLA, on the other hand, we find a striking result: d_{min} takes on its one-dimensional value, $d_{min}=1$, for *all* values of d studied (figure 1). Accordingly, we interpret our results as providing support for the possibility (Witten and Sander 1983) that *for* DLA *there is no upper critical dimension.*

Table 1. Summary of the results of the present work; see equations (2*a*) and (4) for definitions of d_{min} and d_ℓ.

	DLA		CCA	
	d_{min}	d_ℓ	d_{min}	d_ℓ
$d=2$	1.0 ± 0.02	1.69 ± 0.05	1.15 ± 0.04	1.22 ± 0.02
$d=3$	1.02 ± 0.03	2.3 ● 0.2	1.25 ± 0.05	1.42 ● 0.02
$d=4$	1.00 ± 0.04	3.3 ± 0.2	1.35 ± 0.05	1.55 ± 0.05

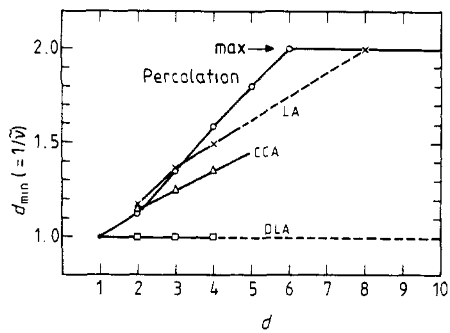

Figure 1. Dependence on lattice dimension d of the fractal dimension d_{min} of the minimum path. The data on DLA and CCA are from the present work; the data on lattice animals and percolation clusters are from Havlin and Nossal (1984), Havlin *et al* (1984a), and Herrmann *et al* (1984) (confirmed by calculations of Herrmann and Stanley (1984)). Note that d_{min} appears to 'stick' at the value two for all models above their respective critical dimensions d_c. The only exception is DLA, for which $d_{min}=1$, within the limits of accuracy, suggesting that there is no critical dimension for DLA.

The DLA clusters were generated using methods which have been discussed previously (Meakin 1983a, b, c). All of the 2D and 3D DLA aggregates contain 25 000 sites and the 4D clusters contained either 20 000 or 25 000 sites. We first choose a 'local origin' somewhere on the cluster. We then measure the radius of gyration of all the points within a path length ℓ of the local origin; this quantity scales with ℓ according to (2*b*). We also measure the *total* cluster mass $M(\ell)$ of all sites within a path length

ℓ; this quantity scales with ℓ according to (4)†. Our results for $d_{\min}$ and d_ℓ are shown in figure 2.

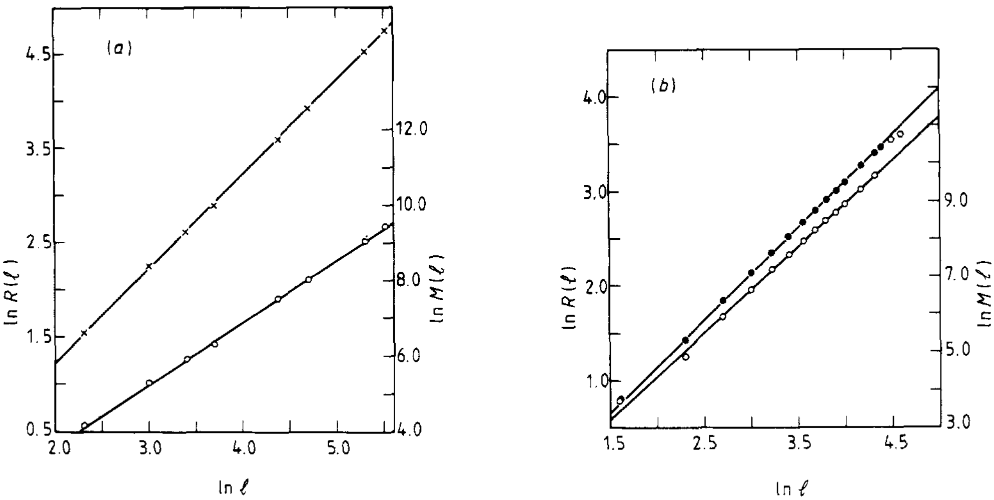

Figure 2. Dependence of $R(\ell)$ and $M(\ell)$ on the minimum path length ℓ for DLA with (*a*) $d=2$ and (*b*) $d=3$. The slopes give the fractal dimensions $d_{\min}$ (equation (2*b*)) and d_ℓ (equation (4)) respectively. (*a*): × refer to left axis and ○ refer to right axis, (*b*) ● refer to left axis and ○ refer to right axis.

The cluster-cluster aggregates were generated on finite hypercubic lattices with non-zero (but small) particle concentrations using periodic boundary conditions. For the 2D, 3D and 4D simulations the lattice sizes were 800^2, 133^3 and 64^4 respectively and the particle densities (ρ) were 0.0158, 0.0034 and 0.00072 particles per lattice site. At these densities the correlation length (within which the clusters have a fractal structure) is approximately equal to the lattice size. Results were obtained from six 4D, eight 3D and ten 2D aggregates. A local origin site was randomly selected from among all of the lattice sites occupied by the aggregates and the mass $M(\ell)$ measured from the local origin site was determined. Results were averaged over a number of such randomly chosen local origin sites in order to obtain an accurate estimate for $d_{\min}$ (the number of local origin sites per cluster was 1000 for $d=2$, 500 for $d=3$ and 1000 for $d=4$). Typical results for 2D and 4D CCA are shown in figures 3 and 3(*a*). Table 1 gives the final values of $d_{\min}$ and d for both models. The DLA results for $d=4$ were obtained by extrapolating the successive slopes obtained by joining successive points of a log-log plot.

Our result that $d_{\min}=1$ ($d_\ell = d_f$) for DLA for $d=2, 3, 4$ has striking implications. The first of these was mentioned above: it provides numerical evidence in support of the idea (Witten and Sander 1983, Witten and Ball 1984) that there is no critical dimension. A second implication is the following: it has been recently shown (Havlin *et al* 1984a) that if rings are irrelevant and if the clusters are finitely ramified (Havlin

† For the $M(\ell)$ calculation, we found the most convincing results if we chose the local origin to be the seed particle. If the local origin was, say, a path length ℓ_0 from the origin, then the results for $\ell \leq \ell_0$ were somewhat different from those for $\ell > \ell_0$.

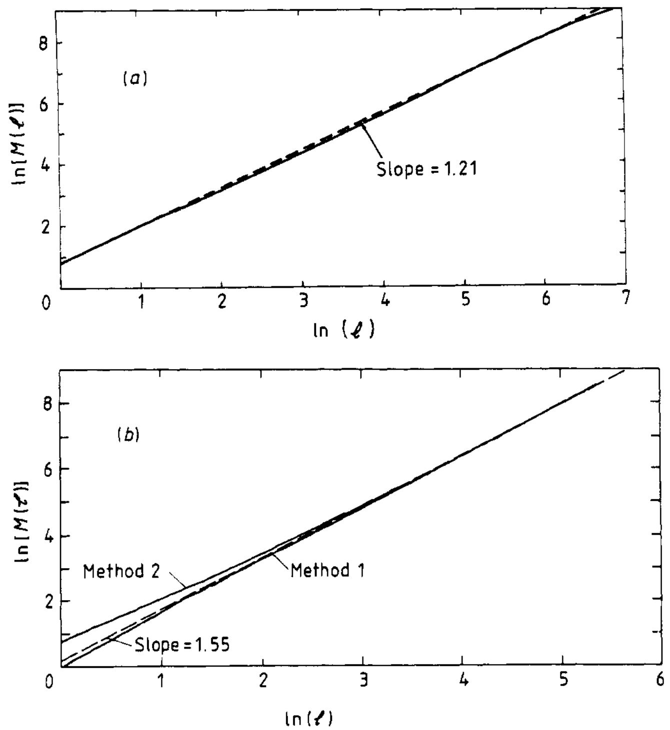

Figure 3. Dependence of the mass $M(\ell)$ included within a minimum distance ℓ for 2D and 4D cluster-cluster aggregates. In method 1 the origin site is considered to be a distance of 1 and the nearest-neighbour sites are considered to be at a distance ℓ of 2. In method 2 both the origin site and its nearest neighbours are included in the mass at distance 1 and the next-nearest-neighbour sites are included in the mass at distance 2. Part (a) shows the results obtained from the 2D CCA model using method 2 ($D(M) \sim M^0$). Part (b) shows the results obtained from the 4D clusters using both methods.

1984b), then the spectral dimension (see Stanley (1984) and references therein) is given by

$$d_s = 2d_\ell/(d_\ell + 1). \tag{6a}$$

If $d_\ell = d_f$, then for *all* values of d_f we have

$$d_s = 2d_f/(d_f + 1). \tag{6b}$$

This result was also presented by Alexander (1983). Very recently, Aharony and Stauffer (1984) (AS) have argued, with one simple assumption, that (6b) should hold for any aggregate *below* the lower critical dimension d_c^-. AS choose d_c^- to be the dimension below which all the growth sites (Leyvraz and Stanley 1983) cannot fit into a thin annulus, and find $d_c^- = 2$ – a result also obtained by Coniglio and Stanley (1984) using quite different methods (see also Sahimi 1984). Recent work has questioned the assumption underlying the AS argument (Stanley *et al* 1984b, Havlin 1984a, Hong 1984), but it does appear that (6b) holds for DLA for d_f *above* d_c^- (as well as below) for the reasons given in deriving (6b): that DLA aggregates are loopless and have the property that $d_{min} = 1$ ($d_\ell = d_f$). It should be mentioned that a direct test of (6b) is provided by extensive simulations (Meakin and Stanley 1983) of the probability of a random walk on a DLA cluster returning to the origin after t steps, $P_0 \sim t^{-d_s/2}$, the mean number of sites covered $\langle s \rangle \sim t^{d_s/2}$, and the mean-square displacement $\langle r^2 \rangle \sim t^{d_s/d_f}$. Averaging the estimates obtained by these three calculations, one obtains $d_s = 1.25 \pm 0.10$ ($d = 2$) and $d_s = 1.35 \pm 0.12$ ($d = 3$). The $d = 2$ result agrees well with (6*b*) ($d_s \simeq 5/4$) while the $d = 3$ result is within the error bars ($d_s \simeq 10/7$).

In summary, we have obtained results for the behaviour of $d_{\min}$ and d_ℓ for DLA and CCA. By doing calculations for $d = 2, 3, 4$ we have found two striking results: (i) $d_{\min} = 1$ for DLA, apparently independent of d, providing support for the conjecture that DLA has no critical dimension, and (ii) $d_\ell = d_f$ for DLA, so that the conjecture (6*b*) appears to be valid for DLA for $d_f > 2$ as well as $d_f < 2$ (table 2).

Table 2. Comparison of different cluster models and their topological properties. 'Aharony-Stauffer' means the result that $d_s = 2d_f/(d_f + 1)$.

Model	Rings irrelevant?	$d_\ell \overset{?}{=} d_f$	Aharony-Stauffer?
DLA	yes (?)	yes	yes
CCA	yes (?)	no	no
Percolation	no	no	no
Lattice animals	yes	no	no
SGA[a]	yes (?)	yes (?)	yes
Trees[b]	yes	no	no

[a] The screened growth aggregates (SGA) and their fractal properties are treated elsewhere (Meakin *et al* 1984a, b).
[b] Trees (defined as a random fractal without loops) are studied in Havlin *et al* (1984b).

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