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A cluster–cluster aggregation model with tunable fractal dimension

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Abstract. A hierarchical cluster–cluster aggregation computer model is introduced which allows one to build random fractal aggregates on a d-dimensional lattice with a fractal dimension fixed *a priori*. The algorithm works iteratively by sticking aggregates of the same number of particles at the correct centre-to-centre distance in order to recover the desired scaling. With the more efficient versions of the model, any fractal dimension ranging from 1 up to a d-dependent upper limit $D_M(d)$ can be obtained. One estimates $D_M(2) \simeq 1.80 \pm 0.03$ and $D_M(3) \simeq 2.55 \pm 0.04$. Calculations up to $d = 6$ show that the ratio $D_M(d)/d$ decreases as d increases.

1. Introduction

Cluster–cluster aggregation models have been introduced in order to explain the fractal structure of several experimental objects such as colloid and aerosol aggregates, clusters of balls floating on water, etc (Jullien and Botet 1987). Different models have been introduced according to their experimental applications. The original diffusion-limited cluster–cluster aggregation model (DLCA) (Meakin 1983, Kolb *et al* 1983), which is appropriate for the rapid aggregation of screened colloidal particles, considers aggregates that undergo Brownian motion and stick irreversibly when they come into contact. The ballistic model (BCA) (Ball and Jullien 1984, Jullien 1984), which is more appropriate for aerosols in the molecular regime, considers independent random straight-line trajectories for the clusters. The chemically limited aggregation model (CLCA) (Jullien and Kolb 1984, Kolb and Jullien 1984), also called 'reaction-limited' aggregation (Brown and Ball 1985, Family *et al* 1985), which applies to the slow aggregation of partially screened colloids, considers that aggregates should collide many times before they stick. These three models, which can be classified according to the fractal dimension d_w of the cluster trajectory lead to aggregates of increasing fractal dimensions, $D = 1.78, 1.98, 2.05$ in dimension $d = 3$, for DLCA, BCA, CLCA, where one can consider that $d_w = 2, 1, 0$, respectively. The observed increase of the fractal dimension corresponds to an increase of the mean penetration of the aggregates when they stick.

It may be noticed that the range of fractal dimensions (1.78–2.05) obtained in the three models is quite narrow. But other cluster–cluster models have been introduced that lead to fractal dimensions out of this range. For example, a lower fractal dimension has been obtained in the 'tip-to-tip' model (Jullien 1985, 1986) which considers polarizable aggregates, and larger fractal dimensions have been obtained when considering restructuring processes in the above models (Jullien and Meakin 1989, Meakin and Jullien 1985, 1988).

All the existing cluster–cluster computer algorithms have already been very useful since they have permitted sucessful reproduction of some physical properties of fractal aggregates

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such as small-angle x-ray and neutron scattering (Axelos *et al* 1986, Hasmy *et al* 1993), light scattering (Shalaev *et al* 1991) etc. However, they are limited to specific fractal dimensions and therefore they do not allow a systematic study of the physical properties as a function of the fractal dimension. In this paper we introduce a generalized cluster–cluster aggregation model, which is hierarchical (as most of the previous models) in the sense that only clusters of the same number of particles can stick together (Botet *et al* 1984) and which has the great advantage of containing the fractal dimension as an input parameter. In its more efficient versions, this model is able to build quite large aggregates with fractal dimensions ranging from 1 up to an upper limit, $D_M(d)$, which we have estimated numerically for space dimensions d ranging from 2 to 6. In section 2 we present the principles of the method and in section 3 we describe the algorithms that we have set up. In section 4 we present analytical investigations of some limiting cases and in section 5 we give the numerical results.

2. Principles of the model

The aggregates are built on a d-dimensional hypercubic lattice with unit lattice parameter. That means that they are made of connected (hyper-)spherical particles of unit diameter centred on the sites of the lattice. We use the hierarchical procedure (Botet *et al* 1984) which starts with a collection of 2^n particles. These particles are grouped into pairs. Each pair generates an aggregate of two particles so that, at iteration $p = 1$, one obtains 2^{n-1} dimers. The dimers are grouped into pairs that generate tetramers and so on. At iteration p one obtains 2^{n-p} aggregates containing 2^p particles each. The procedure stops at iteration $p = n$, where a final aggregate of 2^n particles is obtained. The model is entirely defined giving the rules used to stick together two aggregates of N particles to generate an aggregate of $2N$ particles but, before introducing these rules, let us recall some definitions and derive a useful formula.

The size of an aggregate of N particles is conveniently characterized by its radius of gyration (Guinier 1937) R_N given by

$$R_N^2 = \frac{1}{2N^2} \sum_{i=1}^{N} \sum_{j=1}^{N} (r_i - r_j)^2 \tag{1}$$

where the d-dimensional vector of integer components r_i refers to the position of the ith particle centre. Introducing the position r_G of the centre of mass G,

$$r_G = \frac{1}{N} \sum_{i=1}^{N} r_i \tag{2}$$

the radius of gyration is also given by

$$R_N^2 = \frac{1}{N} \sum_{i=1}^{N} (r_i - r_G)^2 . \tag{3}$$

Following previous reasoning (Ball and Witten 1984, Ball and Jullien 1984, Jullien 1984, Warren 1993), the radius of gyration R_{2N} of an aggregate of $2N$ particles, resulting from the sticking of two aggregates (1) and (2) of N particles, can be expressed as a function

of their radii R_{N1} and R_{N2} after using (1) and splitting the sum into four parts depending on whether i and j belong to the same cluster or not:

$$R_{2N}^2 = \frac{1}{4}(R_{N1}^2 + R_{N2}^2) + \frac{1}{4N^2}\sum_{i_1=1}^{N}\sum_{j_2=1}^{N}(r_{i_1} - r_{j_2})^2 . \tag{4}$$

In this formula i_1 and j_2 refer to particles belonging to aggregates (1) and (2), respectively.

Then, introducing

$$\Gamma = r_{G_1} - r_{G_2} \tag{5}$$

where G_1 and G_2 are the centres of mass of clusters (1) and (2), one can write:

$$r_{i_1} - r_{j_2} = (r_{i_1} - r_{G_1}) - (r_{j_2} - r_{G_2}) + b\Gamma . \tag{6}$$

Inserting into (4) and making use of (2) and (3), one gets

$$R_{2N}^2 = \langle R_N^2 \rangle + \tfrac{1}{4}\Gamma^2 \tag{7}$$

with

$$\langle R_N^2 \rangle = \tfrac{1}{2}(R_{N1}^2 + R_{N2}^2) . \tag{8}$$

Our method is based on equation (7). If one assumes that fractal scaling holds, i.e.

$$N \sim R_N^{\Delta} \tag{9}$$

where Δ is the fractal dimension, Γ should be proportional to the radius of gyration

$$\Gamma = k\sqrt{\langle R_N^2 \rangle} \tag{10}$$

and the constant of proportionality k should be related to Δ by

$$k = 2\sqrt{4^{1/\Delta} - 1} . \tag{11}$$

In practice, instead of (10), we will make use of the following relation:

$$\Gamma^2 = k^2 \frac{R_{N1}^2 + R_{N2}^2}{2} + 1 . \tag{12}$$

Considering the addition of $+1$ ensures that the formula works exactly at the first iteration $p = 1$ when building dimers from individual particles, where $R_N = 0$ and $\Gamma = 1$. The added term becomes negligible when N is large. This introduces 'natural' corrections to scaling of the type of those recently discussed by Warren (1993).

In conclusion, given Δ, which here becomes the input parameter (in addition to the space dimension d), one can calculate k by (11) and if one can manage to ensure (12) during the sticking of two aggregates of N particles, one should be able to build fractal aggregates of the desired fractal dimension. In practice, we calculate the fractal dimension D of the resulting clusters and our program works if the output parameter D is equal to Δ. The main difference between this model and previous models (such as DLCA, BCA and CLCA) is that the natural distribution of the penetration parameter Γ, which depends on the physical aggregation process, is replaced here by a Dirac peak. Thus, in addition to the hierarchical approximation, which is to neglect the polydispersity of aggregates, we introduce another approximation which is to neglect the polydispersity of their penetration distances.

3. Details of the algorithm

As soon as an aggregate has been built, a search is performed to find its surface particles i_s (i.e. particles having at least one unoccupied nearest neighbour) as well as for the unoccupied lattice sites i_n, nearest neigbours to the surface sites that we call neighbour-surface sites, and this information is conveniently stored in addition to the radius of gyration and the coordinates of its particles. Then, given two aggregates of N particles (1) and (2), a double loop is performed over the N_s surface sites of (1) and the N_n neighbour-surface sites of (2). Cluster (2) is translated in order that the current surface site of (1), i_{s1}, coincides with the current neighbour-surface site of (2), i_{n2}, (see figure 1). Then a test is performed to discard overlapping situations, i.e. situations where at least one particle of (1) coincides with a particle of (2). For each remaining configuration, the distance Γ between the centres of mass is calculated as well as the following quantity:

$$\delta\Gamma = \left(\Gamma^2 - \tfrac{1}{2}k^2(R_{N1}^2 + R_{N2}^2) - 1\right)^2. \tag{13}$$

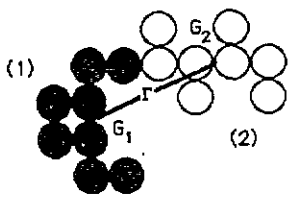

Figure 1. Two-dimensional sketch of the procedure used to build an aggregate of $2N$ particles by sticking two aggregates of N particles.

Among all the sticking positions, one retains the one that minimizes $\delta\Gamma$ and, if there are different positions leading to the same minimum value, one makes a uniform random choice over these possibilities. We have considered several versions of our program.

(i) *Version A.* In version A, only one configuration of cluster (2) is considered for each value of i_s and i_n, which is the configuration obtained by a strict translation of the original cluster. Since at $p = 1$ the dimers are built in random directions, they maintain their orientations at $p = 2$ to build tetramers. Therefore, in this version, orientational randomness is introduced in the early iterations.
(ii) *Version B.* In version B, one considers d configurations of cluster (2) for each value of i_s and i_n, which are all the configurations obtained by rotations around the first diagonal (circular permutations of the coordinates). We will see that this amelioration is essential to recover the lowest fractal dimension $D = 1$ for $\Delta = 1$. Other rotations could have been considered as well as reflections along some coordinate axes, however, we have observed that such modifications slow down the program without providing so much improvement.
(iii) *Case* $\Delta = \infty$. Since Δ is an input parameter of our model, it is not forbidden to raise it to very large (unphysical) values, larger than the space dimension d. In the limit $\Delta = \infty$, one recovers a nice hierarchical model in which the rule of sticking two clusters is to choose the minimum distance between their centres of mass. In both versions A and B we have written special codes for this limiting case that we will call version A′ and B′ in the following. In these codes the only input parameter is the space dimension d.

In all the versions considered, the square of the radius of gyration has been averaged over the 2^{n-p} clusters of $N = 2^p$ particles obtained at each iteration. Then, instead of

making a least-square fit of the log–log plot of N versus $\sqrt{\langle R_N^2 \rangle}$, we have calculated an N-dependent fractal dimension $D(N)$ obtained by comparing the results from one iteration to the next one (Ball and Jullien 1984):

$$D(N) = \frac{\log 4}{\log\left(\langle R_N^2 \rangle - \frac{1}{4}\right) - \log\left(\langle R_{N/2}^2 \rangle\right)}. \tag{15}$$

The term $\frac{1}{4}$ is introduced to take care of the corrections to scaling, as has recently been justified (Warren 1993). We have also estimated the surface fractal dimension D_s from the calculation of the mean number of neighbouring surface sites $\langle N_n(N) \rangle$ at each iteration (one could have used either of the surface sites). Here we have also calculated an N-dependent surface fractal dimension $D_s(N)$ from

$$D_s(N) = 2\frac{\log(\langle N_n(N) \rangle) - \log(\langle N_n(N/2) \rangle)}{\log(\langle R_N^2 \rangle - \frac{1}{4}) - \log(\langle R_{N/2}^2 \rangle)}. \tag{16}$$

4. Analytical results in some limiting cases

4.1. Case $\Delta = 1$

We can show that version B allows exact recovery of a linear chain when the input parameter Δ is set to 1. For a linear chain of N particles, the radius of gyration is given by

$$R_N^2 = \frac{N^2 - 1}{12}. \tag{17}$$

Since from 11 we get $k^2 = 12$, formula (12) gives $\Gamma = N$, which is exactly the centre-of-mass distance needed to stick two chains of N particles end-to-end to obtain a chain of $2N$ particles. However, since the dimers are obtained in random directions at the first iteration, one needs to allow rotations to start to build the linear chain correctly. As we will see in section 5, version A, which considers only translations, is not able to build clusters of fractal dimensions extending down to 1. It is also worth noticing that (15) gives exactly $D(N) = 1$ for all N in this limiting case.

4.2. Case $\Delta = 2$

It might be interesting to see if the model allows one to recover a plane of particles when setting $\Delta = 2$. The sequence of clusters shown at the top of figure 2(*a*), which are obtained by sticking alternately north–east/south–west and east/west, have a radius of gyration given by

$$R_N^2 = \frac{N - 1}{4}. \tag{18}$$

Since $k^2 = 4$ here, equation (12) gives $\Gamma = \sqrt{N}$, which is again exactly what is needed to build a plane of particles in that way. However, this sequence is *not* reproduced by our model since some degeneracies appear as early as iteration $p = 3$. In figure 2(*a*), one has depicted two other configurations obtained, in version B, by sticking $p = 2$ clusters with

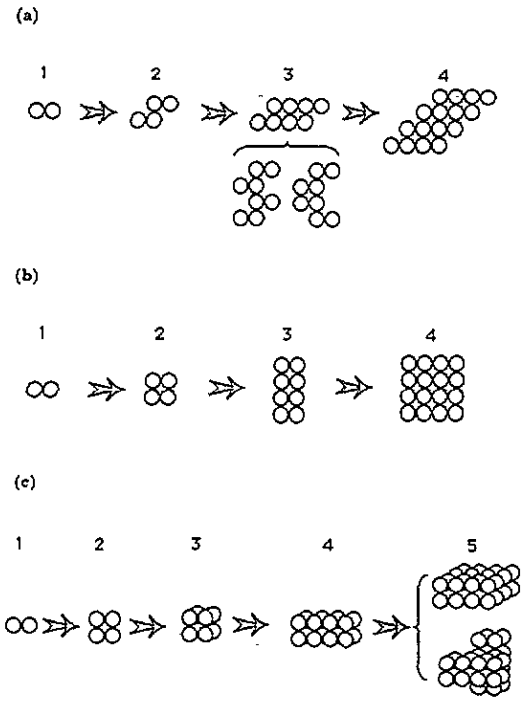

Figure 2. Typical sequences of clusters which can be built with the different versions of the model. The sequence shown in top of (*a*) would lead exactly to an infinite plane, however, due to the degeneracies appearing at iteration $p = 3$, both versions A and B do not build a plane for $\Delta = 2$, $d = 2$. The sequence shown in (*b*), which also leads to a plane, is exactly recovered using version B′ with $d = 2$. However, the corresponding $d = 3$ sequence shown in (*c*) is not recovered by version B′ in $d = 3$.

$\Gamma = 2$ in space dimension $d = 2$ (in version A, the cluster on the right is not obtained). Some more configurations are generated when working in higher space dimensions. One could have built other versions of our model which would have included some restrictions in the rules in order to recover a plane for $\Delta = 2$. However, such restrictions would have led to regular fractals, while our aim here is to build random fractals, closer to what it is seen in the experiments.

Note that, in the large-N limit, the clusters shown on the top of figure 2(*a*) are parallelograms of angle 45° and edge lengths of ratio $\sqrt{2}$. By sticking two of these parallelograms, one obtains a parallelogram of the same type. The renormalization transformation is the same at each step and does not oscillate in contrast with the sequence shown in figure 2(*b*).

4.3. Case $\Delta = 3$

In that case, there is no sequence of clusters which would satisfy our basic equations exactly. In the asymtotic large-N limit such clusters should be rhombohedra with edges proportional to $2^{1/3}$, $2^{2/3}$ and 2. Apparently there is no way to do this on a cubic lattice.

4.4. Case $\Delta = \infty$

The sequence shown in figure 2(*b*), which builds squares every two iterations, satifies the rule of the minimum Γ and we have checked that this sequence is well recovered when

running version B′ in $d = 2$. However, the corresponding sequence, which builds cubes every three iterations, is not recovered when running version B′ in $d = 3$. This is due to the appearence of a degeneracy at iteration $p = 5$ as shown in figure 2(*c*).

5. Numerical results

5.1. Case $d = 2$

In figure 3(*a*) and 3(*b*) we show three typical clusters, containing 8192 particles each, built in two dimensions using versions A and B, respectively, with $\Delta = 1, 1.5$ and 2. One can check that for $\Delta = 1$ a straight-line chain is recovered with version B but not with version A. The corresponding plots for the effective fractal dimensions $D(N)$ and $D_s(N)$ as a function of $1/N$ are given in figure 4. These data result from an average over five independent runs, all ending with aggregates of 8192 particles. In the case $\Delta = 1.5$ the two versions A and B, are quite efficient since $D(N)$ stays equal to 1.5 within less than 0.1 per cent for all $N > 16$. In fact, we have checked that the same efficiency is obtained for all Δ values in the range $1.15 < \Delta < 1.75$ with version A and in the range $1 < \Delta < 1.75$ with version B.

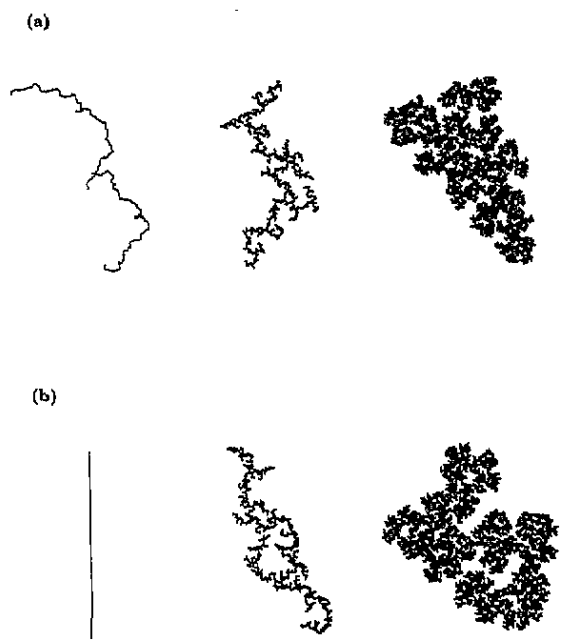

Figure 3. Typical two-dimensional aggregates of 8192 particles obtained for $\Delta = 1$, 1.5 and 2. Cases (*a*) and (*b*) correspond to versions A and B, respectively. The scale has been chosen such that the vertical dimension is the same on all pictures.

When one asks for Δ values out of these ranges, the computer time is increased and the convergence of the effective fractal dimensions when $N \to \infty$ becomes weaker. In version A for $\Delta = 1$, by analysing the convergences of $D(N)$ and $D_s(N)$, one obtains the estimate $D_m(2) = 1.14 \pm 0.02$ which is a lower bound for the fractal dimensions obtained with this

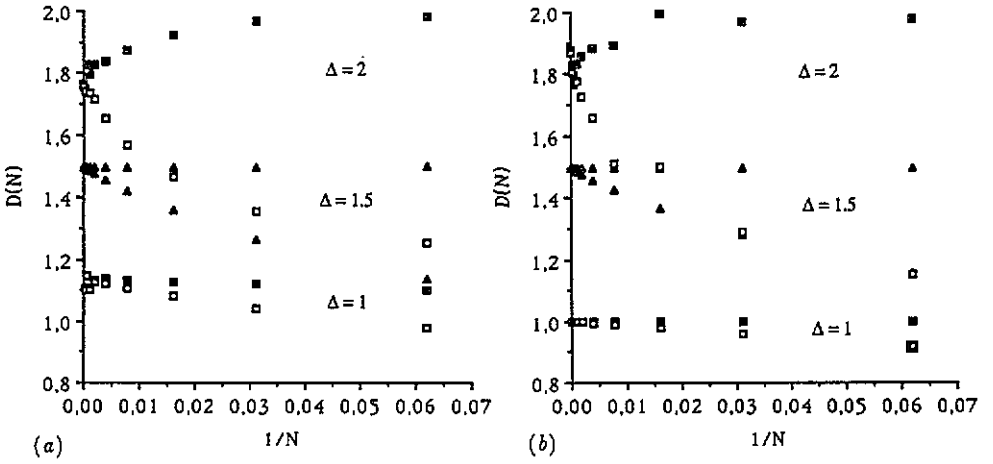

Figure 4. Plot of $D(N)$ and $D_s(N)$ versus $1/N$ in $d = 2$ for $\Delta = 1$, 1.5 and 2, after averaging the results over five independent runs up to 8192 particles. Cases (*a*) and (*b*) correspond to versions A and B, respectively. Filled and open symbols correspond to $D(N)$ and $D_s(N)$, respectively.

version. In both versions A and B, for $\Delta = 2$ the convergence of the fractal dimensions is even poorer. However, $D(N)$ converges to the same value giving the following estimate for the upper bound in two dimensions:

$$D_M(2) = 1.80 \pm 0.03 . \tag{19}$$

It is worth noticing that for $\Delta = 2$, the effective fractal dimensions $D(N)$ and $D_s(N)$ remain quite close to the values 2 and 1, respectively, for small N values, as would be expected for a compact two-dimensional aggregate with a smooth surface. But, as N increases, the hierarchical process cannot avoid building a rough surface. As a result $D_s(N)$ increases and $D(N)$ decreases and finally both $D(N)$ and $D_s(N)$ tend to the same asymptotic value as expected for a mass fractal. Note that the convergence is better with model A, where some disorder exists from the beginning. However, such a tiny difference cannot be seen on the $\Delta = 2$ clusters of figures 3(*a*) and 3(*b*). The observed difference should be attributed to fluctuations. Therefore, the fact that one cannot reach $D = 2$ in $d = 2$ with our model is due to geometrical frustrations associated with the disordered character of our procedure. Note that, if one eliminates disorder by working with rigid rules leading to regular fractals, one can reach $D = 2$. For example, as noticed in section 4.3, the version B′, which builds the sequence of figure 2 (*b*), is able to give $D = 2$. But using version B with a very large, but finite, Δ value, some degeneracies appears as soon as N is sufficiently large and finally $D(N)$ tends to 1.8.

5.2. Case $d = 3$

In figure 5 we show two-dimensional projections of five clusters, containing 8192 particles each, built in three dimensions using version A with $\Delta = 1, 1.5, 2.0, 2.5$ and 3. We have not shown clusters built with version B since, except for $\Delta = 1$, they look pretty much the same.

The corresponding plots for the effective fractal dimensions $D(N)$ and $D_s(N)$ as a function of $1/N$ are given in figures 6 and 7. They result from single runs up to 8192 particles. Here, again, one obtains a good efficiency for intermediate Δ values. As soon

as N is larger than 16, $D(N)$ becomes equal to the desired value within less than 0.1 per cent. The lowest fractal dimension obtained with version A is $D_m(3) = 1.22 \pm 0.05$, a value larger than in two dimensions. As in two dimensions, both versions cannot reach $D = d$. The upper fractal dimension, as deduced from the extrapolation of the $D(N)$ and $D_s(N)$ curves for $\Delta = 3$, can be estimated as

$$D_M(3) = 2.55 \pm 0.04 . \tag{20}$$

The same slower convergence as in $d = 2$ is observed with version B for $\Delta = d$.

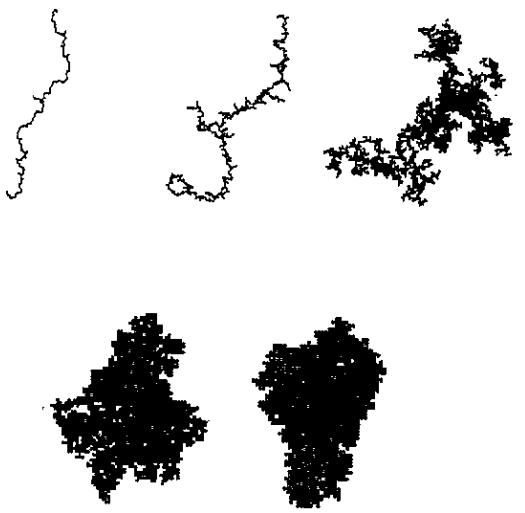

Figure 5. Two-dimensional projections of typical three-dimensional aggregates of 8192 particles built with version A for $\Delta = 1$, 1.5, 2, 2.5 and 3. The scale has been chosen such that the vertical dimension is the same on all pictures.

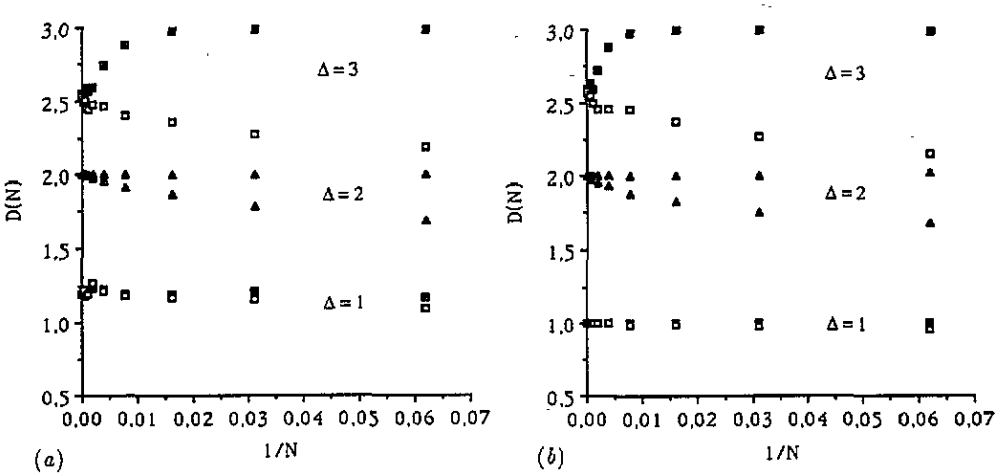

Figure 6. Plot of $D(N)$ and $D_s(N)$ versus $1/N$ in $d = 3$ for $\Delta = 1, 2$, and 3, from single runs up to 8192 particles. Cases (a) and (b) correspond to versions A and B, respectively. Filled and open symbols correspond to $D(N)$ and $D_s(N)$, respectively.

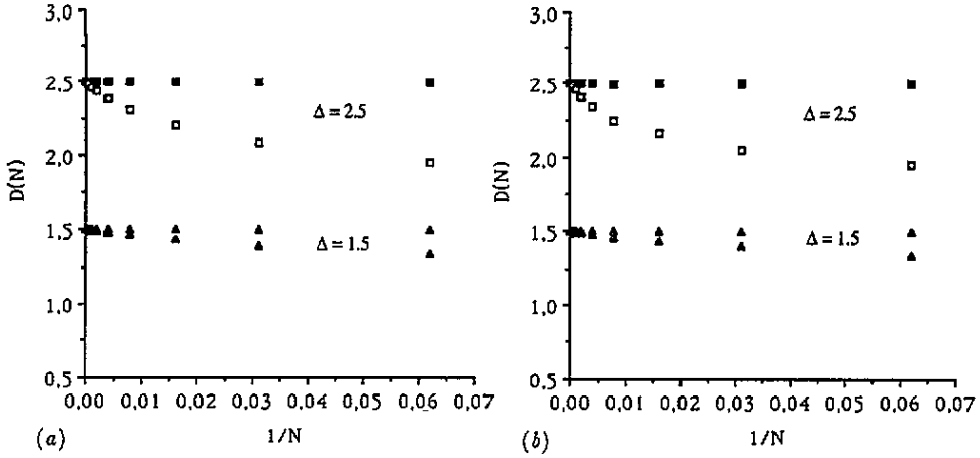

Figure 7. Same as figure 6, but for $\Delta = 1.5$ and 2.5.

5.3. *Estimation of $D_M(d)$ up to $d = 6$*

To study how $D_M(d)$ evolves when increasing d, we have used the quicker version A′ , up to $d = 6$. We have calculated $D(N)$ and $D_s(N)$ up to $N = 4096$ particles after averaging over ten runs, except for $d = 6$ where only five runs have been considered. We have checked that the version A′ gives almost the same estimate for $D_M(3)$ than versions A and B with $\Delta = d = 3$ (formula (20)). In addition, for $d = 4, 5, 6$, one has obtained:

$$D_M(4) = 3.20 \pm 0.06 \qquad D_M(5) = 3.85 \pm 0.075 \qquad D_M(6) = 4.38 \pm 0.15\,. \tag{21}$$

In figure 8, we report the ratio $D_M(d)/d$ as a function of d. This ratio is monotically decreasing as d increases. Therefore, the geometrical frustration effects described in section 5.1 become more and more important as the space dimension increases. However, since we are systematically avoiding overlaps when building a cluster of $2N$ particles from two clusters of N particles, the two clusters can never become transparent to each other and therefore their fractal dimension should stay larger than $d/2$. It may be that the upper fractal

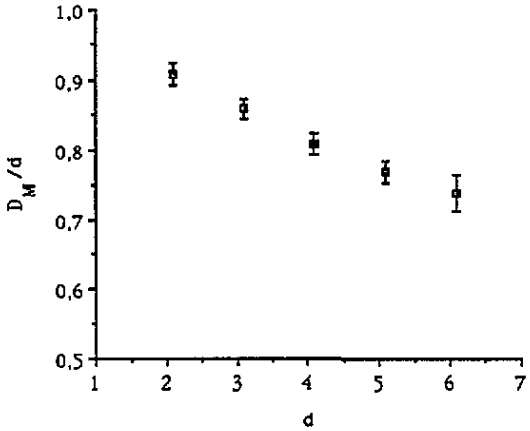

Figure 8. Plot of $D_M(d)/d$ as a function of d.

dimension $D_M(d)$ tends to this lower bound as d tends to infinity, leading to $D_M/d \to \frac{1}{2}$. The results of figure 8 are not inconsistent with this conjecture. However, one needs to have more high-dimensional data to be able to conclude more clearly. Unfortunately, as d increases the CPU time increases and the precision of the results decreases.

6. Conclusion

In this paper we have presented a computer algorithm able to build random fractal aggregates with a tunable fractal dimension. With version B of our algorithm, all fractal dimensions ranging from 1 to an upper limit $D_M(d)$ can be obtained. The fact that one cannot reach fractal dimensions as large as $D = d$ is due to geometrical frustrations induced by disorder: our hierarchical method introduces natural surface roughness while, to reach $D = d$, one needs a smooth surface. A way to avoid this limitation, while preserving randomness, would be to introduce some cluster-size polydispersity in a controlled way. Such an extension of our model is under progress. In the near future, we intend to use our algorithm to make a systematical investigation of some physical properties of fractal aggregates as a function of their fractal dimension.

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