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Aggregation by kinetic clustering of clusters in dimensions $d > 2$

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Résumé. — Le modèle hiérarchique récemment introduit pour décrire le mécanisme d'agrégation cinétique par collage d'amas est étudié numériquement pour des dimensions d supérieures à $d = 2$. La dimension fractale des amas, D, estimée à partir de leur rayon de giration, vaut $1,42 \pm 0,03$; $1,78 \pm 0,05$; $2,02 \pm 0,06$; $2,25 \pm 0,15$ pour $d = 2, 3, 4, 5$ respectivement. Ces résultats sont comparés avec ceux du modèle d'agrégation par diffusion limitée et avec ceux des polymères, linéaires et branchés, à l'équilibre.

Abstract. — The hierarchical model recently introduced to describe the mechanism of kinetic aggregation by clustering of clusters is investigated numerically in dimensions $d > 2$. The fractal dimension of the clusters, D, is evaluated from their radius of gyration and estimated to be 1.42 ± 0.03, 1.78 ± 0.05, 2.04 ± 0.08, 2.30 ± 0.20 for $d = 2, 3, 4, 5$ respectively. These results are compared with those of diffusion limited aggregation and with those of linear and branched polymers at equilibrium.

Several theoretical models have been introduced recently to describe the growth of clusters by aggregation of Brownian particles. The most extensively studied model is the « diffusion limited aggregation » (DLA) model [1-3], in which single particles stick, one by one, on an immobile growing cluster after diffusing in a purely random walk fashion in empty space. Monte Carlo simulations have been performed on this model for space dimensions ranging from $d = 2$ to $d = 6$ [2]. All the numerical results available for the fractal dimension D of the cluster agree very well with the analytical mean-field-like result [3], $D = (d^2 + 1)/(d + 1)$, without an upper critical dimension.

An alternative model, the « kinetic clustering of clusters » (ClCl) model [4, 5] has been proposed to describe the process of flocculation. In ClCl, clusters of particles, as well as single particles, are allowed to diffuse together and any kind of clusters stick on contact. Monte Carlo simulations [4, 5] on ClCl in $d = 2$ show that the resulting clusters are much more ramified than in DLA. More recently we have introduced an idealized version of ClCl, the « hierarchical model » [6], in which only clusters of exactly the same number of particles stick together in a process similar to DLA. We have argued that this model is equivalent to ClCl in its low concentration scaling regime. Results in $d = 2$ agree perfectly well with direct simulations [6]. The advantage of the hierarchical model is that it is numerically more precise and less time consuming than direct simulations.

In this letter, we report the first investigation of ClCl in space dimension larger than $d = 2$, by a systematic use of the hierarchical model up to $d = 5$. Preliminary results of direct simulations on ClCl in $d = 3$ found that the fractal dimension is close to that of linear polymers [4, 7]. With the higher accuracy of the method used here, we can conclude that the numerical values for D stay slightly (but clearly) above those of linear polymers except at $d = 4$ where $D = 2$ is within the error bars. Moreover, it is found that the difference between ClCl and DLA becomes more pronounced when the dimension increases.

In the hierarchical model successive generations of independent clusters of $N = 2, 4, 8... 2^k$ particles are built iteratively starting from a collection of N_0 particles. Let us assume that, at the $(k - 1)$th iteration a generation of $n_{k-1} = N_0/2^{k-1}$ independent clusters of $N_{k-1} = 2^{k-1}$ particles each, is available. These clusters are grouped into $N_0/2^k$ pairs of clusters. Then, each pair generates a new cluster of $N_k = 2^k$ particles by the following growth process. One cluster of the pair is centred at the origin of a hypercubic lattice in d dimension. The other one is released at a site chosen at random on a large hypersphere of radius R_0 centred at the origin. Then, this cluster undergoes a random walk on the lattice, jumping, at each step, to one of the $2d$ neighbouring sites, with equal probability. In this motion, the cluster stays rigid and does not rotate. The diffusion stops when at least one particle of the moving cluster becomes nearest neighbour with a particle of the other. The reunion of the two clusters then forms a cluster of the new generation. If the moving cluster diffuses too far from the origin, say, a distance R_m, it is released again on the hypersphere of radius R_0, and this is repeated.

We have calculated the radius of gyration R of each cluster with the usual formula :

$$R^2 = \frac{1}{N} \sum_{i=1}^{N} (\underset{\sim}{R}_i - \underset{\sim}{R})^2 \qquad \underset{\sim}{R} = \frac{1}{N} \sum_{i=1}^{N} \underset{\sim}{R}_i$$

where the summation covers all the sites of the cluster and where $\underset{\sim}{R}_i$ joins the origin and site i. The square of the radius of gyration is averaged over all the samples of a given generation and an estimation of the statistical error is given by the standard deviation of the results.

Moreover, we have systematically varied the cut-off parameters R_0 and R_m to see their influence on the numerical results. It is enough to choose R_0 only slightly larger than $R_1 + R_2$, where R_1 and R_2 are the maximum radii of the two coalescing clusters. The only source of error is due to the fact that we are obliged to choose a lattice site which is generally not strictly located on the hypersphere. In practice $R_0 = R_1 + R_2 + 8$ is sufficient [8]. Also, the numerical results do not depend on R_m as long as it is chosen sufficiently large. $R_m = 3 R_0$ is a typical value above which the numerical results are reproduced within the error bars.

For $d = 2, 3, 4$, we have performed 20 trials all starting with $N_0 = 4\,096$ particles and we have stopped after five iterations (after seven iterations for $d = 2$ [6]) such that $20 \times 128 = 2\,560$ independent clusters of 32 particles were built. From the standard deviation of R^2 we estimate that the absolute error on R is smaller than 0.02 for $N = 32$. For $d = 5$ we have performed only 5 trials and the error is almost twice as large.

The numerical results for log R versus log N are reported in figure 1. The exact $d = 1$ result $R^2 = (N^2 - 1)/12$ is also reported for the sake of comparison. The fractal exponent D defined by

$$N \underset{N \to \infty}{\sim} R^D$$

must be given by the inverse of the estimated asymptotic slope of those curves ($N \to \infty$). In order to estimate D we have calculated an effective fractal dimension obtained when comparing successive sizes N_1 and $N_2 = 2 N_1$

$$D_{\text{eff}} = \ln 2/\ln (R_2/R_1)$$

and we have reported D_{eff} as a function of N_1^{-1} in figure 2. From these plots, we estimate the
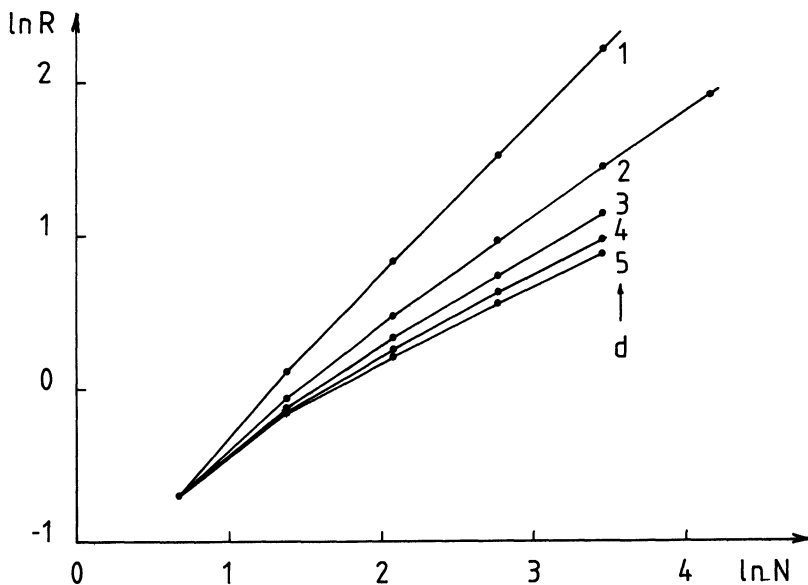
Fig. 1. — Results for the radius of gyration R as a function of the number of particles N (log-log plot). The exact curve $d = 1$ is shown for the sake of comparison.
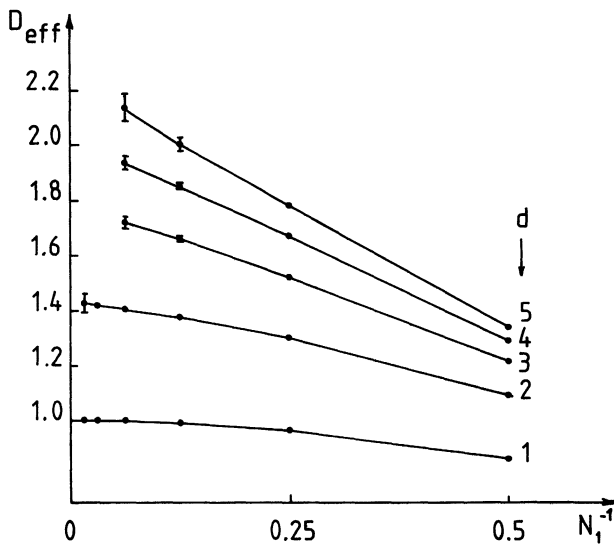

Fig. 2. — Results for the effective fractal exponent D_{eff} calculated by comparing sizes N_1 and $N_2 = 2 N_1$ and plotted against N_1^{-1}. The exact curve $d = 1$ is shown for the sake of comparison.

extrapolated D values to be 1.42 ± 0.03, 1.78 ± 0.05, 2.04 ± 0.08, 2.30 ± 0.20 for $d = 2$, 3, 4, 5 respectively. These results are reported in figure 3. For the sake of comparison we have drawn, on the same figure, the analytical result $D = (d^2 + 1)/(d + 1)$ for DLA [3] and the mean field results : $D = (d + 2)/3$ for $d \leqslant 4$, $D = 2$ for $d \geqslant 4$ for linear polymers at equilibrium [9] and $D = \frac{2}{5}(d + 2)$ for $d \leqslant 8$, $D = 4$ for $d \geqslant 8$ for branched polymers [10].
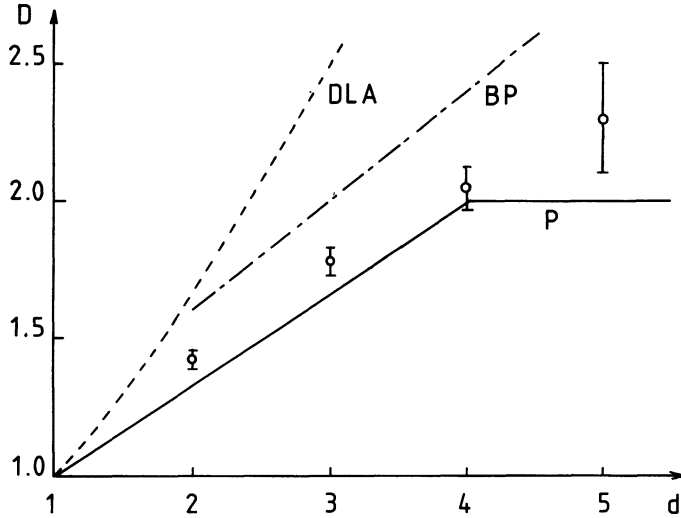
Fig. 3. — The numerical results for the fractal exponent D of the ClCl model (extrapolated values of Fig. 2) are shown by the open circles with statistical error bars. The dotted line corresponds to Muthukumar's result for DLA [3], the full line to the Flory's result for polymers (P) [9] and the dashed-dotted line to the mean-field result for branched polymers (BP) [10].

On figure 3, one observes that the curves $D(d)$ are completely different for ClCl and DLA. While for DLA $D(d)$ shows a positive curvature and tends rapidly to its asymptotic form, $D = d - 1$, the curve for ClCl shows a marked negative curvature [11].

Thus the difference between DLA and ClCl which we already noticed on the $d = 2$ result becomes more and more pronounced when the space dimension increases. For example, a marked difference is observed when considering the 2 d projection of the clusters for $d = 3$ with $D = 1.76 \pm 0.03(2.5)$ for ClCl (DLA) : thus ClCl is still ramified in 2 d projection whereas DLA is compact.

The comparison with linear polymers merits further attention. To get an idea of how well the numerical results for D for ClCl converge, an analogous analysis has been done for the self-avoiding walk. To do so, we have generated independent polymers connecting up to 32 sites on a hypercubic d-dimensional lattice. This was done in the most simple fashion : starting from the origin, we built the polymer iteratively i.e. after occupying $N - 1$ sites, we choose with the same probability the Nth site to be any neighbouring site of the free end, avoiding double occupancy. If this new site is occupied, the chain is discarded and a new polymer is built, again starting from the origin. We performed 10^6, 500 000, 300 000, 40 000 trials for $d = 2$, 3, 4, 5 respectively, for which 16, 483, 2 491, 1 063 independent polymers of 32 sites were built. The average radius of gyration has been calculated in the same way as ClCl and is plotted in figure 4. We have done the same kind of analysis as before by using only the results for $N = 2, 4, 8, 16, 32$. The values for D_{eff} are plotted in figure 5. Even, if the results here are less precise than for ClCl, one can observe a qualitative difference between figure 2 and figure 5. In figure 5, while for $d \neq 4$, the convergence is quite good, in $d = 4$ the extrapolation is much poorer, presumably because $d = 4$ is the upper critical dimension, where logarithmic terms slow down the convergence. From the data of figure 2, no indication is found of slowed convergence for ClCl, except perhaps for the case $d = 5$. In both cases, for all dimensions considered, D_{eff} grows with increasing size, suggesting that the actual $D_{eff}(N)$ represents a lower limit for the true exponent $D = D_{eff}(N = \infty)$.
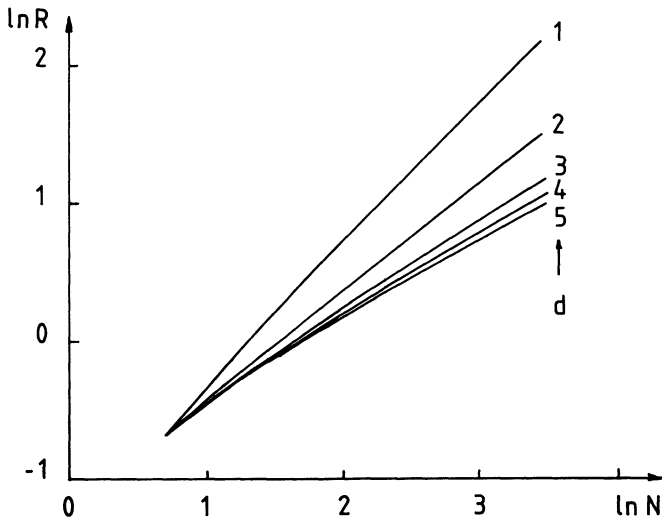
Fig. 4. — Results for the radius of gyration R as a function of the number of particles N (log-log plot) in the case of a direct simulation on linear polymers.
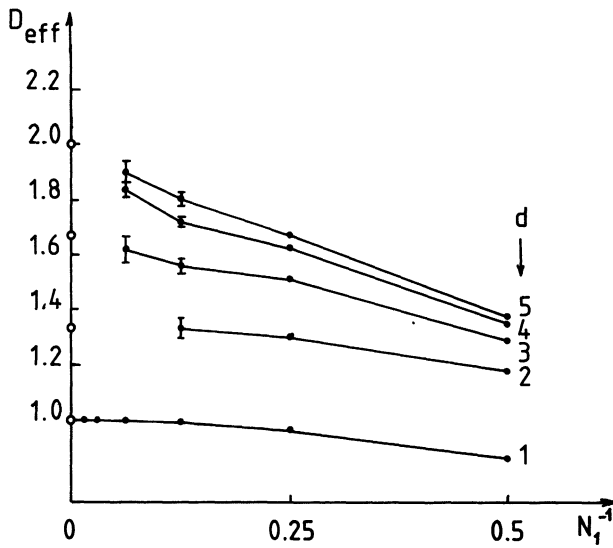

Fig. 5. — Results for the effective fractal exponent D_{eff} in the case of polymers.

In conclusion, we have reported simulations on the hierarchical model for ClCl for $d = 2, 3, 4, 5$. We have found that the fractal dimension behaves completely differently from DLA and D lies closer to the fractal dimension of linear polymers. In the range of d that were modelled, D lies between linear and branched polymers. We hope that these numerical calculations will stimulate the search of an analytical formula for the curve $D(d)$ of ClCl.

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[11] Let us remark that the simple formula $D = \sqrt{d}$ reproduces the numerical results quite well. We do not know of a physical reason why this relation should hold.