Hierarchical model for irreversible kinetic cluster formation

1984 J. Phys. A: Math. Gen. 17 L75

(http://iopscience.iop.org/0305-4470/17/2/009)

J. Phys. A: Math. Gen. **17** (1984) L75–L79. Printed in Great Britain

LETTER TO THE EDITOR

Hierarchical model for irreversible kinetic cluster formation

R Botet, R Jullien and M Kolb

Laboratoire de Phsysque des Solides†, Université Paris-Sud, Centre d'Orsay, 91405 Orsay, France

Received 14 October 1983

Abstract. A model is proposed to describe the growth of clusters by a mechanism of irreversible clustering of clusters. The fractal exponent is extracted by means of numerical simulations on small systems, both directly and in the form of a renormalisation-group analysis. The results are in excellent agreement with previous Monte Carlo simulations. The numerical precision is better due to the simpler (hierarchical) formulation of the model. In the present form the model looks like the diffusion limited aggregation model, which for the sake of comparison is treated by the same renormalisation-group method.

Recently, an irreversible growth model of clustering of clusters (ClCl) has been proposed to describe the process of flocculation (Kolb *et al* 1983, Meakin 1983b). The basic idea is that when particles form clusters one of the most natural ways to do this in an irreversible fashion is to form small clusters which then stick to other small clusters to yield larger clusters etc. By Monte Carlo numerical simulation, it was found that the resulting clusters are very ramified and they have scaling properties which are rather different from all previously studied growth models.

Here we would like to present a two-dimensional hierarchical model which describes this growth mechanism in an idealised way. This permits us to test whether indeed the picture just sketched is correct and to have a formulation that can be dealt with by easier numerical calculations, thus improving the statistical accuracy. Renormalisation groups and scaling are now built into the model through its hierarchical structure.

We start with a collection of $N_0 = 2^{k_0}$ particles. At step 1, we form $N_1 = N_0/2$ clusters of two particles each in the following manner: the two particles diffuse through empty space (random walk) until they meet. Then they stick to each other and form a rigid two-particle cluster. The N_1 clusters are formed independently. At step 2, the N_1 clusters are grouped into $N_2 = N_1/2$ pairs of clusters, each of which leads by the same random diffusion mechanism to a four-particle cluster. The process is repeated (see figure 1). At step k, $N_k = N_0/2^k$ independent clusters of $n_k = 2^k$ particles generate pairwise $N_{k+1} = N_0/2^{k+1}$ independent clusters of 2^{k+1} particles. In practice, the sticking mechanism is realised in a framework very similar to diffusion limited aggregation (DLA) (Witten and Sander 1981, Meakin 1983a). One cluster, say cluster 1, is centred at the origin of a square lattice. The other one, cluster 2, is released at a point chosen at random on a large circle of radius R_0 centred at the origin. Then cluster 2 undergoes a random walk on the lattice, jumping by one lattice spacing at each step. In this motion the cluster stays rigid and does not rotate. The random walk stops when one

† Laboratoire associé au CNRS.

0305-4470/84/020075+05$02.25

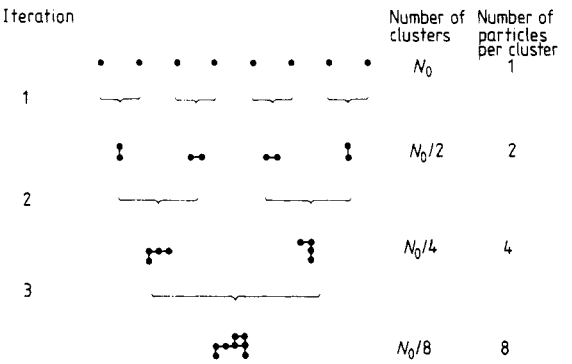

Figure 1. Sketch of the hierarchical model.

particle of cluster 2 becomes the nearest neighbour of one particle of cluster 1 and then a cluster of the new generation is formed by the reunion of clusters 1 and 2. If cluster 2 goes too far away from cluster 1, at a distance R_m, it is released again on the circle and this is done as often as it is necessary. We have checked that the precise choice for R_0 and R_m (unless they are not chosen too small) does not quantitatively affect the results (as already observed for R_m in DLA by Meakin (1983a)). For the results presented in this letter, we have taken $R_0 = 3(R_1 + R_2 + 2)$ and $R_m = 3R_0$, where R_1 and R_2 are the maximum radii of cluster 1 and 2. In figure 2, we provide a typical example of a cluster of 256 particles obtained by this prescription. Note the linear appearance and the few branchings as in figure 1 of Kolb *et al* (1983).

We are interested in the scaling properties of large clusters. That is we wish to know how the radius of gyration R of the clusters varies with their number of particles N in order to extract the fractal dimension $D = 1/\nu$ defined by

$$R \sim N^{\nu}, \qquad N \sim R^{D}, \qquad N \to \infty. \tag{1}$$

For that purpose, we have performed ten independent trials all starting with $N_0 = 4096$ particles and stopping after the seventh iteration after which $10 \times 32 = 320$ independent clusters of 128 particles were built. At each iteration, we have averaged the square of the radius of gyration over all the clusters obtained in all the trials. The numerical

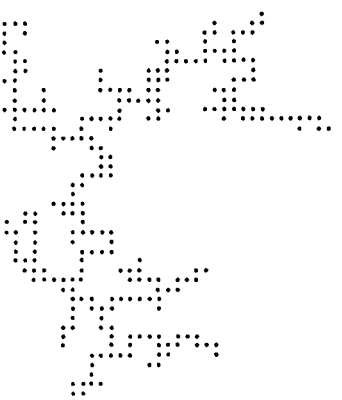

Figure 2. Typical cluster of 256 particles obtained by the present method.

results for the averaged R, with error bars (estimated from the standard deviation of the results), are given by the open circles in figure 3. In this log–log plot, we observe that a linear dependence is quickly recovered for large N and the resulting fractal dimension can be estimated to be

$$D = 1.42 \pm 0.03.$$

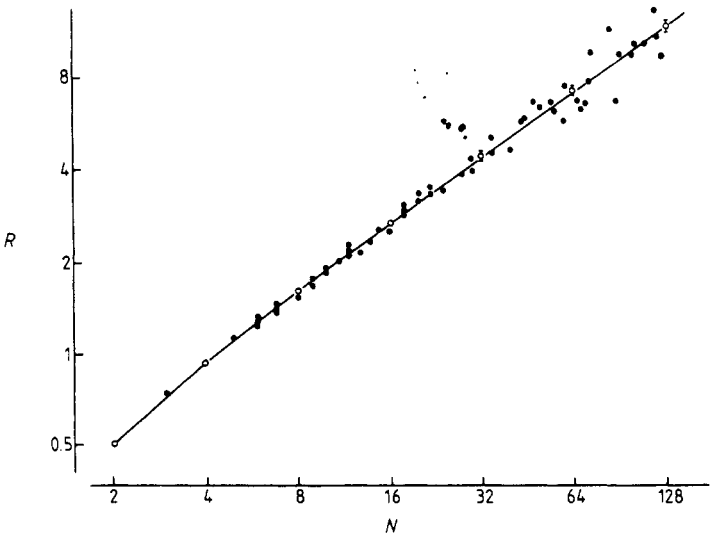

Figure 3. Results for the radius of gyration R as a function of the number of particles N. The open circles, with error bars, are the results of the hierarchical model, starting with $N_0 = 4096$ particles and averaging over all the samples obtained at each step and over ten independent trials. The full circles correspond to values obtained in a direct simulation of ClCl considering a box of 450×450 with 2000 particles.

Before comparing with numerical simulations on the ClCl model (Kolb *et al* 1983, Meakin 1983b) let us recall briefly how they were performed. A large square box of size $L \times L$ is considered on a square lattice, with periodic boundary conditions. At the beginning N_0 particles are distributed randomly on lattice sites. The particles undergo a random walk on the lattice until they meet (when they occupy neighbouring sites), after which they become clusters of two particles which continue to diffuse randomly along with the particles and grow by aggregation when they meet other clusters of particles etc. In their motion, the clusters are rigid and do not rotate. Different prescriptions were adopted for the speed of the clusters as a function of their sizes leading to the same fractal properties for the resulting clusters. From the analysis of the radius of gyration and particle–particle correlation function the fractal dimension was estimated to be

$$D = 1.38 \pm 0.06.$$

In order to perform a more direct comparison we report here some numerical results for the radius of gyration obtained in a simulation with $N_0 = 2000$ and $L = 450$. These results are represented by the full circles in figure 3. Since these values have not been averaged one observes some statistical dispersion. When looking at figure 3

the results of the present hierarchical model could appear as a very good fit of the simulations on ClCl, giving already some confidence in the similarity between both models and suggesting that they could belong to the same universality class.

The differences between the two models clearly concern irrelevant aspects in the language of the renormalisation group. In ClCl it was found that the cluster distribution function in the regime relevant for flocculation peaks at a mean cluster size, implying that the probability is large that two coalescing clusters have about the same size (Kolb *et al* 1983). Here, we have a situation where the distribution is a single δ-function at the average cluster size. Presumably the scaling is not affected by this change. This is also supported by the following: the different kinetic prescriptions adopted in the simulations of ClCl, while they affect the detailed shape of the cluster's size distribution, do not change its character (existence of a peak) nor the resulting D value. In other words, in the present model, a cluster of $n = 2^k$ particles is formed from two clusters of $n_1 = n_2 = 2^{k-1}$ particles as opposed to any combination n_1, n_2 with $n_1 + n_2 = n$. This is the analogue for growth models of the treatment of linear polymers by a hierarchical scheme (Alexandrowicz 1969). The numerical advantage of this model lies in the fact that, by construction, the dilute regime is considered while the direct simulation becomes very time-consuming for low density. Moreover, the present hierarchical model shares some features with the usual treatment of DLA (Witten and Sander 1981, Meakin 1983a) since only two clusters are involved at each step of the calculation. To study time-dependent effects, on the other hand, one has to return to the complete system.

Instead of analysing the growth process directly to test scaling, one can consider a comparative analysis of different sized clusters and then extract than effective scaling exponent D_{eff}, which, in the limit of large clusters, tends to D. This D_{eff} is analogous to the local fractal dimension of Havlin and Ben-Avraham (1982). It can be compared also with an effective exponent, as defined in the phenomenological renormalisation-group (Nightingale 1976, Barber 1983). For this renormalisation-group type analysis, D_{eff} follows from equation (1) applied to clusters with N_1 and N_2 particles respectively:

$$D_{\text{eff}} = \ln(N_2/N_1)/\ln(R_2/R_1). \tag{2}$$

Finite-size scaling considerations indicate that the convergence with N is best if N_1 and N_2 differ very little. We have used this definition to calculate successive approximations for D for the hierarchical model. In figure 4 the full line shows the results for D_{eff} against $1/N_1$, when we always take $N_2/N_1 = 2$, comparing successive steps of the iteation. We observe a rapid convergence to a value consistent with the previous direct estimations of this exponent. In order to compare with single-particle DLA, we have determined D_{eff} for a process where N_1 independent particles diffuse successively towards an N_1-particle cluster to form a $2N_1$-particle cluster. The results are shown by the broken line of figure 4. One can see that for small clusters D_{eff} is close in both models while for larger clusters D_{eff} saturates to a smaller limiting value in ClCl ($D_{\text{eff}} \sim 1.42$ compared with $D_{\text{eff}} \sim 1.67$ in DLA). This deviation quantitatively shows the difference in the screening effects in both models: in particular the more important difficulty to fill the holes when the clusters become larger in ClCl.

An interesting extension of the present hierarchical model is the case where a whole distribution of clusters of various sizes is admitted at each iteration and can interact with one another. This would approach the model to the realistic situation of clusters of any number of particles (instead of $n = 2^k$). We note, however that, even in this situation, some of the geometric aspects are left out (presumably irrelevant),

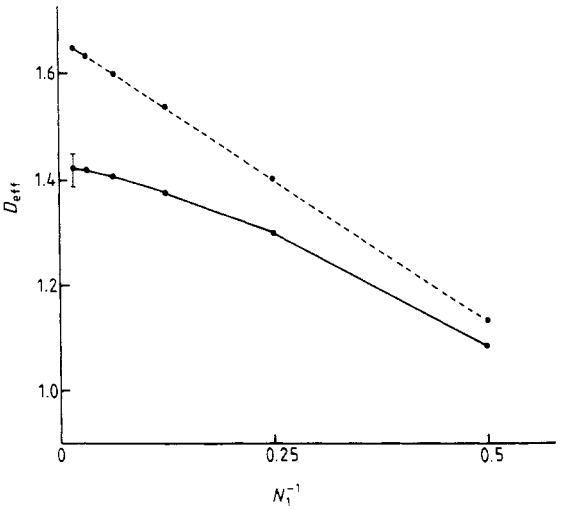

Figure 4. Results for the effective fractal exponent D_{eff} calculated by comparing sizes N_1 and $N_2 = 2N_1$ and plotted against $1/N_1$ for the hierarchical model (full lines) and for the diffusion limited aggregation model (broken lines).

somewhat reminiscent of Smoluchowski's approach (Smoluchowski 1916, 1917, Sutherland 1967).

In conclusion, a new hierarchical model has been presented to describe the aggregation by clustering of clusters. It is very similar to single-particle diffusion limited aggregation in that at each step only two clusters are considered. While in DLA a single particle and a big cluster are always involved, here two clusters of the same size are considered. The net difference in the critical behaviour of the two cases stems clearly from the relative difference in size of the clusters involved at each iteration. Moreover, the results on the hierarchical model are in excellent agreement with previous Monte Carlo simulations on ClCl. Thus, the present work confirms the hypothesis of Kolb *et al* (1983) that in ClCl the relative size of the clusters involved in a collision is the relevant aspect in determing the critical properties. Finally we mention that the present treatment can be extended to higher dimensions.

References

Alexandrowicz Z 1969 *J. Chem. Phys.* **51** 561
Barber M N 1983 *Finite size scaling* review paper to be published
Havlin S and Ben Avraham D 1982 *J. Phys. A: Math. Gen.* **15** L311
Kolb M, Botet R and Jullien R 1983 *Phys. Rev. Lett.* **51** 1123
Meakin P 1983a *Phys. Rev.* A **27** 604
—— 1983b *Phys. Rev. Lett.* **51** 1119
Nightingale M P 1976 *Physica* **83** A561
Smoluchowski M V 1916 *Phys. Z.* **17** 585
—— 1917 *Z. Phys. Chem.* **92** 129
Sutherland D N 1967 *J. Colloid Interface Sci.* **25** 373
Witten T A and Sander L M 1981 *Phys. Rev. Lett.* **47** 1400