Cluster-particle aggregation with fractal (Levy flight) particle trajectories

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The effects of particle trajectories on the structure of aggregates formed by cluster-particle aggregation have been explored by computer simulation. With the use of Levy-flight particle trajectories, clusters with a continuous range of effective dimensionalities between the limits of the Witten-Sander ($D \simeq \frac{5}{3}$ for $d=2$) and Vold-Sutherland models ($D \simeq 2.0$ for $d=2$) can be generated. The effective dimensionality varies continuously with the exponent which describes the distribution of step lengths in the Levy-flight trajectory, but there does not seen to be a unique (1:1) relationship between the dimensionality of the trajectory and the dimensionality of the cluster.

Computer simulation has provided valuable insights into the relationships between growth mechanisms and morphology in a wide variety of systems of scientific and commercial importance. One of the most important phenomena in colloid science is the formation of large particle clusters or flocs from smaller subunits (particles or smaller clusters). In the earliest model of flocculation,[1,2] single particles were added one at a time to a growing cluster of particles, using linear particle trajectories. Two- and three-dimensional models of this type give structures with a Hausdorff[3] or fractal[4] dimensionality (D) which seems to be smaller than, but close to, the ordinary Euclidean dimensionality[1,2,5] (d).

Recently, Witten and Sander[6] have developed a model for particle-cluster aggregation in which the Brownian trajectory followed by the particles is explicitly included in the model. The Witten-Sander (WS) model leads to structures with remarkable scaling and universality properties.[6-8]. In particular, the fractal dimensionality (D) is distinctly smaller than the Euclidean dimensionality of the systems [$D \simeq 1.7$ for $d=2$, (Refs. 6 and 7) $D \simeq 2.45$ for $d=3$ (Refs. 7, 9, and 10)]. In particle-cluster aggregation (unlike cluster-cluster aggregation[11-13]) the trajectory plays a crucial role. It is the trajectory which prevents particles from penetrating into the open structure of the fractal aggregates to form a denser structure with a higher dimensionality. A Brownian trajectory with a dimensionality (D_t) of 2.0 is more effective than a linear trajectory ($D_t=1.0$) in preventing penetration and leads to a structure with a lower dimensionality. One of the most interesting aspects of these models is the relationship between the fractal dimensionality of the particle trajectory and the fractal dimensionality of the aggregate. In this paper, we report some preliminary results obtained from computer simulations of particle-cluster aggregation carried out using Levy-flight trajectories.

To construct a particle trajectory with a well-defined fractal dimensionality, it is necessary to build long-range, power-law correlations into the paths followed by the particles. One way of doing this is to construct a walk in which the length of the step (x) is picked randomly from a distribution which satisfies the conditions

$$P(x \geq U) = U^{-f} , \tag{1a}$$

$$P(x < 1) = 0 , \tag{1b}$$

where $P(x \geq U)$ is the probability that the length of the step will be greater than or equal to U. A trajectory constructed in this way with f in the range $1 < f < 2$ is a Levy-flight with a dimensionality of f ($D_t = f$).[4]

We have carried out cluster-growth simulations using both lattice and nonlattice models in two dimensions. In the lattice model simulations, the step length (x) [picked randomly from the probability distribution given in Eqs. (1a) and (1b)] is converted to the nearest integer step length. The Levy-flight trajectories are started out close (but not too close) to the cluster and terminated if the particles reach a position which is far from the growing cluster. The starting and ending points for the Levy-flight trajectories are important from the points of view of both computation cost and accuracy. In most of our simulations, the trajectory is started off at a random point on a circle of radius $\simeq R_0 = R_{\max} + R_s + 1$ centered on the origin of the cluster. Here, $R_{\max}$ is the distance from the origin of the cluster to the most distant point in the cluster and R_s is selected so that $R_s^{-f} \cong 0.01$. The motivation for the choice of R_0 is to ensure that there is only a small probability that the particle will contact the cluster during the first step in the Levy flight. The "particles" are assumed to stick permanently to the cluster when they first contact the cluster. In the lattice model, the particle is considered to have contacted the cluster if it occupies a site which is a nearest neighbor to an occupied lattice site in the cluster. In both the lattice and nonlattice models the particles are able to stick to the cluster at any point along a step in the Levy flight. Particles are not allowed to pass through any part of the cluster before sticking. Particle trajectories are stopped, and new trajectories started on the circle of radius R_0 if the particles reach a position which is far from the cluster. In most of the simulations reported in this paper, trajectories are stopped at a distance of $2.5R_0$ from the origin. Each trajectory is continued until the particles contact the cluster and the cluster grows or until the trajectory is stopped by reaching a point $2.5R_0$ from the origin. The simulation starts with a single fixed particle at the origin and one mobile particle and ends when a cluster of sufficient size has been grown adding one mobile particle at a time.

Simulations have been carried out with various values for the parameter f which describes the Levy-flight trajectory. Figure 1 shows typical clusters generated using the lattice model with the parameter f set to 2.5, 2.0, $\frac{5}{3}$, and $\frac{4}{3}$. Each of the four clusters in Fig. 1 contains 25 000 particles or oc-

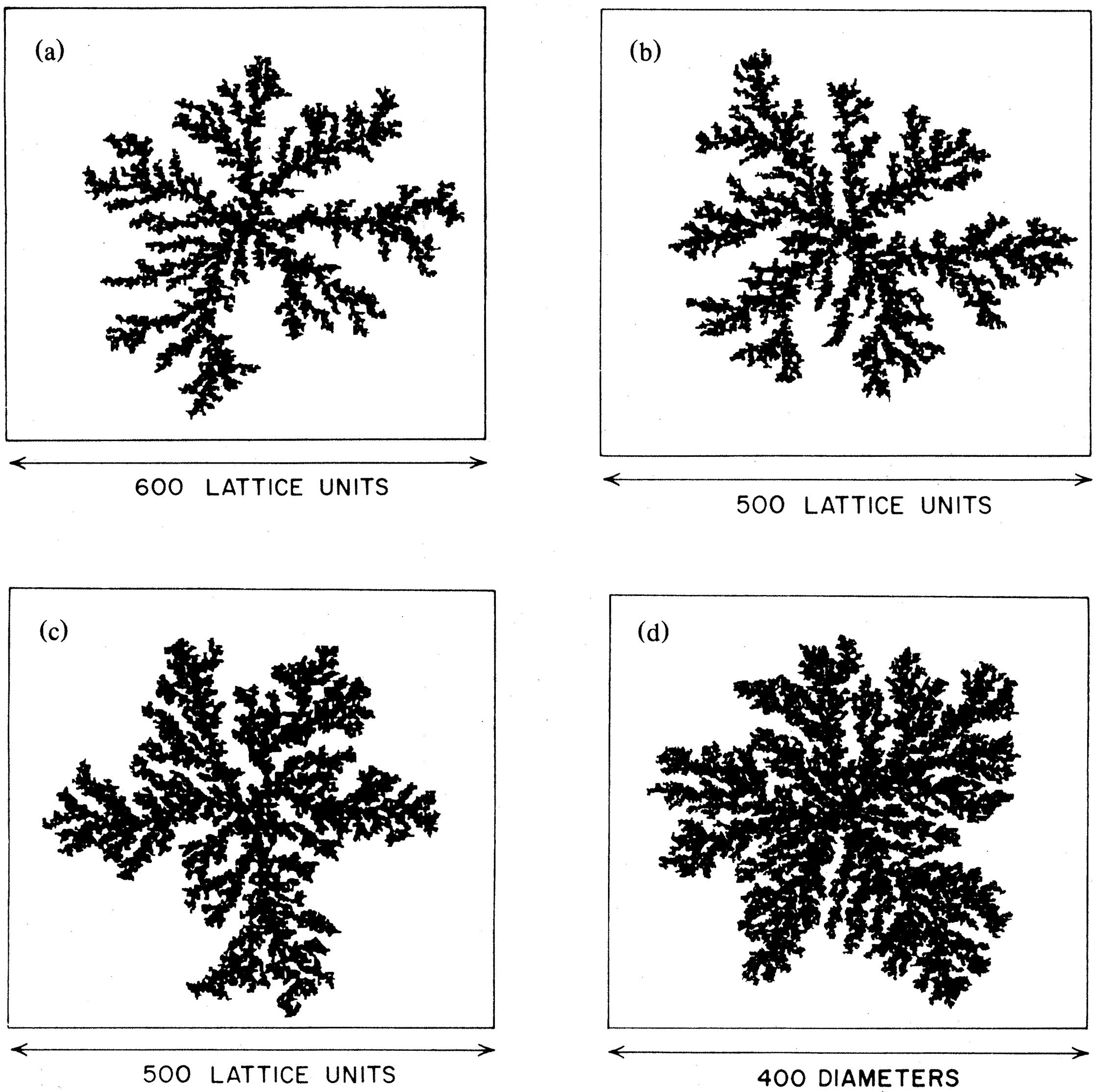

FIG. 1. Clusters consisting of 25 000 occupied lattice sites using a lattice model for particle-cluster aggregation. The clusters were grown using Levy-flight trajectories with the parameter f set to 2.5, 2.0, $\frac{5}{3}$, and $\frac{4}{3}$.

cupied lattice sites. Similar simulations have been carried out with random walks on the lattice ($f \rightarrow \infty$, the Witten-Sander model) and with linear trajectories ($f \rightarrow 0$). The density-density correlation functions $C(r)$ obtained from these simulations and the structures shown in Fig. 1 are given in Fig. 2. Similar simulations have been carried out using the nonlattice model, and Fig. 3 shows the density-density correlation function obtained from clusters containing 25 000 particles each grown using values of 2.5, 2.0, $\frac{5}{3}$ and $\frac{4}{3}$ for the step length exponent (f). Curve A was obtained using a nonlattice version of the Witten-Sander model and curve F was obtained using the Vold-Sutherland model (linear particle trajectories).

For structures with a fractal dimensionality of D and a Euclidean dimensionality of d, we expect plots of $\ln[C(r)]$ vs $\ln(r)$ to be linear with a slope of $-\alpha$, where α is the fractal codimension (d-D). The plots of $\ln[C(r)]$ vs $\ln(r)$ shown in Figs. 2 and 3 show linear behavior over a substan-

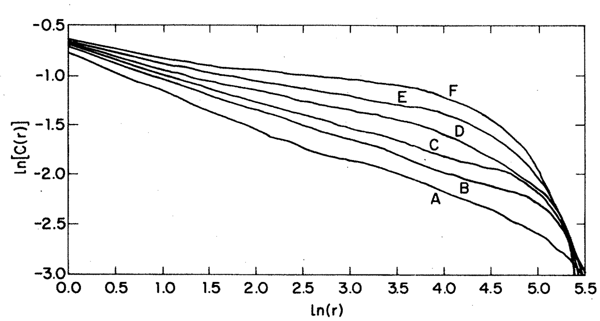

FIG. 2. Density-density correlation functions obtained from clusters of 25 000 occupied lattice sites generated on a square lattice. Curve A was obtained using the Witten-Sander model. Curves B, C, D, and E were obtained using Levy-flight trajectories with the parameter f set to 2.5, 2.0, $\frac{5}{3}$, and $\frac{4}{3}$, respectively, and curve F was obtained using linear trajectories.

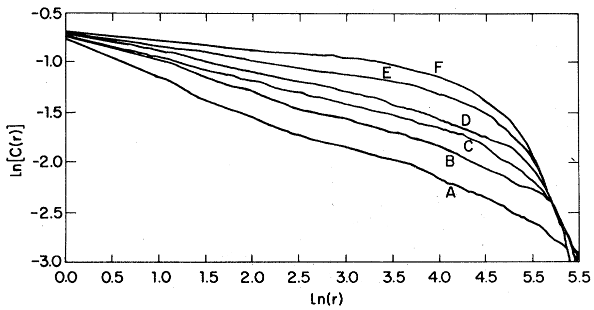

FIG. 3. Density-density correlation functions calculated for clusters of 25 000 particles generated using nonlattice models. Curve A was obtained using a constant step length of 1.0 particle diameters ($f \rightarrow \infty$). This calculation can be regarded as a nonlattice Witten-Sander simulation. Curves B, C, D, and E were generated using Levy-flight trajectories with the parameter f set to 2.5, 2.0, $\frac{5}{3}$, and $\frac{4}{3}$, respectively. Curve F was obtained using linear trajectories.

tial range of length scales with strong deviations at large distances due to the finite size of the aggregates. The results obtained from the lattice models also show deviations at short distances. However, a value for the fractal dimensionality can be estimated from $d(\ln[C(r)])/d(\ln(r))$ at intermediate length scales.

The results shown in Figs. 2 and 3 indicate that the effective fractal dimensionality varies continuously between the limits of $\simeq \frac{5}{3}$ (the Witten-Sander model corresponding to $f \rightarrow \infty$) and $D \simeq 1.92$ (the Vold-Sutherland model corresponding to $f \rightarrow 0$). As expected, both the lattice and nonlattice models give the same values for the fractal dimensionality of the aggregate for a particular f value (within the accuracy of our simulations). Table I shows estimates of the fractal dimensionalities generated by the models discussed in this paper. These results were obtained from the density-density correlation function (D_α) and the dependence of $\ln(R_g)$ on $\ln(N)$, where R_g is the radius of gyration and N the number of particles or occupied lattice sites in the clusters (D_β). Since most of the results given in Table I were obtained from a single simulation of a large ($N = 25\,000$) cluster, we have no estimate of either the statistical or systematic uncertainties both of which may be quite large.

If the parameter f lies in the range 1.0–2.0, then the fractal dimensionality[4] of the particle trajectory D_t is equal to f. For all values of f greater than 2.0, $D_t = 2.0$. Consequently, our results suggest that the aggregate dimensionality decreases continuously as the fractal dimensionality of the trajectories increases but may take on a range of values for $D_t = 2.0$.

Our results are subject to a number of uncertainties associated with finite-size effects and truncation of the particle trajectories to reduce computer time requirements. More extensive simulations are in progress to explore these effects and provide more accurate information on the relationship between the dimensionality of the Levy-flight trajectories and the dimensionality of particle-cluster aggregates formed using their trajectories.

TABLE I. Estimates of the fractal dimensionality of clusters formed by single-particle aggregation using Levy-flight trajectories. The effective dimensionality D_α was obtained from the dependence on $\ln[C(r)]$ or $\ln(r)$ for distances in the range $5 \leq r \leq 30$ (lattice units or particle diameters). The values for D_β were obtained from the dependence of $\ln(R_g)$ on $\ln(N)$ (Ref. 19) for clusters in the size range $2500 \leq N \leq 25\,000$ (except for the nonlattice model with $f = 0$ and the nonlattice model with $f \rightarrow \infty$ where results from earlier work with smaller clusters are given).

	Lattice		Nonlattice	
f	D_α	D_β	D_α	D_β
0	1.93[a]	1.97[a]	1.92[a]	1.95 ± 0.02[a]
$\frac{4}{3}$	1.88	1.85	1.86	1.88
$\frac{5}{3}$	1.80	1.85	1.81	1.85
2	1.75	1.75	1.75	1.82
2.5	1.71	1.70	1.74	1.76
α	1.68 ± 0.04[b]	1.67 ± 0.05[b]	1.68 ± 0.03[c]	1.72 ± 0.05[c]

[a] Reference 5.
[b] Reference 20.
[c] Reference 7.

One possible concern is that the aggregation may be dominated by the small fraction of very long steps in the distribution given in Eq. (1). These very long steps are neglected in our simulations as a result of the termination of the trajectories at a distance of $2.5R_0$ from the origin. It can be shown that if the particles are uniformly distributed, the probability that a particle originating at a distance $\geq R$ from the cluster [where R is much greater than the maximum cluster radius ($R_{\max}$)] will contact the cluster after a single step is proportional to $R^{-(f-1)}$. Clearly, these effects will become very serious as $f \rightarrow 1$. Since neglect of the few very long steps will decrease the apparent value of the fractal dimensionality, the qualitative conclusion reached in this paper should not be influenced by this approximation.

Several theoretical methods have been developed[14–17] which give expressions for the fractal dimensionality of Witten-Sander aggregates. In all cases, the agreement between the theoretical and computer simulation results is good. Results from related models such as the ones discussed above and diffusion-limited aggregation on fractal substrates[18] should provide a more exacting test for the theoretical approaches to diffusion-limited aggregation. The formation of aggregates from particles following fractal trajectories has recently been analyzed by Hentschel[17] using a Flory-type theory. A comparison of our simulation results with theory is contained in Hentschel's paper.

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