Generalized model for the diffusion-limited aggregation and Eden models of cluster growth

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The diffusion-limited aggregation (DLA) model of cluster growth and aggregation has been extended to include both a finite and variable diffusion length for the particle's random walk. The new model generates a variety of different structures similar in geometry to the DLA model; however, they lack the fractal scaling indices normally expected from the DLA model. It is postulated that the DLA and Eden models of cluster growth are both limiting cases of the finite-diffusion-length model where the diffusion length approaches infinity and zero, respectively.

INTRODUCTION

In recent years considerable effort has been directed towards developing models for growth and aggregation processes. Two important and useful models which have received considerable attention are the Eden[1] and diffusion-limited aggregation[2–6] (DLA) models. Both models describe nonequilibrium, kinetic depositions in terms of rate-limited steps where the origin of the rate-limiting step is assumed to differ in both models. For the Eden model, the growth of a cluster is assumed to be restricted by the reaction kinetics at the surface of the growing cluster, while the rate-limiting step for the DLA model is assumed to be the diffusion of a necessary reactant to the surface of the cluster. Important fractal scaling relationships have been found in both models, i.e., relationships of the form $X(N)\propto N^{\nu}$, where $X(N)$ and N are structural properties and $\nu=1/d$ where d is the fractal dimension. In addition to the theoretical significance of the fractal dimensionality, the concept has been successfully applied to a variety of other physical phenomena.[7–11] Until now the Eden and DLA models have been generally considered as similar, but independent theories. In this paper we present a Finite-Diffusion-Length (FDL) model which unifies both models, and shows the Eden and DLA models to both be limiting cases of the more general FDL model.

In a generalized diffusion process, a particle initiates an n-dimensional random walk down a concentration gradient from C_{bulk} to C_0 where C_{bulk} represents the concentration of the particle at $r=\infty$ and C_0 is the concentration at $r=0$. Ideally, the length of the diffusion zone, i.e., the concentration gradient, develops monotonically with time t according to

$$C/C_{\text{bulk}}=\text{erf}\left[\frac{r}{2\sqrt{Dt}}\right], \tag{1}$$

where D is the diffusion constant.[12] A diffusion length L can be defined from Eq. (1) by taking the distance at which the concentration is 99% that of the bulk:

$$C/C_{\text{bulk}}=0.99=\text{erf}\left[\frac{r}{\sqrt{Dt}}\right],$$

$$r_{99\%}=L=3.6\sqrt{Dt}\ . \tag{2}$$

However, in any real diffusion process L is limited in its spatial expansion by randomizing thermal motion and convection, etc.[12,13] After a sufficient time, a steady-state condition develops in which L is virtually constant and serves to define an isoconcentration surface where the concentration of particles is essentially equal to that of the bulk. In a diffusion-based computer model, the isoconcentration surface represents that surface from which there is equal probability to start a particle on its random walk towards the cluster surface. Clusters grown using a finite, constant, and predefined diffusion length would more closely represent reality and would extend the already successful fractal approach of growth and aggregation to encompass a broader spectrum of physical events. It is from this concept that the FDL model was developed. Recent attempts to mimic diffusion-length effects have been reported by Voss *et al.*[14,15] in which a modified DLA cluster grows in a "sea" of randomly walking particles. By altering the particle density in the "sea," varying cluster topologies were obtained. However, the computer logistics of the model still mimic the time-dependent diffusion length of Eq. (1). In FDL the diffusion-length effects are addressed in a considerably simplified computer algorithm which relates the relevant physical properties of the structures to a single independent variable, L [which can also be a function of time as in Eq. (2)]. In addition, the simplification of the model provides a framework in which DLA and Eden models of cluster growth are easily unified.

RESULTS AND DISCUSSION

In the FDL model presented here, a finite and constant diffusion length is used to grow planar structures at various magnitudes of L. A planar configuration was used here, rather than the radial geometries normally used in the DLA and Eden models, because of the applicability of the planar geometry to additional physical phenomena.[10] Definition of the FDL model proceeds as a series of computer simulations which were performed on a periodic, square lattice with an initial surface of 100 lattice points, i.e., the lattice was arranged in a modulo 100, "wraparound" configuration. Simulations proceeded by releasing a particle at a distance of L lattice sites from the surface. The release site of the particle was chosen at random from the locus of all points L lattice units away from the surface. The particle was then allowed to diffuse towards the interface with an on-lattice two-dimensional random walk. When the particle contacted an active site, defined here as an on-lattice site immediately adjacent to the interface, the walk was terminated, the particle was added to the growing structure at the hit site, and the structure's surface was extended by one lattice unit. The process continued with the release of another particle from the newly generated locus of release points L lattice units from the new surface. This process was continued until N particles had been added. N was typically between 10^3 and 10^4 particles. Particles which diffused a distance of more than $2L$ lattice units away from the interface were aborted and a new particle started. Figure 1 shows a typical computer structure generated from the FDL model using a diffusion length of ten lattice units. The line located ten lattice units above the top of the structure is the diffusion surface and represents the locus of points from which the next particle would initiate its random walk. The conformal shape of the diffusion surface in relationship to the structure's interface demonstrates the dynamic interplay between the diffusion length and the growing structure.

The scaling relationships of the FDL-generated structures are best demonstrated in the *linear* density, ρ, versus distance, r, graphs such as the one shown in Fig. 2. Although Fig. 2 presents data for only one $L=10$ structure, all FDL structures showed similar behavior. It is

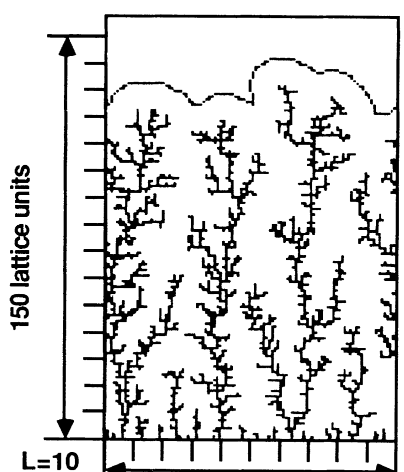

FIG. 1. Typical computer simulation showing a structure formed by the FDL model. The line at the top of the structure is the diffusion surface from which the next diffusing particle would start its random walk. This structure was generated using a diffusion length L of ten lattice units.

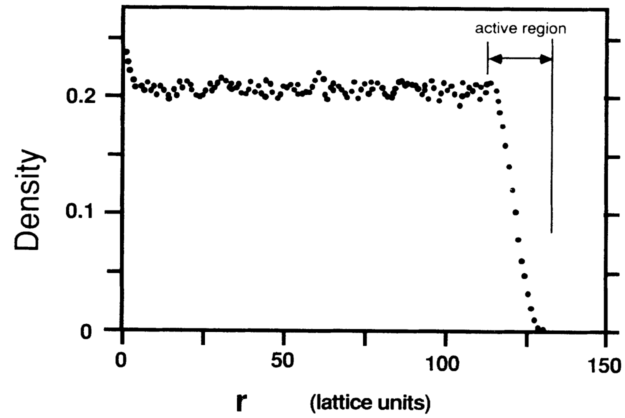

FIG. 2. Typical plot of density vs distance for a FDL structure produced with an L value of ten lattice units. The density is presented as the fractional area occupied by branches between r and $r+1$ lattice units and plotted as a three-point moving average.

clear that, except for the interfacial regions, at $r=0$ and within the active region, the density remains constant throughout the entire thickness of the structure. Using the standard scaling relationship for a constant density structure of 100 lattice points wide and containing a total of N particles gives

$$N^{v}=100\rho r \ , \tag{3}$$

where $v=1$. An integer value of v implies that a nonfractal scaling relationship exists between the density and N

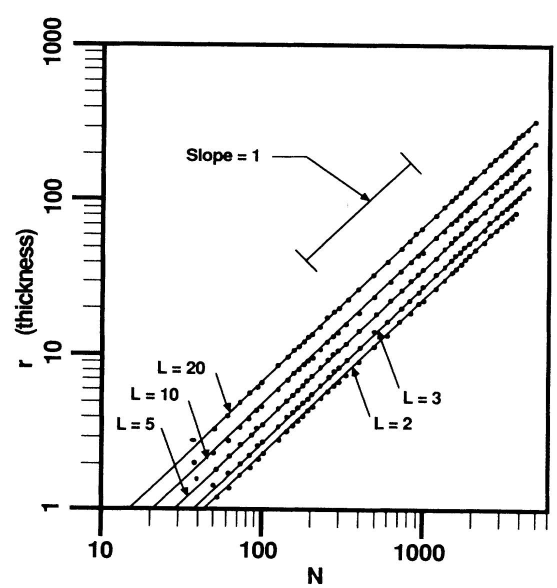

FIG. 3. $\log_{10}$-$\log_{10}$ plot of the average thickness of the FDL structures, r, vs the number of particles added to the structures, N. Plots are shown for $L=2$, 3, 5, 10, and 20 using averages of between three and six samples. All plots were linear with slopes equal to 1, except for small r where interfacial effects were present. Samples with $L=20$ were performed on a 200-lattice unit base.

with $100p$ as the constant of proportionality. Figure 3 demonstrates the validity of Eq. (3) by graphing the $\log_{10}$-$\log_{10}$ plots of a selected number of FDL structures with L values ranging from 2 to 20. All demonstrate the functionality of Eq. (3) with slopes v equal to 1.01 ± 0.09 and extrapolated N intercepts equal to 100 times the fractional density of the FDL structures. The structures generated by the FDL model are clearly nonfractal despite the similarity in appearance to DLA structures [compare Figs. 1 and 6(a)], and the similarity in the computer algorithms. Other common DLA scaling relationships were also investigated including density-density correlations, branch or cluster height, etc., but all lacked the expected fractal dimensionality. Although none of the standard DLA fractal scaling relationships applied to FDL structures, one interesting fractal scaling relationship between the diffusion length and density did appear. Shown in Fig. 4 is the $\log_{10}$-$\log_{10}$ plot of density versus diffusion length. It is clear, at least for the decade change in L shown here, that the graph is linear with a slope of -0.461. A correlation factor of 0.91 indicates that although a strong correlation exists, the error and small statistical sampling is sufficient to suggest that a trivial (nonfractal) scaling exponent of -0.5 might also be possible.

Complete details of the FDL growth characteristics are given elsewhere.[10] However, it is easy to envision the processes responsible for the constant density structures shown in Figs. 1 and 2. In FDL a dynamic "feedback" exists between the conformal diffusion surface and the growing cluster which favors a continual radial expansion of each branch until it fills up all of the available space. However, when two branches begin to approach each another at a distance of about $2L$, the diffusion surfaces begin to "pinch off" and the branches cease to grow further. This implies that one would expect the average interbranch distance to be on the order of $2L$ for FDL structures. This assumption is verified in Table I, where

TABLE I. Average interbranch distance for a number of computer simulations with diffusion lengths between 2 and 20. The mean interbranch distance is given as the average of 13–20 samples and the error is given as the standard deviation from the mean.

Diffuse length L	$2L$	Mean interbranch distance
2	4	3.9 ± 2.1
3	6	6.5 ± 3.7
5	10	9.8 ± 3.8
10	20	16.5 ± 5.4
20	40	39.4 ± 9.5

the average interbranch distances are shown to correlate well with twice the diffusion length.

In addition to the bulk scaling characteristics, the active region of DLA and Eden clusters are known to possess different scaling relationships.[16] In fact, Meakin *et al.* have shown that the surfaces of fractal and nonfractal objects possess scaling relationships which can be described as an infinite hierarchy of critical exponents,[4,17] while Plischke and Racz have performed Monte Carlo calculations for the DLA and Eden interfaces and concluded that the interfacial "hit" distribution of sites is Gaussian.[18,19] From Fig. 2 one can see that the active region of the FDL is clearly different than that of the bulk structure, and functional relationships within the active region are important. However, because of the small size of the computer sampling and lattice, it was not possible to determine a definitive functional relationship for the active region and continued work in this area is needed. However, the widths of the active region are easily obtained from the linear density versus distance graphs

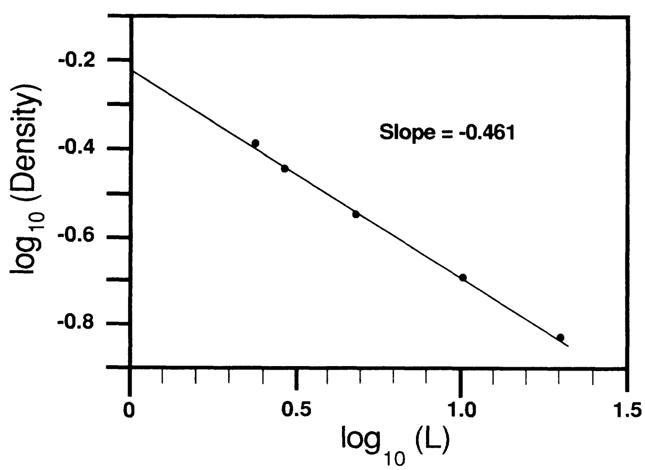

FIG. 4. $\log_{10}$-$\log_{10}$ plot showing the relationship between the density of FDL structures and the value of the diffusion length from which they were formed. A least-squares line is shown drawn through the data points with a correlation coefficient of 0.91.

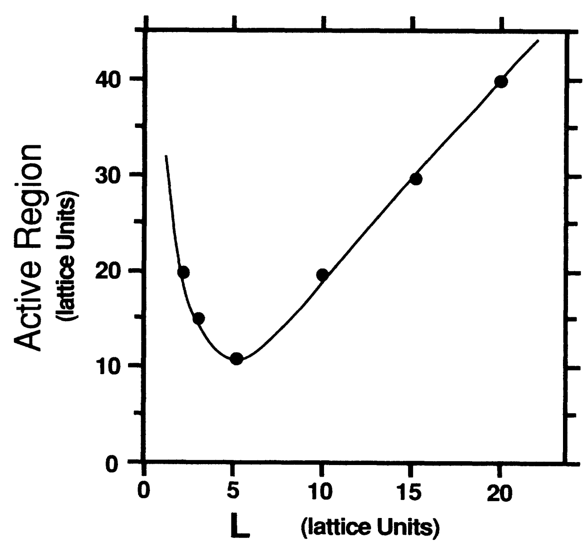

FIG. 5. Relationship between the diffusion length L and the width of the active region of FDL structures formed at various values of L. The width of the active region was taken as the number of lattice units between the largest occupied value of r and the point at which the density became constant. Samples with $L = 15$ and 20 were performed on a 200-lattice unit base.

such as the one shown in Fig. 2. These active region widths are shown graphed in Fig. 5 as a function of the diffusion length from which the structure was generated. Figure 5 shows a minimum in the active region width at L approximately equal to 4. Interestingly, at higher-L values the graph approaches linearity with a slope of 2, suggesting that the width of the active region follows the same functional relationship as that found for the average interbranch distance, i.e., the active region is equal to $2L$.

Figure 6 shows a sequence of FDL structures "grown" using values of L ranging between 10 and 2. In addition, typical DLA and Eden structures are reproduced at the beginning and end, respectively, for comparison.[3,20] The DLA and Eden structures were intentionally placed in their positions relative to the FDL sequence to emphasize their proposed relationship within the FDL model. The single variable L serves to define the topological transition between the various FDL structures, and it is noted that as L decreases, the FDL simulations progress from structures closely resembling DLA morphologies to more dense and compact structures reminiscent of an Eden growth process. From this gross comparison, it is tempting to postulate that the DLA and Eden models are limiting cases of the more general FDL model as L approaches the limiting values of infinity and zero, respectively.

A more convincing argument for the unification of the DLA and Eden growth models within FDL comes from the equivalence between the computer algorithms used to generate DLA or Eden structures with the algorithms of FDL at $L=\infty$ or 0, respectively, i.e., identical computer algorithms will generate identical structures. In comparing DLA with FDL the only difference in the models is the choice of initial conditions as to where to start the particle's random walk. The actual random walk of the particle, definition of active sites, and "sticking" probabilities are all defined identically in both models. In FDL the particle begins its random walk from a dynamic diffusion surface that is finite and conformal to the interface of the growing structure. As L tends toward infinity, the diffusion surface tends towards a straight, parallel line with equally straight and parallel isoconcentration surfaces all the way up to distances approaching the structure's interface. Hence a FDL structure at $L=\infty$

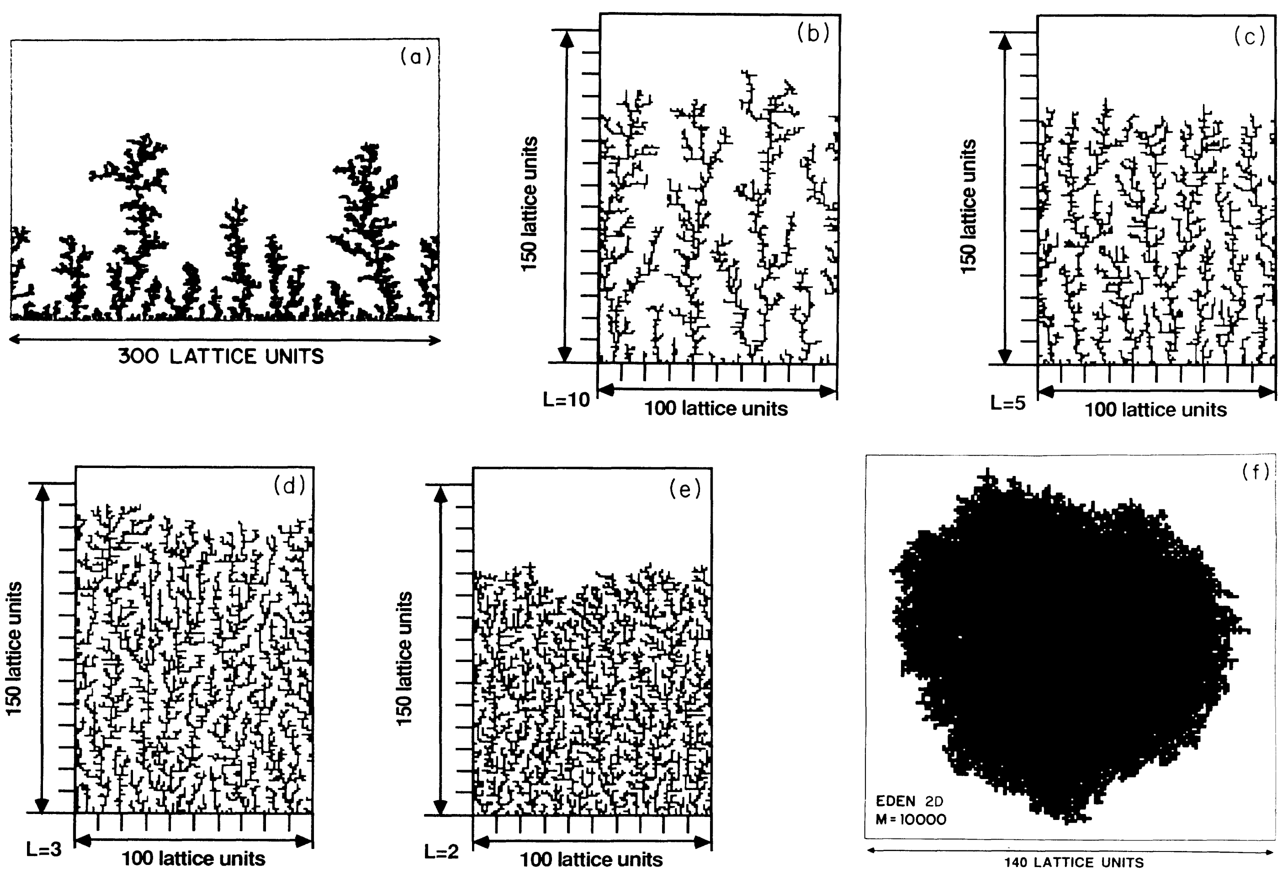

FIG. 6. Figures showing the structural topology obtained in the FDL model for an ordered sequence of L values from 2 to 10. Also shown with the FDL structures is a DLA structure (a) formed on a similar planar substrate as that used in the FDL model (reproduced with permission from Ref. 3), and an Eden model cluster (f) produced from a single-point substrate (reproduced with permission from Ref. 20) for comparison. (a) DLA model. (b) FDL with $L=10$. (c) FDL with $L=5$. (d) FDL with $L=3$. (e) FDL with $L=2$. (f) Eden model.

could be approximated by using a straight diffusion surface which is parallel to the initial surface. In the computer simulation of the DLA model, the initial starting position for the particle's random walk is determined by randomly selecting a lattice position at some arbitrary distance from the upper bound of the cluster. With an isotropic random walk, such a choice is tantamount to assuming a straight and parallel isoconcentration surface and, hence, implies an infinite diffusion length. DLA is, then, best considered as the finite computer approximation of the unobtainable, FDL, $L=\infty$ limit. The physical density of the $L=\infty$, FDL structure can be obtained from Fig. 4 by extrapolating L to infinity. In the limit the density of the FDL structure becomes zero, which is consistent with the density of a DLA cluster in the limit of large N. In addition, extrapolation of Fig. 5 to $L=\infty$, with the assumption of the previously discussed $2L$ relationship, indicates that the active region or penetration depth of a FDL structure also tends to infinity, which is consistent with that found for DLA.[18,19] The DLA structure, then, can best be characterized as the active region of a partially formed FDL structure when L is equal to infinity. For finitely formed structures as are normally found in real physical examples, the normal DLA morphologies are applicable to structures whose thickness is much less than the diffusion length (or whose diffusion length is a function of time).

At the other end of the spectrum, the Eden growth model represents the limiting case of the FDL model for when L becomes vanishingly small. Ignoring for the moment the discrete nature of the growth lattice, a zero diffusion length corresponds to a conformal diffusion surface coincident with the cluster surface. Random selection of any point along the diffusion surface would automatically place the particle at an active site and diffusion, i.e., the random walk, would not occur. Such a condition is exactly the definition of the computer mechanics for a reaction controlled growth at the surface of the cluster, i.e., the Eden growth model, indicating that the Eden and FDL are equivalent structures at the limit of $L=0$. The graph of Fig. 5 shows the width of the active region in FDL increases as L approaches zero, consistent with the expected surface scaling properties of the Eden model as N becomes increasingly large. In reality, however, a FDL zero diffusion length implies that the lattice spacing also tends towards zero. If a finite, nonzero lattice spacing is considered, a condition that more closely resembles nature, then a value of one lattice unit would represent the smallest value of L obtainable. Extrapolation of L to one lattice unit in the graph of Fig. 4 ($\log_{10}L=0$) shows that the density for a structure generated with $L=1$ is 0.595, in close agreement with the density of Eden clusters grown on an infinite percolation network at threshold, 0.593.[1,16]

SUMMARY

The FDL model of cluster growth represents a realistic and general representation of physical growth and aggregation phenomena and contains within its framework the already successful DLA and Eden models as limiting structures. In addition, the FDL model presents the possibility that nonfractal objects of constant physical properties (such as density) may possess an inherent but hidden fractal nature in their generation or existence. Certainly, the density versus distance graphs of Fig. 2 strongly suggest a generalized phase transition between differing dimensional spaces which could possibly represent such diverse phenomena as the electron or energy state distribution at an interface, or the penetration of electromagnetic radiation into an opaque object. Clearly, more work is required, particularly on the functional relationships in the active region, before any such analogies can be regarded seriously. However, the FDL structures are interesting in their own right and deserve further attention.

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