Segregation of fractal aggregates grown from two seeds

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We study the generalized diffusion-limited aggregates, grown from two proximal nucleation seeds placed at distance d lattice units and investigate the probability $p(d)$ that these aggregates get connected. We vary the sticking probability to get a range of aggregate geometry from fractal to compact one. For fractal aggregates, $p(d)$ decays rapidly with d, while for compact ones, the decay is so slow that $p(d)\approx 1$ for all practical distances. We experimentally demonstrate similar behavior for viscous fingering patterns with two injection points and electrochemical deposits grown on two cathodes. Our observations along with previous results on competitive growth suggest a common underlying principle.

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Aggregation of particles is a common phenomenon in many branches of applied sciences and is of interest from commercial and fundamental viewpoints [1–3]. The related studies have been greatly advanced due to novel experiments [1] as well as the rapid development of growth models [4]. Amongst several models, diffusion-limited aggregation (DLA) model developed by Witten and Sander [5] has arguably been the most important one. The morphology of the DLA patterns is very much similar to those obtained in many Laplacian processes such as bacterial growth, solidification, viscous-fingering, electrodeposition, etc. [1,6–8].

In many experiments and simulations, the aggregate are grown from a static nucleation seed, which is not necessarily a naturally occurring situation. This could be a special case of more general circumstances, where, growth starts simultaneously from several proximal nucleation seeds. In this work, we study in detail, the connectivity of aggregates grown from two proximal seeds as a function of their morphology and separation between them. We believe that apart from theoretical interest, these studies could have practical applications in the area of formation of extended network. For instance, if neighboring clusters of conducting particles get connected then one would get a dramatic change in the electrical and thermal conductivity of the structure. Witten and Meakin, to some extent, have attempted a similar issue in their work on multiple seed DLA grown on a two-dimensional (2D) square lattice. It was shown that, DLA clusters simultaneously grown on two seeds separated by nine vacant lattice sites remain unconnected during their simulation time [9]. This issue also finds a passing mention in the subsequent study by Meakin on a model for biological pattern formation [10]. In this model, randomly chosen active site gets occupied with probability $P\sim c^{\varepsilon}$, where c is the local concentration of a nutrient diffusing from a surrounding exterior source and consumed by the growing cluster and ε is a positive parameter. Simulations on 2D square lattice with two seeds separated by ten lattice-sites showed that the clusters remained disjoint for $\varepsilon=1.0$ and 2.0 while get connected for $\varepsilon=0.5$.

Fujikawa *et al.* [11] explored segregation of two bacterial colonies where, bacterial stain, *Bacillus Subtilis*, was simultaneously inoculated at two proximal points on an agar plate. It was observed that bacterial colonies did not join under moderate nutrient concentration but fused together for high nutrient concentration. We recall that under starvation conditions, bacterial colonies show growth similar to a DLA [7].

In case of aggregates grown from two separate seeds, naive expectation would be that they would connect together if one waits long enough. Systematic studies of this problem are required for checking if this is really true. In this work, we present a systematic study of the behavior of probability $p(d)$ that the aggregates generated from two proximal nucleation seeds get connected for a range of interseed distances d using a generalized DLA (GDLA) model. We also demonstrate a similar phenomenon experimentally in viscous fingering with two injection points and electrochemical deposition with two cathodes. All these studies seem to have interesting common underlying features.

While interseed distance obviously affects connectivity of cluster, we find that the morphology or fractal dimension D_F, of the clusters also plays a crucial role. A DLA model on a 2D square lattice can be generalized by introducing a parameter called sticking probability, which allows us to vary D_F of clusters [12,13]. In this model, particles stick to the cluster on visiting active site with sticking probability s, defined as, $s=\alpha^{3-B}$, where α is some positive parameter ($0<\alpha\leqslant 1$) and B is the number of nearest-neighbor occupied sites in the cluster [12]. It is shown that for $\alpha=1$, GDLA patterns are fractal ($D_F\sim 1.7$) while for $\alpha\to 0$, D_F approaches the limiting value 2, since active sites with $B=3$ are more likely to get occupied than those with $B=1$ [13].

We simulated GDLA model on a 2D square lattice with two seeds separated by a d lattice units. They are placed at the lattice points $([-d/2],0)$ and $([d/2],0)$, where $[x]$ stands for largest integer $\leqslant x$. The effects of asymmetric placement of seeds (in case of odd d values) relative to the launching circle were expected to be insignificant since its initial value of radius $r=100$ lattice units was much larger than the distances d. (Radius r was carefully increased as the cluster grows.) A particle was launched from the lattice site nearest

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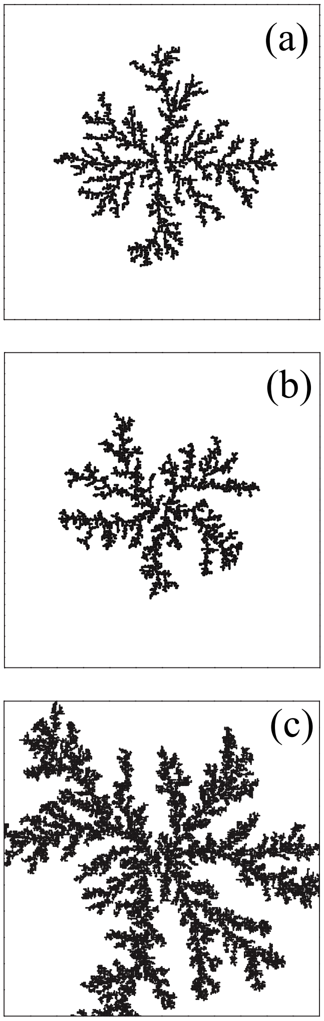

FIG. 1. Representative morphologies of the GDLA patterns generated using two seed configuration for $d=9$ lattice units. (a) $\alpha=1.0$; $D_F=1.67$, (b) $\alpha=0.7$; $D_F=1.74$, and (c) $\alpha=0.3$; $D_F=1.84$.

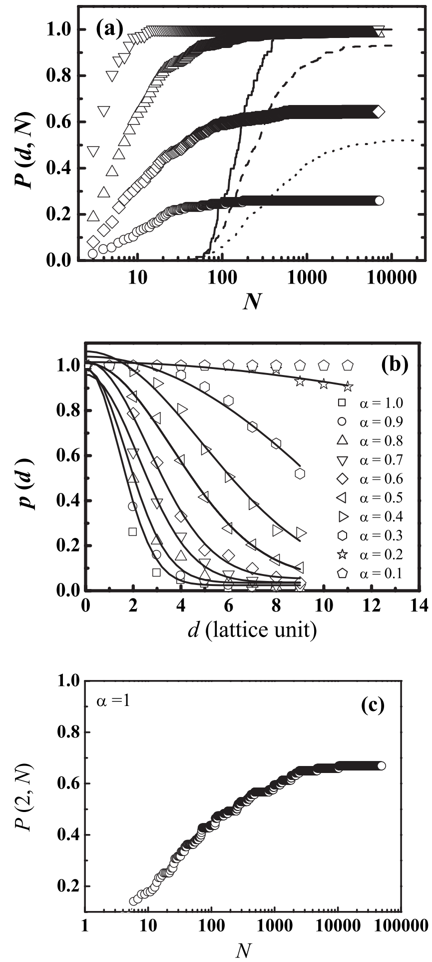

FIG. 2. Plots shows (a) probability of connectivity $P\ (d,N)$ versus number of particles N plotted for $d=2$ lattice units ($\alpha=1.0$ [○]; 0.7 [◇]; 0.4 [△]; 0.1 [▽]) and $d=9$ lattice units ($\alpha=0.3$ [···]; 0.2 [----]; 0.1 [—]), (b) asymptotic probability of connectivity $p(d)$ against interseed distance d for various α values and (c) variation of $P(2,N)$, against N for 3D clusters generated using two-seed configuration on cubic lattice.

to the randomly chosen point on this circle. It carries out a random walk and becomes a part of the cluster with sticking probability $s=\alpha^{3-B}$. If a walker becomes a part of both the clusters, the clusters are said to be connected. We vary α from 1 to 0.1 and d from 2 to 9 lattice units.

First we discuss the representative morphology of GDLA generated from two seed configuration. In Figs. 1(a)–1(c) we show the aggregates for $\alpha=1.0$, 0.7, and 0.3, respectively. Their fractal dimension was $D_F \sim 1.67$, 1.74, and 1.84, respectively. For all the cases, distance between proximal seeds was $d=9$ lattice units. It is observed that as α decreases D_F increases, which is consistent with the result by Banavar *et al.* [13]. Furthermore, we clearly see that, these two seed aggregates remain unconnected. This segregation in the growing cluster is primarily arises due to well-known shielding effect in DLA [12], which prevents a walker from penetrating in the empty spaces between the clusters. However, as $\alpha \to 0$, a walker has more chance to visit the active sites with $B=3$ than 2 or 1, resulting to a compact morphology.

One can argue that the aggregates do not meet at certain stage, could meet on addition of some more particles. Obviously, no simulation can run for infinite time and systematic approach must be employed to find the asymptotic probability for this stochastic growth. We identify a quantity $P(d,N)$ as probability of connectivity between two clusters where, N is the total number of particles in the clusters and d is the distance between seeds. For each set of α and d values, we generate 1000 configurations. In each configuration, clusters were observed for connectivity till $N_{\max}$ particles are added. $P(d,N)$ is then the fraction of 1000 configurations which get connected on or before the accumulation of N particles in the clusters. Figure 2(a) shows a plot of $P(d,N)$ as a function of N on a semi-logarithmic scale for some α and d values. As expected, for larger d values, we do not see any connectivity until adequate particles are aggregated. It is also evident that, the probability $P(d,N)$ saturates in all the cases for large enough N. If the saturation value is not reached for $N_{\max}$ particles, we try a higher value of $N_{\max}$ till saturation is clearly obtained. We denote this asymptotic probability by $p(d)$. As expected, for larger d, saturation occurs at higher value of N and $p(d)$ is smaller. The plot also illustrates that $p(d) \rightarrow 1$ as $\alpha \rightarrow 0$ due to reduction in shielding effect

In Fig. 2(b), we plot asymptotic probability $p(d)$ against d for different α values. We observe that it decays rapidly with d for α values close to 1. However, the decay is slower for smaller values of α. We could fit this data points using a Gaussian form $p(d)=\frac{1}{\sqrt{2\pi}\xi}\exp\left(\frac{-d^2}{2\xi^2}\right)$, where ξ is the root mean square (rms) deviation. The continuous line shows the fitted curve and the fit is excellent. The Gaussian nature of $p(d)$ may be primarily due to that the probability of two clusters getting connected is essentially related to the overlap integral of growth probability distribution or accumulation function [14] of the individual clusters. It is known that the radial growth probability of the individual clusters show Gaussian behavior [15] and for Gaussians, overlap integral is again a Gaussian.

We also conjecture that for compact 2D structures ($\alpha \rightarrow 0$), rms deviation becomes very large ($\xi \rightarrow \infty$) and $p(d) \approx 1$ for all practical values of d. Thus there is a significant difference in the connectivity properties of fractal and compact structures.

We also probed the connectivity properties of compact cluster using another growth model, namely, ballistic driven aggregation (BDA) with four-sided rain [16]. Certainly this model leads to a compact cluster where D_F approaches the limiting value 2. The model with two seeds is as follows: the two seeds separated by distance d are placed at positions on the 2D square lattice. A particle starts at any site on the perimeter and follows ballistically. If perimeter site is on right (left) edge, particle moves towards left (right). Similarly if it is on top (bottom), particle moves to bottom (top). It gets attached to the cluster on visiting any active site. Here, we observe that even for large values of d ($d=50$ lattice units), the clusters connect with probability $p(d) \approx 1$.

These observations also raise an interesting question. Does this merging phenomenon have some critical dimension above which the clusters created from two proximal seeds certainly merge? Since we observe that compactlike 2D clusters do merge with probability 1, would the growing clusters certainly meet each other when underlying dimension is greater than 2? To settle this question, we carried out GDLA simulations in 3D space with two seeds placed at distance $d=2$ lattice unit. We observe that the asymptotic value $p(d) \sim 0.67$ is significantly less than 1 [see Fig. 2(c)]. This implies that the crucial factor that distinguishes between the probability that patterns likely to connect and those remain segregated is whether the underlying clusters are fractal or compact in a given embedding Euclidean space $D_E > 1$. For all the practical distances, compactlike structures are more probable to merge and the fractal structures have small value of merging probability.

We experimentally studied the connectivity in viscous fingering and electrodeposition systems. As mentioned before, patterns generated in several experimental situations appear such as DLA. Recently, it has been confirmed by comparing the growth dynamics that viscous fingering and DLA process indeed belong to the same universal class [8]. The viscous fingering phenomenon is as follows. When a low viscosity fluid is injected into a high viscosity fluid, it advances in the form of fingers. It is of relevance to problems such as indirect recovery of petroleum oil. Our experimental setup is a radial Hele-Shaw cell, which consists of two horizontal parallel plates uniformly filled with a high viscosity fluid. A low viscosity fluid is injected through a hole drilled into a top glass plate. We use glycerine ($\mu=850$ cP), as a high viscosity fluid and air as a low viscosity fluid. Two smooth plexiglass plates of thickness 1.0 cm and lateral dimension 60×60 cm^2 were used to construct the cell. Uniform plate separation of 0.4 mm was maintained using Teflon® spacers. Two holes of diameter 0.2 cm were drilled into top plate at a distance d. Air at constant pressure, was simultaneously injected through both the holes. The viscous fingering patterns were realized for various d values ($d=2$ to 7 cm) and at different air pressures. Fingering events were video recorded using CCD camera and VCR. The process was repeated more than 10 times for each set of parameter values.

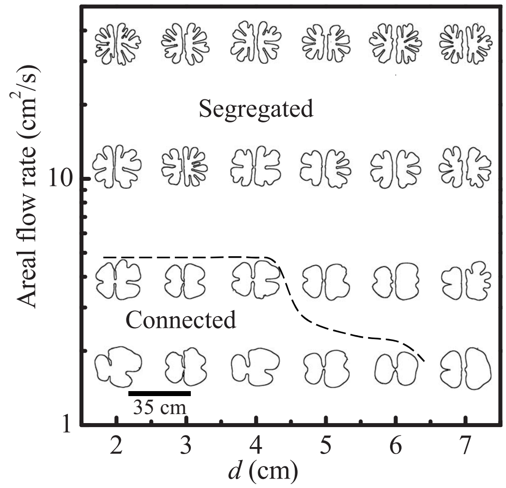

FIG. 3. Connectivity phase diagram for two-point injection viscous fingering in radial Hele-Shaw cell for air-glycerine system. The line is drawn to guide the eye for separation between two regimes.

Figure 3 shows the representative morphologies of the viscous fingering patterns plotted for different areal-flow rate against distance d. It can be clearly seen that there exist two distinct regimes of patterns. When areal flow rate is low, two patterns are connected in all the ten realizations. These patters are certainly compact and their fractal dimension is $D_F \sim 1.93\pm 0.05$. However, at high areal flow rate patterns remain segregated in all the ten trials. We can see that these patters depart from compact morphology. The fractal dimension calculated for the patterns realized at highest areal flow is $D_F \sim 1.77\pm 0.03$. We are unable to examine exact transition regime due to experimental limitations. However, basic phenomenon is illustrated here as well. For segregated viscous fingering patterns, average separation between two clusters is found to be $d/2$. It is interesting to note that this feature was also seen for segregated GDLA clusters.

Electrodeposition is another physical process, which closely resembles DLA [1]. We study Zn electrodeposits in Hele-Shaw cell for two-cathodes geometry. Here, glass plates were 5×5 cm^2 area and were separated by a ring

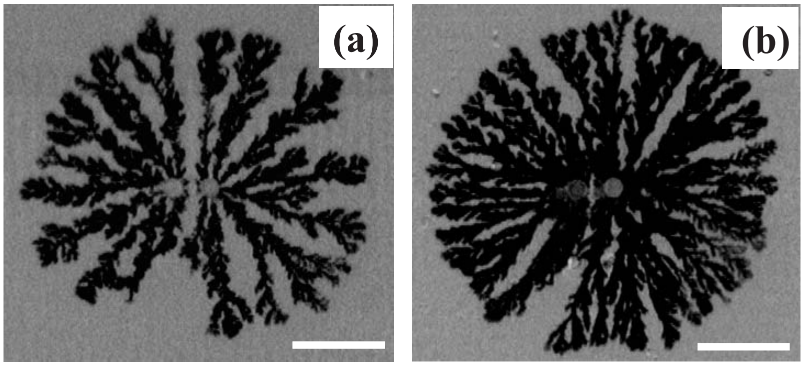

FIG. 4. Zn electrodeposits using aqueous $ZnSO_4$ solution for two-cathodes placed at a distance 0.2 mm and both are connected to a same −5.6 V with respected to ring anode. Segregated and connected patterns are observed for electrolyte concentrations (a) 0.01 M, (b) 0.02 M, respectively. Two wire electrodes (diameter =0.2 mm) can be clearly seen in photograph. Horizontal scale bar is 1 mm long

anode of thickness 0.1 mm. Figures 4(a) and 4(b) show representative electrodeposits from a set of 10 trials. It is seen that two patterns remain segregated for moderate to low electrolyte concentration [Fig. 4(a)] and merge at high electrolytic concentration [Fig. 4(b)]. We have also confirmed the connectivity by measuring resistance between the deposits. In the segregated state, the resistance was of $M\Omega$ order, which drops to order of $k\Omega$ when the clusters get connected. Even in this case, the patterns, which merge are more compact ($D_F \sim 1.92 \pm 0.01$) than ones that remain segregated ($D_F \sim 1.86 \pm 0.01$).

For viscous fingering (electrodeposition) process, capillary (diffusion) length is of the order of mm (nm), which puts lower limit on the separation distance between the injection points (cathodes). Therefore we could not investigate a smooth transition in the connectivity as a function of d. We would, however, like to point out that, to the best of our knowledge, viscous fingering with two-point injection and electrochemical deposition with two cathodes is the first experimental studies of this kind.

These results can be interpreted from a broader perspective. We argue that these are the cases of segregation induced by competition for limited resource. When we have a fractal growth, percentage of occupied sites decreases with increasing cluster radius. (In fact, it is zero asymptotically.) Thus the resource available for growth is limited and is reflected in segregation. For a Hele-Shaw cell, width of advancing finger-tip reduces with increasing fingertip velocity [17]. Thus resource available for growing the tip reduces. The electrodeposits remain segregated for low concentration, i.e., for a limited availability of the resource. We can understand previous experimental and theoretical results in the same spirit. Fujikawa *et al.* showed that under starvation condition, which is a case of limited resource, the bacterial colonies show fractal growth and remain segregated [11]. Meakin's model in which probability of cluster growth is proportional to c^{ε} ($c < 1$), aggregates connect only for $\varepsilon = 0.5$ and not for $\varepsilon = 1$ and 2. Thus segregation occurs at weak growth probability. In all the above cases, segregated clusters are fractal while the connected clusters display a compact morphology.

Growth models have been proposed for phenomena ranging from bacterial growth to the thin film deposition [18]. Thus this study could have important practical implications. On the quantitative side, there are questions, which merit further investigation. The Gaussian dependence of probability of connectivity as a function of distance needs to be studied further. We have also observed that mean distance of separation between two clusters is $d/2$ in simulations as well as in experiments on viscous fingering. These quantitative features could be generic and detailed studies are needed to establish it.

To conclude, we have studied the connectivity properties of several growth phenomena from theoretical and experimental perspective. We observe that, while the compact aggregates definitely get connected with probability $p(d) \approx 1$, fractal patterns grown from proximal seeds may or may not connect and the probability decreases rapidly with distance. We conjecture that the competition for limited resources (which also results in fractal growth) induces segregation in growth processes initiated from proximal seeds. We explain previous studies and put them in a perspective and all these studies seem to point towards the above common underlying principle.

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