Hydrodynamic instability and the structural phase transition in diffusion-limited aggregation with drift

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The Saffman-Taylor instability is extended to diffusive systems with an imposed drift. Under uniform motion of the interface with velocity V , the modified Saffman-Taylor instability is found to occur in the growth process. When the strength of the attractive drift becomes larger than a critical value, the flat interface is stabilized. The structural phase transition from dense aggregation, through diffusion-limited aggregation, to the needle pattern, which occurs when the direction of the drift changes, gives evidence for the presence of a hydrodynamic instability. The morphology diagram is obtained to describe distinct patterns in the diffusion-limited aggregation with drift. The similarity to viscous fingering under a gravitational field is discussed.

I. INTRODUCTION

Pattern formation in systems far from equilibrium is a subject of considerable current interest.¹⁻⁷ A number of models for cluster formation have been proposed, and a variety of computer simulations have been carried out to investigate the relationships between the cluster geometry and growth mechanism.⁸⁻¹³ Examples of pattern formation in a wide variety of diffusive systems include crystal growth,¹⁴⁻¹⁷ electrochemical deposition,¹⁸⁻²² viscous fingering,²³⁻²⁵ and dielectric breakdown.²⁶ Witten and Sander²⁷ proposed the diffusion-limited aggregation (DLA) model to treat pattern formation in the diffusive systems. Simple generalizations of the DLA model have been proposed to take into account the stabilizing force of the surface tension.^{28,29} Meakin³⁰ investigated the effect of particle drift on DLA with the use of a biased random walker and found a crossover phenomenon from the DLA fractal to the dense aggregation. Voss³¹ proposed the multiparticle fractal aggregation model and found a similar crossover with increasing particle concentration. Furthermore, the crossover was found to vary with decreasing sticking probability. Nauenberg, Richter, and Sander³² analyzed the crossover by using the mean-field approximation. Nagatani³³ found a new structural phase transition in the diffusion-limited aggregation with drift model (DLAD). The structural phase transition from the dense aggregation through the DLA fractal to needle pattern occurs when the direction of the drift changes from attraction to repulsion. A question of whether or not the phase transition is related to a hydrodynamic instability,^{34,35} similar to the Saffman-Taylor instability is a worthwhile subject.

Paterson³⁶ first pointed out the relationship between DLA and the Saffman-Taylor instability. The Saffman-Taylor instability occurs when the aggregation process changes from decay to growth. He found the two distinct morphologies: DLA in the growth process and anti-DLA in the decay process. The morphologies are separated by the transition point of the hydrodynamic instability.

In this paper, we investigate the hydrodynamic instability in a diffusive system with an imposed drift. The Saffman-Taylor approach to the stability of the interface is extended and applied to the diffusion-limited aggregation with drift. We perform the simulation for the pattern formation in the diffusion-limited aggregation with drift. The structural phase transition is found with changing direction of the drift. We study whether or not the structural phase transition is related to the hydrodynamic instability. We present the morphology diagram describing the distinct patterns.

The organization of the paper is as follows. In Sec. II we apply the Saffman-Taylor approach to the stability problem of the interface in the diffusion-limited aggregation with drift. We study the hydrodynamic instability in the growth process under uniform motion of the interface with velocity V . The modified Saffman-Taylor instability is found. In Sec. III we present the simulation result. The morphology diagram shows the distinct patterns. In Sec. IV we discuss the similarity with the viscous fingering under a gravitational field. In Sec. V we present the summary.

II. HYDRODYNAMIC INSTABILITY IN DLAD

We consider the problem whether or not the flat interface is stable in two dimensions when the aggregate is forming in the convective diffusion field. The drift is considered in a direction perpendicular to the interface. We apply the Saffman-Taylor approach to the stability problem of the interface. The concentration C of diffusing particles satisfies the convective diffusion equation under the quasistationary approximation (see Fig. 1)

$$u \frac{\partial C}{\partial x} = D \nabla^2 C, \quad (1)$$

where u is the x -directional velocity. The boundary condition for C is the following:

$$C = C_s \text{ at the interface}, \quad (2)$$

where C_s is the saturation concentration. We consider

FIG. 1. Schematic representation of the diffusive system with a drift. To describe a disturbance of the interface, take rectangular coordinates (x, y) , the instantaneous position of the undisturbed interface coinciding with the plane $x = 0$.

that the interface moves with a uniform velocity V . The mass conservation law on the interface is satisfied:

$$uC - D \frac{\partial C}{\partial x} = -C_a V, \quad (3)$$

where C_a is the concentration of the aggregate. We restrict ourselves to the growth process in which the velocity V of interface is a positive value.

To describe a disturbance of the concentration induced by small deviations of the interface, separate the concentration field into the undisturbed and disturbed parts, the instantaneous position of the undisturbed interface coinciding with the plane $x = 0$. Suppose the interface is deformed slightly into a wavelike corrugation of wave number m described by

$$x = \delta(t) \cos(my). \quad (4)$$

The undisturbed part of concentration is governed by

$$u \frac{\partial \langle C \rangle}{\partial x} = D \nabla^2 \langle C \rangle, \quad (5)$$

$$\langle C \rangle = C_s \text{ for } x = 0, \quad (6)$$

$$u \langle C \rangle - D \frac{\partial \langle C \rangle}{\partial x} = C_a V \text{ for } x = 0. \quad (7)$$

The solution of the unperturbed part is given by

$$\langle C \rangle = (C_s + C_a V/u) e^{(ux/D)} - (C_a V/u). \quad (8)$$

The disturbed part of concentration is governed by

$$u \frac{\partial \tilde{C}}{\partial x} = D \nabla^2 \tilde{C}, \quad (5')$$

$$\tilde{C} \rightarrow 0 \text{ for } x \rightarrow \infty. \quad (6')$$

The solution of the perturbed part is given by

$$\tilde{C} = -A (\delta/a) \cos(my) e^{-ax}, \quad (9)$$

where $a = \{-u/D + [(u/D)^2 + 4m^2]^{1/2}\}/2$.

The unknown constant A is given by the mass conservation law on the interface

$$A = C_a / D (1 + u/aD)^{-1}.$$

Adding Eq. (8) to Eq. (9) and taking into account to the

first order of δ , we set $C = C_s$ at the interface. We obtain the governing equation of the amplitude of the deformed interface:

$$\delta/\delta = (a + u/D)(C_s u/C_a + V). \quad (10)$$

In the limit of $u = 0$, Eq. (10) reduces to the Saffman-Taylor instability.^{34,35}

The term $a + u/D$ is always positive, independent of the sign of the drift u . If $C_s u/C_a + V > 0$, the interface becomes unstable. On the other hand, when $C_s u/C_a + V < 0$ the interface is stable. The negative drift represents the drift toward the interface, i.e., the attractive drift. When the strength $|u|$ of the negative drift becomes larger than $C_a V/C_s$, the interface becomes stable. Then the dense aggregate will appear. Figure 4 shows the hydrodynamic instability. The positive V represents the growth process. The hydrodynamic instability appears in the growth process when the strength of the attractive drift toward the interface becomes less than $C_a V/C_s$. The result is similar to the Saffman-Taylor instability for the viscous fingering under a gravitational field.^{34,35} The similarity is discussed in Sec. IV.

III. SIMULATION RESULTS

We perform the simulation of the pattern formation in the diffusion-limited aggregation with drift. We show that the structural phase transition occurs with changing direction of drift in the growth process. The structural phase transition was already found in the aggregation process forming from the single seed with the radial drift. Here we simulate the pattern formation on the flat plate (see Fig. 1) to investigate the connection with the hydrodynamic instability. We can simulate the growth process in the limit of $V = 0$ by using a biased random walker. We have carried out simulations for a range of P ($= u/D$) values on square arrays of size 200×200 . We study the pattern formation in the DLAD by the use of a biased random walker. We start out with the occupied flat plate on the bottom side. Particles are introduced one at a time at a randomly chosen point on the top side (see Fig. 2). We allow particles to follow biased random

FIG. 2. The model used to simulate the effect of the variable drift on the diffusion-limited aggregation. A single particle is introduced at a time at a randomly chosen point on the upper boundary and performs a vertically biased random walk with a probability P . The positive P indicates the upward drift, and the negative drift the downward drift.

FIG. 3. Typical patterns grown with use of the model shown in Fig. 2. (a) Aggregate for $P = -0.1$. (b) Aggregate for $P = -0.05$. (c) DLA fractal for $P = 0.0$. (d) Aggregate for $P = 0.002$. (e) Aggregate for $P = 0.008$. At a negative drift, the crossover occurs from the DLA fractal at small length scales to the dense structure at long length scales. At a positive drift, the crossover occurs from the DLA fractal at small length scales to the one-dimensional structure at long length scales.

walks vertically to the plate. For a positive drift, particles perform a biased Brownian motion with probability P (>0) away from the plate. For a negative drift, particles perform a biased random walk with probability $-P$ ($P < 0$) toward the plate. The boundary condition in the horizontal direction is periodic in order to simulate an infinite half-plane. A particle continues to move until it either reaches a point adjacent to a site already occupied by a particle, or until it reaches the outer boundary. Figure 2 shows a schematic representation of the model used to simulate the effects of the particle drift on the diffusion-limited aggregation. We have carried out calculations for a range of P values ($-1 < P < 0.01$). Figure 3 shows results obtained for a few patterns using the procedures outlined above. The clusters are grown until the height reaches 100 units. The pattern shown in Fig. 3(c) was obtained with no drift. The cluster is the DLA fractal. Figures 3(a) and 3(b) show the patterns obtained for negative drifts $P = -0.1$ and -0.05 . We show the patterns obtained for positive drifts $P = 0.002$ and 0.008 in Figs. 3(d) and 3(e). As the negative drift is lower, the overall shape of the pattern becomes closer to a dense pattern. On the other hand, even a very small value of the positive drift becomes effective for the DLA fractal. The overall shape of the pattern becomes increasingly needlelike with an increase of the positive drift. For the negative drift, the crossover is found from the DLA fractal on short length scales to the dense structure on long length scales. On the other hand, for the positive drift, the crossover is found from the DLA fractal on short length scales to the needle structure on long length scales.

When the drift changes from a negative value to a positive value, the structure of the aggregate varies from a dense structure, through the DLA fractal, to a needlelike structure. It is found that the phase transition can occur with the change of the drift direction in the growth process. Thus we find the distinct morphologies in the different regions of the two parameters u and V . Figure 4 shows the morphology diagram describing the distinct

FIG. 4. Morphology diagram describing the distinct patterns. The vertical and horizontal axes indicate the velocity V of the interface and the drift u . The solid line indicates the hydrodynamic instability in the growth process. When the strength of the downward drift is larger than the critical value, the interface of the aggregate becomes stable and the dense aggregate appears. When the hydrodynamic instability occurs, the morphological transitions occur. The three distinct patterns, the dense aggregate, the DLA fractal, and the needle structure, appear.

patterns. The positive value of V represents the growth process. The line given by $V = -C_s u / C_a$ indicates the marginal line of the hydrodynamic instability. In the region given by $0 < V < -C_s u / C_a$, the dense aggregate occurs. On the point given by $u = 0$ and $V = 0$, the DLA fractal appears. The point represents a critical point of the structural phase transition. In the region given by $V > -C_s u / C_a$, the needlelike structure appears. The structural phase transition is consistent with the hydrodynamic instability.

IV. VISCOUS FINGERING UNDER A GRAVITATIONAL FIELD

We consider viscous fingering under a gravitational field^{34,35} for comparison with the diffusion-limited aggregation with drift. We discuss the similarity of the DLAD to the viscous fingering under gravity. Saffman and Taylor³⁴ and Chouke, van Meurs, and van der Poel³⁵ have studied the stability of the flat liquid-liquid interface under gravity. They have found that a hydrodynamic instability could occur when two superposed viscous fluids were forced by gravity and an imposed pressure gradient through a porous medium. If the steady state was one of uniform motion with velocity V vertically upwards, and the interface between the two fluids was horizontal, then it could be shown that the interface was stable for small deviations if

$$(\mu_2/k_2 - \mu_1/k_1)V + (\rho_2 - \rho_1)g > 0,$$

and unstable if

$$(\mu_2/k_2 - \mu_1/k_1)V + (\rho_2 - \rho_1)g < 0,$$

where the subscript 1 refers to the upper fluid and the subscript 2 to the lower. In the limit of $\mu_2 = 0$, the condition for stabilizing the interface is given by

$$(k_1/\mu_1)(\rho_1 - \rho_2)g + V < 0. \quad (11)$$

When the stabilizing force by gravity exceeds the desta-

bilizing force by the imposed pressure gradient, the interface becomes stable. On the other hand, the stability condition for the interface in the diffusion-limited aggregation with drift is given by

$$(C_s/C_a)u + V < 0. \quad (12)$$

When the strength of the attractive drift becomes larger than $C_a V / C_s$, the interface becomes stable. By comparing the relationship (12) with (11), it is found that the hydrodynamic instability in the diffusion-limited aggregation with drift is similar to the Saffman-Taylor instability in the viscous fingering under a gravitational field.

V. SUMMARY

We investigate the hydrodynamic instability in diffusion-limited aggregation with drift. The question of whether or not the hydrodynamic instability is related to the structural phase transition in the growing aggregate is studied. We apply the Saffman-Taylor approach to the diffusive systems with an imposed drift. The hydrodynamic instability is found to occur when the strength of the attractive drift is less than the critical value. The simulation is performed for pattern formations in the limit of the growing rate $V = 0$. The structural phase transition is found from dense aggregation, through diffusion-limited aggregation, to the needle pattern, through change in the direction of the drift. It is shown that the transition line of the structural phase transition is consistent with the marginal line of the hydrodynamic instability. We find that the hydrodynamic instability is similar to the viscous fingering under a gravitational field.

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