

Electric-field-induced diffusion-limited aggregation

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Electric-field-induced diffusion-limited aggregation (EDLA) fractals formed by the suspended metal microspheres in an oil are observed experimentally. The formation of an EDLA fractal is due to the interaction of the dipole-dipole force induced on the metal microspheres when these microspheres are presented in an external electric field. The EDLA fractal grows as the applied field strength is slowly increased. At a fixed volume fraction ϕ , the dimensions D_f of all fractals formed under the different space fields are in the range of 1.72 ± 0.1 . We find that the fractal dimension D_f changes as the volume fraction ϕ of metal particles varies and satisfies the relationship of $D_f \sim \phi^m$, where m is approximately 1.52. [S1063-651X(97)51103-2]

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Problems of diffusion-limited aggregation (DLA) have been the source of much interest in the past decade [1–4]. Fractal growth based on the mechanism of diffusion-limited aggregation involves the random walk of particles and their subsequent sticking. This mechanism has been tested experimentally in various fields, such as crystal growth, electrochemical deposition, viscous fingering, and dielectric breakdown. All these phenomena are considered to be governed by the same growth mechanism [5–8]. A more interesting model that generates aggregation of particles in clusters through Brownian motion was introduced in 1981 by Witten and Sander [9]. It is well known that diffusion-limited aggregation provides a basis for understanding a large range of natural pattern formation phenomena.

In this paper, we report another kind of fractal pattern produced by electric-field-induced diffusion-limited aggregation (EDLA). The EDLA fractal can be observed in a system made up of a mixture of solid and liquid where the solid phase must be a conducting particle or one with high dc conductivity. In recent electrorheological (ER) experiments performed with conducting microspheres we found some interesting phenomena, for example, that conducting particles such as indium microspheres suspended in silicon oil form a netlike structure if an external high electric field is applied to parallel electrodes [10]. Very recently we observed that some fractal patterns formed by conducting microspheres appeared in the different forms of space field if an applied electric field was increased slowly. After comparing these patterns with

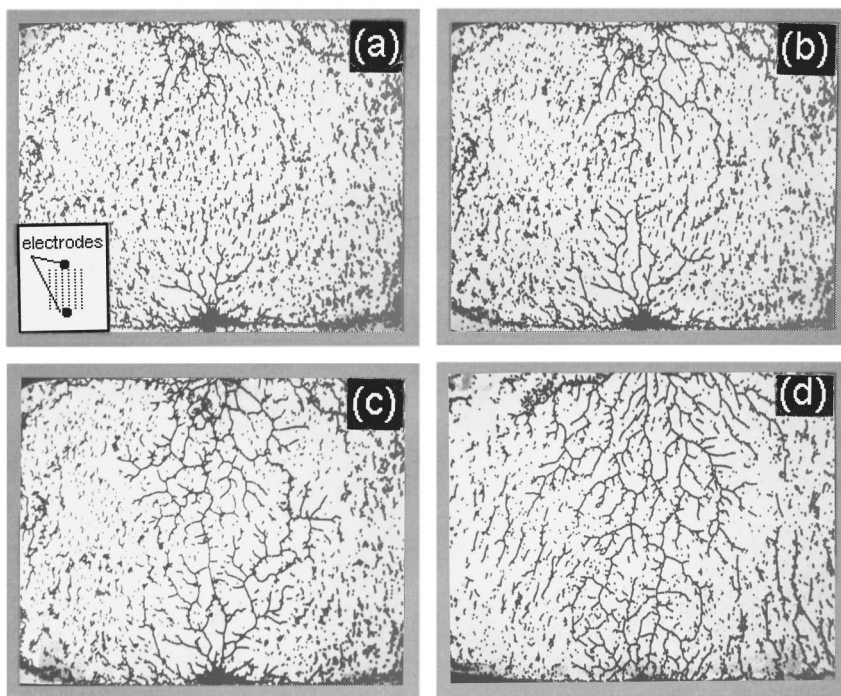


FIG. 1. Development of a fractal formed by metal microspheres as the electric field is increased, where the electric field strengths are (a) 150 V/mm, (b) 250 V/mm, (c) 350 V/mm, and (d) 500 V/mm, respectively.

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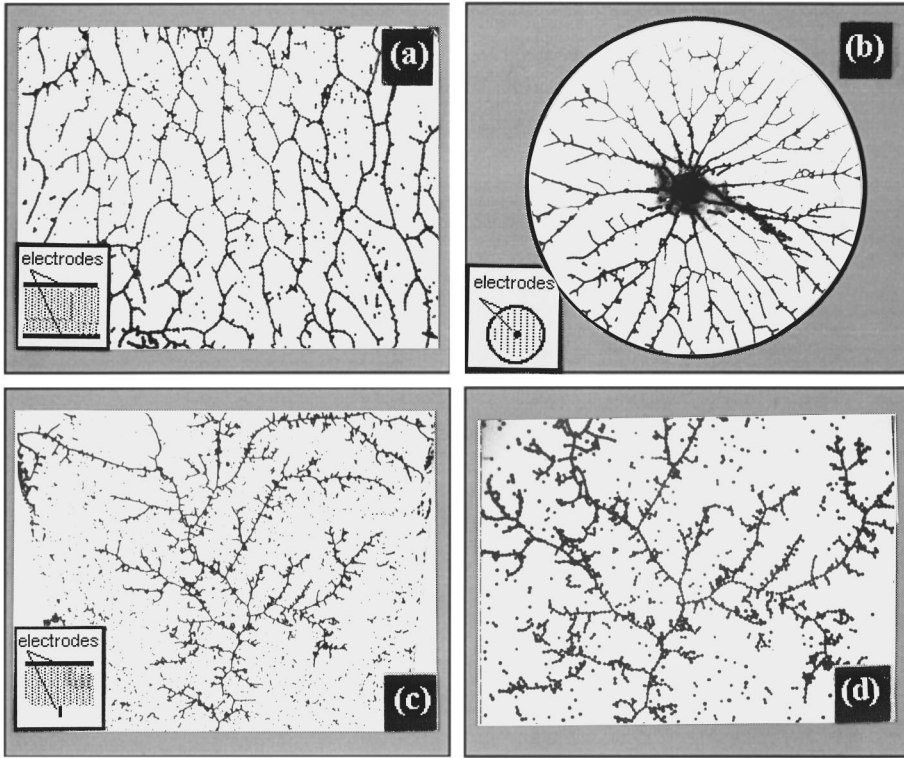


FIG. 2. (a), (b), and (c) show the fractals under different kinds of space field. Insets show the forms of applied electric field. (d) is an enlarged image of part (c).

those described above we found that it is necessary to classify them as another kind of diffusion-limited aggregation induced by high electric field. However, the difference in the physical mechanism between EDLA and those described above is that the former is caused by an electrodynamic interaction and the latter is the effect of Brownian motion.

It is well known that an external electric field E induces a dipole moment on the particle, which is given by

$$p = 4\pi\epsilon_0 a^3 \epsilon_f \beta E, \quad (1)$$

where a is the radius of the sphere and β is a mismatch factor, which is expressed as

$$\beta = \frac{\epsilon_p - \epsilon_f}{\epsilon_p + 2\epsilon_f}, \quad (2)$$

where ϵ is the complex dielectric constant, with the subscripts p and f indicating the particle and fluid, respectively. These dipole moments interact with a force given by

$$f = \frac{6p^2}{4\pi\epsilon_0 \epsilon_f d^4}. \quad (3)$$

It is thought that the force between dipoles will lead to the particles being attracted to each other to create a chainlike structure, and if the electric field strength is increased further some columns suspended across two parallel electrodes can be observed [11–13]. Although these phenomena have been observed in many kinds of ER fluids containing insulating particles, few experiments have been conducted with conducting particles. Thus, the actual pattern formed by conducting particles is unknown to date.

The metal microspheres used in our experiments were made from purified copper grade (Fisher Scientific). At first,

copper particles of about $65 \pm 5 \mu\text{m}$ were selected. Then they were fabricated into the microspheres using a high-temperature spouting device. [14]. It should be pointed out that the surfaces of 20% of the copper microspheres made by this method were oxidized. The surface treatment was performed with dilute HCl and the microsphere was then baked in an oven at a temperature of 60°C . The prepared copper microspheres were mixed with silicon oil with a volume

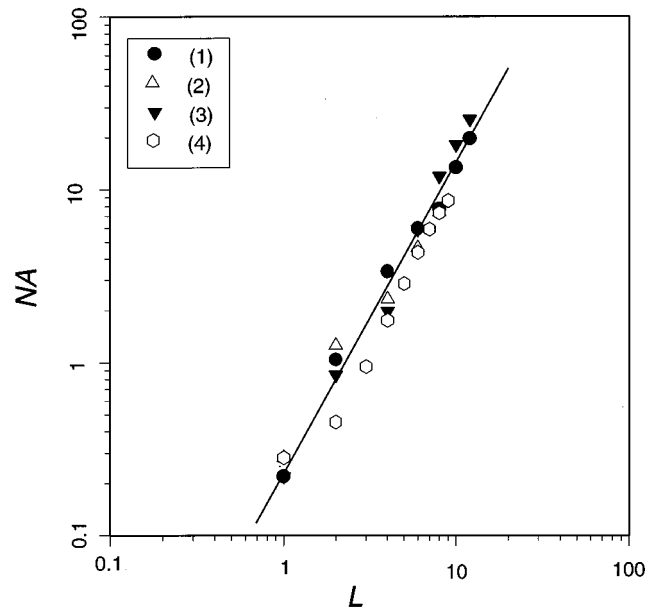


FIG. 3. Schematic log-log plot of NA and L , where symbols (1), (2), (3), (4) represent the data evaluated from Figs. 1(d) and 2(a)–2(c), respectively. The solid line is the dependence of NA on the L obtained from Fig. 2(c).

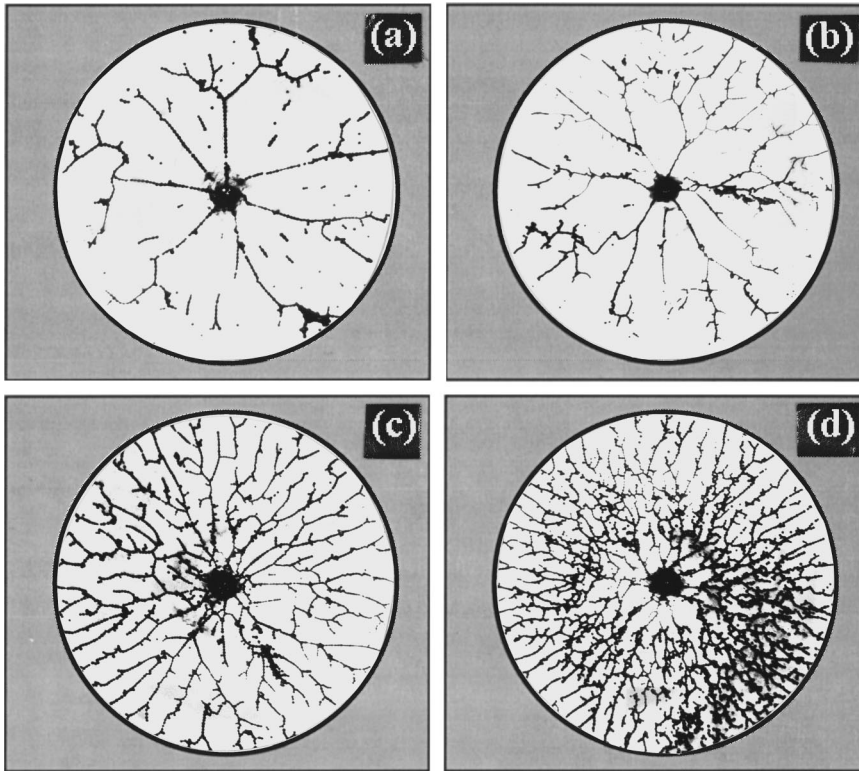


FIG. 4. The changes of fractals when the volume fraction is varied. The volume fractions for (a), (b), (c), and (d) are 0.008, 0.022, 0.102, and 0.163, respectively.

fraction of about 0.082. The experimental setups used in this investigation are shown in the insets of Figs. 1(a) and 2(a)–2(c). The pattern formations were observed *in situ* with a light microscope.

The growth process of two-dimensional EDLA patterns is shown in Fig. 1, where the metal spheres suspended randomly in oil are presented in the field between a two-pin electrode. When an electric field is applied to the electrodes, the nearest one to the pin is first attracted and subsequently sticks to the electrode to form a seed. The new particle undergoes an attractive force until it comes within a fixed “sticking distance” of the seed. At that point, the particle sticks to the seed, and a new particle is attracted in a fixed “sticking distance,” and the process continues to form a short cluster. The “sticking distance” here is decided mainly by the strength of the applied electric field. According to Eq. (3), the force between particles is related to their distance if the electric field E is fixed; thus the growth process stops once the attractive force between the cluster and its neighboring particles exceeds its acting range. Figure 1(a) illustrates the relationship where two short fractals begin to grow separately from two electrodes as the external field is increased slowly but then stabilizes when the electric field is constant. If the electric field strength is increased continuously the fractals begin to grow again until they reach new stable states as shown in Figs. 1(b) and 1(c). Figure 1(d) shows the final state before the fractal breaks down. At this critical point, an electric short would occur if the applied field strength was increased further. This is because passing a large current through a *conducting fractal* leads to electric breakdown. It should be pointed out here that, from observations made with the microscope, we found that the growth rate of two fractals increases rapidly just before contact. This is because the strength of the electric field increases when

two separated *conducting fractals* are close to contact, even though the external electric field is fixed.

Figures 2(a)–2(c) represent the fractal patterns obtained under different space fields. Figure 2(a) shows the situation when the particles are distributed in a parallel field, where more than two fractals begin to grow from two electrodes. This result suggests that the presence of the particle dc conductivity essentially modifies the electrodes from planar to irregular shapes, with sharp corners (single particle) at the points where the electric field is greatest and the force acting on the near particles is largest as well. The situation then becomes similar to that of diffusion-limited aggregation, and the formation of a fractal-like pattern is, therefore, not surprising. A part of the fractal reproduced in Fig. 2(c) is en-

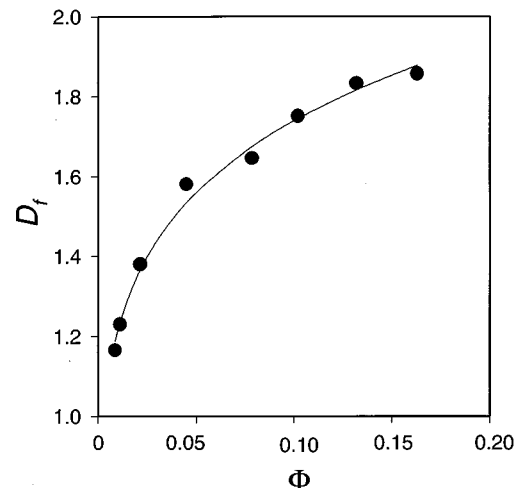


FIG. 5. Dependence of the fractal dimensions on the volume fractions.

larged and reshown in Fig. 2(d) where the branch of a fractal formed by metal microspheres can be seen clearly. In addition, it is found that the fractal is very stable once the microspheres have adhered to it.

The fractal dimension D_f was obtained using the equation $NA = L^{D_f}$ or $D_f = \ln(NA)/\ln L$, where A is the cross-sectional area occupied by each microsphere and N is the number of microspheres in a measured square box with a length L . By varying the length L while counting the enclosed metal microspheres (untouched spheres to the fractal were not included), we obtained the D_f from the slope of $\ln(NA) \sim \ln L$. The results are shown in Fig. 3 for the fractals in Fig. 1(d) and Figs. 2(a)–2(c), which started all from the same volume fraction (0.082) of metal microsphere with different electrode configurations. Surprisingly, the fractal dimensions are all in the range of 1.72 ± 0.1 . This clearly demonstrates the

fact that the fractal dimension only depends on the volume fraction of metal microspheres in the fluid and not on the geometry of the electrodes used.

Figure 4 shows the variation of the pattern formation of metal microspheres in the fluid as a function of the volume fraction. It can be clearly seen from this figure that the lower the volume fraction the fewer the number of fractal branches, and thus the smaller the fractal dimension. The dependence of the fractal dimension D_f on the volume fraction ϕ is plotted in Fig. 5, which varies from about 1.2 at low concentration to 1.8 at a very high concentration of solid phase. By curve fitting, a power-law dependence is found between the D_f and ϕ : $D_f \sim \phi^m$ with the exponent m approximately equal to 0.15 in our experimental system.

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