Simulating fractal pattern formation in metal-oil electrorheological fluids

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Inhomogeneous electric field induced aggregation of metal microspheres suspended in an insulating oil is studied by means of computer simulations. In the present model the metal particles are subjected solely to the forces exerted by the inhomogeneous external electric field and by the fluid; the interparticle interaction and stochastic forces are ignored. The metal clusters grown in the simulation are found to be fractals. At low concentration there is quantitative agreement between the simulations and the experiments of Wen and Lu [Phys. Rev. E **55**, R2100 (1997)]. The fractal dimension of the aggregates has power law dependence on the concentration, and the time evolution of the growth process also shows qualitative behavior similar to that in the experiments. [S1063-651X(98)06703-8]

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I. INTRODUCTION

Fractal growth phenomena have attracted considerable recent scientific interest. These phenomena cover various fields of physics such as crystal growth, electrochemical deposition, viscous fingering, and dielectric breakdown [1]. All these phenomena are considered to be governed by the same growth mechanism that is supported by the fact that stochastic models such as the diffusion limited aggregation (DLA) and the dielectric breakdown model (DBM) give an adequate description of most of the observed features of these processes [1,2].

Very recently a type of fractal pattern formation induced by high electric field in an electrorheological (ER) fluid was observed experimentally by Wen and Lu [3,4]. In their electrorheological experiment, conducting microspheres were suspended in insulating silicon oil. When applying a high external electric field various kinds of patterns formed by the aggregating particles have been observed depending on the actual geometry of the electrodes. For instance, in the case of a homogeneous external field (i.e., between parallel planar electrodes) the suspended metal particles stick to each other, forming a netlike structure [3,4]. Applying an inhomogeneous external field the aggregation of the particles resulted in fractal structures with treelike branching morphology [4]. The authors called this aggregation process electric field induced diffusion limited aggregation (EDLA) in order to distinguish it from the other types of aggregation processes mentioned above.

The most important experimental findings of EDLA processes can be summarized as follows.

(1) The aggregate is a fractal independently of the electrode configuration.

(2) The fractal dimension D of the aggregate depends on the volume fraction ϕ of the microspheres in the solid-liquid mixture and this dependence can be well fitted by a power law, i.e., $D \sim \phi^{\gamma}$, where $\gamma = 0.15$.

(3) At a given volume fraction ϕ the fractal dimension D is independent of the electrode configuration.

(4) The time evolution of the EDLA process also shows interesting features. At a fixed value of the external field $\vec{E}$ the aggregation process slows down considerably after some time and to maintain the aggregation it is necessary to increase slowly the external field strength. The process lasts until the growing cluster connects the two electrodes, giving rise to a short circuit (electric breakdown). It was found that the growth rate of the fractal aggregate increases very rapidly just before the contact.

In this paper we present a two-dimensional dynamical simulation of the aggregation processes induced by an inhomogeneous electric field in metal-oil ER fluids. Our model is based on the assumption that in this type of aggregation the long range attractive forces acting on the metal particles play the primary role for the structure formation instead of the stochastic Brownian motion of the particles. For simplicity, our study is restricted to the case of the inhomogeneous external electric field when, at low volume fraction, the direct particle-particle interaction is negligible compared to the forces exerted by the electrodes. The model is applied to study the geometrical structure of the aggregates focusing on the concentration dependence of the fractal dimension. Our dynamical model also makes it possible to get some insight into the time evolution of the aggregation process.

II. THE MODEL

To work out a model of EDLA processes, first the mechanism leading to the aggregation of the metal particles has to be analyzed. For our investigations we chose the cylindrically symmetric electrode configuration which is the most studied experimental situation of Ref. [4] (see Fig. 4 of Ref. [4]).

During the whole process the electrodes are kept at fixed potential values. The space between the electrodes is filled with an insulating oil in which the metal particles (or particles with high dc conductivity) are suspended. When this ER fluid is exposed to an external electric field $\vec{E}$ the suspended particles obtain an induced dipole moment $\vec{d}$, which

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is proportional to the local effective electric field $\vec{E}^{\rm eff}$:

$$\vec{d}=\alpha\vec{E}^{\rm eff} \quad \text{and} \quad \vec{E}^{\rm eff}=\vec{E}+\vec{E}^{d}, \tag{1}$$

where α is a constant and $\vec{E}^{d}$ denotes the induced dipole field. α is determined by the size of the particles and by the dielectric constants of the particles and the fluid. The induced dipoles interact with the external field and with each other as well, which makes the exact study of the system very complicated. The force $\vec{F}^{\rm el}$ exerted by the external electric field on a dipole $\vec{d}$ is given by

$$\vec{F}^{\rm el}=(\vec{d},\text{grad})\vec{E} \tag{2}$$

or in components

$$F_i^{\rm el}=(\vec{d},\text{grad}\ \ E_i), \tag{3}$$

where $i=x,y,z$ denotes the vector components. Ignoring that the presence of dipoles modifies the external field, it follows that in a homogeneous external field $\vec{F}^{\rm el}=0$. Hence, in this case the dipole-dipole interaction dominates the dynamics of the system. It was revealed in several ER experiments that the dipole-dipole interaction favors the formation of chains of particles along the direction of the external field for insulating particles [5,6] and network of chains with fractal structure for conducting particles [4]. The chain formation of insulating particles has also been reproduced by Monte Carlo [7,8] and by molecular dynamics simulations [9,10].

Following the arguments of Ref. [4], in the experimental situation considered, under the influence of $\vec{F}^{\rm el}$ given by Eq. (2) the metal particles start to move towards the inner electrode and the nearest particles stick to it, forming a seed. Since the particles are conducting the stuck ones change the originally regular electrode shape to an irregular one that results in a considerable modification of the electric field itself. Due to the subsequent sticking of the metal particles, the inner electrode develops branches with sharp tips where the electric field strength is the greatest and hence the force acting on the dipoles is the largest. It may also happen that before reaching the growing metal cluster, due to the dipole-dipole interaction a few particles stick to each other creating short chains moving towards the aggregate. The role of this effect becomes important with increasing concentration.

In the simplest approximation, besides affecting the value of α in Eq. (1), the fluid exerts a hydrodynamic force $\vec{F}^{\rm hydr}$ on the moving particles according to Stock's law

$$\vec{F}^{\rm hydr}=-\beta\vec{v}, \tag{4}$$

where β depends on the particle size and on the viscosity of the fluid, and $\vec{v}$ denotes the velocity of the particles.

In order to investigate the fractal pattern formation in ER fluids under inhomogeneous electric fields we constructed a dynamical model, which takes into account the main ingredients of the growth mechanism described above. Since the growth goes on eventually in a plane our model is two dimensional. Our approach is based on the assumption that in an inhomogeneous external field at low enough concentration of the conducting microspheres the interparticle interaction is negligible compared to the force $\vec{F}^{\rm el}$ exerted by the external field.

In the model starting from a random spatial configuration of the metal particles we follow their trajectories until sticking to the aggregate by solving the equations of motion. The particles are subjected solely to the effect of $\vec{F}^{\rm el}$ and $\vec{F}^{\rm hydr}$, the interparticle interaction is not taken into account (neither dipole-dipole nor hard core interaction), and the induced dipole field $\vec{E}^{d}$ is set to zero. These approximations are consistent at low concentration. It also implies that our model is applicable strictly to the case of inhomogeneous external fields but without any restriction for the electrode geometry. The determination of the trajectories of the particles does not contain a stochastic component, randomness is introduced in the model solely through the random initialization of the positions of the particles. The neglect of the interparticle interaction allows us to perform simulations with a large number of particles.

In order to determine the electric field the Laplace equation

$$\Delta U=0 \tag{5}$$

is solved on a square lattice with the boundary condition that the inner electrode is kept at $U=0$ and the outer one at $U=1$ fixed potential values. The discrete form of Eq. (5) on a square lattice can be written as

$$U_{i,j}=\frac{1}{4}(U_{i+1,j}+U_{i-1,j}+U_{i,j+1}+U_{i,j-1}), \tag{6}$$

which is a system of linear equations where the number of equations is equal to the number of lattice sites considered. In our simulation code a relaxation technique is used to solve Eq. (6) [11]. The electric field strength can be obtained from the potential U as

$$\vec{E}=-\,\text{grad}U. \tag{7}$$

Inserting Eq. (7) into Eq. (1) and using Eq. (2) yields the force exerted by the external field of the electrodes on a particle:

$$F_x^{\rm el}=\alpha\left[\frac{\partial U}{\partial x}\frac{\partial^2 U}{\partial x^2}+\frac{\partial U}{\partial y}\frac{\partial^2 U}{\partial y\,\partial x}\right], \tag{8}$$

$$F_y^{\rm el}=\alpha\left[\frac{\partial U}{\partial x}\frac{\partial^2 U}{\partial x\,\partial y}+\frac{\partial U}{\partial y}\frac{\partial^2 U}{\partial y^2}\right]. \tag{9}$$

Equations (9) and (8) give $\vec{F}_{i,j}^{\rm el}$ at the lattice sites i,j used for the calculation of U.

The motion of the particles is determined in the following way: The particles are considered to move in the continuous coordinate space, the lattice is used only to obtain $\vec{F}_{i,j}^{\rm el}$ and to grow the aggregate. The equations of motion of the particle system are

$$m\ddot{\vec{r}}_k=\vec{F}_{i,j}^{\rm el}+\vec{F}^{\rm hydr}, \quad k=1,\ldots,N \tag{10}$$

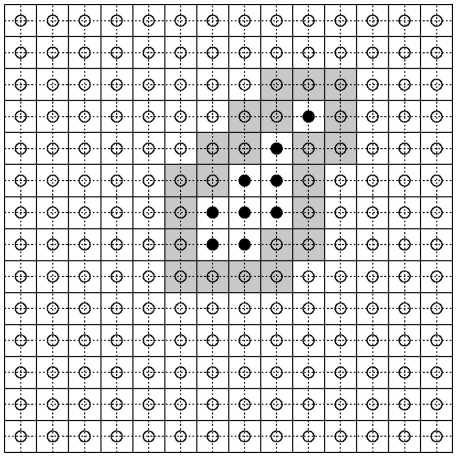

FIG. 1. The lattice structure used in the simulations and the illustration of the growth rule. The open circles denote the empty lattice sites while the filled ones are occupied by the particles of the cluster. The force $\vec{F}^{\rm el}_{i,j}$ is considered to be constant within the plaquette centered around the site i,j (plaquettes bounded by solid lines). The plaquettes belonging to the perimeter of the cluster are shadowed. The example indicates that growth can occur not only in the direction of the four nearest neighbors but also in the direction of the next nearest neighbors of the cluster particles.

where N denotes the total number of particles and $\vec{r}_k$ is the position of the center of the kth particle. In the simulation code Eq. (10) is solved by applying a fifth order predictor-corrector scheme [12]. During the integration of Eq. (10) $\vec{F}^{\rm el}_{i,j}$ is considered to be constant within a plaquette centered around the lattice site i,j.

A moving particle sticks to the growing aggregate when it enters a plaquette belonging to the perimeter of the cluster. Figure 1 gives an explanation of the lattice structure used, and of the growth rule. After each sticking event the potential $U_{i,j}$ of the lattice site occupied by the new stuck particle is set to zero, and the force field $\vec{F}^{\rm el}_{i,j}$ has to be recalculated. This procedure goes on either until there are no suspended particles left (that may be the case at very low concentration) or until the growing cluster reaches the outer electrode.

III. RESULTS

In the framework of the model described above we performed simulations in order to investigate the geometrical structure of the aggregates generated with emphasis on the concentration dependence of the fractal dimension. This dynamical model allows us also to monitor the mass of the growing cluster as a function of time. This way information can be gained about the time evolution of the growth process.

For all the simulations a lattice of size 200×200 sites was used and the total number of particles N was varied in order to change the concentration $\phi=N/N_{\rm plaq}$, where $N_{\rm plaq}$ is the number of plaquettes between the two electrodes. The natural time scale t_0 of the system arises from the force expressions, i.e., $t_0=\beta a/F^{\rm el}_{\rm max}$, where a denotes the lattice unit and $F^{\rm el}_{\rm max}$ is the estimated maximal value of $F^{\rm el}$. The parameter values used in the simulations are $m=1$, $\alpha=1$, $a=1$, $\beta=0.04$.

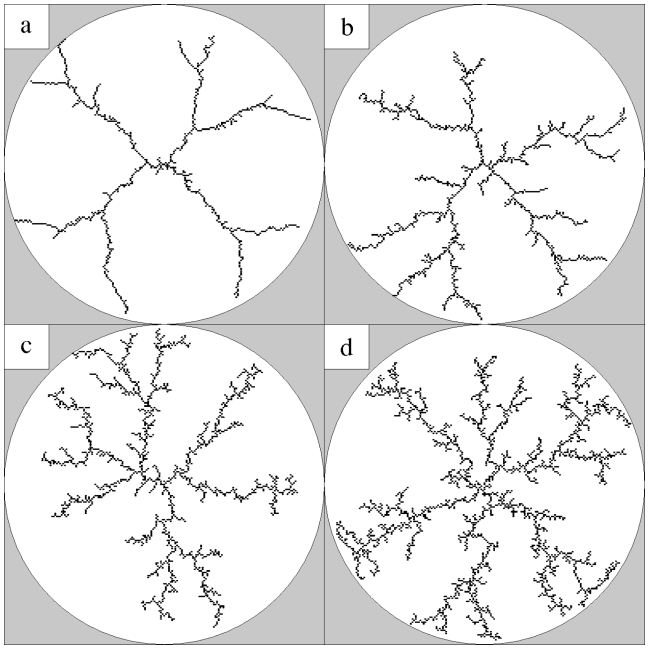

FIG. 2. Representative examples of simulated electric field induced aggregation clusters. The value of N is 1000, 2000, 3500, 5000 for (a), (b), (c), (d), respectively.

Representative examples of clusters generated at different N values are presented in Fig. 2. It can be observed that at lower volume fraction [Figs. 2(a), 2(b)] the metal cluster is composed of a few long branches strongly directed towards the outer electrode, with hardly any longer side branches. It is important to note that when a growing branch is getting closer to the outer electrode the electric field strength $\vec{E}$ is rapidly increasing in the vicinity of the branch tip. When there are no more suspended particles left in front of the tip new particles can reach the branch only from the two sides, which results in splitting of the tips into two side branches. It can also be seen that tip splits may occur serially starting from one branch. The occurrence of tip splits is limited by the geometrical configuration of particles remaining available for the further growth. The comparison of Figs. 2(a) and 2(b) to the experimental results of Ref. [4] [see Figs. 4(a), 4(b) of Ref. [4]] shows that at low concentration qualitative agreement is found between the simulations and the experiments.

By increasing the concentration [see Figs. 2(c), 2(d)], the number of side branches increases and the cluster gets more dense but not as rapidly as was found in the experiments [Figs. 4(c), 4(d) of Ref. [4]]. In the zones lying between the main branches of the aggregate the electric field strength is considerably lowered by screening. Hence, the force acting on the particles between the branches is much smaller than between the cluster tips and the outer electrode. Due to this screening mechanism, the growing cluster can reach the outer electrode, leaving a large number of particles in the fluid that did not have time to become part of the cluster. For clarity, the particles that remained suspended in the fluid are not indicated in Fig. 2.

To give a quantitative description of the geometrical structure of the aggregates, their fractal dimension D was evaluated at several values of the volume fraction ϕ. In order

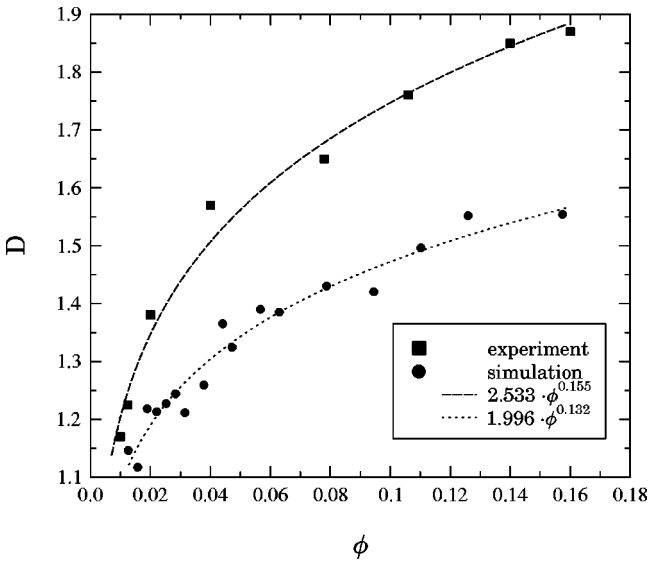

FIG. 3. The fractal dimension D of the aggregate as a function of the volume fraction ϕ. The experimental results are taken from Ref. [4]. The parameter values of the curves fitted to the data points are also given.

to determine the fractal dimension D we counted the number of particles $M(R)$ belonging to the cluster in a circle of radius R centered at the central electrode. At each value of the volume fraction, $M(R)$ was averaged over 15 simulations with different initializations and D was obtained as the slope of $\ln\langle M(R)\rangle$ vs $\ln R$. The results are plotted in Fig. 3 along with the fractal dimension measured experimentally.

As expected, at low concentration there is close agreement between the fractal dimensions obtained experimentally and from the simulations, while for increasing ϕ there is increasing deviation between them. It can be seen in Fig. 3 that similarly to the experimental results, a power law, i.e., $D=B\phi^{\gamma}$, seems to be a reasonable fit also to the simulated data with parameters $B=1.996$ and $\gamma=0.132$ different from the ones extracted experimentally. The value of B falling very close to 2 implies the consistent result that the model would give rise to a space filling cluster only at $\phi=1$.

During the simulations we monitored the cluster mass, i.e., the number of particles M of the cluster as a function of time t. Typical results are presented in Fig. 4 for two different values of ϕ. It can be observed that at low concentration [Fig. 4(a)] the $M(t)$ function is composed of two distinct parts, i.e., the increase of $M(t)$ is much steeper in the later stage of the growth than at the beginning of the process, similarly to the experimental findings. This acceleration of the growth process in the vicinity of the outer electrode is caused by the increase of the electric field strength as the cluster gets closer to the outer electrode. The comparison of Fig. 4(a) to Fig. 4(b) shows that the behavior of $M(t)$ is sensitive to the concentration. At high concentration [Fig. 4(b)] the total amount of time needed for the cluster to reach the outer electrode is much less than at lower concentration [Fig. 4(a)]. The reason is that when the concentration increases the average interparticle distance decreases, which results in decrease of the time elapsed between two subsequent sticking events. Hence, less time is spent for the motion of the particles and this acceleration of the growth is

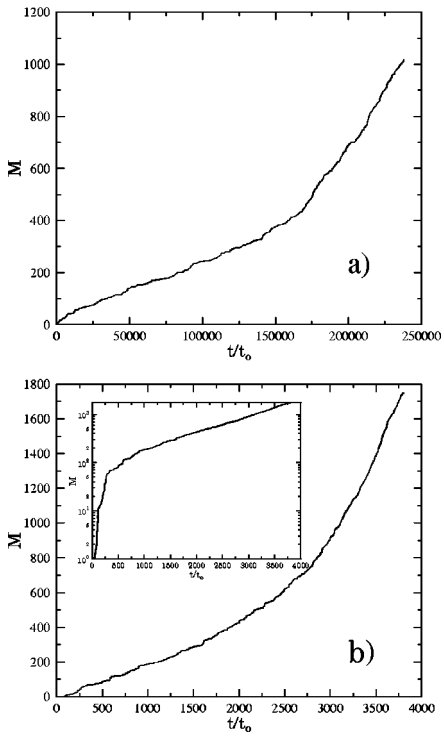

FIG. 4. The number of particles M of the aggregate as a function of time t at volume fractions (a) $\phi=0.04$ and (b) $\phi=0.1$.

amplified further by the increase of the field strength. In Fig. 4(b) the behavior of $M(t)$ seems to be exponential, which is also illustrated in the inset on the semilogarithmic plot.

IV. DISCUSSION

In the present paper we studied the aggregation of metal microspheres induced by an inhomogeneous electric field in an insulating oil. The basic idea of our model is that starting from a random spatial configuration of the metal particles we follow their trajectories until they stick to the aggregate by solving the equations of motion without any stochastic component. For simplicity, the induced dipole field and the interparticle interaction is neglected in the model. It was found that at low concentration the results obtained by means of computer simulations are in good agreement with the experimental observations. Power law dependence of the fractal dimension on the concentration was obtained with an exponent smaller than the experimental one. The time evolution of the growth process is found to be qualitatively similar to the experiments.

The deviation of the simulations and the experiments demonstrates that the interparticle interaction has an important effect on the structure formation at higher concentrations. It results in the faster increase of the fractal dimension and it may also explain the slowing down of the growth at a fixed external field. For a better understanding, neglect of the particle-particle interaction has to be overcome in the future.

In the model calculations the trajectory of the particles is deterministic; randomness is introduced solely by the random initialization of the position of the particles. The results obtained imply that the electric field induced aggregation of metal microspheres in an insulating oil is not a diffusion controlled process in the sense that the Brownian motion of

the particles does not play a crucial role in the structure formation. Instead, the long range attractive forces acting on the metal particles govern the process. In the experiments, by increasing the temperature and decreasing the particle size one may reach a regime where stochastic effects become important.

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[1] T. Vicsek, *Fractal Growth Phenomena* (World Scientific, Singapore, 1989).
[2] L. Niemeyer, L. Pietronero, and H. J. Wiesmann, Phys. Rev. Lett. **52**, 1033 (1984).
[3] W. Wen and K. Lu, Phys. Fluids **8**, 2789 (1996).
[4] W. Wen and K. Lu, Phys. Rev. E **55**, R2100 (1997).
[5] T. C. Halsey, Science **258**, 373 (1992).
[6] R. Tao and J. M. Sun, Phys. Rev. Lett. **67**, 398 (1991).
[7] J. J. Weis, D. Levesque, and G. J. Zarragoicoechea, Phys. Rev. Lett. **69**, 913 (1992).
[8] J. J. Weis and D. Levesque, Phys. Rev. Lett. **71**, 2729 (1993).
[9] D. J. Klingenberg, C. F. Zukosky, and J. C. Hill, J. Appl. Phys. **73**, 4644 (1993).
[10] N. K. Jaggi, J. Stat. Phys. **64**, 1093 (1991).
[11] W. H. Press, S. A. Teukolsky, W. T. Wetterling and B. P. Flannery, *Numerical Recipes in Fortran* (Cambridge University Press, Cambridge, England, 1994).
[12] M. P. Allen and D. J. Tildesley, *Computer Simulation of Liquids* (Clarendon Press, Oxford, 1994).