Monte Carlo simulation of dielectrophoretic particle chain formation

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When an intense radio-frequency field is applied to a suspension of dielectric particles, the suspended phase condenses under the effect of dipolar interactions between particles. We study the interaction energy when losses are present and address the problem of the formation of linear structures, using a Monte Carlo simulation. We find features of a cooperative phenomenon, with a threshold of field intensity. The dependence of the process with concentration, field intensity, and frequency is studied. [S1063-651X(97)04707-1]

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I. INTRODUCTION

The aggregation of particles forming characteristic pearl chains, under the effect of radio-frequency fields is a well known phenomenon and has been investigated in many living and nonliving systems [1–3]. The responsible interaction is the dielectrophoretic force produced by the distortion of the electric field in the vicinity of each individual dielectric particle and it depends on the dielectric characteristics of particle and medium. The general area of dielectrophoresis (DEP) of biological cells is receiving renewed attention due to potential biophysical (characterization of the electrical state of cells and its functional implications) [4–6] and biotechnological applications (alignment, concentration, and manipulation of cells) [7–10]. The subject is also related to other important technical uses in electrorheology and electrooptics [11–13]. While there exist theoretical studies of the basic force experienced by a dielectric particle in a nonuniform field [14,15], as well as elaborated models of diverse bacteria and yeast cells in terms of shelled dielectric spheres or ellipsoids [16,17], the whole process of chain formation including its dependence with frequency, essential in DEP aggregation, has received little attention and only a limited theoretical treatment exits to our knowledge [18]. This is due to the complexity of the phenomenon involving the long-range interaction of many particles. In this paper, we extend the existing theory of chaining to include the presence of dielectric losses and perform a computer simulation that allows us to deal with the complexity of the multiple interactions involved. In the last few years several computer calculations dealing with structural ordering of multiparticle systems interacting via anisotropic van der Waals [19] or dipolar forces [20,21] have been reported. The present simulation shares many of their characteristics, but our spheres are not polar and the chain formation is driven by the polarizing external field only. The study reveals features of importance in the process of dielectrophoretic concentration, such as the dependence of chain length on concentration and the existence and magnitude of a threshold of field intensity. The dynamic characteristics of chaining are somewhat more complicated and would require a time-dependent scheme to be explained.

II. THEORY OF PARTICLE INTERACTION

We consider a two-dimensional system of N spherical particles of radius a in a square of unit side length, representing our sedimented cells. Each particle is polarized by the external field and interacts with this field and other particles. By solving the problem of a dielectric sphere in an external field, when both particle and suspending medium present losses [15], we get that the polarized sphere is equivalent to a dipole with moment

$$\mathbf{p}=\alpha\mathbf{E}, \quad \alpha=4\pi a^3 \mathrm{Re}(\varepsilon_1)\,\frac{\varepsilon_2-\varepsilon_1}{\varepsilon_2+2\varepsilon_1}, \tag{1}$$

where the complex polarizability α is expressed in terms of the permittivities of the particle ε_2 and the external medium ε_1 [Fig. 1(b)], defined as $\varepsilon_{1,2}=\mathrm{Re}(\varepsilon_{1,2})+j\ \mathrm{Im}(\varepsilon_{1,2})$, with $\mathrm{Im}(\varepsilon_{1,2})=-\sigma_{1,2}/\omega$, $\sigma_{1,2}$ being the conductivity and ω the angular frequency. $\mathbf{E}$ is the intensity of the oscillating field.

The average force on the dipole due to a polarizing nonuniform electric field is

$$\overline{\mathbf{F}}=\frac{1}{2}\,\mathrm{Re}(\mathbf{p}\cdot\boldsymbol{\nabla})\mathbf{E}^*=\pi a^3\ \mathrm{Re}(\varepsilon_1)\mathrm{Re}\left(\frac{\varepsilon_2-\varepsilon_1}{\varepsilon_2+2\varepsilon_1}\right)\boldsymbol{\nabla}E^2. \tag{2}$$

The same result has been obtained by Sauer using the integration of the Maxwell stress tensor to the particle surface [14].

We see that, although the system is not conservative, the dielectrophoretic force derives from an average effective potential energy given by

$$\overline{U}=-\tfrac{1}{2}\alpha_{\mathrm{eff}}E_{\mathrm{rms}}^2, \tag{3}$$

with an effective (real) polarizability

$$\alpha_{\mathrm{eff}}=4\pi a^3\ \mathrm{Re}(\varepsilon_1)\mathrm{Re}\left(\frac{\varepsilon_2-\varepsilon_1}{\varepsilon_2+2\varepsilon_1}\right). \tag{4}$$

To model a biological cell, the preceding results are easily generalized to cope with the case of a membrane-covered sphere [Fig. 1(a)]. In this case the particle is shown to be equivalent to a single sphere with an effective permittivity ε_2 given implicitly by [22]

$$\frac{\varepsilon_2-\varepsilon_m}{\varepsilon_2+2\varepsilon_m}=\left(\frac{a}{a+d}\right)^3\frac{\varepsilon_i-\varepsilon_m}{\varepsilon_i+2\varepsilon_m}. \tag{5}$$

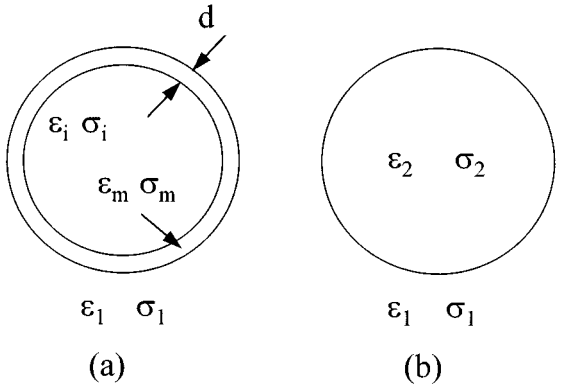

FIG. 1. An electrical model of shelled cell (a) and equivalent spherical dielectric particle (b).

If we now consider the interaction between two polarized identical spheres i and j, near each other, the dipole moment of each of them will be produced by the external field and the field of the other sphere,

$$\mathbf{p}_j = \alpha[\mathbf{E} + \mathbf{E}_i(\mathbf{r}_j)], \tag{6}$$

with

$$\mathbf{E}_i(\mathbf{r}_j) = \frac{1}{4\pi\ \mathrm{Re}(\varepsilon_1)} \frac{3\mathbf{n}(\mathbf{n}\cdot\mathbf{p}_i) - \mathbf{p}_i}{R_{ij}^3}, \tag{7}$$

where $\mathbf{R}_{ij}$ is the position vector between sphere centers and $\mathbf{n} = \mathbf{R}_{ij}/R_{ij}$. We take the dipolar approximation and ignore the contribution from higher multipoles. Due to the symmetry $\mathbf{E}_i(\mathbf{r}_j) = \mathbf{E}_j(\mathbf{r}_i)$, we can solve Eqs. (6) and (7) with $\mathbf{p}_i = \mathbf{p}_j = \mathbf{p}$, and get

$$\mathbf{p} = \frac{\alpha}{1 - 2\alpha F} (\mathbf{n}\cdot\mathbf{E})\mathbf{n} + \frac{\alpha}{1 + \alpha F} (\mathbf{n}\times\mathbf{E})\times\mathbf{n}, \tag{8}$$

$$\mathbf{E}_i = \frac{2\alpha F}{1 - 2\alpha F} (\mathbf{n}\cdot\mathbf{E})\mathbf{n} - \frac{\alpha F}{1 + \alpha F} (\mathbf{n}\times\mathbf{E})\times\mathbf{n} \tag{9}$$

with $F = 1/(4\pi\,\mathrm{Re}(\varepsilon_1) R_{ij}^3)$. The work necessary to bring the sphere i to the proximity of the sphere j will be the sum of two terms: the energy required to take the dipole induced by the external field to the final position inside the field $\mathbf{E}_i$, and the energy to get the additional polarization of the two spheres,

$$\overline{U_{ij}} = -\tfrac{1}{2}\ \mathrm{Re}[\alpha \mathbf{E}\cdot\mathbf{E}_i^*(\mathbf{r}_j)] - \tfrac{1}{2}\ \mathrm{Re}(\alpha|\mathbf{E}_i(\mathbf{r}_j)|^2). \tag{10}$$

The result is

$$\overline{U_{ij}} = \left(\frac{3(1 + 2F^2|\alpha|^2)}{2|1 + \alpha F|^2|1 - 2\alpha F|^2} \sin^2\theta - \frac{1}{1 - 2\,\mathrm{Re}(\alpha)F} + \frac{2\,\mathrm{Re}(\alpha)F}{|1 - 2\alpha F|^2} \right) F|\alpha|^2|E|^2, \tag{11}$$

θ is the angle between vectors $\mathbf{n}$ and $\mathbf{E}$. For both $\mathrm{Re}(\alpha)$ and $\mathrm{Im}(\alpha) \ll 1/F$, we get

$$\overline{U_{ij}} \approx \left(\frac{3}{2} \sin^2\theta - 1 \right) F|\alpha|^2|E|^2. \tag{12}$$

Equation (12) represents the approximation to the interaction energy neglecting the mutual polarization of the spheres. For the closest distance in the chaining direction, $R_{ij} = 2a$, $\theta = 0$, and parameters representative of *Neurospora crassa* cells [23] in a slightly conductive aqueous solution (see results) at a frequency $f = 10$ kHz, the error in the approximation has a value of about 0.8%.

III. MONTE CARLO SIMULATION

A. Numerical model

The system of N spheres will evolve to the minimal energy configuration. Neglecting the charge separation induced by mutual interactions, we can write the total energy in the form

$$\overline{U} = \overline{U_0} + \sum_{i,j>i} \overline{U_{ij}} \tag{13}$$

where $\overline{U_0} = -(1/2)\alpha_{\mathrm{eff}} E_{\mathrm{rms}}^2 N$ is the energy of the induced dipoles in the external field, and

$$\overline{U_{ij}} = 4\pi a^6 \mathrm{Re}(\varepsilon_1) \left| \frac{\varepsilon_2 - \varepsilon_1}{\varepsilon_2 + 2\varepsilon_1} \right|^2 \frac{1 - 3\cos^2\theta_{ij}}{R_{ij}^3} E_{\mathrm{rms}}^2. \tag{14}$$

The attractive van der Waals forces, important in colloidal stability, are not of relevance here and are not considered. The short distance repulsive force is introduced only as a hard sphere condition. In what follows we will assume that the only existing dispersion is that due to the Maxwell-Wagner effects and not to dielectric relaxations. The calculations could be easily extended to include these as well.

Our simulation of the chaining process is based on the classical Metropolis algorithm [24] that has proved to be valuable in the numerical study of similar problems such as the structure of magnetic composites in an external magnetic field [25–27], the ferroelectric ordering of dipolar systems [20,21], or the molecular organization of polymer-dispersed liquid crystal droplets [28]. Here we concentrate on spatial ordering and do not study electrical or thermodynamic observables. The procedure consists of the following steps: (i) we establish an initial random distribution of particles on a two-dimensional square region with adequate concentration. A periodic boundary condition is then imposed in the x and y directions; (ii) individual spheres are moved at random in space; (iii) the energy of each particle U_i is computed according to Eq. (14), and (iv) the new configuration is allowed, either the particle energy is lower than the initial one or the exponential of minus the difference of these energies relative to kT is greater than a random number between zero and one; otherwise the move is rejected and the process repeated with other particle. The whole procedure is performed again for all the particles until self-consistence in the total energy is reached.

B. Results

We used geometrical and electrical parameters representative of a *Neurospora crassa* suspension which has been well characterized [23] ($\varepsilon_1 = 80\varepsilon_0$, $\sigma_1 = 5\times10^{-3}$ S m^{-1},

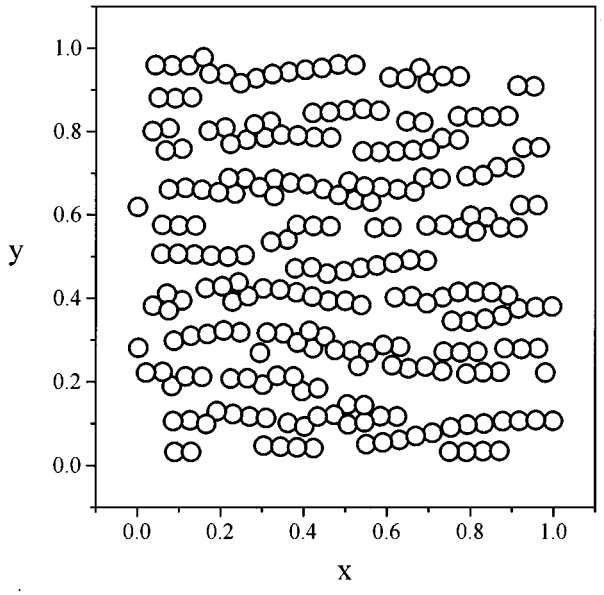

FIG. 2. Simulated final two-dimensional configuration of cells after application of an electric field in x direction (surface fraction $=0.3$, $f=1$ MHz, $E=10^4$ V m^{-1}).

$\varepsilon_i=50\varepsilon_0$, $\sigma_i=0.2$ S m^{-1}, $\varepsilon_m=5\varepsilon_0$, $\sigma_m\approx 0$, $a=10$ μm, $d=10$ nm) and simulated an initially random collection of spheres at different concentrations.

The maximum number of particles included was 238, distributed in a square cell. Several runs were performed with 952 spheres to check that the sample size did not affect significantly our results. In all computations we employed periodic boundary conditions. The long-range dipolar interactions were taken into account by summing the series in the reciprocal space. Using the Poisson summation formula [29] we obtain that the pair potential due to one dipole and all its images can be expressed as

$$\sum \frac{1-3\cos^2\theta_{ij}}{R_{ij}^3}=\frac{2}{w}\sum_n\sum_m e^{j[(2m\pi)/w]x}\left(\frac{2m\pi}{w}\right)^2 \times K_0\left(\left|\frac{2m\pi}{w}\right|(nh-y)\right), \qquad (15)$$

where x,y are relative coordinates of the two dipoles; w,h are the dimensions of the unit cell, and K_0 the modified Bessel function of the second kind. The right-hand side series is rapidly convergent. Each run consisted of 10 000 moves per particle. We checked in a few cases that the results were practically the same when runs extended up to 100 000 moves. Figure 2 shows a final configuration of particles after application of a uniform field, showing the typical chaining appearance of a dielectrophoresis experiment.

With the purpose of quantifying the ordering induced by the external field, we define *connectivity* as the average number of cells contacting a given one. We consider that two cells are connected when the distance between closest points is less than $0.1a$, a being the sphere radius. Figure 3 shows the obtained dependence of connectivity with the occupied surface fraction, the increase being approximately linear for significant concentrations. The ordering field effect for a

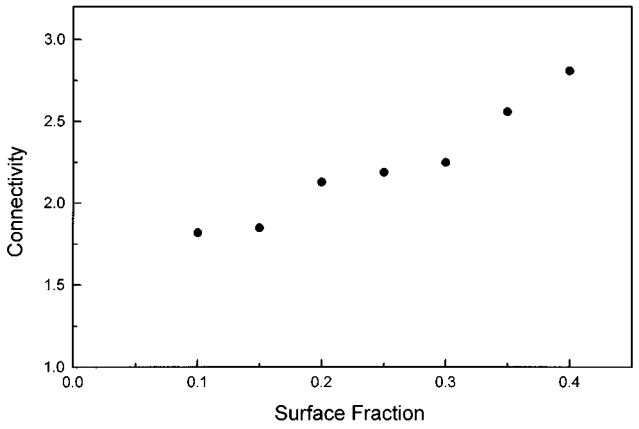

FIG. 3. Influence of cell concentration, represented as surface fraction, on connectivity. ($E=10^4$ V m^{-1}, $f=1$ MHz).

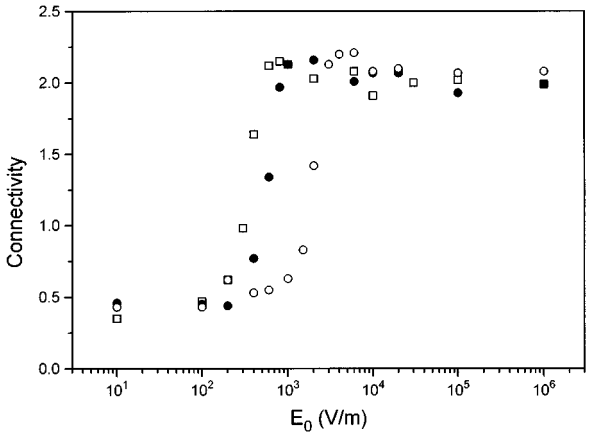

FIG. 4. Dependence of connectivity on field intensity for different frequencies: $f=10^5$ Hz (□), $f=10^2$ Hz (●), and $f=10^9$ Hz (○); surface fraction$=0.3$. The curves show a threshold of field intensity which varies with frequency, for the onset of chaining.

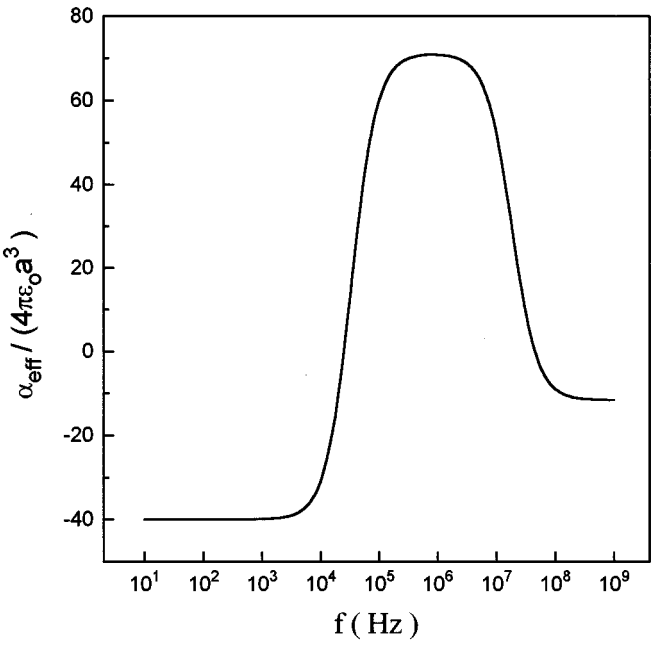

FIG. 5. Theoretical frequency dependence of effective polarizability.

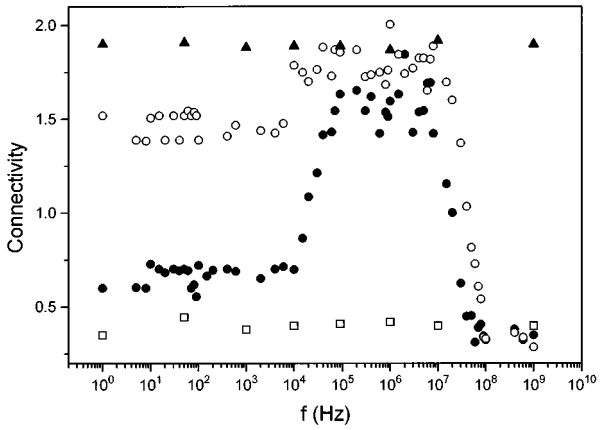

FIG. 6. Computed frequency dependence of connectivity at different field intensities: $E_0=10^2$ V m^{-1} (□), $E_0=5\times10^2$ V m^{-1} (●), $E_0=8\times10^2$ V m^{-1} (○), and $E_0=10^4$ V m^{-1} (▲); surface fraction $=0.3$.

given concentration is strongly dependent on the field intensity. In Fig. 4. we represent connectivity versus field intensity. The curves show the existence of a threshold for a given frequency. We can estimate the theoretically expected critical field by comparing the average thermal translational energy with the interaction energy between spheres Eq. (14). In this equation we take R_{ij} equal to the average distance between closest particles s, and $\overline{\cos^2\theta_{ij}}=1/2$. We get

$$2\pi\frac{a^6}{s^3}\mathrm{Re}(\varepsilon_1)\left|\frac{\varepsilon_2-\varepsilon_1}{\varepsilon_2+2\varepsilon_1}\right|^2 E_{\mathrm{crit}}^2\approx kT, \tag{16}$$

$$E_{\mathrm{crit}}\approx\left(\frac{kTs^3}{2\pi a^6\mathrm{Re}(\varepsilon_1)}\right)^{1/2}\left|\frac{\varepsilon_2+2\varepsilon_1}{\varepsilon_2-\varepsilon_1}\right|. \tag{17}$$

For *Neurospora* data, at $f=10^2$, 10^5, and 10^9 Hz and cell concentration of 0.3, we get $E_{\mathrm{crit}}\approx240$, 140, and 800 V m^{-1}, which agrees reasonably well with the results of our simulation (Fig. 4). A third variable influencing the behavior of the suspension is the frequency. At low frequency the behavior is dominated by medium and particle conductivities, while in the limit of high frequency the real part of permittivity is preponderant. Between both extremes, the dipolar interaction varies due to Maxwell-Wagner relaxations at the interfaces producing a complicated response. We can see the effective polarizability α_{eff}, given by Eq. (4), as a function of frequency, shown in Fig. 5. As a consequence, the process of chain formation depends on the frequency of the alternating field. In Fig. 6 we represent connectivity versus frequency for different field intensities, at a given concentration. The experimental spectra of dielectrophoresis yield, measured as an average length of chains have been used to obtain information about the electrical characteristics of cells [30,31]. We see that both connectivity and effective polarizability follow a similar train with frequency, but only for certain field intensities.

One of the few proposed theoretical models of DEP was that by Pohl and Crane [32,33]; they developed a theory for the dielectrophoretic concentration of cells on electrodes which has been the main reference for this type of studies. Their model however, used electrodes with cylindrical symmetry and assumed that the spheres were collected as individual particles. The experiments show instead, that except at very low concentrations, the bunching starts very quickly after field application and then the chains of variable length drift towards the electrodes contacting other cells or groups along their trajectories. The dielectrophoretic concentration on electrodes is a complex phenomenon and the formation of chains is an essential step in this process. The Monte Carlo simulation provides a correlation between electrical parameters of particles and experimental data of yield or chain length.

IV. CONCLUSIONS

In this work particle aggregation induced by radio-frequency electrical fields has been studied. We have developed a theory for the interaction of lossy dielectric particles immersed in a conductive medium. By incorporating this theory into a Monte Carlo scheme we have found that computer simulations predict the formation of structures that closely correlate with experimental observations.

The model is useful to analyze the influence of diverse parameters, such as the constitutive coefficients of medium and particles and field frequency on the aggregation properties. It predicts the existence of a threshold of field intensity necessary for the chain structure to appear, whose magnitude agrees with the theoretical estimations.

Our work is a step in gaining a more fundamental understanding of the factors involved in the dielectrophoretic aggregation. The approximations used in the model seem reasonable. Two extensions are, however, under study. First, the inclusion in the Hamiltonian of terms produced by higher-order electric moments of the polarized particles. These multipoles could be of importance in the simulation of DEP experiments at very intense field gradients. Secondly, the consideration of dynamic effects, by taking into account the interaction with the suspending medium through a friction coefficient. This will allow us to study time-dependent effects in the particle chaining. Observation and measurement of the chaining characteristics are also at present being carried out in order to make quantitative comparisons between theoretical and experimental results.

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