## ROLE OF HYDRODYNAMIC INTERACTION OF DISPERSED PARTICLES IN STRUCTURIZATION PROCESSES IN ALTERNATING ELECTRIC FIELD

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A study is made concerning the process involved in formation of one-dimensional periodic structures as a result of hydrodynamic interaction of dispersed particles and the dependence of this process on the parameters of the external electric field in which it occurs.

When charged dispersed particles move in a fluid driven by an electric field, such particles interact polarizingly and hydrodynamically. The polarizing interaction is caused by dipole moments which have been induced in the particles. The buildup of these dipole moments is, according to latest theory [1], determined by two competing factors: release of free or bound polarizational charges at the interphase boundary, as a consequence of differences between dispersed particles and dispersing medium with regard to electrical conductivity and dielectric permittivity, and pullout of the double ion layer from its equilibrium state by the external electric field. The hydrodynamic interaction is caused by motion of a particle, which distorts the distribution of fluid velocity around neighboring particles and thus gives rise to additional forces of fluid action on the particles.

According to another study [2], the total energy of dipole-dipole and hydrodynamic interactions for two identical particles oscillating in an ideal fluid due to action of an alternating electric field is

$$\Delta U = \frac{r^6}{2R^3} \left( \pi \rho V_F^2 - 2\varepsilon E_0 \right) \left( 2\cos^2 \varphi - \sin^2 \varphi \right). \tag{1}$$

It has been assumed here that the distance between such particles is much larger than their intrinsic dimensions ( $R \gg r$ ). It follows from expression (1) that particles whose centers lie on a line parallel to the external electric field will attract one another when  $(\pi \rho V_F^2 - 2\epsilon E_0) < 0$ . With the aid of the well-known relation

$$V_F = \frac{\varepsilon \zeta}{4\pi\eta} E_0,\tag{2}$$

this inequality can be reduced to

$$\zeta < 4\eta \sqrt{2\pi/\rho\varepsilon} \,. \tag{3}$$

It follows from relations (1) and (3) that a particle oscillating in a fluid due to action of an alternating electric field has two zones of attraction adjacent to it on the respective two sides relative to the direction of the applied electric field. The zone of repulsion from other particles surrounds a particle in the plane perpendicular to the direction of vibrations and is nearly toroidal in form. Such a configuration of attraction and repulsion zones causes dispersed particles to form chain clusters oriented parallel to the external electric field [2].

In practice, however, the interaction of dispersed particles is not always uniquely determined by properties of the dispersion system, as would follow from inequality (3). It has been discovered [2, 4] that the character of this interaction can depend on the parameters of the external electric field. At certain amplitudes and frequencies of the external electric field the role of the hydrodynamic interaction of particles becomes so significant that particles will begin to cluster in strips oriented perpendicularly to the external electric field. In this study the ordering of quartz particles in a thin bottom layer by an alternating electric field was examined, under conditions of most appreciable hydrodynamic interaction.

The study was made in a miniature flat test cell  $13 \times 13 \times 1$  mm large furnished with two parallel segments of platinum wire spaced 7 mm apart and serving as electrodes. The source of alternating voltage was a model NGPK-3M generator. The structures formed by action of the electric field were examined under a microscope.

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Fig. 1. Periodic structure in a quartz suspension formed by an alternating electric field (amplitude of intensity,  $2.31 \cdot 10^3$  V/m; frequency, 2 Hz).



Fig. 2. Dependence of d ( $\mu$ m) on amplitude of electric field intensity E<sub>0</sub> (V/m) at various frequencies of the electric field (Hz): 1) 0.5; 2) 1; 3) 1.5; 4) 2; 5) 10 (dash line - A<sub>F</sub>).

The suspension was produced with grade KSSh-2 quartz which had been preground and then immersed in water distillate. The mean dimension of particles in fractions used for measurements was 3  $\mu$ m. The concentration of particles was selected so as to ensure that, with all particles spread over one plane near the bottom, the mean distance between them would be of the same order of magnitude as their intrinsic dimension. During measurement the test cell was held in a thermostat at a temperature within 298  $\pm$  1°K.

Within a few minutes after the suspension had been poured into the test cell, all particles settled down and a sedimentation-diffusion equilibrium was established near the bottom. Upon application of an alternating field these particles began to oscillate. In an external electric field with an amplitude and a frequency satisfying the conditions for induced dipole-dipole interaction weaker than hydrodynamic interaction these particles began to attract and repel one another in directions, respectively, perpendicular and parallel to the electric field. A few minutes after the electric field had been applied there appeared on the bottom of the test cell a structure consisting of equidistant strips of quartz particles vibrating synchronously with an amplitude  $A_0 - A_F$ . A typical pattern of such a structure is shown in Fig. 1. It is significant that, in contrast to the random pattern of clusters of dispersed particles oriented parallel to the electric field, in this case the pattern of strips appears ordered and can be characterized by the mean distance d between centers of neighboring strips. Experiments have demonstrated that this distance d is a quite sensitive and reproducible characteristic of one-dimensional periodic structures thus forming, a quantity which depends on properties of the suspension involved as well as on parameters of the external electric field.

The dependence of the mean center-to-center distance between strips on the intensity and the frequency of the electric field is shown in Fig. 2. At fixed frequency of the electric field and amplitude of its intensity the magnitude of d was determined here as the average of twelve readings. To each frequency corresponded a particular amplitude of electric field intensity at which a gradual transition was found to occur from periodic ordering of strips perpendicularly to the external field to formation of chain clusters parallel to that field. The critical electric field intensity at which transition occurs increases as the frequency of the electric field is increased.

The experimental procedure provided also for measuring the amplitude A<sub>F</sub> of oscillations of particles relative to the

fluid (electrophoresis), on the basis of which could then be evaluated the main parameters of polarizing and hydrodynamic interactions between dispersed particles. The intensity of hydrodynamic interaction is determined by the amplitude of the oscillation velocity  $V_F$  of particles, this velocity amplitude being equal to  $\omega A_F$ . Knowing  $V_F$ , one can calculate the  $\zeta$ -potential according to expression (2). This potential yields an estimate of the polarizing interaction [5].

The oscillation amplitude of particles was calculated as follows. The distribution of fluid oscillations over the depth in a flat closed container, oscillations due to action of an alternating electric field, can be described by the expression

$$A_{W}^{*}(z, t) = A_{0} \exp\left(-i\omega t\right) \frac{\sin\beta a \left(1+i\right) - \beta a \left(1+i\right) \cos\beta z \left(1+i\right)}{\sin\beta a \left(1+i\right) - \beta a \left(1+i\right) \cos\beta a \left(1-i\right)},$$
(4)

and the real part of the latter  $A_W(z, t)$  can be put in the form

$$A_{W}(z, t) = A_0 [B_1(z) \cos \omega t + B_2(z) \sin \omega t],$$
(5)

where  $B_1(z)$ ,  $B_2(z)$  are known function of the z-coordinate and  $\beta = \sqrt{\omega \rho/2\eta}$ . Assuming that the oscillations of a particle relative to the fluid due to action of the electric field follow the law  $\tilde{A} = A_F \cos \omega t$ , we obtain for the observable oscillations of a particle

$$A_{\rm H}(z, t) = A_{\rm W}(z, t) + \tilde{A} = A(z) \cos[\omega t + \varphi(z)],$$
(6)

where  $A(z) = \sqrt{[A_0B_1(z) + A_F]^2 + A_0^2B_2^2(z)}$  and  $\varphi(z) = -\arctan\left[B_2(z) \left| \left(B_1(z) + \frac{A_F}{A_0}\right)\right]$ . The quantity A(z) represents the

observable amplitude of particle oscillations at a point with the coordinate z. Upon measurement of A(z) at two arbitrary points with the aid of a microscope and subsequent insertion of these amplitudes into the expression for A(z), we obtain a system of two equations for  $A_F$  and  $A_o$ .

The resulting dependence of amplitude  $A_F$  on the amplitude of the electric field intensity at a frequency of 1 Hz is shown in Fig. 2 by the dash line. Calculation of the electrokinetic potential from  $A_F$  readings indicates a close agreement with inequality (3), but the interaction of particles in an electric field with the given parameters is qualitatively of a different nature than predicted by theory [2].

On the basis of the results obtained here, one can conclude that a description of structurization processes in highconcentration dispersion systems, where distances between particles are comparable with their intrinsic dimensions, must take into account mutual polarization of particles [5] and viscosity of the fluid. Formation of one-dimensional periodic structures is a significant feature which reveals hydrodynamic interaction of particles oscillating due to action of an alternating electric field.

## NOTATION

r, particle radius; R, distance between particles;  $\rho$ , fluid density;  $V_F$ , velocity of electrophoretic motion of a particle relative to the fluid;  $\epsilon$ , dielectric permittivity of the fluid;  $E_0$ , amplitude of the electric field intensity;  $\varphi$ , angle between the centerline of particles and the electric field;  $\zeta$ , electrokinetic potential;  $\eta$ , viscosity of the fluid;  $A_0$ , amplitude of electro-osmotic oscillations of the fluid at the container walls;  $A_F$ , amplitude of oscillations of particles relative to the fluid;  $\omega$ , cycle frequency; and a, half-thickness of the container.

## LITERATURE CITED

- 1. S. S. Dukhin and V. N. Shilov, Dielectric Phenomena and Double Layer in Dispersion Systems and in Polyelectrolytes [in Russian], Naukova Dumka, Kiev (1972).
- 2. T. A. Vorob'eva, "Processes of formation of oriented structures in polymer dispersions due to action of alternating electric field," Candidate's Dissertation, Chemical Sciences, Moscow (1967).
- 3. S. S. Dukhin and B. V. Deryagin, Electrophoresis [in Russian], Nauka, Moscow (1976).
- 4. I. Stauff, "Pearl string formation of emulsions in alternating electric field as relaxation effect," Kolloid. Z., <u>143</u>, 162-171 (1955).
- 5. V. N. Shilov and V. R. Estrela-L'opis, "Multiple theory of electrocoagulation of lyophobic sols," in: Surface Forces in Thin Films [in Russian], Nauka, Moscow (1979), pp. 35-45.