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## Experimental study and mathematical modelling of self-oscillation at the electrode-magnetic fluid interface in an electric field

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### Abstract

The article describes a mathematical model of self-oscillation in the form of a boundary value problem for a nonlinear system of partial differential equations, with a numerical solution. The numerical results were compared to the experimental data to confirm the adequacy of the model. The model uses the classical system of differential equations of material balance, Nernst-Planck and Poisson equations without simplifications or fitting parameters. The aim of the article was to study the parameters of concentration self-oscillation in a layer of the dispersed phase particles of magnetic fluid at the interface with an electrode in an electric field. For this purpose, we developed a mathematical model, the consistency of which was confirmed by the corresponding physical mechanism.

As a result of numerical experiments, we found the critical value of the potential jump after which self-oscillation began. We also determined the oscillation growth period and other characteristics of the process. We developed software called AutoWave01 with an intuitive user interface and advanced functionality for the study of self-oscillation in a thin layer of magnetic colloid.

**Keywords:** Magnetic fluid, Interface, Near-electrode layer, Electric field, Self-oscillation, Mathematical model.

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## 1. Introduction

Colloidal solutions of magnetic composite materials, later called “magnetic fluids”, were synthesised in the 1960s, and they still attract the attention of both experimenters and theorists [1, 2].

Magnetic fluids (MF) are ultradispersed stable colloids of ferro- or ferrimagnetic single-domain particles dispersed in various fluids and exhibiting intense Brownian motion. Small particles of metals such as iron, cobalt, nickel, gadolinium, and their various ferrites and ferromagnetic oxides are used as the dispersed phase. Surfactants are used as stabilisers in order to prevent coagulation of the colloidal solution, which would be unavoidable due to magnetic dipole-dipole and Van der Waals interactions, and to avoid the subsequent particle aggregation. When adsorbed on the surface of microcrystalline dispersed particles, the surfactants form a protective coating, which is a kind of a structural mechanical barrier. The average size of the dispersed particles is  $d_{av} \sim 10$  nm, so the MFs do not stratify and remain homogeneous practically indefinitely.

Dielectric fluid-based MFs have unique electrical and magneto-optical properties related, among other things, to near-surface phenomena. One of these properties of the MFs is their ability to form dense layers at the interface with the electrode in an electric field. These layers consist of the dispersed phase particles with a protective coating. These layers have a significant effect on the macroscopic properties of the colloidal system. They play a major role in obtaining materials with defined properties. This phenomenon is used in the field of indicating instruments [3–5].

Self-organisation was observed in the near-electrode layer and studied, which was self-oscillation and its distributed analogue, autowaves [6].

A magnetic fluid is a unique experimental medium, because the self-oscillation process can not only be observed and studied indefinitely in this medium, but it can also be controlled by means of the electric field.

Mathematical modelling of the self-oscillation process can make it possible to determine its fundamental laws and describe the state of the process at any point in space at any point of time.

In this study, we proposed a mathematical model for the self-oscillation of the MF particle concentration. The model is based on a dynamical approach: the processes occurring in the system are defined by partial differential equations describing the laws of conservation of matter and charge. The model does not include any fitting parameters. The relevant chemical reactions are considered by its boundary conditions.

## 2. Experimental

For the experiment, we used a magnetic fluid of the magnetite in kerosene type. The volume concentration of the solid phase was calculated based on the density of the magnetic fluid. For the MF with a density of  $\rho = 1.14$  kg/m<sup>3</sup>, the concentration of the solid phase was 3.4 vol%. The volume concentration of magnetite was calculated from the density values of the magnetic fluid, the dispersion medium (kerosene), and the dispersed phase (magnetite) by the formula:

$$\varphi_T = \frac{\rho_{MF} - \rho_O}{\rho_T - \rho_O},$$

where  $\rho_{MF}$ ,  $\rho_O$ , and  $\rho_T$  are the density values of the magnetic fluid, kerosene, and magnetite. The density of the MF was determined by the weight method using the buoyant force acting on a brass measuring cylinder immersed in the fluid. The value was calculated by the formula:

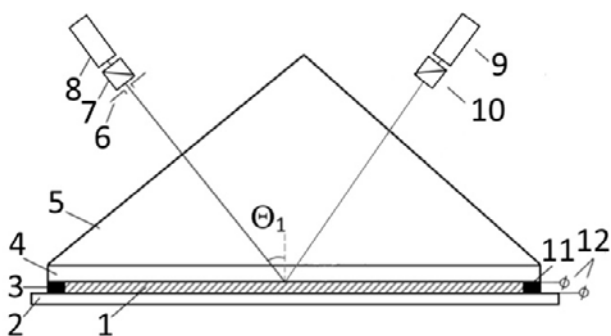
$$\rho = \frac{P_1 - P_2}{gV},$$

where  $P_1$  is the cylinder weight in air;  $P_2$  is the cylinder weight in fluid;  $V$  is the volume of the cylinder; and  $g$  is the gravitational acceleration.

The cylinder was weighed on a VLR-200 analytical balance. The error in determining the density  $\rho$  was  $\sim 0.1$  %.

The relative dielectric constant of the fluid was  $\mu = 2$ , its specific conductivity was  $\bar{A} = 3,8 \cdot 10^{-7}$  Ohm·m<sup>-1</sup>. To determine the dielectric constant and conductivity, we measured the resistance and capacity of the cell containing the MF using an LCR-821 LCR Meter (at a frequency of 1000 Hz). Then we calculated the conductivity  $\sigma$  and the dielectric constant  $\epsilon$ . The average particle size of magnetite was  $\sim 10$  nm, the protective coating consisting of oleic acid molecules was  $\sim 1.5$  nm thick [7, 8].

In order to determine the change in reflectivity (reflection coefficient, following the terminology in [9]) of the conductive electrode-magnetic fluid interface in the electric field, we used the experimental unit shown in Fig. 1. The unit was a plane-parallel cell consisting of two electrodes. One of the electrodes was made of glass with a conductive transparent coating (ITO). A glass prism was placed on the cell surface to avoid the illumination. The magnetic fluid was placed between the electrodes. A beam with a diameter of 2 mm, produced by a laser light source ( $\lambda = 650$  nm), was incident on the face of the prism adhered to glass with immersion liquid, so that the angle of incidence on the glass surface was 45 degrees. The beams reflected from the prism-glass interface and from the conductive coating - MF layer were spatially separated (5 mm). To level the parasitic illumination of the photodiodes, a diaphragm with an aperture of  $\sim 1$  mm was placed in front of polaroid 7. The laser and polaroid could rotate relative to the beam. The polarisation plane of polaroid 10 coincides with the polarisation plane of the laser beam. The polaroid served to reduce the depolarised component of the laser light. Rotating the laser and the polaroid ensured that the s-component (TE wave) was perpendicular to the plane of incidence. The light was reduced by rotating polaroid 7 to make the photodiode operate in linear mode. The surface area of the electrodes was  $S = 36 \times 30$  mm<sup>2</sup>, the



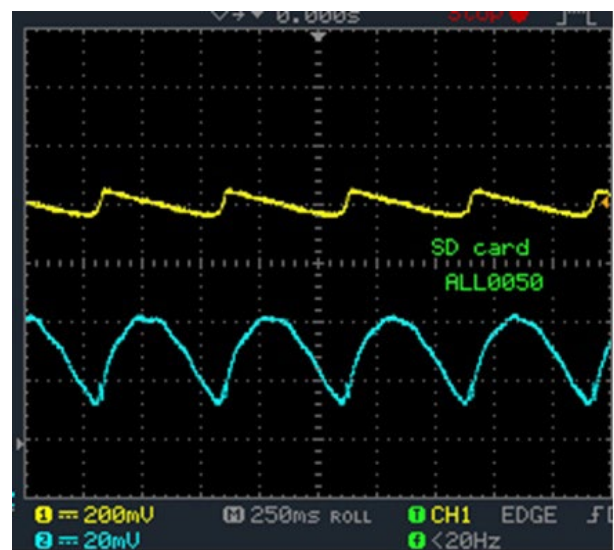
**Fig. 1.** Layout of the experimental unit for measuring reflection coefficient at the electrode-magnetic fluid interface: 1 – magnetic fluid; 2 – foil-clad paper-based laminate plate; 3, 11 – polystyrene insulation pads; 4 – glass with conductive transparent coating; 5 – right isosceles prism; 6 – diaphragm; 7, 10 – polaroids; 8 – photodiode; 9 – laser light source; and 12 – electrodes

thickness of the magnetic fluid layer was 250  $\mu$ m. In our experiments, we used glass samples with an ITO conductive coating (manufactured by OOO Poltek, St. Petersburg).

The experimental technique was as follows: voltage  $U$  was applied to the electrodes and, due to electrophoresis, 100-200 nm thick dense layers were formed. The layers consisted of magnetite particles. The glass with the ITO conductive coating was illuminated by a monochromatic light source. The beam was reflected from two surfaces: glass-conductive coating and conductive coating-magnetic fluid. It interfered, passed through diaphragm 6 and polaroid 7, and hit the FD-256 photodiode. The photodiode was connected to the input of a GDS-71022 dual beam oscilloscope. Since the photodiode operates in linear mode, the voltage at the oscilloscope is directly proportional to the intensity of the reflected beams (reflection coefficient).

When the jump in potential between the electrodes reached the critical value  $\varphi_{crit}$ , we observed a periodic change in the intensity of the reflected beam (Fig. 2, beam 2), which was due to a change in the thickness of the near-electrode dense layer.

When the cell surface was illuminated by light with a wide wavelength spectrum, the change in thickness of the layer was accompanied by a periodic colour change of the electrode surface from green to crimson. The green colour of the



**Fig. 2.** Auto oscillation of the current (1) and optical signal (2) in the cell with the magnetic fluid. The oscillation period is  $T \approx 0.5$  s, the potential jump is  $\Delta\varphi = 5$  B

surface corresponded to the reflection spectrum in the absence of a particle layer, and the crimson colour corresponded to the maximum thickness of the near-surface particle layer. We interpreted the periodic colour change of the surface or the periodic change in the intensity of the reflected light (Fig. 2, beam 2) as self-oscillation of the particle concentration. The electro-optical effect involves a change in the spectrum of reflected light (when illuminated by white light) or its intensity (when illuminated by monochromatic light) due to a thin near-surface layer of a magnetic fluid changing in an electric field. It is called electro-optical interference [10].

### 2.1. Physical model of particle self-oscillation in the cell with a magnetic fluid

*General characteristics of the electrical conductivity of a magnetic fluid.* The electrical conductivity of a magnetic fluid is due to several mechanisms. First of all, it is related to impurity ions, their concentration depends on the quality of washing the dispersed magnetite after coprecipitation during the MF preparation process:

$$\text{FeCl}_2 \cdot 4\text{H}_2\text{O} + 2\text{FeCl}_3 \cdot 6\text{H}_2\text{O} + 8\text{NH}_4\text{OH} \leftrightarrow \text{Fe}_5\text{O}_4 \downarrow + 8\text{NH}_4\text{Cl} + 20\text{H}_2\text{O}.$$

Due to the incomplete dissociation of the salt, the fluid also contains ionic complexes: ionic pairs and triplets [11, 12].

The second type of conductivity is due to contact processes on the electrode surfaces. They are called injection processes [11–14]. In high voltage fields with a medium intensity of about  $10^6$  V/m, electrochemical injection develops. It causes electrohydrodynamic effects in both liquid dielectrics and magnetic fluids [14].

In addition to ionic conduction, charge transfer in a magnetic fluid can be carried out by magnetic particles (MPs) [16].

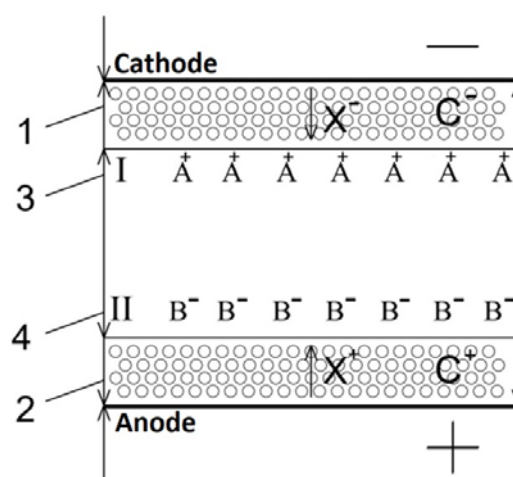
*Space charge formation in a magnetic fluid in an electric field.* In an electric field, a space charge is formed at the electrodes in a magnetic fluid. Many specific effects are associated with it [17–19], including the autowave process. The mechanism of space charge formation in a liquid dielectric under the action of an electric field in the vicinity of conductive surfaces was described in detail. In [13, 19], there is an expression for determining the thickness of the near-electrode layer of the space charge:

$$\delta \sim E_0 \frac{FD^\pm}{RT} \tau_M \sim \frac{\epsilon_0 \epsilon E_0 (b_1 + b_2)}{\sigma}, \quad (1)$$

where  $\tau_M$  is the Maxwell-Wagner relaxation time,  $D^\pm$  is the ion diffusion coefficient,  $E_0$  is the field intensity at the interface of the non-equilibrium dissociation-recombination reaction zone,  $b_1, b_2$  are the mobility values of positive and negative ions,  $F$  is the Faraday constant,  $T$  is the temperature,  $R$  is the molar gas constant,  $\sigma$  is the ohmic conduction,  $\epsilon$  is the dielectric permittivity, and  $\mu_0$  is the vacuum permittivity.

Ratio (1) shows that the thickness of the space charge layer can be estimated as the distance travelled by the ion during the relaxation time. That is why the effect of electrification is greater the lower the electrical conductivity of the liquid, and therefore the longer the Maxwell-Wagner relaxation time. It was shown in [13] that the repulsion of similarly charged ions at the electrodes causes an imbalance in the dissociation-recombination reaction of impurity ions. It results in the formation of a space charge.

Let us consider the layout of the magnetic fluid cell (Figure 3). The electric field transports anions (cations) from the near-cathode (near-anode) region. Since the cathode (anode) does not provide anions (cations), the cathode (anode) region is depleted of anions (cations), resulting in a cathode (anode) region with a positive (negative) charge.



**Fig. 3.** Layout of the cell containing the magnetic fluid: 1, 2 – dense near-electrode layers of magnetic particles; 3, 4 – the regions of the space charge;  $A^+$  and  $A^-$  – impurity ions;  $C^+$  and  $C^-$  – charged particles of the magnetic fluid; and  $X^+$ ,  $X^-$  – injected ions



The size of the localisation region of the space charge in a magnetite-in-kerosene fluid was estimated using the following parameters: the concentration of magnetite was 3.4 vol% and the specific conductivity was  $\sigma \approx 10^{-7} \text{ Ohm}^{-1}\text{m}^{-1}$ . The calculation showed that the width of the regions of space charge is  $\delta \sim 4 \cdot 10^{-7} \text{ m}$ , when the potential jump between the electrodes is  $\Delta\phi = 5 \text{ V}$ , the distance between the electrodes is  $d = 5 \cdot 10^{-5} \text{ m}$ , and the mobility of the impurity ions is  $b_1 \approx b_2 \approx 10^{-8} \text{ m}^2\text{V}^{-1}\text{s}^{-1}$  [13]. The value of the electric field intensity in the near-electrode region varies according to the Poisson equation due to the presence of a space charge with a density of  $\rho$  [21].

According to [22], characteristic time of formation of a space charge in a fluid with a medium magnetite concentration (3.4 vol%) for kerosene-based samples is about 0.05 s. To estimate the space charge formation time in an electric field, the migration polarisation of magnetite particles should be taken into account. In our case the migration polarisation can be described in the framework of Maxwell-Wagner theory. The Maxwell-Wagner polarisation time (2) is much shorter than the space charge formation time:

$$\tau_M \sim \frac{\epsilon_0 \epsilon}{\sigma} \sim \frac{2 \cdot 8.85 \cdot 10^{-12}}{10^{-7}} \sim 1.7 \cdot 10^{-4} \text{ s.} \quad (2)$$

A space charge also occurs at the surface of the dispersed particle. The region of the space charge around the particle is located in that part of the fluid where the ion, resulting from the dissociation of the impurity at the surface of the particle, keeps moving, without recombination.

*Concentration self-oscillation in a magnetic fluid.* In the absence of an electric field, magnetic fluid particles are not charged [22]. In the field  $\vec{E}$ , the particles are charged and acquire an induced dipole moment  $\vec{p}$ . As a result of the polarisation  $\vec{f}_E = (\vec{p} \nabla) \vec{E}$  and the Coulomb  $\vec{f}_q = q\vec{E}$  forces, the particles begin to move towards the corresponding electrode, forming a concentrated layer of particles. At the same time, the surface of the cell containing the MF changes its colour. Once a dense near-electrode layer is formed as a result of the contact of the magnetic particles with the electrode surface, the oleic acid molecules surrounding the particle begin

to interact with the electrode. For example, the reaction releases negative ions  $X^-$  at the cathode, which have almost zero mobility in the dense electrode layer, causing the cathode to charge. When the Coulomb repulsive energy of the layer  $U_q$  exceeds the attraction energy  $U_{im}$ , the layer becomes unstable. The particles change their charge and begin to move towards the opposite electrode. This way the concentration wave occurs. When the particles reach the region of space charge II (Fig. 3), they start moving in the opposite direction. Self-oscillation of the groups of magnetic particles was visualised by electrically controlled interference [10], based on periodic colour changes of the surface of the cell with magnetic fluid and the oscillations of the optical signal (Fig. 2). A dense near-electrode layer of particles was also formed on the opposite electrode (anode). Due to near-electrode reactions,  $X^+$  ions were formed, which resulted in the second concentration wave moving towards the first.

Assuming that all particles in the magnetic fluid have the same size, electric charge, and equal electrophoretic mobility  $b$ . Then in the stationary electric field  $\vec{E}$  the velocity of the particle is:  $v = b(\vec{E}, \vec{n})$ , where  $\vec{n}$  is the direction of the particle motion.

The particle mobility  $b = 5 \cdot 10^{-9} \text{ m}^2\text{V}^{-1}\text{s}^{-1}$  was determined using the formula from [21]. Then the velocity of the particle in the cell is  $v = 5 \cdot 10^{-4} \text{ m} \cdot \text{s}^{-1}$ .

On the other hand, based on the equality of the electrostatic force and the viscous frictional force, we can determine that:

$$q\vec{E} = 3\pi d\eta \cdot \vec{v}, \quad (3)$$

where  $d = 10^{-8} \text{ m}$  is the particle diameter,  $\eta = 1.5 \cdot 10^{-3} \text{ Pa} \cdot \text{s}$  is the dynamic viscosity of the dispersion medium,  $\vec{E}$  is the intensity of the electric field, and the charge of the particle  $q$  is equal to the charge of the electron [22]. According to our experimental data, the period of self-oscillation is  $\sim 1 \text{ s}$ . It is how long it takes the particle to leave the electrode and return.

For  $t = 0.5 \text{ s}$ , the particle covers a distance of  $\approx 5 \cdot 10^{-5} \text{ m}$ , that is, the distance to the opposite electrode. Thus, when the potential jump is  $\Delta\phi = 5$ , there is a periodic movement of particles from one electrode to the other.

Let us explain why the observed concentration oscillation is auto-oscillation. Self-oscillation is a non-damped oscillation in a dissipative dynamical system with nonlinear feedback. It is maintained by the energy of a constant, i.e. non-periodic, external influence [23]. In the laboratory experiments conducted, the source of this constant (non-periodic) influence is a jump in potential at the electrodes. The capacity of the near-electrode layer C serves as the nonlinear regulator. It regulates the accumulation of charge in the layer. In [24] it was shown that due to the formation of a pre-electrode layer, the cell containing the magnetic fluid was a capacitor of variable capacity. The charge in the near-electrode layer is accumulated during the time  $t_1$ . Then, after reaching the critical voltage at the electrodes  $U_{crit}$ , the layer changes its charge during the time  $t_1$ . This way, the constant impact is converted into a variable impact. The oscillating element of the system is the concentration of magnetic particles. The oscillation of the concentration is carried out via a feedback mechanism. It is represented by the layer resistance, which decreases sharply at  $U_{crit}$ . The energy dissipation in a self-oscillating system is compensated by the energy supplied to it from a constant voltage source, so that the oscillation is not damped.

## 2.2. A one-dimensional mathematical model of the oscillation process

We developed a simplified mathematical model of self-oscillation which allowed us to determine the dependence of the process characteristics on the initial parameters of the problem. For example, how the process depends on the potential jump between the electrodes  $\Delta\varphi$ , geometrical dimensions, etc. The proposed model considers the transfer of only positively and negatively charged magnetic particles with concentrations  $C_1, C_2$  and fluxes  $j_1, j_2$ , and the potential jump  $\Delta\varphi$  without taking into account chemical reactions. The influence of impurity and injection ions is taken into account indirectly by the boundary conditions. The basic idea of the model is to investigate the possibility of oscillation due to the change of charge of the MF particles near electrodes or in the regions of localised space charges.

We took  $x=0$  and  $x=H$  as the conditional points where the MF particles are charged. In

certain cases there can be an anode and a cathode. The initial conditions are  $C_1(0, x) = C_{10}(x)$  and  $C_2(0, x) = C_{20}(x)$ . The initial distribution of the neutral MF particles with a known concentration is determined by diffusion [25]. At the initial moment of time  $t=0$ , the MF particles are charged in the electric field. Near the point of  $x=H$  the MF particles are charged negatively, and near the point of  $x=0$  they are charged positively. In the electric field, negatively charged magnetic particles move towards the anode and positively charged particles move towards the cathode. Thus, dense layers are formed at the electrodes.

We used a one-dimensional nonlinear mathematical model with the classical system of differential equations of material balance, Nernst–Planck and Poisson [26–29] without simplifications or fitting parameters:

$$\frac{\partial C_i}{\partial t} = -\frac{\partial j_i}{\partial x}, \quad i = 1, 2, \quad (4)$$

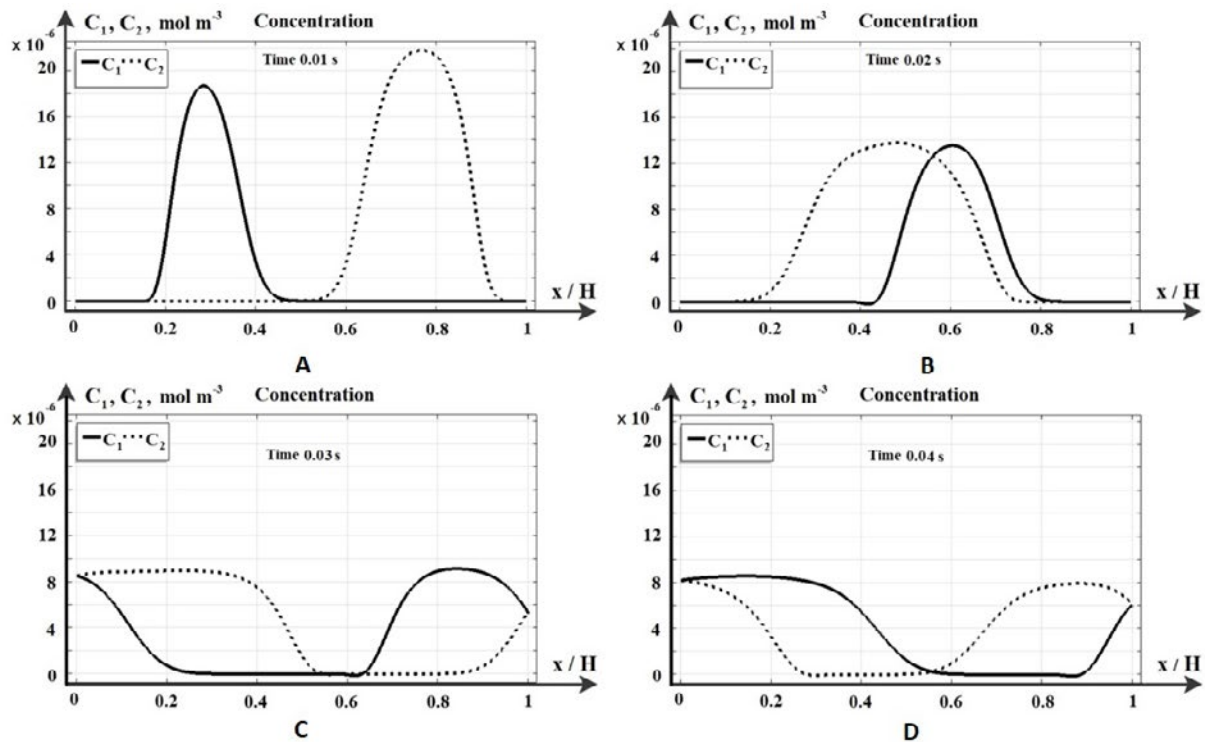
$$j_i = -\frac{F}{RT_0} z_i D_i C_i \frac{d\varphi}{dx} - D_i \frac{dC_i}{dx}, \quad i = 1, 2, \quad (5)$$

$$\frac{d^2\varphi}{dx^2} = -\frac{F}{\epsilon_r} (z_1 C_1 + z_2 C_2), \quad (6)$$

where  $C_i, j_i$  are the concentrations and fluxes of the charged MF particles,  $z_i$  are the charge numbers,  $\varphi$  is the potential,  $\epsilon_r$  is the dielectric permittivity of the magnetic fluid,  $F$  is the Faraday constant,  $D_i$  are the diffusion coefficients of the charged MF particles,  $T_0$  is the temperature, and  $R$  is the molar gas constant.

Our 1D model of self-oscillation of particles in a magnetic fluid considers two types of charged magnetic particles. So, the process of the charge change and corresponding oscillation take place from one to the other region of the space charge.

Assume that the process of the charge change of a wave of charged MF particles occurs gradually at the points  $x=0$  and  $x=H$  as the charged particles approach them. For example, negatively charged particles at the point  $x=0$  receive a positive charge and become positively charged, while positively charged MF particles (if any) reach the point  $x=H$  and become negatively charged (Figure 4b). Note that the charge change process is described by the boundary conditions at  $x=0$  (7) and at  $x=H$  (8), the potential jump  $\Delta\varphi$  is determined by (9).



**Fig. 4.** Concentration distribution of positively ( $C_1$ ) and negatively ( $C_2$ ) charged MF particles at: a)  $t = 0.01$  s, b)  $t = 0.02$  s, c)  $t = 0.03$  s, and d)  $t = 0.04$  s

$$\left( -\frac{F}{RT_0} D_1 C_2 \frac{d\phi}{dx} - D_1 \frac{dC_2}{dx} \right) \Bigg|_{x=0} = j_{1A}, \quad (7)$$

$$\left( \frac{F}{RT_0} D_2 C_1 \frac{d\phi}{dx} - D_2 \frac{dC_1}{dx} \right) \Bigg|_{x=0} = j_{2A},$$

$$-\left( -\frac{F}{RT_0} D_1 C_2 \frac{d\phi}{dx} - D_1 \frac{dC_2}{dx} \right) \Bigg|_{x=H} = j_{1K}, \quad (8)$$

$$-\left( \frac{F}{RT_0} D_2 C_1 \frac{d\phi}{dx} - D_2 \frac{dC_1}{dx} \right) \Bigg|_{x=H} = j_{2K},$$

$$\phi(t, 0) = \Delta\phi, \phi(t, H) = 0, \quad (9)$$

where  $j_{1A}, j_{2A}, j_{1K}, j_{2K}$  are the fluxes of overcharged particles at anode and cathode.

The initial conditions ( $t = 0$ ) determine the initial state of the system. Its further evolution is determined by the equations and boundary conditions. Since thin near-surface layers are formed at both electrodes, we assumed that at the initial moment of time the MF particles are concentrated at each of the electrodes (10):

$$C_1(0, x) = C_{10}(x), C_2(0, x) = C_{20}(x) \quad (10)$$

As will be shown below, this initial state results in two concentration waves moving towards each

other. The waves then interact with each other. For the change in potential, we defined the initial condition as a linear function (11):

$$\phi(0, x) = \phi_0(x) = \frac{H-x}{H} \Delta\phi. \quad (11)$$

Function (11) corresponds to condition (10) and Ohm's law at  $t = 0$ .

### 3. Numerical results and discussion

Numerical study was carried out with the following problem parameters: electrode spacing  $H = 5 \cdot 10^{-5}$  m, calculation for  $t \in [0, 10]$  seconds with a sample of 0.01 s. Mesh parameters: 20,000 elements. The potential jump  $\Delta\phi$  was studied in the range of 1 to 20 V. In our calculations we adopted the initial distribution of magnetic particle concentration corresponding to the experimental data:

$$C_1(0, x) = C_2(0, x) = 3.4 \text{ vol\%}.$$

The finite element method in the Comsol Multiphysics modelling environment was used to numerically solve the boundary value problem of the mathematical model.

The volume concentration of magnetite particles, expressed in  $\text{mol/m}^3$ , was:

$$C = 0.73593 \cdot 10^{-3} \text{ mol/m}^3,$$

$$C_1(0, x) = C_{10}(x) = 0.0074 \cdot e^{-x(0/01 \cdot H)} \text{ mol/m}^3,$$

$$C_2(0, x) = C_{20}(x) = 0.0074 \cdot e^{-(H-x)/(0/01 \cdot H)} \text{ mol/m}^3.$$

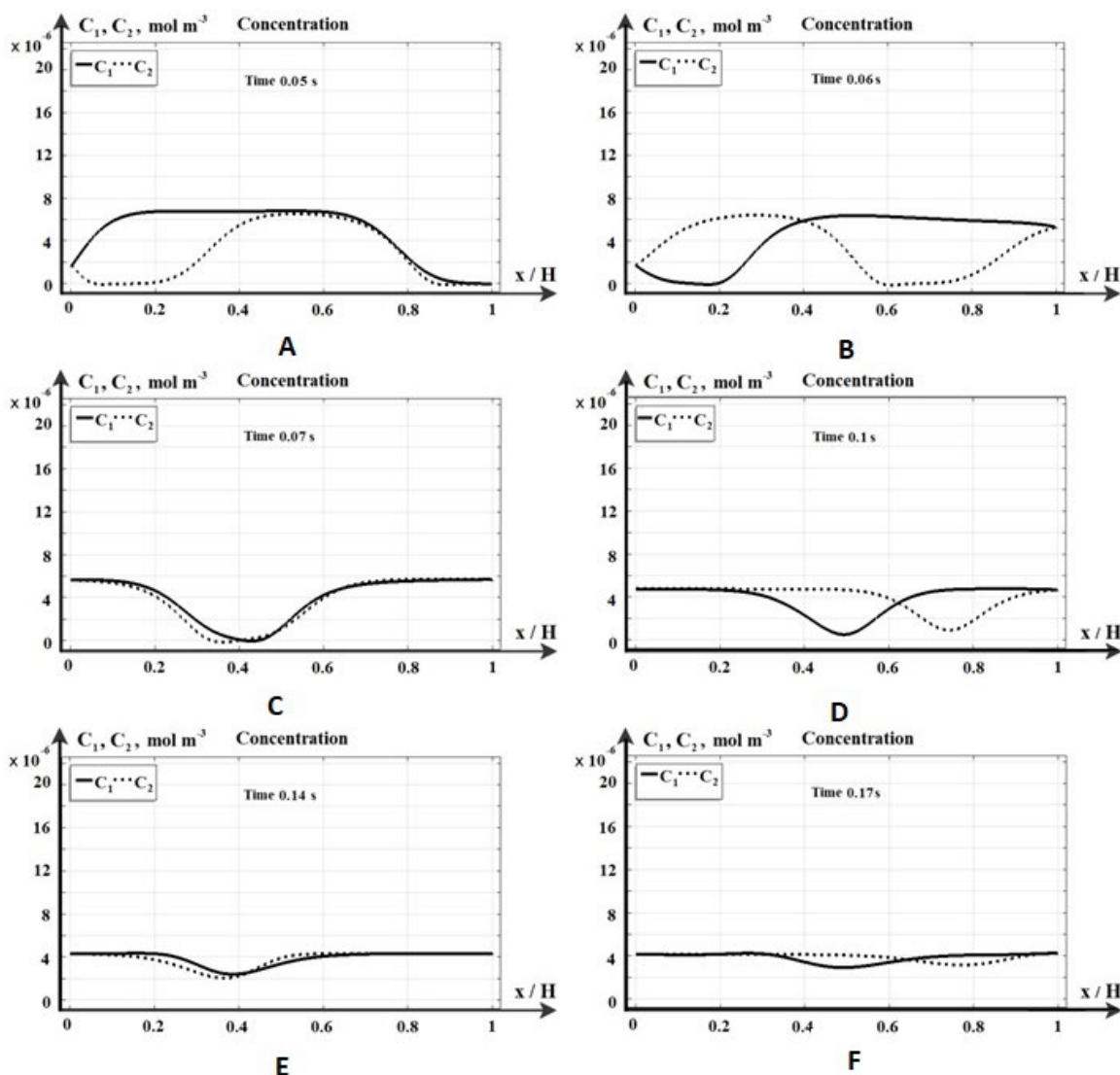
According to these initial conditions, the MF particles were initially distributed quite densely near the electrodes, i.e., a near-surface layer had already been formed.

### 3.1. Numerical analysis results for two concentration waves

Fig. 4a shows the initial concentration distribution of the particles  $C_1(t, x)$  and  $C_2(t, x)$ . At the initial moment of time they are charged positively at the coordinate  $x = 0$  or negatively at  $x = H$ . The positively charged MF particles

begin to move towards  $x = H$ , and the negatively charged magnetic particles move towards  $x = 0$  (Fig. 4b). The dotted line ( $C_2$ ) shows the negatively charged magnetic particles moving to the positively charged electrode (anode). The solid line ( $C_1$ ) shows the positively charged magnetic particles moving to the negatively charged electrode (cathode). Initially, two concentration waves with different amplitude parameters moved towards each other. Their amplitudes then gradually became equal (Fig. 4d). Once the concentration waves reached the corresponding electrodes, the particles changed their charges. Then the cycle was repeated.

At  $t = 0.05$  s (Fig. 5a) a front of positively charged magnetic particles was formed. When



**Fig. 5.** Concentration distribution of positively ( $C_1$ ) and negatively ( $C_2$ ) charged MF particles at: a)  $t = 0.05$  s, b)  $t = 0.06$  s, c)  $t = 0.07$  s, d)  $t = 0.1$  s, e)  $t = 0.14$  s, and f)  $t = 0.17$  s



one cycle ended (Fig. 5b), the next cycle began (Fig. 5c), etc. At  $t = 0.14$  s and  $t = 0.17$  s (Fig. 5e, f), the number of positively and negatively charged particles became almost constant. At a time of 0.29 s (Fig. 6A) the wave of positively charged magnetic particles was moving towards the cathode, and the wave of negatively charged particles was moving towards the anode. At  $t = 0.33$  s (Fig. 6b) the wave at the anode reached its maximum. Then (Fig. 6B) it began to recede.

Thus, the particles move from one electrode to the other and back. They change their charge at the electrodes.

### 3.2. Comparative analysis of the numerical and laboratory experiments

The analysis revealed that the oscillation period in the numerical experiment varies from 0.5 to 0.6 s, which is consistent with the experimental data. The numerical study was carried out for different values of the potential jump  $\Delta\varphi$  in the range from 1 to 20 V. It was determined that the larger the potential jump, the smaller the oscillation period, which was also confirmed experimentally. Moreover, it should be noted that the smaller the potential jump, the more the diffusion process prevails over electromigration, thus suppressing the auto-oscillation process.

We saw that when the potential jump was less than 3 V, concentration waves were not formed in computer simulations and were not observed experimentally. Different charge numbers of the particles from  $\pm 1$  to  $\pm 4$  were considered. It was determined that the larger the charge value, the more chaotic the motion and waveform became.

### 3.3. Software development

We developed software called AutoWave01 with an intuitive user interface and advanced functionality for the study of self-oscillation in a thin layer of magnetic colloid. Using AutoWave01 significantly reduces the cost of expensive experiments. It provides preliminary analysis to find the optimum parameters and all results can be stored in a convenient format.

## 4. Conclusion

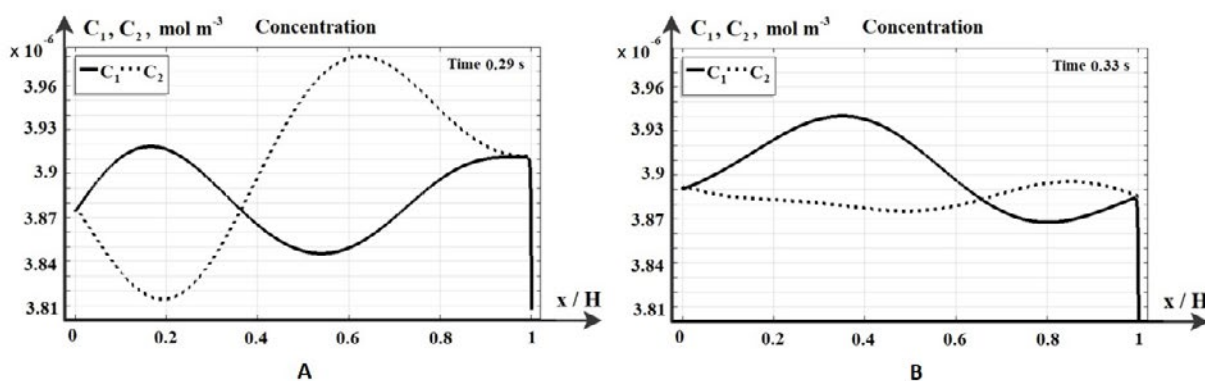
In this study, we carried out mathematical and computer 1D simulations of self-oscillation of particles in magnetic fluids, which are fully consistent with the laboratory experiment at a qualitative level. When the voltage on the electrodes increases to a certain threshold value  $U_{crit}$ , oscillation of the particles of the dispersed phase occurs between the electrodes. This phenomenon is related to the charging of the particles and the change in charge. Our model has a predictive value: it can be used to determine the critical value of the potential jump, to find the oscillation growth period and other characteristics of the process.

### Authors contributions

All authors made an equivalent contribution to the preparation of the publication.

### Conflict of interests

The authors declare that they have no known competing financial interests or personal relationships that could have influenced the work reported in this paper.



**Fig. 6.** Concentration distribution of positively ( $C_1$ ) and negatively ( $C_2$ ) charged MF particles at a)  $t = 0.29$  s, b)  $t = 0.33$  s (enlarged scale)

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