Autowave process in a thin layer of magnetodielectric emulsion

V S Chekanov1* and Yu I Dikansky1

1 North Caucasus Federal University, 355017, st. Pushkin 1, Stavropol, Russia

*oranjejam@mail.ru

Abstract. This paper describes the behavior of a magnetodielectric emulsion "magnetic fluid in oil" in an electric field. Such an emulsion is a functional electrically controlled medium, which may be of practical interest. In an electric field, a layer of microdroplets is formed in the near-electrode region of the magnetic emulsion, and an autowave process is observed at a certain critical strength.

1. Introduction

Research in the development of new materials with controllable properties, the so-called "smart" materials, has recently attracted both theoretical and practical interest [1]. Magnetic (magnetodielectric) emulsions can be classified as such materials. Magnetic emulsions are dispersed systems consisting of two liquid phases, one of which is a magnetic fluid.

Magnetic fluid (MF) is an artificially synthesized material consisting of nanosized particles of a dispersed phase (ferro- or ferrimagnetics) suspended in a carrier liquid (organic solvent, water) and stabilized with a surfactant (oleic acid). On the basis of MF, magnetic emulsions were developed as a magnetically sensitive medium [2, 3].

Such emulsions are a functional material, the properties of which can be controlled by external influences (for example, an electric or magnetic field).

To date, there is a fairly large number of experimental and theoretical works devoted to the study of the properties of magnetic emulsions in a magnetic field.

So, in works [4, 5, 6] it is shown that the action of magnetic field leads to the emulsion structuring – formation of chain aggregates, droplets and dense structures such as columns, also formed by microdroplets. The dependence of the emulsion electrical conductivity on the magnitude of the applied magnetic field has been studied.

In [7, 8], the concentration and temperature dependences of the magnetic susceptibility of an emulsion, the nature of its magnetization in a constant magnetic field, the phenomenon of the emulsion reversal and the associated change in its magnetic permeability were investigated.

When exposed to crossed magnetic and electric fields [9], it was shown, that the emulsion becomes an electrically anisotropic medium under these conditions, which manifests itself in the dependence of the dielectric constant and electrical conductivity on the angle between the directions of the vectors $\vec{E}$ and $\vec{H}$.

The effect of a constant electric field on magnetodielectric emulsions has not been sufficiently studied; therefore, the purpose of this work is to study the near-electrode processes, structure formation and dynamics of particles of the dispersed phase of a magnetic emulsion in a constant field.
electric field. The practical interest of studying the features of the magnetic emulsion interaction with an electric field is important for its application as an electrically controlled medium.

2. Materials and research methods. Experiment technique
The object of the study was a magnetic emulsion, which is a suspension of microdroplets of a magnetic fluid in an oil medium. It was obtained by adding a homogeneous magnetic fluid to the AMG-10 hydraulic oil. Magnetic fluid is a colloidal solution of magnetite particles in LZ-MG-2 oil. The ratio of the volumes of the dispersion medium – emulsion (AMG-10 oil) and the dispersed phase of the emulsion (magnetic fluid) was 9:1. Thus, we have a direct emulsion of the "magnetic fluid in oil" type. Dielectric constant of a magnetic fluid droplet $\varepsilon_1 = 5$, dielectric constant of the dispersion medium $\varepsilon_2 = 2.7$. The average size of magnetite particles in magnetic fluid droplets is $\sim 10$ nm. The mixture was stirred using an electromechanical stirrer, so we get a magnetic emulsion containing microdroplets of a magnetic fluid with a diameter of $d = 2-5 \mu m$.

Table 1 shows the physical parameters of the magnetic fluid used to prepare the emulsion and the dispersion medium parameters.

| Parameter | Value |
|-----------|-------|
| Saturation magnetization of MF, kA/m | 15.9 |
| Interfacial tension N/m | 3.05*10^{-7} |
| Density kg/m^3 | 1326 |
| Conductivity of a drop of MF Ohm·m^{-1} | 4.5 \times 10^{-7} |
| Conductivity of the dispersion medium (AMG-10 oil) Ohm·m^{-1} | 7 \times 10^{-12} |

To study the effect of the electric field on the magnetic emulsion, it was placed in a plane-parallel cell (Figure 1), consisting of two electrodes, one of which was glass with a conductive transparent coating (ITO) – a semiconductor material transparent to visible light. The surface area of the electrodes is $S=60 \times 80$ mm$^2$. The thickness of the glass samples is 4 mm, the thickness of the conductive coating is $h_0 = (280 \pm 5)$ nm (manufactured by Polytech LLC, St. Petersburg). The thickness of the magnetodielectric emulsion layer is 60 microns.

To eliminate the illumination from the outer surface of the glass, a glass prism was glued to it with an immersion liquid.

A beam from the illuminator 8 (incandescent lamp) falls on the edge of the prism 6 (Figure 1) at an angle of $\theta = 45^\circ$. In the absence of voltage across electrodes 9, the beam is reflected from the “glass-ITO” and “ITO-magnetic emulsion” interfaces and interferes in the ITO thin membrane. The reflected light turns out to be colored (interference in a thin membranes). When a voltage is applied ($U=8V$), apparently, the droplets of the magnetic fluid form a layer at the electrodes that changes in thickness depending on the applied field value. The falling beam is reflected from the boundaries "glass - ITO" and "ITO - near the electrode layer", interferes, and the color of the reflecting surface changes.
This observation method is called electrically controlled interference and is described in [10].

*Figure 2.* Change in color of the cell surface with a magnetic emulsion. Voltage at the electrodes $U = 8 \text{ V}$.

If the voltage applied to the electrodes is increased to about 15 V, then an autowave process is observed (Figure 3) with different modes: a single wave, spiral waves, leading centers. A similar process was observed and investigated in magnetic fluids [11, 12], the mechanism of the occurrence of which the authors attributed to the periodic formation and destruction of thin near-electrode layers consisting of close-packed magnetite particles.

*Figure 3.* Experimental observation of auto waves in a magnetic emulsion: a - single wave, b - leading center, c and d - spiral waves. The voltage is $U = 15 \text{ V}$.

3. **Discussion of experimental results**

To understand the physical mechanism of the observed autowave process in the emulsion, it is necessary to investigate the effect of a constant electric field on its structure.

Let's place the emulsion of the magnetic fluid in the cell (Figure 4): a slide on which rectangular plates of metal (copper) foil are glued, so that a narrow gap, 1.3 mm wide, remains between their ends.

A magnetic emulsion is placed in it, and a cover glass is placed on top. The cell is placed on the microscope stage (with the foil facing up) and the field strength is slowly changed in the range $E = 0 \div 150 \text{ kV/m}$.

*Figure 4.* Scheme of a cell for studying the deformation of microdroplets of an emulsion of a magnetic fluid in an electric field: 1 - a glass slide, 2 - metal plates, 3 - cover glass.
As a result of the action of the field, the droplets are transformed from spherical into flattened ellipsoids, the minor axis of which is located along the field (Figure 5 b). Then, smaller drops begin to detach from the drops in streams (Figure 5c). A similar effect can be associated with the fact that a layer of magnetite particles is formed in an electric field at the “drop-oil” interface, the surface tension decreases, and “daughter” droplets are emulsified from the surface of a large drop.

**Figure 5.** Dynamics of changes in microdroplets of an emulsion of a magnetic fluid in an electric field \( \vec{E} = 0 \text{÷} 150 \text{ kV/m} \).

Not initially charged, in an electric field, these droplets acquire an electric dipole moment and, as a result of dipolophoresis, move to the electrode (Figure 5d), forming a near-electrode layer. The formation in an electric field of a layer consisting of emulsion micro drops was reported in [13-15]. Such a layer is a magnetic fluid droplet deformed in the form of oblate and elongated ellipsoids of rotation [16].

MF drops are polarized in an electric field. Since the conductivity of the MF (\( \sigma = 10^{-7} \text{ (Ohm} \cdot \text{m})^{-1} \)) is caused by impurity ions, then their redistribution occurs in the electric field and the microdroplet acquires a dipole moment. As a result of dipolophoresis, the MF drops end up at the electrode, being attracted to it with a force \( \vec{f}_{im} = (\vec{p}\vec{V})\vec{E} \), where \( \vec{p} \) is the dipole moment of the drop. The energy of attraction of the layer to the electrode we call \( U_{im} \).

In addition to the impurity ions contained in the dispersed phase (magnetic fluid), the charge carriers in the magnetic emulsion are injected ions, which are formed as a result of near-electrode chemical reactions. For example, negative ions on the electrodes are formed as a result of the electrochemical act of molecules of oleic acid (surfactant in a magnetic fluid), which has enhanced electron-acceptor properties.

Ion \( X^- \), falling into the surface layer of microdroplets, has practically zero mobility, which leads to the charging of the layer. The total charge \( Q \), per unit area induces the Coulomb energy of the repulsion of the layer, equal to \( U_q = Q\delta \), where \( \delta \) is the layer thickness.

When \( U_q \) exceeds the average energy of attraction \( U_{im} \) the layer of microdroplets will detach from the electrode and, since the voltage in the cell (Fig. 1) is \( \gg 10^5 \text{ V/m} \), apparently, several types of instability of the droplet surface appear: the droplet either divides into several daughter droplets, or small droplets start moving from its end, and it acts as a “micro nozzle” [16,17] (Figure 4c).

Thus, under the condition \( U_q > U_{im} \) an electrohydrodynamic (EHD) flow is generated in a magnetic emulsion [18].

The size of the droplets formed during the disintegration of the near-electrode layer was estimated experimentally by the rate of formation and separation of the layer – \( d \approx 10^{-7} \text{ m} \), which is much smaller than the initial size of the emulsion droplets (2-5\cdot10^{-6} \text{ m}), and the distance between the electrodes, therefore the emulsion can be considered as a continuous medium.

Once disintegrated into small droplets, microdroplets cannot unite; therefore, the nature of the autowave process changes after the first decay of the droplets (the sequence of frames is shown in Figure 6). When the voltage on the electrodes is \( U = 15 \text{ V} \), a single wave passes first (\( t = 1-10 \text{ s} \)), and then we can see the phase waves (\( t = 10-20 \text{ s} \)).
Figure 6. Sequence of frames of the autowave process development in a magnetic emulsion.

4. Conclusion
It was found out that as a result of the action of a constant electric field on a thin layer of a magnetodielectric emulsion "magnetic fluid in oil", a layer of microdroplets is formed near the electrodes, visually detected by a change in the reflection coefficient of the cell surface with the emulsion and a change in its color. When the critical value of the field strength is reached, the layer gets recharged, becomes unstable, and an autowave process is observed with typical modes: a single wave, spiral waves, leading centers. The scientific novelty of the research is due to the fact that the autowave process in a magnetic emulsion was observed for the first time.

5. References
[1] Molchanov V S, Pletneva V A, Klepikov I A, Razumovskaya I V and Philippova O E 2018 Soft magnetic nanocomposites with adaptive matrix composed of wormlike surfactant micelles J. RSC advances 8(21) 11589-97
[2] Romankiw L T Stable emulsion and method for preparation thereof 1976 US Patent No 3981844 (USA)
[3] Chekanov V V and Drozdova V I 1982 Magneto-sensitive emulsion A.C. No 966735 Discoveries. Inventions. Prom. samples. Trademarks (USSR) 38
[4] Drozdova V I and Chekanov V V 1981 Diffusion of ferrofluid particles in a magnetic field Magnetic hydrodynamics 161-3
[5] Ivey M, Liu J, Zhu Y and Cutillas S 2000 Magnetic-field-induced structural transitions in a ferrofluid emulsion Phys. Rev. E 63 011403
[6] Zakinyan A R and Zakinyan A A 2020 Electrical conductivity of magnetic emulsions with a chain microstructure in a magnetic field Science. Innovations. Technologies
[7] Dikansky Yu I, Bedzhanyan M A and Kiselev V V 1995 Features of magnetization of magnetic emulsions Magnetic hydrodynamics 31(1-2) 79–84
[8] Dikansky Yu I, Zakinyan A R and Konstantinova N Yu 2008 On the magnetic permeability of a magnetodielectric emulsion ZhTF 78(1) 21-6
[9] Dikansky Y I, Zakinyan A R and Tkacheva E S 2013 Electrical properties of magnetodielectric emulsions in magnetic field Modern science and innovations 130-6
[10] Chekanov V V, Kandaurova N V, Chekanov V S and Romantsev V V 2019 The use of electrically controlled interference in an electrically tunable light filter and for observing the autowave process in the near-electrode layer of a magnetic fluid Optical Journal (Saint Petersburg) 86(1) 21-6
[11] Chekanov V V, Kandaurova N V and Chekanov V S 2017 Phase autowaves in the near-electrode layer in the electrochemical cell with a magnetic fluid Journal of Magnetism and Magnetic Material 431 38-41
[12] Chekanov V V, Kandaurova N V and Chekanov V S 2018 Observation of the autowave process in the near-electrode layer of the magnetic fluid. Spiral waves formation mechanism Journal of Molecular Liquids 272 828–33
[13] Kozhevnikov V M, Larionov Yu A, Chuenkova I Yu and Antonova A A 2018 Particle Aggregation in Magnetic Fluid Layer in Electric Field Magnetohydrodynamics 54(1-2) 85–90
[14] Kozhevnikov V M, Larionov Yu A, Chuenkova I Yu and Danilov M I 2004 Obtaining the structured magnetic fluids in an electric field and their technical applications Magnetohydrodynamics 40(3) 269-80
[15] Dikansky Yu I and Nechaeva O A 2003 Structural transformations in magnetic fluid in electric and magnetic fields Colloid Journal 65(3) 1-5
[16] Grigoriev A I 2000 Capillary electrostatic instabilities Soros educational journal 6(6) 37–43
[17] Shiryaeva S O 2000 On some regularities of polarization and dispersion of a drop in an electrostatic field ZhTF 70(6) 20–6
[18] Stishkov Yu K and Bogdanov D V 2017 Computer simulation of the injection mechanism of the occurrence of EHD flows in liquids with an increased level of low-voltage conductivity, Electronic Materials Processing 53(1) 16–22