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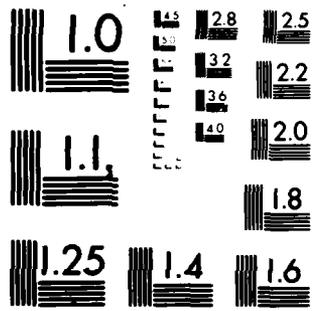
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by

Yu. F. Deinega

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SOME PROBLEMS OF THE ELECTRO-RHEOLOGY OF DISPERSED SYSTEMS

[NEKOTORYE PROBLEMY ELEKTROREOLOGII DISPERSNYKH SISTEM]

by

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Masliny i Tekhnol. Pererab. Kauchukov
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EDITOR'S SUMMARY

A qualitative review of some of the problems confronting a clear understanding of the electro-rheological effect and of some potential new applications of the phenomenon.

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LIST OF CONTENTS

	<u>Page</u>
Text	3
References	9
Illustrations	Figures 1-2

1 TEXT

Electrical methods have formed the classical basis for the investigation of the structure of dispersed systems for more than 100 years. Their precise value is difficult to assess but it is probably sufficient to recall the role played by electrical methods in the formation of the contemporary representatives of dispersed systems.

The main thrust of these investigations was directed towards aqueous systems with non-aqueous system receiving scant attention. Recently the situation has changed noticeably with an ever increasing interest into the electrical properties of non-aqueous, particularly hydrocarbon, dispersed systems. This is linked directly to the discovery of new phenomena in such systems together with the possibility of practical applications.

There are a number of distinct differences between the behaviour of hydrocarbon dispersed systems in electric fields and the aqueous counterpart. Firstly there is the opportunity to sustain much higher field strengths by virtue of the low conductivity and high breakdown strength of such systems. Secondly, there are a number of special features involved in the construction of the surface layer. In particular the electrical double layer thickness, which in a hydrocarbon media is some two orders of magnitude greater than for the aqueous system. On the surface of a hydrophilic phase in a hydrocarbon medium, as a general rule, a thin hydrated layer is formed which plays an important part in the charge formation and polarisation process. Finally, a hydrocarbon medium creates conditions favourable for the creation of surface electrical heterogeneity.

In the final analysis, all the behavioural multiplicity of dispersed systems in an electric field is determined by a combination of electro-kinetic, polarisation and electro-chemical phenomena. The formulation of a generalised picture of the behaviour of dispersed systems in electric fields depends upon the electrical characteristics of the system, its concentration and the field strength. The formulation of such a picture is the most important problem relating to the electro-chemistry of dispersed systems.

The phenomena arising from the behaviour of dispersed systems under electric fields are finding application in electro-dyeing, oil dehydration, electro-filtration, electro-separation, electro-phoretic coatings and the production of anisotropic materials. Of particular interest is the opportunity to change the rheological properties of dispersed systems by the application of an electric field. This area of investigation is frequently referred to as the electro-rheological effect (ERE). The basic problem in electro-rheology is the establishment of the interaction between the structural, electrical and rheological properties of dispersed systems so that the effect can be used either to control the mechanical properties by an electric field and varying charge on the particles, or to regulate the electrical properties of the system by mechanical means.

The overall view of electro-rheology can be broken down into three fields:

- (1) A study of the effect of an electrical charge on particles of the dispersed phase, and of macromolecules, on the rheological properties of the system. This is essentially a study of the first, second and third electro-viscous effect.

- (2) A study of the effect of deformation on the structure and dielectric properties of dispersed systems. Changes in the dielectric constant, electro-conductivity and loss tangent resulting from deformation, the rheoelectric effects, can provide important information concerning the deformation process - the process of orientation, creation and destruction of structures, thereby giving rise to electrically anisotropic structures.
- (3) A study of the effect of electric fields on the structure and rheological properties of dispersed systems.

Strong electric fields can, depending on the nature of the change in the structure, either raise or lower the effective viscosity of dispersed systems. As Yu. F. Deinega and S.V. Vinogradov¹ demonstrated, the resistance of a dispersed system can decrease ten-fold as a result of phase separation in an electric field. This is associated with the formation of both boundary and volume layers in the dispersion medium.

On the other hand, however, in a series of hydrocarbon dispersed systems subjected to an electric field, a significant increase in viscosity can be observed. This phenomena of a rapid reversible increase in viscosity under the influence of an electric field has been given the name of the electro-rheological effect². The ERE is a completely unique phenomenon: usually a change in a property by tens of per cent is considered significant, but the effective viscosity increase of the ERE is hundreds or even thousands of times, so the fluid system effectively turns into a solid body. The ERE can provide the basis for a series of new and unique designs and devices.

Important investigations into the ERE were carried out by Winslow⁶, Klass and Martinek⁴ and Shul'man and Matsepuro⁷. The influence of various factors, intensity and form of electric field, the rate of deformation, temperature, disperse phase concentration and different additives, on the effect were studied.

The central question to the electro-rheology of dispersed systems is the study of the nature of the ERE. The increase in viscosity of such systems has been attributed to the orientation of particles³, the deformation of the electrical double layer¹, inter-electrode circulation⁵ and particle interactions⁶⁻⁹.

The orientation of particles in an electric field imposes a limit on the freedom of their hydrodynamic rotation and affects the dissipation of mechanical energy. Chaffey and Mason³ have shown that a retardation of the rotation of spherical particles in the shear flow leads to an increase in the coefficient in the Einstein formula for the effective viscosity from 2.5 to 4; the effect of the electric field on the viscosity is not, therefore, great. However, with an increase in the anisotropy of the particles, it can increase significantly.

The deformation of the electrical double layer, superimposed on the hydrodynamic deformation, can according to Klauss and Martinek⁴, increase the first and second electro-viscous effects. This hypothesis in the opinion of the authors, is reinforced by a series of experimental facts, in particular the rapid response of a system to the electric field. No quantitative support for this hypothesis has yet been obtained however.

In dilute dispersed systems an electric field can result in an interelectrode circulation or oscillation of the particles. In a shear flow oscillation of particles transverse to the flow can account for the difference in velocity of the particle and the liquid, *i.e.* where the flow is concerned, forces will act on the particle imparting an additional acceleration to it. This leads to a supplementary dissipation of energy and consequently an increase in the effective viscosity⁵. Such particle motion in dilute systems leads only to a small increase in viscosity. However, such oscillation can occur in concentrated suspensions where the structure is disturbed. The increase here can be more pronounced.

One reason for the increase in viscosity is the formation of a structure, determined by the interaction between adjacent particles and the electrodes. Dielectric polarisation lies at the root of particle interactions. The imposition of dipole moments to the particles in the system leads to the generation of forces between particles which disappear when the field is turned off. An investigation into the nature of the dielectric polarisation, its dependence on hydration, temperature, frequency of the electric field and the role played by conductivity in structure formation represents a formidable problem.

In polar media, the production of aggregate chains is usually associated with polarisation of the ionic sheath; self polarisation of materials is negligible. In non-polar, media, particularly, hydrocarbon media where the charge concentration is very small, an important part is played by the hydration of particles in the dispersed phase, and also by film polarisation.

In order to establish the relationship between the polarisation of disperse systems and the ERE, complex investigations are required into the dielectric and electro-rheological properties of dispersed systems. Of particular importance are measurements of the dielectric parameters at high field intensities used in the study of the electro-rheological effect.

The nature of the polarisation of a system in a strong electric field is essentially different from polarisation in weak fields, this is illustrated in Fig 1. With an increase in water content, the dielectric constant rises, obtains a maximum and then falls. Such a characteristic differs from measurements at low field intensities where no maximum is observed. As the field strength increases the maximum shifts to lower water contents.

The ERE shows an analogous dependence, Fig 2. As the water content is increased, the effect increases and then falls sharply. The higher the field strength, the lower the moisture content at which the maximum occurs and the more strongly is the maximum defined.

Experimentally, variations in the ERE are observed with changes in temperature, frequency of the applied field and in association with the particular features of the polarisation of hydrated disperse systems in strong electric fields.

Particular aspects of the polarisation of hydrated systems characterise the appearance of a maximum in both dielectric and electro-rheological effect versus moisture content curves. Water molecules adsorbed onto particles form a thin hydrated layer which increases their conductivity and lead to a polarisation of the particles. The force of

attraction between particles due to polarisation increases and the ERE becomes stronger. The change of water from the free state to the bound state is accompanied by an ordering of the structure of the water and by the surface formation of chains along which proton migration can occur. With such a mechanism, charges will be transported along the surface of the particles, but charge transfer between particles will be difficult. As the moisture content increases, free water appears with 'collars' of water forming between particles. The electrical conductivity rises sharply as shown in the lower portion of Fig 1. This results in a reduction in the polarisation of the system and leads to a reduction in the force of attraction due to polarisation and even to a change in sign of the force⁹.

Therefore, an increase in the conductivity of the particles will produce an increase in the forces of polarisation, so long as free charges are displaced only within the bounds of the particle. Charge transfer between particles is accompanied by a reduction in the polarisation interaction and by a reduction in the ERE. These findings explain the temperature and frequency dependence of the electro-rheological effect.

An important role in the breakdown of a structure in the process of its destruction is the part played by the interelectrode oscillation of particles which determines the interaction of particles with the electrode and the charge transfer of the particle. Investigations into the oscillation of particles by recording of electrical pulses with the charge transfer at the electrodes has made it possible to put forward the proposition concerning the presence in the absorbed layer of the activator two types of charge carrier, electrons and ions¹². It should be noted that an important part in the charge transfer of hydrated particles may be played by the electro-chemical decomposition of water. Further investigations into the interaction of particles with the electrodes are of interest where the study of the kinetic and the mechanism of structure formation in the electric field is considered. A central problem in studies of the ERE is the relationship between such structure formation and the rheological parameters of dispersed systems. Two approaches can be considered: in the first case, one can examine electro-rheograms on the basis of a suitable rheological equation of state and quantify the results with solid phase concentration, concentration of surface-adsorbed water, field strength and temperature^{13,14}. Such results can be used for engineering calculations of the hydraulic characteristic in flow of electro-rheological systems.

The second approach is more complex and is based on the calculation of the change in the force of interaction between particles of the dispersed phase in an electric field and the effect of this interaction on the structure and rheological properties of dispersed systems^{8,9}.

The interaction of particles in an electric field leads to the formation of chain structures which show a resistance to the flow. The steady state dimension of the chain in the flow is determined by the competition between the structure forming effect of the field and the destructive effect of the liquid flow. Consequently, the problem can be divided into two parts:-

- (1) Determination of the force of interaction between polarised particles.

- (2) Determination of the steady state dimensions of chains for given forces of interaction between particles and between particle and flow.

Knowing the moment of the viscous forces applied to the chain it is possible to evaluate the change in viscosity.

The polarisation interaction is the result of the induced dipole moments. Induced dipole moments are difficult to calculate for a broad class of systems polarised by various mechanisms. The interaction of polarised particles can be calculated quite simply if the distance between them is great. If, however, the distance between them coincides with their dimensions, each particle will be subjected to the effect of both an outer homogeneous and inhomogeneous polarisation field of a neighbouring particle. To take into account the mutual polarisation even of spherical particles presents a very complex problem. Up to the present, a solution has only been obtained for the simplest case of a highly conducting particle in the dielectric¹⁵. Taking into account the mutual polarisation of the particles increases the energy of attraction by almost six times¹⁶.

The question concerning the effect of chains formed in an electric field on the viscosity was first investigated by Bibik and Skobochkin⁸. When the mutual polarisation is taken into account and when the conditions of chain equilibrium are refined, the following expression for the change in viscosity in an electric field is found⁹.

$$\Delta\eta = \frac{P\epsilon}{\pi D} E^2 (F - g) , \quad (1)$$

where P is the volume fraction of the dispersed phase
 ϵ is the dielectric constant of the dispersion medium
 D is the displacement velocity or shear rate
 E is the field intensity
 F and g are constants introduced into the Kransy-Ergen formula for the energy of polarisation interaction of particles. The value of F and g is determined by the mutual distribution of particles and their polarisability.

This expression gives a change in viscosity which is eight times greater than that obtained in Ref 8. The value of the viscosity calculated from equation (1) was found to be 2.0 to 2.5 times the experimental value.

The discrepancy lies in the assumption of conducting, non-touching spheres suspended in a non-conducting medium, i.e. a model was examined for which the polarisation interaction would be a maximum for a given particle radius and field strength. In addition, the distance between particle surfaces was taken to be infinitely small, therefore the formula obtained overestimates the change in viscosity associated with the formation of chains not making contact with the electrodes. Further investigations in this direction must be aimed at refining the structural model, in particular taking into account the contact between chains and electrodes and including a broad spectrum of dielectric characteristics of the dispersed phase and the dispersion medium.

In conclusion it is necessary to indicate yet another new scientific approach germane to the influence of electric fields on the structure and rheological properties of dispersed systems. The discussion so far has been related to the formation of coagulated structures in electric fields. With the help of electric fields it is possible for condensation crystallisation structures to form¹⁷. The idea of such investigations is based on the fact that, upon formation of the dispersed phase in the process of cooling, a series of systems pass through a liquid crystalline state which is very sensitive to the effect of an electric field. The electrical effect on this stage of the formation of the system leads to an essential change in its structure and in its mechanical, optical and electrical properties. This opens up new possibilities for controlling the structure of dispersed systems and the change in its rheological properties.

The use of electric fields for changing the rheological properties of dispersed systems has only just begun. Further investigations along these lines are doubtless important, both from the point of view of the theory of structure formation in dispersed systems and for the practical application of electric fields in various areas of technology.

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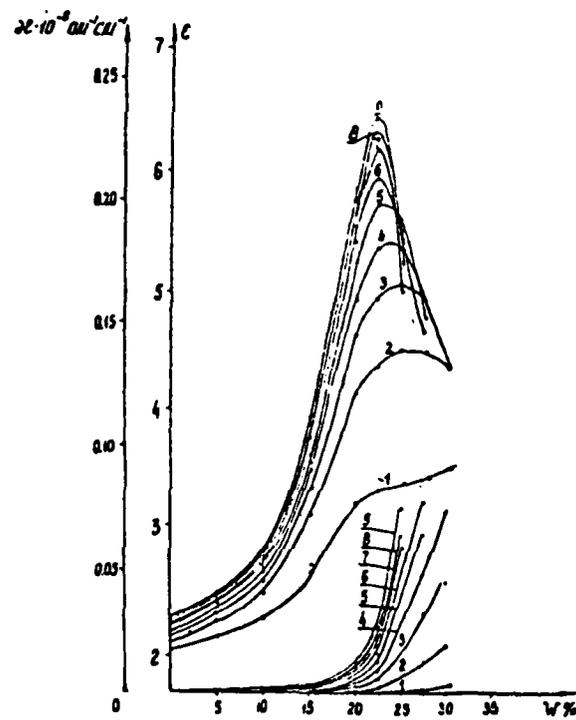


Fig 1 The dependence of ϵ and χ on the moisture content of the dispersed phase in a starch-vaseline oil suspension ($30s^{-1}$); $f = 50$ Hz; intensity of variable electrical field;

1-1; 2-4; 3-8; 4;12; 5-16; 6-20; 7-24; 8-28; 9-32; kV/cm

(χ in units of $\chi \cdot 10^{-8} \text{ ohm}^{-1} \text{ cm}^{-1}$)

Fig 2

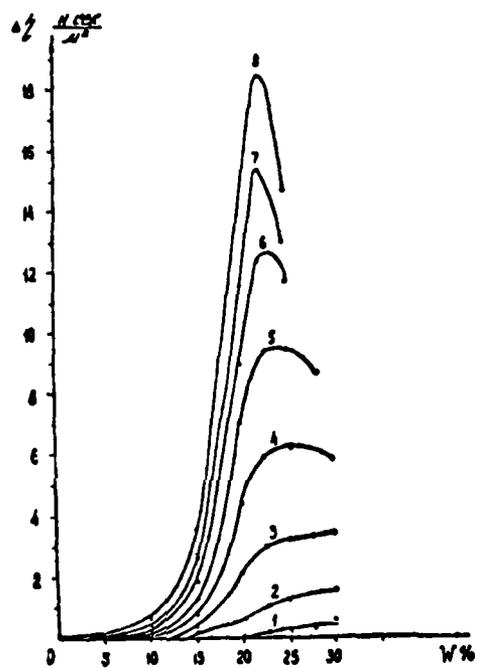


Fig 2 The dependence of the electrorheological effect on the moisture content of the dispersed phase for a starch-vaseline oil suspension (30s^{-1}); $f = 50\text{Hz}$; intensity of electrical field.

1-4; 2-8; 3-12; 4-16; 5-20; 6-24; 7-28; 8-32 kV/cm

($\Delta\eta$ in N s/m^2)

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