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The Rheological Behaviour and Thixotropy of Electrorheological Suspensions in Differing Temperature/Velocity Conditions

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R O Y A L   A E R O S P A C E   E S T A B L I S H M E N T

THE RHEOLOGICAL BEHAVIOUR AND THIXOTROPY OF ELECTRORHEOLOGICAL SUSPENSIONS
IN DIFFERING TEMPERATURE/VELOCITY CONDITIONS

[REOLOGICHESKOE POVEDENIE I TIKSOTROPIYA ELEKTROREOLOGICHESKIKH SUSPENZII
V RAZLICHNYKH TEMPERATURNO-SKOROSTNYKH REZHIMAKH]

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AUTHORS' SUMMARY

Measurements are made of the dynamic moduli of strain development G' and losses G'' in highly concentrated electrorheological aerosil and diatomite in transformer oil under low-amplitude strain conditions. The moduli G' and G'' are compared, do not depend on the angular frequency ω up to high values of ω, and increase with electric field strength, particle concentration and temperature.

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The practical application of the electrorheological effect (ERE) depends to a decisive extent on the mechanical, thermo-physical and structural properties of the electrorheological suspensions (ERS)[1]. It is convenient to categorise ERSs by their particle concentration ranges: very dilute ones – up to 1% by volume (for dielectric motors), medium concentrations (working medium for hydro-systems, dampers, vibrators) and high concentrations (for strengthening of components, for systems with controlled slipping).

ERS are made up of a non-conducting dispersion medium, particles of a disperse phase and sometimes a surface-active substance (SAS). The activity of the suspensions in the electrical field is chiefly dependent upon the specific polarizability and conductivity of the particles.

At the present time the most comprehensive study has been made of suspensions of moistened silica in a non-conducting medium (*eg* in hexadecane or in transformer oil)[2]. Dried silica powders are unsuitable for ERS, the low water content (*eg* for aerosil and diatomite 1-3 wt%) being insufficient to activate them. The ERE starts to appear only beyond a certain threshold water content adsorbed on the particles in suspension. With further increase in the quantity of adsorbed water the effective viscosity of the ERS in the electrical field at first increases sharply, and then falls rapidly. In the same way the strength of the interactions between silica-gel particles varies with varying water content[3].

In the silica suspensions examined the evidence of ERE was linked to the presence of adsorbed water on the particles. Instead of water, amines may be used (for example, diethylamine, ethylenediamine) or organic acid (for example, lactic or glutamic acid). These substances, which give the disperse phase of ERS specific properties, are called activators. Usually these substances are capable of forming hydrogen bonds. The surface of the silica should have a sufficient number of silanol groups. Stripping off the hydroxyl cover weakens the effectiveness of the disperse phase as compared with an identical quantity of physically adsorbed water. The following characteristics of ERS particles have been formulated as a result of studying large numbers of such hydrated dispersed materials: in a strong electric field separated particles behave like conducting ones: they become strongly polarized, distorting the initial homogeneous field, they are displaced toward the electrodes and they begin a movement back and forth along the field direction from one electrode to another. Such self-vibrating movement is called interelectrode oscillation. The moving particles are displaced along the field vector and are concentrated in certain regions of the gap. After a certain time they form into a small immovable bridge which connects the electrodes.

If the small bridge is subsequently mechanically destroyed, then the displaced particles, oscillating energetically, again form a compact structure. The ERS at rest in a powerful electrical field constitutes an anisotropic solid structure, filled with a disperse medium.

The particle oscillating across the gap transports charge from one electrode to the other. The magnitude of the borne charge is proportional to the size of the particle and the square of the field strength. In this respect a small sphere of silica gel (dielectric) with a water content of 5-6 wt% does not differ from a platinum one (a good conductor). However a small metallic sphere gives up (or receives) a charge at each contact; the overcharge characteristic of a silica gel one is much more complex: the small sphere contacts each electrode several times and breaks away from it[4,5]. The particles joined in the small interelectrode bridge ensure a small but indispensable conductivity and do not differ from dielectrics. The electric field compresses the particles in the region of the contacts between them and the electrodes so as to ensure transfer of the charge across the small bridges. At the same time the particles should have a certain number of mobile charge carriers and transfer paths for them. In the adsorption layer of water on the surface of the silicas the charge carriers are formed by electrons and protons. In order to create transfer paths for protons by a relay mechanism, the adsorption layer should be dense and continuous. In the case of aerosil this is approximately 3 monolayers of water (BET) at 20°C.

The increase in effective viscosity of the ERS is highest during that transfer of the charge over the surface of the particles which is not accompanied by a displacement of the activator itself (*eg* molecules of water lightly bonded to the surface). In a strong electric field any excess water migrates to the inter-particle contact region, yielding a parasitic conductivity (sometimes electrolysis is possible), which sharply reduces the cohesive forces in the small bridge.

Transfer of the charge by ERS particles is a basic aim of electrorheology but fairly difficult to achieve. The capacity to accept a large charge and the low electroconductivity of particles, together with the complex behaviour of their overcharging at the electrodes, are evidence of the presence of at least two types of interconnected charge carriers of differing mobility. The oscillating particle transports electrons through the interelectrode gap (the oscillation frequency and the transferable charge being independent of time, *ie* assuming the activator molecules are not electrolysed). The multistage overcharging of particles at the electrodes, the variation in current via the ERS with time, and the temperature

dependencies of the mechanical and electrophysical properties of the suspensions may all be explained by the limiting role of ion (proton) transfer. It is probable that at the places of contact of the particles in the small bridge a cut-off layer of the charge carriers occurs and that the structures of the ERS in a powerful electrical field demonstrate semi-conductor properties.

A variation in the temperature of the ERS affects the quantity and especially the mobility of the charge-carriers. Measurements were made of the effective viscosity and conductivity (η_{ef} and σ) in an electric field on a Rheotest-2 viscometer having a special measuring cylinder for suspensions of aerosil and diatomite with water, diethylamine, or polyethylenepolyamine as activators at temperatures between 293 and 363 K. The relationship of η_{ef} and σ to the quantity of activator and to temperature takes an identical form: with an increasing quantity of activator η_{ef} increases to a maximum, then falls rapidly, then increases exponentially.

Increase of the ERS temperature also causes an increase of η_{ef} to a certain maximum, the position of which depends upon the rate of increase of the conductivity σ. The value of η_{ef} at high temperatures may decrease in the case of a continuous increase in current, due to transfer of activator, or when using a low-molecular-weight activator, as a result of desorption of the latter from the surface of the particles. The use of an activator with a high molecular weight (*eg* polyethylene-polyamine - PEPA) renders its desorption impossible. Measurements of η_{ef} and σ for an ERS made with a PEPA content up to 30 wt% of the particles of aerosil at temperatures from room to 373 K have shown that the very same values of η_{ef} may be obtained by increasing either the quantity of PEPA or the temperature. The conclusion was drawn that the higher the application temperature of the ERS, the smaller should be the quantity of activator on the particles. Earlier[6] it was demonstrated that the mobility of the charge carriers is the limiting factor in transfer of the charge in ERS particles. The quantity of activator guarantees paths for the charge transfer; the electric field strength and the heat supply provide energy for it. The higher the electric field strength and the temperature, the more the optimal value η_{ef} is achieved at lower activator content.

Highly concentrated ERS, which are intended for operation in stabilizing appliances with controlled slip have a structured disperse phase both in the initial state and also in the electric field. The number of interparticle contacts depends on the particle concentration and the process of deformation. Conditions at the particle/liquid boundary, *ie* the nature of the disperse medium,

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the activator, the SAS and to a large extent the strength of the external electric field, affect the strength of the contacts. Any study of the rheological properties of thixotropic structured disperse systems is complicated by the dependence of these properties on time[7-9]. Some idea of the mechanical properties of highly concentrated ERS is obtained by measurement of their dynamic moduli under cyclic deformation.

Measurements have been made of the dynamic moduli of the gain (- G') and the losses (G") in suspensions of diatomite (between 10 and 60 wt%) and aerosil (between 13 and 27 wt%) in transformer oil at various temperatures. (Activators: 6.8 wt% of water was adsorbed on diatomite, and 30 wt% of polyethylene polyamine plus traces of water on aerosil. Measuring instruments: mechanical spectrometer DKhP-2, frequency range between 10^{-2} and 10^{2} Hz and vibrorheometer VR-74, frequencies between 10^{-1} and 1 Hz.)

Investigation of suspensions of aerosil showed that only low-amplitude cyclic deformation is measurable; at high amplitudes the oscillations of the internal cylinder of the vibrorheometer become non-sinusoidal. The strength of the ERS structure is considerably greater than the magnitude of its adhesion to the surfaces of the measuring instrument both with or without an electrical field.

The values of the gain - moduli G' and of the losses G" of the ERS without a field are comparable (Fig 1, curves 1). At lower concentrations of the disperse phase the value of G" is greater than G' , while with an increase in the concentration of particles G' begins to exceed G" . In aerosil suspensions up to a concentration of particles of the order of 20 wt%, G' and G" grow with increased angular frequency ω . However, at higher concentrations of particles this relationship of the moduli G' and G" to frequency is not followed. The application of the electric field strengthens the structure, which leads to a progressive increase in moduli G' and G" with increase of the field strength (Fig 1, curves 2 and 3). In the investigated suspensions of aerosil, G' and G" are independent of the frequency in an electric field. The greater the field strength, the less the gain of the moduli G' and G" per unit of field strength.

For diatomite suspensions the relationships of the elasticity moduli and the losses from frequency oscillations at various field strengths are characterized by two sections: a linear one with constant values of G' and G" and a nonlinear one, in which G' decreases, while G" increases with increase of ω . It may be assumed that the nonlinear section of the curves G'(ω) and G"(ω) is caused by disruption of the ERS structure, which starts at a certain value of the

angular mechanical frequency $\omega_n{}^{10}$. As is shown by calculations of the magnitudes $|G*| = \sqrt{(G')^2 + (G'')^2}$, at $\omega < \omega_n$, the values of $G*$ are close to the flow limits τ_0 , calculated from the flow curves.

The results obtained demonstrate that an increase in the electric field strength moves the threshold point for the break-up of the ERS structure to higher frequencies. The achievement under these conditions of the necessary level of visco-elastic characteristics of ERS requires the application of a mechanical field of greater intensity, and as a result, of greater losses of the mechanical energy.

Therefore, in the region of the non-disrupted structure the moduli G' and G'' of the ERS are independent of the angular frequency ω . Disruption of the structure occurs at lower values of ω in ERS of lower concentration and starts at $\lg \omega \geq 1.9$ in very highly concentrated ERS, which is characterized by a fall in G' and a rise in G'' with the frequency.

Analysis of the limits of the region in which the deformation is linearly dependent on the amplitude of the velocity of deformation γ_{max} is of great significance in the experiments. The observed decline in the curves $G'(\gamma_{max})$ and $G''(\gamma_{max})$, which reflects the nonlinearity of the rheological behaviour, appears much more markedly on the $G'(\gamma_{max})$ curve. This feature is generally valid for disperse and polymer systems alike, although the physical conditions of the process occurring may differ. Thus, unlike fluid polymer systems undergoing deformation, ERS during transition to the nonlinear region of deformation oscillation systems have been kept sinusoidal (to a first approximation), which enabled all calculations to use the relationships of the linear theory of visco-elasticity. With increase in electric field strength, the transition from linear to nonlinear deformation is observed at higher values of γ_{max} . The latter may be explained by the decisive contribution of the electrical forces in the formation of the structure. For suspensions of aerosil, the linear region of deformation was not attained in the range of frequencies investigated.

The values of G' and G'' with an increased concentration of particles without a field at first increase smoothly (up to 20 wt% of aerosil), then increase very abruptly (Fig 2, curves 1). The application of an electric field leads to a material strengthening of the structure of the ERS at lower concentrations of the particles, while the general nature of the relationships and the comparable magnitudes of G' and G'' are retained (Fig 2, curves 2 and 3). In the case of the softer diatomite disperse phase (skeletal residues of fossils), the modulus of elasticity is variable in the absence of a field. However, it

increases 1.5 to 2 times in heavily-loaded ERS with a 10-fold increase in field strength (from 0.06 to 0.6 kV/mm). A change in the diatomite content has a stronger effect on the mechanical properties of ERS in the concentration range 5-20 wt%. With compositions having a concentration $\overset{\bullet}{C}$ of more than 40%, the function $G'(\overset{\bullet}{C})$ begins to level off. In this concentration range the values for the loss modulus G'' continue to increase. This property of the concentration relationships of the dynamic characteristics of diatomite suspensions is obviously explained by the differing defectiveness of the structure at the maximum stage of its organization and, as a consequence, by a larger contribution to the values of the complex modulus $G*$ of the component, caused by the elastic interaction of the contacts and depending upon the number of particles in each element of the structure.

As shown above, heating to a certain range of temperatures intensifies the ERE. Measurement of the dynamic moduli G' and G'' at temperatures of 295-366 K demonstrated that in highly concentrated ERS heating does not in principle alter the character of the relationships of the dynamic mechanical properties (Fig 3). In the indicated temperature range the gain modulus G' exceeds the loss modulus G'', ie the elastic properties of the network of ERS particles in the electric field predominate.

The value G' increases with temperature more sharply than G''. In the investigated range of angular frequencies with low-amplitude deformation, G' and G'' are independent of the frequency up to 366 K. At a higher field strength $(1 \times 10^6$ V/m) a sharp increase of G' and G'' occurs with heating between 295 and 315 K, further heating resulting in a more modest increase in modulus. A lowering of the viscosity of the disperse medium is shown indirectly in the form of an increase of the elasticity of the structures due to a reduction of the loosening action on the fine layers of fluid between the particles.

The tests performed on the cyclical deformation of ERS at various temperatures and electric field strengths demonstrate that at low amplitude deformation the mechanical properties of ERS are completely determined by the properties of the interelectrode structures of the particles of the disperse phase.

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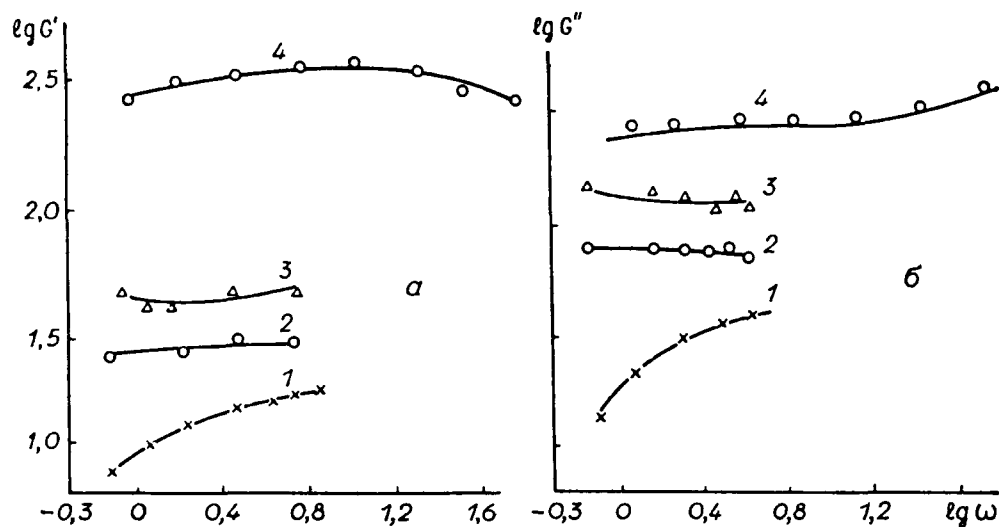
Fig 1   Relationship of the moduli of gain G' (a) and losses G"
        (b) to the angular frequency ω for suspensions of aerosil
        (30% PEPA) in transformer oil with a particle concentration
        of 18.7% (deformation amplitude 0.287):

        1 - without field; 2,3 - E = 0.5;   1 kV/mm respectively;
        4 - suspension of diatomite at 20% concentration,
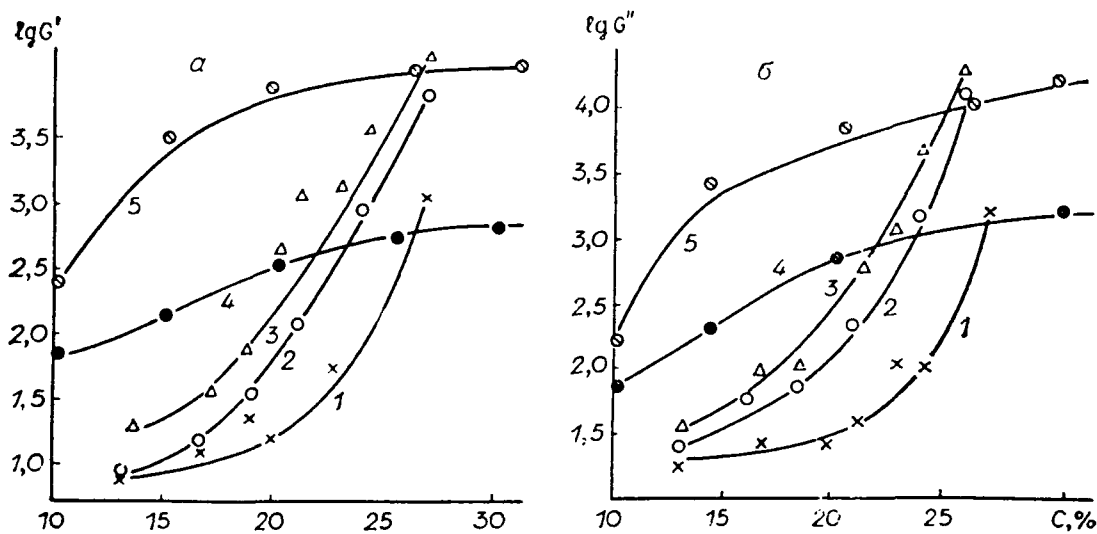   E = 0.13 kV/mm

Fig 2   Relationship of the moduli of gain G'   (a) and loss G"
        (b) to the filler content in suspensions of aerosil (1-3) and
        diatomite (4,5) in transformer oil (ω = 3.142):

        1 - without field; 2,3 - E = 0.5; 1 kV/mm; ω = 1; 4 - E = 0.1;
                  5 - E = 2.6 kV/mm

Fig 3
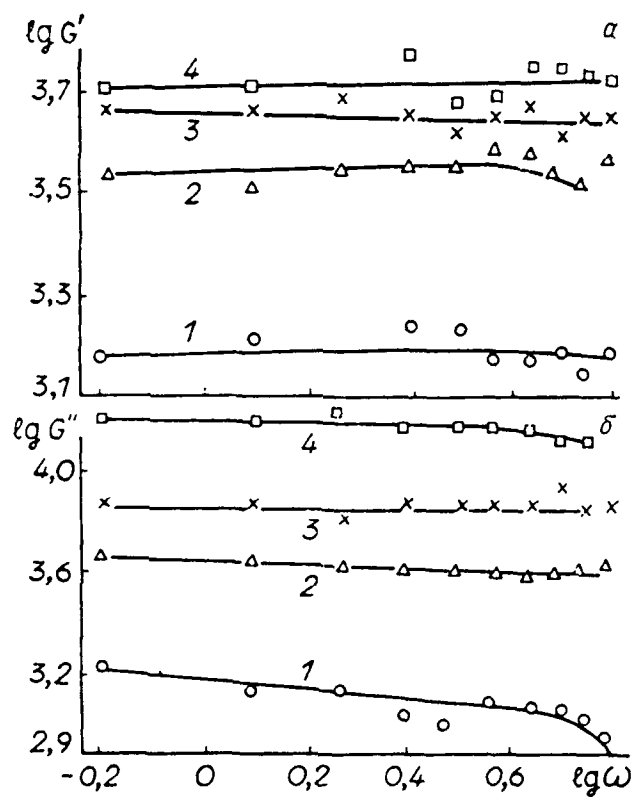

Fig 3   Relationship of the moduli of gain G' (a) and loss G"

(b) to the angular frequency ω   for suspensions containing
24.2 wt% aerosil at various temperatures;
K: 1 - 295; 2 - 315; 3 - 333; 4 - 366. Deformation amplitude
0.28, E ≈ 0.5 kV/mm