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THIXOTROPIC PROPERTIES OF ELECTRORHEOLOGICAL SUSPENSIONS IN CONTINUOUS DEFORMATION

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The article studies the peculiarities of the mechanical behavior of suspensions, structure-sensitive to electrical effects, in steady and transient regimes under continuous shear strain. It was found that the destruction of the structural carcass is preceded by an induction period, and the dependence of its time on the shear stress is described by the ratio between time to rupture and load for solids.

The authors of [1-4] showed that on the basis of the electrorheological effect (ERE) it is possible to devise various kinds of devices and elements: relays, regulators, adjusting mechanisms, dampers, braking devices, locking devices, stopping elements of hydraulic systems, electromotors, kilovoltmeters, resonators, etc. The use of the ERE for fastening mechanically unstable, pliable, weak materials to be machined, which are widely used in practice [5], is extremely important in machine and instrument construction.

At the Institute of Heat and Mass Exchange extensive research has been carried out for the last 35 years to determine the suitability of dielectric disperse systems to respond to electric impulses [6-9]. The investigations were complex and concerned a wide range of problems, aiming at the discovery of the inherent regularities of ERE and at working out physical concepts of its nature. It was shown that the magnitude and kinetic traits of the ERE are determined by the structural and rheological state of the medium. This state is characterized by the number of particles of the solid phase included in the interaction, by the detectability of the structure, the strength of the necks determined by a certain magnitude of mutual adhesion of particles and adhesion to the electrodes, and also by the length of their existence [7-9].

The regularities of the effect of an electric field on the processes of transfer in electrorheological suspenisons (ERS) were studied by many authors [6, 10-14], the research being carried out with the aid of capillary, rotational, and vibration viscometers. The following features were discovered: nonlinearity of the hydraulic characteristics of flow rate vs. head in an electric field, pseudoplastic behavior of the medium, an increase of the effective viscosity and of the modulus of elasticity by several orders of magnitude, the appearance of initial shear stress on the flow curves.

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All the hitherto known results were obtained for regimes of steady-state flow where the process of structure formation had been completed up to a degree determined by the ratio of the electrical and mechanical effects. Problems of the rheological behavior of ERS at the stage of formation and destruction of structures, and also under nonsteady conditions in the transition from one level of mechanical or electrical effect to another have so far not been studied in spite of their special urgency and importance for the control of hydraulic, robot, damping devices, etc. The investigation of regularities of the kinetics of ERS is also important on a general theoretical level for devising mathematical models of their behavior with a view to relaxation properties.

As object of study we chose typical ERS based on a customary dielectric dispersion medium, viz., desiccated transformer oil with high electrical resistivity and a readily available modification of silica, viz., diatomite. The siliceous hydrated skeleton of its particles consists of the finest variously shaped cells that are visible only with 10^5 times magnification. The diatomite was first subjected to wet crushing to obtain dispersity of 1-10 μm and specific surface ~27 m^2/g. Water was used as activator, and oleic acid as surfactant. We investigated suspensions with weight concentration of the solid phase from 2 to 60%.

A study of the processes of restructurization in ERS with low concentration (1-3%) showed that the particles of solid phase react in various ways to the switching on and off of an electric field. If the structure formation or "capture" of particles proceed rapidly (according to estimates of [15] within ~0.001 sec), then the process of their destruction extends over a long period of time (at least more than one day) on condition that there are external force effects, i.e., the process is actuated by Brownian motion only.

To study the time-dependent traits of the behavior of ERS with medium (>5%) and high (>20%) concentration, opaque in transmitted light, we use the rheological method whose information content was confirmed in the practice of many investigations, in particular in the mechanics of polymers [16]. The method consists in the following: changes of the structural state of a substance subjected to mechanical action (shear, torsion, continuous flow, cyclic action) are judged by rheological characteristics which are variable in time.

The investigations were carried out on a rotational viscometer operating in the regime of constant shear stress [17], designed at the Institute NKhS of the Academy of Sciences of the USSR and modified for use in constant electric fields with intensities up to 4 kV. The operative unit of the viscometer is a motor-driven rotating bell situated in an annular gap at the depth h. Together with the cylinders of the stator it forms two gaps with size Δr = 2.2 mm, which are small compared with the radius of the bell R = 50.2 mm. Such a design ensures uniform shearing rate on both sides of the bell-shaped rotor. Voltage is supplied to the rotor through a sliding contact from a stabilized current source, the stator is grounded. Electrical uncoupling of the stator from the rotor is effected with the aid of insulating pads.

The bell of the operative unit is mounted on the shaft of a biphase induction motor with linear control characteristic from which it receives its torque. In the motor there are small precision bearings in which the friction moment is small in comparison with the specified torque. The absence of spring elements makes it possible to reduce the error at high shear stresses and to measure the shearing rate and the effective viscosity in the range $\dot{\gamma} = 0.04\text{-}10^5\ \mathrm{sec}^{-1}$, $\eta = 0.001\text{-}10^7$ Pa·sec, respectively, at shear stresses $\tau = 0.1\text{-}10^2$ Pa. A mirror reflecting the laser beams is mounted on the shaft. When the mirror is turned, the beam strikes a photoresistor mounted on the saddle of the recording potentiometer. A signal in the electric circuit of the potentiometer causes the saddle to move, and the development of strain vs. time, i.e., the creep curves, are recorded on the oscillogram.

The method of calculating shearing rates and viscosity from the creep curves, and also of determining the absolute values of shear strain and stress was described in detail in [17]. The experiments were carried out at room temperature T = 18 ± 2°C.

In the first series of experiments the investigated medium was placed in the gap of the working cell and was held in an electric field for a certain time t_E, then the field was switched off and at the same time a minimal mechanical stress τ_0 was applied in a "step," leading within some fractions of a second to shear of the medium ascertained by the laser beam. Such experiments were also conducted without switching off of the field.

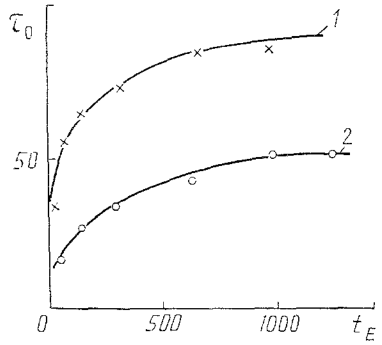

Fig 1. Dependence of the minimal shear stress inducing flow of the ERS in an electrical field with intensity E = 1.5 kV/mm (1), and with the field switched off (2) at the instant of shear on the time of preliminary holding in the field, C = 20%, τ_0, Pa; t_E, sec.

Figure 1 presents the data for ERS with C = 20% situated in an electric field with intensity E = 1.5 kV/mm. It can be seen that with longer holding time in the electric field t_E prior to shear the carcass of the suspension becomes stronger and τ_0 increases. However, when the time exceeds 600 or 700 sec, τ_0 tends to a constant value which indicates that the formation of the carcass has ended. Similar results were also obtained with other values of E and concentrations of ERS. It was found that the characteristic time of attaining saturation on the dependence τ_0–t_E decreases with increasing concentration of the suspension and increasing electric field intensity. This time is hundred thousands of times longer than the time of primary response of particles of the disperse phase of the ERS to an electric impulse (~10^{-3} sec) [15]. It can be seen that in the suspension, together with an instantaneous reaction (rotation, capture of particles of the disperse phase), there proceeds some lengthier structurization leading to increased strength of the ERS. Figure 1 also shows the dependence τ_0–t_E for the suspension after the electric field has been switched off, at the instant of application of the shear stress. This dependence is analogous to the curve τ_0–T_E with the electric field switched on. It characterizes the strength of the structural carcass of the ERS which is retained after the field has been switched off. The difference of τ_0 in both types of experiment expresses obviously the contribution of electric forces of interaction to the strength of the structural carcass. With $t_E \geq 120$ sec, this difference attains a constant value equal to ~40 Pa. It decreases with decreasing E. Experiments showed that with E = 0.75 kV/mm its value is five times smaller than in the case E = 1.5 kV/mm. Except for cases specially pointed out, all the results presented below were obtained after completion of the formation of the structural carcass of ERS in an electric field.

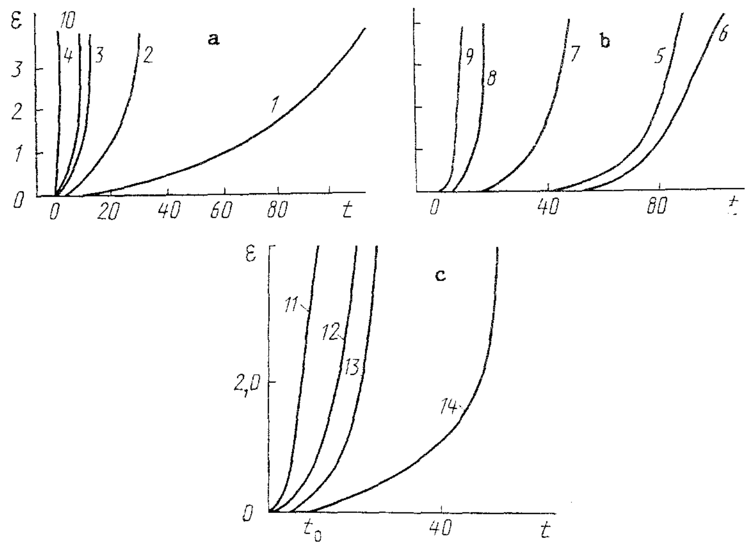

Fig. 2. Development of strain of ERS in time upon change of mechanical stress (a, b) and voltage (c): 1, 10) τ = 0.48 Pa; 2) 1.12; 3, 11-14) 2.24; 4, 5) 3.0; 6) 2.6; 7) 3.47; 8) 4.46; 9) 5.598; 10) E = 0; 11) 0.05 kV/mm; 12) 0.125; 13, 1-4) 0.25; 14) 0.5; 6-9) 0.75, C = 40%, a, c) t_E = 300 sec, b) 60; t, sec.

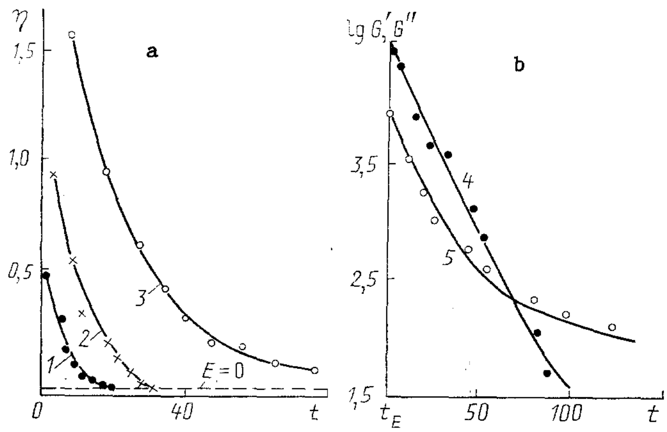

Fig. 3. Time dependence of the effective viscosity of ERS in continuous shear (a) and of the modulus of elasticity G' and of the loss modulus G" (b) in periodic strain after the field with the following intensity has been switched off: 1) E = 0.12 kV/mm; 2) 0.25; 3) 0.5; 4, 5) 2.6; a) τ_0 = 2.24 Pa, C = 40%, t_E = 300 sec; b) C = 60%, γ_0 = 0.027, f = 0.16 Hz; 4) σ', 5) σ". η, Pa·sec; G', G", Pa.

The rheological investigations in transient regimes of continuous strain were conducted in the second series of experiments after switching off of the electric field at the instant of application of the mechanical load. We studied the development of strain in time with different values of the shear stress and magnitudes of E of the preliminarily applied electric field on the example of ERS containing 40% diatomite.

Figure 2a demonstrates the development of strain with E = 0.25 kV/mm and an increase of τ from 0.75 to 3.0 Pa upon the transition from curve 1 to curve 4, and also of ε(t) with E = 0 and τ_0 = 0.75 Pa (curve 10). Figure 2b presents the data of ε(t) with E = 0.75 kV/mm and an increase of τ from 2.6 to 6.0 Pa upon the transition from curve 6 to curve 9. Figure 2c shows the results with τ = 2.4 Pa and the increase of E from 0.05 to 0.5 kV/mm upon the transition from curve 11 to curve 14. It can be seen that with zero value of E, the dependence of strain on time is of a linear nature right from the beginning (curve 10). In the remaining cases we find at first slow, then accelerated development of strain in time which manifests itself the more strongly, the higher the shear stress and the smaller the value of E is. With sufficient duration of strain, which is the longer, the smaller the shear stress and the larger E is, these curves change into straight lines; this corresponds to complete destruction of the structural carcass formed under the effect of the electic field. The slope of these straight-line segments does not depend on the value of E (Fig. 2c). It is determined solely by the shear stress and increases as ε increases (Fig. 2b). We note that the intensity of the action of the preliminarily applied field [its duration (Fig. 2b) or intensity (Fig. 2a)] did apparently not suffice for the completion of the formation of the structural carcass of the ERS.

From the data of Fig. 2 we calculated the effective viscosities and their change in dependence on time, with fixed shear stress (τ = 2.24 Pa) induced at the instant the electric field was switched off (Fig. 3a). It can be seen that these viscosities decrease rapidly in time because of the enhanced disintegration of the structural carcass of the ERS. Here, the rate of decrease of viscosity and the degree of destruction of the structural carcass are the lower, the larger the value of E of the preliminarily applied electric field is. For instance, with E = 0.125 kV/mm the viscosity of ERS after application of τ for ~25 sec already attains the viscosity of ERS not subjected to the effect of an electric field and flowing in this case practically like a Newtonian liquid. On the other hand, with E = 0.5 kV/mm a noticeable difference of viscosities is retained even for 60 sec. Analogous regularities were also obtained under conditions of dynamic deformation by the method of [18] after the electric field had been switched off. The changes of the modulus of elasticity G' and of the loss modulus G" in time, at frequencies corresponding to the range of the carcass not destroyed in the field, are shown in Fig. 3b. It was noted that the moduli G' and G" decreased rapidly by about two orders of magnitude already within the first minute of strain.

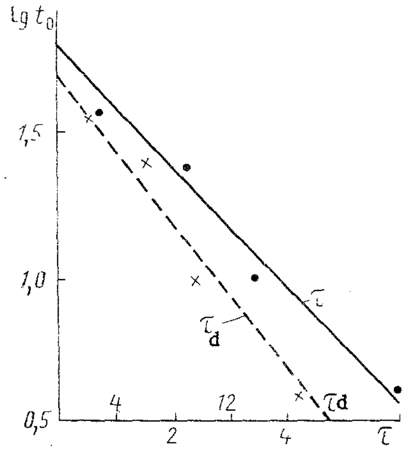

Fig. 4. Dependence of endurance on mechanical stress with an electric field with intensity E = 0.75 kV/mm, t_E = 60 sec, C = 40% acting. t_0, sec.

The obtained results also show that with increasing intensity of the preliminarily applied electric field the strength of the structure of the ERS increases and the drop of G' and G" in time slows down, i.e., the structural carcass disintegrates at a decreasing rate. These data indicate that the mentioned level of the mechanical load the medium retains its properties when the frequency of the acting electric field is higher than $1/t_p$, where t_p is the time determined from the corresponding dependence G', G"—t.

Reverting to Fig. 2, we have to point out that in case of zero or low value of E (less than 0.25 kV/mm) (or insufficient time of preliminary action of the electric field) the ERS becomes fluid within the limits of the experimental time scale (0.1 sec) as soon as shear stress is applied (see, e.g., Fig. 2a). However, in the remaining cases flow is preceded by the induction period t_0 within which the ERS is not subjected to appreciable strain. At that t_0 increases with decreasing τ and increasing E. Delayed development of fluidity upon load application had been observed earlier in some disperse systems of organic origin (bitumens), and also in the case of chemical flow of elastomer with sparse three-dimensional network [19, 20].

It can be seen from Fig. 4, which was plotted according to data of Fig. 2 and other analogous results, that the dependence of t_0 on τ can be described by a straight line in semilogarithmic coordinates. The same applies to the dependence of endurance Θ, i.e., the time from the instant of application of the load P to failure of the material, on the magnitude of P in the case of low-molecular and polymer bodies [21, 22]. At that Zhurkov's equation

$$\Theta = \Theta_0 \exp\left(\frac{U - \alpha P}{RT}\right) \tag{1}$$

is satisfied, where $\Theta_0 = 10^{-12}$-10^{-13} sec; U is the activation energy of the process of failure; α is a structure-sensitive parameter; T is the temperature, K; R is the universal gas constant. Obviously, to the parameter t_0 we can also ascribe the meaning of endurance in connection with the onset of brittle failure of the structural carcass of the ERS and the transition to the fluid state. In that case, with $P = \tau S$, the calculation of the dependence in Fig. 4 makes it possible to determine the activation energy of failure of the structural carcass. But here it must be taken into account that the applied shearing force P should not be calculated for the entire area of the electrodes S but only for the area of contacts of the structures S_d (the area of the bases of the necks). For 40% ERS a simple calculation taking the volume content of the disperse phase into account yields

$$\varphi = \frac{40/d_d}{40/d_d + 60/d_o} = 0{,}21,$$

where d_d, d_0 is the specific weight of diatomite and of transformer oil, respectively. Then

$$S_d = \varphi^{2/3} = 0{,}53, \ \tau_d = \frac{P}{S_d} = 2{,}83\tau.$$

The values of the activation energy U, with a view to this correction for E = 0.75 kV/mm, are equal to 77-80 kJ/mole (in dependence on the chosen magnitude of Θ_0). When we go over to the data obtained for E = 0.5 kV/mm, we obtain U several per cent lower. Such values of activation energy are close to the energy of intermolecular bonds. The destruction of the carcass of ERS is apparently connected with overcoming the forces of intermolecular interaction ensuing both from contacts of diatomite particles and their interaction through molecules of the activator and of the surfactant. On the ther hand, the values of τ in Fig. 4 can be ascribed the meaning of yield points depending on the time of application of the load, as had been assumed earlier on in the case of bitumens and the chemical flow of cross-linked elastomers with sparse three-dimensional network [19, 20]. In this connection there arises the question of the meaning of yield points established from data in strain with constant shearing rate. Since the duration of action of the load is inversely proportional to the strain rate, these values of yield points have to have the meaning of some effective parameters depending on the chosen range of low shearing rates $\dot{\gamma}$. However, in view of the exponential nature of the dependence of the duration of action of the load on its magnitude, we have to expect a weak dependence of these parameters on the strain rate. An approximate calculation shows that a change of $\dot{\gamma}$ from a few units to some tenths changes the magnitude of the effective yield point by 10-30% only. Besides, according to present-day notions [22], Eq. (1) does not apply at high values of Θ depending on the nature of the material, and the magnitude P assumes the sense of strength not depending on the duration of loading of the material. The same is obviously also bound to occur with the magnitude of the yield point determined in the region of very low values of $\dot{\gamma}$.

Thus, the results that were discussed show that it is necessary to take the time factor into account when considering the peculiarities of the mechanical behavior of ERS. Its role is determined by the existence of a structural carcass in ERS which forms in dependence on time under the effect of an applied electric field. The strength of this carcass is determined both by intermolecular interaction in ERS and by the Coulomb forces acting between particles of the disperse phase in the presence of an electric field. The ERS with the forming structural carcass behave like nonlinear plastic bodies with a yield point that may depend on the duration of the load on the suspension. Switching off of the electric field weakens the structural carcass and leads to its rapid destruction, already at small strain amplitudes.

NOTATION

$\dot{\gamma}$, shearing rate, sec^{-1}; E, electric field intensity, kV/mm; τ_0, initial shear stress; G', modulus of elasticity, Pa; G", loss modulus, Pa; ε, strain; Θ, t_0, endurance, sec; U, activation energy, kJ/mole.

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