Elastoviscoplastic behavior model of electrorheological fluids in various deformation modes

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Abstract. The results of investigations of rheological properties of electrorheological fluids in various deformation modes: oscillatory, continuous flow, constant applied loading – are presented in the article. A generalized rheological model on the basis of a consecutive combination of viscoplastic, elastic and two viscoelastic elements with differing relaxation times, taking into account time dependence of retarded elastic deformation, is suggested.

1. Introduction
Numerous rheological investigations in different deformation conditions of the broad spectrum of electrorheological fluids (ERF) compositions with the components having the various chemical nature and structural characteristics have revealed distinctive features of mechanical behaviour, characteristic for the materials possessing properties both of Newton fluids, and quasi-solid bodies. Rheological behaviour of ERF in stationary or quasi-stationary conditions under the influence of large viscoplastic strains is most studied. In connection with complexity of the structural phenomena the elementary linear and power phenomenological models are applied to their description [1 – 5]. They do not allow to show real relaxation processes at change of ERF properties under the influence of shear forces in time, that excludes possibility of their adequate application in non-stationary dynamic conditions. Now there is no the conventional theory of ERF deformation based on basic researches and considering rheological, physical and strength properties of its structural skeleton and layers of dispersion medium or systems as a whole on macro- and micro-levels.

Depending on type of the structure created by the field, its pre-history, loading and additional external conditions (temperature), elastic, viscous or plastic properties can prevail in ERF. Features of process of deformation will be defined by a complex of characteristics of structural elements of a material: their arrangement in the bulk, character of their molecular and polarization interaction, distribution of the internal stresses depending on presence of heterogeneities, etc. Character of deformation is defined also by time of action of force and the electric field in comparison with time relaxation of medium. Taking into account the analysis of relaxation phenomena for various elastoviscoplastic materials the modeling analysis which is based on comparison of deformation behavior of real bodies and idealized mechanical models is used. As a whole, ERF is among complex structural materials and therefore they can be presented in the form of multi-element rheological elastoviscoplastic models consisting of four and more elements.

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The choice of adequate model by means of mathematical modeling of the mode of deformation of medium is based on use of results of rheological tests of specific compositions of ERF which allow to reveal the main features of mechanical behavior of medium. The object of present work is construction of the generalized model of viscoelastic behaviour ERF at various deformations on the basis of measurement of viscoelastic characteristics and the time mechanical response in non-stationary shear conditions.

2. Materials and equipment
The dispersion of goethite (goethite is iron needle ore \(\alpha\)-FeOOH; rhombic crystal system, rhombobipyramidal kind of a symmetry) with content of a solid component 58 wt.% with the addition of 0.6 wt.% of a surfactant (monooleate of glycerin) was used for investigation of rheological properties. Transformer oil served as dispersion medium.

Rotational rheometer "Physica MCR301" by Anton Paar with measuring cell of plate – plate type with diameter of plates 50 mm and gap between plates of 1 mm and cell of coaxial cylinders type (diameter of inner cylinder is 26.7 mm, of outer one is 28.9 mm, gap between cylinders is 1.13 mm) was used for rheological experiments.

3. Experimental technique
Tests of sample were carried out in following deformation modes:

1) harmonic oscillations at fixed angular frequency \(\omega=6.283 \, \text{s}^{-1} (f=1 \, \text{Hz})\) with a variation of amplitude of oscillations from 0.01 to 100 % that has allowed to define components of complex shear modulus (storage modulus \(G'\) and loss modulus \(G''\)) depending on deformation amplitude, as well as limits of linear viscoelasticity area;

2) harmonic oscillations in a range of angular frequencies \(\omega\) from 0.314 to 314 \(\text{s}^{-1}\) (a range of frequencies \(f\) from 0.05 to 50 Hz) at small amplitudes (0.1 %) that corresponds to linear viscoelasticity area;

3) constant shear rate in a range from 0.00001 to 1000 \(\text{s}^{-1}\), that allowed to get flow curves on the measured values of shear stress;

4) tests for creep and relaxation: constant shear stress \(\tau\) was applied, it was maintained some time, then \(\tau=0\) was established, dependence of strain \(\varepsilon\) on time was fixed (before shear the electric field with the same intensity as at shear was applied to ERF during 5 min for completion of processes of internal structure realignment). Electric field intensity \(E\) varied in range up to 2 kV/mm.

4. Results and discussion
ERF on a goethite basis under investigation show elastic properties in the range of strains to 1 – 2 % without electric field, at larger strains a transition into viscoplastic condition occurs. In the electric field the elastic limit is shifted to area of 0.1 %, the range of resiliency decreases by order, but at that the elasticity modulus increases more than by 3 order (to 1.2 MPa) at applying of electric field of 2 kV/mm (figures 1, 2). The structure becomes more rigid, however level of strain for transition to irreversible flow decreases. The components of complex shear modulus practically do not depend on deformation frequency in the field in the range of frequencies 0.05 – 50 Hz. In the absence of electric field loss modulus grows with frequency (from 27 to 250 Pa at \(E = 2 \, \text{kV/mm}\)).

Examples of creep curves are given in figures 3, 4. Initially there is an instantaneous elastic deformation, further deformation increases to some value, after loading removal there is an elastic restoration on value of instant deformation and relaxation process begins. After some time interval deformation decreases to residual value.

Rheological behavior of ERF can be described by complex model (figure 5), containing two viscoelastic elements of Kelvin – Voigt, having essentially differing characteristic relaxation times. This generalized model is seems to the most preferable to the analysis though the issue about kind of rheological model cannot be considered as closed, and the made choice as strictly founded. Further we check its adequacy to experimental data.
In the context of present work we confine oneself to research of only viscoelastic properties of ERF at various, but constant in time shear stresses and electric field intensities. The choice of values of shear stress was made so that they did not exceed yield stress for given level $E$. For determination of value $\tau_0(E)$ the experimental technique of linear growth of shear stress was used.

The received dependence $\tau_0(E)$ is shown in figure 6 and well approximated by exponential dependence (a continuous line)

$$\tau_0 = \tau_{00} \exp(-E/E_0),$$  \hspace{1cm} (1)

at values of $\tau_{00} = 8.93$ Pa, $E_0 = 0.35$ kV/mm.

Appreciable more precise definition of $\tau_0(E)$ at $E > 1.5$ kV/mm is provided with small adjustment of dependence (1) in the form of

$$\tau_0 = \tau_{00} \exp\left(-\frac{E}{E_0} - \frac{E}{E_1}\right)$$  \hspace{1cm} (2)

at values of $\tau_{00} = 8.93$ Pa, $E_0 = 0.35$ kV/mm, $E_1 = 70$ kV/mm (hatch line in figure 6).

Approximately and indirectly about structure of rheological model we can judge on correlation between values of relative deformation at loading stage $\varepsilon^l(t)$ and unloading stage $\varepsilon^u(\Delta t)$, where
Δt = t − t_u, t_u is absolute value of the moment of time at which loading was removed (unloading and retardation of system have begun). In figures 7(a)-(d), corresponding to various levels of loading, the correlation between ε^{II}(Δt) and ε'(t) at Δt ≡ t is illustrated by insets in each figure. With increase in value of constant loading relation between ε^{II}(Δt) and ε'(t) at Δt ≡ t comes nearer to linear as though at the larger τ action only of one element of Kelvin – Voigt starts to prevail. Continuous lines in figure 7 show result of approximation of experimental data with generalized (two-tier) model of Kelvin – Voigt at τ=const, i.e.

\[ ϵ^{I}(t) = ϵ_0 + ϵ_1(1 - e^{-t/θ_1}) + ϵ_2(1 - e^{-t/θ_2}), \]

\[ ϵ^{II}(Δt) = ϵ_{u1}e^{-Δt/θ_1} + ϵ_{u2}e^{-Δt/θ_2}, \]

\[ ϵ_k = \frac{τ}{G_k}, k=0, 1, 2; \ θ_k = \frac{η_k}{G_k}; \ ϵ_{uk} = (1 - e^{-τ_u/θ_k}), k=1, 2. \]

Adjustment of calculated values of relative deformation to the measured values was carried out by minimization of function

\[ F(ϵ_0, ϵ_1, ϵ_2, θ_1, θ_2) = \sum_n (ϵ_{exp}(t_n) - ϵ_{calc}(t_n))^2 = \text{min}. \]

Here indexes exp and calc designate accordingly measured and calculated values of deformations. Adjustment to experimental data was carried out on all curve of deformation. By separate consideration of curves ε^{II}(Δt) and ε'(t) received parameters of model (3) – (4) would be different for each stage.

Results of data treatment without field are presented in figures 8 and 9. We will note approximate proportionality between ϵ_k and τ, argumentative of weak dependence of mechanical properties of ERF on value of shear stress at small deformations in the absence of electric field.
Figure 7. Deformation curves of ERF at different values of shear stress in the absence of electric field: (a) $\tau = 1$ Pa, (b) 1.5, (c) 2 and (d) 2.5.

Figure 8. Dependence of parameters of model (3) – (4) on shear stress at $E = 0$ kV/mm.
Average values of mechanical constants are equal 631, 1997, and 898 Pa for $G_0$, $G_1$, and $G_2$, and also 19.6 and 258 kPa⋅s for $\eta_1$ and $\eta_2$ with the maximum relative deviations from average value 17, 14, 20, 26, and 30%.

In the presence of electric field it is possible to observe both quantitative and qualitative changes in behavior of deformation curves. In case of independence of mechanical properties on $\tau$ all curves $s(E, \tau, t) = \varepsilon/\tau$ at the same level of intensity of field $E$ and different $\tau$ must coincide (as it follows from (3)–(4)). However it does not occur. It is possible to note only essential rapprochement between curves $s(E, \tau, t)$ at increase in intensity of electric field to 2 kV/mm. Among the considered levels of intensity of electric field only the data at $E=2$ kV/mm are close to a condition of constancy of mechanical properties of ERF in time. Expressions (3) and (4) can be right only for this field intensity. We give the further analysis of curves of deformation for this reason only for case $E=2$ kV/mm.
Figure 11. Viscoelastic characteristics of ERF at $E=2$ kV/mm according to model (3) – (4).

Final results of treatment of experimental data are illustrated in figures 10 and 11. They show that with increase of loading at a set value of electric field intensity rigidity of Hook element increases, and rigidity of units of model Kelvin – Voigt decreases. Internal viscosity of these units with growth of $\tau$ also increases.

5. Conclusion
Thus, on the basis of the model analysis it is suggested multi-element composite rheological model for calculation of mechanical behaviour of electrorheological fluids, considering a complex of structural properties and loading conditions. Its further development will allow to establish correlation between structural-mechanical properties (elasticity, viscosity, plasticity) and composition, physical and chemical structure of electrorheological fluids in the external electric field, will make possible calculation of flow of electrically controlled media in technical devices (the adaptive shock-absorbers, fixing devices).

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