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MESOSCOPIC EQUATION OF STATE OF POLYMER SYSTEMS AND DESCRIPTION OF THE DYNAMIC CHARACTERISTICS BASED ON IT

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A rheological constitutive relation used for description of the components of the dynamic shear modulus in superposition of small oscillating vibrations on stationary shear flow has been obtained based on the dumbbell model.

Worldwide production of polymers has long been developed much more intensely than the production of such traditional structural materials as cast iron and aluminum. Polymer materials (fibers, plastics, and rubbers) are as popular and necessary in our everyday life as the existing materials (metals and nonmetals) from small molecules. Polymers are used as independent structural materials, not as substitutes.

At the same time, to obtain products from polymer materials one must process them in a viscous-fluid state. However, the behavior of polymer materials significantly differs from the behavior of the traditional objects of study of physicists — liquids and solid bodies — which is attributed to the complexity of the structure of polymer materials which combines the order of solid bodies and the chaos of liquids. By virtue of the distinctive features of their structure, polymer materials possess unique properties: the capacity for large irreversible strains in the state of high elasticity and also hardness and fluidity as functions of the strain time (frequency). On this basis, it may be inferred that study of the motion of a polymer system in different units of technological equipment is a very important practical problem; its solution requires that a rheological constitutive relation enabling one to describe the rheological (mechanical) properties of a polymer be constructed.

We know of two methods of construction of the rheological equation of state: a phenomenological method and a mesoscopic, or statistical, one. In the first case, the theory of motion of macroscopic bodies is constructed on the basis of the general (found from experiment) regularities; in the second case one describes the object, allowing for the molecular structure of a substance in a certain approximation and for fairly complex processes of intermolecular interaction. Thereafter, using probabilistic methods, one introduces characteristics averaged over the ensemble of all kinds of realizations; they are identified with the quantities determined by experiment. The rheological constitutive relations obtained by any of these approaches or rheological models must be checked for correspondence to the actual properties of polymer liquids.

The basis for the mesoscopic approach to description of the dynamic characteristics of polymer media is formed by the equations of macromolecular dynamics to write which one uses model representations. If only the slowest relaxation processes in the case of flow of a polymer medium are considered, the model of a "dumbbell" is very convenient: two Brownian particles linked by an elastic force; the equations of macromolecular dynamics for it have the form [1]

$$\begin{gathered} -2T\mu\,(\mathbf{r}_i^1-\mathbf{r}_i^2)+\zeta_{ij}\,(\nu_{jk}\mathbf{r}_k^1-\boldsymbol{\omega}_j^1)-T\frac{\partial}{\partial\mathbf{r}_i^1}\ln W=0\,, \\ -2T\mu\,(\mathbf{r}_i^2-\mathbf{r}_i^1)+\zeta_{ij}\,(\nu_{jk}\mathbf{r}_k^2-\boldsymbol{\omega}_j^2)-T\frac{\partial}{\partial\mathbf{r}_i^2}\ln W=0\,. \end{gathered} \tag{1}$$

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With allowance for the induced anisotropy, the anisotropy of mobility of the macromolecule in question can be prescribed using the tensor coefficient of friction [2]:

$$\zeta_{ik} = \zeta \left(\delta_{ik} + 3\beta \left(a_{ik} - \frac{a_{ll}}{3} \delta_{ik} \right) + \kappa a_{ll} \delta_{ik} \right)^{-1} .$$

To find and investigate the analytical solutions of system (1) we conveniently pass to normal coordinates:

$$\rho_i = (\mathbf{r}_i^2 - \mathbf{r}_i^1)/\sqrt{2} , \quad \rho_i^0 = (\mathbf{r}_i^2 - \mathbf{r}_i^1)/\sqrt{2} ; \tag{2}$$

then the diffusion rate of a bead in the coordinate system (2) takes the form

$$\psi_i = \frac{1}{\sqrt{2}} (\omega_i^2 - \omega_i^1) , \quad \psi_i^0 = \frac{1}{\sqrt{2}} (\omega_i^2 + \omega_i^1) .$$

From Eq. (1), we obtain

$$\psi_j = \nu_{jl} \rho_l - 4T\mu \zeta_{jl}^{-1} \rho_l - T\zeta_{jl}^{-1} \frac{\partial}{\partial \rho_l} \ln W , \quad \psi_j^0 = \nu_{jl} \rho_l^0 - T\zeta_{jl}^{-1} \frac{\partial}{\partial \rho_l^0} \ln W . \tag{3}$$

After determination of ψ_i and ψ_i^0 we can write the diffusion equation for the distribution function on the basis of the Smoluchowski equation:

$$\frac{\partial W}{\partial t} + \nu_{jk} \rho_k \frac{\partial W}{\partial \rho_j} - 2T\mu \zeta_{jk}^{-1} \rho_k \frac{\partial W}{\partial \rho_j} - 4T\mu \zeta_{jj}^{-1} W - T\zeta_{jk}^{-1} \frac{\partial^2 W}{\partial^2 \rho_j \rho_k} = 0 .$$

It describes the diffusion of dumbbell beads relative to each other and after its substitution into (3) enables us to obtain relaxation equations for correlation moments:

$$\frac{d}{dt} \langle \rho_i \rho_k \rangle - \nu_{il} \langle \rho_l \rho_k \rangle - \nu_{kl} \langle \rho_l \rho_i \rangle + \frac{\zeta_{il}^{-1}}{2\tau} \langle \rho_l \rho_k \rangle + \frac{\zeta_{kl}^{-1}}{2\tau} \langle \rho_l \rho_i \rangle + \frac{\zeta_{ik}^{-1}}{2\mu\tau} = 0 .$$

In this case the shape and orientation of dumbbells in the flow can be characterized by the following tensor:

$$a_{ik} = \frac{\langle \rho_i \rho_k \rangle}{\langle \rho^2 \rangle_0} - \frac{1}{3} \delta_{ik} ,$$

whence we obtain, after transformations, the rheological constitutive relations

$$\sigma_{ik} = - p\delta_{ik} + 3 \frac{\eta_0}{\tau_0} a_{ik} , \tag{4}$$

$$\frac{d}{dt} a_{ik} - \nu_{ij} a_{jk} - \nu_{kj} a_{ji} + \frac{1 + (\kappa - \beta) a_{jj}}{\tau_0} a_{ik} = \frac{2}{3} \gamma_{ik} - 3 \frac{\beta}{\tau_0} a_{ij} a_{jk} . \tag{5}$$

System (4) and (5) contains four unknown parameters η_0, τ_0, β, and κ. The dimensional parameters η_0 and τ_0 have been evaluated by the formulas

$$\eta_0 (c, M) = \eta_0 (c^*, M^*) \left(\frac{c}{c^*} \right)^{4.5} \left(\frac{M}{M^*} \right)^{3.4} , \quad \tau_0 = \frac{\eta_0}{nT} .$$

It is noteworthy that the rheological model (4) and (5) was obtained earlier as a zero approximation of the available small parameters, related to the internal viscosity and the environmental aftereffect, and is a generalization of the Pokrovskii–Vinogradov structural-phenomenological model. This model can be used for numerical investigation of the behavior of polymers under difficult conditions of their straining; such conditions are characteristic of technological processes of processing: outflow of a polymer jet from a circular pipe, flow with a free surface, etc.

Based on the rheological model (4) and (5) obtained, we numerically investigated in [2–4] stationary viscosimetric functions: the viscosity, the first and second difference of normal stresses in simple shear, and the viscosity in uniaxial tension as functions of a constant velocity gradient. Also, we found the ratio of the stationary viscosity in uniaxial tension to the stationary viscosity in shear as a function of the first invariant of the tensor of additional stresses. In [5, 6], we calculated flows in a circular pipe, which enables us to refine corrections to the Poiseuille law, and in cylinders with a rotary end and a free surface, which made it possible to describe differences in the motion of Newtonian and polymer fluids. The influence of the molecular weight on shear and longitudinal viscosities was found in [7]. We note that if the condition of independence of the asymptotic behavior of the shear viscosity from the molecular weight is satisfied, there is the following relationship (found in [7]) between the anisotropy parameters introduced into the equations of macromolecular dynamics: $\kappa = 1.2\beta$.

Straining of polymer solutions and melts in the regime of superposition of stationary shear flow on periodic strain with small amplitudes is one efficient method of checking the rheological equations of state. Therefore, in this work, we will study the following types of flow: superposition of small oscillating vibrations on stationary flow in the directions parallel to the shear and orthogonal to it.

The field of velocity gradients in parallel superposition has the form

$$\nu_{ik} = 2\begin{bmatrix} 0 & \gamma_1 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} + 2\omega \begin{bmatrix} 0 & \gamma_2 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} \exp(i\omega t)\,; \tag{6}$$

whereas in modeling of the orthogonal superposition it appears as

$$\nu_{ik} = 2\begin{bmatrix} 0 & \gamma_1 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} + 2\omega \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & \gamma_2 \\ 0 & 0 & 0 \end{bmatrix} \exp(i\omega t)\,. \tag{7}$$

For computation from Eqs. (4) and (5) we make the quantities used dimensionless, which results in

$$\tilde{\sigma}_{ik} = -\tilde{p}\delta_{ik} + \tilde{a}_{ik}\,, \quad \mathrm{De}\frac{d}{d\tilde{t}}\tilde{a}_{ik} - \mathrm{We}\tilde{\nu}_{ij}\tilde{a}_{jk} - \mathrm{We}\tilde{\nu}_{kj}\tilde{a}_{ji} + \left(1 + \frac{\mathrm{We}(\kappa - \beta)\,\tilde{a}_{jj}}{3}\right)\tilde{a}_{ik} = 2\tilde{\gamma}_{ik} - \mathrm{We}\beta\tilde{a}_{ij}\tilde{a}_{jk}\,. \tag{8}$$

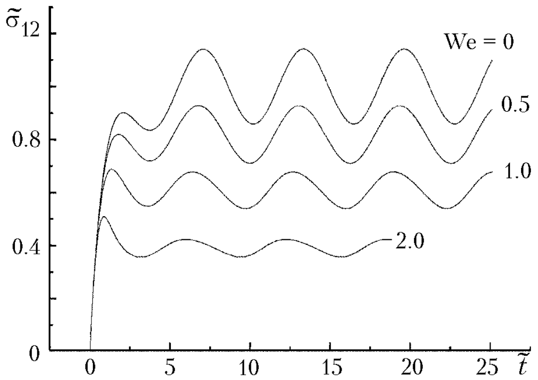
The dimensionless quantities in (8) are found from the following expressions:

$$\tilde{\sigma}_{ik} = \frac{3R}{U\eta_0}\sigma_{ik}\,, \quad \tilde{p} = \frac{3R}{U\eta_0}p\,, \quad \tilde{a}_{ik} = \frac{3R}{\tau_0 U}a_{ik}\,, \quad \tilde{\nu}_{ik} = \frac{U}{R}\nu_{ik}\,, \quad \mathrm{We} = \frac{\tau_0 U}{R}\,, \quad \mathrm{De} = \omega\tau_0\,.$$

The parameters of the models obtained are the following dimensionless quantities: $\tilde{t}$, $\tilde{\gamma}_1$, and $\tilde{\gamma}_2$, and the numbers We and De, i.e., $\tilde{a}_{ik} = \tilde{a}_{ik}(\tilde{t}, \tilde{\gamma}_1, \tilde{\gamma}_2, \mathrm{We}, \mathrm{De})$.

Numerical solution of the superposition of small oscillating vibrations of simple shear flow in the directions parallel to the flow and orthogonal to it has been carried out from model (8) using the Runge–Kutta method of fourth order of accuracy (Fig. 1). For the Cauchy problem of the systems of equations, we selected the zero initial conditions $\tilde{a}_{ik}(0) = 0$, which corresponds to straining from the state of rest. The stress grows with time, subsequently reaching the level of steady-state straining (steady-state flow).

From the dependences presented in Fig. 1, it is clear that, as the We number grows, i.e., as the elastic properties of a polymer increase, the dimensionless amplitude of oscillation of the stress decreases and the level of variations of the stresses in steady-state flow is reduced. Furthermore, a delay of the medium's vibrations appears with



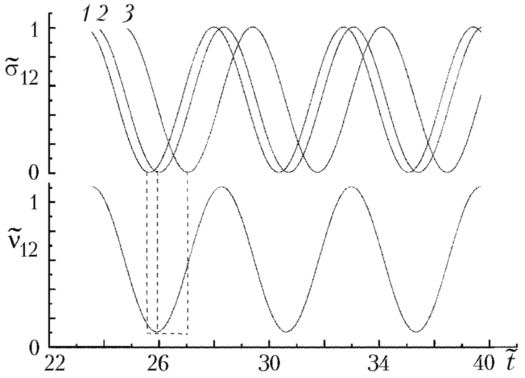


Fig. 1. Shear stresses vs. time for parallel superposition, $\kappa = 0.7$, $\beta = 0.5$, $\tilde{\gamma}_1 = 0.5$, $\tilde{\gamma}_2 = 0.1$, and De = 1.

Fig. 2. Dependences of $\tilde{v}_{12}$ and $\tilde{\sigma}_{12}$ for parallel superposition (We = 1, $\kappa = 0.7$, $\beta = 0.5$, $\tilde{\gamma}_1 = 4$, and $\tilde{\gamma}_2 = 0.001$): De = $10^{0.125}$ (1), 10 (2), and 10^2 (3).

increase in We. If, for example, we compare the curves obtained in Fig. 1 for Newtonian (We = 0) and viscoelastic liquids (We = 2), we see a phase shift between them. An increase in the dimensionless rate $\tilde{\gamma}_1$ leads to a decrease in the oscillation amplitude and an increase in the dimensionless stress $\tilde{\sigma}_{12}$. Larger We numbers correspond to a lower steady-state stress.

The results obtained are qualitatively consistent with the existing experimental data.

Also, we have considered the processes of straining of a medium under forced vibrations for parallel and orthogonal superpositions; it has been established that if $D \neq 0$, we have a delay of stress oscillations from forcing oscillations and the value of the delay increases with De number. These results were obtained for both the parallel and orthogonal superpositions. However, the difference in these types of superposition was established, namely, an advance of the vibrations of the medium strained relative to the forcing oscillations was observed (Fig. 2) for certain critical values of the rate of shear $\tilde{\gamma}_1 > \tilde{\gamma}_1^0$ in modeling of parallel superposition for frequencies De < 1; when De = 1, the vibrations of the medium strained coincided with forcing oscillations, and we had a delay of the medium's vibrations from the forcing oscillations for De > 1. Curve 1 in the figure is shifted to the left and curve 3 is shifted to the right from the forcing-oscillation curve.

The results of calculations of orthogonal superposition have shown the absence of a change in the phase shift between stress oscillations and forcing oscillations.

To compare the solutions of the system of equations (8) to the experimental data available in the literature we expand σ_{12} for the parallel type of superposition and σ_{23} for the orthogonal type in Fourier series in time t, leaving the terms with a fundamental frequency ω:

$$\sigma_{12}(t) = \overset{0}{\sigma}_{12} + \overset{1}{\sigma}_{12}\cos\omega t + \overset{2}{\sigma}_{12}\sin\omega t\,, \quad \sigma_{23}(t) = \overset{0}{\sigma}_{23} + \overset{1}{\sigma}_{23}\cos\omega t + \overset{2}{\sigma}_{23}\sin\omega t\,. \tag{9}$$

Next we transpose $\overset{0}{\sigma}_{12}$ and $\overset{0}{\sigma}_{23}$ in expressions (9) to the left-hand side and, multiplying both sides of the equations by $-i\omega/\gamma_2$, we obtain that the right-hand sides of the equations represent equations for the complex shear modulus of the parallel and orthogonal superpositions respectively:

$$G(\omega, \gamma_1) = \frac{\omega\overset{2}{\sigma}_{12}}{\gamma_2} - i\,\frac{\omega\overset{1}{\sigma}_{12}}{\gamma_2}\,, \quad G(\omega, \gamma_1) = \frac{\omega\overset{2}{\sigma}_{23}}{\gamma_2} - i\,\frac{\omega\overset{1}{\sigma}_{23}}{\gamma_2}\,.$$

From these expressions, separating the real and imaginary sides $G(\omega, \gamma_1) = G'(\omega, \gamma_1) - iG''(\omega, \gamma_1)$, we can determine the elastic and loss moduli for the parallel and orthogonal types of superposition. Using them, one can find such characteristics as the dynamic viscosity $\eta' = G''/\omega$ and the phase angle (angle of mechanical loss) $\delta = \arctan\,(G''/G')$.

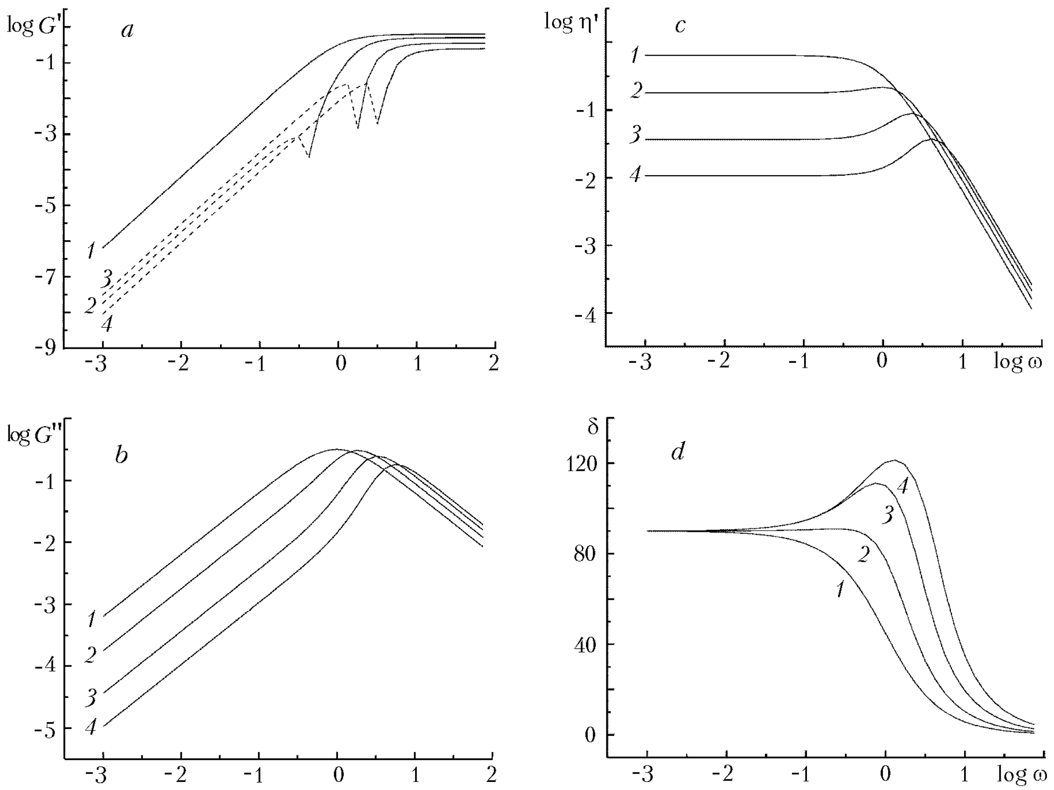


Fig. 3. Elastic modulus G' (a), loss modulus G'' (b), dynamic viscosity η' (c), and phase angle δ (d) vs. frequency ω in parallel superposition: 1) $\gamma_1 = 0.2$; 2) 0.5; 3) 1.5; 4) 4.

The components of the complex shear modulus depend on both the frequency ω and the rate of shear γ_1. In the case where $\gamma_1 \to 0$, one obtains the shear modulus $G_0(\omega) = G_0'(\omega) - iG_0''(\omega)$ determined by the linear-viscoelasticity relations.

Figure 3 gives results of the numerical experiment on parallel superposition of small oscillating vibrations on simple shear flow for different rates of shear γ_1 and the following set of parameters: $\eta_0 = 1$, $\tau_0 = 1$, $\kappa = 0.7$, $\beta = 0.5$, and $\gamma_2 = 0.001$.

From the above dependences, it is clear that the results of modeling of parallel and orthogonal superpositions, which have been noted earlier, lead to the fact that, for certain values of the frequency $\omega < \omega_0$, the shear modulus G' for the parallel superposition takes, unlike the orthogonal superposition, negative values, shown on the plot by dashed curves. Comparing the curves in Fig. 3a and d, we note that the frequency ω_0 corresponds to the phase angle $\delta = 90^{\text{o}}$. Also, it has been established that the shear modulus takes negative values when we have a delay of the forcing oscillations from the medium's vibrations.

The results obtained are qualitatively consistent with experimental data [8, 9]; therefore, the next step of the numerical experiment was to study their quantitative correspondence. In modeling the parallel superposition, we took the values of the parameters from [8]: $\eta_0 = 501.19$ P, $\tau_0 = 0.453$ sec, and $\gamma_2 = 0.001$ for the rates of shear γ_1 equal to 0.056, 0.179, 0.563, and 1.790 sec^{-1}. For numerical calculations from model (4) and (5), we selected κ and β from the condition of best agreement between the experimental data and the theoretical curves; We took $\kappa = 0.7$ and $\beta = 0.5$. Since the samples used in the experiments had a high polydispersity, we were unable to select the values of the rheological parameters for accurate description of the linear modulus $G_0'(\omega)$ obtained for $\gamma_1 = 0.056$ sec^{-1}; therefore, in the work, we calculated the deviations (Fig. 4a) $G_0'(\omega) - G_i'(\omega)$ of the elastic modulus $G_0'(\omega)$ and of the following moduli: $G_1'(\omega)$ obtained for the rate of shear $\gamma_1 = 0.179$ sec^{-1} (curve 1), $G_2'(\omega)$ for $\gamma_1 = 0.563$ sec^{-1} (2), and $G_3'(\omega)$ for $\gamma_1 = 0.790$ sec^{-1} (3). Curves $1'$–$3'$ are the corresponding deviations of experimental data [8].

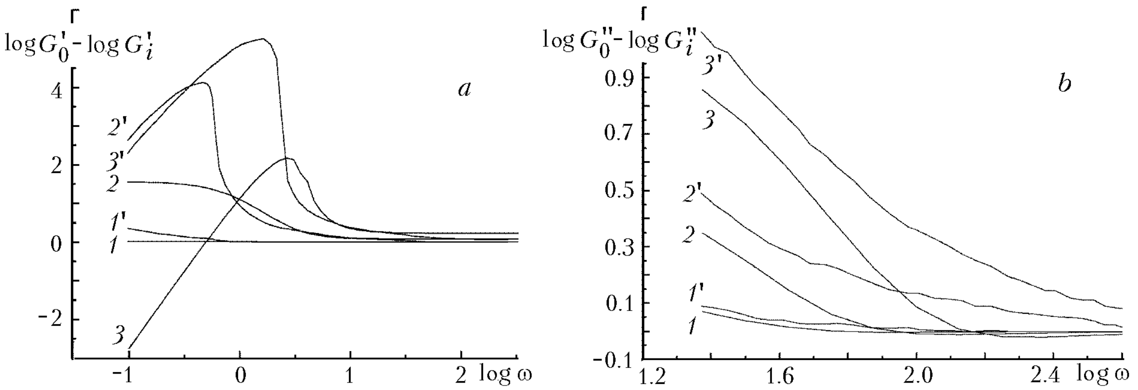


Fig. 4. Comparison of theory and experiment for parallel (a) and orthogonal (b) superpositions.

Data for modeling of the orthogonal superposition were taken from [9]: η_0 = 30 P, τ_0 = 0.03532 sec, and γ_2 = 0.001 for the rate of shear γ_1 equal to 0.2, 25.4, 102.4, and 407 sec^{-1}. The values of the parameters κ and β of model (4) and (5) were taken to be 0.7 and 0.5, respectively. The results on the orthogonal superposition are also presented in Fig. 4b in the form of the deviations $G_0'(\omega)$ for γ_1 = 0.2 sec^{-1} and the moduli $G_1'(\omega)$ for γ_1 = 25.4 sec^{-1} (curve 1), $G_2'(\omega)$ for γ_1 = 102.4 sec^{-1} (2), and $G_3'(\omega)$ for γ_1 = 407 sec^{-1} (3). Curves $1'$–$3'$ are the corresponding deviations of experimental data [9].

We note that the attempt at describing the dependence of the dynamic characteristics of polymer solutions in orthogonal superposition based on the Leonov–Prokunin model, which has been made in [10], cannot be accepted as successful, since the deviation from experiment attains three times in certain cases, despite the accurate description of the initial value of the moduli.

Thus, a qualitative agreement between the theoretical dependences of the components of the complex shear modulus in different regimes of straining and the experimental data has been shown. The existing differences are due to the fact that the relaxation character of interaction of the macromolecule with its environment (environmental aftereffect) and the long-scale interaction of the beads along the chain, associated with the presence of topological restrictions in macromolecular motion (internal viscosity), have not been allowed for within the framework of the rheological model (4) and (5). There are approaches to allowance for these factors at present, which makes it possible to investigate the influence of the environmental aftereffect and the internal viscosity on the dynamic characteristics of solutions and melts of linear polymers in shear. Another reason for the existing differences is the high polydispersity of the samples used in the experiments [8, 9], which can also be allowed for within the framework of the approach presented by averaging of the components of the complex shear modulus over molecular-weight distribution.

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NOTATION

a_{ik}, second-rank symmetric tensor describing the deviation of a statistically nonequilibrium system from the equilibrium state; $\tilde{a}_{ik}$, dimensionless anisotropy tensor; c, concentration of the solution; c^*, certain constant; De, Deborah number; $G(\omega, \gamma_1)$, complex shear modulus; G', elastic modulus; G'', loss modulus; G_0, complex shear modulus for $\gamma_1 \to 0$; G_0', elastic modulus for $\gamma_1 \to 0$; G_0'', loss modulus for $\gamma_1 \to 0$; n, number of macromolecules per unit volume; M, molecular weight; M^*, certain constant; p, pressure; $\tilde{p}$, dimensionless pressure; R, characteristic scale; $\mathbf{r}_i^1$ and $\mathbf{r}_i^2$, Cartesian coordinates of the radius vector of dumbbell beads; T, temperature in energy units; $2T\mu$, coefficient of elasticity of the model spring; $\tilde{t}$, dimensionless time; U, scale velocity; W, probability density function related to the individual molecules being in a prescribed configuration in the polymer chain; We, Weissenberg number; β and κ, scalar coefficients of anisotropy; δ, phase angle (mechanical-loss angle); δ_{ik}, Kronecker symbol; $\gamma_{ik} = (\nu_{ik} + \nu_{ki})/2$, symmetrized tensor of velocity gradients; $\tilde{\gamma}_{ik}$, dimensionless symmetrized tensor of velocity gradients; γ_1, rate of shear; γ_2, oscillation amplitude; $\tilde{\gamma}_1$, dimensionless rate of shear; $\tilde{\gamma}_2$, dimensionless oscillation amplitude; $\tilde{\gamma}_1^0$, critical value of the dimensionless rate of shear; η', dynamic viscosity; η_0 and τ_0, initial values of the shear viscosity and the relaxation time; μ, proportionality factor; ν_{jk}, velocity-gradient tensor; $\tilde{\nu}_{ik}$, dimensionless tensor of velocity gradients; ρ_i, relative motion of beads; ρ_i^0, motion of the center of gravity of the dumbbell; σ_{ik}, stress tensor; $\tilde{\sigma}_{ik}$, dimensionless stress; $\overset{0}{\sigma}_{12}$, $\overset{1}{\sigma}_{12}$, $\overset{2}{\sigma}_{12}$, $\overset{0}{\sigma}_{23}$, $\overset{1}{\sigma}_{23}$, and $\overset{2}{\sigma}_{23}$, Fourier coefficients; τ, relaxation time of the orientational process; ω, frequency; ω_j^α, average velocity of a bead with No. α; ψ_i, relative rate of diffusion of beads; ψ_i^0, rate of diffusion of the center of gravity of the dumbbell; ζ, coefficient of friction; ζ_{ik}, tensor coefficient of friction; $\langle \rho_i \rho_k \rangle$, moments of second order, characterizing dumbbell orientation on the average; $\langle \rho^2 \rangle_0$, equilibrium value of the expression $\langle \rho_1^2 \rangle + \langle \rho_2^2 \rangle + \langle \rho_3^2 \rangle$. Subscripts: i, j, k, and l, Cartesian coordinate Nos. (they take values of 1, 2, and 3); 0, initial or equilibrium value. Superscripts: α, bead Nos. in the macromolecule model (they take values of 1 or 2); 0, center of gravity.

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