RHEOKINETICS OF MOMENTUM TRANSFER IN MICROSTRUCTURED MEDIA

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A theory for momentum transfer in chemically active microstructured media is proposed. This theory is based on the assumption of the existence of a continuous internal parameter, which is determined by the mean statistical structure of the medium and obeys the main principles of rheology and physical kinetics. Expressions for the stress-tensor components are obtained, which permit estimation and analysis of the influence of physico-mechanical properties of the medium and external factors on momentum-transfer characteristics. As an example, spatially uniform flows are considered (unsteady shear, oscillating, and elongation ones). The results can be used in studying flows with complex deformation).

The constitutive elements of multiphase media differ considerably in their sizes and physical properties. The transfer processes in such heterogeneous media, where each individual phase occupies a certain part of their volume ($V = \Sigma V_j$, where $j = 1, \ldots, n$), is described in [1] in the approximation of "multivelocity (temperature) mutually penetrating and interacting continua" taking into account averaging of thermomechanical characteristics and physical properties over the variety of their values for individual locally equilibrium phases that occupy a volume V_j. The transfer in these media can be caused by the nonequilibrium character of mass, force, energy, and physicochemical interaction between the phases inside a representative volume V of the mixture.

At the same time, there is a great variety of finely and ultra-dispersed incompressible multicomponent media and their flows [2] for which the transfer problem may be considered from the viewpoint of a quasi-homogeneous approach, invoking methods of rheology and physical (structural) kinetics. In a homogeneous medium, each component can occupy its entire volume ($V = V_j$, where $j = 1, \ldots, n$). The momentum (substance and energy) transfer and the corresponding fluxes in such media caused by external and internal forces can be described by deviation of the distribution function of the probability density of physicochemical (structural) properties of the mixture components from their equilibrium state.

We assume that the contribution of reaction forces to the change in momentum depends on the rate of change of the distribution function of the probability density of chemically active reacting components.

We consider a rather broad class of rheological media that have common structural-mechanical properties.

It is known that a medium with small particles that do not interact with each other (the volume concentration of particles is $\varphi \leqslant 0.02$) in the laminar regime shows no rheological properties. In this case, the multi-component medium is modeled with a spatial fluctuating network structure [3–6] filled with a nonrheological medium (such as, for instance, a low-molecular solvent, an oxidant, etc.). The model of a microstructured medium is described in [6].

The inertia-free Kun segment [7] can serve as a characteristic kinetic parameter of the structural network. This quantity is a minimum macroscopic linear scale b_i of a subchain of length $b_i S < H$ which,

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for a given macroscopic perturbation, has a random mobility (of the ith physicomechanical component) with a linear velocity v_i. Here S is the number of segments between neighboring nodes of the net and H is a characteristic macroscopic scale on which thermodynamic and statistical flow parameters of the medium change appreciably. Continuity of the random quantity (component) is ensured by consideration of a distribution function for the probability density $W_i(x, v_i, t)$ for which x, v_i, and t are independent variables. The mean velocity of a component with the number (concentration) density $n_i(x, t) = \int W_i(x, v_i, t) dv_i$ is the quantity $\bar{v}_i(x, t) = n_i^{-1} \int v_i W_i(x, v_i, t) dv_i$ averaged over the distribution of v_i. The flow velocity of the mixture is defined as $v_0(x, t)$. The thermal velocity of its ith component is defined as $v_i - v_0$, and the diffusional velocity of the flow of several components is $\bar{v}_i - v_0$. In this situation, the coordinate system moves with the macroscopic velocity of the mixture flow.

Because of the complexity of taking into account information about the motion of component-segments (such as intersegmental rigidity, orientation of the components, etc.), we simplify the model. In what follows, we assume that the phase carrier does not interact with an irregular ($Sb_i \neq$ const) three-dimensional free-joint network structure in its nodes (x_i^α, where $\alpha = 1, \ldots, n$), where Stokes hydrodynamic, elastic (entropic) and thermal diffusion forces are concentrated. Hence, we assume that an effective node (of diameter d_i) interacts hydrodynamically with the carrier medium (gas, liquid, fuel oxidant, etc.) in the same manner as a set of segment-components of a subchain of density $n_i \approx Sb_i$. The chemical-reaction rate is modeled by the rate of variation of the number of active network nodes entering the reaction.

According to the kinematic theorem of transfer [8], the continuity equation for the distribution function of the probability density W_i for the ith reacting component (node) has the form

$$\frac{\partial W_i}{\partial t} + \mathrm{div}\,(W_i \bar{v}_i) = M_i. \tag{1}$$

where $\bar{v}_i$ is the velocity of the node relative to a moving coordinate system fitted to the center of mass and $M_i = \partial W_i / \partial t$ is the rate of change of the probability density of the number of nodes that entered the reaction (or the rate of the chemical reaction of energetically active nodes).

We find the relation for $\bar{v}_i$ in Eq. (1) from the condition of inertia-free balance of forces acting on a chosen node: $\Sigma f \approx 0$. The Stokes resistance force of the node with respect to the undisturbed macroscopic velocity of the medium at the point where the node is located is

$$f_1 = -\xi(\bar{v}_i - \dot{v}_{ij}x_j). \tag{2}$$

where ξ is the friction coefficient of a node of diameter d_i.

In a subchain extended by a distance h, an entropic force $F = 3kTh/(Sb_i^2)$ contracting the subchain operates. Denoting the coordinates of the neighboring nodes by x_i^α ($\alpha = 1, 2, \ldots, n$) and taking into account that the origin of the coordinate system is chosen at the center of mass, we obtain $\sum_{\alpha=1}^{n} x_i^\alpha \approx 0$. The effective mass of the node is $m_\alpha = $ const. In view of this, we obtain the elastic force (for the most probable value $\alpha \approx 4$)

$$f_2 = \frac{3kT}{Sb_i^2} \sum_{\alpha=1}^{n} (x_i^\alpha - x_i) \approx -\frac{12kTx_i}{Sb_i^2}. \tag{3}$$

where k is the Boltzm... constant.

The diffusion force caused by thermal motion of the ith node moving with velocity $\bar{v}_i$ relative to the coordinate system is [3-5]

$$f_3 = -D\xi\nabla \ln c = -kT\frac{\partial \ln W_i}{\partial x_i}. \tag{4}$$

where D is the diffusivity and T is the temperature. From the condition of inertia-free balance of forces we have

$$\bar{v}_i = \xi^{-1}(\xi \dot{v}_{ij}x_j + f_2 + f_3). \tag{5}$$

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From (1)–(5) we obtain

$$\frac{\partial W_i}{\partial t} + x_j \dot{v}_{ij} \frac{\partial W_i}{\partial x_j} = \ae^{-1}[\Delta W_i + \nabla(x W_i)] \pm \frac{\partial \bar{W}_i}{\partial t},\tag{6}$$

where $\ae = \xi S b_i^2/(12kT)$ is the time of transformation of the structure to its most probable configurational state (time of dynamic relaxation). The coordinates $x_{i,j}$ in Eq. (6) are normalized to $(Sb_i^2/12)^{0.5}$.

For the equilibrium state, all derivatives in the left part of Eq. (6) and the last term in the right part are equal to zero. With account for the normalization $\int_{-\infty}^{\infty} W_i(x)\,dx = 1$, Eq. (6) transforms to

$$W_0 = (12/(2\pi S b_i^2))^{3/2} \exp\left(-12x^2/(2 S b_i^2)\right).$$

In this situation, the root-mean-square deviation of the node from the origin (configuration space) is $\langle x^2 \rangle_0 = \int x^2 W_0\,dx = S b_i^2/4$. Hence, the equilibrium state of the medium is characterized by an internal structural scale (root-mean-square radius of inertia) of order $\langle x^2 \rangle_0 = 3\ae D$. It should be noted that W_0 depends on $S b_i^2$, which has a definite physical meaning (root-mean-square radius of inertia of the interaction forces).

Multiplying Eq. (6) by $x_i x_j$ and integrating it, we obtain the following equation for the first moments of the distribution function $\left(\langle x_i x_j \rangle = \int x_i x_j W_i\,dV_i\right)$:

$$\frac{d\langle x_i x_j \rangle}{dt} = \langle x_i x_k \rangle \dot{v}_{kj} + \langle x_j x_k \rangle \dot{v}_{ki} - 2\ae^{-1}(\langle x_i x_j \rangle - \delta_{ij}) \pm \frac{\partial \overline{\langle x_i x_j \rangle}}{\partial t}.\tag{7}$$

In deriving Eq. (7), we used the condition of incompressibility ($\dot{v}_{kk} = 0$) and the above-indicated normalization of the quantity W_i and passed from the integral over the volume to the integral over the surface under the condition that $W_i \to 0$ at infinity. Here δ_{ij} is a unit matrix.

In what follows, in considering particular cases with various conditions of deformation, we assume that the chemical-reaction rate (in the presence of a source or drain) depends on the quantity $\langle x_i x_j \rangle$, i.e., $\partial\langle x_i x_j \rangle/\partial t = \pm k \langle x_i x_j \rangle$ [or $\partial\overline{\langle x_i x_j \rangle}/\partial t = \pm k(\langle x_i x_j \rangle - \delta_{ij})$]. The temperature dependence of the reaction rate constant may obey the Arrhenius law $k = k_0 \exp(-E/(RT))$. Here E is the activation energy and R is the universal gas constant. The reaction time τ (duration of chemical relaxation or the lifetime of nodes in the scale $\langle x^2 \rangle_0$) for a first-order reaction is inversely proportional to the reaction rate: $k \sim \tau^{-1}$. The quantity $k_0 \sim \tau_0^{-1}$ is the scale of the total time of the chemical reaction.

The momentum transfer occurs only in a nonequilibrium state of the medium, where the distribution function of the probability density differs from the equilibrium one. It follows from relation (7) that this difference is caused by local or spatial nonuniformity of the quantity $\langle x_i x_j \rangle$ or by chemical reactions.

Writing Eq. (7) in a dimensionless form, we obtain

$$\text{De}_0 \frac{d\langle x_i x_j \rangle}{d\bar{t}} = \text{We}_0[\langle x_i x_k \rangle \bar{v}_{kj} + \langle x_j x_k \rangle \bar{v}_{ki}] - \beta(G.T)[\langle x_i x_j \rangle - \delta_{ij}].\tag{8}$$

Here $\bar{t} = t/t_*$ (t_* is the time required for the physicochemical changes in the configuration space $\langle x^2 \rangle_0 = S b_i^2/4$ to be completed) and G is the velocity gradient.

The rheokinetics of the process is seen to be characterized by the Deborah and Weissenberg numbers ($\text{De}_0 = \ae_0/t_*$ and $\text{We}_0 = G\ae_0$, respectively) and also by physical nonlinearity of the type $\beta(G.T) = 2\ae_0/\ae(G) \pm (\ae_0/\tau_0)\exp(-E/(RT))$, where $\ae_0$ is the maximum relaxation time for $G \to 0$, and $\beta^{-1}(G.T)$ is the dimensionless time of physicomechanical relaxation in the presence of a drain or source of nodes in the reaction space (accordingly, either plus or minus are taken in the expression for β).

It follows from Eq. (8) that, under the action of external forces (G and T), the macroscopic transfer of momentum in the media under consideration is to be accomplished not instantaneously but in accordance with the times of physical and chemical relaxation.

At a low temperature of the reaction, we have $\ae \ll \tau = \tau_0 \exp(E/(RT))$ and $\beta = (2\ae \pm \tau)/(\tau\ae) \approx 1/\ae$; therefore, chemical kinetics is the determining (slowest) mechanism of momentum transfer. For a sufficiently

high temperature, we have $\beta \approx 1/\tau$, and the diffusion mechanism can be the determining one. When the times of physical and chemical relaxation are comparable, the transfer occurs in the transition (diffusion-kinetic) regime.

We obtain the vector of the momentum flux density of the nodes across an elementary surface in the direction x_j per unit time relative to v_0 as foll [8]. If one node interacts with the medium with a force $f_1 = -\xi(\bar{v}_i - \dot{v}_{ij}x_j)$ and there are n_i nodes in a unit volume, then we have

$$P_{ij} = -P_0\delta_{ij} + 2\mu\dot{e}_{ij} - 0.5n_i \int x_j(f_2 + f_3)W_i\, dV_i, \tag{9}$$

where $\dot{e}_{ij} = 0.5(\partial v_{ij} + \partial v_{ji})$ is the symmetric part of the strain-rate tensor and μ is the viscosity of the carrier medium.

According to (3) and (4), from (9) we obtain

$$P_{ij} = -P_0\delta_{ij} + 2\mu\dot{e}_{ij} + \varepsilon(\langle x_i x_j\rangle - \delta_{ij}). \tag{10}$$

In deriving Eq. (10), we used the Ostrogradskii–Gauss theorem and normalization. The quantity $\varepsilon = 0.5n_i kT$ is the elasticity modulus of the medium, which can be determined experimentally. The moments of the distribution function of the probability density of the nodes are normalized to $Sb_i^2/12$.

It follows from Eq. (10) that the well-known relation for the stress tensor in the model under study is supplemented by an anisotropic part $\varepsilon(\langle x_i x_j\rangle - \delta_{ij})$, which gives a measure of deviation of the stressed state of the medium from its equilibrium value.

Using the relations obtained, we consider the influence of rheokinetic factors of momentum transfer for several particular cases.

For the case $\dot{v}_{21} = G\sin(\omega t)$, the system of equations for the moments of the distribution function (8) is

$$\frac{d\langle x_1^2\rangle}{dt} + \beta[\langle x_1^2\rangle - 1] = 2G\langle x_1 x_2\rangle\sin(\omega t), \quad \frac{d\langle x_1 x_2\rangle}{dt} + \beta\langle x_1 x_2\rangle = G\langle x_2^2\rangle\sin(\omega t),$$

$$\frac{d\langle x_2^2\rangle}{dt} + \beta[\langle x_2^2\rangle - 1] = 0, \quad\quad\quad \frac{d\langle x_1 x_3\rangle}{dt} + \beta\langle x_1 x_3\rangle = G\langle x_2 x_3\rangle\sin(\omega t), \tag{11}$$

$$\frac{d\langle x_3^2\rangle}{dt} + \beta[\langle x_3^2\rangle - 1] = 0, \quad\quad\quad \frac{d\langle x_2 x_3\rangle}{dt} + \beta\langle x_2 x_3\rangle = 0.$$

Here G is the amplitude of deformation, $\omega = 2\pi/t_*$, where t_* is the period of deformation (characteristic time scale), and $\beta = 2/æ + (1/\tau_0)\exp(-E/(RT))$.

Using the method of variation of arbitrary constants, from the system of inhomogeneous equations (11) we find the corresponding moments $\langle x_i x_j\rangle$. In doing so, we use the initial conditions for $t = 0$: $\langle x_1^2\rangle = \langle x_2^2\rangle = \langle x_3^2\rangle = 1$ and $\langle x_3 x_1\rangle = \langle x_1 x_2\rangle = \langle x_2 x_3\rangle = 0$.

A successive solution of system (11) yields the following dimensionless expressions for nonzero moments:

$$\langle x_2^2\rangle = \langle x_3^2\rangle = 1, \quad \langle x_1 x_2\rangle = \frac{G}{\beta^2 + \omega^2}[\beta\sin(\omega t) - \omega\cos(\omega t) - \beta\exp(-\beta t)],$$

$$\langle x_1^2\rangle = 1 + \frac{2G^2\beta}{\beta^2 + \omega^2}\left[\frac{1}{2\beta} - \frac{0.5\beta\cos(2\omega t) + \omega\sin(2\omega t)}{\beta^2 + 4\omega^2}\right.$$

$$\left. -\frac{\omega}{2\beta}\left(\frac{1}{\beta} + \frac{\beta\sin(2\omega t) - 2\omega\cos(2\omega t)}{\beta^2 + 4\omega^2}\right) + \frac{\exp(-\beta t)\cos(\omega t)}{\omega}\right] + A\exp(-\beta t), \tag{12}$$

$$A = -\frac{G^2}{\beta^2 + \omega^2}\left[1 - \frac{3\omega\beta}{\beta^2 + 4\omega^2} - \frac{\omega}{\beta^2}\right].$$

For $\omega = 0$, from (12) we have

$$\langle x_1 x_2\rangle = \frac{G}{\beta}(1 - \exp(-\beta t)), \quad \langle x_1^2\rangle = 1 + \frac{2G^2}{\beta^2}(1 - \exp(-\beta t) - \beta t\exp(-\beta t)). \tag{13}$$

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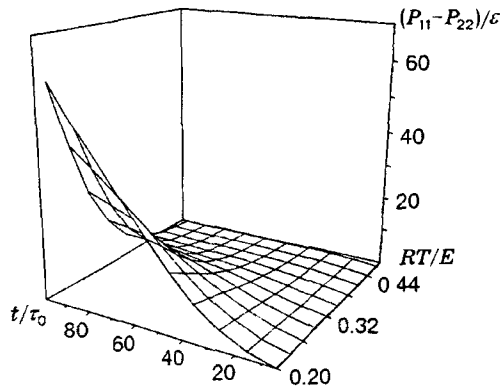
Fig. 1

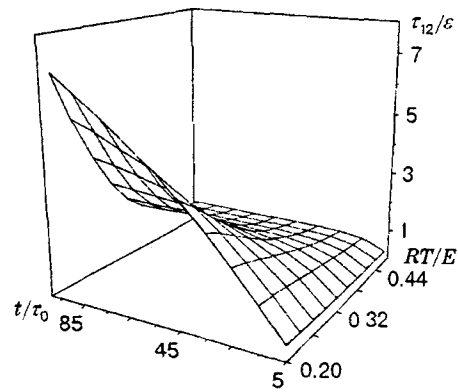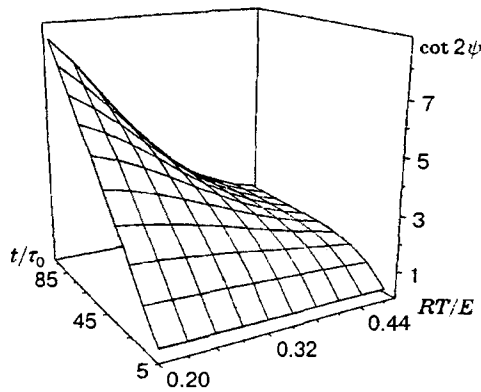
Fig. 2

Fig. 3.

Expressions (13) for $t \to \infty$ can be written as

$$\|\langle x_i x_j \rangle\| = \left\| \begin{array}{ccc} 1 + 2G^2/\beta^2 & G/\beta & 0 \\ G/\beta & 1 & 0 \\ 0 & 0 & 1 \end{array} \right\|. \tag{14}$$

Substituting (14) into (10), we obtain the stress tensor

$$P_{ij} = \left\| \begin{array}{ccc} -P_0 + 2\varepsilon G^2/\beta^2 & \mu G + \varepsilon G/\beta & 0 \\ \mu G + \varepsilon G/\beta & -P_0 & 0 \\ 0 & 0 & -P_0 \end{array} \right\|. \tag{15}$$

It follows from (15) that, for a shear flow specified with a constant velocity gradient $G = \dot{v}_{12}$, the stressed state is characterized by a stress tensor with tangent and normal components $\tau_{12} = \tau_{21}$ and P_{ii}, respectively, with $\tau_{ij} \sim G$ and $(P_{11} - P_{22}) \sim G^2$. This viscoelasticity is evidenced by the tests of [9-11] and also by various phenomenological models of both differential and integral [2] types.

For the media under consideration, the momentum fluxes and the chemical-reaction rates are anisotropic. Figures 1 and 2 show the difference in normal $(P_{11} - P_{22})$ and tangential (τ_{12}) components of stress as a function of the dimensionless time and the quantity RT/E for a transitional flow $(G\tau_0 = 0.1)$. At first, the difference in normal stresses varies in time more slowly than tangential stresses and, afterwards, faster. With increasing RT/E, the difference $P_{11} - P_{22}$ decreases faster than τ_{12}. For short times of external action, the momentum transfer is practically independent of RT/E.

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v_1, v_2, v_3, v_4

2.0

1.5

1.0

0.5

0

4

3

2

1

0   2   4   6   8 βt

Fig. 1

$\tau_{12} \cdot 10^m$, N/m²

6

4

2

0   2   4   6 $(P_{11}-P_{22}) \, 10^n$, N/m²

Fig. 5

P_{11}/ε

30

20
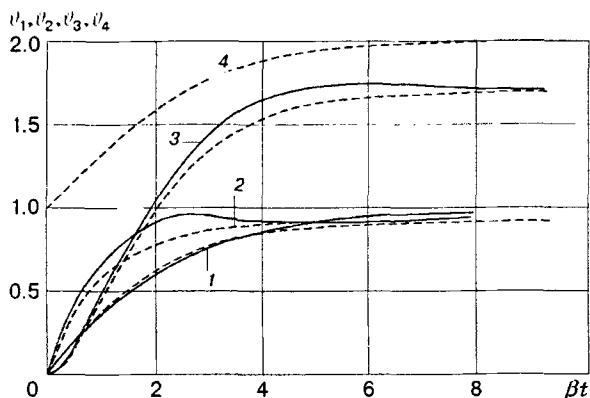
10

RT/E

0.52

t/τ_0 0.50   0.42   0.34   0.26   0.20   0.36
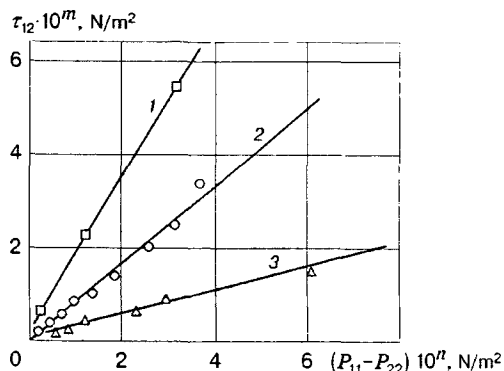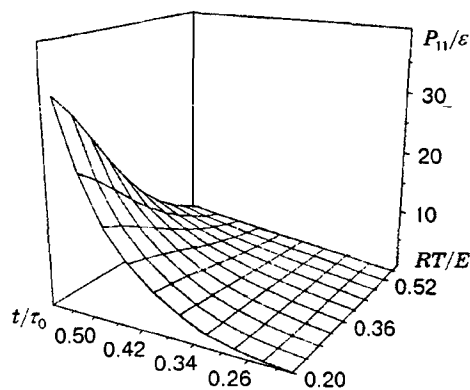
Fig. 6

It is known that not all collisions of molecules whose energy is greater than the activation energy result in chemical transformation. For a reaction to proceed, in addition to the required activation energy, the colliding molecular structures should be properly packed and oriented and their momenta properly ordered. For complex structures, orientation can significantly affect both the rate and direction of chemical reactions.

By means of orthogonal transformations [12] of matrix (14) we obtain

$$\frac{\langle x_1^2 \rangle - \langle x_2^2 \rangle}{2\langle x_1 x_2 \rangle} = \frac{Gx_0}{2x_0/x \pm (x_0/\tau_0) \exp(-E/(RT))} = \cot 2\psi. \tag{16}$$

Relation (16) gives the probability of the case in which the principal axes of the ellipse of revolution for the moments are aligned with the flow. In the case $G \to 0$ and $\psi = 45°$, we obtain $P_{11} - P_{22} = 2\tau_{12} = 0$. For $\psi = 22.5°$, we have $P_{11} - P_{22} = 2\tau_{12}$. It follows from Eq. (16) that, as the value of G increases, the angle of orientation and the activation energy of the medium decrease. This is caused by increasing frequency of collisions between the components. Hence, the reaction rate increases with increasing shear velocity. The dependence $\cot 2\psi = f(RT/E.t/\tau_0)$ for $G\tau_0 = 0.1$ is shown in Fig. 3. Orientation exerts a profound influence on flow anisotropy and the rate of chemical reactions at low temperatures.

Figures 4 and 5 show experimental data and results predicted by the above described model for $\tau_0 \to \infty$ in transient and steady deformation regimes of high-molecular media, respectively. The parameters x and ε of the model were determined from the experimental data of [13-16] in accordance with the predictions of the proposed theory for steady deformation. The solid curves in Fig. 4 show the experimental results of [13] obtained for an 8%-polystyrene solution in tricresyl phosphate [$\vartheta_1 = \cot 2\psi$ (curve 1), $\vartheta_2 = \tau_{12}/\varepsilon$ (curve 2), and $\vartheta_3 = (P_{11} - P_{22})/\varepsilon$ (curve 3)]. Figure 4 also shows the calculated dependence $\vartheta_4 = \varepsilon(P_{11} - P_{22})/\tau_{12}^2 = f(\beta t)$ (curve 4). The expression $\varepsilon(P_{11} - P_{22})/\tau_{12}^2$ is the ratio of the elastic energy accumulated during shear

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to the dissipated energy. A close consideration shows that, unlike the expressions for τ_{12}, $P_{11} - P_{22}$, and $\cot 2\psi$, the ratio $\varepsilon(P_{11} - P_{22})/\tau_{12}^2$ does not depend on the deformation amplitude and only weakly depends on RT/E.

The points in Fig. 5 show the experimental data of [11] for $m = -3$ and $n = -4$ (points 1), the data of [12] for $m = -2$ and $n = -2$ (points 2), and the data of [13] for $m = -1$ and $n = -3$ (points 3). The calculated dependences are shown by solid curves (m and n are scaling factors).

In the case of uniaxial elongation under the action of a constant velocity gradient, we have $\dot{v}_{22} = \dot{v}_{33} = -0.5\dot{v}_{11}$ by virtue of incompressibility, and all nondiagonal components of (7) are equal to zero. For diagonal components, we obtain the relation

$$\frac{dx_i^2}{dt} + \beta[\langle x_i^2 \rangle - 1] = 2\dot{v}_{ii}\langle x_i^2 \rangle. \tag{17}$$

Solving Eq. (17) in a manner similar to that used above, we obtain

$$\langle x_i^2 \rangle = (1 - 2\dot{v}_{ii}\beta^{-1})^{-1}\{1 - 2\dot{v}_{ii}\beta^{-1}\exp\left[-\beta t(1 - 2\dot{v}_{ii}\beta^{-1})\right]\}. \tag{18}$$

In this case, the stress tensor has the form $P_{ii} = -P_0 + \varepsilon(\langle x_i^2 \rangle - 1)$, and its deviator part for P_{11}, by virtue of Eq. (18), is

$$P_{11} = \frac{2\varepsilon\dot{v}_{11}}{\beta(1 - 2\dot{v}_{11}/\beta)}\{1 - \exp\left[-\beta t(1 - 2\dot{v}_{11}/\beta)\right]\}.$$

The dependence $P_{11}/\varepsilon = f(RT/E, t/\tau_0)$ for $v_{11}\tau_0 = 0.1$ under conditions of tension is shown in Fig. 6.

The approach described above allows one to estimate and analyze the influence of the physicochemical properties of the medium, external factors acting on it, and structural (orientation) changes in the medium on momentum-transfer characteristics in the presence of a source or drain of the substance.

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