

## REVIEWS

## DIFFERENTIAL MODELS OF RHEOLOGICALLY NONSTATIONARY FLUIDS

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UDC 532.135+51-72:532

*An analysis has been made of basic differential models of rheologically nonstationary (viscoelastic) fluids as well as of their development and interrelation. The considered models cover a rich variety of viscoelastic media: polymer solutions and melts, natural formations (glaciers), and others. Among these models, a key role is played by the Maxwell upper convection model: it provides a theoretical basis for experimental determination of the dynamic characteristics of viscoelastic fluids and for development of new rheological models. It has been shown that to improve the reliability of results obtained with the aid of a complex rheological model, it is expedient to ensure a possibility of reducing it to the existing models and thus finding analytical solutions for a number of the simplest flows. Examples of use of the models in question when results of rheometric investigations are approximated and viscoelastic-fluid flows are calculated have been given. Special emphasis has been placed on an analysis of the correspondence of the derived solutions to the physical essence of the described processes, and also of the correctness of interpretation of some results or others.*

**Keywords:** mathematical models, viscoelastic fluids, determination of dynamic characteristics.

**Introduction.** Among the non-Newtonian media occurring widely in nature and technology, it is common practice to recognize stationary (non-Newtonian purely viscous and viscoplastic media) and rheologically nonstationary (viscoelastic) media [1–3]. The first are characterized by the one-to-one relationship of the stress and the rate of strain (in the simplest case of shear); their ratio which is called the effective viscosity  $\mu_{ef}$  of a medium remains constant with time at a constant value of

$$\mu_{ef} = \mu_{ef}(\dot{\gamma}) . \quad (1)$$

For the second, the effective viscosity depends, in addition to the shear rate, on the time  $t$  of action of applied stresses

$$\mu_{ef} = \mu_{ef}(\dot{\gamma}, t) . \quad (2)$$

Information on purely viscous and viscoplastic non-Newtonian media is widely presented in foreign scientific literature [4–6] and in the monographs published in the Russian language [7, 8] alike, but actively developing models of viscoelastic media are of the greatest interest.

Note that a change in the fluid's viscosity is associated with a number of hydrodynamic and physicochemical processes (orientation of particles of the dispersed phase in the flow, the disruption and restoration of bonds between them, etc.) and always takes some time. However, if the time of this reconstruction is shorter than the relaxation time, such fluids may be considered stationary. Thus, low-molecular-weight fluids at strain frequencies as low as  $10^6 \text{ s}^{-1}$  exhibit no viscoelastic properties; the time of their relaxation is shorter than  $10^6 \text{ s}$ , but at higher frequencies (to  $3 \cdot 10^8 \text{ s}^{-1}$ ), relaxation phenomena appear in them, too [9]. Thus, the notion of viscoelasticity is relative: as long as strains manage to follow stresses, no viscoelastic properties of the fluid are manifested, but with increase in the frequency of external actions or decrease in the mobility of macromolecules of the fluid with temperature strains begin to lag behind the actions and relaxation processes appear in the fluid medium.

Rheological models of a viscoelastic fluid always constrain the time parameter. Mathematically, they can be subdivided into two groups: differential models relating instantaneous values of stresses to the gradient of the fluid's velocity and integral ones reflecting the dependence of stresses in the fluid on the prehistory of flow.

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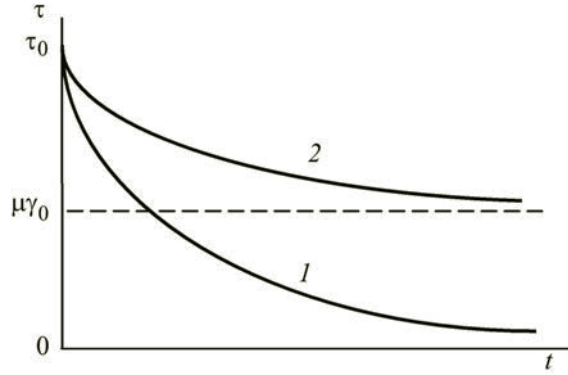


Fig. 1. Variation in the stress in the Maxwell viscoelastic fluid under constant strain (1) and at a constant strain rate (2).

**Differential Models.** In the general case the differential rheological model is of the following form [10]:

$$\lambda \frac{d\mathbf{T}}{dt} + \mathbf{T} + f(\nabla\mathbf{U}, \mathbf{T}) = \mu(\nabla\mathbf{U} + \nabla\mathbf{U}^T). \quad (3)$$

Here  $f(\nabla\mathbf{U}, \mathbf{T})$  is the nonlinear function which is invariant to the coordinate system.

*Maxwell convection models.* Expression (3) is a nonlinear generalization of Maxwell's linear theory of viscoelasticity. A concrete form of the rheological law must satisfy the principle of material frame indifference, i.e., the independence of the properties of a fluid from the selection of a coordinate system. It yields two natural rheological models [11]:

the Maxwell lower convection model

$$\mathbf{T} + \lambda \left( \frac{d\mathbf{T}}{dt} + \nabla\mathbf{U} \cdot \mathbf{T} + \mathbf{T} \cdot \nabla\mathbf{U}^T \right) = \mu(\nabla\mathbf{U} + \nabla\mathbf{U}^T) \quad (4)$$

and the Maxwell upper convection model

$$\mathbf{T} + \lambda \left( \frac{d\mathbf{T}}{dt} - \nabla\mathbf{U} \cdot \mathbf{T} + \mathbf{T} \cdot \nabla\mathbf{U}^T \right) = \mu(\nabla\mathbf{U} + \nabla\mathbf{U}^T). \quad (5)$$

In actual practice, much wider use is made by the second model, since results obtained with it are in better agreement with experimental data and with the molecular theory of viscosity [12].

In the case of one-dimensional strain Eqs. (4) and (5) are reduced to the linear equation

$$\gamma = \frac{d\varepsilon}{dt} = \frac{\tau}{\mu} + \frac{1}{G} \frac{d\tau}{dt}. \quad (6)$$

A physical analog of Eq. (6) represents a shock absorber and a spring connected in series and allowing for unlimited strain (of flow), which corresponds to a fluid dispersion medium with elastic dispersed-phase elements. When the values of the parameters  $\mu$  and  $G$  are constant Eq. (6) has analytical solutions. Thus, with the initial condition  $\tau(0) = \tau_0$  and the strain  $\varepsilon(t) = \varepsilon_0 = \text{const}$ , we have

$$\tau = \tau_0 \exp(-t/\lambda). \quad (7)$$

With the same initial condition and a constant strain rate  $d\varepsilon/dt = \gamma_0 = \text{const}$ , the integration of Eq. (6) yields

$$\tau = \mu\gamma_0 + (\tau_0 - \mu\gamma_0) \exp(-t/\lambda). \quad (8)$$

In Eqs. (7) and (8),  $\lambda = \mu/G$  is the relaxation constant of the viscoelastic fluid. As follows from (7) and (8), under the instantaneous strain  $\varepsilon_0$ , the initial stress  $\tau_0$  in a Maxwell fluid decreases exponentially to zero with time, and in the case of constant strain with a rate  $\gamma_0$ , to the quantity  $\mu\gamma_0$  which is proportional to this rate (Fig. 1).

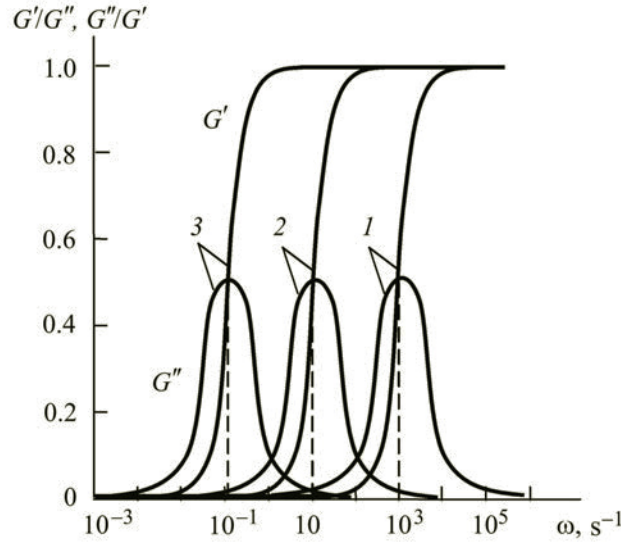


Fig. 2. Plots of  $G'(\omega)$  and  $G''(\omega)$  for the Maxwell fluid with a varying relaxation time:  $\lambda = 0.001$  (1),  $0.1$  (2), and  $10$  (3).

The Maxwell model provides a theoretical basis for determination of the dynamic characteristics of a viscoelastic fluid. Substitution, into (6), of the expression of the harmonic strain  $\varepsilon = \varepsilon_0 \sin \omega t$  produced in a rotary rheometer during its operation in an oscillatory regime yields the following solution to the indicated equation [13, 14]:

$$\tau = G \frac{(\lambda\omega)^2}{1 + (\lambda\omega)^2} \sin \omega t + G \frac{\lambda\omega}{1 + (\lambda\omega)^2} \cos \omega t . \quad (9)$$

Here the amplitudes of harmonic functions are the elastic (storage)  $G'$  and loss (dissipation)  $G''$  moduli of the energy of the viscoelastic fluid:

$$G'(\omega) = G \frac{(\lambda\omega)^2}{1 + (\lambda\omega)^2} \quad \text{and} \quad G''(\omega) = G \frac{\lambda\omega}{1 + (\lambda\omega)^2} . \quad (10)$$

Their geometric sum determines the value of the complex or dynamic modulus

$$G = \sqrt{G'^2 + G''^2} . \quad (11)$$

Figure 2 gives the plots of the relative value of the moduli  $G'$  and  $G''$  versus the frequency  $\omega$  at different values of the relaxation constant  $\lambda$ .

As we can see from Fig. 2, with growth in  $\lambda$  the  $G'(\omega)$  and  $G''(\omega)$  curves shift symbatically to a low-frequency region, i.e., viscous properties are more pronounced at a low frequency, whereas elastic ones, at high frequencies. For each fixed  $\lambda$  value, the moduli  $G'$  and  $G''$  vary with frequency in different directions: with increase in the frequency, the elastic modulus grows monotonically tending to a value of the dynamic modulus, whereas the loss-modulus curve is symmetric about the vertical axis and has a pronounced maximum. The intersection of the  $G'$  and  $G''$  curves yields the condition of determination of the relaxation constant  $\lambda$  of the viscoelastic fluid: setting the function  $G'(\omega)$  equal to the function  $G''(\omega)$ , we obtain

$$\lambda \Big|_{G'=G''} = 1/\omega . \quad (12)$$

The frequency at which  $G''(\omega)$  peaks is found from the condition

$$\partial G'' / \partial \omega = 0 . \quad (13)$$

Substituting the expression of the function  $G'(\omega)$  into (13), on rearrangements we have

$$\omega \Big|_{G''=G''_{\max}} = 1/\lambda . \quad (14)$$

Here, the value itself of the loss modulus will be

$$G''_{\max} = 0.5G , \quad (15)$$

i.e., the loss modulus of the Maxwell fluid peaks at a frequency reciprocal of its relaxation constant and its value is equal to half the dynamic modulus.

According to the theory of periodic functions, the tangent of the displacement angle  $\delta$  resulting from the summation of two harmonic oscillations of the same frequency, which are phase-shifted by  $90^\circ$  from each other, is analytically determined as the ratio of the amplitudes of these oscillations. In this case for the shear stress expressed by Eq. (9) we have

$$\tan \delta = G''/G' = 1/\lambda\omega . \quad (16)$$

In actual practice, the components of the dynamic modulus of the Maxwell fluid can be computed from the measured value of the displacement angle  $\delta$ . The dynamic modulus  $G$  itself is defined as the ratio of the measured amplitudes of the stress  $\tau_0$  and the strain  $\varepsilon_0$  [13, 9]

$$G = \tau_0/\varepsilon_0 . \quad (17)$$

The above equations correspond to an ideal (homogeneous) Maxwell viscoelastic fluid with one relaxation constant  $\lambda$ . Conversely, actual viscoelastic fluids which are a multicomponent inhomogeneous system as a rule, demonstrate a more complex dependence of viscous and elastic properties on the frequency of acting strains. A mechanical model of such fluids can be provided by the model of a generalized Maxwell fluid, which is a number of parallel-connected viscoelastic elements, each characterized by the eigenvalue of the relaxation constant  $\lambda_i = \mu_i/G_i$  [1, 13, 15]. In this model, the strain  $\varepsilon$  (harmonic in this case) is the same for all the parallel elements, and the stress  $\tau$  is equal to the sum of the stresses on the elements [13, 9]

$$\tau = \sum_{i=1}^N G_i \frac{(\lambda_i\omega)^2}{1 + (\lambda_i\omega)^2} \sin \omega t + \sum_{i=1}^N G_i \frac{\lambda_i\omega}{1 + (\lambda_i\omega)^2} \cos \omega t . \quad (18)$$

For the spectrum of relaxation times  $\lambda_i$ , each corresponding to its own dynamic modulus  $G_i$ , the expressions of the moduli  $G'(\omega)$  and  $G''(\omega)$  will take the form

$$G' = \sum_{i=1}^N G_i \frac{(\lambda_i\omega)^2}{1 + (\lambda_i\omega)^2} \quad \text{and} \quad G'' = \sum_{i=1}^N G_i \frac{\lambda_i\omega}{1 + (\lambda_i\omega)^2} . \quad (19)$$

Here, the value of the dynamic modulus itself will be equal to

$$G = \sum_{i=1}^N G_i . \quad (20)$$

Notice that the relaxation spectrum is a set of the moduli  $G_i$  to be calculated and relevant  $G'_i$ ,  $G''_i$ , and  $\delta_i$  at assigned values and the number  $\lambda_i$  [16]. The parameters ( $\lambda_i$  and  $G_i$ ) have been called adjustable parameters in [16], which stresses their artificial nature. The objective of obtaining the relaxation spectrum is to simplify computations due to the representation of the complex function  $G(\omega)$  of an actual viscoelastic fluid as a superposition of a finite number of simpler functions.

Frequency characteristics of viscoelastic fluids, which are used as the initial data of many rheological models (in particular, of those considered below), are determined on the basis of Maxwell models. Thus, in [17], using the Papanastasiou–Scriven–Macosko (PSM) model, a numerical study has been made of capillary-extrusion flow of a fluoropolymer melt; these model's constants  $\lambda_i$  and  $G_i$  were determined from oscillation-test data by the method of nonlinear regression analysis. The same parameters involved in the storage function  $m(t)$  of the Wagner model were determined in [18] experimentally from the results of oscillatory shear flow of a polyethylene melt at frequencies of 0.1 to 200 Hz, from which values for the moduli  $G'$  and  $G''$  were obtained.

In [19], the generalized Maxwell model was used to determine constants of the relaxation function in simulating the viscoelastic response of angle ligaments to a cyclic external action. In [20], a study has been made of the viscoelastic properties of aqueous solutions of erucyl bis (2-hydroxyethyl) methylammonium chloride (EHAC) and their mixtures with hydrophobized polyacrylamide (PAA). The data obtained for EHAC solutions were in good agreement with the Maxwell model with one relaxation time, whereas their mixtures with PAA had more complex properties and spectrum of relaxation times similarly to semidiluted polymer solutions. In [21], the elastic and loss moduli of EHAC solutions were determined in

the frequency range 0.01–100 rad/s at a temperature of 25°C, and also were calculated from Eqs. (10) of the Maxwell model with one constant  $\lambda$ . Good agreement was obtained between experimental and calculated results ([21], Fig. 2).

*Oldroyd models* follow, respectively, from the Maxwell lower and upper convection models (Oldroyd-A and Oldroyd-B models):

$$\mathbf{T} + \lambda \left( \frac{d\mathbf{T}}{dt} + \nabla\mathbf{U} \cdot \mathbf{T} + \mathbf{T} \cdot \nabla\mathbf{U}^T \right) = 2\mu \left[ \mathbf{D} + \theta \left( \frac{d\mathbf{D}}{dt} + \mathbf{D} \cdot \nabla\mathbf{U}^T + \nabla\mathbf{U}^T \cdot \mathbf{D} \right) \right], \quad (21)$$

$$\mathbf{T} + \lambda \left( \frac{d\mathbf{T}}{dt} - \nabla\mathbf{U} \cdot \mathbf{T} - \mathbf{T} \cdot \nabla\mathbf{U}^T \right) = 2\mu \left[ \mathbf{D} + \theta \left( \frac{d\mathbf{D}}{dt} - \mathbf{D} \cdot \nabla\mathbf{U}^T - \nabla\mathbf{U}^T \cdot \mathbf{D} \right) \right]. \quad (22)$$

For a one-dimensional flow, nonlinear equations (21) and (22) are reduced to the linear one-dimensional Jeffreys model [22]

$$\tau + \lambda \frac{d\tau}{dt} = \mu \left( \gamma + \theta \frac{d\gamma}{dt} \right). \quad (23)$$

The physical meaning of the delay time  $\theta$  is that on stress relieving, shear rates will decrease proportionally to  $\exp(-t/\theta)$ . At  $\theta = 0$ , Oldroyd and Maxwell models turn out to be identical. Just as for Maxwell models, in actual practice, more frequent use is made of the Oldroyd upper convection model. In [23], a new differential-difference model of a viscoelastic fluid with a constant relaxation time has been proposed; the model gives a fin velocity of propagation of perturbations and is in good agreement with Maxwell and Oldroyd models.

In [24], a numerical study has been made of the notion of bubbles in a polymer solution whose rheological properties were described by the Oldroyd-B model. The total viscosity of the solution  $\mu_0$  at a low shear rate was represented as the sum of the viscosities of the solvent  $\mu_s$  and the polymer  $\mu_p$

$$\mu_0 = \mu_s + \mu_p = (1 + c) \mu_s. \quad (24)$$

The introduction of the parameter  $c = \mu_p/\mu_s$  made it possible to relate the time of relaxation  $\lambda$  and delay  $\theta$ :

$$\theta = \lambda/(1 + c). \quad (25)$$

The calculation results confirmed the existence of the critical volume of a bubble in which its velocity sharply grows, and the afterbody acquires a pointed shape, which was repeatedly observed experimentally [25]. Also, of interest are the results of calculation of the velocity of bubbles in an unsteady regime (Fig. 3).

It has been shown that the bubble velocity reverses its direction for a short period (0.025–0.045), after which tends to a constant positive value. The phenomenon is attributed to the change in the velocity field near the bubble on reaching a precritical dimension by it when at some instant there occurs fluid motion past the bubble, which is reverse with respect to the lift, and is confirmed by experimental data [26, 27].

The *Coleman–Noll model* represents the general equation of state of a viscoelastic fluid with fading memory by the asymptotic series [28, 29]

$$\mathbf{T} = -P\boldsymbol{\delta} + \mathbf{S}_1(\mathbf{U}) + \mathbf{S}_2(\mathbf{U}) + \dots + \mathbf{S}_n(\mathbf{U}). \quad (26)$$

Here  $\mathbf{S}_i(\mathbf{U})$  are the nonlinear components of the stress tensor [30]

$$\begin{aligned} \mathbf{S}_1 &= \mu(\nabla\mathbf{U} + \nabla\mathbf{U}^T) = \mu\mathbf{A}_1, \\ \mathbf{S}_2 &= \mathbf{S}_1 + \alpha_1 \frac{\partial\mathbf{A}_1}{\partial t}, \\ &\dots\dots\dots, \\ \mathbf{S}_n &= \mathbf{S}_{n-1} + \frac{(-1)^{n-1}}{(n-1)!} \frac{\partial^{(n-1)}}{\partial t^{(n-1)}} \mathbf{A}_1. \end{aligned}$$

Another form of representation of Eq. (26) is of the form [31]

$$\mathbf{T} = -P\boldsymbol{\delta} + \alpha_0\mathbf{A}_1 + \sum_{k=1}^{n-1} \alpha_k \frac{d^{(k)}}{dt^{(k)}} \mathbf{A}_1. \quad (27)$$

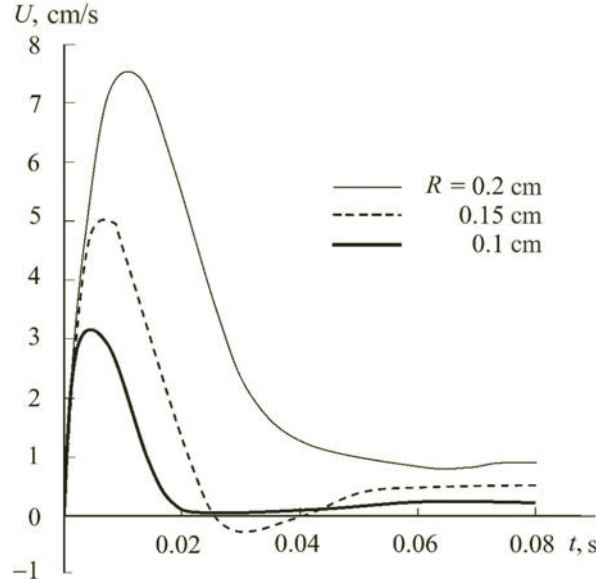


Fig. 3. Variation in the velocity  $U(t)$  of bubbles of radius  $R$  in the viscoelastic fluid:  $\mu_0 = 1.025$  Pa·s,  $\lambda = 0.1$  s, and  $c = 19.5$  [24, Fig. 20].

The constants  $\alpha_k$  characterize the rheological properties of the fluid (in particular,  $\alpha_0 = \mu$ , i.e., the shear viscosity), and the tensor  $\mathbf{A}_1$  is coincident up to a numerical coefficient with the strain-rate tensor  $\mathbf{D}$ :  $\mathbf{A}_1 = 2\mathbf{D}$ .

Since each term of series (26) and (27) that follows makes a smaller contribution to the value of the stress tensor, it is expedient to limit the number of terms without loss of the model's substantial properties, primarily, the reflection of a nonzero difference of the normal-stress components. Following on from this idea, we propose a number of viscoelastic-fluid models preceding the Coleman–Noll generalization.

The *Rivlin–Ericksen model* uses the series of first terms of Eq. (27) to describe the rheological properties of the so-called second-order fluids [32]

$$\mathbf{T} = -P\boldsymbol{\delta} + \mu\mathbf{A}_1 + \mu_1\mathbf{A}_2 + \mu_2\mathbf{A}_1^2, \quad (28)$$

where  $\mu$ ,  $\mu_1$ , and  $\mu_2$  are the shear viscosity, the extensional viscosity, and the cross viscosity of a fluid respectively and  $\mathbf{A}_2 = \frac{d\mathbf{A}_1}{dt} + \mathbf{A}_1 \cdot \nabla \mathbf{U} + \nabla \mathbf{U}^T \cdot \mathbf{A}_1$  is the Rivlin–Ericksen tensor [33]. The constants  $\mu_1$  and  $\mu_2$  are the scalar functions of the invariants of the strain-rate tensor:  $\mathbf{A}_1$ . At  $\mu_2 = 0$  (absence of viscoelastic properties), Eq. (28) becomes identical to the equation of the Reiner–Rivlin model of a stationary non-Newtonian fluid [9]. The Rivlin–Ericksen model gives the basic properties of a viscoelastic fluid: the nonzero difference of the components of normal stresses and the delay of the change in the strain rate on stress relieving [28].

By the values of  $\mu$ – $\mu_2$ , there are expressed the shear stress  $\tau_{12}$  and the differences of the components of normal stresses  $\tau_{11}$ ,  $\tau_{22}$ , and  $\tau_{33}$  of the viscoelastic fluid [30]

$$\tau_{12} = \mu\gamma, \quad (29)$$

$$\tau_{11} - \tau_{22} = N_1 = -2\mu_1\gamma^2, \quad (30)$$

$$\tau_{22} - \tau_{33} = N_2 = (2\mu_1 + \mu_2)\gamma^2. \quad (31)$$

The values of the rheological constants  $\mu$ – $\mu_2$  of a second-order fluid are bounded by the relations following from the thermodynamic condition of the minimum free energy in an equilibrium state [33]

$$\mu \geq 0, \mu_1 \geq 0, \mu_1 + \mu_2 = 0. \quad (32)$$

However, experimental data demonstrate that in many viscoelastic fluids, the values of  $\mu_1$  and  $\mu_2$  do not satisfy the constraints (32). Thus, it was shown in [34] that if  $\mu_1 + \mu_2 > 0$ , the condition  $\mu_1 < 0$  is not observed, and a theorem was also proved that the condition

$$\mu > 0, \mu_1 < 0, \mu_1 + \mu_2 \neq 0 \quad (33)$$

confirmed by experiments yields results not detectable in any fluids at all.

The Rivlin–Ericksen model was used in calculating the flow of a viscoelastic polymer out of the extruder onto a horizontal plane [35]. It has been established that the stresses drop with distance from the extruder axis, and the growth in the  $\mu_1/\mu$  ratio (i.e., in the viscoelastic properties) causes the polymer layer to thicken. Here, the cross viscosity  $\mu_2$  does not affect the velocity field, but affects the pressure distribution in the layer. Using such a model, an analytical solution was obtained for gentle flow of a viscoelastic medium in a curved rectangular duct [36].

The constants of the Rivlin–Ericksen model for polycrystalline ice were determined in experiments on compression of its samples initiating creeping ice mass flow:  $\mu = 4.5 \cdot 10^{13}$  Pa·s,  $\mu_1 = -1.0 \cdot 10^{19}$  Pa·s<sup>2</sup>, and  $\mu_2 = 3.4 \cdot 10^{21}$  Pa·s<sup>2</sup> [37]. A calculation of the creep of an ice mass from a steep slope using the obtained data showed a decrease in the compression of the ice at the boundaries of the duct and a growth in its stretching. The formation of crevasses is contributed to by the normal stresses  $N_1$  and  $N_2$ , with the angle of their action on the duct boundary decreasing, which leads to a slight turn of the direction of formation of crevasses.

Unsteady motion of a Rivlin–Ericksen fluid over an inclined plane when its free surface is acted upon by the tangential force for a finite time has been investigated in [38]. Analytical expressions have been obtained for the fluid velocity as a function of time. In the absence of viscoelasticity, the velocity at the end of the calculation portion tended to zero, and in the presence of it became negative, with the absolute values of the velocity growing with viscoelasticity. This is attributable to the action of internal forces "pulling" the viscoelastic fluid back once the external shearing force ceases to act.

In [39], a modification of the Rivlin–Ericksen model has been proposed in which the rheological coefficients  $\mu_1$  and  $\mu_2$  are dependent in a power-law manner on the shear rate  $\gamma$ :

$$\mathbf{T} = -P\boldsymbol{\delta} + \mu\mathbf{A}_1 + K|\gamma|^{n-1}(\mu_1\mathbf{A}_2 + \mu_2\mathbf{A}_1^2). \quad (34)$$

Equation (34) was used in investigating numerically one-dimensional flow of a viscoelastic fluid in a circular pipe of constant cross section. For a transient regime of flow, the relation between the pressure gradient on the pipe's end portion and the volume flow rate of the fluid was obtained.

Another modification of the Rivlin–Ericksen model has been proposed for description of a mass of crystal ice creeping over the plane with a uniform slope under gravity [40];

$$\mathbf{T} = -P\boldsymbol{\delta} + \mu(\mathbf{A}_1)\mathbf{A}_1 + \alpha(\mathbf{A}_2 + \mathbf{A}_1^2) + \beta(\bar{\mathbf{E}})\bar{\mathbf{E}}. \quad (35)$$

Here  $\mu(\mathbf{A}_1) = K \left( \frac{1}{2} \text{tr} \mathbf{A}_1^2 \right)^{n/2}$  is the shear viscosity of the ice, Pa·s,  $\alpha = \mu_1 = -\mu_2$  is the rheological coefficient reflecting, with account of condition (33), the viscoelasticity and cross viscosity of the ice, Pa·s<sup>2</sup>,  $\beta(\bar{\mathbf{E}}) = \beta_0 \exp \left( -c \frac{1}{2} \text{tr} \bar{\mathbf{E}}^2 \right)$  is the variable shear modulus, Pa,  $\bar{\mathbf{E}} = \mathbf{E} - \frac{1}{3} \text{tr} \mathbf{E} \boldsymbol{\delta}$  is the deviator of the strain tensor, and  $\beta_0$  and  $c$  are the empirical constants characterizing the initial rigidity and rate of damping of elastic strains of the ice [40]. Nonlinear differential algebraic equations of ice motion were solved by the finite-element method. It has been established that model (35) is capable of reflecting complex rheological properties of a moving ice mass that are observed under actual conditions.

The *Giesekus model* is based on a network theory of polymers and takes account of the relation of the strain of polymer molecules to their mobility of a polymer chain [41, 42]:

$$\mathbf{T} + \lambda \left( \frac{d\mathbf{T}}{dt} - \nabla\mathbf{U} \cdot \mathbf{T} - \mathbf{T} \cdot \nabla\mathbf{U}^T \right) + \frac{\alpha\lambda}{\mu} \mathbf{T}^2 = \mu(\nabla\mathbf{U} + \nabla\mathbf{U}^T). \quad (36)$$

In one-dimensional shear flow, expression (36) takes the form

$$\tau + \lambda \frac{d\tau}{dt} + \frac{\alpha\lambda}{\mu} \tau^2 = \mu\gamma. \quad (37)$$

Here  $0 < \alpha < 1$  is the dimensionless factor of anisotropy of the polymer. At  $\alpha = 0$ , Eqs. (36) and (37) become Eqs. (5) and (6) of Maxwell models.



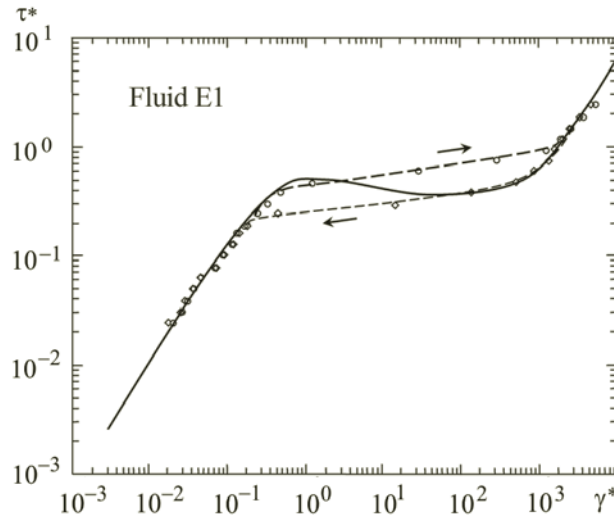


Fig. 4. Stress ( $\tau^* = \tau/G_0$ ) vs. shear rate ( $\gamma^* = \gamma\lambda$ ) of the polymer solution E1: circles/rhomb, experiment on the rise and reduction in the shear rate; solid curve, calculation from the Giesekus model [21, Fig. 5b].

The Giesekus model reflects qualitatively the existing properties of viscoelastic fluids: reduction in the viscosity after the shear, the nonzero second difference of the normal stresses  $N_2$ , and growth in the stress during the shear flow. Notice that the quantity  $N_2$  depends on the structure of the polymer chain: for the linear orientation of polymer molecules, we have  $N_2 = 0$ , and for their entanglement to form loops,  $N_2 \neq 0$  [43].

Shear and elongational or extensional flows of a polymer solution with wormlike micelles have been presented in [21]. The hysteresis of viscosity observed at different temperatures and concentrations of the salt ( $\text{NH}_4\text{Cl}$ ) was manifested as the presence of a portion with negative viscosity ( $d\tau/d\dot{\gamma} < 0$ ) on the flow curves, which corresponded to an unstable state of the solution's internal structure ([21], Fig. 1). The obtained rheometry data were compared with the theoretical prediction of the rheological properties of the solution on the basis of the Giesekus model. Experimental and calculated dependences of the stress on the shear rate are given in Fig. 4. As we can see, the model quite aptly describes the solution's rheological properties, in particular, thixotropy.

Also, the model reflects the expansion of a jet at exit from the nozzle, which is characteristic of viscoelastic fluids. With growth in the velocity of the fluid at entry into the nozzle, the exit expansion of the jet grows; here, for shorter nozzles, the jet expansion turns out to be higher, which is in complete agreement with numerous experimental data [44, 45]. This is due to the fact that when a pre-compressed viscoelastic fluid leaves the nozzle the stored elastic-strain energy is liberated. In short nozzles, the fluid better keeps "memory" of the previous velocity field (the outflow time is shorter or comparable with the relaxation period  $\lambda$ ). Therefore, the expansion turns out to be greater.

The *Johnson–Segalman model* is based on the hypothesis on the presence of two mechanisms of strain in polymer media due to the motion of polymer and solvent molecules. The first mechanism enhances strains, and the second smoothes them out; here, the total strain of the medium can be nonaffine-type [46]. The two motions (of the polymer and of the solvent) are integrated into the state equation

$$\mathbf{T} + \lambda \left\{ \frac{d\mathbf{T}}{dt} - \nabla\mathbf{U} \cdot \mathbf{T} - \mathbf{T} \cdot \nabla\mathbf{U}^T + \frac{1-a}{2} \left[ \mathbf{T}(\nabla\mathbf{U} + \nabla\mathbf{U}^T) + (\nabla\mathbf{U} + \nabla\mathbf{U}^T)\mathbf{T} \right] \right\} = \mu(\nabla\mathbf{U} + \nabla\mathbf{U}^T). \quad (38)$$

We know of various forms of representation of the equation of the Johnson–Segalman rheological model, in particular, using the upper  $\overset{\nabla}{\mathbf{T}}$  and lower  $\underset{\Delta}{\mathbf{T}}$  convection derivatives of the stress tensor  $\mathbf{T}$  [47, 48]

$$\mathbf{T} + \lambda \left[ \left( 1 - \frac{\xi}{2} \right) \overset{\nabla}{\mathbf{T}} + \frac{\xi}{2} \underset{\Delta}{\mathbf{T}} \right] = \mu(\nabla\mathbf{U} + \nabla\mathbf{U}^T). \quad (39)$$

On the substitution of the expressions of  $\overset{\nabla}{\mathbf{T}}$  and  $\underset{\Delta}{\mathbf{T}}$  into (39) and rearrangements, we have



$$\mathbf{T} + \lambda \left[ \frac{d\mathbf{T}}{dt} - \left(1 - \frac{\xi}{2}\right) (\nabla\mathbf{U} \cdot \mathbf{T} + \mathbf{T} \cdot \nabla\mathbf{U}^T) + \frac{\xi}{2} (\nabla\mathbf{U}^T \cdot \mathbf{T} + \mathbf{T} \cdot \nabla\mathbf{U}) \right] = \mu(\nabla\mathbf{U} + \nabla\mathbf{U}^T). \quad (40)$$

The parameters  $a$  or  $\xi$  in Eqs. (38) and (39) are called slip parameters. They are taken to be a measure of nonaffinity of strain manifested as the stretching (slip) of a polymer chain relative to the field of continuum flow. For reduced forms of representation of the model, we have  $|a| < 1$  and  $0 \leq \xi \leq 2$ . Boundary values of the parameters  $a$  and  $\xi$  reduce the Johnson–Segalman model to Maxwell convection models: to the lower model at  $a = -1$  or  $\xi = 0$  and to the upper one at  $a = 1$  or  $\xi = 2$ . In this case the strain nonaffinity disappears and the motions of the polymer and solvent molecules are coincident. At  $|a| < 1$  or  $0 < \xi < 2$ , the polymer chains are "shifted" from the dispersion medium and the viscosity change becomes nonmonotonic. On the stationary flow curve, this corresponds to the portion with negative differential viscosity.

As noted in [49], the Johnson–Segalman model is the simplest model which gives the nonmonotony of the curve of flow of micellar polymer solutions and is in agreement with the observed phenomenon of their shear banding [50].

Using Eq. (39), an analytical dependence has been obtained in [47] for dimensionless components of normal and shear stresses occurring in one-dimensional shear flow of a viscoelastic fluid. This dependence has local extrema, which corresponds to the presence of a maximum and a minimum on the stationary curve of viscoelastic-fluid flow. Thus, the model reflects the nonstability of flow of polymer solutions on their internal restructuring.

A sharp growth in the velocity of flow of micellar polymer solutions (spurt effect), which is observed on flow-induced phase separation [50–52] was numerically modeled by the radial-basis-functions method [5]. The obtained results were in complete agreement with experiments in which a study was made of the velocity jump of a viscoelastic fluid in flowing through extrusion nozzles and correlate with the Johnson–Segalman hypothesis on the nonaffine strain of a viscoelastic medium.

Flow of a Johnson–Segalman fluid in a sharply convergent axisymmetric channel (extrusion nozzle) at high shear rates was investigated numerically by the defect-correction method and the continuation method [54]. Stable equations were obtained at maximum values of the parameter  $\lambda$  of the order of 5 (defect-correction method) and of 10 (continuation method).

The *Phan-Thien–Tanner model* is based on a network theory of polymers and is represented in general form as [55, 56]

$$\mathbf{T} + \lambda \left[ \frac{d\mathbf{T}}{dt} - \nabla\mathbf{U} \cdot \mathbf{T} - \mathbf{T} \cdot \nabla\mathbf{U}^T + \xi(\mathbf{T} \cdot \mathbf{D} + \mathbf{D} \cdot \mathbf{T}) \right] + \mathbf{T}f(\text{tr } \mathbf{T}) = 2\mu\mathbf{D}. \quad (41)$$

The physical meaning and the range of variation of the parameter  $\xi$  ( $0 \leq \xi \leq 2$ ) are in complete agreement with those of an analogous parameter of the Johnson–Segalman model. For slow flows at a low rate of strain of the polymer molecules  $\xi = 0$  [57], the model itself is simplified.

The function  $f(\text{tr } \mathbf{T})$  from Eq. (41) in the original study [55] is represented by the linear relation for the stress tensor  $\mathbf{T}$

$$f(\text{tr } \mathbf{T}) = 1 + \frac{\varepsilon\lambda}{\mu} \text{tr } \mathbf{T}, \quad (42a)$$

where  $\varepsilon$  is the dimensionless parameter of extensibility of the polymer ( $\varepsilon \geq 0$ ). The expression of the function  $f(\text{tr } \mathbf{T})$  in the form [56]

$$f(\text{tr } \mathbf{T}) = \exp\left(\frac{\varepsilon\lambda}{\mu} \text{tr } \mathbf{T}\right) \quad (42b)$$

has become a generalization of formula (42a). Use is also made of the quadratic approximation of the dependence  $f(\text{tr } \mathbf{T})$  [58]

$$f(\text{tr } \mathbf{T}) = 1 + \frac{\varepsilon\lambda}{\mu} \text{tr } \mathbf{T} + \frac{1}{2} \left( \frac{\varepsilon\lambda}{i} \text{tr } \mathbf{T} \right)^2. \quad (42c)$$

At  $\varepsilon = 0$ , in all the cases we have  $f(\text{tr } \mathbf{T}) = 1$  and the Phan-Thien–Tanner model is identical to the Johnson–Segalman model. The Phan-Thien–Tanner model is traditionally used in the calculation of polymer flows. Thus, with it, a comparison was made of the influence of the function  $f(\text{tr } \mathbf{T})$  defined by expressions (42a)–(42c) on the shear and extensional viscosities [58].

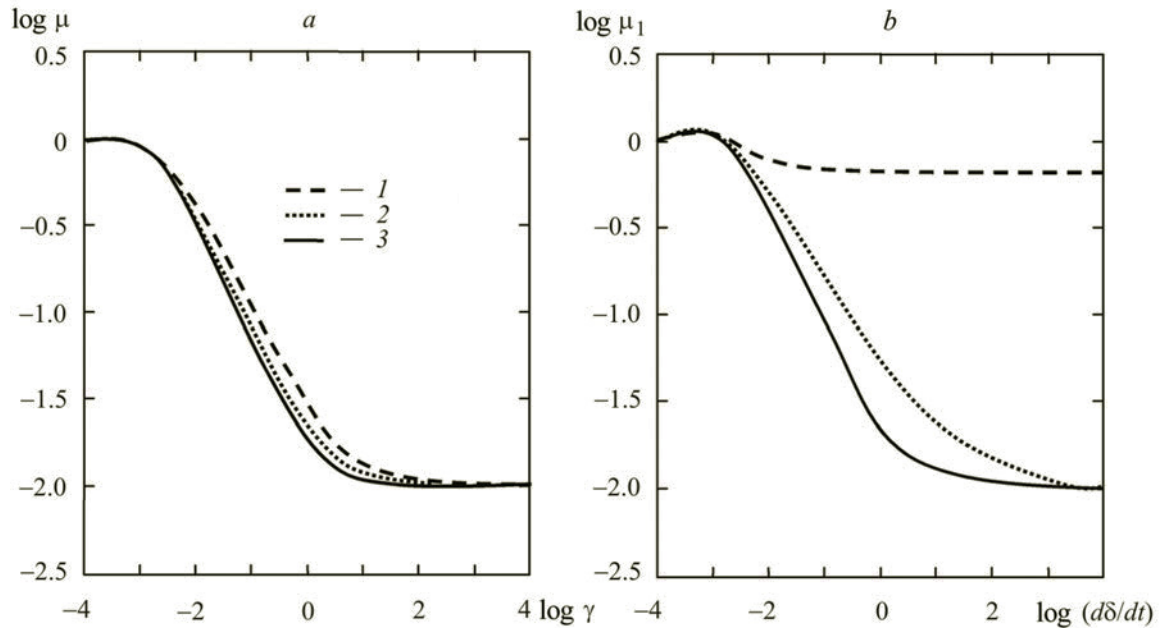


Fig. 5. Variation in the shear  $\mu(\gamma)$  (a) and extensional  $\mu_1(d\epsilon/dt)$  viscosities of the Phan-Thien–Tanner fluid in linear (1), quadratic (2), and exponential (3) approximations of the  $f(\text{Tr } \mathbf{T})$  function. The rheological parameters are  $\varepsilon = 1$ ,  $\xi = 0$ , and  $We = 200$  [58, Figs. 2d and 3d].

Notice that although the shear and extensional viscosities alike represent the ratio of the stresses to the rate of strain of a medium, however, there is a fundamental difference between them: the first is defined in the case of one-dimensional shear strain, and the second, under volume strain. In [58], the values of  $\gamma$  and  $d\epsilon/dt$  were calculated from the formula

$$\gamma = 2\sqrt{I_{2D}} \text{ ,} \quad (43)$$

$$d\epsilon/dt = 3I_{3D}/I_{2D} \text{ .} \quad (44)$$

As we can see from Fig. 5, the linear and quadratic approximations of the function  $f(\text{tr } \mathbf{T})$  provide a good approximation in calculating the shear viscosity throughout the shear-rate range. At the same time, the linear approximation overstates the values of extensional viscosity by nearly two orders of magnitude with increase in the strain rate. The result is attributed to the distinctive features of the response of the viscoelastic fluid to shear (one-dimensional) and volume (three-dimensional) strains. In the first case the linear function quite accurately describes three-dimensional stresses arising in the fluid. In the case of volume flow with a sufficient velocity this approximation cannot take correct account of the nonlinear relation of nonisotropic stresses to the strain rate. Thus, selection of the form of the function  $f(\text{tr } \mathbf{T})$  in the Phan-Thien–Tanner model must be consistent with the character of viscoelastic-fluid flow.

In [59], visualization of the velocity field and numerical modeling with a simplified Phan-Thien–Tanner model (the parameter  $\xi$  in Eq. (41) was taken to be zero) were used in the comparative investigation of flow of a viscoelastic fluid (0.075% polyethylene oxide (PEO) solution in a glycerin/water mixture (60/40%)) in T-shaped microchannels of cross section  $50 \times 50 \mu\text{m}$ .

Determination of the constants of the Phan-Thien–Tanner model in [59] is of special interest. The value of  $\mu$  was traditionally found from the flow curve as the value of viscosity before its decrease with growth in the shear rate ( $\mu_{\text{PEO}} = 19.5 \text{ mPa}\cdot\text{s}$  at  $\gamma \leq 15 \text{ s}^{-1}$ ). The relaxation time  $\lambda$  was determined by the method of capillary longitudinal rheometry ( $\lambda_{\text{PEO}} = 66 \pm 4 \text{ ms}$ ), and also from the flow curve from the shear rate  $\gamma$  at the beginning of viscosity reduction ( $\lambda_{\text{PEO}} = 1/\gamma = 1/15 \approx 67 \text{ ms}$ ). The model's third parameter, i.e., the extensibility of a polymer  $\varepsilon$  which characterizes its extensional viscosity was evaluated from the representation of polymer chains as thinning fibers whose radius decreases linearly with time when their full extension is attained. The range of variation in the values of the parameter  $\varepsilon$  was determined by the laser-micrometry method and was  $0 \leq \varepsilon \leq 7.0 \cdot 10^{-6}$  [59].

Not only are numerical solutions of viscoelastic-fluid flows found using the Phan-Thien–Tanner model, but also analytical ones [57, 60–65], which makes it possible to assess the correctness of results of numerical calculations. Thus, steady-state flow in a plane channel and a pipe was calculated in [60, 63] at the linear and exponential function  $f(\text{tr } \mathbf{T})$ . It has been shown that shear stresses on channel walls are substantially lower than those for a Newtonian or viscoelastic fluid whose properties are approximated by the Maxwell upper convection model.

In [61, 62], consideration was given to annular flows of a viscoelastic fluid using the linear function  $f(\text{tr } \mathbf{T})$ . Expressions have been obtained for the axial velocity, the shear and normal stresses, and the dynamics of their change in the radial direction with account taken of the flow rate of the fluid and its rheological parameters, and also of the geometry of the flow zone.

Steady-state flow of a Phan-Thien–Tanner fluid in the gap between rotating coaxial cylinders was considered in [64]. The obtained velocity field takes account of the ratios of the radii of the inner and outer cylinders and of their angular velocities. It has been established that a growth in the fluid's elasticity (parameter  $\varepsilon$ ) increases the gradient of tangential velocity in the inner cylinder and reduces the value of the viscosity  $\mu$ , i.e., the fluid exhibits pseudoplasticity.

The problem of flow of a Phan-Thien–Tanner fluid between plane plates arranged at a small angle has been solved analytically in [65]. A motive force of the flow was provided by the movement of the lower horizontal plate with a constant rate, which is characteristic of friction assemblies of a number of mechanisms. The obtained results point to the strong influence of viscoelastic properties of the fluid on the profile of horizontal velocity and on the variation in the pressure gradient along the channel length. Also, the pressure gradient is dependent on the angle of mutual inclination of the plates: when they are parallel it is equal to zero and it grows with inclination angle and has, along the channel length, a maximum shifting from the center of the channel to its convergent end [65, Fig. 3].

**Conclusions.** Differential models of rheologically nonstationary fluids, which reflect an instantaneous relation of the stresses to the strain rate, were historically developed on the basis of the phenomenological approach for a certain "universal" viscoelastic fluid (Maxwell and Coleman–Noll models). Today's differential models of rheologically nonstationary fluids are being developed more specifically for concrete types of viscoelastic fluids (primarily, polymer melts and solutions). These models (of Giesekus, Johnson–Segalman, Phan-Thien–Tanner, and others), preserving a traditional mathematical form, are based on a network theory of polymers and take account of the mechanisms of strain of macromolecules in polymer media through the introduction of additional parameters. The above trend is due to the increasing and growing use of polymer and composite materials; this requires that the processes and technologies of their processing with conversion into products be upgraded and accordingly the theory, in particular, of rheological models of polymers, be developed [66].

On the whole, the development of differential models of rheologically nonstationary fluids follows the path of their complication, which makes it possible to allow for increasingly subtler effects. Here, it is expedient to ensure the possibility of reducing a complex rheological model to existing models and thus finding analytical solutions for a number of the simplest flows. This approach ensures the verification of the novel model and improves the reliability of results obtained with it.

## NOTATION

$\mathbf{D}$ , strain-rate tensor,  $s^{-1}$ ;  $\mathbf{E}$ , Cauchy–Green strain tensor;  $G$ , dynamic (complex) modulus of the viscoelastic fluid, Pa;  $G'$  and  $G''$ , storage (elastic) and energy-loss (dissipation) moduli of the viscoelastic fluid, Pa;  $I_{2D}$  and  $I_{2T}$ , second invariants of the strain-rate ( $s^{-2}$ ) and stress ( $\text{Pa}^2$ ) tensors;  $K$ , measure of consistency of a non-Newtonian fluid,  $\text{Pa}\cdot s^n$ ;  $N_1$  and  $N_2$ , first and second difference of normal stresses of the viscoelastic fluid, Pa;  $n$ , index of non-Newtonian fluid flow;  $P$ , pressure, Pa;  $\mathbf{T}$ , stress tensor, Pa;  $\nabla \mathbf{U}$  and  $\nabla \mathbf{U}^T$ , velocity-gradient tensor and its conjugate tensor,  $s^{-1}$ ;  $t$ , time, s;  $We$ , Weber number;  $\gamma$ , shear rate (one-dimensional analog of the strain-rate tensor),  $s^{-1}$ ;  $\delta$ , Kronecker symbol (unit tensor);  $\delta$ , angle of displacement of the stresses from the strain of the viscoelastic fluid, rad;  $\varepsilon$ , relative strain;  $\lambda$  and  $\theta$ , constants of relaxation and delay of the viscoelastic fluid, s;  $\mu$  coefficient of dynamic viscosity of a Newtonian fluid,  $\text{Pa}\cdot s$ ;  $\tau$ , shear stress (one-dimensional analog of the stress tensor), Pa;  $\tau_0$ , initial shear stress, Pa;  $\omega$ , angular velocity,  $s^{-1}$ .

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