COMPUTATIONAL
CONTINUUM MECHANICS

ВЫЧИСЛИТЕЛЬНАЯ
МЕХАНИКА СЛОШНЫХ СРЕД

Computational Continuum Mechanics. 2021. Vol. 14. No. 1. pp. 12-29 DOI: 10.7242/1999-6691/2021.14.1.2

REFINING RHEOLOGICAL MODEL FOR DESCRIPTION OF LINEAR AND NONLINEAR VISCOELASTICITY OF POLYMER SYSTEMS

A.A. Laas[1], M.A. Makarova[2], A.S. Malygina[2], G.O. Rudakov[2] and G.V. Pyshnograi[1,2]

[1]*Altai State University, Barnaul, Russian Federation*
[2]*Altai State Technical University named after I.I. Polzunov, Barnaul, Russian Federation*

The article reviews the current mesoscopic modeling of flows for polymer solutions and melts of various structures. It also demonstrates the unified structure of the rheological constitutive relation, up to a certain dissipative function. Linear and nonlinear effects are considered for the simple shear and uniaxial tension of polymer melts based on the modified Vinogradov and Pokrovskii rheological model. The model can describe with adequate accuracy the linear viscoelasticity of fluid polymer systems, as well as transient processes at the shear flow and uniaxial elongation. It was compared with other experimental data for the melt of an industrial polyethylene sample. The nonlinear viscoelastic response of a polymer material was modeled at large periodic deformations. The calculation results were obtained via Runge–Kutta approach with integrated subprograms of MATLAB IT environment. They were later compared with the experimental data for the solution of polyethylene oxide in dimethyl sulfoxide. The nonlinear behavior of the polymer sample manifested itself in the distortion of the viscoelastic response of the material at sinusoidal oscillations. In this case the shear stresses are no longer the correct harmonicas and a "step" appears at the leading edge of the response. Besides, their amplitude is not proportional to the amplitude of the shear. The paper also considers the superposition of the oscillating shear flow on the simple shear. The distortion of the viscoelastic response is observed here though, unlike the high-amplitude periodic deformation, the curvatures of the upper and lower half-waves of the response differ. Since there is still no experimental data for this case, this work expects to encourage the researchers working in the field of nonlinear viscoelastic properties of polymer solutions and melts for further endeavors.

Key words: rheology, polymers, mesoscopic approach, viscoelasticity, nonlinear effects, simple shear, uniaxial elongation, solutions, melts, multimode rheological model

1. Introduction

Studying the nonlinear behavior of viscoelastic liquids is an essential task in the mechanics of continua. Even the basic experiments reveal nonlinear effects, for instance, in the simple shear flow, oscillating shear flow or at uniaxial elongation. The results of the research are often of purely practical meaning since they provide optimal modes of production storage, processing and delivery. Theoretically, the data on nonlinear effects is valuable as one may test the employed rheological models, choose the most adequate one and later implement the estimate for the liquid flow under study.

Several rheological models are mentioned in literature. They describe nonlinear effects at uniaxial elongation and simple shear quite precisely. However, there is a need for carrying out some calculations and further interpreting the results of rheological measurements in the flows of a more complex structure, namely, at shear oscillations of large amplitude or at the superposition of stationary shear and oscillating flows. For this purpose studying complicated modes requires applying new mathematical methods for processing the measurement data within the framework of employed rheological models.

2. Mathematical modeling in the rheology of polymer media

The application of the data accumulated in science on the rheology of polymer solutions and melts to the analysis of their production processes is a complex and substantial task both scientifically and practically. Even its partial solutions provide for the products of specified physical-mechanical and structural characteristics. Polymer liquids (solutions or melts) possess a complex entangled structure. Thus, for their modeling they use rheological defining relations of several time and spatial scales. Until now there the defining equations that could generalize all the practicalities of polymer media flows have not yet been created. Researchers and engineers use the equations which describe either the polymer flow peculiarities they are keen on or some

---

Email addresses for correspondence: aleksandr_laas@mail.ru

particularcases. It must be noted that numerous defining relations have been offered, but only some of them are applied to solve the tasks on polymer processing.

Traditionally, rheological models verification [1–9] begins with considering the modes of stationary shear deformation on their basis and periodic oscillations of small amplitude. Nevertheless, these simple deformation types are not sufficient to represent the processes in the industrial processing of polymers, when substantial non-stationary deformation occurs [10–16]. Therefore, the next stage to develop a rheological model meant including nonlinear effects, namely, large periodic deformations [10, 11], which greatly expands the possibilities to explore various types of flows of polymer solutions and melts in industrial equipment units. The development of models describing nonlinear viscoelasticity is getting more and more sought-after [13–16].

Dynamic shear oscillating tests have been widely used in rheology of liquids with special features such as polymer solutions and melts, biopolymers, polyelectrolytes, suspensions, emulsions, gels etc. Just to name a few, measuring rheological features of low-amplitude oscillating flows Small-Amplitude Oscillatory Shear (SAOS) is a classical way to construct linear viscoelasticity relations for liquids. It has been studied thoroughly both theoretically and practically [2]. However, as it was stated above, the majority of real processes are accompanied by serious non-stationary deformation, which requires studying nonlinear dynamic features of the systems under study. In order to picture better the nonlinear behavior of the polymer liquid, Large-Amplitude Oscillatory Shear (LAOS) can be applied. Experimentally both SAOS and LAOS are characterized by two parameters at the entrance: relative amplitude of shear oscillations γ_0 and their frequency ω. However, there are substantial differences in the response of the polymer system to periodic oscillations defined by the features of the material. For SAOS the response looks like a sinusoid and the amplitude of shear vibrations is directly proportional γ_0. SAOS major features are $G'(\omega)$ — elasticity modulus and $G''(\omega)$ — loss modulus. For LAOS the response of the system does not look like a proper sinusoid and the amplitude for such vibrations does not grow strictly proportionally at growing γ_0. That is confirmed by experiments [17–29].

The first works devoted to LAOS method [17–29] date back to the middle of the XXth century. While studying shear stresses, Fourier analysis of nonlinear viscoelastic response was carried out [18–25] and researchers faced significant technical challenges: when calculating high-order harmonics it was noticed that first signal harmonics being the components of the dynamic shear module, decrease at increased vibration amplitude. At that time, they presented a clear way to describe nonlinear viscoelastic deformations using Lissajous patterns [24]. This visualization technique is an alternative to the construction of time dependences of shear stresses at various deformation values [11]. Nowadays LAOS method is applied to verify rheological models while studying nonlinear deformations [11, 27].

Among numerous rheological models, it is worth noting those that provide for adequate dependences when studying both linear viscoelasticity at small periodical deformations and nonlinear effects at large deformations. These are Oldroyd-B models [30], Phan-Tien–Tanner ones (PTT) [7], Giesecus model [3], Leonov-Prokunin model [4, 31], «Pom–Pom» model [5, 8, 9], as well as the modified Vinogradov-Pokrovskii model [1, 6, 13–15]. We will carry out calculations in this work based on the latest model in order to check its applicability to describe the changes of polymer liquid behavior at increased deformation.

The major adequacy criterion for the rheological model is the ability to mathematically depict viscometric flows. Multimode model modifications which consider the multitude of relaxation processes provide for the most accurate representation. In a simple case of calculating linear viscoelasticity at low-speed deformation all the above mentioned models are reduced to identical expressions for the components of the dynamic module. Thus, no significant differences are observed between models in the linear area. Nevertheless, at increased deformation speed and transferring to the nonlinear area the models demonstrate different results.

First, there are different approaches to the construction of a rheological model: phenomenological and microstructural. Within the first approach, the object in question is considered apart from its physical nature and the experiment is the only way to define its rheological characteristics and ties between them. For instance,

- Maxwell model [32] shows a spring (an elastic element) connected in sequence with a force plunger (a viscous element) [2]. Within this model for the polymer exposed to constant load, the stress relaxation – time dependence looks exponential. Maxwell model can be applied only in the area of linear viscoelasticity;
- Oldroyde model [30] generalizes Maxwell model for linear viscoelastic liquid flows, which is accompanied by a transfer of viscoelastic stresses towards the flow. This model is also applicable to describe only linear viscoelasticity;
- Leonov-Prokunin model [4, 31] is based on the principles of non-equilibrium thermodynamics. Elastic deformations are characterized by the equilibrium thermodynamic state of the polymer system. Today the model is employed less frequently than others due to a complex transfer to the 3D flow calculations.

The second approach to the construction of rheological defining relations is based on the mesoscopic approximation which rests upon the dynamics of the isolated macromolecule moving in the medium of solvent and other macromolecules. The following models are mesoscopic: «Pom–Pom» model (PP), PTT models, Gysecus model etc. Mathematically, they are models in the differential form.

Giesecus model [3] presented in 1982 is widely used nowadays. The model describes the polymer medium flow by a nonlinear differential equation that contains one parameter, which limits its applicability.

PTT rheological model [7] is one of the first models to adequately represent stationary and non-stationary viscosimetric functions at simple shear and uniaxial elongation. Namely, when setting the simple shear flow, non-zero components of the stress tensor demonstrate non-monotonous behavior.

A lot of differential equations (for instance, PTT model, Gysecus model, extended «Pom–Pom» (extended PP), Leonov-Prokunin model) presuppose the following tie between the extra stresses tensor $\boldsymbol{\tau}$ and symmetric tensor $\mathbf{c}$ which shapes the structure of the studied material at some approximation:

$$\boldsymbol{\tau} = G(\mathbf{c} - \mathbf{I}),$$

where G is a scalar (null tensor) equal to the dimension of the elasticity module and $\mathbf{I}$ — a unit tensor.

Generally, for one relaxation process the rheological model equation can be written in tensor form (tensor-like):

$$\frac{d\mathbf{c}}{dt} - \nabla\mathbf{v}\mathbf{c} - \mathbf{c}\nabla\mathbf{v}^{\mathrm{T}} - \frac{\xi}{2}(\boldsymbol{\gamma}\mathbf{c} + \mathbf{c}\boldsymbol{\gamma}) + \frac{1}{\lambda}\mathbf{H}(\mathbf{c}) = 0, \qquad (1)$$

where $\mathbf{v}$ — a vector of liquid flow speed, $\boldsymbol{\gamma}$ — a symmetric tensor of deformation speed, ∇ — a vectorial differential operator, $\mathbf{H}(\mathbf{c})$ — a dissipative term dependent on structural tensor $\mathbf{c}$, λ — relaxation time. Parameter ξ describes the non-affine deformation ($0 \le \xi \le 2$). When reaching peak values 0 and 2 the tensor derivative possesses the contravariant ($\xi = 0$, the highest convective derivative) and covariant ($\xi = 2$, the lowest convective derivative) forms correspondingly. For many polymer systems parameter ξ takes the zero value. On the one hand it reduces the number of nonlinear model parameters as well as obtains the oscillations of transient responses at high shear rates. On the other hand, nonzero value ξ contributes to a more drastic increase in viscosity at uniaxial elongation. For example, Giesecus and Leonov-Prokunin models under study in this work presuppose that congruence $\xi = 0$, unlike the exponential PTT model. The particular forms of dissipative terms $\mathbf{H}(\mathbf{c})$ for the models under study are given in Table 1.

Table 1. Dissipative term $\mathbf{H}(\mathbf{c})$ for some models (presupposing $\xi=0$).

	The form of the dissipative term	The number of parameters together with linear	Nonlinear parameters
Oldroyd [28]	$(\mathbf{c}-\mathbf{I})$	2	–
Exponential PTT [7]	$\exp\left[\zeta\left(\operatorname{tr}\mathbf{c}-3\right)\right]\left(\mathbf{c}-\mathbf{I}\right)$	3	ζ
Lonov-Prokunin [4]	$\frac{b}{2}\left(\mathbf{c}^2-\frac{\operatorname{tr}\mathbf{c}-\operatorname{tr}\mathbf{c}^{-1}}{3}\mathbf{c}-\mathbf{I}\right)$	4	ζ , ν
Giesecus [3]	$\beta(\mathbf{c}-\mathbf{I})^2+\mathbf{c}-\mathbf{I}$	3	β
Pom–Pom (PP) [5, 9]	$F_c\mathbf{c}-\mathbf{I}$	3	β
Extended Pom–Pom (XPP) [8]	$\beta\left(\mathbf{c}-\mathbf{I}\right)^2+F_x\mathbf{c}-\mathbf{I}$	5	β , λ_S , q
Modified Vinogradov-Pokrovskii (mVP) [6]	$\beta\left(\mathbf{c}-\mathbf{I}\right)^2+\left(1+\left(\kappa-\beta\right)\operatorname{tr}\left(\mathbf{c}-\mathbf{I}\right)/3\right)\left(\mathbf{c}-\mathbf{I}\right)$	4	κ , β

Table 1 also uses the following labeling: $\operatorname{tr}\mathbf{A}$ — matrix/tensor trace $\mathbf{A}$; β and κ — induced anisotropy parameters; $b=\exp\left[-\zeta\sqrt{\operatorname{tr}\mathbf{c}-3}\right]+\dfrac{\operatorname{sh}\left[\nu\left(\operatorname{tr}\mathbf{c}-3\right)\right]}{\nu\left(\operatorname{tr}\mathbf{c}-3\right)+1}$, where ζ and ν — viscosities of the polymer melt (solution) under study; $F_x=2\dfrac{\lambda}{\lambda_S}\exp\left[\dfrac{2}{q}(\mathbf{\Lambda}-1)\right]\left(1-\dfrac{1}{\mathbf{\Lambda}}\right)+\left(\dfrac{1}{\mathbf{\Lambda}}\right)^2\left[1-\beta-\dfrac{\beta}{3}\operatorname{tr}\left(\mathbf{c}^2-2\mathbf{c}\right)\right]$, $F_c=\left(2-\dfrac{1}{\sqrt{\mathbf{\Lambda}}}\right)\exp\left[\beta\left(\mathbf{\Lambda}-1\right)\right]$, where $\mathbf{\Lambda}=\sqrt{\operatorname{tr}\mathbf{c}/3}$.

The contents of table 1 mean that the majority of rheological defining relations are of similar structure. The rheological models themselves differ due to dissipative term $\mathbf{H}(\mathbf{c})$ and, despite some discrepancies, describe the following experimental phenomena: shear viscosity anomaly, gradient dependences of the first and second-order differences for shear stresses, non-monotonous (going through the maximum) decrease of stationary viscosity at uniaxial elongation. As stated in [16], the estimation of dimensional parameters G and λ is carried out quite easily though the values of nonlinear parameters have to be sorted out “manually”. It is easy for one parameter, but when there are more, some difficulties may arise while applying models to particular polymer liquids [9, 16, 33, 34].

3. Modeling in linear viscoelasticity

Linear viscoelasticity is one of the most studied areas in theoretical and applied rheology [1–5, 35]. Traditionally, the research means formulating equations for classical viscoelastic Kelvin-Voight [36] and Maxwell [32] bodies and analyzing their behavior in the simplest transient modes, including a) deformation changes at constant preset stress; b) stress changes at constant preset deformation or constant deformation rate; c) periodic deformation due to the preset harmonic law [1–5]. Besides, the analysis does not present any challenge since the obtained equations (or their systems) are common linear differential equations that assume analytical solutions.

To fully describe the responses of continuous medium to applied impacts, the notion of relaxation spectrum [1–3] is introduced. It appears to be a discrete set of relaxation times and their corresponding elasticity modules. The task is to define the relaxation spectrum components according to the results of rheological measurements. For the relaxation times and elasticity modules one may formulate expressions that depend on the number of the relaxation process, molecular mass and polymer concentration. Calculations due to such expressions comply well with the experimental data responding to the mode of small oscillating deformations for the samples of various molecular masses [3, 16, 34]. The number of considered relaxation processes may be quite high, within the order of hundreds. However, it does not mean any difficulty with calculations as we deal with analytical expressions. Although, trying to generalize results from the linear viscoelasticity area for the nonlinear effects, one faces some obstacles [14]. In nonlinear cases the equations are quite complex and require a numerical solution.

At a large number of relaxation processes, it consumes a lot of operating time. Therefore, now it is getting topical to model the relaxation spectrum for polymer systems when the number of relaxation processes under consideration is finite [37, 38].

As a rule, to consider several relaxation processes in the polymer system they resort to the multimode approximation of the rheological model [12–18]. Thus, the point is to define the number of modes that will be sufficient for evaluating the constructed relations of linear viscoelasticity.

All the parameters included into the rheological model can be divided into two groups: those responsible for linear viscoelastic behavior of the system and those responsible for nonlinear effects. Low deformation rates correspond to the areas of linear viscoelasticityю Besides, in many rheological models the expressions are similar [1, 2, 34] and may contain a large number of components.

The relations for the linear viscoelasticity area are easily obtained from (1). The majority of multimode rheological models in their tensor form are presented as following in linear approximation due to speed gradients and preciseness to the used values [1, 11]:

$$\begin{gathered} \boldsymbol{\sigma} = -p\mathbf{I} + 3\sum_{\alpha} \frac{\eta_\alpha}{\tau_\alpha} \boldsymbol{a}^\alpha, \\ \frac{d}{dt}\boldsymbol{a}^\alpha + \frac{1}{\tau_\alpha}\boldsymbol{a}^\alpha = \frac{2}{3}\boldsymbol{\gamma}. \end{gathered} \qquad (2)$$

Here: $\boldsymbol{\sigma}$ — a total stress tensor; p — hydrostatic pressure; η_α и τ_α — sets of initial values for shear viscosities and relaxation times of the considered processes ($\alpha = 1, 2, \ldots, n$); $\boldsymbol{a}^\alpha$ — a set of dimensionless symmetrical second-rank tensors. In fact, these equations are the multimode approximation of Maxwell model [2, 38].

At a simple oscillating shear flow the components of tensor $\boldsymbol{\gamma}$ are connected with the amplitude of shear rate v_{12}^* as following:

$$(\gamma_{ij}) = \begin{pmatrix} 0 & v_{12}^*/2 & 0 \\ v_{12}^*/2 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \cos \omega t,$$

where t — is current time, ω — oscillation frequency. After substituting this expression into (2) and solving the obtained differential equations one will easily find the expressions for frequency dependences of the dynamic module components: shear module and loss module which are important features for linear viscoelasticity and look as following [1, 2, 14, 15]:

$$G'(\omega) = \sum_{\alpha=1}^{n} \frac{\omega^2 \tau_\alpha \eta_\alpha}{1 + (\omega\tau_\alpha)^2}, \qquad G''(\omega) = \sum_{\alpha=1}^{n} \frac{\omega \eta_\alpha}{1 + (\omega\tau_\alpha)^2}. \qquad (3)$$

To compare the data calculated due to expressions (3) with the experimental data, we should refer to works [33, 38], which studied rheological features of the industrial polyethylene melt and which provided the details of the experiments held. Rheological parameter values g_α and τ_α will be taken from these works (see Table. 2). Necessary values η_α are defined according to formula $\eta_\alpha = g_\alpha \tau_\alpha$.

Table 2. Dimensional rheological parameters g_α and τ_α for the sample of LDPE Bralen polyethylene melt.

α	1	2	3	4	5	6	7	8	9	10
g_α, Pa	121440	35292,2	33442,9	19480,3	11923,4	5763,63	2574,63	800,865	213,412	34,6864
τ_α, c	0,00134	0,0052	0,02015	0,07804	30,23	1,17104	4,53626	17,5722	68,0695	263,681

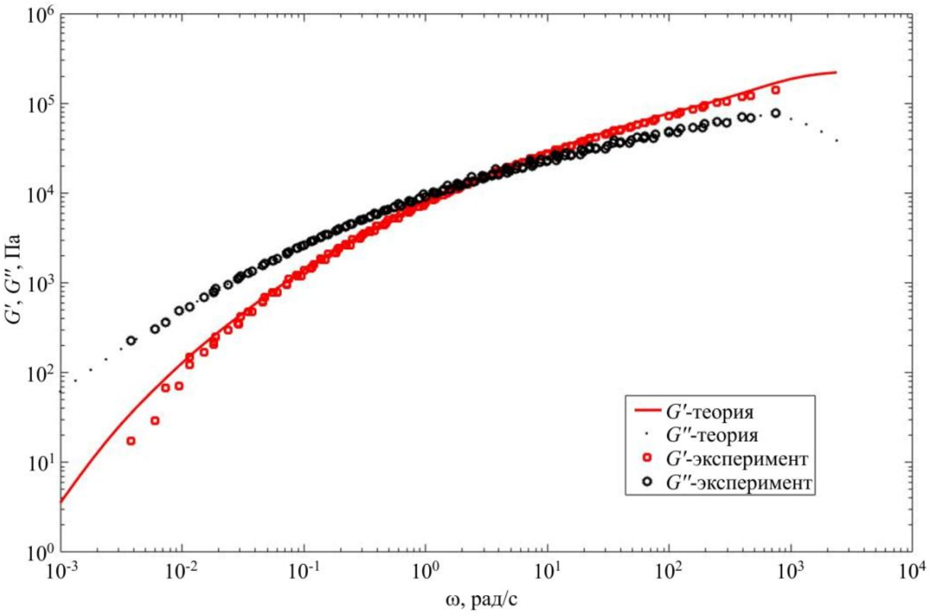

Fig. 1. To the comparison of calculated frequency dependences of elasticity module G' and loss module G'' with the experimental data for the sample of LDPE Bralen polyethylene melt.

The results are presented in Figure 1. Figure 1 shows that expressions (3) describe the dependences of dynamic module components in quite a wide impact frequency range.

4. The multimode modified Vinogradov-Pokrovskii rheological model

One of the major tasks in polymer rheology is to consider the multiplicity of relaxation times. Employing the multimode approximation is required even when studying the dynamics of melted solutions of monodisperse polymers [31, 36–41], which is due to the structure of the polymer molecule resembling a long chain. According to calculations, the mode relaxation time reduces as the square of its number, so it has so-called “Rouse” behavior. However, as stated in [1, 2, 34], the higher the concentration of the polymer solution, the more tangled and coupled the molecules-chains are. The dynamics of such a system gets considerably more complicated [1, 2] since extra “long-range” interactions are formed between the macromolecule segments. As a result, “super-slow” relaxation processes appear in the system and new components emerge in the expression for the stress tensor [35, 36, 40–42].

Should the polymer be polydisperse or branched, it is getting essential to consider the multitude of relaxation times. Besides, the dependence type of the relaxation time for each process deviates from the “Rouse” type and gets more complex. Therefore, it is particularly topical to opt for a reliable and quite a basic model in order to describe the rheology of concentrated polymers. Within the mesoscopic approach, a polymer molecule is presented as a number of beads (macromolecule segments) tied by elastic springs. Such a system will be characterized by the beads friction coefficient and the balanced size of the macromolecule coil. When studying the dynamics of concentrated polymer solutions and melts, the model needs to be extended with the parameters that consider if molecules are entangled or “hooked”.

One of these models seems to be the modified Vinogradov-Pokrovsky rheological model [12–15, 42–44] or so-called mVP model [46–48]. The model of the bead friction coefficient includes the tensor considering anisotropy induced by flow and defined by sizes and shapes of macromolecule coils. Works [13, 47] contain the multimode generalization of the mVP model and it is defined by the following expressions:

$$\boldsymbol{\sigma} = -p\mathbf{I} + 3\sum_{\alpha} \frac{\eta_\alpha}{\tau_\alpha}\left(\boldsymbol{a}^\alpha - \frac{1}{3}\left(\operatorname{tr}\boldsymbol{a}^\alpha\right)\mathbf{I}\right), \tag{4}$$

$$\frac{d}{dt}\boldsymbol{a}^\alpha - \nabla\mathbf{v}\boldsymbol{a}^\alpha - \boldsymbol{a}^\alpha\nabla\mathbf{v}^{\mathrm{T}} + \frac{1+(\kappa-\beta)\operatorname{tr}\boldsymbol{a}^\alpha}{\tau_\alpha} = \frac{2}{3}\boldsymbol{\gamma} - \frac{3\beta_\alpha}{\tau_\alpha}\boldsymbol{a}^\alpha\boldsymbol{a}^\alpha .$$

Here: p — hydrostatic pressure; $\boldsymbol{\sigma}$ — a stress tensor of the polymer system; $\nabla\mathbf{v}$ — a velocity gradient tensor; α — the mode reference number; n — a number of relaxation modes or processes under consideration; $\boldsymbol{a}^\alpha$ — a symmetrical anisotropy tensor corresponding to the contribution of the relaxation mode number α; $\operatorname{tr}\boldsymbol{a}^\alpha$ — a trace of a dimensionless tensor of secondary stresses $\boldsymbol{a}^\alpha$; η_α — a shear viscosity coefficient for mode number α; τ_α — relaxation time for mode number α .

In the multimode generalization the dependence of induced anisotropy parameters β_α and κ_α on $\operatorname{tr}\boldsymbol{a}^\alpha$ is defined by the following expressions [46, 47]:

$$\beta_\alpha\left(\operatorname{tr}\boldsymbol{a}^\alpha\right) = \frac{f_\alpha + p_\alpha \operatorname{tr}\boldsymbol{a}^\alpha}{1+\sqrt{p_\alpha \operatorname{tr}\boldsymbol{a}^\alpha}}, \qquad \kappa_\alpha = 1{,}2\,\beta_\alpha . \tag{5}$$

Obviously, model (4), (5) differs from the one applied in [12–15, 37] by the way induced anisotropy is presented. First works [6, 12, 14, 15, 43] supposed that β and κ are constant and their values do not depend on the molecular mass and polymer concentration. Later (see [13]) when noting the multimode approximation of the model, these parameters were supposed to be tied with the first invariant of the tensor of secondary stresses by correlations different from (5):

$$\beta_\alpha\left(\operatorname{tr}\boldsymbol{a}^\alpha\right) = \frac{f_\alpha + p_\alpha \operatorname{tr}\boldsymbol{a}^\alpha}{1+p_\alpha \operatorname{tr}\boldsymbol{a}^\alpha}, \qquad \kappa_\alpha = 1{,}2\beta_\alpha . \tag{6}$$

According to the calculations, expressions (6) though providing for non-monotonous (going through the maximum) dependence of stationary viscosity at uniaxial elongation on elongation velocity, wrongly predict its asymptotic behavior. Due to (6), stationary viscosity at uniaxial elongation goes to its stationary value at increased velocity, while using expressions (5) given in [46] it goes through the maximum and decreases which corresponds better to experimental data [16, 31, 37, 38]. Now we deal with a new version of mVP model, it is necessary to recalculate viscometric functions considering expressions (5), which will be implemented in the next paragraph of the paper. Thus, model (4), (5) contains a set of four parameters: η_α, τ_α, f_α, p_α, which values should be chosen proceeding from experimental data.

In order to choose the parameters at uniaxial elongation mode the impact of each particular mode has been analyzed. Stationary viscosity dependence was split into separate areas and parameter values f_α and p_α [13, 48] were defined for each area. This research has shown that f_α and p_α are monotonous functions of mode α, and they may be described as following:

$$f_\alpha = \frac{B}{1+(\alpha-\alpha_0)^2}, \qquad p_\alpha = \frac{P}{1+(\alpha-\alpha_0)^2}. \tag{7}$$

Here α_0 — the parameter which rules the monotonous or non-monotonous behavior of f_α and p_α dependence on mode number α. Consistent with work [46], opt for $\alpha_0 = 1{,}5$. Applying expression (5) narrows the number of non-linear parameters for the rheological model down to two: B and P. Besides, mode number α cannot be directly connected to molecular polymer features as for the majority of multimode generalizations for rheological correlations. Therefore, the acceptance of expressions (5) must be evaluated by the theoretical data correlation with experimental data.

5. Viscometric functions at simple shear and uniaxial elongation

In expressions (7) new parameters B and P are introduced. Paper [13] presents the evaluation of their impact on stationary shear viscosity and viscosity at uniaxial elongation and it is shown that stationary viscosity at elongation is the non-monotonous function of shear velocity: the value of its maximum increases at the decrease of parameter B. However, at $P = 0$ the function becomes monotonous. Therefore, the values of parameters B and P impact considerably the dependence types of stationary shear viscosity on shear velocity and stationary viscosity at elongation. Sorting out these values one may get quite a good correlation between calculated and experimental curves [33, 38].

For Bralen polyethylene samples and for further calculations due to model (4), (5) the following parameter values were chosen: $B = 0{,}95$, $P = 0{,}005$, $\alpha_0 = 1{,}5$. They have been calculated due to expression (7) which provides for a more reliable result than intuitive sorting out applied earlier in [13]. Dimensional parameters η_α и τ_α are presented in table 1.

As Figure 2 shows, model (4), (5) describes quite precisely gradient dependences of viscosity at simple shear and uniaxial elongation. Specifically, one can see well the non-monotonous behavior of the dependence of elongation viscosity $\lambda(\varepsilon) = (\sigma_{11} - \sigma_{22})/\varepsilon$, and the effect of anomaly for shear viscosity $\eta(v_{12}) = \sigma_{12}/v_{12}$, where ε — elongation velocity, a v_{12} — shear velocity. Besides, Koks-Mertz rule is carried out [2, 45], according to which (at given $v_{12} = \omega$) complex viscosity measured in SAOS experiment and the set shear viscosity can be united:

$$\eta(v_{12}) = \left|\eta^*(\omega)\right| = \sqrt{\left(G'(\omega)\right)^2 + \left(G''(\omega)\right)^2} \Big/ \omega .$$

Figure 3 demonstrates the results of calculations for setting shear viscosity in a sample of Bralen industrial polymer material. Also, for comparative purposes Figure 4 contains the experimental data and corresponding non-stationary dependences of the first-order difference of normal stresses.

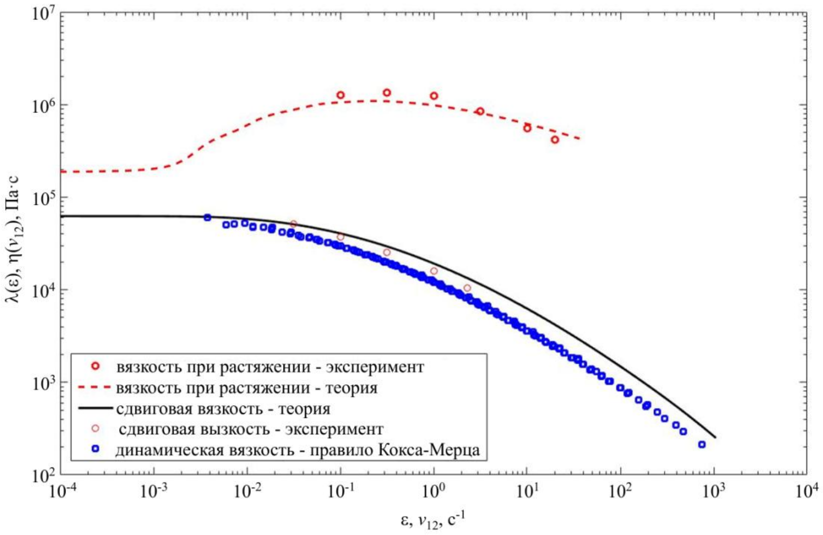

Fig. 2. Comparing the calculated gradient dependences of stationary viscosity at elongation and simple shear with experimental data.

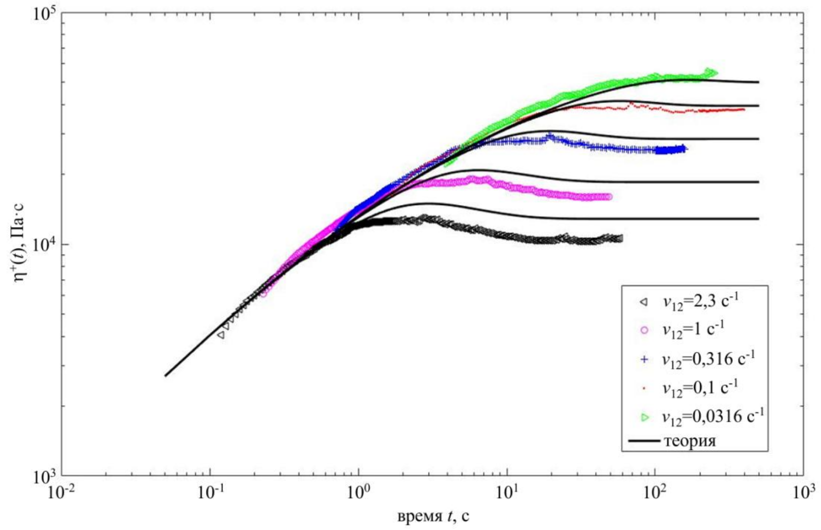

Fig. 3. Shear viscosity dependences on time at varied values of shear velocity: calculations (unbroken lines) and experiment (symbols).

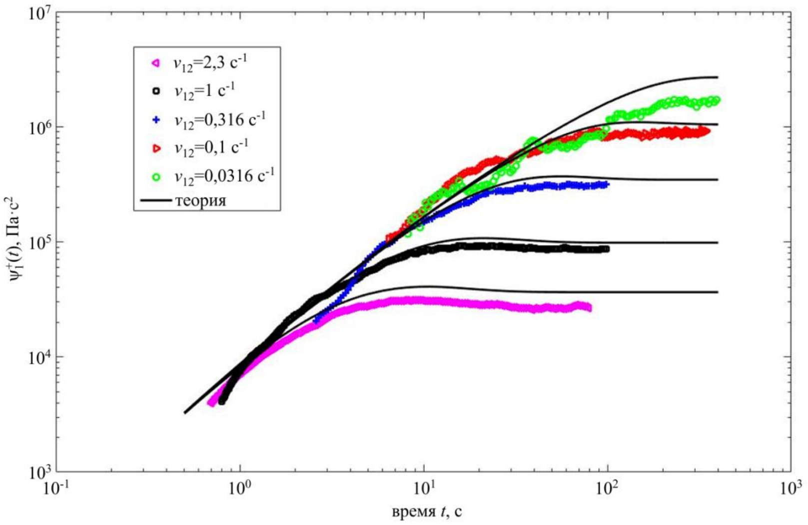

Fig. 4. Dependences of the first-order difference of normal stresses on time at varied values of shear velocity: calculations (unbroken lines) and experiment (symbols).

The results of calculations for time dependences of setting viscosity at uniaxial elongation are presented in Figure 5. Increasing monotonously, the curves level off. It can be noticed that the higher the elongation velocity, the faster viscosity reaches its ultimate value. As deformation starts, the calculated curves correspond well with the graphs built due to the experimental data. However, later the experimental curves experience a drastic drop which may be connected with sample destruction at extended elongation. Notably, this drawback remains in other rheological models [3, 5, 47].

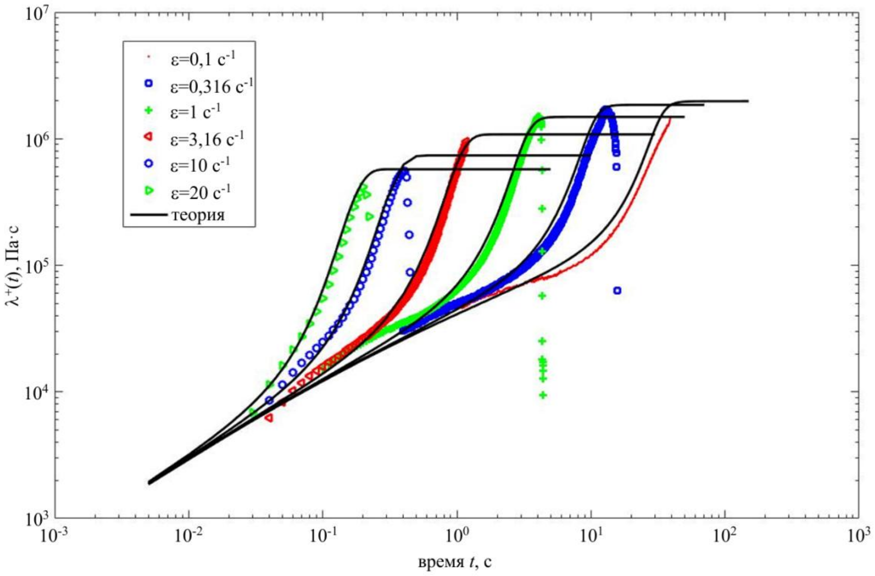

Fig. 5. Dependences of setting viscosity at uniaxial elongation and various values of elongation velocity: calculations (unbroken lines) and experiment (symbols).

In order to compare the modified Vinogradov-Pokrovsky model (4), (5) with other famous rheological models, the results of works [11, 16, 33, 48] were studied. There obtained experimental dependences are collated with the calculated ones due to the modified Leonov-Prokunin model [4], «Pom–Pom» model [5] and XPP model [8]. The data analysis stated that the model described by correlations (4), (5) possess a better prognostic ability than for Leonov-Prokunin and «Pom–Pom» models and comes close to that of XPP model. Besides, its predictive accuracy increases significantly compared to the single-mode approximation of mVP model [6, 14–15].

Also when comparing the calculated data shown in Figures 2-5 and those taken from work [13] which were calculated due to formulas (6) it shows that viscometric functions differ insignificantly. As it was stated above, correlations (5) are preferable since they describe well the behavior of elongation viscosity at high elongation velocities [49].

Since studying viscometric functions does not help to finally opt for a particular rheological model, it makes sense to refer to other types of shear flows. This may be the high-amplitude shear deformation [9–12, 17–24] or superposition of small oscillating motions over the simple shear flow [25–29].

6. High-amplitude shear deformation

According to the calculations given in paragraphs 3-5 and their comparisons with experimental data, it can be stated that model (4), (5) is feasible to be applied as an initial approximation at forecasting nonlinear and viscoelastic properties of fluid polymer media with linear or branched structures [13, 47 ,48]. The calculations due to model (4), (5) have been carried out via method Runge-Kutta and integrated MATLAB subprograms.

Now we will consider the deformation process, which follows the harmonic law: $\gamma\left(t\right)=\gamma_0\sin\left(\omega t\right)$, where γ_0 — a set amplitude, ω — oscillation frequency. At such deformation the tensor of velocity gradients has only one nonzero component $v_{12}\left(t\right)=\gamma_0\omega\cos\left(\omega t\right)$.

Basing on equations (4), (5), the modeling results demonstrate that should deformation start from the rest state $\left(\sigma_{ik}\left(0\right)=0\right)$, the oscillations in the polymer become periodical rather quickly. Theoretical dependences obtained within time $10\tau_0$, are presented in Figure 6. At small oscillations amplitude one can observe the direct proportion of shear stresses popping up in the system and the deformation scale. Thus, the material response presents itself as a proper harmonica. Should the oscillations be of large amplitude, the polymer media does not reveal this property and the time dependence of shear stress becomes more complex. These results are proved by experimental data (see e.g. [10]).

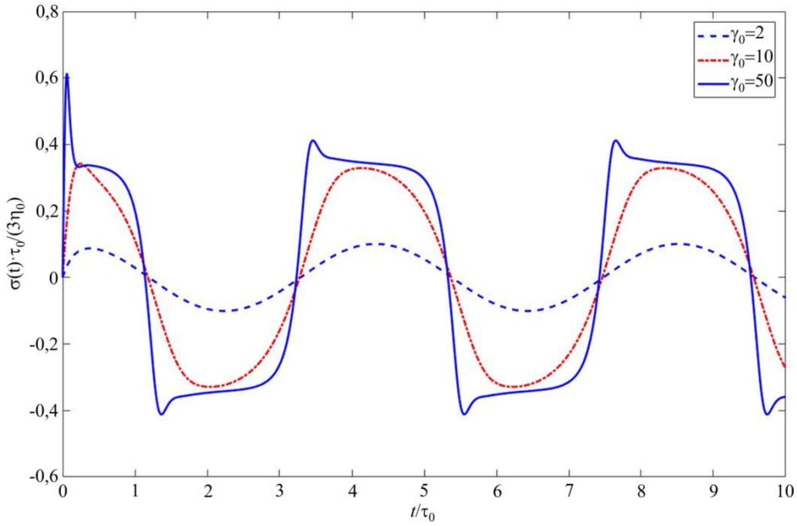

Fig. 6. Dependences on stress time at simple shear for various values of a deformation amplitude.

Numerous works are dedicated to the research into periodic deformations impact on the polymer system (see references [10, 11, 47, 49]). For example, work [11] shows the results of the experimental study for periodic deformations of large amplitude (between 50% and 4000%) frequency being $\omega = 0,2\,\text{Hz}$. Concentrated solution of polyethylene oxide (mass concentration of 5%) in dimethyl sulfoxide. The experiment details are given in [11].

In order to hold the calculations they applied single-mode approximation for model (4), (5). While modeling of periodic deformations with small amplitude, first they found the optimal parameter values: $\tau_0 = 0,21\,\text{с}$, $\eta_0 = 2,76\,\text{Pa·с}$, $\beta_0 = \left(0,037 + p_0\,\text{tr}\left(a\right)\right)\Big/\left(1+\sqrt{p_0\,\text{tr}\left(a\right)}\right)$, $p_0 = 0,003$.

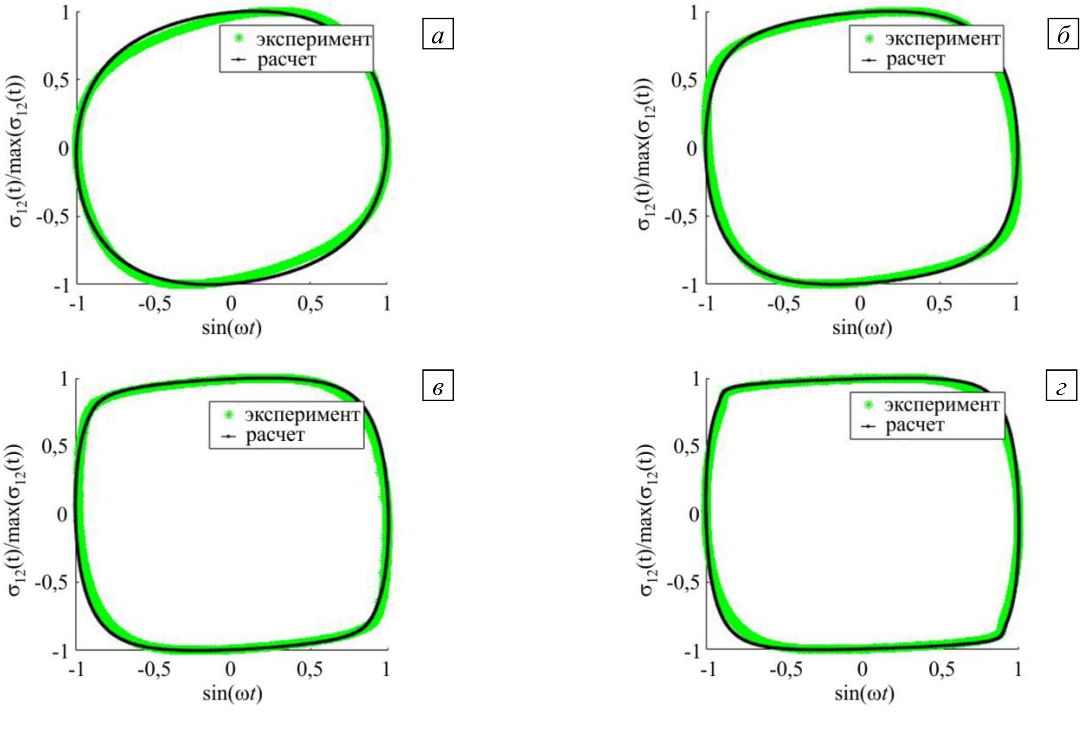

Fig. 7. Calculated and experimental Lissajous figures for shear stress dependence on deformation; relative deformation amplitudes are equal correspondingly: 5 (*а*), 10 (*б*), 20 (*в*), 40 (*г*).

Later, considering these values, they found theoretical dependences for large amplitude deformations which were compared with experimental curves [11].

To present the results graphically Lissajous figures were constructed, i.e. phase portraits of the stress dependence on deformation at various amplitudes and frequencies (Fig.7). For better visual clarity, normalization was done (current values were referred to maximal ones) both for shear stresses and shear velocity. It is observed that at small deformation amplitudes Lissajous figures get ellipsoid-shaped whereas at larger amplitudes they get “corners” and even contrary flexures which correspond well with the experimental data.

Based on the results obtained, the following conclusions can be drawn:

– the larger the harmonic oscillations amplitude, the stronger the nonlinear response is from the material under study to the applied impact [11];

– the modified Vinogradov-Pokrovsky model (4), (5) is capable of describing precisely the rheology of fluid polymer media under periodic deformation of large amplitude.

7. The superposition of oscillating and shear flows

In paragraphs 3-6 the behavior of polymer fluids was studied at oscillating shear deformations and at simple shear and uniaxial elongation. Also at shear deformation the processes with fluids of complex structure can be characterized by simultaneous stationary and oscillating deformation. Such processes are widely spread in various areas e.g. 3D printing, coating at polymer processing, blood coagulating which impacts clot forming and functioning [51]. It might seem that their solution requires solving a complete hydrodynamic task. However, it may be limited by the case when velocity gradients are a combination of a simple shear and oscillations, while the nonzero component of the tensor of velocity gradients looks as following:

$$\nu_{12}(t) = \gamma_1 + \gamma_0 \omega \cos(\omega t) .$$

This corresponds with the superposition of the simple shear flow and forcing oscillations in parallel directions. Initial characteristics are stationary shear rate γ_1 and relative amplitude of sinusoidal oscillations γ_0. Let τ_0 be the maximal relaxation time of the polymer system. At $\gamma_0 << \tau_0 \gamma_1$ the experimental implementation of such flows does not cause any difficulties [25–30].

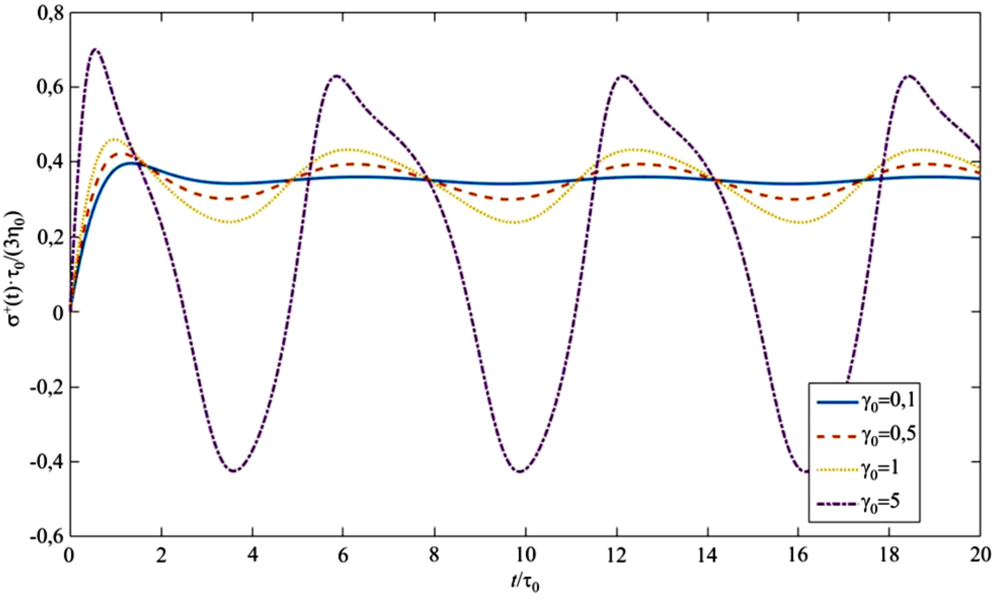

Fig. 8. The impact of the relative amplitude of forcing oscillations γ_0 on the dependence of shear stress on time at $\beta_0 = 0,2$; $p_0 = 0,005$ and fixed stationary shear velocity $\gamma_1 = 2$ c^{-1}.

Due to the obtained data one constructs frequency dependences of components of shear dynamic module at various values of γ_1. Nevertheless, the following case is quite curious: when velocity γ_0 corresponds with value $\tau_0\gamma_1$ or exceeds it.Let us consider the behavior of nonlinear response of the polymer system basing on the single-mode version of mVP model (4), (5). Since it was not possible to find the experimental data for this particular case, to hold calculations we will opt for the following model parameters [47]: $\tau_0 = 1$ c; $\beta_0 = \left(0,2 + p_0 \operatorname{tr}(\boldsymbol{a})\right) \Big/ \left(1 + \sqrt{p_0 \operatorname{tr}(\boldsymbol{a})}\right) = 0,2$; $p_0 = 0,005$; $\omega = 1$. Value η_0 is not required as we are going to consider the dependence of dimensionless value $\sigma_{12}^{+}(t)\tau_0 / (3\eta_0)$ on dimensionless time t/τ_0 at variations of parameters γ_1 γ_0.

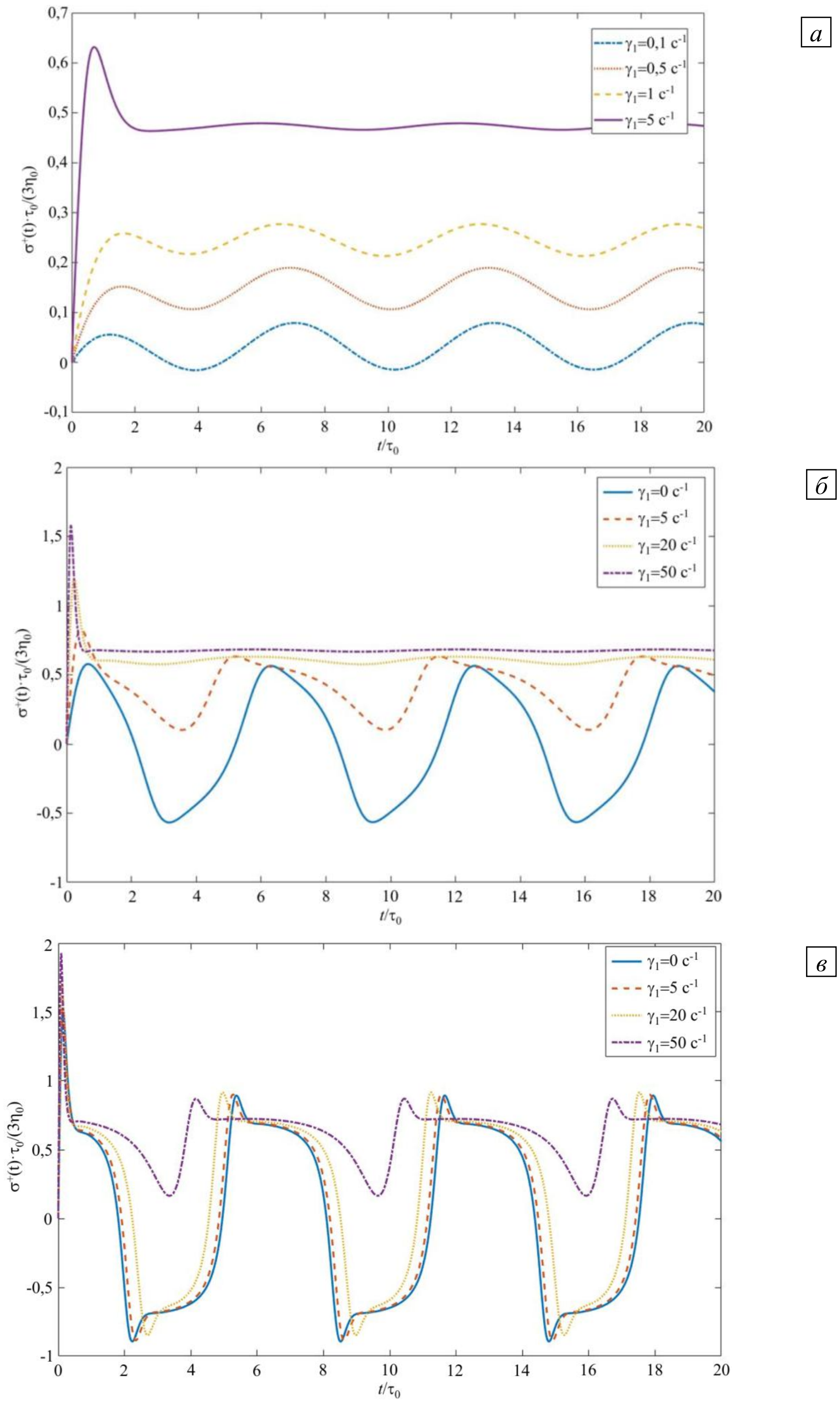

Fig. 9. The impact of the size of stationary shear velocity γ_1 on the stress-time dependence at $\beta_0 = 0,2$; $p_0 = 0,005$ and small ($\gamma_0 = 0,2$) (*a*), medium ($\gamma_0 = 5$) (*б*), large ($\gamma_0 = 50$) (*в*) amplitude of forcing oscillations.

When deformation appears due to external impact periodically changing in time, the polymer system goes from the state of rest into its stationary state characterized by the accumulated tangent stresses, and it executes forced oscillations close to this state. Should the stationary shear velocity be fixed ($\gamma_1 = \text{const}$), the amplitude of forced oscillations depends nonlinearly on the relative amplitude of forcing impact. Besides, at growing amplitude the shape of forced oscillations deviates from the regular harmonica in a non-symmetric way (Fig.8).

When the relative amplitude of forcing oscillations is fixed ($\gamma_0 = \text{const}$) and the stationary shear velocity is alternating, the curves of stresses-time dependences are stacked up. At low velocities of stationary shear the amplitude of forced oscillations does not change, whereas at high velocities of stationary shear the amplitude of forced oscillations reduces. At the start of forced oscillations we observe broken time in polymer motion which is well seen in graphs. Further on this oscillation process gets set and has a steady period and constant amplitude.

At low values of stationary shear velocity the stresses behavior gets sinusoidal. At growing γ_1 the sinusoid starts curving smoothly and flat-lining. At quite high values of γ_1 and low value of the amplitude of forcing oscillations γ_0 graph gets almost straight and parallel to X-axis (Fig. 9*a*).

We also have considered how the shear stresses are influenced by stationary shear velocity within the range between 0 and 50. Figures 9*б*, *в* demonstrate the graphs for medium and large values of the forcing oscillations amplitude. The given curves reveal the transition and later the steady process. However, there are significant differences between fragments (*б*) and (*в*).

Therefore, the smaller the forcing oscillations amplitude, the closer to X-axis the peaks of stresses curves are located and the transition process is less noticeable. When the stationary shear velocity γ_1 increases, graphs start curving, but, unlike Figures 8 and 9*a*, they are less sinusoid-like. At the same time, the larger the forcing oscillations amplitude γ_0 and the higher the stationary shear velocity γ_1, the more significantly the curves get deformed.

8. Conclusion

The work reviews the current information in mesoscopic modeling of flows, solutions and melts of polymers with various structures. The majority of currently popular rheological defining relations are of similar structure accurate to some dissipative function. The researchers have suggested a new version of the modified Vinogradov-Pokrovsky rheological model (4), (5). Based on it, linear and nonlinear effects have been studied for the industrial polyethylene sample at the simultaneous impact of simple shear and uniaxial elongation. The calculations for the authors' model were carried out via Runge-Kutta model with the help of integrated subprograms of the computational environment MATLAB. The new model was proved to adequately describe frequency dependences of the components of the shear dynamic module and transition processes at setting stresses in the shear fluid and uniaxial elongation. The written sources provided for the experimental data on the polyethylene melt taken for comparative reasons. Basing on the suggested model (4), (5), we studied the behavior of nonlinear viscoelastic response of the polymer material at large periodic deformations (LAOS).

The results of calculations for solution curves within coordinates "normalized shear stresses – normalized deformations" were compared with the experimental data obtained for the polyethylene oxide solution in dimethyl sulfoxide. The polymer sample behaves nonlinearly as it reveals the distorted viscoelastic material response to sinusoidal oscillations. Besides, the shear stresses curves do not look like regular harmonicas any more since the leading edge of the response demonstrates the so-called "step", and the proportionality factor between the stresses amplitude and shear amplitude is not constant: it is a decay function of the shear amplitude.

Considering the superposition of the oscillating shear flow over the simple shear, we can observe the distortion of viscoelastic response. However, unlike LAOS deforming, the distortion for upper and lower half-waves happens differently. The lower half-wave appear to be smoother and the distinctive "step" resides only in the upper half-wave. As no experimental data was found for this case, one should hope the results of this work will stimulate the researchers who study nonlinear viscoelastic properties of polymer solutions and melts.

The new version of mVP model (4), (5) goes in line with the experimental data. It also describes the stationary and non-stationary dependences of viscometric functions at the simultaneous impact of the simple shear and uniaxial elongation on the polymer with appropriate practical accuracy. This model provides for a more adequate presentation at higher elongation velocities and demonstrates high precision at LAOS deforming, which may become the basis for further research of rheologically more complex flows, e.g. at the superposition of the oscillating shear flow over the simple shear or flows in the areas with complex geometry.

References

1. Pokrovskii V.N. *The mesoscopic theory of polymer dynamics*. Springer, 2010. 256 p. https://doi.org/10.1007/978-90-481-2231-8
2. Malkin A.Ya., Isaev A.I. *Reologiya: kontseptsiya, metody, prilozheniya* [Rheology: concepts, methods, applications]. St. Petersburg, Profession, 2007. 560 p.
3. Giesekus H. A simple constitutive equation for polymer fluids based on the concept of deformation-dependent tensorial mobility. *J. Non-Newtonian Fluid Mech.*, 1982, vol. 11, pp. 69-109. https://doi.org/10.1016/0377-0257(82)85016-7
4. Leonov A.I. Analysis of simple constitutive equations for viscoelastic liquids. *J. Non-Newtonian Fluid Mech.*, 1992, vol. 42, pp. 323-350. https://doi.org/10.1016/0377-0257(92)87017-6
5. Inkson N.J., McLeish T.C.B., Harlen O.G., Groves D.J. Predicting low density polyethylene melt rheology in elongational and shear flows with "pom-pom" constitutive equations. *J. Rheol.*, 1999, vol. 43, pp. 873-896. https://doi.org/10.1122/1.551036
6. Altukhov Yu.A., Golovicheva I.E., Psyhnograi G.V. Molecular approach in linear polymer dynamics: Theory and numerical experiment. *Fluid Dyn.*, 2000, vol. 35, pp. 1-9. https://doi.org/10.1007/BF02698779
7. Thien N.P., Tanner R.I. A new constitutive equation derived from network theory. *J. Non-Newtonian Fluid Mech.*, 1977, vol. 2, pp. 353-365. https://doi.org/10.1016/0377-0257(77)80021-9
8. Verbeeten W.M.H., Peters G.W.M., Baaijens F.P.T. Differential constitutive equations for polymer melts: The extended Pom-Pom model. *J. Rheol.*, 2001, vol. 45, pp. 823-843. https://doi.org/10.1122/1.1380426
9. Bishko G., McLeish T.C.B., Harlen O.G., Larson R.G. Theoretical molecular rheology of branched polymers in simple and complex flows: The Pom-Pom model. *Phys. Rev. Lett.*, 1997, vol. 79, pp. 2352-2355. https://doi.org/10.1103/PhysRevLett.79.2352
10. Ilyin S.O., Malkin A.Ya., Kulichikhin V.G. Application of large amplitude oscillatory shear for the analysis of polymer material properties in the nonlinear mechanical behavior. *Polym. Sci. Ser. A.*, 2014, vol. 56, pp. 98-110. https://doi.org/10.1134/S0965545X14010039
11. Zelenkova J., Pivokonsky R., Filip P. Two ways to examine differential constitutive equations: Initiated on steady or initiated on unsteady (LAOS) shear characteristics. *Polymers*, 2017, vol. 9, 205. https://doi.org/10.3390/polym9060205
12. Koshelev K.B., Pyshnograi G.V., Tolstykh M.Yu. odeling of the three-dimensional flow of polymer melt in a convergent channel of rectangular cross-section. *Fluid Dyn.*, 2015, vol. 50, pp. 315-321. https://doi.org/10.1134/S0015462815030011
13. Merzlikina D.A., Pyshnograi G.V., Pivokonskii R., Filip P. Rheological model for describing viscometric flows of melts of branched polymers. *J. Eng. Phys. Thermophy.*, 2016, vol. 89, pp. 652-659. https://doi.org/10.1007/s10891-016-1423-7

14. Gusev A.S., Makarova M.A., Pyshnograi G.V. Mesoscopic equation of state of polymer systems and description of the dynamic characteristics based on it. *J. Eng. Phys. Thermophy*., 2005, vol. 78, pp. 892-898. https://doi.org/10.1007/s10891-006-0009-1
15. Pyshnograi G.V., Gusev A.S., Pokrovskii V.N. Constitutive equations for weakly entangled linear polymers. *J. Non-Newtonian Fluid Mech.*, 2009, vol. 164, pp. 17-28. https://doi.org/10.1016/j.jnnfm.2009.07.003
16. Pivokonsky R., Filip P., Pyshnograi G. On the reduction of nonlinear parameters in the eXtended Pom-Pom differential constitutive model. *Acta Technica*, 2018, vol. 63, pp. 1-15.
17. MacDonald I.F., Marsh B.D., Ashare E. Rheological behavior for large amplitude oscillatory shear motion. *Chem. Eng. Sci.*, 1969, vol. 24, pp. 1615-1625. https://doi.org/10.1016/0009-2509(69)80101-6
18. Onogi S., Masuda T., Matsumoto T. Nonlinear behavior of viscoelastic materials. I. Disperse systems of polystyrene solution and carbon black. *Trans. Soc. Rheol.*, 1970, vol. 14, pp. 275-294. https://doi.org/10.1122/1.549190
19. Dodge J.S., Krieger I.M. Oscillatory shear of nonlinear fluids. I. Preliminary investigation. *Trans. Soc. Rheol.*, 1971, vol. 15, pp. 589-601. https://doi.org/10.1122/1.549236
20. Matsumoto T., Segawa Y., Warashina Y., Onogi S. Nonlinear behavior of viscoelastic materials. II. The method of analysis and temperature dependence of nonlinear viscoelastic functions. *Trans. Soc. Rheol.*, 1973, vol. 17, pp. 47-62. https://doi.org/10.1122/1.549319
21. Komatsu H., Mitsui T., Onogi S. Nonlinear viscoelastic properties of semisolid emulsions. *Trans. Soc. Rheol.*, 1973, vol. 17, pp. 351-364. https://doi.org/10.1122/1.549285
22. Tee T.T., Dealy J.M. Nonlinear viscoelasticity of polymer melts. *Trans. Soc. Rheol.*, 1975, vol. 19, pp. 595-615. https://doi.org/10.1122/1.549387
23. Cho K.S., Hyun K., Ahn K.H., Lee S.J. A geometrical interpretation of large amplitude oscillatory shear response. *J. Rheol.*, 2005, vol. 49, pp. 747-758. https://doi.org/10.1122/1.1895801
24. Klein C., Venema P., Sagis L., van der Linden E. Rheological discrimination and characterization of carrageenans and starches by Fourier transform-rheology in the non-linear viscous regime. *J. Non-Newtonian Fluid Mech.*, 2008, vol. 151, pp. 145-150. https://doi.org/10.1016/j.jnnfm.2008.01.001
25. Simmons J.M. Dinamic modulus polyisobutylene solutions in superposed steady shear flow. *Rheol. Acta*, 1968, vol. 7, pp. 184-188. https://doi.org/10.1007/BF01982380
26. Faitel'son L.A., Yakobson É.É. Components of the complex modulus during the periodic shear flow of a viscoelastic fluid. *Mech. Compos. Mater.*, 1981, vol. 17, pp. 193-202. https://doi.org/10.1007/BF01039136
27. Siddigui A.M., Hayat T., Asghar S. Periodic flows of a non-Newtonian fluid between two parallel plates. *Int. J. Non Lin. Mech.*, 1999, vol. 34, pp. 895-899. https://doi.org/10.1016/S0020-7462(98)00063-8
28. Vermant J., Walker L., Moldenaers P., Mewis J. Orthogonal versus parallel superposition measurements. *J. Non-Newtonian Fluid Mech.*, 1998, vol. 79, pp. 173-189. https://doi.org/10.1016/S0377-0257(98)00105-0
29. Wong C.M., Isaev A.I. Orthogonal superposition of small and large amplitude oscillations upon steady shear flow of polymer fluids. *Rheol. Acta*, 1989, vol. 28, pp. 176-189. https://doi.org/10.1007/BF01356978
30. Oldroyd J.G. On the formulation of rheological equations of state. *Proc. R. Soc. Lond. A*, 1950, vol. 200, pp. 523-541. https://doi.org/10.1098/rspa.1950.0035
31. Leonov A.I., Prokunin A.N. *Nonlinear phenomena in flows of viscoelastic polymer fluids*. Chapman and Hall, 1994. 475 p. https://doi.org/10.1007/978-94-011-1258-1
32. Maxwell J.C. IV. On the dynamical theory of gases. *Phil. Trans. R. Soc.*, 1867, vol. 157, pp. 49-88. https://doi.org/10.1098/rstl.1867.0004

33. Pivokonsky R., Zatloukal M., Filip P. On the predictive/fitting capabilities of the advanced differential constitutive equations for branched LDPE melts. *J. Non-Newtonian Fluid Mech.*, 2006, vol. 135, pp. 58-67. https://doi.org/10.1016/j.jnnfm.2006.01.001
34. Pokrovskii V.N. Dynamics of weakly-coupled linear macromolecules. *Sov. Phys. Usp.*, 1992, vol. 35, pp. 384-399. https://doi.org/10.1070%2FPU1992v035n05ABEH002236
35. Soulages J., Schweizer T., Venerusa D.C., Kröger M., Öttinger H.C. Lubricated cross-slot flow of a low density polyethylene melt. *J. Non-Newtonian Fluid Mech.*, 2008, vol. 154, pp. 52-64. https://doi.org/10.1016/j.jnnfm.2008.02.007
36. Thomson W.W. On the elasticity and viscosity of metals. *Proc. R. Soc. Lond.*, 1865, vol. 14, pp. 289-297.
37. Pyshnograi G., Merzlikina D., Filip P., Pivokonsky R. Mesoscopic single and multi-mode rheological models for polymeric melts viscometric flows description. *WSEAS Transactions on Heat and Mass Transfer*, 2018, vol. 13, pp. 49-65.
38. Abbasi M., Ebrahimi N.G., Nadali M., Esfahani M.K. Elongational viscosity of LDPE with various structures: employing a new evolution equation in MSF theory. *Rheol. Acta*, 2012, vol. 51, pp. 163-177. https://doi.org/10.1007/s00397-011-0572-z
39. Pokrovsky V.N., Pyshnograi G.V. Simple forms of the defining equation of concentrated solutions and polymer melts as a consequence of the molecular theory of viscoelasticity. *Fluid Dyn.*, 1991, vol. 26, pp. 58-64. https://doi.org/10.1007/BF01050113
40. Altukhov Yu.A., Gusev A.S., Pyshnogray G.V., Koshelev K.B. *Vvedeniye v mezoskopicheskuyu teoriyu tekuchikh polimernykh sistem* [Introduction to the mesoscopic theory of fluid polymer systems]. Barnaul, AltGPA, 2012. 121 p.
41. Pyshnograi G.V., Pokrovsky V.N., YanovskyYu.G., Karnet Yu.N., Obraztsov I.F. Opredelyayushcheye uravneniye nelineynykh vyazkouprugikh (polimernykh) sred v nulevom priblizhenii po parametram molekulyarnoy teorii i sledstviya dlya sdviga i rastyazheniya [Constitutive equation of non-linear viscoelastic (polymer) media in nought approximation by parameter of molecular theory and conclusions for shear and extension]. *Dokl. Akad. Nauk*, 1994, vol. 339, no. 5, pp. 612-615.
42. Leonov A.I. *A brief introduction to the rheology of polymeric fluids*. Coxmoor Publishing Company, 2008. 257 p.
43. Golovicheva I.E., Zinovich S.A., Pyshnograi G.V. Effect of the molecular mass on the shear and longitudinal viscosity of linear polymers. *J. Appl. Mech. Tech. Phys.*, 2000, vol. 41, pp. 347-352. https://doi.org/10.1007/BF02465279
44. Rouse P.E. A theory of the linear viscoelastic properties of dilute solutions of coiling polymers. *J. Chem. Phys.*, 1953, vol. 21, pp. 1272-1280. https://doi.org/10.1063/1.1699180
45. Graessley W.W. *The entanglement concept in polymer phenology*. Springer-Verlag Berlin Heidelberg, 1974. 182 p. https://doi.org/10.1007/BFb0031036
46. Makarova M.A., Malygina A.S., Pyshnograi G.V., Rudakov G.O. Modeling of rheological properties of polyethylene melts at their uniaxial tension. *Vychisl. mekh. splosh. sred – Computational Continuum Mechanics*, 2020, vol. 13, no. 1, pp. 73-82. https://doi.org/10.7242/1999-6691/2020.13.1.6
47. Pyshnograi G.V., Cherpakova N.A., Al Joda H.N.A. Special features of nonlinear behavior of a polymer solution on large periodic deformations. *J. Eng. Phys. Thermophy.*, 2020, vol. 93, pp. 617-625. https://doi.org/10.1007/s10891-020-02159-8
48. Kuznetsova Yu.L., Skul’skiy O.I. Effect of different flows on the shear branding of a liquid with a non-monotonic flow curve. *J. Appl. Mech. Tech. Phy.*, 2019, vol. 60, pp. 22-30. https://doi.org/10.1134/S0021894419010048
49. Kuznetsova Yu.L., Skulskiy O.I., Pyshnograi G.V. Presure driven flow of a nonlinear viscoelastic fluid in a plane channel. *Vychisl. mekh. splosh. sred – Computational Continuum Mechanics*, 2010, vol. 3, no. 2, pp. 55-69. https://doi.org/10.7242/1999-6691/2010.3.2.17
50. Curtis D.J., Badiei N., Holder A., Claypole J., Deganello D., Brown M.R., Lawrence M.J., Evans P.A., Williams P.R., Hawkins K. Assessment of the stress relaxation characteristics of critical

gels formed under unidirectional shear flow by controlled stress parallel superposition rheometry. *J. Non-Newtonian Fluid Mech.*, 2015, vol. 222, pp. 227-233. https://doi.org/10.1016/j.jnnfm.2014.12.004

51. Altukhov Yu.A., Pokrovskii V.N., Pyshnograi G.V. On the difference between weakly and strongly entangled linear polymer. *J. Non-Newtonian Fluid Mech.*, 2004, vol. 121, pp. 73-86. https://doi.org/10.1016/j.jnnfm.2004.05.001
52. Rolón-Garrido V.H., Pivokonsky R., Filip P., Zatloukal M., Wagner M.H. Modelling elongational and shear rheology of two LDPE melts. *Rheol. Acta*, 2009, vol. 48, pp. 691-697. https://doi.org/10.1007/s00397-009-0366-8

The authors declare no conflict of interests.

The paper was received on 11.01.2021.
The paper was accepted for publication on 08.02.2021.