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A KINETIC APPROACH TO THE RHEOLOGY OF INHOMOGENEOUS SYSTEMS WITH ACCOUNT FOR THE INTERACTION OF DISPERSE-PHASE PARTICLES

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By measuring the optical characteristics of a flow the rheology of a viscous non-Newtonian fluid — an aqueous solution of hydrolyzed polyacrylonitrile — has been studied. The loss of stability by the system with chaotic fluctuations of transparency has been established. A kinetic model, as well as kinetic equations that describe the flow of heterogeneous systems, have been suggested. It is expected that this approach will be used in calculations in oil- and gas production and filtration of non-Newtonian systems in porous media.

For disperse systems with rheological properties dependent on time, e.g., thixotropic ones, nonlinear effects, in particular, fluctuations of rheological characteristics, are connected with the interaction of structural elements [1]. The interaction of the particles of a dispersed phase in purely viscous non-Newtonian systems does not lead to a noticeable temporal change of the rheological properties because of the small size of the particles (~10 μm) and the absence of a developed three-dimensional structure but has its effect on their steady-state rheology. An explanation of the rheology of purely viscous non-Newtonian systems is possible by determining the optical characteristics of flow [2, 3].

Investigations were carried out on an experimental rig similar to that used in [2, 3]. Through a cylindrical layer of a fluid flowing with a constant rate of shear, light was transmitted, the intensity of which was measured by a photoelectronic multiplier. If voltage on the latter was proportional to the system transparency, a recorder registered the graph of its dependence on time. The transparency increases on formation of aggregates of particles of the dispersed phase and decreases on their breakdown [2]. From a change in the transparency one can assess the dynamics of the interaction of particles.

We investigated a 2% solution of partially hydrolyzed polyacrylonitrile (PAN) with a degree of hydrolyzation of 65%. The rheological curve for the solution was plotted from the results of rotational viscosimetry on a "Rheotest-2.1" at a temperature of 20°C. From Fig. 1 one can see the starting length of a pseudoplastic flow, which, after an increase in the threshold rate of shear (γ = 600 1/sec), is replaced by a dilatant one. While a pseudoplastic flow is encountered rather frequently, a dilatant flow is a rather rare and inadequately studied phenomenon, as noted by the majority of researchers engaged in studying this flow [4–7]. According to [5, 8], dilatancy is observed in systems with a high concentration of a solid phase and in rough dispersions. For the systems indicated, this is explained on the basis of the theory of an "excluded volume" [7] on the assumption that in the process of flow the concentration of a solvent constantly decreases due to its higher mobility, and this leads to an increase in dry friction between the solid-phase particles and, as a consequence, to an increase in the apparent viscosity of the system.

In [4] it is shown that strong dilatancy may be observed in disperse systems with rather small particles (smaller than 5 μm). This can be explained by their migration to the walls of the capillary, which is expressed as an increase in the apparent viscosity of the system with the rate of shear [9, 10]. According to [11], a dilatant flow can be observed in polymer solutions (in particular, of polymethacrylic acid), which is explained by the unfolding of the chains of macromolecules with increase in the rate of shear and in the strength of interaction between them.

It should be noted that for the majority of dilatant systems one fails to observe a dilatant flow at all rates of shear [4]. Transition from a low to a high rate of shear is accompanied by transition from a pseudoplastic flow to a dilatant one (for example, for polyvinyl chloride [5]). As is seen from Fig. 1, a similar rheology is typical, in particular, of hydrolyzed PAN.

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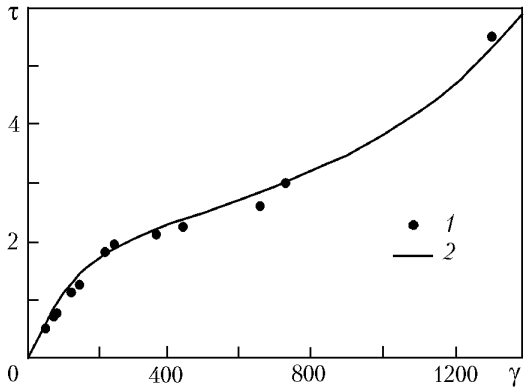


Fig. 1. Rheological curve [1) experimental; 2) theoretical] of an aqueous solution of partially hydrolyzed PAN having a 2% concentration, τ, Pa; γ, sec^{-1}.

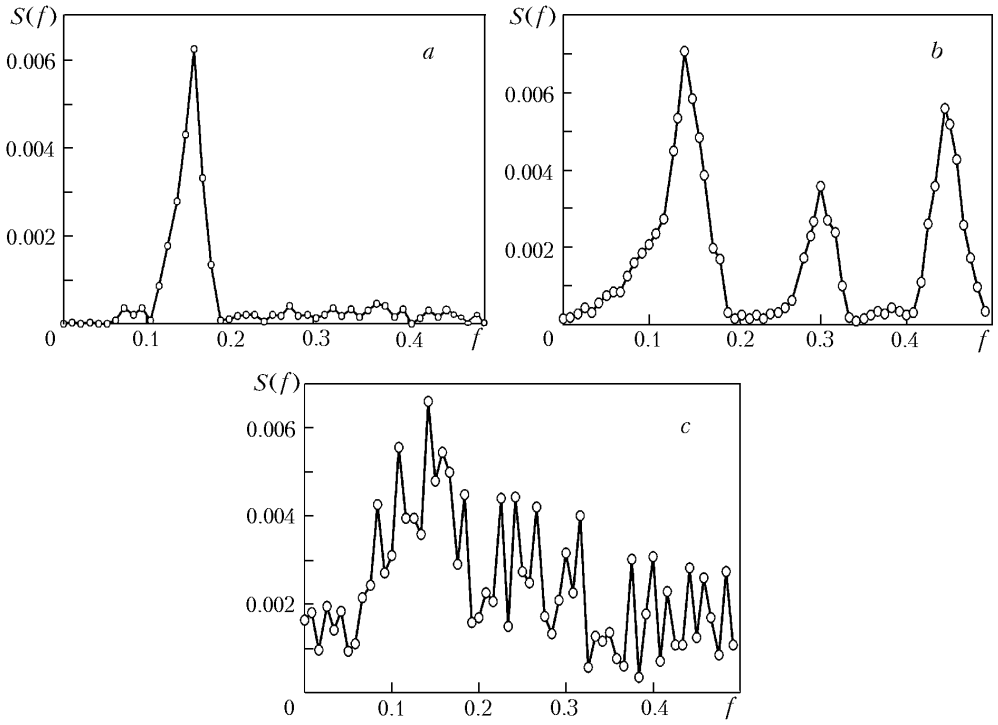
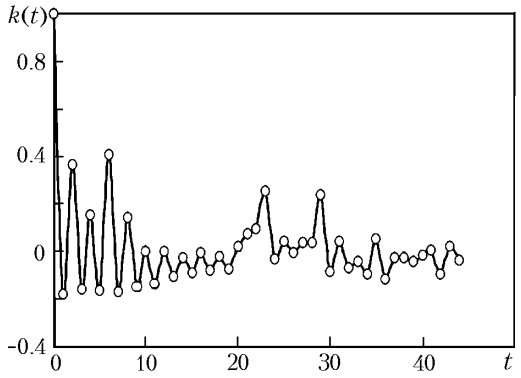


Fig. 2. Power spectrum $S(f)$ at γ = 600 (a), 800 (b), and 1200 sec^{-1} (c). f, sec^{-1}.

An experiment on determination of the dynamics of transparency of the system in the process of flow was carried out at a constant rate of shear. Investigations were carried out for four regimes: in the region of a pseudoplastic flow (γ = 400 1/sec), at the inflection point (γ = 600 1/sec), near the inflection point in the region of the dilatant flow (γ = 800 1/sec), and in the region of a developed dilatant flow (γ = 1200 1/sec). The results were presented as the dependences of stresses on time and thereafter for these dependences spectral Fourier analysis was made by a standard algorithm [12].

In the first case, the transparency does not change in time, i.e., dispersion flow is stable; in the second case, the flow stability is disturbed, and there are almost periodic fluctuations of transparency (Fig. 2a); in the third case, a quasi-periodic change in transparency with three characteristic frequencies are observed (2b), and in the fourth case, the transparency undergoes a chaotic change (the power spectrum $S(f)$) (Fig. 2c), indicative of which is rapid damping of the autocorrelation function $k(t)$ (Fig. 3) [13].



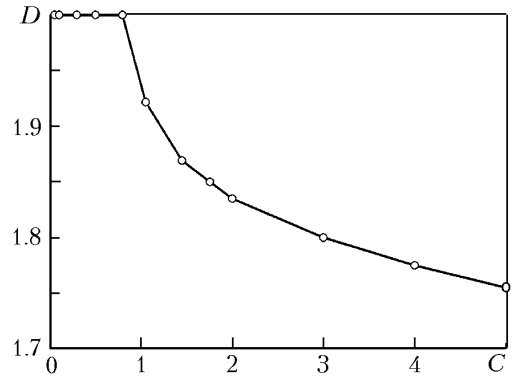


Fig. 3. Autocorrelation function $k(t)$ at $\gamma = 1200$ sec^{-1}. t, sec.

Fig. 4. Fractal dimensionality D vs. the concentration C of the polymer.

Based on experimental investigations, we may conclude that the disturbance of the dispersion flow stability leads to an increase in the hydraulic resistance and correspondingly to an increase in the apparent viscosity of the system investigated. Here, for each new level of interaction there is a new value of the apparent viscosity of the system.

It should be noted that the disturbance of the dispersed-phase-flow stability is observed at a concentration of partially hydrolyzed PAN in an aqueous solution of not less than 1%. Therefore, the reason for the disturbance of flow stability seems to be due to the strengthening of the interaction between particles, which is usually observed on increase in the dispersed-phase concentration and manifesting itself in the formation of associates of the dispersed-phase particles [14–16]. To check the assumption made, investigations of the dispersion structure were carried out with the aid of a Carl Zeiss optical microscope at different concentrations of the polymer. It has been established that at low concentration (0.05%) the dispersed phase consists of separate deformed, randomly oriented elongated (in the form of sticks) inclusions of size about 1 μm. As the concentration increases up to 2.0% or higher, larger associates are formed due to aggregations that predominantly have a round or oval shape of size 5–10 μm. Moreover, as is seen from Fig. 2c, with increase in the rate of shear, the average absolute value of transparency (of stress) is decreased, indicating breakdown of the associates and increase in the number of elementary inclusions.

In order to determine the uniformity of the distribution of the dispersed-phase particles in the system from microphotographs, the dependences of the number of particles present in an arbitrary selected circle on its radius r, $n(r)$, were constructed for different concentrations of the active mass in a solution. Based on the dependences constructed, the fractal dimensionality of the geometrical structure of the system was determined by analogy with [14]. Figure 4 shows the dependence of the fractal dimensionality D on the polymer concentration. As is seen from the figure, at concentrations of up to 1% the geometrical structure of the system is homogeneous and the fractal dimensionality corresponds to the Euclidean dimensionality of the surface. A further increase in the concentration leads to a decrease in the fractal dimensionality of the inhomogeneities, but at the 2% concentration it insignificantly (by 8%) differs from the Euclidean dimensionality of the surface. Thus, in the first approximation the spatial distribution of the dispersed phase may be considered homogeneous.

The foregoing allows the following kinetic model of the process observed to be suggested. With increase in the rate of shear, the number of elementary inclusions increase at the expense of the breakdown of associates. As the number of elementary inclusions increases, the interaction between them and the apparent viscosity of the system increase.

The processes proceeding in concentrated dispersed systems are mainly of a dynamic character. Their theoretical study with account for the effects of coagulation even for the simplest cases is very complicated mathematically [15, 16]. Therefore, the construction of simpler models ensuring an accuracy sufficient for practice is of definite interest.

In [1–21], based on the kinetic approach, thixotropic processes in complex systems were investigated, and it was shown that within the framework of that approach it is possible to predict the rheological properties of a dispersion. However, in those investigations the influence of interaction of the dispersed-phase particles on the evolution of

the rheological characteristics of the dispersion and the dynamics of motion of the disperse medium, which is of certain importance, were not taken into account. We will consider the motion of a dispersion in a tube of radius R.

Let N_1 and N_2 be the concentrations of elementary inclusions and associates, respectively. The names "elementary inclusions" and "associates" are conventional; the former is understood to be the particles that are not broken under any experimental conditions the latter denotes larger particles formed as a result of aggregation of elementary inclusions. Then, on the basis of the Lotka–Volterra [22] and Ferhülster models [23], neglecting, in the first approximation, the spatial inhomogeneity because of the relative smallness of the concentration of particles, the system of kinetic equations that describe the evolution of the dispersed phase may be presented in the form [1]

$$\frac{dN_1}{dt} = (a_2\,\gamma - \beta_1)\,N_2 - a_1\,\gamma N_1\,(1 - \alpha_1 N_1) - (\beta_1 + \beta_2)\,N_1 + \beta_1 N\,,\quad \frac{dN_2}{dt} = -\,a_2 N_2\,\gamma\,(1 - \alpha_2 N_2) + \beta_2 N_1\,, \tag{1}$$

$$N_1\,(0) + N_2\,(0) \le N = \text{const}\,.$$

For an axisymmetrical case, the equation of liquid motion in a cylindrical tube has the form

$$\rho\,\frac{\partial u}{\partial t} = \eta\left[\frac{\partial^2 u}{\partial r^2} + \frac{1}{r}\,\frac{\partial u}{\partial r}\right] + \frac{\Delta P}{l}\,. \tag{2}$$

In the first approximation, to estimate the influence of the dynamics of motion of the liquid itself on the system rheology we will assume that the rate of shear over the cross section is $\gamma = -\partial u/\partial r = \text{const}$. Then we have

$$u = \gamma\,(R - r)\,. \tag{3}$$

From Eq. (2), subject to (3), with the assumption adopted we obtain

$$\rho\,\frac{d\gamma}{dt}\,(R - r) = -\,\frac{\eta}{r}\,\gamma + \frac{\Delta P}{l}\,. \tag{4}$$

Having multiplied each term of Eq. (4) by $2\pi r dr$ and divided by πR^2, we average both of its sides over the cross section of the tube:

$$\frac{d\gamma}{dt} = -\,\frac{6\eta}{\rho R^2}\,\gamma + 3\,\frac{\Delta P}{\rho l R}\,. \tag{5}$$

The viscosity of the system η changes with time and can be determined from the formula [1]

$$\eta = \eta_0 + \eta_1\,\frac{N_2}{N}\,. \tag{6}$$

From Eq. (5), subject to (6), we find

$$\frac{d\gamma}{dt} = -\,\frac{6\gamma}{\rho R^2}\left(\eta_0 + \eta_1\,\frac{N_2}{N}\right) + 3\,\frac{\Delta P}{\rho l R}\,. \tag{7}$$

From the system of equations (1) and (6), we many determine the dynamics of the change in the rheological parameters of the disperse system. The stationary values of the parameters N_1, N_2, and γ can be found from Eqs. (1) and (7), which yield

$$q_2 = \overline{N}_2 = \frac{\beta_1 + \dfrac{\beta_1}{\beta_2}\,a_2\,\gamma_0 + \dfrac{a_1 a_2}{\beta_2}\,\gamma_0^2 + \sqrt{\left(\beta_1 + \dfrac{\beta_1}{\beta_2}\,a_2\,\gamma_0 + \dfrac{a_1 a_2}{\beta_2}\,\gamma_0^2\right)^2 - 4\beta_1 N a_0}}{2a_0}\,, \tag{8}$$

$$a_0 = \frac{a_1 a_2 \gamma_0^2}{\beta_2}\alpha_2 + \frac{a_1 a_2^2 \alpha_1 \gamma_0^3}{\beta_2^2}\alpha_2 + \frac{\beta_1 + \beta_2}{\beta_2} a_2 \gamma_0 \alpha_2 , \quad q_1 = \overline{N}_1 = \frac{a_2 \gamma_0 q_2 (1 - \alpha_2 q_2)}{\beta_2} , \tag{9}$$

$$\gamma_0 = \frac{\Delta P}{l} \frac{P}{2\left(\eta_0 + \eta_1 \dfrac{q_2}{N}\right)} . \tag{10}$$

If α_1 and α_2 are much less than a_1 and a_2, then from Eq. (8) we obtain

$$q_1 = \frac{\dfrac{a_2 \gamma_0}{\beta_2} N}{1 + \dfrac{a_2}{\beta_2}\gamma_0 + \dfrac{a_1 a_2}{\beta_1 \beta_2}\gamma_0^2} , \quad q_2 = \frac{N}{1 + \dfrac{a_2}{\beta_2}\gamma_0 + \dfrac{a_1 a_2}{\beta_1 \beta_2}\gamma_0^2} . \tag{11}$$

From Eq. (10), subject to (11), we have

$$\gamma_0^3 - \left(\frac{\Delta P R}{2\eta_0 l} - \frac{\beta_1}{a_1}\right)\gamma_0^2 - \left[\frac{\Delta P R}{2\eta_0 l}\frac{\beta_1}{a_1} - \left(1 + \frac{\eta_1}{\eta_0}\right)\frac{\beta_1 \beta_2}{a_1 a_2}\right]\gamma_0 - \frac{\Delta P R}{2\eta_0 l}\frac{\beta_1 \beta_2}{a_1 a_2} = 0 . \tag{12}$$

Equation (12) yields

$$\gamma_0 = \sqrt[3]{-\frac{n}{2} + \sqrt{\left(\frac{n}{2}\right)^2 + \left(\frac{m}{3}\right)^3}} + \sqrt[3]{-\frac{n}{2} - \sqrt{\left(\frac{n}{2}\right)^2 + \left(\frac{m}{3}\right)^3}} + \frac{1}{3}\left(\frac{\Delta P R}{2\eta_0 l} - \frac{\beta_1}{a_1}\right), \tag{13}$$

where

$$m = -\frac{1}{3}\left(\frac{\Delta P R}{2\eta_0 l} - \frac{\beta_1}{a_1}\right)^2 - \frac{\Delta P R}{2\eta_0 l}\frac{\beta_1}{a_1} + \left(1 + \frac{\eta_1}{\eta_0}\right)\frac{\beta_1 \beta_2}{a_1 a_2} ;$$

$$n = -\frac{2}{27}\left(\frac{\Delta P R}{2\eta_0 l} - \frac{\beta_1}{a_1}\right)^2 - \frac{1}{3}\left(\frac{\Delta P R}{2\eta_0 l} - \frac{\beta_1}{a_1}\right)\left[\frac{\Delta P R}{2\eta_0 l}\frac{\beta_1}{a_1} - \left(1 + \frac{\eta_1}{\eta_0}\right)\frac{\beta_1 \beta_2}{a_1 a_2}\right] - \frac{\Delta P R}{2\eta_0 l}\frac{\beta_1 \beta_2}{a_1 a_2} .$$

The values of q_1 and q_2 are determined from Eq. (11), subject to Eq. (13).

In the first approximation, we assume that there are slight fluctuations of ν_1, ν_2, and ε around the stationary position of q_1, q_2, and γ_0 [22, 23]:

$$N_1 = q_1 (1 + \nu_1) , \quad N_2 = q_2 (1 + \nu_2) , \quad \gamma = \gamma_0 + \varepsilon . \tag{14}$$

Then, from (1) and (7), with (14) taken into account, we obtain an equation of a perturbed motion of the system from the first approximation:

$$\frac{d\nu_1}{dt} = -b_1 \nu_1 + \frac{q_2}{q_1}(a_2 \gamma_0 - \beta_1)\nu_2 + \left(\alpha_1 q_1 + \frac{a_2 q_2}{a_1 q_1} - 1\right) a_1 \varepsilon ,$$

$$\frac{d\nu_2}{dt} = \frac{q_1}{q_2}\nu_1 + (2 a_2 q_2 - 1)\, a_2 \gamma_0 \nu_2 + (\alpha_2 q_2 - 1)\, a_3 \varepsilon ,$$

$$\frac{d\varepsilon}{dt} = -\frac{6}{\rho R^2}\gamma_0\eta_1\frac{q_2}{N}\nu_2 - \frac{6}{\rho R^2}\left(\eta_0 + \frac{\eta_1 q_2}{N}\right)\varepsilon\,. \tag{15}$$

The threshold values of the parameters at which the loss of stability of the nonperturbed motion (Eqs. (1) and (2)) of the system occurs can be determined from Eq. (15). For the equation of perturbed motion (15) the characteristic equation has the form

$$\begin{vmatrix} -b_2 - \lambda & \frac{q_2}{q_1}(a_1\gamma_0 - \beta_1) & \left(a_1 q_1 + \frac{a_1 q_2}{a_1 q_1} - 1\right)a_1 \\ \frac{q_1}{q_2}\beta_2 & (2\alpha_2 q_2 - 1)\,\alpha_2\gamma_0 - \lambda & (\alpha_2 q_2 - 1)\,a_3 \\ 0 & -\frac{6}{\rho R^2}\gamma_0\eta_1\frac{q_2}{N} & -\frac{6}{\rho R^2}\left(\eta_0 + \frac{\eta_1 q_2}{N}\right) - \lambda \end{vmatrix} = 0\,, \tag{16}$$

whence

$$\lambda^3 + c_1\lambda^2 + c_2\lambda + c_3 = 0\,, \tag{17}$$

where

$$c_1 = a_2\gamma_0(1 - 2a_2 q_2) + \frac{6}{\rho R^2}\left(\eta_0 + \frac{\eta_1 q_2}{N}\right) + b_1\,;\quad c_2 = (1 - 2a_2 q_2)\,a_2\gamma_0\frac{6}{\rho R^2}\left(\eta_0 + \frac{\eta_1 q_2}{N}\right) +$$

$$+ b_1(1 - 2a_2 q_2)\,a_2\gamma_0 + b_1\frac{6}{\rho R^2}\left(\eta_0 + \frac{\eta_1 q_2}{N}\right) + (1 - aq_2)\,a_3\frac{6}{\rho R^2}\gamma_0\eta_1\frac{q_2}{N} + (a_2\gamma_0 - \beta_1)\,\beta_2\frac{6}{\rho R^2}\,;$$

$$c_3 = b_1(1 - 2a_2 q_2)\,a_2\gamma_0\frac{6}{\rho R^2}\left(\eta_0 + \frac{\eta_1 q_2}{N}\right) + b_1(1 - aq_2)\,a_3\frac{6}{\rho R^2}\gamma_0\eta_1\frac{q_2}{N} +$$

$$+ (\beta_1 - a_2\gamma_0)\,\beta_2\frac{6}{\rho R^2}\left(\eta_0 + \frac{\eta_1 q_2}{N}\right) + \left(a_1 q_1 + \frac{a_2 q_2}{a_1 q_1} - 1\right)\frac{a_1 q_1}{a_2 q_2}\,;$$

$$b_1 = a_1\gamma_0(1 - 2\alpha_1 q_1) + \beta_1 + \beta_2\,.$$

The Hurwitz matrix for Eq. (17) has the form

$$A = \begin{pmatrix} c_1 & 1 & 0 \\ c_3 & c_2 & c_1 \\ 0 & 0 & c_3 \end{pmatrix}. \tag{18}$$

According to the Lyapunov theorem, for asymptotic stability of nonperturbed equations of motion (Eqs. (1) and (2)) of the system it is necessary and sufficient that Eq. (17) could have negative real parts. Then, according to the Hurwitz criteria, all major diagonal minors of matrix (18) must be positive, i.e.,

$$c_1 > 0\,,\quad c_1 c_2 c_3 > 0\,,\quad c_3(c_1 c_2 - c_3) > 0\,. \tag{19}$$

Moreover, according to the Lyapunov theorem, if at least one of the minors in (18) is negative, then the nonperturbed motion of the system is unstable at any nonlinear terms on the right-hand sides of Eqs. (1) and (2). Thus, the resulting conditions (19) make it possible to determine the threshold values of the parameters at which an unstable state of the system sets in. Since the shear stress τ in the system can be determined from the formula

$$\tau = \eta\gamma, \tag{20}$$

from (20), subject to (6), (11), and (14), for the limiting case $t \to \infty$ we have

$$\tau = \gamma_0 \left(\eta_0 + \frac{\eta_1}{1 + \dfrac{a_2}{\beta_2}\gamma_0 + \dfrac{a_1 a_2}{\beta_1 \beta_2}\gamma_0^2} \right). \tag{21}$$

Numerical calculation of the value of τ from Eq. (20) has been performed at the following values of the parameters (that were determined from the experimental curves of the flow with the aid of the least-squares method [24]): η_0 = 4 mPa·sec; η_1 = 20 mPa·sec; $a_1 = a_2 = 10^{-8}$; $\beta_1 = \beta_2 = 10^{-6}$ sec^{-1}. The results of calculation together with the results of experimental investigations are presented in Fig. 1. As is seen from the figure, the results of the theoretical investigations agree well with experimental data. At the above-indicated values of the parameters and a value of the rate of shear γ above 600 sec^{-1}, the stability of the system is perturbed. Next, as is shown in [12, 22, 25–27], in a system similar to that under study, at values of the parameters above the threshold one, chaos sets in because of the 2-, 3-, or 5-fold increase in the period of fluctuations, which agrees with the results of the investigations carried out.

The results obtained can be used for hydrodynamic calculations in oil and gas production, as well as to describe filtration of non-Newtonian systems in porous media.

NOTATION

A, Hurwitz matrix; a_1 and a_2, nonnegative numbers characterizing the intensity of destruction of elementary inclusions and associates; C, polymer concentration; c_1, c_2, and c_3, matrix elements; D, fractal dimensionality; f, frequency, sec^{-1}; $k(t)$, autocorrelation function; l, length of the tube, m; N, concentration; N_1 and N_2, concentration of elementary inclusions and associates, respectively; $n(r)$, integers; ΔP, pressure drop, MPa; q_1, stationary value of N_1; q_2, stationary value of N_2; R, radius of the tube, m; r, coordinate; $S(f)$, power spectrum; t, time, sec; u, velocity of the liquid at any point of the tube cross section in the axial direction, m/sec; α_1 and α_2, negative numbers characterizing the retardation of the intensity of destruction of particles on increase in their number; β_1 and β_2, nonnegative numbers that determine the rate of recovery of the concentration of particles of both species, sec^{-1}; γ, rate of shear, sec^{-1}; η, dynamic viscosity of the system, mPa·sec; η_0 and η_1, constants; λ, characteristic numbers; ν_1 and ν_2, small fluctuation; ρ, density, kg/m^3; τ, shear stress, Pa.

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