## SPECIFIC FEATURES OF MOTION OF RHEOPECTIC FLUIDS

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Results of an experimental and theoretical investigation of rheopectic fluid flows in tubes are presented. Loss of stability and appearance of periodic and random self-oscillations are shown to be possible during motion of media with shear-strengthened structures.

Introduction. Rheopectic (or antithixotropic) is the name given to media having the characteristic feature (in contrast to thixotropic fluids) of gradual structure formation during flow [1, 2]. In rheological properties these media are nearest to dilatant fluids, differing from the latter in that the dependence of tangential stress on shear rate in rheopectic media is a function of time. Experiments show that the property of rheopexy is detected only at small enough shear rates; if the rate of shear strain exceeds a certain critical value, no structure formation occurs. Rheopectic media are encountered in technological processes quite often, but theoretical problems associated with simulation of their motion have not been worked out adequately so far.

The present work is concerned with investigation of nonstationary processes occurring during motion of rheopectic fluids. In it results of laboratory experiments are presented for an investigation of tube flow of a suspension of quartz sand in a bentonite solution. This material may be regarded as a rheopectic medium by virtue of the fact that if the pressure drop at the ends of the tube is small enough, then after having started to flow the velocity of the suspension decreases gradually until it becomes equal to zero. But as soon as the pressure drop starts to grow, the flow resumes. As is known, this kind of phenomenon typifies rheopectics [1, 2]. Experiments showed that the approach to steady-state modes of flow has an oscillatory character. In some cases steady-state modes of flow lose stability, and sustained oscillations of the suspension flow rate at a constant pressure drop are observed. This effect is similar to the phenomenon of elastic turbulence found earlier in experiments on the motion of thixotropic media [1, 3-5].

In the present work a kinetic equation is suggested that describes the processes of the restoration and destruction of the structure of rheopectic media under the action of shear strain. Based on this, a simple mathematical model is obtained that makes it possible to give a qualitatively correct description of the effects observed in experiments. It is shown that at some values of the parameters loss of stability of the stationary modes of flow is possible, which is accompanied by the appearance of random oscillations of a deterministic nature (deterministic chaos). Earlier, this phenomenon, i.e., unpredictably complex motion in relatively simple systems without sources of random noise, was observed in a number of real and model physical, chemical, and biological systems [6-9]. In [10] the appearance of random oscillations during motion of thixotropic media is shown to be possibile, and thereby a connection between elastic turbulence and deterministic chaos is revealed. Note that this connection is supported once again by results of the present work.

1. Experimental Study of the Laws Governing Motion of Rheopectic Media in Tubes. Let us consider results of laboratory experiments on the investigation of tube flow of a suspension of quartz sand (with the diameter of a sand grain  $\le 2 \cdot 10^{-5}$  m) in a bentonite solution (data of G. M. Panakhov and N. M. Safarov). The tube had a length L = 3.0 m, and its internal diameter was d = 0.016 m.

In the course of the experiments the dependence of the mass flow rate of the fluid G on time was determined at a constant pressure drop  $\Delta p$ . Before each series of experiments the tube was carefully flushed out with water

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Fig. 2. Functions G = G(t) for a 10%-suspension.

and dried. By letting the suspension settle for a long time it was confirmed that it was stable, i.e., there was no deposition of sand (ove a time much greater than the time of the experiments). Figure 1 presents curves G = G(t) plotted for a flow of a suspension containing 20 wt.% sand (the density of the mixture is  $1.32 \cdot 10^3 \text{ kg/m}^3$ ) at pressure drops equal to  $\Delta p = 1.1 \cdot 10^4$ ,  $1.3 \cdot 10^4$ ,  $1.5 \cdot 10^4$ , and  $1.6 \cdot 10^4$  Pa (curves 1, 2, 3, and 4, respectively). As we see, the behavior of the suspension depends substantially on the magnitude of the pressure drop applied. If the pressure drop is small, the fluid flow rate decreases with time until it becomes equal to zero. To explain this fact, we may assume that during slow flow of the suspension rheopectic structure formation occurs, which increases the limiting shear stress of the fluid. The inclusion of the suspension in the class of rheopectic media is also justified by the fact that bentonite solutions also display rheopectic properties by themselves [2]. However, it should be noted that it would be desirable to confirm the rheological classification of the suspension at hand in independent metrological experiments. Unfortunately, such data are lacking at present, and therefore we limit the discussion to the arguments given above.

When the pressure drop attains a certain critical value, no "choking" of the tube occurs (see curve 4). This agrees with the fact that the rheopexy property is detected only at sufficiently low shear rates; as the rate of shear strain increases, processes of structure breakdown start to prevail. It is paradoxical that in a certain region an increase in the pressure drop leads to a more rapid "choking" of a tube. This indicates that at low flow velocities the rate of structure formation increases with the rate of shear strain.

Note also the fact that upon approach to steady states the flow rate values experience decaying oscillations whose amplitude exceeds the possible errors of measurements.

Experiments carried out at other concentrations of sand showed that an increase of its content up to 40% does not lead to a qualitative change in the picture observed. During flow of less concentrated suspensions the effect of "choking" can be absent, but one can see nondecaying oscillations of rather substantial amplitude compared to the mean value of G (see Fig. 2, in which measurements of the flow rate of a 10%-suspension are presented).

The regularities noted above can lead to complications in various technological processes. For example, transportation of non-Newtonian media that display rheopectic properties in tubes may lead to emergency shutdown of the pipeline. Academician A. Kh. Mirzadzhanzade assumed that the "choking" of a rheopectic fluid flow may cause the jamming of tools during drilling and the formation of hanging plugs during operation of sandy wells.

2. Simulation of the Flow of Rheopectic Media through Tubes. Let us write out equations that describe rheopectic fluid flow through a tube. First of all, let us estimate the contribution of inertial forces, which is determined by the ratio  $\delta = (\rho w_*/t_*)/\theta_*$ . Assuming, according to the conditions of the experiment, that  $t_* = 30$  sec,  $\rho = 10^3 \text{ kg/m}^3$ ,  $R = 10^{-2} \text{ m}$ ,  $G_* = 5 \cdot 10^{-2} \text{ kg/sec}$ , and  $\theta_* = 10^3 \text{ Pa/m}$ , we obtain  $w_* \approx 0.1 \text{ m/sec}$  and  $\delta \approx 10^{-3} \ll 1$ . Thus, we may neglect the effect of inertial forces. We also neglect the compressibility of the fluid and consider the tube to be infinitely long. The first of the latter two assumptions is quite justified because  $L/ct_* \ll 1$ , where L is the tube length, c is the velocity of sound. However, the assumption of an infinitely long tube is a substantial idealization, since the time of unsteady-state processes  $t_*$  is of the same order of magnitude as the characteristic time of passage of fluid particles through the tube  $L/w_*$ . However, it can also be adopted for a qualitative description of the phenomena observed.

Within the framework of the structural-kinetic approach [11-13] the rheology of rheopectic media can be determined by processes of restoration and disintegration of the structure, representing them schematically as direct and inverse chemical reactions whose total effect is described by a certain kinetic equation in the concentration of structural bonds *s*. Analysis of experimental data shows that rheopectic fluids are nonlinear viscoelastic media with a variable value of the limiting shear stress. Therefore, the rheological equation for them can be adopted in the form

$$\tau = \tau_0 + f(\dot{\varepsilon}) , \qquad (1)$$

where  $f(\dot{\varepsilon})$  is a certain function that determines the structural viscosity;  $\tau_0 = \tau_0(s)$  is the limiting shear stress, which depends on the concentration *s* of structural bonds formed during flow. For simplicity, we approximate this relation by the linear function

$$\tau_0 = \tau_1 \left( 1 + \gamma s \right) \,.$$

We assume that at low shear rates the structure formation of the fluid takes place at a rate proportional to  $\dot{\varepsilon}$ , whereas with an increase in  $\dot{\varepsilon}$  processes of destruction of bonds start to show up, which occur with a certain lag  $\lambda$ . Then the kinetic equation that determines the relative rate of change of the concentration of bonds can be written in the form

$$\frac{1}{s} \frac{ds(t)}{dt} = \alpha \dot{\varepsilon}(t) - \beta [s \dot{\varepsilon}^2]_{\lambda}, \qquad (2)$$

where  $\alpha$ ,  $\beta$  are positive coefficients that determine the rates of restoration and disintegration of bonds;  $|g|_{\lambda} = g(t-\lambda)$ . The use of an equation with a divergent argument makes it possible to overcome difficulties that arise in attempting to give an explicit description of multistage processes of structural transformations. In the absence of needed information on properties of individual elements of the structure and details of the interaction between them, this approach permits one, in a rather simple form in a certain integral manner, to take into account the effects of retardation and to give an adequate description of certain aspects of the motion of rheophysically complex media [14, 15].

Averaging, for simplicity, all of the quantities considered over the tube cross section, we set

 $\dot{\varepsilon} = \nu w$ ,

which, after nondimensionalization, leads to the model

$$\frac{ds}{dt} = s \left( \dot{\varepsilon} - B \left[ s \dot{\varepsilon}^2 \right]_{\lambda} \right), \tag{3}$$

 $w = \dot{\varepsilon} = \varphi(s)$ .

Here,  $\varphi(s) = (1/\dot{\epsilon}_*)\varphi_1(\tau_1(A-s))$ ,  $A = R\theta/2\tau_1 - 1$ ,  $B = \dot{\epsilon}_*\beta/\alpha\gamma$ ,  $s \to \gamma s$ ,  $\dot{\epsilon} \to \dot{\epsilon}/\dot{\epsilon}_*$ ,  $t \to t/t_*$ ,  $w \to w/w_*$ ,  $\lambda \to \lambda/t_*$ , where  $\dot{\epsilon}_* = \nu w_*$ ,  $t_* = 1/\alpha\dot{\epsilon}_*$  is the characteristic time of rheopectic structure formation,  $\nu \approx 4/R$ ,  $\dot{\epsilon} = \varphi_1(z)$  is the function inverse to  $z = f(\dot{\epsilon}) \ (\varphi_1(0) = 0)$ .

The stationary points of Eq. (3) can be found from the conditions

$$s = s_1 = 0,$$
  

$$w = \varphi(s) = 0$$
(4)

and

$$B\varphi(s) = 1/s.$$
<sup>(5)</sup>

Condition (4) determines the quiescent point  $s = s_0 = A$  corresponding to flow "choking." Solutions of Eq. (5) depend on the mutual disposition of the functions  $B\varphi(s)$  and 1/s.

We can easily see that at rather small values of A and B (i.e., at small pressure gradient and rate of disintegration of bonds) Eq. (5) does not have solutions. An increase in these parameters leads to the appearance of additional equilibrium points corresponding to stationary fluid flow. To investigate the stability of the equilibrium points  $s_i$  (i = 0, 1, ...), we set  $s = s_i + \tilde{s}$  in Eq. (3) and linearize in  $\tilde{s}$ . Thus, for the equilibrium point  $s = s_1 = 0$  we obtain

$$\frac{d\tilde{s}}{dt_1} = \varphi \ (0) \ \tilde{s}$$

Since  $\varphi(0) > 0$ , this point is unstable. At  $i \neq 1$  we have the equation

$$\frac{d\widetilde{s}}{dt} = -\widetilde{s} + C \ [\widetilde{s}]_{\lambda} ,$$

where

$$t_{1} = s_{i} |\varphi_{i}^{'}| t, \quad C = B\varphi_{i}s_{i} \left(2 - \frac{\varphi_{i}}{s_{i} |\varphi_{i}^{'}|}\right), \quad \varphi_{i} = \varphi(s_{i}).$$

(It is assumed that  $\varphi(s)$  is a monotonically decreasing function).

Since C = 0 when  $s = s_0$ , this point is always stable. The remaining equilibrium points are stable for |C| < 1, aperiodically unstable for C > 1, and oscillatingly unstable for C < -1 and rather large values of  $\lambda$  [7].

To specify the form of the function  $\varphi(s)$ , the relation  $w_0 = w_0(\tau)$  determined from results of the experiments described above was used  $(w_0 = w|_{t=0})$  is the value of the mean velocity of the fluid at the instant of the start of motion under the action of the pressure gradient  $\theta$ ). Since before plotting each curved of w = w(t) the system was brought into the initial state,  $s|_{t=0} = 0$ . Therefore from the form of the curve of  $w_0(\tau)$  we can judge the form of the function  $\varphi_1(\tau - \tau_1)$ .

Below some results obtained during investigation of the model are presented. Figure 3 gives curves of w = w(t) obtained by numerical integration of Eq. (3) under the condition that s, w = 0 (t < 0) and at the values of the



Fig. 3. Plots of flow velocity vs time.

Fig. 4. Strange attractor at A = 2.65 (w, mean velocity at time t;  $[w]_{\lambda}$ , mean velocity at time  $t - \lambda$ ).

parameters B = 0.5,  $\lambda = 2$ . As is seen, the model suggested describes qualitatively correctly the regularities found experimentally (compare with Fig. 1). Calculations showed that with an increase in the pressure gradient the steady-state modes of flow can lose stability and periodic self-oscillations can appear in the system. Model (3) also admits deterministic random oscillations, which are established through a cascade of bifurcations of successive doubling of the period following M. Feigenbaum's universal scenario [7-9]. Thus, in Fig. 4 in the projection on the plane  $[w]_{\lambda} - w$  a strange attractor is shown that corresponds to the values of the parameters B = 2,  $\lambda = 5$ , and A = 2.65. With a further increase in the pressure gradient the motion first is ordered and then becomes random again.

Conclusion. Thus, the specific features of the flow of rheopectic fluids observed experimentally can be explained by the competition of two opposite processes: structure formation at low shear rates and disintegration of the structure upon an increase in the shear rate. It is shown that the presence of a retardation time in the processes of structure disintegration can lead to the appearance of nondecaying oscillations of the flow rate of a fluid moving under the action of a constant pressure gradient. Analytical and numerical analyses of the model suggested make it possible to elucidate the structure of the phase space of the system, which can have several stable equilibrium points as well as attracting sets in the form of limiting cycles and strange attractors. The presence of such a complex phase pattern broadens the possibility of controlling the motion of a rheopectic fluid by "bringing" it to the required mode of flow.

## NOTATION

G, mass flow rate of the fluid; w, mean flow velocity;  $\theta$ , pressure gradient; t, time;  $\rho$ , fluid density; R, L, radius and length of the tube; c, velocity of sound;  $\tau$ , shear stress;  $\tau_0$ , limiting shear stress; s, concentration of structural bonds;  $\dot{\epsilon}$ , rate of shear strain;  $\gamma$ , coefficient determining the dependence of  $\tau_0$  on s;  $\lambda$ , time of retardation in the process of breakdown of structural bonds;  $\alpha$ ,  $\beta$ , coefficients of restoration and destruction of structural bonds. Variables with asterisks denote certain characteristic values of the quantities.

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