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SELF-EXCITED AND STOCHASTIC OSCILLATIONS IN THE HYDRODYNAMICS OF NON-NEWTONIAN FLUIDS†

I. SH. AKHATOV, M. M. KHASANOV and I. G. KHUSAINOV

Ufa

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An approach is developed in accordance with which the rheological behaviour of non-Newtonian fluids is governed by the non-linear kinetics of processes involving the restoration and disruption of bonds between the structural elements of the medium. The corresponding kinetic equations are used as closing relationships in describing the unsteady motions of such fluids in the gap between the cylinders of a rotational viscosimeter. It is shown, by a numerical analysis of the method proposed, that the self-excited and stochastic oscillations when measuring the viscosity of non-Newtonian fluids may be the result of the joint action of the non-linear kinetics of structural changes in the fluid and the inertial properties of the viscosimeter.

EXPERIMENTAL investigations of the rheological properties of fluids with a complex internal structure (clay suspensions, paraffin and polymer melts, heavy crude oils, etc.), carried out using a rotating cylinder viscosimeter, show that, in a number of cases, at a constant rate of rotation of the viscosimeter motor the magnitude of the measured tangential stress may vary with time in a quite complex manner. A qualitative description of this effect has been given in [1]. It has been shown experimentally [2] that, under certain conditions, it is possible to realize an oscillatory outflow of a polymer melt from a capillary tube at a constant velocity of the displacement piston. This phenomenon has come to be known as elastic turbulence and is explained, in particular, by the hypothesis of the slip of the fluid close to the wall of the viscosimeter or the capillary [3–5].

The dependence of the rotational viscosimeter readings on time, obtained during measurements of the tangential stress **R** of a paraffin (nonadecane) melt at a temperature close to its temperature of crystallization,‡ is shown in Fig. 1. The characteristics of the irregular oscillatory process, that is, the correlation scale and the Kolmogorov entropy were calculated for the curve shown. The correlation integral was calculated using the formula [6]

$$C_M(r) = \frac{1}{N^2}\sum_{i=1}^{N}\sum_{q=1}^{N} H(r - \|R_i - R_q\|)$$

where N is the total number of points in the processed time interval, H is the Heaviside function, r is the length of a cell in phase space, R_i is a point of the M-dimensional phase space with coordinates $\{R(t_i), \ldots, R(t_i+(M+1)\Delta t)\}$ and $\|R_i - R_q\|$ is the distance between the ith and qth points of phase space. The correlation scale was calculated using the formula [6]

$$W = \ln C_M(r) / \ln r$$

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‡ The experiment was carried out by S. A. Konev.

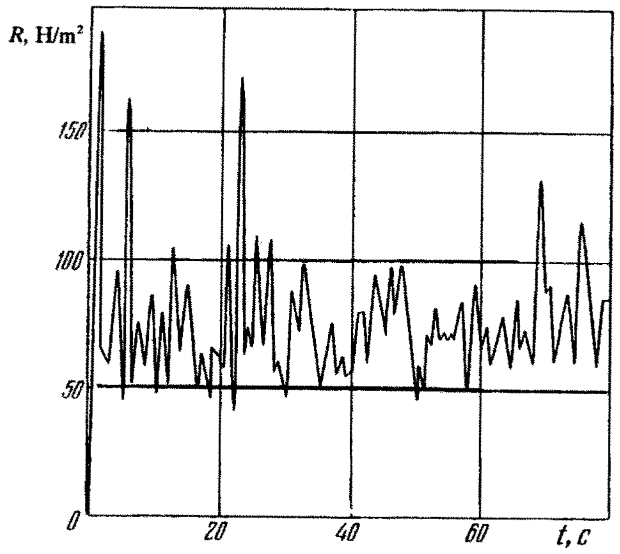

FIG. 1.

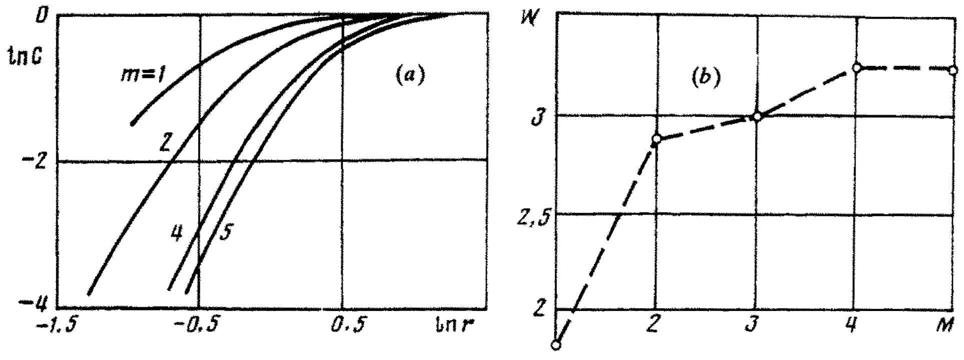

FIG. 2.

The dependences of the correlation function $C_M(r)$ and the correlation scale W on the number of phase variables M are shown in Fig. 2(a, b), respectively. The lower bound for the Kolmogorov entropy was calculated using the formula [7]

$$K_2 = \lim_{r\to 0} \lim_{M\to\infty} \ln[C_M(r)/C_{M+1}(r)]$$

Calculations show that $K = 0.1$ in the given case. This means that the Kolmogorov entropy is positive. This suggests that the process shown in Fig. 1 is chaotic. It is seen in Fig. 2(b) that, when $M = 4$, the value of the correlation scale W reaches a stationary level. This means that the chaos is determinate and that the minimum number of variables required to model the process of the deformation of the paraffin melt in the rotational viscosimeter is equal to four. Consequently, the process under consideration can be adequately described by a fourth-order non-linear dynamic system. This suggests the idea of the fundamental possibility of the existence of certain finite kinetics for the processes involved in the change in the internal structure of the fluid.

The proposal has been put forward in [8] that structural rearrangements occurring during the flow process are the cause of the occurrence of elastic turbulence. Linear kinetic equations were used in [9, 10] in order to simulate the processes involved in the breakdown and restoration of the structure. It was shown that many of the special features of the steady-state rheological characteristics of a fluid can be satisfactorily described within the framework of such an approach. It was proposed in [1] that non-linear models, which generalized classical models of the "predator–prey" type, should be used when describing the kinetics of the restoration–disruption processes. It was noted that new possibilities for describing complex, unsteady (self-excited and chaotic) conditions are opened up when using non-linear models in addition to their ability to describe steady-state rheological characteristics.

In what follows we present a simple non-linear kinetic equation for describing the processes involved in the restoration and disruption of the bonds between the structural elements of the medium. This is used when simulating the motion of the fluid in the gap between the cylinders of a rotational viscosimeter.

Assuming that the width of the gap in the viscosimeter is small compared with the radii of the cylinders, let us consider the following planar scheme for the flow of the fluid between two parallel infinitely extending plates separated from one another by a distance h. The lower fixed and upper moveable plate also possess sufficiently large areas so that it is possible to neglect edge effects. The upper plate is brought into translational motion by means of a spring, one end of which is connected to the plate while the other moves at a constant velocity v_0.

The shear flow of the fluid between the plates is described by the equation

$$\rho\frac{\partial v}{\partial t} = \frac{\partial}{\partial t}\left(\mu\frac{\partial v}{\partial y}\right), \quad 0 < y < h \tag{1}$$

(v, ρ and μ are the velocity, density and viscosity of the fluid, respectively). The equation of motion of the upper plate can be written in the form

$$m\frac{d^2x}{d^2t} - Q\mu\frac{\partial v}{\partial y}\bigg|_{y=h} + fx = 0 \tag{2}$$

where x is the absolute elongation of the spring, f is the coefficient of rigidity of the spring and m and Q are the mass and area of the upper plate. The system of equations of motion of the fluid and the plate (1) and (2) is closed using boundary and kinematic conditions of the form

$$v(0, t) = 0, \ v(h, t) = v_0 - dx/dt \tag{3}$$

As the quantitative characteristic of the degree of structuring of the fluid, which determines its viscosity, we shall use the concentration of bonds s which are disrupted during the flow process. The dependence of the viscosity of the fluid on the concentration s may be taken in the form

$$\mu(s) = \mu_0\frac{\xi - \xi_*}{1 - \xi_*} - \mu_*\frac{1 - \xi}{1 - \xi_*} \tag{4}$$

$$\xi = \exp(-\sigma s^{\beta}), \ \xi_* = \exp(-\sigma s_*^{\beta})$$

where α and β are certain positive constants. In accordance with this parametrization, the viscosity of the fluid is the greatest and equal to $\mu|_{s=0} = \mu_0$ at a concentration of disrupted bonds equal to zero. As more bonds become disrupted (that is, as s increases), the viscosity decreases exponentially and reaches its minimum value $\mu|_{s=s_*} = \mu_*$ when all of the bonds are disrupted ($s = s_*$).

Under shear flow, the bonds between the structural elements of the medium can be disrupted and restored. In order to describe the bond disruption and restoration process, let us introduce the following non-linear kinetic equation

$$ds/dt = -\alpha\,\{s - s_*[1 - \exp(-\gamma s\mu(s)\,\dot{\epsilon}^2)]\} \tag{5}$$

where α and γ are positive constants and $\dot{\epsilon} = \partial v/\delta y$ is the shear rate. In accordance with this equation, the concentration of disrupted bonds at a constant value of the shear rate must tend to a certain equilibrium value s_0 which is determined using the formula

$$s_0 = s_*[1 - \exp(-\gamma s_0 \mu(s_0)\epsilon^2)]$$

It is seen from this that, as $\dot{\epsilon}$ increases, the concentration of disrupted bonds increases and approaches its maximum value s_* exponentially. Moreover, the expansion of the right-hand side of Eq. (5) in series

$$s_*[1 - \exp(-\gamma s \mu(s)\,\dot{\epsilon}^2)] \approx s_* \gamma s \mu(s)\,\dot{\epsilon}^2$$

shows that, at small values of the shear rate, the rate of bond disruption is directly proportional to the intensity of the viscous dissipation of energy in the flow.

After introducing the dimensionless variables

$$V = \frac{v}{v_0}, \quad \tau = \alpha t, \quad \eta = \frac{y}{h}, \quad S = \frac{s}{\sigma^{1/\beta}}, \quad X = \frac{\alpha x}{v_0}$$
$$\nu(S) = \mu(S)\,\frac{1 - \exp(-\sigma s_*^{\beta})}{\mu_0 - \mu_*}$$

the system of equations (1)–(5) acquires the form

$$\begin{aligned}
&\kappa \frac{\partial V}{\partial \tau} = \frac{\partial}{\partial \eta}\left(\nu(S)\frac{\partial V}{\partial \eta}\right), \quad \kappa = \rho\alpha h^2 \frac{1 - \exp(-\sigma s_*^{\beta})}{\mu_0 - \mu_*}\\
&\frac{d^2 X}{d\tau^2} - \lambda\nu(S)\left.\frac{\partial V}{\partial \eta}\right|_{\eta=1} + FX = 0\\
&V(0, \tau) = 0, \quad V(1, \tau) = 1 - \frac{dX}{d\tau}\\
&\nu(S) = P + \exp(-S^{\beta})\\
&\frac{\partial S}{\partial \tau} = -S + A\left[1 - \exp\left(-GS\nu(S)\left(\frac{\partial V}{\partial \eta}\right)^2\right)\right]\\
&\lambda = \frac{Q[\mu_0 - \mu_*]}{m\alpha h(1 - \exp(-A^{\beta}))}, \quad F = \frac{f}{m\alpha^2}, \quad P = \frac{\mu_* - \mu_0\exp(-A^{\beta})}{\mu_0 - \mu_*}\\
&A = S_*\sigma^{1/\beta}, \quad G = \frac{\gamma}{\sigma^{1/\beta}}\,\frac{v_0^2}{h^2}\,\frac{\mu_0 - \mu_*}{1 - \exp(-A^{\beta})}
\end{aligned} \tag{6}$$

The formulation of the problem which has been given can be simplified when account is taken of the smallness of the parameter $\kappa(\kappa \sim h^2)$. In this case, one can neglect the term $\kappa\partial V/\partial\tau$ in the first equation (6) and then

$$T = \nu(S)\,\partial V/\partial\eta = \text{const}$$

The solution of this equation is not unique. It can be constructed as a set of spatial structures, "domains" [12, 13], which are regions with different values for the concentration of disrupted bonds s_i and shear rates $(\partial V/\partial\eta)_i$ (the concentration of disrupted bonds and the shear rate are independent of η within each domain). In this case, the boundary conditions (the fourth and fifth relationships of (6)) are satisfied if

$$\sum_{j=1}^{J}\left(\frac{\partial V}{\partial \eta}\right)_j Z_j = 1 - \frac{dX}{d\tau}, \quad \sum_{j=1}^{J} Z_j = 1$$

where J is the number of domains and Z_j is the thickness of the jth domain.

The system of equations (6) then reduces to a non-linear dynamic system of the form

$$\frac{d^2X}{d\tau^2} - \lambda B + FX = 0, \quad B = \left(1 - \frac{dX}{d\tau}\right)\left(\sum_{j=1}^{J} \frac{Z_j}{1/\nu(S_j)}\right)^{-1}$$

$$\frac{dS_j}{d\tau} = -S_j + A\left[1 - \exp\left(-GS_j\frac{B^2}{\nu(S_j)}\right)\right]$$

$$\nu(S_j) = P + \exp(-S_j^{\beta}), \quad j = 1, \ldots, J$$

It has been shown above that, according to the experimental data, it is sufficient to use a fourth-order non-linear dynamic system to simulate deterministic chaos in a rotational viscosimeter. System (7) was therefore solved numerically for the case of two domains. Here, the following values of the dimensionless parameters were fixed: $\beta = 0.25$, $F = 17$, $A = 4$, $P = 0$, 1, $Z_1 = Z_2 = 0.5$ and $\beta = 10$. The effect of the dimensionless value of the shear rate $E = \sqrt{G}$ on the structure of the solutions of the system was investigated.

The results of the calculations are summarized in Fig. 3 where the dependence of the dimensionless tangential stress T on $\dot{E}$ is shown. The dependence characterizes the equilibrium positions of the dynamic system being considered to which, in the case of stability, the solution tends with time. In Fig. 3, these stable branches are plotted using bold lines. At small values of the shear rate ($\dot{E} < 0.95$), the structural bonds in the fluid are not disrupted. A simple shear flow of a fluid with a high viscosity occurs in which there is no separation of the flow field into domain structures. If, for any reason whatsoever, the disruption of some of the structural bonds occurs at the initial instant of time, that is, $S_1|_{t=0} = S_{1*} \neq 0$, then those disrupted bonds are completely restored in the course of time, that is, $S_1|_{t\to\infty} = 0$.

When the shear rate $\dot{E}$ increases ($\dot{E} \geqslant 0.95$) separation of the flow field into domain structures occurs, with the disruption of some of the bonds close to the moving wall. Here, the zeroth state $S_1 = 0$ loses stability with the production of a new equilibrium position $S_1 \neq 0$ which, in its turn as $\dot{E}$ increases still further, loses stability with the formation of the limit cycle. The range of oscillations in the magnitude of the dimensionless tangential stress ($T^{\max}(\dot{E})$ and $T^{\min}(\dot{E})$) is shown in Fig. 3 by the dashed lines. The time average values of T accompanying these self-excited oscillations are shown in Fig. 3 by the thin solid line.

When the shear rate is increased still further, the process of the successive doubling of the period of the self-excited oscillations occurs. This sequence of bifurcations involving the doubling of the period when $\dot{E} \geqslant \dot{E}_\infty = 1.52$ passes into chaos. Analysis of the one-dimensional Lorentz mapping (Fig. 4) corresponding to this chaos shows that, in the system investigated, the transition to chaos occurs in accordance with the classical Feigenbaum scenario. This is also suggested by the fact that the values of the parameter $G_n = \dot{E}_n^2$, at which doubling of the period of the self-excited oscillations occurs, obey the Feigenbaum law [14] $G_n - G_\infty = c/\delta^n$,

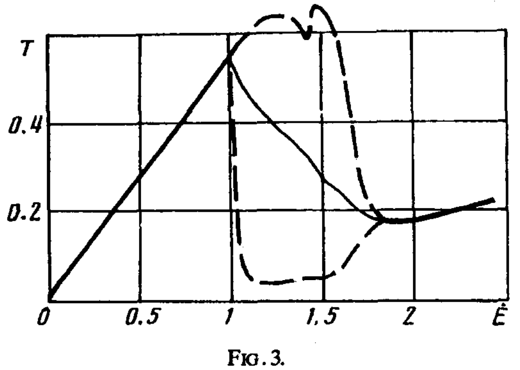

FIG. 3.

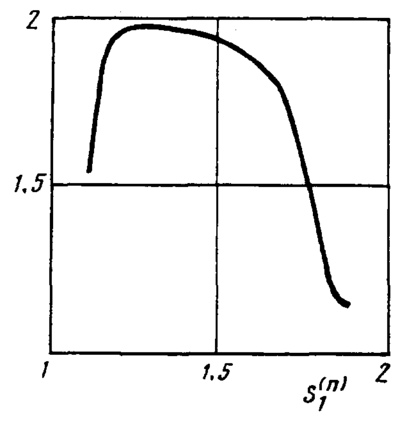

FIG. 4.

where c is a constant (in the case considered, $c = -6.54$) and δ is the universal Feigenbaum constant.

$$K = \tau^{-1} \ln(L(\tau)/L(0))$$

For stochastic behaviour to exist it is necessary that mixing conditions are satisfied which is ensured by the exponential divergence of the trajectories at each point of the attractor. The nature of this divergence may be evaluated by investigating the Kolmogorov entropy [7] which is defined by the expression
where $L(0)$ and $L(\tau)$ are the distances between the two points in the phase space (X_1, X_2, S_1, S_2), respectively, at the initial instant of time and after a time period τ. Calculations show that, in the course of time, the value of the Kolmogorov entropy reaches a positive stationary level $K_\infty = 0.057$.

When the shear rate is increased further $\dot{E} > 1.67$, a reverse cascade of the period doubling bifurcations occurs which, at $\dot{E} = 1.87$, leads to the disappearance of self-excited oscillations with the formation of a stable equilibrium. This equilibrium is characterized by a high degree of disruption of the structural bonds close to the mobile wall of the domain. As $\dot{E}$ increases, the number of the disrupted bonds increases and asymptotically tends to its maximum value.

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