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SHEAR INSTABILITY OF THE STRUCTURE OF MEDIA POSSESSING VISCOUS FLUIDITY

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Instability of the structure of viscous fluids in the Couette flow regime (spontaneous formation of bands, which are tangential discontinuities in terms of viscosity) is experimentally found. This process is demonstrated to be analogous to formation of shear bands in polymethylmethacrylate (Plexiglas) under plastic shear strains.

Key words: *structural viscosity of fluids, shear strains, instability of the structure.*

The mechanisms of fracture of deformable solids has been fairly well studied in mechanics of fracture of condensed media [1, 2], though the general theory of fracture of fluid and fluid-type (dye, bitumen, paste, gel, etc.) media has not bee constructed yet.

It is known from numerous experiments that volume extension of low-viscosity fluids (water, emulsions and suspensions with a low-viscosity matrix, etc.) with arbitrary geometry and flow regime leads to unlimited cavitation (intense growth of bubbles from cavitation seeds) with subsequent fragmentation of the foamy medium under the action of capillary forces. According to [3, 4], however, such a process does not develop under pulsed volume extension of high-viscosity fluid samples. Two types of fragmentation are typical of these media. In the divergent flow regime, the thickness of a spherical or cylindrical layer of the high-viscosity fluid decreases, and the primary disturbances develop into jet structures on the inner and outer surfaces, which then decompose into individual fragments under the action of capillary forces. In the case of pulsed transportation of a high-viscosity fluid sample in a cylindrical channel, the medium is finally stratified into "fibrous" fragments stretched in the streamwise direction [3]. As in the former case, fragmentation of the high-viscosity fluid is not accompanied by the development of unlimited cavitation in the entire volume, which is attributed in [5] to dissipation of energy in the vicinity of expanding cavitation seeds owing to high shear viscosity of the fluid. Thus, the question on the mechanism of splitting of a high-viscous fluid volume into "fibrous" fragments arises.

A series of experiments with glycerin filling the gap between two counter-rotating coaxial cylinders was performed in [6]. It was found that a tangential discontinuity in terms of viscosity (TDV) is formed in glycerin along the line of contact of the opposing coaxial flows, where a local maximum of shear strain is observed: the shear viscosity μ drastically decreases in a narrow annular layer, and the medium behaves as a low-viscosity fluid. A zone of a local discontinuity in the shear strain rate $\dot{\varepsilon}_\tau$ was artificially generated in these experiments to initiate TDV formation.

Nevertheless, as "fibrous" stratification of a cylindrical glycerin sample under uniaxial radial loading was experimentally observed in [3], the following question arises: Is spontaneous origination of a region with a tangential discontinuity of the viscosity field μ possible in a viscosimetric flow with a constant-strain history (the Couette flow type) and with a monotonic distribution of $\dot{\varepsilon}_\tau$ in the direction perpendicular to the main flow?

1. To study the stability of the structure of fluids in a shear flow, we used the experimental setup described in [6]. The setup was tuned to an operation regime illustrated in Fig. 1. The transparent bottom of the outer cylinder allowed video filming of the structure of the cylindrical fluid layer from the side of its bottom.

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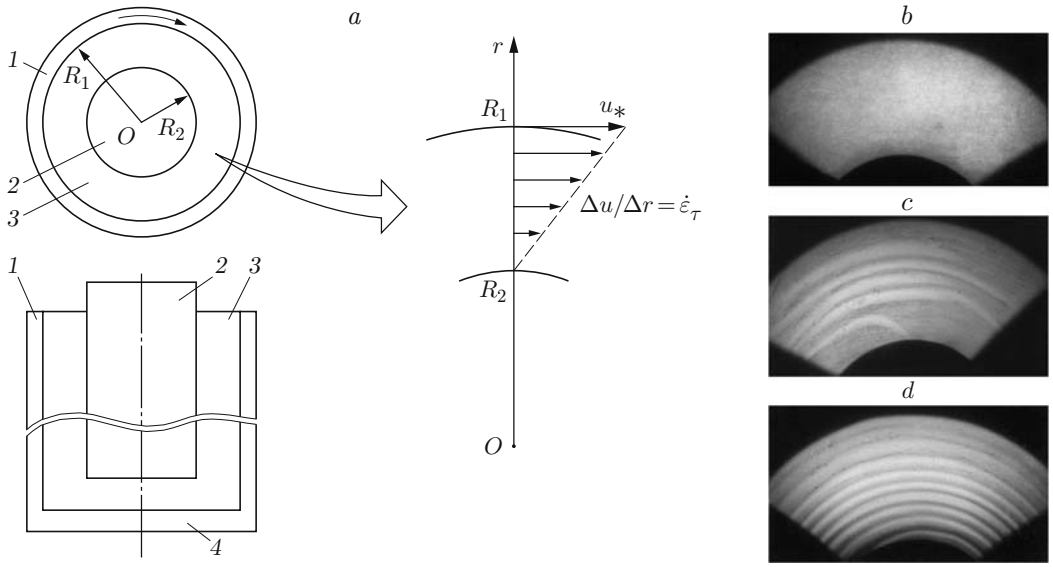

Fig. 1. Layout of the experiment (a) and evolution of the structure of the medium in a shear flow in a cylindrical fluid layer (b–d): (a) elements of the setup [rotating outer cylinder (1), motionless inner cylinder (2), examined fluid (3), and bottom of the outer cylinder (4)]; (b) water; (c) aqueous solution of sodium chloride; (d) glycerin.

The experiments were performed with the following fluids arranged in order of increasing their maximum (initial) values of shear (structural) viscosity μ_*: water ($\mu_* = 10^{-3}$ Pa·sec and $T = 17°$C), 9.2% solution of sodium chloride in water ($\mu_* = 1.3 \cdot 10^{-3}$ Pa·sec and $T = 15°$C), and glycerin ($\mu_* = 0.85$ Pa·sec and $T = 25.6°$C; $\mu_* = 1.48$ Pa·sec and $T = 20°$C; $\mu_* = 1.9$ Pa·sec and $T = 17.7°$C; $\mu_* = 2.9$ Pa·sec, $T = 12.8°$C, $\mu_* = 4$ Pa·sec, and $T = 10°$C). The series of experiments yielded the following results. In pure water, at least at $T \geqslant 3°$C, no visible violation of optical homogeneity of the medium in the Couette flow regime occurs (Fig. 1b). In the 9.2% aqueous solution of sodium chloride at $T \leqslant 8°$C we can see spontaneous incipience of arc-shaped lines (optical inhomogeneities, Fig. 1c) corresponding to a local violation of the order of the medium structure [7] in 4–5 min after the Couette flow is formed. As a consequence, the structural viscosity decreases [6]. In 3–5 sec, these inhomogeneities are "healed" and then appear in other regions of the flow. In the case of the Couette flow in glycerin, stable TDVs are formed (Fig. 1d). (The decrease in μ in TDVs was registered by the technique described in [6].) In this case, the time of TDV formation decreases with increasing μ_*. As the shear strain rate $\dot{\varepsilon}_\tau$ increases, the zones of tangential discontinuities expand and straps are formed between them; this resembles the evolution of the slipping lines in the shear strain zone in solid-plastic materials [2].

Figure 2 shows the experimentally measured time of formation of tangential discontinuities in terms of structural viscosity Δt_* as a function of $\dot{\varepsilon}_\tau$ in the Couette flow in a wide range of initial values of structural viscosity.

2. It follows from the experimental results obtained that the stability of the structure of a viscous polar fluid in the shear flow regime depends on the initial value of structural viscosity μ_* and on the shear strain rate $\dot{\varepsilon}_\tau$. In low-viscosity fluid flows, no visible changes in their structure are observed. Signs of spontaneous origination of TDVs are observed with increasing μ_*, and stable TDVs are formed at $\mu_* \geqslant 1.5$ Pa·sec. The higher the values of μ_* and $\dot{\varepsilon}_\tau$, the shorter the time of formation of tangential discontinuities, i.e., the time of development of structural instability of polar fluids in the Couette flow regime. To find the reason for this phenomenon, we performed a qualitative analysis of disturbances of the medium parameters by an example of a plane Couette flow (Fig. 3a and b). As the fluid possesses a certain viscosity μ, it moves with a velocity u_* at the boundary with the upper moving plate, and a shear flow with the linear velocity in the y direction being distributed as $u = u_* y/h$ [8] is formed toward the lower motionless plate in the fluid band owing to diffusion of momentum with the diffusivity $\eta = \mu/\rho$ (ρ is the fluid density). Hence, the shear strain rate has the same value over the entire flow regime in an undisturbed shear flow in the fluid with homogeneous distributions of its physical parameters (including structural

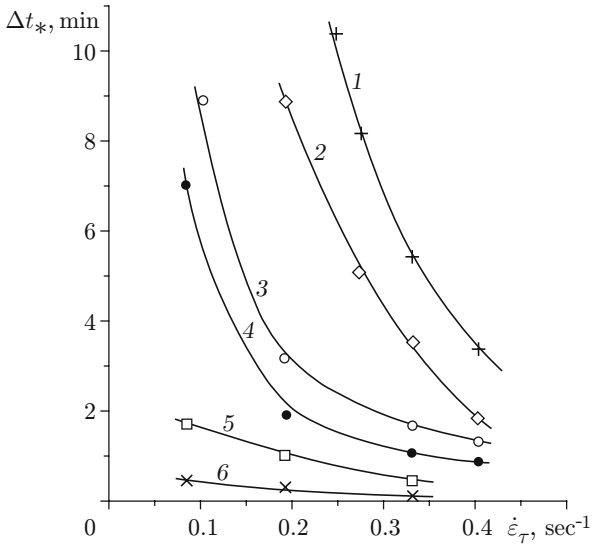

Fig. 2. Time of TDV development in fluids as a function of the shear strain rate and structural viscosity of the medium: $\mu_* = 0.612$ (1), 0.895 (2), 1.48 (3), 1.9 (4), 2.9 (5), and 4 Pa·sec (6).

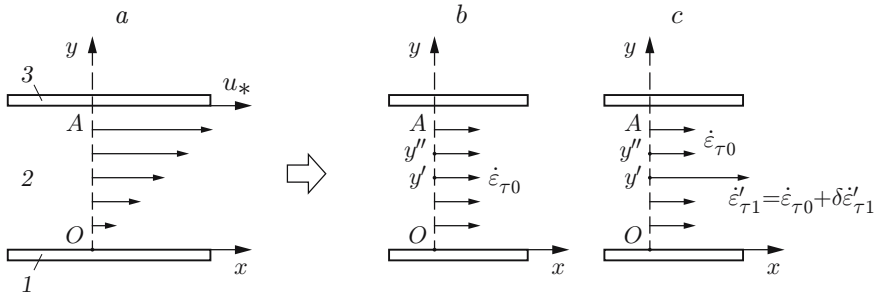

Fig. 3. Development of instability of the structure of viscous fluids in the plane Couette flow: velocity distribution in the Couette flow (a), diagram of the shear strain rate (b), and spontaneous local disturbance of the field of shear strain rates (c); lower motionless solid plane boundary (1), viscous polar fluid (2), and upper solid plane boundary moving with a constant velocity u_* (3); $OA = h$ is the distance between the parallel plates.

viscosity) over its volume and a homogeneous temperature field: $\dot{\varepsilon}_\tau = \delta u/\delta y = [u''(y'') - u'(y')]/(y'' - y') = u_*/h$ (Fig. 3b). As polar fluids at rest have an ordered structure [7], which is destroyed in the shear strain region, we can use the Powell–Eyring formula [9] in the approximation of the generalized Newtonian fluid:

$$\mu = \mu_0 + (\mu_* - \mu_0)\,\frac{\operatorname{arsinh}\sqrt{s}\lambda}{\sqrt{s}\lambda}. \tag{1}$$

Here μ_* is the initial value of structural viscosity of the medium, μ_0 is the asymptotic value of viscosity (Newtonian viscosity of the fluid with a fractured structure), $s = 2D : D$ (D is the strain-rate tensor), and λ is a constant with a dimension of time. For the Couette flow considered in the present paper $s = \dot{\varepsilon}_\tau^2$, λ is the shear stress relaxation time, and Eq. (1) acquires the form

$$\bar{\mu} = \bar{\mu}_0 + (1 - \bar{\mu}_0)\,\frac{\operatorname{arsinh}\mathrm{De}}{\mathrm{De}}, \tag{2}$$

where $\bar{\mu} = \mu/\mu_*$, $\bar{\mu}_0 = \mu_0/\mu_*$, and $\mathrm{De} = \dot{\varepsilon}_\tau\lambda$ is the Deborah number. According to Eq. (2), if $\bar{\mu}_0$ is of the order of unity, i.e., $(\mu_* - \mu_0)/\mu_* \ll 1$, then we have $\bar{\mu} \approx \bar{\mu}_0$ for all values of De, which is actually observed in experiments with water: no visible violation of homogeneity of the structure of water is observed with increasing $\dot{\varepsilon}_\tau$. As $\bar{\mu}_0$ decreases, the effect of the Deborah number on structural viscosity of the medium becomes more pronounced. Thus,

it was found [6] that $\bar{\mu}_0 \approx 0.32$ for glycerin at $T = 19°\mathrm{C}$ which is the reason for formation of clearly expressed TDVs in the Couette flow.

The Powell–Eyring formula, nevertheless, does not take into account the time evolution of μ with changing $\dot{\varepsilon}_\tau$. Thus, if the shear strain rate instantaneously acquires a certain value $\dot{\varepsilon}'_\tau$, Eq. (2) predicts that the structural viscosity instantaneously decreases to the corresponding value $\mu' = \bar{\mu}\mu_*$; if $\dot{\varepsilon}_\tau$ instantaneously vanishes, the structural viscosity immediately recovers the initial value μ_*. According to the experimental results of [6], however, TDV "healing" in glycerin occurs within tens of seconds (15 min at $T = 19°\mathrm{C}$). It follows from the experimental data plotted in Fig. 2 that visible TDVs are formed after a certain latent process governing the initial stage of fracture of the ordered structure of the fluid, which lasts during a certain time Δt_* depending on μ_* and $\dot{\varepsilon}_\tau$ (the higher μ_* and $\dot{\varepsilon}_\tau$, the shorter the latent period of TDV formation). It should be noted that TDVs are also formed at rather low values of $\dot{\varepsilon}_\tau$ (at least, at $\dot{\varepsilon}_\tau \leqslant 0.1\ \mathrm{sec}^{-1}$).

Origination of this phenomenon can be qualitatively explained by an example of a plane Couette flow (see Fig. 3) as follows. Let the medium have the following parameters in an undisturbed steady-state flow: temperature T_0, density ρ_0, structural viscosity μ_*, and kinematic viscosity (momentum diffusivity) $\eta_0 = \mu_*/\rho_0$. We fix the points y' and $y'' = y' + \delta y$ within the segment OA (Fig. 3b and c) so that the condition $\delta y/y' \ll 1$ is satisfied. The flow velocities at these points are $u'_0(y') = u_* y'/h$ and $u''_0(y'') = u_* y''/h$, respectively; hence, the shear strain rate at the point y' is $(u''_0 - u'_0)/\delta y = u_*/h$. If a temperature fluctuation occurs at a certain time at the point y' and the temperature increases to $T_1 = T_0 + \delta T'$ ($\delta T' \ll T_0$), the structural viscosity decreases by a certain value $\delta\mu_1$ at the point y', because the structural viscosity decreases with increasing T. According to numerous experimental data (see [10]), a change in ρ with changing T for fluids is a second-order quantity, as compared to a change in μ; therefore, the momentum diffusivity at the point y' decreases to $\eta_1 \approx (\mu_* - \delta\mu_1)/\rho_0$ with increasing T. A decrease in η leads to a decrease in the flow velocity at the point y' to a certain value $u'_1 = u'_0 - \delta u_1(\eta_1)$, which means that the shear strain rate at this point increases to

$$\varepsilon'_{\tau 1}(y') = \frac{u''_0 - u'_1}{\delta y} = \frac{1}{\delta y}\left[y''\,\frac{u_*}{h} - \left(y'\,\frac{u_*}{h} - \delta u_1\right)\right] = \dot{\varepsilon}'_{\tau 0} + \frac{\delta u_1}{\delta y} = \dot{\varepsilon}'_{\tau 0} + \delta\dot{\varepsilon}'_{\tau 1}.$$

As a result, the parameter De at the point y' increases to $\mathrm{De} = \lambda(\dot{\varepsilon}'_{\tau 0} + \delta\dot{\varepsilon}'_{\tau 1})$ and, according to Eq. (1), a further decrease in structural viscosity is observed. This, in turn, leads to a decrease in η_1, a decrease in $u'_1(y')$, an increase in $\dot{\varepsilon}'_{\tau 1}(y')$ and De, a further decrease in structural viscosity, etc. We can easily show that the same process occurs for negative values of $\delta T'$.

Similar experiments performed with 3, 5, and 10% aqueous solutions of glycerin ($\mu_* = 0.82$, 0.54, and 0.11 $\mathrm{Pa}\cdot\mathrm{sec}$, respectively, at $T = 20°\mathrm{C}$) show that the structure of the medium in the shear flow is also unstable in the case of solutions of polar fluids. TDVs are formed in these solutions in the Couette flow regime, but their latent time Δt_* is substantially smaller than that in a homogeneous viscous fluid. As the condition of macrofracture of the structure of a condensed medium is the time of recovery of intermolecular bonds Δt_+ exceeding the time of bond breakdown Δt_- [11], i.e., the condition $\Delta t_- < \Delta t_+$ should be satisfied for TDV formation under shear strains, apparently, the ratio $\Delta t_-/\Delta t_+$ in glycerin solutions is smaller than that in the case of homogeneous fluids (components of the solution).

Thus, if there are some minor spontaneous fluctuations of temperature or other physical parameters of the process affecting the value of η (e.g., a change in $\dot{\varepsilon}_\tau$ or ρ, and hence, $\eta = \mu/\rho$) in a local region of the shear flow of a fluid possessing structural viscosity ($\mu_* > \mu_0$), this leads to the development of instability of the medium structure. At the initial stage, this process has a latent character: the initially arising local increase in $\dot{\varepsilon}_\tau$ grows with time and finally reaches a value at which an intense decrease in structural viscosity begins, i.e., the decrease in μ occurs in the so-called regime with aggravation [12].

3. To study the behavior of the structure of media with different rheological properties under shear loading, we performed a series of experiments to examine the evolution of the structure of Plexiglas (polymethylmethacrylate) under shear strains. The ultimate strength of polymethylmethacrylate (PMMA) under shear loading is $\tau_* = 490\ \mathrm{kg/cm^2}$ at $T = 20°\mathrm{C}$. The experiments were performed by the scheme shown in Fig. 4. The experimental setup contained the following components: polished PMMA sample (rectangular parallelepiped $1.7 \times 1.0 \times 0.6$ cm), article fixing the PMMA sample in the bed frame, sample segment $ABCD$ where shear deformation occurs under the action of the force F applied to the rectangular insert (the force is initiated by loading the insert by a special spring device equipped with a dynamometer). The entire experimental unit was placed into a water bath, which

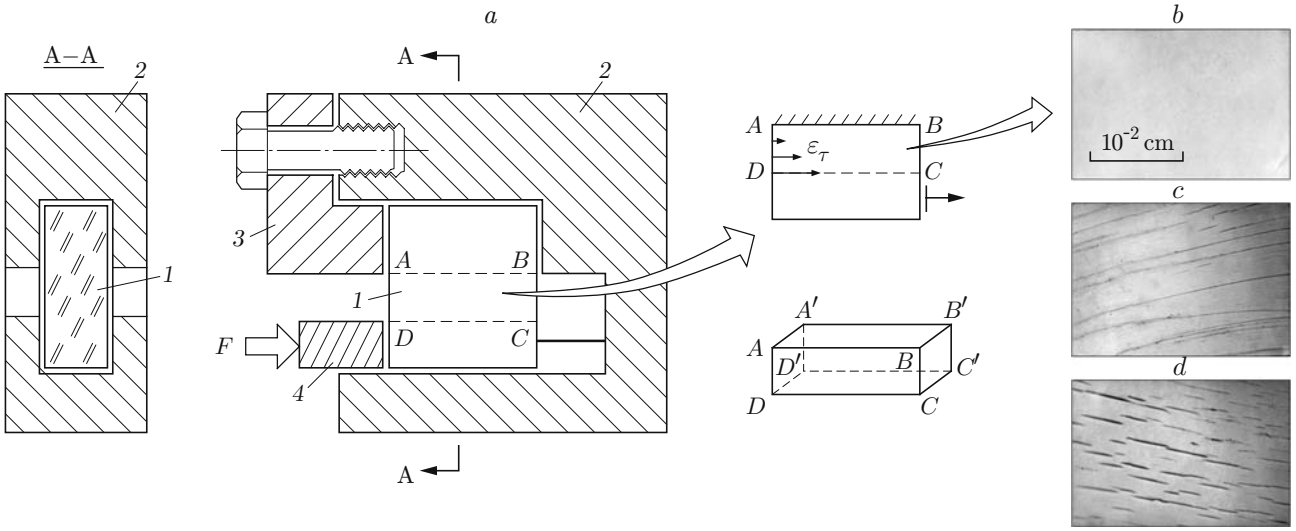

Fig. 4. Scheme of the experiment (a) and shear instability of the PMMA (Plexiglas) structure (b–d): (a) elements of the setup: 1) polished PMMA sample; 2) bed frame; 3) element fixing the PMMA sample in the bed frame; 4) insert; $ABCD$ is the segment of the sample where shear deformation occurs; (b) initial state of the material structure at $\tau = 0$; (c) material structure at $T = 70^\circ$C in 10 sec after the stress $\tau = 362$ kg/cm^2 is applied to the sample; (d) material structure at $T = 23^\circ$C in 3.5 h after the stress $\tau = 362$ kg/cm^2 is applied to the sample.

allowed us to vary the temperature of the loaded sample from +18 to +90°C. The action of the force F formed the field of shear stresses τ in the sample and, hence, shear strains ε_τ responsible for the evolution of the sample structure, which was registered by a microscope equipped with a camera adapter.

Figure 4 also shows the microphotographs of the PMMA sample segment. An analysis of the microphotographs showed the following. For values of T close to the PMMA melting point, the structure of the sample during plastic shear deformation (creep mode) is unstable; it acquires a layered character (Fig. 4c) and resembles the structure of the glycerin sample in the shear flow regime (see Fig. 1d). With decreasing T, the structural viscosity of the "solidifying" PMMA sample increases; correspondingly, the degree of stratification of the sample material in the zone of the plastic shear flow decreases, and the slip limes become interrupted. Finally, at $T = 23^\circ$C the sample behaves as a solid: the slip lines transform into a network of microcracks (Fig. 4d). It is known that shear strains in plastic materials also form shear bands under shear strains; subsequent coalescence of these bands leads to formation of cracks [2, 13, 14].

Thus, in the Couette flow regime, the structure of high-viscosity fluids is unstable: a system of tangential discontinuities in terms of viscosity is formed in the medium. Moreover, a comparative analysis shows that the character of the evolution of the medium structure under shear strains in fluids possessing structural viscosity is similar to changes in the structure of polymers and plastic metals subjected to shear strains.

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