

HYDRODYNAMICS OF FLUIDS OF VARIABLE VISCOSITY

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An invariant rheological equation is proposed for the flow of fluids with a variable viscosity independent of time. It is shown to be desirable to distinguish a subclass of fluids with a linear fluidity law.

1. Fluid media with variable viscosity are widely used in the chemical and processing industries. Here the term "variable viscosity" denotes that the viscosity is a function not only of the thermodynamic parameters of state (p, T) but also of certain other parameters. It is usual, in this case, to speak of a fluid with anomalous viscosity. However, the multitude of fluid media with variable viscosity makes it virtually impossible to treat them all as anomalous.

As a rule, the variable viscosity is related with the dispersion phase, which has a certain structure. We shall describe as fluids with structural viscosity those media whose viscosity for given p and T is a single-valued function of the tangential stresses\*

$$\mu = \mu(\tau). \tag{1.1}$$

There are a large number of fluids whose viscosities are a complex function of shearing stress and the time during which this stress acts (thixotropic and rheoplectic), as well as fluids with elastic properties (viscoelastic). The hydrodynamics of such fluids will not be considered here.

It should be noted that viscosity is a more complex function of the tangential stress than its reciprocal—fluidity. Therefore it is convenient to construct a phenomenological theory of the flow of fluids with structured viscosity relative to  $\varphi(\tau)$ , which in a one-dimensional flow is defined as

$$\varphi = \frac{w}{\tau - \tau_0} \quad (\tau_0 \text{ is the yield point}). \tag{1.2}$$

2. We shall consider the relation between fluidity and tangential stresses. A considerable number of complex empirical expressions based on more or less successful approximations of the nonlinear flow curve in a certain interval of stresses or shear rates [1, 3] have failed to enter into general use, whereas Ostwald's widely accepted power formula [4], which tempts by the simplicity of the operations involved, will not stand criticism either from the viewpoint of correspondence with the limit properties of  $\varphi(\tau)$  nor from the viewpoint of the dimensionality of the coefficients it contains.

Figure 1 shows schematically the general relation  $\varphi(\tau)$  for the case  $d\varphi/d\tau > 0$  when  $\tau > \tau_1$  according to the experimental data of a series of authors.\* In the region  $\tau < \tau_0$ ,  $\varphi = 0$  and the fluid exhibits so-called conditional elasticity. In the region  $\tau_0 < \tau < \tau_1$  the behavior of the fluid is characterized by constant fluidity  $\varphi_0$ . Naturally, there is some arbitrariness in determining  $\tau_1$ , since the transition from constant fluidity  $\varphi_0$  to variable fluidity is realized smoothly on a certain segment  $\Delta\tau$ .

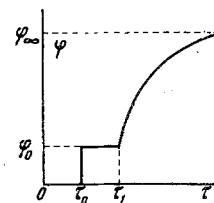


Fig. 1. The nature of the function  $\varphi = \varphi(\tau)$ .

We shall call the quantity  $\tau_1$  the limit of stability of the macrostructure of the fluid, and  $\varphi_0$  the zero fluidity; we shall denote the fluidity as  $\tau \rightarrow \infty$  by  $\varphi_\infty$ .

As the scale of variation of fluidity it is natural to take the difference  $\varphi_\infty - \varphi_0$ , and as the unknown variable the fluidity defect  $\varphi_\infty - \varphi$ . Then from the quantities  $\varphi$ ,  $\varphi_0$ ,  $\varphi_\infty$  and  $\tau - \tau_1$  it is possible to form two dimensionless complexes, by introducing a certain quantity  $\varphi_*$ , which can serve as a measure of the structural stability of the fluid:

$$\varphi_* = \frac{\varphi_\infty - \varphi}{\varphi_\infty - \varphi_0}, \quad \tau_* = \theta \frac{\tau - \tau_1}{\varphi_\infty - \varphi_0}. \tag{2.1}$$

The relation  $\varphi_*(\tau_*)$  has the same character for fluids with both increasing ( $\varphi_\infty > \varphi_0$ ), and decreasing ( $\varphi_\infty < \varphi_0$ ) fluidity. In this case  $d^2\varphi_*/d\tau_*^2 > 0$  and for the fluidity we have

$$d\varphi_* = -\varphi_*^n d\tau_*. \tag{2.2}$$

Correspondingly,

$$\varphi_* = 1 \text{ when } \tau_0 < \tau < \tau_1, \tag{2.3}$$

$$n = 1, \quad \varphi_* = \exp(-\tau_*) \text{ when } \tau > \tau_1, \tag{2.4}$$

$$n \neq 1, \quad \varphi_* = [1 - \tau_*(1-n)]^{\frac{1}{n-1}} \text{ when } \tau > \tau_1. \tag{2.5}$$

Whence it follows that the fluidity for  $n = 1$  is characterized by a set of five quantities  $\{\tau_0, \tau_1, \varphi_0, \varphi_\infty, \theta\}$ .

\*Other terms are also used for such fluids: "non-Newtonian fluids with rheological characteristics independent of time" [1], "generalized newtonian fluids" [2].

\*For so-called dilatant fluids  $d\varphi/d\tau < 0$ .

In strongly structured fluids the break-up of the structure under the influence of shearing stress leads to a considerable change in fluidity. Such fluids include polymer melts, concentrated suspensions, pastes, bitumens, etc.

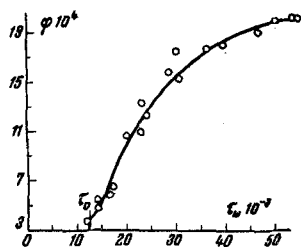


Fig. 2. Bitumen M III,  $t = 20^\circ \text{C}$ ,  $\varphi_0 = 3.8 \cdot 10^{-4} \text{P}^{-1}$ ,  $\varphi_\infty = 20 \cdot 10^{-4} \text{P}^{-1}$ ,  $\tau_0 = 12.5 \cdot 10^3 \text{ dyne/cm}^2$ .

For weakly structured fluids (polymer solutions, low-concentration suspensions, emulsions, latexes) the change in fluidity under the action of shearing stress is relatively slight.

At sufficiently small  $\tau_*$  the region of  $\tau$  of practical interest is satisfactorily described by the first two or three terms of the expansion of Eq. (2.4). Rewriting the latter in the form\*

$$\varphi = \varphi_\infty - (\varphi_\infty - \varphi_0) \exp\left(-\theta \frac{\tau - \tau_1}{\varphi_\infty - \varphi_0}\right), \quad (2.6)$$

we get

$$\varphi = \varphi_0 + \theta(\tau - \tau_1) - \frac{\theta^2}{2} \frac{(\tau - \tau_1)^2}{\varphi_\infty - \varphi_0} + \dots \quad (2.7)$$

or, confining ourselves to the first two terms of the expansion,

$$\varphi = \varphi_0 + \theta(\tau - \tau_1). \quad (2.8)$$

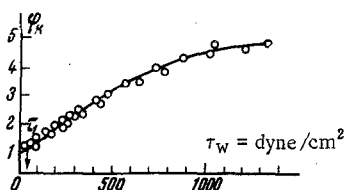


Fig. 3. Solution of 1.69% rubber in toluene,  $t = 24^\circ \text{C}$ ,  $\varphi_0 = 0.7 \text{P}^{-1}$ ,  $\varphi_\infty = 5.1 \text{P}^{-1}$ .

Equation (2.8) is very convenient for constructing approximate methods of calculating the flow of fluids with structural viscosity. Therefore, in the theory, it is desirable to distinguish a special subclass of fluids with a linear fluidity law. For a dilatant fluid a minus

sign should be inserted in front of the second term of (2.8).

3. For a laminar isothermal fluid flow in a circular cylindrical channel the tangential stress distribution has the form

$$\tau = \tau_w \xi, \quad \xi = r/r_0. \quad (3.1)$$

Here  $\xi$  is the dimensionless radius,  $\tau_w$  is the value at the wall. When  $\tau_0 = 0$  and  $\tau < \tau_1$  as a result of integration of the equation

$$\frac{dw}{dr} = -\varphi(\tau - \tau_0) \quad \left(\tau = -\frac{r}{2} \frac{dP}{dx}\right), \quad (3.2)$$

we get the velocity profile

$$w = 1/2 \tau_w \varphi_0 r_0 (1 - r^2/r_0^2). \quad (3.3)$$

At  $\tau_1 = 0$  and with a linear law of fluidity  $\varphi = \varphi_0(1 + \beta\tau)$  we get

$$w = 1/2 \tau_w r_0 \varphi_0 [(1 - \xi^2) + 2/3 \beta \tau_w (1 - \xi^3)], \quad (3.4)$$

$$w_{\max} = 1/4 \tau_w r_0 \varphi_0 (1 + 2/3 \beta \tau_w) \quad \left(\beta = \frac{\theta}{\varphi_0}\right), \quad (3.5)$$

$$\langle w \rangle = 1/4 \tau_w r_0 \varphi_0 (1 + 4/5 \beta \tau_w) \quad (3.6)$$

$$\omega = \frac{w}{\langle w \rangle} = 2 \frac{1 - \xi^2 + 2/3 \beta \tau_w (1 - \xi^3)}{1 + 4/5 \beta \tau_w}, \quad (3.7)$$

$$\omega_{\max} = \frac{w_{\max}}{\langle w \rangle} = 2 \frac{1 + 2/3 \beta \tau_w}{1 + 4/5 \beta \tau_w}. \quad (3.8)$$

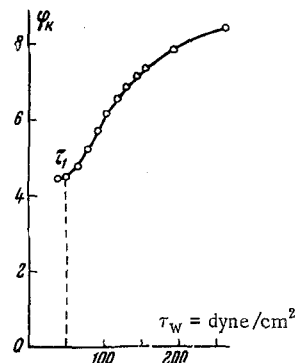


Fig. 4. Solution of sodium-carboxymethyl cellulose in water—1%,  $t = 20^\circ \text{C}$ ,  $\varphi_0 = 4.5 \text{P}^{-1}$ ,  $\varphi_{00} = 9 \text{P}^{-1}$ ,  $\tau_1 = 50 \text{ dyne/cm}^2$ .

Here  $w_{\max}$  is the maximum velocity at the tube axis, and  $\langle w \rangle$  is the mean flow rate.

From (3.8) it is clear that  $\omega_{\max} < 2$  when  $\beta > 0$ , i.e., the velocity profile is fuller than for the flow of ordinary Newtonian fluids ( $\beta = 0$ ); conversely,  $\omega_{\max} > 2$  when  $\beta < 0$ . Evidently, in the given case it is convenient to construct the Reynolds number of the flow in the form  $R_D = \langle w \rangle D \varphi_0 \rho$ .

Here  $\varphi_0$  is the zero fluidity ( $\tau \rightarrow 0$ ). Taking into account (3.6), we find that

$$\zeta = \frac{8\tau_w}{\rho \langle w \rangle^2} = \frac{5}{\beta} \left[ \left(1 + \frac{128\beta}{5R_D}\right)^{1/2} - 1 \right] \left( \beta = \frac{\theta}{\varphi_0} \rho \langle w \rangle^2 \right). \quad (3.9)$$

4. For longitudinal isothermal flow past a plate

\*One should also keep in mind that for the majority of fluids  $\tau_1 \approx 0$ .

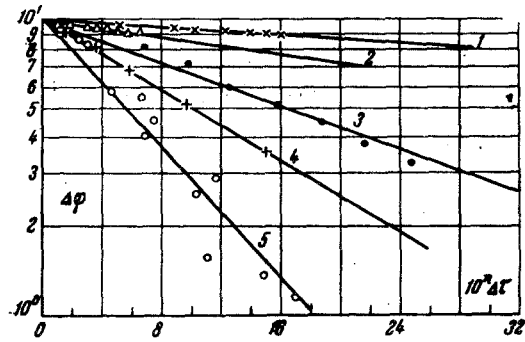


Fig. 5. 1) Polyvinyl alcohol in water 2.5%,  $n = -1$ ,  $\tau_1 = 0$ ,  $\tau_0 = 0$ ; 2) poly-methacrylate in water 0.025%,  $n = 1$ ,  $\tau_1 = 0$ ,  $\tau_0 = 0$ ; 3) carboxymethyl cellulose in water 1%,  $n = 0$ ,  $\tau_1 = 50$  dyne/cm<sup>2</sup>,  $\tau_0 = 0$ ; 4) rubber in toluene 1.69%,  $n = -1$ ,  $\tau_1 = 25$  dyne/cm<sup>2</sup>,  $\tau_0 = 0$ ; 5) bitumen M III,  $n = -6$ ,  $\tau_0 = 12.5$  dyne/cm<sup>2</sup>.

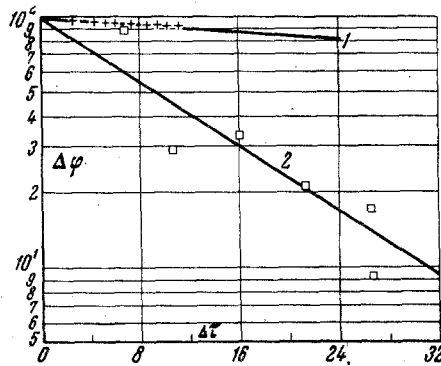


Fig. 6. 1) Carboxymethyl cellulose in water 0.25%,  $n = 1$ ,  $\tau_1 = 0$ ,  $\tau_0 = 0$ ; 2) starch in glycol 47.4%,  $n = 2$ ,  $\tau_1 = 0$ ,  $\tau_0 = 0$ .

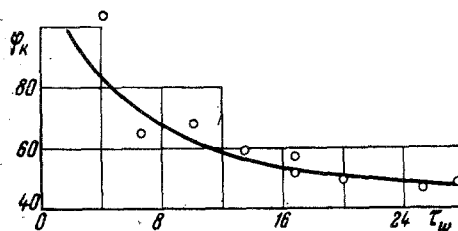


Fig. 7. Starch in glycol 47.4%,  $t = 25.5^\circ \text{C}$ ,  $\tau_0 = 111 \text{ P}^{-1}$ ,  $\varphi_{00} = 46 \text{ P}^{-1}$ .

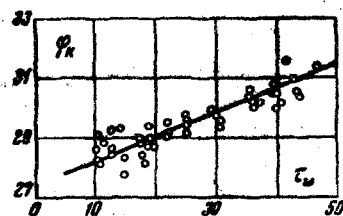


Fig. 8. Carboxymethyl cellulose in water 0.25%,  $t = 45^{\circ}-48^{\circ} \text{C}$ ,  
 $\varphi_0 = 27.5 \text{ p}^{-1}$ ,  $\varphi_{\infty} = 71 \text{ p}^{-1}$ .

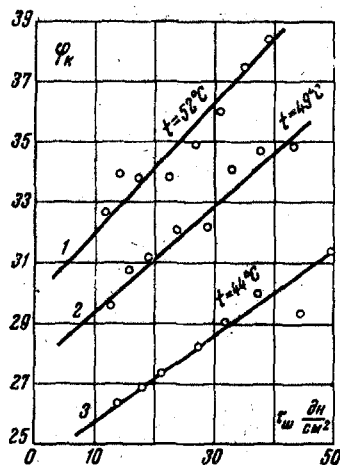


Fig. 9. Polymethacrylate in water 0.25%, 1— $\varphi_0 = 29.5 \text{ p}^{-1}$ ,  $\varphi_{\infty} = 108 \text{ p}^{-1}$ ;  
 2— $\varphi_0 = 27.5 \text{ p}^{-1}$ ,  $\varphi_{\infty} = 105 \text{ p}^{-1}$ ; 3—  
 $\varphi_0 = 24 \text{ p}^{-1}$ ,  $\varphi_{\infty} = 100 \text{ p}^{-1}$ .

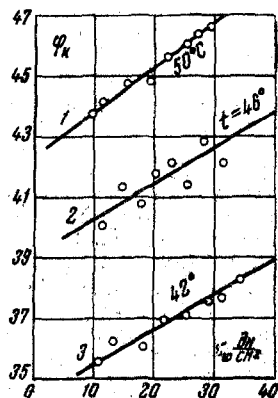


Fig. 10. Polyvinyl alcohol in water 2.5%, 1— $\varphi_0 = 42 \text{ p}^{-1}$ ,  
 $\varphi_{\infty} = 70 \text{ p}^{-1}$ ; 2— $\varphi_0 = 39 \text{ p}^{-1}$ ;  
 $\varphi_{\infty} = 64.5 \text{ p}^{-1}$ ; 3— $\varphi_0 = 34.5 \text{ p}^{-1}$ ,  
 $\varphi_{\infty} = 56.5 \text{ p}^{-1}$ .

the tangential stress profile can be represented in the form [5]

$$\tau \approx \tau_w (1 - 3\xi^2 + 2\xi^3) \quad (\xi = y/\delta). \quad (4.1)$$

Here  $\xi$  is the dimensionless distance along the normal from the wall,  $\delta$  is the thickness of the boundary layer. For a linear law of fluidity the distribution of the longitudinal component of the velocity vector is given by

$$dw_x / dy = \varphi_0 \tau + \theta \tau^2. \quad (4.2)$$

Integrating (4.2) with allowance for (4.1), we get

$$w_x = \tau_w \varphi_0 \delta \left[ \xi - \xi^3 + \frac{1}{2} \xi^4 + \theta \tau_w \xi - 2\theta \tau_w \xi^3 + \theta \tau_w \xi^4 + \frac{\theta}{5} \tau_w \xi^5 - 2\theta \tau_w \xi^6 + \frac{4}{7} \theta \tau_w \xi^7 \right]. \quad (4.3)$$

When  $\xi = 1$ , we have  $w_x = w_0$ , where  $w_0$  is the velocity of the oncoming flow and, consequently,

$$w_0 = \frac{1}{2} \tau_w \varphi_0 \delta \left( 1 + \frac{2\theta}{35} \tau_w \right). \quad (4.4)$$

For the friction coefficient, using (4.4), we have

$$C_f = \frac{2\tau_w}{\rho w_0^2} = \frac{35}{2\theta\beta} \left[ \left( 1 + \frac{208\beta}{35R_\delta} \right)^{1/2} - 1 \right] R_\delta = w_0 \delta \varphi_0 \rho, \quad (4.5)$$

$$\beta = \frac{\theta}{\varphi_0} \rho w_0^2.$$

5. For the experimental determination of the rheological characteristics of a fluid we compute directly not the true law  $\varphi(\tau)$ , but the relation between the so-called apparent fluidity  $\varphi_k$  and the tangential shearing stress at the wall  $\tau_w$ . Thus, for example, in a capillary viscometer from the measured flow  $Q$  and the pressure drop  $\Delta P$  we find the quantities

$$\varphi_k = \frac{8LQ}{\pi r_0^4 \Delta P}, \quad \tau_w = \frac{r_0 \Delta P}{2L}, \quad (5.1)$$

where  $r$  and  $L$  are the radius and length of the capillary. In the region  $\tau < \tau_1$  we have  $\varphi = \varphi_0$ , and in the region  $\tau > \tau_1$  the function  $\varphi(\tau) \neq \varphi_k(\tau)$ . From (5.1) there follows

$$\langle w \rangle = \frac{1}{4} \varphi_k \tau_w r_0. \quad (5.2)$$

Comparing (5.2) with (3.6), we have

$$\theta_k = 0.80. \quad (5.3)$$

6. We shall make a comparison of the results obtained with experiment data. Above it was shown that the character of the functions  $\varphi(\tau)$  and  $\varphi_k(\tau)$  is analogous, while the quantity  $\vartheta$  can be determined from  $\vartheta_k$  in accordance with (5.3); therefore the rheological equation (2.4) can be checked from the data of viscometric measurements, i.e., in the form  $\varphi_k(\tau_w)$ .

Figures 2-4 give experimental data for bitumen according to Mikhailov [6], for a solution of rubber in toluene according to Reiner [2], and also for a 1% aqueous solution of carboxymethyl cellulose according to our measurements. In the first case the yield point is distinctly observed, and in the other two cases the limit of structural stability. In Figs. 5 and 6 the same data and data on a series of other fluids are presented in semilogarithmic anamorphosis, where

$$\Delta\varphi = \frac{\varphi_\infty - \varphi_k}{\varphi_\infty - \varphi_0}, \quad \Delta\tau = \frac{\tau - \tau_1}{\varphi_\infty - \varphi_0}.$$

It should be noted that in Figs. 6 and 7 the solution of starch in glycol is [7] a fluid of dilatant type;  $\Delta\varphi = 1$  when  $\Delta\tau = 0$ .

Clearly, within the limits of accuracy of the experiments and the estimates of  $\varphi_0$  and  $\varphi_\infty$ , the rheological law (2.4) is quite well expressed.

Considering Figs. 5 and 6, it is possible to conclude that the scatter of the experimental points about the straight lines, especially in the region of large  $\Delta\tau$ , is quite large. However, it should be kept in mind that with increase in  $\tau$  a small error in the experimental determination of  $\varphi_k$  corresponds to a considerable error in the quantity  $\Delta\varphi$  represented in the graphs. In a number of cases the accuracy of the experimental data is hard to estimate; however, the fluidity curves calculated by us from Eq. (2.4) with values of  $\vartheta$  obtained from Figs. 5 and 6 ( $\vartheta$  is the slope of the straight line) deviated from the experimental curves shown in Figs. 2-4 and 7 by not more than 5%.

Figures 8-10 show experimental data [8] that confirm the existence of fluids with a linear fluidity law in the interval of  $\tau$  of practical interest.

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