

FLOW OF A POLYMER SOLUTION AS THE MOTION  
OF A MEDIUM WITH ADDITIONAL INTERNAL  
DEGREES OF FREEDOM

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The method for calculating the flow of dilute polymer solutions near walls is derived on the basis of an analysis of the interaction of elastic polymer particles with a turbulent newtonian solvent.

If foreign (e.g., polymer) particles are suspended in a newtonian liquid, and if the moment of inertia of these particles is negligible, the physical properties of the resulting two-phase medium, treated as a continuum, differ from those of the original newtonian liquid only by the Einstein correction to the viscosity [1].

If the polymer particles have a significant moment of inertia, on the other hand, the two-phase medium has more complicated properties, due to the appearance of additional internal degrees of freedom in rotational motion [2, 3]. This freedom corresponds to a continuous exchange of angular momentum between the polymer particles and the surrounding newtonian liquid; the rate of these relaxation processes depends on the inertia of the polymer particles. As a reaction to the change in the internal moment of such a medium, antisymmetric stresses arise [2-4]; such stresses are of course not involved in classical models of continuous media. The relaxation processes which occur in polymer solutions are accompanied by a dissipation of part of the kinetic energy of the flow, and this dissipation can be thought of as the work associated with antisymmetric stresses [4].

On the basis of these arguments we can write the equation for the average turbulent motion of polymer solutions for a constant boundary-layer stress as

$$\frac{dU_1}{dx_2} (\nu_0 + \varepsilon + \nu_E + \nu_{ra}) = u_*^2, \quad (1)$$

$$\varepsilon = - \frac{\tau_*}{u_1 u_2} \left( \frac{dU_1}{dx_2} \right)^{-1}; \quad \nu_E = \frac{D_E}{\rho} \left( \frac{dU_1}{dx_2} \right)^{-2}; \quad \nu_{ra} = \frac{D_{ra}}{\rho} \left( \frac{dU_1}{dx_2} \right)^{-2}. \quad (2)$$

The total relaxational dissipation can be thought of as the sum of the relaxational expenditures of the average and turbulent motion,  $D_R = D_{ra} + D_{rt}$ . Although the turbulent relaxational viscosity  $\nu_{rt}$  corresponding to relaxational dissipation  $D_{rt}$  does not appear explicitly among the parameters in (2) which govern the motion in (1), it must be involved in the problem, since it governs the vortical viscosity, given by

$$\frac{\varepsilon}{\nu_0} \approx \frac{x_2 u_*}{\nu_0} \left\{ 1 - \exp \left[ - \frac{\theta}{\alpha} \left( \frac{\nu_w}{\nu_t} \right)^{3.2} \frac{(x_2 u_* / \nu_0)^2}{(\nu_c / \nu_0)^2} \right] \right\}. \quad (3)$$

Equation (3) matches the laws governing the behavior in the immediate vicinity of the wall ( $\varepsilon \lesssim \nu_w$ ) and in the turbulent zone ( $\varepsilon \gg \nu_w$ ). These laws are obtained through a dimensional analysis, through the use of the continuity condition and the boundary conditions at the wall, and through an analysis of a simplified energy balance for the Kline eddy structures near the walls, which govern the rate at which turbulent energy is generated [5].

To determine the dissipation rates  $D_E$ ,  $D_{ra}$ , and  $D_{rt}$  and the corresponding functions  $\nu_E$ ,  $\nu_{ra}$ , and  $\nu_{rt}$ , we analyzed the interaction of polymer particles with a newtonian solvent undergoing turbulent

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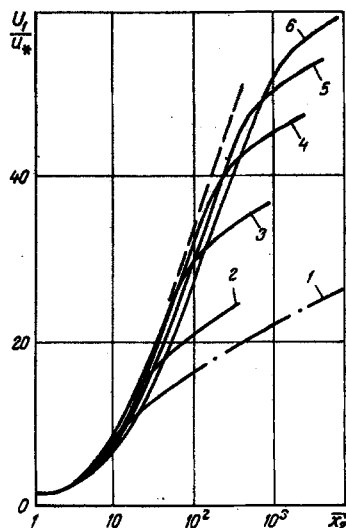


Fig. 1. Average-velocity profile as a function of the Reynolds number. 1)  $\log R_d = 3.66$ ; 2) 4.19; 3) 4.88; 4) 5.37; 5) 6.63; 6) 5.86.

fluctuations in a flow with a shear average velocity. By "polymer particle" we mean either a macromolecule or a supermolecular formation (association). We do not place any limitations on the dimensions of these particles, and we assume them to be ellipsoidal, like chain macromolecules, with an arbitrary ratio  $l$  of the axes of the ellipsoid.

The Einstein viscosity  $\nu_E$  is calculated from the known [6] functions of the relative shear  $\sigma_0 = (dU_1/dx_2)W/k_B T$  for laminar flow. In this generalization to the case of turbulent flow we take into account, along with the disorientation of the particles resulting from their thermal (Brownian) bombardment by solvent molecules, the disorientation because of turbulent fluctuations.

We expressed the inertial retardation of the particles during their rotation in the flow in terms of the rotational relaxation time  $\tau_R = (J + j)/W$ . By determining the relaxational-dissipation rate, which is known to be proportional to the mean square difference between the angular velocities of the particles and the surrounding solvent, we find the following equations for the relaxational viscosities:

$$\frac{\nu_{ra}}{\nu_0} = \frac{cN_A h^3}{M} \cdot \frac{l-1}{8(l^2+1)} f(l) \left[ 1 + \frac{1}{1+g^2\tau_R^2} - \frac{2}{(1+g^2\tau_R^2)^{2/3}} \right]; \quad (4)$$

$$\begin{aligned} \frac{\nu_{rt}}{\nu_0} = \frac{cN_A h^3}{M} f(l) & \left[ \frac{2}{3} k_R^4 + \frac{4}{3} k_R^3 + 2k_R^2 + 2k_R + 1 \right] \\ & \times \exp \left[ -2(k_R + \omega_R) \right], \end{aligned} \quad (5)$$

where  $M$ , in the case of an association, is to be understood as the factor by which the molecular weight of the association is greater than that of the polymer, or the factor by which the volume of the association is greater than that of the macromolecule. It follows from (4) that the energy of the average motion is dissipated in a relaxational manner among nonspherical particles ( $l \neq 1$ ) alone.

The parameters  $\nu_E$ ,  $\nu_{ra}$ , and  $\nu_{rt}$  are affected by the size  $h$  and shape  $l$  of the polymer particles, which can be deformed as a result of the dynamic effects of the flow. By analyzing these effects and specifying the elastic forces in the polymer particle after Frenkel' [4], we find the following differential equation for the deformation of the particle in a shear flow:

$$\frac{dh}{d\varphi} = \pi P(\varphi) \left[ \sin 2\varphi - \frac{32\sqrt{2}}{3\pi\zeta} \cdot \frac{k_B T}{g\rho\nu_0 h_0^3} \left( \frac{h_0}{h} - \frac{h_0^3}{h} \right) \right]. \quad (7)$$

We cannot use the familiar Peterlin equation [6] for  $P(\varphi)$  here, since it was derived on the basis of assumptions of laminar flow, a small relative shear  $\sigma_0$ , and inertialess particles — assumptions which do not hold for our problem. When these assumptions are not used in solving the Fokker-Planck diffusion equation, which the function  $P(\varphi)$  obeys, the following expression is found [7]:

$$P(\varphi) = \pi^{-1} [P_I(\varphi) + \Phi(\sigma, B) P_{II}(\varphi)] [1 + \Phi(\sigma, B)]^{-1}, \quad (8)$$

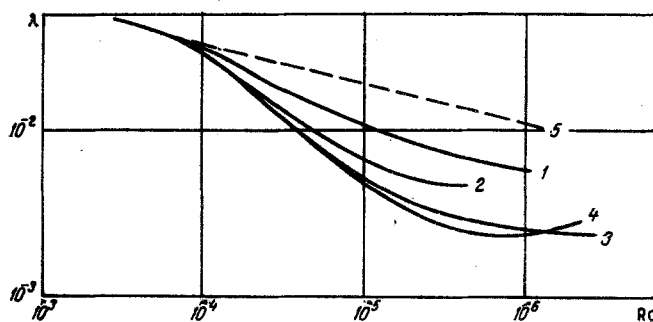


Fig. 2. Frictional drag for a tube. 1)  $c = 10^{-6}$  g/cm<sup>3</sup>; 2)  $5 \cdot 10^{-6}$ ; 3)  $10^{-5}$ ; 4)  $10^{-4}$ ; 5) water ( $c = 0$ ).

where

$$\Phi = \frac{1+B}{4} \frac{1}{\sqrt{1-B^2}} \left( \frac{\sigma B}{4} \right)^2; \quad P_I = 1 + \frac{\sigma B}{4} \sin(2\varphi + \psi); \quad (9)$$

$$P_{II} = \frac{\sqrt{1-B^2}}{1-B \cos(2\varphi + \psi)},$$

$$B = [(l^2 - 1)/(l^2 + 1)] \cos \psi; \quad \operatorname{tg} \psi = g\tau_R \quad (10)$$

and  $\sigma$  is the generalized relative shear, which takes into account both Brownian and turbulent disorientation. It turns out that the turbulent disorientation is more important than the Brownian disorientation for the type of flow under consideration here. As a result, in a shear turbulent flow the active wall turbulence produces a predominant orientation of the polymer particles such that these particles are subjected to tensile stresses from the flow for a longer period of time than that over which they are subjected to compressional stresses.

The rest of the calculation can be outlined as follows: through a numerical integration of Eq. (7) using (8) and a subsequent averaging, we determine the average statistical lengths and shapes of the polymer particles at various points  $\bar{x}_2 = x_{2u*}/\nu_0$  in the flow at which the values of the shear are  $g = dU_1/dx_2$  and we determine the turbulence levels in a first approximation as if there were no polymer additives in the flow. Then, for the same points  $\bar{x}_2$ , we determine the parameters  $\nu_E$ ,  $\nu_{ra}$ , and  $\nu_{rt}$ , which we use to calculate the vortical viscosity in (3); then a numerical integration of Eq. (1) leads to a profile of the average velocity in the second approximation. This new state of the turbulent flow is used to analyze the behavior of the polymer particles in it, and the entire calculation is repeated until the desired steady state is reached. A computer is quite suitable for this calculation, and an ALGOL program has been written for this problem. Some calculated results are shown in Figs. 1-3.

Figure 1 shows the evolution of the average-velocity profile as a function of the Reynolds number  $R_d$  for the flow of a WSR-301 polyox solution at a concentration of  $c = 10^{-5}$  g/cm<sup>3</sup> in a tube 35 mm in diameter. It is easy to see that with  $\log R_d = 3.66$  the velocity profile in the solution is the same as that in the pure solvent; i. e., this type of flow is "subthreshold." At high Reynolds numbers the profiles approach the saturation asymptote (the dashed line) which has been observed in many experiments.

Figure 2 shows the dependence of the frictional drag on the Reynolds number for various concentrations  $c$  of the WSR-301 polyox solution for a tube of the same diameter.

The results of these and other calculations, whose results are similar to those shown in Figs. 1 and 2, agree well with the available experimental data, correctly predicting the most important behavior: the nature of the effect of the solution concentration and the Reynolds number on the shape of the average-velocity profile and the frictional drag (and the absence of such an effect at Reynolds numbers below threshold); the dependence of the threshold Reynolds numbers on the tube diameter; and the saturation in the effect of the polymer at certain concentrations and certain Reynolds numbers. The calculated results agree well with the experimental data for the following initial dimensions  $h_0$  and shapes  $l_0$  of the polymer particles:  $l_0 \approx 5-14$  and  $h_0 = 1100-1700 \mu$  for WSR-301 polyox;  $l_0 \approx 1.5-4.0$  and  $h_0 = 870-980 \mu$  for guar resin. These dimensions are about two-thirds the values observed experimentally by Kalashnikov and Kudin [8], but both are much larger than the macromolecular values, so that the hydrodynamic effects of polymer additives observed in these experiments are apparently due to associations.

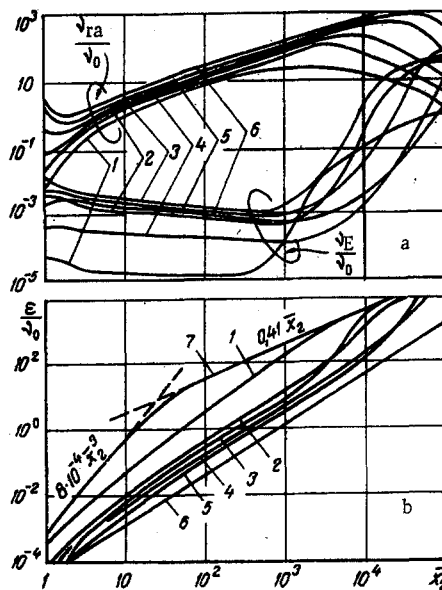


Fig. 3. Einstein and average relaxational viscosity (a) and Boussinesq eddy viscosity (b) as functions of the distance to the wall, in dimensionless units. 1)  $c = 10^{-6}$  g/cm<sup>3</sup> and  $\log R_x = 8.36$ ; 2)  $10^{-5}$  and 8.78; 3)  $5 \cdot 10^{-5}$  and 8.40; 4)  $10^{-5}$  and 8.54; 5)  $5 \cdot 10^{-5}$  and 8.80; 6)  $5 \cdot 10^{-5}$  and 8.88; 7) water.

Figure 3a shows the distribution of the Einstein and average relaxational viscosities over the thickness of the boundary layer, and Fig. 3b shows the Boussinesq eddy viscosity, for various concentrations of the WSR-301 polyox and for various Reynolds numbers. We see that for the dilute solutions the Einstein viscosity has essentially no effect on the flow, since it is much smaller than the molecular, the average relaxational, and the eddy viscosities in any part of the flow. On the other hand, the average relaxational viscosity dominates nearly everywhere, except in the zone of pronounced turbulence, where it is less important than the eddy viscosity. This eddy viscosity is so weak in polymer solutions (Fig. 3b) that the decrease in the associated Reynolds stresses dominates over the additional relaxational stresses which arise in the average flow, which are ultimately responsible for the reduction of the frictional drag.

#### NOTATION

$U_1$	is the component of the average velocity along the wall;
$x_2$	is the distance to the wall;
$g = dU_1/dx_2$ ;	
$\nu_0$	is the viscosity of the solvent;
$\rho$	is the density of the solution;
$u_*$	is the dynamic velocity;
$\overline{u_1 u_2}$ and $\epsilon$	are the fluctuation correlation and Boussinesq eddy viscosity;
$D_E, D_{ra}, D_{rt}$	are the Einstein, average relaxational, and turbulent relaxational dissipations;
$\nu_E, \nu_{ra}, \nu_{rt}$ ;	
$\nu_w = \nu_0 + \nu_E + \nu_{ra}$ ;	
$\nu_E = \nu_0 + \nu_{rt}$	are the corresponding effective viscosities;
$\kappa = 0.41$	is the von Karman constant;
$b = 8 \cdot 10^{-4}$	is a constant;
$W$	is the coefficient of rotational friction between the particle and the solvent;
$k_B$	is the Boltzmann constant;
$T$	is the absolute temperature;
$N_A$	is Avogadro's number;
$J, j$	are the intrinsic and additional moments of inertia of the particle;
$h_0, h$	are the unperturbed and deformed lengths of the particle;
$f(l)$	is the function of the shape of the ellipsoid;
$\varphi$	is the angle specifying the particle orientation;
$P(\varphi)$	is the probability for this orientation;
$\xi$	is the viscous-friction coefficient for flow around the particle;
$\sigma = \sigma_0(1 + \pi \rho \epsilon g h^3 / 12 l^2 k_B T)^{-1}$	is the generalized relative shear of the average velocity;

$R_d$  is the Reynolds number along a tube diameter;  
 $R_x$  is the Reynolds number along the longitudinal coordinate on a plate;  
 $\lambda$  is the friction coefficient in the tube;  
 $k_R = 3.2(\nu_w/\nu_0)W(\nu_0/hu_*)$ ;  $\omega_R = 0.8(\nu_w/\nu_0)W(\nu_0/u_*^2 \tau_R)$ .

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