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The rheological properties of suspensions of fibres in Newtonian fluids subjected to extensional deformations

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Data are presented for the extensional flow of suspensions containing 0·1–1·0 % of fibres by volume. The aspect ratio of the fibres was varied from 280 to 1260. The observed stress levels were between one and two orders of magnitude greater than in shearing flow, in agreement with the *a priori* predictions of Batchelor (1971).

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1. Introductory comments and review of theory

Suspensions of non-spherical particles in Newtonian liquids exhibit an interesting variety of rheological phenomena, including finite normal stresses in steady laminar shearing flows and a viscosity function which decreases with increasing shear rate (Giesekus 1962; Jeffery 1922; Nawab & Mason 1958; Scheraga 1955). The rheological properties of suspensions in extensional flows are also of significance in a number of areas, including the spinning of multiphase synthetic textile fibres and, possibly, turbulent drag-reduction processes. Recent analyses by Batchelor (1970, 1971) provide explicit predictions of these properties for suspensions of long slender particles in well-developed extensional flows. These predictions are not only applicable to macroscopic suspensions but may also be employed for modelling the properties of macromolecular systems. Such polymeric fluids exhibit extraordinarily high resistances to extensional deformations (Astarita & Nicodemo 1970; Kanel 1972; Spearot & Metzner 1972).

For the uniaxial extension of a cylindrical rod under steady-state and isochoric conditions, the components of the rate-of-strain tensor are, in Cartesian co-ordinates,

$$\begin{pmatrix} d_{11} & 0 & 0 \\ 0 & -\tfrac{1}{2}d_{11} & 0 \\ 0 & 0 & -\tfrac{1}{2}d_{11} \end{pmatrix},$$

in which the stretch direction is that of the x_1 axis.

If the components of the total stress tensor are denoted as S_{ij} we may define a deformation resistance coefficient (an apparent 'extensional viscosity') as

$$\lambda = \frac{S_{11}-\frac{1}{2}(S_{22}+S_{33})}{d_{11}}.$$

For long slender particles of appreciable size the Brownian disorientation of the aligned particles or fibres will be very small (see, for example, Takserman-Krozer & Ziabicki 1963*a*, *b*). Consequently, perfect alignment in the stretch direction may be assumed when a steady state has been attained. If the suspensions are sufficiently dilute so that the velocity field surrounding one particle is unaffected by neighbouring particles one obtains from the Batchelor (1970) analysis, for the case of long circular cylinders,

$$\lambda = \mu\left[3+\frac{2}{3}\frac{c\,(L/D)^2}{\ln(2L/D)}f\left(\frac{1}{\ln(2L/D)}\right)\right]. \tag{1}$$

In this equation μ denotes the viscosity of the continuous phase and c the volumetric concentration (volume fraction) of the particles having length L and diameter D. The function f is given by

$$f(\alpha) = \frac{1+0{\cdot}640\alpha}{1-\frac{3}{2}\alpha}+1{\cdot}659\alpha^2+O(\alpha^3). \tag{1a}$$

The restriction of sufficient diluteness requires

$$n(\tfrac{1}{2}L)^3/\ln(2L/D) \ll 1, \tag{1b}$$

in which n denotes the number concentration of particles.

The second term on the right-hand side of (1) expresses the change due to the presence of the fibres. As $f(\alpha)$ is of order unity, (1) suggests that large values of the deformation resistance coefficient λ could be achieved at small values of the particle concentration c in the case of particles of large aspect ratio. However, the restriction on the concentration levels under which (1) is valid (equation (1*b*)) is such as to permit only modest perturbations of the stresses required to deform the suspending liquid.

In a second paper Batchelor (1971) has considered more concentrated suspensions having interacting velocity and stress fields. The governing equations were solved subject to two restrictions. First, the geometry of the particles and their concentration levels must satisfy the inequality

$$L \gg H \gg D,$$

in which H is the average distance between fibres. Second, the effect on one particle of all the others is replaced by a cylindrical boundary condition, i.e. a cell model is employed. The resulting expression for the constant apparent extensional viscosity is

$$\lambda = \mu\left[3+\frac{4}{3}\frac{c\,(L/D)^2}{\ln(\pi/c)}\right]. \tag{2}$$

Very high values of the ratio λ/μ may be predicted within the range of validity of this theory. It does not reduce to the dilute suspension theory at lower concentrations; however, the two theories predict an identical result at an intermediate concentration level and one may interpolate between (1) and (2) in this intermediate region, as shown in figure 3 of Batchelor (1971).

2. Design of experiments

A device similar to that employed in spinning textile fibres was used; it was patterned after an earlier model built by Weinberger (1970). A spinneret connected to the fluid reservoir was mounted on a torsion bar whose rotation could be measured precisely by means of a displacement transducer. The suspension leaving the spinneret as a vertical column or jet was taken up, and stretched, by a rotating cylinder. The strain rates within the column were computed from the known flow rate, the geometry of the column as measured photographically and the assumption of a flat velocity profile. This latter assumption has been treated in detail by Baid (1973), who showed it to be excellent except in the region within a few column diameters of the spinneret, a region excluded from consideration in any event, as will be noted later. The experimental apparatus is also described in detail by Baid.

Glass fibres of constant length were available as chopped strands supplied by Owens–Corning. The individual fibres were separated by washing away the adhesive between them in concentrated hydrochloric acid. The single fibres were rinsed repeatedly in water and dried in an oven at 100° C. A low molecular weight polybutene (Indopol H-100, from Amoco) constituted the continuous phase. Its density was 0·89 g/cm^3, its surface tension 34·3 dyne/cm and its viscosity 283 P at 24·0 °C.

The desired quantities of the two components were mixed at about 35° C. The temperature was kept above ambient to avoid excessive dispersion of air in the suspension and to facilitate de-aeration; mixing speeds were kept low to minimize fibre fracture. The suspensions showed a Weissenberg effect which increased with fibre concentration and with fibre length. Additionally, as noted by Nawab & Mason (1958), long fibres are subject to appreciable centripetal forces in Couette flows. This creates a demixing which interferes with particle dispersion, especially for particles of high aspect ratio and for high fibre concentrations. These complications were minimized by use of small sample volumes and by restricting the study to suspensions of modest concentration. These several difficulties severely restricted the conditions under which measurements could be made; the suspensions which actually could be used are described later (table 1) where the experimental results are also given.

3. Analysis of data

A differential axial balance of linear momentum gives

$$\delta\{R^2[S_{11}-\tfrac{1}{2}(S_{22}+S_{33})]\} = (Q_M/\pi)\,\delta V-\sigma\,\delta R-\rho g\,R^2\,\delta z, \qquad (3)$$

in which V denotes the axial fluid velocity, R the column radius, Q_M the mass flow rate of the suspension, σ the surface tension, ρ the suspension density, g the acceleration due to gravity and z the length along the column. In deriving (3) the longitudinal curvature of the liquid column is neglected in comparison with the radius R. This is properly negligible except near the spinneret, where well-developed flow conditions with aligned fibres may also not be obtained. Therefore only the lower end of the liquid column is used for evaluation of (2). Equation (3) was integrated from the spinneret (on which the forces were measured) to any position z along the column, the last term being evaluated numerically.

The experimental measurements and the data-reduction method were checked using a Newtonian polybutene oil having a viscosity of 240 P as determined from independent rheogoniometric measurements. The predicted behaviour in extension was observed with a standard deviation of less than 10 % over the full range of conditions studied: two spinneret diameters, 10 runs and deformation rates between 1·6 and 5 s^{-1}. Since the stress levels of the Newtonian oil are lower than those encountered using the suspension (by a factor varying between 2 and 40) this is a sensitive and useful evaluation of the accuracy of the measurements.

The viscosity of Newtonian oils is very sensitive to small temperature variations. Therefore the ratio λ/μ (given in (2)), which is nearly independent of temperature, was computed and used to normalize differences from run to run as perturbations in room temperature occurred.

The assumption that the column is a homogeneous continuum requires the presence of a large number of fibres at any cross-section. At the lowest concentration level and the highest stretch rates used in this work 90 fibres were present at the lower end of the column and 280 at the upper. Superficially, these numbers appear to be large but it is not clear that they are large enough, as is shown by the following considerations. All concentration levels studied were such that there was strong hydrodynamic interaction between the fibres. Under such conditions those fibres which are closer to the outer surface than the average fibre-to-fibre distance will experience hydrodynamic forces which differ from those on a fibre in the interior of the suspension. The interfibre distance H may be approximated by

$$H \approx D/\sqrt{c}.$$

For a suspension containing 1 % of fibres by volume one finds the 'fibre free' surface region to occupy between 7 and 13 % of the column cross-section. At a 0·1 % concentration level the corresponding values are 21 and 36 %. While these numbers appear to be unacceptably large a more accurate estimate of the effect of the free surface is obtained if its contribution to the total force on a particle is calculated. Indeed, even those fibres closest to the outer surface still have approximately half of their sphere of interaction with other fibres intact. In addition the stresses due to the presence of other particles decay rapidly with distance. Thus, the effects of the free surface will be much smaller than is indicated by geometric arguments alone. However, their influence, if any, and problems arising out of the unbalanced stress fields to which the particles nearest the surface are subjected, will need to be evaluated experimentally.

One additional experimental difficulty should be mentioned. In viscoelastic fluids spinning rates may be limited severely by the phenomenon of 'draw

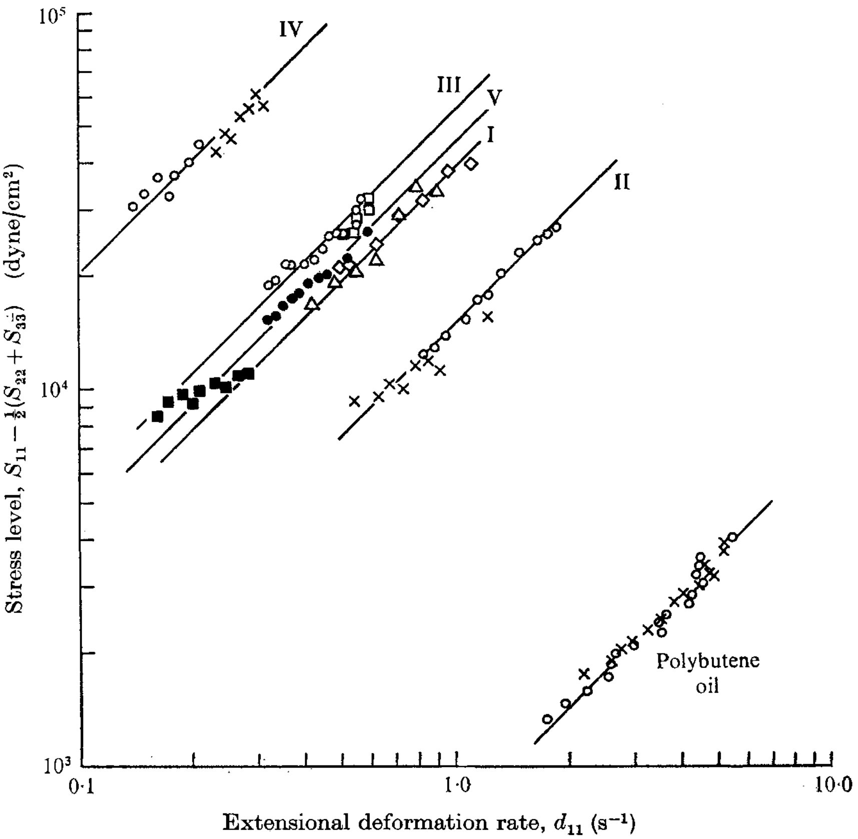

FIGURE 1. Stress–deformation-rate relationships for fibre suspensions. Concentration levels and length: diameter ratios of fibres are given in table 1. △, ◇, sample I; ×, ○, sample II; ○, □, sample III; ○, ×, sample IV; ●, ■, sample V.

resonance': oscillations of low frequency but large amplitude which develop as the stretch rate of the fluid is increased (Miller 1963; Bergonzoni & Dicresce 1966; Zeichner 1973). Evidently suspensions are subject to similar instabilities. This was most serious for the longest fibres and the highest concentrations, i.e. at the largest stress levels. In some cases these stress (and diameter) oscillations could not be eliminated entirely. To reduce their effect three independent runs were made under each set of conditions and two filament photographs were taken of each run. The radius–distance curves used were a smoothed average of the six determinations. Nevertheless, the accuracy obtained is less than that obtainable with Newtonian fluids at comparable stress levels.

4. Results

Figure 1 shows, for each sample, the results of two independent sets of runs carried out at stretch rates varying as widely as possible in view of the restrictions discussed above. Within the limits of the experimental error the viscosity is independent of stretch rate, or equivalently, of position along the jet, with the

Fluid system	L (mm)	L/D	c	H/D	λ_s	$\lambda_s/100c$
I	3·10	282	0·00930	10·4	51	55
II	6·375	586	0·00099	31·8	17·5	180
III	6·375	586	0·00287	18·7	74	260
IV	6·375	586	0·00890	10·6	260	290
V	12·065	1259	0·00096	32·3	59	620
W†	0·2	57	0·013	8·8	—	—

† Suspension used by Weinberger (1970); included for purposes of comparison and completeness.

TABLE 1. Effect of fibres on resistance to extensional deformations (experimental).

possible exception of one set of runs for system V. This indicates, first, that no substantial change of orientation takes place in the region of nearly pure extensional straining. The residence time in the fluid column was usually close to $1/d_{11}$, which is also the approximate time interval required for substantially complete parallel alignment of the fibres. Since the shearing flow in the connecting tubing and the converging flow in the spinneret will already have pre-oriented the fibres significantly such constancy of alignment is to be expected. Second, the free-surface effects discussed previously appear to be absent at the level of discrimination possible with these data.

Although the particle concentrations were always less than 1 % the apparent extensional viscosities are much larger than that of the continuous phase and in this respect the important qualitative predictions of the Batchelor analysis are supported. The exact increase in the resistance to deformation arising from the presence of the fibres may be ascertained from a specific extensional viscosity λ_s defined as

$$\lambda_s = (\lambda - 3\mu)/3\mu.$$

Table 1, which also describes the systems studied, shows that the fibres increased this deformation resistance by a factor of between 17·5 and 260; the ratio $\lambda_s/100c$, which rises to 620, gives the magnitude of the increase which would occur in a suspension containing 1 % of fibres by volume assuming a linear extrapolation to be valid in all cases.

The theoretical value of λ_s is given by (2) as

$$(\lambda_s)_{\text{th}} = \frac{4}{9}\frac{c\,(L/D)^2}{\ln(\pi/c)}. \tag{4}$$

The shape of the particles enters this expression only through the square of the aspect ratio while the concentration effect is determined by the ratio $c/\ln(\pi/c)$. Thus the contributions of volume and shape can easily be separated and compared with the experimental results. Values of the quantity λ_s have been taken from the curves of figure 1 and a plot of $\lambda_s/(L/D)^2$ *vs*. concentration is shown in figure 2.

It may be seen that the variations with concentration are, within experimental accuracy, precisely those predicted by theory. With respect to changes in aspect

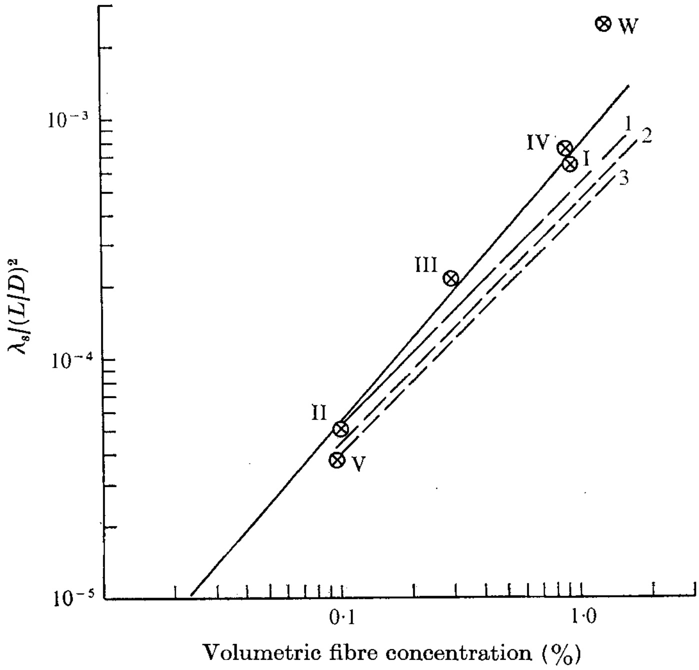

FIGURE 2. Comparison of the experimental and theoretical effects of fibre L/D and concentration level upon resistance to deformation. ——, theory for concentrated suspension; – – –, theory for dilute suspension; 1, $L/D = 282$; 2, $L/D = 586$; 3, $L/D = 1259$.

ratio the data are somewhat less consistent. The differences at the 1 % concentration level are within experimental error and a four-fold change in the square of the aspect ratio does not give rise to a measurable change in $\lambda_s/(L/D)^2$, as predicted by (2). For a similar change at a 0·1 % particle concentration, but with absolute particle lengths twice as great, a measurable decrease with increasing aspect ratio is found although it must be noted that the experimental accuracy also decreases with increasing fibre length. Figure 2 additionally depicts the curves for dilute suspensions (equation (1)); these show that an effect of L/D close to that observed at the 0·1 % concentration level is predicted with considerable precision by this dilute suspension theory. This must be fortuitous since the inequality (1*b*) is violated by several orders of magnitude. If a similar 'shift' is introduced into the analysis for closely spaced particles, but is based upon H/D instead of L/D, which is intuitively reasonable as noted by Batchelor (1971), the results fail to agree with theory by several orders of magnitude.

The only other experimental results available to the authors are those obtained by Weinberger (1970). The aspect ratio of his particles was much smaller than that of ours (table 1) and his sample concentration level may lie between those required for either (1) or (2) to be applicable. However, his result is included in figure 2 for completeness. As already noted by Batchelor (1971) the resulting value for the extensional viscosity is even higher than that predicted by the close-particle theory. Batchelor noted possible empirical modifications of the concentrated suspensions formula which would bring it into better agreement with

Weinberger's data point, but such formulae predict stresses 100–400 % above our measured values. A less sensitive effect of the aspect ratio could be developed empirically but the data are not extensive enough to give much quantitative information on the form of such a correction.

5. Conclusions

The experimental data show extensional viscosities which are independent of strain rate and as much as 260 times greater than that of the suspending fluid. Over the range of conditions studied the effects of particle concentration are precisely those predicted by the Batchelor analysis (equation (2)); the effect of particle geometry is in close agreement with theory for the particles having length/diameter ratios of 282 and 586; for the $L/D = 1259$ suspension the experimental results fall about 30 % lower than expected.

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