A theory of anisotropic fluids

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A theory is proposed in which the stress tensor is a function of the components of the rate of deformation tensor and a symmetric tensor describing the microscopic structure of a fluid. The expression for the stress tensor can be written in closed form using results from the Hamilton–Cayley theorem. This theory is shown to contain Prager's theory of dumbbell suspensions as a special case. By limiting the type of terms in the constitutive equations, various stress components can be evaluated for simple shear. These exhibit non-Newtonian behaviour typical of certain higher polymer solutions.

Some of the results of the anisotropic fluid theory are compared with experimental measurements of normal stress and apparent viscosity. Certain high polymers in solution show good agreement between theory and experiment, at least for low enough values of the rate of shear.

1. Introduction

Several fluid mechanical theories have been formulated which take into account the fluid's microscopic structure. Jeffery (1922), for example, calculated the fluid motion in the vicinity of a suspended ellipsoid and used the results to find the increase in viscosity due to the presence of the ellipsoidal particle in an otherwise Newtonian fluid. Prager (1957) considered a suspension of non-interacting dumbbell particles and found a constitutive equation for stress and equations determining the 'preferred' direction adopted by the particles.

We formulate here a theory of anisotropic fluids which is rather general in interpretation. We shall introduce a symmetric tensor a_{ij} that will describe the microscopic structure of a fluid and find the most general expression for the stress tensor as a function or the components of a_{ij} and rate of deformation d_{ij}. We shall consider the fluid so represented as being incompressible, and its properties independent of temperature. With these simplifications it will not be necessary to consider energy conservation explicitly, and the continuity equation reduces to $d_{ii} = 0$.

It will be shown that Prager's (1957) theory is contained in this theory of anisotropic fluids, at least for a special case. In later papers the author will show that Jeffery's (1922) theory and Ericksen's (1959, 1960) theory are also contained in this theory. The problem of simple shear will be considered in particular since

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it is the most tractable. In the last section, comparisons will be made with the results of some experimental measurements.

We use cartesian tensor and matrix notation throughout the paper. The Einstein summation convention is observed.

2. Formulation of the theory

In this section we formulate the theory of anisotropic fluids. We introduce a symmetric tensor a_{ij} general in interpretation which describes the microscopic structure of a fluid. We postulate that the components of the stress tensor t_{ij} are expressible as polynomials in the components of a_{ij} and the rate of deformation tensor d_{ij} which is defined as

$$d_{ij} = \tfrac{1}{2}(\dot{x}_{i,j} + \dot{x}_{j,i}), \tag{1}$$

where the dot denotes material derivative and $_{,j}$ is short for $\partial/\partial x_j$. We further assume that the stress is symmetric.

Rivlin (1955) has found the most general expression for a 3×3 symmetric matrix expressed as a polynomial in two other symmetric 3×3 matrices. He uses the Hamilton–Cayley theorem and identities derived from it to find $\mathbf{t} = \mathbf{t}(\mathbf{a}, \mathbf{d})$ in closed form. This general expression is

$$\begin{aligned}\mathbf{t} = \beta_0 \mathbf{I} + \beta_1 \mathbf{a} + \beta_2 \mathbf{d} + \beta_3 \mathbf{a}^2 + \beta_4 \mathbf{d}^2 + \beta_5(\mathbf{ad} + \mathbf{da}) \\ + \beta_6(\mathbf{a}^2\mathbf{d} + \mathbf{da}^2) + \beta_7(\mathbf{ad}^2 + \mathbf{d}^2\mathbf{a}) + \beta_8(\mathbf{a}^2\mathbf{d}^2 + \mathbf{d}^2\mathbf{a}^2),\end{aligned} \tag{2}$$

where $\mathbf{I}$ is the unit matrix. We consider that the isotropic pressure is contained in β_0. The β's are scalar polynomials in the elements of $\mathbf{a}$ and $\mathbf{d}$ and are invariant under orthogonal transformations; they are functions of a complete set of simultaneous invariants of $\mathbf{a}$ and $\mathbf{d}$, for example

$$\left.\begin{array}{c}\operatorname{tr}\mathbf{a}, \quad \operatorname{tr}\mathbf{a}^2, \quad \operatorname{tr}\mathbf{a}^3, \quad \operatorname{tr}\mathbf{d}, \quad \operatorname{tr}\mathbf{d}^2, \quad \operatorname{tr}\mathbf{d}^3, \\ \operatorname{tr}\mathbf{ad}, \quad \operatorname{tr}\mathbf{a}^2\mathbf{d}, \quad \operatorname{tr}\mathbf{ad}^2, \quad \operatorname{tr}\mathbf{a}^2\mathbf{d}^2,\end{array}\right\} \tag{3}$$

where $\operatorname{tr}\mathbf{a}$ means the trace of $\mathbf{a}$. We consider that $\mathbf{t}$ represents stress, $\mathbf{d}$ the rate of deformation tensor, and $\mathbf{a}$ the general tensor describing the structure of the fluid. Since $\mathbf{t}$, $\mathbf{d}$, and $\mathbf{a}$ are all symmetric, we have a constitutive equation for stress. Equation (2) is rather complicated, but we can simplify it by stipulating that the components of stress are linear in the components of the rate of deformation. This gives

$$\begin{aligned}\mathbf{t} = (\sigma_0 + \sigma_1 \operatorname{tr}\mathbf{ad} + \sigma_2 \operatorname{tr}\mathbf{a}^2\mathbf{d})\,\mathbf{I} + (\sigma_3 + \sigma_4 \operatorname{tr}\mathbf{ad} + \sigma_5 \operatorname{tr}\mathbf{a}^2\mathbf{d})\,\mathbf{a} \\ + \sigma_6 \mathbf{d} + \sigma_7(\mathbf{ad} + \mathbf{da}) + \sigma_8(\mathbf{a}^2\mathbf{d} + \mathbf{da}^2),\end{aligned} \tag{4}$$

where the σ's are functions of the invariants of $\mathbf{a}$ only. We have used the relation for incompressible fluids, $\operatorname{tr}\mathbf{d} = 0$.

We use this stress tensor in the standard form of the equations of motion,

$$\rho \ddot{x}_i = F_i + t_{ij,j}, \tag{5}$$

where ρ is density and F_i is the body force. This with the continuity equation provides us with four equations. We have ten unknowns, however, the $\dot{x}_i$, the a_{ij}, and the isotropic pressure. We must introduce six additional equations in order to have a complete set.

We assume a constitutive equation of the form

$$\dot{a}_{ij} = F_{ij}(a_{kl}, \dot{x}_{p,q}). \tag{6}$$

We are motivated in choosing this functional relationship by results obtained by other authors mentioned in the introduction who considered microscopic fluid structure. The reason for this form will become evident when a comparison is made in the next section.

Noll (1955) has shown the correct form equation (6) must take in order to be invariant under time-dependent orthogonal transformations. He found that the velocity gradients must be replaced by the rate-of-deformation tensor d_{ij} and $\dot{a}_{ij}$ must be replaced by $\hat{a}_{ij}$ defined by

$$\hat{a}_{ij} = \dot{a}_{ij} - \omega_{ik}a_{kj} + a_{ik}\omega_{kj}, \tag{7}$$

where the vorticity is

$$\omega_{ij} = \tfrac{1}{2}(\dot{x}_{i,j} - \dot{x}_{j,i}). \tag{8}$$

Using the results of Rivlin and the replacement theorem of Noll, we find the most general expression for $\dot{a}_{ij}$ is

$$\begin{aligned}\dot{\mathbf{a}} = \boldsymbol{\omega}\mathbf{a} - \mathbf{a}\boldsymbol{\omega} + \alpha_0\mathbf{I} + \alpha_1\mathbf{a} + \alpha_2\mathbf{d} + \alpha_3\mathbf{a}^2 + \alpha_4\mathbf{d}^2 + \alpha_5(\mathbf{ad} + \mathbf{da}) \\ + \alpha_6(\mathbf{a}^2\mathbf{d} + \mathbf{da}^2) + \alpha_7(\mathbf{ad}^2 + \mathbf{d}^2\mathbf{a}) + \alpha_8(\mathbf{a}^2\mathbf{d}^2 + \mathbf{d}^2\mathbf{a}^2),\end{aligned} \tag{9}$$

where the α's are functions of the invariants listed in expression (3). Equations (5), (9), and the continuity equation together with (2) provide the ten equations to determine a_{ij}, $\dot{x}_i$, and the isotropic pressure. We now have a complete set of equations to describe the mechanics of an anisotropic fluid.

3. Comparison with Prager's theory

Prager obtains a constitutive equation for stress for a suspension of rigid dumbbells in a fluid. He restricts the theory to a suspension so dilute that there is no hydrodynamic interaction between dumbbells or between the two ends of the same dumbbell. Prager introduces the angular distribution function W for the orientation of the dumbbells in the flow field, where W satisfies the rotational diffusion equation

$$D_r\frac{\partial^2 W}{\partial\rho_k\,\partial\rho_k} = \dot{W}, \tag{10}$$

the conditions that W is periodic in spherical co-ordinates and

$$\oint_s W\,ds = 1, \quad \oint_s \dot{W}\,ds = 0. \tag{11}$$

Here D_r is the rotational diffusion coefficient and s represents the surface of a unit sphere centred on the centre of the dumbbell. He lets ρ_i be a unit vector which lies along the radial direction of the dumbbell.

Prager considers the velocity of the end of the dumbbell relative to co-ordinate axes centred on the centre of the dumbbell. This can be represented by $\dot{\rho}_i L$ where L is the dumbbell half-length. Since $|\rho_i| = 1$, we note that $\rho_i\rho_i = 1$ and $\rho_i\dot{\rho}_i = 0$. The velocity $\dot{\rho}_i L$ equals the fluid velocity in the neighbourhood of the end of the dumbbell, minus the radial component of the fluid velocity, plus the diffusional velocity of the dumbbell. For this special case Prager does not consider translation of the dumbbell relative to the fluid.

The fluid velocity for homogeneous flow is given by

$$\dot{x}_i = \dot{x}_{i,j} x_j = (d_{ij} + \omega_{ij}) x_j. \tag{12}$$

The radial velocity v_{ri} of the fluid is found by Prager to be

$$v_{ri} = L d_{kl} \rho_k \rho_l \rho_i, \tag{13}$$

and the diffusional velocity is found by Kuhn & Kuhn (1945) to be

$$v_{Di} = -\frac{LD_r}{W}\frac{\partial W}{\partial \rho_i}. \tag{14}$$

Thus the end of the unit vector lying along the dumbbell moves with a velocity given by

$$\dot{\rho}_i = (d_{ij} + \omega_{ij}) \rho_j - d_{kl} \rho_k \rho_l \rho_i - \frac{D_r}{W}\frac{\partial W}{\partial \rho_i}, \tag{15}$$

having noted that $\rho_i L = x_i$.

Now if we let a_{ij} have the form

$$a_{ij} = \oint_s \rho_i \rho_j W \, ds, \tag{16}$$

we can find an expression for $\dot{a}_{ij}$ to compare with equation (9). Taking the total derivative of equation (16) and substituting from (10) and (15) we have

$$\begin{aligned} \dot{a}_{ij} = \oint_s \Big[& (d_{ik} + \omega_{ik}) \rho_k \rho_j W + (d_{jk} + \omega_{jk}) \rho_k \rho_i W \\ & - 2 d_{kl} \rho_k \rho_l \rho_i \rho_j W - D_r \rho_j \frac{\partial W}{\partial \rho_i} \\ & - D_r \rho_i \frac{\partial W}{\partial \rho_j} + \rho_i \rho_j D_r \frac{\partial^2 W}{\partial \rho_k \partial \rho_k} \Big] ds. \end{aligned} \tag{17}$$

We have assumed homogeneous fluid flow, so the components of d_{ij} and ω_{ij} are constants on the surface of the unit sphere. Thus

$$\oint_s (d_{ik} + \omega_{ik}) \rho_k \rho_j W \, ds = (d_{ik} + \omega_{ik}) a_{kj}. \tag{18}$$

Now we note that

$$\int_v F \, dV = \frac{1}{3} \oint_s F \, ds, \quad \int_v F r^2 dV = \frac{1}{5} \oint_s F \, ds, \tag{19}$$

if F is not a function of radius. Using the divergence theorem and the results of (19), we can simplify (17). After suitable algebra, we have

$$\left. \begin{aligned} \oint_s \rho_j \frac{\partial W}{\partial \rho_i} ds &= 3 a_{ij} - \delta_{ij}, \\ \oint_s \rho_i \rho_j \frac{\partial^2 W}{\partial \rho_k \partial \rho_k} ds &= 2 \delta_{ij} - 6 a_{ij}. \end{aligned} \right\} \tag{20}$$

Using these results, equation (17) becomes

$$\dot{a}_{ij} = (d_{ik}+\omega_{ik})\,a_{kj}+a_{ik}(d_{kj}-\omega_{kj})+4D_r\delta_{ij}-12D_r a_{ij}-2d_{kl}\oint_s \rho_k\rho_l\rho_i\rho_j W\,ds. \quad (21)$$

This is almost in the form of equation (9) except for the last term. We need to know the solution of (10) for W in order to reduce this term from fourth order in $\boldsymbol{\rho}$ to terms of second order in $\boldsymbol{\rho}$. Since this is not available, we resort to an approximation.

As a first-order approximation we assume that the integral in equation (21) can be expressed as a quadratic polynomial in a_{ij}. We only require that the conditions

$$a_{ii} = 1, \quad \dot{a}_{ii} = 0 \quad (22)$$

be satisfied and that the expression for the integral be completely symmetric. This gives

$$\oint_s \rho_k\rho_l\rho_i\rho_j W\,ds = \tfrac{1}{7}[\delta_{ij}a_{kl}+\delta_{ik}a_{jl}+\delta_{il}a_{jk}+\delta_{kl}a_{ij}+\delta_{jl}a_{ik}+\delta_{jk}a_{il}] \\ -\tfrac{1}{35}[\delta_{ij}\delta_{kl}+\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}]. \quad (23)$$

Here we note that the coefficients of the quadratic terms must be zero. Using (23) in (21) and noting that $\operatorname{tr}\mathbf{d} = 0$, we have

$$\dot{a}_{ij} = \omega_{ik}a_{kj}-a_{ik}\omega_{kj}+\delta_{ij}(4D_r-\tfrac{2}{7}\operatorname{tr}\mathbf{ad}) \\ -12D_r a_{ij}+\tfrac{3}{7}(a_{ik}d_{kj}+d_{ik}a_{kj})+\tfrac{4}{35}d_{ij}. \quad (24)$$

We compare this with equation (9) and find that

$$\left.\begin{aligned} \alpha_0 = 4D_r-\tfrac{2}{7}\operatorname{tr}\mathbf{ad}, \quad \alpha_1 = -12D_r, \\ \alpha_2 = \tfrac{4}{35}, \quad \alpha_5 = \tfrac{3}{7}, \\ \alpha_3 = \alpha_4 = \alpha_6 = \alpha_7 = \alpha_8 = 0. \end{aligned}\right\} \quad (25)$$

In order to find the constitutive equation for stress, Prager assumes there is a frictional force $\mathbf{F}$ acting on the ends of the dumbbell which is proportional to the difference between the fluid velocity at the dumbbell end and the dumbbell velocity, i.e.

$$F_i = \zeta(\dot{x}_i-\dot{\rho}_i L), \quad (26)$$

where ζ is a friction factor. He assumes that this force adds a contribution to the stress tensor normally used for an incompressible Newtonian fluid with viscosity η_0 such that the stress is

$$t_{ij} = -P\delta_{ij}+2\eta_0 d_{ij}-\frac{\sigma}{\zeta}\oint_s F_i\rho_j W\,ds, \quad (27)$$

where p is pressure, σ is a scalar depending on the friction factor ζ, the dumbbell concentration, and the dumbbell size, and s is the surface of a unit sphere. Using (26), (12) and (15) the expression for stress is

$$t_{ij} = -p\delta_{ij}+2\eta_0 d_{ij}+\sigma\left(-D_r\delta_{ij}+3D_r a_{ij}+d_{kl}\oint_s \rho_k\rho_l\rho_i\rho_j W\,ds\right), \quad (28)$$

having evaluated terms as above. If we evaluate the integral in (28) as was done above in the expression for $\dot{a}_{ij}$, we can find a more simple constitutive equation for the stress,

$$t_{ij} = \delta_{ij}\left(-p-\sigma D_r+\frac{\sigma}{7}\operatorname{tr}\mathbf{ad}\right)+3\sigma D_r a_{ij} + (2\eta_0-\tfrac{2}{35}\sigma)\,d_{ij}+\tfrac{2}{7}\sigma(a_{ik}d_{kj}+d_{ik}a_{kj}). \tag{29}$$

Comparing this with equation (2) gives

$$\left.\begin{aligned} \beta_0 &= -p-\sigma D_r+\tfrac{1}{7}\sigma\operatorname{tr}\mathbf{ad}, \quad \beta_1 = 3\sigma D_r,\\ \beta_2 &= 2\eta_0-\tfrac{2}{35}\sigma, \quad \beta_5 = \tfrac{2}{7}\sigma,\\ \beta_3 &= \beta_4 = \beta_6 = \beta_7 = \beta_8 = 0. \end{aligned}\right\} \tag{30}$$

So we see that Prager's theory is a special case of the theory proposed in § 2 at least for the simplification introduced in equation (23).

4. Simple shear

In this section we consider the fluid motion given by $\dot{x}_1 = Kx_2$, $\dot{x}_2 = \dot{x}_3 = 0$, which is referred to as simple shear. We let K be a constant. The rate of deformation d_{ij} and the vorticity ω_{ij} reduce to

$$d_{12} = d_{21} = \omega_{12} = -\omega_{21} = \tfrac{1}{2}K, \tag{31}$$

and all other components are zero. We will simplify the expressions (2) and (9) and examine the microscopic motion and the resulting stress components.

We consider that the a_{ij} are the coefficients of the quadratic form that describes the ellipsoidal shape of a particle suspended in a fluid. We assume that when the fluid is at rest, these particles tend to a spherical shape and for large times become ellipsoidal in shape when the fluid is sheared. We consider that inertial forces are negligible so that equation (9) describes the motion of the ellipsoid.

In order to make our calculations tractable we assume that the expression for $\dot{a}_{ij}$ is linear in the components of d_{ij} and that the ellipsoidal particle never varies much from spherical shape. That is, we assume

$$a_{ij} = a\delta_{ij}+a'_{ij}, \tag{32}$$

where all elements of a'_{ij} are small compared to a. We substitute (32) into (9) and linearize, keeping products of the components of a'_{ij} and d_{ij}. The motivation for retaining these particular terms is due to a comparison with the results from Prager given in (24). We let a be a constant, and dropping the prime we have

$$\dot{a}_{ij} = \omega_{ik}a_{kj}-a_{ik}\omega_{kj}+\delta_{ij}(\gamma_0+\gamma_1\operatorname{tr}\mathbf{a}+\gamma_2\operatorname{tr}\mathbf{ad}) + a_{ij}\gamma_3+d_{ij}(\gamma_4+\gamma_5\operatorname{tr}\mathbf{a})+(a_{ik}d_{kj}+d_{ik}a_{kj})\,\gamma_6, \tag{33}$$

where the γ's are constants and $\operatorname{tr}\mathbf{d} = 0$. The six components of (33) are, in detail,

$$\left.\begin{aligned} \dot{a}_{11} &= \gamma_0+(\gamma_1+\gamma_3)\,a_{11}+\gamma_1 a_{22}+\gamma_1 a_{33}+K(\gamma_6+\gamma_2+1)\,a_{12},\\ \dot{a}_{22} &= \gamma_0+\gamma_1 a_{11}+(\gamma_1+\gamma_3)\,a_{22}+\gamma_1 a_{33}+K(\gamma_6+\gamma_2-1)\,a_{12},\\ \dot{a}_{33} &= \gamma_0+\gamma_1 a_{11}+\gamma_1 a_{22}+(\gamma_1+\gamma_3)\,a_{33}+K\gamma_2 a_{12},\\ \dot{a}_{12} &= \tfrac{1}{2}K\gamma_4+\tfrac{1}{2}K(\gamma_6+\gamma_5-1)\,a_{11}+\tfrac{1}{2}K(\gamma_6+\gamma_5+1)\,a_{22}+\tfrac{1}{2}K\gamma_5 a_{33}+\gamma_3 a_{12},\\ \dot{a}_{13} &= \tfrac{1}{2}K(\gamma_6+1)\,a_{23}+\gamma_3 a_{13},\\ \dot{a}_{23} &= \tfrac{1}{2}K(\gamma_6-1)\,a_{13}+\gamma_3 a_{23}. \end{aligned}\right\} \tag{34}$$

In order that the components of a approach stable values for large times, we find from the last two expressions of (34) that the rate of shear must satisfy $K < -2\gamma_3/(\gamma_6^2-1)^{\frac{1}{2}}$, requiring that $\gamma_3 < 0$ and $|\gamma_6| > 1$. It is interesting to note here from (24) and (25) that no steady-state solution exists for a_{ij} in Prager's theory, i.e. there is no statistical tendency for a suspension of dumbbells to become oriented in simple shear. We arrive at this conclusion with reservation because of the simplification introduced in equation (23).

We also note that if γ_0 and γ_4 are zero in **(34)**, the particle remains a sphere for large times until a critical rate of shear is reached. At this point the sphere begins to stretch until the linearization utilized is no longer valid. The case of γ_0 and γ_4 being zero would describe a fluid which behaves as a Newtonian fluid for low rates of shear and becomes non-Newtonian above a certain shear rate.

We can solve equation (34) for the components of a_{ij} for steady-state flow by putting $\dot{a}_{ij} = 0$. We assume that a stable solution exists, i.e. that we have not exceeded the critical rate of shear and the conditions on the y's hold. We find that

$$\left.\begin{aligned}
a_{11} &= d^{-1}\{-\gamma_0\gamma_3^3+\tfrac{1}{2}K^2\gamma_3[\gamma_3\gamma_4(\gamma_6+\gamma_2+1)\\
&\quad +\gamma_1\gamma_4(\gamma_6+3)-2\gamma_0(\gamma_6+\gamma_5+1)-\gamma_0\gamma_5(\gamma_6+1)]\},\\
a_{22} &= d^{-1}\{-\gamma_0\gamma_3^3+\tfrac{1}{2}K^2\gamma_3[\gamma_3\gamma_4(\gamma_6+\gamma_2-1)\\
&\quad +\gamma_1\gamma_4(\gamma_6-3)+2\gamma_0(\gamma_6+\gamma_5-1)-\gamma_0\gamma_5(\gamma_6-1)]\},\\
a_{33} &= d^{-1}\{-\gamma_0\gamma_3^3+\tfrac{1}{2}K^2\gamma_3[\gamma_3\gamma_4\gamma_2-2\gamma_1\gamma_4\gamma_6\\
&\quad +\gamma_0(\gamma_6+\gamma_5-1)(\gamma_6+1)+\gamma_0(\gamma_6+\gamma_5+1)(\gamma_6-1)]\},\\
a_{12} &= \tfrac{1}{2}K\gamma_3^2d^{-1}\{2\gamma_0\gamma_6+3\gamma_0\gamma_5-\gamma_3\gamma_4-3\gamma_1\gamma_4\},\\
a_{13} &= a_{23} = 0,
\end{aligned}\right\} \qquad (35)$$

where

$$\begin{aligned}
d = \gamma_3^2\{\gamma_3(\gamma_3+3\gamma_1)-\tfrac{1}{2}K^2[(\gamma_6+\gamma_5+1)(\gamma_6+\gamma_2-1)\\
+(\gamma_6+\gamma_5-1)(\gamma_6+\gamma_2+1)+(2\gamma_1/\gamma_3)(\gamma_6^2-6)+\gamma_2\gamma_5]\}.
\end{aligned}$$

Now if $K = 0$, we have a sphere of radius $A = \{-(\gamma_3+3\gamma_1)/\gamma_0\}^{\frac{1}{2}}$. We non-dimensionalize the components of a_{ij} by multiplying by the radius squared to give

$$\left.\begin{aligned}
a_{11}A^2 &= 1+K^2d^{-1}\{\tfrac{1}{2}\gamma_3A^2[\gamma_3\gamma_4(\gamma_6+\gamma_2+1)+\gamma_1\gamma_4(\gamma_6+3)\\
&\quad -2\gamma_0(\gamma_6+\gamma_5+1)-\gamma_0\gamma_5(\gamma_6+1)]+k\},\\
a_{22}A^2 &= 1+K^2d^{-1}\{\tfrac{1}{2}\gamma_3A^2[\gamma_3\gamma_4(\gamma_6+\gamma_2-1)+\gamma_1\gamma_4(\gamma_6-3)\\
&\quad +2\gamma_0(\gamma_6+\gamma_5-1)-\gamma_0\gamma_5(\gamma_6-1)]+k\},\\
a_{33}A^2 &= 1+K^2d^{-1}\{\tfrac{1}{2}\gamma_3A^2[\gamma_3\gamma_4\gamma_2-2\gamma_1\gamma_4\gamma_6\\
&\quad +\gamma_0(\gamma_6+\gamma_5-1)(\gamma_6+1)+\gamma_0(\gamma_6+\gamma_5+1)(\gamma_6-1)]+k\},\\
a_{12}A^2 &= \tfrac{1}{2}A^2K\gamma_3^2d^{-1}\{2\gamma_0\gamma_6+3\gamma_0\gamma_5-\gamma_3\gamma_4-3\gamma_1\gamma_4\},
\end{aligned}\right\} \qquad (36)$$

where

$$\begin{aligned}
k = \tfrac{1}{2}\gamma_3^2\{(\gamma_6+\gamma_5+1)(\gamma_6+\gamma_2-1)+(\gamma_6+\gamma_5-1)(\gamma_6+\gamma_2+1)\\
+(2\gamma_1/\gamma_3)(\gamma_6^2-6)+\gamma_2\gamma_5\}.
\end{aligned}$$

We see that a,,, a_{22} and a_{33} remain small for large rates of shear if $-A^2\gamma_3/2k$ times the respective square brackets remain close to unity.

We consider now the orientation of the ellipsoid assuming that the components of a_{ij} remain small compared to A^{-2}, the spherical radius at $K = 0$. The eigenvalues of a_{ij} are given by

$$\left.\begin{aligned} \lambda_1 &= \tfrac{1}{2}(a_{11}+a_{22})+\tfrac{1}{2}\{(a_{11}-a_{22})^2+4a_{12}^2\}^{\frac{1}{2}},\\ \lambda_2 &= \tfrac{1}{2}(a_{11}+a_{22})-\tfrac{1}{2}\{(a_{11}-a_{22})^2+4a_{12}^2\}^{\frac{1}{2}},\\ \lambda_3 &= a_{33}. \end{aligned}\right\} \tag{37}$$

Using (35) we find the normalized eigenvectors to be

$$\hat{e}_1 = \frac{\pm[\{B+(1+B^2)^{\frac{1}{2}}\}\hat{x}_1-\hat{x}_2]}{\sqrt{2}\,[B^2+B(1+B^2)^{\frac{1}{2}}+1]^{\frac{1}{2}}},\quad \hat{e}_2 = \frac{\pm[\{B-(1+B^2)^{\frac{1}{2}}\}\hat{x}_1-\hat{x}_2]}{\sqrt{2}\,[B^2-B(1+B^2)^{\frac{1}{2}}+1]^{\frac{1}{2}}},\quad \hat{e}_3 = \pm\hat{x}_3, \tag{38}$$

where $B = K/\gamma_3 < 0$, assuming K to be a positive number. The $\hat{e}$'s are the unit vectors lying along the principal axes of the ellipsoid, and the x's are unit vectors lying along the co-ordinates axes. From (37) we note that $\lambda_3 \geqslant \lambda_1 \geqslant \lambda_2$ or $\lambda_1 \geqslant \lambda_2 \geqslant \lambda_3$. We exclude the equality signs since these would require special values for the γ's appearing in (35). If λ_3 is the largest eigenvalue, we have an ellipsoid whose major axis is in the $\hat{e}_2$ direction and minor axis in the $\hat{e}_3$ direction. If λ_3 is the smallest eigenvalue, we have an ellipsoid whose major axis is in the $\hat{e}_3$ direction and minor axis in the $\hat{e}_1$ direction. For either case we see from (38) that the larger of the two principal axes of the ellipsoid in the (x_1, x_2)-plane points 45° counterclockwise from the x_1 axis for zero rate of shear. This angle becomes smaller as the shear rate increases. For very large shear rates the ellipsoid aligns itself with the co-ordinate axes.

Now we consider the stress given by equation (2). We linearize the stress with respect to the components of a_{ij} and d_{ij} as was done above with $\dot{a}_{ij}$, again retaining the products of the components of a_{ij} and d_{ij}. The motivation for retaining these particular terms is again due to a comparison with the results from Prager given in equation (29). The stress takes the simplified form

$$t_{ij} = (\delta_0+\delta_1\,\mathrm{tr}\,\mathbf{a}+\delta_2\,\mathrm{tr}\,\mathbf{ad})\,\delta_{ij}+\delta_3 a_{ij}+(\delta_4+\delta_5\,\mathrm{tr}\,\mathbf{a})\,d_{ij}+\delta_6(a_{ik}d_{kj}+d_{ik}a_{kj}), \tag{39}$$

where the δ's are considered constants. Writing out the components of (39) gives

$$\left.\begin{aligned} t_{11} &= (\delta_0+\delta_1\,\mathrm{tr}\,\mathbf{a}+\delta_2\,\mathrm{tr}\,\mathbf{ad})+\delta_3 a_{11}+\delta_6 a_{12}K,\\ t_{22} &= (\delta_0+\delta_1\,\mathrm{tr}\,\mathbf{a}+\delta_2\,\mathrm{tr}\,\mathbf{ad})+\delta_3 a_{22}+\delta_6 a_{12}K,\\ t_{33} &= (\delta_0+\delta_1\,\mathrm{tr}\,\mathbf{a}+\delta_2\,\mathrm{tr}\,\mathbf{ad})+\delta_3 a_{33},\\ t_{12} &= \delta_3 a_{12}+\tfrac{1}{2}(\delta_4+\delta_5\,\mathrm{tr}\,\mathbf{a})\,K+\tfrac{1}{2}\delta_6(a_{11}+a_{22})\,K,\\ t_{13} &= \delta_3 a_{13}+\tfrac{1}{2}\delta_6 a_{23}K,\\ t_{23} &= \delta_3 a_{23}+\tfrac{1}{2}\delta_6 a_{13}K. \end{aligned}\right\} \tag{40}$$

We see from (35) that, for steady state, $t_{13} = t_{23} = 0$ for all values of K as long as the ellipticity remains small. We are interested in various combinations of the stress components, namely the normal stress differences $t_{11}-t_{22}$ and $t_{11}-t_{33}$, and the apparent viscosity t_{12}/K. From (40) these are

$$\left.\begin{aligned} t_{11}-t_{22} = \delta_3(a_{11}-a_{22}),\quad t_{11}-t_{33} &= \delta_3(a_{11}-a_{33})+\delta_6 a_{12}K,\\ t_{12}/K &= \delta_3 a_{12}/K+\tfrac{1}{2}(\delta_4+\delta_5\,\mathrm{tr}\,\mathbf{a})+\tfrac{1}{2}\delta_6(a_{11}+a_{22}). \end{aligned}\right\} \tag{41}$$

Substituting from (35) we have

$$\left.\begin{aligned} t_{11}-t_{22} &= \delta_3\gamma_3 eK^2/d, \quad t_{11}-t_{33} = \tfrac{1}{2}\gamma_3 eK^2[(\gamma_6+1)\,\delta_3-\gamma_3\delta_6]/d,\\ t_{12}/K &= \tfrac{1}{2}\delta_4+\tfrac{1}{2}\{-\delta_3\gamma_3^2 e-3\delta_5\gamma_0\gamma_3^3-2\delta_6\gamma_0\gamma_3^3\\ &\quad+\tfrac{1}{2}K^2\gamma_3[\delta_5(2\gamma_3\gamma_4\gamma_6+3\gamma_3\gamma_4\gamma_2-6\gamma_0+2\gamma_0\gamma_6^2)\\ &\quad+\delta_6(2\gamma_3\gamma_4\gamma_6+2\gamma_3\gamma_4\gamma_2+2\gamma_1\gamma_4\gamma_6-4\gamma_0-2\gamma_0\gamma_5\gamma_6)]\}/d, \end{aligned}\right\} \tag{42}$$

where

$$e = \gamma_3\gamma_4+3\gamma_1\gamma_4-2\gamma_0\gamma_6-3\gamma_0\gamma_5$$

and d is defined in (35).

We see that the normal stress differences are proportional to K^2 for low values of K and ultimately approach limiting constants as K gets very large. The apparent viscosity has two different limiting values at low and high rates of shear. From the theory it is not possible to determine whether the apparent viscosity at high rates of shear is greater or lower than that at low rates of shear. In the next section these results will be compared with some experimental measurements.

5. Experimental comparisons

Many investigators have measured the properties of non-Newtonian fluids, but few have measured normal stress and apparent viscosity over large ranges of rate of shear for any given material. Ideally, we would like measurements of the normal stress differences $t_{11}-t_{22}$ and $t_{11}-t_{33}$ and the apparent viscosity t_{12}/K for a given material at a fixed temperature over a range of rate of shear from say $K = 0$ to $K = 10^4\,\mathrm{sec}^{-1}$. Also, measurements on more than one type of instrument would be of great benefit. The comparisons to be made in this paper are in the author's opinion amongst the better experimental results. Instruments commonly in use are the cone-and-plate and parallel-plate rheogoniometers. The most common material tested is polyisobutylene in solution which is available in a variety of molecular weights. We consider comparing the results of the anisotropic theory with experiments on high polymers such as polyisobutylene, because it is believed that very long chain molecules in solution tend statistically to be spherical in shape when the fluid is at rest and to elongate when the fluid is sheared. This is the model proposed in the last section.

Many authors consider that Weissenberg's conclusion holds, i.e. that $t_{22}-t_{33} = 0$. Others believe that this quantity is small compared with the stress difference $t_{11}-t_{22}$ but not zero. The behaviour of $t_{22}-t_{33}$ is not well understood for even the most commonly tested materials. If we assume that Weissenberg's conclusion holds, we see from equation (42) that

$$\delta_6 = \delta_3(\gamma_6-1)/\gamma_3. \tag{43}$$

There is no reason why equation (43) should or should not hold, since the theory of anisotropic fluids does not specify values for the δ's. We will only consider the normal stress difference $t_{11}-t_{22}$ in the following paragraphs since so little is known about the other normal stress differences.

Now looking at equation (42), we see that these equations have the form

$$t_{11}-t_{22} = \frac{K^2}{p+qK^2}, \quad \frac{t_{12}}{K} = \frac{m+nK^2}{p+qK^2}. \tag{44}$$

Here m, n, p, and q are constants and K is the rate of shear. We see that the normal stress difference approaches K^2/p as $K \to 0$ and $1/q$ as K gets sufficiently large. The apparent viscosity approaches m/p as $K \to 0$ and n/q as K gets sufficiently large, two constants which are unequal in general. We will attempt to fit experimental curves by evaluating the constants in equations (44).

Greensmith & Rivlin (1953) performed very precise experiments using a parallel-plate rheogoniometer. Figure 1 represents data found for polyisobutylene dissolved in Tetralin at 25 °C. Curve A is for a 34·8 % by weight solution of B 15 and curve B is for a 6 % solution of B 120 polyisobutylene. Curve A is a plot of the formula

$$\bar{h}_0 - \bar{h} = \frac{K^2}{2213 + 8{\cdot}96 \times 10^{-2} K^2} \tag{45}$$

and curve B is a plot of the formula

$$\bar{h}_0 - \bar{h} = \frac{K^2}{262 + 2{\cdot}42 K^2}. \tag{46}$$

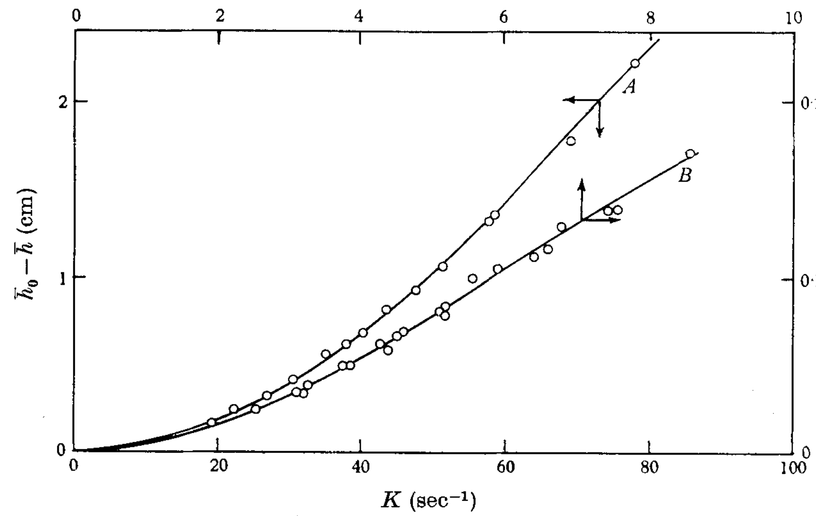

FIGURE 1. Normal stress *vs* rate of shear for two polyisobutylene solutions (from Greensmith & Rivlin 1953).

Here $\bar{h}_0$ is the average height of rise of fluid at the apparatus centre-line and $\bar{h}$ the average height of rise of fluid at other points. Generally $\bar{h}_0 - \bar{h} = (t_{220} - t_{22})/\rho g$, where ρ is fluid density, g the gravitational constant, and $t_{220} = t_{110} = t_{330}$. The theoretical curves agree with the data for the low rates of shear of the experiment.

Markovitz & Williamson (1957) used a cone-and-plate instrument with polyisobutylene. Figure 2 represents values for Vistanex B-100 polyisobutylene dissolved in Decalin. A variety of concentrations and temperatures were used, but the data were reduced to standard conditions using the formulas

$$K_r = \frac{K\eta T_0}{F(c)\,T}, \quad \nu_r = \frac{\nu T_0}{F(c)\,T}, \tag{47}$$

where η is the apparent viscosity at zero shear rate, $T_0 = 298\,°\mathrm{K}$, T is the temperature, $F(c) = c/c_0$ for $c \leqslant c_0$, $F(c) = (c/c_0)^2$ for $c \geqslant c_0$, c is the weight concentration, $c_0 = 0{\cdot}08$, $\nu = \frac{1}{3}(t_{11}-t_{22})$, and the subscript r stands for reduced. In figure 2 curve A is given by the formula

$$\nu_r = \frac{K_r^2}{1513+3{\cdot}03\times 10^{-3}K_r^2} \tag{48}$$

and curve B by the formula

$$\nu_r = \frac{K_r^2}{1483+4{\cdot}93\times 10^{-3}K_r^2}. \tag{49}$$

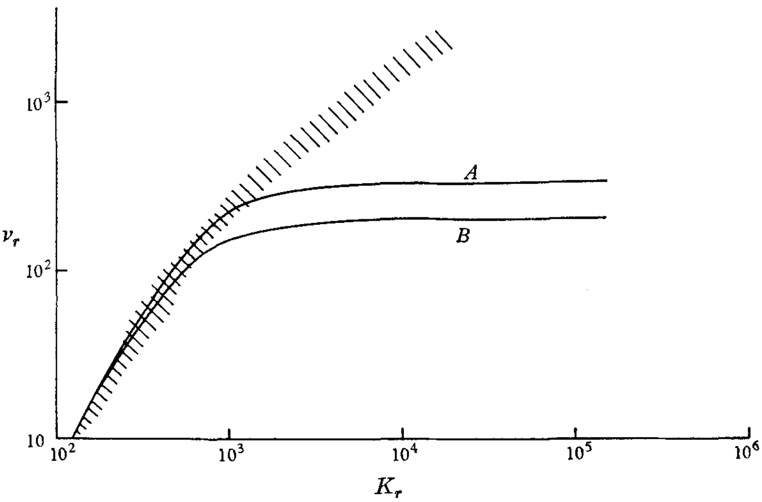

FIGURE 2. The hatched lines represent the spread of data of reduced normal stress *vs* reduced rate of shear for a large variety of polyisobutylene solutions (from Markovitz & Williamson 1957).

We see that the theoretical curves are close to the experimental curves for K sufficiently small, but the curves deviate widely at high rates of shear.

Kotaka, Kurata & Tamura (1957) used a parallel-plate instrument to measure the normal stress difference and the apparent viscosity of polystyrene dissolved in Decalin. Their experiments were performed at 25 °C. In figure 3 we have the results of the normal stress measurements. Curve A is for a 19·8 % solution by weight and is given by the formula

$$t_{11}-t_{22} = \frac{K^2}{4{\cdot}65+2{\cdot}74\times 10^{-4}K^2}. \tag{50}$$

Data for a 24·5 % solution are represented by the theoretical curve B given by

$$t_{11}-t_{22} = \frac{K^2}{0{\cdot}445+3{\cdot}71\times 10^{-4}K^2}, \tag{51}$$

or by curve C given by

$$t_{11}-t_{22} = \frac{K^2}{0{\cdot}457+1{\cdot}08\times 10^{-4}K^2}. \tag{52}$$

Data for a 30·8 % solution are represented by the theoretical curve D given by

$$t_{11}-t_{22} = \frac{K^2}{3{\cdot}73\times 10^{-2}+1{\cdot}98\times 10^{-4}K^2}. \tag{53}$$

We again find that we can approximate the experimental curves fairly well for sufficiently low values of the rate of shear.

Kotaka *et al.* measured the apparent viscosity for the same solutions mentioned in the above paragraph. In figure 4, data for a 19·8 % solution are given by

$$\frac{t_{12}}{K} = \frac{66{\cdot}4 + 3{\cdot}06 \times 10^{-3} K^2}{4{\cdot}65 + 2{\cdot}74 \times 10^{-4} K^2}. \tag{54}$$

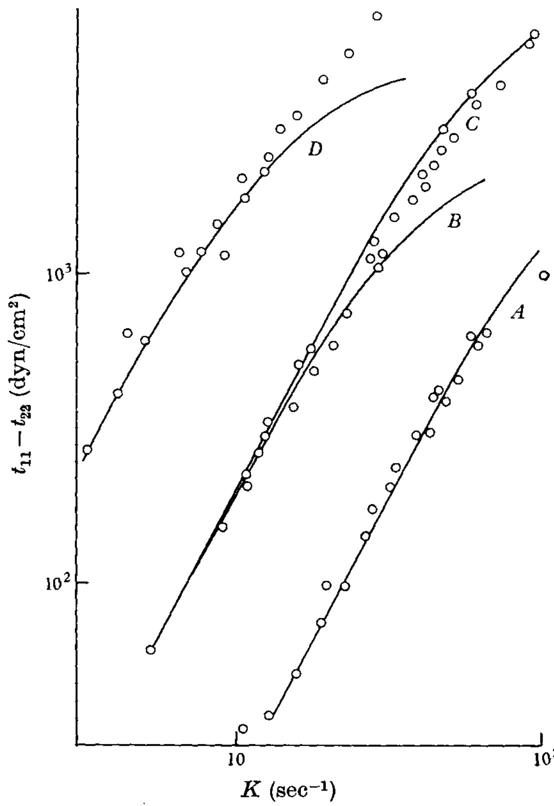

FIGURE 3. Normal stress *vs* rate of shear for three polystyrene solutions (from Kotaka *et al.* 1957).

In figure 5, curve A is the theoretical curve for a 24·5 % solution and is given by

$$\frac{t_{12}}{K} = \frac{27{\cdot}0 + 1{\cdot}61 \times 10^{-2} K^2}{0{\cdot}445 + 3{\cdot}71 \times 10^{-4} K^2}. \tag{55}$$

Curve B is the theoretical curve for a 30·8 % solution and is given by

$$\frac{t_{12}}{K} = \frac{12{\cdot}64 + 3{\cdot}38 \times 10^{-2} K^2}{3{\cdot}73 \times 10^{-2} + 1{\cdot}98 \times 10^{-4} K^2}. \tag{56}$$

From the above comparisons with experiment we see that, at least for sufficiently low rates of shear, we can approximate data fairly well by the formulas given in (44) for certain materials. The values of the constants in (44) depend on the material and on concentration. At high rates of shear the comparison is not close, but it must be noted that the experimental results at high rates of shear are in doubt due to heating, secondary flow, and other non-ideal effects in the apparatus.

It is generally accepted that the apparent viscosity has two limiting values at high and low rates of shear for certain materials as predicted in (44).

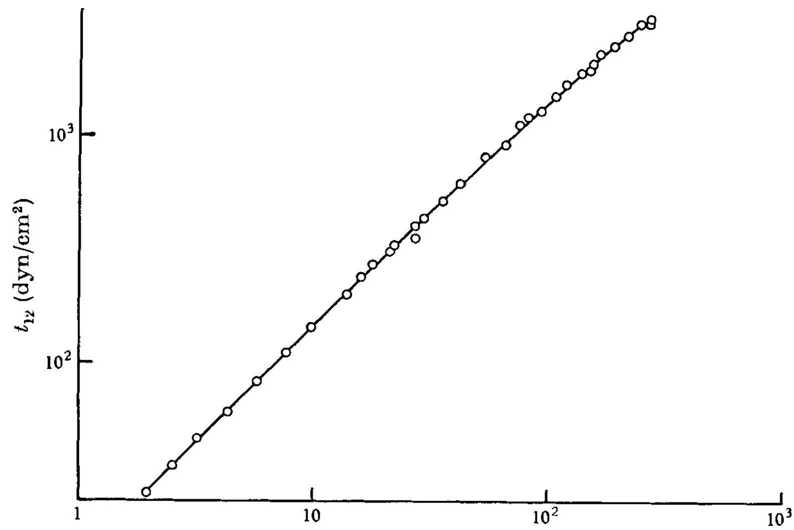

FIGURE 4. Apparent viscosity *vs* rate of shear for a 19·8 % solution of polystyrene (from Kotaka *et al.* 1957).

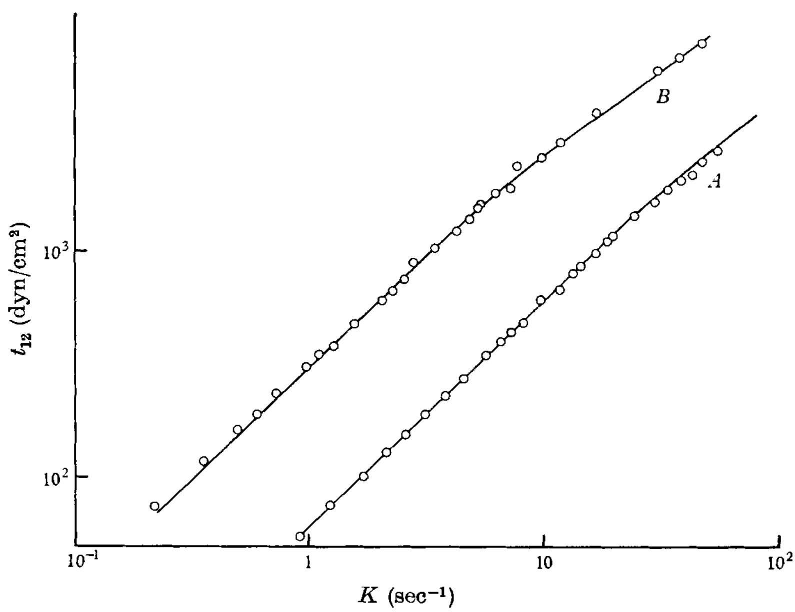

FIGURE 5. Apparent viscosity *vs* rate of shear for 24·5 and 30·8 % solutions of polystyrene (from Kotaka *et al.* 1957).

In comparing the results found in equations (35) and (38) for shape and orientation of suspended particles, there is little experimental information other than of a qualitative nature. Visual observation of immiscible drops suspended in a fluid indicates that the drops elongate in simple shear. The higher the shear rate the greater is the elongation of the drop. The drops tend to line up with their major axis at 45° to the flow direction at low rates of shear, and this angle decreases with increasing rates of shear. The theory agrees with these observations.

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