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On the modeling of a microstructural pair of constitutive equations to describe the motion of elastic fluids

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Abstract

Scaling arguments based on a balance between viscous drag and restoring Brownian forces are used in order to generate a nonlinear dumbbell model with finite spring correction and a drag correction for a dilute polymer solution. The microstructure coupled equations leads to the most commonly Oldroyd-like constitutive equation. We show that the investigated model is able to capture generalized nonlinear response of viscoelastic fluids. The approach is used to investigate the nonlinear response of an elastic liquid under strong extensional flows. We characterize the nonlinear effect of the fluid in terms of an extensional viscosity that is found to be a function of three relevant parameters of the problem: Deborah number, the macromolecule extensibility (or anisotropy) and the macromolecule volume fraction. We verify the predictions of the model with Batchelor's calculation of extensional viscosity based on a slender body theory for suspensions composed of long rigid rods. The analysis may explain from a phenomenological point of view why few ppms of macromolecules of high molecule weight or a small concentration of long fibers may produce drastic changes in the pressure drop of robust turbulent flows. In addition, the proposed analysis may be useful for direct computer simulations of turbulent flows in the presence of macromolecule .

Keywords: extensional viscosity, anisotropic fluid, elasticity, Spring-Dumper model

1 Introduction

The addition of small amounts of polymer to a fluid can drastically enhance the extensional viscosity of the solution while just slightly affecting the shear viscosity [7, 8]. The large increase in the extensional viscosity of dilute polymer solutions reduces the pressure drop in flows through porous media, observed by [5], and the drag in turbulent pipe flow, observed by a vast number of work e.g. [1, 2, 10, 13]. For polymer melts, the measurements of the extensional properties have been carried out at either constant deformation rates or constant stresses, and steady state have been reported. For polymer solutions, however it is only recently that the technique of filament stretching at constant extension rates, that has been introduced by [14], allowed comparable extensional viscosity measurements.

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Due to the important role of the extensional viscosity in these and many other flow phenomena, it is of great interest to be able to measure the extensional viscosity of dilute polymer solutions. However, it is difficult to create a well defined extensional flow field for dilute polymer solutions and so difficult to develop models describing the behavior of the macromolecules in the flow. The rheological properties of polymer solutions are strongly dependent upon the concentration regime of the polymer under study. In semi-dilute and concentrated systems, entanglement effects can have a predominant role in defining the behavior of the system. In dilute solutions, the rheological properties of the system are a reflection of the mechanical properties of the individual polymer chains and the number of chains in the bulk solution. However, even in such dilute systems, the mechanical properties of the individual chains and, hence, the bulk solution are strongly dependent on the type of flow. In shear flow, the mechanical properties change gradually with the shear rate while in extensional flow the variations can be relatively sharp. Close to the critical strain rate, a coil stretch transition occurs where the coil quickly reaches its full stretched state. Such stretching significantly increases the hydrodynamic volume of the polymer chain, which leads to an increase in the extensional viscosity of the solution.

The rheological behavior of the common Oldroyd constitutive equation is well-known e.g. [4]. In steady simple shear flow, the effective viscosity is constant, the first-normal-stress difference increases quadratically with shear-rate, and there is no second-normal-stress differences. In steady pure straining motion, the extensional viscosity increases with strain-rate, becoming infinite at the finite critical Deborah number ($De \sim 1$).

To understand the physics in this constitutive equation for elastic liquids, it is helpful to look at a micro-structural model which leads to an Oldroyd-like-constitutive equation. Rather than studying the rheological performance and mathematical structure of the coupled flow equations, we retreat to some physical model of the microstructure which generates the constitutive equation. In this article, it is considered a bead-and-spring model of polymer solutions. The simplest version of the bead-and-spring model (i.e. dumbbell model) was introduced by [9] and discussed in details by [7] and [11] with a finite spring correction, i.e. a FENE (i.e. finitely extensible non-linear elastic) model. The original empirical FENE spring force was first proposed for dumbbells by [15].

We present scaling arguments based on a balance between viscous drag and a restoring Brownian force which lead to a time dependent partial differential equation for the conformation tensor of the macromolecule. In particular, this tensor gives an explicit information about the internal structure of the fluid. We then describe a highly anisotropic fluid flow with a nonlinear spring model which gives a finite limit of macromolecule extension and a nonlinear damper in the way suggested by [11]. The present analysis has been attractive because deformable particles almost always become rod-like in shape when greatly deformed by the bulk flow, and this anisotropy may act as a stabilizing factor even in the flow of very dilute polymer solution.

2 Dimensional analysis

Dilute polymer solutions (no overlapping of the macromolecules) behavior can be idealized as being a polymer chain composed of two beads representing drag forces, linked to a Hookean spring representing elastic forces (see figure 1). This configuration bead-and-spring is called a dumbbell e.g. [4]. In this model, a macromolecule is composed of N rigid segments (each corresponding to an individual monomers) of length δ, with each individual monomer randomly oriented with respect to the adjoining segments. The equilibrium configuration distribution of the macromolecule is then given by a random walk of N steps, each of length δ. In this model, the mean end-to-end distance of the polymer, a is proportional to $N^{1/2}\delta$ as required by the central limit theorem, which is smaller than the length of the polymer chain by a factor of $N^{1/2}$. In figure (1a), the vector $\mathbf{r}$ between the centres of the two beads represents the end-to-end distance of the polymer molecule.

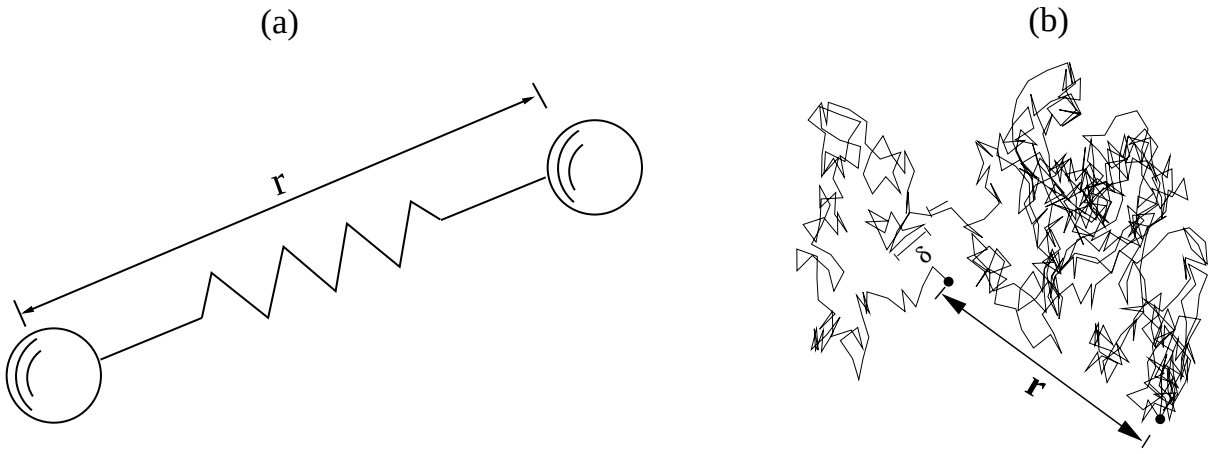

Figure 1: Sketch of a polymer molecule. (a) The elastic dumbbell model and (b) Polymer configuration corresponding a chain of N individual manometer randomly orientated.

In a flow, the distribution of polymer configurations will be affected by the local velocity gradient. In an extensional flow the difference in the fluid velocity acting on the two halves of the polymer will cause the molecule to extend. The extension is opposed by Brownian motion which tend to restore the equilibrium distribution.

Let us examine the equilibrium condition between Brownian and viscous forces and consider, for instance, the simplest linear Hookean spring for modelling the restoring Brownian force. We express the elastic force as being $F_B = Gr$, where G denotes the spring constant given by $G = 3KT/a^2$ e.g. [6]. Here, a denotes the equilibrium length of the macromolecule (i.e. a typical value of $|\mathbf{r}| = r$ for the elastic regime where a linear spring model makes sense.), K is the Boltzmann constant and T the absolute temperature.

The hydrodynamic interactions between the molecule and the solvent is represented by the

viscous drag on the beads. So, in a dumbell model, the viscous force is defined as the force exerted over an isolated spherical particle of ratio a by the solvent with viscosity μ. For a low Reynolds number, the expression is given by the Stokes law, namely, $F_V = 6\pi\mu a dr/dt$. At the thermodynamic equilibrium state, the balance between the viscous and the restoring forces acting on the macromolecule leads to

$$\frac{dr}{dt} = \frac{G}{C_v} r, \tag{1}$$

where $C_v = 6\pi\mu a$. Integrating equation (1) fort he initial condition $r(0) = r_0$, one obtains:

$$r(t) = r_0 \exp\left(\frac{t}{\tau}\right), \tag{2}$$

where $\tau = C_v/G = 2\pi\mu a^3/KT$ denotes the relaxation time of an extended macromolecule to the randomly-coiled state. Since N is equal to the molecular weight of the polymer (M) divided by the molecular weight of a single monomer (M_i), $N = M/M_i$ it results that the macromolecule relaxation time is proportional to $M^{3/2}$.

2.1 Physical parameters of the flow system

In defining the Reynolds number, Re, for a polymer solution, we need to define carefully what is meant by the viscosity μ, since both shear and extensional viscosity vary with shear-rate. Conventionally, μ is taken to be the shear viscosity in the limit of small shear rates. Most polymeric fluids are highly viscous and so we consider the limit where Re is small. The flow of a fluid with density ρ and viscosity μ flowing through a body of length scale a with velocity scale u_c, Re is defined as $Re = \rho u_c a/\mu$.

In the context of elastic liquids, a fundamental parameter is the Deborah number, defined as the ratio of the relaxation time and a characteristic time of the flow, i.e. $De = \tau/t_f$, where τ denotes the relaxation time of the polymer. The relaxation time measures the ability of the macromolecules to recoil to their equilibrium condition after stretching. The flow time scale t_f is defined as the ratio of a velocity u_c and a length a, characteristics of the flow. $De << 1$ means weak flow and the fluid behaves closely to a Newtonian fluid. In this limit, Brownian motion prevents the polymer from becoming highly extended and the macromolecule distribution remains close to the randomly coiled distribution. We denote this limit as being the elastic regime, with $r \sim a$. For large values of Deborah (i.e. $De \sim 1$), however, the flow is strong enough to overcome the Brownian relaxation and so the polymer may become greatly extended, so that the mean end-to-end distance becomes large compared to its values in the randomly-coiled distribution, i.e. $r \gg a$. This second regime will correspond to the anisotropic limit of the macromolecule. For strong flows, $De \sim 1$, the macromolecule stretching produces anisotropy in the flow and non-linear effects as normal-stress-difference.

A second important dimensionless parameter is the extensibility of the polymer, L, defined as being the ratio of the full stretched length ($\ell = N\delta$) to the average length corresponding to

the end-to-end distance in the randomly coiled state of the macromolecule, a, i.e. $L = N\delta/a$. In the present work, while Deborah can be interpreted as a measure of the polymer elasticity on the flow, L will denote a measure of the flow anisotropy created by the macromolecule orientation. We will examine the effect of both contribution to the flow.

The final dimensionless group is a measure of the effective volume fraction of the macromolecule evaluated at the randomly coiled state, namely $\phi = 4\pi a^3 n/3$, where n is the number of molecules, N_m per unit of volume , i.e. $n = N_m/V$. For the solution to be dilute, ϕ must be much less than unit, therefore the polymer contribution to the fluid stress will be negligible unless the polymer become highly extended.

3 Mathematical model

The balance equations are the conservation of mass and of momentum

$$\nabla \cdot \mathbf{u} = 0, \qquad \nabla \cdot \boldsymbol{\Sigma} = \mathbf{0} \tag{3}$$

where inertia is neglected.

3.1 Material description

In general, the stress tensor for an elastic liquid is given by the following constitutive equation

$$\boldsymbol{\Sigma} = -p\boldsymbol{I} + \boldsymbol{\sigma} \tag{4}$$

with the deviatoric stress $\boldsymbol{\sigma}$ given by

$$\boldsymbol{\sigma} = 2\mu_s(\phi, \dot{\gamma})\mathbf{D} + \overline{\boldsymbol{\sigma}}_B, \tag{5}$$

where p is the mechanical pressure, $\boldsymbol{I}$ is the unit tensor, $\mu_s(\phi, \dot{\gamma})$ is the shear viscosity as a function of the volume fraction, ϕ, and the shear rate, $\dot{\gamma}$, $\overline{\boldsymbol{\sigma}}_B$ is the elastic stress contribution due to the presence of macromolecules and $\mathbf{D}$ is the rate of the strain tensor, $\mathbf{D} = (\nabla\mathbf{u}+\nabla\mathbf{u}^T)/2$.

The macroscopic behavior of a dilute solution of macromolecules is obtained by averaging a property for a singe molecule over a large ensemble of identical molecules. We then derive an equation for $\overline{\boldsymbol{\sigma}}_B$ which correlates the average microscopic behavior of macromolecules with the macroscopic response of the fluid.

3.2 Nonlinear Stress-Rate of Strain relation

Since we are treating with dilute polymer solutions, the shear viscosity may be considered as being a linear function of the volume fraction only, i.e. $\mu_s(\phi, \dot{\gamma}) \approx \mu(1+c\phi)$. For a dilute aqueous solution of polyacrylamide-PAMA, the best fitting constant is found to be $c = 1.4 \times 10^4$ [2].

Now, the non-Newtonian average contribution stress due to the macromolecules is given by a volume average over a volume V sufficiently large to contain a meaningful number of macromolecules, that is

$$\overline{\boldsymbol{\sigma}}_B = \lim_{V\to\infty} \frac{1}{V} \int_0^V \boldsymbol{\sigma}_B d\mathbf{x} = n\langle \boldsymbol{\sigma}_B \rangle, \tag{6}$$

where $\langle\rangle$ represents an ensemble average.

According to the scaling analysis presented in §1, the elastic restoring force can be written as

$$\mathbf{F_B} = G\mathbf{r}, \tag{7}$$

and the associated stress tensor for one macromolecule is given by the dyadic $\boldsymbol{\sigma}_B = G\mathbf{rr}$. Taking the average over N_m macromolecules within a sufficiently large volume V, one obtains

$$\langle \boldsymbol{\sigma}_B \rangle = G \left(\frac{1}{N_m} \sum_{s=1}^{N_m} \mathbf{rr} \right). \tag{8}$$

Since $n = N_m/V$, $\overline{\boldsymbol{\sigma}}_B = n\langle \boldsymbol{\sigma}_B \rangle = nG\langle \mathbf{rr} \rangle$.

We define the conformation tensor or the moment of inertia tensor of the deformable macromolecule as being

$$\boldsymbol{B}(t) = \langle \mathbf{rr} \rangle = \frac{1}{N_m} \sum_{s=1}^{N_m} \mathbf{rr}. \tag{9}$$

Note that $\boldsymbol{B}$ is, by definition, a symmetric and positive definite tensor.

Rewriting Eq. (5) in terms of the conformation tensor leads to:

$$\boldsymbol{\sigma} = 2\mu_s(\phi)\mathbf{D} + nG\boldsymbol{B}(t). \tag{10}$$

3.3 Conformation Time Evolution

The closure problem of the constitutive equation, Eq. (10), requires a time evolution equation for the conformation tensor $\boldsymbol{B}(t)$. Turning back to the scaling analysis given in §(2), the balance between elastic and viscous forces gives

$$6\pi\mu a \frac{d\mathbf{r}}{dt} + G\mathbf{r} = 0. \tag{11}$$

Now, multiplying both sides of Eq. (11) by $\mathbf{r}$, subtracting the rigid body translation and adopting a frame of reference rotating and deforming with the macromolecule, after averaging, one obtains:

$$3\pi\mu a\frac{\delta \mathbf{B}}{\delta t} + G\mathbf{B} - KT\mathbf{I} = 0. \tag{12}$$

Here $\delta/\delta t$ is the upper convective time derivative or the Oldroyd derivative that is material frame indifferent. It is simply the rate of change of $\mathbf{B}$ seen by an observer translating and deforming with the macromolecule

$$\frac{\delta \mathbf{B}}{\delta t} = \frac{D\boldsymbol{B}}{Dt} - \nabla \boldsymbol{u}\cdot\boldsymbol{B} - \boldsymbol{B}\cdot\nabla \boldsymbol{u}^T, \tag{13}$$

Now, writing Eq. (12) in terms of the Oldroyd derivative given by Eq. (13) and after rearranging the equation, we find

$$\frac{D\boldsymbol{B}}{Dt} = \nabla \boldsymbol{u}\cdot\boldsymbol{B} + \boldsymbol{B}\cdot\nabla \boldsymbol{u}^T - \frac{2}{\tau}\left[\mathbf{B} - (a^2/3)\mathbf{I}\right], \tag{14}$$

where D/Dt denotes the translational material derivative. Here, the first term on the right-hand side in equation (14) represents the stretching of the macromolecule by the flow and the second term represents the relaxation of the polymer due to Brownian motion.

It is important to note that few algebraic manipulation, taking the Oldroyd derivative of the stress Eq. (10) and using the microstructure equation (12), show that the pair of constitutive equation given by Eqs. (10) and (14), reduces to the most common Oldroyd-B fluid [4], namely:

$$\hat{\boldsymbol{\sigma}} + \lambda\frac{\delta \hat{\boldsymbol{\sigma}}}{\delta t} = 2\mu(\phi)\left(\boldsymbol{D} + \lambda\frac{\delta \boldsymbol{D}}{\delta t}\right), \tag{15}$$

with $\hat{\boldsymbol{\sigma}} = \boldsymbol{\sigma} - nKT\boldsymbol{I}$ and the material constant $\lambda = \tau/2$.

In this work, we will keep the form of a pair of constitutive equations in order to explore the explicit dependence of the microstructure given by the behavior of the second moment tensor $\mathbf{B}$.

3.4 An equivalent non-linear Dumper-and-Spring model.

A dumbbell model with a drag correction and a non-linear spring force which gives a finite limit to the spring extension is considered. A Hookean spring may produce unlimited elongation of the dumbbell. Such a behavior is not only unrealistic but can also lead to numerical difficulties as unlimited values for the stress tensor. These difficulties can, at least partially, be eliminated by limiting the extensibility of the macromolecule [11]. Changing the linear spring law to one with a finite extension, i.e. $Gf(r)$, yields a dumbbell-FENE model, where $f(r)$ is the nonlinear spring law. Actually, the correct spring law for a random-chain model was proposed by [6] as being $f(r) = (\ell/3r)F^{-1}(r/\ell)$, where $F(z) = coth(z) - 1/z$ is the Langevin function. Because this law is difficult to work mathematically, we use instead a simpler law with the same qualitative behavior such as first proposed by [15],

$$f(r) = \frac{l^2}{l^2 - r^2} \quad \text{with} \quad r^2 = tr(\boldsymbol{B}). \tag{16}$$

Note that, as r tends to the rigid fibre limit ℓ, the polymer reaches its limits of maximum extensibility, whereas in the case of small distortions of the macromolecules $r \sim a$ (i.e. randomly-coiled state), $f(r) \sim 1$ (i.e. linear spring law would be appropriated). In addition, the FENE dumbbell model described above needs also to take account of the variation in the hydrodynamic drag for $r \sim \ell$. In this respect, this model is strictly valid only for small values of r. As the polymer expands, the size of the object on which the frictional force of the fluid acts increases. The viscous drag increases with the largest linear dimension and so the size of the beads should increase roughly with $r = tr(\mathbf{B})^{1/2}$ [7].

After incorporating into the model the corrections discussed above, the constitutive equations for the stress tensor and the conformation tensor are written, respectively, as

$$\boldsymbol{\sigma} = 2\mu_s(\phi)\mathbf{D} + nGf(r)\boldsymbol{B}(t), \tag{17}$$

$$\frac{d\boldsymbol{B}}{dt} = \nabla \boldsymbol{u} \cdot \boldsymbol{B} + \boldsymbol{B} \cdot \nabla \boldsymbol{u}^T - \frac{2af(r)}{\tau[tr(\mathbf{B})]^{1/2}} \left[\mathbf{B} - (a^2/3)\mathbf{I}\right]. \tag{18}$$

Eqs. (17) and (18) can be made dimensionless with appropriate scales. Using u_c and a as the reference speed and the reference length respectively, the flow is characterized by the Deborah number $De = 2\pi\mu a^2 u_c/KT$; for a simple shearing motion with shear rate $\dot{\gamma}$ the characteristic velocity is $u_c = a\dot{\gamma}$, whereas for a pure extensional flow with rate of strain $\dot{\epsilon}$, $u_c = a\dot{\epsilon}$. As mentioned before, the other parameters of interest are the particle volume fraction ϕ and the extensibility of the polymer, $L = \ell/a$. The dimensionless constitutive equations expressed in terms of the stress $\widetilde{\boldsymbol{\sigma}}$ and the conformation tensor $\widetilde{\boldsymbol{B}}$ of the polymer are respectively

$$\widetilde{\boldsymbol{\sigma}} = \widetilde{\mu}(\phi)\widetilde{\mathbf{D}} + \frac{9\phi f(R)}{2De}\widetilde{\mathbf{B}}, \tag{19}$$

$$\frac{d\widetilde{\mathbf{B}}}{d\widetilde{t}} = \widetilde{\nabla}\widetilde{\mathbf{u}} \cdot \widetilde{\mathbf{B}} + \widetilde{\mathbf{B}} \cdot (\widetilde{\nabla}\widetilde{\mathbf{u}})^T - \frac{2f(R)}{De[tr(\widetilde{\mathbf{B}})]^{1/2}} \left(\widetilde{\mathbf{B}} - \frac{\mathbf{I}}{3}\right), \tag{20}$$

where $\widetilde{\boldsymbol{\sigma}} = a\boldsymbol{\sigma}/\mu u_c$, $\widetilde{\mu} = \mu_s/\mu$, μ is the solvent viscosity, $\widetilde{\mathbf{B}} = \mathbf{rr}/a^2$, $R = r/a$ and the spring function in terms of the dimensionless quantities R and L is given by $f(R) = L^2/(L^2 - R^2)$. By examining the right-hand side term in equation (20), it becomes clear that for large Deborah, the contribution for the polymer relaxation is negligible compared to the contribution of the first term responsible for the stretching of the polymer. One can see that this is possible only if one considers high relaxation time macromolecule compared to the time scale of the flow. The above pair of constitutive equations should be a reasonable approximation for describing flows of very dilute polymer solutions such as those used in turbulent drag reduction.

4 Results

4.1 Two asymptotic limits of the fluid system

A physical interpretation of the role of the anisotropy and the elasticity on a flow due to the presence of additives can be gained by examining two limiting cases of equation (20) for steady extensional flows. In a steady uniaxial extensional with strain-rate $\dot{\epsilon}$ the velocity field is given by, $\mathbf{u} = (\dot{\epsilon}x_1, -\frac{1}{2}\dot{\epsilon}x_2 - \frac{1}{2}\dot{\epsilon}x_3)$ with the dimensionless extensional viscosity being calculated as

$$\frac{\mu_e}{\mu} = \frac{2\widetilde{\sigma}_{11} - \widetilde{\sigma}_{22} - \widetilde{\sigma}_{33}}{6}. \tag{21}$$

In the above equation, we denote by 1, 2 and 3 the streamwise, transverse and spanwise directions, respectively. The definition for μ_e given in equation (21) is normalized by using Trouton's viscosity which corresponds to the extensional viscosity of a Newtonian fluid experimenting a steady extensional flow, $\mu_e/\mu = 3$.

4.1.1 The elastic regime

In the case of elastic limit, when $f(R) = 1$, $De \ll 1$ and $R \sim 1$, the governing equation (20) of $\mathbf{B}$ evolution reduces simply to

$$\frac{d\widetilde{B}_{11}}{d\widetilde{t}} = 2\left[\left(1 - \frac{1}{De}\right)\widetilde{B}_{11} + \frac{1}{3De}\right], \quad \frac{d\widetilde{B}_{22}}{d\widetilde{t}} = \frac{2}{3De} - \left(1 - \frac{2}{De}\right)\widetilde{B}_{22}, \quad \widetilde{B}_{33} = \widetilde{B}_{22}. \tag{22}$$

The stationary solution of equation (22) gives $B_{11} = (1 - De)^{-1}/3$ and $B_{22} = 2(De + 2)^{-1}/3$. A prediction of the extensional viscosity for this elastic limit is obtained by inserting $\widetilde{B}_{11}$ and $\widetilde{B}_{22}$ into the stress equation (19) and using (21), which yields

$$\mu_e/\mu = \mu_s + \frac{3\phi}{2}(De + 2)^{-1}(1 - De)^{-1}. \tag{23}$$

Thus, at leading order

$$\mu_e/\mu = \mu^* + \frac{3}{8}\phi De, \tag{24}$$

where $\mu^* = \mu_s + 3\phi/4$ represents a modified shear viscosity as a function of the particle volume fraction. The result indicates that in a dilute polymer solution for which $\phi \ll 1$ under weak flow ($De \ll 1$), the Non-Newtonian contribution $O(\phi De)$ on the extensional viscosity is a negligible effect and the behavior of dilute polymer solutions in this regime is approximately Newtonian and so is of less interest.

4.1.2 The anisotropic regime

Provide that $R \sim L \gg 1$, in the steady extensional flow, the extension of the macromolecule may be considered predominantly in the 11 - direction. At high Deborah numbers (i.e. strong

flows, $De \sim 1$), $\widetilde{B}_{11}$ will be of order L^2, whereas $\widetilde{B}_{22}$ and $\widetilde{B}_{33}$ will be of order one. Making the approximations $R^2 = tr(\widetilde{\mathbf{B}}) \approx \widetilde{B}_{11}$ and $\widetilde{\nabla}\widetilde{\mathbf{u}} \cdot \widetilde{\mathbf{B}} = \widetilde{\mathbf{B}} \cdot (\widetilde{\nabla}\widetilde{\mathbf{u}})^T \approx \widetilde{B}_{11}\mathbf{e_1}\mathbf{e_1}$, and taking the trace of equation (20) reduces it to

$$\frac{dR}{dt} = R - \frac{2f(R)}{R^2 De}(R^2 - 1). \tag{25}$$

Define the equilibrium extension of the macromolecule, R_L, as being the saturated value of R for $De \sim 1$. Then R_L satisfies

$$R_L - \frac{2f(R_L)}{R_L^2 De}(R_L^2 - 1) = 0 \tag{26}$$

At equilibrium, the component of the dimensionless non-Newtonian stress in the 11-direction given by equation (19) corresponds to the magnitude of the dimensionless extensional viscosity, namely

$$\frac{\mu_e}{\mu} = \frac{9\phi}{2De} R_L^2 f(R_L) = \frac{9\phi}{2} \frac{R_L^5}{R_L^2 - 1}. \tag{27}$$

The result indicates that, in the limit of highly extended polymer, the extensional viscosity is independent of the polymer relaxation time (i.e. the Deborah number), and when R_L tends to $L \gg 1$ (i.e. the rigid fibre limit) the extensional viscosity is proportional to ϕL^3, having the same scaling predicted by slender body theory for the extensional viscosity of a suspension of rigid rods [3].

Now, we can also use this calculation in order to estimate the Deborah number, De_L, corresponding to the asymptotic limit $R_L \to L$ for a given $L \gg 1$. Writing $R_L = cL$, with c being a constant very close to the unit and inserting this condition into equation (26), we obtain

$$De_L \sim \frac{1}{c(1 - c^2)} L^{-1}. \tag{28}$$

Equation (28) yields $De_L \approx 0.6$ for $c = 0.99$ and a typical value of $L = 80$. Therefore, in a polymer solution for which $\phi \ll 1$ but $\phi L^3 > 1$, the presence of polymer molecules will yield a potentially strong effect due to the anisotropy produced by the highly extended particles in flows with $De \sim 1$.

4.2 Computer simulation results

In this section, we present numerical simulation results for the extensional viscosity as a function of the parameters: Deborah number, polymer volume fraction and the extensibility of the macromolecule. The numerical integration of Eq. (20) was performed by using a fourth order Runge-Kutta scheme. The time step was carefully controlled as a function Deborah number, i.e. $dt = min\left\{10^{-3}, 10^{-2}De\right\}$ to keep the integration accurate. The bulk stress is computed using equation (19).

Figure (2) depicts the response of the dimensionless extensional viscosity to a steady extensional flow for different values of the parameter De, $L = 80$ and $\phi = 200ppm$. It is seen a stationary state of the extensional viscosity for all Deborah number simulated. The insert shows behaviour for values of De corresponding to the elastic regime. The results indicate that, for a typical value of extensibility of a high molecular-weight chains, $L = 80$ corresponds to the anisotropic regime of highly extended molecules. The stationary values of the relative extensional viscosity $\hat{\mu}_e - \mu^*$ are approximately 10^6 order of magnitude greater than its magnitude in the elastic regime for $De = 0.55$.

In contrast, the stationary value of the extensional viscosity shown in the inset of figure (2) for $De = 0.35$ is almost indistinguishable from the corresponding Newtonian value $\mu^* \approx 1$. In this regime of De the polymer molecule does not give reliable stresses because the molecules are only slightly distorted by the flow. This result is supported by the scaling discussed in §(4.1) which has found the polymer contribution to the extensional viscosity $O(\phi De)$ for $R \sim 1 \ll L$ and $\phi \ll 1$. This is a clear indicative that only when the molecular distortion is large will the polymer contribution to the bulk stress become significant, providing a strictly non-Newtonian regime as predicted by the asymptotic limit ($R \to L$) giving by (27).

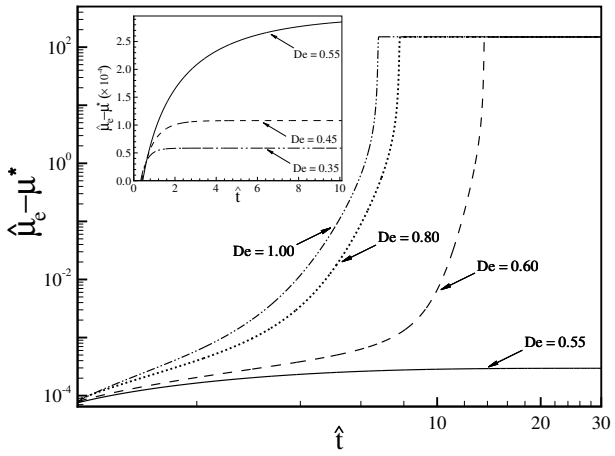

Figure 2: Dimensionless relative extensional viscosity $\hat{\mu}_e - \mu^*$ as a function of the dimensionless time for Deborah ranging from 0.35 to 1, with $L = 80$ and $\phi = 200ppm$. The insert show the behavior of the extensional viscosity for the elastic limit (weak flows).

Figure (3) shows the dimensionless extensional viscosity as a function of Deborah number for $L = 3.5, 15, 25, 35$ and a particle volume fraction $\phi = 200ppm$. The numerical integration results show that for $De < 0.5$ the dimensionless extensional viscosity depends on De and is only weakly affected by L. The insert shows a comparison between the numerical results and the elastic asymptotic solution given in (23) (solid curve) for the elastic behaviour of the extensional viscosity with $De < 0.15$. The straight dashed line is strictly valid for $De \to 0$. For $De \sim 0.1$, the entire elastic behavior of the extensional viscosity is accurately described by the

approximation (23).

According to the analysis presented in §(4.1), the leading order dependence of $\hat{\mu}_e$ is $(3/8)\phi De$. We see, however, that the accuracy of this linear approximation is reasonable for values of $De < 0.05$. The deviation of the elastic approximation shown in the insert of figure (3) for increasing De from the numerical results are a manifestation of the transition of the anisotropy caused by the extended molecules, clearly visible in (3) for $D > 0.5$.

The result changes completely when the molecules are highly extended. In this case, the anisotropy, which is always a very small contribution to the flow in the elastic regime, becomes the dominant mechanism and the molecules behaves nearly as long fibres. In addition, the plot shows that, for smaller L corresponding to polymer of low molecular-weight, the anisotropic regime is gradually attained, whereas for higher L the extensional viscosity jumps from values $O(1)$ to $O(10^2)$ for a typical $L = 50$. The transition to the anisotropic regime of the flow is observed for $De \approx 0.6$ at all L simulated. In this regime, the extensional viscosity is independent of De and so achieved uniform values which depends only on L such as predicted by the asymptotic formula (27). This behavior of the extensional viscosity determined from the simulations are in qualitative agreement with experiments carried out by [12].

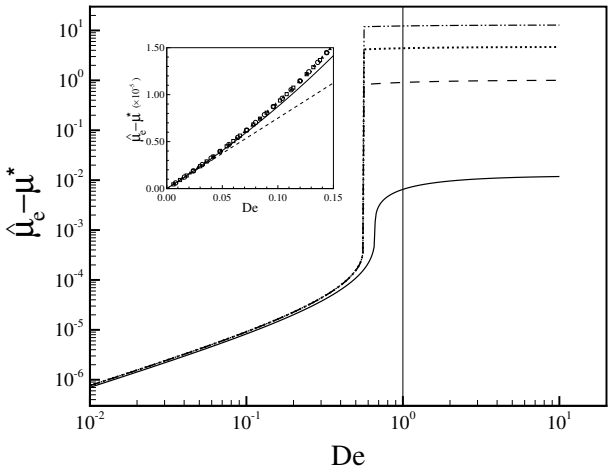

Figure 3: Dimensionless relative extensional viscosity $\hat{\mu}_e - \mu^*$ as a function Deborah number for $L = 3.5$ (continuous line),15 (dashed line),25 (dotted line) ,35 (dashed-dotted line). A comparison between analytical predictions and numerical simulation for the elastic regime is plotted in the insert. Solid line represents the solution (23) and the dashed line the leading order approximation, $(3/8)\phi De$.

The dimensionless relative extensional viscosity as a function of anisotropic parameter ϕL^3 for Deborah numbers ranging from $De = 0.6$ to $De = 2.0$ is presented in Figure (4). Several values of L were computed with $\phi = 200ppm, 250ppm, 300ppm, 350ppm$ and $400ppm$. The observed linear ϕL^3 dependence of the extensional viscosity was previously shown by Batchelor's theory of elongated particles suspended in a Newtonian solvent (Batchelor, 1970) for rigid rods. The

asymptotic limit corresponding to the macromolecule conformation $R \to L$ for strong flows was described in §(4.1). In the case of very dilute solutions considered here, the observed linear dependence between μ_e and ϕL^3 shown in (4) may be attributed to the anisotropy introduced by the molecule long-fibre shape like, corresponding to the fully extended molecule configurations by the flow. A final important result is shown in the insert of figure (4). It is seen that for $De \sim 1$, variations in the extensional viscosity with De are not noticeable and all results collapse to the straight line $(9/2)\phi L^3$, according to (27). Since re-orientation of the extended molecules induced by hydrodynamic interactions are extremely small in very dilute suspension and polymer relaxation at the time scale of a strong flow has a negligible effect when $De \sim 1$, we speculate that the anisotropy may be the key factor behind the observed attenuation of flow instabilities in the presence of such additives. In addition, there is certainly the possibility that, even for very dilute polymer solutions such as those used in turbulent drag, reduction of the polymer contribution to the total stress should be a strong contribution to the flow compared to the contribution from the solvent, if the molecules are in the anisotropic regime $R \to L$. Experimental studies are unavailable for a quantitative comparison with the predictions presented in this article.

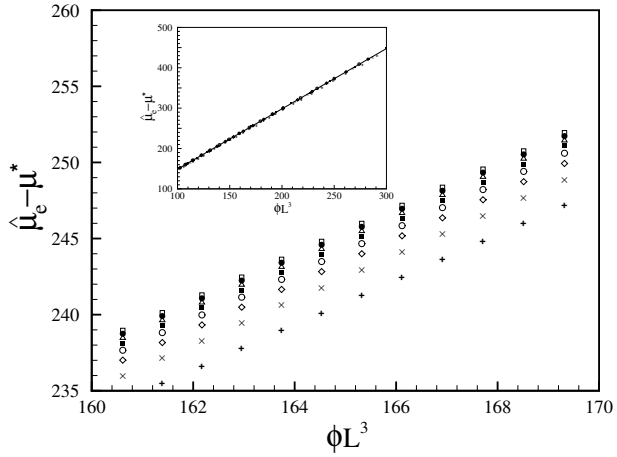

Figure 4: Dimensionless relative extensional viscosity $\hat{\mu}_e - \mu^*$ as a function of the anisotropic parameter ϕL^3, for different values of De. The values of concentration used were 200, 250, 300, 350 and 400ppm with L ranging form 70 to 80. Points + $De = 0.6$; × $De = 0.8$; open diamonds: $De = 1.0$; open circles $De = 1.2$; solid squares: $De = 1.4$; open triangle $De = 1.6$; solid circles: $De = 1.8$; open squares: $De = 2.0$. The fully anisotropic regime for 5 values of $De > 1$ is shown in the insert.

5 Final remarks

In this article, we have generated a pair of constitutive equations to describe anisotropic fluids through scaling arguments based on a balance between viscous drag and restoring Brownian

forces. The physics involved in the constitutive equations of elastic liquids has been better understood. We have shown theoretical results for extensional viscosity in terms of Deborah number De, the volume fraction ϕ and the extensibility of the polymer L. The studies have revealed that, once Deborah number reaches an critical value around $De_c = 1$, the macromolecules behave closely to rigid fibres capturing the same scaling given by viscous slender body theory of rigid rods. That is $\mu_\epsilon/\mu \sim= \phi L^3$, regardless of Deborah number.

In this nonlinear flow regime, the macromolecule elasticity becomes irrelevant and the flow depends mainly on the anisotropy produced by the alignment of the highly stretched macromolecules with the flow. The present studies have suggested that this anisotropy should greatly dominates retardation or elastic effect in strong flows like turbulence. Thus, close to the rigid rod limit, the anisotropy produced by the extended macromolecule would be the key mechanism to explain dilute polymer solution yielding turbulent drag reduction.

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