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EFFECT OF INTERNAL VISCOSITY OF POLYMERIC FLUIDS UNDER STRONG EXTENSIONAL FLOWS*

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Abstract The dumbbell model with internal viscosity for a dilute polymer solution is investigated based on a balance of viscous drag and restoring Brownian forces. An approximate method is used to obtain the solution of extensional stress in closed form in the case of steady flow. For different internal viscosities, this parametric study shows different asymptotic regimes of the extensional viscosity as a function of strain rate. This analysis may explain the attenuation of pressure drop in strong flows from a phenomenological point of view.

Keywords: Polymeric fluid; Internal viscosity; Extensional viscosity.

INTRODUCTION

Extensional strains are involved in many industrial processes, such as extrusion of polymer melts and coatings and drag reduction of polymer solutions. It is known that small amounts of polymer can enhance the extensional viscosity of the solution while just slightly affecting the shear viscosity. The extensional viscosity of dilute polymer solutions can reduce the drag in turbulent pipe flows[1, 2]. Recently, the technique of filament stretching at constant extension rates has been introduced by Tirtaatmadja and Sridhar[3], made possible of extensional viscosity measurements.

The simplest, albeit useful in applications, model for polymer solutions is the Hookean dumbbell model proposed by Kuhn[4], where a polymer molecule in dilute solution is modeled by two beads connected by a spring force. The mathematical simplification of this model has contributed substantially to developing constitutive equations and to investigating the solutions of polymer fluid dynamics problems[5, 6]. To match the empirical results, a few additions have been incorporated into the standard dumbbell model. Finitely extensible nonlinear elastic (FENE) property and internal viscosity of the spring are among them. For both cases, the governing equations of conformation tensors are nonlinear and have no closed form solutions without approximations. Brownian dynamic simulation has been used widely in the study of dynamics of polymers for the FENE dumbbell model[7−9] and dumbbell model with internal viscosity[10−12]. Analytical methodologies make the explanation of the physical phenomena more straightforward. The authors have investigated the effect of internal viscosity in the dumbbell model for polymers in shear flow by numerical method[13] and analytical method[14]. The main idea we pursue in this contribution is to apply the ensemble averaging method to some non-second moment terms and to obtain closed form solutions for polymers in extensional flow. In this paper, a new approximation scheme is proposed to solve the dumbbell model analytically. The forth moment tensor is approximated by an expression of the second moment tensor to obtain a set of nonlinear algebraic equations. Based on the analytical solutions, the extensional viscosity of the polymeric fluid in steady extensional flow is

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discussed.

MODEL DESCRIPTION AND APPROXIMATIONS

For the polymers of bead-spring-bead dumbbell model in a Newtonian solvent with viscosity η_s considered here, we assume that there is no interaction between the beads. Let us denote viscous drag coefficient due to the resistance of the flow by ζ. For the dumbbell model with internal viscosity (IV), the spring force is a function of the configuration vector and configuration velocity. A force law can be expressed in the following form:

$$F(\boldsymbol{Q},\dot{\boldsymbol{Q}}) = H\boldsymbol{Q} + K\left(\frac{\boldsymbol{Q}\otimes\boldsymbol{Q}}{Q^2}\right)\dot{\boldsymbol{Q}} \tag{1}$$

where Q is the length of vector $\boldsymbol{Q}$, H is the spring coefficient of the dumbbell model and K is a constant denoting the measurement of the IV. Since the dot means differentiation with respect to time t, $\dot{\boldsymbol{Q}}$ means the velocity vector of the dumbbells. Substituting Eq. (1) and the equation of motion of one bead into the continuity equation

$$\frac{\partial\psi}{\partial t} = -\frac{\partial}{\partial\boldsymbol{Q}}\cdot\dot{\boldsymbol{Q}}\psi\ , \tag{2}$$

the diffusion equation is derived

$$\frac{\partial\psi}{\partial t} = -\frac{\partial}{\partial\boldsymbol{Q}}\cdot\left\{\left(\boldsymbol{\delta} - g\left\langle\frac{\boldsymbol{Q}\otimes\boldsymbol{Q}}{Q^2}\right\rangle\right)\cdot\left([\boldsymbol{\kappa}\cdot\boldsymbol{Q}]\psi - \frac{2kT}{\zeta}\frac{\partial\psi}{\partial\boldsymbol{Q}} - \frac{2H}{\zeta}\boldsymbol{Q}\right)\right\}, \tag{3}$$

where $\boldsymbol{\delta}$ is unit matrix, $g = 2\varepsilon/(1 + 2\varepsilon)$ and ε is the relative internal viscosity, $\varepsilon = K/\zeta$, which ranges from zero to infinity. For $g = 0$, Eq. (3) recovers the form of the diffusion equation for Hookean dumbbells without IV.

The second moment conformation tensor $\langle\boldsymbol{Q}\otimes\boldsymbol{Q}\rangle$ is of the most interest when calculating the stress tensor. The governing equation of conformation tensor can be developed by multiplying the diffusion equation by the dyadic product $\boldsymbol{Q}\otimes\boldsymbol{Q}$ and integrating over the entire configuration space:

$$\langle\boldsymbol{Q}\otimes\boldsymbol{Q}\rangle_{(1)} = \frac{4kT}{\zeta}\left(\boldsymbol{\delta} - 3g\left\langle\frac{\boldsymbol{Q}\otimes\boldsymbol{Q}}{Q^2}\right\rangle\right) - \frac{4H}{\zeta}(1-g)\langle\boldsymbol{Q}\otimes\boldsymbol{Q}\rangle - 2g\boldsymbol{\kappa}:\left\langle\frac{\boldsymbol{Q}\otimes\boldsymbol{Q}\otimes\boldsymbol{Q}\otimes\boldsymbol{Q}}{Q^2}\right\rangle \tag{4}$$

The subscript (1) denotes convected derivative. In homogeneous flows the convected derivative is defined as

$$\boldsymbol{A}_{(1)} = \frac{\partial}{\partial t}\boldsymbol{A} - \left\{\boldsymbol{\kappa}\cdot\boldsymbol{A} + \boldsymbol{A}\cdot\boldsymbol{\kappa}^T\right\}. \tag{5}$$

We can not calculate the second moment tensor $\langle\boldsymbol{Q}\otimes\boldsymbol{Q}\rangle$ because there are other moment terms, *e.g.*, $\langle(\boldsymbol{Q}\otimes\boldsymbol{Q})/Q^2\rangle$ and $\langle\boldsymbol{Q}\otimes\boldsymbol{Q}\otimes\boldsymbol{Q}\otimes\boldsymbol{Q}\rangle$. In order to put the governing equation into solvable form, the high order terms can be approximated as follows,

$$\left\langle\frac{\boldsymbol{Q}\otimes\boldsymbol{Q}}{Q^2}\right\rangle = \frac{\langle\boldsymbol{Q}\otimes\boldsymbol{Q}\rangle}{\langle Q^2\rangle_{\text{eq}}}, \tag{6}$$

$$\left\langle\frac{\boldsymbol{Q}\otimes\boldsymbol{Q}\otimes\boldsymbol{Q}\otimes\boldsymbol{Q}}{Q^2}\right\rangle = \frac{\langle\boldsymbol{Q}\otimes\boldsymbol{Q}\rangle\otimes\langle\boldsymbol{Q}\otimes\boldsymbol{Q}\rangle}{\langle Q^2\rangle_{\text{eq}}}. \tag{7}$$

These two equations are key approximations to make the governing equation analytically solvable. Equation (6) is similar to the Perterlin approximation used in FENE dumbbell model. Using the approximation Eqs. (6) and (7), we cast the governing Eq. (2) into:

$$\langle \boldsymbol{Q}\otimes\boldsymbol{Q}\rangle_{(1)} = \frac{4kT}{\zeta}\left(\boldsymbol{\delta}-3g\frac{\langle \boldsymbol{Q}\otimes\boldsymbol{Q}\rangle}{\left\langle Q^2\right\rangle_{\text{eq}}}\right)-\frac{4H}{\zeta}(1-g)\langle \boldsymbol{Q}\otimes\boldsymbol{Q}\rangle-2g\boldsymbol{\kappa}:\frac{\langle \boldsymbol{Q}\otimes\boldsymbol{Q}\rangle\otimes\langle \boldsymbol{Q}\otimes\boldsymbol{Q}\rangle}{\left\langle Q^2\right\rangle_{\text{eq}}} \tag{8}$$

Equation (8) is a nonlinear algebraic equation of <$\boldsymbol{Q}\otimes\boldsymbol{Q}$> if all the time-dependent terms are neglected in the steady state flow case. We will seek a closed form solution to this governing equation, followed by the extensional viscosity discussion in the case of steady extensional flow.

Now we consider the steady extensional flow with velocity vector

$$\boldsymbol{v}=\left(v_x, v_y, v_z\right)=\left(-\frac{1}{2},-\frac{1}{2},1\right)\dot{\varepsilon} \tag{9}$$

where $\dot{\varepsilon}$ is the extensional rate. The transpose of velocity vector gradient is

$$\boldsymbol{\kappa}=\left(\nabla\boldsymbol{v}\right)^T=\begin{pmatrix}-\frac{1}{2} & 0 & 0\\ 0 & -\frac{1}{2} & 0\\ 0 & 0 & 1\end{pmatrix}\dot{\varepsilon} \tag{10}$$

The average value of the square of the end-to-end distance in equilibrium for the Hookean dumbbell model in silent flow is (see p73 in Ref. [5])

$$\left\langle Q^2\right\rangle_{\text{eq}}=\frac{3kT}{H} \tag{11}$$

For convenience, we introduce notation for the conformation tensor

$$\boldsymbol{A}=\begin{pmatrix}A_{xx} & A_{xy} & A_{xz}\\ A_{yx} & A_{yy} & A_{yz}\\ A_{zx} & A_{zy} & A_{zz}\end{pmatrix}=\langle \boldsymbol{Q}\otimes\boldsymbol{Q}\rangle \tag{12}$$

and its convected differentiation in steady state where the time-dependent term is neglected in Eq. (5)

$$\boldsymbol{A}_{(1)}=-\left\{\boldsymbol{\kappa}\cdot\boldsymbol{A}+\boldsymbol{A}\cdot\boldsymbol{\kappa}^T\right\} \tag{13}$$

Substituting Eqs. (11)–(13) into (8) yields

$$\left\{\boldsymbol{\kappa}\cdot\boldsymbol{A}+\boldsymbol{A}\cdot\boldsymbol{\kappa}^T\right\}=\frac{4kT}{\zeta}\boldsymbol{\delta}-\frac{4H}{\zeta}\boldsymbol{A}-2gH\boldsymbol{\kappa}:\frac{\boldsymbol{A}\otimes\boldsymbol{A}}{3kT} \tag{14}$$

Using Eq. (8), we can cast Eq. (14) into matrix form:

$$-\dot{\varepsilon}\begin{pmatrix}-A_{xx} & -A_{xy} & \frac{1}{2}A_{xz}\\ -A_{yx} & -A_{yy} & \frac{1}{2}A_{yz}\\ \frac{1}{2}A_{zx} & \frac{1}{2}A_{zy} & 2A_{zz}\end{pmatrix}+\frac{4H}{\zeta}\begin{pmatrix}A_{xx} & A_{xy} & A_{xz}\\ A_{yx} & A_{yy} & A_{yz}\\ A_{zx} & A_{zy} & A_{zz}\end{pmatrix}=\frac{gH\dot{\varepsilon}}{3kT}\begin{pmatrix}B_{xx} & B_{xy} & B_{xz}\\ B_{yx} & B_{yy} & B_{yz}\\ B_{zx} & B_{zy} & B_{zz}\end{pmatrix}+\frac{4kT}{\zeta}\boldsymbol{\delta} \tag{15}$$

where $B_{ij}=A_{ix}A_{xj}+A_{iy}A_{yj}-2A_{iz}A_{zj}$, $i, j = x, y, z$. Equating every element in Eq. (15) and taking into account that tensor $\boldsymbol{A}$ is symmetric and positive definite we can get one set of the solutions:

$$A_{xy}=A_{xz}=A_{yz}=0 \tag{16}$$

$$A_{xx} = A_{yy} = \frac{3 + 3\dot{\varepsilon}\lambda_H - \sqrt{(3 + 3\dot{\varepsilon}\lambda_H)^2 - 12g\dot{\varepsilon}\lambda_H}}{2g\dot{\varepsilon}\lambda_H} \cdot \frac{kT}{H} \tag{17}$$

$$A_{zz} = \frac{3\dot{\varepsilon}\lambda_H - \frac{3}{2} - \sqrt{\left(\frac{3}{2} - 3\dot{\varepsilon}\lambda_H\right)^2 + 6g\dot{\varepsilon}\lambda_H}}{2g\varepsilon\lambda_H} \cdot \frac{kT}{H} \tag{18}$$

For convenience, we can use the Kramers equation of stress tensor for spring model

$$\tau_p = -nH\langle \boldsymbol{Q} \otimes \boldsymbol{Q} \rangle + nkT\boldsymbol{\delta} \tag{19}$$

In this model the stress tensor does depend on the internal viscosity force, which has been involved in the governing equation of conformation tensor, as indicated by Wedgewood [12]. Substituting Eqs. (16–18) into (19), we obtain the extensional stress tensor components as:

$$\tau_{xx} = \tau_{yy} = -nkT \frac{3 + 3\dot{\varepsilon}\lambda_H - 2g\dot{\varepsilon}\lambda_H - \sqrt{(3 + 3\dot{\varepsilon}\lambda_H)^2 - 12g\dot{\varepsilon}\lambda_H}}{2g\dot{\varepsilon}\lambda_H} \tag{20}$$

$$\tau_{zz} = -nkT \frac{3\dot{\varepsilon}\lambda_H - \frac{3}{2} - 2g\dot{\varepsilon}\lambda_H - \sqrt{\left(\frac{3}{2} - 3\dot{\varepsilon}\lambda_H\right)^2 + 6g\dot{\varepsilon}\lambda_H}}{2g\varepsilon\lambda_H} \tag{21}$$

where shear stresses vanish. The average value of the square of end-to-end extension distance of the polymers can be obtained by the trace of the deformation tensor $\boldsymbol{A}$:

$$\langle Q^2 \rangle = \mathrm{trace}\boldsymbol{A} \tag{22}$$

by using Eqs. (17) and (18). In the steady uniaxial extensional flow with strain-rate $\dot{\varepsilon}$, the extensional viscosity can be calculated as Ref. [3]

$$\mu_e = \frac{2\tau_{zz} - \tau_{xx} - \tau_{yy}}{6} \tag{23}$$

which can be evaluated by Eqs. (20) and (21).

EXAMPLES AND REMARKS

Based on Eqs. (22) and (23), we can calculate the square of end-to-end extension distance of the polymers and the extensional viscosity for different internal viscosities as presented in Fig. 1. With the increase of the extension rates, the end-to-end distance of the polymers and the extensional viscosity reach a plateau asymptotically. This result can explain the attenuation of the pressure drop in strong extensional flows. Both plots show that the contribution of the internal viscosity is more drastic when the flow is stronger. However on the other hand it can be seen from the figures that the plateau phenomenon becomes less obvious with increase of internal viscosity. The non-equilibrium state must be considered for better results. The plots have good agreement with experimental result, qualitatively, before the strain hardening happens[15].

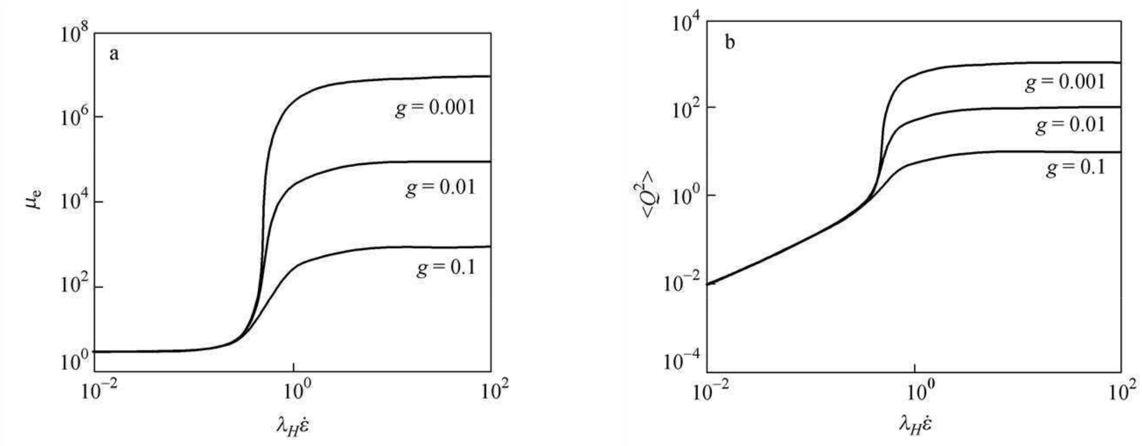

Fig. 1 The contribution of internal viscosity
a) The viscosity coefficient; b) The end-to-end distance

In summary, we have proposed an approximate approach for obtaining the closed form solution of the governing equation of extension tensor and hence the closed form of stress tensor. The contribution of internal viscosity to the extensional viscosity has been demonstrated.

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