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# A New Constitutive Model for the Analysis of Semi-flexible Polymers with Internal Viscosity

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**Abstract.** The analysis of dynamics of semi-flexible polymers, such as DNA molecules, is an important multiscale problem with a wide range of applications in science and bioengineering. In this contribution, we show how accounting for internal viscosity in dumbbell-type models may render physically plausible results with minimal computational cost. We focus our attention on the cases of steady shear and extensional flows of polymeric solutions. First, the tensors with moments other than the second order moment are approximated. Then, the nonlinear algebraic equation for the second moment conformation tensor is solved. Finally, substituting the resulting conformation tensor into the Kramers equation of Hookean spring force, the constitutive equations for the model are obtained. The shear material properties are discussed in the context of different internal viscosities and our computational results are compared with the results of other methods applicable for high shear or extensional rates.

**Keywords:** Polymeric fluid; Dumbbell model; Internal viscosity.

## 1 Introduction

The dynamics of polymeric fluids is an important multiple time scale problem, involving the quick speed of the small solvent molecules, fast movement of atomic particles that constitute the polymers and slow orientation of the polymer deformation [1]. Brownian dynamic simulation is the technique taking into account all the motions in the polymeric fluids. Coarse-grained models are often used in the analysis of the rheological properties of polymeric fluids. From the mesoscopic view, the fast movements of the atomic particles of the polymers are neglected in order to relieve the complexity. In the investigation of the mathematical problems for the coarse-grained models, multiscale method is also an efficient technique to obtain the approximate solutions [2].

The simplest, albeit extremely useful in applications, model for polymer solutions is the Hookean dumbbell model proposed by Kuhn [3], where a polymer molecule in dilute solution is represented by two beads connected by a spring

force. This mathematical simplification has contributed a lot to the development of constitutive models for investigating properties of polymeric fluids with dumbbell-type models [14,15]. In order to obtain a better agreement with experimental results, a few additions have been incorporated into the standard dumbbell model. Of particular importance for our further discussion are two of them: finitely extensible nonlinear elastic (FENE) property and internal viscosity of the spring. Taking into account these properties leads to a situation where the governing equations of conformation tensors become nonlinear and the resulting problem has no closed form solution unless an appropriate approximation is made.

Brownian dynamic simulations have been used widely in applications of the FENE dumbbell model to the analysis of fluids and semi-flexible polymers. The results obtained on the basis of such simulations are often more accurate compared to approximate theoretical methodologies discussed in [6,7,8]. Nevertheless, Brownian dynamic simulations are time consuming and do not render a straightforward technique for the explanation of the underlying physical properties of fluids and polymers. A consequence of this is intensive research efforts in developing semi-analytical methodologies which should be combined with computational experiments in order to shed further light on such properties. From a mathematical point of view, these efforts are reducible to the construction of some closure forms for the equation written with respect to the conformation tensor in such a way that the governing constitutive equations for the system (after certain approximations) become analytically solvable. The main idea is to apply an ensemble averaging methodology to non-second moment terms so that closed form solutions could be obtained [2,9,10]. However, for the dumbbell model with internal viscosity there is a forth moment tensor in the governing equation which makes it difficult to find the closed form solutions. Therefore, a numerical methodology should be developed to simulate the dynamics of the polymeric system. In the context of rheological properties of complex fluids, this issue was previously discussed by a number of authors (e.g., [11,15]). Booij and Wiechen [12] used a perturbation solution approach to calculate the first order approximation of the internal viscosity parameter. More recently, based on the Gaussian closure method, the second moment tensor and higher moment tensors have been calculated by integration over the conformation distribution space [13,14,15].

In this contribution, a new approximation scheme is proposed to solve the governing equations analytically. The forth moment tensor is approximated by an expression involving the second moment tensor in order to obtain a set of nonlinear algebraic equations. Based on the analytical solutions of such equations, the material properties of the polymeric fluids in steady-state shear flows and extensional viscosities in extensional flows are discussed. The phenomena of shear thinning and attenuation of pressure drop have been found and the results have been compared to the results obtained by Brownian dynamics simulations and the Gaussian closure method in the context of high shear or extensional rates. Our results can explain the phenomenon of shear thinning by introducing the

internal viscosity and obtaining better predictions compared to the traditional technique.

## 2 The Governing Equations

For the polymers in a Newtonian solvent with viscosity  $\eta_s$  described with the bead-spring-bead dumbbell model it is assumed that there is no interaction between the beads. Let us denote the viscous drag coefficient due to the resistance of the flow by  $\zeta$ . For the dumbbell model with internal viscosity (IV), the spring force is a function of the configuration vector,  $\mathbf{Q}$ , and configuration velocity  $\dot{\mathbf{Q}}$ . The force law in this case can be expressed as:

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

where  $Q$  is the length of vector  $\mathbf{Q}$ ,  $H$  is the spring coefficient of the dumbbell model and  $K$  is a constant denoting the measurement of the IV. The dot indicates differentiation with respect to time  $t$ , so that  $\dot{\mathbf{Q}}$  represents the velocity vector of the dumbbells. By substituting equation (1) and the equation of motion of one bead into the continuity equation [1]

$$\frac{\partial \psi}{\partial t} = -\frac{\partial}{\partial \mathbf{Q}} \cdot \dot{\mathbf{Q}} \psi, \quad (2)$$

we can derive the diffusion equation

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

where  $\delta$  is a unit matrix,  $g = 2\varepsilon/(1+2\varepsilon)$ , and  $\varepsilon$  is the relative internal viscosity. Note that  $\varepsilon = K/2\zeta$  and it can formally range from zero to infinity. Furthermore, for  $g = 0$ , equation (3) recovers the form of the diffusion equation for Hookean dumbbells without IV.

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

$$\begin{aligned} \langle \mathbf{Q} \otimes \mathbf{Q} \rangle_{(1)} = & \frac{4kT}{\zeta} \left( \delta - 3g \left\langle \frac{\mathbf{Q} \otimes \mathbf{Q}}{Q^2} \right\rangle \right) - \frac{4H}{\zeta} (1-g) \langle \mathbf{Q} \otimes \mathbf{Q} \rangle \\ & - 2g\kappa : \left\langle \frac{\mathbf{Q} \otimes \mathbf{Q} \otimes \mathbf{Q} \otimes \mathbf{Q}}{Q^2} \right\rangle. \end{aligned} \quad (4)$$

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

$$\mathbf{A}_{(1)} = \frac{\partial}{\partial t} \mathbf{A} - \{ \kappa \cdot \mathbf{A} + \mathbf{A} \cdot \kappa^T \}. \quad (5)$$

Unfortunately, it is not possible to calculate the second moment tensor  $\langle \mathbf{Q} \otimes \mathbf{Q} \rangle$  directly because there are other moment terms, e.g.,  $\left\langle \frac{\mathbf{Q} \otimes \mathbf{Q}}{Q^2} \right\rangle$  and  $\langle \mathbf{Q} \otimes \mathbf{Q} \otimes \mathbf{Q} \otimes \mathbf{Q} \rangle$ . Hence, in order to cast the governing equation into a form amenable to the analytical solution, the higher order terms should be approximated. This is done as follows:

$$\left\langle \frac{\mathbf{Q} \otimes \mathbf{Q}}{Q^2} \right\rangle \approx \frac{\langle \mathbf{Q} \otimes \mathbf{Q} \rangle}{\langle Q^2 \rangle_{eq}}, \quad (6)$$

$$\left\langle \frac{\mathbf{Q} \otimes \mathbf{Q} \otimes \mathbf{Q} \otimes \mathbf{Q}}{Q^2} \right\rangle \approx \frac{\langle \mathbf{Q} \otimes \mathbf{Q} \rangle \otimes \langle \mathbf{Q} \otimes \mathbf{Q} \rangle}{\langle Q^2 \rangle_{eq}}. \quad (7)$$

The equations (6), (7) are key approximations allowing us to make the governing equation analytically solvable. Note that equation (6) is similar to the Perterlin approximation used in FENE dumbbell model. By using equations (6) and (7), the governing equation (2) can be cast in the following form:

$$\begin{aligned} \langle \mathbf{Q} \otimes \mathbf{Q} \rangle_{(1)} = & \frac{4kT}{\zeta} \left( \delta - 3g \frac{\langle \mathbf{Q} \otimes \mathbf{Q} \rangle}{\langle Q^2 \rangle_{eq}} \right) - \frac{4H}{\zeta} (1-g) \langle \mathbf{Q} \otimes \mathbf{Q} \rangle \\ & - 2g\kappa : \frac{\langle \mathbf{Q} \otimes \mathbf{Q} \rangle \otimes \langle \mathbf{Q} \otimes \mathbf{Q} \rangle}{\langle Q^2 \rangle_{eq}}. \end{aligned} \quad (8)$$

In the steady state flow case, when all the time-dependent terms can be neglected, equation (8) becomes a nonlinear algebraic equation with respect to  $\langle \mathbf{Q} \otimes \mathbf{Q} \rangle$ . In the next section, we will seek a closed form solution to this governing equation, followed by the material properties discussion in the case of steady state shear flow.

### 3 Results and Examples

#### 3.1 The Material Coefficients in Steady Shear Flows

First, we consider the steady state shear flow with the velocity vector given by

$$\mathbf{v} = (v_x, v_y, v_z) = (\dot{\gamma}y, 0, 0), \quad (9)$$

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

$$\boldsymbol{\kappa} = (\nabla \mathbf{v})^T = \begin{pmatrix} 0 & \dot{\gamma} & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}. \quad (10)$$

The average value of the square of the end-to-end distance in equilibrium in shear flow with shear rate  $\dot{\gamma}$  can be represented as [11]

$$\langle Q^2 \rangle_{eq} = \frac{3kT}{H} + \frac{2kT}{H} (\lambda_H \dot{\gamma})^2, \quad (11)$$



where the time constant  $\lambda_H$  is defined by  $\lambda_H = \zeta/4H$ . For convenience, we use the following notation for the conformation tensor

$$\mathbf{A} = \begin{pmatrix} A_{xx} & A_{xy} & A_{xz} \\ A_{yx} & A_{yy} & A_{yz} \\ A_{zx} & A_{zy} & A_{zz} \end{pmatrix} = \frac{H}{kT} \langle \mathbf{Q} \otimes \mathbf{Q} \rangle, \quad (12)$$

so that its convected differentiation in steady state situations gives

$$\mathbf{A}_{(1)} = -\{\boldsymbol{\kappa} \cdot \mathbf{A} + \mathbf{A} \cdot \boldsymbol{\kappa}^T\}. \quad (13)$$

Substituting equations (11)-(13) into (8) and equating corresponding tensor elements in the result, we conclude that the nonzero elements of  $\mathbf{A}$  can be calculated as follows

$$A_{xx} = \frac{2\lambda_H \dot{\gamma} A_{xy} + 1}{1 - \frac{2(\lambda_H \dot{\gamma})^2 - 2\lambda_H \dot{\gamma} A_{xy}}{3 + 2(\lambda_H \dot{\gamma})^2} g}, \quad A_{yy} = A_{zz} = \frac{1}{1 - \frac{2(\lambda_H \dot{\gamma})^2 - 2\lambda_H \dot{\gamma} A_{xy}}{3 + 2(\lambda_H \dot{\gamma})^2} g}, \quad (14)$$

where  $A_{xy}$  is the real root of

$$\begin{aligned} & \frac{4(\lambda_H \dot{\gamma})^2}{[3 + 2(\lambda_H \dot{\gamma})^2]^2} g^2 A_{xy}^3 + \frac{4\lambda_H \dot{\gamma}}{3 + 2(\lambda_H \dot{\gamma})^2} \left[ 1 - \frac{2(\lambda_H \dot{\gamma})^2}{3 + 2(\lambda_H \dot{\gamma})^2} g \right] A_{xy}^2 \\ & + \left[ 1 - \frac{2(\lambda_H \dot{\gamma})^2}{3 + 2(\lambda_H \dot{\gamma})^2} g \right]^2 A_{xy} - \lambda_H \dot{\gamma} = 0. \end{aligned} \quad (15)$$

For convenience, we use the Kramers equation for the stress tensor in the spring model:

$$\frac{\tau_p}{nkT} = -\frac{H}{kT} \langle \mathbf{Q} \otimes \mathbf{Q} \rangle + \boldsymbol{\delta} = -\mathbf{A} + \boldsymbol{\delta}. \quad (16)$$

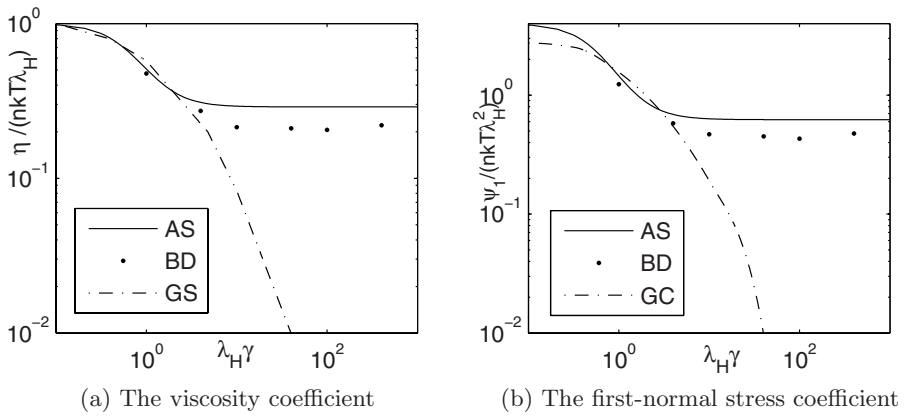
The three material functions of interest, namely, the viscosity  $\eta(\dot{\gamma})$ , the first-normal stress coefficient  $\Psi_1(\dot{\gamma})$ , and the second-normal stress coefficient  $\Psi_2(\dot{\gamma})$  are connected with the stress components by the following relationships:

$$\begin{aligned} \tau_{xy} &= -\eta(\dot{\gamma}) \dot{\gamma}, \\ \tau_{xx} - \tau_{yy} &= -\Psi_1(\dot{\gamma}) \dot{\gamma}^2, \\ \tau_{yy} - \tau_{zz} &= -\Psi_2(\dot{\gamma}) \dot{\gamma}^2. \end{aligned} \quad (17)$$

Substituting equation (16) into (14), we obtain the material coefficients via the following representations:

$$\begin{aligned} \frac{\eta(\dot{\gamma})}{nkT\lambda_H} &= \frac{A_{xy}}{\lambda_H \dot{\gamma}}, \\ \frac{\Psi_1(\dot{\gamma})}{nkT\lambda_H^2} &= \frac{2A_{xy}}{\left[ 1 - \frac{2(\lambda_H \dot{\gamma})^2 - 2\lambda_H \dot{\gamma} A_{xy}}{3 + 2(\lambda_H \dot{\gamma})^2} g \right] \lambda_H \dot{\gamma}}, \\ \Psi_2 &= 0. \end{aligned} \quad (18)$$

Based on equations (15) and (18), we calculate the material properties for different internal viscosities. The plots presented in Figure 1 demonstrate comparison results between our approximate solutions (AS), the results obtained by Brownian dynamics simulations (BD), and the results obtained with the Gaussian closure technique (GC) [15]. The internal viscosity was chosen as  $\varepsilon = 1$ . In Figure 1, the solid lines represent our algebraic solutions to the governing equations for the material coefficients, the dots represent the data obtained by Brownian dynamics simulations, and the dot-dash lines represent the results obtained by the Gaussian closure method. For both viscosity and first-normal stress coefficients, our algebraic solutions exhibit the plateau values appearing also in the case of Brownian dynamics simulations. Observe that compared to the Gaussian closure method, our methodology has a wider range of applicability.



**Fig. 1.** Comparison of analytical results with Brownian dynamics simulations and the Gaussian closure methodology ( $\varepsilon = 1$ )

### 3.2 The Extensional Viscosity in Steady Extensional Flows

Next, we consider the steady extensional flow with the velocity vector given by

$$\mathbf{v} = (v_x, v_y, v_z) = \left(-\frac{1}{2}, -\frac{1}{2}, 1\right) \dot{\varepsilon}, \quad (19)$$

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

$$\kappa = (\nabla \mathbf{v})^T = \begin{pmatrix} -\frac{1}{2} & 0 & 0 \\ 0 & -\frac{1}{2} & 0 \\ 0 & 0 & 1 \end{pmatrix} \dot{\varepsilon}. \quad (20)$$

In the steady uniaxial extensional flow with strain rate  $\dot{\varepsilon}$ , the extensional viscosity is defined as [16]

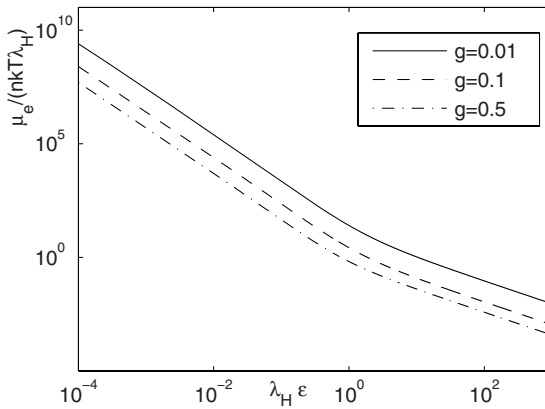
$$\mu_e = \frac{2\tau_{zz} - \tau_{xx} - \tau_{yy}}{6\dot{\varepsilon}}. \quad (21)$$

By using the equation (8), we get the solutions of the equation with respect to the conformation tensor by the same procedure used in the steady state shear flow case. Then the extensional viscosity is calculated as

$$\frac{\mu_e}{nkT\lambda_H} = \frac{2A_{zz} - A_{xx} - A_{yy}}{6\dot{\epsilon}}, \quad (22)$$

where  $A_{xx}$ ,  $A_{yy}$  and  $A_{zz}$  are determined by the following set of algebraic equations:

$$\begin{aligned} (\lambda_H \dot{\epsilon} + 1) A_{xx} + \frac{4}{3} g \lambda_H \dot{\epsilon} (A_{zz} - A_{11}) &= 1, \\ (-2\lambda_H \dot{\epsilon} + 1) A_{xx} + \frac{4}{3} g \lambda_H \dot{\epsilon} (A_{zz} - A_{11}) &= 1, \\ A_{yy} &= A_{xx}. \end{aligned} \quad (23)$$



**Fig. 2.** The viscosity coefficient in the steady extensional flow

Figure 2 demonstrates decrease in extensional viscosity with higher rates of extensional flows. This explains the attenuation of the pressure drop in strong extensional flows.

## 4 Conclusions

In this contribution, we developed a set of approximate semi-analytical solutions for the dumbbell model with IV without integration of the Gaussian distribution. Our concise equations can predict the material coefficients of polymeric fluid well, qualitatively and also quantitatively. The shear thinning phenomena are described well with the new developed model deduced from the dumbbell model with internal viscosity. The effect of internal viscosity in the extensional flow case has also been demonstrated. By comparing our computational results with Brownian dynamic simulations and the Gaussian closure methodology, we demonstrated the efficiency of the proposed approximate technique for a wider range of high shear or extensional flow rates.

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