On the selection of parameters in the FENE-P model

A.P.G. van Heel *, M.A. Hulsen, B.H.A.A. van den Brule

Delft University of Technology, Laboratory for Aero and Hydrodynamics, Rotterdamseweg 145, 2628 AL Delft, The Netherlands

Received 22 May 1997; received in revised form 17 July 1997

Abstract

We compare the FENE and FENE-P models in different flow situations. We start with Brownian dynamics simulations of start-up of shear and uniaxial elongational flow. The FENE-P model predicts the FENE behaviour unsatisfactorily. However, we will show that its performance can be substantially improved. We propose to determine the parameters in the FENE-P model such that important FENE flow characteristics are recovered. The three resulting models (FENE, FENE-P and the proposed FENE-P model) will first be compared in the above-mentioned rheometrical flows. To show that the improvement persists in a complex flow field, we also simulate the flow of the FENE fluid past a cylinder using the Brownian configuration field technique. We then compare the results with those of the proposed FENE-P model and those of the original FENE-P model. © 1998 Elsevier Science B.V. All rights reserved.

Keywords: FENE dumbbell model; Peterlin approximation; Brownian dynamics; Brownian configuration fields

1. Introduction

In studying dilute polymeric solutions, the polymer molecules are often modelled as dumbbells consisting of two beads connected by a spring. In a flowing Newtonian solvent the dumbbells are convected and distorted by the viscous force exerted on the beads by the solvent. In the simplest model, the elastic force between the beads is taken to be proportional to the separation between the beads. This is the so-called Hookean dumbbell model. In addition to the forces mentioned above, the beads experience a randomly fluctuating force due to the thermal agitation by the surrounding solvent molecules. It can be shown that the constitutive equation associated with the Hookean dumbbell model is identical to the macroscopic Oldroyd-B equation. Due to its simplicity the model has some serious drawbacks, the most important being the fact that the shear viscosity is constant and that the dumbbells can be stretched infinitely. In elongational
flow for example, this leads to an unbounded value of the elongational viscosity at high strain rate.

A way to overcome these problems is to replace the Hookean spring by a non-linear spring to limit the dumbbell extension to a maximum value. An important example of such a non-linear spring is the finitely extensible non-linear elastic (FENE) spring introduced by Warner [1]. In this model the connector force $F^c$ takes the following form

$$F^c = \frac{HQ}{1 - Q^2/Q_0^2}, \quad (1)$$

where $Q$ is the connector vector between the beads, $H$ is the spring constant and $Q_0$ denotes the maximum possible spring extension. A major drawback of the FENE model is that it does not yield a closed-form constitutive equation for the polymer stress. For this reason it is not suited for a macroscopic flow calculation. However, if the denominator in the FENE expression for the connector force is replaced by its ensemble averaged value, i.e.

$$F^c = \frac{HQ}{1 - \langle Q^2 \rangle/Q_0^2}, \quad (2)$$

it is possible to close the model. This pre-averaging is known as the Peterlin approximation and the resulting model as the FENE-P model.

In flow simulations the FENE model is often replaced by the FENE-P model because the latter model can be used in macroscopic flow calculations. However, Brownian dynamics simulations of start-up of shear and elongational flow of FENE and FENE-P chains [2] and dumbbells [3,4] demonstrate that the pre-averaging can cause significant changes in rheological behaviour. The results of these rheometrical flows provide us with useful information about the quality of the Peterlin approximation, but they cannot be extrapolated to complex flow fields in which shear and elongational flow occur simultaneously. Indeed, it is only with the introduction of recent techniques like CONFFESSIT [5] and the Brownian Configuration Field method [6], that the FENE model itself can also be used in complex flow calculations, and that the changes in behaviour due to the pre-averaging can be assessed in general flow fields. As an example, we shall compare the FENE and FENE-P models in the complex flow field past a cylinder. The discrepancy in behaviour, observed in shear and elongational flow simulations, is also found to persist in this case. It will be shown for example, that the Peterlin pre-averaging can lead to polymer stresses that are larger than the FENE values by a factor of 1.5. The observed discrepancy may be caused by the difference in important linear viscoelastic properties of the FENE and FENE-P model. For this reason we repeated the flow calculations mentioned above, selecting the parameters in the FENE-P model such that it has the same time constant, the same zero-shear-rate viscosity and the same shear modulus as the FENE model that we want to approximate. The resulting model is investigated both in simple flows like shear and uniaxial elongational flow as well as in the complex flow field past a cylinder. It is found that this FENE-P model predicts the start-up of shear flow of the FENE model much more accurately than the original FENE-P model does. Also in elongational flow for small strains the model is seen to approximate the FENE results better than the FENE-P model. However, for large strains the original FENE-P model appears to be more appropriate. In the complex flow field past a cylinder, the proposed FENE-P model shows good agreement with the results of the FENE model.
2. Basic dumbbell theory and simulation method

The dynamics and material properties of a suspension of dumbbells in a Newtonian solvent can either be obtained using an approach based on configuration distribution functions [7], or equivalently from an approach based on stochastic differential equations for the dumbbell connector vector [8]. In this section we will follow the latter approach. The time evolution of the internal configuration of a FENE dumbbell is governed by the following stochastic differential equation [8]

\[ d\mathbf{Q} = \left[ \mathbf{k} \cdot \mathbf{Q} - \frac{2}{\zeta} \mathbf{F}^c \right] dt + \sqrt{\frac{4kT}{\zeta}} dW(t). \]  

(3)

In this equation \( \mathbf{Q} \) is the connector vector between the beads, \( \mathbf{k} \) is the transpose of the velocity gradient, \( \mathbf{k} = (\nabla \mathbf{v})^T \); \( \zeta \) is the friction coefficient of a bead, \( \mathbf{F}^c \) is the connector force given by Eq. (1), \( t \) is the time, \( k \) is Boltzmann’s constant, \( T \) is the absolute temperature and \( W \) represents a three-dimensional Wiener process. The first term incorporates the distortion of the beads due to the velocity field, the second term represents the effect of the restoring spring force, and the last term models the Brownian motion of the beads.

In the following it is convenient to introduce a new unit of time, viz. \( \zeta / 4H \), which will be abbreviated by \( \lambda_H \), and a new measure of length, \( \sqrt{kT/H} \). Quantities expressed in these units will be accented, thus \( \mathbf{Q} = \mathbf{Q} / (\sqrt{kT/H}) \) and \( t = t / \lambda_H \). Expressed in the new units, the time evolution of the FENE dumbbell becomes

\[ d\hat{\mathbf{Q}} = \left[ \hat{\mathbf{k}} \cdot \hat{\mathbf{Q}} - \frac{1}{2} \hat{\mathbf{F}}^c \right] d\hat{t} + dW(t), \]  

(4)

where \( \hat{\mathbf{k}} = \lambda_H \mathbf{k} \), \( \hat{t} \) denotes the dimensionless time and \( \hat{\mathbf{F}}^c \) is the dimensionless connector force, given by

\[ \hat{\mathbf{F}}^c = \frac{\mathbf{Q}}{1 - \mathbf{Q}^2/b}, \]  

(5)

where \( b \) denotes the finite extensibility parameter \( b = HQ_0^2 / (kT) \). The three-dimensional Wiener process \( W(t) \) is a Gaussian stochastic process and is characterised by its expectation value and variance, i.e.

\[ \langle W(t) \rangle = 0, \]  

(6)

and

\[ \langle W(t_1)W(t_2) \rangle = \min(t_1, t_2) \mathbf{1}. \]  

(7)

The time evolution of the connector vector of a dumbbell is obtained by integrating its stochastic differential equation, Eq. (4). Macroscopic expectation values are determined from ensemble averages. For example, the polymer contribution to the stress, \( \tau^P \), is obtained from the Kramers expression

\[ \tau^P / nkT = \langle \hat{\mathbf{Q}} \hat{\mathbf{F}}^c \rangle - \mathbf{1}, \]  

(8)
where the brackets denote an averaging over the dumbbell configuration distribution function and \( n \) is the number of dumbbells per unit volume. In equilibrium \( \tau^p \) vanishes.

To investigate the change in rheological behaviour due to the Peterlin approximation in simple well-defined flows, similar to the approaches taken in [2–4], we commence with Brownian dynamics simulations of start-up of shear and uniaxial elongational flow. In the simulations we use a large ensemble of \( N \) dumbbells, and for each dumbbell, its stochastic differential equation Eq. (4), is integrated using the Euler forward scheme

\[
\dot{\mathbf{Q}}(\hat{t} + \Delta \hat{t}) = \dot{\mathbf{Q}}(\hat{t}) + \left[ \mathbf{\kappa} \cdot \mathbf{Q}(\hat{t}) - \frac{1}{2} \tilde{\mathbf{F}}(\hat{t}) \right] \Delta \hat{t} + \Delta \mathbf{W}(\hat{t}),
\]

and for the \( \tilde{\mathbf{F}} \) we take either the FENE or FENE-P connector force Eq. (1) or Eq. (2), respectively. Although the Euler forward scheme may lead to dumbbells whose length exceeds the maximum allowable dumbbell length, especially when the time step \( \Delta \hat{t} \) is chosen too large, in our simulations we have never encountered this situation. To avoid the problem of over-extension altogether, one could alternatively use semi-implicit integration schemes [2,8]. The components of the random vector \( \Delta \mathbf{W}(\hat{t}) \) are independent Gaussian random variables with an expectation value 0 and variance \( \Delta \hat{t} \). In analogy with Eq. (8), the polymer contribution to the stress can be obtained from the ensemble average, i.e.

\[
\tau^p/nkT \approx \frac{1}{N} \sum_{i=1}^{N} \dot{\mathbf{Q}} \cdot \tilde{\mathbf{F}}^\varepsilon_i - 1.
\]

In our simulations we use ensembles consisting of 64 000 dumbbells with an extensibility parameter \( b = 50 \) and the stochastic differential equations were integrated using a time step \( \Delta \hat{t} = 10^{-3} \). Unless otherwise specified, all subsequent equations will be expressed in these new units of time and length, but to make the notation less cumbersome the accents will be no longer be used. Thus, the values of the shear and elongation rate given below are the values of the dimensionless shear and elongation rate. The results are shown below.

Fig. 1. Brownian dynamics simulations of the shear stress of FENE (solid line) and FENE-P dumbbells (dash-dot line) after inception of shear flow. The results were obtained using ensembles of 64 000 dumbbells with an extensibility parameter \( b = 50 \). The curves correspond to (dimensionless) shear rates \( \dot{\gamma} = 1 \) (lower curves), \( \dot{\gamma} = 2 \) and \( \dot{\gamma} = 4 \) (upper curves).
Fig. 2. Brownian dynamics simulations of the first normal stress difference of FENE (solid line) and FENE-P dumbbells (dash-dot line) after inception of shear flow. The results were obtained using ensembles of 64,000 dumbbells with an extensibility parameter $b = 50$. The curves correspond to shear rates $\dot{\gamma} = 1$ (lower curves), $\dot{\gamma} = 2$ and $\dot{\gamma} = 4$ (upper curves).

From the results for the shear stress $\tau_{xy}$, shown in Fig. 1 two important observations can be made. Firstly, at the inception of flow, the FENE-P model is seen to underpredict the FENE shear stress, and its overshoot occurs too late and is too high. Secondly, the Peterlin approximation yields higher steady state values. The results of the first normal stress difference, Fig. 2, show that the FENE-P model overpredicts the FENE values by a factor of 1.5 for a (dimensionless) shear rate $\dot{\gamma} = 4$. Fig. 3 shows the evolution of the elongational viscosity $\tilde{\eta} = (\tau_{zz}^p - \tau_{yy}^p)/\dot{\epsilon}$ during start-up of elongational flow, using elongation rates $\dot{\epsilon} = 1, 2$ and 4. It is seen that at an elongation rate $\dot{\epsilon} = 1$ for the FENE-P model overpredicts the FENE steady state plateau value. Moreover, for elongation rates $\dot{\epsilon} = 2$ and $\dot{\epsilon} = 4$ the FENE-P model exhibits an almost instantaneous jump to its plateau value, while the FENE displays a much more gradual increase. In conclusion, both the shear flow and the elongational flow simulations indicate that there are serious differences in the behaviour of the FENE and FENE-P model. This is in agreement with earlier Brownian dynamics simulations [3,4].

Fig. 3. Brownian dynamics simulations of the elongation viscosity of FENE (solid line) and FENE-P dumbbells (dash-dot line) after inception of elongation flow. The results were obtained using ensembles of 64,000 dumbbells with an extensibility parameter $b = 50$. The curves correspond to (dimensionless) elongation rates $\dot{\epsilon} = 1$ (lower curves), $\dot{\epsilon} = 2$ and $\dot{\epsilon} = 4$ (upper curves).
3. Calculation of the FENE shear modulus

When a viscoelastic material is subjected to a very small deformation, so as to remain in the linear regime, that is also applied fast enough to neglect relaxation phenomena during the deformation process itself, the material essentially behaves like a linear elastic solid. This means that the resulting extra-stress is proportional to the deformation,

$$\tau(0^+) = G_0 \gamma(0^+),$$

where $\tau(0^+)$ and $\gamma(0^+)$ are the extra-stress tensor and infinitesimal strain tensor immediately after deformation, and $G_0$ is the shear modulus. The shear modulus can also be obtained from the relaxation modulus $G(t)$,

$$G_0 = \lim_{t \to 0} G(t).$$

In order to determine the shear modulus of a suspension of FENE dumbbells we subject the ensemble to a step-strain experiment and calculate the resulting extra-stress. We simulate this experiment by applying a large and constant velocity gradient during a very small period of time $\delta t$, such that the resulting deformation remains small. The time evolution of the configuration of each dumbbell in the ensemble is governed by a stochastic differential equation, Eq. (4). However, because of the delta-like behaviour of the velocity gradient, the effects of the connector force and of the Brownian motion during the deformation process are negligible, and the stochastic differential equation effectively reduces to an ordinary differential equation. This is equivalent to assuming that each dumbbell moves affinely with the fluid, thus

$$\mathbf{Q}^+ = \mathbf{Q} + \mathbf{Q} \cdot \mathbf{Q} \gamma \delta t,$$

where $\mathbf{Q}$ is the dumbbell connector vector in equilibrium and $\mathbf{Q}^+$ denotes the connector vector of the dumbbell at $t = \delta t$, that is, immediately after the deformation has been applied. The polymer contribution to the stress immediately after the deformation is given by

$$\tau^+ / nkT = \langle F^+ \mathbf{Q}^+ \rangle^+ - 1.$$

In this expression, $1$ is the unit tensor and the brackets denote an averaging over the probability distribution, that is,

$$\langle \cdot \rangle^+ = \int \psi^+(\mathbf{Q}^+ \cdot) \mathbf{d}Q^+, \tag{15}$$

where $\psi^+(\mathbf{Q}^+ \cdot) \mathbf{d}Q^+$ is the probability to find a connector vector in the range $\mathbf{Q}^+$ and $\mathbf{Q}^+ \cdot \mathbf{d}Q^+$, immediately after deformation.

Substituting the dimensionless form of the connector force Eq. (5) into Eq. (14), it follows that the polymer contribution to the stress immediately after deformation is given by

$$\tau^+ / nkT = \left( \frac{\mathbf{Q}^+ \mathbf{Q}^+}{1 - (\mathbf{Q}^+ \gamma / \beta)^2} \right)^+ - 1. \tag{16}$$

Furthermore, in view of the assumed affine motion, Eq. (13), we have

$$\psi^+(\mathbf{Q}^+) \mathbf{d}Q^+ = \psi(\mathbf{Q}) \mathbf{d}Q,$$  

$$\tag{17}$$
where $\psi(Q)$ is the equilibrium distribution, $Q$ is the orientation of a dumbbell in equilibrium and $Q^+$ denotes the orientation of the same dumbbell after the deformation has been applied. Using this relation, and substituting for $Q^+$ from Eq. (13), Eq. (16) can be transformed into an integration over the equilibrium distribution function:

$$\tau^+/nkT = \left( \frac{\langle \mathbf{Q} + \kappa \cdot \mathbf{Q} \mathbf{Q} + \kappa \cdot \mathbf{Q} \rangle}{1 - \mathbf{Q} + \kappa \cdot \mathbf{Q}^2/b} \right) - 1.$$

(18)

Since we consider infinitesimal deformations we have $|\kappa|\delta t \ll 1$, and Eq. (18) can be approximated by

$$\tau^+/nkT \approx \kappa \mathbf{T} \delta t + \kappa^T \delta t + \left\langle \mathbf{Q} \mathbf{Q} \left( \frac{\kappa + \kappa^T}{b(1 - Q^2/b)^2} \right) \right\rangle \delta t,$$

(19)

which is exact up to terms linear in $|\kappa|\delta t$. Furthermore, in going from Eq. (18) to Eq. (19) we made use of the fact that in equilibrium we have

$$\left\langle \mathbf{Q} \mathbf{Q} \right\rangle = 1.$$

(20)

In evaluating the last tensor in Eq. (19) it is convenient to calculate its matrix representation relative to the principal axes of the tensor $\kappa + \kappa^T$, and because the FENE equilibrium distribution [7] is isotropic, this matrix is diagonal. The calculation of the resulting integrals, which can be evaluated exactly, is given in Appendix A. The final result is

$$\left\langle \mathbf{Q} \mathbf{Q} \left( \frac{\kappa + \kappa^T}{b(1 - Q^2/b)^2} \right) \right\rangle = \frac{1}{(b - 2)} \{ \text{tr}(\kappa + \kappa^T) \mathbf{I} + 2(\kappa + \kappa^T) \},$$

(21)

where ‘tr’ denotes the trace operator. Assuming incompressibility the first term vanishes and Eq. (19) reduces to

$$\tau^+/nkT = \frac{b}{b - 2(\kappa + \kappa^T)} \delta t.$$

(22)

To compare this expression with the definition of the shear modulus $G_0$ in Eq. (11), note that the infinitesimal strain tensor $\gamma$ is given by $\gamma = (\kappa + \kappa^T) \delta t$. Thus, we obtain

$$G_0 = \frac{b}{b - 2} nkT.$$

(23)

By an analogous calculation the shear modulus of the FENE-P model can be shown to be $G_0 = nkT$. Clearly, Eq. (23) is not applicable for $b < 2$, and it even diverges for $b \to 2$. This is a consequence of the linearisation with respect to $|\kappa|\delta t$ and in particular of the assumed affine motion. For small values of $b$, however, a large fraction of the dumbbells have a length near the maximum possible extension, and the assumption of affine motion is then no longer justified, because affine motion would violate the requirement that $Q^2 < b$. We note that Eq. (23) is in accordance with the expression for the FENE relaxation modulus $G(t)$, derived along other lines in [9], if we take the limit $t \to 0$ of this expression. Alternatively it can be obtained from the expression for the complex FENE viscosity $\eta''$ given in [7,10], by taking the limit $G_0 = \lim_{\omega \to \infty} \omega \eta''(\omega)$. 
Fig. 4. Time evolution of the polymer contribution to the shear-stress, for FENE (solid line) and FENE-P dumbbells (dash-dot line) using ensembles of 64,000 dumbbells with an extensibility parameter $b = 12$. A large, constant shear rate $\dot{\gamma} = 1000$ is imposed during a very short period of time $\delta t = 10^{-7}$. Subsequently the flow is turned off and the system is allowed to relax.

Because the appearance of the factor $b/(b-2)$ in the shear modulus of the FENE model is most easily visualised for small values of the extensibility parameter, we conducted simulations of step-strain experiments of shear flow using FENE and FENE-P dumbbells with $b = 12$. For each simulation, an equilibrium ensemble of 64,000 dumbbells was subjected to shear flow with a constant shear rate $\dot{\gamma} = 1000$ that was imposed during a very short period of time $\delta t = 10^{-7}$, after which the flow was stopped. The stochastic differential equations were solved using the Euler forward scheme from Eq. (9) and using a time step of $\Delta t = 10^{-9}$. To reduce the noise in the shear stress, we apply a simple but very effective form of variance reduction [11]. This means that starting from one and the same equilibrium ensemble, we calculate two shear stress signals. First we determine the shear stress $\tau_{xy}$ while applying the shear flow. Next, using exactly the same sequence of random numbers we repeat the calculation in absence of flow. This equilibrium signal (with a vanishing expectation value) is then subtracted from the shear flow signal. Since by construction the two signals are correlated, this operation suppresses the noise component in the shear flow signal dramatically. The results in Fig. 4 correspond to the case $b = 12$ and are seen to agree with the predicted slope of 1.2 for the FENE model and a slope of 1 for the FENE-P model. Due to the shear modulus of the FENE-P model $G_0 = n k T b$ being smaller than the FENE value $G_0 = n k T b/(b-2)$, the stress predicted by the FENE-P model turns out to be too low. This behaviour was also observed in the simulations of the shear stress in the previous section, where during start-up of the flow the FENE-P model was seen to underpredict the stress obtained by the FENE model.

4. Selection of the parameters in the FENE-P model

The Brownian dynamics simulations of start-up of shear and elongational flow demonstrate that in a given flow the FENE-P response can be very different from the FENE behaviour. In the previous section we have shown that the underprediction of the stress of the FENE model by the FENE-P during the start-up of flow is a consequence of the fact that the shear modulus
of the FENE-P model is smaller than the shear modulus of the FENE model. Similarly, it can be shown that the FENE-P model results in a zero-shear-rate viscosity \( \frac{nkT \lambda_H b}{(b + 3)} \), [12], while the FENE value is \( nkT \lambda_H b/(b + 5) \), [7]. Therefore, if the parameters used in the FENE-P model have exactly the same values as their FENE counterparts, the behaviour of the FENE-P model will necessarily differ from the FENE behaviour. In view of this it seems to be a more natural approach to select the parameters in the FENE-P model such that at least the linear viscoelastic properties of the resulting FENE-P model are close to those of the FENE model to be approximated.

Similar to the approach followed by Bird et al. [7], where effectively the zero-shear-rate viscosity of the FENE and an approximative FENE model are taken to be equal, we construct a FENE-P model which has the same shear modulus and the same zero-shear-rate viscosity as the FENE model we want to approximate. The motivation behind this particular choice is to obtain flow behaviour identical to that of the FENE model in two distinct flow regimes. The shear modulus determines the response to very fast changes in flow, while the zero-shear-rate viscosity plays an important role in steady flows. The resulting FENE-P model will be denoted by FENE-P* and the parameters in this model are denoted as \( \lambda_H^*, b^* \) and \((nkT)^*\), to distinguish them from the FENE values \( \lambda_H \), \( b \) and \( nkT \). Equating the shear moduli gives

\[
(nkT)^* = \frac{b}{b - 2} nkT,
\]

while the condition of equal zero-shear-rate viscosities leads to

\[
\frac{b}{b^* + 3} (nkT)^* \lambda_H^* = \frac{b}{b + 5} nkT \lambda_H.
\]

Combining these two equations, we obtain a single relation that involves both the finite extensibility parameters and the microscopic relaxation times of the two models. This relation is insufficient to express \( b^* \) and \( \lambda_H^* \) uniquely in terms of their FENE counterparts \( b \) and \( \lambda_H \). The resulting freedom can be exploited to impose another condition to obtain optimal agreement with the FENE behaviour, for example \( b^* = b \) or \( \lambda_H^* = \lambda_H \). We have determined the optimal choice of parameters from the position of the overshoot peak in the shear stress at moderate and elevated shear rates. The choice \( b^* = b \) was found not to yield the correct peak position. In order to investigate the effect of changing the value of \( \lambda_H^* \) under the constraint of equal shear moduli and equal zero-shear-rate viscosity, we set \( \lambda_H^* = c \lambda_H \), varying \( c \) near 1. The choice \( c = 1 \) was found to be optimal. The results of shear flow simulations using shear rates \( \dot{\gamma} = 10 \) and \( \dot{\gamma} = 20 \) are presented in Table 1. As can be seen, small changes in \( c \) have a great impact both on the value of \( b^* \) and on the position of the overshoot peak relative to that of the FENE model. Because the overshoot peak position is best predicted if \( c = 1 \), we take \( \lambda_H^* = \lambda_H \). Substituting this in Eq. (25), along with Eq. (24), we arrive at the following relation between the extensibility parameters

\[
b^* = 3(b - 2)/7.
\]

Thus, the extensibility parameter to be used in the FENE-P* model is much smaller than the FENE value, and the relative difference with the FENE extensibility persists in the limit \( b \to \infty \). This should not be too surprising, since the meaning of the extensibility parameter \( b \) in the
Table 1
Shear flow simulation results, using $\lambda_H^* = c \lambda_H$, where $c$ varies near 1 and the resulting values for $b^*$ are shown in column 2. The last two columns show the resulting overshoot peak positions relative to the overshoot peak position in the FENE simulations, using shear rates $\dot{\gamma} = 10$ and $\dot{\gamma} = 20$, respectively.

<table>
<thead>
<tr>
<th>$c$</th>
<th>$b^*$</th>
<th>$t_{\text{peak}}/(t_{\text{peak,FENE}})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.90</td>
<td>96</td>
<td>1.91 ± 0.03</td>
</tr>
<tr>
<td>0.95</td>
<td>33.88</td>
<td>1.22</td>
</tr>
<tr>
<td>1.00</td>
<td>20.57</td>
<td>1.00</td>
</tr>
<tr>
<td>1.05</td>
<td>14.77</td>
<td>0.84</td>
</tr>
<tr>
<td>1.10</td>
<td>11.52</td>
<td>0.72</td>
</tr>
</tbody>
</table>

FENE model is very different from that in the FENE-P model. In the FENE model the length of an individual dumbbell is constrained by the condition $Q^2 < b$, whereas in the FENE-P model only the average dumbbell length is constrained, i.e. $\langle Q^2 \rangle < b$. Thus, in the FENE-P model it will be possible to find dumbbells with a length $Q^2 > b$. As a first indication of the performance of the FENE-P* model, we will repeat the Brownian dynamics simulations of start-up of shear and elongational flow, and the results will be contrasted with those of the FENE-P model. In the FENE simulations we take, $b = 50$. From Eq. (24) and Eq. (25) we obtain $b^* = 20.57$ and $(nkT)^* = 1.04 nkT$. If we concentrate on the shear stress results in Fig. 5 first, the FENE-P* appears to be a much better approximation of the FENE model than is the original FENE-P. As the shear modulus of the FENE-P* model was designed to be equal to that of the FENE model, the start-up of the shear stress is improved significantly and its initial slope coincides with the FENE slope. Furthermore, while the overshoot of the original FENE-P model occurs too late and is too large, the FENE-P* model predicts the right position and magnitude. Similarly, since the FENE-P* model has the same zero-shear-rate viscosity as the FENE model,

![Fig. 5. Shear stress FENE (solid line), FENE-P (dash-dot line) and FENE-P* model (dashed line) in start-up of shear flow. The extensibility parameter in the FENE and FENE-P calculations is $b = 50$. In the FENE-P* model we use $b^* = 20.57$ and $(nkT)^* = 1.04 nkT$. The results were obtained using ensembles of 64000 dumbbells. The curves correspond to shear rates $\dot{\gamma} = 1$ (lower curves), $\dot{\gamma} = 2$ and $\dot{\gamma} = 4$ (upper curves).](image-url)
the steady state values of the stress are considerably closer to the FENE values than the original FENE-P model; for a shear rate $\dot{\gamma} = 4$ a slight underprediction is observed. The results of the first normal stress differences are shown in Fig. 6. The large differences between the steady state values of the original FENE-P and the FENE almost disappear when using the FENE-P* model. Note that the good agreement between the shear flow results of the FENE model using $b = 50$ and the FENE-P* using $b^* = 20.57$ also indicates the difficulty of the molecular interpretation of the extensibility parameter when using the FENE-P model to fit experimental shear data.

As can be seen from Figs. 7 and 8, at higher shear rates the results of the FENE-P* model and those of the FENE model deviate more than at the shear rates considered above, but the FENE-P* model is still an improvement compared to the FENE-P model, especially for the first normal stress difference.
When comparing the elongation simulations a completely different behaviour is observed, cf. Fig. 9. The FENE-P* model fails to reach the FENE plateau values of the elongation viscosity. The reason for this is demonstrated most easily by considering the plateau value of the elongational viscosity in the limit of infinite elongation rate $\dot{\varepsilon} \to \infty$. In this limit the elongational viscosity of the FENE-P* model becomes \[ \tilde{\eta}^* = \frac{2b^*(nkT)^*\lambda}{H}. \] (27)

Expressing the FENE-P* quantities in terms of FENE values we obtain

\[ \tilde{\eta}^* = \frac{3}{7} \tilde{\eta}. \] (28)

Therefore, in contrast to the FENE-P model, the FENE-P* cannot reach the FENE plateau values. In this respect the original FENE-P model clearly outperforms the FENE-P* model.
Fig. 10. Elongation viscosity as a function of strain for the FENE (solid line), FENE-P (dash-dot line) and FENE-P* model (dashed line) in start-up of elongational flow. The extensibility parameter in the FENE and FENE-P calculations is $b = 50$. In the FENE-P* model we use $b^* = 20.57$ and $(nkT)^* = 1.04nkT$. The results were obtained using ensembles of 64,000 dumbbells. The applied elongation rate $\dot{\varepsilon} = 1$.

On the other hand, however, it is seen that the start-up behaviour of the FENE-P* model is in better agreement with the FENE start-up results than the conventional FENE-P model. This is shown in Figs. 10 and 11; for strain values $\dot{\varepsilon}t < 1.5$, the FENE-P* curve is closer to the FENE results than the FENE-P curve. From a Lagrangian point of view this is important because in more complex flows, along its trajectory, a dumbbell may temporarily experience a flow field with a dominant elongational behaviour. Unless the residence time in this region is very long and the strain rate is large, it is not important to recover the complete elongation curve up to its plateau, but it is rather the behaviour as a function of accumulated strain that matters.

Fig. 11. Elongation viscosity as function of strain FENE (solid line), FENE-P (dash-dot line) and FENE-P* model (dashed line) in start-up of elongational flow. The extensibility parameter in the FENE and FENE-P calculations is $b = 50$. In the FENE-P* model we use $b^* = 20.57$ and $(nkT)^* = 1.04nkT$. The results were obtained using ensembles of 64,000 dumbbells. The applied elongation rate $\dot{\varepsilon} = 2$. 
5. Simulation of flow past a cylinder

Until now, our investigation and comparison of the FENE, FENE-P and FENE-P* models has been restricted to their behaviour in shear and elongational flow. In a typical simulation, however, the character of the flow field will embrace the whole range from shear to elongation-like, it is necessary to test and compare the models also in complex flow situations. As an example of such a complex flow field, we consider the start-up of flow past a cylinder between two parallel plates and compare the behaviour of the FENE, FENE-P and the FENE-P* models. We use the same configuration as in [6]: the cylinder of radius \( a \) is placed at the origin of a Cartesian coordinate system with parallel plates at \( y = \pm 2a \), cf. Fig. 12. The flow is generated by imposing a constant flow-rate and is started at \( t = 0 \). The characteristic dimensionless quantities of the flow problem are the Reynolds number \( Re = \rho Ua/(nkT\lambda_H) \), the Deborah number \( De = \dot{\lambda}_H U/a \) and the viscosity ratio \( \eta_s/(nkT\lambda_H) \), where \( \rho \) is the density, \( \eta_s \) is the solvent viscosity and \( U \) is the mean fluid velocity. The results will be presented in dimensionless form using the radius of the cylinder as the new unit of length, and \( a/U \) as the new unit of time. In the FENE calculations we take \( Re = 0.01125 \), \( De = 0.6 \), \( \eta_s/(nkT\lambda_H) = 0.125 \), and use a finite extensibility parameter \( b = 50 \). The parameters in the FENE-P* model are determined from Eqs. (24) and (25) thus, \((nkT)^* = 1.04nkT\) and \( b^* = 20.57 \).

The flow problem is solved by means of the finite element method. The continuity and momentum equations are discretised by means of the DEVSS method [13]. In combination with the DEVSS method, the discontinuous Galerkin method (DG) is used to discretise the closed form constitutive equation, [14], in the case of the FENE-P model and the equations for the Brownian configuration fields in the case of the FENE model. We use an Euler forward time stepping scheme for obtaining the polymer stress either from the constitutive equation or from the Brownian configuration fields. Subsequently, the velocity field is calculated from the momentum equation, where all terms are taken implicitly, except for the nonlinear convective term, which is taken explicitly. The details of the implementation can be found in [6].

The FENE results are obtained using 4000 configuration fields. The results shown below are averaged over four independent calculations on mesh2 of [6]. The convergence upon mesh refinement in the case of the FENE, FENE-P and FENE-P* is found to be much faster than the convergence behaviour in the case of Hookean dumbbells. The differences between the results of the mesh1 and mesh2 calculations of the FENE, FENE-P and FENE-P* model are found to be very small in contrast to the results for the Hookean dumbbell model [6].
Fig. 13 shows the drag coefficient $C_d = F_d / (nkT \gamma_H U)$, as a function of time, where $F_d$ is the drag force per unit length on the cylinder. In accordance with our earlier shear flow simulations, the FENE-P model is seen to underpredict the drag coefficient during the start-up phase and it monotonically grows to a steady state somewhat higher than the FENE value. In contrast, the FENE-P* model shows the correct start-up behaviour and predicts a modest overshoot. The quality of the steady state behaviour of drag coefficient of the FENE-P* and FENE-P models is seen to be similar.

The stresses obtained by the FENE-P* and FENE-P models, however, are very different. Fig. 14 shows the $xx$-component of the polymer stress, $\tau_{xx}^p$, at the symmetry axis ($y = 0$) and at the surface of the cylinder. The FENE-P* model is seen to approximate the FENE results very well. At the surface of the cylinder the FENE-P* model slightly underpredicts the FENE results, but in the wake behind the cylinder, the results coincide with the FENE results. In contrast, the FENE-P model is seen to result in stresses that are too large, not only at the surface of the cylinder but also in the wake. At the surface of the cylinder it overpredicts the FENE results by a factor of nearly 1.5. Fig. 15 shows the $xy$-component of the polymer stress, $\tau_{xy}^p$, at the symmetry axis ($y = 0$) and at the surface of the cylinder. Similar to the results for the $xx$-component of the
6. Conclusions

Although the FENE-P model is often used to replace the underlying FENE model, the FENE-P model has different rheological properties than the FENE model. This has been demonstrated by means of simulations of simple shear and elongation flow, as well as for a complex flow viz. flow past a cylinder. We constructed a FENE-P model in which the parameters were selected such that it has the same shear modulus and the same zero-shear-rate viscosity as those of the FENE model to be approximated. Making the extra assumption of
equal microscopic time constants $\lambda_H$, we obtain a FENE-P model with a smaller extensibility parameter than that which is used in the FENE model. This model is referred to as the FENE-P* model. The Brownian dynamics simulations of shear flow demonstrate that the FENE-P* model reproduces the FENE shear stress and first normal stress differences much better than the original FENE-P model, particularly at moderate shear rates. These results indicate that one should exercise care when interpreting the value of the parameters in an approximative model, since they may only be indirectly related to molecular quantities. For example, our results may partly explain the small value of the $b$ (or $L^2$) parameter that have been used in simulations to obtain satisfactory agreement with experimental data [15]. A consequence of the smaller extensibility parameter, however, is that for large elongation rates the FENE-P* elongational viscosity is smaller than the FENE and FENE-P elongational viscosity by a factor of $3/7$, and in this case the FENE model is approximated better by the FENE-P model. In start-up of elongational flow, however, for strain values $<1.5$ the results of the FENE-P* model are closer to the FENE elongational viscosity than those of the FENE-P model. Finally, the simulations of the complex flow field past a cylinder for the FENE, FENE-P and FENE-P* models demonstrate that the FENE-P model results in an overprediction of the FENE stress values, while the FENE-P* model shows good agreement with the FENE results. This behaviour is also expected to hold for other complex flows at moderate Deborah numbers.

Acknowledgements

Part of the work was funded by the European Commission under contract number BRPR-CT96-145. Further financial support by Akzo-Nobel, Philips Electronics, Shell and Unilever is gratefully acknowledged.

Appendix A. Shear modulus of the FENE model

In this section we will elaborate on the derivation of the FENE shear modulus, specifically on the steps made in going from the LHS to the RHS of Eq. (21). In order to evaluate the tensor

$$\left\langle \mathbf{QQ} \left( \frac{\mathbf{Q} \cdot (\kappa + \kappa^T) \cdot \mathbf{Q}}{b(1 - Q^2/b)^2} \right) \right\rangle,$$

we represent it relative to the orthogonal principal axes of the symmetric tensor $(\kappa + \kappa^T)$. Relative to this basis, the tensor $(\kappa + \kappa^T)$ has the following diagonal matrix representation

$$\kappa + \kappa^T = \begin{pmatrix} \hat{\lambda}_1 & 0 & 0 \\ 0 & \hat{\lambda}_2 & 0 \\ 0 & 0 & \hat{\lambda}_3 \end{pmatrix},$$

where $\hat{\lambda}_1$, $\hat{\lambda}_2$ and $\hat{\lambda}_3$ are the eigenvalues of $(\kappa + \kappa^T)$. Thus, the matrix representation of Eq. (29) becomes
where \( Q_k \) is the component of \( Q \) in the direction of the \( k \)th principal axis of \((\kappa + \kappa^T)\). In order to calculate the averages, we need the equilibrium distribution \( \psi(Q) \). The latter is given in [7], and in spherical coordinates it reads

\[
\psi(r, \theta, \phi) = \frac{1}{N}[1 - r^2/b]^{\nu/2},
\]

valid for \( 0 \leq r \leq \sqrt{b} \) and \( \psi(r, \theta, \phi) = 0 \) otherwise. The normalisation factor \( N \) is given by

\[
N = 2\pi b \sqrt{b B \left( \frac{3}{2}, \frac{b + 2}{2} \right)},
\]

where \( B \) denotes Euler's Beta-function.

Since the equilibrium distribution \( \psi(r, \theta, \phi) \) is isotropic, the non-diagonal integrals vanish because their angle-integrals integrate to zero. Therefore, we only have to calculate the diagonal integrals, which are essentially forms like

\[
\left\langle \frac{Q_1^4}{b(1 - Q^2/b)^2} \right\rangle,
\]

and

\[
\left\langle \frac{Q_1^2 Q_2^2}{b(1 - Q^2/b)^2} \right\rangle.
\]

In evaluating these expressions, it is convenient to express the Cartesian components of \( Q \) in spherical coordinates. The final result is

\[
\left\langle \frac{Q_1^4}{b(1 - Q^2/b)^2} \right\rangle = \frac{3}{b - 2}.
\]

By a similar calculation it can be shown that Eq. (35) transforms into

\[
\left\langle \frac{Q_1^2 Q_2^2}{b(1 - Q^2/b)^2} \right\rangle = \frac{1}{(b - 2)}.
\]

Therefore Eq. (29) becomes

\[
\frac{1}{b - 2} \left\{ (\lambda_1 + \lambda_2 + \lambda_3) \left[ \begin{array}{ccc} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{array} \right] + 2 \left[ \begin{array}{ccc} \lambda_1 & 0 & 0 \\ 0 & \lambda_2 & 0 \\ 0 & 0 & \lambda_3 \end{array} \right] \right\}.
\]

The first term in this expression is isotropic while the second term is exactly the matrix representation of the tensor \((\kappa + \kappa^T)\). Therefore, expressing Eq. (38) in tensor form, we finally obtain

\[
\frac{1}{b - 2} \{ \text{tr}(\kappa + \kappa^T) \mathbf{1} + 2(\kappa + \kappa^T) \},
\]
where $\mathbf{1}$ is the unit tensor, and ‘tr’ denotes the trace operator.

References