Numerical Simulation of Viscoelastic Start-up Flow between Parallel Plates Using Stochastic Calculation*

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The start-up flow of polymeric liquids between parallel plates was numerically simulated using a stochastic approach. The extra stress was calculated by solving the stochastic equation of a reptation model instead of using a constitutive equation. The numerical results showed that the present numerical simulation was capable to describe typical macroscopic behaviors of polymeric liquids such as the change in velocity profile with time, the growth in shear stress, and overshoot phenomena of velocity and wall shear stress. Moreover, the stochastic approach can predict microscopic behaviors such as the time development of polymer orientation. It is advantage of the present method that information related to the behavior of microscopic polymer structures in a flow can be obtained. In addition, the comparison with the numerical prediction using an approximated constitutive model showed that a closure approximation used led to a remarkable error just after the onset of the flow.

Key Words: Non-Newtonian Fluid, Viscoelastic Fluid, Unsteady Flow, Stochastic Calculation, Startup Flow, Parallel Plates, Curtiss-Bird Model, Doi-Edwards Model

1. Introduction

In the traditional approach to the numerical solution of viscoelastic flow problems, the balance equations, i.e. the equation of continuity, the equation of motion, and the energy equation, and the constitutive equation are simultaneously solved by appropriate numerical methods. The constitutive equation describes the behavior of viscoelastic fluids and the polymeric contribution to the stress field is computed by solving a constitutive equation. Thus the constitutive equation plays an important role in the analysis of viscoelastic flows. Constitutive equations derived from kinetic theory models usually express the molecular orientation in terms of an orientation distribution function and hence some approximations are required to obtain a closed-form of constitutive equa-

tions represented by an orientation tensor. On the other hand, in recent studies, stochastic approaches have been applied to compute the stress field. In the stochastic approach, the stress field is determined using a large number of polymer configurations evolving according to stochastic differential equations and thus no closed-form constitutive equation is required.

The CONNFESSIT (Calculation of Non-Newtonian Flow: Finite Elements & Stochastic Simulation Techniques) approach^{(1),(2)} combines a finite element solution of the balance equations with stochastic simulation techniques for computing polymer stress and hence can connect a description of the microstructure of a polymeric liquid using kinetic theory with a macroscopic description of the flow. This characteristic is advantageous for the simulation of the flow of polymeric liquids. It is well known that the microscopic configuration such as molecular orientation is important information in the flow analysis of viscoelastic fluids⁽³⁾. The numerical simulations using the CONNFESSIT approach have been performed for dumbbell models⁽⁴⁾⁻⁽¹²⁾ and reptation models⁽¹³⁾⁻⁽¹⁵⁾;

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The former is a model for dilute polymer solutions and the latter is derived for concentrated polymer solutions and polymer melts.

In the present paper, we numerically simulated the viscoelastic startup flow at constant mean velocity between parallel plates using the reptation model as a kinetic model of polymer dynamics. The startup flow relates to many flow problems observed in polymer processing and hence it is important to study the viscoelastic behavior in the startup flow. Although stochastic approaches were employed in the analysis of transient behavior of polymeric liquids in simple shear^{(12)–(14)} and uniaxial elongation flows⁽¹³⁾, it has not been adopted to the startup flow considered in the present study. In the startup flow in channels, the shear rate spatially distributes and temporally changes, while the shear rate is constant in simple shear flows. Thus it is an interesting issue to investigate the capability of the present approach in the startup flow. We therefore examined the effectiveness of the numerical simulation using a stochastic approach by investigating both macroscopic behaviors such as velocity and stress fields and microscopic behaviors such as transient motion of polymers in the startup flow. In novel industrial processing such as micro polymer processing, the microscopic information is as important as the prediction of the macroscopic flow field. In addition, we compared the numerical results with the counterparts computed with an approximated constitutive equation to investigate the advantage of the stochastic approach in the analysis of transient flows. In the present simulation, we considered the startup flow in a simple channel geometry as a basic study for investigating the advantage of the stochastic calculation in the analysis of the transient behavior of polymeric liquids in complex flow fields appeared in polymer processing.

2. Theory

2.1 Reptation model

A class of molecular models of polymer dynamics in concentrated solutions and melts are derived applying the idea of “reptation” proposed by de Gennes⁽¹⁶⁾. This class includes the Doi-Edwards model^{(17)–(21)} and the Curtiss-Bird model^{(22),(23)}.

The original version of the Doi-Edwards⁽¹⁹⁾ and Curtiss-Bird⁽²²⁾ models is based on the following diffusion equation of a probability density $f(\mathbf{u}, s, t)$ of finding a link at position s along the chain having orientation $\mathbf{u}$ at time t :

$$\frac{\partial f}{\partial t} = \frac{1}{\lambda} \frac{\partial^2 f}{\partial s^2} - \frac{\partial}{\partial \mathbf{u}} \cdot (\mathbf{L} \cdot \mathbf{u} - \mathbf{L} : \mathbf{uuu}) f, \quad (1)$$

where $\mathbf{u}$ is a unit vector and s a number between 0 and 1, λ a characteristic time constant. The velocity

gradient tensor $(\nabla \mathbf{v})^T$ is denoted by $\mathbf{L}$, where $\mathbf{v}$ is the velocity vector. The boundary conditions for $s=0$ and $s=1$ supplementing the diffusion equation (1) are

$$f(\mathbf{u}, 0, t) = f(\mathbf{u}, 1, t) = \frac{1}{4\pi}. \quad (2)$$

The stress tensor $\boldsymbol{\tau}$ is described by

$$\boldsymbol{\tau} = nNk_B T \left[\langle \mathbf{uu} \rangle + \lambda \varepsilon_i \mathbf{L} : \langle s(1-s) \mathbf{uuuu} \rangle - \frac{1}{3} \mathbf{I} \right], \quad (3)$$

where n is the number density of beads, N the number of beads in a chain, k_B the Boltzmann constant, T the absolute temperature, and $\mathbf{I}$ the unit tensor. ε_i is link tension coefficient having a value between 0 and 1, which characterizes anisotropic frictional properties of the polymer chains in a melt. When $\varepsilon_i=0$, this model reduces to the Doi-Edwards model. The brackets $\langle \dots \rangle$ denote average with respect to the probability function $f(\mathbf{u}, s, t)$.

2.2 Stochastic approach

The diffusion equation (1) can be transformed into the following stochastic differential equations^{(2),(13)}:

$$d\mathbf{u} = (\mathbf{L} \cdot \mathbf{u} - \mathbf{L} : \mathbf{uuu}) dt, \quad (4)$$

$$ds = \sqrt{2/\lambda} dW, \quad (5)$$

where W is the Wiener process. The Stochastic differential equations (4) and (5) are discretized as follows:^{(2),(13)}

$$\mathbf{u}_{i+1} = \mathbf{u}_i + (\mathbf{L} \cdot \mathbf{u} - \mathbf{L} : \mathbf{uuu})_i \Delta t \approx \frac{\mathbf{u}_i + \mathbf{L} \cdot \mathbf{u}_i \Delta t}{|\mathbf{u}_i + \mathbf{L} \cdot \mathbf{u}_i \Delta t|}, \quad (6)$$

$$s_{i+1} = s_i + \sqrt{\frac{2\Delta t}{\lambda}} W_i, \quad (7)$$

where the subscript i indicates the value at the i th time step. Δt is the time interval. W_i s are random numbers generated from a Gaussian distribution with unit variance and zero mean.

The tensor $\mathbf{L}$ is evaluated at $t=i\Delta t$. In the calculation of s , if the value of s_{i+1} lies outside the interval $[0, 1]$, the value is replaced by the value obtained via reflection at the boundary passed in leaving the interval $[0, 1]$, i.e. $s_{i+1} \rightarrow -s_{i+1}$ when $s_{i+1} < 0$ and $s_{i+1} \rightarrow 2 - s_{i+1}$ when $s_{i+1} > 1$. When the reflection occurs, the orientation vector is randomly chosen because of the boundary condition (2). In the present simulation, an improved algorithm is applied in numerical simulations to consider unobserved reflections. The detail of the algorithm is available in the literature^{(2),(9)}.

For evaluating the stress tensor, the stochastic differential equation is solved for a large number of trajectories, and the average $\langle \dots \rangle$ is evaluated as an ensemble average of the result for each trajectory:

$$\int f(\mathbf{u}, s, t) \mathbf{uuu} d\mathbf{u} \rightarrow \frac{1}{N_T} \sum_{i=1}^{N_T} (\mathbf{uu})_i, \quad (8)$$

where i means the results of the i th trajectory and N_T is the number of the total trajectories.

Viscometric functions evaluated using the stochastic calculation show typical rheological properties of polymeric liquids such as the shear-thinning property of both the steady shear viscosity and the first normal stress coefficient^{(2),(13)}: The shear-thinning property intensifies with increasing ε_i . In addition, elongational properties have been also investigated to indicate that steady and unsteady elongational properties of polymeric liquids can be described by the stochastic approach⁽¹³⁾.

2.3 Start-up flow between parallel plates

We numerically calculated a two-dimensional start-up flow of the Curtiss-Bird fluid between parallel plates. In the calculation, the stochastic approach with the improved algorithm^{(2),(9)} was adopted. In addition, we employed an approach used by Isayev⁽²⁵⁾ in a simulation of the start-up flow of the Leonov model. The start-up flow of constant mean velocity was considered; This problem corresponds to "Constant- U case" in his simulation⁽²⁵⁾.

The gap distance between the plates is $2b$. The flow and the gap directions are x and y , respectively and the plates place at $y = \pm b$. The inertial effect was neglected in the present case. The equation of motion then reduces to

$$\frac{\partial \tau_{xy}}{\partial y} - \frac{\partial p}{\partial x} = 0, \quad (9)$$

$$\frac{\partial}{\partial y}[\tau_{yy} - p] = 0, \quad (10)$$

where p is the pressure and τ_{ij} the ij component of the extra stress tensor $\boldsymbol{\tau}$. The mean velocity is constant at $\bar{U}$ and the following relation is obtained.

$$\int_{-b}^b v dy = 2b\bar{U}, \quad (11)$$

where v is the velocity in the flow direction (x). Integrating equation(11) by parts together with boundary conditions, $v=0$ at $y = \pm b$ and $\partial u/\partial y=0$ at $y=0$, yields

$$-\int_{-b}^b \frac{\partial v}{\partial y} y dy = 2b\bar{U}. \quad (12)$$

By integrating equation(9) with respect to y under the condition of $\partial \tau_{xy}/\partial y=0$ at $y=0$, one obtains

$$\tau_{xy} = \frac{\partial p}{\partial x} y \equiv -\Lambda y. \quad (13)$$

The extra stress is divided into a Newtonian and a polymer contributions, i.e. $\boldsymbol{\tau}^N$ and $\boldsymbol{\tau}^p$, respectively. The polymer contribution is calculated with the stochastic method. When τ_{xy} is divided into a Newtonian and a polymeric parts, it can be expressed by $\tau_{xy} = \tau_{xy}^N + \tau_{xy}^p = \eta_N(\partial v/\partial y) + \tau_{xy}^p$. Substituting this equation into equation(13) yields

$$\frac{\partial v}{\partial y} = \frac{-\Lambda y - \tau_{xy}^p}{\eta_N}. \quad (14)$$

From equations(12) and (14), one obtains

$$\Lambda = \frac{3\eta_N \bar{U}}{b^2} - \frac{3}{2b^3} \int_{-b}^b \tau_{xy}^p y dy. \quad (15)$$

The governing equation was non-dimensionalized according to the following rule: $y = Ly^*$, $t = (L/U)t^*$, $p = (nNk_B T)p^*$, $\tau_{xy}^p = (nNk_B T)\tau_{xy}^{p*}$, and $v = Uv^*$, where L is a characteristic length and U a characteristic velocity. In the present calculation, we chose $L=b$ and $U=\bar{U}$. Consequently, the non-dimensional governing equations are denoted by

$$-\frac{\partial p^*}{\partial x^*} = \frac{3}{\alpha} - \frac{3}{2} \int_{-1}^1 \tau_{xy}^{p*} y^* dy^* \equiv \Lambda^*, \quad (16)$$

$$\frac{\partial v^*}{\partial y^*} = -\alpha(\Lambda^* y^* + \tau_{xy}^{p*}), \quad (17)$$

where α is a parameter defined by $\alpha = nNk_B TL/(\eta_N U)$ and means the ratio of the contribution to $\boldsymbol{\tau}$ between $\boldsymbol{\tau}^p$ and $\boldsymbol{\tau}^N$: When $\boldsymbol{\tau} = \boldsymbol{\tau}^N + \boldsymbol{\tau}^p = \eta_N\{\nabla \mathbf{v} + (\nabla \mathbf{v})^T\} + \boldsymbol{\tau}^p = 2\eta_N \mathbf{D} + \boldsymbol{\tau}^p$ is normalized according to $\boldsymbol{\tau} = (nNk_B T)\boldsymbol{\tau}^*$, $\boldsymbol{\tau}^p = (nNk_B T)\boldsymbol{\tau}^{p*}$, and $\mathbf{D} = (U/L)\mathbf{D}^*$, one obtains $\boldsymbol{\tau}^* = (2/\alpha)\mathbf{D}^* + \boldsymbol{\tau}^{p*} = \boldsymbol{\tau}^{N*}/\alpha + \boldsymbol{\tau}^{p*}$.

At the time $t^*=0^+$, the gapwise shear rate distribution is assumed that⁽²⁵⁾

$$\frac{\partial v^*}{\partial y^*} = \begin{cases} 0, & 0 \leq |y^*| < 1 - \delta^*, \\ \frac{3}{\delta^*}(1 - \delta^* - y^*), & 1 - \delta^* \leq |y^*| \leq 1, \end{cases} \quad (18)$$

The value of δ^* is not determined a priori. Thus the calculation is repeated by changing the value of δ^* as $\delta^* \rightarrow 0$ until the computational results become independent of δ^* .

The non-dimensional constitutive equations are as follows:

$$\tau_{xy}^{p*} = \langle u_1 u_2 \rangle + \varepsilon_i We \frac{\partial v^*}{\partial y^*} \langle s(1-s)u_1^2 u_2^2 \rangle, \quad (19)$$

$$s_{i+1} = s_i + \sqrt{2\Delta t^*/We} W_i, \quad (20)$$

$$\mathbf{u}_{i+1} = \frac{\mathbf{u}_i + \mathbf{L}^* \cdot \mathbf{u}_i \Delta t^*}{|\mathbf{u}_i + \mathbf{L}^* \cdot \mathbf{u}_i \Delta t^*|}, \quad (21)$$

where We is the Weissenberg number defined by $We = \lambda U/L$. The subscript i indicates the value at the i th time step.

We applied the no-slip condition on the channel walls as the boundary condition. The scheme for computation of the start-up flow is as follows: (a) the flow is at rest at $t^*=0$, i.e. $v^*=0$ and $\tau_{xy}^{p*}=0$, (b) the pressure gradient Λ^* is computed by equation(16), (c) the shear rate $\dot{\gamma}^* = \partial v^*/\partial y^*$ is evaluated by equation(17), (d) the velocity field is obtained by integrating $\dot{\gamma}^*$, (e) the shear stress τ_{xy}^{p*} is obtained by solving equations(19) - (21) with the stochastic method, (f) the time step is progressed, i.e. $t^* \rightarrow t^* + \Delta t^*$, and (g) the procedures (b) - (f) are repeated until the flow fully develops. The computational domain is divided into a large number of grids and the trapezoidal rule is applied for the numerical integration of $\dot{\gamma}^*$.

2.4 Approximated model of Doi-Edwards equation

We also computed the start-up flow using an approximated Doi-Edwards equation for comparison. Marrucci⁽²⁶⁾ and Larson⁽²⁷⁾ individually derived a differential approximation of the Doi-Edwards model:

$$\overset{\nabla}{\tau}^p + \frac{2}{3G}(\mathbf{D} : \tau^p \tau^p + \frac{1}{\lambda_R}(\tau^p - G\mathbf{I})) = 0, \quad (22)$$

where G is the elastic modulus, λ_R is the relaxation time, and the notation $[\cdot]$ denotes the upper-convected derivative. A quadratic closure approximation for the term of $\langle uuuu \rangle$ is applied to obtain the closed-form equation.

The steady shear viscosity and the first normal stress coefficient of the approximated equation are compared to the results of the stochastic calculation at $\varepsilon_l=0$ in Fig. 1. Although the predictions of the approximated equation slightly differ from the results of the stochastic calculation, this approximation seems to be appropriate over wide range of the shear rate.

In the case of the flow between parallel plates, the non-dimensional form of equation(22) reduces as follows:

$$\frac{\partial \tau_{xx}^{p*}}{\partial t^*} = 2\dot{\gamma}^* \tau_{xy}^{p*} - \frac{2}{3\beta} \dot{\gamma}^* \tau_{xx}^{p*} \tau_{xy}^{p*} - \frac{\lambda}{\lambda_R} \frac{1}{We} (\tau_{xx}^{p*} - \beta), \quad (23)$$

$$\frac{\partial \tau_{xy}^{p*}}{\partial t^*} = \dot{\gamma}^* \tau_{yy}^{p*} - \frac{2}{3\beta} \dot{\gamma}^* (\tau_{xy}^{p*})^2 - \frac{\lambda}{\lambda_R} \frac{1}{We} \tau_{xy}^{p*}, \quad (24)$$

$$\frac{\partial \tau_{yy}^{p*}}{\partial t^*} = -\frac{2}{3\beta} \dot{\gamma}^* \tau_{xy}^{p*} \tau_{yy}^{p*} - \frac{\lambda}{\lambda_R} \frac{1}{We} (\tau_{yy}^{p*} - \beta), \quad (25)$$

where $\beta \equiv G/(nNk_B T)$. It can be proved that the ratios λ/λ_R and β are 10 and 1/6, respectively: See Appendix A. The numerical procedure is the same as

Fig. 1 Shear viscosity η/η_0 and the first normal stress coefficient ψ_1/ψ_{10} of the Doi-Edwards model. Lines: stochastic calculation, circles: differential approximation

that for the stochastic approach except for the calculation of the stress. The equations (23) – (25) were solved using the finite difference method.

3. Results and Discussion

3.1 Transient behaviors of velocity and shear stress

In the present simulation, the division number in the y^* direction is 100, the time interval Δt^* is 0.000 1, and the number of trajectory N_T is 10 000 for each computational grid. Preliminary computations confirmed that these values of Δt^* and N_T are enough for the simulations carried out in the present study. The computation was carried out for various values of We and two ε_l ; The parameter α was fixed to 10 for every simulation. We show the results up to $We=10$ in the present paper. The computation is available at $We=20$ in the present scheme; however, more trajectories may be necessary for a computation at higher We .

The dependence of the fully-developed velocity profile on We is shown in Fig. 2. The results for $\varepsilon_l=0.5$ at $t^*=5$ are indicated in the figure. The velocity profile becomes flatter near the channel center with increasing We , which agrees with the behavior of non-Newtonian fluids having shear-thinning viscosity. The velocity profile depends on ε_l only slightly and the maximum velocity decreases as ε_l becomes small; hence the effect of ε_l is not discussed in the present paper.

We analyzed the development of the centerline velocity v^* and the wall shear stress τ_{xy}^* with time to investigate the transient behavior in the startup flow. Figure 3 shows the time development of v^* on the centerline for $\varepsilon_l=0.5$ at several values of We . The centerline velocity increases at the early time step,

Fig. 2 Dependence of the velocity profile on We at $t^*=5$ for $\varepsilon_l=0.5$

Fig. 3 Development of the centerline velocity with time for $\varepsilon_l = 0.5$

Fig. 4 Development of the wall shear stress with time for $\varepsilon_l = 0.5$: $We = 0.5, 1, 3, 5, 8, 10$

takes a maximum value, and decreases to a constant value. This phenomenon corresponds to the change in the velocity profile from a plug-like profile just after the onset of the flow (equation(18)) through a Newton-like parabolic profile to a profile that is flatter near the centerline.

The amount of the overshoot becomes larger because the centerline velocity at the steady state decreases more as the Weissenberg number increases as shown in Fig. 2. The velocity overshoot in the start-up flow of polymeric liquids has been observed experimentally in flows of polymer melts in a rectangular channel⁽²⁵⁾ and of polymer solutions in a circular tube⁽²⁸⁾.

Next, we analyzed the shear stress. It is confirmed the computational results by the stochastic approach satisfy the linear distribution derived by equation(13). Figure 4 shows the time development

of the wall shear stress for $\varepsilon_l = 0.5$ at several values of We . The stress rapidly increases at the onset of the flow, overshoots, and gently approaches to a steady state value. The overshoot phenomenon appears notably at high Weissenberg numbers. This result qualitatively agrees with the numerical prediction for polymer melts⁽²⁵⁾. At larger We , the amount of the overshoot is larger and it takes more time for developing the stress to the steady state. The numerical predictions agree with natural behavior of polymeric liquids because the absolute value of velocity gradient near the wall becomes larger with increasing We . For other values of ε_l , the same tendency was obtained; the shear stress decreases for small ε_l because the shear-thinning property is stronger for smaller ε_l . These results indicate that the stochastic approach can simulate the transient behavior of viscoelastic startup flow between parallel plates.

3.2 Comparison with the prediction of an approximated constitutive equation

It is interesting to compare predictions of the approximated constitutive equation (22) with the counterparts of the stochastic calculation. The results for the differential approximation of the Doi-Edwards model and those for the stochastic approach at $\varepsilon_l = 0$ are compared in Fig. 5. Temporal changes in both the centerline velocity and the wall shear stress at $We = 1, 5$ are indicated in the figure. As shown in Fig. 5(a), the predictions of v^* by the two methods are qualitatively the same, while they are quantitatively different just after the onset of the flow at higher Weissenberg number.

In the case of the wall shear stress, the difference between the results of the two methods is large as compared to the case of v^* . The disparity in the wall shear stress at $We = 5$ is about 5% at the steady state, while that in the shear viscosity in Fig. 1 is about 10% at high shear rate. In addition, the difference in τ_{xy}^* between the two predictions decreases at smaller We ($We = 1$) i.e. under a condition of lower shear rate. Thus, it can be said that the difference at the steady state is not caused by the difference in the computational scheme but originates from the difference of characteristics of the two models.

Similarly to the case of the centerline velocity, the transient behavior at early time step shows a difference between the stochastic calculation and the calculation using the differential approximation. This is probably caused by the closure approximation. It is known that the validity of the approximation depends on the degree of alignment of polymer chains⁽²⁹⁾. For example, the quadratic closure, one of the simplest approximation, is exact only for perfectly aligned molecules and decreases the accuracy as the align-

ment departs from the perfect condition. In the start-up flow, the polymer orientation changes from a random condition to a highly oriented state with time. Consequently, the difference appears strongly just after the onset of the flow when the polymer orientation is close to a random condition, and becomes small at the steady state when the polymer chains align well. Moreover, the validity of approximated equations spatially changes because the velocity gradient varies with the position and the time development of molecular orientation depends on the velocity gradient as shown in later. We need to investigate time-dependent characteristics of the approximated model to identify the cause of the difference in transient behaviors of v^* and τ_{xy}^* at early time step. However, it is not the objective of the present study. The accuracy of approximated equations depends on both

time and position. This fact affects numerical predictions of the approximated equation in both time-dependent flows and complex flows. It is advantageous to the stochastic approach that this approach can avoid such a problem.

3.3 Molecular orientation

It is a characteristic of the stochastic approach that information of microstructures such as molecular orientation can be obtained. Information related to the microstructure is useful for the analysis of behaviors of polymeric liquids in complex flows. Finally, we investigated the change in an orientation vector $\mathbf{d}$, which is defined by a unit eigenvector for the maximum eigenvalue of an orientation tensor $\langle \mathbf{u}\mathbf{u} \rangle - \mathbf{I}/3$. The direction of $\mathbf{d}$ means the mean orientation of $\mathbf{u}$.

Figure 6 shows the time development of $\mathbf{d}$ at $\varepsilon_t = 0.5$ and $We = 5, 10$. The changes in the x^* , y^* , and z^* -components of $\mathbf{d}$ at $y^* = -0.5, -1$ are indicated; the x^* , y^* , and z^* -components are denoted by d_1 , d_2 , and d_3 , respectively. For help of understanding, the projections of $\mathbf{d}$ on the $x^* - y^*$ plane are also depicted in the figure.

The vectors change their directions just after the onset of the flow and have a constant orientation at the steady state. Polymer chains lie almost in the $x^* - y^*$ plane because the z^* -component d_3 is nearly equal to zero. The vector changes its direction more quickly nearer the channel wall since the absolute value of velocity gradient is larger in this region. The orienta-

Fig. 5 Comparison of the predictions between the stochastic calculation and the differential approximation for the Doi-Edwards model at $We=1, 5$: (a) centerline velocity and (b) wall shear stress

Fig. 6 Development of the orientation vector with time for $\varepsilon_t=0.5$ at $We=5$. The upper figure indicates the projection of the vector on the $x - y$ plane

Fig. 7 Development of the orientation vector with time at $y^* = -1$ for $\varepsilon_t = 0.5$: $We = 1, 5, 10$

tion angle with the x^* -axis decreases to a constant, and the angle is smaller at $y^* = -1$ than that at $y^* = -0.5$. The difference in the angle is also caused by the difference in the velocity gradient at each position.

The behaviors of the orientation vector at $y^* = -1$ for three Weissenberg numbers are compared in Fig. 7. Since the value of the z^* -component only fluctuates around zero as shown in Fig. 6, we do not indicate data of d_3 in Fig. 7. It takes more time for polymer chains in a flow at larger We to reach a steady state orientation. This tendency is consistent with the dependence of the stress growth on We shown in Fig. 4.

4. Conclusion

We applied the stochastic approach to numerically simulate the start-up flow of viscoelastic fluids between parallel plates. The flow of the Curtiss-Bird fluid was considered and the polymeric stress was computed by solving the stochastic differential equation equivalent to the diffusion equation of the model. The present numerical simulation can describe not only macroscopic behavior such as the velocity and stress overshoots but also microscopic information such as the transient motion of molecular orientation. It is advantage of the present method that information related to microscopic polymer structures in a flow can be obtained. In addition, the comparison of the stochastic results with the numerical prediction of the approximated closed-form constitutive equation indicated that the transient prediction of the approximated equation showed errors when the molecular orientation changed largely. Consequently, the stochastic approach predicts the transient flow behavior such as the startup flow more accurately. Although the stochastic approach requires high computational performance as compared to the usual method using

constitutive equations, this approach has an advantage in the simulation of unsteady flow of polymeric liquids. We are planning to numerically simulate the transient flow of polymeric liquids in complex-shaped channels using the present approach in future study.

Appendix A

We derived here the relations, $\lambda/\lambda_R = 10$ and $\beta = 1/6$. It is known that $\eta_0 = nNk_B T \lambda (1 + 2\varepsilon_t/3)/60$ and $\phi_{10} = nNk_B T \lambda^2/300$, where η_0 and ϕ_{10} respectively indicate η and ϕ_1 when $\dot{\gamma} \rightarrow 0^{(24)}$. For the Doi-Edwards model ($\varepsilon_t = 0$), the zero-shear-rate viscosity η_0 is $nNk_B T \lambda/60$ and the zero-shear-rate first normal stress coefficient ϕ_{10} is $nNk_B T \lambda^2/300$. On the other hand, $\eta_0 = \lambda G$ and $\phi_{10} = 2G\lambda^2$ for the approximated differential model, equation (22). Comparing the counterparts of each model yields the following results:

$$\frac{\lambda}{\lambda_R} = \frac{60G}{nNk_B T}, \quad (26)$$

$$\left(\frac{\lambda}{\lambda_R}\right)^2 = \frac{600G}{nNk_B T}. \quad (27)$$

Consequently, one obtains $\beta \equiv G/(nNk_B T) = 1/6$. Substituting this result into equation (26) yields $\lambda/\lambda_R = 10$.

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