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Modeling of Viscoelastic Fluid Flow Behavior in the Circular Die Using the Leonov-Like Conformational Rheological Constitutive Equations

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Summary: In this paper, the flow behavior of Leonov-Like conformational rheological model, which has root in the generalized Poisson bracket formalism based on the conformation tensor, have been studied in the circular die flow. Prediction of the normal stress differences during the flow of these fluids lets us to follow and calculate relaxation dependent phenomena such as die swell. The model predictions have been compared for the four families of mobility expressions. The Study of the model prediction sensitivity to its mobility term shows that model predictions can cover a wide range of rheological behaviors generally observed for polymer melts and solutions in the circular die flow.

Keywords: circular die; conformation tensor; modeling; rheology

Introduction

The study of the flow behavior of polymeric fluids with relatively long molecular chains is very complex because their rheological behavior lies between the Newtonian fluids and the ideal elastic solids. Therefore, a useful instrument to understand the rheological behavior of these fluids, commonly used in industrial polymer processing, is the theoretical modeling of their rheological behavior. There are many developed models which usually relate the stress tensor to the rate of the strain tensor or its derivatives, where conformational rheological models relate the stress tensor to the molecular conformation tensor changes during flow. In these types of rheological models a microstructural state variable called conformation tensor c shows the state of deformations of polymer molecules during flow.[1,2] In conformation tensor model, if the volume of polymer chains during flow remain constant, this is called VPCR or "Leonov-like" model in which $det\ c = constant$ must be satisfied. [3]

When a polymer melt and solution flows in a circular die, the storage and dissipation of elastic energy will result in die swell phenomenon. In general, die swell phenomena is attributable to the elastic behavior of the fluids. On the other hand, there are some correlations between die swell phenomena and the normal stress differences in literatures. Several researchers have studied die swell phenomena of polymer fluids as well as they attempted to relate it to the first normal stress difference (N_1) and proposed some theoretical or semitheoretical equations.[4–7] Hence determination of the first normal stress difference is very important to explain die swell phenomena and to compute die swell ratio. In the following, the Leonov-like model has been used for prediction of the normal stress differences in the circular die.

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Rheological Model Presentation

For a non-compressible polymer fluid with a microstructure represented by a second order symmetric tensor, c, the Poisson bracket formalism leads to the following equations for the time evolution of c and the stress tensor, σ:[6,8–10]

$$\frac{\partial c}{\partial t} = \frac{1}{2}(\dot{\boldsymbol{\gamma}} \cdot c + c \cdot \dot{\boldsymbol{\gamma}}) - \frac{1}{2}(\boldsymbol{\omega} \cdot c - c \cdot \boldsymbol{\omega}) - \Lambda : \frac{\partial \Phi}{\partial c} \tag{1}$$

$$\boldsymbol{\sigma} = -2\left(c \cdot \frac{\partial \Phi}{\partial c}\right) \tag{2}$$

Where Λ is a fourth-order tensor, called the mobility tensor which is essentially an inverse relaxation time to the polymer fluids, $\dot{\boldsymbol{\gamma}}$ is the rate of strain tensor, c is conformation tensor, $\boldsymbol{\omega}$ is the vorticity tensor, $\boldsymbol{\sigma}$ is the extra tensor, and Φ is the Helmholtz free energy function. The first term $\left(\frac{1}{2}(\dot{\boldsymbol{\gamma}} \cdot c + c \cdot \dot{\gamma}) - \frac{1}{2}(\omega \cdot c - c \cdot \omega)\right)$ is a convective term describing the conservative effects derived from the Poisson bracket which is only related to the conformation tensor in specific radius of die. The second term $\left(\Lambda : \frac{\partial \boldsymbol{\Phi}}{\partial c}\right)$ is dissipative term accounting for non-conservative phenomena derived from the dissipative bracket in the generalized Poisson bracket formalism.

For the Hookean based model, the Helmholtz free energy function can be written as:[9,10]

$$\boldsymbol{\Phi} = \frac{\lambda}{2}\left[tr\left(\frac{\delta - c^{-1}}{2}\right)\right]^2 + M\left[tr\left(\frac{\delta - c^{-1}}{2}\right)^2\right] \tag{3}$$

In the above equation, M and λ are the model parameters related to the molecular weight and relaxation time respectively, and δ is a unit tensor. To ensure that this model remains volume preserving for different mobility tensors, Ramazani et al. introduced the following form for the dissipative term of the time evolution of the conformation tensor.[11]

$$\Lambda : \frac{\partial \boldsymbol{\Phi}}{\partial c} = \Lambda^1 : \frac{\partial \boldsymbol{\Phi}}{\partial c} - \frac{1}{3} tr\left(c^{-1} \cdot \Lambda^1 : \frac{\partial \boldsymbol{\Phi}}{\partial c}\right) c \tag{4}$$

Where Λ^1 could be any combination of the generalized form of the fourth order mobility tensors presented by Beris and Edwards as follows:[3]

$$\begin{aligned}\Lambda_{\alpha\beta\varepsilon\gamma} = {} & a_1 \delta_{\alpha\beta}\delta_{\gamma\varepsilon} + a_2(\delta_{\alpha\gamma}\delta_{\beta\varepsilon} + \delta_{\alpha\varepsilon}\delta_{\beta\gamma}) + a_3(c_{\alpha\beta}\delta_{\gamma\varepsilon} + \delta_{\alpha\beta}c_{\gamma\varepsilon}) \\ & + a_4(c_{\alpha\gamma}\delta_{\beta\varepsilon} + c_{\alpha\varepsilon}\delta_{\beta\gamma} + \delta_{\alpha\gamma}c_{\beta\varepsilon} + \delta_{\alpha\varepsilon}c_{\beta\gamma}) + a_5 c_{\beta\alpha}c_{\gamma\varepsilon} \\ & + a_6(c_{\alpha\gamma}c_{\beta\varepsilon} + c_{\alpha\varepsilon}c_{\beta\gamma}) + a_7(c_{\alpha\mu}c_{\mu\beta}\delta_{\gamma\varepsilon} + \delta_{\alpha\beta}c_{\gamma\mu}c_{\mu\varepsilon}) \\ & + a_8(c_{\alpha\mu}c_{\mu\gamma}\delta_{\beta\varepsilon} + c_{\alpha\mu}c_{\mu\varepsilon}\delta_{\beta\gamma} + \delta_{\alpha\gamma}c_{\beta\mu}c_{\mu\varepsilon} + \delta_{\alpha\varepsilon}c_{\beta\mu}c_{\mu\gamma}) \\ & + a_9(c_{\alpha\mu}c_{\mu\beta}c_{\gamma\varepsilon} + c_{\alpha\beta}c_{\gamma\mu}c_{\mu\varepsilon}) + a_{10}(c_{\alpha\mu}c_{\mu\gamma}c_{\beta\varepsilon} + c_{\alpha\mu}c_{\mu\varepsilon}c_{\beta\gamma} \\ & + \delta_{\alpha\gamma}c_{\beta\mu}c_{\mu\varepsilon} + c_{\alpha\varepsilon}c_{\beta\mu}c_{\mu\gamma}) + a_{11}c_{\alpha\eta}c_{\eta\beta}c_{\gamma\mu}c_{\mu\varepsilon} \\ & + a_{12}(c_{\alpha\eta}c_{\eta\gamma}c_{\beta\mu}c_{\mu\varepsilon} + c_{\alpha\eta}c_{\eta\varepsilon}c_{\beta\mu}c_{\mu\gamma})\end{aligned} \tag{5}$$

Where c_{ij} are components of the conformation tensor as well as $a_1, a_2 \ldots$ and a_{12} can be constants or functions of the scalar invariants of c.

We used above mentioned Helmholtz free energy functions with different mobility tensors; therefore a large family of volume preserving models can be defined. Four types expression which have actually investigated in this paper are specified in Table 1.

Modeling Development

Because of the shear rate dependency on the radius of die in the circular die flow, the momentum relation has been used to solve the model. Consequently the model and the relevant motion equation have been solved simultaneously.

Table 1.
Four families of the mobility tensor.

$\Lambda^1_{\alpha\beta\gamma\varepsilon}$	order 1	$\Lambda_0(c_{\alpha\gamma}\delta_{\beta\varepsilon} + c_{\alpha\varepsilon}\delta_{\beta\gamma} + \delta_{\alpha\gamma}c_{\beta\varepsilon} + \delta_{\alpha\varepsilon}c_{\beta\gamma})$
	order 2	$\Lambda_0(c_{\alpha\gamma}c_{\beta\varepsilon} + c_{\alpha\varepsilon}c_{\beta\gamma})$
	order 3	$\Lambda_0(c_{\alpha\mu}c_{\mu\gamma}c_{\beta\varepsilon} + c_{\alpha\mu}c_{\mu\varepsilon}c_{\beta\varepsilon} + c_{\alpha\gamma}c_{\beta\mu}c_{\mu\varepsilon} + c_{\alpha\varepsilon}c_{\beta\mu}c_{\mu\gamma})$
	order 4	$\Lambda_0(c_{\alpha\eta}c_{\eta\gamma}c_{\beta\mu}c_{\mu\varepsilon} + c_{\alpha\eta}c_{\eta\varepsilon}c_{\beta\mu}c_{\mu\gamma})$

We considered a cylindrical coordinate system (r, θ, z) with the z axis in the axial direction.

For a steady shear flow in the circular die, velocity components are given by:

$$V_1 = V_z = V_z(r),\ V_2 = V_r = 0,\\ V_3 = V_\theta = 0 \tag{6}$$

Therefore, the rate of strain tensor and the vorticity tensor have the following forms:

$$\dot\gamma(r) = \dot\gamma(r)\begin{pmatrix} 0 & -1 & 0 \\ -1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}\\ \omega(r) = \dot\gamma(r)\begin{pmatrix} 0 & -1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \tag{7}$$

The rate of strain tensor, $\dot\gamma(r)$, and the vorticity tensor, $\omega(r)$, are presented in a convective term of the time evolution of the conformation tensor [see equation 1].

The relevant equation of motion in z-direction are given by[12]

$$\frac{\partial P}{\partial z} + \frac{1}{r}\frac{\partial}{\partial r}(r\tau_{rz}) = 0 \tag{8}$$

Where $\frac{\partial P}{\partial z}$ is a constant, therefore,

$$\tau_{21} = \tau_{rz} = -\frac{r}{2}\frac{\partial P}{\partial z} \tag{9}$$

Finally, the model in the form of a differential equation system with the motion equation was solved using MATHEMATICA software.

Result and Discussion

The shear rate, viscosity, the first and the second normal stress differences (N_1, N_2) have been predicted by the model with four types of mobility tensor in steady shear flow and fully developed conditions. All predictions were obtained using the following set of parameters: $\Lambda_0 = 1/19500$, $M = 7000$, $\lambda = 500$, $\frac{\partial P}{\partial z} = -5000$ kpa, $R = 0.005$ m (R is radius of die)

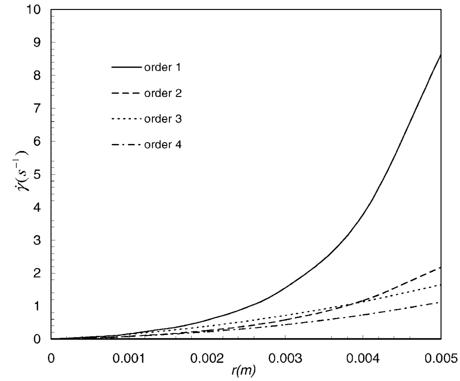

Figure 1.
Effect of the mobility tensor on the shear rate in the circular die.

Figure 1 and 2 show the effects of the mobility tensor on the shear rate and the viscosity, respectively. The predictions indicate that the choice of mobility term has a pronounced effect on the shear rate and the viscosity. It can be resulted that the flexibility in the choice of the mobility tensor permits a good fit of a whole class of experimental data presented in literature.[9–11]

The results presented in Figure 1 and 2 show that the shear rate increases with increasing the radius of die and, conversely,

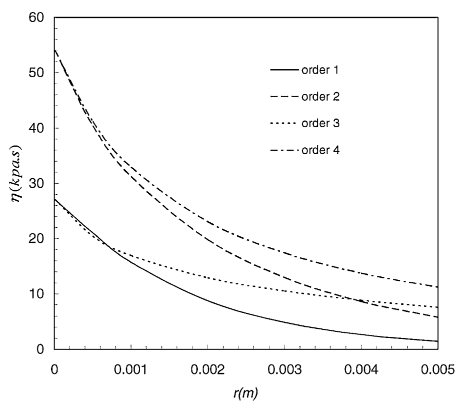

Figure 2.
Effect of the mobility tensor on the viscosity in the circular die.

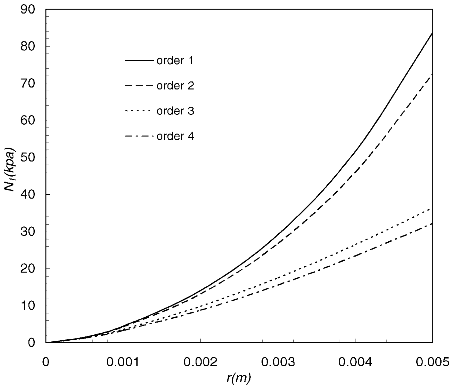

Figure 3.
Effect of the mobility tensor on the first normal stress difference.

the viscosity reduces. The model prediction for the shear rate using the first-order mobility tensor will be considerably different from the model predictions using other mobility tensors while the viscosity prediction is not so much different with the different mobility tensor across die. However for all models, viscosity reduction across of the die becomes more significant as approaching to the center of die.

Figure 3 and 4 show the model predictions for the effect of the mobility tensor term on the N_1 and N_2 in steady die flow, respectively.

The use of different expressions of the mobility tensor resulted in a strong effect

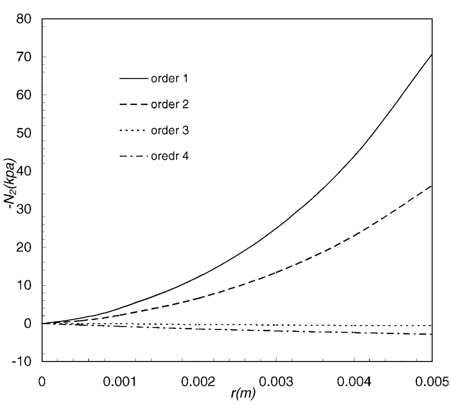

Figure 4.
Effect of the mobility tensor on the second normal stress difference.

on the first and the second normal stress differences in the die flow. When the first and the second normal stress differences compared with different mobility tensors, these show a great sensitivity to the choice of the mobility tensor.

As a result the choice of mobility tensor has a pronounced effect on the prediction of the first and the second normal stress differences.

Also the second normal stress difference increases with the first and the second-order mobility tensor despite it reduces with the third and the fourth-order mobility tensor in the die.

Conclusion

An internal microstructural model with different mobility tensors have been studied to predict of the rheological behavior of polymer melts and solutions in the circular die in steady shear flow. Moreover, how the choice of the mobility tensor will affect on the rheological behavior predictions of the polymer fluids have been evaluated in the circular die. The predicted result for the normal stress differences can be used to calculate die swell ratio by existing relations in literature.

Use of different mobility tensors cause that the model predictions can cover an extended range of rheological behaviors generally observed for polymer melts and solutions in the circular die.

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