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START-UP OF FLOW THROUGH A 4:1:4 CONSTRICTION IN A TUBE USING THE ROUSE-CCR TUBE MODEL FOR LINEAR ENTANGLED POLYMERS WITH FINITE EXTENSIBILITY

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ABSTRACT

Rouse-CCR tube model for linear entangled polymers with finite extensibility

Recently, Graham et al. [1] proposed a complete molecular theory for fast flows of entangled polymer melts that does not need decoupling approximation, which leads to the averages for orientation tensor and chain stretching. The resulting model includes the processes of reptation, convective constraint release (CCR), reptation-driven constraint release, chain stretch and contour length fluctuations (CLF). The theory, however, is too complicated to be used as such within numerical codes for complex flow simulations. Likhtman and Graham [2] derived from the full theory a simplified constitutive equation, which they called the Rolie-Poly equation, standing for Rouse linear entangled polymers. We want to emphasize here that neither theories have finite extensibility included, which would limit the degree of strain hardening in the stretching regime. In fact, non-Gaussian behavior cannot be ignored in fast flows, when chains stretch significantly. In order to account for finite extensibility of polymer chains into the original Rolie-Poly equation, we require that, in the absence of any other mechanisms, the trace of the original equation leads to the relaxation for the stretch similar to the MLD model [3]. We then write the non-Gaussian Rolie-Poly constitutive equation, which account for finite extensibility of polymer chains, in the form

$$\frac{\partial \boldsymbol{\sigma}}{\partial t} + (\boldsymbol{u} \cdot \nabla)\boldsymbol{\sigma} = \boldsymbol{L} \cdot \boldsymbol{\sigma} + \boldsymbol{\sigma} \cdot \boldsymbol{L}^T + \boldsymbol{f}(\boldsymbol{\sigma}), \qquad (1)$$

where $\boldsymbol{L} = \nabla \boldsymbol{u}^T$ is the transpose of velocity gradient tensor and the tensor function, $\boldsymbol{f}$, is now given by

$$\boldsymbol{f}(\boldsymbol{\sigma}) = -\frac{1}{\tau_d}(\boldsymbol{\sigma} - \boldsymbol{\delta}) - \frac{2}{\tau_R} k_s(\lambda)\left(1 - \sqrt{\frac{3}{tr\boldsymbol{\sigma}}}\right)\left(\boldsymbol{\sigma} + \beta\left(\frac{tr\boldsymbol{\sigma}}{3}\right)^{\delta}(\boldsymbol{\sigma} - \boldsymbol{\delta})\right). \tag{2}$$

Here τ_d is the fixed-tube disengagement time or reptation time, τ_R is the longest Rouse time or stretch time, β is the CCR coefficient analogous to the coefficient introduced by Marrucci in his original CCR paper [4], δ a negative power which can be obtained by fitting to the full theory, $\lambda = \sqrt{tr\boldsymbol{\sigma}/3}$ is the chain stretch ratio and $k_s(\lambda)$ is the nonlinearity of the spring coefficient accounting for the finite extensibility of polymer chains, equals unity for linear springs and becomes much greater than unity as the spring becomes nearly fully stretched. In the limit of large stretch, in the absence of any other mechanisms, retraction (the trace of Eq. 1) leads to the desired following relaxation for the stretch

$$\frac{d\lambda}{dt} = -\frac{1}{\tau_R} k_s(\lambda)(\lambda - 1). \tag{3}$$

This form was used in the MLD model with finite extensibility [3]. Note that in the limit of linear spring, $k_s(\lambda)$ remains unity, Eqs. 1-2 reduce to the original Rolie-Poly constitutive equation. The nonlinear spring coefficient is approximated by the normalized Padé inverse Langevin function [3], i.e.,

$$k_s(\lambda) = \frac{(3 - \lambda^2/\lambda_{\max}^2)(1 - 1/\lambda_{\max}^2)}{(1 - \lambda^2/\lambda_{\max}^2)(3 - 1/\lambda_{\max}^2)}, \tag{4}$$

where, λ_{max}, is the maximum stretch ratio. In extensional flows at large strain rates, when λ approaches $\lambda_{\max}$, the linear spring, k_s, grows rapidly, approaching a singularity. Thus, the effective stretch relaxation time, $\tau_{eff} = \tau_R/k_s$, is considerably reduced and the evolution equation for the conformation tensor $\boldsymbol{\sigma}$ becomes stiff. The constitutive equation is completed by specifying the relationship between the polymer stress $\boldsymbol{\tau}_p$ and the conformation tensor $\boldsymbol{\sigma}$. For the non-Gaussian chain we write

$$\boldsymbol{\tau}_p = \frac{\eta_0}{\tau_d} k_s(\lambda)(\boldsymbol{\sigma} - \boldsymbol{\delta}), \tag{5}$$

where η_0 is the zero-shear rate viscosity. As noted by Ghosh et al. [5], the inverse Langevin force law, obtained from equilibrium statistical mechanics, should be used with caution in nonequilibrium situations. Only when the time scale of the deformation is much longer than the relaxation time scale of the entire chain will the chain unravel reversibly. In strong extensional flows, this condition is violated, and stress-birefringence hysteresis is observed.

Matrix-logarithm of the conformation tensor

There is growing numerical evidence that catastrophic breakdown of viscoelastic problems may be caused by the failure of polynomial based approximations to properly represent exponential profiles developed by the conformation tensor near stagnation points or in regions of high deformation rate. Fattal and Kupferman [6] resolved that difficulty through the transformation of differential constitutive models into an equation for the matrix logarithm of the conformation tensor, which we now give in the general context of the Rolie-Poly model with finite extensibility.

To derive an evolution equation for the logarithm of the conformation tensor $\boldsymbol{\sigma}$, which we denote, $\boldsymbol{\psi} = \log \boldsymbol{\sigma}$, we follow the method of Hulsen et al. [7] based on the evolution of the principal axes of the deformation tensor and derive the equations in the tensorial form. The diagonalizing transformation of the conformation tensor $\boldsymbol{\sigma}$, can be written as

$$\boldsymbol{\sigma} = \boldsymbol{R}\tilde{\boldsymbol{\sigma}}\boldsymbol{R}^{\mathrm{T}} = \boldsymbol{R}\begin{pmatrix} \tilde{\sigma}_{11} & 0 & 0 \\ 0 & \tilde{\sigma}_{22} & 0 \\ 0 & 0 & \tilde{\sigma}_{33} \end{pmatrix}\boldsymbol{R}^{\mathrm{T}}. \tag{6}$$

Here $\tilde{\boldsymbol{\sigma}}$ is a diagonal matrix whose diagonal elements, $\tilde{\sigma}_{\mathrm{kk}}$, are eigenvalues of $\boldsymbol{\sigma}$ and $\boldsymbol{R}$ is an orthogonal tensor whose columns are eigenvectors of $\boldsymbol{\sigma}$. The matrix logarithm of the conformation tensor can then be written as

$$\boldsymbol{\psi} = \log \boldsymbol{\sigma} = \boldsymbol{R}(\log \tilde{\boldsymbol{\sigma}})\boldsymbol{R}^{\mathrm{T}}, \tag{7}$$

form which we derive (after some manipulations) the evolution equation for the matrix logarithm of the conformation tensor, $\boldsymbol{\psi}$, in the following form

$$\dot{\psi}_{\mathrm{ij}} = \mathrm{R}_{\mathrm{ik}}\tilde{\mathrm{N}}_{\mathrm{km}}\mathrm{R}_{\mathrm{jm}}. \tag{8}$$

The diagonal and off-diagonal components of the tensor $\tilde{\boldsymbol{N}}$ are defined, respectively, by

$$\tilde{N}_{mm} = 2\tilde{L}_{mm} + \tilde{\sigma}_{mm}^{-1}\tilde{f}_{mm}, \tag{9}$$

and

$$\tilde{N}_{km} = \frac{\log\tilde{\sigma}_{mm} - \log\tilde{\sigma}_{kk}}{(\tilde{\sigma}_{mm} - \tilde{\sigma}_{kk})}\left[(\tilde{L}_{km}\tilde{\sigma}_{mm} + \tilde{L}_{mk}\tilde{\sigma}_{kk})\right], \text{ with } \mathrm{k} \neq \mathrm{m}. \tag{10}$$

In the limit that $\tilde{\sigma}_{\mathrm{mm}} \to \tilde{\sigma}_{\mathrm{kk}}$, we get from Eq. (10)

$$\tilde{N}_{km} = (\tilde{L}_{km} + \tilde{L}_{mk}), \text{ with } \mathrm{k} \neq \mathrm{m}. \tag{11}$$

Finally, the equations governing the conservation of mass and transport of momentum are, for viscoelastic incompressible flow,

$$\nabla \cdot \boldsymbol{u} = 0, \tag{12}$$

$$-\nabla \mathrm{p} + \nabla \cdot (2\eta_{\mathrm{s}} \boldsymbol{d} + \boldsymbol{\tau}_{\mathrm{p}}) = \rho \left[\frac{\partial \boldsymbol{u}}{\partial \mathrm{t}} + (\boldsymbol{u} \cdot \nabla)\boldsymbol{u} \right]. \tag{13}$$

Here $\boldsymbol{u}$ is the fluid velocity, ρ the fluid density and p the hydrodynamic pressure. The extra stress tensor has been split into a polymeric contribution $\boldsymbol{\tau}_{\mathrm{p}}$ and a solvent contribution with η_{s} the solvent viscosity and $\boldsymbol{d}$ is the rate-of-deformation tensor defined by

$$\boldsymbol{d} = \frac{1}{2}(\nabla \boldsymbol{u} + \nabla \boldsymbol{u}^{\mathrm{T}}). \tag{14}$$

The problem is now to solve the set of nonlinear partial differential equations consisting of Eqs. (12)-(14) and (8) with appropriate boundary conditions.

At each time step the resolution is performed in a decoupled fashion using an iterative scheme. The momentum and the continuity equations are first solved as a saddle-point problem, with the viscoelastic stress treated as a given body force, and the constitutive equation is then integrated with known kinematics. Implicit Euler approximation to the time derivative in the momentum and constitutive equations is used. A fixed-point algorithm is used to iterate between the solution of the momentum and constitutive equations at each time step, thereby making the overall algorithm coupled in a segregated manner. The stabilized discrete elastic viscous stress splitting (DEVSS-G) method [8] and stabilized finite element methods (GLS, SUPG) are used to carry out three-dimensional time-dependent simulations. Such stabilized method allows the use of velocity and pressure interpolants that do not satisfy the Babuska-Brezzi condition such as the linear equal order interpolation functions. These elements are both computationally effective and easy to implement, especially for three-dimensional applications. The resulting formulation is well posed. Computations are conducted using our parallel computation framework. The practical utility and effectiveness of the proposed numerical scheme is demonstrated by solving fully three-dimensional constriction flow.

Uniaxial extensional flow

We here perform a quantitative comparison with the original Rolie-Poly model. We further include in these comparisons results of the MLD model from Ye et al. [3] and experiments [3]. The polymer used is a nearly monodisperse linear polystyrene sample (L289) [3]. The maximum stretch ratio was adjusted arbitrarily to 10. The steady state values of the extensional stress are shown in Fig. 1. In addition, we show the prediction of the MLD model. At low strain rate for which $\dot{\varepsilon}\tau_R \leq 1$, predictions are in good agreement with experimental data. In the stretching regime, $\dot{\varepsilon}\tau_R > 1$, the original Rolie-Poly fails, since it omits chain finite extensibility effects. Comparison with original Rolie-Poly model for the steady state extensional viscosity is shown in Fig. 2. As noted by Ye et al. [3], the initial drop from the Newtonian value is due to tube orientation in the elongation direction. As strain rates increase ($\dot{\varepsilon}\tau_R > 1$), chain stretch makes the viscosity increase. It is shown in Fig. 2 that the extensional viscosity is

constant since stress is linear in the rate of deformation. The extensional viscosity reaches a final constant value due to finite extensibility.

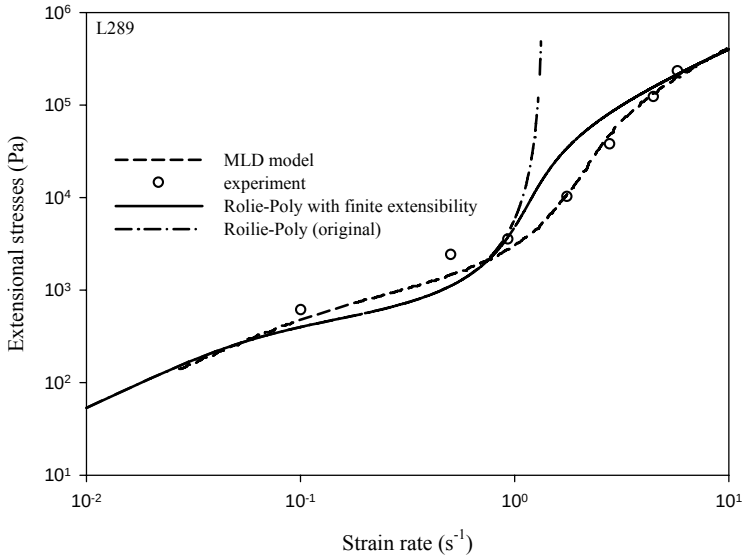


Fig. 1. Prediction of steady state extensional stress compared to original Rolie-Poly model, MLD model [3] and experimental data [3].

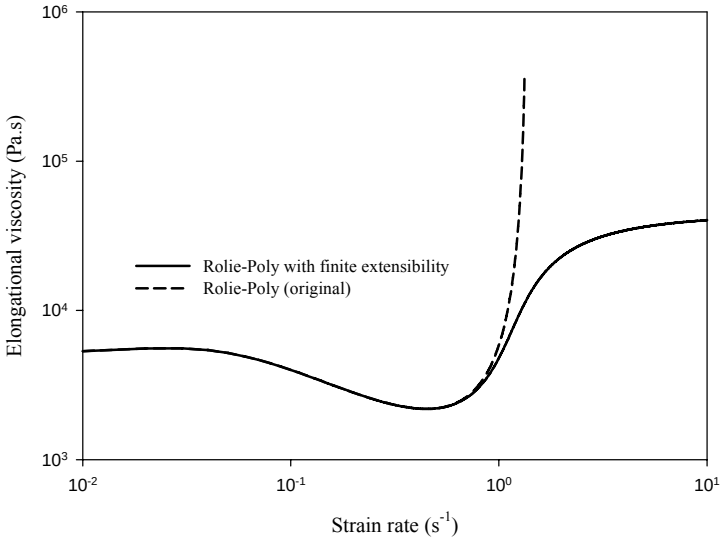
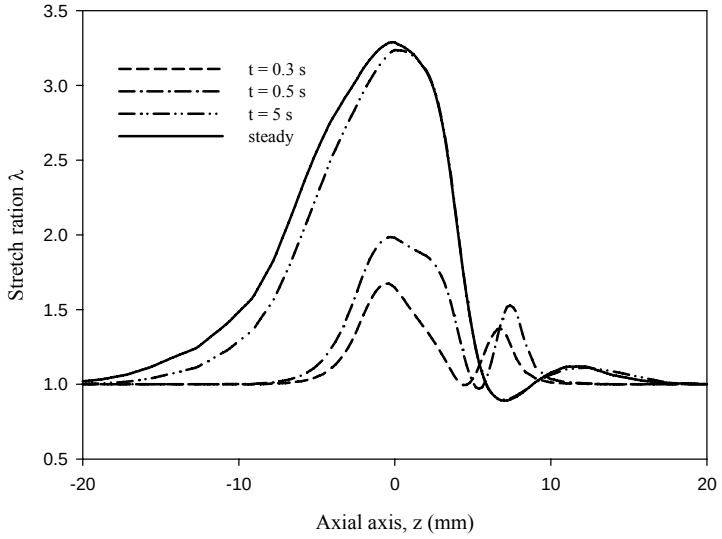


Fig. 2. Steady-state extensional viscosity predictions of the Rolie-Poly models with and without finite extensibility.

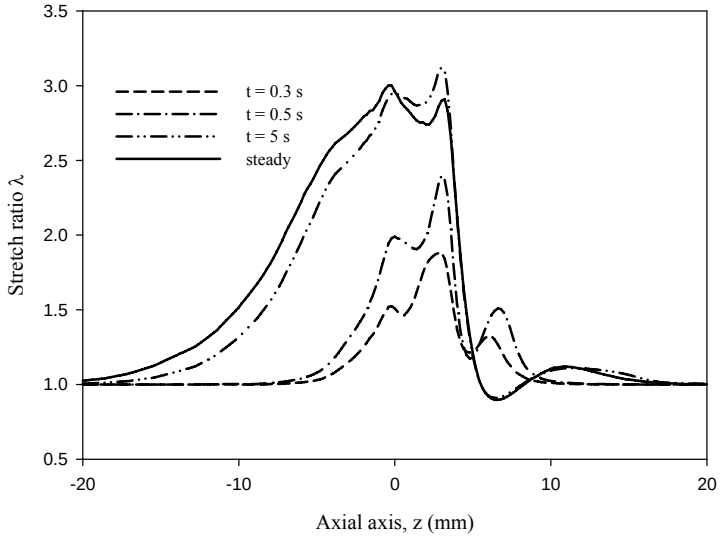
Star-tup of flow through an axisymmetric 4:1:4 constriction

The material used is the same monodisperse polystyrene used in the uniaxial extensional flow. The maximum stretch ratio was arbitrarily reduced to 5. The choice of a small value was made in order to mimic dissipative stress after only a small degree of stretch. Starting from rest and using time-independent (static) inlet boundary conditions, we obtained results through a true three-dimensional transient development. We are therefore able to capture possible flow bifurcations and all the important flow

features. The temporal evolution of the stretch ratio and the vortex are shown in Figs. 3 and 4. The overshoot in the stretch is clearly exhibited in Fig. 3(b) in the constriction region, dominated by strong shear flow. We also observe that the stretch ratio becomes smaller than unity downstream of the constriction, as a result of biaxial extension and relaxation. There is also a clear evidence of a lip vortex growth (Fig. 3, t = 0.3 s and 0.5 s), stretching from re-entrant corner. Our simulations suggest that inertia is not an essential ingredient for the appearance of the lip vortex in this constriction flow.



(a)



(b)

Fig. 3. Transient and steady-state stretch ratio during the start-up flow through an axisymmetric 4:1:4 constriction: (a) along the center line and (b) near the wall. z=0 corresponds to the upstream re-entrant corner.

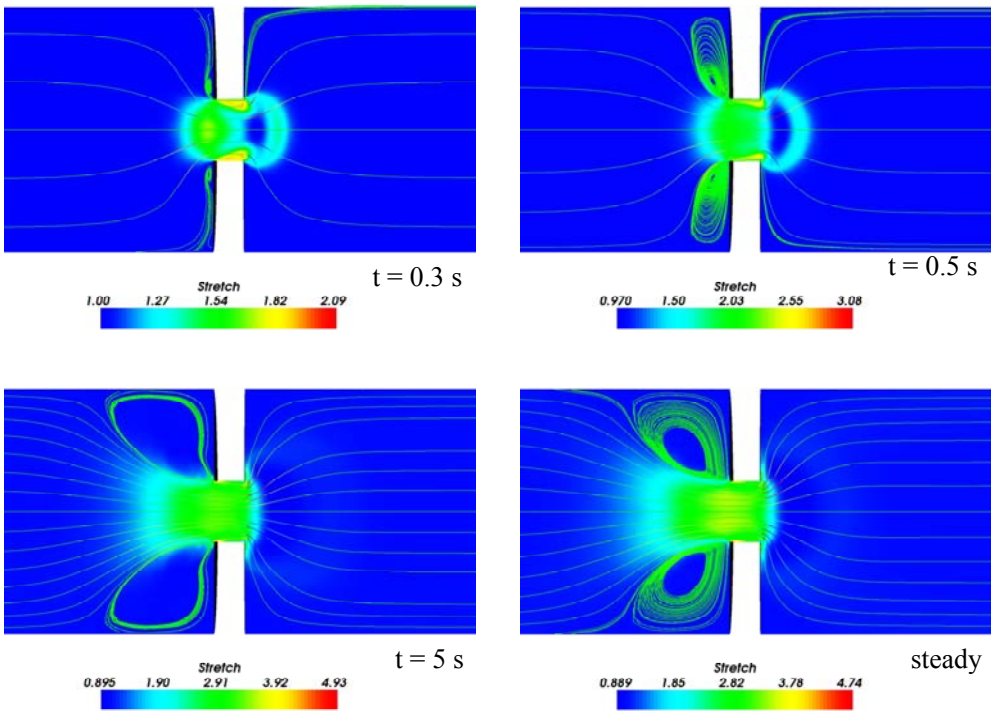


Fig. 4. Temporal evolution of the stretch ratio and vortex during the start-up flow.

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