Numerical simulation of transient flow of polymer melts

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1 Introduction

For describing the rheological behaviour of entangled polymers like polymer melts and concentrated solutions, the reptation theory [1] has become a basic tool. It is based on the conceptually simple idea that a polymer chain can move more easily in the direction of its backbone than perpendicular to it, because in that direction the motion is hindered by neighbouring polymer chains. The constraints resulting from the neighbouring chains effectively confine the movements of a polymer chain to a surrounding tube-like region. Although successful in predicting, for example, the damping function and the plateau modulus of linear viscoelasticity, the model also shows some deficiencies like an excessive shear thinning in fast shearing flows. Recent progress in the modeling of polymer melts has alleviated the shortcomings in the Doi–Edwards model. Recent modifications of the original model include for example convective constraint release and fulfilling a force balance on the nodes [2], [3]. With these modifications, a better agreement with experimental data of the shear stress and normal stress ratio could be obtained. How the improved model behaves in a more complex flow, however, is still an open question.
Below, we discuss the behaviour of the new model in a benchmark complex flow, the 4:1:4 constriction flow. For this, we have extended the Backward-tracking Lagrangian Particle Method [5], previously developed for computing differential constitutive equations and kinetic theory models for dilute solutions, to handle constitutive equations of integral type that result from reptation theory.

2 Governing equations

The usual conservation laws of mass and momentum for incompressible and isothermal viscoelastic flow are,

$$\nabla \cdot \boldsymbol{v} = 0, \tag{1}$$

$$\rho \frac{D\boldsymbol{v}}{Dt} = -\nabla p + \nabla \cdot (2\eta_s \boldsymbol{d} + \boldsymbol{T}), \tag{2}$$

where ρ is the fluid density, $\boldsymbol{v}$ the fluid velocity, p the hydrodynamic pressure, and D/Dt denotes the material derivative. The extra-stress tensor has been split in a polymeric contribution $\boldsymbol{T}$ and a solvent contribution with η_s the solvent viscosity and $\boldsymbol{d} = (\boldsymbol{\kappa} + \boldsymbol{\kappa}^T)/2$ the rate-of-deformation tensor, where $\boldsymbol{\kappa}^T$ denotes the velocity gradient. The extra-stress tensor $\boldsymbol{T}$ may either be obtained by a micro or macrorheological model. Here, we consider macrorheological models for polymer melts. In that case the polymeric stress is governed by either an integral or differential equation.
For integral models, the polymeric stress can be written in the general form

$$\boldsymbol{T} = G \int_{-\infty}^{t} \mu(t;t')\boldsymbol{Q}(t;t')\ \mathrm{d}t', \tag{3}$$

where G is the shear modulus, μ a memory function that may depend on the flow conditions and $\boldsymbol{Q}$ denotes an orientation tensor of the tube segments. The memory function μ takes the general form

$$\mu(t;t') = \frac{1}{\tau(t')} \exp\left(- \int_{t'}^{t} \frac{\mathrm{d}t''}{\tau(t'')} \right), \tag{4}$$

where τ is a relaxation time that may depend on the flow. In the Doi–Edwards model, τ equals the reptation or disengagement time τ_d. As τ_d is a constant, the integral can be calculated analytically and only the integral in Eq. (3) remains to be computed.
Recently, Ianniruberto and Marrucci [2] have introduced in the reptation model the idea that constraints surrounding a polymer chain are more rapidly swept away in fast flows. To take into account this so-called convective constraint release (CCR), they proposed to take for the overall relaxation time τ

$$\frac{1}{\tau} = \frac{1}{\tau_d} + \frac{\beta}{G} \boldsymbol{\kappa} : \boldsymbol{T}, \tag{5}$$

where β is a positive numerical coefficient that ensures an increasing shear stress as a function of

shear rate. Note that τ is approximately equal to τ_d for slow flows, and that only for fast flows the relaxation time is decreased considerably. An initially overlooked drawback of Eq. (5) is the non-positiveness of the stress work $\boldsymbol{\kappa} : \boldsymbol{T}$, which may lead to negative relaxation times. For this we also consider an ad hoc alternative as suggested to us by Marrucci [4] that guarantees $0 < \tau \leq \tau_d$,

$$\frac{1}{\tau} = \frac{1}{\tau_d} + \frac{\beta}{2G} \left(\boldsymbol{\kappa} : \boldsymbol{T} + |\boldsymbol{\kappa} : \boldsymbol{T}| \right). \tag{6}$$

In the Doi–Edwards model, the orientation tensor $\boldsymbol{Q}$ equals the average orientation of the tube segments. In a recent paper, Marrucci [3] argued that a force balance on the nodes should be fulfilled, resulting in a modified $\boldsymbol{Q}$ tensor. In this manner, it was possible to obtain a better agreement with experimental data for the normal stress ratio. The modified $\boldsymbol{Q}$ tensor is given by

$$\boldsymbol{Q} = \frac{\sqrt{\boldsymbol{B}}}{\mathrm{tr}\,\sqrt{\boldsymbol{B}}}, \tag{7}$$

where $\boldsymbol{B}$ denotes the Finger tensor that is a measure of the deformation of a fluid element and fulfills the evolution equation

$$\frac{D\boldsymbol{B}}{Dt} = \boldsymbol{\kappa} \cdot \boldsymbol{B} + \boldsymbol{B} \cdot \boldsymbol{\kappa}^T. \tag{8}$$

As integral models are more computationally intensive than differential equations, a differential approximation of Eqs. (3), (4), and (8) has been derived [3]. The resulting equation is a constitutive equation for the square of the stress,

$$\begin{aligned} \frac{D\boldsymbol{T}^2}{Dt} &= \boldsymbol{\kappa} \cdot \boldsymbol{T}^2 + \boldsymbol{T}^2 \cdot \boldsymbol{\kappa}^T - 2\boldsymbol{T}^2 \left(\boldsymbol{\kappa} : \boldsymbol{T}/G \right) \\ &\quad - \frac{2}{\tau} \left(\boldsymbol{T}^2 - \frac{G}{3} \boldsymbol{T} \right) \end{aligned} \tag{9}$$

where the total relaxation time τ is again given by Eq. (5) or (6).

3 Numerical method

In Lagrangian particle methods, at each time step, the Eulerian solution of the conservation equations is decoupled from the Lagrangian computation of the polymer stress. In this manner, we allow for different solution methods well-suited for diffusion-dominated equations and transport equations, respectively.

The Eulerian form of the equations of motion (1), (2) are discretised with the aid of the finite element method. To increase the stability of the numerical scheme, the well-known Discrete Elastic-Viscous Stress Splitting (DEVSS) method [6] has been used. DEVSS involves a separate discretisation of the velocity gradient, which is obtained by projecting the piecewise discontinuous finite element velocity gradient $\nabla \boldsymbol{u}$ on a continuous linear field. In the momentum equation, an extra stabilizing term is then included that contains the difference between $\nabla \boldsymbol{u}$ and its projection, multiplied with an auxiliary viscosity $\overline{\eta}$. For reasons of efficiency, the resulting matrix-vector equation is solved with the aid of an LU factorisation, which only has to be computed once, before the start of the actual flow simulation.

For the computation of the stress integral (3), we use the deformation field method as proposed in [7]. This method involves a large number of N_d deformation fields, for the Marrucci model that we consider here the Finger tensor fields $\boldsymbol{B}_i$ with $i = 1, \cdots, N_d$, measuring the deformation between current time t and various reference times t'_i in the past. At every time step all fields age by Δt, and for all N_d fields $\boldsymbol{B}_i$ the evolution equation (8) is solved. Instead of solving a double integral, which is computationally expensive, we solve the equivalent evolution equation for μ

$$\frac{D\mu}{Dt}(t; t') = -\frac{\mu(t; t')}{\tau}, \tag{10}$$

with initial condition

$$\mu(t'; t') = \frac{1}{\tau(t')}. \tag{11}$$

This is similar to the approach of Peters *et al.* [8] for the Mead–Larson–Doi model. In case of the Marrucci model the evolution equation for the memory function becomes

$$\begin{aligned} \frac{D\mu^{CCR}}{Dt} &= -\mu^{CCR} \frac{\beta}{G} \boldsymbol{\kappa} : \boldsymbol{T}, \\ \mu^{CCR}(t'; t') &= 1 + \tau_d \frac{\beta}{G} \boldsymbol{\kappa} : \boldsymbol{T}, \end{aligned} \tag{12}$$

where we have split the constant part and the contribution of convective constraint release to the memory function, $\mu = \mu^{CST} \mu^{CCR}$, with $\mu^{CST}(t', t) = \exp\left(-(t - t')/\tau_d\right)/\tau_d$. In case we use Eq. (6) instead of Eq. (5), the evolution equation and initial condition (12) are modified accordingly.

The evolution equations for the Finger tensor fields $\boldsymbol{B}_i$ are solved by means of a Lagrangian method, the Backward-tracking Lagrangian Particle Method (BLPM) [5]. For Lagrangian particle methods the transport equations, Eqs. (8), (12) for the integral and Eq. (9) for the differential model, are solved along the trajectories of the Lagrangian particles that are convected by the flow. In the Lagrangian Particle Method (LPM) [9] particles are dropped in the flow at initial time, and next these particles are convected by the flow through the whole flow domain. A drawback of LPM is that a large amount of particles is needed

in highly graded meshes, resulting in excessive memory and CPU requirements.
To circumvent these problems, in BLPM a small number of particle locations at the current time are specified a priori in each element of the mesh. This results in a significant reduction of memory and CPU time requirements, which is particularly important for calculations with integral models. Here, we take for the fixed particle locations the nodal points of a quadratic discontinuous finite element representation. At each time step, the particle trajectories leading to these nodes are calculated by tracking one time step Δt back in time. At the obtained initial positions, the quantity to be integrated has to be initialised. This is performed by interpolation from the stored finite element field at the corresponding time level $t-\Delta t$. Finally, to obtain the values at the fixed particle positions, the evolution equations are integrated with a semi-implicit predictor-corrector scheme. Further details about this numerical method are given in [5].

4 Results

As a complex flow geometry we consider the flow through a planar 4:1:4 constriction with rounded corners. Around the constriction, the geometry and mesh for which we show numerical results, are displayed in Fig. 1. The mesh consists

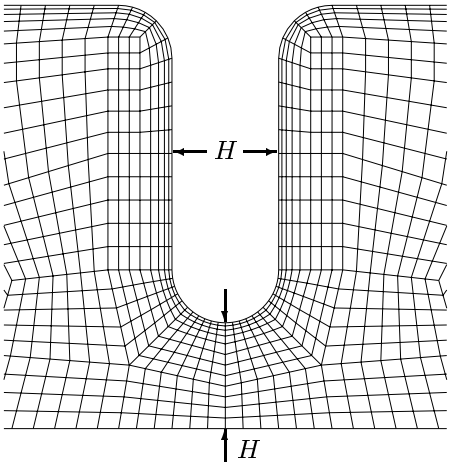

Figure 1: Mesh and geometry specification of the 4:1:4 constriction flow with rounded corners.

of 1288 quadrilateral finite elements. Note the fine meshing near the rounded constriction wall, where very large stress gradients develop for high Weissenberg flows.
To guarantee an increasing shear viscosity with increasing shear rate, we employ $\beta = 3.8$ as proposed by [3]. We only consider creeping flow, so that in absence of a solvent viscosity, we have one characteristic number. For this Weissenberg number, which is a measure for the amount of elasticity in the flow, we take $We = \tau_d U/H$. The characteristic velocity U is taken as the average velocity at the smallest gap space H. For the DEVSS method we take the auxiliary viscosity $\overline{\eta} = \eta_p$.
For the integral model using $N_d = 100$ deformation fields, the computer requirements are 130 MB memory consisting of approximately 75 MB for the 100 deformation fields and 40 MB to store the LU factorisation in the solver of the momentum equation. The CPU time was 1.5 hours per 1000 iterations on a 533 MHz ev56 processor of a DEC Alpha workstation, which results in an overall CPU time of 15 hours per run for the used time step $\Delta t = 10^{-3}$ and final time $T = 10$. The differential approximation only contains one field instead of the 100 deformation fields, so needing considerably less memory, approximately 50 MB including the 40 MB for the LU factorisation. The CPU time was typically 30 minutes for 1000 iterations, resulting in an overall CPU time of about 5 hours per run for the used $\Delta t = 10^{-3}$ and $T = 10$.
For the integral Marrucci model, the isolines for $\beta\boldsymbol{\kappa} : \boldsymbol{T}/G$, the extra contribution to the overall relaxation time due to convective constraint release, are displayed in Fig. 2.

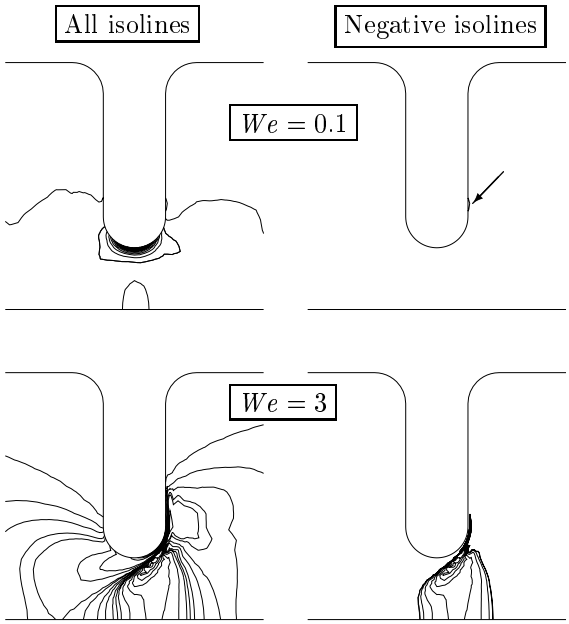

Figure 2: Isolines of $\beta\boldsymbol{\kappa} : \boldsymbol{T}/G$ for integral Marrucci model at low and medium Weissenberg number; for clarity negative isolines are also displayed separately on the right-hand side; $We = 0.1$: [-0.002,0.163], $We = 3$: [-4.89,48.4].

At the low Weissenberg number, the flow is almost Newtonian. Consequently, the isolines are almost symmetric in the flow direction, and the CCR contribution practically remains positive everywhere, since $\boldsymbol{\kappa} : \boldsymbol{T} \simeq 2G\tau\boldsymbol{\kappa} : \boldsymbol{\kappa}$, which is pos-

itive by definition. At the higher Weissenberg number, however, the situation changes dramatically. The maximum is shifted downstream along the wall of the constriction. A large region of negative values develops in the expansion part of the flow, extending from the axis of symmetry till close to the wall. Note that the maximum negative values are very close to the maximum positive values at the wall, leading to very large gradients. Figure 3 shows the impact of the enforcement (6) of positiveness in the CCR contribution of the re-

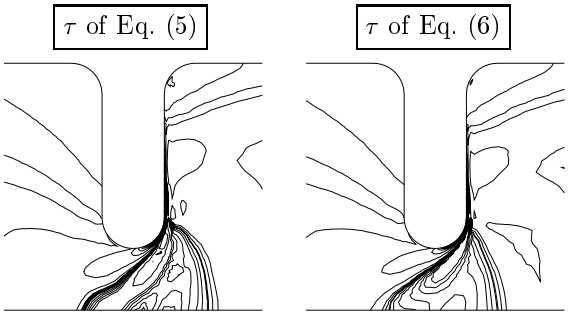

Figure 3: Isolines of $\beta\boldsymbol{\kappa} : \boldsymbol{T}/G$ for Marrucci differential approximation at $We = 10$; τ of Eq. (5): [-20.5, 109.0], τ of Eq. (6): [-10.6, 94.1].

laxation time. Particularly in and near the region of negative isolines, the solution differs considerably. We remark in passing that a comparison at this Weissenberg number was not possible for the integral model, because the numerical scheme diverged due to negative relaxation times when using Eq. (5). Thus, further theoretical developments seem to be necessary to properly include a negative $\boldsymbol{\kappa} : \boldsymbol{T}$ in convective constraint release.
A comparison between the CCR contribution to the relaxation time for the Marrucci integral model and its differential approximation at $We = 10$ is provided in Fig. 4. Although, there are some

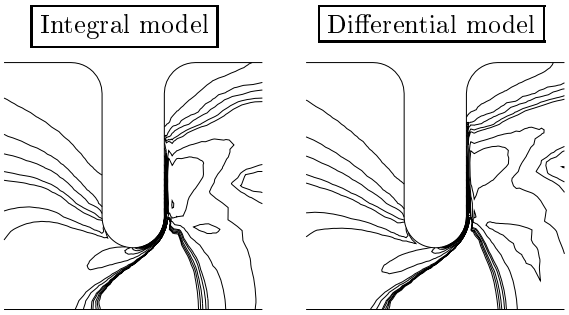

Figure 4: Isolines of $\beta\boldsymbol{\kappa} : \boldsymbol{T}/G$ for integral Marrucci model and its differential approximation at $We = 10$ using Eq. (6); integral model: [0, 112], differential approximation: [0, 94.1].

differences in the numerical values, the overall pattern of the isolines is very similar, even near the steep boundary layer downstream at the constriction wall. Concluding, the differential model is a very good approximation of the integral version, even in complex flow at high Weissenberg numbers.

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