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A COMBINED NUMERICAL/EXPERIMENTAL METHOD FOR ESTIMATING PARAMETERS AND STRUCTURE OF VISCOELASTIC CONSTITUTIVE EQUATIONS USING COMPLEX FLOWS

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Abstract: A combined numerical/experimental method is described that is used for evaluation of viscoelastic constitutive equations. The method is applied to a three dimensional stagnation flow which exhibits strong elongational deformations. The material investigated is a polymer solution (2.5% Pib/C14). Laser Doppler Anemometry and flow induced birefringece are used to measure respectively pointwise velocities and linewise (in the depth of the flow cell) stresses. Since numerical simulations of 3D viscoelastic flows are to massive yet, the deformation history of a fluid element is calculated with the viscous Carreau Yasuda model and this history is used to calculate the stresses with the viscoelastic Giesekus and PTT model. The optical birefringence signal is calculated from the integrated stresses, using the *small retardance approximation*. The approach is first tested for a slit flow (pure shear). For both models, numerical results agree well with measurements. For the stagnation flow, stresses along streamlines near the inflow and outflow symmetry plane are calculated. Using the parameters determined in simple shear flow, both viscoelastic models predict much too low integrated normal stresses in the vicinety of the outflow centreline when compared with measured values. It is demonstrated that with a proposed new constitutive model both, viscometric data and the strong elongational flow characteristics can be described at the same time.

1 Introduction

The parameters of constitutive equations for viscoelastic fluids are usually determined in simple shear only, as measurements in elongational flows are difficult to perform. Many constitutive models can fit these viscometric data equally well, though they predict different behaviour in extensional flows. It is thought that a solution to this problem is the evaluation of the constitutive equations in complex flows which are reasonably well to realize, but harder to analyse than elongational

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flows. However, the development of reliable numerical tools make this type analyses of complex viscoelastic flows possible ((1),(2)). Besides, as these complex flows also contain (Lagrangian transient) mixed shear and elongational deformation they are even a more severe test for the constitutive equations then separate shear and elongation experiments.

The purpose of this study is the development of a combined experimental/numerical method that enables to test constitutive equations in complex flows. Figure 1 shows schematicallly our approach. Basically, the approach is the same as others used in our group to characterize anisotropic, inhomogenuous materials ((3),(4),(5)). However, in this study the residues between locally measured and predicted properties are primarily used to evaluate or even adjust constitutive models, instead of obtaining an optimal parameterset.

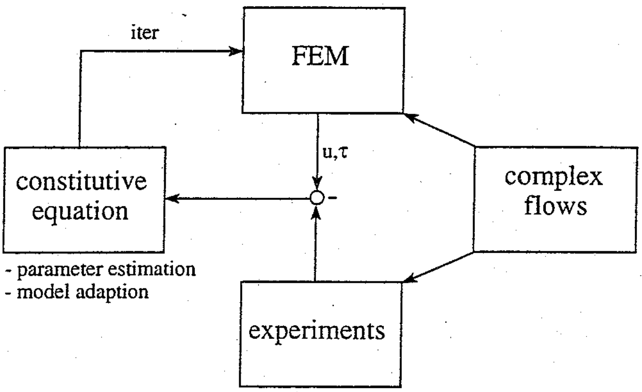

Figure 1. Schematic representation of the mixed experimental-numerical method.

The flow of a polymer solution in a stagnation die, which exhibits high elongational strains is investigated. Due to the walls of the flow cell, also shear strains will be created. Pure elongational flow is only present at the centre of the device.

Optical techniques are used to study the behavior of polymeric liquids locally. Laser Doppler anemometry (LDA) (6) to determine pointwise the velocities in the complex flow field and, following the approach of Burghardt (7), flow induced birefringence (FIB) to determine the stresses. The polymeric liquid investigated is rheologically characterized in simple shear flow first. The parameters of two nonlinear constitutive models (PTT and Giesekus) are estimated with oscillatory shear data and steady shear data. Also the parameters of a nonlinear viscous model (Carreau Yasuda) are fitted on steady shear data.

Since three dimensional finite element calculations with nonlinear viscoelastic models are to extended yet, the calculation of velocities and stresses are done separately. The flow field has been computed with the finite element package SEPRAN (8), using the Carreau Yasuda model. The velocity gradients determined are used to calculate the three dimensional stresses with the nonlinear viscoelastic models.

This approach is first tested in a fully developed rectangular slit flow (pure shear) (section 3). Then, calculations and measurements are performed on the stagnation die (section 4). In section 5 a new model is introduced and compared with measurements. Finally, the conclusions are summarized in the last section.

2 Methods and materials

2.1 Laser Doppler anemometry

Velocities were measured with a dual beam Laser Doppler anemometry system (Dantec, flow velocity analyzer 58N20), equiped with a 300 mW argon ion laser and fiber optics. The system was operated in backscatter mode, and a single velocity component was measured with the green beam ($\lambda = 514.5[nm]$). The measuring volume dimensions are ($50 \cdot 50 \cdot 200 \mu m$) and the resolution of the measured velocity field for this system is 0.13 [mm/s].

2.2 Flow induced birefringence

Flowing polymer liquids are optically anisotropic. Using the Rheo Optical Analyzer (ROA) developed by Fuller (9), the phase retardation δ and the orientation of the optic axis χ for a birefringent medium can be measured simultanuously. With the empirical stress optic rule (10), which correlates the deviatoric parts of the refractive index tensor ($\hat{\mathrm{n}}$) and the stress tensor ($\hat{\sigma}$) within a polymer flow, stresses can be calculated:

$$\hat{\mathrm{n}} = C\hat{\sigma} \tag{1}$$

where C is the stress optical coefficient. When the retardation and orientation angle are assumed constant throughout the flow cell, i.e. planar flow, the measured quantities can directly be converted into shear stress (τ_{xy}) and first normal stress difference ($N_1 = \tau_{xx} - \tau_{yy}$) (11). However, in a flow cell, three dimensional effects will always be present. Therefore the first normal stress difference and the shear stress will not be constant over the depth of the flow cell. Following Li and Burghardt (7) it can be shown that two intensities are measured for materials with a small total retardation (*small retardance approximation*).

$$R_1 = -\frac{2\pi C}{\lambda} \int N_1(z) dz \tag{2}$$

$$R_2 = \frac{4\pi C}{\lambda} \int \tau_{xy}(z) dz \tag{3}$$

This can be compared to calculations by integration of the local stresses along the depth of the flowcell (direction of the FIB lightbeam). The spatial resolution of the measuring system is 0.4 mm.

2.3 Constitutive equations

Generalized Newtonian

A generalized Newtonian model (Carreau Yasuda) (12) is used to fit the rheological steady shear data:

$$\eta = \eta_0 \left(1 + \left(\lambda_{cy}\sqrt{II_D}\right)^a\right)^{\frac{n-1}{a}} \tag{4}$$

Baaijens (11) showed that this model can accurately describe the velocity field of the flow past a cylinder of PIB/C14 solutions , indicating that, for these fluids, there is no strong influence of elongational stresses on the velocity field.

Differential viscoelastic constitutive equations

Two multimode differential constitutive equations are evaluated, both stemming from a molecular background (12). The Giesekus model is given by:

$$\lambda_i \overset{\nabla}{\tau_i} + \frac{\alpha}{G_i}(\tau_i \cdot \tau_i) + \tau_i = 2\eta_i D \tag{5}$$

and the PTT model by:

$$\overset{\nabla}{\tau_i} + \xi(D \cdot \tau_i + \tau_i \cdot D) + \frac{1}{\lambda_i}\tau_i = 2G_i D; \qquad \lambda_i = \frac{\lambda_{0i}}{1 + \frac{\epsilon}{G_i} tr(\tau_i)} \tag{6}$$

where i is the index for each mode. Giesekus has just one parameter (α), whereas PTT has two (ϵ, ξ), when all parameters are set to zero the Upper Convected Maxwell equation is recovered.

2.4 Rheological characterization in simple shear

The investigated liquid is a solution of 2.5 % PIB (Oppanol B200, BASF; $M_w \approx 4.7 \cdot 10^6[-]$) in C14 (tetradecane). Characterisation in simple shear is performed on a rotational viscometer (Rheometrics RFS-II), with a cone-plate geometry (diameter 50 [mm], cone angle 0.0199 [rad]). Both steady and dynamic measurements are performed. The Carreau Yasuda model is fitted on the viscosity curve at a reference temperature of 25^oC. Further, four Maxwell modes (η_i, λ_i) were fitted on the dynamic measurements using a Levenbergh-Marquardt algorithm. Next, the parameters of the two nonlinear viscoelastic equations were determined using steady shear measurements, this is shown in figure 2. For the PTT model two parametersets are determined, one with parameter $\xi = 0$.

3 Steady shear flow in a rectangular channel

In this section our approach will be tested for a fully developed slit flow, where elongational forces are not present. Two channels have been investigated, which

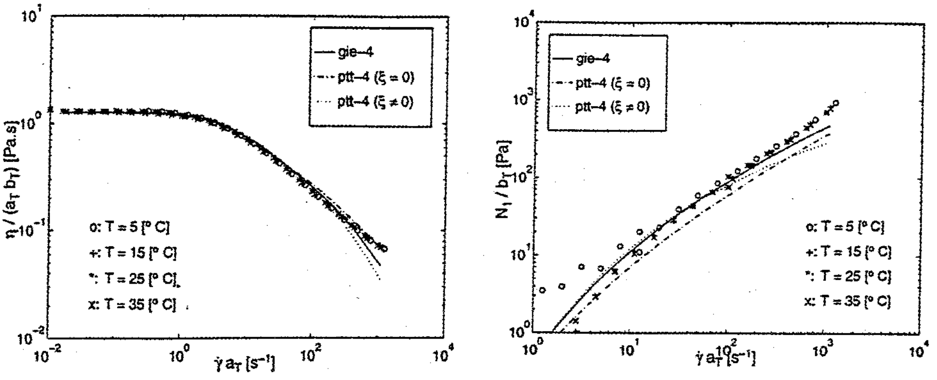

Figure 2. Characterisation in simple shear of the 2.5% Pib/C14 solution together with fits for the Giesekus and PTT model. On the left $\eta(\dot{\gamma})$, on the right $N_1(\dot{\gamma})$.

differ in their depth/height ratio (8:1 and 2:1), here only results will be presented for the small channel. The computed and measured 3D velocity profiles are shown in figure 3. Besides the velocity gradient $\dot{\gamma}_{xy}$ which in 2D flow is only present, here also $\dot{\gamma}_{xz}$ is important. As can be seen the Carreau Yasuda model accurately predicts the measured velocity field.

The 3D stresses are predicted with the viscoelastic models using the velocity gradients and are shown in figure 4. The predicted integrated stresses are shown in figure 5 together with the FIB results. The stresses also show a good agreement for all models, the largest deviation is found for the PTT model with $\xi = 0$. It can be concluded that the results for the slit flow support our approach of decoupling velocity and stress calculations in this case.

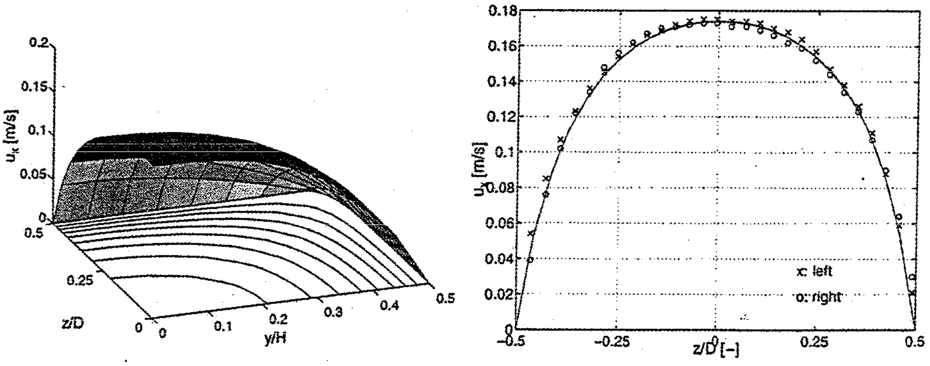

Figure 3. On the left: 3D prediction of velocity field by the Carreau Yasuda model, on the right: comparison to measured velocties along the line $y/H = 0$.

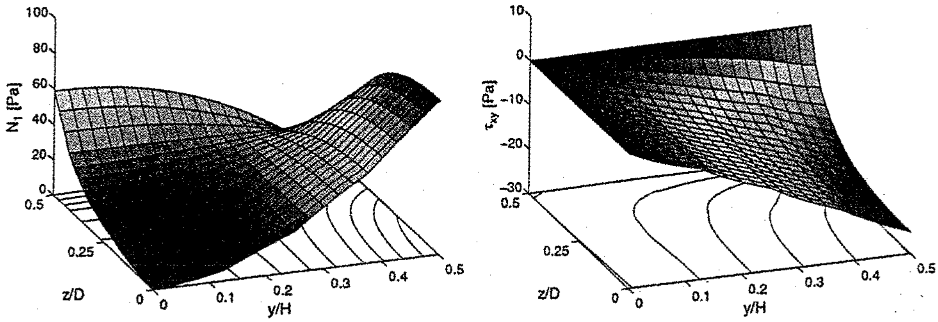

Figure 4. 3D predictions of stresses by the nonlinear viscoelastic models. On the left: first normal stress difference ($N_1 = \tau_{xx} - \tau_{yy}$), on the right: shear stress (τ_{xy}).

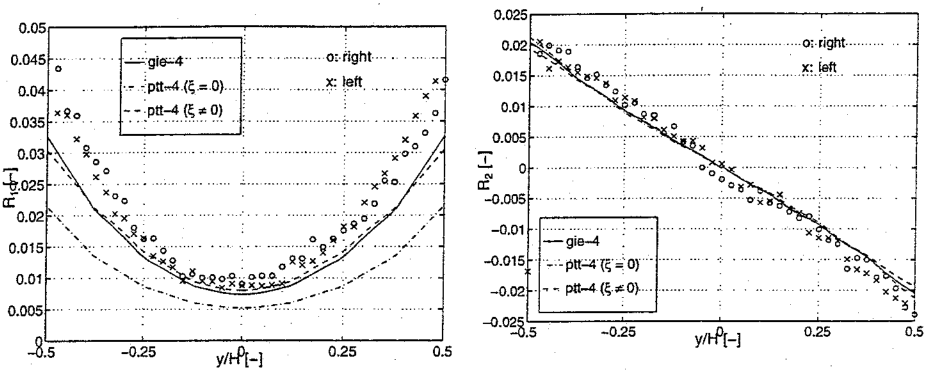

Figure 5. Comparison of measurements (intensities R_1 and R_2) with calulations (using equations 2 and 3).

4 Stagnation flow

The flow cell with its main dimensions is shown in figure 6. It consists of two perpendicular channels. In this picture the inflows are from the top and the bottom channel towards the middle of the flow cell, and the outflows are on the left and right hand side. In this way a stagnation line is created in the middle of the flow cell, causing a strong elongational flow from this line towards the outflows.

The same approach as for the slit flow is followed, the finite element mesh used to calculate the velocity field is shown in figure 6. Again, as can be seen in figure 7, using the Carreau Yasuda model gives an excellent agreement of the predicted velocities with the LDA measurements. The deformation history is determined along 24 streamlines starting from different positions along the depth at the inflow plane. Stresses are calculated along the streamlines with the nonlinear viscoelastic models using an Euler integration scheme. The integrated stresses are compared with FIB measurements in figure 8. Near the stagnation point ($x/R = 0$ in figure 8), the area where a strong elongational flow is present, all models fail to predict the large increase in stress (R_1). This discrepancy remains present towards the outflows.

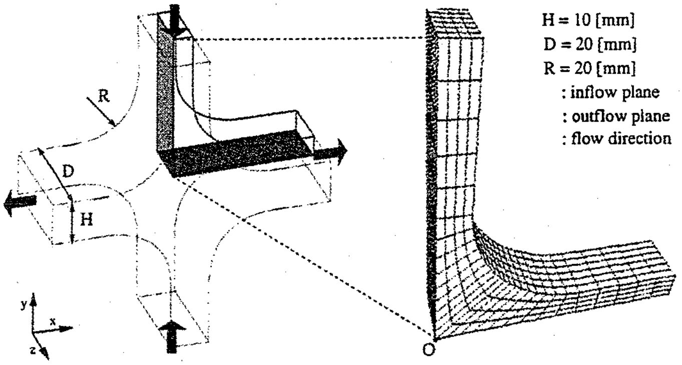

Figure 6. Schematic presentation of the flow cell together with finite element mesh.

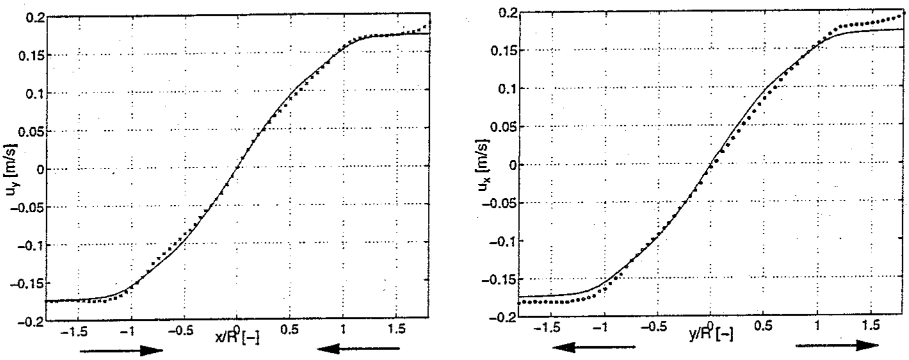

Figure 7. Comparison of the calculated velocities and LDA measurements. On the left: along the line (x,z)=(0,0) in the inflow plane as indicated in figure 6, on the right: along the line (x,y)=(0,0) in the outflow plane (figure 6).

5 New model

In the previous section it is shown that the evaluated constitutive models can not predict the stresses in the stagnation flow device with the parameters determined in simple shear flow. Changing the parameters might give a better resemblance in the elongational flow part, but on the other hand the agreement with steady shear data will become worse. Therefore constitutive equations are sought that are more flexible, preferebly with parameters that indepently control the shear and elongational behavior. An example of this, is an adjustment of the PTT model with $\xi = 0$ eq. 2.3. The structure of the model is changed by prescribing the

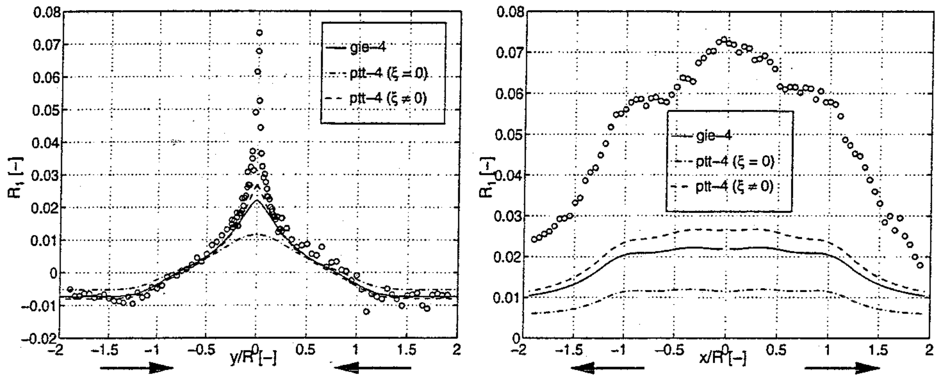

Figure 8. Comparison of the integrated stresses with FIB measurements. On the left: in the inflow plane as indicated in figure 6, on the right: in the outflow plane (figure 6).

viscosity η with an Ellis function ((13)):

$$G\lambda = \eta(\tau) = \eta_0(1 + (\frac{II_\tau}{k})^r)^{-1} \tag{7}$$

with λ defined in equation 2.3 In this way the viscosity is always predicted correctly, further the parameter ϵ can be adjusted for the predictions in elongational flow. When this is done for the 2.5% Pib/C14 fluid, the results for 3 different parameters ϵ are displayed in figure 9 for the slit flow, and in figure 10 for the opposed jets flow. Varying the model parameter does not change the predicted shear stress (R_2) in the slit, and the predicted first normal stress difference (R_1) only slightly changes. The predicted first normal stress difference (R_1) in the stagnation flow changes more drastically and the parameter can be adjusted to obtain an optimal fit.

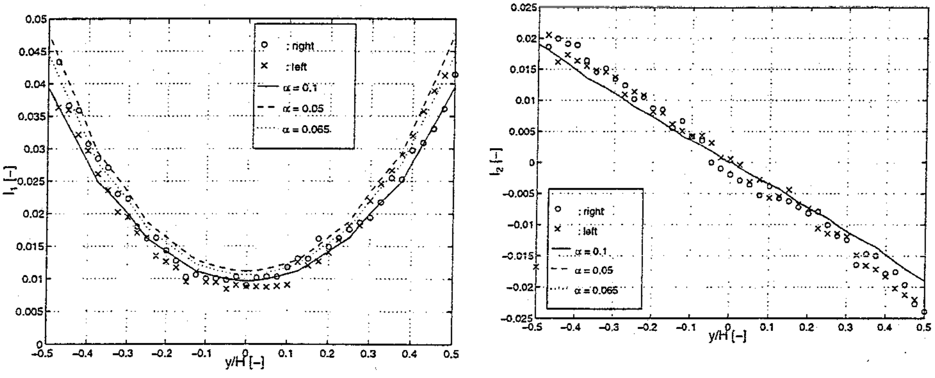

Figure 9. Comparison of the integrated stresses (intensities R_1 and R_2) from modified PTT model with FIB measurements in a 2:1 slit flow.

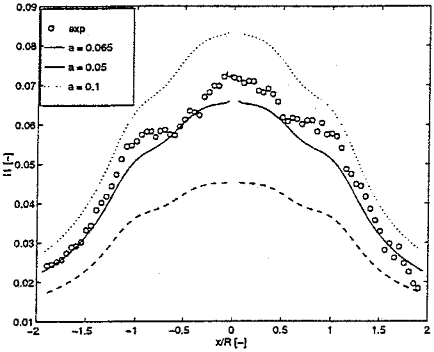

Figure 10. Comparison of the integrated stresses from modified PTT model with FIB measurements. On the left: in the inflow plane as indicated in figure 6, on the right: in the outflow plane (figure 6).

6 Conclusions

A numerical/experimental method is described which allows for the evaluation of nonlinear constitutive equations in complex flows. With the aid of optical techniques (LDA and FIB), velocities and stresses can locally be determined in the flow. It is shown that decoupling velocity (viscous model) and stress field calculation (nonlinear viscoelastic models) to do 3D flow simulations is acceptable. The evaluated Giesekus and PTT model can not predict the stresses in the stagnation flow (strong elongation) with the parameters determined in simple shear flow. With a proposed new constitutive equation, both the slit as the stagnation flow can be accurately predicted. The latter model exhibits more flexibility compared to the Giesekus and PTT model, and can almost independently adjust the material behavior in shear and elongational flow.

Acknowledgement

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