Measurements of Transient Elongational Viscosity

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INTRODUCTION

While material functions in shear flow are fairly easy to obtain experimentally, material functions in extensional flow are somewhat harder to come about. This has led to the development of a new instrument for measuring the transient elongational viscosity of polymeric solutions (Sridhar et.al.[1]). The instrument is called a Filament Stretching Rheometer, and has appeared in different modifications around the world. In this work a modified Filament Stretching Rheometer currently in use at the Danish Polymer Centre is investigated.

The instrument used in this work was constructed by the workshop in the Department of Chemical Engineering based on a design by S. F. Christensen of NKT A/S.

IDEAL ELONGATIONAL FLOW

To define start-up of ideal elongational flow, we consider a fluid held between two circular discs. The fluid is kept in a way that it forms an ideal cylinder with length L_0 and radius R_0. At time t>0 the plates separate such that length L(t) and radius R(t) are given by

$$L(t) = L_0 e^{\dot{E}t}, R(t) = R_0 e^{-\frac{1}{2}\dot{E}t} \qquad (1)$$

where $\dot{E}$ is the imposed strain rate. If we now assume the cylinder to keep its shape and only change its aspect ratio $\Lambda = L/R$, we get ideal elongation flow:

$$v_r = -\frac{1}{2}\dot{\epsilon}r, v_\theta = 0, v_z = \dot{\epsilon}z \qquad (2)$$

where the elongation rate $\dot{\epsilon}$ equals the imposed strain rate $\dot{E}$. In this flow, we can define the transient elongation viscosity:

$$\overline{\eta}_1^+(\dot{\epsilon}, t) = -\frac{\tau_{zz} - \tau_{rr}}{\dot{\epsilon}} \qquad (3)$$

We also define the Hencky strain ϵ:

$$\epsilon = \int_{t'=0}^{t} \dot{\epsilon} dt' = \dot{\epsilon}t \qquad (4)$$

and finally the Trouton ratio

$$Tr = \frac{\overline{\eta}_1^+(\dot{\epsilon}, t)}{\eta_0} \qquad (5)$$

where η_0 is the zero shear viscosity.

APPARATUS
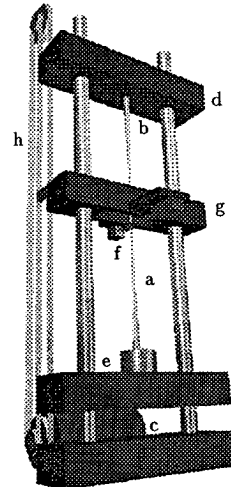
A diagram of the rheometer is shown in figure 1.

Figure 1: Schematic diagram of the Filament streching rheometer.

The sample (a) undergoing investigation is placed between two parallel discs (b). The experiment begins by separating the discs in the axial direction with an exponentially increasing velocity. This is done by a motor (c) that pulls a sled (d) on which the disc is mounted.
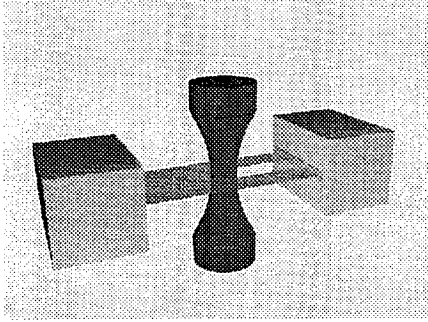

Figure 2: Measurement of filament diameter by a laser micrometer.

During the stretching of the material, the axial force on the bottom plate is measured by a load cell (e). At the same time, the diameter at the middle of the filament is recorded by a laser micrometer (f) (see figure 2). The laser micrometer is kept mid between the two discs by a second sled (g) which moves at half the speed of the upper sled (d). The ratio of speed between the two sleds is governed by the two timing belts (h) driving them. As the mid sled is moving with half the velocity of the upper sled, the sled will always be in the middle of the upper and lower discs. Finally a CCD camera can be put on the mid sled, enabling visual inspection of filament break up, evolution of radius etc.

Because of the no-slip condition at the end discs, the flow in the sample will not be ideal uniaxial elongation throughout the entire filament. Still, mid between the two discs the filament is considered to deform as an ideally deforming cylinder (Kolte et. al. [3]). Thus, by monitoring the area of the cross-section throughout the experiment, the strain rate $\dot{\epsilon}_0$ in the mid-area can

be determined. By neglecting surface tension, gravity and inertia, the force acting in the axial direction is considered constant, and by measuring the force in the bottom of the sample, the force in the mid section in axial direction is known. Knowing the evolution in midsection area, and the force acting in the axial direction, the normal stress and elongation rate can be determined and the transient elongation viscosity $\overline{\eta}_1^+$ can be calculated. Because of the geometry, the second viscosity function is zero at all times, $\overline{\eta}_2^+ = 0$.

FORCE BALANCE

The transient elongational viscosity is calculated by combining the force balances in axial and radial directions. Neglecting inertia, a force balance in axial direction for the lower half of the filament can be written as (Spiegelberg et. al. [2]):

$$F_B + (\tau_{zz} - p)\pi R^2 + 2\pi R\sigma + p_a\pi R^2 = \tfrac{1}{2}\rho g L_0 \pi R_0^2 \qquad (6)$$

with nomenclature as in figure 3. In the same way, a balance can be written in the radial direction at the center of the filament:

$$p = \frac{\sigma}{R} + p_a + \tau_{rr} \qquad (7)$$

Combining these equation and neglecting gravitation and surface tension, we obtain an expression for the normal stress difference in the middle of the filament, and we can find the elongational viscosity $\overline{\eta}$:

$$\overline{\eta}_1^+ = -\frac{\tau_{zz} - \tau_{rr}}{\dot{\epsilon}(t)} = \frac{F_B}{\dot{\epsilon}(t)\pi R^2} \qquad (8)$$

Analyzing the effect of neglecting gravitation and surface tension, the dimensionless numbers of interest are the Bond number $Bo = \rho g R_0^2/\sigma$ rating gravity to surface tension and the capillary number $\eta_0\dot{\epsilon}(t)R_0/\sigma$, balancing viscous forces and surface tension. As the rheometer is designed for very viscous fluids, the capillary number will usually be larger than 100, the bond number will usually be in the range of 1-5. Thus, the effect of gravity and surface tension seems negligible.

The effect of neglecting inertia has been examined by different authors and can shortly be presented by:

$$\frac{\rho \dot{\epsilon} L^2}{8\bar{\eta}} \ll 1 \qquad (9)$$

where L is the distance between the two plates. As L is growing at an exponential rate, the criterion will not be met at some point in the experiment. On the other hand, as the sample is stretched, some fluids will exhibit strain hardening, delaying the onset of effects from inertia. Hence, inertia can usually be neglected, but the effect has to be checked in each experiment.
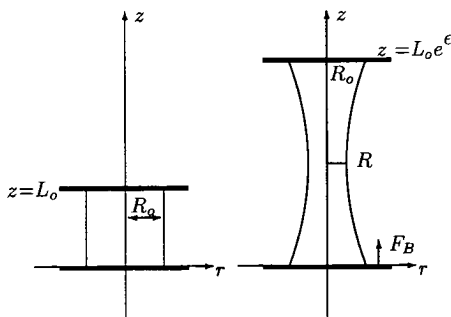

Figure 3: Uni-axial elongation in a cylindrical coordinate system.

TESTING THE RHEOMETER

Using the Trouton ratio between shear viscosity and elongational viscosity, the rheometer could be tested using a simple Newtonian fluid. When stretching Newtonian fluids, forces in the axial (stretching) direction, will originate solely from flow in the filament. Therefore, measurements on Newtonian fluids would be a good test of the rheometer. If the apparatus could catch the small forces acting in the axial direction during stretching of a Newtonian fluid, the larger forces during stretching of viscoelastic fluids would be much easier to measure.

A polybutene (HYVIS 150) was used for the measurements, shear measurements

showed that the fluid did not behave newtonian at very high shear rates. Thus, at higher strain, the Trouton ratio was expected to grow. The zero shear viscosity was η_0=578 Pa s.

Because of the no slip condition at the end plates, forces are a little elevated, predicting a too large Trouton ratio. This overshoot in predicted Trouton ratio is a function of Hencky strain and initial aspect ratio $\Lambda_0 = L_0/R_0$ (Spiegelberg et. al.[1]):

$$Tr_{calc} = \frac{F}{\mu \dot{\epsilon} \pi R^2} = 3 + \frac{1}{\Lambda_0} exp\left(-\frac{7}{3}\epsilon\right) (10)$$

In figure 4 measured Trouton ratios are shown along with the prediction defined in equation 10. The two series is measured for Λ_0=0.40 and 0.66.

Good agreement is found between measured and theoretical values. Small disagreement is found at the beginning of the experiment, where the upper plate has to be accelerated from 0 velocity to a final velocity in the time between 0 and 0^+. Also at the end of the experiments, the calculated Trouton ratio rises because of strain hardening in the filament.
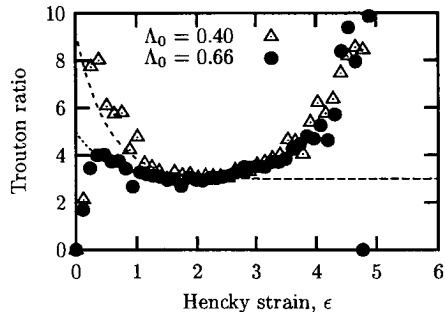

Figure 4: Measured and theoretical Trouton ratios for two initial aspect ratios, both experiments performed with strain rates of $2s^{-1}$

CONCLUSIONS

The validation of the rheometer was compared to other groups measurements of Trouton ratios, and our rheometer seems to

be at the same level as these other rheometers, both in terms of precision and reproducability.

The apparatus is now being modified, so that measurements at elevated temperatures will be possible. The goal is to measure viscosities of polymeric melts at high temperature, and thereby have an alternative to the Meissner rheometer (Meissner et. al.[4]), currently being the best source of elongational viscosities at high temperatures. The advantage of the filament stretching rheometer over the Meissner rheometer is the possibility of measuring at strain rates up to $10s^{-1}$, whereas the Meissner rheometer can measure until $1s^{-1}$.

ACKNOWLEDGEMENTS

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