Birefringence measurements on polymer melts in an axisymmetric flow cell

Abstract The stress-optical rule relates birefringence to stress. Consequently, measurement of flow birefringence provides a non-intrusive technique of measuring stresses in complex flows. In this investigation we explore the use of an axisymmetric geometry to create a uniaxial elongational flow in polymer melts. In axisymmetric flows both birefringence and orientation angle change continuously along the path of the propagating light. The cumulative influence of the material’s optical properties along the light’s integrated path makes determination of local birefringence in the melt impossible. One can nevertheless use birefringence measurements to compare with predictions from computer simulations as a means of evaluating the constitutive equations for the stress. More specifically, in this investigation we compare the light intensity transmitted through the experimental set-up vs entry position, with the theoretically calculated transmitted intensity distribution as a means of comparing experiment and simulation. The main complication in our experiments is the use of a flow cell that necessarily consists of materials of different refractive indices. This introduces refraction and reflection effects that must be modeled before experimental results can be correctly interpreted. We describe how these effects are taken into account and test the accuracy of predictions against experiments. In addition, the high temperatures required to investigate polymer melts mean that a further complication is introduced by thermal stresses present in the flow cell glass. We describe how these thermal-stresses are also incorporated in the simulations. Finally, we present some preliminary results and evaluate the success of the overall method.

Key words Viscoelastic · Axisymmetric flow · Birefringence · Polymer melt

Introduction

A major goal of non-Newtonian fluid mechanics is to determine constitutive models for polymeric fluids that can accurately predict the stresses in flows of arbitrary kinematic complexity. The constitutive models can be used to simulate diverse fluid mechanical problems. Examples are found in industrial polymer forming and processing applications and also in bio-mechanics where the flow of viscoelastic fluids are involved in the body’s mechanics. Furthermore, from a purely scientific perspective, the accuracy of these models can reveal clues regarding the fundamental nature of the molecular dynamics of polymeric materials. However, numerous constitutive models have been proposed and until the recent introduction of the “Pom-Pom” model (McLeish and Larson 1998; Verbeeten et al. 2001) no one model has proved superior to all the others in every flow
In an axisymmetric flow the stresses and consequently the optical properties change continuously along the propagation path of the light beam. The two measured optical properties, retardance $\delta'$ and orientation angle $\psi'$, reflect the cumulative effect of the 3-D stress distribution on the polarization-state of the light. Integration along the light-beam’s path maps three independent quantities: shear stress $\tau_{s\theta}$ and first and second normal stress differences $N_1(r,z) = \tau_{zz}(r,z)-\tau_{rr}(r,z)$, $N_2(r,z) = \tau_{\theta\theta}(r,z)-\tau_{\phi\phi}(r,z)$ respectively. These three quantities cannot be retrieved from the two experimentally accessible data. This reflects the main disadvantage of the use of birefringence in axisymmetric flow: if one has no a priori information of the stress distribution, direct conversion of $\delta'$ and $\psi'$ into stress components, through the stress optical rule, is not feasible. Rather than attempting to invert the optical data to yield the radial stress distribution, the optical property distribution serves as a basis for comparison with numerical simulations, as proposed by Li and Burghardt (1995). Because they are derived from simulated stress fields, the theoretically determined optical properties are highly sensitive to the choice of constitutive equation.

In their investigations on polymer solutions, Li and coworkers (Li et al. 1998, 1999, 2000; Li and Burghardt 1995) used refractive index-matched materials within the flow cell in order to prevent complications due to refraction of the sampling light beam. The fundamental difference with that work is that we model the effects of refraction on the laser light as it passes through the flow cell. In so doing, we remove the index-matching restriction and significantly extend the range of materials that can be investigated. Furthermore, once the effects of refraction are modeled it becomes possible to surround the flow-geometry with heated transparent silicon oil in order to investigate polymer melts at elevated temperatures. In this paper we describe the flow cell construction, the optical modeling required to interpret the results, and some preliminary comparisons between numerical and experimental results.

**Experimental set-up**

Extruder line and flow cell

The line-up of the most important units in the experimental set-up is schematically illustrated in Fig. 1. Details regarding the manufacturer and type of components can be found in Janssen (2000). A twin-screw extruder is used as a pump, with a gear-pump in line to prevent a fluctuating flow-rate. A by-pass is inserted between the extruder-nozzle and the gear pump to prevent pressure build up at low rotation speeds of the gear-pump. A static mixer containing four flow-diverting elements is used as a thermal equilibration zone.