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Lubricated optical rheometer for the study of two-dimensional complex flows of polymer melts

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Abstract

We describe a novel optical cross-slot channel rheometer generating two-dimensional and isothermal complex flows of polymer melts. This is made possible by lubricating the channel front and back viewing windows. Flow-induced birefringence and particle tracking velocimetry are reviewed and used to investigate the cross-slot flow of a low density polyethylene melt involving mixed shear and planar extensional deformations. This new device solves the issue of end effects in flow birefringence experiments where no variations of the optical properties along the light path are expected. It greatly facilitates the interpretation of stress field data by providing reliable measurements of the polymer melt extinction angle χ and retardation δ, with a spatial resolution of one tenth of a millimeter. At the same time, it offers an enhanced temperature control and an increased optical accuracy due to an improved laser beam shaping. The capabilities and performances of this unique type of lubricated rheometer are discussed in detail and compared with previous approaches.

Keywords: Lubrication; End effects; Stagnation flow; Flow-induced birefringence; Particle tracking velocimetry; Two-dimensional flow; Polymer melt

1. Introduction

The viscoelastic properties of polymer melts are of importance as they govern the flow behavior whenever plastics are processed in the molten state. In industry, they play for instance a central role in the design of extruder screws or in the development of efficient molds for injection molding applications. In blow molding operations, the process of parison sag and swell is another example of flow governed entirely by the rheological properties of the melt. During industrial processes, complex flows that involve a mixture of shear, extension and/or compression generally occur. The importance of these flows in processing applications has been indicated by many authors including Cogswell and Lamb [1] as well as Dealy and Wissbrun [2]. Today, there exist powerful rheological codes which can be used to design and optimize dies and molding tools. Furthermore, continuous efforts are made to develop them so that they can better describe the processing operations. Powerful as these codes are or will become, they will be useful and rigorous designs only if they are fed with accurate rheological data. If conventional rheometers offer a detailed knowledge in standard shear or elongational flows, they fail in providing useful information regarding more realistic flows that are met in industrial applications. However, a thorough understanding of the viscoelastic properties of polymeric fluids in mixed flows involving both shear and elongational deformations is essential for improving equipment design procedures and for optimizing industrial processes. In this context, the cross-slot flow channel appears to be a valuable tool to gain insight into the rheology of complex flows close to industrial processing conditions.

1.1. Review of past studies on flow-induced birefringence and cross-slot flows

Stagnation flows of polymer melts or solutions are commonly recognized as valuable flows for obtaining extensional data at high elongation rates, typically in the range 1–500 s^{-1}. Mackay et al. [3] even reported a maximum elongation rate of about

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$1000\,s^{-1}$ in their study of polyethylene and polypropylene melts using opposed orifices. These flows are not homogeneous: a fluid element close to the stagnation point will experience much higher strain than one further away. The existence of a center of symmetry provides long residence times necessary for macromolecules to highly stretch out. This feature is particularly suitable for flow-induced birefringence measurements, where the strong orientation of the macromolecules is probed. In this respect, flow birefringence applied to cross-slot geometries has received a lot of attention over the years, as illustrated by the work of Kalashnikov et al. [4] who studied polyethylene oxide and polyisobutylene solutions between crossed polarizers in order to probe supermolecular structures in polymeric solutions. They observed the onset of localized fibrils birefringence in planar elongational flow at a critical Weissenberg number close to unity. Since the study of Kalashnikov, flow-induced birefringence measurements in cross-slot devices have mainly been used to assess the performances of constitutive modeling. The group of Prof. H.E.H. Meijer at the Eindhoven Technical University is considered to be one of the pioneers of the use of a cross-slot channel for polymer melts and solutions, as indicated by the recent work of Schoonen et al. [5]. In his work, the Giesekus and Phan-Thien Tanner models have been applied to simulate the cross-slot flow of a polyisobutylene solution and he proposed a modification of the Phan-Thien Tanner model to better predict the elongational data. Using the same flow cell, Peters et al. [6] further extended this analysis to a polyethylene melt and developed a new class of viscoelastic constitutive models offering an enhanced control of shear and extensional properties. The three-dimensional (3D) simulations of the latter constitutive relations together with the Giesekus and Phan-Thien Tanner models have been compared by Bogaerds and coworkers to the experimental results obtained for a polyisobutylene solution in the same flow geometry [7]. In a similar cross-slot flow cell, Verbeeten et al. [8,9] examined the performances of the multi-mode extended Pom-Pom, Giesekus and exponential Phan-Thien Tanner models and Swartjes et al. [10] studied flow-induced crystallization using flow birefringence and compared their results with numerical predictions using the Leonov and the extended Pom-Pom models. Very recently, Odell and coworkers [11] developed a new type of extensional rheometer by combining oscillatory and cross-slot flows. They were able to measure the birefringence response of dilute polymeric solutions to characterize macromolecules in terms of molecular flexibility and ultra high molecular weight distribution. Finally, cross-slot flows have also been generated using a two-piston multipass rheometer designed and developed at the University of Cambridge [12]. First results were obtained for a polydisperse polystyrene melt in [13], where isochromatic birefringence patterns were compared with numerical predictions of various constitutive equations including the integral Wagner, Rolie-Poly and Pom-Pom models.

In an attempt to optically map the stress field in stagnation flows for subsequent constitutive model validation, most rheo-optics researchers have used so far large aspect ratios to neglect the influence of the confining walls on both the stress and velocity fields. Indeed, there should be no variations of the optical properties along the laser beam propagation direction as the birefringence signals are integrated along the light path. In reality however, large aspect ratios are not sufficient to guarantee an approximate planar flow, as will be discussed in the next section.

1.2. End effects in flow-induced birefringence experiments

A reliable optical mapping of the stress distribution by flow-induced birefringence requires a 2D flow field, as no variations of the optical properties along the optical path are expected [14]. In reality however, such a requirement is never fulfilled: the flow is actually confined by viewing windows which bring about end effects. For highly birefringent polymer melts, the early study of Wales [15] was often cited as an experimental evidence that end effects in birefringence experiments are negligible for channels with aspect ratios greater than 10, namely the channel depth divided by its width. In recent years, Wales assumption has been extensively used in rheo-optics in order to satisfy the condition of two-dimensionality [16–22]. However, little attention has been given so far to the effective role of end effects in flow birefringence experiments. Indeed, few studies [23–25] have shown that even large aspect ratios of order 8–10 were not sufficient to approach planar flows. The first systematic study on end effects in flow-induced birefringence experiments was conducted by McHugh et al. [26]. These authors attributed isoclinic band-spreading patterns to the inevitable stress field gradient close to the confining viewing windows of their slit-flow geometry. Later, Burghardt and Fuller [23] modeled the optical properties of an upper-convected Maxwell fluid undergoing confined shear flow in a planar Couette geometry. They found considerable errors in the determination of the extinction angle near the side walls, which is consistent with the observations made by Janeschitz-Kriegl et al. [27]. Galante and Frattini [24] used the differential propagation Müller matrix formalism to compute the apparent retardation and extinction angle of an an upper-convected Maxwell fluid in nominally 2D channel flows. They concluded that 3D effects led to large errors in the measurement of the extinction angle at positions near the transitions in retardation order. Kajiwara et al. [28] analyzed the side wall effects in their finite element numerical study of the flow of a low density polyethylene melt through a tapered slit die. They found that the average stresses over the channel depth were closer to the experimental data than those computed assuming an ideal planar flow. Recently, Schoonen [21] further simulated the influence of end effects on both isochromatics and isoclinics measurements for a fully developed Newtonian slit flow. For an aspect ratio of 8, the parasitic shear gradients at the front and back slit wall caused a decrease in the retardation of 6% at the lateral walls and an increase of 5% at the symmetry plane. The influence on isoclinics experiments was much larger and made the measured birefringence data useless without 3D analysis. This result is in line with the work of Öttinger [29], who considered a sandwich composed of three homogeneous pure retarders to illustrate end effects in flow-induced birefringence experiments: a 2D core surrounded by two outer edge regions. He demonstrated that the small retardation approximation for