Polymer 104 (2016) 193–203

Contents lists available at ScienceDirect

Polymer

journal homepage: www.elsevier.com/locate/polymer

First normal stress difference and in-situ spectral dynamics in a high sensitivity extrusion die for capillary rheometry via the 'hole effect'

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ARTICLE INFO

Article history:
Received 31 March 2016
Received in revised form
14 July 2016
Accepted 22 July 2016
Available online 26 July 2016

Dedicated to the memory of our colleague and friend; Víctor Hugo Rolón Garrido, *14. April 1979 – †8. June 2015.

Keywords:
Normal stress differences
Extrusion
Instabilities

ABSTRACT

The development of a high sensitivity polymer melt extrusion flow instability detection die capable of estimating the normal stress differences of polymer melts during capillary rheometry tests via the 'hole effect' is presented here as proof-of-principle. Two polymer melts, a low density polyethylene (branched topology) and a high density polyethylene (linear topology), were tested in a variety of experimental configurations with the purpose of determining optimal conditions for performing normal stress difference measurements. The data was compared with rotational rheometry oscillatory shear measurements analyzed via the Laun rule. It is shown that it is possible to estimate the first normal stress difference in the capillary die, whereas the second normal stress difference cannot be determined within the experimental errors using the current configuration design. Furthermore, the influence of the induced streamline curvature via the 'hole effect' on the onset and development of melt flow instabilities is simultaneously assessed. It is shown that the hole depth has a stabilizing influence, i.e. the onset of instabilities occurs at higher shear rates, in the long chain branched polymer tested, whereas for the linear polymer tested it has a destabilizing effect on the stick-slip instability i.e. the onset of stick-slip occurs at lower shear rates.

1. Introduction

The determination and understanding of polymer melt rheological properties are of utmost importance for the design and operation of polymer melt processing operations. In particular, processing takes place at high shear and elongation rates, which are typical for extrusion and injection molding. A major challenge of polymer rheology is the investigation of the relationship between the molecular architecture of a material, how it can be processed and the ensuing properties [1]. Rheological polymer melt properties, from the most basic, such as the shear viscosity function in the linear regime, to advanced oscillatory shear nonlinearities are readily available using rotational rheometry [2]. However, when it comes to relatively high shear rate rheometry, that would match processing conditions, the most common measurement available is the determination of the shear rate dependent viscosity function of a polymer measured on capillary rheometers. The phenomenon of die swelling, i.e. the relative increase in the extrudate dimensions following the passage of a material through a shaping die, and polymer melt instabilities, i.e. surface and volume distortions of extrudates [3,4], are other examples of characteristic phenomena that occur in the extrusion of polymer melts at high shear rates [5]. Furthermore, such phenomena are the result of the viscoelastic nature of polymer melts, which makes their rheological investigation difficult however of interest. To address these difficulties, a high-sensitivity pressure-based system for the simultaneous determination of polymer melt normal stresses and detection of instabilities via capillary rheometry is investigated in this study.

1.1. Normal stresses and polymer melt flows

The existence of non-zero normal stress components in the extra stress tensor of polymer melts differentiates viscoelastic from pure viscous flows [6], where the normal components are zero. The normal stresses are directly related to flow phenomena that impact polymer melt processing, e.g. die-swelling and polymer melt flow instabilities. Normal stresses in polymeric liquids are quantified

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http://dx.doi.org/10.1016/j.polymer.2016.07.061
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using the first and second normal stress differences, $N_1 = t_{11} - t_{22}$ and $N_2 = t_{22} - t_{33}$, respectively, where $t_{ii}, i = 1,2,3$, are the normal components of the extra stress tensor. For polymers, it is generally acknowledged that $N_1 > 0$, $N_2 < 0$ and that typically $|N_1| \approx 10|N_2|$ [7–9].

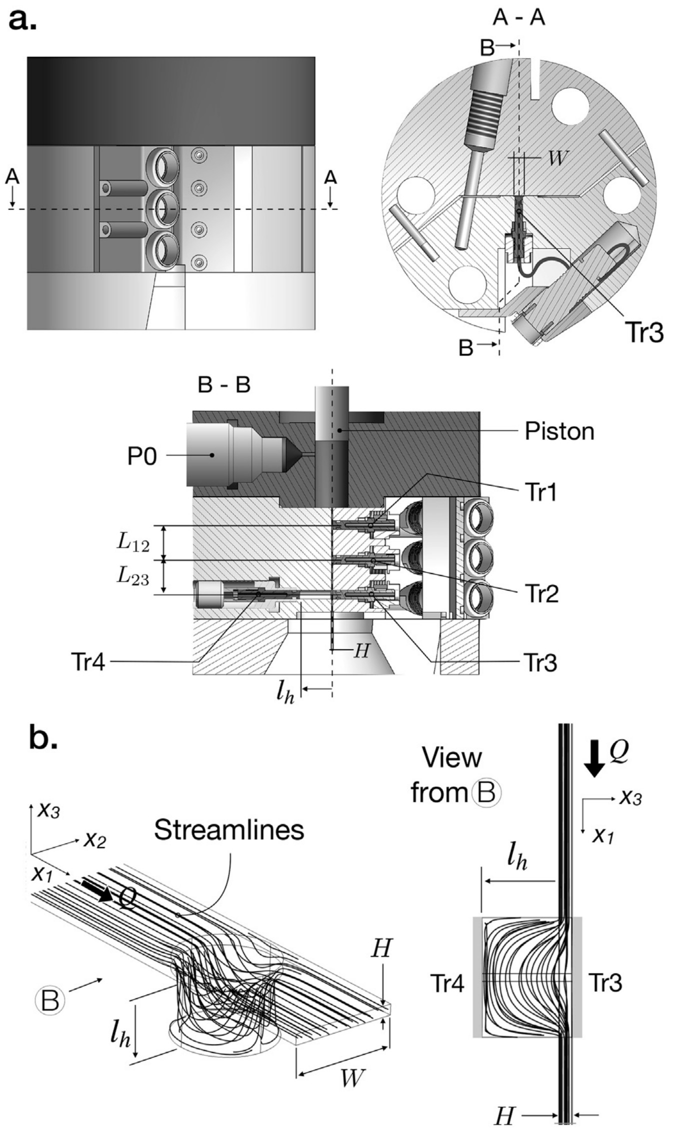

Fig. 1. (a) Virtual prototype of the high sensitivity die equipped with four piezoelectric pressure transducers, Tr1-4. The channel height and width are $H = 0.3$ mm and $W = 3$ mm. The distance between Tr4 and the flow channel, i.e. the hole depth l_h, can be varied to induce streamline curvature, whereas the distance between transducers Tr1-3 is $L_{12} = L_{23} = 10$ mm. (b) Detailed view of the streamline curvature induced when Tr4 is offset to the flow channel. The streamlines were computed using Comsol Multiphysics.

The determination of normal stress differences can be achieved using a rotational rheometer, however the second normal stress difference is a non-trivial measurement [10]. The first normal stress difference, N_1, can be determined using a cone-plate measurement system in steady shear [10]. In addition, when using a plate-plate geometry, the normal force is converted into the simultaneous contribution of the two normal stress differences [10], i.e. $N_1 - N_2$. A more accurate, albeit a more difficult to implement method, is the use of pressure gauges positioned at various places on the plate (reversed cone plate geometry), from which $N_1 + 2N_2$ is determined [10]. An indirect method of determining the first normal stress difference from oscillatory shear measurements is the so-called Laun rule [11]:

$$N_1^{\mathrm{Laun}}(\dot{\gamma}) = 2G'(\omega)\left(1+\left(\frac{G'(\omega)}{G''(\omega)}\right)^2\right)^a \tag{1}$$

where $\dot{\gamma}$ is the shear rate, ω is the angular frequency and G' and G'' are the dynamic moduli determined from oscillatory tests. Several values for the exponent a are reported in the literature [12,13], including $a = 0$, i.e. $N_1^{\mathrm{Laun}}(\dot{\gamma}) = 2G'(\omega)$, $a = 0.2$ and $a = 0.7$. Regardless, the determination of normal stress differences from rotational rheometry are usually limited to shear rates that are well below the values used in processing due to edge fracture effects [14,10]. To compensate for these limitations, measurements in pressure driven (Poiseuille) flows have been considered via two methods [9,15]: (i) an extrapolation procedure to determine the exit wall pressure in a slit die and (ii) via the so-called 'hole effect'. More recent testing and overviews for the extrapolation procedure can be found in Baird, 2008 [12] and Teixeira et al., 2013 [13]. For the 'hole effect' approach, systematic pressure measurement errors using pressure gauges mounted in the walls with taps in the instrument have been put to use [16,17] for the purpose of determining the normal stress differences. The pressure errors arise due to the local streamline curvature where pressure gauges are offset from the flow channel walls, see also Fig. 1. For viscoelastic fluids, including polymer melts, the streamline curvature will induce an additional force due to non-zero normal components in the extra-stress tensor compared to the situation where the flow is undisturbed [15]. Thus, it is possible to estimate the normal stress differences by placing two transducers facing each other with one positioned flush with the channel and the other one in an offset position and then measuring the pressure difference between the two, i.e. the 'hole pressure', $p_h = p_4 - p_3$ in Fig. 1, where p_3 and p_4 are the pressure readings from transducers Tr3 and Tr4. A straightforward estimation of the normal stress differences from hole pressure data is given in the form proposed by Tanner and Pipkin [18]:

$$p_h = a_1 N_1 + a_2 N_2. \tag{2}$$

where, for a circular opening, $a_1 \approx 0.25$, and the constant $a_2 \ll a_1$, i.e. $p_h \approx 0.25 N_1$ [15]. A more comprehensive framework that includes simplifying assumptions concerning the symmetry of the streamlines and stress distribution in the hole region was proposed by Higashitani and Pritchard, 1972 [12,13,19] and further enhanced by Baird, 1975 [20]. To summarize, in the case of a circular opening, the following relationship was deduced that relates the hole pressure and the normal stress differences:

$$(N_1 - N_2) = 3p_h \frac{d \ln p_h}{d \ln \sigma_w} \tag{3}$$

where p_h is the hole pressure and σ_w is the wall shear stress. It should be noted that the theory can be applied to rectangular slots placed perpendicular or parallel as well as transverse to the flow direction, to determine N_1 and N_2, respectively [12,13,19]. Recent results showed that these measurements on polymer melts were feasible using conventional melt pressure transducers and are therefore applicable to lab-scale processing and rheological characterization methods, e.g. extruders [12,13,21,22]. However, the most convenient 'hole pressure' method construction version remains the circular opening. In this respect, a careful error estimation procedure was applied by Teixeira et al. [13] for varying slots with the conclusion that N_2 is in fact 0 within experimental error.