First normal stress difference measurements for polymer melts at high shear rates in a slit-die using hole and exit pressure data

Donald G. Baird *

Department of Chemical Engineering and Macromolecular and Interfaces Institute, Virginia Polytechnic Institute and State University, Blacksburg, VA 24061, United States

Abstract

In this paper we determine the ability to obtain values of the first normal stress difference, \( N_1 \), at relative high shear rates (1.0–100.0 \( s^{-1} \)) in a slit-die for several polymer melts of varying degrees of fluid elasticity using hole pressure data and the Higashitani–Pritchard–Baird (PHB) equation. Accurate measurements of the hole pressure, \( P_h \), which is defined as the difference between the pressures measured by means of a flush-mounted pressure transducer and a transducer mounted at the base of rectangular slot (i.e. the hole), are extremely difficult to make because \( P_h \) is typically less than 2\% of the absolute pressure measured. By calibrating the pressure transducers once mounted in the slit-die, accounting for viscous heating, and ensuring the transducers were properly aligned, precise measurements of \( P_h \) were made even with commercial pressure transducers. Values of \( N_1 \) obtained from \( P_h \) data agreed well with values of \( N_1 \) and twice the storage modulus (2\( G' \)) for four different polymer melts for shear rates approaching 70 \( s^{-1} \) and wall shear stresses approaching 80 kPa. Furthermore, exit pressure measurements were made simultaneously in the same slit-die device, and values of \( N_1 \) extracted from the data were found to be as much as 100\% higher than values measured by means of a cone-and-plate device and the hole pressure which was consistent with the reports of others.

Keywords: Hole pressure; First normal stress difference; Exit pressure; Slit-die; High shear rates; Higashitani–Pritchard–Baird equation

1. Introduction

In order to assess the performance of polymer melts in processing operations such as extrusion, where shear rates are known to reach values in the range of 100 \( s^{-1} \) and more, a complete rheological understanding of the fluid is needed. Although viscosity measurements are obtainable at these shear rates, measurements of the first normal stress difference, \( N_1 \), are not, and, yet, phenomena such as extrudate swell and molecular orientation are related to this quantity. Furthermore, relying on constitutive relations to provide this knowledge is not possible without an experimental verification of the predicted values of \( N_1 \). Hence, there is a need to be able to measure \( N_1 \) at high shear rates or shear stresses, at least up to the onset of melt fracture, and in geometries such as slit-dies common to polymer processing operations.

Measurements of the first normal stress difference, \( N_1 \), for polymer melts using conventional devices such as cone-and-plate and parallel-plate rheometers are typically limited to shear rates of the order of 1 \( s^{-1} \) because of edge fracture at the free surface [1,2]. Two methods which might have the potential for obtaining \( N_1 \) at high shear rates are based on measurements of the hole pressure [3–5] and exit pressure [6,7]. It has been shown by several researchers that the exit pressure method systematically over predicts values of \( N_1 \) based on comparison to \( N_1 \) data at low shear rates [8,9]. The method for measuring \( N_1 \) based on the hole pressure has been shown to agree well with independent measurements at low shear rates for polymer melts [9,10]. However, at high shear rates, >10 \( s^{-1} \), it is unclear whether it is possible to extract values of \( N_1 \) from hole pressure data, because of the inability to obtain independent values of \( N_1 \) for comparison and because of a breakdown in the theory itself. The purpose of this paper is to assess the ability to obtain values of \( N_1 \) for polymer melts at shear rates in the range of 1.0 to about 70 \( s^{-1} \) or shear stresses up 80 kPa using the hole pressure and exit pressure methods. Before doing this, we first review the theory and the key assumptions on which the method for extracting \( N_1 \) from hole pressure data is based and then review briefly some of the studies concerned with factors affecting the hole pressure measurements and assessment of the theory. Although the main emphasis of this paper is on the hole pressure method, we also summarize the key findings on the exit pressure method.
2. The hole pressure

2.1. Correlation to \( N_1 \)

In 1968 Broadbent et al. [3] first reported systematic errors in the measurement of pressures in flowing viscoelastic fluids using pressure taps (i.e. pressure transducers mounted at the base of small holes) versus flush-mounted pressure transducers. As shown in Fig. 1, the difference between the pressure measured by the flush-mounted pressure transducer (PT1) and that measured by a transducer mounted at the base of a hole (PT2) is referred to as the hole pressure (\( P_h \)). We note at first it was referred to as the hole pressure error, but once it was realized that it was directly related to \( N_1 \), then it was referred as the hole pressure. For viscoelastic fluids, \( P_h \) was large and positive and increased with increasing shear stress while for Newtonian fluids under the same conditions \( P_h \) was negligible [4]. Tanner and Pipkin [11] estimated the hole pressure using second order fluid theory and found that it was 0.25\( N_1 \). Broadbent and Lodge [4] observed a simple relation between \( P_h \) and \( N_1 \) for a polymer solution, but the coefficient was less than 0.25 and varied with the given polymer system [5]. Higashitani and Pritchard [12] carried out a more general analysis of flow over a hole and derived an expression for \( P_h \), which when differentiated lead directly to an expression for obtaining \( N_1 \) from \( P_h \) data and directly accounted for the variation of the coefficient with the given fluid [13].

2.2. Summary of the Higashitani and Pritchard theory

As the purpose of this paper is to extract values of \( N_1 \) from hole pressure data at high shear rates or stresses, then we begin with a brief summary of the key assumptions in the Higashitani and Pritchard theory. They assumed that the flow over a slot as shown in Fig. 2 was curvilinear shear flow and that there was negligible flow in the cavity. They introduced a general curvilinear coordinate system in which \( X_1 \) was the coordinate along the shear direction, \( X_2 \) was along the direction perpendicular to the shearing surfaces and \( X_3 \) was the third coordinate (not shown but going into the plane of the figure). The equations of motion for components 1 and 2 became

\[
\frac{1}{h_1} \frac{\partial \Pi_{11}}{\partial X_1} + \frac{1}{h_2} \frac{\partial \Pi_{12}}{\partial X_2} - \frac{2 \Pi_{21}}{r_23} = 0 \quad (1)
\]

\[
\frac{1}{h_2} \frac{\partial \Pi_{22}}{\partial X_2} + \frac{1}{h_1} \frac{\partial \Pi_{21}}{\partial X_1} + \Pi_{11} - \Pi_{22} = 0 \quad (2)
\]

where the \( \Pi_{ij} \) are the components of the total stress and a tensile stress is taken as positive, \( h_i \) the scale factors associated with the curvilinear coordinates \( X_i \) and \( r_23 \) is the radius of curvature of the coordinate curve formed by the intersection of surfaces with \( X_2 \) and \( X_3 \) constant. Eqs. (1) and (2) were combined and integrated to give

\[
(-\Pi_{22})_1 - (-\Pi_{22})_2 = \int_2^1 \frac{N_1}{2\sigma} \frac{\partial \sigma}{\partial X_2} \, dX_2 + h_2 \int_2^1 \left[ \frac{N_1}{2\sigma} \frac{\partial \sigma}{\partial X_1} + \frac{\partial \sigma}{\partial X_1} \right] \, dX_2 \quad (3)
\]

where \((-\Pi_{22})_1 \) and \((-\Pi_{22})_2 \) are the pressures at the upper wall and slot bottom, respectively. With the assumption that the flow is symmetric about the slot center-line, then \( \partial \Pi_{ij}/\partial X_1 = 0 \) and, hence, after a change of variable

\[
P_h = \int_0^\sigma \frac{N_1}{2\sigma} \, d\sigma \quad (4)
\]

Even though it was assumed that the second term in Eq. (3) is small compared to the first term, it must be noted that in pressure driven flow \( \partial \Pi_{11}/\partial X_1 \) may not be zero due to the large isotropic pressure gradient. It was recognized by Baird [13] that differentiation of the integral in Eq. (4) with respect to \( \sigma_w \) lead to the desired relation (Eq. (5) below) for extracting \( N_1 \) from \( P_h \) data for flow over a rectangular slot placed perpendicular to the flow direction:

\[
N_1 = 2P_h \frac{d \ln P_h}{d \ln \sigma_w} \quad (5)
\]
Higashitani and Pritchard carried out similar analyses for a round hole \((P_h = P_{ho}, \text{ in this case})\) and a rectangular slot placed parallel to the flow direction \((P_h = P_{ho||})\) when differentiatied gave expressions involving the second normal stress difference, \(N_2\), respectively [13]:

\[
N_1 - N_2 = 3P_h \frac{d \ln P_{ho}}{d \ln \sigma_w} \tag{6}
\]

\[
N_2 = P_h \frac{d \ln P_{ho||}}{d \ln \sigma_w} \tag{7}
\]

Provided these relations are valid, then these equations give, in principle, a remarkable method for providing the complete rheological characterization of polymeric fluids in a slit-die at high shear rates. Our particular interest in this work is Eq. (5), for flow over a rectangular slot, but we include the expressions for a round hole and a slot placed parallel to the flow for completeness.

2.3. Evaluation of the HPB equation

Numerical simulation [9,14,15] and flow birefringence [10] both showed that the major assumptions (no flow in the hole and symmetry of the stress tensor about the center-line of the hole) in the HP theory are violated even at low shear rates. However, both types of studies showed that even with secondary flow in the hole, the stresses cancelled out in the integration of the stress field along the center-line (Eq. (4)). Furthermore, the stress field was slightly asymmetric even at low shear rates, but did not significantly influence the integration process. Pike and Baird [10] using flow birefringence, evaluated quantitatively the effect of asymmetry, but at low shear rates the effect was insignificant. However, the fact that the major assumptions are violated at low shear rates raises the question as to whether the theory is valid at shear rates significantly higher than 10 s\(^{-1}\).

At high shear rates greater than about 10 s\(^{-1}\), numerical and flow birefringence analyses are not applicable because of loss of numerical convergence and merging of isochromatic fringe patterns. Therefore, it is necessary to rely on experimental measurements to assess the validity of Eq. (5). Measurements of \(N_1\) for polyisobutylene/decalin solutions at shear rates in the range of 10\(^6\) s\(^{-1}\) were carried out by Lodge [16] using a slit-die rheometer and a transverse slot. The measurements were limited to wall shear stress values of about 16 kPa. Comparison was made against independently measured values obtained in a parallel plate device, and the agreement was excellent up to a shear rate of 29,000 s\(^{-1}\), as this was the limit of the rotational device measurements. Shear stress measurements agreed up to shear rates of almost 10\(^5\) s\(^{-1}\), which again was the limit of the torsional device. The Reynolds numbers reached levels of about 8.0 which meant there were negligible effects of inertia on the \(P_h\) measurements and, hence, \(N_1\). Using time–temperature superposition, agreement between \(N_1\) values obtained from \(P_h\) data and the torsional device was found up to shear rates of 1.2 \times 10^6 s\(^{-1}\) confirming the validity of the HPB equation for a polymer solution.

Padmanabhan and Bhattacharya [17] reported measurements of \(N_1\) in a slit-die using \(P_h\) and exit pressure data for two LDPE’s of different molecular weight, MW. Independent values of \(N_1\) were obtained by means of steady shear data in a cone-and-plate device and by means of dynamic mechanical data. Shear rates from 40 to 700 s\(^{-1}\) and wall shear stress values from 2 \times 10\(^4\) to 10\(^5\) kPa were reached. The viscosity measured by means of the slit-die agreed well for one of the resins with that measured by means of a capillary rheometer and estimated by means of the complex viscosity but not for the other. The values of \(N_1\) obtained from \(P_h\) data did not agree well with those measured independently or estimated from dynamic data. Part of this disagreement was due to using Tanner’s equation (i.e. \(N_1 = 4P_h\)) which was based on assuming the fluid was second order rather than the HPB equation. Furthermore, although values of \(N_1\) determined by means of the cone-and-plate rheometer varied with the resin, the values obtained from \(P_h\) data were the same. Hence, there is a need to confirm the ability to obtain \(N_1\) from \(P_h\) data and the \(P_hB\) equation for polymer melts at high shear rates and wall shear stresses.

3. The exit pressure

Although the primary purpose of this paper is to report the ability to measure \(N_1\) values at high shear rates for polymer melts using the hole pressure, we also summarize the exit pressure method. The original analysis of the exit pressure was reported by Han [7], and later corrected by Walters [1]. In essence a macroscopic momentum balance was performed over the exit region of a slit-die as shown in Fig. 3. This analysis lead to the following equation for the exit pressure, \(P_x\):

\[
P_x = \Pi_{yy}(h, 0) = -\frac{1}{h} \int_0^h (\Pi_{zz} - \Pi_{yy}) dy - \frac{1}{h} \int_0^h y \frac{\partial \tau_{zy}}{\partial z} dy \tag{8}
\]

where \(h\) is one-half of the die height, \(H\). The second term on the right side of the equation is related to the disturbance of the flow as the exit region is approached [18,24]. It was assumed by Han that the flow remained steady shear flow until the die exit in which case the second term was assumed to be negligible.

![Fig. 3. Die exit region over which a macroscopic momentum balance is performed to obtain the exit pressure.](image-url)
Eq. (8) was differentiated with respect to the wall shear stress to give the following expression for obtaining $N_1$ from $P_x$ data.

$$N_1 = P_x + \sigma_0 \frac{\partial P_x}{\partial \sigma_w}$$

(9)

However, it was routinely observed that values of $N_1$ obtained from Eq. (9) and $P_x$ data were systematically higher than independently measured values [8]. Read and coworkers [24] using birefringence data showed that no matter how small the rearrangement distance was at the die exit, the contribution from the second term was significant, being as much as 50–75% of the exit pressure. Padmanabhan and Bhattacharya [17] measured values of $P_x$ simultaneously with $P_h$ measurements and reported values of $N_1$ obtained from $P_x$ and Eq. (9) varied with the resin but were significantly lower than those obtained from $P_h$ data but still higher than independently measured values of $N_1$. The inaccuracy of the exit pressure method rests in two sources. One, of course, is the assumption that the flow is steady shear flow up until the die exit and the rearrangement of stresses are neglected. The other involves the extrapolation process for obtaining $P_x$. Laun [19] pointed out that the magnitude and sign of $P_x$ depended on the type of pressure profile used (linear or quadratic). Because $P_x$ is only a small fraction of the measured pressures (1%), significant errors of the measured exit pressure can occur through the extrapolation process. Because in this work great care was taken to measure highly precise values of pressure in a slit-die, we report measurements of values of $P_x$ and values of $N_1$ obtained by means of Eq. (9) in an effort to further assess this technique.

4. Experimental

4.1. Materials

The four polymers used in this paper and some of their characteristics are listed in Table 1. They were selected because they cover a wide range of rheological responses from fluids with a high degree of memory and long relaxation times to those with very short relaxation times and little memory. The two commercial polystyrenes, Styron 678 (PS-1) and Styron 685 (PS-2), have somewhat different molecular weights and significantly different zero shear viscosities, $\eta_0$, and relaxation times. A commercial low density polyethylene (LDPE), NP-952, with relatively high molecular weight and a significant degree of long chain branching was chosen because of its high degree of viscoelasticity with a relaxation time of 16.3 s. The polycarbonate (PC) resin exhibits very little viscoelasticity and has a short relaxation time behaving nearly as a second order fluid. In addition to the molecular weight (MW) and MW distribution, the zero shear viscosity along with the relaxation time (as determined by fitting the Bird–Carreau model to the viscosity data) are reported for each resin in Table 1.

4.2. Rheological characterization

Steady shear and dynamic oscillatory measurements were carried out on a RMS 800 using a 25 mm cone-and-plate attachment with a 0.1 rad/s. The magnitude of the complex viscosity, $|\eta^*|$, was obtained from the values of $G'$ and $G''$. Dynamic oscillatory measurements were carried out in the linear viscoelastic range of strain (typically <10% strain) to obtain the storage ($G'$) and loss ($G''$) moduli over an angular frequency ($\omega$) range of 0.1–100 rad/s.

4.3. Slit-die rheometer

A schematic of the slit-die used in this research is shown in Fig. 4 in which the pertinent dimensions and spacing of the pressure transducers are shown. The die was constructed from two rectangular blocks of 316 stainless steel bolted together by eight 5/16 in. and two 1/8 in. bolts. The slit height, $H$, to width, $W$, ratio was 10 eliminating the effect of side-walls on the flow. The rectangular slots placed perpendicular to the flow direction could be readily interchanged by inserting them into the bottom block of the die. The dimensions of the various slots used in this work are summarized in Table 2, but, in particular, it should be noted that the slot depth to width ratio, $dB/W$, was greater than 4 in all cases in order to eliminate the effect of hole depth on $P_h$.

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**Table 1**

<table>
<thead>
<tr>
<th>Polymer</th>
<th>Trade name</th>
<th>Source</th>
<th>Melt index</th>
<th>MW (MWD)</th>
<th>$\eta_0$ (Pa s)</th>
<th>$\lambda$ (s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Polystyrene PS-1</td>
<td>Styron-678</td>
<td>Dow Chem.</td>
<td>12.0</td>
<td>255,000 (2.55)</td>
<td>10,446 (190 °C)</td>
<td>2.76</td>
</tr>
<tr>
<td>Polystyrene PS-2</td>
<td>Styron-685</td>
<td>Dow Chem.</td>
<td>2.40</td>
<td>285,000 (2.70)</td>
<td>48,340 (190 °C)</td>
<td>7.35</td>
</tr>
<tr>
<td>LDPE</td>
<td>NP-952</td>
<td>Equistar</td>
<td>2.0</td>
<td>232,000 (17.1)</td>
<td>15,961 (190 °C)</td>
<td>16.3</td>
</tr>
<tr>
<td>Polycarbonate PC</td>
<td>Lexan-141</td>
<td>General Electric</td>
<td>30,000 (2.5)</td>
<td>1,600 (260 °C)</td>
<td></td>
<td>&lt;0.01</td>
</tr>
</tbody>
</table>
Three pressure transducers (PT1, PT3, PT4) of the strain gauge type (Dynisco PT 422a) were mounted flush with the upper slit wall while the fourth pressure transducer (PT2) was mounted at the bottom of a cylindrical cavity which was below the base of the slot. In order to increase the sensitivity of the measurements of $P_h$, the lowest pressure range transducer (0–3450 kPa) was selected for transducers PT1 and PT2, while the pressure ranges of transducers PT3 and PT4 were 0–5170 and 0–10,340 kPa, respectively.

Precise temperature control is crucial to the measurement of accurate values of $P_h$. The temperature of the melt entering a gear pump from the extruder (Killion, 2.54 cm diameter) was controlled by a temperature controller which actuated a tape heater wrapped around a pipe which transported the melt from the extruder to the gear pump. Furthermore, a static mixer was used to produce a homogeneous temperature profile in the pipe. A separate temperature controller was used to control the temperature of the pipe transporting the melt from the gear pump to the slit-die. Four sets of cartridge heaters (Industrial heaters Inc.) controlled by four independent temperature controllers were used to maintain the wall temperature of the die. In order to minimize the temperature gradients at the die exit, a fifth set of cartridge heaters placed in a steel plate bolted to the face of the die exit and controlled by a temperature controller was used. The sensing thermocouples were mounted as close to the slit wall as possible. Once the system was operating, thermocouple probes were inserted into the slit to monitor the distribution of the melt temperature along the slit. The temperature controllers were then reset to maintain the melt temperature to within ±0.5 °C throughout the entire length of the die. At high shear rates (>60 s⁻¹) the average temperature rise at the die exit was measured to be about ±1.5 °C.

In order to maintain stable and uniform flow into the slit-die, a gear pump (Zenith HPB 5556, 1.72 cm³/rev) was used. For a given flow rate the speed of the extruder (RPM’s) was adjusted until the flow rate was stabilized. A bleed valve was used to balance the flow whenever the throughput from the extruder exceeded the limits of the gear pump, especially at the lowest flow rates. Although one could obtain the flow rate from the speed of the gear pump, the most accurate method was to measure the mass flow rate at the die exit by collecting extrudate for a fixed time.

### 4.4. Pressure transducer calibration

Accurate measurement of the hole pressure requires precise pressure measurements. Calibration of the pressure transducers was done on an in situ basis because the torque applied to the transducer on mounting it into its adaptor changed the calibration and to insure calibration was executed at the temperature of the measurements. The slit-die containing polymer melt was allowed to equilibrate at the operating temperature for at least 3 h to allow the stress in the melt to relax which was caused by thermal expansion of the melt. The exit of the die was then sealed by bolting a steel plate to the die exit, and the melt was then pressurized by pumping a test oil into the die from a dead weight tester (Chandler Engineering, model no. 23-1) until the pressure was equilibrated. Hence, the pressure versus voltage curve for each transducer was then obtained under conditions used in making the pressure measurements in the flowing melt.

### 4.5. Slit-die viscosity measurements

Obtaining viscosity data from pressure measurements in a slit-die was done by well-known methods, but they are repeated here for the sake of completeness [20]. The wall shear stress was obtained from the pressure gradient and the equation of motion as

$$\sigma_w = \frac{H}{2} \left(-\frac{dP}{dx}\right) = \frac{H}{2L_1} (PT3 - PT1) = \frac{H}{2L_2} (PT4 - PT3)$$

(10)

where the dimensions $H$, $L_1$, and $L_2$ and the pressure transducer positions are shown in Fig. 4. In principle, the values of wall shear stress ($\sigma_{w3}$ or $\sigma_{w43}$) calculated by either set of values should be identical. The wall shear rate for a Newtonian fluid or the apparent shear rate is given by

$$\dot{\gamma}_a = \frac{6Q}{H^2W}$$

(11)

where $Q$ is the volumetric flow rate. The true wall shear rate was obtained by correcting for the non-parabolic velocity profile and is given by

$$\dot{\gamma}_w = \dot{\gamma}_a \left[\frac{2}{3} + \frac{d \ln \dot{\gamma}_a}{3 d \ln \sigma_w}\right]$$

(12)

The viscosity was, of course, obtained from $\sigma_w/\dot{\gamma}_w$ using Eqs. (10)–(12).

### 5. Results and discussion

The key aspect of this paper is determining whether it is possible to obtain accurate values of $\dot{N}_1$ from hole pressure data at relatively high shear rates (i.e. >10 s⁻¹). However, before discussing this aspect, it is important to assess the accuracy of the given measurements as many sources of error can interfere with these measurements. Furthermore, we must emphasize that the hole pressure is less than 2% of the measured pressure and, hence, any source of error can significantly influence the accuracy of the measurements. In particular, errors can come from the measurement of pressures, dimensional precision of the slit-die, viscous heating, and disturbance of pressure measurements due to the hole (i.e. slot). We discuss each of these items and their influence on the measurement of the hole pressure first. Then
we compare values of viscosity measured in the slit-die with those measured by independent means and finally values of \( N_1 \) determined from hole pressure data are compared with directly measured values and those estimated from \( 2G' \). We note that Laun [25] has proposed an empiricism for relating \( N_1 \) to dynamic data at high shear rates and angular frequencies, but the exponent in this relation changes slightly and, hence, we still would have some uncertainty in making the comparison between \( P_h \) data and estimated \( N_1 \) values at high shear rates. Finally, we compare values of \( N_1 \) obtained by means of the exit pressure with those obtained by means of the hole pressure and those directly measured using a cone-and-plate device.

5.1. Accuracy of the pressure measurement system

The accuracy of the pressure transducers supplied by the manufacturer was listed at \( \pm 1\% \) of the maximum range of the transducer which would have \( \pm 34.5 \) kPa for PT1 and PT2. This would have precluded their use in the measurement of the hole pressures which ranged from 3 to 70 kPa for the polymers used in this research. However, by calibrating the transducers after being mounted in the slit-die using the procedure described in the experimental section, it was possible to measure pressures (PT1 and PT2) with considerably more accuracy as summarized in Table 3. In Table 3 the accuracy of the transducer is listed in terms of the precision as defined by Meissner [21] and was obtained experimentally by repeated measurements using static pressures. In particular, at high pressure ranges the precision was 3.5 kPa while at low pressures the precision was 0.35 kPa for PT1 and PT2. The pressures recorded by means of PT3 (0–5170 kPa) and PT4 (0–10,340 kPa) could be done with precisions of 5.2 and 10.3 kPa, respectively, at high shear stresses (high pressures) and a factor of 10 less at low shear stresses. The precision following Meissner’s approach [21] of \( P_h \) at low pressures was found to be 0.6 kPa while at high pressures it was 8 kPa. This was determined by using static pressures applied to both pressure transducers, PT1 and PT2, and recording the differences in the pressures (which for static pressures should have been zero).

Besides errors associated with the accuracy of the pressure measurements, numerous other potential sources of error existed. They could come from misalignment of pressure transducers such that PT1 and PT2 were not centered, from non-parallel walls, and from placement of the transducers relative to the die exit or entry. One way to estimate this was to compare the wall shear stress values determined by using PT4–PT3 and PT3–PT1. As an example, values of \( \sigma_{w43} \) and \( \sigma_{w31} \) are compared in Table 4 for LDPE at 150 °C. Here it can be seen that the difference between the two values is random with increasing shear rate. Hence, if the plates were not parallel, then one might expect a consistent difference between the values of wall shear stress. Another point of importance is that these pressure drops were measured in the presence of a slot of width 3.18 mm and, hence, there appears to be no systematic error due to the disturbance of flow caused by the slot. The placement of the transducers must also have been such that die entry and exit effects were negligible.

5.2. Viscosity measurements

Accurate viscosity measurements in the slit-die were the first check that meaningful pressure measurements and, hence, hole pressures could be measured. Viscosity versus shear rate for two of the polymer melts, PS-1 and LDPE are presented in Figs. 5 and 6, respectively, as representative data. As one can see the agreement between viscosity obtained from steady and dynamic shear data and the slit-die device is excellent. Even when pressure measurements were made opposite slots of different widths, the agreement was still excellent which suggested
the flow over the slot did not influence the flow on the opposite die wall. However, because the geometric factor multiplying the pressure drop was small (i.e., $H/2L_1 = 0.05$), errors in the pressure measurements could be disguised in the conversion of pressure measurements to viscosity. Hence, accurate viscosity measurements were necessary but certainly not sufficient for ensuring pressure measurements which would give accurate hole pressure values.

5.3. Hole pressure measurements

We next assessed the ability to directly measure accurate hole pressure values. In Fig. 7 we show values of $P_h$ versus wall shear stress for PS-2 obtained by means of two independent runs. Here we see at low wall shear stresses that there is about a 20% variation in $P_h$ measurements from one run to another, and at high shear stress values it is about 20% also while at the intermediate shear stresses the variation in values is less than 5%. In
results above that the pressure measurement system was operating properly, and we could expect to obtain accurate values of \( P_h \).

6. First normal stress difference measurements

Our primary interest here was to determine whether we could obtain accurate measurements of \( N_1 \) from \( P_h \) data and Eq. (5) at high shear rates or wall shear stresses. In Figs. 10–12 values of \( N_1 \) obtained from \( P_h \) data and Eq. (5) (HPB equation) are compared to values of \( N_1 \) obtained by means of a cone-and-plate rheometer and as estimated from \( 2G' \) for three polymer melts. Furthermore, data is presented in Fig. 10 for two different slot widths. The agreement between values of \( N_1 \) determined by means of \( P_h \) data and independently measured values for PS-1 (Fig. 10) is remarkably good. In particular, we observe in Fig. 10 for PS-1 that the \( N_1 \) values obtained by means of the HPB equation agree extremely well with the cone-and-plate values and those estimated by \( 2G' \) for shear rates from about 1.0 to about 70 s\(^{-1}\). For LDPE (NP-952) the agreement between \( 2G' \) and \( N_1 \) obtained by means of the HPB equation was excellent over the range of shear rates or frequency up about 70 s\(^{-1}\) (this was the limit of the \( P_h \) measurements), but \( N_1 \) values obtained by means of the cone-and-plate rose above \( 2G' \) as the shear rate or angular frequency increased. In Fig. 12 is shown data for PS-2 at 190 °C, and we see the agreement between \( 2G' \) and \( N_1 \) obtained from \( P_h \) data is very good for shear rates greater than 5 s\(^{-1}\). However, below 5 s\(^{-1}\) there is significant disagreement between the values. We note also that \( N_1 \) tends to rise above \( 2G' \) as the shear rate is increased. Although there is disagreement between \( N_1 \) values obtained from \( P_h \) data and \( 2G' \), the values obtained from \( P_h \) data are repeatable as shown by measurements made using two different slot widths. In general, the agreement between cone-and-plate values of \( N_1 \) and those obtained from \( P_h \) data was very good. It was unclear why for three of the resins...
the agreement was very good, but for one of the resins there was disagreement at the low shear rates.

At high shear rates the effect of inertia must also be considered as it tends to reduce the hole pressure. In other words, because of secondary flows in the hole, the pressure in the hole could be greater than at the flush-mounted pressure transducer. For Newtonian fluids Jackson and Finlayson and Jackson [22] found the following equation for the hole pressure for flow over a rectangular slot:

$$P_h = -0.033\sigma_w Re,$$

where the Reynolds number is defined as

$$Re = \frac{\rho BH\dot{\gamma}}{4\eta},$$

where $B$ is the slot width and $H$ is the slit-die height. Using Eqs. (13) and (14) we estimated the inertial contribution to the hole pressure for PS-1 to be about $1.4 \times 10^{-4}$ kPa at a wall shear rate of $63 \text{s}^{-1}$ and $\sigma_w = 56.5$ kPa. Hence, contributions from inertia due to secondary flows in the cavity were negligible for all the measurements reported here which was in agreement with the results of Lodge [16] for measurements on dilute polymer solutions up to shear rates of $10^6 \text{s}^{-1}$.

7. Exit pressure measurements

Although the exit pressure method for obtaining $N_1$ has been discussed in detail elsewhere [8,9] and has been reported to consistently give values of $N_1$ higher than those measured by means of the cone-and-plate device, we present results measured in the same device as used for the hole pressure measurements for the sake of completeness. Typical pressure profiles for a polymer melt (in this case PS-1) as measured by means of pressure transducers PT4, PT3, and PT1 are shown in Fig. 13. In the figure the lines represent linear fits of the pressure profiles at various shear rates. Both linear and quadratic fits of the data were used and the correlation coefficients in both cases were greater than 0.99. Representative $P_x$ values for PS-1 and LDPE (NP-952) using both linear and quadratic extrapolation are shown in Figs. 14 and 15, respectively. $P_x$ values obtained by extrapolation of the quadratic function to the die exit were always greater than those based on extrapolation of the linear function. The difference could be as large as 20–30 kPa at low shear rates and 80–100 kPa at high shear rates. These results were consistent with those reported by Laun [19] for a polymer melt. In Figs. 16 and 17 values of $N_1$ obtained by means of $P_x$ data and Eq. (9) are compared against those obtained by means of the cone-and-plate and the hole pressure. At low shear rates there was actually reasonable agreement between $N_1$ values measured by means of a cone-and-plate device and those obtained by means of $P_x$ values and Eq. (9) especially for PS-1. However, as the shear...
rate was increased, the values obtained from \(P_x\) data were significantly higher than the other \(N_1\) values reaching levels of about 100% higher. These results were consistent with those reported by others [8,9]. The large discrepancy was mostly attributed to the lack of validity of the original assumption in the derivation of Eq. (9) (i.e. that the flow remains steady shear flow until the die exit) due to the fact that there was a strong rearrangement of the flow and stress fields at the die exit. No matter how small the rearrangement region is, the second term always is present in Eq. (8) and can lead to a significantly higher value of the exit pressure [9]. For the most part the measurements reported here were devoid of many of the errors which could contaminate the exit pressure measurements, and, yet, the values of \(N_1\) obtained from \(P_x\) data were still consistently higher than values of \(2G'\) and \(N_1\) values obtained from \(P_h\) data. Factors such as viscous heating (when precautions are not taken) and pressure dependence of viscosity can also affect the linearity of the pressure profiles and, hence, the extrapolation to the die exit to obtain the exit pressure. Again the exit pressure was typically less than 1% of the pressure measured.

8. Conclusions

In this paper we showed that, in general, consistent and accurate measurements of the hole pressure could be obtained at relative high shear rates for polymer melts when the pressure transducers were calibrated after mounting in the die and great care was taken to ensure accurate placement of the transducers. Even at shear rates as high as about 70 s\(^{-1}\) and wall stress values approaching 10\(^5\) Pa, hole pressure data coupled with the Higashitani–Pritchard–Baird (HPB) relation (Eq. (5)) gave values of \(N_1\) which agreed well with values of \(2G'\) for the four polymer melts used in this work. Consistent with the precision of the hole pressure measurements, values of \(P_h\) for a melt of low elasticity were found to be negligible. Only for one of the melts (a polystyrene) was there any significant disagreement between the values of \(N_1\) obtained by means of \(P_h\) and that obtained by means of a cone-and-plate device, and this was only at low shear rates (<5 s\(^{-1}\)). It is remarkable that the HPB relation is apparently still valid at conditions where the stress is asymmetric about the center-line (this is true just because of the pressure gradient across the slot alone) and the motion in the hole is significant [23]. Finally, the exit pressure method for obtaining \(N_1\), even when measured under conditions in which highly precise measurements were made, gave values which were consistently higher than \(2G'\) and those measured using the hole pressure.

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References

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