Determination of the entrance pressure drop in capillary rheometry using Bagley correction and zero-length capillary

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ABSTRACT

In capillary rheometry, the extra pressure drop associated with the contraction flow at the capillary entrance can be corrected afterwards by using the well-known Bagley correction. Another method for correcting the entrance effect is to use a nominally zero-length capillary (also called the orifice die). These two methods were compared by performing viscosity measurements for two polymers: LDPE and PS. The aim was to find out how well the entrance pressure drop measured by zero-length capillary corresponds to that obtained from the Bagley correction.

INTRODUCTION

Contraction flow at the capillary entrance region in the capillary rheometer causes an extra pressure drop due to stretching of fluid elements. In principle, the significance of the entrance pressure drop compared to the pressure drop across the capillary decreases with increasing L/D (length to diameter ratio). The large L/D however can lead to other errors; the longer the capillary is, the greater the pressure effect becomes. Increased pressure can have a significant effect on viscosity of some polymers and therefore too large capillary L/D should be avoided. Also the role of viscous heating gets more pronounced with longer capillaries. Hence it is preferable to perform the viscosity measurements using capillaries with relatively small L/D to avoid the effects of pressure and viscous heating. This requires a correction for the entrance pressure drop.

Hatzikiriakos et al.[1] have investigated the differences in the entrance pressure drop using conventional Bagley correction and zero length capillary. They concluded that when the extrudate flows freely without sticking to the walls of the capillary exit region, the pressure drop obtained with zero length capillary was almost equal to that of Bagley plot extrapolation.

Also Kim and Dealy[2] have made a comparison between different die geometries ending up to similar results. They found out that the entrance angle has to be greater than 90° and the exit section must be wide enough to prevent the melt sticking to its wall. With a capillary design fulfilling these requirements they discovered that the entrance pressure drop from zero-length capillary was very close to that extrapolated from linear Bagley plots.

Sunder and Goettfert[3] used the zero-length capillary for evaluating extensional viscosity and compared these results to Bagley correction. They emphasized the importance of the right choice of capillaries in Bagley correction: the lengths of the chosen capillaries must not be too close to each other because otherwise the possible small measurement errors will be accumulated. According to them an appropriate choice is three capillaries with L/D ratios of 5, 10 and 20. Entrance pressure drop obtained by zero-length capillary appeared to be greater than those extrapolated from Bagley plots. The difference was most significant, even 50 %, at low shear rate region. Also they stated that the main reason for these differences is the fact that the melt sticks to the capillary exit area.

Mitsoulis and Hatzikiriakos[4] investigated the effect of the capillary entrance angle on the pressure drop and concluded that the effect is significant but rather ambiguous; the pressure drop decreased with increasing contraction angle from 10° to approximately 30°-45° but then slightly increased as the contraction angle was increased up to 150°.

METHODS

The correction for the extra pressure drop at capillary entrance can be performed in two different ways:

<u>Entrance pressure drop from Bagley correction</u>

Conventionally the entrance pressure drop is evaluated experimentally from the measurement data of at least two, preferably three, capillaries of same diameter but different length. This is accomplished by plotting the pressure drop (Δp) at constant shear rate versus capillary L/D and fitting a linear fit on the data. The linear Bagley plot is then extrapolated to zero and the point at which the line intersects with y axis is the entrance pressure drop (Δp_e). True shear stress (τ_w) can be calculated as

$$\tau_w = \frac{\Delta p - \Delta p_e}{2L/R} \qquad (1)$$

where L/R represents the capillary length to radius ratio.

However, sometimes the points do not fall in the linear fit but tend to curve either upwards or downwards. If the Bagley plot shows a curvature upwards it is probably due to pressure effects. Therefore sometimes assuming a parabolic pressure profile across the die instead of linear one gives a better fit[5]. Nevertheless at very low pressure level the second order fit can give unrealistic results due to the errors which are significant compared to the pressure. A non-linear fit should be used if it describes the measured data better than a linear fit[3].

<u>Entrance pressure drop from zero-length capillary</u>

A zero-length capillary theoretically has a length-to-radius ratio of zero. Thus as the polymer melt flows through a zero-length capillary contraction, it converges and diverges immediately again. The flow length is not sufficient to allow the formation of fully developed flow. In theory the only pressure drops occurring in a zero-length capillary are the ones due to converging of the flow at the entrance of the capillary and diverging at its exit.

Based on this assumption, the pressure drop obtained from zero-length capillary measurements is directly comparable to the entrance pressure drop in longer capillaries. The advantage of the zero-length capillary over the conventional Bagley correction with three or more capillaries is that the number of tests needed to complete a set of measurements for one material is smaller. Only measurements with zero-length capillary and with one longer capillary are required for a sufficient amount of data for the entrance effect correction.

However, certain factors can affect so that the entrance pressure drop values obtained using zero-length capillary become unrealistically high. Probably the most important one is the fact that if the melt extrudate sticks to the walls of the capillary exit region, it causes an extra viscous flow and thus extra pressure drop in that area[1, 2]. This leads to excess estimations of the entrance effect in capillary flow. Secondly, as the pressures associated with the zero-length capillary are very low, an experimental error can be relatively high compared to the measured values[6]. Two geometrical failures also cause additional pressure drop in zero-length capillary: In order to manufacture a rigid capillary, it must have a real length greater than zero. This length, even a small one, causes an additional pressure drop. Neither it is possible to manufacture a capillary as short with both inlet and outlet angle of 180° [3].

Despite of these facts some authors have found that the entrance pressure drop from zero-length capillary corresponds well with the one extrapolated from the Bagley plots[2,6].

EXPERIMENTAL

In this study the entrance pressure effect was evaluated for two polymers. A first one was low density polyethylene (LDPE) Lupolen 1840 H supplied by Basell and the second one polystyrene (PS) Polystyrol 143E by BASF. Two different approaches for evaluating the entrance pressure drop were compared. First it was extrapolated from Bagley plots and second the entrance pressure drop was evaluated using a zero-length capillary. The aim was to find out whether a zero-length capillary geometry used here is a suitable device for determining entrance pressure drop in capillary flow and whether it can be used instead of Bagley correction.

Measurements were carried out using Göttfert Rheograph 6000 capillary rheometer. Tests were performed at shear rates from 50 1/s up to 2000 1/s.

Various different geometries of zero-length capillaries exist. Kim and Dealy[2] used one with a wider expansion region thus avoiding the melt sticking to the wall during the extrusion. The die design used in our experiments deviates to a degree from the ones used by Kim and Dealy[2] or by Hatzikiriakos et al[1]. In our zero-length capillary the contraction is abrupt (180°) but the exit area has a conical shape. The true capillary length is 0.2 mm thus it fills the condition that the real length must be below 0.5 mm as stated in earlier study[2]. The geometry of the zero-length capillary is presented in Fig. 1. Due to the geometry of the capillary it is almost impossible to prevent the melt from sticking to the exit area.

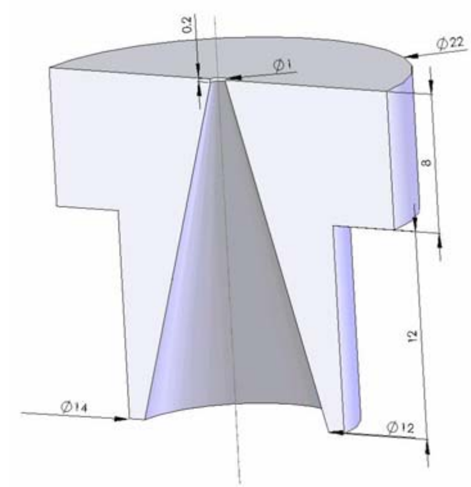

Figure 1. Zero-length capillary

Sunder and Göttfert[3] found out that the sticking was lesser at higher shear rates but we didn't observe any significant difference between low and high shear rates.

RESULTS

The entrance pressure drop was evaluated in two ways:

1) By extrapolation of linear Bagley plots. For this viscosity measurements with three capillaries having the diameter of 1 mm and lengths of 5, 10 and 20 mm were performed.

2) By direct measurements with zero-length capillary.

<u>LDPE</u>

For Bagley plots, the pressure drop was obtained from capillaries having L/D of 5, 10 and 20. The extrapolation of the linear Bagley plots of LDPE showed a good fitting (Fig. 2).

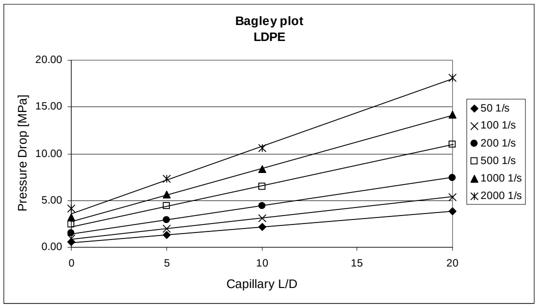

Figure 2. Bagley plot for LDPE at shear rates 50-2000 1/s (Linear fit for capillary L/Ds of 5, 10 and 20)

The data from zero-length capillary is not used in generating the Bagley plot but is included in the figure for comparison. As expected, the zero-capillary measurements give somewhat higher pressure drop than estimation from Bagley plot. Entrance pressure drop values from different methods are given in Table 1. $\dot{\gamma}$ is the apparent shear rate and Δp_{eLin}, $\Delta p_{e0/1}$ and relative error denote the entrance pressure drop evaluated from linear Bagley plot, zero-capillary measurements and the relative difference between them respectively.

Table 1. Entrance pressure drop form different methods for LDPE

$\dot{\gamma}$ [1/s]	Δp_{eLin} [MPa]	$\Delta p_{e0/1}$ [MPa]	relative error [%]
50	0.5081	0.5349	5.28
100	0.8183	0.8940	9.25
200	1.3642	1.4631	7.25
500	2.1177	2.4573	16.03
1000	2.7430	3.1998	16.65
2000	3.5698	4.1597	16.53

From the Table 1 it can be observed that the relative error of the entrance pressure drop from zero-length capillary measurements is higher at high shear rates. This observation deviates from the earlier study[3], in which the relative error was found to be very high, even 50 %, at low shear rates and to decrease to approximately 10 % at higher shear rates. In the earlier study, a gas-driven capillary rheometer enabling an accurate measurement of shear rates as low as 0.1 1/s was used[3]. In our tests the lowest shear rate was 50 1/s. This shear rate was chosen for a lower limit in order to ensure the sufficient resolution of the pressure transducer within the measured pressure range.

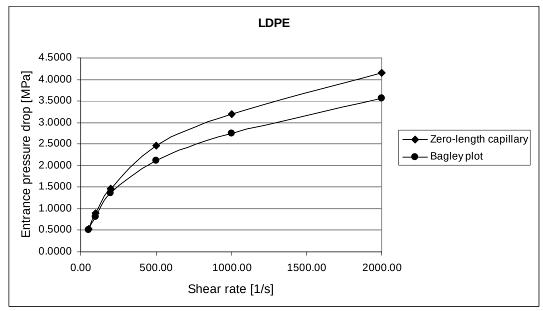

Figure 3. Entrance pressure drop vs. shear rate for LDPE

PS

Also the Bagley plots of polystyrene showed good linearity (Fig. 3).

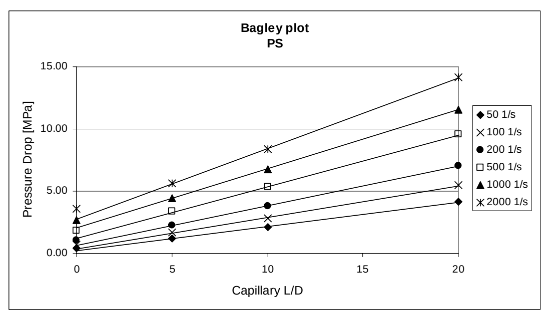

Figure 4. Bagley plot for PS at shear rates 50-2000 1/s (Linear fit for capillary L/Ds of 5, 10 and 20)

Zero-capillary measurements gave significantly larger overestimation of entrance pressure drop for PS than for LDPE as can be seen in Table 2. The results are now in accordance with those of Sunder and Göttfert[3] showing a greater relative error at low shear rates.

Table 2 Entrance pressure drop form different methods for PS

$\dot{\gamma}$ [1/s]	Δp_{eLin} [MPa]	$\Delta p_{e0/1}$ [MPa]	relative error [%]
50	0.1881	0.4568	142.83
100	0.3542	0.6937	95.85
200	0.6351	1.0552	66.15
500	1.2311	1.8173	47.62
1000	2.0078	2.6576	32.36
2000	2.7650	3.6077	30.48

The difference in the zero-capillary measurements for PS is very high: Even at lowest it is almost twice as high as the largest error for LDPE.

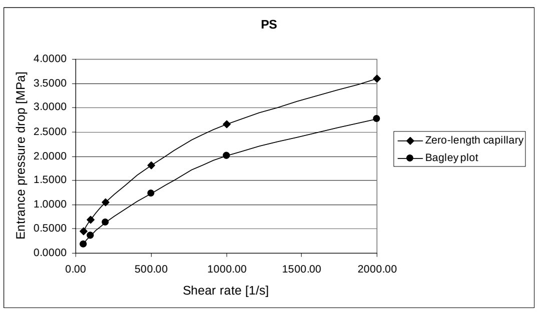

Figure 5. Entrance pressure drop vs. shear rate for PS

CONCLUSIONS

Zero-length capillary measurements gave higher entrance pressure drop values than Bagley correction. This result was expected due to the fact that the geometry of the capillary caused the melt sticking to its exit area. For LDPE the error was smaller at low shear rates whereas for PS the error decreased as the shear rate increased.

The results for both LDPE and especially for PS prove that the geometry of the zero-length capillary used here is not best possible because it is impossible to prevent the melt sticking to the conical exit area. Another reason for high entrance pressure drop values might be the finding of Mitsoulis and Hatzikiriakos[4], that the entrance pressure drop increases slightly when the inlet angle is equal to or larger than 150°. As it is technically impossible to manufacture a zero-length capillary having both inlet and outlet angle of 180°, it seems to be more reasonable to use a geometry similar to that developed by Kim and Dealy[2]. The exit area should be wide enough with an outlet angle of 180° and inlet angle small enough to allow a manufacture of rigid capillary geometry but large enough not to cause extra pressure drop.

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