Evaluation of Different Methods for Determining the Entrance Pressure Drop in Capillary Rheometry

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Abstract:
Two approaches for determining the entrance pressure drop in capillary rheometry were compared with low-density polyethylene and polystyrene melts as test fluids. Direct measurements with the orifice die were found to yield higher values for the entrance pressure drop, and hence lower values for the wall shear stress, than the Bagley correction method. This was postulated to be caused by the sticking of the melt to the wall of the outlet region of the orifice die. The additional pressure drop created in the outlet region of the orifice die, when the flowing material fills it completely, was also evaluated by means of numerical flow simulation.

Zusammenfassung:
Für die Bestimmung des Eintrittsdruckverlusts im Kapillarrheometer werden für zwei Polymerschmelzen (Polyethylen niedriger Dichte und Polystyrol) zwei Ansätze miteinander verglichen. Messungen mit einer Drosseldüse (nominelle Kapillarlänge null) ergaben im Vergleich zur Bagley Korrektur höhere Werte für den Einlaufdruckverlust und damit geringere Werte für die Wandschubspannung. Es ist anzunehmen, dass dieser Unterschied durch das Anhaften der Schmelze im Düsenaustrittsbereich der Drosseldüse entsteht. Dieser zusätzliche Druckverlust wurde mit Hilfe einer numerischen Fließsimulation untersucht, wobei angenommen wurde, dass der gesamte Austrittsbereich der Drosseldüse mit Schmelze bedeckt ist.

Résumé:
Deux approches ont été comparées afin de déterminer la chute de pression d'entrée en rhéomètrie capillaire en utilisant des fondus de polyéthylène basse densité et de polystyrène comme fluides tests. Les mesures directes en sortie de filière ont conduit à des valeurs plus élevées pour la chute de pression en entrée, et donc à des valeurs plus petites pour la contrainte à la paroi, que les valeurs obtenues avec la méthode corrective de Bagley. Ceci a été potentiellement attribué à l'adhésion du fondu à la paroi de la région externe de la filière. La chute de pression additionnelle créée dans cette région, lorsque le fluide en écoulement la remplit entièrement, a été également évaluée au moyen d'une simulation numérique de l'écoulement.

Key words: capillary rheometer, Bagley correction, orifice die, entrance pressure drop

1 INTRODUCTION

In the capillary rheometer, a piston moving in a cylindrical reservoir drives the test fluid through a small capillary. The pressure drop across the capillary is typically determined by a pressure transducer mounted in the reservoir just above the capillary entrance. To obtain a true wall shear stress, i.e., a wall shear stress corresponding to the fully developed flow in the capillary, the measured pressure drop must be corrected for the additional pressure drop caused by the passage of the fluid through a contraction at the entrance to the capillary. In principle, the significance of the entrance pressure drop compared to the total pressure drop across the capillary decreases with increasing length-to-diameter (L/D) ratio of the capillary. Indeed, it is common practice to use a single long capillary ($L/D \geq 30$) in the hope that this overwhelms the entrance effects. Unfortunately this approach may introduce other error sources. That is, both the pressure effect on viscosity and viscous heating tend to become more pronounced as the L/D ratio of the capillary increases. Hence, it is preferable to use capillaries with small to moderate L/D ratios. This, however, increases the importance of performing the correction for the entrance pressure drop.

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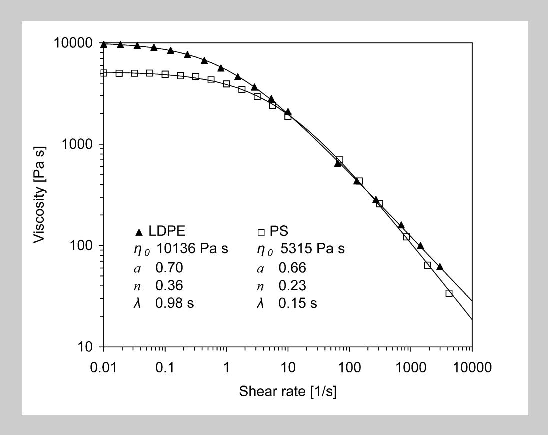

Figure 5: Viscosity data for LDPE and PS measured by capillary rheometer (high shear rates) and rotational rheometer (low shear rates), and the best fits to the data sets according to the Carreau-Yasuda model, Eq. 6 (fitting parameters are shown in the figure).

Table 2: Entrance pressure drop measured with orifice die, Δp_{eo}, extrapolated from Bagley plots, Δp_{eB}, difference between them, $\Delta p_{eo} - \Delta p_{eB}$, and calculated pressure drop values for the outlet region of the orifice die, Δp_{Calc}.

	$\dot{\gamma}$ [1/s]	Δp_{eo} [MPa]	Δp_{eB} [MPa]	Δp_{eo}-Δp_{eB} [MPa]	Δp_{Calc} [MPa]
LDPE	50	0.5471	0.4820	0.0651	0.2367
	100	0.9038	0.7954	0.1084	0.3322
	200	1.4753	1.3061	0.1692	0.4572
	500	2.4231	2.0412	0.3819	0.6806
	1000	3.1778	2.6434	0.5344	0.9064
	2000	4.2257	3.3831	0.8426	1.1952
PS	50	0.4568	0.2125	0.2443	0.2224
	100	0.7010	0.3786	0.3225	0.3224
	200	1.0625	0.6009	0.4616	0.4500
	500	1.8417	1.2384	0.6033	0.6663
	1000	2.6746	1.9761	0.6985	0.8704
	2000	3.6175	2.7650	0.8525	1.1136

these data points are nevertheless shown in the Bagley plot of Figure 4 for illustrative purposes.

As anticipated, the orifice die produces higher values for the entrance pressure drop, and hence lower values for the wall shear stress, than the extrapolation from the Bagley plot. The values of τ_w obtained through these two methods as well as the values obtained by neglecting the correction for the entrance pressure drop with the capillary of L/D = 30 are listed in Table 1. The overestimation of Δp_e with the orifice die is attributable to the sticking of the melt to the wall of the outlet region, which is practically impossible to prevent in our orifice die. Note, however, that the values of τ_w obtained using the orifice die match more closely the Bagley corrected values than those obtained from a single L/D = 30 capillary without the correction for the entrance pressure drop. In the case of PS the pressure apparently affects the viscosity with the capillary of L/D = 30, as already inferred on the basis of Figure 4. Indeed, contrary to common belief, the accuracy of single capillary measurements can not necessarily be improved by increasing the L/D ratio of the capillary, because other factors, such as the viscosity dependence on pressure and viscous heating, are more likely to come into play.

To estimate the parameter values of the Carreau-Yasuda viscosity model, Eq. 2, used in the numerical flow simulation, the viscosity values for LDPE and PS were determined from the present capillary rheometry data. The Bagley as well as Rabinowitsch corrections were applied to the raw data. In order to improve the fit, additional viscosity data for the low shear rate region were taken from the rotational rheometer measurements with the cone-and-plate geometry. The measured viscosity data and the resulting Carreau-Yasuda model fits are given for both materials in Figure 5.

Table 2 presents the pressure drop values measured directly with the orifice die, Δp_{eo}, the Bagley plot extrapolated values, Δp_{eB}, and the differences between them, $\Delta p_{eo} - \Delta p_{eB}$. In addition, the calculated pressure drops in the conical outlet region of the orifice die, Δp_{Calc}, are given in this table. If this outlet region is full of melt during the experiment, the values obtained for $\Delta p_{eo} - \Delta p_{eB}$ should approximately correspond to those of Δp_{Calc}. This seems to be more closely the case for PS implying that it sticks more to the outlet wall than LDPE. For LDPE the sticking seems to increase with increasing apparent shear rate, whereas for PS an opposite behaviour can be observed. This apparently also explains why the ratio $\Delta p_{eo}/\Delta p_{eB}$ remains almost unchanged for LDPE ($\approx$ 1.2), but decreases with increasing apparent shear rate for PS (from 2.1 to 1.3). It is worth noting, however, that the differences in the sticking behaviour between PS and LDPE are hard to observe visually. Moreover, the majority of the pressure drop in the outlet region of the orifice die develops right at the beginning, where the sticking is particularly difficult to see during the experiment.

The smaller relative difference between Δp_{eo} and Δp_{eB} for LDPE may also be attributed to the extensional flow properties. Owing to the long-chain-branched structure of LDPE it has a relatively high extensional viscosity, which, on the other hand, largely determines the pressure drop in the contraction flow at the capillary entrance. Thus, a larger entrance pressure drop for LDPE means a smaller relative difference between Δp_{eo} and Δp_{eB}. No attempts were made to simulate directly the contraction flow. It is well known that the purely viscous constitutive equation used here (Eq. 2) is incapable of providing realistic descriptions for the extensional viscosity, which makes such simulations quite useless. The numerical flow simulations for estimating the entrance pressure drop in capillary rheometry have been performed by some authors, notably by Mitsoulis and Hatzikiriakos [10] using the integral-type K-BKZ constitutive relation. They attained reasonable entrance pressure drop predictions for low contraction angles (up to 30°), where the flow is apparently shear-dominated, but for larger contraction angles the entrance pressure drop was under-predicted.

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For achieving a free flow of an extrudate with no sticking to the outlet wall, the orifice die geometry similar to that of [7] would definitely work better than the present one. However, for rigidity reasons, such a wide exit area is evidently only possible when the contraction at the entrance to the capillary is non-abrupt, i.e., the contraction angle is less than 180°. We, however, prefer to use the orifice die which has an abrupt contraction like our other capillaries. By exploiting the results of the flow simulations, the entrance pressure drops obtained from the orifice die could possibly be corrected to some extent at least for the case when the outlet region of the orifice die is full of melt. In this case, as already described above, the overestimation with the orifice die should be close to Δp_{Calc}. When we calculated the ratio $(\Delta p_{eB} + \Delta p_{Calc})/\Delta p_{eB}$ we found that it approximately varies from 1.3 to 1.5 for LDPE and from 1.4 to 2.1 for PS. Thus, as an ad hoc overall correction, one might divide the pressure drop values obtained from the orifice die by 1.5 (the applicability of this value for other materials is of course not known). This kind of correction might possibly be applied to the measurements of viscosity under elevated pressures by means of a capillary rheometer equipped with a pressure chamber. These measurements are typically time-consuming and therefore the use of the orifice die would be of great benefit. Moreover, in this type of measurement the outlet region of the orifice die is indeed full of melt.

A factor which may also be present in the capillary flow of polymer melts is the wall slip. The occurrence of slip would, of course, complicate the interpretation of the measured data. As is well known, particularly linear polyethylenes like LLDPE and HDPE are prone to slip. On the other hand, with the polymers used in this study (PS and LDPE), the degree of wall slip is generally small [11]. Consequently, we postulate that the wall slip is unlikely to play a significant role in the present experiments.

4 CONCLUSIONS

Direct measurements with the orifice die gave higher entrance pressure drop values than the Bagley correction method. This result was anticipated due to the fact that the geometry of the orifice die caused the melt sticking to its outlet region. Generally, the relative difference to the Bagley corrected values was larger with PS than with LDPE. Nevertheless, the wall shear stress values obtained using the orifice die were always closer to the Bagley corrected values than those obtained from a single $L/D = 30$ capillary without the correction for the entrance pressure drop. Numerical flow simulation was also used to elucidate the additional pressure drop arising in the outlet region of the orifice die and the simulation results suggested that PS sticks more to the outlet wall than LDPE.

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