Influence of the Die Entry Angle on the Entrance Pressure Drop, Recoverable Elastic Energy, and Onset of Flow Instability in Polymer Melt Flow

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Synopsis

An experimental study has been carried out to investigate the effects of die entry angle on the entrance pressure drop, recoverable elastic energy, and onset of melt fracture in the flow of viscoelastic polymeric melts through a capillary die. For the study, capillaries with an L/D ratio of 4 and with varying die entry angles, 15°, 30°, 60°, 90°, 120°, and 180°, were used for extruding low-density polyethylene and high-density polyethylene. Measurements were taken of wall normal stresses along the upstream reservoir section, tapered conical section, and straight capillary section.

INTRODUCTION

It has been a major concern of the polymer-processing industries to develop a better means of designing extrusion dies to permit smoother flow and higher productivity. Although there has been some limited success, the techniques available so far have been based to a great extent on trial and error, and they are among the most carefully guarded secrets in the industry. The difficulty of undertaking a fundamental study lies mainly in the often complicated flow geometry to be considered.

Even in the case of circular capillary dies, the design problem is far from solved, because a slight change in the upstream die configuration may affect the pressure drops of the melt considerably and hence the extrusion conditions. Furthermore, because of the viscoelastic nature of the melts, the flow behavior of such materials in the entrance region is not fully understood at present, although a number of explanations have been reported in the literature.¹⁻⁴

Many extrusion dies of commercial importance, for instance, dies for fiber spinning and film extrusion, have a much more complicated entrance geometry than the straight tube with a flat entrance. In order to illustrate the point, some typical entrance geometries for spinnerettes are shown in Figure 1. There are primarily two processing reasons for using tapered entrance sections in extrusion dies. One is to avoid the dead space at the corners of the reservoir section above the die entrance, which is created by



Fig. 1. Typical entrance geometries for spinnerettes.

converging melt flow streamlines. It has been a well-established fact that there is no flow of polymer melts outside the converging streamlines. In industrial polymer processing, some polymers (e.g., PVC resins and nylon 6) form crosslinkages and/or degrade when they stay in the stagnant zone, behavior which is detrimental to smooth operation. Therefore dies with tapered entry would eliminate such a problem.

The other reason, equally as important as the first, is that a die with tapered entry can yield an increase in productivity by permitting one to operate with higher values of the critical shear rate. This is the shear rate at which melt flow instability, otherwise known as "melt fracture," starts to occur. In the past, some interesting studies have been reported by several investigators on the effect of die entry geometry on the critical shear rate.⁵⁻¹¹

In this paper, we present our recent experimental study carried out to investigate the effects of die entry angle on the entrance pressure drops, on the elastic properties of the polymer melts investigated, and on the critical shear rates at which melt fracture starts to occur. For the study, capillaries with a fixed L/D ratio and with varying die entry angles were used for extruding low-density polyethylene and high-density polyethylene. Measurements were taken of wall normal stresses along the upstream reservoir section, tapered conical section, and capillary section. These measurements were used to determine the entrance pressure drops, pressure gradients, and exit pressures, which then permitted us to determine both the viscous and the elastic properties of the melts investigated.

EXPERIMENT

The apparatus used for the present study is essentially the same as that used for previous studies by the author,¹²⁻¹⁵ except for the design of the capillary dies themselves. In the present study, six capillary dies were used having entry angles of 15°, 30°, 60°, 90°, 120°, and 180° (flat entrance). Figure 2 gives a schematic of a typical die, and Table I gives the dimensions of the dies and also the positions of pressure tap holes in individual dies. Note that the capillary diameter of all dies was 0.125 in., giving an L/D ratio of about 4 for the straight capillary section. Note also that the upstream reservoir section was 0.750 in. in diameter, giving a reservoir-to-capillary diameter (D_R/D) ratio of 6.



Fig. 2. Schematic of the capillary die with tapered die entry.

TABLE I											
Dimensions of Dies and Positions of Pressure Tap H	oles										

Die	En- try an- e gle	<i>L</i> _c , ^a in.	L_{D} , ^b in.	Positions of pressure tap holes,° in.						
				(1)	(2)	(3)	(4)	(5)	(6)	(7)
A	15°	2.374	0.512	-2.475	-1.070	-0.376	0.046	0.252	0.337	0.411
В	30°	1.166	0.507	-1.186	-0.421	-0.216	0.020	0.189	0.250	0.423
С	60°	0.541	0.512	-1.040	-0.368	-0.184	0.037	0.249	0.334	0.409
D	90°	0.321	0.512	-0.812	-0.212	-0.111	0.039	0.245	0.331	0.413
\mathbf{E}	120°	0.180	0.512	-0.682	-0.129	-0.072	0.040	0.250	0.330	0.406
F	180°		0.512	-0.508	_		0.038	0.137	0.267	0.421

• The reservoir diameter is 0.750 in.

^b The capillary diameter is 0.125 in.

• The entrance of the capillary is chosen as reference position (x = 0).

The experimental procedure is straightforward, as described in earlier papers by Han.^{12,13} Briefly stated, we measured the wall normal stresses (sometimes referred to as wall pressures) of polymer melts flowing from a reservoir section into a circular die. This was done with melt pressure transducers (Dynisco, Model PT422), using a potentiometer and a null detector. The entire system was electrically heated with Calrod heaters, and the temperature was controlled within $\pm 0.5^{\circ}$ F by a Thermistoroperated thermal regulator.

Polymers used for the study were low-density polyethylene (Union Carbide, PEP 211), which is believed to have much long-chain branching, and high-density polyethylene (Union Carbide, DGNB 3825). Extrusion experiments were carried out at 154° C.

RESULTS AND DISCUSSION

Wall Pressure Measurement and Flow Curves

Figure 3 shows representative axial pressure profiles for high-density polyethylene at 154°C, in the die having an entrance angle of 90°. From

HAN



Fig. 3. Representative profiles of wall normal stresses in flow of high-density polyethylene melts at 154° through a tapered conical die ($\alpha = 90^{\circ}$).

these profiles one can evaluate three important variables, namely, the entrance pressure drops, constant pressure gradients, and the exit pressures, all as functions of shear rate. Similar plots were constructed for the other dies having entrance angles of 15° , 30° , 60° , 120° , and 180° . However, space limitation does not permit us to present those plots here.

The constancy of the pressure gradient, $-\partial p/\partial x$, permits one to calculate the true wall shear stress τ_w by

$$\tau_w = \left(-\frac{\partial p}{\partial x}\right) \frac{D}{4} \tag{1}$$

in which D is the capillary diameter. It is to be noted in eq. (1) that no end-correction is necessary in this approach.



Fig. 4. Flow curves of high-density polyethylene at 154°C.

Figure 4 gives plots of shear stress versus shear rate, the so-called flow curves, for high-density polyethylene at 154° C, with die entry angle as a parameter. It can be seen that, as expected, the entrance angle does not affect the flow curves. This indicates that we can give credence to the experimental technique employed and the accuracy of the measurements. Similar plots for low-density polyethylene at 154° C are shown in Figure 5. Note, however, that the data for dies having entrance angles of 120° and 180° are not shown in Figure 5. This is because melt fracture was observed with these two dies, and therefore measurements of wall pressure were not recorded. As noted earlier in studies by Han and Lamonte,^{14,15} when melt fracture occurred there were wild fluctuations in pressure both in the capillary and upstream of the reservoir section. More will be said later about melt fracture as affected by the entrance angle.



Fig. 5. Flow curves of low-density polyethylene at 154°C.





Fig. 6. Plots of entrance pressure drop vs. shear rate for high-density polyethylene.

Effects of Die Entry Angle on the Entrance Pressure Drops

Figure 6 shows plots of the entrance pressure drop versus shear rate for high-density polyethylene at 154°C, with die entry angle as a parameter. Two things are worth noting. One is that, for a fixed die entry angle, the entrance pressure drop increases with shear rate. Another is that the entrance pressure drop first decreases as the entrance angle is increased from 15° to 60° and then levels off as the entrance angle is increased further. Similar observations can be made for low-density polyethylene, as shown in Figure 7.

The entrance pressure drops are larger for small die entry angles than for large ones because the conical section is longer for small angles. For instance, the conical section is 2.374 in. long for the entry angle of 15°,



Fig. 7. Plots of entrance pressure drop vs. shear rate for low-density polyethylene.



Fig. 8. Plots of exit pressure vs. shear rate for high-density polyethylene.

whereas it is only 0.541 in. for the entry angle of 60° and 0.321 in. for the entry angle of 90° . Note that when the entry angle is as small as 15° , the conical section of such a die can almost be considered as an extension of the capillary section.

However, in the commercial extrusion of polymeric materials, dies having smaller entry angles are preferred because the critical shear rate at which melt fracture starts to occur is greater, and, hence, the throughput rate can be greater. There are several studies reported on the effect of die entry angle on the critical flow conditions, notably, by Tordella^{7.8} and Bagley.^{5.6} Later in this paper, we shall elaborate further on this matter in view of our own data.

Effects of Die Entry Angle on the Recoverable Elastic Energy

Figure 8 shows plots of exit pressure versus shear rate for high-density polyethylene at 154°C, with die entry angle as a parameter; and Figure 9 shows similar plots for low-density polyethylene at 154°C. It is seen in these figures that, for a change in entry angle between 180° and 60°, there is virtually no significant change in exit pressure. However, a further decrease from 60° to 15° results in an appreciable reduction of exit pressure. This drop in exit pressure appears to be primarily a consequence of the increased length of the conical section. To better understand this, the reader is referred to an earlier study by Han et al.,¹² who showed that exit pressure first decreases with die length and then starts to level off at a certain critical value of the die length.

At this point, it is appropriate to consider the physical origin of exit pressure. In previous studies,^{3,12} Han contended that the existence of exit pressure is a manifestation of the elastic behavior of the material and that exit



Fig. 9. Plots of exit pressure vs. shear rate for low-density polyethylene.

pressure results from the residual elastic energy stored in the melt. His contention was based on the argument that exit pressure is the elastic internal energy which was originally put into the melt as it underwent excessively large pressure drops at the die entrance. Earlier, LaNieve and Bogue⁴ presented the view that for viscoelastic fluids the total entrance pressure drop may be divided into two parts, the viscous loss and the elastic loss, and that a portion of the elastic loss at the die entrance will be retained in the fluid and will then be recoverable at the die exit. Following the view of LaNieve and Bogue, Han³ has recently noted that, for several polymer melts investigated, 90% and more of the total entrance pressure drops were attributable to the elastic loss and that a little over 10% of the elastic loss was recovered at the die exit as exit pressure. Note that in the particular study referred to above, Han³ used dies of flat entry (180°).

Therefore, in order to observe the effect of die entry angle on the recoverable elastic energy from a different point of view, plots of the ratio of the exit pressure to entrance pressure drop $(P_{\text{exit}}/\Delta P_{\text{ent}})$ versus shear rate are given in Figure 10 for high-density polyethylene and in Figure 11 for low-density polyethylene, with die entry angle as a parameter. It is seen in these figures that, for a change in entry angle between 180° and 60°, there is no significant change in $P_{\text{exit}}/\Delta P_{\text{ent}}$. However, for entry angles of 15° and 30°, an appreciable drop in $P_{\text{exit}}/\Delta P_{\text{ent}}$ is noticeable. Figures 10 and 11 indicate to us that the fraction of the total entrance pressure drop recoverable at the die exit decreases rapidly at and below a certain critical value of die entry angle. This also indicates to us that die swell will also be reduced, in view of the fact that both the die swell and exit pressure have the same physical origin.¹² As a matter of fact, Poller and Reedy (16) have shown that die swell ratio varies with die entry angle and have thus corroborated the results presented in Figures 8 to 11.



Fig. 10. Plots of $P_{\text{exit}}/\Delta P_{\text{ent}}$ vs. shear rate for high-density polyethylene.

Effects of Die Entry Angle on the Critical Flow Conditions

It has long been known to the polymer processing industry that dies having small entry angles help to reduce the severity of extrudate distortion. To explain this, Bagley and Schreiber⁵ have stated that as the die entry angle decreases, there is less chance for the melts, relatively unoriented, to be introduced into the main streamlines from the "dead" space, and hence yielding less of a degree of extrudate distortion which is due to a more homogeneous state in strain. This view was based on the experimental evidence of Bagley and Birks¹ that, in polymer melt flow, materials start to build converging streamlines in the upstream of the reservoir section, leaving the material outside the main streamlines (i.e., at the corner) less oriented.

More recently, Ballenger and White² have corroborated the earlier observations by Bagley and Birks¹ and have pointed out further that the shape of flow patterns at the entrance of a die varies from one material to another. For instance, they have shown that branched (low-density) polyethylene creates a relatively large "dead" space outside the converging flow streamlines (i.e., yields a small characteristic entrance flow angle), whereas linear (high-density) polyethylene creates a fairly small "dead" space (i.e., yields a large characteristic entrance flow angle). These authors also observed that as the flow rate is increased, the well-established



Fig. 11. Plots of $P_{\text{exit}}/\Delta P_{\text{ent}}$ vs. shear rate for low-density polyethylene.

converging flow streamlines get distorted and are eventually ruptured, yielding severely distorted extrudate.

While there has been some controversy over the cause (or causes) of melt fracture, there appears to have been general agreement among researchers that the elastic properties of a melt are primarily responsible for its occurrence. For instance, Hutton¹⁷ gave his view on the matter by stating that there is a limit to the amount of elastic shear strain energy that can be held in the shear field. If this limit is exceeded, a fraction of the elastic energy is converted into surface free energy, yielding a distorted extrudate. More recently, Han and Lamonte^{15,16} have supported this criterion of the limit in elastic shear strain, by measuring the elastic properties of polymer



Fig. 12. Pictures of extrudate samples of low-density polyethylene extruded through a die having the entrance angle of 120°: (a) at $\dot{\gamma} = 99.8 \text{ sec}^{-1}$; (b) $\dot{\gamma} = 294.1 \text{ sec}^{-1}$; (c) $\dot{\gamma} = 852.5 \text{ sec}^{-1}$.

melts in extrusion below and above a critical shear rate at which the inception of melt fracture occurs.

In the experimental study reported here, we have observed an *apparent* dependence of melt fracture on die entry angle. Figure 12 shows pictures of representative extrudate samples of low-density polyethylene extruded in the die having an entry angle of 120° . It is seen that at low flow rate, the extrudate is smooth, but as the flow rate is increased, extrudate starts to be distorted, and the severity of distortion increases as the flow rate is increased further. In our experimental study, there was no extrudate distortion observed for dies having entry angles smaller than 120° . However, even though there was no visible extrudate distortion for small entry-angle dies, fluctuations in wall pressure were noticed. As noted in earlier studies by Han and Lamonte,^{15,16} pressure flucutation at the die wall indicates a non-uniform flow in the die. It should be noted also that the pressure fluctuation becomes more severe as the melt approaches the entrance of the capillary section, and it decays as the melt travels through the straight section of the capillary.

On the other hand, when high-density polyethylene was extruded, no distorted extrudate was observed for all the die entry angles tested, though some fluctuation in wall pressure was observed. It can therefore be said that the extent of the defects associated with melt fracture depends not only on the die entry angle but also on the molecular structure of the material, which in turn would govern the critical value of elastic shear strain energy.

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