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Extrudate distortion studies of polystyrene using an extrusion rheometer

ANTHONY A. COLLYER, GEOFFREY H. FRANCE
Department of Applied Physics, Sheffield City Polytechnic, Sheffield, UK

Uncorrected and corrected logarithmic flow-curves for a general purpose polystyrene (MW = 261 000 and MW/MN = 4.4) obtained using a Davenport Extrusion Rheometer are shown for the range 160 to 200° C. The uncorrected flow curves show a change in slope, but at the lower extrusion temperatures this change occurs after the appearance of distorted extrudates. The onset of extrudate distortion obtained from observation does not coincide with the change in slope of the graph. The corrected logarithmic flow curves show no change in slope. Values of $\dot{\gamma}_c$ and η_c from both sets of graphs show that $\dot{\gamma}_c$ is inversely proportional to η_c, and for the higher melt temperatures the corrected τ_c values increase with temperature. The high value of critical wall stress at 160° C is attributed to the increase in melt elasticity with decreasing temperature being a greater effect than the decrease in elasticity due to a decrease in $\dot{\gamma}_c$.

1. Introduction

In the extrusion of polymer melts through dies there is a critical shear rate above which the surface of the extrudate becomes roughened, and further increases in shear rate will cause a severe distortion of the extrudate. This critical shear rate, $\dot{\gamma}_c$, represents the shear rate at the wall of the die. The corresponding shear stress at the wall and the apparent melt viscosity are denoted by τ_c and η_c respectively.

Earlier observers discovered that graphs of log τ against log $\dot{\gamma}$ often consisted of two straight lines, giving a change in slope of the flow curve as shown in Fig. 1. It was originally believed that the point of intersection of the lines was an indicator of the onset of extrudate distortion [1, 2]. However, Schott and Kaghan [3] and Metzner *et al.* [4] found no such changes of slope on their graphs.

Later Tordella [5] stated that, even though in some cases there may be no visible extrudate distortion corresponding to the conditions represented on the flow curves by a change in slope, his bi-refringence studies on high-density polyethylene and TFE–HFP co-polymer showed that unstable flow commenced at this shear rate.

Bagley [2] suggested that the large pressure drop at the entrance of the die may be regarded as an imaginary extension of the die and that plots of total pressure drop against length-to-diameter (L/D) ratio would be linear. The correction required to the L/D ratio of the die is found from the intercept on the (L/D) axis of such a graph, and the corrected value is used to calculate τ. Later observers [6–8] found that these Bagley plots are not straight when the apparent melt viscosity is pressure sensitive, but become straight when corrections for this pressure dependence are applied.

Den Otter [9, 10] used another method for correcting the shear stress. He calculated the shear stress, τ, from

$$\tau = \frac{\Delta p - \Delta p_0}{4(L/D)}, \qquad (1)$$

where Δp is the total pressure drop and Δp_0 is the entrance pressure.

Another refinement adopted by more recent observers was to correct the shear rate at the wall by use of the Rabinowitch correction for shear-thinning fluids. The corrected shear rate is given by

$$\dot{\gamma}_{\mathrm{CORR}} = \left(\frac{3n+1}{4n}\right)\dot{\gamma}, \qquad (2)$$

where n is the shear-thinning index.

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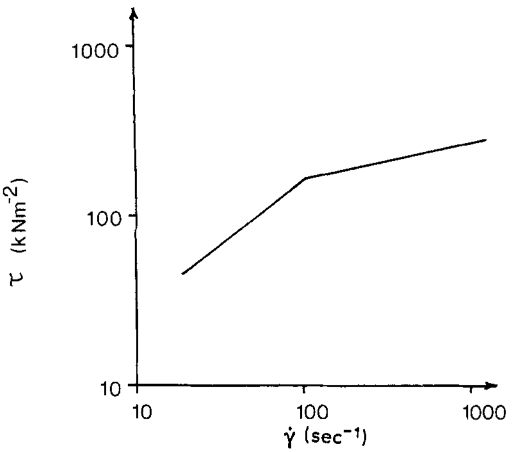

Figure 1 Plot of Log τ against log $\dot{\gamma}$ showing the discontinuity of the graph at the onset of extrudate distortion.

With these corrections Bartos [11], Clegg [12], Bialas and White [13] and Ramamurthy [14] found no changes of slope in their logarithmic flow-curves, and the last observer noted that the idea of predicting extrudate distortion from logarithmic flow-curves is highly suspect. Nakajima and Collins [8] made the former corrections and in addition allowed for pressure loss in the barrel and the pressure dependence of the melt viscosity. No change in slope occurs in their logarithmic flow-curves. It is generally accepted that the corrections above remove the change in slope in logarithmic flow-curves.

The present work is concerned with both corrected and uncorrected graphs for a general purpose polystyrene at the lower end of its extrusion range. Higher temperatures were also used to give a method of comparison with previous work. It was felt that despite the loss in favour on theoretical grounds of uncorrected logarithmic flow-curves, they may still be a useful and simply obtained indicator of extrudate distortion.

2. Experimental procedures

The present results were obtained on a Davenport Extrusion Rheometer, which consists of a vertical die connected to a heating barrel. The melt is extruded through the die by a piston, which moves at a pre-set rate. Temperature and pressure measurements are made at the entrance to the die.

For uncorrected shear stresses, the shear stress at the wall was calculated from the total pressure drop, Δp, across the die, by

$$\tau = \frac{\Delta p}{4(L/D)}. \qquad (3)$$

Corrected shear stresses were obtained using a short die of 2 mm diameter and 2 mm length and measuring the total pressure drop, $\Delta p'$, across it. The corrected value of τ for each shear rate was obtained from

$$\tau = \frac{(\Delta p - \Delta p')}{4(L'/D)}, \qquad (4)$$

where L' is the difference in length between the short die and the test die used in the experiments.

Any pressure fluctuations that occurred were averaged, but this proved necessary only at the higher shear rates, above the observed onset of extrudate distortion. No corrections were considered necessary for the variation of melt viscosity with hydrostatic pressure because the maximum hydrostatic pressures at the onset of extrudate distortion in the present work were considerably lower than those used on a similar material in the experiments of [7] and [8], even at the lowest melt temperature used.

The uncorrected shear rate at the wall was obtained from the piston velocity, S, by

$$\dot{\gamma} = \frac{S}{16.53\, r^3}, \qquad (5)$$

where S is in cm min^{-1} and the die radius, r, is in cm. Corrected values of shear rate were obtained from the Rabinowitch correction.

The experiments were carried out on a general purpose polystyrene (MW = 261 000 and MW/MN = 4.4) using a die of diameter 2 mm and length 19 mm. One set of results was obtained for a die of the same length but of half the diameter. In every case at least 20 readings were taken, and initially the material was heated for 15 min.

3. Results

The uncorrected logarithmic flow curves are shown in Fig. 2. In all cases a small, discernable change in slope is visible, from which an estimated onset of melt distortion can be made. A visual check on the extrudates was carried out, and thus two estimates of the onset were made for each temperature.

The changes in slopes of the graphs are indicated by the arrows in Fig. 2. These points lie on the straight line A and give values of τ_c, $\dot{\gamma}_c$ and η_c shown in Table I. This interpretation of the onset of extrudate distortion suggests that τ_c decreases markedly with increasing melt temperature, which is not in agreement with other observers. However,

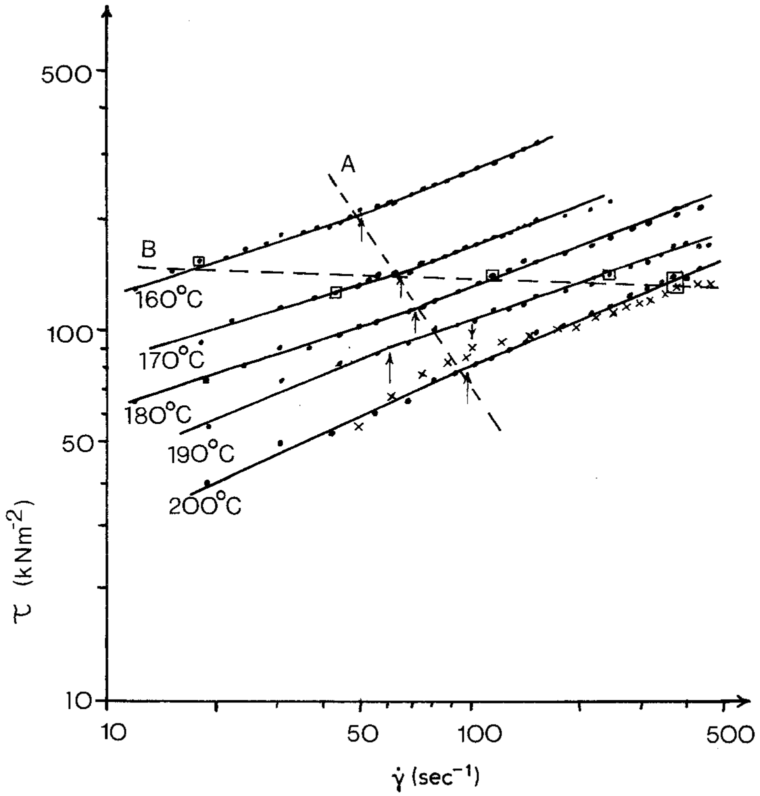

Figure 2 Logarithmic flow curves for polystyrene using a 2 mm diameter die. The crosses show results for a 1 mm diameter die of the same length for a melt temperature of 200° C.

$\dot\gamma_c$ is inversely proportional to η_c in agreement with [1, 15–18].

The square boxes on the graphs correspond to the visual occurrence of a distorted extrudate, which in this case was slightly wavy and typical of polystyrene. These points lie on the straight line B in Fig. 2. These points give a value of τ_c which is constant within experimental error. Values of τ_c, $\dot\gamma_c$ and η_c from line B are shown in Table I, and again $\dot\gamma_c$ is inversely proportional to η_c.

Values of τ_c obtained from a 1 mm diameter die (shown as crosses in Fig. 2) are similar to those for the larger die, but the change in slope of the graphs is more apparent.

The unusual aspect of the uncorrected graphs is that the onset of extrudate distortion appears before the change in slope of the graphs for melts at 160 and 170° C, the cross-over occurring between 170 and 180° C.

The graphs for 160, 170 and 180° C show a change to a greater slope as Westover [19] found in the extrusion of an ultra-high molecular weight high-density polyethylene through a die of small aspect ratio, $L/D = 5$. In the present work $L/D = 9.5$. At the higher temperatures the change in slope is of the usual kind, with a cross-over temperature for this of between 180 and 190° C. Far more accurate data is needed to determine whether the intersection of lines A and B does coincide with the temperature at which the change

TABLE I Values of critical shear stress, shear rate and viscosity for extrudate distortion from discontinuities and from observation of the extrudates for a 2 mm diameter die, and values of shear-thinning index

Temp. (° C)	From discontinuity			From observation						Shear-thinning index
				Uncorrected values			Corrected values			
	τ_c (kN m^{-2})	$\dot\gamma_c$ (sec^{-1})	η_c (N sec m^{-2})	τ_c (kN m^{-2})	$\dot\gamma_c$ (sec^{-1})	η_c (N sec m^{-2})	τ_c (kN m^{-2})	$\dot\gamma_c$ (sec^{-1})	η_c (N sec m^{-2})	n
160	210	52	4000	155	18	8610	100	44	2270	0.15
170	140	64	2270	130	43	3020	74	99	750	0.16
180	115	70	1570	145	115	1260	83	250	330	0.18
190	98	72	1360	145	240	600	89	490	180	0.21
200	88	78	810	140	360	400	88	610	140	0.26

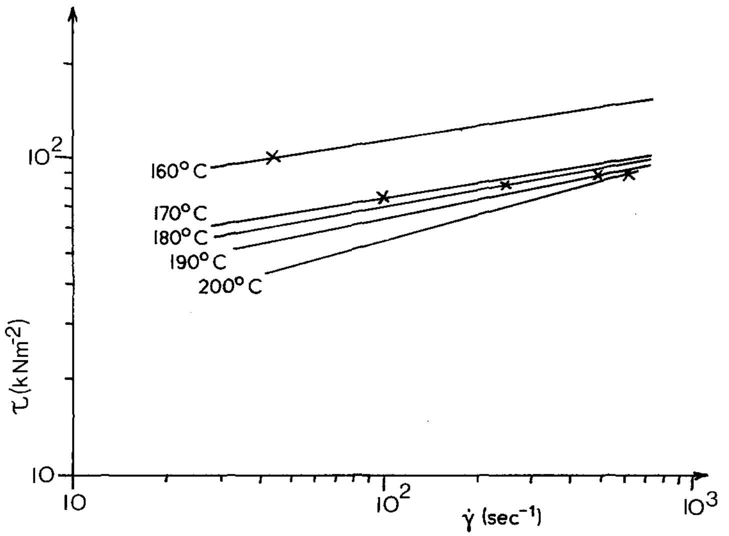

Figure 3 Corrected logarithmic flow curve for polystyrene using a 2 mm diameter die. The crosses mark the onset of extrudate distortion.

in slope (after the discontinuity) starts to be of the usual kind.

From the preceeding observations, it seems that predictions of extrudate distortion from the point at which a change in slope of the graph occurs are even less reliable at the lower extrusion temperatures, with extrudate distortion likely to occur before the change in slope in the graphs.

Values of the shear-thinning index, n, of the melts are shown in Table I. The index increases with the melt temperature, showing that the melts are less shear-thinning as their temperature increases. These values of n were used to calculate the Rabinowitch correction for shear rate.

Fig. 3 shows corrected logarithmic flow-curves, and as noted by many observers no change in slope is apparent. The onset of extrudate distortion is marked by a cross on each graph. The best straight line is obtained through the crosses of the four highest temperatures, giving values of τ_c that increase slightly with temperature in agreement with previous observers. The values of $\dot{\gamma}_c$ and η_c shown in Table I are inversely proportional, with the point for the lowest melt temperature slightly offline.

4. Discussion

The results show that uncorrected graphs are not reliable in predicting extrudate distortion from the change in slope, particularly at the lower temperatures, when the distortion occurs before the change in slope.

Correction to shear stress and shear rate appear to have a larger effect on the results at 160° C than on those for the higher temperatures. The corrected values indicate an increase in τ_c with temperature in agreement with previous authors, but the critical shear stress for the 160° C melt is higher than the others.

A possible explanation of this enhancement of τ_c at the lowest temperature depends on the way in which the melt elasticity, and hence τ_W, varies with shear rate and temperature. Melt elasiticity increases with increasing shear rate and decreases with increasing melt temperature. As is apparent from Table I, the greater changes in $\dot{\gamma}_c$ occur at the higher temperatures, and it would be expected that increase in melt elasticity due to the increase in shear rate would be greater than the decrease in melt elasticity due to an increase in melt temperature. This gives an overall increase in τ_c with melt temperature. At the lower melt temperatures the variation of $\dot{\gamma}_c$ is much smaller, and it is suggested that in this case the increase in melt elasticity due to the reduction in temperature is now the greater effect, giving an increase in τ_c at the lower melt temperatures.

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