

# Effects of Die Temperature on Extrudate Swell in Screw Extrusion

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## Synopsis

Extrusion of a hot polymer melt through a cooler die zone substantially increases the extrudate swell of some thermoplastics. This effect was examined for commercial samples of low-density polyethylene, polypropylene, and polystyrene. Two conflicting effects come into play during extrusion of a thermoplastic. Colder melt temperatures promote increased extrudate swell, but the same conditions also facilitate molecular disentanglement and reduced melt elasticity and die swell. Since the extrusion process itself may affect the relation between die swell and melt temperature, laboratory-scale measurements for the design of processes like blow molding are better carried out with small-scale screw extruders than with capillary rheometers. For some applications it may be advantageous to use a polymer whose die swell is particularly responsive or unresponsive to die temperature variations. The procedure described in this article can be used effectively to monitor this characteristic.

## INTRODUCTION

Extrudate swell, or die swell as it is more commonly known, is the property of expansion of viscoelastic fluids on extrusion from a constriction.<sup>1-5</sup> There is general agreement that extrudate swell results from relaxation of elastic stresses in the fluid.<sup>6</sup> Extrudate swell is dependent on shear rate<sup>7</sup> (or more universally on shear stress<sup>6</sup>), polymer molecular weight distribution,<sup>5,7</sup> the length-to-diameter ( $L/D$ ) ratio of the die,<sup>3,8</sup> and the difference in diameters of the extrusion reservoir and orifice.<sup>9-11</sup>

The swelling of polymer melts must be taken into account in the design of extrusion and blow molding processes. Wall thickness of blow-molded products is significantly affected by extrudate swell. Adjustment of process conditions to produce a particular wall thickness is difficult because neither extrudate swell nor the polymer flow in the annular parison die can be analyzed in a completely quantitative manner. To increase wall thickness of blow-molded objects, the annular parison die can be opened up. This increases flow rate and parison weight.<sup>12</sup> Another simpler route to achieve this purpose would be just to increase extrudate swell. Colder melt temperatures or higher shear rates are used to increase extrudate swell.<sup>13</sup> This, however, requires more extrusion work and therefore greater energy input.

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It is interesting also that forcing a hot melt through a cooler die zone substantially increases extrudate swell of some thermoplastics. This may be significant in blow molding because it indicates that it could be possible to control such parameters as product wall thickness by the die temperature alone. The effect that extruding a hot melt through a cool die zone has on extrudate swell was examined here in detail for commercial samples of polystyrene, low-density polyethylene, and polypropylene. Extrudate swell was correlated to die temperature and shear rate in the die, in this work.

### EXPERIMENTAL

Extrusions were carried out with a 0.5-in. diameter extruder, designed in our laboratory, with a 27:1 length-to-diameter ratio barrel. Drive was provided with a 0.5-hp shunt-wound motor and a 30:1 worm gear reducer. The extruder barrel had three temperature-controlled zones, and temperatures of the interchangeable head and die were also independently controlled. A pressure transducer was mounted in the die head. The screw had a 4:1 compression ratio. Die face temperatures were monitored with a thermocouple clamped to the surface of the die.

The initial melt temperature was monitored by inserting a thermocouple into the die opening as the melt was extruding. If the band heater controlling the die was disconnected for any length of time, the melt extrudate swell was observed to increase as the die cooled. The phenomenon of increasing extrudate swell with decreasing melt temperature is well known<sup>13,14</sup> but the experimental conditions employed here differ from those usually employed. Instead of utilizing a melt rheometer and varying its temperature, an extruder was used with constant temperature settings in its four heating zones from feed to head. Only the die was cooled.

The experiment consisted of obtaining constant flow conditions in the extruder, then unplugging the die heater and monitoring the die temperature drop and melt pressure. Extrudate swell was measured on samples of extrudate collected at different temperatures.

After a period, the die temperature would remain relatively constant as the rate of heat absorption from the flowing melt nearly equaled the heat losses. At this point the final melt temperature was measured by inserting the thermocouple into the die orifice as the melt was extruding. The die temperature decay was monitored throughout the experiment by clamping the thermocouple to the die face close to the die orifice. Melt temperatures corresponding to intermediate die temperatures were obtained by interpolating between the initial and final melt temperatures by assuming a direct correspondence between die temperature decay and melt temperature decrease.

The polystyrene and polyethylene samples were extruded with the four heating zones set at 140°C and the polypropylene with corresponding settings of 170°C. The thermoplastics were extruded through a series of circular cross-section dies with different length-to-diameter ratios. The dies were either 1 or 2 in. long with 0.5-in. diameter channels that abruptly closed into 1/8-in. or 1/4 in. diameter capillaries. The length of the narrow capillaries determined the  $L/D$  ratio. These dies are referred to as flat entry

orifices. The melt pressure, temperature, and mass extruded in a given time were measured at different extruder screw speeds. Flow curves of apparent shear stress against apparent shear rate were plotted.

The shear stress at the capillary wall  $\tau_w$ , is given by

$$\tau_w = \frac{\Delta P}{2(L/R + n)} \quad (1)$$

where  $\Delta P$  is the pressure drop,  $L/R$  is the length-radius ratio of the die capillary, and  $n$  is a correction factor for pressure drop in the die entrance, called the Bagley end correction.<sup>15</sup> The Bagley correction is obtained by plotting pressure drop  $\Delta P$  against  $L/R$  for several flow rates and extrapolating to a constant intercept at zero pressure. The shear rate at the wall ( $\dot{\gamma}_w$ ) is given by

$$\dot{\gamma}_w = \left( \frac{3n' + 1}{4n'} \right) \frac{4Q}{\pi R^3} \quad (2)$$

where

$$n' = \frac{d[\log(R\Delta P/2L)]}{d[\log(4Q/\pi R^3)]} \quad (3)$$

where  $Q$  is the volumetric flow rate. This is the Mooney-Rabinowitsch equation,<sup>16,17</sup> which corrects for flow with nonparabolic or nonideal velocity profiles.

Extrudates were sampled by using a razor knife to cut the melt at the die exit cleanly. After a suitable time, the sample was snipped at the die exit with a pair of tweezers. The samples were held vertically in air so that they were straight when they hardened. It was important to make sure the initial cut was quick and clean and gave a circular sample end cross-section. This allowed extrudate swell measurements to be made near this end where drawdown effects were minimal. Some polypropylene samples had elliptical cross sections. In these cases, the diameter was taken as the average of the maximum and minimum size readings.

Residence time ( $T_r$ ) calculations:

$$T_r = \frac{\text{die volume}}{\text{melt volumetric flow rate}} = \frac{V}{Q} \quad (4)$$

required knowledge of the melt specific volumes. The specific volumes used were as follows: polystyrene, 1.02 cm<sup>3</sup>/g at 186°C;<sup>18</sup> polyethylene, 1.295 cm<sup>3</sup>/g at 170–190°C;<sup>19</sup> and polypropylene was taken as 1.31–1.32 cm<sup>3</sup>/g at 190–200°C.<sup>19</sup>

Extrudate swell, or diameter percentage increase, was calculated from

$$\text{Diameter \% increase} = \left( \frac{\text{melt diameter} - \text{capillary diameter}}{\text{capillary diameter}} \right) \times 100 \quad (5)$$

To obtain a better qualitative picture of the variation in extrudate swell with melt temperature and shear rate, three-dimensional perspective block diagrams were plotted using the Surface II graphics system<sup>20</sup> and an IBM-370 computer. This system also outputs the slope of the surface at the grid nodes, which gives an estimate of the rate of change of extrudate swell in a particular region.

Molecular weight distributions of the polymers were measured by size exclusion chromatography in trichlorobenzene solutions using a low-angle laser light scattering detector for molecular weights and a differential refractive index detector to measure corresponding concentrations. Methods used to prepare aggregate-free solutions of the polyolefins have been described elsewhere.<sup>21-24</sup> Universal calibration was based on a hydrodynamic volume method.<sup>25,26</sup>

## RESULTS

Molecular weights of the polymers used are recorded in Table I.

Bagley end-correction<sup>15</sup> plots for the three polymers studied are shown in Figures 1 through 3. The pressure drop- $L/D$  plots are given here at fixed screw speed instead of the more usual representation in terms of apparent shear rates in the die. The two forms are equivalent. End-correction plots were linear for these polymers under our experimental conditions.

True flow curves for the three polymers were obtained after application of the Bagley and Mooney-Rabinowitsch corrections, as described above. These plots are shown in Figures 4 through 6. The polyethylene and polystyrene viscosities coincide at low shear rates, but the former is evidently more shear thinning in the particular experimental temperature range.

Typical extrudate swell-screw speed raw data are recorded in Figure 7 for the polystyrene sample. Extrudate swell is not a single-valued function of die  $L/D$  ratio at a given screw speed and temperature. It depends also on the ratio of the die and barrel diameters, with the smaller die producing the greater extrudate swell.<sup>9-11</sup> As expected, die swell increases with screw speed, in the absence of melt fracture effects.

The polyethylene and polypropylene samples show much less variation of die swell with temperature and with screw speed, as shown, for example, in Figure 8.

Three-dimensional perspective diagrams allow easy assessment of the effects of screw speed on extrudate swell. Figure 9 depicts plots of polystyrene extrudate swell against screw speed and melt temperature for the two die geometries of Figure 7.

A qualitative difference between the melt behavior of dies with different diameters is immediately apparent. The block plot for the larger (0.25 in.) diameter (Fig. 9b) has a wedge shape with a face of approximately constant slope; the smaller diameter die (0.125 in.) plot of Figure 9a exhibits a plateau region in low screw speed regions followed by a sharp rise at higher rpm values. These trends are general for this polystyrene with dies of different  $L/D$  ratios. The performance of this melt is evidently more difficult to predict and control in extrusions where the orifice diameter differs greatly from that of the diameter of reservoir from which the polymer issues.

TABLE I  
Polymer Molecular Weights

Sample	$M_n$	$M_w$	$M_z$	$M_{z+1}$	SD (N) <sup>a</sup>	SD (W)	SKEW (N) <sup>b</sup>	SKEW (W)
Polyethylene	32,700	173,200	860,000	1,350,000	68,000	345,000	17	7
Polystyrene	58,000	186,000	612,000	986,000	86,000	282,000	13	8
Polypropylene	37,000	178,000	715,000	1,250,000	72,000	309,000	15	8

<sup>a</sup> SD = standard deviation of the number  $N$  or weight distribution.<sup>27</sup>

<sup>b</sup> SKEW = skewness of the number  $N$  or weight distribution.<sup>27</sup>

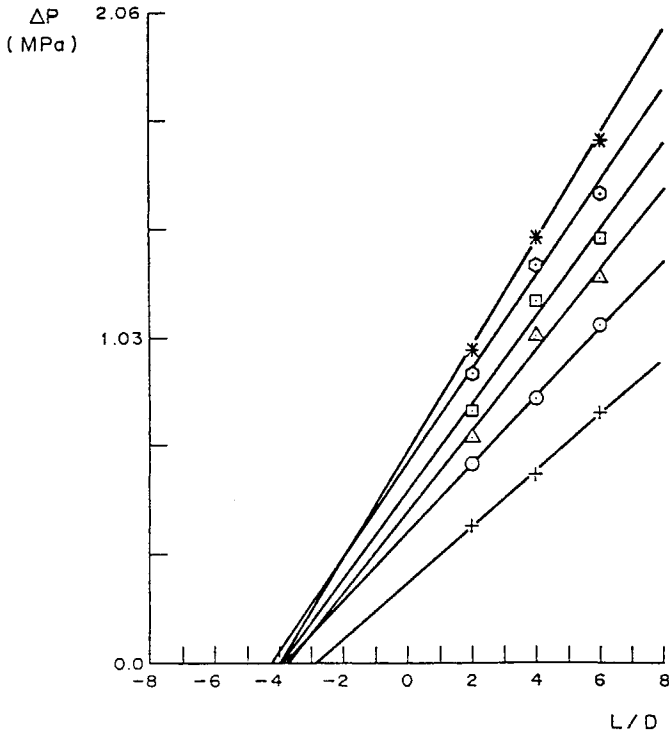


Fig. 1. Bagley<sup>15</sup> plot for polystyrene at 178–180°C melt temperature; 0.125-in. diameter dies: (+) 10 rpm, (○) 20 rpm, (△) 30 rpm, (□) 40 rpm, (◇) 50 rpm, (\*) 60 rpm screw speed.

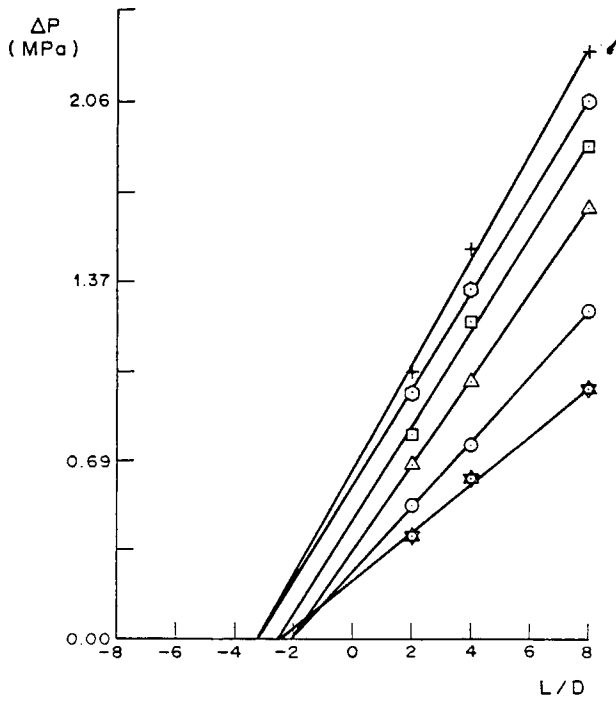


Fig. 2. Bagley<sup>15</sup> plot for polyethylene at 175–179°C melt temperature; 0.125-in. diameter dies: (☆) 10 rpm, (○) 20 rpm, (△) 30 rpm, (□) 40 rpm, (◇) 50 rpm, (+) 60 rpm screw speed.

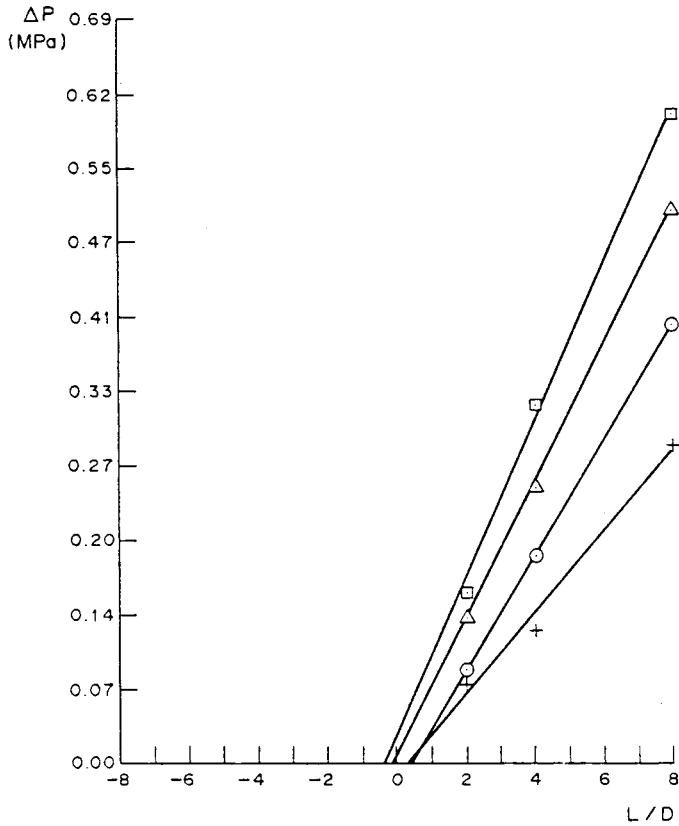


Fig. 3. Bagley<sup>15</sup> plot for polypropylene at 190–205°C melt temperature; 0.125-in. diameter dies: (+) 10 rpm, (○) 30 rpm, (△) 50 rpm, (□) 60 rpm screw speed.

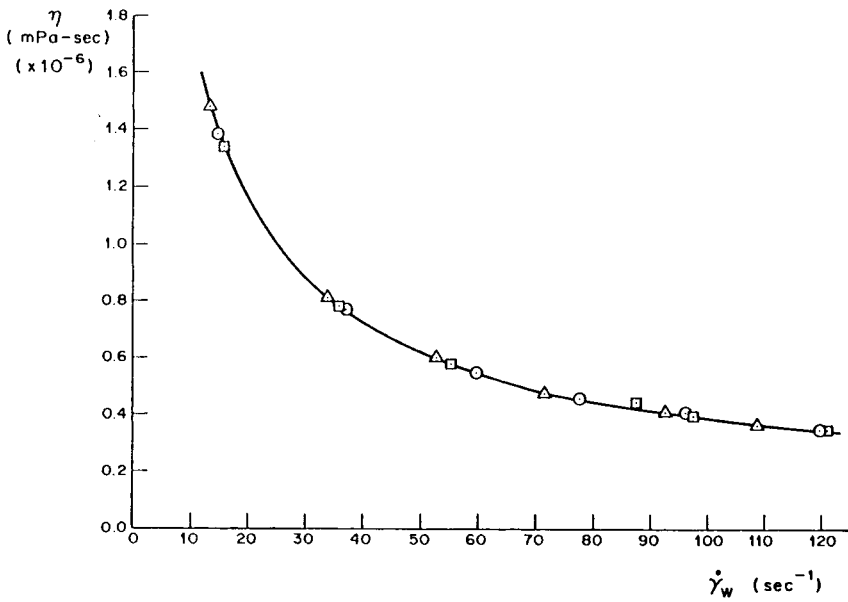


Fig. 4. True flow curve (viscosity versus wall shear rate) for polystyrene polymer at 178–180°C. (○) Die  $L/D = 2$ ; (△) Die  $L/D = 4$ ; (□)  $L/D = 8$ .

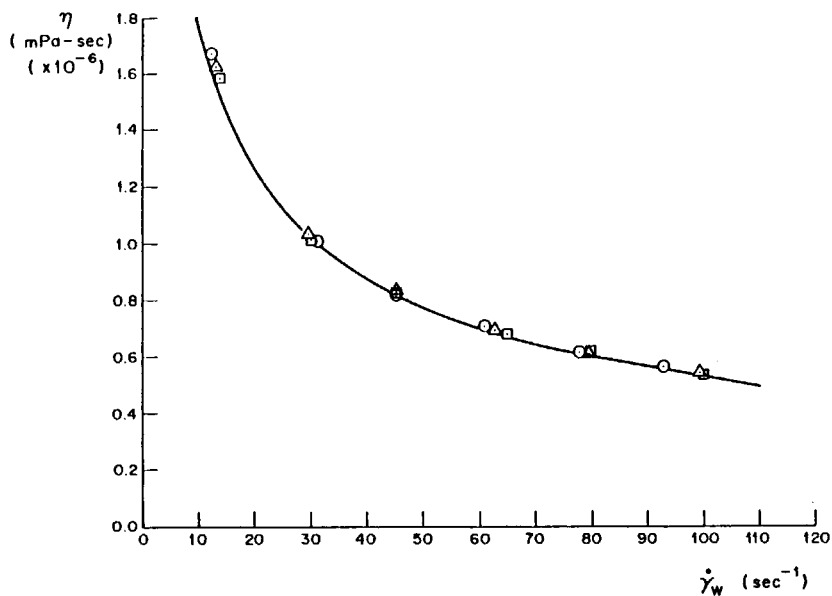


Fig. 5. True flow curve (viscosity versus wall shear rate) for polyethylene polymer at 175-179°C. (○)  $L/D = 2$ ; ( $\Delta$ )  $L/D = 4$ ; ( $\square$ )  $L/D = 8$ .

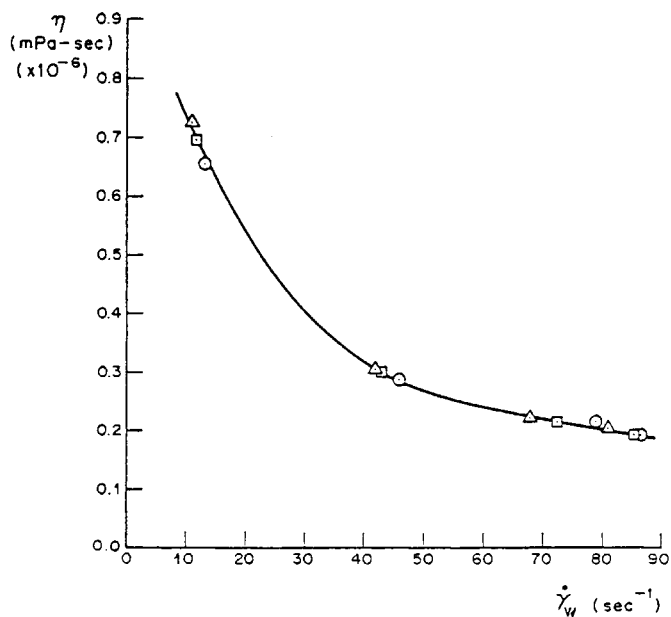
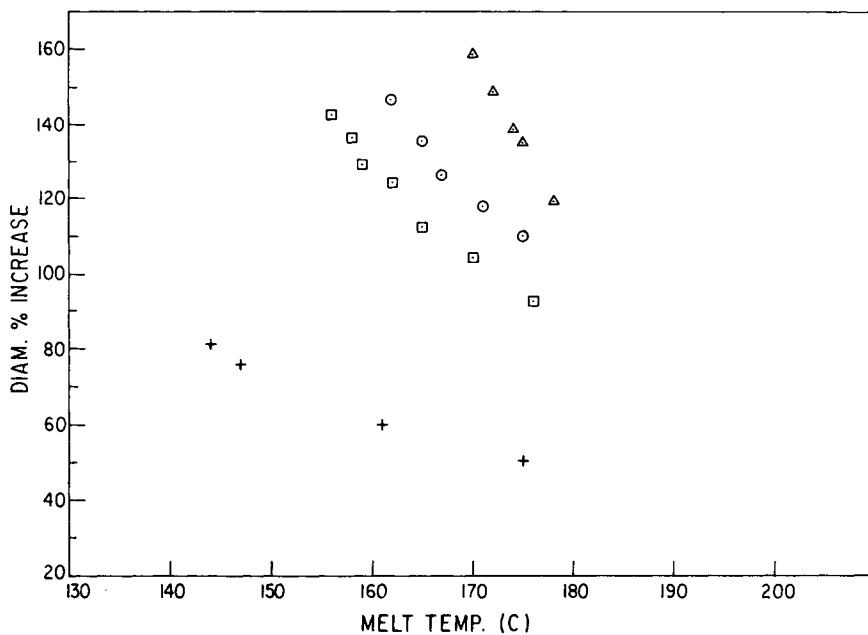
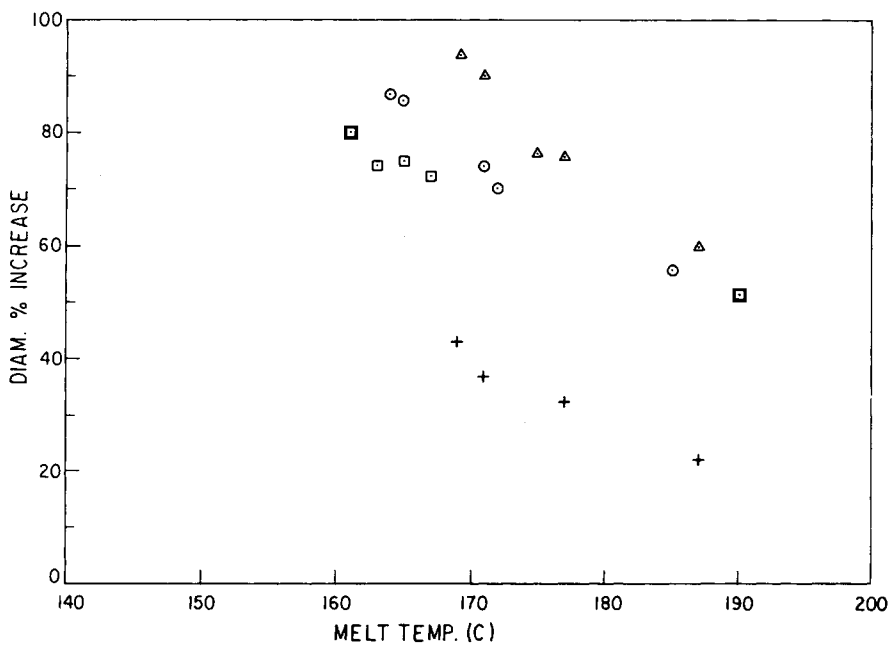


Fig. 6. True flow curve (viscosity versus wall shear rate) for polypropylene sample at 190-205°C. (○) Die  $L/D = 2$ ; ( $\Delta$ )  $L/D = 4$ ; ( $\square$ )  $L/D = 8$ .



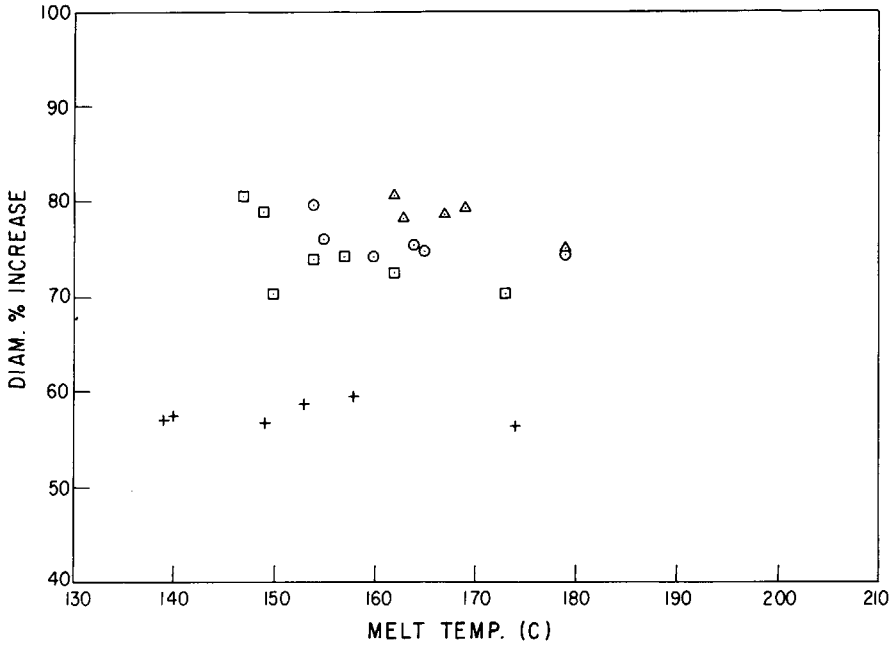


(a)

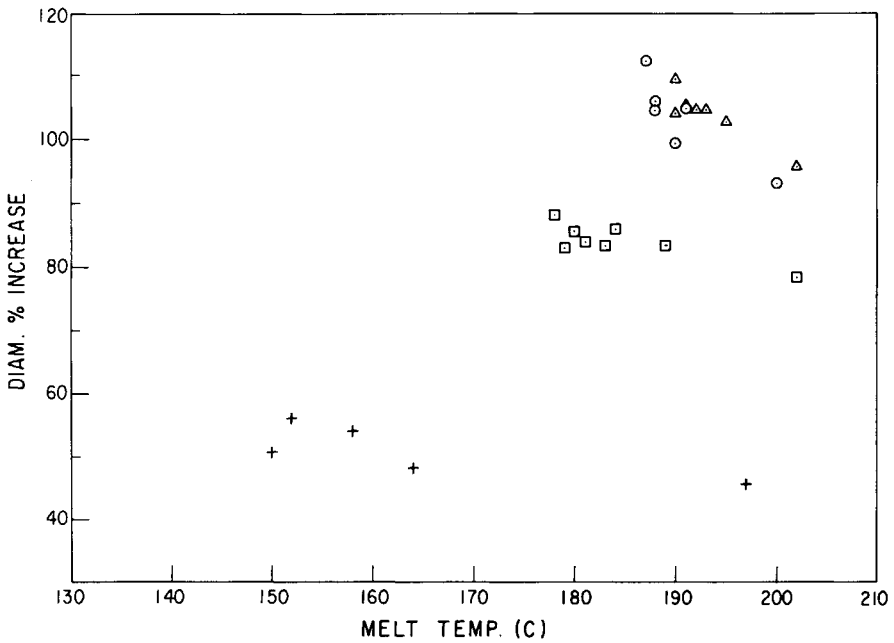


(b)

Fig. 7. Variation of polystyrene extrudate swell with melt temperature. (a) Die  $L = 0.5$  in.,  $D = 0.125$  in.,  $L/D = 4$  (b) Die  $L = 1.0$  in.,  $D = 0.25$  in.,  $L/D = 4$ . (+) 10 rpm screw speed, (△) 30 rpm, (○) 50 rpm, (△) 60 rpm.

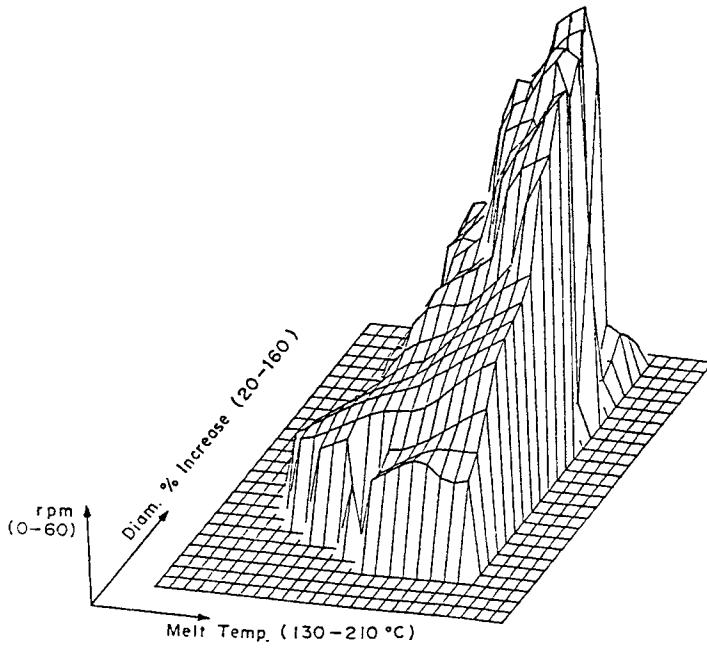


(a)

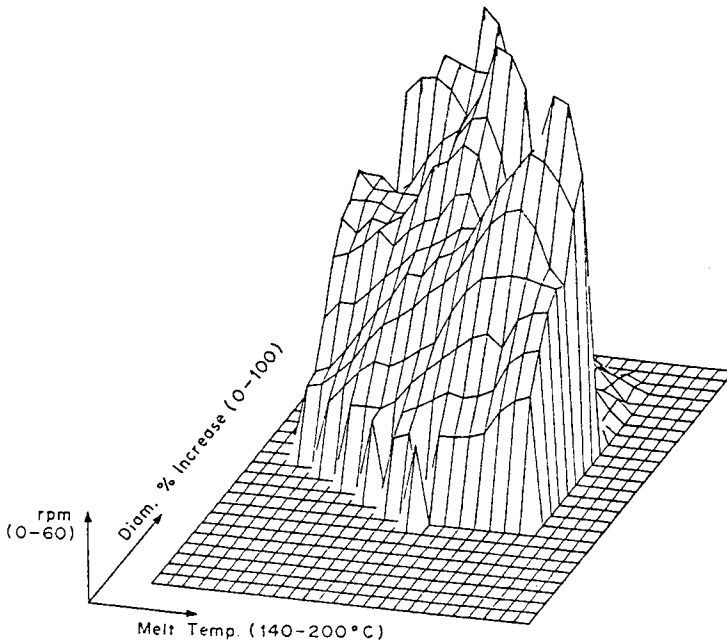


(b)

Fig. 8. (a) Variation of polyethylene extrudate swell with melt temperature. Die  $L = 0.5$  in.,  $D = 0.125$  in.,  $L/D = 4$ . (+) 10 rpm, (□) 30 rpm, (○) 50 rpm, (△) 60 rpm. (b) Variation of polypropylene extrudate swell with melt temperature. Die  $L = 0.25$  in.,  $D = 0.125$  in.,  $L/D = 2$ . Symbols as in Figure 2a.



(a)



(b)

Fig. 9. (a) Block diagram of polystyrene sample extrudate swell with melt temperature and screw speed. (a) Die  $L = 0.5$  in.,  $D = 0.125$  in.,  $L/D = 4$ ; (b) Die  $L = 1.0$  in.,  $D = 0.25$  in.,  $L/D = 4$ .

## DISCUSSION

The die swells of the polyethylene and polypropylene samples responded less strongly to temperature variations than the polystyrene polymer. This behavior probably reflects the results of two conflicting effects. There is a general tendency for increased extrudate swell at colder melt temperatures, as noted. On the other hand, extrusion at colder temperatures results in increased molecular disentanglement and reduced melt elasticity and die swell. The latter phenomena have been described for linear low-density polyethylene,<sup>28</sup> high-pressure process polyethylene,<sup>29</sup> and plasticized poly(vinyl chloride).<sup>30</sup>

Polystyrene responds less to shear history variations, in general. The reason for this behavior is not entirely clear. The melt is evidently quite elastic, judging from the extrudate swell values listed above. At normal processing temperatures, however, the polystyrene chain is much less flexible than the polyolefins examined here. Intramolecular entanglements in polystyrene are therefore less susceptible to shear disentanglement.

Since the extrusion process itself may apparently affect the relation between die swell and melt temperature laboratory scale measurements for design of processes like blow molding are better carried out with small-scale extruders than with capillary rheometers in which the shear and thermal history of the polymer may differ greatly from that in full-scale operations.

The actual extrudate swell that is observed in a given operation will depend on the shear rate in the die, the melt temperature in the die, the shear history of the polymer, and the length-to-diameter ratio of the die, as well as its absolute diameter and the geometry of the lead-in from the extruder barrel to the die. These various factors will not all be equally significant, and their relative importance will vary with different polymers, as shown. This work has demonstrated a convenient system for evaluating these variables. It has also shown that die swell may be controlled to some extent by varying the die temperature independent of the rest of the extruder control settings.

One further comment is needed on the general effect of melt temperature on extrudate swell. As noted, die swell decreases with increased melt temperature. Rubber elasticity theory and experience indicate that the modulus of a cross-linked elastomer increases at higher temperatures. A simple picture of an element of an entangled polymer melt envisages the material subjected to a given stress level as it enters the die from an extruder operating at fixed screw speed and melt temperature, as in our experiments. If the die itself is colder, the melt may be cooled during its passage. The modulus of the polymer that issues from the machine is therefore lower, and its recoverable shear strain and die swell are correspondingly greater. As mentioned above, this effect is counteracted to some extent by the effect of shearing on polymer melts. In general, this causes a reduction in melt elasticity and die swell and is more marked for polyolefins than for polystyrene.<sup>31</sup> Possibly for this reason, the polyethylene and polypropylene melts studied are less elastic than the polystyrene initially (Figs. 7 and 8), and their response to temperature changes is correspondingly lower. This

behavior is not necessarily typical of all polyolefins. Variations in molecular weight distribution would be expected to affect the temperature dependence of melt elasticity.

For some applications, it may be advantageous to use a polymer whose die swell is particularly responsive or unresponsive to die temperature variations. The procedure described here can be used effectively to monitor and control this property.

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