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A Study of Coextrusion in a Circular Die

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Synopsis

An experimental study has been carried out of concentric and eccentric two-phase flow of polymer melts in a circular die. For the study, a special die was constructed such that two separate streams of molten polymer could be supplied to the die inlet. Materials used for study were low-density polyethylene, high-density polyethylene, and polystyrene. In the experiment, two different capillary length-to-diameter (L/D) ratios were employed: 4 and 18. For a die having an L/D ratio of 18, wall normal stresses were measured, permitting the determination of the pressure gradient and hence the viscous property. For each set of extrusion conditions (L/D ratio, flow rate, component ratio), extrudate samples were collected. These were later carefully cross sectioned and photographed in order to examine the shape of the interface between the two components. It has been found that the lower-viscosity components tends to wrap around the higher-viscosity component, which is consistent with the previous observations reported in the literature.

INTRODUCTION

In recent years, rheologists have become increasingly interested[1–6] in research leading to an understanding of the various complicated flow phenomena which occur when two viscoelastic fluids flow side by side through an extrusion die. Of particular interest in this connection is the shape of the interface. As shown in an earlier study by Han[3] and more recently by Lee and White,[6] the interfacial configuration in an extrudate is influenced by the shape of the die cross section, the manner in which the two fluids are introduced at the die entrance, the component ratio of the two fluids, the length-to-diameter ratio of a capillary, and perhaps, most significantly, the viscoelastic properties of the individual fluids concerned.

Notwithstanding the quite widespread use of coextrusion techniques in the plastics industry (e.g., multilayered film) and in the fiber industry (e.g., bicomponent fiber), there remain many as yet unanswered questions which need to be studied at a more fundamental level.

In the present paper, the author presents some new experimental results concerning the shape of the interface as affected by the manner in which two fluids meet at the die inlet.

EXPERIMENTAL

Materials chosen for the experimental study were: general-purpose polystyrene (Dow Chemical Co., Styron 686), low-density polyethylene (Union Carbide Corp., DYNF 1), and high-density polyethylene (Union Carbide Corp., DMDJ 4309). Two sets of bicomponent systems were used for the coextrusion experiment, namely, polystyrene/low-density polyethylene (PS/LDPE) and polystyrene/high-density polyethylene (PS/HDPE).

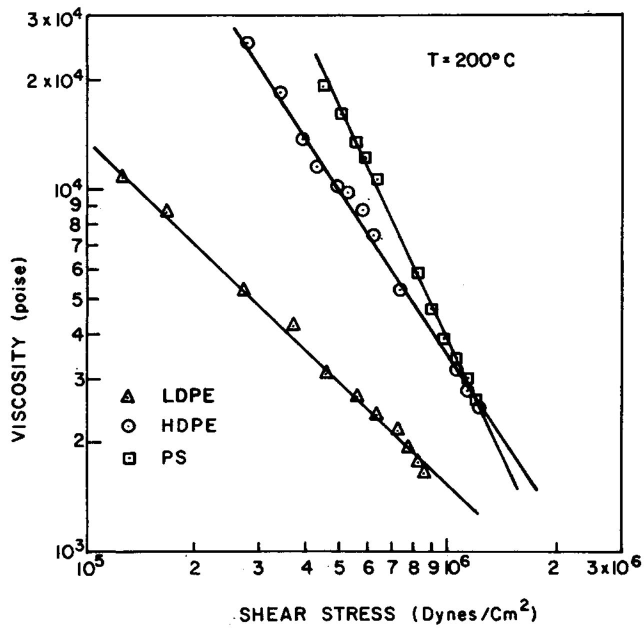

Fig. 1. Viscosity vs. shear stress for the polymer melts investigated.

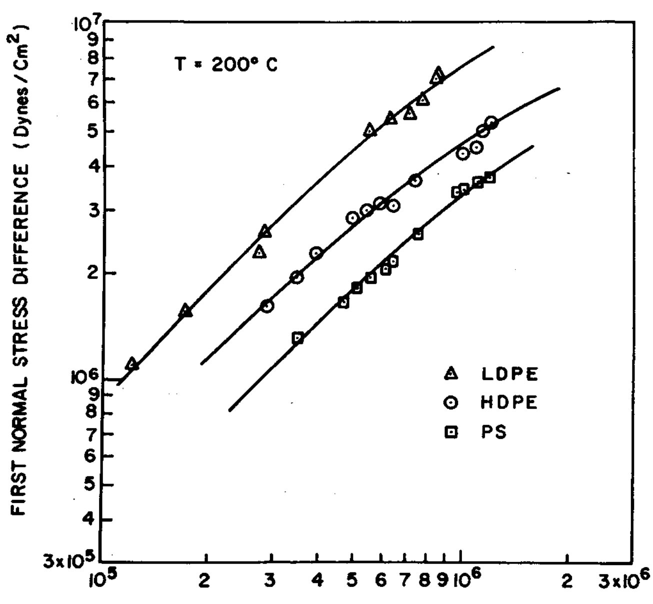

Fig. 2. First normal stress difference vs. shear stress for the polymer melts investigated.

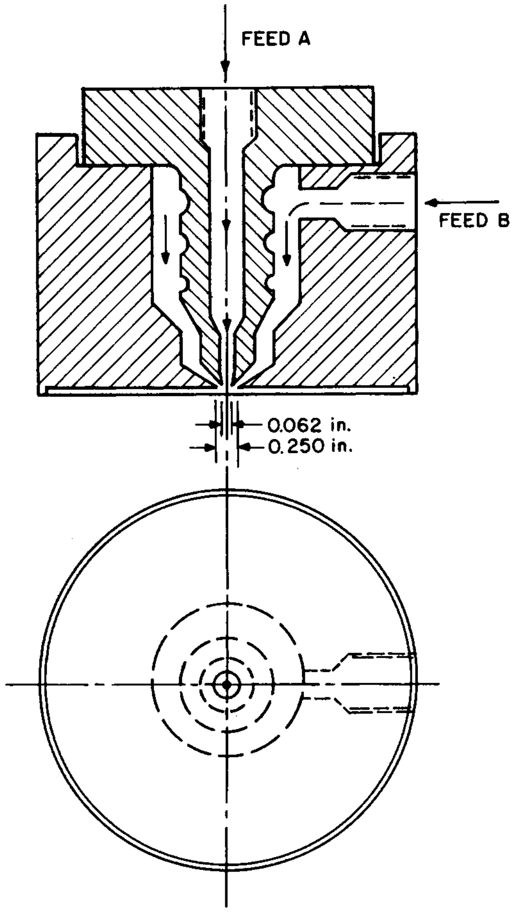

Fig. 3. Detailed layout of the die assembly.

The viscoelastic properties of the individual components in the molten state had been determined in connection with a previous study[7] and are given in Figures 1 and 2.

The apparatus consists of two 1-in. Killion extruders, a feeding section which combines the two melt streams, and a die section. Figure 3 shows a schematic of the feeding section, describing how the two molten polymers meet at the die inlet.

Insofar as the feeding of two polymers is concerned, two different experiments were carried out: one used concentric flow and the other, eccentric flow. For the experiment using eccentric flow, the center portion of the feed section (i.e., stream A) was adjusted to be at about 0.025 in. off the center.

In the coextrusion experiment, two capillary dies were used having L/D ratios of 4 and 18, the capillary diameter being 0.25 in. Details of die design are the same as those given in an earlier paper by Han.[3] Different L/D ratios of capillary dies were employed in order to observe the effect of L/D ratio on the shape of the interface. Measurements of wall normal stress were also taken on the die having an L/D ratio of 18.

For each flow rate, extrudate samples were collected. Later, these samples were cross sectioned and photographed in order to examine the shape of the interface.

RESULTS AND DISCUSSION

Pictures of extrudate cross section showing the shape of the interface are given in Figure 4 for the polystyrene/high-density polyethylene (PS/HDPE) and in Figure 5 for the polystyrene/low-density polyethylene (PS/LDPE). Note in these pictures that the two melts were fed concentrically to the die inlet. It is seen that when the polystyrene was fed inside the other component, the interfacial shape was little affected whether the die was short ($L/D = 4$) or long ($L/D = 18$). However, when the polystyrene was fed outside the other component, the long die ($L/D = 18$) tended to yield a slightly eccentric interface, particu-

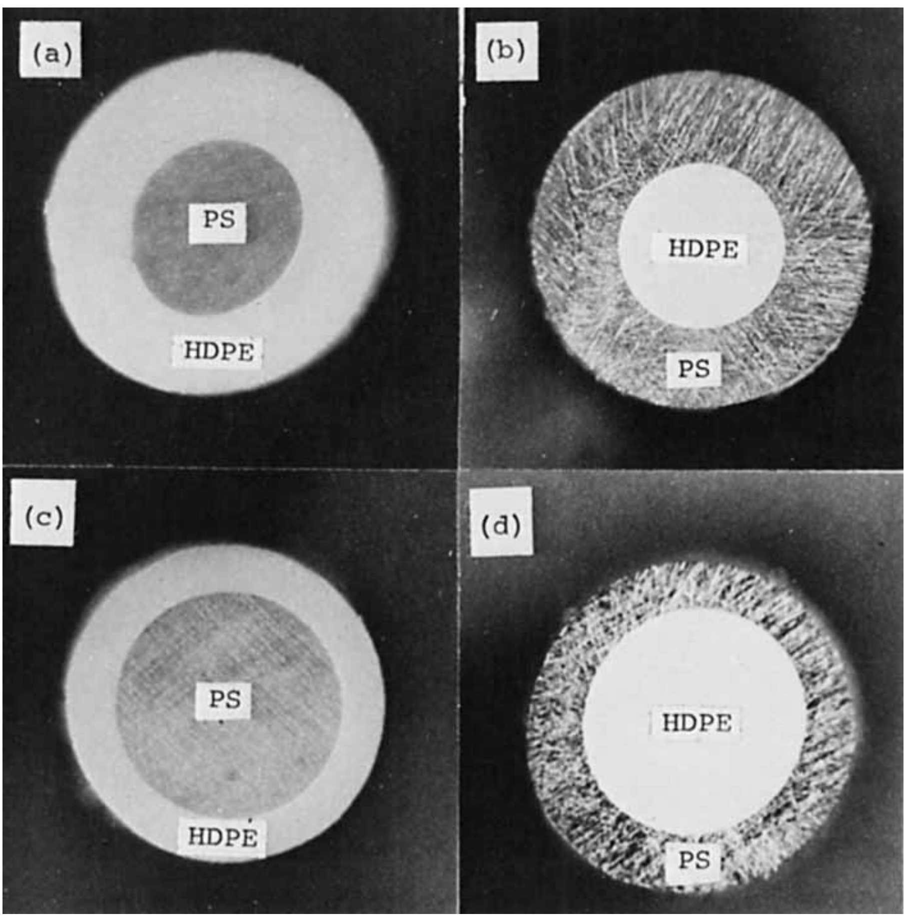

Fig. 4. Interfacial shape of the PS/HDPE two-phase system extruded concentrically: (a) PS/HDPE with $L/D = 4$ and $Q = 89.9$ cc/min; (b) HDPE/PS with $L/D = 4$ and $Q = 75.8$ cc/min; (c) PS/HDPE with $L/D = 18$ and $Q = 68.5$ cc/min; (d) HDPE/PS with $L/D = 18$ and $Q = 85.0$ cc/min.

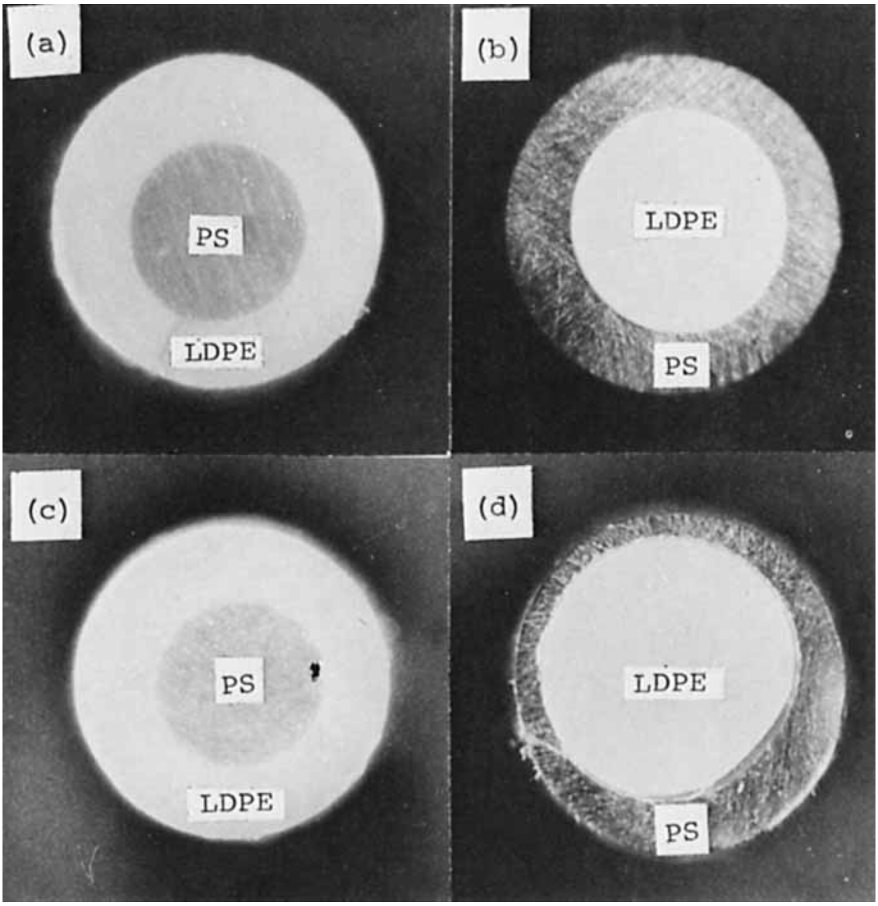

Fig. 5. Interfacial shape of the PS/LDPE two-phase system extruded concentrically: (a) PS/LDPE with $L/D = 4$ and $Q = 53.6$ cc/min; (b) LDPE/PS with $L/D = 4$ and $Q = 27.9$ cc/min; (c) PS/LDPE with $L/D = 18$ and $Q = 78.3$ cc/min; (d) LDPE/PS with $L/D = 18$ and $Q = 108$ cc/min.

larly for the LDPE/PS system. The difference between these instances may be attributable to the fact that the less viscous component (HDPE in the HDPE/PS system and LDPE in the LDPE/PS system) tends to wrap around the more viscous component.

Perhaps a more dramatic observation which may support the theory set forth above can be seen in Figure 6 for the HDPE/PS system and in Figure 7 for the LDPE/PS system. Note in these pictures that the two melts were fed eccentrically to the die inlet. It is seen that, when the polystyrene (more viscous) was fed outside the other component (less viscous), the shape of the interface in the extrudate was completely different from that at the die inlet.

Let us try now to offer a quantitative interpretation of the experimentally observed interface shape in terms of the rheological properties of bicomponent systems investigated (see Figs. 1 and 2). Over the range of extrusion conditions tested (i.e., apparent shear rate of 90 to 600 $\sec^{-1}$ and wall shear stress of 0.3×10^6 to 0.9×10^6 dynes/cm^2), we have the following relationships:

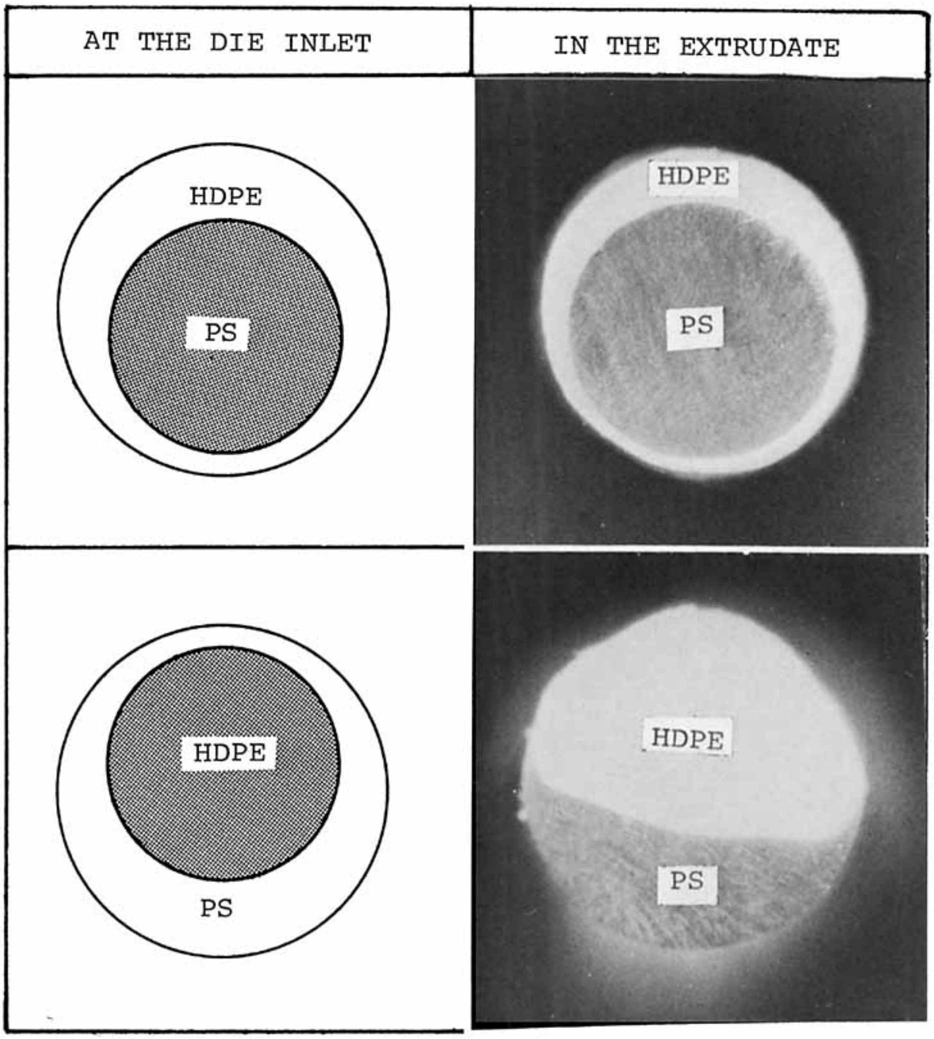

Fig. 6. Interfacial shape of the PS/HDPE two-phase system extruded eccentrically in the capillary having an $L/D = 18$. For PS/HDPE, $Q = 55.1$ cc/min; for HDPE/PS, $Q = 78.4$ cc/min.

For the LDPE/PS system,

$$\eta_{LDPE}/\eta_{PS} = 0.14 \text{ to } 0.34$$

$$(\tau_{11} - \tau_{22})_{LDPE}/(\tau_{11} - \tau_{22})_{PS} = 2.53 \text{ to } 2.37$$

$$(\tau_{22} - \tau_{33})_{LDPE}/(\tau_{22} - \tau_{33})_{PS} = 1.61 \text{ to } 1.78$$

For HDPE/PS system,

$$\eta_{HDPE}/\eta_{PS} = 0.52 \text{ to } 0.89$$

$$(\tau_{11} - \tau_{22})_{HDPE}/(\tau_{11} - \tau_{22})_{PS} = 1.59 \text{ to } 1.39$$

$$(\tau_{22} - \tau_{33})_{HDPE}/(\tau_{22} - \tau_{33})_{PS} = 1.17 \text{ to } 1.19$$

Note that the second normal stress difference data are taken from an earlier publication[7] and that the component which is less viscous (LDPE in the LDPE/

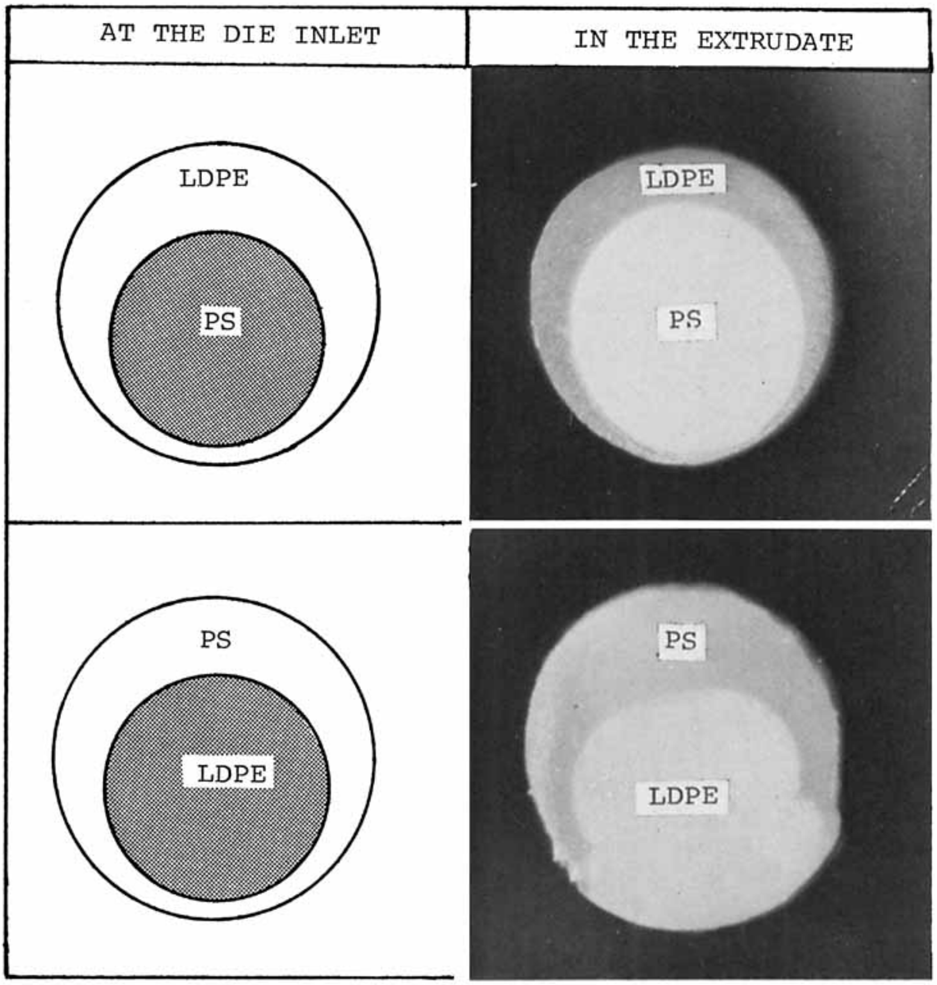

Fig. 7. Interfacial shape of the PS/LDPE two-phase system extruded eccentrically in the capillary having L/D = 18. For PS/LDPE, Q = 41.1 cc/min; for LDPE/PS, Q = 112 cc/min.

PS system and HDPE in the HDPE/PS system) is more elastic than the other component.

It appears that the interface shape is strongly influenced more by a difference in the viscosity of two components than by a difference in the elasticity. For instance, it is seen in Figure 7 that when the low-density polyethylene (less viscous) was fed outside the polystyrene (more viscous), there is hardly seen any change in the interface shape. This may be attributable to the fact that in the fully developed region, it is the viscosity, and not the elasticity, which governs the pressure drop across the capillary. On the other hand, when the low-density polyethylene was fed inside the polystyrene, the less viscous component (i.e., LDPE) tends to wrap around the more viscous component, regardless of the substantial difference in the elasticity of the two components.

Wall normal stresses were measured along the axis of the capillary, and the pressure gradients were determined. Plots of pressure gradient versus volumetric flow rate are given in Figure 8 for the HDPE/PS system, and in Figure 9

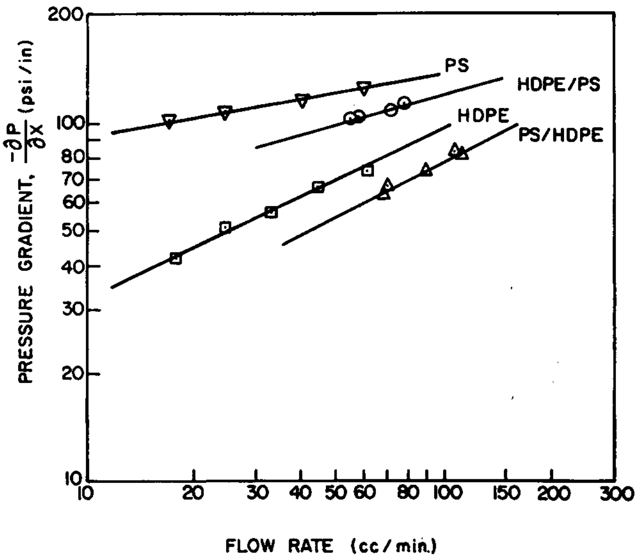

Fig. 8. Pressure gradient vs. volumetric flow rate for the PS/HDPE two-phase system in the die having $L/D = 18$.

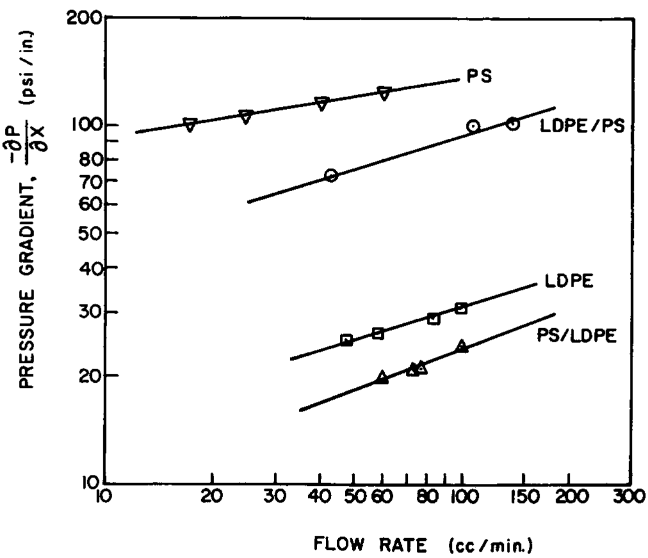

Fig. 9. Pressure gradient vs. volumetric flow rate for the PS/LDPE two-phase system in the die having $L/D = 18$.

for the LDPE/PS system. Note in Figure 8, for instance, that HDPE/PS denotes that the high-density polyethylene (HDPE) was fed inside the polystyrene (PS), and PS/HDPE denotes that polystyrene (PS) was fed inside the high-density polyethylene (HDPE). It is interesting to note that, when the more viscous component (i.e., PS) flows outside the less viscous component (i.e., LDPE or HDPE), the pressure gradients of the two-phase system are somewhere between the pressure gradients of the two individual components. However, when the more viscous component (i.e., PS) flows inside the less viscous component (i.e., HDPE or LDPE), the pressure gradients of the two-phase system are lower than that of the less viscous component. This observation has also been reported in earlier publications by Han.[2,3]

To summarize, an experimental study has been carried out to investigate the shape of the interface by varying the manner in which two molten polymer streams meet at the die inlet. The results reported in this paper give a new insight into a better understanding of a fundamental aspect of the coextrusion operation.

The study reveals that considerable reduction in pressure drops may occur in coextruding two materials when the less viscous component wraps around the more viscous component. This finding is highly significant in the extrusion of very viscous polymers, where the exceedingly high-pressure drop in a die is a bottleneck to production.

In view of the fact that careful control of the shape of the interface is very important to the success of the coextrusion process (e.g., bicomponent fiber), the results presented above provide some guidance to a better design for coextrusion dies (e.g., L/D ratio) and to a choice of optimal processing conditions (e.g., temperature and flow rate) in coextrusion.

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