

A Study of Bicomponent Coextrusion of Molten Polymers

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Synopsis

An experimental study has been carried out of coextruding polystyrene with low-density polyethylene and polystyrene with high-density polyethylene, using both slit and circular dies. Two melt streams were separately fed to the die entrance and forced to flow side by side through a die. When using the slit die, wall normal stresses were measured with three melt pressure transducers flush-mounted on each of the rectangle's long sides, directly opposite each other. When using the capillary die, three different capillary length-to-diameter (L/D) ratios were employed: 4, 11, and 18. Wall normal stresses were measured for dies having L/D ratios of 11 and 18 only. The measurements of wall normal stresses permitted one to determine the pressure gradient, and hence the viscous property. For each set of extrusion conditions (L/D ratio, flow rate, and 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. At the phase interface of the polystyrene/low-density polyethylene system, it has been observed that the polystyrene, which is more viscous and yet less elastic than the low-density polyethylene, has a convex surface. However, at the phase interface of the polystyrene/high-density polyethylene system, the high-density polyethylene, which is more viscous and also more elastic than polystyrene, is seen to be convex. This then appears to indicate that the viscosity ratio of two components is primarily responsible for the final shape of the interface.

INTRODUCTION

In recent years, coextrusion in either a flat film die or a circular die has emerged into the polymer processing industry as an attractive means of economically producing bilayered and multilayered, composite films, by combining two or more different melt streams. The success of a coextrusion process depends very much on the die design, which should provide ways of combining different melt streams and of controlling the thickness of the layers of an extrudate. There are a number of patents that describe die designs for coextrusion. A few recent publications^{1,2} summarize many of the patents and discuss various technical problems involved in the coextrusion process.

In addition to the economics involved, the coextrusion process offers the advantage of producing very thin films, less than 0.3 mil thick, say, which otherwise would be very difficult to achieve by conventional lamination and coating. Since in coextrusion two or more melt streams flow side by side through a die, such flow may be defined as stratified flow.

Besides the practical importance, the multiphase stratified flow of polymeric melts through a conduit is of theoretical importance as well. One of the fundamental questions which arises, for instance, from bicomponent stratified flow through a thin slit die is the shape of the interface between the two components. In the flow of polymeric melts which are viscoelastic, a theoretical analysis of the problem can become quite complicated due to melts having elastic properties in addition to viscous properties. Recently, White et al.³ made a theoretical attempt at predicting the shape of the interface, using some simplifying assumptions. A recent study by Yu and Han⁴ indeed shows that the interface is not flat when polystyrene and polypropylene melts are coextruded through a thin slit die. They also found that polystyrene, which is more viscous, tends to be convex at the interface and that polypropylene, which is less viscous, tends to preferentially wet the wall of the short side of the rectangular slit cross section.

Circular dies have also been employed in the production of bicomponent (or conjugate) fibers.⁵⁻⁸ An advantage of coextruding two components in a side-by-side semicircular configuration lies in that the technique can produce unique fiber properties which resemble natural wools, the so-called crimped fibers or wool fibers. The crimping characteristics result from the different thermal expansion coefficients of individual components, leading to the buckling of the filament while, upon exiting from the spinnerette, it is either being cooled or coagulated along the length of the spinline.

It may be surmised that the distribution of the two components and their interfacial configuration in the molten state within a spinnerette hole is of paramount importance in ultimately controlling the amplitudes and frequencies of crimps in the solidified fiber. Despite the technological importance of this subject, however, there seems to have been relatively little work reported in the literature on the fundamental nature of the flow characteristics of two viscoelastic polymer melts in circular dies.

A number of theoretical studies have been reported on the stratified flow of two immiscible Newtonian fluids in a circular tube. Gemmel and Epstein⁹ used a numerical technique to solve the equations of motion, and Bentwitch¹⁰ and Yu and Sparrow¹¹ transformed the original system partial differential equations into a new set of equations in the complex plane.

These authors assumed that the interface between the components is smooth and ripple free, and that the effects of preferential wetting of the duct wall are negligible. It should be noted, however, that the theoretical analysis of polymer melt flow is much more complicated than that of Newtonian flow. It can indeed be said that previous attempts made at theoretically analyzing Newtonian fluids do not seem directly applicable to viscoelastic fluids.

Very recently, Southern and Ballman¹² made an interesting observation of the interfacial shape when two commercial polystyrenes were extruded through a circular hole (0.050-in. diameter and 3-in. length). Although these authors made an attempt to correlate the interfacial shape with the

rhological properties (viscosity and elasticity) of the individual components in the molten state, their attempt has a few shortcomings due to insufficient data. First, there was no quantitative measurement taken of the flow properties of the bicomponent system while it was being extruded. Therefore, it was not possible to evaluate, for instance, how much the pressure drop across the tube for a more viscous component is affected by the presence of a less viscous component. Second, the differences in die swell ratio between the two components were too small to be considered as an effective measure of the difference in their melt elasticities. That is, the die swell ratios of the individual components differed from each other only by 1-3%, which could be well within the measurement error.

In the present paper, we shall discuss the results of our recent experiments on bicomponent coextrusion through a slit die and circular dies. For the study, two sets of two-phase systems were used, polystyrene/low-density polyethylene and polystyrene/high-density polyethylene. The primary objectives of this study were (a) to investigate the effects of the viscous and elastic properties of individual components on flow behavior in the bicomponent coextrusion process, and (b) to investigate the shape of the interface in the extrudate as affected by both the viscous and elastic properties of individual components.

EXPERIMENTAL

Apparatus and Experimental Procedure

The apparatus used for the present study is essentially the same as that described in a previous paper by the author.⁴ Basically, the apparatus consists of three parts, a feeding system which combines two melt streams, a die section, and a panel board for pressure measurement. In order to supply two separate streams of molten polymer, an extruder and a melt pump were used. The two melt streams, one from the extruder and the other from a hydraulic storage tank, were combined at a point very close to the inlet of a die which is divided by a knife-edged flow divider. Figure 1 shows how the two molten polymers meet at the die inlet.

In the coextrusion experiment with the slit die, three melt pressure transducers were mounted, opposite to each other, flush with the die wall on each side along the longitudinal axis. For given flow rates in both phases,

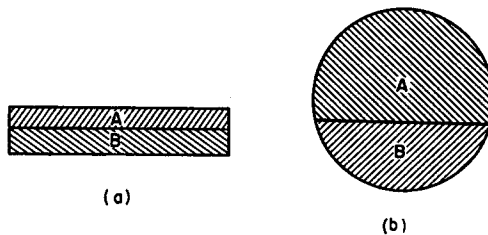


Fig. 1. Schematic which shows how two molten polymers meet at the die inlet.

TABLE I
Dimensions of Capillary Dies and Locations of Pressure Tap Holes

Die	D , in.	L , in.	L/D	Locations of pressure tap holes, in.		
				P_1	P_2	P_3
A	0.250	1.000	4	—	—	—
B	0.250	2.750	11	2.370	2.497	2.545
C	0.250	4.500	18	4.015	4.120	4.255

wall normal stresses were measured by means of the melt pressure transducers. The outputs of the transducers were read in millivolts by a potentiometer and null detector. The readings were then converted into pressure (psig) with the aid of calibration curves, which had previously been determined using a dead weight tester. The entire die section was electrically heated by use of Calrod heaters and heavily insulated with asbestos. The temperature was controlled within $\pm 1^\circ\text{F}$ by Thermistor-operated thermal regulators.

In the coextrusion experiment with circular dies, three capillary dies were used, having L/D ratios of 4, 11, and 18, the capillary diameter being 0.25 in. Details of die design are essentially the same as those given in earlier papers by Han et al.^{13,14} (See Table I.) 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 dies having L/D ratios of 11 and 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.

Materials

Two sets of bicomponent polymer systems were used for the coextrusion experiment, namely, polystyrene/low-density polyethylene (PS/LDPE) and polystyrene/high-density polyethylene (PS/HDPE). Note that polystyrene is incompatible with either of the polyethylenes. In other words, when combining melt streams of polystyrene with either low-density polyethylene or high-density polyethylene there is no adhesion, and thus identification of each component in the extrudate samples is very easy. This helps one to observe the interface between the two components.

The viscoelastic properties of the individual components in the molten state are given in Figures 2 and 3, which had been determined in connection with a previous study.¹⁵

RESULTS AND DISCUSSION

Wall Pressure Measurements

Figure 4 shows representative plots of the axial distribution of wall pressures of the polystyrene/high-density polyethylene two-phase system in the

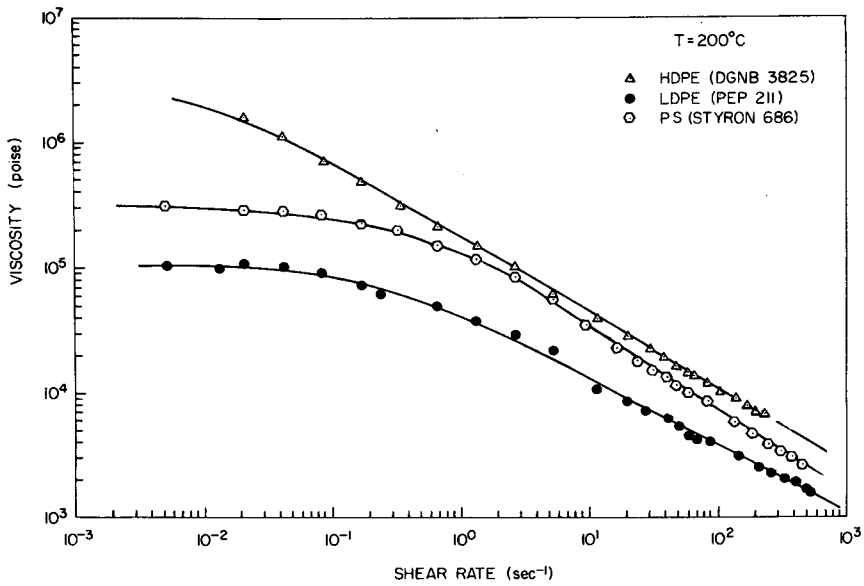


Fig. 2. Viscosity vs. shear rate for three polymer melts investigated.

slit die, with total volumetric flow rate as a parameter. Similar plots were prepared for the polystyrene/low-density polyethylene two-phase system, but the limitation of space does not permit us to present those plots here. It is seen in Figure 4 that for a given flow rate, the axial pressure gradient is a straight line over the distance in which measurements were taken. The pressure gradient may be plotted against volumetric flow rate, as

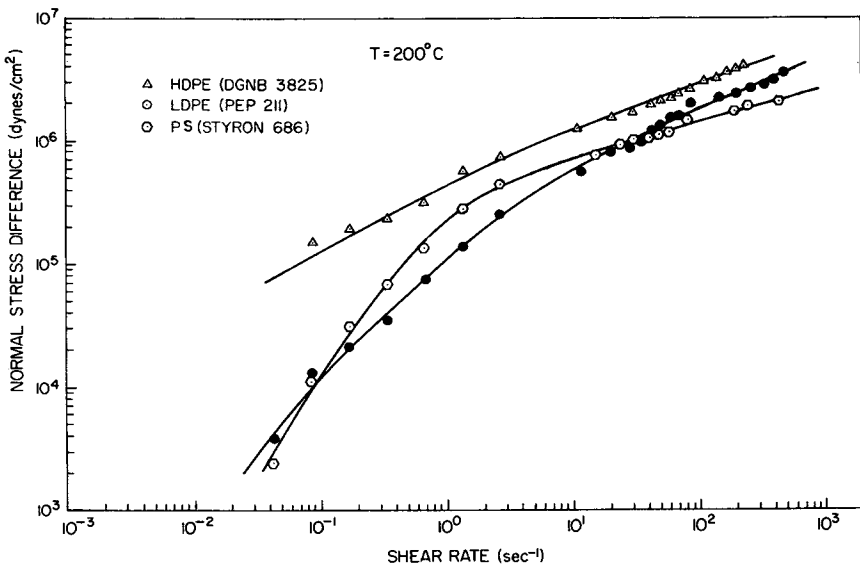


Fig. 3. Normal stress difference vs. shear rate for polymer melts investigated.

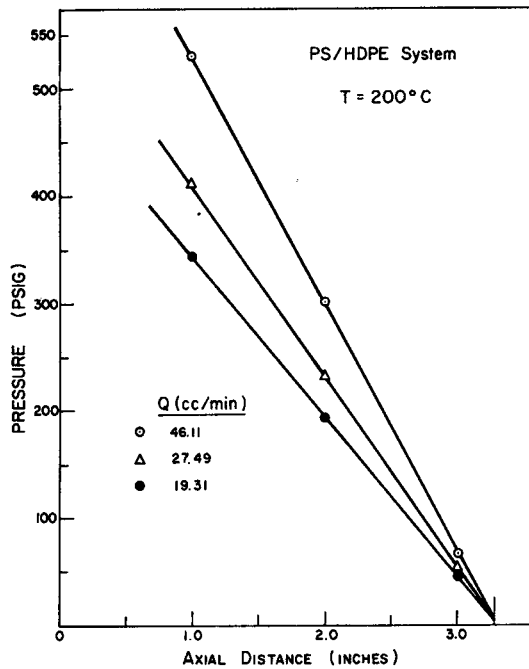


Fig. 4. Representative pressure profiles of PS/HDPE two-phase system in the slit die.

given in Figure 5. Also given in Figure 5 are plots for the pure components, polystyrene, low-density polyethylene, and high-density polyethylene. Note that the plots in Figure 5 may be considered as equivalent to flow curves.

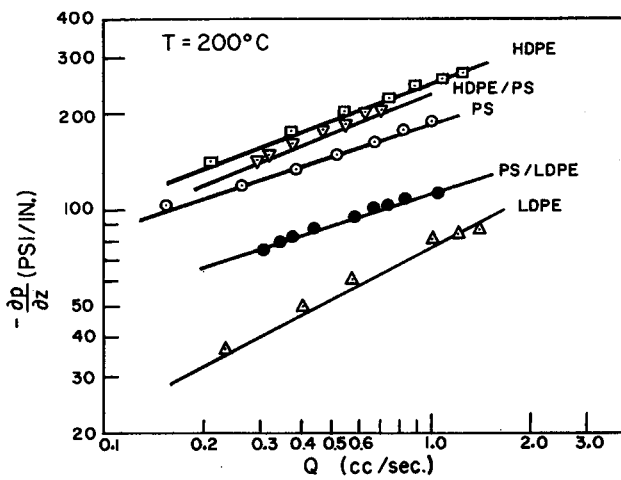


Fig. 5. Pressure gradient vs. volumetric flow rate in the slit die.

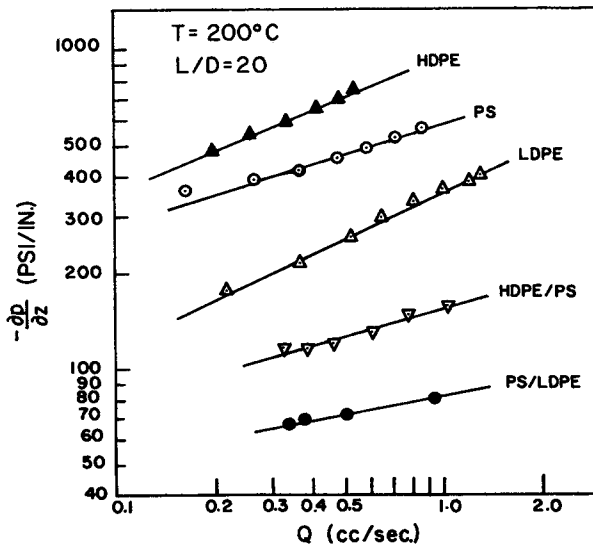


Fig. 6. Pressure gradient vs. volumetric flow rate in the circular die ($L/D = 18$).

Figure 6 gives plots of pressure gradient ($-\partial p/\partial z$) versus flow rate for the two-phase systems investigated in the circular dies.

Reduction in Pressure Gradient

Three things are worth noting from the plots of Figure 5. First, the pressure gradient increases with total volumetric flow rate. Second, regardless of the differences in the component ratio for a given two-phase system (either PS/LDPE or PS/HDPE), plots of $-\partial p/\partial z$ versus Q follow a single line. Third, the presence of the less viscous component brings a reduction in the pressure gradient of the more viscous component. It is seen in Figure 5 that plots of $-\partial p/\partial z$ versus Q of two-phase systems lie between those of individual components.

In order to observe the relationship between the fraction of the less viscous component and the reduction in the pressure gradient of a two-phase system, it is convenient to define a quantity called the pressure gradient reduction factor (P.G.R.F.):

$$\text{P.G.R.F.} = \left(-\frac{\partial p}{\partial z} \right)_{A/B} / \left(-\frac{\partial p}{\partial z} \right)_A$$

in which the subscript A denotes the more viscous component, and B the less viscous component. Plots of P.G.R.F. versus vol-% of the less viscous component are given in Figures 7 and 8 for the two-phase systems investigated in the slit die and circular dies, respectively. It should be noted that polystyrene is more viscous than low-density polyethylene, but less viscous than high-density polyethylene over the range of the variables investigated (i.e., temperature and shear rate), as given in Figure 2.

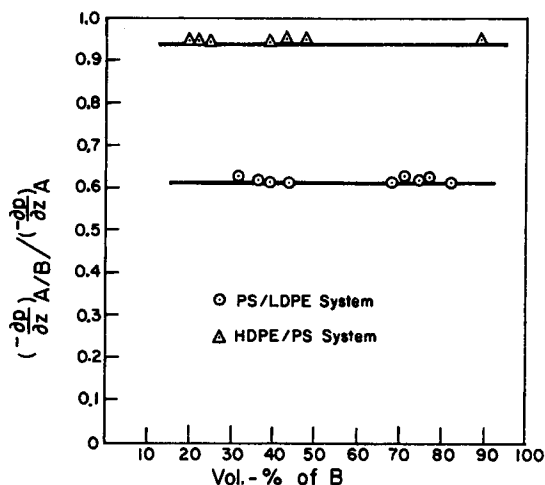


Fig. 7. Pressure gradient reduction factor vs. vol-% of the less viscous component in the slit die.

The reduction in pressure gradient has a practical significance for polymer processing. For instance, an addition of lubricants or low molecular weight can significantly reduce the extrusion pressure of very viscous polymeric melts. The application of this concept has been long known in the polymer processing industry, but little quantitative discussion of it has been given in the literature. The reduction in the pressure gradient is attributable to the tendency for the less viscous material to preferentially wet the tube wall. A similar observation has been reported earlier by a group of investigators^{9,16,17} who were concerned with the transportation of heavy crude oil in pipelines. If water was added, the water, being less viscous, was found to preferentially wet the wall, giving rise to an increase in the

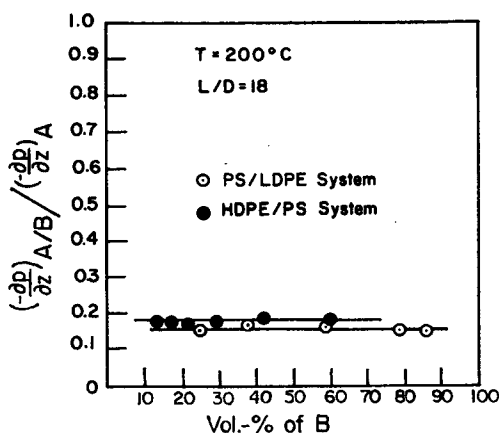


Fig. 8. Pressure gradient reduction factor vs. vol-% of the less viscous component in the circular die ($L/D = 18$).

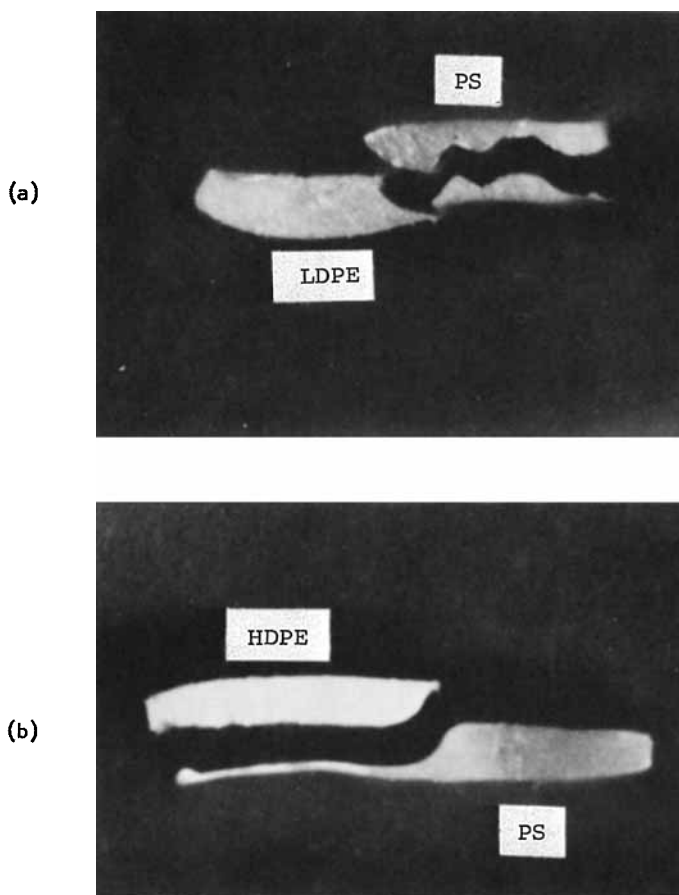


Fig. 9. Interfacial shape of the two-phase systems extruded through the slit die: (a) PS/LDPE systems; (b) HDPE/PS system.

pumping rate as great as tenfold. This concept may be applicable to such polymer processing as wire coating.

In the section to follow, we shall discuss the final distribution of two components present in the extrudate

The Shape of the Interface

In the present study, an interesting observation was made of the shape of the interface. Some representative results are given in Figure 9 for the PS/LDPE and HDPS/PS systems investigated using a slit die. Note that these pictures were taken of the extrudate samples collected during the extrusion experiment and later cross sectioned. Note that the extrudate samples initially forming a single piece have split apart into separate pieces when completely frozen.

Two things are worth mentioning about the shape of interface shown in Figure 9. First, despite the fact that the two polymers were initially fed

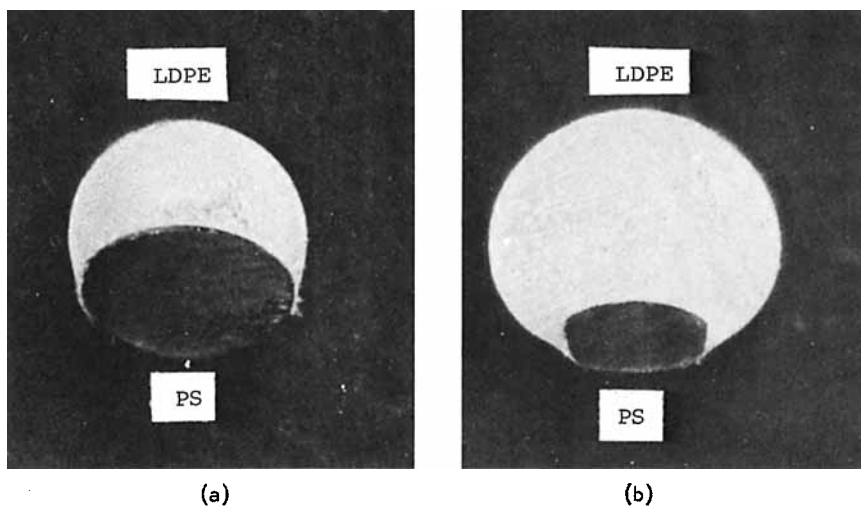


Fig. 10. Interfacial shape of the PS/LDPE two-phase system extruded through the circular die having an $L/D = 4$: (a) $Q_{PS} = 10.9$ cc/min, $Q_{LDPE} = 9.9$ cc/min; (b) $Q_{PS} = 8.3$ cc/min, $Q_{LDPE} = 53.8$ cc/min.

to a die inlet side by side, parallel to the long side of the rectangle of the slit die (see Fig. 1a), the pictures in Figure 9 show that the less viscous component tends to preferentially wet the wall, giving rise to an interfacial shape far from flat. Second, the interface shows some severe corrugation. Since the less viscous component tends to preferentially wet the wall in both two-phase systems (i.e., LDPE in the PS/LDPE system and PS in the HDPE/PS system), it appears that the viscosity ratio of the individual components is a most important factor governing the shape of the interface.

As one may suppose, the occurrence of such interfacial shapes as shown in Figure 9 can be detrimental to producing bicomponent films. The question therefore arises as to how one can avoid getting these shapes when coextruding. Although in the past there have been some theoretical attempts¹⁸⁻²⁰ made to explain possible interfacial instabilities in the stratified two-phase flow of Newtonian and viscoelastic fluids, the type of interfacial instabilities observed in the present study is somewhat different from those reported. An explanation must await further research.

Figure 10 gives pictures of the extrudate cross section of the polystyrene/low-density polyethylene (PS/LDPE) system, coextruded through a circular die having an L/D ratio of 4. It is seen that the polystyrene, which is more viscous and yet less elastic than the low-density polyethylene, has a convex surface. Moreover, as the flow rate is increased, the low-density polyethylene tends to surround polystyrene which is more viscous.

Figure 11 gives pictures of the extrudate cross section of the PS/LDPE system, coextruded through three circular dies for different L/D ratios,

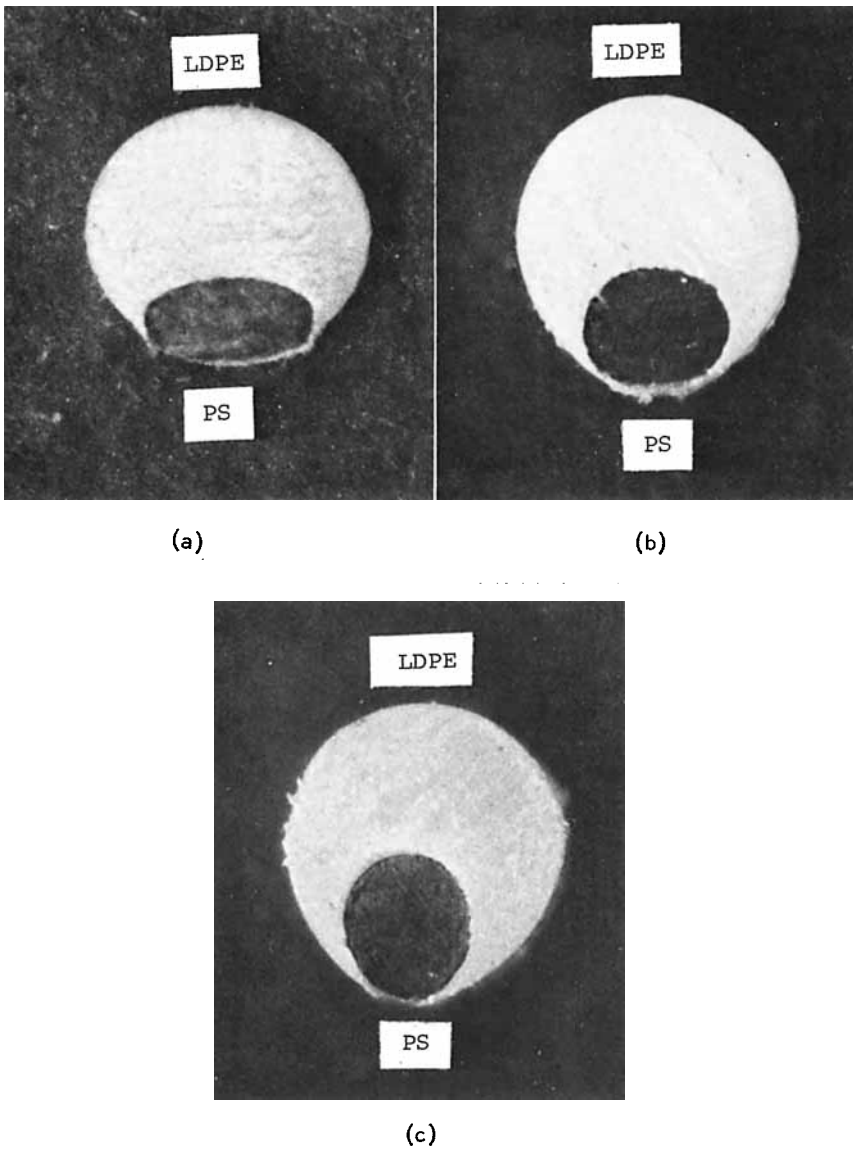


Fig. 11. Interfacial shape of the PS/LDPE two-phase system extruded through circular dies: (a) $L/D = 4$, $Q_{PS} = 8.3$ cc/min, $Q_{LDPE} = 33.2$ cc/min; (b) $L/D = 11$, $Q_{PS} = 8.3$ cc/min, $Q_{LDPE} = 30.8$ cc/min; (c) $L/D = 18$, $Q_{PS} = 8.3$ cc/min, $Q_{LDPE} = 30.8$ cc/min.

at approximately the same flow rate. It is seen that the shape of the interface becomes almost completely circular at L/D ratios of 11 and 18, giving rise to a sheath-core configuration. It can be then said that an L/D ratio of 11 is large enough to achieve an equilibrium in stress inasmuch as it affects the interfacial curvature.

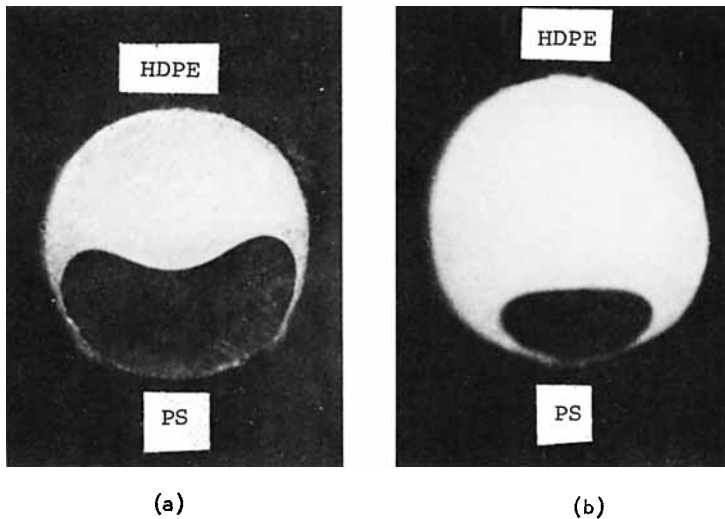


Fig. 12. Interfacial shape of the HDPE/PS two-phase system extruded through the circular die having an $L/D = 18$: (a) $Q_{PS} = 8.3$ cc/min, $Q_{HDPE} = 11.3$ cc/min, (b) $Q_{PS} = 8.3$ cc/min, $Q_{HDPE} = 54.2$ cc/min.

Figure 12 shows pictures of the extrudate cross section of the HDPE/PS system, coextruded through a die having an L/D ratio of 18. It is seen that a approximately the same flow rate of individual components (Fig. 12a), the high-density polyethylene, which is more viscous and also more elastic than polystyrene, has a convex surface. However, as the flow rate is increased, the high-density polyethylene tends to surround polystyrene which is less viscous, as shown in Figure 12b. This then appears to indicate that the ratio of the individual components also is responsible for determining interfacial curvature.

Now, in order to investigate the effects of viscous and elastic properties on the interfacial curvature, cross plots of Figures 2 and 3 are given in Figure 13 for the melt viscosity versus shear stress, and in Figure 14 for normal stress difference versus shear stress. It is interesting to compare Figure 13 with Figure 2, and Figure 14 with Figure 3. It is seen that relative values of the melt viscosity are not changed whether plotted against shear rate or against shear stress. However, relative values of the melt elasticity are changed by the way they are plotted. In other words, it can be said from Figure 14 that low-density polyethylene is the most elastic of the three materials tested. Earlier, Han²¹ has suggested that shear stress, instead of shear rate, be used for determining the relative elasticity of different materials.

It can now be said that whether or not a component tends to be convex or concave at the phase interface appears to be governed by the viscosity ratio and the composition of the two components involved. A similar conclusion has been drawn earlier by Southern and Ballman,⁷ who co-

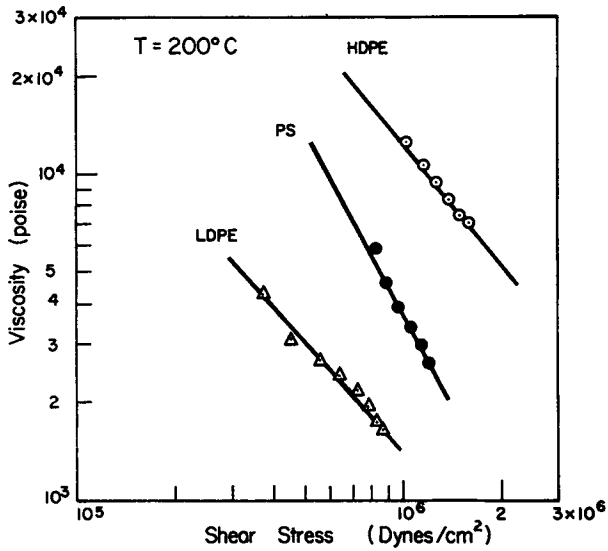


Fig. 13. Viscosity vs. shear stress for the polymer melts investigated.

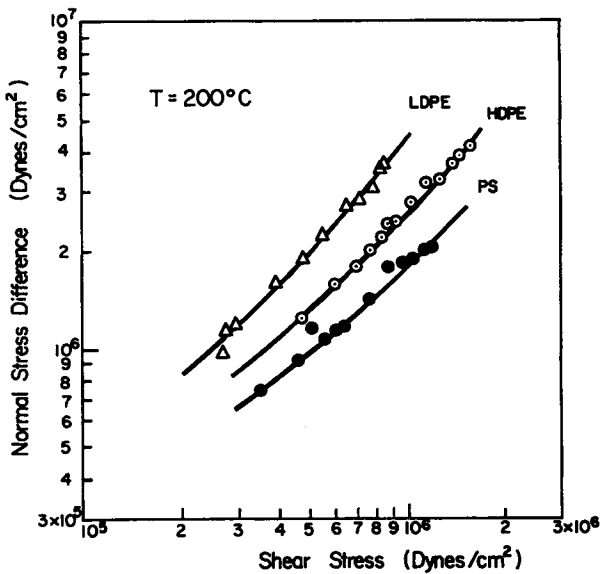


Fig. 14. Normal stress differences vs. shear stress for the polymer melts investigated.

extruded two commercial polystyrenes having different molecular weight distributions. In their study, however, die swell ratio was used as a measure of the elastic properties of the two polystyrenes used. Unfortunately, the die swell ratios of the two polystyrenes differed from each other only by 1-3%, which could as well be within measurement error. As may be seen

from Figure 14, however, the normal stress differences of polystyrene and low-density polyethylene differ from each other by more than 200%, which is beyond any possible measurement error.

What role, then, if any, do the elastic properties of the two components involved play insofar as the interfacial curvature is concerned? Very recently, White et al.,³ using some simplifying assumptions, tried to determine theoretically the role which the elastic properties of two-phase viscoelastic fluids might play in determining interfacial curvature. Their analysis shows that the fluid with the greater second normal stress difference will tend to be convex into the other fluid. It should be noted, however, that the analysis is based on the assumption that the fluid viscosities in both phases are the same. Therefore, their analysis does not seem directly applicable to two-phase systems which have widely different melt viscosities, as was the case in the study reported here.

It should be noted also, in Figures 10 to 12, that the phase interface is very smooth indeed (i.e., ripple free). It appears that the geometry of a die cross section might be a factor in the interfacial curvature of two-phase systems. The present author also is of the opinion that the melt elasticity may be responsible for the occurrence of *unstable* (i.e., corrugated) phase interfaces. However, it is merely a conjecture at the present time, and a theoretical study is under way to explain the various experimental observations reported above.

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