Melt Transformation Coextrusion of Polyethylene and Polybutylene
Measurements of Streamline Flow and Pressure Effect on Birefringence and
Interface Visualization in The MTCE Process

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Abstract

Chemical Engineering


Director of Thesis: Dr. John R. Collier.

In this investigation of melt transformation coextrusion of polyethylene and polybutylene, two single screw extruders were used, one processing the core polymer, polyethylene, the other, the shell polymer, polybutylene. The main purpose of this investigation using a new streamlined die with glass windows was to measure streamline flow in the converging section of the die, to visualize interface features in the coextrusion process and to characterize the die pressure effect on the orientation of the melt molecules by measuring birefringence and extrudates' thermal and mechanical properties.

The results of the study under different conditions (die pressure and die temperature) showed that streamline flow was reached using this die. A quite steady and clear interface was observed, and pressure had considerable effect on birefringence and further on the extrudates' final thermal and mechanical properties.

The samples produced by the coextrusion process exhibited a melting point elevation by about 5 degree celsius at die pressure 5000 psi. When the
same core polymer was extruded (single layer extrusion) alone, it had a melting point 1 degree celsius lower than that of coextrusion process at the same die pressure. It was also noted that the coextrudates' yield strength was slightly higher than that of single layer extrudates'.
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1. Introduction

During the past decade, a number of new synthetic polymeric materials have been introduced into markets. Synthetic fibers, rubbers and plastics for packaging, beverage and building materials are some typical examples. Meanwhile, much effort has been done in the polymer processing industry on the improvement of their existing processing conditions.

One of these processing techniques developed at Ohio University is the Melt Transformation Extrusion (MTE) (1) process. This process is able to produce polymer extrudates with tensile strength as high as that of aluminum. But one disadvantage is the low extrusion rate. It was found that frictional drag in the die played a critical role. In order to overcome this problem, fluoropolymer coating was applied on the die surface. This helped increase the extrusion rate, but not sufficiently.

Melt Transformation Coextrusion (MTCE) (2) was tried in the same laboratory to increase the extrusion rate. This improved process dramatically increased the extruding rate. Unfortunately, the pressure built in the die was not high enough to facilitate the desired flow orientation of the melt molecules. As a result, thermal and mechanical properties of these extrudates were not as good as those of the MTE process.

One approach to solving this problem was to alter the processing facilities. Craig Shoemaker (3) used a twin extruder for the core polymer, and successfully produced the orientation result similar to the MTE process. All of these investigation results indicated that the pressure built in the die is the most important parameter. This pressure dominates the flow induced orientation degree and, in turn, determines the extrudates' final thermal and mechanical properties. Flow induced orientation is related to the flow patterns in the die, Therefore studying flow patterns in the die is of
importance both in the processing industry and in the theoretical description.

A streamlined die with glass windows, whose design was based on theoretical analysis, was used in this investigation. The main purpose of this investigation by using this new streamlined die was to build up high pressure, to measure flow streamline, to visualize interface features in the coextrusion process and to characterize die pressure effect on orientation of the melt molecules by measuring birefringence and extrudates' thermal and mechanical properties.
2. Literature Review

The following gives a general review of the literature in the fields of polymer orientation, coextrusion and birefringence.

2.1. Achievement of High Modulus in Oriented Polymers

In practice, the following methods are available to produce high modulus oriented polymers: cold-drawing, hydrostatic extrusion, solid state extrusion, solution spinning and melt transformation. In a cold-drawing process the polymer is mechanically stretched. The formation of a neck is frequently observed in many polymers, which undergo large plastic deformation. The amount of plastic deformation is expressed as the draw ratio, which is defined as the ratio of the final length of the material drawn divided by its original length. The draw ratio is approximately constant for a given polymer, therefore, it is often called the 'natural draw ratio', and has values in the range of 4-10 for all cold-drawing polymers.

Cold-drawing has been widely studied. Andrews and Ward (4) systematically worked on the influence of molecular weight and molecular weight distribution on the cold-drawing of linear polyethylene (LPE). They concluded that the draw ratio is sensitive to the weight average molecular weight of the polymer. Capaccio and Ward (5) continued these experiments and found that drawing at temperatures appreciably above ambient but still well below the polymer melting point minimized the production of microvoids in the oriented samples. In addition, it was shown (6,7) that high effective deformation and hence high moduli can been achieved over a
very wide range of molecular weight, Mw, by an appropriate combination of processing conditions. For the high Mw polymers, molecular entanglement is the predominant features and the morphology plays a minor role. On the other hand, for low Mw polymers, with few entanglements and high crystallinity, morphology is the prevailing factor. Clark and Scott (8) produced high modulus polyoxymethylene (POM) by a two-stage drawing process. A comparison of one-stage and two-stage drawing indicated the superiority of single-stage over two-stage.

Hydrostatic extrusion has been used to produce highly oriented polymers from ultra-high molecular weight, such as linear polyethylene. In this process, a billet of polymer is forced to flow through a converging die by application of pressure to the fluid. One advantage of this process is that the friction between the billet and the container walls is eliminated and the friction between the billet and the die is very low because of an entrained fluid film.

In the investigation of the hydrostatic extrusion behavior of linear polyethylene (9), two sets of experiments were made: a small number of polymer grades in large-diameter (12.5 mm) extrusions, and Mw effects in small-diameter extrusions. The experimental results observed from large-diameter extrusions indicated that the ultra-high molecular weight samples showed no increase in modulus as a result of tensile deformation, whereas the low molecular weight grades showed a considerable increase. The relationship between modulus and deformation ratio is qualitatively similar to that observed in the drawing process. While the extensional moduli of the small-diameter extrudates increased rapidly with deformation ratio (10).

Hydrostatic extrusions of polypropylene, PP, and Polyoxymethylene,
POM, have also been conducted (11). One problem which was encountered in the PP process was the sticking of the polymer to the die during extrusion. This problem was remedied by coating the die with polytetrafluoroethylene. POM is known to be highly deformable in the solid. In the investigation of the extrusion behavior of POM, it was found that the lower Mw polymer gave lower pressures and a higher overall value of extrusion ratio, but the effect was not as pronounced as with linear polyethylene.

Solid state extrusion is another process which has produced very high modulus samples. Solid state deformation of semicrystalline polymers was studied (12-15) by extruding high density polyethylene (HDPE) through conical dies. Crystallization of the polymer in the conical entrance of the die and in the rheometer barrel was observed. Southern and Porter (12) reported that pressure rapidly increased after crystallization had occurred and the extrusion could not be maintained at constant velocity.

Deformation mechanisms and structural models received much attention in the past. Semicrystalline polymers, in order to orient, elongate and pack each chain at the highest density per cross sectional area. The spherulite and isotropic structures were believed to undergo a series of changes during deformation. A spherulite (Fig. 1) may be considered as a structure which consists of folded chain lamellae arranged radially about its center. The spherulites are surrounded by an amorphous phase, and they are connected to each other through tie molecules (17). In the process of deformation, the spherulites change from a spherical to ellipsoidal shape (18). As the spherulites deform, lamellae reorient along the stress directions and at the yield stress, when necking occurs, the spherulitic structure is replaced by a fibril structure (17).

The spherulite deformation of polyethylene has been also studied by
Fig. 1 Spherulite Polymer Crystal Structure (16)
other investigators (19-26). Stern (24) has found that even though the overall spherulite length may vary upon stretching, the microstructure may not. Peterlin (27-29) and others (30,31) proposed that the strongest element of the oriented polymer is the microfibril. The microfibril consists of highly oriented folded chain molecules connected by many tie-molecules within the amorphous phase separating the lamellae (Fig. 2) (27). According to these investigators, the increase in fiber stiffness with draw ratio results from the increase in the number of tie-molecules that occurs as lamellae blocks slip past each other during necking. The modulus may increase either by increasing the number of tie-molecules or by reducing the number of chainfolds. Fully extented chain molecules can not be formed if the draw ratio is less than the ratio of random coil to extented chain dimension (32,33). The extended chain morphologies are shown in Figure 3 (34).

Solution spinning is another process which is able to produce high modulus polymer products. It has become clear that high modulus is attributed to a high degree of extended chains and molecular orientation, and this high degree of orientation can be achieved by two different methods. For rigid polymers, they are formed from anisotropic solutions, and for the flexible polymers, mechanical deformations can be applied. Some experimental work has been done on rigid polymers by Du Pont, while theoretical developments and experimental findings related to flexible polymers have been reported by many (5,35-38). It should be pointed out that the process for producing high modulus polymer products from solution spinning is still under development.

The last method of producing high modulus oriented polymers was developed at Ohio University by Collier et al (1). This process is known as Melt Transformation Extrusion and it will be discussed separately in the
Fig. 2 Schematic Structure of Drawn Crystalline Polymer (27)
Fig. 3 Schematic Representation of Extended Chain-Lamellar Block Crystal Structure
following section.

2.2. **Melt Transformation Extrusion**

The process was first reported in 1976 by Collier et al (1). The techniques of this process basically involve the following sequence:

1). To melt and pressurize a polymer in an extrusion process,

2). To condition the molten polymer between a die and the extruder under controlled conditions of pressure and temperature for a certain period of time,

3). To accelerate the oriented melt in an elongational flow field of a specially designed die, and

4). To flow-induced crystallize the oriented melt thereby locking in the orientation under a steep temperature gradient imposed at the tip of the die.

In general the MTE process has the following three advantages over solid state extrusion process:

1). Lower extrusion pressure. The MTE process since it uses a polymer melt needs only a moderate extrusion pressure, usually in the range of 2,000-8,000 psi, while the solid state extrusion process requires four to fifteen times this pressure (39).

2). Continuous extrusion process and higher production rates. The MTE process is continuous, and higher production rates usually occur because of an extruder continuously supplying polymer to the die, while the solid state extrusion process is a batch one and has limited production rates.

3). Samples with higher modulus. The MTE process is capable of producing slightly higher modulus samples than the solid state extrusion process at equivalent values of draw ratios (40).
Among all the orientation methods, as mentioned earlier, a high elongational flow field is the common feature. Large deformation ratios and/or converging angles imply a greater acceleration in the converging section of the die. As a result, the folded molecular chains are unfolded at a fast rate when they accelerate during flow through this converging section (Fig. 4), so that a high degree of orientation of the melt is achieved (1, 39, 41).

In addition to the effect of the elongational flow field on the orientation of the melt, pressure plays a profound role also. It has been shown that the higher the pressure used in extrusion, the stronger and more transparent the samples become (43). However, the mechanism of the pressure effect on orientation is not well known. It has been proposed that the transformation from the polymer melt to polymer liquid crystalline form, like a nematic form (Fig. 5 and 6), may exist under certain conditions of temperature and pressure (44, 45). As mentioned earlier, the techniques of the MTE process basically involve four steps. The second one is to condition the molten polymer between a die and the extruder for a certain period of time. It has been found (44, 46-49) that at least 15 minutes are necessary for the polymer melt to be under the effect of the pressure and temperature conditions in the pipe between the extruder and the die in order to obtain highly oriented samples. This pipe was referred to "conditioning zone". So, conditioning in the MTE process means that the polymer has to reside in the pipe for at least 15 minutes before exiting through the die in order to produce highly oriented extrudates.

It is suspected that based on this fact, either a liquid crystal formation or unsteady state cooling is the reason for this conditioning time (3). It has been found (40) that about 10-15 minutes are needed for cooling the polymer melt from the extruder temperature to 90 per cent 0
Fig. 4 Elongational Flow in the Converging Section of a Die (42)
Fig. 5 Temperature versus Pressure for a Polymer which Exhibits a Liquid Crystalline
Fig. 6 Free Energy versus Temperature for Pressures Above and Below the Critical
f the die temperature. But the birefringence measurement results recently done by Ghosh (50) indicated possible liquid crystal formation. Thus, the transformation from an isotropic polymer melt to an anisotropic liquid crystal melt, like a nematic form, may exist in the MTE process. In the nematic liquid crystalline form, only the molecular chains are aligned in the flow direction, while their ends are randomly oriented in the axial direction, see Figure 7 (51).

As a distinct extrusion method, the MTE process has produced ultra-modulus polymer samples, for instance, PE and PP, which have also shown the enhancement in optical and thermal properties. The optical property enhancement arises from the microstructure of the fibril crystals, The diameter of the fibril is enough less than the wave length of light that scattering at the interface is negligible. The samples are more transparent than the original bulk polymer resin. The thermal property enhancement, for example, the elevation of melting point, relies on the changes of thermodynamic functions in the system.

In fundamental thermodynamics, the Gibbs energy, $G$, was defined as

$$G = H - TS$$ \tag{1}

where

$G =$ Gibbs energy,
$H =$ enthalpy,
$S =$ entropy,
and
$T =$ absolute temperature.

Upon differentiating equation (1),
Fig. 7. Nematic Liquid Crystalline Form
\[ dG = dH - TdS - SdT \quad (2) \]

It is well known that melting process is isothermal for any substance, so the third term on right-hand side of equation (2) disappears, and equation (2) is simplified as,

\[ dG = dH - TdS \quad (3) \]

After integrating equation (3),

\[ \Delta G = \Delta H - T\Delta S \quad (4) \]

The Gibbs energy change is of zero value at first order transitions such as a normal melting point, i.e. \( \Delta G = 0 \), thus,

\[ 0 = \Delta H - T\Delta S \]

or

\[ T = \Delta H / \Delta S \quad (5) \]

In order to identify the process at the melting point, equation (5) is written as,

\[ T_m = \Delta H_f / \Delta S_f \quad (6) \]

where


\( T_m = \) melt point,

\( \Delta H_f = \) heat of fusion,

and

\( \Delta S_f = \) energy change upon crystallization.

Now, taking equation (6) as a starting point to analyze why the melting point has been increased after crystallization. Here only two cases are discussed:

Case 1. polymer melt is in a random state, i.e. before crystallization, equation (6) in this case is treated as,

\[
T_{mr} = \frac{\Delta H_f}{\Delta S_f} \tag{7}
\]

where the subscript ' \( r \)' stands for polymer melt in a random state.

Case 2. polymer melt is in state of orientation, i.e. after crystallization, equation (6) in this case is treated as

\[
T_{m0} = \frac{\Delta H_f}{\Delta S_f} \tag{8}
\]

where

Subscript ' 0 ' stands for polymer melt in a state of orientation.

Generally, the heat of fusion does not change too much for both case 1 and case 2, so the heat of fusion can be assumed to be approximately the same for the case 1 and case 2 (2,3,50),

\[
\Delta H_{f0} = \Delta H_f \tag{9}
\]

thus, from equation (7) and (8),
The entropy change upon oriented crystallization is well below that for a random polymer melt, that is,

\[ \Delta S_{fr} = \Delta S_{f0} \]  \hspace{1cm} (11)

From the equations above, it is obvious,

\[ \Delta S_{fr} > \Delta S_{f0} \]  \hspace{1cm} (11)

The difference between \( T_{m0} \) and \( T_{mr} \) is the melting point elevation,

\[ \Delta T_m = T_{m0} - T_{mr} \]  \hspace{1cm} (13)

Equation (13) indicates that the samples with more extended chain crystal structure have a higher melting point elevations than those samples with less extended chains.

2.3. Methods of Coextrusion

2.3.1. General Review
During the past decade, coextrusion has become an important commercial process for manufacturing multilayered plastic products, such as film sheet, and tubing. In many cases, simultaneous extrusion of two or more molten polymers through a single die has made it possible to combine the desirable properties of several polymers, which were previously produced by lamination and coating. Through coextrusion, the manufacturers not only save the cost of handling individually fabricated film piles and combining with primers, adhesives, heat and pressure, but they can also handle much thinner layers. Films can be made less than 0.3-mil, which would otherwise be very difficult to achieve by conventional lamination and coating.

Coextrusion is a process which consists of coupling two or more extruders to a single die head. Tighter control of the extrusion process variables such as screw speed, melt pressure, melt temperatures and power input to the extruder motors is necessary. Basically, there are three methods of carrying out this operation classified according to die types.

The first is the multislit die system where polymers are individually fed to separate manifolds in the die then passed through separate slits and combined after they leave the die. In the blown film extrusion process, for example, the separate films are brought together below the 'freeze line' of both bubbles, i.e., while still molten (Fig. 8). Provision can be made in the die for the space between the films to be saturated with an active gas (such as ozone), whose function is to treat the adhering surfaces and improve the bond strength. This method is particularly useful when combining unlike materials, e.g., nylon and polyethylene.

The second is the multimanifold die system where various polymers are again fed to separate manifolds in the die but here they are combined in the final land section prior to leaving the die. Figure 9 shows a typical set up
Fig. 8 Coaxial Extrusion with Provision for Treating with Active Gas to Improve Adhesion
Fig. 9 Coextrusion Using Multiple Manifold Die(52)
for a three film process. This kind of die performs well with polymers which do not have closely matched viscosities and flow properties. If more than three layers are needed, it does not work well because the die will be difficult to design and hard to adjust.

The third one is the feedblock type die. This is the most common method for the coextrusion of flat films. In this method, the separate melts are brought together in a common manifold (Fig. 10). The flow passages are so designed as to minimize turbulence and avoid mixing of the separate streams. Generally, the viscosities of the various polymers should be closely matched when using this type of die. It is also desirable that the viscosities match at about the same temperature to eliminate problems associated with maintaining different melt temperatures in the die. This method allows great flexibility in practical operation. Using it two or more layers can be extruded relatively easily and it can extrude a polymer in much thinner layers than would be possible if it were extruded as a single film. But it is not possible to maintain a large temperature difference between layers, which is possible in the multislit process.

Table 1 lists the comparison of the capabilities of coextrusion methods for the last two coextrusion methods (52). Notice that the feedblock die method is more versatile than the multimanifold die method except for coextruding polymers with widely different viscosities.

From the market point of view, resin consumption for coextruded sheet increased from 41,000 in 1977 to 110,000 metric tons in 1980, the percentage of increase in this period is 168%. And coextruded films increased from 65,000 in 1977 to 210,000 metric tons in 1980, the percentage of increase is 223% in the same period (53).

The success of coextrusion depends very much upon the die design.
Fig. 10 Coextrusion by Laminar Flow Process Using Single manifold Die(52)
Cryovac Division designed and produced shrink and stretch films manufactured by combined oriented and nonoriented layers of ethylene-propylene copolymer coextruded with a heat sealable material on the outer layers in order to overcome the problem encountered when using polyvinyl chloride and polyethylene shrink monofilms. Composite Containers Company has developed a new approach to sheet coextrusion. The feedblock was designed with a backflow entry of the skin layer polymers. This design allows for the combination of a much wider range of dissimilar viscosity polymers and reduces interfacial turbulence, variations in layer thickness and imperfect bonding (53). Spiderless dies have been used to produce better quality pipes because spiderless dies do not have mandrel supports interrupting the polymer flow. These spiderless dies are almost turbulence free. (54). The Cloeren Company has designed a new die by using both feedblock and multimanifold technology in the same die. This new feedblock has three adjustment which can be made on each layer going through the feedblock. This feedblock then feeds a multimanifold die which has movable vanes to change the angle at which the feedblock layers come in contact with the multimanifold die layers. This new die is reported to be able to handle viscosity differences up to 400:1 (55).

2.3.2. Flow Patterns and Interfacial Stability

For years, a lot of work has been done in research leading to an understanding of various complicated flow phenomena which occur when two viscoelastic fluids flow side by side through an extrusion die (56,57). Of particular interest in this area is flow patterns and interfacial stability.

It was noted early in the transportation of heavy viscous crude oil through a pipeline that adding a small amount of water to the crude oil
Table 1. **Capability of Coextrusion Methods** (52)

<table>
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<tr>
<th>Requirement</th>
<th>Multimanifold die method</th>
<th>Feedblock method</th>
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<tbody>
<tr>
<td>Application to existing die</td>
<td>poor</td>
<td>good</td>
</tr>
<tr>
<td>suitability for more than three layers</td>
<td>poor</td>
<td>excellent</td>
</tr>
<tr>
<td>ability to increase number of layers</td>
<td>poor</td>
<td>excellent</td>
</tr>
<tr>
<td>ability to change layer thickness</td>
<td>good</td>
<td>good</td>
</tr>
<tr>
<td>ability to change layer position</td>
<td>fair</td>
<td>excellent</td>
</tr>
<tr>
<td>suitability for very thin (&lt;0.001 in) adhesive</td>
<td>good</td>
<td>good</td>
</tr>
<tr>
<td>ability to coextrude wide viscosity difference</td>
<td>good</td>
<td>poor</td>
</tr>
<tr>
<td>suitability for heat sensitive</td>
<td>poor</td>
<td>excellent</td>
</tr>
<tr>
<td>ease of die adjustment</td>
<td>fair</td>
<td>excellent</td>
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reduced the pressure gradient. The reduction in pressure gradient is attributable to the tendency for the less viscous material to preferentially wet the tube walls (58).

Serious theoretical attempts have been made to study the mechanisms of the interface in the stratified bicomponent flow. Maclean (59) and Everage (62) showed, by using the principle of minimum energy dissipation, in the stratified flow of two newtonian fluids that the sheath core configuration requires less energy than a flat interface, with the less viscous component surrounding the more viscous component. This theoretical approach result is consistent with the experimental phenomena observed by Charles and co-workers (61,62). The effect of the elastic properties of the components on the phase interface has also been studied. White et al. (63), who used some simplifying assumptions, tried to determine theoretically the role which the elastic properties of two phase viscoelastic fluids might play in determining interfacial curvature. They concluded that the fluid with the greater second normal stress difference will tend to be convex into the other fluid. However, their analysis was based on such an assumption that the fluid viscosities in both phases are the same. Therefore, their conclusion may not be applicable to the system which has widely different viscosities. White et al. (63) also conducted an experimental study, using polyethylene and polystyrene extruded through a capillary rheometer. The interface observed was distorted with the less viscous polyethylene trying to encapsulate the more viscous polypropylene. It is very interesting to note that the movement of interface, as the two polymer melts flow through a long slit section, was observed. Han and Khan (64) reported that when high density polyethylene (HDPE) and polystyrene (PS) were coextruded, the interface moved from its initial position on the HDPE side to its final position
on the PS side, and when low density polyethylene (LDPE) and PS were coextruded, the interface moved from the LDPE side to the PS side.

In an earlier study (57) the interfacial configuration in an extrudate was found to be 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, the most significant, the viscoelastic properties of the individual fluids concerned.

In coextrusion processes, the stability of the interface between two polymer melts draws much attention. Figure 11 shows the types of interfaces possible (66). A very rugged interface between the components under certain extrusion conditions was observed (57-65). The rugged interface is detrimental to the final extrusion products. Khan and Han (64) made a hydrodynamic stability analysis in order to better understand the phenomenon of interfacial stability in a plane Poiseuille stratified flow, using the Coleman-Noll second order fluid as the constitutive equation. They found that elasticity is as important as viscosity in causing interfacial instability in three dimensional flow. Southern and Ballman (65) also concluded that the interfacial instability is caused by the difference in the elasticities of the coextruded polymer melts, and with a larger difference in elasticity, the more jagged the interface between the polymer melts.

Shrenk et al. (66) found that there is a critical interfacial shear stress required for melt fracture to occur. When the output rates of extrusion increased, a wave-like form appeared. Later, they made a fractional factorial experiment to determine which variables are more important to reduce the interfacial shear stress between layers. It was found that the most important variables are: the skin-to-core layer thickness, the skin layer
Fig. 11 Interface Stability and Instability (66)
temperature and viscosity, total extrusion rate and the die gap. Thus, stable interfacial flow can be obtained by controlling these following factors: increasing the skin layer thickness, reducing the total extrusion rate, and reducing the gap of the die.

2.3.3. Theoretical Flow

It is of importance to derive flow equations for the flow rates and velocities that develop in the coextrusion process. Some work in this area has been done (67-71), including cases where both non-slippage and slippage at the interface or wall is considered. Slippage at the interface for the MTCE process could happen if there is a big difference between melting points of the two polymers being extruded. Interfacial slippage is the mechanism for a melt viscosity reduction in polymeric systems. Some fluids, such as rigid PVC often exhibit wall slippage (69,70). High density polyethylene and silicon fluid can also show wall slippage (71). In most cases, polymer-polymer interfaces have no slippage if the two polymer melts have a moderate difference in their melting points and viscosities. The derivation will be done only for non-slippage case. The following assumptions are used in the derivation:

a). Both polymer melts obey the power law constitutive equation for their shear dependent viscosity,

b). No flow instability,

c). No slippage at the interface and the wall,

d). Steady flow has been reached,

e). The melt flow is isothermal and isotropic,

f). The lower viscosity melt is flowing around the higher viscosity
polymer,
g). The width and length of the gap is much larger than the height of the gap, so that the parallel plate geometry can be assumed, see Figure 12: Die channel Velocity Distribution (2).

The symbols used in the derivation are defined as:
Fluid A = shear layer polymer melt,
Fluid B = core layer polymer melt
$K_i$ = viscosity of the polymer melt i,
$n_i$ = constants of the power law viscosity equation for polymer i
$V_i$ = velocity of the component i,
$Q_i$ = flow rate of polymer i,
$T$ = shear stress,
$P$ = pressure,
$X$ = x axis of the Cartesian coordinate,
$Y$ = y axis of the Cartesian coordinate,
$Z$ = z axis of the Cartesian coordinate,

For the coordinate system defined in Fig. 12, the equation of motion in the Z-component is

$$- \frac{\partial P}{\partial Z} = \frac{\partial T_{YZ}}{\partial Y} \quad (14)$$

Let

$$k = - \frac{\Delta P}{\Delta Z} = \frac{(P_0 - P_1)}{\Delta Z} \quad (15)$$

so

$$dT_{YZ} = kdY$$
Fig. 12 Die Channel Velocity Distribution

Z- Extrusion Direction
\[ T_{YZ} = kY + C_1 \quad (16) \]

The integrating constant, \( C_1 \), can be evaluated by the boundary condition at the center line of flow,

\[ Y = 0, \quad T_{YZ} = 0, \quad C_1 = 0 \quad (17) \]

thus,

\[ T_{YZ} = kY \quad (18) \]

for Fluid A, when the power law applied to Fluid A,

\[ T_{YZ} = -K_a \left( \frac{dV_{Z,a}}{dY} \right)^{n_a} \quad (19) \]

Substituting equation (19) into (18),

\[ -K_a \left( \frac{dV_{Z,a}}{dY} \right)^{n_a} = kY \quad (20) \]

Integrating equation (20) and solving for \( V_{Z,a} \),

\[ V_{Z,a} = - \left( \frac{k}{K_a} \right)^{\left(\frac{1/n_a}{n_a/(n_a+1)}\right)} (Y)^{\left(\frac{1/n_a}{n_a+1}\right)+1} + C_2 \quad (21) \]

No slip assumption at the wall is used here,
\[ Y = H, \quad V_{Z,a} = 0 \] (22)

Thus,

\[ C_2 = \left( \frac{k}{K_a} \right)^{1/n_a} \left( \frac{n_a}{n_a+1} \right) \left( \frac{H}{n_a} \right)^{1/n_a} + 1 \] (23)

The velocity distribution is,

\[ V_{Z,a} = \left( \frac{k}{K_a} \right)^{1/n_a} \left( \frac{n_a+H}{n_a+1} \right) \left( 1-\left( \frac{Y}{H} \right)^{1/n_a} + 1 \right) \] (24)

The volumetric flow rate per half channel width (W) can be calculated using equation (24),

\[ \frac{Q_a}{W} = \int_{H}^{H} V_{Z,a} \, dY \] (25)

\[ Q_a/W = \left( \frac{k}{K_a} \right)^{1/n_a} \left( \frac{n_a H^2}{(2n_a+1)} \right) \left( 1-(1/H)(1+2n_a)/ (1+n_a) \right) + \left( n_a/(1+n_a) \right) \left( h/H \right) \left( 1+n_a/2 \right) \] (26)

For Fluid B,

\[-K_b \left( \frac{dV_{Z,b}}{dY} \right)^{n_b} = kY \] (27)

Solving for \( V_{Z,b} \),
\[ V_{Z,b} = -(k/K_b)^{1/n_b} (n_b/(n_b+1))(Y)^{(1/n_b)+1} + C_3 \]  

(28)

The constant, \( C_3 \), can be evaluated by the boundary condition of

\[ Y = h, \quad V_{Z,b} = V_{Z,a} \]  

(29)

so,

\[ C_3 = (k/K_a)^{1/n_a} (n_a H/(n_a+1)) (1-(Y/H)(1/n_a)+1) + \\ (k/K_b)^{1/n_b} (n_b/(n_b+1)) (h^{(1/n_b)+1} \]  

(30)

The velocity distribution of the Fluid B is,

\[ V_{Z,b} = (k/K_a)^{1/n_a} (n_a H/(n_a+1))(1-(Y/H)(1/n_a)+1) + \\ (k/K_b)^{1/n_b} (n_b H/(n_b+1))(1-(Y/H)(1/n_b)+1) \]  

(31)

The volumetric flow rate

\[ Q_b/W = V_{Z,b} dY \]  

(32)

\[ =(k/K_b)^{1/n_b} (n_b h^2)/(2n_b+1) + (K/K_a)^{1/n_a} (n_a H)/ \]
The total volumetric flow rate of the entire die gap becomes

\[ Q_{\text{total}} = 2 \left( Q_a + Q_b \right) \quad (34) \]

Therefore, the velocity distribution and the flow rates can be determined according to the expressions derived above.

2.4. The Theoretical Background for Birefringence Measurement

As we know, an ordinary polymer melt is isotropic in character since its molecules are randomly oriented in all directions, and therefore no directional property characteristics are present. But, if such a polymer melt is set in motion, it may no longer be isotropic, that is the molecules in the melt will become more oriented in the direction parallel to the motion, and therefore the properties of the polymer melt become different in different directions.

When a light beam is directed at the melt, the refractive index is different in the direction perpendicular to the motion and the direction parallel to the motion. The refractive index difference in the two directions is called birefringence, thus,

\[ \Delta n = n_{pa} - n_{pe} \quad (35) \]

where
\[ \Delta n = \text{birefringence}, \]
\[ n_{pa} = \text{refractive index in the direction parallel to the motion}, \]
\[ n_{pe} = \text{refractive index in the direction perpendicular to the motion}. \]

From simple theories of photoelasticity (72,73), it has been shown that the birefringence, \( \Delta n \), is directly proportional to the wave length of the light and inversely to the products of \( 2\pi \) and the thickness of the sample, by means of following relation,

\[ \Delta n = \frac{\lambda}{2 \pi d} \alpha \]  
\[ \text{(36)} \]

where
\[ \lambda = \text{wave length of the light used}, \]
\[ d = \text{thickness of the sample inside the die}, \]
and
\[ \alpha = \text{phase difference of the transmitted light between the principal directions}. \]

It has been also found that for a stressed model under the dark field arrangement of a circular polariscope (Figure 13), the phase difference, \( \alpha \), is given by the following expression,

\[ \alpha = 2 \arcsin \left( \frac{I}{I_0} \right)^{1/2} \]  
\[ \text{(37)} \]

where
\[ I = \text{intensity of the emergent beam when the sample is flowing through the die}, \]
and when the analyser is crossed with respect to the laser beam.
Fig. 13 A Stressed Photoelastic Model in a Circular Polariscope Arrangement
\( I_0 \) = intensity of the emergent beam with no sample in the die and with the analyser parallel to the laser beam. Thus, after measuring \( I_0 \) and \( I \), the phase difference, \( \alpha \), can be calculated by equation (37). Then substituting the calculated value of \( \alpha \) into equation (36), the birefringence, \( \Delta n \), can be evaluated. For details of the derivation, readers are referred to reference (52).
3. Experimental

This section describes the equipment and procedures used in this investigation.

3.1. Equipment

The equipment used is listed below.

A. Die

A side-view, internally cooled 416 stainless steel die was used. The die has a length of 7.0 inches, width of 2.8 inches and thickness of 2.85 inches. The flow channel has a length of 5.2 inches, width of 1.0 inch and height of 0.7 inch.

The ribbon thickness and deformation ratio (DR) can be changed by changing the inserts which determine the final thickness of the ribbon.

The two glass windows were inserted in each half of the die and held by two hollow window pieces, which make it possible for the investigator to measure birefringence and visually inspect the melt flow inside the die. The die is shown in Figures 14 and 15, and the insert in Figure 16.

B. Differential Scanning Calorimeter (DSC)

The Differential Scanning Calorimeter (DSC), with a chart recorder Model 56-1003, both made by Perkin Elemer Instruments, Norwalk, Connecticut was used. The DSC was used to measure the melting points of extrudates.

C. Extruders
Fig. 14 Front of the Die Section
Fig. 16 Front and Top of Land Insert

FRONT OF LAND INSERT

TOP OF LAND INSERT
Two single screw extruders were used. One single screw extruder with a 3/4 inch diameter barrel and a length to diameter ratio of 20 manufactured by Brabender Instrument Incorporated, South Hackensack, New Jersey, was used. This extruder is a model 200 with a screw having a compression ratio of 4:1. The extruder is supplied with 1.5 horsepower 220 volt motor, model PL-V180, with a maximum speed of 2400 RPM, a gear reduction box with a reduction of 20.4:7. The speed controller for the motor is a Fincor model 2402 MKII DC motor controller, manufactured by Fincor Incom International Incorporated, York, Pennsylvania. This extruder was used for processing the core polymer, polyethylene.

The other single screw extruder also had a 3/4 inch diameter and a length to diameter ratio of 20 manufactured by Seiscor, a division of Seismograph Service Corporation. This extruder is driven by a 3/4 horsepower D.C. motor manufactured by General Electric Corporation. This motor is provided with a single reduction worm gear reducer, 321-c series, manufactured by Boston Gear Corporation. This extruder was used for processing the shell polymer, polybutylene.

D. **Metering Pump**

A 0.297 cc/Rev. high temperature gear pump with a maximum input pressure of 1,000 psi was utilized. It is driven by a 1/4 horsepower D.C. motor manufactured by General Electric Corporation and a gear reducer box, model WC313-100-GS, manufactured by Boston Gear Corporation. The high temperature metering pump was manufactured by Zenith Corporation. The metering pump described above was employed to stabilize and control flow rate.
E. Heaters

The 800 watt heaters in the Brabender extruder were provided by Brabender. The 800 watt heaters in the barrel of the shell side extruder and the 500 watt heaters for the line between the extruder and gear pump were provided by Seiscor. The 1,600 watt heaters for the die were obtained from the Industrial Heater Company, New York City, New York. All other heaters used for the piping, connections, tees, couplers and elbows were provided by the Industrial Heater Company. The heaters range from 300 to 600 watts depending upon the specific use of the heater and the power density needed for the specific design.

F. Pipes and Fittings

All pipes and fittings utilized in this investigation were supplied by Autoclave Engineers Incorporated. These medium pressure pipes have a maximum working pressure of 10,000 psi at 200 °F. The pipe threads are left handed, with 16 threads per inch. The piping has an outside diameter of one inch and an inside diameter of 0.678 inches. All threaded joints were treated with anti-seize lubricant and thread sealant in order to make removal of the joints easier and for better sealing.

G. Tensile Testing

All tensile tests of samples were done on an Instron Universal Testing Instrument, model TT-D manufactured by Instron, Canton, Massachusetts. The load cell is a GR type, model D 30-20, with a maximum capacity of 20,000 pound. All samples were tested on the machine at a chart speed of 2.0 inches per minute and the crosshead speed of 1.0 inch per minute.

H. Temperature Controllers
The two heaters in the Brabender single screw extruder and the die heaters were regulated by three West model JPC controllers manufactured by West Instruments Corporation and provided by Brabender Instrument Incorporated. The two API Instruments controllers were provided with the Seiscor single screw extruder made by the API Instruments Corporation. The other heaters on the piping, fittings and couplers were controlled by three Love proportional controllers, Model 52, manufactured by Love Controls Corporations. Two out of the three controllers regulated more than one heater, but only one thermocouple was used to measure the temperature at the end of the heating section.

I. Pressure Transducers

Three Dynisco pressure transducers were used to measure the pressures at three points in the system. All of which were the same model, TPT 432 A-10M-6/18. They were located at the die and the ends of both extruders, and shared one pressure indicator. The pressure transducer at the die was usually connected to the pressure indicator. If it is necessary, the pressure indicator was shifted to connect one of other two pressure transducers.

J. Cooling Chamber

A copper cooling chamber was located just in front of the extrudates' exit to cool the molten extrudates, so that the flow-induced orientation in the melt was frozen in place. The cooling chamber has the dimensions of 120 x 25 x 18 mm. Laboratory compressed air was used as the cooling fluid.

K. Microscope:
The magnified pictures of the flow interface were taken using a Wild stereo-microscope, model M5A, with a Wild camera, model MP5II. A Wild semi-photomat, model MPS15, was used together with the microscope. The Wild stereo-microscope was also utilized in measuring the streamline flow.

M. Optical Accessories:
M.1. Laser
A Helium-Neon 15 mw, Class 3 B Laser Product, was used. The model, 120s Helium-Neon laser, was made by Spectra-Physics Stabilite.

M.2. Light Detecting Device
The light detecting device used was an Oriel 7068 photomultiplier detector head and an Oriel 77340 uv-visible photomultiplier tube.

M.3. Quarter wave Plate
The quarter wave plates used were L 293-6328 series 1/4 wave plates, manufactured by Gaertner Scientific Corporations.

3.2. Experimental Procedure

This section describes in detail the experimental work and its procedure.

3.2.1. Coextrusion
A. Die Preparation
In operation, the die often needed to be dismantled and mantled repeatedly. Whenever it was needed, the following steps were followed. First, the transducer and the cooling chamber were removed from the die
while the die was still on line. Then, the die was taken off line, the heaters and the die were dismantled, and as much of the molten polymer cleaned off of the inside of the die as soon as possible while the polymer was still in the molten state. Usually some polymer hardened before it was cleaned out. In this case, it had to be heated above its melting temperature while continuously cleaning. In this investigation, the die parts were heated in an oven. Attention had to be taken in this step. The parts could not be heated too much over the melting temperature to prevent excessive degradation and carbonation of the molten polymer. If this happens, it is removed with difficulty using brass brushes. Second, special care to prevent damage had to be taken while scraping the surface parts, especially, the surfaces of the insert and the die itself. Brass brushes were also used in this step. Next, after almost all the solid polymer was cleaned out, the last step of complete removal of dirt and polymer residuals was taken. This was done by rubbing with water soaked fine grade emery paper (600) until the surfaces were shiny. Finally, the die was then reassembled, including fitting the insert, glass windows, cooling chamber, the die heaters and the die adaptor. The die was tightened to 100 ft-lbs while the glass window retainers were tightened to 85 ft-lbs using an Allen wrench.

**B. Connection of the Die to the Extrusion Line**

After assembling the die, it was put on the die support, which was specially designed for later measurement of flow birefringence. The core extruder was adjusted to the same height as that of the adaptor, so that the die adaptor could be connected to the transfer line. This line is called the "conditioner" and was already connected to the extruder. All the piping fittings on the line were tightened to 100 ft-lbs torque using the same Allen wrench.
wrench, after all the joints were treated by anti-seize lubricant and thread sealant for easier removal of the joints and better sealing.

The shell extruder was connected to the system either before or after the die was put on the line. The important thing was that before connecting the shell extruder with the metering pump to the system, to make sure that it ran well independently.

After connecting the shell extruder to the line, the pressure-temperature probe was installed in the die. Teflon tape was used to make removal of the probe easier and to improve sealing. Then, insulation of the die, the conditioner, and the piping was accomplished using fiber glass insulation. Thermocouples were installed wherever they were needed.

C. Running Coextrusion

Following the assembly steps described above, the system was ready to be heated up. The heaters were turned on and heating occurred in 50 C° steps until the temperatures reached their first set points. Approximately 20 minutes was needed for the die temperature to equilibrate and one half of that time was needed for the other parts to equilibrate. Then, the set points were increased up another 50 C° again, the same procedure was repeated until all the temperatures reached their desired set points. Approximately 90 minutes were needed for the system to obtain thermal equilibrium. It should be pointed out that after temperatures reached their final set points, retightening was made at all the joints, glass windows and the die. This step was essential for the die. It was noticed that if no retightening was done, the die tended to leak at the pressure over 3,500 psi. The torque on the die was usually as high as 120 ft-lbs, and did not exceed 130 ft-lbs since higher values may damage the gasket in the die.
The general temperature distribution is shown in Figure 17. Once the thermal equilibrium was reached, the core extruder was started first. After the extrudate was coming out from the die, the shell extruder was started. The shell polymer melt was joined with the core one. At the same time, the laboratory compressed air was allowed to flow through the internal cooling chamber. The air flow rate was measured by the flow meter which was provided with the shell extruder.

At this point, the run was started, data was taken. When a run was finished, the shell extruder was turned off first, then the core extruder. All heaters were shut off when a zero pressure reading was reached in the die. The data was recorded and the samples were collected.

3.2.2. Single Layer Extrusion

In this investigation, single layer extrusion was also made in order to compare the results to coextrusion. The polymer used for single layer extrusion was the same as the core one used in two component trilayer coextrusion.

The procedure for getting the system ready for single layer extrusion was the same as that of trilayer coextrusion except that the shell extruder was taken off line and the system was run for a certain period of time to purge all of the skin polymer out. After all the skin polymer was purged, the single layer extrusion was started, following the procedure described in the section above.

3.2.3. Streamline Flow Measurement

As was mentioned in the introduction, one task of this investigation was to measure streamline flow in the converging section of the die. The
Fig. 17 Temperature Distribution in the Coextrusion Process

Core Extruder

195 °C
190 °C

185 °C

Skin Extruder

190 °C

180 °C

Die

175 °C
200 °C
purpose of doing this was to verify by experiment whether the new side-viewing die could give streamline flow or not since streamline flow is capable of enhancing orientation of the melt molecules efficiently.

A. Preparations of Tracer and Transparent Coordinate:

Originally, aluminum oxide (Al₂O₃), whose particle size is smaller than 10 μm, was intended to be used in the measurement. It turned out that these particles were too small to be traced, so a reasonable size of copper tracer was prepared in the laboratory where the investigation was performed. The tracer size is about 0.5x0.5x0.05 mm.

The term "transparent coordinate" means that a coordinate grid was copied to a transparency, so that a transparent coordinate grid was produced. This was placed on the window and used to locate the particle flowing through the die.

B. Measurement of Particle Flow Paths

This was done in the single extrusion process. Before performing the streamline measurement, two important things needed to be done. First, the die was leveled using a level. Second, the transparent coordinate was placed on the surface of the glass window using a double sided tape. The second is a critical step and special care should be taken. The coordinate should be oriented properly. This has to be checked prudently and patiently to make sure that the x-axis of the coordinate was parallel to the tracer flow path along the center line of the channel, otherwise, the particle flow paths recorded were meaningless.

The microscope was used to enlarge the image, so that the investigator was able to follow each particle easily and accurately. Nine readings were
recorded for each particle flowing through the channel of total length 5.2 inches. This is how the measurement of streamline flow was done.

3.2.4. **Birefringence Measurement**

The instrumental set up for birefringence measurement is shown in Figure 18. The Helium-Neon laser beam came out horizontally and entered the chopper, which produced a controlled frequency loss of the laser beam. The first 1/4 wave plate was placed vertically in the path of the incident laser beam. One of the major axes of the wave plate had an angle of 45° with respect to the optical axis of the laser beam. After passing through the die, the laser beam entered another 1/4 waveplate, which was also placed vertically but was cross with respect to the first wave plate. The next optical element which the laser beam entered was an analyzer, which is a polarizer whose optical axis was perpendicular to that of the original beam. The last optical element is called a detector, which was used to amplify and convert the remaining light signal into an electrical one for measurement of birefringence.

The measurement of birefringence was done in two steps:

A. **Measurement of Io**

As was mentioned earlier that Io is the intensity of emergent beam with no sample in the die (empty die). For measurement of it, the die was dismantled and thoroughly cleaned. Special attention was paid to the two pieces of glasses in the glass windows. No polymer chips or dirt were allowed on them, otherwise the result would be affected. After that, the die was mantled and put back on the line. Once this was done, the measurement Io was taken without running the system.

B. **Measurement of I:**
Fig. 18 The Instrumental Set up for Birefringence Measurement
I is the intensity of the emergent beam with the polymer flowing through the die. After \( I_0 \) was taken, extrusion was started, and \( I \) was measured. \( I \) is a function of orientation of the melt molecules, so \( I \) has different values at different operating conditions.

### 3.2.5. Measurement of Melting Points of Extrudates

A Perkin Elmer Differential Scanning Calorimetry (DSC) 1-B was used to determine melting points of the extrudates. The samples were placed in an aluminium pan, covered and crimped. The samples were tested at a scan rate of 20 °C per minute and a range of 8 millivolts. The chart recorder was set at a full range of 10 millivolts and the chart speed of 40 millimeters per minute.

### 3.2.6. Measurement of Mechanical properties of Extrudates

The Instron TTD Universal Testing Instrument was utilized to determine yield and ultimate strengths. The test samples were cut to be 63.5x9.53 in mm according to ASTM D368-86. The tests were made at a cross-head speed of 1.0 inch per minute and a chart speed of 2.0 inches per minute. The samples were tested until fracture occurred.
4. Results

In this section, the experimental results are presented. According to the experimental design, the results are divided into five groups: streamline flow, flow fields and pressure effects on birefringence, pressure effect on extrudate melting point, pressure effect on yield and ultimate strength of the extrudates, and interface features.

4.1. Streamline Flow

This was done in the single layer extrusion process. Figure 19 is the plot of streamline flow measured at the die temperature of 190 °C, pressure of 2,000 psi and the extrusion speed of 5 RPM. Three particles, A, B, and C, were traced. A, B, and C indicated in the figure were also the initial locations of these particles. Figure 20 shows another plot of streamline flow measured at the die temperature of 190 °C, pressure of 3,000 psi and the extrusion speed of 10 RPM. In this run a total of five particle were traced, A, B, C, D, and E. Identically, A, B, C, D, and E indicate in the plot the initial locations of these particles.

4.2. Flow Fields and Pressure Effects on Birefringence

This was done in the trilayer coextrusion process. Figure 21 shows the birefringence changes with locations along the center line of the insert starting with the melt reservoir and progressing down through the converging section. Measurements were made at a die temperature of 190 °C, a pressure of 1,600 psi, a ratio of the core polymer to the skin polymer about 9.0 and the core extruder speed of 1.0 RPM. Figure 22 shows the
Fig. 19 Streamline Flow

Die Temperature 190 ºC
Die Pressure 2,000 psi
Speed 5 RPM

Flow Direction

1.25 mm
56.52 mm
75.56 mm
54.0 mm

17.78 mm
Fig. 20 Streamline Flow

Die Temperature 190 °C
Die Pressure 3,000 psi
Speed 10 RPM

Flow Direction
Fig. 21 Flow Field Effect on Birefringence

Ratio of Core/Skin = 9.0
Speed = 1.0 RPM

$T = 190 \degree C$
$P = 1600 \text{ psi}$

Position along Center Line of the Insert, mm

Birefringence $\times 10^7$
Fig. 22 Flow Field Effect on Birefringence

Ratio of Core/Skin=6.0
Speed=5.0 RPM

P=3200 psi
T=175°C

Position along Center Line of the Insert, mm

Birefringence x E7
birefringence changes with location, which is the same as that in Figure 21 except that the measurement for the latter was done at a die temperature of 175 ºC, a pressure of 3,200 psi, a ratio of the core polymer to the skin polymer about 6.0 and the core extruder speed of 5.0 RPM. Figure 23 is the comparison of the results shown in figures 21 and 22. In figure 24 birefringence changes with pressure at three fixed points are presented. The three fixed points were measured along the vertical plane at the start of convergence. All three points were located in the core polymer region. Curve 1 is 2.615 mm away from the top edge, and curves 2 and 3 are 5.229 mm and 7.844 mm away from the top edge, respectively as shown in figures. The interface is estimated to be about 1.976 mm from the top edge according to the ratio of the core polymer to the skin polymer. And the center line is 8.89 mm either from top or bottom edge.

4.3. Pressure Effect on Extrudate Melting Point

Figures 25 and 26 show the pressure effects on melting point of the extrudates in the single layer and trilayer coextrusions, respectively.

4.4. Pressure Effect on Yield and Ultimate Strengths

Figures 27 and 28 are the results of pressure effect on yield strength of the extrudates, and Figure 29 and 30 indicate the results of pressure effect on ultimate strength of the extrudates both in the single layer and the trilayer coextrusions. Figure 31 presents the comparison of pressure effect on yield and ultimate strengths of the extrudates in the single layer extrusion and Figure 32 is the comparison of pressure effect on yield and ultimate strengths of the extrudates in the trilayer coextrusion.

4.5. Interface Features
Fig. 23 Flow Field Effect on Birefringence

Position along Center Line of the Insert, mm

Birefringence x E7

- □ 1600 psi
- ● 3200 psi
Fig. 24 Pressure Effect on Birefringence

- 2.615 mm from top edge
- 5.229 mm from top edge
- 7.844 mm from top edge

Birefringence x E7

Pressure, psi
Fig. 25 Pressure Effect on Melting Point

Speed: 1.0-8.0 RPM
T: 230-160°C

Single Layer Extrusion

Melting Point

Pressure, psi
Fig. 26 Pressure Effect on Melting Point

Ratio of Core/Skin = 9.0-6.5
Speed < 1.0-10.0 RPM
T = 220-155 °C

Pressure, psi

Melting Point, K

Trilayer Coextrusion
Fig. 27 Pressure Effect on Yield Strength

Speed <1.0-8.0 RPM
T=230-160 C

Pressure, psi

Yield Strength x 10^3 psi
Fig. 28 Pressure Effect on Yield Strength

- Ratio of Core/Skin = 9.9-6.0
- Speed < 1.0-10.0 RPM
- T = 220-155 °C

Yield Strength x E-3 psi
Pressure, psi

Trilayer Coextrusion
Fig. 29 Pressure Effect on Ultimate Strength

Ultimate Strength x E-3, psi

Pressure, psi
Fig. 30 Pressure Effect on Ultimate Strength

- Ratio of Core/Skin = 9.0-6.0
- Speed ≤ 1.0-10.0
- T = 220-155°C

Ultimate Strength x 10^3 psi
Fig. 31 Pressure Effect on Strengths

- Single Layer Extrusion
- Yield Strength
- Ultimate Strength

Strengths $\times 10^3$ psi

Pressure, psi
Fig. 32 Pressure Effect on Strengths

- Yield Strength
- Ultimate Strength

Trilayer Coextrusion

Pressure, psi

Strengths x E-3, psi
In the study of phase interface features, two sets of material systems were employed: polybutylene and polyethylene (high density) and polybutylene and polyethylene (low density). The polybutylene was used as the skin component in both systems. Fig. 33 is a side-viewing photograph showing the phase interface which was taken in the convergent section at the die pressure of 600 psi, temperature of 175 °C, the core extrusion speed of 2 RPM, and using polybutylene and polyethylene (high density). Fig. 34 is another side-viewing photograph showing the phase interface. In Fig. 34, all of the conditions were the same as those of Fig. 33 except that the photograph was taken at a die pressure of 2,000 psi, temperature of 138 °C, and the core extruder extrusion speed of 3 RPM. Fig. 35 is the side-viewing photograph indicating the phase interface. This photograph was also taken in the convergent section, at the die pressure of 1,500 psi, temperature of 190 °C and the core extrusion speed of 1,500 psi, temperature of 190 °C and the core extrusion speed of 5 RPM, but the material system was polybutylene and polyethylene (low density).
Fig. 33 Side-Viewing Photograph Showing the Phase Interface

P=600 psi  Core/Skin=9.0
T=175 °C  Polybutylene/Polyethylene(high)
Speed=2 RPM  Multiplication: 5
Fig. 34 Side-Viewing Photograph Showing the Phase Interface

P=2000 psi  Core/Skin=9.0
T=138 °C    Polybutylene/Polyethylene(high)
Speed=3 RPM Multiplication: 5
Fig. 35 Side-Viewing Photograph Showing the Phase Interface

P=1500 psi  
T=190 °C  
Speed= 5 RPM

Core/Skin=9.0  
Polybutylene/Polyethylene(low)  
Multiplication: 5
5. Discussion of Results

This section is a discussion of the experimental results in this investigation.

5.1. Streamline Flow

With the aid of the tracer particles, specific flow pathlines were determined. They are illustrated in Figures 19 and 20. In Figure 19, the streamline flow was measured at a die pressure of 2,000 psi and temperature of 190 °C, and in Figure 20, it was made at a die pressure of 3,000 psi and temperature of 190 °C.

As it is seen that all the individual flow pathlines both in Figures 19 and 20 indicate a streamline flow pattern. The flow changes in nature from a unidirectional shear field in the upper reservoir to a significant elongational one in the converging flow region near the land section.

It seems by comparison of Figures 19 and 20 that the extrusion parameters (die pressure and die temperature) do not alter the flow pathlines or flow pattern, at least no appreciable change in the flow pattern is observed. Increasing the extrusion pressure by combination of decreasing the die temperature and increasing the extrusion speed only leads to changing the elongational flow rate and accelerates the melt molecular orientation in the converging section. It was noticed that flow acceleration occurred in the converging section as indicated by the recorded resident time. An immediate, rapid decrease in transverse cross-sectional area of the converging section resulted in a rapid initial acceleration of the melt as it passed through it.

It was interesting that the "die swell" phenomenon was observed in the uppermost reservoir of the die where polymer melt exited from the piping.
This is because of a slight change in cross-sectional area between the piping exit and the reservoir entrance, so that the viscoelastic melt is able to exhibit elastic stress relaxation.

5.2. Flow Field Effect on Birefringence

Flow field effect on birefringence is plotted in Figures 21, 22 and 23. In Figures 21 and 22, the flow fields were measured from the melt reservoir down to the converging section along the center line of the insert at the pressure of 1,600 and 3,200 psi, respectively. Figure 23 is the comparison of Figures 21 and 22. Figure 24 shows the pressure effect on birefringence at three fixed points on a line perpendicular to the center line at the start of convergence. All the three points were located in the core polymer region. Specific locations were described in the previous chapter.

It can be seen clearly from Figures 21 and 22 or 23 that birefringence changes with positions along the center line of the insert. Specifically, it rapidly starts to increase from the beginning of the convergence. The data in Figure 23 indicates the effect of the pressure on birefringence at the same flow field. An important phenomenon depicted in Figures 21 and 22 is that the birefringence values are different at the two points of x=0 and x=24.09 mm. It should be kept in mind that the point x=24.09 mm in this coordinates locates the focus of the convergence, whereas x=0 is 24.09 mm upstream from this location, i.e, x=0 is located in the melt reservoir. Introducing a skin layer in the coextrusion process not only enhances some properties of the extrudates, but also improves the processing conditions. Generally speaking, a much lower viscosity skin layer reduces the friction between the core polymer melt and the die walls, and further lessens the pressure drop in
the die. There seems to be a general agreement that a much lower viscosity skin layer functions as a "lubricant" in the coextrusion process, so that the shear stress is concentrated in it. As a result, the flow pattern in the die could be changed. Particularly, if the shear stress can be completely taken away from the core, the flow in the reservoir section (equal cross-sectional area) will become a plug flow one. It is known that plug flow does not affect the molecular orientation of the polymer melts. In Figures 21 and 22 or 23, the two points at \( x=0 \) (upstream of convergence) and \( x=24.09 \) mm (start of convergence) have different values of birefringence which may imply that a certain amount of shear flow still existed and a velocity profile adjustment occurred. It may also mean that the elongational flow is transmitted upstream as suggested by Fig. 35, thereby affecting the molecular orientation that developed when the polymer melt flowed passing from the points \( x=0 \) and \( x=24.09 \) mm (74). Figure 24 indicates that birefringence increases linearly with the die pressure.

5.3. Pressure Effect on Melting Point of Extrudates

The effect of pressure on the melting point of the extrudates is illustrated in Figures 25 and 26. Figures 25 and 26 stand for the single layer extrusion and the trilayer coextrusion systems, respectively. From the plots of 25 and 26, three characteristics are observed. First, both in the single layer and the trilayer extrusion systems, the melting points increase with the die pressure. Second, beyond the die pressure at 4,500 psi, the increase in the die pressure does not enhance the elevation of the melting point in the single layer extrusion. However, a different dependence is noted in the coextrusion system. Third, in the single layer extrusion, the melting point
gradually increases with the die pressure, and the increase in the melting point continues to some extent after the die pressure reached at 3,500 psi, then stops increasing at the die pressure of 4,500 psi. But in the coextrusion system, the melting point rapidly increases initially with the die pressure, then it changes slightly when the die pressure increases from 1,500 to 4,500 psi; after that, it continues to increasing until the die pressure reaches 5,500 psi.

There are two important points worth paying attention to. First, at the die pressure about 1,700 psi, the melting point of the extrudates in the single layer extrusion system is about 400.7 K, while the melting point of the extrudates in the trilayer coextrusion system is about 402.7. The difference of 2 °C is big. This fact indicates that at this pressure the molecular orientation in the trilayer coextrusion system has developed considerably. This degree of the molecular orientation in the trilayer coextrusion system corresponds to the one in the single layer extrusion at the pressure of 4,200 to 4,300 psi. From the above analysis, it can be concluded that the measured pressure at 1,700 psi in the trilayer coextrusion system is a "superficial" pressure. In fact, this pressure on a birefringence basis is equivalent to the pressure at 4,200 to 4,300 psi in the single layer extrusion. Second, when the die pressure reaches 4,500 psi, the melting point of extrudates in the single layer extrusion no longer increases while the opposite result is observed in the coextrusion system. This phenomenon reflects that some shear distortion happened in the single layer extrusion system in this range of the die pressure, which results in distortion of further molecular orientation (75). But in the trilayer coextrusion, this shear distortion is remedied by the "lubricant" of the skin layer, therefore, the melting point in the trilayer coextrusion system still increases with the die pressure.
5.4. **Pressure Effect on Yield and Ultimate Strengths**

**A. Pressure Effect on Yield Strength**

The die pressure effect on yield strength is shown in Figures 27 and 28. Figure 27 is for the single layer extrusion while Figure 28 is for the trilayer coextrusion. It is easily seen that yield strength of the extrudates increases with the die pressure either in the single layer extrusion or in the trilayer coextrusion. It is noted from these two figures that yield strength increases slightly with die pressure in the range of 1,500-3,500 psi; after that, the increase in yield strength with die pressure becomes more obvious, especially in the coextrusion system. It seems that the die pressure effect on yield strength in the single layer extrusion system is not as profound as in the trilayer coextrusion system. From Figure 28, a transition is observed in the trilayer coextrusion system, from which yield strength sharply increases with the die pressure until 5,500 psi. It should be mentioned that when the die pressure exceeded 3,500 psi, the extrudates' color changed from ivory-like to colorless, and the extrudates became more transparent. By comparison of these two figures with Figures 25 and 26, it is immediately noted that the die pressure effect on the extrudates' yield strength is similar to that of the die pressure effect on the melting point of extrudates. So, the mechanism for explanation of the die pressure effect on extrudates' yield strength is the same as that of the die pressure effect on melting point of the extrudates.

**B. Pressure Effect on Ultimate Strength**

The pressure effect on ultimate strength of the extrudates is presented in Figures 29 and 30. Figure 29 is for the single layer extrusion system and
Figure 30 is for the trilayer coextrusion system. It can be seen from the trilayer coextrusion system that the die pressure effect on ultimate strength of the extrudates has a similar dependence to that of the die pressure effect on the yield strength of the extrudates. This is compared in Figure 32, which is a combination of Figures 28 and 30. But in the single layer extrusion, this does not occur as shown in Figure 31, which is a combination of Figures 27 and 29.

It is noted in Figure 29 that the die pressure effect on ultimate strength of the extrudates has less regularity. When the take-up device was used, it frequently caused the extrudates to twist. Thus, the pull of the extrudates (while it came out of the die) was in fact done by hand. Although care was taken, non-uniform extension existed and may be the possible reason which would explain the ultimate strength effect.

5.5 Phase Interface Features

As mentioned earlier in chapter 4, photographs 33 and 34 were taken using polybutylene/polyethylene (high density) while photograph 35 was taken using polybutylene/polyethylene (low density). It should be pointed out that the phase interface in the former system was easy to recognize, but this was not true in the latter system since only a little color contrast existed in this system. In order to observe the phase interface of the polybutylene/polyethylene (low density), the aluminum oxide (Al2O3), whose particle size is smaller than 10 m, was added to the core polymer, polyethylene (low density). As a result, obvious color differences between the two fluid phase interface occurred, so that the phase interface could easily be observed and photographed.

As shown in Fig.s 33, 34 and 35, a clear phase interface was noted and
the phase interface curvature seems not to be altered by the die pressure (note that photograph 33 was taken at a die pressure of 600 psi and photograph 34 at a die pressure of 2,000 psi). Also, a steady phase interface was observed during the experiment.
6. Conclusions

Based upon the experimental results and observations in this investigation, the following conclusions are drawn.

A. The die pressure and temperature were found to be important parameters in determining the extrudates' properties, and orientation of the melt molecules was also found to depend very much on the die pressure.

B. A higher die pressure was reached by a combination of increasing the extrusion rate and decreasing the die temperature.

C. A streamline flow pattern was observed by using this side-view die, and it seems to not be altered by variations in the die pressure and temperature.

D. Shear flow existed throughout the trilayer of coextrusion in the reservoir section. This indicated that the shear stress between the core polymer melt and the die walls were not completely restricted to the skin layer.

E. A steady interface was observed.
7. Recommendations

These suggestions are recommended for further study.

A. From the convenient operation point of view, this current side viewing die is too heavy. For future die design, this should be taken into account.

B. The cooling chamber consists of two separate sections which often leaked along their interface. A one piece cooling chamber is suggested for future use.

C. In high die pressure operation (pressure > 3,500 psi), retightening the die is essential to avoid die leakage. The retightening should be done after the system has reached the desired temperature profile and before extruding through it.

D. The take-up device was not used throughout this experiment, partially because the speed of the device was hard to control uniformly. Improved speed control of the take-up device is suggested for future use.

E. According to the results obtained in this study, the die pressure effect on the extrudates' properties is profound up to an achievable pressure of 5,500 psi. This implies that the ultimate orientation of the polymer melt may not have been completely reached at this high die pressure. If possible, die pressures higher than 5,500 psi are suggested to be used to test the dependence of orientation of the polymer melt on higher die pressures.
F. In the coextrusion system, shear flow was found to exist in the core region at the ratio of the core polymer to the skin polymer equal to 9.0. In order to evaluate the effect of the ratios of core polymer to skin polymer on shear flow, more work needs to be done on this part. Specifically, this ratio should be varied and the dependence of birefringence on these ratios is measured.

G. In this study, the flow pattern was observed to apparently have no dependence on the die pressure and temperature. This conclusion is limited to a narrow pressure range, from 2,000-3,000 psi. A wider die pressure range is suggested to further determine if a pressure dependence exists and if so to characterize it.

H. The investigator strongly recommends the use of inserts having different convergence angles at the same operation conditions to seek the dependence of orientation of the polymer melt on those different inserts.
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