THE FLOW STABILITY OF LINEAR LOW-DENSITY POLYETHYLENE
AT POLYMER AND METAL INTERFACES

by

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(ABSTRACT)

The role of the single component instability of surface melt fracture on the interface behavior in stratified bicomponent flow has been examined. First, the factors and conditions leading to the onset of surface melt fracture in linear low-density polyethylene (LLDPE) were identified using fluoro-elastomer (FE) as a blending additive and as a die coating in two visualization dies. A visualization die was constructed so that subsequent experiments examining the joining flow behavior of two stratified flows could be examined. Experiments were conducted in the joining flow die over a range of upstream conditions corresponding to surface melt fracture behavior and the resulting flow birefringence patterns and the interface of the extrudate were examined. It was determined from the single component studies that the role of FE in eliminating surface melt fracture behavior for LLDPE was to introduce slip at the melt/metal interface in the dies. Additionally, it was determined that the coupling of a critical stress with a critical acceleration of the melt as it exits the die, suggested by Kurtz [19], was an accurate description of the behavior observed experimentally. Under upstream conditions corresponding to surface melt fracture behavior, no irregular distortions were observed in the bicomponent interface. It was therefore concluded that the single component instability of surface melt fracture does not play a role in irregular distortions of the interface.

Numerical simulations employing the Phan-Thien Tanner (PTT) constitutive model and the finite element method (FEM) were conducted to examine the influence of relaxation times and extensional viscosity on the developing flow region in joining flow die. Numerical predictions employing material constants fit to the rheological properties of LLDPE were compared with the ex-
Experimental results to establish the reliability of the numerical method. Qualitative agreement between the predictions and the experimental behavior was observed. However, the magnitude of the stresses predicted by the model were not quantitatively accurate. It was concluded that the numerical method was capable of predicting trends in behavior, but was not quantitatively accurate. Given this limitation, it was suggested by the results of the numerical studies that the relaxation behavior has a pronounced effect on the developing stress field, while the impact of the extensional viscosity is minimal. Simulations were also performed to evaluate the 'stick-slip' behavior of LLDPE. The results provided additional support to the supposition of the role of FE in eliminating surface melt fracture behavior in LLDPE.
to my father
Once in a while
you get shown the light
in the strangest of places
if you look at it right.

from *Scarlet Begonias*

by R. Hunter & J. Garcia
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Original Contributions

The author considers the following as his original contributions:

1. The design of an experimental die for performing flow visualization experiments on the joining flow behavior of two polymer melts.

2. The finding that the role of fluoro-elastomer in eliminating surface melt fracture in LLDPE is to introduce slip at the melt/metal interface. This impacts both the critical stress and critical acceleration in the mechanism for surface melt fracture proposed by Kurtz [19].

3. The finding that the single component instability of surface melt fracture does not lead to irregular distortions in the bicomponent interface.
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1.0 INTRODUCTION

The primary goal of polymer scientists and engineers is to develop materials which meet the needs of the consumer. Due to the impracticality, if not impossibility of developing a single polymer which exhibits all of the desired qualities, scientists have become adept at modifying existing polymers to meet the required demands. The blending of polymers, as well as incorporating fillers, additives, stabilizers, colorants and processing aids has become so common that one would be hard pressed to find a product that is 'pure polymer'. Even with the advanced state of blending and copolymerization technology, there are still limits to the extent that a single system can be modified to meet the physical and chemical requirements.

The limitations of single polymer systems were addressed in the 1950's by incorporating multiple layers of different materials into a single composite. Films of different materials with specific properties were bonded together to form a single laminate. While this technique was successful, it required additional handling and processing of the films after they were extruded. In the early 1960's, coextrusion technology started to develop and eliminated the post processing lamination step. In coextrusion, different layers of materials are brought together while in the melt state to form multi-layer films when cooled. The application of this technology over the last 25 years has grown considerably. One of the most common applications is in the packaging industry, where a combination of both barrier properties and physical strength is required. Typical of most technol-
ogies, the application of the coextrusion process has increased at a much faster rate than the the basic understanding of the process. It is the goal of the experimental research objectives in the current work, to help extend the basic knowledge of the coextrusion process.

There are two common methods currently in use for producing coextruded sheet or film. The first is blown film, and the second is cast sheet. The blown-film process is the most common method for producing thin polymeric films. This technique involves extruding the melt, either single layer or multi-layer, through an annular die to form a tube. This tube is then drawn in the machine direction, while simultaneously being inflated, causing it to extend in the transverse or hoop direction. This method is capable of forming films (single-layer) less than 1 mil in thickness. The second common method for producing films is flat die extrusion. As the name implies, the polymer is extruded through a thin, wide, flat die where it is then drawn in the machine direction at different rates depending on the desired thickness. This method, while not capable of the extrusion rates of blown film, is capable of producing thicker gage films.

The coextrusion process commonly involves bringing two or more polymer flows together in a stratified laminar flow field prior to exiting the die, irrespective of whether it is an annular die for blown film or a flat die for cast film. In any extrusion process, there is a rearrangement of the flow field between the exit of the extruder and the exit of the die. A melt pool or reservoir is typically formed at the end of the extruder or the beginning of the die to dampen the periodic discharge from the screw. The material passing through the pool is then channeled in such a way as to provide uniform distribution across/around the die face. The fluid is then typically accelerated through a contraction of some kind prior to exiting the die. The question then is: where is the best place to bring the two or more layers of fluid together? In the discussion that follows, the focus will be on flat die geometries but may be readily extended to annular configurations.

Early coextrusion dies brought the layers together just upstream of the die exit. This resulted in a multi-manifold configuration shown in Fig. 1. The transition of the flow from the pool to the wide, flat exit configuration was accomplished independently for each material in its own manifold

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prior to joining the layers. As a result, an easy transition was made from single layer to multi-layer flows with a minimum of rheological concerns. Despite its success, the complexity involved in machining multi-manifold dies for more than a few layers provided the impetus to develop a method capable of joining a large number of layers in a single configuration.

The approach used to resolve the limitations of the multi-manifold dies was to bring the materials together in a feed-block and then feed the superposed fluids into a single manifold for distribution across the die. The feed-block method did indeed allow for the production of films with a large number of layers (over 100), unfortunately this technique has its own problems and limitations. The transitional flow in the manifold is quite complex. In the transition channel, the materials experience simultaneously shearing and extensional forces. Unlike the multi-manifold dies, where the different layers are in contact for only a short period with a minimum of flow rearrangement, in the feed block design the materials are in contact well ahead of the die exit and are also required to undergo a complex rearrangement. As a result, the rheological properties of the individual layers play a significant role in the behavior of the film in this transition region. Unlike the multi-manifold dies, where success is realized even for fluids with vastly different viscosities, the quality of the films produced using the feed-block configuration is highly dependent on the relative rheological properties of the different layers. Consideration of what rheological properties are important to the success of coextruding different materials in the different geometries is quite important. This question leads to the objectives of the experimental portion of the present research.

The experimental objectives which are to be addressed in the current research are concerned with the interfacial stability where the two different layers join together (Fig. 2). Prior to introducing what rheological properties are expected to play a role in the interfacial stability, the type of instabilities which are of primary interest need to be defined.

Two types of instabilities are common in bicomponent flow. The first and most commonly studied instability is called ‘viscous encapsulation’ or ‘wrap around’. This phenomenon initially involves the curvature of a flat interface and then a slow encapsulation of the more viscous material
Figure 1. Die Configurations: (a) multi-manifold, (b) feedblock
by the less viscous material (Fig. 3). This behavior has been explained by a two step mechanism. The initial convex curvature developed by the more viscous phase is a means to balance the pressure gradients required to drive the two flows. This distortion of the interface occurs rapidly, usually in 1 or 2 \( L/D \) 's upon entering the channel. The second step, which is the actual 'encapsulation', is the result of a driving force of the flow toward the configuration which requires the least amount of energy to maintain it. This preferred configuration has the less viscous material in the regions of highest shear, i.e. the sheath-core configuration in capillary flow. While this type of instability is not of primary interest in the proposed study, its influence on the observed behavior of the interface for materials with different viscosities can not be ignored.

The second type of instability found in coextrusion is that of an irregular interface. Unlike viscous encapsulation, where there is a migration of the interface while remaining smooth, the distorted interface may maintain a constant average position, but severe distortions along the machine and/or transverse directions occur (Fig. 4). Little work, experimental in nature, has been reported concerning this behavior. Speculation as to the mechanism leading to this instability has related it to the 'melt fracture' behavior observed in single component flows. Unfortunately, 'melt fracture' itself is not well understood and is still a topic of debate in the literature. It is this type of instability which will be the focus of the current experimental work.

In order to determine what rheological properties are important for a particular geometry, the kinematics associated with the geometry must be determined. Both the multi-manifold and the feed block configurations involve accelerating the fluids as they join together. The fluids go from a zero velocity at the walls to a finite velocity at the interface. In the feed block, the materials are further accelerated/decelerated as the transition from a tall narrow geometry to a short wide geometry is realized. Extensional properties of the fluids will then play a significant role in the behavior of the system. Furthermore, in order to maintain a stable interface as the fluids flow side by side through the die, continuity of the velocity and the total stress of the fluids at the interface must be maintained. While there is a relatively short contact time in multi-manifold systems, in the feed block geometry the materials experience this phenomenon for an extended period of time. As a result,
Figure 3. Viscous Encapsulation
the importance of the shear properties, such as the extra normal stresses of the fluids, need to be considered. A thorough characterization of the materials' extensional and shear properties is paramount if experiments and numerical simulations of bicomponent flows are to be of any value.

Given the rheological properties of two fluids, it would be of obvious benefit to be able to predict whether or not the materials would form a compatible or stable bicomponent flow. In order to accomplish this, the behavior of materials with different rheological properties in a joining flow geometry needs to be examined. The simulation of the joining flow behavior of materials with different rheological characteristics is the numerical objective of the current research.

The success of any numerical simulation depends on its ability to accurately predict real behavior. The first step toward this end in viscoelastic simulations is the appropriate choice of a constitutive equation. The upper-convected Maxwell (UCM) model is the most commonly used constitutive equation which exhibits the desired fading-memory, viscoelastic behavior. The model predicts a Newtonian viscosity behavior in shear and exhibits singular stress behavior at a critical rate in extension. Unfortunately, few fluids are adequately described by this behavior. An alternative to the UCM is the Phan-Thien Tanner (PTT) model. The PTT model incorporates the desired fading memory aspects of the UCM model without the singular extensional behavior and in addition, provides the capability to adjust the extensional behavior of the fluid apart from the shear behavior. In the current study, it is the PTT model which will used to describe the rheological behavior of the fluids.

Consideration of previous experimental investigations of multi-layer extrusion behavior shows that the majority of efforts have been directed toward understanding the viscous encapsulation behavior, and relatively little work has been directed toward irregular interface distortions. Additionally, simulations of multi-layer flows have failed to address the influence of extensional behavior as materials are brought together and accelerated in a single flow channel. It is the purpose of the current study to address two basic areas of understanding in the coextrusion process. First, to determine if the single component instability of surface melt fracture plays a role in irregular interface
distortions in multi-layer flows and second, to examine the effect of bounded versus unbounded extensional stress growth behavior on the stress development in the joining flow geometry. The first objective requires an investigation of the factors leading to the surface melt fracture behavior in single component flows, prior to determining if this behavior carries over into multi-layer flows. The second objective will be addressed from both an experimental and numerical tact. Experimentally, it is practically impossible to vary the extensional behavior of a melt independently of other rheological properties. As a result, other factors affecting the behavior of different melts need to be considered. Numerical simulations incorporating the PTT constitutive model do allow for isolating the extensional behavior apart from other rheological properties and should provide valuable insight to stress development in the joining flow geometry. The need to compare the predictions of the simulations with experimentally observed behavior is of obvious concern and will be included in the present study.
2.0 LITERATURE REVIEW

The experimental objective of the current research is to examine the interfacial behavior of two polymeric fluids as they join together to form a stratified bicomponent flow. Any investigation of the interfacial stability in bicomponent flows requires an understanding of first, the type of instability observed and second, the factors which can influence the observed stable/unstable behavior. In section 2.1, the interfacial instability referred to as 'viscous encapsulation' or 'wrap-around', which is associated with a mismatch in the viscosities of the two fluids will be examined. Corrugated or wavy type interface instabilities will then be considered in section 2.2. These instabilities have been associated with a difference in normal stress properties [1,2] and the single component instability referred to as melt fracture [3]. It is this type of instability which is of primary interest in the proposed investigation. Heuristic reasoning suggests that factors which influence the stability of single component flows (e.g., vortex formation on melt fracture) may well have an impact on the interfacial stability of bicomponent flows. In section 2.3, the phenomena of melt fracture and vortex formation in single component flows will be briefly discussed. The utility of stability analyses in the study of melt fracture behavior will also be discussed. A review of the stability analyses pertaining to the response of the interface in bicomponent flows to periodic disturbances will be given in section 2.4.
The numerical objective of the present research is to examine the influence of differing rheological properties on the joining flow behavior of viscoelastic fluids. The problems encountered in viscoelastic simulations have been well documented [4] and the pertinent details are reviewed in section 2.5. Particular attention will be given to the methods used in solving high Deborah number flow problems with boundary singularities. In the last section of this chapter, the research objectives of this thesis are clearly stated and the approach taken to meet them defined.

2.1 Visco Encapsulation

While the phenomenon of viscous encapsulation is not a primary focus of the current research, any investigation of the interfacial stability in bicomponent flows must consider the effect of the two fluids having different viscosities. The phenomenon of 'wrap around' is best described by a smooth distortion of the interface between the two fluids and the subsequent migration of one of the fluids to a configuration where it fully envelops the other. This phenomenon is currently accepted to be a result of a mismatch in viscosity of the two materials. This mismatch results in a drive toward a minimum energy or dissipation configuration, with the more viscous material in the regions of lowest shear, i.e. the less viscous material encapsulating the more viscous material in capillary flow. Key investigations leading to the explanation of this behavior are discussed in this section. Observations made during the study of this phenomenon which are pertinent to the current research are highlighted.

2.1.1 Effect of Viscosity Ratio

Initial investigations into the encapsulation behavior were motivated by its importance to the fiber spinning industry, where it was desired that the materials remain in a side-by-side rather than
a sheath-core configuration (Fig. 5). Experimental studies showed that the encapsulating behavior was present in both planar [5-7] and pipe/capillary [5,6,8,9] geometries. Southern and Ballman [10] investigated the effect of differing viscosities in capillary flow by examining the interfacial behavior of two polystyrenes (PS-A & PS-B) which exhibited a cross-over in their viscosities as a function of shear rate. At low shear rates ($\dot{\gamma} < 12$), PS-B had a higher viscosity than PS-A, while at higher shear rates ($\dot{\gamma} > 12$) PS-A had the higher viscosity. The two materials exhibited common die swell ratios over the range of experiments and as a result, the influence of elastic differences between the two materials was assumed to be negligible. By varying the total flow rate (wall shear rate), they were able to demonstrate that the less viscous material (depending on shear rate) tended to encapsulate the more viscous material. Experiments conducted at low flow rates, where PS-B had the higher viscosity, showed that the PS-B interface was convex and that the contact points of the interface with the wall had moved such that PS-A was preferentially wetting the capillary wall. When the experiments were performed at higher shear rates, where PS-A exhibited the higher viscosity, similar results were observed for the interface, but with the roles of the two fluids being reversed. The results of Southern and Ballman clearly demonstrated the importance of viscosity ratio on the interface configuration in bicomponent flows. Subsequent work by these investigators [2] highlighted more dramatically the role of viscosity ratio on the interface shape. Using two polystyrenes similar to those used in the earlier investigation, they found that near the cross-over point a curvate cycloid interface configuration was achieved (Fig. 6). In the regions of highest shear near the wall, PS-B with the lower local viscosity moves to preferentially wet the walls and encapsulate the locally higher viscosity PS-A. However, in the region of low shear near the center of the capillary, PS-A, which has the locally lower viscosity, has moved to encapsulate the PS-B material. These results unequivocally demonstrate the effect of viscosity ratio on the encapsulation phenomenon. Similar results have been reported in slit flow geometries by Han [7] and in capillary flows by Han [5] and Everage [9].
Figure 5. Side by Side and Sheath Core Configurations
Figure 6. Effect of Viscosity Cross-Over on the Interface [2]: (a) $\gamma = 149.5\text{s}^{-1}$, (b) $\gamma = 74.5\text{s}^{-1}$
2.1.2 Effect of L/D

The initial work of Southern and Ballman provided a great deal of insight into the behavior of two fluids with a non-unity viscosity ratio, as demonstrated above. A major limitation of their initial work however, was the use of a single capillary L/D for their experiments. Their experiments clearly indicated trends in the transient interface configuration behavior, but did not provide any specific measure of the final/steady-state configuration. Attempts to address this limitation were presented in their subsequent work [2], as well as by Han [5], White and Lee [6] and Everage [8,9]. Only the work of Everage was successful in attaining steady-state profiles. His initial investigation involved the study of two different bicomponent systems: a nylon/nylon system and a xylene/water system. Both systems had a viscosity ratio of approximately 6:1 and achieved a sheath-core steady-state configuration. However, the axial distance required to achieve these configurations were substantially different. The nylon/nylon system with viscosities on the order of 10⁵ poise required 120 L/D's, while the xylene/water system with viscosities of the order on 10⁻² poise required only 3 L/D's. No explanation for the drastic differences in L/D's required for steady-state configurations was presented. It is apparent from Everage's results and other investigations that a mismatch in the viscosities of the two fluids provides the driving force for the encapsulation phenomenon. However, the magnitude of the driving force is not simply a function of the viscosity ratio of the two fluids but is dependent also on the actual magnitudes of the respective viscosities.

2.1.3 Additional Observations

Prior to moving on to a discussion of proposed mechanisms explaining the observed viscous encapsulation phenomenon, there are two points not previously discussed in this section which need to be addressed. First is the effect of the elasticity difference or ratio of the two fluids on the behavior of the bicomponent interface and second is the phenomenon of extrudate bending for
superposed fluids exiting a die. So far, the discussion of encapsulation has centered on the influence of viscosity ratio. This is the result of '20/20 vision' regarding what is currently known regarding this phenomenon. Initial studies on this behavior did not focus solely on viscous behavior, but also considered elastic effects [1,2,5]. The results of these studies showed that elasticity differences played an insignificant role in the encapsulation phenomenon, but did result in irregular distortions along the interface. This behavior is more closely related to the type of interface instabilities discussed in the next section and will be discussed there in detail accordingly. The phenomenon of extrudate bending has been observed in both capillary [2,9] and slit die [10] coextrusion studies. As the superposed fluids exit the die, the extrudate bends in the direction of the more viscous component. This phenomenon is addressed in the proposed mechanism of Southern and Ballman in the following paragraphs and has direct implications on the concepts of recoverable shear strain discussed in the melt fracture literature (section 2.3).

### 2.1.4 Mechanisms for Viscous Encapsulation

In an attempt to explain the encapsulation phenomenon, MacLean [11] used a variational approach to analyze the interface shape between two power-law fluids based on minimizing viscous dissipation. Two configurations were considered in both slit and capillary geometries to determine which was energetically preferred. The procedure was to assume an interface, solve for the velocity field and evaluate the entropy production or viscous dissipation of the system. The first case considered in the slit-die geometry was a single interface with each material contacting one boundary of the die. The second configuration consisted of layers of the less viscous material contacting both boundaries and a layer of the more viscous material between them, resulting in two interfaces. A comparison of the two configurations showed that the second, with three layers rather than two, was energetically preferred. Configurations considered for the pipe flow geometry were the materials side-by-side with a flat interface compared with the less viscous material fully encapsulating the more viscous fluid in a sheath-core configuration. Results of this comparison showed that the
sheath-core configuration was energetically preferred. Everage [8] extended the work of MacLean for capillary flow by considering four interface configurations using a Newtonian model. In addition to the two configurations studied by MacLean for tube flow, an interface in the form of an arc with a radius \( R_I \) equal to \( 1.5R_o \) (tube radius) and a circular interface \( (R_I = 0.569R_o) \) tangent to the tube wall were studied. The results of Everage’s study confirmed that of MacLean in that the sheath-core configuration was the energetically preferred case.

There are clear limitations to the work presented by MacLean and Everage. First, only special cases of interface configurations are considered. Analyses of configurations which are expected to behave poorly, such as a sheath-core configuration with the less viscous material at the center or the three layer case with the more viscous material at the boundaries are not considered. While it is expected that these would prove unfavorable, for the sake of completeness they should be considered as a limiting ‘worst case’. Recently, Joseph, Nguyen and Beavers [12] have been able to generate flows which violate the minimum dissipation principle. While the conditions required to generate these flows (a large surface tension contribution) are well outside the range of conditions encountered in polymer flows, it is necessary to point out that the viscous dissipation principle is not universally applicable. Second, the work of MacLean and Everage applies to a comparison of steady-state configurations only and does not treat the transient behavior known to exist for the case of two materials flowing side-by-side in a pipe.

In an attempt to explain the transient behavior of the encapsulation phenomenon, Southern and Ballman [2] proposed a two step mechanism. First, the interface curved to achieve a balance of the pressure gradients required to drive the flow in each layer. This step was considered to occur very rapidly (in less than \( 2L/D's \)) and reversibly and was responsible for the bending of the extrudate. The second step involved the irreversible migration of the lower viscosity material to encapsulate the higher viscosity material to achieve a more energetically preferred state. This step was perceived to be very slow, requiring a great deal of time (greater than the \( 60L/D's \) available). Everage [9] demonstrated the irreversible nature of this second step by its effect on the observed exit angle (degree of bending). He was able to increase the \( L/D \) in his experiments beyond the point.
of achieving the steady-state sheath-core configuration. The loss of exit angle (reversible deformation) as the less viscous material moves to preferentially wet the walls (irreversible deformation) is clearly indicated in Fig. 7. The results of Everage indicate that the two step mechanism proposed by Southern and Ballman is valid and probably is in fact what happens. The problem with the proposed mechanism is that it is purely qualitative and does not provide any insight to the capillary lengths required for either step.

White and Lee [6] presented an analytical development of the movement of the interface of two fluids between parallel plates and the distortion of the interface between two fluids side-by-side in a pipe. The driving force in both cases was the imbalance of the pressure gradients required to maintain equal flow rates of fluids with different viscosities through identical geometries. This driving force was proportional to the difference in the gradients and was opposed by viscous effects in the fluids. A differential equation was posed in these terms and a solution, exponential in form, was found. The results for the slit geometry indicated a movement of the interface toward the less viscous component with no distortion in the direction transverse to the flow direction. Results of the pipe flow geometry predicted a curving of the interface, but with no movement of the contact points at the wall (the interface showed no preferential wetting). This analysis is consistent with the first phase of the two-step mechanism suggested by Southern and Ballman [2] and Everage [9]. Additionally the importance of the magnitudes of the viscosity, as observed by Everage, could be taken into account. However, it does not account for the encapsulation of the more viscous phase observed by previous investigators.

There has been no explanation offered in the literature for any quantitative mechanism for the second step in the viscous encapsulation process. Work in this area has tapered off and is no longer a popular topic in current literature. More recently, the experimental work presented in the literature on bicomponent flows has focused on the behavior of the fluids flowing through contractional geometries. These flows result in instabilities that are quite different from the encapsulation phenomenon discussed in this section and are more closely related to the types of instabilities associated
Figure 7. Exit Angle versus L/D [9].
with single component flows. This type of behavior is discussed in the next section of this chapter along with the instabilities associated with elasticity differences.

2.1.5 Conclusions

It is clear from the discussion in this section that the response of the interface in bicomponent flows to mismatched viscosities will have an effect on its shape. The contact lengths which will be examined in the present research are relatively short (less than 10 \(L/D\)'s) and as a result, the preferential wetting associated with second step of the encapsulation phenomenon should not be a major factor. However, as indicated by the work of Southern and Ballman [2] and Everage [9], the smooth distortion of the interface associated with the first step of the encapsulation behavior occurs in less than 3 \(L/D\)'s even for highly viscous fluids. This result has a direct impact on the proposed investigation. The two materials incorporated in the study have similar viscosities at low shear rates, but exhibit different degrees of shear-thinning behavior within the range of shear rates being examined. As a result, while encapsulation is not the phenomenon of interest, its impact on the observed interface behavior will have to be accounted for in the final analysis.

2.2 Irregular Interface Distortions in Bicomponent Flow

The primary experimental objective of the present research is to examine the interfacial stability in the joining flow of two non-Newtonian fluids. The type of instability which is of interest, manifests itself in the form of wavy or irregular distortions shown in Fig. 4. Unlike the encapsulation instability discussed in the previous section, where the interface remains smooth and is a result of viscosity mismatch, the instability considered in this section results in irregular distortions of the interface and is not well understood. This type of instability has been observed in both capillary
[2] and planar [3, 5] geometries and has been attributed to a difference in the elasticities of the two fluids [2, 5] and to a critical interfacial shear stress [3], similar to the phenomenon of melt fracture observed in single component flows. The limited literature dealing with this instability is discussed in this section. Additionally, the work of Binding, et. al. [13], on the observed behavior of dissimilar polymer solutions (e.g., elastic with inelastic) will also be discussed.

2.2.1 Elastic Effects in Polymer Melts

The effect of elasticity on the shape of the interface in bicomponent flows was initially studied by White and co-workers [1]. As alluded to in the previous section, this study was motivated by the desire to determine the role of elasticity on the encapsulation phenomenon. The authors presented a theoretical analysis of the flow of two non-linear viscoelastic fluids in stratified Poiseuille flow for both parallel plates and tubes. The analysis was limited to materials with the same viscosity functions, but different elasticities. The initial shape of the interface in both cases was assumed to be flat. The equations of motion for the two fluids were solved and the stresses at the interface were examined. In the case of tube flow, it was determined that the influence of the second normal stress difference ($N_2$) caused each fluid to protrude into the other at decreasing values of the radial position along the interface. This indicated that the material with larger absolute $N_2$ will be the one to protrude into the other. The authors do not present any information on the relative magnitude of $N_2$ effects compared with viscosity ratio effects. Due to the relatively small magnitude of $N_2$ compared with the shear stress or the first normal stress difference, it would be expected that any affects associated with differences in $N_2$ between two materials would be small compared to viscosity difference affects.

The accompanying experimental results presented with the theoretical work of the White and co-workers was in the form of flow visualization studies in a capillary. A single binary system composed of low-density polyethylene (LDPE) and atactic polystyrene (PS) was examined. For
the rates studied, no conclusive results were obtained for the shape of the interface. This indicates that if there is indeed an effect on the interface from a difference in $N_1$ of the materials, it is quite small. Han [5], in a study of PS/LDPE and PS/HDPE bicomponent systems, concluded that the effect elasticity differences between the two phases was to cause a corrugated instability at the interface. The only evidence that was presented for this statement was a picture of cross-sectioned extrudate taken from a slit die, showing a corrugated interface. Unfortunately, while normal stress - shear rate data are given, there is a cross-over of the $N_1$ behavior of the two materials, and the conditions experienced for the pictured extrudate are not given (the magnitude of the elastic difference is unknown).

The best evidence for the role played by elasticity in affecting the interface of bicomponent flows was given by Southern and Ballman [2]. The authors used two polyethylenes having similar viscosities, but significantly different primary normal stress differences (Fig. 8). As shown in Fig. 8, the elasticity difference has a dramatic effect on what was initially a smooth flat interface. Furthermore, the distortions appear to be worse in regions of higher shear as shown near the wall. The authors presented no explanation for this behavior nor have any been given since their work.

2.2.2 Elastic Effects in Polymer Solutions

Further work on the effect of elasticity has been concerned primarily with solutions, and as a result cross-sectioning of the emerging extrudate has not been possible. This has limited observation techniques to flow visualization studies. The most significant work on the bicomponent flow of solutions has been performed by Binding [14] and Binding with co-workers [13]. Binding's initial studies examined the behavior of three types of solutions in two different planar geometries. The first fluid was a Newtonian solution (N) of water and maltose syrup. The second solution was a constant viscosity, elastic Boger (B) fluid made up of polyacrylamide in maltose and water. The last fluid used was a shear-thinning elastic fluid (P). The two geometries used in the study were a
Figure 8. Influence of Elasticity Mismatch on the Interface [2].
4:1 standard planar contraction and a 4:1 T contraction (Fig. 9). Binding first examined the behavior for each fluid individually in both geometries. His results for the standard 4:1 contraction showed that the P fluid exhibited strong vortices in the corners at the exit, while both the N and B fluids exhibited only small stagnant regions in the corners. For the T geometry, no vortices were observed for the Newtonian fluid or for the Boger fluid when the inlet flow rates in both arms were equal. Vortices were always observed in both branches of the T at the lip of the contraction for the P fluid, regardless of relative flow rates in either arm. A vortex was observed to form in the lower Reynolds number branch of the T when Boger fluids were fed at different rates in each arm.

Studies using different test fluids together in the two geometries were limited to Newtonian-Boger combinations. Due to the difference in densities between the P fluid and either the Newtonian or Boger fluids, Binding was unable to generate the desired stratified behavior for these combinations. However, startling results were obtained using the B/N combination. In the standard contraction, large vortices were observed in the Boger fluid at low Reynolds number flows. This is surprising considering that neither the Newtonian or Boger fluid was observed to form vortices independently in this geometry. At higher flow rates, the vortices in the Boger fluid disappeared, but an asymmetry in the flow field was still observed. This is again in contrast to the expected behavior of vortices in single component flows, where vortex behavior is observed to increase with flow rate (see section 2.3). In the T contraction, Binding was able to generate vortices in either the Newtonian branch or the Boger branch depending on conditions, but not in both. As a result of these observations, Binding concluded that the behavior of the individual components alone was not sufficient to predict the interfacial behavior in bicomponent flows. The boundary conditions at the interface were seen to play a significant role in determining the patterns of the flow field.

Binding and co-workers [13] extended Binding's previous work by examining two additional geometries as well as the planar T geometry. An axisymmetric contraction as well a 4:1 planar step contraction were considered (Fig. 10). The results of the T contraction in this study confirmed the previous observations of Binding. Single component studies for the axisymmetric geometry showed
Figure 9. Standard 4:1 Planar and 4:1 T Constructions [14].
Figure 10. Planar Step and Axisymmetric Contractions [13].
that the Boger fluid formed vortices in this geometry, while the Newtonian fluid did not. Similar results were observed for the Newtonian fluid in the planar step geometry while the Boger fluid was observed to form or not form vortices depending on conditions of the flow. These results are in agreement with the observed behavior of single component flows discussed in section 2.3. Similar to the previous study by Binding, Binding and co-workers were able to generate vortices in the Newtonian fluid and in the Boger fluid where single component studies of these fluids were not able to do so. The results of these studies further substantiate the conclusion of Binding that the boundary conditions at the interface of bicomponent flows play at least as significant role in the behavior of the flow as the rheological properties of the individual components.

2.2.3 Melt Fracture Instabilities at the Interface

Unlike the previous investigations discussed in this section, the last study to be considered was not motivated by a desire to ascertain the role of elasticity in the instability of bicomponent interfaces. The instabilities to be studied had been observed in the coextrusion of multi-layer sheet and were observed in the form of planar waves (Fig. 4). A discussion of the investigation of this type of instability follows.

Schrenk and Alfrey [3] investigated the onset of interfacial instability in an ABS-HIPS-ABS system. The instability that was observed was in the form of long waves and the onset was determined qualitatively by eye. The apparatus consisted of a three-layer feedblock and a single-manifold die. The layers join after exiting the feedblock and are subsequently spread and thinned in the manifold prior to exiting through the land section of the die. Four variables, including total flow rate, layer thickness ratio, skin-layer viscosity and die gap, were found to have the most influence on the stability of the interface. Further results showed that the instability was initiated in the land section of the die. The authors hypothesized that the interface instability was analogous to the type of melt fracture associated with gross distortion behavior (section 2.3) and calculated a
critical shear-stress value of 0.5 MPa based on a power-law viscosity model. This was about half of the value reported for the onset of melt fracture in polyethylene. The determination of the land section of the die as the site initiating the instability is not conclusive. This determination was made based on blocking the die exit, cooling the system and sectioning the material inside the die. Observation of the presence of the instability in the land section by itself is insufficient to determine the site of initiation. The entry region to the die-land could well be the site of initiation, with the growth of the waves not observable until the land section.

2.2.4 Conclusions

The conclusions of Binding and co-workers have direct implications on the current experimental study. In addition to the factors leading to observed single component instabilities, such as rheological properties and geometric singularities, the influence of the boundary conditions at the interface will have to be considered. Additionally, discontinuities in rheological properties at the interface will have to be accounted for.

Further consideration of the literature discussed in this section reveals one undeniable fact: the behavior of the bicomponent interface, whether considering solutions or melts, is inextricably tied to the types of instabilities associated with single component flows. It has also been demonstrated that an understanding of the single component behavior alone does not suffice in the explanation of bicomponent flows, but it does provide the necessary foundation. With this in mind, the next section of this chapter will deal specifically with the types of single component instabilities which are expected to play a role in the present investigation.
2.3 Melt Fracture

In the previous section, the phenomenon of melt fracture was alluded to in connection with the interfacial instability observed by Schrenk and Alfrey [3]. The term melt fracture has been loosely applied in the literature to cover three different types of extrudate irregularities of varying degrees of severity. Surface melt fracture, also called matte or sharkskin, is characterized by fine scale surface irregularities, typically less than one-tenth the extrudate diameter, during steady pressure flow conditions. Spurt or slip-stick fracture is described by oscillating pressure gradients in constant velocity experiments or by fluctuating mass flow rates under constant pressure conditions. Associated with this behavior is a discontinuity in the flow curve for the material. Finally, gross melt fracture is characterized by severe distortions in the extrudate, typically of the same order as the extrudate itself. While the extrudate may be severely distorted, it may still exhibit a glossy finish, free of fine-scale surface irregularities. In this section, a review of surface and gross melt fracture behavior and the implications for bi-component behavior will be discussed.

Surface fracture has been attributed to both the exit region [15-19] and the land region [21-23] of the die. Additionally, surface fracture is exhibited by relatively few polymers and the behavior is observed prior to the onset gross fracture. Conversely, gross fracture has been observed to occur in a broad range of polymers [24-27] and has been associated with flow disturbances initially observed in the entry region of the die. An excellent introduction to melt fracture behavior, as well as a comprehensive review of the early experimental work in this area has been presented by Tordella [16]. Dennison [17] presented a more recent discussion dealing specifically with the sharkskin phenomenon, while Petrie and Denn [15] have updated Tordella’s review and have incorporated the use of stability analyses.

The intent of this section is not to duplicate existing reviews of melt fracture, but rather to highlight the pertinent portions and to incorporate the more recent work which has been presented.
in the literature. Specifically, the melt fracture behavior of low-density polyethylene (LDPE), high-density polyethylene (HDPE) and linear low-density polyethylene (LLDPE) are considered. Molecular considerations and possible mechanisms are also addressed in the current section. Finally, the implications of single component melt fracture behavior on bi-component instability are discussed.

2.3.1 LDPE - Experimental

Low-density polyethylene (LDPE) does not undergo surface fracture, but rather a direct transition from smooth extrudate to gross fracture behavior is observed. Prior to the onset of gross fracture in LDPE, vortices are observed to form in the entry region of the die, creating a symmetric 'wine-glass' entry pattern. This phenomenon is discussed in detail at the end of this section, but the point of interest here, is the behavior of the recirculating regions during gross fracture. At the onset of fracture, the symmetry of the entry flow is lost and a periodic surging of material from the recirculating vortices into the primary flow at the lip of the entrance occurs. This behavior has been observed during flow visualization studies [28,29], where material from one of the recirculating regions would surge into the main flow. As the material in one region was depleted and its contribution to the main flow diminished, surging from the adjacent recirculating region would increase to compensate for the loss. While this local surging of the flow was observed, no surging was observed down stream in the main part of the capillary (the land section). Flow birefringence studies further supported the results of visualization experiments in citing the entry region as the point of initiation for gross melt fracture in LDPE. The experiments demonstrated that asymmetries in the stress field first appeared in the entry region and then carried over into the land region.

Studies on the effect of capillary length on gross fracture [15,16] have shown that the severity of the fracture decreases with increased length for LDPE. This supports the visualization and birefringence experiments in citing the entry region as the site of initiation. If the land region was
responsible for the instability, it would be expected that at the very least, the instability would not diminish with land length and more likely, it would increase. Birefringence experiments examining the effect of capillary length [16] have shown that stress levels within the capillary diminish along its length. The reduction in stress with length suggests that relaxation behavior of the material plays an important role in the severity of the fracture, but not necessarily in its initiation.

Investigations of the influence of inlet geometry on gross melt fracture [16] have shown that the severity of fracture in LDPE can be reduced by stream-lining or tapering the entrance region of the die. This again supports the entry region as the most probable site for initiating the instability in LDPE.

2.3.2 Instability: Initiation vs. Propagation

Prior to discussing the observed behavior of HDPE and LLDPE, it is important to stress the difference between the initiation and the propagation of an instability. This point may be well made by considering the behavior of LDPE discussed above. The flow visualization studies showed that the entry region experienced unsteady flow during gross melt fracture, while flow within the capillary itself remained steady. The birefringence studies found that the stresses became unstable in the entry region prior to the development of asymmetric stresses in the capillary itself. Both studies indicate that an instability was present in the entry region when there was none observed in the land region. Only the birefringence study indicated a disturbance in the land section and that was observed after the onset of entry instability. This behavior suggests that, while the instability is present in the entry region, it is damped in the land section, how else could it not be present in the land section. This conclusion is supported by the L/D studies, where the severity of the fracture is observed to decrease with increased land length.
The conclusion that the entry region is the site of initiation of the instability is only valid if it is assumed (there was no evidence given) that the flow prior to entering the entry region was stable. When studying the flow behavior through a die composed of an entry, land and exit section this is a reasonable and necessary assumption. It could be argued legitimately, that the instability was already present prior to entering the die entry region (though indiscernibly) and that the entry region only provided a site for rapid propagation or growth, just as the land section served to provide a region for dampening it. It is not meant to suggest that this is the actual case, but only that it is a possibility, no matter how small.

Currently, there are no methods available for theoretically predicting the initiation of a flow instability, but stability analyses do provide the means for predicting the damping or propagation behavior in particular flows. Introductions to the application of stability analyses with specific applications to polymer flows have been given by Denn [30] and Middleman [31]. Unfortunately, analytical solutions are limited to simple geometries and require the use of simple constitutive equations. Predictions for the response of a system in a mildly complex geometry (a simple contraction) or a material described by a moderately complex constitutive equation require the use of numerical algorithms and as a result, are frequently avoided. The utility of these analyses in terms of melt fracture, however, will be discussed in the following paragraphs, while their application in predicting bi-component flow instabilities is given in section 2.4.

2.3.3 HDPE - Experimental

High-density polyethylene (HDPE), unlike LDPE, exhibits both surface and gross fracture behavior. While the surface fracture behavior has been generally attributed to the exit region of the die [15,17], studies on the gross fracture behavior have led to no such widely accepted site. There is, however, general agreement in the literature that flow within the entry region is stable during gross fracture, suggesting that the entry region be ruled out. The effect of increasing land length on
the severity of gross fracture for HDPE is also contrary to that of LDPE. The severity of gross fracture has been observed [32] to increase with increasing land length suggesting that the land region may be responsible for initiating gross fracture in HDPE. It is appropriate to reiterate the difference between initiation and propagation. While the lack of disturbance in the entry and subsequent increase in severity with increasing land length strongly suggest that it is the land region which is responsible for initiating gross fracture in HDPE, it is possible that the instability is initiated in the entry and only propagated in the land region.

The most controversial point regarding the behavior of HDPE is whether or not a slip mechanism is present at the wall of the land region in the die. Arguments for the presence of slip at the wall are based on flow curves (flow or shear rate vs. shear stress) and Mooney equation plots (Fig. 11). In terms of flow curves, it is argued that when the flow is on the lower branch of the curve, the material adheres to the wall and there is no slip present. When the flow is on the upper branch of the curve, the material is slipping at the wall and the discontinuity in the curve is indicative of the onset of the slip behavior. As reported by Tordella [16], below point A, the extrudate was observed to be stable. Between A and B, surface distortions (matte) were observed in the extrudate. At the discontinuity in the flow curve (B and C), the flow resulted in a 'bamboo' or rippled appearance of the extrudate and was associated with 'stick-slip' behavior. The flow was believed to oscillating between the lower branch (stick) and the upper branch (slip). The Mooney equation plots [33] allow for the determination of the slip velocity at the wall. The apparent shear rate, $\dot{\gamma}_w$, is plotted against the reciprocal of the capillary radius, $1/R$, at constant stress, $\tau_w$. If slip is present, these plots result in a straight line and the intercept is related to the actual velocity gradient at the wall.

Both flow curves and Mooney equation plots provide only indirect evidence for the presence or absence of slip of the material at the wall. As a result, the conclusions drawn from these methods are subject to interpretation. It can be argued in the case of the Mooney plots, that due to the unstable nature of the flow, the simple relations used in developing the theory are not applicable. Furthermore, the direct observation of HDPE during melt fracture by den Otter [26,27] failed to
Figure 11. Mooney Equation Plot [33].
find any evidence of slip. While failure to observe slip does not directly refute the phenomenon, den Otter's success in observing slip for other materials [34] lends strong support to his observations of HDPE. The influence of the die entrance geometry on gross fracture in HDPE was also considered by den Otter. The frequency of the extrudate distortions was observed to be a function of the inlet geometry. This suggests that the entry region of the die does play a role in the gross fracture behavior, but is by no means conclusive.

The question of the initiation site for gross fracture in HDPE is as yet unresolved. Even accepting slip as a real phenomenon, it is not clear whether this behavior is the cause or the result of gross melt fracture. As discussed earlier, the fact that the severity of fracture increases with increasing land length only serves to confirm the land section as a region where the instability can propagate and does not indicate conclusively that this region is the site of initiation. However, the lack of observed disturbances in the entry region for HDPE presents a strong argument for the land region as the initiating site and must be considered.

2.3.4 LLDPE - Experimental

The study of the melt fracture behavior of linear low-density polyethylene (LLDPE) has been a relatively new topic of discussion in the literature. LLDPE is similar to HDPE in that they both exhibit surface and gross fracture behavior, but the emphasis in the literature has centered on the surface fracture behavior of LLDPE. This is understandable considering the prime application of LLDPE in the blown film market, where optical clarity is a primary concern. It is the onset of matte or sharkskin which affects the quality of the films. In the paragraphs that follow, it is the surface fracture behavior of LLDPE which will be considered.

In order to determine the criteria for the onset of surface fracture in LLDPE, Kurtz [19] carried out a set of experiments in which the diameter of the capillary was varied while holding the L/D
ratio constant. The results indicated that, for capillaries less than 1 mm in diameter, the onset of sharkskin was determined by a critical exit velocity ($\gamma D$), and for capillaries greater than 1 mm, a critical shear stress was the determining factor. The critical shear stress required corresponded to the value where the slope changed in the flow curve for the material (\( \pm 0.1 \) MPa). It was concluded that both a critical exit velocity and wall shear stress were required for initiating surface fracture.

Thus, a combination of the land and exit regions was responsible for initiating the surface fracture behavior in LLDPE. These findings were consistent with the earlier studies of HDPE. Unfortunately, only one capillary less than 1 mm was used by Kurtz, and this cannot be overlooked in terms of the reliability of his results citing a critical exit velocity.

An extensive study by Ramamurthy [21] examined the influence of molecular weight, temperature, die diameter, die L/D, die entry angle, and materials of die construction on the surface fracture behavior of LLDPE. The results of the capillary studies indicated that, of the parameters studied, only the materials of construction had any impact on the fracture behavior, and that the influence of materials of construction in capillary studies was weak. The most interesting result of Ramamurthy's capillary studies was the presence of slip velocities in the land region of the die. The results of a Mooney type analysis indicated an onset of slip velocities at a wall shear stress value of 0.10 MPa. This occurred prior to the onset of sharkskin, which was observed to be coincident with the change in slope of the flow curve at 0.14 MPa. This led to the conclusion that slip at the wall was the cause of sharkskin rather than the result of it. The presence of wall slip velocities during surface fracture was later confirmed by Kalika and Demi [22], but the results of their work determined a critical shear stress of 0.23 MPa for sharkskin, significantly higher than that found by Ramamurthy. Both investigations suggested that sharkskin was the result of a failure of adhesion at the melt/metal interface as a result of the high stresses present. This conclusion has direct implications on the most probable sites for initiating surface fracture. As suggested by Cogswell [18] and later by Lipscomb and co-workers [23], the stresses present at boundary singularities, even for Newtonian fluids, are orders of magnitude greater than those in regions of fully developed flow. If wall slip is indeed the cause of surface fracture and results from a failure of adhesion at the
melt/metal due to high stresses, it seems more reasonable that slip would initiate in the entry or the exit rather than in the land region.

The experimental studies of the melt fracture behavior of LLDPE cite the land and/or the exit region for initiating surface melt fracture behavior in LLDPE, eliminating the entry region as a possible site for initiating this behavior. A more detailed discussion of the experiments regarding the surface melt fracture behavior of LLDPE is reserved for the discussion of possible mechanisms explaining this phenomenon. Prior to that discussion, the molecular considerations affecting melt fracture behavior in polyethylenes is considered.

2.3.5 Molecular Considerations in Melt Fracture

When considering the melt fracture behavior of LDPE and HDPE, it is tempting to attribute the difference in behavior to the fact that LDPE is a branched material and HDPE is linear. While similar distinctions hold for the behavior of some materials (linear vs branched polydimethylsiloxanes [26,27]), there are a number of materials which do not follow the linear/branched classification. Neither polypropylene [25] nor linear silicones [24] exhibit the discontinuity in the flow curve associated with HDPE and some linear low molecular weight silicones have been shown [27] to exhibit the recirculating vortices associated with LDPE. As a result, correlation of certain flow behavior with linear or branched structure must be treated with appropriate caution.

The most comprehensive work examining the effect of structure on melt fracture in polyethylene melts was presented by Blyler and Hart [33]. The authors examined the behavior of five different linear polyethylenes ($M_w$ ranging from 40,000 to 200,000 and $M_w/M_n$ ranging from 2 to 32), two different branched materials, blends of the high molecular weight linear polyethylenes with low molecular weight polyethylene waxes and blends of linear and branched polyethylenes. The results
of their work with the branched materials were typical of the behavior discussed previously: no discontinuity in flow curves and the formation of recirculating vortices prior to entering the capillary.

The flow curves generated for the different molecular weight linear polyethylenes are quite informative (Fig. 12). The effect of molecular weight is clearly evident on the magnitude of the discontinuity (no discontinuity is observed for the lowest molecular weight material) as well as its effect on the hysteresis loop. The influence of average molecular weight on the flow curve is further evidenced by examining blends of the highest molecular weight material with the wax (Fig. 13). It is important to note that the effect of molecular weight is only evidenced below the critical shear stress prior to the discontinuity in the flow curve and that above this stress, molecular weight has negligible influence.

The effect of branching was examined by the blending of different amounts of linear and branched materials. Results of Blyler and Hart's blends of linear and branched materials are shown in Fig. 14. The blends exhibit flow characteristics of both the parent polymers in proportion to the content of each. It is worth noting the regular increase in critical shear stress for the onset of fracture as a function of branched content.

Blyler and Hart concluded that, for linear polyethylenes above a certain critical molecular weight, the onset of melt fracture occurred at a fairly constant stress value of approximately $2 \times 10^9$ Pa. Furthermore, the influence of molecular weight was limited to the region of the flow curve prior to the onset of melt fracture. The effect of increased branching was concluded to increase the value of the observed critical shear stress at which melt fracture occurred.
Figure 12. Effect of $M_s$ on Flow Curves [33].
Figure 13. Flow Curves for Blends of Different $M_w$ Materials [33].
Figure 14. Flow Curves for Blends of Linear and Branched Polyethylenes [33].
2.3.6 Mechanisms for Gross Melt Fracture

The onset of gross melt fracture in both LDPE and HDPE has been typically associated with a critical value of the recoverable shear, $S$, in the fluid [15]. The recoverable shear being defined as the ratio of the wall shear stress in the capillary to the elastic modulus of the fluid, $S = \tau_w/G$. The reported range of values for $S$, being between 1 and 10, with the majority of the values falling between 5 and 8 [15]. With the exception of the work by Everage and Ballman [35], the use of an elastic shear modulus has been used in evaluating $S$. By default, this results in explaining gross fracture in terms of shear flow behavior. The exclusive use of shear flow properties in explaining the gross fracture behavior of HDPE, where the instability is observed to propagate, if not initiate in the land region, is reasonable: the flow in the land region is dominated by shear flow behavior. The sole use of shear flow properties to explain the gross fracture behavior of LDPE, however, is not acceptable. A more detailed explanation of the factors leading to the onset of unstable flow in the entry region is reserved for the last part of this section and will not be developed here. For now, it is sufficient to point out that the mechanisms involved in the entry region instability, where there is a considerable extensional contribution to the flow, are more complex than the simple case of shear behavior discussed below.

The simplest explanation for the discontinuity in the flow curves for HDPE has been related to an inherent constitutive instability [20,36]. At high levels of stress, the behavior of the melt has shifted to the rubbery plateau region, resulting in a 'high elastic state' in the fluid. It was suggested that under the appropriate conditions, this shift in behavior could result in the observed discontinuity in the flow curve for HDPE. No explanation was offered for why LDPE, which is branched (appearing more cross-linked, with a longer plateau on a short time-scale), does not exhibit the same discontinuity.

Tordella [16] presented a mechanism for gross fracture based on a molecular mechanism. The flow at low rates was seen as Newtonian and the coils of the polymer chains were deformed at a
rate slower than the thermal motion of the chains. As a result, the flow merely imposed a drift or bias to the random motion. At higher rates, the structure was thought to be no longer random and semi-discrete cluster formation began. Flow occurred by a rolling motion of the clusters. The motion of the clusters was still thought to be slow compared with the Brownian motion of the chains and inter-cluster penetration of the chains occurred continuously, providing the mechanism for transmitting stress. As the shearing rate of the fluid was increased, the rolling motion became more rapid relative to the thermal motion of the chain segments. The clusters became more and more discrete with fewer inter-cluster chains present. At a critical shear-rate, there were too few chains bridging the different clusters to transmit the stress and local failure of the flow occurred. This resulted in high localized stresses at the interfaces and the failure propagated through out the fluid. Tordella noted that the mechanism suggested was for failure in the bulk of the fluid and not at the interface with the capillary wall. He reasoned that the surface of the wall was quite coarse on a molecular scale and that there is probably a semi-stagnant layer of polymer which formed the 'effective capillary surface'. As a result, failure at the capillary surface was viewed as a polymer-polymer failure, rather than a polymer-metal failure. This mechanism seems to be a reasonable explanation for the fracture behavior in HDPE. It should be noted here however, that a recent investigation [21] indicates that the materials of construction of the die has a discernible effect on the onset of melt fracture in both HDPE and LLDPE. This suggests that the failure of the melt is influenced by the die wall, if not initiated at the die wall. This is contrary to the mechanism proposed by Tordella.

2.3.7 Mechanisms for Surface Melt Fracture

The mechanisms which have been proposed in the literature for surface melt fracture, specifically for LLDPE, embrace the constitutive instability discussed previously for gross fracture. That is to say, that transition from a viscous flow state to a 'high elastic state' [20] is necessary for surface
fracture to occur. It is the behavior of the melt after this transition which differ in the proposed mechanisms.

Kurtz [19] proposed a two step mechanism for the surface fracture behavior of LLDPE. The first step involved 'pre-stressing' the melt along the wall in the land region of the die and the second step was a critical acceleration of the 'pre-stressed' material as it exited the die. The 'pre-stressing' is directly analogous to the 'high elastic state' suggested by Vinnogradov and co-workers [20] previously. Once the material is 'pre-stressed', the melt along the wall is extended as it exits the die and accelerates from a near zero velocity within the die up to the average plug-flow value outside the die, resulting in a brittle failure of the material on the extrudate surface. It is important to emphasize that both a critical stress and acceleration are required for surface fracture to occur. Furthermore, Kurtz suggested that it is materials which undergo a brittle failure under these conditions that exhibit surface fracture, while materials which undergo ductile failure do not. This is perhaps a convenient way of addressing the material dependence of surface fracture behavior and has not been supported by any experiments. The final point to be made regarding this mechanism is explaining the increased severity with increasing flow-rate. Quite simply, as the rate is increased, the position of the required critical stress moves further into the melt and the acceleration required to obtain the plug-flow profile outside the die is increased. Both these factors contribute to increasing the depth of material at the surface meeting the requirements for surface melt fracture.

A more recent mechanism for the surface fracture behavior of LLDPE has been proposed by Ramamurthy [21]. His experiments with different materials of construction for a blown film die suggested that there was an interaction between the melt and the metal die surface. This lead to the idea that surface fracture results from a failure of adhesion at the melt/metal interface. Again, a transition to the 'high elastic state' at a critical stress was believed to occur, but Ramamurthy suggested that this lead to the onset of slip at the wall. As the rate is increased, the slip velocity is increased resulting in more severe surface fracture. Arguments supporting the presence of slip during surface fracture [21,22] are based on the flow curves for LLDPE. It has been suggested that there is a change in the slope prior to the onset of surface fracture and well before the discontinuity as-
sociated 'stick-slip' fracture. A critical examination of these curves finds one hard pressed to observe said change in slope. While slip at the wall may very likely occur during gross fracture, the evidence for slip during surface fracture is not conclusive or convincing.

2.3.8 Entry Flow Behavior

The importance of the entry region in the melt fracture behavior of LDPE was emphasized earlier in this section. The initial studies on the entry flow behavior of polymer solutions and melts were motivated by the influence of this region on the melt fracture phenomenon, but in the early 1970's the entry flow behavior became the focus of attention in its own right. Unfortunately, the predilection to correlate the observed behavior with shear flow phenomenon carried over from melt fracture studies. Everage and Ballman [35] were among the first to recognize the importance of the extensional nature of the flow in this region and suggested that the onset of melt fracture should be correlated with a recoverable extensional strain rather than recoverable shear strain. An excellent review of the literature relating to the entry flow behavior has been presented by White, Gotsis and Baird [37] and it is not intended to duplicate their work here. In this section, only points of their review concerning the onset of unstable flow which are relevant to the proposed research will be given.

Studies of entry flow behavior have used both polymer solutions and melts. There are distinct advantages and disadvantages associated with either fluid. Solutions offer the advantage of being able to conduct the experiments at room temperature as well as being able to tailor the rheological properties of the fluid under investigation. This can be accomplished through the use of Boger fluids [38,39]. Typical Boger fluids consist of 0.02−0.1% PAA, 1-4% water and 96-99% maltose or glucose syrup. The Newtonian behavior of the syrup dominates the shear-viscosity behavior of the fluid, while the PAA provides elasticity, the magnitude of which can be controlled by composition. These solutions are ideal for examining the effect of fluid elasticity on entry flow.
behavior. While the elasticity of Boger fluids is comparable with that of melts, the viscosity is typically two orders of magnitude lower. This, in addition to the lack of chain interactions present in melts, limits the applicability of representing melt behavior by that of solutions.

The advantage of using polymer melts as the test fluid is obvious: it is the behavior of melts that is of practical interest. Problems associated with the use of melts are experimental in nature. Rheological properties of melts can not be easily tailored in the same manner as those of Boger fluids. As a result, it is more difficult to isolate a particular characteristic of the melt and determine its effect on the flow behavior in the entry region. Additionally, experiments involving melts must be carried at temperatures in excess of 150°C and due to the highly viscous nature of melts at elevated pressures. The test apparatus must be able to handle these conditions, while still providing the investigator the means to directly observe the flow behavior.

Historically, thorough characterization of the rheological properties of either solutions or melts used in entry flow experiments has been lacking. This was due in part to a lack of available techniques, part to a lack of understanding of what properties were important or in part to poor presentation of results. Characterization of the shear viscosity and normal stress behavior for solutions can be accomplished up to very high shear-rates. Determination of the solution's extensional properties is limited. For melts, the extensional behavior is more readily characterized, but measurement of the normal stress behavior at shear-rates above 10 s⁻¹ has not been possible.

Initial attempts to correlate vortex formation and size (Fig. 15) with rheological properties relied on the use of the elasticity of the material. The Weissenberg number, which is a ratio of the normal stress to twice the shear stress (i.e. $We = N_t/2\sigma$), was used as the measure of the material's elasticity. Reasonable success was achieved using this correlation for materials which formed vortices, but no explanation was given for why some fluids which were elastic formed vortices, while others did not. These results indicated that, while elasticity may have played a role in the behavior of vortex growth, it was not solely responsible for their formation.
Figure 15. Schematic of Entry Flow Behavior for LDPE [44].
Contradictory reports of particular materials forming vortices or not forming vortices were made in the literature adding to the confusion of what factors were important in vortex formation and growth. It appears that these contradictory reports were the result of the particular geometry used in the experiments. It has been shown [37,39-41] that materials which form vortices in axisymmetric contractions may not exhibit this behavior in a planar contraction. Boger fluids are a typical example. Furthermore, the contraction ratio used in a particular experiment has been shown to have an influence on vortex behavior. A study of the pressure loss in sink flow experiments [42,43], similar to the flow through an orifice, provided a relative measure of the extensional stresses encountered in the different geometries. Results indicated that the extensional stresses encountered in axisymmetric contractions are substantially higher than those in a corresponding planar contraction. This is a clear indication that the extensional properties of the fluids will play a significant role in the flow behavior in the entry region.

Investigations of the flow of polymer melts [44] presented the first attempts to correlate the behavior in the entry region with extensional behavior. The flow was broken down into a shear component and an extensional component. Estimates of the extensional behavior of different materials were made from capillary flow data and used to correlate the flow behavior in the entry region. Results indicated that vortices were observed for materials that exhibited increasing extensional viscosity with stress. Following this work, a mechanism was suggested to explain the formation of vortices. Vortices were seen to result as a stress-relief mechanism for materials which exhibited strain hardening behavior. This has become the accepted explanation for formation of vortices in the entry region.

2.3.9 Concluding Remarks

Prior to moving away from the melt fracture discussion of this section, there are a few points which need to be emphasized. First, the factors leading to the onset of melt fracture in single
component flows are certainly not diminished in bi-component flows. If surface fracture is solely
the result of slip at high stresses, it is likely that downstream, where the stresses at the interface are
greatly reduced, this would not pose any serious problems. If, however, surface fracture results from
a critical acceleration of the melt, the required acceleration of the two different materials at the
interface could very possibly play a significant role in the stability of the interface. The importance
of the extensional behavior of the fluids as they join together and are accelerated is self evident.
Additionally, the importance of the extensional behavior on the entry flow behavior of LDPE and
its impact on gross fracture behavior must be stressed.

The impact of the types of possible instabilities leads to the second point to be emphasized: the
need to thoroughly characterize the rheological properties of the materials to be used in extension
as well as in shear. This is necessary if any type of meaningful interpretation of observed bi-
component instabilities is to be achieved, either to rule out melt fracture as a cause or to support
claims of its importance.

The final point to be reiterated from this section is the utility of stability analyses in predicting
whether or not a particular region of flow will propagate or damp an imposed disturbance for a
given set of conditions. The ability to examine the influence of particular constitutive parameters,
such as increased elasticity, is of practical importance in understanding observed behavior. It is this
point which leads into the discussion of the next section of this chapter: the stability analyses of the
interface in bi-component flows.

2.4 Stability Analyses of Bicomponent Flow

In the previous section, the use of stability analyses in establishing regions where disturbances
may propagate has been shown to be quite useful. In this section, a review of the literature con-
cerned with stability analyses of the interface in bicomponent flows is presented. The importance of viscosity ratio, elasticity ratio and depth ratio of the fluids are highlighted.

Yih [45] presented the first detailed stability analysis of two superposed Newtonian fluids. The one-dimensional flow between parallel plates was examined subject to two-dimensional disturbances. Using a linear stability analysis, the perturbed equations of motion were developed and the appropriate eigenvalue problem formulated. By appropriate choice of boundary conditions, plane Poiseuille and Couette flow were examined. Regions of stability were given in terms of the viscosity ratio, \( m = \mu_1/\mu_2 \) and depth ratio, \( n = d_1/d_2 \) of the two fluids.

Results of Yih's analysis for Couette flow indicated that the interface between the two fluids was unstable for all values of \( m < 1 \), when \( n > 1 \). For \( m < 1 \) and \( n < 1 \) there were regions of stability and instability present, depending on the values of \( m \) and \( n \). These results were presented graphically. Typical results show that for a value of \( n = /2 \), the flow is stable for values of \( m \) between 1 and 9. Due to the way the equations were non-dimensionalized, the imaginary part of the eigenvalue, which determined stability, was cast in terms of \( aR \), where \( a \) was the wave number and \( R \) was the Reynolds Number of the flow. Therefore, when an instability appeared, it was present for all values of the Reynolds number (as \( R \to \infty, a \to 0 \)). Thus, even for creeping flows, the instability will be present, although its magnitude approaches 0. The correctness of the analysis was tested by interchanging the fluids and examining the results. As expected, when the fluids are reversed, the regions of stability for \( 1/m \) and \( 1/n \) are the same as those for \( m \) and \( n \) determined initially.

Yih also examined the stability of Poiseuille flow for the special case of \( n = 1 \). Results of this analysis indicated that the interface was unstable for all values of the viscosity ratio. The conclusions which are drawn for both types of flow suggest that instabilities, when they are present, will manifest themselves in the form of long waves, rather than in turbulent short waves. This is due to the fact that they are present for all values of the Reynolds number, even for creeping flows where turbulence is not observed.
Hooper and Boyd [46] examined the linear stability of two Newtonian fluids in unbounded Couette flow. This work was motivated by the desire to determine if the instabilities reported by Yih [45,47] were strictly a result of the viscosity jump at the interface or whether the walls confining the flow had an impact on the stability of the flow. Following Yih's development of the perturbation equations, the system was analyzed for the case of infinite fluids on either side of the interface. Results of Hooper and Boyd's analysis indicate that, in the absence of surface tension, the interface is always unstable. They did not restrict their findings to long-wave disturbances, but suggested that short-wave disturbances were probably damped by surface tension. They also found that variation in density between the two fluids could have a stabilizing or destabilizing effect on the interface depending on the configuration of the fluids. In the short-wave limit, they found that the disturbances were present only locally at the interface and that spatial variations occur on the scale of the wavelength. Therefore, the analysis can be extended to any laminar shear flow, such as Poiseuille flow where, at the interface, the flow locally resembles Couette flow. The growth rates of the disturbances observed were slow and were sensitive to relatively small values of surface tension. As a result, the authors concluded that they were not likely to be observed experimentally.

Joseph, Renardy & Renardy [48] examined the stability of two Newtonian fluids flowing through a circular pipe. The impetus for the work was to evaluate the 'viscous-dissipation principle', which states that the preferred configuration of two fluids undergoing Poiseuille flow is the one that minimizes the viscous-dissipation for a given flow-rate or maximizes the flow-rate for a given pressure gradient. This is the suggested mechanism which leads to a less viscous fluid encapsulating a more viscous fluid. The previous work of Hickox [49] only considered the stability of the configuration with the less viscous fluid at the core. As suggested by the minimization principle, this arrangement was found to be unstable. Joseph, Renardy & Renardy extended the analysis to study the stability of the interface when the more viscous material is at the core. The results of their analysis show that the stability of the preferred configuration, as suggested by the minimization principle, is dependent on the ratio of the interfacial radius, \( R_i \), and the pipe radius, \( R_p \). They find that for \( R_i/R_p < 0.7 \), the flow is unstable for all modes and that for \( R_i/R_p > 0.7 \), the flow is predominantly
stable. This is in qualitative agreement with Yih's finding that in plane-Poiseuille flow, the stability of the interface is dependent on the depth ratio of the fluids. In contrast to single phase flows, where the viscosity acts to dampen the short-wave disturbances, it was the surface-tension which dampened these disturbances at the interface in bicomponent flows. The authors concluded that flows with large amounts of the less viscous fluid on the outside were weakly unstable to long-wave disturbances. All flows with the less viscous fluid on the outside were weakly unstable to short-wave disturbances, but that these disturbances were probably damped by the influence of surface tension. These results indicate that the 'viscous-dissipation principle' is not valid universally.

Renardy and Joseph [50] further studied the stability of two Newtonian fluids between concentric cylinders in Couette flow. The work was motivated by the experimental work of Joseph, Nguyen & Beavers [12], who observed stable configurations of the flow with thin layers of less viscous material lying next to either cylinder. The authors found that reason for this stability was a result of the influence of surface tension. They did report, however, that the configuration with the less viscous layer next to the inner cylinder was more stable (the decay rates for this configuration are larger). These results are in further contradiction to the 'viscous-dissipation principle'. In the absence of surface tension, the flows are unstable to short-wave disturbances and the authors suggest that this will lead to the formation of emulsions. Moreover, density differences in these flows result in centrifugal force effects. In the absence of surface tension, the stable configuration is the one with the more dense fluid in the outer layer. However, if the density difference is small, surface tension can have a stabilizing effect on the reverse configuration.

Renardy [51] examined the stability of two Newtonian fluids in Couette flow for intermediate values of the wave number \( \alpha \). Her results indicated that the response of the interface to this range of disturbances could not be directly interpolated from the long-wave and short-wave limits. However, the effects of density, viscosity and depth ratios on the behavior of the interface qualitatively follow the results given previously.
Wong and Jeng [52] studied the effect of the shear-thinning behavior of two Ellis fluids on the stability of flow through a circular pipe. Similar to the results for Newtonian fluids, the flow was found to be predominantly stable if the more viscous fluid was at the core and predominantly unstable if the less viscous fluid was at the core. When the more viscous material was at the core, stronger dependence of the viscosities on the shear-rate had a stabilizing influence. When the outer fluid was more viscous, stronger dependence of the viscosities on the shear-rate was destabilizing. Unlike previous analyses for Newtonian fluids, the authors found the influence of surface tension had a destabilizing effect on the flow.

Li [53] examined the effect of elasticity on the stability of two Oldroyd fluids in plane Couette flow. Following the method of Yih [45], the equations governing the primary flow were developed and the appropriate eigenvalue problem was formulated. Again, two-dimensional perturbations were applied to the steady one-dimensional flow and their effects were studied. The zeroth-order approximation solution is identical to that of Yih, indicating that the method was correctly formulated. The author found that the effect of elasticity was evidenced only through continuity of shear and normal force conditions at the interface. The results of the analysis are given in terms of the depth ratio, \( n \), the viscosity ratio, \( m_n \), and the elasticity ratio, \( m_\gamma \).

The results of Li's analysis showed that for \( n < 0.5 \), elasticity had a stabilizing effect on the flow for \( m_n < 1 \), regardless of the value of \( m_\gamma \). For \( n \) increasing from 0.5 to 1, the range of values of \( m_n \) for which the elasticity of the fluids stabilizes the flow becomes narrower for \( m_n < 1 \) and wider for \( m_n > 1 \). The analysis further indicates that in the absence of viscosity variation (\( m_n = 1 \)), elasticity has no effect on stabilizing or destabilizing the flow. The author concludes that in general, elasticity can have either a stabilizing or destabilizing effect on the flow depending on the values of \( n, m_n, \) and \( m_\gamma \).

In a subsequent paper, Li [54] investigated the stability of two superposed Oldroyd fluids flowing down an inclined plane. In addition to the interface stability, the effect of the elasticity ratio on the stability of the free surface was examined. The analysis is a direct extension of his previous analysis.

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of plane Couette flow. The results showed that the effect of elasticity on the free-surface was destabilizing, regardless of the viscosity ratio. The elasticity ratio has little effect on the stability except at very small values, where it has a slight stabilizing effect. The depth ratio has no influence except to magnify the effects of stable or unstable behavior. It does not act to stabilize or destabilize the flow as is the case for Couette flow.

Khan and Han [55] studied the linear stability of two Coleman-Noll second-order fluids flowing through a rectangular duct. A pseudo-three-dimensional perturbation analysis was performed for a one-dimensional flow. This was accomplished by examining disturbances in the \( x_1 - x_2 \) plane (along the flow-direction) and then the \( x_2 - x_3 \) plane (transverse to the flow-direction) respectively. Based on the work of previous authors, who have determined that the second-order fluid does not yield physically meaningful results except in the long-wave limit (small \( \alpha \)), the authors restricted their analysis to values of \( \alpha \) in this range. The perturbation equations for the analysis are developed in the typical manner and the eigenvalue problem formulated.

The results of the Khan and Han's analysis in the flow-direction showed that for equal depths of fluids, the flow was always unstable. The analysis neglects the effect of surface tension, which the authors assume to be inconsequential, contrary to previous investigators. The results for the equal depths of fluids is thus in agreement with the earlier work of Yih for Newtonian fluids. For the case of unequal depths, the results indicated that the elasticity could have either a stabilizing or destabilizing effect, depending on the value of the viscosity ratio and was independent of the elasticity ratio. The results for the plane transverse to the flow direction show that the stability is dependent on both the elasticity and viscosity ratio. It should be noted that the effect of the side walls on the boundary of this plane were not considered. The analysis breaks down for the limiting values of the elasticity and viscosity ratios of 1 (single component) and cannot be compared to other analyses of single-component flows. The authors concluded that, in general, linear stability analyses are more stable than zeroth-order analyses.
Waters and Keeley [56] presented the results of their stability analysis of two four-constant Oldroyd fluids in Couette flow. By appropriate choices of constants, limiting cases for Newtonian, power-law and Oldroyd-B (constant viscosity, elastic) fluids were also presented. The results are presented in terms of a depth ratio, $\delta = d_2/d_1$, a viscosity ratio, $\beta = \eta_2/\eta_1$, and a power-law index ratio, $n_2/n_1$.

The Newtonian analysis showed that for $\delta < 1$ and $\beta > 1$ or $\delta > 1$ and $\beta < 1$ the flow was unstable. This is in agreement with the previous work of Yih [45,47] and Renardy [51]. Results for the case of power-law fluids, which in Couette flow exhibit constant viscosities, indicated a deviation from Newtonian stability contours, even when the apparent viscosity ratio was used ($\beta_s = \eta_1/\eta_2$). The index ratio of the fluids had a profound effect in shifting the stability contours.

The effect of the elasticity for Oldroyd-B fluids could be either stabilizing or destabilizing. This is in agreement with the previous results of Li [54]. The authors found that elasticity had little effect for viscosity ratios of $\beta = 1 \pm 2$ or if either layer was much larger than the other. The stability of Oldroyd 4-constant fluids is influenced by the shear-thinning behavior of only the less viscous layer. Its effect on stability is quite pronounced for $\beta < 1$ and $\delta > 10$. Again, the effect of the index ratio is not limited solely to its effect on $\beta_s$.

It is obvious from this survey of stability analyses that a great deal effort has been devoted to predicting the onset of interface instabilities. However, experimental testing of these predictions has been limited primarily to polymer solutions, rather than to melts. As a result, the utility of these methods in predicting real behavior for use in the design of coextrusion processes is still questionable. The current study should provide results suitable for determining the accuracy of these methods under realistic processing conditions.
2.5 Viscoelastic Flow Simulation

The second objective of the present research is to examine the effects of different rheological characteristics on the the joining flow behavior of viscoelastic fluids. In this section, a brief review of the problems encountered in modelling viscoelastic flows in general will be given, as well as recent developments in the literature for handling these problems.

2.5.1 Governing Equations

The flow of isothermal fluids are governed by the conservation of mass (continuity equation) and the conservation of momentum (equation of motion). In the absence of inertial effects (low Reynolds number flows) and the assumption that the fluid is incompressible, these equations are given by

\[ \nabla \cdot \mathbf{u} = 0 \quad \text{(continuity)} \]  \hspace{1cm} 2.5.1

\[ \nabla P + \nabla \cdot \mathbf{f} = 0 \quad \text{(motion)} \]  \hspace{1cm} 2.5.2

In addition to these two equations, a third equation (constitutive), which relates the stress in the fluid to its deformation, is needed. One of the simplest constitutive equations for a viscoelastic fluid is the upper-convected Maxwell model, which in non-dimensional form is given by

\[ \dot{\mathbf{\varepsilon}} + \dot{\mathbf{e}} \mathbf{\varepsilon} = -\frac{1}{\eta} \quad \text{(UCM)} \]  \hspace{1cm} 2.5.3

---

1 Variables underscored once indicate a vector, while those underscored twice indicate a second rank tensor. The sign convention used for the stresses in this text is \( \tau_{ij} > 0 \) in compression.
where the Deborah number is defined by \( De = \lambda \dot{\gamma} \), and \( \tau_1 \) is the upper convected derivative of the stress given by

\[
\tau_1 = \frac{\partial q}{\partial t} + (u \cdot \nabla) q - (\nabla u)^T q + q \cdot (\nabla u)
\]

Due to the coupling of the equation of motion with the constitutive equation and the addition of irregular boundaries or boundary conditions, the system of equations described above needs to be solved numerically. A review of the application of the finite difference and finite element methods used for solving this type of system has been given in Pearson and Richardson [57] and by Crochet, Davies and Walters [58].

### 2.5.2 Finite Elements

The finite element method (FEM), which has been the method of choice in simulating viscoelastic flows, involves the discretization of the domain into sub-domains (finite elements) and locally approximating the governing equations by polynomials. The error of the approximation is then in some sense minimized, depending on the type of finite element method employed. In the case of non-linear systems, such as those encountered in non-Newtonian flows, the approximating system is linearized and solved iteratively. Excellent introductions to the various types of finite element methods and the use of iterative schemes have been presented by Reddy [59] and Davis [60].

The most common FEM employed in the simulation of viscoelastic flow is Galerkin’s method. Galerkin’s method involves casting the governing differential equations into an equivalent integral form by the use of a multiplying test function. Higher order derivatives in the governing equations are reduced by transferring an order of differentiation to the test functions, using integration by parts. This results in the ‘weak’ form of the governing equations. The dependent variables and the test functions are then approximated locally (over each element) by polynomial functions, resulting
in an algebraic system of equations for the element. The element equations are then assembled into a global system of algebraic equations, which is solved simultaneously, producing the approximated solution to the problem. In the case of non-linear systems, the local equations are linearized prior to assembly and the solution is found iteratively subject to some convergence criterion.

2.5.3 The High Deborah Number Problem

One of the standard geometries used in evaluating the performance of a FEM code is the 4:1 contraction. This is a simple geometry which highlights the differences between Newtonian and non-Newtonian flow behavior. The ability to predict the vortex behavior discussed in previous sections is one of the factors used in the evaluation of the code. More recently, the emphasis in evaluating performance has been based on the degree of elasticity that can be incorporated into the flow and still result in a converged solution. The level of elasticity is typically indicated by the Deborah number. A typical upper limit for simulations using the UCM constitutive equation is $De = 1$. The inability of codes to converge above a limiting elasticity level has led to what is called the 'high Deborah number problem'. A second test problem which has been used in examining the performance of a code at high elasticity levels is the 'stick-slip' problem. This involves the flow through a rectangular or circular duct with the 'no-slip' boundary condition at the wall relaxed at some fixed distance along its length (Fig. 16). The limiting behavior in both the contraction and 'stick-slip' problem is thought to be the result of non-integrable stresses developed at boundary singularities in the flow. The critical value for different simulations has been found to be dependent on the type of constitutive equation used in the simulation and also on the mesh refinement (discretization). Thorough reviews of this problem have been presented elsewhere [4,37,61] and will not be duplicated here.

The crux of the 'high Deborah Number' problem is the inability of the numerical simulation to handle the large stress gradients which result near boundary singularities, as a result of the elastic
Figure 16. Stick-Slip Geometry.
nature of the fluid. Mendelson et al. [61] identified two possible causes for this loss of convergence. The first was accumulated approximation error near the singularities. This is the result of the approximating method failing to accurately describe the behavior near the singularity, e.g. linear or quadratic approximations of steep stress gradients. The second cause for the lack of convergence was the loss or the multiplicity of the solution above a critical characteristic elasticity. In other words, the schemes failed because there was no solution for the described problem or there were multiple solutions (bifurcation) and the codes oscillated between them. Reports of bifurcation points have been reported for the upper-convected Maxwell, second-order fluid and the Phan-Thien Tanner models, but these are currently believed to be artifacts of the discretization method [62].

The loss of convergence in viscoelastic flows has also been attributed to a mathematical change-of-type from elliptic to hyperbolic in the system of equations governing the flow [63-65]. The change-of-type is associated with the increased importance of the lower order derivative term, \( De (\mu \Sigma) \), in the constitutive equation, as the Deborah number is increased. The problem of changing from elliptic to hyperbolic form, as a result of lower derivatives, has been considered in detail in other fields of fluid mechanics [66-69]. The basic problem is that finite element methods, which are well suited for elliptic problems, are not well suited for hyperbolic. Thus, the change of type can lead to the anomalous behavior of the solution generated by the method being used.

Approaches used in handling the problems posed by change-of-type, as well as dealing with non-integrable stresses resulting from the elastic nature of the fluids, will be discussed in the following paragraphs.

2.5.4 Convection Dominated Flows: SUPG

The problem of lower order terms affecting the behavior of numerical solutions is typical of high Reynolds number flows, where inertial effects are important. The inertia terms in the equation of
motion are of the same order as the convective stress term in viscoelastic constitutive equations. In high Reynolds flows, the problem is manifested in the form of spurious oscillations or 'wiggles' in the velocity field, while in viscoelastic flows, the oscillations or 'wiggles' are seen in the stress field. The obvious parallel between the behavior in the two systems suggests that, methods which are helpful in one case, may well be helpful in the other.

Brooks and Hughes [66,69] working with high Reynolds number flows, developed a method which has been successfully adapted to viscoelastic flow simulation [67,70-72]. This method was termed the streamline-upwind Petrov-Galerkin (SUPG) formulation. This method is the result of two modifications to the classical Galerkin formulation: streamline upwinding and the Petrov-Galerkin formulation. Detailed discussions of the development of this procedure have been given elsewhere [67,69] and only an outline of the SUPG method is presented here.

The development of the SUPG method was motivated by trying to control the effect of the convective term on the behavior of the solution in numerical simulations. The success of upwinding methods in one-dimensional, convection dominated flows led to their extension into two-dimensional flow problems. Unfortunately, the effect which was desired and realized in one-dimensional problems was carried over into the second direction in two-dimensional problems, leading to undesired results termed 'cross-wind diffusion'. In addition, traditional upwinding methods were applied exclusively to the convective term only, leading to a non-consistent formulation of the problem. This led to criticism that the method incorporated artificial diffusion into the formulation and that the true physics of the problem was lost. The unwanted cross-wind diffusion and the incorporation of artificial diffusion into the problem represented legitimate concerns in the use of upwinding techniques and has led to a great deal of skepticism in regarding its application. Both of these concerns have been addressed in the SUPG method presented by Brooks and Hughes.

The SUPG methodology is most clearly described by contrasting its application with the traditional Galerkin (GAL) formulation. The UCM model described by equation 2.5.3 will serve as the basic equation in the following comparison.
The first step in both the GAL and SUPG methods is to generate the equivalent integral form of the governing equation as follows:

\[
\int_{\Omega} \left\{ [\mathbf{u} + D\mathbf{e} \cdot (\mathbf{u} \cdot \nabla) \mathbf{r} - (\nabla \mathbf{u})' \mathbf{r} - \frac{1}{2}(\nabla \mathbf{u})'] \right\} w \, d\Omega
\]

2.5.5

The first difference between the SUPG and GAL methods is in the choice of the approximating functions for \( w \) and \( u \) or \( r \). Let the approximating functions over an element be described by three classes of behavior. In the first, \( C^\infty \), the functions are continuous and smooth across the element boundary, \( \Gamma \). In the second class, \( C^0 \), the function is only required to be continuous across \( \Gamma \). In the final group, \( C^{-1} \), the functions are discontinuous across \( \Gamma \). In the classical GAL formulation, the weighting function, \( w \), is approximated using the same class of functions used for \( u \) or \( r \), typically \( C^0 \) for viscoelastic flow problems. In the Petrov-Galerkin formulation, if the approximating function for the stress or velocity is given by \( S \in C^0 \), the weighting function is given by \( \hat{S} = S + P \), \( S \in C^0 \) & \( P \in C^{-1} \). The influence of the \( C^{-1} \) contribution of \( P \) is limited to the weighting function's behavior within the element (discontinuous at the boundary) and as result lends itself to weighting upstream behavior more than downstream behavior (Fig. 17). Additionally, if \( P \) is cast in terms of \( S \), such that \( P = k \mathbf{u} / \| \mathbf{u} \| \nabla S \), the influence of upwinding in an element can be directed along the streamline, eliminating the problem of cross-wind diffusion. The application of this technique has been shown to be quite successful in high Reynolds number flows by Brooks and Hughes [87].

2.5.5 Stress Sub-Elements in Viscoelastic Flows

Marchal and Crochet [71] investigated the method of formulating viscoelastic flow problems and its affect on the behavior of the solution. Starting with the simulation of a Newtonian fluid in the stick-slip problem, the authors investigated the solution using a primitive formulation (velocity and
Figure 17. Weighting Functions: (a) Galerkin, (b) Petrov-Galerkin.
pressure as primary variables in equation 2.5.2 and a mixed formulation (stress and pressure in equation 2.5.2 in conjunction with the Newtonian constitutive equation). The results showed the primitive formulation to yield a smooth solution for the stress behavior around the boundary singularity, while the mixed formulation exhibited oscillations in the stress field. While either formulation, prior to discretization and approximation, represents an identical problem, the behavior of the respective formulations in simulation are dramatically different (Fig. 18). The spurious oscillations of the mixed method were attributed to the coupling of the stresses in the EOM with the constitutive equation. This led the authors to develop an optimal mixed method for the Stokes flow prior to attempting a viscoelastic mixed method.

Marchal and Crochet found that by using nine-noded Lagrangian elements for the velocity, with the corner nodes used for bilinear approximations of the pressure, in conjunction with $n^2$ ($n > 2$) bilinear sub-elements for the stresses (Fig. 19), the spurious oscillations of the mixed method could be eliminated (Fig. 20). Application of this method to viscoelastic flows was not successful. The authors concluded that due to the hyperbolic nature of the viscoelastic flow problem, the SUPG method described previously was required to handle the dominance of the convective derivative. Success was realized with the SUPG method provided that the velocity field was predetermined prior to the solution of the stress field. Unfortunately, when the EOM was coupled with the constitutive equation, the SUPG method failed to converge. With the failure of the consistent application of the SUPG method, Marchal and Crochet attempted simulations applying the SUPG method exclusively to the convective term in the constitutive equation. The authors reported that this method achieved success at all values of the Deborah number considered ($De > 45$) and did not appear to suffer from the limiting elasticity level phenomenon. While it is tempting to herald this work by Marchal and Crochet as having overcome the high Deborah number flow problem, at least for the 'stick-slip' problem, there are discomforting aspects of their work which prevent one from doing so. First and foremost is their use of a non-consistent formulation of the problem to accomplish these results. If the non-consistent formulation is examined and the constitutive equation

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Figure 18. Primitive vs Mixed Formulations [71]:
(a) u-v-P velocity, (b) mixed velocity,
(c) mixed shear-stress.
required to achieve the same formulation, using a consistent formulation is backed out, the results are disconcerting. The results of such an analysis yield the following constitutive equation:

\[ z + De ((μ·γ) z - (γμ)^T y - z·(γμ)) = -\dot{z} + De \gamma ((\beta·γ)·y) \]  \hspace{1cm} 2.5.6

where \( \beta = k / (μ·y) \)

With no justification for the last term on the right hand side of the equation other than it offsets the increasing importance of the convective term on the left hand side and is helpful in obtaining a converged solution. It would seem to make more sense to simply eliminate the convective term on the left because it causes problems. Even neglecting this arbitrary inclusion to the constitutive equation, the fact this method requires more than 70 \( L/Ds \) to achieve damped behavior of the calculated stresses at high \( De \) numbers, does not seem reasonable. This fact tends to support the argument that the non-consistent application of upwinding methods serves to add artificial to the physics of the problem and that the convergent algorithms are obtained at the expense of distorting the true physics of the problem.

In conclusion, while the method presented by Marchal and Crochet is tempting in its apparent ability to solve the high Deborah number problem, it does so at the expense of the actual physics of the problem.

2.5.6 The Modified Upper-Convected Maxwell Model (MUCM)

The non-convergent behavior of the UCM with mesh refinement near boundary singularities was addressed in the recent work of Apelian, Armstrong and Brown [70]. The authors examined the influence of the constitutive on the non-integrable stresses near the boundary singularity in the 'stick-slip' problem. These large stresses result from the elastic term in the UCM, \( De_{1} \), the
Figure 19. Velocity/Pressure Element, Stress Sub-Elements [71].
Figure 20. Results for Different Sub-Elements [71]:
(a) 2x2 velocity, (b) 3x3 velocity, (c) 4x4 velocity,
(d) 2x2 stress, (e) 3x3 stress, (f) 4x4 stress.
Deborah number being defined as $De = \lambda \dot{\gamma}_\infty$. $\lambda$ and $\dot{\gamma}_\infty$ are the relaxation time and wall shear rate respectively. A modified upper-convected Maxwell model (MUCM) is developed from network theory which results in the Deborah number being a function of the trace of the stress tensor, $tr(\tau)$.

The development of the MUCM is based on the assumption that the creation and destruction rates of network segments are increasing functions of the average extension of the network. The average extension is related to the $tr(\tau)$ At high levels of $tr \tau$, the junctions exist for shorter periods of time resulting in a decreasing function for $\lambda$ with increasing $tr \tau$. This development leads to Newtonian behavior of the model in regions of extreme $tr \tau$ ($\lambda \to 0$). The resulting MUCM is given by

$$\dot{\lambda} + De \, tr \dot{\lambda} = -\dot{\gamma}$$

where $De(tr) = \frac{\dot{\gamma}_\infty \lambda_0}{1 + (F(\lambda)tr)^{\kappa} - 1}$

$$F(\lambda) = \frac{(\kappa^{-1} - 1)^{1/(\kappa - 1)}}{2 De^\kappa} , \quad \kappa = 0.99$$

The behavior of this model in steady shear flow and steady elongational flow is shown in Fig. 21. An added benefit of the MUCM, is that the unbounded extensional viscosity predicted by the UCM at $e = 1/2 \lambda_0$ is no longer present.

The results of the Galerkin FEM analysis by Apelian, Armstrong and Brown indicated that the use of the MUCM allowed converged solutions to a significantly higher Deborah number than those using the UCM ($De = 1.9$ vs 0.6 for the same mesh). The loss of convergence at $De = 1.9$, was reported to be coincident with the change-of-type of the system, discussed earlier in this review. The influence of the MUCM is observed only in the stress levels near the boundary.

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Figure 21. Comparison of UCM and MUVM Rheological Predictions [70]: (a) shear flow, (b) extensional flow.
singularity. The behavior of the flow away from this point is nearly identical to that predicted by the UCM model.

The use of the MU CM model has been obviously demonstrated to improve the performance of the standard UCM. The model is developed from network theory and there are no inconsistencies in its application. However, the problem of the limiting Deborah resulting from the change-of-type still persists. This problem is addressed in the following paragraphs.

2.5.7 The Explicitly Elliptic Momentum Equation (EEME)

The problem of change-of-type is the most dominant factor in limiting the success of numerical simulations for viscoelastic flows. The change from elliptic to hyperbolic form was shown by Joseph, Renardy and Saut [63] using a Maxwell model to result from the vorticity of the system. The criterion that this change-of-type would not occur was given by

\[
\text{det}[\delta - D\varepsilon] > 0
\]

2.5.8

The violation of this criterion has been observed just prior to the failure of simulations by a number of authors. This problem has been recently addressed by King, Apelian, Armstrong and Brown [72] using what they call an explicitly elliptic momentum equation (EEME).

The development of the EEME by King et al., was motivated by Renardy's [73] formulation of the momentum equation, which makes its elliptic nature explicit. This was done by Renardy in proving the existence of solutions for problems using the UCM constitutive equation. An outline of the development of King et al.'s EEME is given here. The equations governing the steady-state, boundary value problem for an UCM fluid were given previously by equations 2.5.1-2.5.3. The divergence operator is applied to the constitutive equation (2.5.3), resulting in the following expression,
\[ \Sigma \cdot \hat{\Sigma} + D e [(u \cdot \Sigma) (\Sigma \cdot \hat{\Sigma}) - (\Sigma u) (\Sigma \cdot \hat{\Sigma})] = \sum_j \frac{\partial^2 u_j}{\partial x_j^2} + D e \sum_j \sum_k \tau_{jk} \frac{\partial^2 u_l}{\partial x_j \partial x_k} \]  

2.5.9

Equation 2.5.2 is then solved for \( \nabla \cdot \hat{\Sigma} = -\nabla P \) and substituted into the previous equation, yielding,

\[ -\nabla P + D e [(u \cdot \Sigma) (\Sigma \cdot \hat{\Sigma}) - (\Sigma u) (\Sigma \cdot \hat{\Sigma})] = \sum_j \frac{\partial^2 u_j}{\partial x_j^2} + D e \sum_j \sum_k \tau_{jk} \frac{\partial^2 u_l}{\partial x_j \partial x_k} \]

After sufficient manipulation, this equation is reduced to the EEME,

\[ \Sigma \cdot (\hat{\Sigma} - \Sigma u) + (\Sigma u) \cdot (\Sigma \cdot \hat{\Sigma}) = \nabla q \]  

2.5.10

where \( \hat{\Sigma} = \hat{\Sigma} - D e \hat{\Sigma} \)

and \( q = P + D e (u \cdot \Sigma) P \)

In this form, \( q \) is considered to be a modified or 'substantial' pressure and \( \chi \) is symmetric and positive definite, satisfying the change-of-type criterion of equation 2.5.7.

Formulations of the EEME and the MUCM, using standard Galerkin (GAL), consistent streamline-upwinding / Petrov-Galerkin (SUPG) and the non-consistent SUPG (AD) method of Marchal and Crochet [71], were solved for the 'stick-slip' problem and the journal bearing problem. Only the results of the 'stick-slip' will be discussed here. The order of the approximating functions for the variables \((u, \tau, q)\) in different simulations are designated as either QQL or QLL. Q indicates
the use of biquadratic Lagrangian elements and L represents the use of bilinear Lagrangian elements.

The results using fixed kinematics (a Newtonian velocity field) for the QLL/GAL, QLL/AD and QLL/SUPG formulations show that converged solutions are obtained for all three methods up to a limiting $De$ of nearly 5. Oscillations were observed in the stress field downstream from the singularity in the QLL/GAL formulation that were smoothed by the other formulations. These oscillations were attributed to the QLL/GAL methods inability to approximate the stresses using linear elements.

The use of the QLL/GAL method in viscoelastic flow failed at a $De$ of 3.2. The failure was a result of large oscillations developed in the stress field near the singularity. Both the QQL/GAL and the QLL/AD formulations converged to a limiting $De$ of 6.75. The behavior of these two formulations is almost identical. Results for the QQL/SUPG formulation converged up to $De = 16$ in the mesh used for the other analyses and upon refining the mesh, results were obtained for $De > 25$. An important point in the behavior of the SUPG and the AD methods is the effect of mesh refinement on the behavior of the solution. The solutions obtained using the SUPG methods converge to the same solution with increasing mesh refinement. In the case of the AD method of Marchal and Crochet, variation in the solution with refinement persisted for as much as 40 half-widths past the singularity.

The work presented by King et al. clearly demonstrates the utility of the EEME in obtaining converged solutions at high values of $De$ in the 'stick-slip' problem. Furthermore, its use in conjunction with SUPG formulations is superior to the AD method suggested by Marchal and Crochet. Its application to the contraction flow problem has yet to be demonstrated, but it is anticipated that it will prove useful for this problem.
2.5.8 Viscoelastic Simulations: Final Considerations

The discussion in this section has been limited primarily to discussing methods employing the upper-convected Maxwell model or a variation of it. Aside from the numerical considerations of the success or failure of a particular method, its utility must also be measured by its ability to predict 'real' flow behavior. The success of predicting 'real' flows starts with the constitutive equation used to model the fluid. The constant shear viscosity behavior represented by the UCM, as well as its unbounded extensional viscosity at a critical extension rate, are not representative of many polymer melts. The importance of extensional behavior discussed in the section on melt fracture suggests that a successful formulation should be able to incorporate different rheological behavior in both shear and extension. A constitutive model that has been used successfully [62] to approximate this behavior is the Phan-Thien Tanner (PTT) model. It is the PTT model which will be used exclusively in the present work.

2.6 Research Objectives

2.6.1 Experimental

In the first three sections of this chapter it was seen that a considerable amount of work has been done in the area of examining multi-layer flows. Additionally, a great deal of work investigating the single component instability of melt fracture has also been accomplished. However, with the exception of viscous encapsulation, little experimental work in the area of interface instabilities in coextruded polymer melts has been reported. It seems reasonable to expect that factors leading to the onset of single component instabilities would not be diminished in multi-layer configurations
and in fact, differences in rheological properties of the two materials, may lead to a more pronounced response at the melt/melt interface. In the fourth section of this chapter, the results of stability analyses of the bicomponent interface were presented, but experimental verification for the response of polymer melts was found to be lacking. The need to establish or eliminate factors leading to the onset interface instabilities in bicomponent flows leads to the overall experimental objective of the proposed investigation: to determine if the single component instability of surface melt fracture plays a role in the interface behavior as two separate fluids join together to form a single stratified flow.

In order to achieve the overall objective stated above, it will be broken down into three parts. First, establish the region or regions in an extrusion die responsible for initiating surface melt fracture. Second, determine the factors and conditions leading to the onset of surface melt fracture. Finally, duplicating the conditions required for surface melt fracture in the single component streams prior to the joining of the two fluids, examine the bicomponent interface behavior.

Two different materials will be used in this study: linear low-density polyethylene (LLDPE) and low-density polyethylene (LDPE). As discussed in the third section of this chapter, LLDPE exhibits the desired surface melt fracture behavior and will be used in the single component studies. The joining flow studies will incorporate both materials in an effort to establish whether or not the single component instability carries forward to disrupt the interface. The rheological behavior of the materials will be fully characterized in both shear and extensional fields.

2.6.2 Numerical

The benefit of being able to accurately predict the behavior of multi-layer flows a priori is self-evident. The fifth section of this review demonstrated the problems with trying to accurately predict the behavior of a single viscoelastic fluid at high levels of the De number. A great deal of effort has
been devoted to overcoming this limitation with varying degrees of success. The use of streamline
upwinding, while allowing for solutions to high values of De, were shown to require unrealistic die
lengths to obtain smooth solutions and in addition, changed the problem being solved by the ad-
dition of a new term to the constitutive equation. The explicitly elliptic momentum equation shows
promise in overcoming the problems associated with the system of equations changing type, but
success of the method has been restricted to geometries not possessing singular boundary behavior.
Taking these problems into account, the utility in predicting flow response to variations in geometry
or material properties at lower levels of the De number, still provides useful insight to the behavior
of the system under more severe conditions, where converged solutions are not yet attainable. This
leads to the numerical objective of the proposed study: to examine the influence of the relaxation
and extensional behavior of different fluids on the stress field development in the joining flow geo-
metry described previously. An understanding of how these properties affect the developing
stresses should provide additional insight into the factors which may lead to irregular interfaces in
bicomponent flow.

In order to separate the extensional behavior response from the shear behavior, the Phan-Thien
Tanner (PTT) constitutive equation will be used in the numerical simulations. The prediction of
the extensional behavior of the PTT model can be varied independently of its shear behavior.
Comparison of the simulations with the experimentally observed behavior for both low-density and
linear low-density will be made to judge the success of the numerical method. In addition, the limits
of convergence for the geometry examined will also be considered.
3.0 SURFACE MELT FRACTURE STUDIES OF LINEAR LOW-DENSITY POLYETHYLENE

The experimental objective of this thesis is to establish whether or not the single component instability of surface melt fracture plays a role in bicomponent interface instability. Toward this end, it was first necessary to establish the region of an extrusion die where this behavior is initiated in single component flows and determine the factors leading to its onset. In this chapter, the work performed in isolating the site of initiation and the conditions for surface melt fracture to occur in linear low-density polyethylene (LLDPE) is presented.

Fluoro-elasticomer (FE), which eliminates surface melt fracture in LLDPE, was used as a primary investigative tool in this portion of the experimental study. By determining the role of FE in eliminating surface fracture, it was possible to identify the site where surface fracture is initiated and the necessary conditions for its occurrence. The effect of blending FE with LLDPE at four different concentrations on the rheological behavior and on the quality of extrudate from a capillary device was examined. In addition to the blend studies, FE coatings were applied to different regions of two...
slit dies and the effect of the coatings on the observed flow birefringence patterns and extrudate quality was established.

The presentation of the surface melt fracture studies is divided into four parts. First, a brief introduction to the melt fracture behavior of LLDPE and the phenomenon under investigation is clearly identified. This is followed by a description of the apparatus and procedures used to meet the objectives of the single component study addressed in this chapter. The results of the experiments are presented and discussed in the third part of this chapter and in the fourth part, the conclusions are drawn from the results and recommendations for future studies are given.

3.1 Introduction

Linear low-density polyethylene (LLDPE) exhibits the entire range of melt fracture behavior discussed in Chapter 2. A transition from a smooth, glossy extrudate surface to a dull finish with fine-scale surface irregularities occurs at a value of the wall shear stress in the neighborhood of 0.14 MPa for capillary die geometries. These fine-scale surface defects, typically less than one-tenth the extrudate diameter or film thickness, are termed matte or sharkskin depending on the severity and in the current study will be termed surface melt fracture. At a shear stress level of 0.40 MPa, the extrudate is completely ruptured, with the magnitude of the irregularities being of the same order as the diameter of the extrudate itself. This type of behavior is referred to as gross melt fracture. Prior to the onset of gross fracture, the extrudate is observed to oscillate between regions of surface melt fracture and gross melt fracture. This type of behavior is typically termed spurt or slip-stick fracture. The different regions of melt fracture behavior for the two materials used in this study are shown in Fig. 22 along with corresponding examples of the extrudate. It is emphasized here that, in the present study, it is the surface melt fracture behavior of LLDPE which is of interest.
Figure 22. Melt Fracture Behavior of LLD-1 and LLD-2, L/D = 75: (a) smooth, (b) surface fracture, (c) slip-stick fracture, (d) gross fracture.
The literature concerning the surface melt fracture behavior of LLDPE, discussed in Chapter 2, was by no means conclusive as to the site where the fracture initiates or to the factors leading to its onset. There was general agreement that a transition from viscous flow behavior to a 'high elastic state' is a necessary condition, but whether or not it is sufficient is still open to debate. Kurtz [19] suggested that a critical extension rate, provided by the acceleration of the melt as it exited the die, was required in addition to the transition to the 'high elastic state' in order for surface melt fracture to occur. Alternative arguments [21,22] suggested that the transition to the 'high elastic state' lead to a failure of adhesion at the melt/metal interface and the resulting slip of the melt at the surface was the cause of surface melt fracture. The intent of the present study is to identify the region of an extrusion die where surface fracture initiates. Once the region is identified, knowledge of the kinematic field in conjunction with the rheological behavior of the material should provide additional insight into the factors ultimately responsible for the surface melt fracture behavior of LLDPE.

Traditional approaches to isolating the entry, land and exit regions of extrusion dies have relied on varying the L/D ratio of the die and the geometry of the entry and exit region. If the behavior under investigation became more pronounced with increasing L/D, this indicated that the land region of the die was important. Diminished behavior with increasing L/D suggested that the entry region was important, while no change in behavior with L/D was indicative of the exit region. The response or lack of response of the behavior to variations in inlet or exit geometry provided additional insight for the respective regions. When possible, the use of flow birefringence to monitor changes in the stress field during these experiments was also useful. Unfortunately, these methods are generally restricted to evaluating changes in the degree or severity of the behavior and do not provide direct evidence for the cause of the particular behavior. The utility of comparing the response of one material which exhibits a specific behavior with one that does not, is subject to a host of rheological considerations which are not readily resolved. That is to say, it is all but impossible to vary a single rheological characteristic of a polymer melt to investigate its impact on modifying or eliminating the behavior in question. However, for the specific case of the surface melt fracture
behavior of LLDPE, the use of fluoro-elastomer (FE) as an additive/die coating does provide this unique opportunity. The addition of FE to LLDPE completely eliminates the surface melt fracture behavior and was used as a primary tool in the current study.

The objectives of this part of the experimental investigation were to first, identify the region of an extrusion die where surface melt fracture initiates and second, to determine the conditions necessary required for the onset of this behavior. These objectives were resolved by determining the role of FE in eliminating surface melt fracture. The effect of FE on the rheology and extrudate quality of LLDPE was studied at four different concentration levels. FE coatings were then applied to different regions of two slit dies and the effect of the coatings on the flow birefringence patterns and extrudate quality established. These experiments represent the substance of the work reported in this chapter.

3.2 Experimental Apparatus and Materials

The materials used in the surface fracture studies of LLDPE and the experimental apparatus and procedures are presented in this section. The materials incorporated in the study and the method of blending are presented first. The equipment and methods used to perform the rheological characterization are described next, followed by the procedure used in the capillary studies. A description of the procedure followed in the slit die experiments, including the preparation of the FE coatings is presented next. Finally, a description of the optical arrangement used in the slit die studies for observing the flow birefringence is given.
3.2.1 Materials

Two different LLDPE's were used in this study. The first was a non-commercial grade provided by the Mobil Chemical Company with a molecular weight, $\bar{M}_w$, of 140,000 and $M_w/M_n$ of 4.1. This material has been designated LLD-1 in the present study. The second material was a standard commercial grade (NTA-101), also provided by Mobil with a $M_w$ of 110,000 and $M_w/M_n$ of 4 and has been designated LLD-2. All experiments involving LLD-1 were performed at 190 °C, while those incorporating LLD-2 were conducted at 170 °C. A comparison of the steady shear viscosity behavior of LLD-1 and LLD-2 is shown in Fig. 23 and comparison of the dynamic behavior is shown in Fig. 24. LLD-1 exhibits a higher viscosity and elastic modulus than LLD-2 at their respective test temperatures, but the differences are not significant enough to anticipate drastically different behavior in similar experiments.

The LLDPE/FE blends used in this study were comprised of LLD-1 and a commercial fluoro-elastomer (Dynamar L-10040, 3M Company). Blends of 200, 500, 1000 and 2000ppm FE were made for evaluation. These blends were prepared by weighing the appropriate amounts of FE and LLD-1 on a Mettler model PE3600 balance and initially mixing in a covered container by hand. The mixture was then passed through a Killion 25.4mm single screw extruder and the extrudate pelletized and collected. The material was then re-extruded to provide a more thoroughly mixed blend.

3.2.2 Rheological Characterization

Shear flow characterization of LLD-1, LLD-2 and the LLD-1/FE blends was performed in a Rheometrics Mechanical Spectrometer, model RMS-800. Steady-shear viscosities were measured from 0.01 to 5.0 s$^{-1}$. Beyond this region, measurements were not possible due to loss of the melt from between the plates. Further characterization of the shear properties was obtained in the dy-
Figure 23. Steady Shear Viscosity: LLD-1 190 °C, LLD-2 170 °C.
Figure 24. Dynamic Shear Behavior: LLD-1 190 °C, LLD-2 170 °C.
namic mode. The complex viscosity \( \eta^* \) and the elastic modulus \( G' \) were measured from 0.1 to 100 rad/s. In the limit of low shear rate, the first normal stress difference is given by \( N_1 = 2G' \). This relation holds at higher shear rates for some materials and was used in this investigation to provide a measure of the materials' elastic behavior in shear.

The characterization of LLD-1, LLD-2 and LLD-1/FE blends in extension was performed in a Rheometrics model RER 9000 extensional rheometer. The materials were molded into cylinders and attached to platens at each end by epoxy. The sample was then inserted into the rheometer's oil bath and brought up to test temperature. The density of the oil was chosen to match that of the sample to eliminate buoyancy effects. The sample was then extended at a constant extension rate and the force measured. Knowing the extension rate, the initial sample dimensions and assuming uniform deformation, the extensional stress and viscosity \( \bar{\eta}_e \) were obtained.

### 3.2.3 Capillary Apparatus

The experiments examining the surface melt fracture of LLDPE as a function of die \( L/D \) and FE concentration were performed using an Instron model 3211 capillary rheometer. The basic principles involved in the design of the instrument are quite simple. First, the plunger forcing the material in the barrel through the capillary is mechanically driven at a constant velocity. The force required to drive the material through the capillary is measured by a load cell at the top of the plunger. Knowing the force and the barrel diameter, the pressure exerted on the fluid by the plunger can be calculated. If the diameter of the barrel is much larger than the capillary diameter \( D_b/D_c > 10 \), then the pressure drop from the plunger to the capillary exit is assumed to occur solely in the capillary. Given the pressure drop and the capillary dimensions the apparent shear-rate, \( \dot{\gamma}_a \), and wall shear stress, \( \tau_w \), can be calculated. If desired, Rabinowitsch corrections for power-law behavior can be applied to obtain a corrected wall shear-rate, \( \dot{\gamma}_w \). In addition to the Rabinowitsch correction, Bagley corrections for entrance pressure losses can be applied. This is accomplished by
using a series of capillaries with the same diameter, but different L/D's. A plot of the pressure versus L/D at constant shear-rate is constructed and a linear regression performed. Extrapolation of the fit to zero L/D yields the entrance pressure loss for a given shear-rate. In the current study, the bulk of the results are presented in the uncorrected (apparent) form. The reason for this is two-fold. First, the method used to determine shear-rates in the companion slit die experiments were based on calculations using the flow rate and not pressure drops. As a result, there was no way to correct for any entrance pressure losses in these experiments. Second, the question of whether or not slip is present at the wall during melt fracture is still open to debate. If slip is present, then the assumptions used to correct for non-Newtonian behavior (no wall slip) are no longer valid. These two considerations lead to the adoption of using apparent values, rather than corrected values.

The capillaries used in all experiments had a 90° included entry angle, a diameter of 0.686mm and were constructed of tungsten carbide. Three different capillary L/D's (12.5, 37.1 and 75.1) were used to evaluate the surface melt fracture behavior of all the materials studied. Each experiment was run three times and between runs involving different LLD-1/FE blends, the barrel and capillary were purged by running two full barrels of unblended LLD-1 through the system to minimize the effects of residual FE on the walls. The order of runs for the blend studies was from lowest to highest concentration of FE, in order to minimize any residual FE effects.

3.2.4 Slit Die Apparatus

It was suggested by the effectiveness of blending in small amounts of fluoro-elastomer in eliminating surface melt fracture in LLDPE to explore the use of FE coatings as a tool for examining the role of the entry and exit regions in surface melt fracture behavior. Two different slit die geometries were used in this study. The first was a planar 4:1 contraction die designed by White [84] and the second was an exit flow die designed by Read [85]. These dies are shown schematically in Fig. 25. Because windows were incorporated in the design of the dies, it was possible to monitor
the effect of the FE coating on the stress field in the specific region, as well as its effect on the surface melt fracture behavior. In this sub-section, a description of the apparatus, as well as the procedure used in performing the slit die experiments are given.

The arrangement of the equipment used in the slit die experiments is shown in Fig. 26. The melt was fed to the dies using a Killion 25.4mm single screw extruder. Flow rates were determined by collecting samples of the extrudate for a fixed period of time (two minutes) and weighing them. Apparent shear rates in the die were determined from the mass flow rate, melt density and die cross-section dimensions by \( \dot{\gamma}a = 6\dot{m}/\text{WHP} \). Melt temperatures of the material exiting the extruder were monitored by a melt thermocouple located at the end of the extruder barrel. The exit die contained two independently controlled heating zones, while the planar contraction die contained three zones. The die temperature was maintained within 2 °C of the melt by Omega model CN9000 temperature controllers. A complete description of the optical arrangement and alignment is reserved for the last part of this section.

The fluoro-elastomer coatings used in the slit die studies were prepared by dissolving 2 grams of 3M Dynamer L-10040 in 13 grams of acetone. The acetone was measured and placed in a glass beaker containing a magnetic stirring bar. The FE was measured and added gradually over the course of 20 minutes to the stirred solution. It was necessary to add the FE slowly to avoid clumping of the material. The solution was then allowed to continue mixing for 4 hours. A thin coating of the solution was then applied to the appropriate region of the die using a fine brush and allowed to dry for 24 hours prior to assembling the die. The coating was approximately 0.005 inch thick, but an accurate measurement was not possible. Prior to running any material through the coated dies, the coating was allowed to bake at test temperature for 2 hours. In separate experiments, the coating was applied to the re-entrant corner, the exit corner and the entire flow region of the planar contraction die. Similarly, coatings were applied to the exit corner, in symmetric transverse strips in the land region, and the last 5 L/H's on one side of the die only. The coating locations used in both geometries are shown in Fig. 27. Between experiments using coatings in
Figure 25. Slit Die Geometries: (a) 4:1 planar contraction, (b) exit.
Figure 26. Slit Die Apparatus: (a) extruder, (b) slit die, (c) optics.
different regions, the disassembled die was soaked in acetone overnight and scrubbed to remove the previous coating.

The operating procedure used in conducting the slit die experiments was as follows:

1. The FE coating is applied to the desired region of appropriate die.

2. The die, including the windows, is assembled. The bolts should be snug, but not tight.

3. The die is attached to the extruder and brought up to the desired test temperature gradually (no more than 1 °C per minute to avoid cracking the quartz windows).

4. After the die has reached the desired temperature, all of the bolts holding the die halves together are tightened. The bolts holding the window frames to the die should be hand tight, a ratchet or wrench should not be used.

5. The coating on the die is allowed to bake at test temperature for 2 hours.

6. The power to the extruder is turned on and the barrel is brought up to test temperature.

7. The optics are aligned as described at the end of this section.

8. The extruder drive motor is engaged and the desired screw rpm selected.

9. When steady-state is reached (melt temperature is a good indicator), a sample of the extrudate is collected for a fixed period of time (two minutes is recommended) and weighed. The screw rpm is adjusted as required to obtain the desired flow rate. It may be necessary to adjust the temperature in the last zone of the extruder to maintain the proper melt temperature at different flow rates.
Figure 27. Fluoro-elastomer Coating Locations: (a) planar contraction die: re-entrant corner, exit corner, entire region. (b) exit flow die: exit corner, symmetric strips, asymmetric land and exit region.
10. When the desired flow rate and melt temperature are achieved, an extrudate sample is collected for analysis and the birefringence pattern is recorded on the VCR. It is recommended that after the sample has been collected and the steady-state birefringence recorded, the relaxation of the stress field be recorded by turning off the extruder with the video still running.

11. The screw rpm for the next flow rate is selected and steps 9 and 10 repeated.

3.2.5 Optical Apparatus and Alignment

The use of the visualization dies discussed previously provided the opportunity to examine the effect of the FE coatings on the stress field in the flow region as well as the coating's effect on the quality of the extrudate. Addition of FE to LLDPE as a blend caused the material to become optically opaque in birefringence measurements, not allowing the role of the FE on stress field behavior to be determined. However, using FE as a coating in the die and using the optically clear, unblended LLDPE, it was possible to examine the effect of the FE coating on the stress field development in various regions of the die. In this sub-section, the optical apparatus and procedure used for aligning the optics is presented. A description of the theory involved in birefringence measurements is presented in Appendix A. It is sufficient at this point to note that only isochromatic patterns were obtained in the slit die experiments.

The optical apparatus used in the slit die experiments is shown schematically in Fig. 28. The light source used for the birefringence experiments was a monochromatic laser provided by Spectra Physics (model 155, \( \lambda = 632.8 \) nm). The beam was enlarged by a 10x objective microscope lens, so that the desired flow region was illuminated. The enlarged beam then passed through a collimating lens prior to entering the plane polarizer and 1/4 wave plate. In the slit die experiments a combination polarizer-1/4 wave plate lens (Oriel circular film polarizer) was used. The beam then
passed through the sample and a second pair of polarizers and the image recorded on video tape using a Panasonic WV-3100 video camera. The optical arrangement resulted in isochromatic birefringence patterns with half-order fringes (1/2, 3/2, etc...) being generated.

In order to obtain quality birefringence patterns, proper alignment of the optics was of paramount concern. The procedure used in aligning the optics was as follows:

1. The optical rail, with no lenses attached, is placed on the table beneath the die.

2. The laser is aimed at the nearest metal edge of the die near the desired flow region. *THE BEAM SHOULD NOT PASS THROUGH BOTH WINDOWS OF THE DIE!*.

3. The reflected beam is located and the laser direction adjusted so that the reflected beam passes directly back into the laser, while the incident beam is still aimed at the desired edge. A piece of plain white paper facilitates the location and adjustment of the reflected beam.

4. The collimating lens, supported independently of the optical rail, is placed such that the light beam passes through the focal point. A ring stand is suitable for supporting the lens and should be placed so that there is room on the optical rail between the lens and the laser for mounting the objective lens.

5. The collimating lens is aligned laterally and vertically so that the incident beam is again located at the edge of the die and the reflected beam is passing directly back into the laser source. Stray beams must be eliminated by proper alignment of the collimating lens.

6. The optical rail is adjusted so that it is in line with the laser beam and the collimating lens.

7. The 10x objective lens is attached to the optical rail between the laser and the collimating lens. The lens is adjusted until the broadened beam illuminates the desired flow region.
Figure 28. Optical Apparatus: (a) monochromatic laser, (b) 10x objective lens, (c) collimating lens, (d) plane polarizer, (e) 1/4 wave plate, (f) die, (g) video camera, (h) optical rail.
8. The shutter on the laser is closed and the video camera is attached to the optical rail.

9. Using an external light source to illuminate the desired flow region, the position and zoom of the camera are adjusted until the flow region is at the desired magnification and in focus.

10. The external light source is turned off and the laser shutter opened.

11. The polarizing lenses are attached to the optical rail on either side of the die. The lenses should be normal to the light beam and the white marks on the lenses should be facing each other.

12. The experiments are the run as described earlier.

3.3 Results and Discussion

The results of the surface melt fracture studies of LLDPE are presented and discussed in this portion of the chapter. The effect of capillary L/D is examined first, followed by the capillary studies and the rheological characterization of the LLD-1/FE blends. The fourth and fifth sections address the effect of FE coatings on the extrudate quality and flow birefringence, respectively. In the final section the implications of the individual results to the mechanism of surface melt fracture is discussed.

3.3.1 Capillary L/D Experiments

Three capillary dies with a diameter of 0.686mm and L/D were used to examine the effect of capillary length on the surface melt fracture behavior of LLD-1. The experiments were conducted
over a range of shear-rates from 37$r^{-1}$ to 2250$r^{-1}$ and each experiments was repeated a total of three times. In general, the repeatability of the results was excellent as indicated by the error bars on the plots. The effect of capillary length on the flow curves and the quality of the extrudate are presented and discussed in the following paragraphs.

The effect of capillary length on the flow curve for LLD-1 is shown in Fig. 29. The apparent shear stress for the shortest capillary appears to be slightly higher than the stresses for the two longer capillaries over the entire range of shear rates, but except for the highest three rates, the slight difference is within the experimental error of the measurements. The slightly higher shear stresses observed for the shortest capillary are indicative of entrance pressure losses, and when Bagley corrections are applied, the apparent differences are eliminated (Fig. 30). It is suggested by these results, that the entry region behavior for LLD-1 can not be overlooked.

A comparison of the effect of L/D on the quality of the extrudate surface at 37.5$r^{-1}$, 75$r^{-1}$ and 225$r^{-1}$ is shown in Fig. 31. At 37$r^{-1}$, the onset of surface fracture has occurred for the shortest capillary, while the extrudate from the longer capillaries is still smooth. As the shear rate is increased, the appearance of the extrudates become more similar, corresponding to the shear stress behavior shown in the apparent flow curve. At a shear rate of 225$r^{-1}$, there is no significant difference in the severity of the surface fracture or the shear stress in the apparent flow curves. It is indicated by these results that the influence of capillary L/D is only significant at the onset of surface fracture and this influence diminishes as the shear-rate and severity of fracture increases. Additionally, while the slight differences observed in the apparent flow curve are questionable in terms of the experimental error, they more accurately reflect the quality of the extrudate surface. That is to say, for shorter capillary lengths, the contribution of the entry region to the stress in the fluid should not be eliminated when considering the surface melt fracture behavior.

It has been suggested that for shorter capillary lengths, the entry region of the die may play a significant role in determining the onset of surface melt fracture. Viewing the results in terms of the mechanism proposed by Kurtz [19], the observed behavior is not surprising. The impact of the entry
Figure 29. Effect of L/D on Flow Curve, Apparent Values.
Figure 36. Effect of L/D on Flow Curve, Bagley Corrections Applied.
Figure 31. Effect of L/D on Extrudate Surface: (a) L/D = 12.5, (b) L/D = 37.1, (c) L/D = 75.1.
region should be manifest in the 'pre-stressing' or transition to the 'high elastic state' required prior to the critical acceleration of the melt upon exiting the die. It is well established that the stresses generated in a melt as it passes through a contraction are substantially higher than those realized downstream of the contraction in the land region. With increasing capillary length, the stresses generated in the entry region have sufficient time to relax, and the stress in the melt as it prepares to exit the die is equivalent to the stress developed in the land region of the die. The stress in the melt as it prepares to exit a short die, however, would be higher than the equivalent stresses developed in the land region and more heavily weighted toward the stresses developed in the entry region.

### 3.3.2 LLD-1/FE Blend Capillary Studies

The effect of fluoro-elastomer (FE), when blended with LLD-1, on the surface melt fracture behavior was examined in the capillary apparatus. Experiments were conducted with FE concentrations of 200, 500, 1000 and 2000ppm, over a range of shear-rates from 37.5\(\text{s}^{-1}\) to 2250\(\text{s}^{-1}\). The three capillary dies used in the previous set of experiments were also incorporated in the LLD-1/FE blend studies. The experiments were conducted at 190°C and each experiment was repeated three times. The effect of blending FE with LLD-1 on the quality of the extrudate surface and on the flow curves is presented in the following paragraphs.

As shown in Fig. 32, the surface melt fracture behavior of LLD-1 was completely eliminated for all levels of FE examined. In addition, the onset of gross melt fracture was also delayed as evidenced by comparing the quality of the extrudates at 1125\(\text{s}^{-1}\). The pure material has clearly undergone gross fracture, while the blend extrudates are still free of any fracture. The question which needs to be addressed is how the FE affects the behavior of LLD-1 to eliminate the surface melt fracture behavior.
Figure 32. Effect of FE on Extrudate, $\dot{\gamma} = 1125 \text{s}^{-1}$: 0, 200, 500, 1000 and 200 ppm FE.
(a) $L/D = 12.5$, (b) $L/D = 75.1$. 

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The effect of FE on the flow curve for LLD-1 is shown in Fig. 33 for an L/D of 12.5 and in Fig. 34 for an L/D of 37. Over the range of shear rates associated with surface melt fracture for the pure material, there is a marked reduction in the apparent shear stress levels for the LLD-1/FE blends. It was suggested in the previous section, that for the pure material at low shear-rates, that the entry region contributed to the surface melt fracture behavior through the entrance pressure. This raises the question of whether or not the observed reduction in the apparent stress values is the result of decreasing the entrance pressure contribution. The answer to this question is that it does not, as shown in Fig. 35. Above an apparent shear-rate of 225 s\(^{-1}\), the addition of FE actually serves to increase the entrance pressure contribution to the apparent shear stress. This being the case, then the question of whether or not the appropriate criteria for the onset of surface melt fracture should be the corrected, rather than the uncorrected, shear stress. The effect of the FE on the corrected shear stress values is shown in Fig 36. For all levels of FE, there is a corrected stress level higher than that required for the onset of surface melt fracture in the pure LLDPE. This indicates that corrected stress levels do not explain the elimination of surface melt fracture in LLD-1.

It is obvious from the results of the LLD-1/FE blend capillary studies, that while FE serves to reduce the stress levels in the pure material, the elimination of the surface melt fracture cannot be explained simply in terms of this effect. In addition, at moderate shear-rates, the FE actually serves to increase the entrance pressure for LLD-1 eliminating this behavior as a possible explanation. The question still remains of how the FE affects the LLD-1 to eliminate the surface melt fracture behavior. In the sections that follow, the results of the experiments conducted to determine whether FE affects the bulk rheological properties of LLD-1 or the melt/metal interface are presented and discussed. It is hoped that by identifying the role of FE in eliminating surface melt fracture, the factors and conditions leading to the onset of this behavior can be identified.
Figure 33. Effect of FE on the Apparent Flow Curve of LLD-1, L/D = 12.5; 0, 200, 500, 1000 and 2000 ppm FE.
Figure 34. Effect of FE on the Apparent Flow Curve of LLD-1, L/D = 37.1: 0, 200, 500, 1000 and 2000 ppm FE.
Figure 35. Effect of FE on the Entrance Pressure: 0, 200 and 2000 ppm FE.
Figure 36. Effect of FE on the Corrected Flow Curve of LLD-1, L/D = 37.1: 0, 200, 500, 1000 and 2000 ppm FE.
3.3.3 LLD-1/FE Blend Rheology

Results of the LLD-1/FE capillary study clearly showed the effectiveness of FE in eliminating the surface melt fracture behavior of LLD-1, but failed to provide any explanations. The question of FE affecting the rheology or boundary behavior was raised and in this section, the effect of FE on the melt rheology of LLD-1 is addressed.

The behavior of the LLD-1/FE blends was evaluated in steady and dynamic shear in the mechanical spectrometer. The effect of FE on the steady shear viscosity is shown in Fig. 37 and the complex viscosity in Fig. 38. A slight reduction in both the steady and complex viscosity is observed with the addition of FE to LLD-1. This result is consistent with the observed reduction of the shear stresses discussed in the previous section, but again, the observed differences in the viscosities are by no means significant enough to explain the observed difference in melt fracture behavior. This is also true of the effect of FE on G', shown in Fig. 39. Clearly, there is no significant difference in the elastic modulus with the addition of FE. It is indicated by these results that, in a region of flow dominated by shear behavior, there should be no difference in the rheological behavior of the fluids.

Characterization of the LLD-1/FE blends in extension was performed at extension rates of 0.06, 0.10 and 0.3 s⁻¹. The effect of FE on the extensional stress growth behavior is shown in Fig. 40. At the lower two rates, there is no observed difference in the extensional behavior with the addition of FE. At the highest rate, the FE serves to increase the extensional stress of LLD-1. The observed increase in the extensional stress growth behavior at higher extension-rates is consistent with the effect of FE on the entrance pressure discussed in the previous section. The entrance pressure provides a measure of the energy required for the flow to rearrange as it accelerates through the contraction. Thus, if the FE serves to increase the magnitude of the extensional stress growth behavior at higher rates, it should be expected to increase the entrance pressures observed in the capillary experiments. While the FE does increase the magnitude of the extensional stresses at higher exten-
Figure 37. Effect of FE on the Steady Shear Viscosity: 0, 200, 500, 1000 and 2000 ppm FE.
Figure 38. Effect of FE on the Complex Viscosity: 0, 200, 500, 1000 and 2000 ppm FE.
Figure 39. Effect of FE on the Elastic Modulus: 0, 200, 500, 1000 and 2000 ppm FE.
sion rates, there is no observed change in the type of extensional behavior with the addition of FE. That is to say, a transition from bounded to unbounded stress growth is not observed. The results of the extensional characterization indicate that, for regions of flow with a weak extensional component, the impact of FE on the rheological behavior should not be significant. For regions of flow with a strong extensional component, the FE would serve to increase the magnitude of the stress in the fluid, but not drastically enough to explain the observed differences in extrudate quality. In fact, the increase in stress at higher extension rates would be expected to increase the severity of melt fracture rather than serve to eliminate the phenomenon.

The results of the characterization demonstrate that FE does not affect the rheology of LLD-1 sufficiently to explain the elimination of surface melt fracture behavior. This points to role of FE as affecting the melt/metal boundary behavior for LLD-1. If the elimination of surface melt fracture by the addition of FE is the result of boundary interaction, it is reasonable to expect that the use of FE as a die coating, rather than as a blended additive, should also serve to eliminate surface melt fracture. Finally, if FE coatings do eliminate surface melt fracture, they should serve as a useful tool for determining the region of a die responsible for the initiation of surface melt fracture. The use of FE as a die coating is the subject of the next section of this chapter.

3.3.4 Effect of FE Die Coatings on Surface Melt Fracture

The previous studies examining the effect of blending FE with LLD-1 to eliminate surface melt fracture and the lack of effect of FE on the bulk rheological properties suggested using FE as coating in a die to further study the surface melt fracture behavior of LLDPE. If the role of FE is to influence the melt/metal boundary behavior, then applying a FE coating to the die should effect the same result. By applying the FE coating to different regions of a die and examining the quality of the extrudate, it should be possible to identify the region of the die responsible for initiating the surface melt fracture behavior of LLDPE. The effect of FE coatings on the extrudate quality is
Figure 40. Effect of FE on the Extensional Stress Growth Behavior of LLD-1: 0, 200, 500 and 1000ppm FE, (a) $\dot{\varepsilon} = 0.06\text{s}^{-1}$, (b) $\dot{\varepsilon} = 0.10\text{s}^{-1}$, (c) $\dot{\varepsilon} = 0.30\text{s}^{-1}$.
presented in this section. Presentation of the effect of these coatings on the birefringence patterns is reserved for the next section.

Experiments incorporating FE coatings were conducted in both a planar 4:1 contraction die and an exit flow die. The locations of the coatings were shown previously in Fig. 27. In the entry die studies, the influence of the coatings on both LLD-1 and LLD-2 surface melt fracture behavior was examined, while experiments in the exit die were restricted to LLD-2 only. The range of shear rates examined in the entry die was between 10 and 60 s⁻¹, while the upper limit for the exit flow die was 50 s⁻¹. In both cases, the limiting factor was the maximum flow rate developed by the extruder feeding the dies. Prior to applying coatings to either the entry or exit dies, the onset of surface melt fracture as a function of shear rate in the uncoated dies was determined for both LLD-1 and LLD-2. These corresponded to an apparent shear rate of 29 s⁻¹ for LLD-1 in the entry die and 30 s⁻¹ for LLD-2 in both the entry and exit dies.

The results of coating the re-entrant corners of the planar contraction indicate, that for both LLD-1 and LLD-2, surface melt fracture is eliminated over the entire range of the experiments. A comparison of the extrudate from the uncoated and coated re-entrant corner experiments for LLD-1 is shown in Fig. 41. Similar results were observed when the exit region of the planar contraction die was coated: surface melt fracture was eliminated over the entire range of rates for both materials. A comparison of the LLD-1 extrudate from the uncoated and coated exit region is shown in Fig. 42. The results of the entry die experiments support the earlier results of LLD-1/FE blend studies in finding that the role of FE in eliminating surface melt fracture is manifest at the melt/metal boundary rather than in affecting the rheology. However, these results present conflicting arguments for the site responsible for initiating surface melt fracture. A detailed discussion of the observed behavior follows.

The results of the entry die coating studies appear to present arguments for both the entry region and the exit region as the site responsible for initiating surface melt fracture in LLDPE. The elimination of surface fracture when the re-entrant corners are coated, in conjunction with the earlier
Figure 41. Effect of Coating the Re-Entrant Corners in the Entry Die: (a) uncoated die, $\dot{\gamma}_d = 42\text{s}^{-1}$, (b) uncoated die, $\dot{\gamma}_d = 52\text{s}^{-1}$, (c) re-entrant corners coated, $\dot{\gamma}_d = 42\text{s}^{-1}$, (d) re-entrant corners coated, $\dot{\gamma}_d = 54\text{s}^{-1}$.
Figure 42. Effect of Coating the Exit Region in the Entry Die: (a) uncoated die, $\gamma_e = 42s^{-1}$, (b) uncoated die, $\gamma_e = 52s^{-1}$, (c) exit corners coated, $\gamma_e = 44s^{-1}$, (d) exit corners coated, $\gamma_e = 55s^{-1}$. 
studies examining the influence of capillary length on surface fracture, present a strong argument for the entry region being the most probable die region for initiating fracture. Unfortunately, the elimination of surface fracture when the exit region is coated argues against the entry region. If the entry region were responsible for initiating surface fracture, the only explanation for eliminating the fracture when the exit is coated, is that coating the exit attenuates the fracture. Considering that the coating applied to the exit region is less than a single die height in length, it is unreasonable to assume that existing fracture could possibly be attenuated in this short a length. Clearly, this suggests that the exit region of the die is the most probable site for initiating fracture, but how then, are the conflicting entry coating results resolved. The best explanation is that as the LLDPE flows over the coated re-entrant corner, FE from the surface is picked up by the melt and carried downstream to the exit region where surface fracture is initiated. While this explanation appears speculative, results of surface analysis (ESCA) of the extrudate does indeed show a 0.1% concentration of fluorine, which is absent from the extrudate of the uncoated experiments. Judging by the LLD-1/FE blending experiments, as little as 0.02% FE is a sufficient concentration to eliminate surface melt fracture behavior. Additionally, experiments in the exit flow die with thin transverse strips of coating upstream of the exit, in the land region, also suggest that the pick-up of FE by the melt is not an unreasonable explanation. This will be discussed in more detail after the results of the exit die have been presented in the following paragraph.

The results of applying a FE coating to the exit region of the exit flow die were identical to the results found for coating the same region of the entry die. Surface fracture was eliminated over the entire range of shear rates attainable. In addition to the exit region being coated, experiments were conducted with FE coatings applied in thin, symmetric transverse strips 5 L/H's upstream of the exit and to the last 5 L/H's of one side of the die only (see Fig. 27). The result of the symmetric transverse coating was to eliminate surface fracture over the entire range of rates. The elimination of fracture in this coating scheme provides additional support to the argument for the melt picking up FE as it passes over a coated region and being carried downstream to the exit. The results of the asymmetric land/exit coating were the most interesting of the exit die experiments. The surface of
the extrudate on the side that was coated, remained free of surface fracture over the entire range of rates examined. The surface of the extrudate from the uncoated side of the die still exhibited surface fracture, however the onset of fracture was delayed to an apparent shear rate of 40 s\(^{-1}\). The significance of this result will be discussed thoroughly in the next section dealing with the effect of coatings on the observed birefringence, but for now it is sufficient to point out that a reduction of the stress alone in the exit region is not sufficient to eliminate surface melt fracture in LLDPE.

The results of the FE coating studies on the quality of the extrudate surface for both LLD-1 and LLD-2 confirm that the role of FE in eliminating surface melt fracture is felt at the melt/metal boundary of the flow. Furthermore, the experiments point to the exit region of the die as being the site responsible for initiating surface melt fracture in LLDPE. These results do not, however, provide any insight as to how the FE effects the behavior of LLDPE at the melt/metal boundary. Toward this end, experiments monitoring the effect of FE coatings on the observed flow birefringence patterns were conducted. The results and implications of these experiments is presented in following section.

3.3.5 Effect of FE Die Coatings on Flow Birefringence

The previous experiments confirm that the effect of FE in eliminating surface melt fracture in LLDPE is manifest at the melt/metal interface in the die. Additionally, the FE coating experiments strongly point to the exit region as the site for initiating surface melt fracture in LLDPE. In this section, the effect of FE coatings in the different die regions on the stress field in the entry, land and exit region of the die is presented. The results are limited to the behavior observed for LLD-2, due to insufficient quantities of LLD-1.

Flow birefringence patterns of the entry region in the planar contraction die were recorded for the uncoated, re-entrant corners coated and exit region coated experiments described previously.
Under all conditions examined, the patterns in the entry were steady, i.e. there were no oscillations in the patterns even during surface fracture conditions in the uncoated die. A comparison of the entry region birefringence at an apparent shear rate of $22r^{-1}$ is shown in Fig. 43. A similar comparison at an apparent shear rate of $f_{42.42}$ s sup -1 is shown in Fig. 44. These two comparisons correspond to behavior prior to and well after the onset of surface melt fracture in the uncoated die. The asymmetry in the uncoated patterns was observed over the entire range of shear rates and again, there was no observed fluctuations in the entry patterns as has been reported for conditions of gross fracture for other materials. An explanation for the cause of the asymmetry is not apparent. The asymmetry in the stress field was eliminated when coatings were applied to either the re-entrant corners or the exit region and indicates that the asymmetry is in some way tied to the factors which lead to the surface melt fracture behavior of LLDPE. Comparing the coated re-entrant corner and coated exit corner entry region patterns shows there is no significant difference between the two. These results support the earlier suggestion that the benefit of coating the re-entrant corner is not felt at the entry, but downstream at the exit. If coating the re-entrant corners was affecting some behavior uniquely associated with the entry region, some kind of difference in the different coating entry patterns would be expected. This is not the case.

The birefringence patterns observed in the entry region do not provide any direct evidence suggesting that it is the flow in this region which leads to surface melt fracture. The results do show that there is an asymmetry in the stress field in the entry region when the die is uncoated (Figs. 43(a) and 44(a)), but the asymmetry is decreased at the higher rate (corresponding to the condition of surface melt fracture) and completely eliminated by coating either the re-entrant or exit corners. A lack of observed difference between the birefringence patterns for the different coating locations again suggest that the exit region is of paramount concern in the surface melt fracture behavior of LLDPE. Experiments examining the birefringence in the exit region under different coating conditions is the next topic to be addressed.

The experimental results of the current study, to this point, collectively indicate that it is the exit region in a die which is ultimately responsible for initiating surface melt fracture in LLDPE.
Figure 43. Effect of FE Coatings on the Entry Region Birefringence: (a) uncoated die, $\gamma_x = 22s^{-1}$, (b) re-entrant corners coated, $\gamma_x = 21s^{-1}$, (c) exit corners coated, $\gamma_x = 23s^{-1}$.
Figure 44. Effect of FE Coatings on the Entry Region Birefringence: (a) uncoated die, $\dot{\gamma}_e = 43 s^{-1}$, (b) re-entrant corners coated, $\dot{\gamma}_e = 41 s^{-1}$, (c) exit corners coated, $\dot{\gamma}_e = 44 s^{-1}$. 
Additionally, the experiments show that the role of FE in eliminating surface melt fracture is to affect the melt/metal boundary behavior of the flow. The remaining question is in what manner does the FE affect the boundary behavior to eliminate surface melt fracture. Toward this end, two sets of experiments were conducted using the exit flow die. The first set examined the effect of coating the exit corner with FE on the birefringence patterns in the exit region and the second examined the effect of an asymmetric coating of FE applied to the land and exit region on the birefringence. The results of these experiments are discussed in the following paragraphs.

The effect of coating the exit region on the flow birefringence was examined at apparent shear rates corresponding to those previously studied in the planar contraction die. In addition to monitoring the steady-state patterns, the relaxation of the stress field upon cessation of flow was also recorded. This allowed for an accurate determination of the number of shear and extensional fringes at a particular set of conditions. A comparison of the steady-state exit region birefringence for the uncoated and coated exit corners at rates below and above the onset of surface fracture is shown in Fig. 45. In addition to determining the number of shear and extensional bands at a given rate, qualitative measures of the upstream distance where the flow first started to re-arrange and the length over which the flow re-arranged were made. The point for initiating re-arrangement was taken to be where the shear bands in the center of the flow started to merge. A measure of the distance required to re-arrange was taken to be from the point where the flow first started re-arrange to the center of the maximum extensional band region. The results of these measurements are tabulated in Table 1. The results provide only a trend of behavior and are by no means conclusive in regards as to how the presence of FE serves to eliminate surface melt fracture. The most significant trend is the distance upstream that the flow starts to re-arrange. The FE coating causes the flow to start re-arranging earlier than in the uncoated case, but in terms of the overall length, there is no significant difference in the behavior. It is shown by examination of the birefringence patterns that there is a tendency of slightly reduced extensional band behavior, but again, the observed differences are not significant enough to explain the role of the FE in eliminating surface fracture behavior. The question of how the FE coatings influence the boundary behavior is not resolved by
the exit birefringence patterns. However, results of the asymmetric land/exit region coating do provide more conclusive evidence and are discussed in the following paragraph.

It was indicated by the experiments examining the effect of the coating on the exit corner on the exit birefringence patterns that only slight trends were observable and these were not significant to the point of establishing the role of FE in eliminating surface fracture behavior. In a final attempt to address this question, a FE coating was applied to the last 5 L/H's of the exit die on one side only (see Fig. 27). The birefringence at the point where the flow first encounters the coating was recorded over the range of shear rates used in the previous studies. The benefit of examining the interaction in this region was that the flow field is well defined, i.e. the flow was not undergoing the complex re-arrangement of entering a contraction or exiting the die. As a result, the interaction between the FE coating and the melt was isolated and could be examined directly. The results of the effect of the asymmetric coating on the flow birefringence patterns are shown in Fig. 46. The stresses in the flow field start to relax about 0.5 L/H upstream on the side of the die which is coated. In addition to a reduction in the stress field, examination of the down field fringes shows an asymmetry in the band patterns across the die. The distance between the bands is increased on the coated side of the die and the spacing is also more uniform. These results are indicative of a reduced gradient at the wall and point directly to FE introducing slip at the wall. This result is interesting in its implication on the factors which ultimately lead to the onset of surface melt fracture in LLDPE and will be discussed in more detail in the section. For now, it is sufficient to conclude that the role of FE at the melt/metal boundary is to introduce slip. The failure to observe this behavior in the entry and exit region coating experiments is most likely the result of the complex nature of the flows in these areas. The reduction of the stresses at the walls in the earlier experiments was most probably masked by the increase in the extensional components of the total stress resulting from the re-arrangement of the flow.
Figure 45. Effect of FE Coatings on the Exit Region Birefringence: (a) uncoated die, $\dot{\gamma}_x = 24 s^{-1}$, (b) exit corners coated, $\dot{\gamma}_x = 24 s^{-1}$, (c) uncoated die, $\dot{\gamma}_x = 35 s^{-1}$, (d) exit corners coated, $\dot{\gamma}_x = 35 s^{-1}$. 
### Table 1. Summary of Exit Coating Experiments on Exit Region Birefringence

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Figure 46. Effect of Asymmetric FE Coatings on the Flow Birefringence:  
(a) $\dot{\gamma}_s = 8 \, s^{-1}$,  
(b) $\dot{\gamma}_s = 11 \, s^{-1}$,  
(c) $\dot{\gamma}_s = 17 \, s^{-1}$  
(d) $\dot{\gamma}_s = 22 \, s^{-1}$.
3.3.6 Final Considerations

The discussion of results presented so far has focussed primarily on specific points regarding the melt fracture behavior of LLDPE and the role of FE in eliminating this phenomenon. In this section, the implications of the specific points on the overall understanding of surface melt fracture behavior is discussed.

The results of all the experiments conducted in this study were consistent with the two step mechanism suggested by Kurtz [19] for the surface melt fracture behavior of LLDPE. The results indicate that a transition to the 'high elastic state' in conjunction with a critical acceleration as the melt exits the die is necessary for surface melt fracture to occur. The coupling of the two requirements is best demonstrated by considering the results of LLD-1/FE blend capillary study. When 200ppm FE was blended with LLD-1, the effect was to lower the capillary stresses and completely eliminate surface melt fracture over the entire range of shear rates. Even applying the normal corrections to the apparent stress values, the extrudate was free of surface defects to above 0.30MPa, well above any critical shear stress values reported in the literature. This indicates that the LLD-1/FE blends were sufficiently 'pre-stressed' but failed to undergo surface fracture. The most probable reason the material failed to exhibit melt fracture was because the critical acceleration condition was not met. The coated slit die experiments demonstrated that the role of FE at the melt/metal boundary is to introduce a slip condition. The critical acceleration required for the onset of surface fracture occurs as the melt is accelerated rapidly from a zero wall velocity to finite value upon exiting the die. When slip at the wall is introduced, as in the case of FE blends, the required acceleration is reduced and the length over which it may occur is increased.

The second point to be made is in respect to the role of wall-slip in surface melt fracture. It was suggested by Ramamurthy [21] that surface melt fracture was the result of a failure of adhesion at the melt/metal interface and that the resulting wall-slip lead to this behavior. While there was no direct evidence in the uncoated dies under the conditions where surface melt fracture occurred
for the presence or absence of slip, the results of the coating experiments indicate that the role of
the FE coating in eliminating surface melt fracture is to introduce slip at the melt/metal boundary.
This result is in direct contradiction of the mechanism suggested by Ramamurthy.

The final point to be made is in respect to the effect of die L/D on surface melt fracture. While
an earlier onset of surface melt fracture was observed for the shortest capillary L/D, the entry region
region of the die is only important in terms of the stress level in the fluid as it exits the die. This is
best explained by considering the ‘pre-stressing’ step discussed earlier. The role of the entry for
‘short’ dies is to provide the ‘pre-stressing’ of the melt prior to the critical acceleration of the ma-
terial as it exits the die. The ‘pre-stressing’ of the melt in the entry by itself is not sufficient to initiate
melt fracture by itself. As the length of the die is increased, the residual stresses developed in the
entry region relax and the stresses incurred in the land region provide the necessary ‘pre-stressed’
condition.

3.4 Conclusions and Recommendations

In the previous section, the results of the surface melt fracture studies were presented and dis-
cussed. The conclusions resulting from this study and recommendations for future work are pre-
sented in the current section.

3.4.1 Conclusions

1. The exit region of the die is ultimately responsible for initiating surface melt fracture in
LLDPE. The success of coating the exit region with fluoro-elastomer (FE) in eliminating the sur-

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face melt fracture behavior of linear low-density polyethylene (LLDPE) unequivocally isolates the exit region as the site ultimately responsible for initiating this behavior, regardless of mechanism.

2. The role of fluoro-elastomer in eliminating the surface melt fracture of LLDPE is to introduce slip at the melt/metal boundary. It is strongly suggested by the observed birefringence patterns that wall-slip is present in the asymmetric coating experiments. This conclusion is consistent with the reduction of the observed shear stress behavior in the LLD-1/FE blend study.

3. The coupling of a transition to the 'high elastic state' with a critical acceleration of the melt as it exits the die, as suggested by Kurtz, is an accurate description of the experimental behavior observed in this study. The level of stresses generated in the LLD-1/FE blends in the capillary experiments is well above the level required for surface fracture in the samples without FE or any critical values reported in the literature for the onset of surface fracture. As a result, a critical stress by itself is insufficient to initiate surface melt fracture.

4. The role of the entry region in the surface melt fracture behavior of LLDPE is limited to its effect on the 'pre-stressing' step in the overall mechanism. The elimination of surface melt fracture by coating the exit region in the planar contraction coupled with the earlier onset of fracture observed for the shortest capillary die indicate that, for 'short' dies, the high stresses encountered in the entry region can provide the 'pre-stressing' necessary for surface melt fracture, but with increasing length it is the stresses developed in the land region which provide this function.

3.4.2 Recommendations

While some of the questions regarding the site and factors responsible for initiating surface melt fracture in LLDPE have been addressed in this study, there are still questions which remain to be answered. Recommendations for future work in this area are given below.
1. Further examine the role of the entry region on surface melt fracture behavior. An exhaustive study incorporating capillary dies with a range of L/D's from 0.5 to 5 and varying entry angles should provide additional insight to the role of the entry region in this phenomenon. Additionally, examination of the behavior in these dies up to rates resulting in gross melt fracture, which is believed to be an entry driven phenomenon, may determine if any relation exists between the two types of melt fracture.

2. Examine the exit region birefringence when only one of the exit corners is coated with FE. An asymmetric coating of the last 2 or 3 L/H's of the exit region may possibly provide additional information on the surface fracture behavior of LLDPE. The effect of the FE coating on the stress field as the flow encounters the coating as well as the effect on the exit region birefringence could be examined concurrently.

3. Examine the surface melt fracture behavior of high density-polyethylene (HDPE). Establish whether or not the factors influencing the surface melt fracture behavior of HDPE are the same as those for LLDPE.

4. Examine the effect of FE coatings on the wall shear stress by direct measurement of the pressure drop in the slit die. By incorporating pressure transducers in the channel of the slit die, direct measurement of the role of FE on the wall shear stress levels could be determined.
4.0 THE JOINING FLOW BEHAVIOR OF LOW-DENSITY AND LINEAR LOW-DENSITY POLYETHYLENE

The experimental objective of this thesis is to first, understand the factors and conditions leading to the onset of surface melt fracture behavior in single component flows and second, to establish whether or not these factors lead to instabilities in bicomponent interfaces. In the previous chapter, the work executed in establishing the site for initiating surface melt fracture in linear low-density polyethylene (LLDPE), as well as the factors leading to the onset of this behavior, was presented. In this chapter, the work performed in examining the role of surface melt fracture in bicomponent interface instability is presented. In addition to the primary objective, the influence of material elasticity on interface behavior is considered by comparing the joining flow behavior of LLDPE with low-density polyethylene (LDPE).

Two different methods of evaluating interface behavior were employed in this study. The first was to examine the flow birefringence of the fluids as two isolated streams joined together to form a single stratified flow (Fig. 2). Three different layer combinations: LLDPE/LLDPE,
LDPE/LLDPE and LDPE/LDPE, were studied using flow birefringence. The second technique used to study the interface behavior was to color the material in one of the streams and examine the interface of the extrudate using optical microscopy. Layer combinations of LLDPE/LLDPE and LDPE/LLDPE were studied in this manner.

The presentation of the joining flow studies is divided into four parts. First, a brief introduction to bicomponent interface instability is presented and the objectives of the current study clearly defined. This is followed by a description of the apparatus and methodology used to meet the objectives. The results of the joining flow studies are presented and discussed in the third part of this chapter and in the fourth part, the conclusions are drawn from the results and recommendations for future studies are given.

4.1 Introduction

The interface behavior of multi-component stratified flows has been a topic of discussion in the literature since the late 1960’s. Initial interest stemmed from the importance of interface behavior in bi-component fiber spinning processes and focused on the encapsulation of one of the materials by the other. A second type of interface behavior, which is of more concern in the film casting process, is the onset of irregular distortions of the interface at critical processing rates. Unlike the viscous encapsulation phenomenon, where a smooth transition from a side-by-side arrangement to a fully encapsulated one is observed, irregular interface distortions eventually lead to a break-up of the stratified configuration, as shown in Fig. 4. It is this second type of interface behavior, the onset of irregular interface distortions, which is of interest in the present study.

The literature concerning irregular interface behavior, discussed in Chapter 2, is sparse in comparison to the literature addressing the encapsulation phenomenon. The onset of irregular interface
behavior has been attributed to both a difference in the elasticities of the melts [2,5] and to a critical interfacial shear-stress of the melts [3], similar to the single component instability of melt fracture. In regions where there is a re-arrangement of the flow, such as the entry or exit region of an extrusion die, it is reasonable to expect that differences in the relaxation behavior of two materials will have a pronounced effect on the interface, as the stresses at the interface try to balance and the interface reaches an equilibrium configuration. Thus, the suggestion that fluid elasticity plays a role in irregular interface behavior is reasonable. In addition, it is reasonable to expect that factors leading to the onset of instabilities in single component flows would not necessarily be diminished in bi-component flows. Specifically, a material which is subjected to conditions resulting in melt fracture, prior to joining with a second fluid to form a stratified bi-component flow, may well affect the stability of the downstream interface. These considerations lead to the heart of the work presented in this chapter: examination of the influence of upstream conditions, corresponding to surface melt fracture behavior, on the interface behavior as two layers join together and the effect of different relaxation times on interface behavior.

In order to investigate the effects of differing relaxation times and the role of surface melt fracture on bi-component interface behavior, two different polyethylenes were incorporated in the study. The first material was a linear low-density polyethylene (LLDPE) which exhibits the entire range of melt fracture behavior. This material was identified in the surface melt fracture studies of Chapter 3 as LLD-2. Not only does this material provide the desired melt fracture characteristics, but also has a relatively short relaxation time (0.125s). The second material studied was a low-density polyethylene (LDPE) with a much longer relaxation time of 1.4s. Using these two materials in different layer combinations, the effects of upstream conditions and different relaxation behavior were investigated. A visualization die was constructed so that the effect of these variables on the flow birefringence as the two layers joined together could be evaluated. Additionally, experiments were conducted with one of the layers colored so that the extrudate could be examined to identify the interface behavior. The birefringence and optical microscopy studies represent the substance of the work reported in this chapter.
4.2 Experimental Apparatus and Materials

The materials used in the joining flow studies and the experimental apparatus and procedures are presented in this part of the chapter. The materials incorporated in the study and the method of characterization are presented first. A description of the joining flow die and operating procedure for the extruders/die system is presented next. The description of the optical apparatus and operating procedure used in the birefringence studies was presented in Chapter 3 and will not be duplicated here. The final section of this part of the chapter describes the methods used in preparing and examining the extrudate samples for the optical microscopy work.

4.2.1 Materials

The objectives of the current study were to determine the role of surface melt fracture and the effect of differing relaxation times on the interface behavior in a joining flow geometry. To accomplish these goals it was necessary to first identify a material which exhibited the desired melt fracture behavior and fully examine this behavior in single component geometries prior to incorporating the material in a bi-component study. A commercial linear low-density polyethylene (LLDPE, Mobil Chemical Company NTA-101) was chosen and the surface melt fracture behavior thoroughly examined (Chapter 3). A second material was required to examine the role of differing relaxation times on the joining flow behavior. To minimize the effects of viscous encapsulation, a material with similar shear viscosity was desired, unfortunately matching shear viscosity behavior over a wide range of shear-rates for materials with vastly different relaxation times is not practical for polymer melts. As a result, it was decided to match the viscosities at a shear-rate of $0.1 \text{s}^{-1}$. This rate is intermediate in terms of the upstream single component wall shear-rate range and the downstream interface shear-rate range. The material chosen based on these criteria was a commercial low-density
polyethylene (LDPE) provided by Quantum Chemicals (NPE-953). A comparison of the complex shear viscosities is shown in Fig. 47. A description of the rheological characterization methods and results follow.

4.2.2 Rheological Characterization

Shear flow characterization of the LLDPE and LDPE was performed in a Rheometrics Mechanical Spectrometer, model RMS-800. Steady-shear viscosities ($\eta$) and first normal stress differences ($N_1$) were measured from 0.01 to 5.0 s$^{-1}$ and are shown in Fig. 48. Beyond this range, measurements were not possible due to the sample fracturing between the plates. Further characterization of the shear properties was obtained in the dynamic mode. The complex viscosity ($\eta^*$) and the elastic modulus ($G'$) were measured from 0.1 to 100 rad/s. A quantitative measure of the relaxation times of the two materials was obtained by fitting the complex shear data to a single relaxation mode Phan-Thien Tanner Model constitutive equation (see Chapter 5). The results of the fits gave values of the relaxation time ($\lambda$) of 0.125 seconds for the LLDPE and 1.40 seconds for the LDPE.

The characterization of LLDPE and LDPE in extension was performed in a Rheometrics model RER 9000 extensional rheometer. The materials were molded into cylinders and attached to platens at each end by epoxy. The sample was then inserted into the rheometer's oil bath and brought up to test temperature. The density of the oil was chosen to match that of the sample to eliminate buoyancy effects. The sample was then extended at a constant extension rate and the force measured. Knowing the extension rate, the initial sample dimensions and assuming uniform deformation, the extensional stress and viscosity were obtained. The differences in the extensional stress growth behavior of LLDPE and LDPE are clearly demonstrated in Fig. 49. LLDPE exhibits bounded stress growth behavior over the range of extension-rates examined (0.02 – 0.20 s$^{-1}$), while the unbounded behavior of LDPE is evident even at an extension-rate of 0.053 s$^{-1}$.
Figure 47. Complex Shear Viscosity of LLDPE and LDPE, 170 °C.
Figure 48. Steady Shear Properties of LDPE and LLDPE, 170 °C: (a) steady-shear viscosity, \( \eta \), (b) first normal stress difference, \( N_1 \).
Figure 49. Extensional Stress Growth behavior of LDPE and LLDPE, 170 °C:
0.020, 0.053 and 0.20 s⁻¹.
A final comparison of the behavior of LLDPE and LDPE concerns their respective melt fracture behavior. A detailed discussion of the behavior of LLDPE was presented in the previous chapter and will not be duplicated here. The point of interest here can be made by considering the flow curves of the two materials shown in Fig. 50. First, unlike LLDPE, LDPE does not exhibit surface melt fracture behavior. A transition from smooth extrudate to gross fracture occurs at an apparent shear stress, $\tau_a$, of approximately 0.1 MPa. This is at a lower level of stress than that corresponding to the onset of surface melt fracture in LLDPE. The point to be made is that the corresponding shear-rate where gross melt fracture occurs in LDPE is about a full decade beyond the rate corresponding to surface melt fracture in LLDPE. As a result, for equivalent flow rates of the materials, the conditions required for surface melt fracture in LLDPE are well below the onset of gross melt fracture in LDPE.

Prior to moving away from the discussion of the characterization of LDPE and LLDPE there are a few final points to be made. First, while the materials have similar shear viscosities at low shear-rates, at higher shear-rates there is an increasing difference. Thus, while efforts have been made to minimize the influence of viscosity mismatch, the role of viscous encapsulation in experiments conducted at higher shear-rates cannot be overlooked. Second, while the two materials were chosen based on different characteristic relaxation times, the difference in the extensional stress growth behavior will need to be accounted for in considering the flow behavior as the materials in the separate upstream channels accelerate to form a single stratified flow. Finally, experiments examining the role of surface melt fracture behavior will be compared based on equivalent shear-rates rather than equivalent stress levels.

4.2.3 Joining Flow Die

In order to meet the objectives of the current study, it was necessary to construct a die suitable for conducting joining flow experiments. Several factors were taken into consideration prior to the
Figure 50. Flow Curves for LDPE and LLDPE, 170 °C.
design and construction of the joining flow die. First and foremost was the ability to study the stress 
field development as the two layers joined to form a single stratified flow using flow birefringence. 
In order to obtain quality birefringence patterns, a compromise had to be made between making 
the channel wide enough so that the flow was approximately two-dimensional and not so wide that 
the laser beam could not fully penetrate the flow. For the optical apparatus available in the labora-

tory, a one inch channel width has been successfully employed in previous visualization studies 
[84,85] and was taken as the maximum allowable width for the joining flow die. To be able to im-
pose a two-dimensional flow assumption, a 10:1 width-to-height ratio was required. Because the 
behavior of the materials as they joined was of interest, the downstream height dimension was set 
to one-tenth of an inch. This resulted in very thin channels upstream, which had both positive and 
negative implications, which will be discussed in the next part of this chapter. The second factor 
concerning the design of the die was to have fully developed flow of the single component streams 
prior to their joining together. This, coupled with having the ability to measure pressure drops both 
upstream and downstream of the contact point, lead to fixing the upstream and downstream length 
dimensions of the die. To minimize the entry/exit effects, ports for pressure transducers were placed 
5 L/H's away from these regions. The die design which resulted from these considerations is dis-
cussed in the following paragraphs.

The die which was constructed for the joining flow experiments is shown in Fig. 51. The die 
was constructed of 316 stainless steel and consisted of three basic sections: the top plate, the bottom 
plate and the divider/channel section. The top and bottom sections are identical and provide the 
feed ports, pressure taps and form the top and bottom walls of the flow channels (Fig. 52). The 
pressure taps were constructed using custom inserts so that transducers or blanks would be flush 
mounted in the channel. The third section of the die is made of two plates which provide channel 
walls for the top and bottom streams and a divider plate to separate the two streams (Fig. 53). This 
construction allows for varying the height of the two upstream channels by varying the thickness 
of the channel wall plates. Two additional plates were required to provide the channel walls at the 
exit of the die.
Figure 51. Design of the Joining Flow Die.
Figure 52. Top/Bottom Section of the Joining Flow Die.
Figure 53. Channel Wall/Divider section of the Joining Flow Die.
Two windows were positioned on opposite sides of the die so that observation of the streams as they joined could be made. The length of the windowed region was chosen such that the developing flow region upstream and downstream could be viewed. The windows were custom made to specifications by Dell Optics. Initially, the windows incorporated slots to support the divider in the viewing area, but under experimental conditions the edges of the slots chipped and distorted the birefringence patterns. Subsequently, the slots were eliminated and the divider was left unsupported in the joining region. The windows were held in place by frames attached to the top and bottom sections of the die.

The die temperature was controlled by two independently controlled sets of heater bands custom built for the die by Industrial Heater Company. The zones were divided upstream/downstream of the contact point in the die. For the experiments conducted, flush-mount thermocouples were placed in the pressure taps and the melt temperature in the die used for controlling the heater bands via Omega, model CN9000, temperature controllers. A complete description of the temperature control of the incoming melt streams is described in the following section.

### 4.2.4 Extruders/Die Operating Procedure

Two different types of experiments were conducted using the joining flow die. In the first set of experiments, the influence of upstream surface melt fracture conditions and differing relaxation behavior on the flow birefringence patterns in the developing flow region at the contact point in the die was examined. In the second, the influence of these parameters on the shape of the interface in the extrudate was examined. In terms of the apparatus employed and the operating procedure for the system, these two types of experiments differed only in the inclusion/exclusion of the flow birefringence apparatus. In this section, the system and operating procedure for the birefringence experiments are given: the extrudate interface experiment procedure is obtained by neglecting the optical descriptions presented.
The system used in performing the joining flow experiments consisted of 3 basic parts: the feed system, comprised of two Killion 25.4mm single screw extruders, the joining flow die and the optics, shown schematically in Fig. 54. Melt from the extruders was fed to the die through 3/4 inch, schedule 40, galvanized pipe. The piping between the extruders and the die was constructed in two sections, one from the die and the second from the extruders. The two sections were then joined by a union fitting and the entire piping system heated by standard 1 inch heater bands (Wallow, 100W and 200W, 240V) and the steel temperature controlled by the die zone controllers in the extruders. The die used in the experiments was described in the previous section and the optical apparatus was the same as described in Chapter 3.

Experiments conducted in the joining flow die were performed at 170 °C. During the experiments, the temperature of the melt exiting the extruders was maintained at approximately 165 °C and the steel temperature of the piping maintained at 170 °C. Both zones of the joining flow die were controlled to keep the temperature of the melt at 170 °C (± 2°). The melt thermocouples in the die were located in the top layer of the entry zone and at the bottom in the exit zone. The first and second zones in both extruders were set to 90°C and 150°C respectively. The temperature in the third zone of each extruder was adjusted to maintain an exit melt temperature of 165°C. For low screw speeds, a typical set point was 165°, while for higher speeds a set point as low as 155° was required.

Total mass flow rates for the experiments were determine by collecting extrudate samples for a fixed period of time (2 minutes) and weighing the samples. In order to determine the flow rates in the individual layers, it was necessary to construct a calibration curve relating screw rpm’s in the extruders to total mass flow rate. This was accomplished using a bi-linear regression of the two screw rpm’s and the total mass flow rate. Knowing the individual mass flow rates, \( \dot{m} \), melt density, \( \rho \), die height, \( H \), and die width, \( W \), Newtonian wall shear-rates, \( \dot{\gamma}_w \), in the upstream portion of the die could be estimated by \( \dot{\gamma}_w = \frac{6\dot{m}}{\rho WH} \).
Figure 54. Joining Flow Experimental Apparatus: (a) extruder, (b) die, (c) optics.
The procedure used in setting up the experimental system was as follows:

1. The joining flow die, minus the quartz windows, window frames or transducers is assembled. The bolts holding the die together should be snug, but not tight.

2. The two heater band assemblies are slid over the die and secured. The thermocouples are inserted into the die and connected to the temperature controller.

3. The die is heated to experimental operating temperature and allowed to equilibrate for 30 minutes.

4. One zone at a time, the temperature controllers are turned off, the thermocouple and heater bands rapidly removed and the cap screws holding the die together tightened. The heater bands and thermocouples are then replaced.

5. While allowing the die to cool, the adaptors are attached to the extruders and the extruder portions of the piping with heater bands and thermocouple wells are assembled.

6. After the die has cooled down, the two die portions of the piping system with heater bands are assembled and attached to the die.

7. Supporting the die on a stand, it is attached to the two extruders by connecting the die and extruder sections of the piping system.

8. The die is aligned such that the channel width is parallel to the table supporting the stand. It will probably be necessary to adjust the position of the extruders and the rotation of the piping to accomplish this task. While it is tedious, it is IMPORTANT if the optics are to be properly aligned later.
9. The quartz windows are inserted into the die with the gaskets and window frames. DO NOT TIGHTEN THE WINDOW FRAMES WHEN THE DIE IS COLD!

After assembling the feed and die sections of the system and prior to running experiments, it was necessary to generate the flow rate calibration curves. The operating procedure used to generate the calibration curve is basically the same as that used in the actual experiments minus the optical set-up. The following procedure serves to describe both the calibration and experimental operation.

1. The joining flow die temperature controllers are turned on and die is brought up to temperature gradually (no more than 1°C per minute to avoid cracking the windows).

2. After the die has reached the desired temperature, the bolts holding the window frames are tightened 'hand-tight'. DO NOT USE A RATCHET OR WRENCH!

3. The extruders are turned on and the barrels and piping system brought up to test temperature.

4. While the extruders are heating up, the optics are set up as described in Chapter 3. This step is neglected for the calibration and experiments employing a colored layer.

5. When the system has reached the desired temperature, it will need to be purged for approximately one hour to eliminate and residual oil in new pipes or degraded polymer in old pipes. Until melt has reached the die from both extruders, it is recommended to keep the rpm’s below 15. After the die is being filled by both extruders, increase the rpm’s to 25 and purge the system.

6. After the system has been purged, the rpm’s on each extruder are set to the desired rate and the system is allowed to equilibrate. A reasonable indicator is the melt temperatures exiting the extruders.
7. The temperature in the third zone of the extruder is adjusted until the desired melt temperature is achieved.

8. A sample of the extrudate is collected for a fixed time period (2 minutes is recommended) and the sample weighed. The screw speeds are adjusted until the desired flow rate is achieved.

9. Once the desired flow rate and melt temperatures have been achieved, the birefringence patterns are recorded and a sample of the extrudate collected. If possible, it is recommended that after collecting the sample and recording the steady-state patterns, the extruders be stopped and the stress relaxation patterns be recorded.

10. The next screw speeds are selected and steps 7-9 repeated as necessary.

11. After completing the experimental runs, the extruders should be run dry prior to shutting them down.

12. The drive motors on the extruders, the heaters on both the extruders and the piping are then turned off.

13. The bolts holding the window frames on the die should be loosened prior to turning off the die heaters.

4.2.5 Optical Microscopy

In order to determine the effects of relaxation and melt fracture behavior on the bicomponent interface in the joining flow geometry, experiments were conducted using pigmented LLDPE. Layer combinations of LLDPE/LLDPE, with only one layer colored, and LLDPE/LDPE with the
LLDPE layer colored were examined. The experimental conditions and flow rates for the colored experiments were matched with the conditions and flow rates used to examine the flow birefringence. The extrudates collected from the colored layer experiments were sectioned and examined using a Zeiss Axioskop microscope. Samples, approximately 0.05 inch thick, were cut along and transverse to the machine direction using a microtome. The samples were mounted between glass slides and examined at 12.5x and 25.0x magnifications using cross polarizers. Photographs of the samples were obtained using the camera attachment on the microscope.

4.3 Results and Discussion

The results of the joining flow behavior experiments are presented and discussed in this section of the chapter. The effect of upstream conditions corresponding to surface melt fracture in LLDPE on the interface behavior is presented first, followed by the effect of different characteristic relaxation times on the interface. The results are presented in the form of optical micrographs of cross-sectioned extrudate and flow birefringence patterns of the joining flow region.

4.3.1 The Role of Surface Melt Fracture

Two different layer combinations, LLDPE/LLDPE and LLDPE/LDPE, were investigated to examine the role of upstream surface melt fracture conditions in the LLDPE layer on the bicomponent interface behavior. In both layer combinations, the LLDPE from one of the extruders was pigmented and samples of the extrudate were collected for examination by optical microscopy. Equal flow rates were maintained in both layers to eliminate any affects resulting from layer thickness ratio. The range of flow rates studied corresponded to apparent upstream shear-rates
between 20r⁻¹ and 100r⁻¹. This range of shear-rates covered conditions well below and well above the onset of surface melt fracture behavior of the LLDPE in slit die geometries (see Chapter 3). All of the experiments were performed at 170°C as described in the previous section of this chapter.

The presentation and discussion of the LLDPE/LDPE layer combination in this part of this section is in the context of LDPE as a material which does not exhibit surface melt fracture behavior, rather than as a material with a different characteristic relaxation time. It is not, however, intended to dismiss the role of relaxation times on the interface behavior, but rather, reserve that discussion for the second part of this section.

The results of the LLDPE/LLDPE layer combination at upstream apparent shear-rates of 35r⁻¹ and 100r⁻¹ are shown in Fig. 55. The interface at both upstream shear-rates is smooth and free of any irregularities. Even at the higher upstream shear-rate, where there is evidence of surface melt fracture at the external edges (Fig. 56), there is no evidence of irregular interface behavior. Micrographs of the LLDPE/LDPE layer combination interfaces at 35r⁻¹ and 93r⁻¹ are shown in Fig. 57. While there is an obvious distortion of the interface, the interface remains smooth and lacks the fine-scale distortions associated with surface melt fracture behavior.

It is suggested by the results of the optical microscopy studies that conditions corresponding to surface melt fracture in LLDPE prior to the joining together of two layers does not play a role in irregular interface behavior. The smooth distortion observed in the LLDPE/LDPE layer combination is more typical of viscous encapsulation than of surface melt fracture. A more detailed discussion of this behavior is reserved for the third part of this section. There is however, another point to be considered prior to moving away from the discussion of the role of surface melt fracture: the effect of the downstream die length. The die was designed to allow for the possibility of downstream pressure drop measurement capabilities and, as a result, the downstream length, as constructed, was over 30 L/H's. While this construction does not play a role in the birefringence experiments (the original intent of the die), it may have a significant effect on the observed interfaces of the extrudate. In terms of the interface, the necessary conditions of high stress coupled with a
Figure 55. Micrographs of the LLDPE/LLDPE Interface: (a) $\dot{\gamma}_{\text{app}} = 35 \text{s}^{-1}$, (b) $\dot{\gamma}_{\text{app}} = 100 \text{s}^{-1}$.
Figure 56. Micrographs of the LLDPE/LLDPE External Surface: (a) $\dot{\gamma}_{\text{up}} = 35 \text{s}^{-1}$, (b) $\dot{\gamma}_{\text{up}} = 100 \text{s}^{-1}$. 

THE JOINING FLOW BEHAVIOR OF LOW-DENSITY AND LINEAR LOW-DENSITY POLYETHYLENE
Figure 57. Micrographs of the LLDPE/LDPE Interface: (a) $\dot{\gamma}_{app} = 35s^{-1}$, (b) $\dot{\gamma}_{app} = 100s^{-1}$.
critical acceleration of the melt required for surface melt fracture are restricted to the actual contact point in the joining flow die as designed. The long downstream length may serve to attenuate any irregular interface distortions generated as the fluids first join together. Examination of the flow birefringence pattern for the LLDPE/LLDPE layer combination at the highest shear-rate, shown in Fig. 58, shows that the stress field is nearly fully developed after only 3 L/H's. While it is not likely that any irregular distortions would attenuate in a manner such that the colored and uncolored layers return to a smooth interface configuration, the possibility cannot be completely eliminated.

4.3.2 The Role of Relaxation Times

Three different layer combinations were used to explore the influence of relaxation time on the joining flow behavior: LLDPE/LLDPE, LLDPE/LDPE and LDPE/LDPE. The purpose of examining the behavior of the same material in each layer was to establish the individual material behavior prior to examining the effect of combining the two materials. The primary investigative tool employed in the relaxation study was flow birefringence. While optical microscopy was required to establish if differences in characteristic relaxation times lead to irregular distortions in the interface, flow birefringence provided the means to examine the influence of relaxation behavior on the stress field development as the materials first join to form a multi-layer stratified flow. The results of these experiments are presented in this part of the current section.

A comparison of the flow birefringence behavior of LLDPE/LLDPE and LDPE/LDPE layer combinations is shown in Fig. 59. The first point to be made is in regard to the stress levels in the two layer combinations. The stresses upstream in the LLDPE/LLDPE combination are substantially higher than those in the LDPE/LDPE combination. This is a result of the shear-thinning behavior of LDPE and matching the viscosities at a lower shear-rate to minimize viscous encapsulation effects. The second point concerns the die length required to achieve a fully developed
Figure 58. Flow Birefringence of LLDPE/LDPE: $\dot{\gamma}_{\text{bire}} = 77 \text{s}^{-1}$.
stress field. For LLDPE, a fully developed field is realized after 1.5 L/H's, while for LDPE the field is not fully developed even after 3 L/H's. This is consistent with the anticipated difference expected from a longer relaxation time for LDPE. The third and final point to be made from examining Fig. 59 is the shape of the extensional bands coming off the tip of the divider plate. The bands for the LLDPE/LLDPE combination are triangular or arrow shaped, while those for the LDPE/LDPE are much more square. It is suggested by this comparison that the extensional contribution to the complex flow in this region is much greater for LDPE than for LLDPE. While it is tempting to attribute this behavior strictly to a slower relaxation of the high upstream shear-stresses, it is necessary to consider the differences in extensional rheology of the two fluids. There is a marked difference in the extensional stress growth behavior of LLDPE and LDPE (Fig. 49) and the unbounded behavior exhibited could also be used to explain the differences in the extensional band behavior. That is to say that the high extensional stresses resulting from a rapid acceleration of LDPE cause the acceleration from a zero wall velocity upstream to a finite value downstream to occur much more gradually. As a result, the extensional contribution to the flow in the joining flow region persists over a longer length for LDPE, preventing the rapid domination of shear-flow type bands as observed for LLDPE.

Based on the differences observed in the LLDPE and LDPE homogeneous layer combinations, it was anticipated that these differences would lead to some type of irregular behavior in the LLDPE/LDPE layer combination. This was not the case. Comparisons of the three layer combinations at shear-rates of 40s⁻¹ and 70s⁻¹ are shown in Figs. 60 and 61, respectively. The birefringence patterns for the LLDPE/LDPE layer combinations are a compromise between the two homogenous layer combinations at equivalent shear-rates. The length required to achieve a fully developed stress field in the LLDPE/LDPE combination is intermediate to the single fluid lengths. Additionally, the extensional stress band pattern coming off the tip of the divider is an average of that observed in the two individual layers. It is noted here that the observed birefringence patterns were steady for all three layer combinations. That is to say, combination of the two different fluids did not lead to any type of periodic oscillations in the observed birefringence patterns.
Figure 59. LLDPE/LLDPE vs LDPE/LDPE Flow Birefringence:  
(a) LLDPE, $\dot{\gamma}_{\text{app}} = 36 \text{s}^{-1}$.  
(b) LDPE, $\dot{\gamma}_{\text{app}} = 35 \text{s}^{-1}$. 

THE JOINING FLOW BEHAVIOR OF LOW-DENSITY AND LINEAR LOW-DENSITY POLYETHYLENE
Prior to moving to a final discussion of the joining flow experiments, it is important to highlight that the lack of observed irregular birefringence behavior is consistent with the lack of observed irregular distortions in the interface of the extrudate discussed in the previous part of this section. The regular distortion of the interface observed in the LLDPE/LDPE extrudate is the focus of next part of this section.

4.3.3 Regular Interface Distortion: Viscous Encapsulation

In the first part of this section, optical micrographs of the LLDPE/LDPE extrudate interface were presented and discussed in terms of surface melt fracture behavior (Fig. 57). It was suggested that the observed regular distortion or curvature of the interface was not the result of surface fracture, but rather due to viscous encapsulation. While it was not the intention of this work to address encapsulation behavior, the results of the experiments require some discussion of the observed behavior.

The shape of the extrudate interfaces for LLDPE/LDPE layer combinations with increasing shear-rate is shown in Fig. 62. At an apparent downstream shear-rate of $7 \text{s}^{-1}$, there is already a slight curvature present in the interface. As the shear-rate is increased, the contact point of the interface at the edge of the extrudate is moving such that the LDPE layer (uncolored) is preferentially encapsulating the LLDPE layer (colored). At a shear-rate of $26 \text{s}^{-1}$, the interface contact point has moved from the edge of the extrudate to the top of the sample, continuing to force the LLDPE away from the wall region of the die (the region of highest shear). This behavior is consistent with the viscous encapsulation behavior discussed in Chapter 2. As suggested by the work of Everage [8,9], the initial curvature of the interface occurs very rapidly, while for one material to fully encapsulate another requires a much longer die length. In addition to the encapsulation of the LLDPE by the LDPE, the ‘gull-wing’ shape of the interface is similar to that reported by Southern and Ballman [10] for materials which exhibit a cross-over in viscosity behavior. While their exper-
Figure 60. Birefringence Comparison of the Different Layer Combinations: (a) LDPE/LDPE, $\dot{\gamma}_{\text{res}} = 39 \text{s}^{-1}$, (b) LDPE/LLDPE, $\dot{\gamma}_{\text{res}} = 40 \text{s}^{-1}$ (c) LLDPE/LLDPE, $\dot{\gamma}_{\text{res}} = 39 \text{s}^{-1}$. 
Figure 61. Birefringence Comparison of the Different Layer Combinations: (a) LDPE/LDPE, $\gamma_{ZP} = 69 s^{-1}$, (b) LDPE/LLDPE, $\gamma_{ZP} = 70 s^{-1}$ (c) LLDPE/LLDPE, $\gamma_{ZP} = 70 s^{-1}$. 
periments were conducted in a capillary die and the current investigation incorporated a slit-die, their results are directly applicable here. As soon as the initial curvature of the interface in the slit-die occurs (within 3 L/D's [8,9]), the assumed two-dimensional nature of the slit-die is no longer valid. That is to say that there is no longer a plane defining the shear-rate separating the two layers. As a result, the viscosity cross-over exhibited by the LLDPE/LDPE system (Fig. 48) comes into play, resulting in a 'gull wing' interface, directly analogous to the 'curtate cycloid' interface reported by Southern and Ballman. Thus, the interface behavior which was observed in the LLDPE/LDPE experiments is readily explained based purely on viscous encapsulation arguments. Furthermore, previous experiments attributing interface distortions to either relaxation behavior [2] or melt fracture behavior [3] have cied irregular distortions, rather than smooth regular distortions. These previous studies lend additional support to encapsulation being responsible for the observed interface behavior.

4.3.4 Final Considerations

The primary experimental objective of this thesis was to establish whether or not, under upstream conditions corresponding to those required for surface melt fracture in single component flows, surface melt fracture lead to irregular distortions in the bicomponent interface. It is suggested by the results of the joining flow studies presented in this chapter that it does not. In addition to the role of surface melt fracture, the effect of differing characteristic relaxation times in the individual layers was also examined. While differences in the stress field development as the two layers joined together were observed, these differences did not lead to irregular interface distortions in the extrudate. If differing relaxation behavior did lead to irregular behavior, this behavior was attenuated by the excessive downstream length of the joining flow die or was minuscule in comparison with the viscous effects resulting from the choice of materials incorporated in the study.
Figure 62. LLDPE/LDPE Viscous Encapsulation Behavior (LDPE uncolored): (a) $\dot{\gamma}_a = 7 s^{-1}$, (b) $\dot{\gamma}_a = 10 s^{-1}$, (c) $\dot{\gamma}_a = 15 s^{-1}$, (d) $\dot{\gamma}_a = 26 s^{-1}$.
4.4 Conclusions and Recommendations

In the previous section, the results of the joining flow studies of LLDPE and LDPE were presented and discussed. The conclusions resulting from this study and recommendations for future work are presented in the current section.

4.4.1 Conclusions

1. Conditions corresponding to surface melt fracture behavior in the upstream channels do not play a role in the interface behavior downstream. It is strongly suggested by the results of examining the interface of the extrudate for both LLDPE/LLDPE and LLDPE/LDPE layer combinations that conditions corresponding to the surface melt fracture behavior of the LLDPE stream(s) does not lead to irregular distortions of the multi-layer interface. While it is possible that the long land length of the die, downstream of the contact point, may serve to attenuate generated irregular distortions, it is not likely to eliminate all evidence when one of the layers is colored.

2. The smooth regular distortion of the interface observed in the LLDPE/LDPE extrudate is the result of viscous encapsulation behavior. The transition from a smooth flat interface at low shear-rates to a smooth ‘gull wing’ shaped interface is the result of viscous encapsulation and viscosity crossover of the two materials. The encapsulation of the LLDPE by the LDPE is clearly evident with increasing shear-rate (viscosity difference). Additionally, the ‘gull wing’ shaped interface at the center of the extrudate is directly analogous to the ‘curtate cycloid’ interface reported by Southern and Ballman [2] and results from the viscosity cross-over behavior of the LLDPE and LDPE used in the study.
3. Differences in the characteristic relaxation times for the materials incorporated in the investigation do affect the development of the stress field in the joining region of the die, but have little effect on the observed interface in the extrudate. It is shown by the results of the homogeneous layer combinations’ birefringence that increasing the characteristic relaxation time does increase the die length required to attain a fully developed stress field as expected, but any effects on the extrudate interface shape is minimal in comparison with the viscous effects for the materials examined.

4.4.2 Recommendations

While the role of surface melt fracture behavior on bicomponent interface behavior in a joining flow geometry has been addressed in the current study, there are still many questions which remain to be answered in regards to the factors which affect multi-layer interface behavior. Recommendations for future work in this area are given below.

1. Further examine the role of melt fracture by studying the exit flow behavior of fully developed stratified flows. In the current study, the focus centered on conditions corresponding to surface melt fracture in the upstream channels. The role of surface and gross melt fracture conditions on the exit flow behavior of stratified fluids should be addressed in future studies.

2. Examine the role of differing extensional rheology on the contractional flow of superposed fluids. It was suggested in the current work that differing extensional behavior affected the stress field development as the materials accelerated off the divider. This raises the question of how differing extensional behavior would affect the interface of two fluids under going an abrupt contraction. This geometry is analogous to the acceleration of fluids through the lips of a film die and may provide valuable practical insight.
3. Further verify the results of this study, by conducting similar experiments with a much shorter downstream L/H die. The downstream length of the die used in the current studies was too long to definitively establish interface behavior based on examination of the extrudates. Future studies involving multi-layer flows should incorporate a 'shortest distance' length from the region of the die being examined to the exit to minimize possible attenuating effects.

4. Investigate the role of gross melt fracture on the joining flow behavior of two different materials, again incorporating a shorter downstream die length. The maximum upstream shear-rates in the experimental die were determined by the maximum developed pressure in the extruders. As a result of die geometry, principally the excessive length of both the upstream and downstream portions, the pressure drop required to drive the flow limited the rates. A shorter die would allow for the study of the influence of conditions corresponding to gross melt to be examined.
5.0 NUMERICAL STUDIES

The numerical objective of this thesis is to investigate the role of differing rheological properties on the flow behavior in the geometries studied experimentally. Specifically, the effects of characteristic relaxation times and different extensional behavior are considered. To accomplish this goal, two different requirements needed to be satisfied. First, a numerical method capable of handling the geometries examined experimentally was required and second, a constitutive equation with the ability to predict the desired rheological behavior was needed. The finite element method (FEM) was chosen to meet the first requirement and the Phan-Thien Tanner (PTT) constitutive model the second. Finite element methods allow for the solution of a complex system of equations in complex geometries with a minimum degree of difficulty in their use. The PTT model provides the necessary degree of freedom in tailoring the rheological response of the fluid in both shear and extensional flow fields. The work performed in numerically evaluating the flows studied experimentally is presented in this chapter.

Two different geometries were investigated in the studies presented in this chapter. The first was a parallel plate geometry used to examine the ‘stick-slip’ behavior observed in the fluoro-elastomer coating exit die study discussed in Chapter 3. This is a simple geometry (rectangular) with discontinuous boundary conditions applied to one edge (Fig. 16, Chapter 2). The joining flow die examined in Chapter 4 provided the second geometry studied in the current work. This geometry is more
complex than the 'stick-slip' geometry and contains a geometrical discontinuity (the tip of the divider). The discontinuities associated with either die are expected to lead the 'high Deborah Number' convergence problems discussed in Chapter 2. As a result, in addition to the results of the simulations, the limits of convergence in the two geometries will also be presented in this chapter.

The presentation of the numerical studies is divided into four parts. First, a brief introduction to the problems being investigated and the finite element method as it applies to viscoelastic flow simulations is presented. This is followed by a description of the PTT constitutive equation and how model constants affect the rheological predictions. In addition, the implementation of the PTT model with the FEM is also presented in the second part of this chapter. The results of the numerical simulations and the limits of convergence in the two geometries are presented and discussed in the third part of this chapter. In the fourth part, the conclusions are drawn from the results and recommendations for future studies are given.

5.1 Introduction

The benefit of being able to predict the flow behavior of a polymer melt in any given geometry by numerical simulation is self evident. Potential problems arising from a specific die design could be anticipated without the time and expense required for building and testing a prototype model. In addition to die design, the utility of being able to numerically tailor the rheological response of a fluid provides the means to isolate and examine fundamental behavior resulting from a specific characteristic, which is inherently difficult, if not impossible, experimentally for polymer melts. Unfortunately, the simulation of 'real behavior' for viscoelastic fluids is still in the developmental stage.
Solutions to problems involving the flow of simple fluids through complex geometries or complex fluids through simple geometries require the use of numerical techniques. Both finite differences (FD) and finite element methods (FEM) have been successfully used to overcome these problems. The difficulties associated with incorporating irregular and/or curved geometries in FD techniques and the ease of incorporating these irregularities in FEM has led to the FEM becoming the technique of choice in viscoelastic flow simulations. In this method, the domain of the problem is discretized into a set of sub-domains (finite elements) and the governing equations approximated locally by low order polynomials. The error of the approximation is then in some sense minimized, depending on the type of finite element method employed. In the current study, Galerkin’s method [59] is used exclusively (see Chapter 2.5).

The difficulties associated with the high Deborah number problem, discussed in Chapter 2, are well documented and currently represent the major limitation of viscoelastic simulations. Recently, the majority of effort in this area has focused on increasing the limits of convergence of viscoelastic simulations, rather than on solving flows through new geometries within the existing limits. It is not intended to suggest that these efforts are frivolous, but rather that they are mathematical in nature, and that the original intent of solving a fluid mechanics problem has been neglected. This point is best made by considering the non-consistent application of streamline upwinding techniques presented by Marchal and Crochet [71]. As discussed in Chapter 2, this technique results in converged solutions to a different problem than was intended by affecting the constitutive equation. Other methods of overcoming the high Deborah number problem, such as employing the explicitly elliptic momentum equation [72], have remained honest in terms of the problem being solved, but their success to date has been limited to problems without the singularities resulting from corners, as found in abrupt contraction simulations.

Given the current state of numerical flow simulations, a choice between employing a constitutive model which accurately describes the viscoelastic nature of a melt, but is limited in terms of the Deborah number of the flow, employing methods which alter the physics of the problem or a simple, inelastic model was required. For the current investigation, it was decided to employ a
viscoelastic constitutive model which more accurately describes the rheological properties of the fluid, specifically the model of Phan-Thien and Tanner (PTT). The choice of the model was based on the ability to vary the extensional rheological predictions of the model, separate from the shear flow behavior. Employing the PTT model in the FEM formulation of the problems considered should provide an accurate description of the flow behavior, but the range of solutions will be limited by the high Deborah number problem discussed previously. Accepting this, accurate solutions at lower Deborah numbers should provide valuable insight into the behavior of flows at higher rates.

The numerical objectives of this thesis are to examine how differing rheological properties affect the developing flow in a joining flow geometry and to examine the role of wall slip in flow between parallel plates. These objectives were resolved by examining the effect of different parameter constants on the rheological predictions of the PTT model and incorporating specific behavior into the simulations in the two geometries. These investigations represent the substance of the work presented in this chapter.

5.2 Numerical Methods and Constitutive Model

In this part of the chapter, descriptions of the finite element formulation, the constitutive equation and the implementation of the finite element method used in the current study are presented. In the first section, the equations governing the ‘slow’ flow of incompressible fluids are cast in terms of the finite element formulation. In the second section, the Phan-Thien Tanner constitutive model and the predictions of the model in shear and extensional flow fields are presented. The finite element code used in the solution of the problems is described in the last section. Additionally, programs used to generate the necessary boundary conditions and the appropriate input file are also described in the third section.
5.2.1 Penalty Formulation of the 'Slow' Flow Problem

The flow of isothermal fluids are governed by the conservation of mass (continuity) and the conservation of momentum (motion). In the absence of inertial effects ('slow' flow) and the assumption that the fluid is incompressible, these equations are given by

\[ \nabla \cdot \mathbf{u} = 0 \quad \text{(continuity)} \tag{5.1} \]

\[ \nabla P + \nabla \cdot \mathbf{\tau} = 0 \quad \text{(motion)} \tag{5.2} \]

In addition to these two equations, a third equation (constitutive), which relates the stress in the fluid to its deformation is required. In general terms, this can be expressed as

\[ \mathbf{\tau} = \mathbf{\tau}(\dot{\mathbf{\gamma}}, \mathbf{\xi}, ...) \quad \text{(constitutive)} \tag{5.3} \]

Adopting indicial notation, Eqs. 5.1 - 5.3 become

\[ u_{a, a} = 0 \quad \text{(5.1a)} \]

\[ p_{, a} + \tau_{a\beta, \beta} = 0 \quad \text{(5.2a)} \]

\[ \tau_{a\beta} = \tau_{a\beta}(\gamma_{a\beta}, \dot{\gamma}_{a\beta}, ...) \quad \text{(5.3a)} \]

For two dimensional problems, in the absence of a constitutive equation explicit in \( \tau \), this represents a set of six coupled equations in six unknowns: \( P, u_1, u_2, \tau_{11}, \tau_{12}, \) and \( \tau_{22} \).

The Galerkin finite element formulation is obtained by first generating the variational form of Eqs. 5.1a - 5.3a. This is accomplished by the multiplication of each equation by an arbitrary test function.

---

1 Variables underscored once indicate a vector, while those underscored twice indicate a second rank tensor. The sign convention used for the stresses in this text is \( \tau_{u} > 0 \) in compression.
function, \( w_i \), and setting the integral over the element domain, \( \Omega^e \) to zero. For Eq. 5.2a, this corresponds to:

\[
\int_{\Omega^e} [P_{\alpha} + \tau_{\alpha\beta}] w d\Omega^e = 0
\]

Reducing the order of differentiation by shifting it to the test function (applying the gradient theorem), the following equation results:

\[
\int_{\Omega^e} [P w_i + \tau_{\alpha\beta} w_i] d\Omega^e = \int_{\Gamma^e} [w_i \tau_{\alpha}] d\Gamma^e
\]

In the above equation, \( \tau_i \) is the traction vector along the element boundary, \( \Gamma^e \). The pressure term in Eq. 5.5 can be eliminated by applying the continuity equation (Eq. 5.1) as a constraint on the momentum equation [59]. This results in the elimination of Eq. 5.1 from the system of equations, and the pressure term in Eq. 5.5 is replaced by

\[
P = -\gamma_P u_{i,a}
\]

The choice of the penalty parameter, \( \gamma_P \), is made such that

\[
\gamma_P = \eta \times 10^4 \leq 10^{13}
\]

A value of \( \gamma_P = 10^{13} \) was used in the current study. Upon elimination of the pressure, Eq. 5.5 becomes

\[
\int_{\Omega^e} [\gamma_P \eta_{\beta, \beta} w_i + \tau_{\alpha\beta} w_i] d\Omega^e = \int_{\Gamma^e} [w_i \tau_{\alpha}] d\Gamma^e
\]

and Eq. 5.1 is eliminated.

The impact of using the penalty formulation, rather than the full 'mixed' formulation is two-fold. First, the number of nodal unknowns is reduced from six down to five. Second, the differentiability

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constraint on the approximating functions is reduced. A brief discussion of this later aspect in terms of the final step of generating the Galerkin formulation is discussed in the following paragraph.

After the variational form of the equations has been generated, the second and final step of the Galerkin formulation is the incorporation of approximating functions for the unknowns and test functions. The choice of approximating functions is constrained by the order of differentiation present in the equations. Examination of Eq. 5.7 shows that interpolating functions for \( u_p \) and \( \tau_{ij} \) need to be once differentiable (i.e. linear). If the full 'mixed' method is employed (Eq. 5.5), it has been shown [58] that the order of the pressure approximating functions should be one order lower than that of the velocity. This would lead to a second-order interpolation function being required for \( u_p \). Thus, not only is the number of degrees of freedom per node reduced by incorporating the penalty method, but linear elements (4 nodes) rather than quadratic elements (8 or 9 nodes) can be employed, further reducing the total number of degrees of freedom for a given mesh. The nodal unknowns are approximated by interpolation functions of the form

\[
{u}_p = \sum_{j=1}^{n} \psi_j u_{pj} \tag{5.8}
\]

\[
\tau_{ij} = \sum_{j=1}^{n} \psi_j \tau_{ij} \tag{5.9}
\]

and the weighting function by \( w_i = \psi_i \). The Galerkin penalty formulation of the momentum equation is then given by:

\[
\sum_{j=1}^{n} \left\{ \int_{\Omega} [\gamma_p \psi_j \psi_{\beta, \beta} \psi_{L, \alpha} + \tau_{\alpha \beta j} \psi_j \psi_{L, \beta}] d\Omega - \int_{\Gamma_p} [\psi_j \psi_{L, \alpha}] d\Gamma_p \right\} = 0 \tag{5.10}
\]

Linear approximations (\( n = 4 \)) were used in this study for \( \psi_i \).
So far, no specific mention of the form of the constitutive equation has been made. It is sufficient to state at that this point that the Galerkin formulation of the constitutive equation follows that of the momentum equation exactly. An additional consideration for the constitutive equation is that, unlike Eq. 5.2, it is not linear in $\xi$ and $\eta$. As a result, it must be linearized prior to implementation and a non-linear solution algorithm employed. The FEM code used in this investigation is a slight modification of the one developed by Gotsis [62] and a complete description of the linearization procedure for the Phan-Thien Tanner model and solution of the non-linear problem is presented there. It is sufficient to note that a Picard iteration method is employed to solve the system of non-linear equations.

5.2.2 Phan-Thien Tanner Model

In the introduction of this chapter, the Phan-Thien Tanner (PTT) model was described as being able to accurately predict real rheological behavior. Specifically, the PTT model allows for independently varying the shear and extensional properties. It is this feature which makes the use of the model desirable. For a single relaxation mode, the model is given by

$$Z(tr\bar{\tau}) \bar{\tau} + \dot{\lambda}_{(1)} + \frac{\dot{\xi}}{2} \left( \bar{\tau} \bar{\tau} + \bar{\tau} \bar{\tau} \right) + \eta \bar{\tau} = 0 \tag{5.11}$$

where $\bar{\tau}(0)$ is the convected derivative given for symmetric $\tau$ by

$$\tilde{\tau}(1) = \frac{D}{D\tau} \bar{\tau} - (\bar{\tau} \cdot \nabla \mu)^T - (\nabla \nabla \mu) \tag{5.12}$$

and $Z(tr\bar{\tau})$ has two forms given by

$$Z(tr\bar{\tau}) = 1 - \varepsilon \lambda tr(\bar{\tau})/\eta \tag{5.13a}$$

$$Z(tr\bar{\tau}) = \exp(-\varepsilon \lambda tr(\bar{\tau})/\eta) \tag{5.13b}$$
The model was derived from molecular considerations, the details of which are presented elsewhere [62,84,86], with the key features being the inclusion of terms which allow for the rate of creation/destruction of network junctions, $Z(t\gamma)$, and the role of slip in affecting the relaxation behavior via the $\lambda \xi/2$ term. The most significant impact of the form of the $Z(t\gamma)$ expression is on the extensional viscosity ($\bar{\eta}_e$) predictions. For the linear form (Eq. 5.13a), $\bar{\eta}_e$ is equal to the Trouton value at low extension rates and rises to a plateau value at higher rates. Similarly, at low extension rates, the exponential form also predicts a Trouton value, but as the extension rate is increased, $\bar{\eta}_e$ increases and passes through a maximum. The value of the plateau in the linear case and the maximum in the exponential form are determined by the value of $\xi$. The latter form of the term predicts behavior in agreement with experimental data for most polymers and was the form used in the current study.

One problem with the PTT model is that it predicts a much greater degree of shear-thinning behavior than is reasonable (the shear stress passes through a maximum at high rates). In order to overcome this limitation, a retardation term is added to the extra stress. The extra stress is then viewed as having two components: a viscoelastic portion, $\bar{\tau}^1$ and a purely viscous portion $\bar{\tau}^2$. The total extra stress is then given by

$$\bar{\tau} = \bar{\tau}^1 + \bar{\tau}^2$$

where $\bar{\tau}^1$ is given by Eq. 5.11 with $\bar{\tau}$ replaced by $\bar{\tau}^1$ and $\eta$ by $\eta_1$. $\bar{\tau}^2$ is given by

$$\bar{\tau}^2 = \eta_2 \dot{\varepsilon}$$

In order to prevent the shear stress from passing through a maximum, $\eta_2$ must be chosen such that $\eta_1/\eta_2 \leq 8$. The result of adding the retardation term is that at high shear-rates, the viscosity decreases to the plateau value of $\eta_2$ and the stress continues to increase monotonically. A ratio of 7.5 was used in the numerical studies presented in this chapter.
Analytical expressions for the shear-flow predictions of the PTT model require an assumption as to the contribution of the $Z(n_{2})$ term in Eq. 5.11. If the magnitude of the strain is assumed to be small, then the contribution from the creation/destruction of network junctions can be considered negligible to the extra stress of the system ($\varepsilon = 0$). This is a reasonable assertion for shear flow, and eliminates the exponential coefficient in Eq. 5.11. As a result, the predictions of the model in steady shear are given by

$$\eta = \frac{\eta_1 \xi(2 - \xi)(\dot{\gamma})^2}{1 + \xi(2 - \xi)(\dot{\gamma})^2} + O(\varepsilon)$$  \hspace{1cm} 5.16$$

$$N_1 = \frac{2\eta_1 \dot{\gamma}^2}{1 + \xi(2 - \xi)(\dot{\gamma})^2} + O(\varepsilon)$$  \hspace{1cm} 5.17$$

$$N_2 = -\frac{\xi}{2} N_1$$  \hspace{1cm} 5.18$$

The term $O(\varepsilon)$ was included to indicate that for large values of $\varepsilon$ there will be an additional contribution. In small amplitude oscillatory shear, the model predicts

$$G' = \frac{\eta \omega}{1 + (\omega \lambda)^2}$$  \hspace{1cm} 5.19$$

$$G'' = \frac{\eta \omega}{1 + (\omega \lambda)^2}$$  \hspace{1cm} 5.20$$

The assumption of small strains applied in obtaining analytical expressions for the shear flow predictions of the PTT model are not valid in extension. As a result, in order to determine the steady-state extensional viscosity, $\eta_e$, and the transient stress growth behavior, $\sigma_\tau^+$, it is necessary to solve two pairs of coupled differential equations. For the steady-state viscosity

$$\exp\left[\frac{\xi}{\eta_1} (\tau_{11} + 2\tau_{22})\right] \tau_{11} - 2\dot{\gamma}(1 - \xi)\tau_{11} = 2\eta \dot{\gamma}$$  \hspace{1cm} 5.21$$

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\[
\exp\left[ \frac{\lambda}{\eta_1} (\tau_{11} + 2\tau_{22}) \right] \tau_{22} + \lambda \dot{\varepsilon}(1 - \xi) \tau_{22} = -\eta \dot{\varepsilon} \quad 5.22
\]

For transient stress growth
\[
\lambda \frac{d\tau_{11}}{dt} + \exp\left[ \frac{\lambda}{\eta_1} (\tau_{11} + 2\tau_{22}) \right] \tau_{11} - 2\lambda \dot{\varepsilon}(1 - \xi) \tau_{11} = 2\eta \dot{\varepsilon} \quad 5.23
\]
\[
\lambda \frac{d\tau_{22}}{dt} + \exp\left[ \frac{\lambda}{\eta_1} (\tau_{11} + 2\tau_{22}) \right] \tau_{22} + \lambda \dot{\varepsilon}(1 - \xi) \tau_{22} = -\eta \dot{\varepsilon} \quad 5.24
\]

Three fortran programs were written to examine the effects of the material constants in the PTT model on the rheological predictions. The first program, CMPTTSHR, solves Eqs. 5.16 and 5.17 over an input range of shear-rates. The values predicted by a set of constants can be compared with an input set of experimental values or with the predictions from another set of constants. The second program, CMPTTEST, generates the extensional stress growth behavior for a particular set of material constants by solving Eqs. 5.23 and 5.24. The IML routine DIVPAG is used to solve the initial value problem. Again, the predictions can be compared with sets of experimental data or other predictions. The last program, EXETAPTT, solves Eqs. 5.21 and 5.22 for the steady-state extensional viscosity.

In order to compare numerical simulations with experimental results, it is necessary to fit the rheological data with model constants for use in the simulations. The program FITPTT was written for this purpose and uses the IML non-linear least-squares routine DUNLSF to fit experimental data with Eq. 5.16. The results of the fit for \( \eta_1 \) were insensitive to initial guesses, but this was not the case for \( \xi \) or \( \lambda \). Depending on the initial guesses, unrealistic values for both \( \xi \) and \( \lambda \) were determined. To overcome this limitation, a range of fixed \( \xi \) values were used and the best fit values for \( \lambda \) determined. The choice of \( \lambda \) and \( \xi \) was then made based on the minimum error of the fits with the data. This procedure was successful for determining the values of \( \eta_1 \), \( \lambda \), and \( \xi \). As indicated earlier, \( \varepsilon \) primarily affects the extensional behavior and, based on the small strain assumptions, is not accounted for in Eq. 5.16. Attempts to fit \( \varepsilon \) to extensional data using a program were not
successful. As a result, after determining the shear-flow constants, different values of \( \varepsilon \) were used to predict the extensional behavior and a choice was made based on comparing the predictions with actual data.

### 5.2.3 Implementation of the Finite Element Method

The simulation of the problems presented in this chapter were accomplished using the finite element program PTTFEM. This code is a modified version of NEWT developed by Gotsis [62] and a full description of the routines will not be presented here. The two primary modifications by the author in PTTFEM were the restriction of available constitutive models to the PTT model and the incorporation of multiple fluid constant sets. The later modification was made in hopes of simulating bicomponent interface behavior, the results of which were unsuccessful. A description of the work performed toward this end is presented in Appendix D and a listing of PTTFEM is given in Appendix E. In addition to these two modifications, the ability to track nodal variables during the iterative solution procedure was also incorporated.

PTTFEM is a finite element code which solves the penalty formulation of the momentum equation in conjunction with the PTT constitutive model. This formulation results in five degrees of freedom per node: \( u_1, u_2, \tau_{11}, \tau_{12}, \) and \( \tau_{22} \). These variables are solved for iteratively using a Picard iteration scheme. After the nodal solutions have been generated, the pressure at the center of each element are recovered using Eq. 5.6. In addition, the shear-rate, extension-rate and vorticity at the center of each element is also calculated. The nodal and element solutions are then written to an output file and plotting files. If desired, the program is then re-run to generate the stream function solution. This solution is stored in a separate file for plotting. The software used to generate the mesh and boundary conditions for the simulation and the plotting software are described in the following paragraphs.
Generation of the meshes used in the simulations was accomplished using the pre-processor portion of FIDAP, a commercial FEM package developed by Fluid Dynamics International, Inc. The procedure used in generating the FIDAP input file is described in the user's guide and is quite easily mastered. Once a specific mesh has been developed, mesh refinement can be accomplished with a minimum of effort. The output file from FIPREP, filename FD1NP, is copied twice and edited to include the nodal x-y data in one file and the element connectivity data in the second file. It is these two files which are used in preparing the input file to PTTFEM.

After the appropriate mesh files are generated, it is necessary to generate inlet and outlet profiles for use in specifying boundary conditions. This is accomplished using either CXPROF for joining flow simulations or SSPROF for stick-slip problems. Both programs were developed by the author and solve for the velocity, stress and streamline profiles for the inlet(s) and outlet plane. The output file, filename PROPRO, is then used in generating the input file for PTTFEM, described in the next paragraph.

The input file for PTTFEM is generated by either CXPREP or SSPREP, depending on the type of problem. In addition to the mesh and profile files discussed earlier, an input file containing parameters for the simulation is required. These consist of the nodes in the mesh located on the different portions of the boundary (entry plane, etc.), values for the iteration acceleration parameter and the penalty parameter. The programs then generate the complete input file in the format appropriate for PTTFEM.

By using a series of programs to generate the input file for PTTFEM, series of experiments examining different effects are greatly facilitated. Once a particular mesh or boundary profile has been generated, there is no need to re-run FIDAP or CX/SSPROF to investigate the role of acceleration parameter or global tolerance on the solution. Similarly, to examine the effect of increasing flow-rate, only the input file to CX/SSPROF needs to be modified.
The final point to be addressed in this section is the plotting of the results. In addition to generating a complete solution file, PTTFEM generates a series of plotting files for the velocities, pressure, birefringence and streamlines. These files are compatible with the SURFACE III contour plotting package developed by Interactive Concepts Inc.. This package was used to generate the contour plots presented in this chapter. In addition to the contour plots, the program VPLT was written to generate line plot files from a modified output file. The files were then plotted using SAS/GRAF, developed by SAS Institute, Inc..

5.3 Results and Discussion

The objectives of this thesis pertaining to the numerical work are to examine how differing rheological properties affect the developing flow in a joining flow geometry and to examine the role of wall slip in flow between parallel plates. The incorporation of the Phan-Thien Tanner (PTT) model into the finite element code, discussed in the second part of this chapter, provides the necessary means to accomplish these objectives. By adjusting the material constants, a wide range of rheological behavior can be examined numerically. However, the success of any simulation is ultimately determined by whether or not the results accurately predict real behavior. By fitting the rheological behavior of low-density polyethylene (LDPE) and linear low-density polyethylene (LLDPE) to the PTT model, the results of the simulations for these fluids can be compared with the experimentally observed behavior discussed in Chapters 3 and 4. Based on these comparisons, confidence in the simulation of the numerical fluids, exhibiting a broad range of rheological behavior, can be established. The results of the work in accomplishing the experimental objectives of this thesis are presented and discussed in this part of the chapter.

The presentation and discussion of the results of the numerical studies is divided into four sections. In the first section, the effect of the material constants in the PTT model on the rheological
Predictions is presented. In addition, the PTT fits for LDPE and LLDPE are given in this section. Results and discussion of the joining flow and stick-slip simulations are presented in the second and third sections, respectively. The fourth section provides some final overall considerations of the numerical studies. Specifically, the limits of convergence in the two test geometries are addressed.

5.3.1 PTT Constitutive Model: Fits and Predictions

In the earlier discussion, it was stated that comparison of the numerical simulations with experimental results provides a true measure of the simulations' success. To accomplish this comparison, it was necessary to fit the material constants for LDPE and LLDPE using the programs described in the second part of this chapter. The constants determined for the two fluids are given in Table 2. Comparison of the predicted shear viscosity and extensional stress growth for LLDPE with experimental data are shown in Figs. 63 and 64, respectively. Similar comparisons for LDPE are shown in Figs. 65 and 66. For both materials, a much greater degree of shear thinning is predicted by the model than is observed experimentally. This deficiency was discussed previously in conjunction with the need to add a retardation term to the constitutive equation. The effect of the retardation viscosity is clearly evidenced by the high shear-rate plateau for LDPE (Fig. 65). The most significant difference in the shear predictions for the two materials is the onset of shear-thinning behavior. This is a result of the relaxation constant, $\lambda$, which can be roughly estimated by the reciprocal the shear-rate at the onset of shear-thinning. The impact of the relaxation time on the predictions for the two fluids is also evident in the extensional stress growth behavior. The higher relaxation time for LDPE delays the onset of the stress growth plateau in extension. The extensional behavior predicted for LLDPE is in excellent agreement with the data for the lower two extension rates and only qualitatively correct at the higher rate. For LDPE, the predictions at low strains are accurate, while the predicted plateau at higher strains does not agree with the data. The model qualitatively depicts the differences observed experimentally for LDPE and LLDPE through
the appropriate choice of constants. A discussion of how the different constants affect the rheological predictions of the model is presented in the following paragraphs.

Prior to determining how different rheological properties affect the behavior in a particular geometry, it was necessary to establish how to generate specific behavior using the constants in the PTT model. Toward this end, the influence of the five parameters \( (\eta_1, \lambda, \xi, \varepsilon\) and \(\eta_1/\eta_3) \) on the shear and extensional behavior was examined. Table 3 lists the 'numerical fluids' examined in this study. The fluid specified as NFL00 represents the basic fluid and the variations of this fluid are named according to the parameter varied. The complete results of this study are presented in Appendix C and are only summarized here.

Increasing \( \eta_1 \) in the PTT model serves to increase the magnitude of the predicted shear viscosity and elastic modulus (or \( N' \)), as expected. Similarly, the magnitude of the plateau in extensional stress growth is increased, but the behavior during the transient development is not affected. The normalized steady-state viscosity remains unaffected by varying \( \eta_1 \). These results are consistent with the anticipated effects of increasing the viscosity.

The role of increasing the relaxation time was alluded to in the previous discussion of fitting the PTT model to rheological data for LLDPE and LDPE. The onset of shear-thinning behavior occurs earlier with increasing relaxation time. \( \lambda \) also has a pronounced effect on the extensional stress growth and steady-state extensional viscosity behavior, shown in Figs. 67 and 68, respectively. At lower relaxation times, the effect of increasing \( \lambda \) is to delay the onset of the steady-state plateau (Fig. 67). The point of interest is the shape of the curves for 5 and 10 s. An inflection point is observed in the response, similar to the behavior shown for LDPE in Fig. 66. In addition, the extension-rate for the onset of the deviatory response (departure from the normalized value of 1) in the extensional viscosity curve is reduced with increasing \( \lambda \) (Fig. 68), similar to the onset of the shear-thinning behavior. It is obvious from these results that the role of the relaxation is not limited to the shear flow behavior, but also has a pronounced effect on the extensional predictions.
<table>
<thead>
<tr>
<th>MATERIAL</th>
<th>η_1 (PaS)</th>
<th>λ (S)</th>
<th>ζ</th>
<th>μ</th>
<th>η_1/η_1</th>
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<td>9730</td>
<td>0.125</td>
<td>0.200</td>
<td>0.100</td>
<td>7.500</td>
</tr>
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<td>1.390</td>
<td>0.150</td>
<td>0.010</td>
<td>7.500</td>
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Figure 63. Viscosity Shear Rate Data for LLDPE vs PTT Fit
Figure 64. Extensional Stress Growth Data for LLDPE vs PTT Fit: (a) $\dot{\gamma} = 0.020 s^{-1}$, (b) $\dot{\gamma} = 0.053 s^{-1}$, (c) $\dot{\gamma} = 0.200 s^{-1}$.
Figure 65. Viscosity Shear Rate Data for LDPE vs PTT Fit
Figure 66. Extensional Stress Growth Data for LDPE vs PTT Fit: 
(a) $\dot{i} = 0.020 \text{s}^{-1}$, 
(b) $\dot{i} = 0.053 \text{s}^{-1}$, (c) $\dot{i} = 0.200 \text{s}^{-1}$. 

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<table>
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<th>FLUID</th>
<th>( \eta_1 ) (PaS)</th>
<th>( \lambda(S) )</th>
<th>( \zeta )</th>
<th>( \kappa )</th>
<th>( \eta_1/\eta_1 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>NFL-00</td>
<td>10000</td>
<td>1.000</td>
<td>0.200</td>
<td>0.100</td>
<td>7.500</td>
</tr>
<tr>
<td>NFL-11</td>
<td>8000</td>
<td>1.000</td>
<td>0.200</td>
<td>0.100</td>
<td>7.500</td>
</tr>
<tr>
<td>NFL-12</td>
<td>12000</td>
<td>1.000</td>
<td>0.200</td>
<td>0.100</td>
<td>7.500</td>
</tr>
<tr>
<td>NFL-21</td>
<td>10000</td>
<td>0.125</td>
<td>0.200</td>
<td>0.100</td>
<td>7.500</td>
</tr>
<tr>
<td>NFL-22</td>
<td>10000</td>
<td>0.250</td>
<td>0.200</td>
<td>0.100</td>
<td>7.500</td>
</tr>
<tr>
<td>NFL-23</td>
<td>10000</td>
<td>0.500</td>
<td>0.200</td>
<td>0.100</td>
<td>7.500</td>
</tr>
<tr>
<td>NFL-24</td>
<td>10000</td>
<td>2.500</td>
<td>0.200</td>
<td>0.100</td>
<td>7.500</td>
</tr>
<tr>
<td>NFL-25</td>
<td>10000</td>
<td>5.000</td>
<td>0.200</td>
<td>0.100</td>
<td>7.500</td>
</tr>
<tr>
<td>NFL-26</td>
<td>10000</td>
<td>10.00</td>
<td>0.200</td>
<td>0.100</td>
<td>7.500</td>
</tr>
<tr>
<td>NFL-31</td>
<td>10000</td>
<td>1.000</td>
<td>0.150</td>
<td>0.100</td>
<td>7.500</td>
</tr>
<tr>
<td>NFL-32</td>
<td>10000</td>
<td>1.000</td>
<td>0.250</td>
<td>0.100</td>
<td>7.500</td>
</tr>
<tr>
<td>NFL-41</td>
<td>10000</td>
<td>1.000</td>
<td>0.200</td>
<td>0.010</td>
<td>7.500</td>
</tr>
<tr>
<td>NFL-42</td>
<td>10000</td>
<td>1.000</td>
<td>0.200</td>
<td>0.001</td>
<td>7.500</td>
</tr>
<tr>
<td>NFL-43</td>
<td>10000</td>
<td>1.000</td>
<td>0.200</td>
<td>0.020</td>
<td>7.500</td>
</tr>
<tr>
<td>NFL-44</td>
<td>10000</td>
<td>1.000</td>
<td>0.200</td>
<td>0.050</td>
<td>7.500</td>
</tr>
<tr>
<td>NFL-51</td>
<td>10000</td>
<td>1.000</td>
<td>0.200</td>
<td>0.100</td>
<td>6.000</td>
</tr>
</tbody>
</table>
Figure 67. Effect of $\lambda$ on Extensional Stress Growth: (a) $\lambda = 0.125\pi$, (b) $\lambda = 0.250\pi$,
(c) $\lambda = 0.500\pi$, (d) $\lambda = 1.00\pi$, (e) $\lambda = 2.50\pi$, (f) $\lambda = 5.00\pi$, (g) $\lambda = 10.0\pi$. 
Figure 68. Effect of $\lambda$ on Extensional Viscosity: (a) $\lambda = 0.125$, (b) $\lambda = 0.250$, (c) $\lambda = 0.500$, (d) $\lambda = 1.00$, (e) $\lambda = 2.50$, (f) $\lambda = 5.00$, (g) $\lambda = 10.0$. 

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The influence of $\xi$ on the rheological predictions is trivial when compared with the effect of the relaxation time or viscosity. The point of interest stems, not from the magnitude of its effect, but rather on its manner. As mentioned in the second part of this chapter, the inclusion of $\xi$ in the model is to account for the effect of slip between chains in the network on the relaxation behavior. As a result, it is anticipated that the effect of varying $\xi$, no matter how small, would correspond to variations resulting from relaxation time behavior. While this expectation is realized in shear behavior and extensional stress growth, there is an additional effect observed in the steady-state extensional behavior. The effect on the onset of deviatory behavior is similar to the effect observed with differing relaxation behavior but, an additional effect on the magnitude of the maximum is also realized, as shown in Fig. 69. While the effect is minimal, it is none the less interesting.

Exclusion of the $Z(\tau_v)$ term in Eqs. 5.16 and 5.17 obviously renders the effect of $\zeta$ on shear-flow predictions inconsequential. Similarly, the effect of $\zeta$ on extensional stress growth is trivial compared with the impact of relaxation time. However, the value of $\zeta$ has a dramatic effect on the steady-state extensional viscosity behavior shown in Fig. 70. While $\zeta$ has no effect on the onset of deviatory behavior, its impact on the magnitude is dramatic. The magnitude of the maximum increases with decreasing $\zeta$, until it becomes unbounded for values of 0.050 or less. This result is consistent with the intent of the parameter to describe the rate of network junction formation/destruction discussed in the previous part of this chapter. As the time scale of the deformation exceeds the ability of the network to respond, the stress required to accomplish the deformation becomes unbounded. The impact of this parameter is to affect strain-rate extensional behavior of the flow.

The final parameter to be examined was the ratio of $\eta_1/\eta_2$. The impact of this parameter provides no surprises. It affects the magnitude of the rheological values in a fashion similar to that of increasing $\eta_1$. As expected, the most significant effect is to increase the magnitude of the high shear-rate viscosity plateau with decreasing $\eta_1/\eta_2$.

It is suggested by the results of this study that the relaxation time, $\lambda$, and the $\zeta$ parameter should have the most pronounced effect on the results of the simulations for a given set of conditions. As
Figure 69. Effect of $\xi$ on Extensional Viscosity: (a) $\xi = 0.15$, (b) $\xi = 0.20$, (c) $\xi = 0.25$. 

EXTENSION RATE (1/sec)

EXTENSIONAL VISCOSITY / 3 ETA0

XI: — 0.20 — 0.15 — 0.25
Figure 70. Effect of $\varepsilon$ on Extensional Viscosity: (a) $\varepsilon = 0.001$, (b) $\varepsilon = 0.010$, (c) $\varepsilon = 0.020$, (d) $\varepsilon = 0.050$, (e) $\varepsilon = 0.100$. 

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a result, numerical studies examining the effect of rheological behavior will focus on these two parameters.

5.3.2 Joining Flow Simulations

The first objective of this part of the thesis is to examine the effect of differing rheological behavior on the development of the flow in a joining flow geometry. In the previous section, it was determined that the $\lambda$ and $\varepsilon$ parameters in the PTT model should have the most impact on this behavior. In this section, the results of the joining flow simulations are presented and discussed. First, the three meshes used in the simulations are presented. This is followed by the simulations incorporating model constants fit to the rheological properties for LLDPE and LDPE. The results of these simulations are then compared with the behavior observed experimentally (Chapter 4). The results of the simulations examining the impact of $\lambda$ and $\varepsilon$ on the developing behavior are presented in the third part, followed by some final comments.

5.3.2.1 Joining Flow Meshes

Three different finite element meshes were used in the joining flow studies presented in this section and are shown in Fig. 71. The physical domain for all three meshes is the same, with the only difference being the degree of mesh refinement. Linear elements (4 nodes per element) were incorporated in all cases, with Mesh CX1 being comprised of 288 elements and 335 nodes, Mesh CX2 having 480 elements and 540 nodes, and Mesh CX3 having 640 elements and 718 nodes. These correspond to 1440, 2700 and 3590 degrees of freedom per mesh, respectively.
Figure 71. Joining Flow Meshes: (a) Mesh CX1, (b) Mesh CX2, (c) Mesh CX3.
5.3.2.2 LLDPE and LDPE Simulations

In order to test the quality of the numerical simulations, experiments incorporating model constants fit to the rheological properties of LLDPE and LDPE were conducted. Simulations of LLDPE were conducted in all three of the joining flow meshes. As shown in Table 4, the limit of convergence decreased with increasing mesh refinement. Confidence in these limits was established by varying the iteration tolerance requirement and the initial guesses to the solution. This later test was accomplished by varying the Newtonian viscosity and the boundary conditions for the first iteration of the solution procedure. The appropriate degree of mesh refinement was determined by comparing the critical solution values listed in Table 4 at equivalent flow rates ($Q = 1.5$). It is shown by this comparison that when the mesh is refined from 288 elements to 480 elements different values of the upstream wall shear-rate and shear-stress are predicted. In addition, the extension rate at the end of the divider is also changed. Further refinement of the mesh from 480 elements to 640 elements produces no change in these predicted values. Additional comparisons were made between the meshes by examining the centerline solutions downstream of the divider. Comparisons of $\gamma_4$, $\tau_{11}$ and $\tau_{13}$ for the three meshes are shown in Figs. 72, 73 and 74, respectively. Consideration of these plots support the earlier comparison of critical values. Therefore, it was determined that Mesh CX2 provides a sufficient degree of refinement while allowing for the maximum level of convergence. As a result, Mesh CX2 was used in the remaining joining flow simulations.

The birefringence pattern predicted by numerical simulation for LLDPE is compared with the experimentally observed pattern in Fig. 75. The numerical pattern is qualitatively in good agreement with the experimental pattern. The relaxation of the upstream dinges follows those observed experimentally and the downstream length required for full development of the patterns are also in agreement. The only real difference between the patterns is the shape of the extensional bands at the tip of the divider. The numerical patterns are much more rounded than those observed experimentally. Quantitatively, the predictions are not accurate. The numerical flow rate is less than half of the experimental value. This indicates that the numerical method is predicting stress levels that
<table>
<thead>
<tr>
<th>MESH</th>
<th>$Q$ (cc/s/m)</th>
<th>$D_e$</th>
<th>$\dot{\gamma}_w$ (s$^{-1}$)</th>
<th>$\tau_w$ (MPa)</th>
<th>$\dot{\gamma}_w$ (s$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CX1</td>
<td>1.50</td>
<td>0.406</td>
<td>3.10</td>
<td>0.06</td>
<td>4.50</td>
</tr>
<tr>
<td>CX1</td>
<td>4.00</td>
<td>1.381</td>
<td>9.00</td>
<td>0.60</td>
<td>12.5</td>
</tr>
<tr>
<td>CX2</td>
<td>0.25</td>
<td>0.067</td>
<td>0.53</td>
<td>0.06</td>
<td>0.81</td>
</tr>
<tr>
<td>CX2</td>
<td>1.50</td>
<td>0.408</td>
<td>3.31</td>
<td>0.33</td>
<td>5.20</td>
</tr>
<tr>
<td>CX2</td>
<td>3.00</td>
<td>0.994</td>
<td>7.00</td>
<td>0.52</td>
<td>15.1</td>
</tr>
<tr>
<td>CX3</td>
<td>1.50</td>
<td>0.405</td>
<td>3.32</td>
<td>0.33</td>
<td>5.21</td>
</tr>
</tbody>
</table>
Figure 72. Effect of Mesh Refinement on Centerline $u_2$: (a) Mesh CX1, (b) Mesh CX2, (c) Mesh CX3.
Figure 73. Effect of Mesh Refinement on Centerline $\tau_{11}$: (a) Mesh CX1, (b) Mesh CX2, (c) Mesh CX3.
Figure 74. Effect of Mesh Refinement on Centerline $\tau_{ij}$: (a) Mesh CX1, (b) Mesh CX2, (c) Mesh CX3.
are much too high. While the choice of the stress optical coefficient used in the calculations may affect the predicted patterns, the magnitude of the disparity is too great to attribute to this possibility.

Attempts to simulate the joining flow behavior of LDPE were not successful. The upper limit of convergence for LDPE corresponded to a flow rate of only 0.25cc/s/m. This is less than one-twentieth of the lowest value obtained experimentally. At this level, the predicted birefringence pattern shown in Fig. 76, does not have enough characteristics to even qualitatively compare with the experimental behavior. The low flow-rate level at the limit of convergence results from the large relaxation time constant. The role of $\lambda$ in determining the limit of convergence is discussed in more detail in the next part of this section.

It is suggested by the qualified success of the LLDPE simulations, that over the range of converged solutions, the predictions of the numerical model qualitatively represent the behavior observed experimentally. As a result, the model should be able to accurately predict trends in behavior associated with different rheological behavior. It is also indicated by the low flow rate at the upper level of convergence for LDPE, that the range of flow rates for which the method converges is strongly influenced by the relaxation behavior incorporated in the model.

5.3.2.3 Effect of $\lambda$ and $\epsilon$

The simulations conducted to examine the influence of $\lambda$ and $\epsilon$ on the joining flow behavior are summarized in Table 5. In the experiments, the effect of independently varying $\lambda$ and $\epsilon$ for NFL00 (Table 3) was examined. Relaxation times ranged from 0.125s to 2.0s and $\epsilon$ was varied from 0.01 to 0.10. All simulations were conducted using Mesh CX2. The upper limit of convergence for these runs corresponded to a Deborah number of approximately 1. As a result, in order to compare the results at equivalent flow rates, a low value of $Q$ was chosen to accommodate the solution for a 2.0s relaxation time.
Figure 75. Numerical and Experimental Joining Flow Birefringence for LLDPE
(a) Experimental - Q = 6.69 cc/s/m, (b) Numerical - Q = 3.0 cc/s/m.
Figure 76. Numerical Joining Flow Birefringence for LDPE: $Q = 0.25 \text{cc/s/m.}$
**Table 5. Summary of Parameter Effect Simulations**

<table>
<thead>
<tr>
<th>RUN</th>
<th>PARAMETER</th>
<th>Q</th>
<th>De</th>
<th>$\hat{\varepsilon}(r^{-1})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>406</td>
<td>BASIC (NFL00)</td>
<td>0.20</td>
<td>0.436</td>
<td>0.60</td>
</tr>
<tr>
<td>405</td>
<td>BASIC (NFL00)</td>
<td>0.30</td>
<td>0.713</td>
<td>1.50</td>
</tr>
<tr>
<td>417</td>
<td>$\lambda = 2.000$</td>
<td>0.20</td>
<td>0.929</td>
<td>0.60</td>
</tr>
<tr>
<td>418</td>
<td>$\lambda = 1.500$</td>
<td>0.20</td>
<td>0.673</td>
<td>0.60</td>
</tr>
<tr>
<td>419</td>
<td>$\lambda = 0.500$</td>
<td>0.20</td>
<td>0.214</td>
<td>0.60</td>
</tr>
<tr>
<td>420</td>
<td>$\lambda = 0.250$</td>
<td>0.20</td>
<td>0.170</td>
<td>0.60</td>
</tr>
<tr>
<td>421</td>
<td>$\lambda = 0.125$</td>
<td>0.20</td>
<td>0.053</td>
<td>0.60</td>
</tr>
<tr>
<td>424</td>
<td>$\varepsilon = 0.010$</td>
<td>0.30</td>
<td>0.728</td>
<td>1.40</td>
</tr>
<tr>
<td>425</td>
<td>$\varepsilon = 0.020$</td>
<td>0.30</td>
<td>0.729</td>
<td>1.40</td>
</tr>
<tr>
<td>426</td>
<td>$\varepsilon = 0.050$</td>
<td>0.30</td>
<td>0.732</td>
<td>1.40</td>
</tr>
</tbody>
</table>
The effects of $\lambda$ on $u$, and $\tau_{11}$ along the downstream centerline are shown in Figs. 77 and 78, respectively. $\lambda$ has only a slight effect on the developing velocity field, but its impact on $\tau_{11}$ is quite significant. From a numerical standpoint, this effect is real, i.e. it is insensitive to initial guess and solution tolerance, but whether or not the magnitude is representative of real fluid behavior is suspect. The previous discussion of the unrealistic stresses generated in the LLDPE simulation have a direct bearing on these results. The problems associated with viscoelastic simulations in geometries involving corners, discussed in Chapter 2, would also suggest that the predicted magnitude is unrealistic. Further discussion of this point is reserved until the final section of this portion of the chapter. While the magnitude of the effect of $\lambda$ on $\tau_{11}$ may be suspect, the trend is probably correct.

The lack of influence of $s$ on the centerline velocity and stress is shown in Figs. 79 and 80. This result is quite surprising based on the effect of $s$ on the extensional viscosity (Fig. 70). The extension rate at the tip of the divider for these simulations is $1.4 s^{-1}$ (Table 5) which roughly corresponds to the maximum difference in the extensional viscosity shown in Fig. 70. The simulations do predict a rapid decrease in the extension rate moving away from the divider, but it would seem reasonable that the large difference in extensional viscosity would still have more of an effect on $\tau_{11}$.

5.3.2.4 Final Discussion

The relative success of the model to predict the joining flow behavior of LLDPE is tempered by its inability to converge at higher flow rates. A more detailed discussion of the limiting convergence problem is reserved until after the discussion of the slip-stick simulations in the next section. For the range of converged flow rates, the relaxation time clearly has more impact on the joining flow behavior than does the extensional viscosity. To a certain degree, this is an unexpected result. It is suggested by these results, that in terms of the joining flow behavior, matching materials based on their shear behavior alone should be sufficient to avoid problems with interface irregularities in bicomponent flow. It is parenthetically stated that this is based on results from a limited range of flow rate calculations.
Figure 77. Effect of $\lambda$ on Centerline $u_1$: $Q = 0.20 \text{cc/s/m.}$
Figure 78. Effect of \( \lambda \) on Centerline \( \tau_{11} \): \( Q = 0.20 \text{cc/s/m} \).
Figure 79. Effect of $\varepsilon$ on Centerline $u_1$: $Q = 0.30$ cc/s/m.
Figure 86. Effect of $\varepsilon$ on Centerline $\tau_{11}$: $Q = 0.30 \text{cc/s/m}$. 

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5.3.3 Stick-Slip Simulations

It was suggested by the results of the asymmetric coating experiments in Chapter 3, that the role of fluoro-elastomer in eliminating surface melt fracture behavior in LLDPE was to introduce a finite slip velocity at the wall. This supposition relied heavily on the interpretation of the birefringence patterns and lacked any direct observation of wall slip. Based on the success of the numerical model to qualitatively predict the joining flow behavior of LLDPE, it was decided to examine the effect of introducing finite wall slip velocities in flow between parallel plates for LLDPE numerically. The success or failure of the model to simulate the experimentally observed birefringence should provide additional insight into the behavior observed in Chapter 3. The results of these numerical studies are presented and discussed in this section.

5.3.3.1 Stick-Slip Meshes

Three different meshes were used in the stick-slip studies and are shown in Fig. 81. The arrows indicate the point in the meshes where slip velocities were introduced. Mesh SS1 contains 253 nodes, Mesh SS2 has 341 nodes and Mesh SS3, 585 nodes. Linear elements were used in all simulations. The physical domains of the meshes are identical, the only difference being the degree of refinement. The meshes extend for 5 \( \frac{L}{H} \)'s upstream and downstream of the point where slip is introduced. This provides a reasonable distance for imposing fully developed inlet and exit plane boundary profiles. Unlike typical stick-slip simulations, where slip is introduce symmetrically across the channel width, in the current study, slip is introduced only on the top wall.
Figure 81. Stick-Slip Meshes: (a) Mesh SS1, (b) Mesh SS2, (c) Mesh SS3.
5.3.3.2 *LLDPE Simulations*

Prior to evaluating the role of wall slip with respect to the experimental behavior, the effect of mesh refinement on the solutions was evaluated. The comparison of the different meshes was made at a flow rate of 16.0 cc/s/m and a ratio of the wall slip velocity, $u_s$, to the maximum velocity, $u_m$, of 0.12. This flow rate corresponds to an upstream Deborah number ($De = \dot{\gamma} \lambda$) of 2.3. The results of the mesh comparison for the velocity and birefringence development are shown in Figs. 82 and 83, respectively. It is suggested by these comparisons, that there is no significant difference in the predicted velocity field and only a slight difference on the birefringence predicted at the wall ($Y/H = 1.00$). As a result, it was decided to incorporate Mesh SS2 in the remaining simulations. It’s slightly higher mesh refinement than Mesh SS1, in the down channel direction was desirable.

The role of wall slip behavior for LLDPE was evaluated over a range of flow rates from 1.0 to 70 cc/s/m. This corresponds to Deborah numbers between 0.10 and 14.1. In addition to varying the flow rate, the ratio of $u_s$ to $u_m$ was examined over a range from 0.04 to 1.0. A $u_s/u_m$ value of 1.0 corresponds to complete slip at the wall, i.e. the velocity profile reaches a maximum at the wall. For this condition, the upper limit of convergence is reduced from a Deborah number of 14.1 to 5.0. The high level of convergence for $u_s/u_m$ is not unexpected: the problem approaches the trivial one-dimensional case of no wall slip for flow between parallel plates. What is surprising is the relatively high Deborah number for the case of complete slip. This flow is almost directly analogous to the joining flow behavior discussed in the previous section. The limiting value of $De$ in that geometry was determined to be approximately 1. The limits of convergence for the different geometries are discussed in more detail in the next section of this part of the chapter.

Comparisons of the numerical and experimental birefringence patterns for three different flow rates are shown in Figs. 84, 85 and 86. For the first two rates, a $u_s/u_m$ value of 0.08 was used in the numerical simulation, while for the third rate, a value of 0.12 was required. The simulations are qualitatively in excellent agreement with the experimental results. The asymmetric downstream
Figure 8.2. Effect of Meshes on $\nu$, Development: (a) $Y/H = 0.50$, (b) $Y/H = 1.00$. 
Figure 83. Effect of Meshes on Birefringence Development: (a) $Y/H = 0.50$, (b) $Y/H = 1.00$. 
fringe behavior is accurately predicted as well as the upstream and downstream rearrangement behavior. Quantitatively, the stick-slip simulations suffer from the same problem as the joining flow simulations: the flow rate in the numerical simulations are much lower than those observed experimentally. Again, it is suggested by these results that model predicts stresses which are much higher than those realized experimentally. Given this limitation, the supposition of wall slip in the experimental observations for the asymmetric coating experiments in Chapter 3 is strongly supported by the results of these simulations.

5.3.3.3 NFL-41 Simulations

The final set of numerical experiments compared the stick-slip behavior of NFL-41 (Table 3) with the behavior for LLDPE. NFL-41 has a longer relaxation time and lower ε value (this latter difference is not expected to play a significant role). The flow rate and Deborah number corresponding to the upper limit of convergence were both reduced for these constants. For a $\mu_r/\mu_a$ ratio of 0.12, the maximum Deborah number realized for NFL-41 was 11.5, compared with a value of 14.1 for LLDPE. Comparisons of the birefringence development at two different cross-channel positions are shown in Figs. 87 and 88. The magnitude of the birefringence is higher for NFL-41 at both positions and the degree of overshoot at the wall is also higher for NFL-41, but the down-channel lengths involved in the rearrangement of the flows are equivalent. It is suggested by these results, that relaxation time does affect the overshoot behavior as expected, but has little effect on the overall rearrangement behavior.

5.3.4 Final Considerations: Limits of Convergence

In the previous two sections, the limits of convergence in the joining flow and slip-stick geometries were presented. Prior to moving on to the last part of this chapter, a few final points regarding
Figure 84. Experimental vs Numerical Birefringence Patterns: (a) Experimental - $Q_E = 8.7$,
(b) Numerical - $Q_N = 3.5$. 

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Figure 85. Experimental vs Numerical Birefringence Patterns: (a) Experimental - $Q_\varepsilon = 12.2$, (b) Numerical - $Q'_{\varepsilon} = 5.0$. 
Figure 86. Experimental vs Numerical Birefringence Patterns:  
(a) Experimental - $Q_E = 15.3$. 
(b) Numerical - $Q_N = 6.0$. 

NUMERICAL STUDIES
Figure 87. Birefringence Development - LLDPE constants vs NFL-41 \((Q = 8.0, Y/H = 1.00)\)

- (a) LLDPE
- (b) NFL-41
Figure 38. Birefringence Development - LLDPE constants vs NFL-41 (Q = 8.0, Y/H = 0.83)
(a) LLDPE, (b) NFL-41.
these limits need to be made. First, the higher limit for the stick-slip problem compared with the joining flow limit is as expected: the severity of discontinuity in the joining flow problem is greater. In addition, the abrupt acceleration of the fluid along the wall in the stick-slip problem is imposed as a boundary condition rather than being determined by solution. For the lower values of $\nu_{\infty}/\nu_{m}$, the acceleration is minimal. That the solutions converge up to Deborah numbers above 10 is not surprising. The second point concerns the convergence limit for the 'full slip' condition compared with the joining flow limit. Physically, full slip is analogous to the joining flow problem with a infinitesimal divider. The major difference between the two problems numerically is that the velocity field is specified in one problem and implied in the other. It would seem that the acceleration required for satisfying the imposed velocity condition (the flow accelerates from 0 to $u_{m}$ over 1 element) would lead to substantially higher stresses, than when the flow is allowed to rearrange over a greater distance. Failure of viscoelastic simulations have been typically associated with the inability of the method to handle the high stresses generated at singularities and would suggest that the 'full slip' condition should fail to converge at a lower value. This was not the case. The final point to made is the limits of convergence compared with the abrupt contraction geometry. Gotsis [62] cited a limiting value of 4.1 for the Deborah number in a planar 4:1 contraction. This value corresponds well with the 'full slip' result determined in the current work. The effect of the geometric discontinuity in the joining flow problem at the divider was expected to be less severe than the corner discontinuity in the contraction problem. Unfortunately, this apparently is not the case.

5.4 Conclusions and Recommendations

In this, the final part of the chapter, the conclusions drawn from the results discussed previously and suggestions for future studies are given.
5.4.1 Conclusions

1. The incorporation of the Phan-Thien Tanner constitutive model in the finite element code PTTFEM provides a useful tool for qualitatively predicting the joining and stick-slip behavior of linear low-density polyethylene. The results of both the joining flow and stick-slip simulations of LLDPE are qualitatively in good agreement with the behavior observed experimentally, i.e. the predicted shapes of the birefringence patterns match those observed experimentally. However, the success of the model is restricted to qualitative predictions: in both geometries the predicted magnitudes of the stresses were more than twice those observed for an equivalent experimental flow rate.

2. Within the limits of converged solutions, shear flow properties are sufficient for predicting the flow behavior in the joining flow geometry. It was shown by the results of the experiments examining the influence of the parameter ε, that the difference in extensional properties predicted for the extension rates observed at the divider had no significant effect on the developing stress field. It is emphasized here, that this conclusion is drawn from a limiting range of flow rates.

3. The supposition that wall slip resulted in the observed birefringence patterns for the asymmetric coating experiments is correct. The patterns observed experimentally were qualitatively predicted by the incorporation of wall slip in the numerical simulations of LLDPE. It is important to again highlight that the results are qualitative and not quantitative.

4. The joining flow problem provides a more severe test for numerical simulations than either the stick-slip or planar contraction problem. It was shown by comparing the limits of convergence for the ‘full slip’ condition in the stick-slip problem, the planar 4:1 contraction problem and the joining flow problem, that the solution for the joining flow problem diverges at a much lower value of the Deborah number.
5.4.2 Recommendations

1. Compare the influence of differing relaxation and extensional properties on the bicomponent interface in the joining flow geometry directly. While the results of the single component simulations provide valuable insight into the factors affecting the flow behavior in a joining flow geometry, they cannot replace the information from direct multi-component simulations.

2. Compare the influence of two materials exhibiting different vortex growth behavior on the bicomponent interface in a planar contraction. Bicomponent simulations in the planar contraction geometry may well provide additional insight into bicomponent interface stability.

3. Simulate the joining flow behavior in a ‘Y’ configuration with the upstream channels having the same dimension as the downstream channel. The high upstream Deborah number problem could be circumvented by increasing the upstream channel dimensions allowing for higher total flow rates for equivalent values of De or $\tau_w$. 

NUMERICAL STUDIES
References


17. Dennison, M.T., Plastics Polymers, 35 (1967)


References


References


74. Mitsoulis, E., Int. J. Num. Meth. Fluids,


References

230

Appendix A. Flow Birefringence

The optical properties of a polymer molecule are locally anisotropic along the chain, i.e. the refractive index along the chain axis is different than the index transverse to the axis direction. When the polymer is in the melt state under quiescent conditions, the random orientation of the chains leads to globally isotropic optical behavior within the system. However, the stresses acting on the melt under flow conditions lead to global orientation of the molecules and anisotropic optical behavior. The method of flow birefringence takes advantage of this optical anisotropy to determine the level of stress in the melt under conditions of flow. In this appendix, a brief overview of the applied flow birefringence theory is presented. A more theoretical development has been presented by White [84] and will not be duplicated here.

There are three basic assumptions in the birefringence theory applied in the current study: first, the flow field is assumed to be two-dimensional; second, the real polymer chain in the melt is accurately described by a statistical chain; and third, only small deformations of the chain are occurring. There are then two principal stresses, $\Pi_{t}$ and $\Pi_{n}$, acting on the fluid at an angle $\chi'$ relative to the local direction of flow. The principal stress difference, $\Pi = \Pi_{t} - \Pi_{n}$, is related to the extra stresses by

$$\pi \sin 2\chi' = 2\tau_{12}$$

$A1$
\[ \Pi \cos 2\chi' = \tau_{11} - \tau_{22} \quad A2 \]

In a similar fashion, the birefringence, \( \Delta n \), is related to the components of the refractive index tensor, \( g \), by

\[ \Delta n \sin 2\chi = 2n_{12} \quad A3 \]

\[ \Delta n \cos 2\chi = n_{11} - n_{22} \quad A4 \]

where \( \chi \) is the extinction angle. Expressions for the refractive index behavior of a material in terms of the applied stresses can be obtained from the stress optic law. Specifically, these expressions are given by

\[ n_{12} = C\tau_{12} \quad A5 \]

\[ n_{11} - n_{22} = C(\tau_{11} - \tau_{22}) \quad A6 \]

where \( C \) is the material dependent stress optical coefficient. Incorporation of Eqs. A5 and A6 into Eqs. A1-A4 results in expressions for the applied stresses in terms of the birefringence:

\[ \Delta n \sin 2\chi = 2C\tau_{12} \quad A7 \]

\[ \Delta n \cos 2\chi = C(\tau_{11} - \tau_{22}) \quad A8 \]

In order to resolve the shear stress and the normal stress difference from the birefringence, it is necessary to determine the extinction angle \( \chi \) through experiments which involve generating isoclinic birefringence patterns. These experiments were not conducted in the work presented in this thesis, rather, isochromatic fringe patterns were obtained. In order to analyze the isochromatic patterns, Eqs. A7 and A8 are combined to obtain the following expression for the state of stress in the fluid:

\[ \Delta n = C \sqrt{4\tau_{12}^2 + (\tau_{11} - \tau_{22})^2} \quad A9 \]
The final point to be made in terms of flow birefringence is in regard to the order of the observed fringes. The retardation, $N$, is related to the birefringence by the following relationship

$$N = \frac{\Delta n L}{\lambda}$$

In this equation, $L$ is the width of the sample and $\lambda$ is the wave-length of the laser light source. For the apparatus described in Chapter 3, half-order fringes are observed, e.g. $1/2, 3/2, 5/2$, etc....
Appendix B. Rheological Data
### Steady Shear Data

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<th>$N_t$ (Pa)</th>
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Table 7. Capillary Data - MJA 042, 190 °C

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Appendix B. Rheological Data
Table 8. Extensional Data - MJA 042, 190 °C

\[ \varepsilon = 0.060 \text{s}^{-1} \]

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Appendix B. Rheological Data
\( \dot{\varepsilon} = 0.100 \text{ s}^{-1} \)

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Table 10. Extensional Data - MJA 042, 190 °C

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Appendix B. Rheological Data
Table 11. Shear Data - MJA 042/200ppm FE, 190 °C

### Steady Shear Data

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<th>( \eta ) (PaS)</th>
<th>( N_t ) (Pa)</th>
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<td>0.1213e+04</td>
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<tr>
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<td>0.4568e+03</td>
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<tr>
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### Dynamic Shear Data

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Appendix B. Rheological Data
Table 12. Capillary Data - MJA 042/200pmm FE, 190 °C

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<th>(\dot{\gamma}_s \text{ (1/S)})</th>
<th>(\tau_s \text{ (Pa)})</th>
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Table 13. Extensional Data - MJA 042/200pmm FE, 190 °C

\[ i = 0.060 s^{-1} \]

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Appendix B. Rheological Data
Table 14. Extensional Data - MJA 042/200pm Fe, 190 °C

\[ \dot{\varepsilon} = 0.100 \text{s}^{-1} \]

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<th>( \sigma_{\varepsilon} (\text{Pa}) )</th>
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Table 15. Extensional Data - MJA 042/200 ppm Fe, 190 °C

\[ \dot{\varepsilon} = 0.3 \text{ s}^{-1} \]

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<th>( \sigma_\text{e}(\text{Pa}) )</th>
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<td>0.0000E + 00</td>
<td>0.0000E + 00</td>
<td>0.0000E + 00</td>
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Table 16. Shear Data - MJA 042/500ppm FE, 190 °C

Steady Shear Data

<table>
<thead>
<tr>
<th>$\dot{\gamma}$ (s$^{-1}$)</th>
<th>$\eta$ (PaS)</th>
<th>$N_1$ (Pa)</th>
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<tbody>
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<td>0.1000e-01</td>
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<td>0.1616e+02</td>
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<tr>
<td>0.2154e-01</td>
<td>0.1394e+05</td>
<td>0.1396e+02</td>
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<tr>
<td>0.4641e-01</td>
<td>0.1182e+05</td>
<td>0.2425e+02</td>
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<tr>
<td>0.1000e+00</td>
<td>0.1311e+05</td>
<td>0.1469e+03</td>
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<tr>
<td>0.2154e+00</td>
<td>0.1201e+05</td>
<td>0.8993e+03</td>
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<tr>
<td>0.4641e+00</td>
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<td>0.2729e+04</td>
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<tr>
<td>0.1000e+01</td>
<td>0.8209e+04</td>
<td>0.7457e+04</td>
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<td>0.1167e+05</td>
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Dynamic Shear Data

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<tr>
<th>$\omega$ (rad/S)</th>
<th>$\eta''$ (PaS)</th>
<th>$G'$ (Pa)</th>
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<td>0.1000e+00</td>
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<td>0.1778e+00</td>
<td>0.1232e+05</td>
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Table 17. Capillary Data - MJA 042/500pmm FE, 190 °C

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<th>( \dot{\gamma} ) (1/S)</th>
<th>( \tau_s ) (Pa)</th>
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<tr>
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<th>( \tau_s ) (Pa)</th>
<th>( \dot{\gamma} ) (1/S)</th>
<th>( \tau_s ) (Pa)</th>
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<th>( \dot{\gamma} ) (1/S)</th>
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Table 18. Extensional Data - MJA 042/500pmm FE, 190 °C

\[ \dot{\epsilon} = 0.060 s^{-1} \]

<table>
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<th>( \sigma ) (Pa)</th>
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</table>

Appendix B. Rheological Data 248
<table>
<thead>
<tr>
<th>TIME (S)</th>
<th>STRAIN</th>
<th>$\bar{\eta}$ (PaS)</th>
<th>$\sigma_z$ (Pa)</th>
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Table 20. Extensional Data - MJA 042/500ppm FE, 190 °C

\[ \dot{\varepsilon} = 0.300s^{-1} \]

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<th>( \sigma(\text{Pa}) )</th>
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Table 21. Shear Data - MJA 042/1000ppm FE, 190 °C

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<tr>
<th>γ (s⁻¹)</th>
<th>η (PaS)</th>
<th>N₁ (Pa)</th>
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<tr>
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<tr>
<td>0.2154e-01</td>
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<tr>
<td>0.4641e-01</td>
<td>0.1431e+05</td>
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Dynamic Shear Data

<table>
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<th>G' (Pa)</th>
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Table 22. Capillary Data - MIA 042/1000μm FE, 190°C

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<th>( \tau_e (\text{Pa}) )</th>
<th>( \dot{\gamma}_e (1/S) )</th>
<th>( \tau_e (\text{Pa}) )</th>
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<table>
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<th>( \tau_e (\text{Pa}) )</th>
<th>( \dot{\gamma}_e (1/S) )</th>
<th>( \tau_e (\text{Pa}) )</th>
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<th>( \tau_e (\text{Pa}) )</th>
<th>( \dot{\gamma}_e (1/S) )</th>
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Appendix B. Rheological Data
Table 23. Extensional Data - MJA 042/100×9mm FE, 190 °C

\[ \dot{\gamma} = 0.060 s^{-1} \]

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Appendix B. Rheological Data 253
Table 24. Extensional Data - MJ0 042/1000 ppm FE, 190 °C

\[ \dot{\epsilon} = 0.100 \text{s}^{-1} \]

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<th>( \sigma_x (\text{Pa}) )</th>
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Appendix B. Rheological Data
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<th>$\sigma_E$ (Pa)</th>
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### Table 26. Shear Data - MJA 042/2000ppm FE, 190 °C

#### Steady Shear Data

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<td>0.1670e+05</td>
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<td>0.1440e+05</td>
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#### Dynamic Shear Data

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<td>0.1024e+05</td>
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Appendix B. Rheological Data
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<th>$\tau_s$ (Pa)</th>
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<td>$\dot{\gamma}$ (s$^{-1}$)</td>
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<th>$G'$ (Pa)</th>
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Table 29. Capillary Data - NTA 101, 170 °C

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<th>( \dot{\gamma} ) (1/S)</th>
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### Table 30. Extensional Data - NTA 101, 170 °C

\( \dot{\gamma} = 0.020 \text{s}^{-1} \)

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<td>0.0000E + 0</td>
<td>0.0000E + 0</td>
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Table 31. Extensional Data - NTA 101, 170 °C

\[ \dot{\varepsilon} = 0.353 s^{-1} \]

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<tr>
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<td>0.1120E + 01</td>
<td>0.3792E + 05</td>
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Appendix B. Rheological Data
Table 32. Extensional Data - NTA 101, 170 °C

\( \dot{\varepsilon} = 0.200 s^{-1} \)

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<th>( \sigma_e (\text{Pa}) )</th>
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<td>0.00000E+00</td>
<td>0.00000E+00</td>
<td>0.00000E+00</td>
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Appendix B. Rheological Data
## Table 33. Shear Data - NPE 953, 170 °C

### Steady Shear Data

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### Dynamic Shear Data

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Table 35. Extensional Data - NPE 953, 170 °C

\[ \dot{\gamma} = 0.020 s^{-1} \]

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**Table 36. Extensional Data - NPE 953, 170°C**

\( \varepsilon = 0.053 \)
### Table 37. Extensional Data - NPE 953, 170 °C

\[ \dot{\zeta} = 0.200 \tau^{-1} \]

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</table>
Appendix C. PTT Rheological Predictions
Figure 39. Effect of $\eta_1$ on Shear Viscosity
Figure 90. Effect of $\eta_1$ on $G'$
Figure 91: Effect of $\eta_1$ on Extensional Stress Growth: $\dot{\gamma} = 0.20 \text{s}^{-1}$. 

Appendix C: PTT Rheological Predictions
Figure 92. Effect of $\eta_1$ on Extensional Viscosity
Figure 93. Effect of $\lambda$ on Shear Viscosity
Figure 94. Effect of $\lambda$ on $G'$
Figure 95. Effect of $\dot{\gamma}$ on Extensional Stress Growth: $\dot{\gamma} = 0.20 \text{ s}^{-1}$. 

Appendix C. PTT Rheological Predictions
Figure 96. Effect of $\lambda$ on Extensional Viscosity
Figure 97. Effect of $\xi$ on Shear Viscosity
Figure 93. Effect of $\xi$ on $G'$
Figure 99. Effect of $\xi$ on Extensional Stress Growth: $\dot{i} = 0.20s^{-1}$. 

Appendix C. PTT Rheological Predictions
Figure 100. Effect of $\zeta$ on Extensional Viscosity
Figure 191. Effect of $\varepsilon$ on Shear Viscosity
Figure 102. Effect of $\varepsilon$ on $G'$
Figure 103. Effect of $\varepsilon$ on Extensional Stress Growth: $i = 0.20 \text{s}^{-1}$. 

Appendix C. PTT Rheological Predictions
Figure 104. Effect of $\varepsilon$ on Extensional Viscosity
Figure 105. Effect of \( \eta_1/\eta_2 \) on Shear Viscosity.
Figure 106. Effect of $\eta_1/\eta_2$ on $G'$

Appendix C. PTT Rheological Predictions
Figure 107. Effect of $\eta_1/\eta_2$ on Extensional Stress Growth: $\dot{e} = 0.20$ s$^{-1}$.
Figure 108. Effect of $\eta_1/\eta_2$ on Extensional Viscosity
Appendix D. Multi-Fluid Simulations

The single fluid simulations of Chapter 5 provided insight into the factors which play a significant role in joining flow behavior. Based on the results of these simulations, it was suggested that the shear-flow properties alone were sufficient for determining compatibility of different materials as they came into contact in this geometry. Unfortunately, the single fluid simulations did not provide any insight into how differences in the shear-flow behavior of the individual layers would affect the interface behavior. In order to address this point, it is necessary to directly simulate the bicomponent behavior. In this appendix, the efforts made to accomplish the bicomponent simulations are presented.

D.1.1 Introduction

The equations which govern the flow of stratified fluids are the same as those for single component flows (Eqs. 5.1-5.3), but are written for each layer (Fig. 109) of the flow as follows:

\[ \nabla \cdot u^a = 0 \quad D1 \]

\[ \nabla p^a + \nabla \cdot x^a = 0 \quad D2 \]
where \( \nu^*, \tau^* \) and \( P^* \) are the velocity, extra stress and pressure in fluid \( a \), respectively.

Solution of Eqs. D1-D3 requires boundary conditions at the interface for \( \nu^*, \tau^* \) and \( P^* \). The appropriate conditions at the interface for the velocity are: no flow across the interface in either layer and the velocity in each layer is the same at the interface (no slip). These conditions are expressed by

\[
\nu^I \cdot n^I = \nu^I \cdot n^I = 0 \quad D4a
\]

\[
\nu^I \cdot l^I + \nu^I \cdot l^{II} = 0 \quad D4b
\]

where \( n^* \) and \( l^* \) are the unit normal and tangent vectors at the interface in layer \( a \).

The appropriate conditions for \( P^* \) and \( \tau^* \) are found by performing a force balance at the interface. At steady state it is required that

\[
\mathcal{I}^I + \mathcal{I}^{II} = 0
\]

where \( \mathcal{I}^* = \mathcal{g}^* \cdot n^* \)

and \( \mathcal{g}^* = P^* \hat{n}^* + \tau^* \)

is the total stress in fluid \( a \). Letting \( \mathcal{g} = \mathcal{g}^I = -\mathcal{g}^{II} \), the force balance at the interface becomes

\[
\mathcal{g}^I \cdot n = \mathcal{g}^{II} \cdot n \quad D5
\]

Eq. D5 is readily decomposed into its normal and tangent component form

\[
(\mathcal{g}^I \cdot n) \cdot n = (\mathcal{g}^{II} \cdot n) \cdot n \quad D5a
\]

\[
(\mathcal{g}^I \cdot n) \cdot t = (\mathcal{g}^{II} \cdot n) \cdot t \quad D5b
\]

where \( t \equiv l^I = -l^{II} \).
Figure 109. Typical Interface in Bicomponent Flow
While Eq. D5 requires continuity of the total stress, $\sigma$, at the interface it does not imply continuity of the pressure or the extra stress. This is most easily demonstrated by considering the case of two Newtonian fluids. For this case, Eq. D5b becomes

$$- p^I + 2\mu^I \frac{\partial u^I}{\partial n} \bigg|_I = - p^II + 2\mu^II \frac{\partial u^II}{\partial n} \bigg|_II \quad D6$$

Conservation of mass requires that at the interface

$$\frac{\partial u^I}{\partial n} \bigg|_I = \frac{\partial u^II}{\partial n} \bigg|_II = - \frac{\partial u^I}{\partial t} \bigg|$$

As a result, unless $\mu^I = \mu^II$, the pressure and the extra stress are discontinuous across the interface.

D.1.2 Implementation of the Finite Element Method

There were three primary considerations in implementing the finite element method for bicomponent simulations. First, it was necessary to incorporate different material constants for each fluid. Second, a method for handling the discontinuity in the total stress along the interface was required. Finally, a means of determining the interface position, which is unknown a priori, was needed. A discussion of these considerations is presented in the following paragraphs.

Incorporation of different material constants into the simulations was trivial if an initial shape of the interface was assumed. The mesh was constructed so that the assumed interface was coincident with element boundaries. Elements were then assigned to the appropriate material constant sets, depending on which layer of the flow they were in. The element contributions to each node were then calculated using the proper constants. The method of updating the interface position will be discussed in the last part of this section.
The discontinuities in $\tau$ and $P$ required special treatment in the implementation of the finite element method for simulating bicomponent flows. Discontinuous pressures were readily handled by the penalty formulation of the equation of motion, i.e. the pressure was eliminated as a nodal unknown. The discontinuity of the extra stress across the interface was addressed by incorporating double nodes along the interface (Fig. 110). Each pair of nodes on the interface had the same physical coordinates, but were assigned to different elements (fluids). As a result, the extra stress in one fluid could be calculated independently of the other fluid. The assumption of an interface position was required for this method, similar to the incorporation of different material constant sets. Imposition of the appropriate continuity conditions and subsequent interface location updating is presented in the following paragraphs.

The implementation of both double nodes and multiple material constant sets required that the interface location, which was unknown a priori, be assumed prior to the solution of the problem. As a result, unless the correct interface position is chosen, the required conditions at the interface, discussed in the introduction, cannot be satisfied. By relaxing one of the required conditions and generating a solution, the interface position may be updated by comparing the solution with the relaxed condition. This point will be clarified directly.

There are three conditions which must be satisfied at the interface: continuity of the velocity, no normal flow across the interface and continuity of the shear stress. These are stated mathematically as

\begin{align*}
  u_i' &= u_i'' & \text{(D7a)} \\
  u_n' &= u_n'' &= 0 & \text{(D7b)} \\
  \tau_{12} &= \tau_{12}' & \text{(D7c)}
\end{align*}
Figure 110.  Double-Node Description of $\tau$ Along the Interface.
This last condition results from expanding Eq. D5: while the extra normal stresses are discontinuous across the interface, the shear stress is not. The methodology for handling the interface location is as follows:

1. An initial shape of the interface was assumed.

2. The mesh for the problem was developed based on the interface shape.

3. The solution was generated after imposing two of three interface conditions (Eqs. D7a-D7c).

4. The solution along the interface was compared with the relaxed condition.

5. If the relaxed condition was satisfied, the process was terminated. If not, a new interface shape was assumed and steps 2-4 were repeated.

Implementation of the interface conditions was accomplished by the subroutine DBLNOD in the program PTTFEM (Appendix E). Simulations were conducted using modified versions of meshes CX1, CX2 and CX3 (Chapter 5). A discussion of the results of the bicomponent simulations is presented in the next section of this appendix.

D.1.3 Results

The majority of the bicomponent simulations which were conducted, incorporated the same material constants for both fluids. The reason for this was so that performance of the bicomponent simulations could be evaluated compared with single component predictions. While the discussion here will focus on these simulations, the results are similar for simulations employing fluids with different material constants. In short, the bicomponent simulations were not successful.
The results of imposing continuity of $\mu$ and $\tau_{12}$ (Eqs. D7a and D7c) are compared with the single component solution at equivalent conditions in Figs. 111 to 115. Severe oscillations in the velocity and shear-stress along the entire interface were predicted by the bicomponent simulations (Figs. 111 and 112). Results for $\tau_{11}$ are slightly better, but still show severe oscillations just after the contact point (Fig. 113). The failure of the simulations was not restricted to the interface behavior, but was also evident in the individual layers upstream of the contact point (Fig. 114) and across the full channel width downstream (Fig. 115). The results of imposing these two continuity conditions are representative of the results observed for imposing the other two combinations of interface conditions. The oscillations in the solutions were indicative of an over constrained system and as result, simulations employing only one of the appropriate interface conditions were conducted. For all three individual conditions, the results were similar to those shown in Figs. 111 to 115, i.e. the solutions did not improve.

In order to determine if the equation solver was responsible for the failure, the code was modified to employ a canned IMSL linear system solver. Favorable results were not realized. In a final attempt to obtain reasonable solutions, the stresses at the exit plane were specified. This also did not improve the results.

To summarize the bicomponent simulations, they did not work. Recommendations for future approaches is given in the next section of this appendix.

D.1.4 Recommendations

The failure of the author to obtain reasonable solutions for the bicomponent simulations makes it difficult to provide specific recommendations for future work. It is immediately obvious that if specific approaches to overcoming the problems encountered here were evident, they would have
Figure 111. Interface \( u_1 = u \) and \( \tau_{12} \) continuous.
Figure 112. Interface \( \tau_{12} \), \( \mu \) and \( \tau_{13} \) continuous.
Figure 113. Interface \( \tau_{ll} \), \( \mu \) and \( \tau_{ll} \) continuous.
Figure 114. Upstream $u$ Cross-Channel Profile: $y$ and $v_{13}$ continuous.
Figure 115. Downstream $u_4$ Cross-Channel Profile: $u$ and $\tau_{12}$ continuous.
been pursued. Having appropriately preaced the following recommendations, they are presented in this last section of this appendix.

1. **Develop the appropriate methods for handling bicomponent behavior with a less sophisticated constitutive model.** Eliminate any problems associated with viscoelastic simulations by first developing the appropriate methodology using Newtonian or power-law model. The model incorporated should be formulated using the mixed method rather a primitive u-v-p formulation. It is important to keep the extra stresses as primary variables if it is to be adapted to viscoelastic models.

2. **Reduce the geometric complexity of the simulations to eliminate problems associated with discontinuities.** In Chapter 5, the limits of convergence for the joining flow geometry were shown to be substantially lower than for these associated with either the 4:1 planar contraction or stick-slip geometry. By incorporating a simpler geometry, such as a slowly converging channel, the complexity of the problem can be reduced so that the focus is restricted to handling interface discontinuities rather than geometric singularities.
Appendix E. PTTFEM

PTTFEM FORTRAN  last revised by rhra 12/09/89
(add restart capability 12/09/89)
(add tracking capability 12/10/89)

INPUT VARIABLES:

-------------------

amul(i)...... PTT eta1 for fluid i
amun(i)...... 1st iteration newtonian eta for fluid i
beta......... acceleration parameter for piccard iteration
dksi(i)...... PTT ksi parameter for fluid i
epsil(i)...... PTT epsilon parameter for fluid i
ibf(nsdf)..... array of spec. NON-ZERO s.d.o.f. (strm-fnc)
ibdf(nsdf)..... array of spec. p.d.o.f.
ibs(nsdf)..... array of spec. p.d.o.f. (strm-fnc)
ibs(nbf)..... array of spec. NON-ZERO s.d.o.f.
istrt ....... restart condition for run
  0: normal entry, no restart file generated
  1: normal entry, restart file generated
  2: restart entry, no restart file generated
  3: restart entry, restart file generated
itmax........ maximum number of iterations for run
itrack ...... number of tracking files generated during run
itrk(i) ...... number of dof tracked in file i
iweci(4)...... 4 nodes at which the We & De numbers are calc
ndn(i,j)...... i = 1 to ndmn,
  vels at node j = 1 equal to vels at node j = 2
nem......... number of elements in the mesh
nfl......... number of fluids in the problem
ndnm........ number of nodes with common velocities
nnd........... number of nodes in the mesh
nod(i,j)...... j = i to npe, connectivity for element i
  j = npe + 1, fluid constant set for element i
npe......... number of nodes per element
npmt......... print flag for linear solution plots (uxl&u12)

    = 0 : no plot files created
    = 1 : plot files created
nsbf......... number of specified NON-ZERO secondary d.o.f
usbf......... number of specified primary d.o.f
nsf......... num of spec NON-ZERO secondary d.o.f (strm-fnc)
nsf5......... number of specified primary d.o.f (strm-fnc)
ntrk(i,j)..... dof j tracked in file i
pp......... penalty parameter
retrat(i)...... ratio of eta1/eta2 in PTT model for fluid i
den(i)......... density of fluid i
rxi(i)......... PTT relaxation time for fluid i
soc(i)......... stress optic coefficient for fluid i
toliter....... iteration tolerance
title......... title of run
vbf(nsf)...... values of ibf(nsf)
vbdf(nsd)..... values of ibdf(nsd)
vbd2f(nsd).... values of ibdf(nsd) for 1st iter (Newtonian)
vbs(nsf)...... values of ibs(nsf)
vbsf(nsbf)..... values of ibsf(nsbf)
vpf........... value for normalizing stream-function
x(i)........... x-coordinate of node i
xc(i)......... x-coordinate of the center of element i
y(i)........... y-coordinate of node i
yc(i)......... y-coordinate of the center of element i

INTERNAL VARIABLES:

icomi......... array of comments generated during execution
numc......... number of comments generated during execution
ndtype(i)..... fluid associated with node i
vai(npe*ndf).... element piccard updated solution vector

INPUT FILE FORMATS:

          card 1 (1000) title
card 2 (1010) nmm,npe,nfl,itmax,ntmp,isttr,itrack
card 3's (1010) (nod(n,j),i = 1,npt)
card 4's (1020) (x(i),y(i),i = 1,nmm)
card 5's (1020) amui(nfl),rxi(i),dks(i),epsil(i),den(i),
            retrat(i),amun(i)
card 6's (1020) (soc(i) i = 1,nfl)
card 7's (1020) pp,beta,toliter,vpsi
card 8 (1010) nsdf
card 9's (1010) (ibdf(i),i = 1,nsdf)
card 10's (1020) (vbdff(i),i = 1,nsdf)
card 11's (1020) (vbd2f(i),i = 1,nsdf)
card 12 (1010) nsbf
card 13's (1010) (ibsf(i),i = 1,nsbf)
card 14's (1020) (vbsf(i),i = 1,nsbf)
card 15 (1010) nsf
  card 16's (1010) (ibsf(i),i = 1,nsf)
card 17's (1020) (vbs(i),i = 1,nsf)
card 18 (1010) nsf
card 19's (1010) (ibf(i),i = 1,nsf)
card 20's (1020) (vb(i),i = 1,nsf)
card 21 (1010) ndn
cdcd card 22's (1010) (ndn(i),i, j = 1,2)
card 23 (1010) (iwe(i),i = 1,4)
card 24 (1010) (itrk(i),i = 1,itrack) if itrack > 0
card 25's (1010) (atrk(i,j),j = 1,itrack) if itrack > 0
card 26's (1040) (gfi,i = 1,neq) if istrt > 1

OUTPUT FILES:

-------------
unit 7 ............ contains complete solution
unit 8 (3e20.4)..... Ux plotting file
unit 9 (3e20.4)..... Uy plotting file
unit 10 (3e20.4)..... pressure plotting file
unit 11 (3e20.4)..... fluid 1 birefringence plotting file
unit 12 (3e20.4)..... fluid 2 birefringence plotting file
unit 13 (3e20.4)..... stream-function plotting file
unit 14 (10e13.5)... restart file
unit 15 (10e13.5)... tracking file #1
unit 16 (10e13.5)... tracking file #2
unit 17 (10e13.5)... tracking file #3
unit 18 (10e13.5)... tracking file #4
unit 19 (10e13.5)... tracking file #5
unit 21 (3e20.4)..... linear Ux plotting file
unit 22 (3e20.4)..... linear T12 plotting file

COMMENTS:

-------------
1. if the density is set to zero, then the inertial terms in the equation of motion are neglected (Stokes Flow)
2. if itmax is set to 1, only the Newtonian case is solved
3. if vpsl is set to zero, the stream-lines are not normalized

implicit real*8(a-h,o-z)
nodal variables
dimension ndtype(661),t11(661),t12(661),t22(661)
previous solution vectors
dimension gf(3305),gp(3305)
element variables
dimension xc(601),yc(601),shrt(601),extr(601),
boundary condition variables
dimension ibdf(351), vbdf(351), vbdf2(351), ibsf(199), vbsf(199),
1 ibs(151), vbs(151), ibf(101), vb(101)
tracking variables
dimension itrk(10), ntrk(10,10), vtrak(10)
extraneous variables
dimension icom(10), itime(0), ndn(50,2), iwei(4), wei(4), deb(4),
1 vel(45), title(20), soc(2)
mesh common block
common/msh/x(661), y(661), nod(601,10)
current element common block
common/elstil(45,45), elsy(9,2), f(45)
rheological constants common block
common/rheol/amu(2), rtx(2), dksi(2), epsil(2), den(2), retrast(2)
1 amnu(2)
global stiffness matrix common block
common/gstif/gstil(3305,301)
set number of generated comments to 0
ncom = 0

..............................................................
.
  preprocessor unit.
.
..............................................................
read input file over logical unit 5
call timeon
read(5,1000) title
read(5,1010) nem,nmm,npe,nfl, itmax, nprint, istrt, itrack
cset total element info to npe + 1 (add fluid constant set number)
npt = npe + 1
cread element connectivity & fluid constant set
do 10 n = 1, nem
read(5,1010) (nod(n,i), i = 1, npt)
10 continue
cread nodal x,y data
read(5,1020) (x(i), y(i), i = 1, nmm)
cread PTT constants for nfl sets
do 20 i = 1, nfl

Appendix E. PTTFEM
20 read(5,1030) amu1(i),rlx(i),dksi(i),epsil(i),den(i),
 1 retrat(i),amun(i)
c
  read stress optic coefficients for nfi sets
  read(5,1020) (soc(i),i = 1,nfl)
c
  read numerical solution parameters
  read(5,1020) pp,beta,toliter,vps

c
  read specified primary degrees of freedom
  read(5,1010) nsdf
  if (nsdf.eq.0) go to 30
  read(5,1010) (ibdf(i),i = 1,nsdf)
c
  if run is a restart skip newtonian b.c.'s
  if (istrt.ge.2) goto 25
  read(5,1020) (vbdfl(i),i = 1,nsdf)
25 read(5,1020) (vbdfl2(i),i = 1,nsdf)
c
  read specified non-zero secondary degrees of freedom
30 read(5,1010) nsbf
  if(nsbf.eq.0)goto 40
  read(5,1010) (ibsf(i),i = 1,nsbf)
  read(5,1020) (vbcs(i),i = 1,nsbf)
c
  read specified primary d.o.f. for the stream function
40 read(5,1010) nsdf
  if (nsdf.eq.0) go to 50
  read(5,1010) (ibdf(i),i = 1,nsdf)
  read(5,1020) (vbdf(i),i = 1,nsdf)
c
  read specified non-zero secondary d.o.f for the stream function
50 read(5,1010) nsf
  if (nsf.eq.0) go to 60
  read(5,1010) (ibf(i),i = 1,nsf)
  read(5,1020) (vbf(i),i = 1,nsf)
c
  read pairs of nodes with common velocities
60 read(5,1010) nddn
  if (nddn.eq.0) go to 80
  do 70 i = 1,nddn
   70 read(5,1010) (ndn(i,j),j = 1,2)
c
  read 4 nodes for calculation of De & We
80 read(5,1010) (swei(i),i = 1,4)
c
  read tracking information
  if (itrck.gt.0) then
   read(5,1010) (itrk(i),i = 1,itrck)
   do 90 i = 1,itrck
    90 read(5,1010) (iltrk(i,j),j = 1,itrk(i))
   endif
c
  end of data input (restart data read after line 320)

c
Appendix E. PTTFEM
check input data for errors or unreasonable values

c check for number of nodes greater than 661 (FATAL!)
  if (nnm.gt.661) goto 900

c check for number of elements greater than 601 (FATAL!)
  if (nem.gt.601) goto 910

c check for number of nodes per element (FATAL!)
  if ((npe.ne.4).and.(npe.ne.8).and.(npe.ne.9)) goto 920

c check for lack of boundary conditions (FATAL!)
  if (nsdf + nstf.eq.0) goto 930

c check for mis-match of nfi and nmdn (FATAL!)
  if ((nfl.gt.1).and.(nmdn.eq.0)) goto 940

c check for bogus number of fluids (FATAL!)
  if ((nfl.ne.1).and.(nfl.ne.2)) goto 950

c check for unreasonable tolerance (COMMENT)
  if (toliter.lt.1.d-8) then
    ncom = ncom + 1
    icom(ncom) = 1
    toliter = 0.01
  endif

c check for zero acceleration parameter (COMMENT)
  if (beta.lt.1.d-5) then
    ncom = ncom + 1
    icom(ncom) = 2
    beta = 1.
  endif

c check for ratio of retardation viscosity (COMMENT)
  do 100 i = 1,nfl
    if (retrat(i).gt.8.) then
      ncom = ncom + 1
      icom(ncom) = 3
      retrat(i) = 7.5
    endif
  100 continue

c set internal variables needed for computations

c set degrees of freedom per node to 5 (ux,uy,t11,t12,t22)
  ndf = 5

c calculate number of equations and degrees of freedom per element
  neq = nnn*ndf
  nn = npe*ndf

c set appropriate value of icl for use in gauss point determination
  icl = 2
if (npe.eq.4) iel = 1

c  set global tolerance
tol = (toiter*amul(1)**2*3.*dfloat(nnm))
c  reset current and previous error to 0
er1 = 0.
er2 = 0.
c  compute the half band width
nhbw = 0
do 210 n = 1,nem
do 210 i = 1,npe
do 210 j = 1,npe
   nw = (iabs(nod(n,i)-nod(n,j)) + 1)*ndf
210 if (nhbw.lt.nw) nhbw = nw

c  set full band width
nbw = 2*nhbw

c  determine node/fluid relation
do 220 i = 1,nem
   itype = nod(i,npt)
do 220 j = 1,npe
   k = nod(i,j)
220 ndtype(k) = itype

c  end of internal variable determination

c  check internal variables for errors

c  check for nbw greater than 301 (FATAL!)
if (nbw.gt.301) goto 960

c  check for constrained nodal data not within half band width
ndnmax = 0
do 230 i = 1,nndn
   nmax = iabs(ndn(i,1)-ndn(i,2))
if (nmax.gt.ndnmax) ndnmax = nmax
230 continue
if (ndnmax*ndf.gt.nhbw) goto 970

c  evaluate the time needed for read-in of input
call timeck(time(1))
c  write run parameters and constants

write(7,2000) title
write(7,2010) nnum,nxdf
write(7,2020) nem,nsbf
write(7,2030) neq,nsf
write(7,2040) nbw,nsf
```c
write(7,2050) nfl,nndn

write(7,2060) itmax,pp
write(7,2070) toliter,beta
write(7,2080) tol

if (nfl.eq.1) goto 240
write(7,2090)
write(7,2100) (amun(i),i = 1,nfl)
write(7,2110) (amu1(i),i = 1,nfl)
write(7,2120) (rlx(i),i = 1,nfl)
write(7,2130) (dksi(i),i = 1,nfl)
write(7,2140) (epsil(i),i = 1,nfl)
write(7,2150) (retrat(i),i = 1,nfl)
goto 250

240 write(7,2160) amun(1)
write(7,2170) amu1(1)
write(7,2180) rlx(1)
write(7,2190) dksi(1)
write(7,2200) epsil(1)
write(7,2210) retrat(1)

check for generated comments

250 if (ncom.eq.0) goto 270
write(7,8000)
do 260 i = 1,ncom
if (icom(i).eq.1) write(7,8010) i
if (icom(i).eq.2) write(7,8020) i
if (icom(i).eq.3) write(7,8030) i
260 continue

write solution heading
270 write(7,3000)

end of pre-processing

.................................................................

. processor unit . ...........................................

.................................................................

initialize the global stiffness matrix and force vector

do 300 i = 1,neq
gf(i) = 0.0
gp(i) = 0.0
300 gstmt(i,nbw) = 0.0

set istr to 0 (not currently performing stream-line calculations)
istr = 0

initialize iteration counter
```

Appendix E. PTTFEM
iter = 0

begin iteration loop for the non-linear problem

update iteration number and current values of previous errors

310 iter = iter + 1
err2 = err1
err1 = err
err = 0.

start clock for non-linear solution
if (iter.eq.2) call timeck(time(2))

update solution vectors, initialize global stiffness matrix

do 320 i = 1,neq
   gp(i) = gf(i)
gf(i) = gstiff(i,nbw)
do 320 j = 1,nbw
320    gstiff(i,j) = 0.0

if run is a re-start, read previous solution
if ((istrt.gt.2).and.(iter.eq.1)) then
   read(5,1040) (gf(i),i = 1,neq)
endif

loop over number of elements begins here

do 330 n = 1,nem

set acceleration parameter to 1 for first iteration
   b = beta
if(iter.eq.1) b = 1.

set itype to appropriate fluid set
   itype = nod(n,upt)

update nonlinear terms for element and set current elxy
call UPDATE(n,vel,b,df,gp,elxy,ndf,ape)

calculate element contributions to global stiffness matrix
call STIFFFQ(npe,ndf,iel,itype,pp,vel,istr,iter)

calculate element contributions in the global matrix
call ASMBLE(n,nbew,npe,ndf)

loop over elements ends here
330 continue

impose b.c.'s according to iteration & re-start condition
if ((iter.eq.1).and.(istr.le.1)) then
   call bound(nsbf,nsdf,ibsf,ibdf,vbsf,vbdf,nhbw)
goto 335
endif

endf
   call bound(nsbf,nsdf,ibsf,ibdf,vbsf,vbdf2,nhbw)

Appendix E. PTTFEM
continue

if single fluid, skip interface continuity
   if (nil.eq.1) goto 340
endif

if first iteration, skip forced continuity at the interface
   if (iter.eq.1) goto 340
endif

impose continuity of velocity at fluid/fluid interface
   call DBLNO(niter,ndn,ndn,nhbwn)
enddo
continue

check clock to determine time required for matrix inversion
   if (iter.eq.2) call timeck(ietime(3))
endif

solve assembled matrix for primary degrees of freedom
   (solution for i is returned in gestf(i,nbw))
   call SOLVER(neq,nhbw)
endif

check clock again and determine time required for matrix inversion
   if (iter.eq.2) call timeck(iitime(4))
endif

if desired, write tracking files at current iteration
   if (itrack.eq.0) goto 810
endif

if (iter.eq.1) write(15,2250) (ntrk(1,i),i = 1,itrk(1))
do 710 i = 1,itrk(1)
710 vr(1,i) = gestf(ntrk(1,i),nbw)
   write(15,1050) (vr(1,i),i = 1,itrk(1))
endif

if (itrack.eq.1) goto 810
if (iter.eq.1) write(16,2250) (ntrk(2,i),i = 1,itrk(2))
do 720 i = 1,itrk(2)
720 vr(2,i) = gestf(ntrk(2,i),nbw)
   write(16,1050) (vr(2,i),i = 1,itrk(2))
endif

if (itrack.eq.2) goto 810
if (iter.eq.1) write(17,2250) (ntrk(3,i),i = 1,itrk(3))
do 730 i = 1,itrk(3)
730 vr(3,i) = gestf(ntrk(3,i),nbw)
   write(17,1050) (vr(3,i),i = 1,itrk(3))
endif

if (itrack.eq.3) goto 810
if (iter.eq.1) write(18,2250) (ntrk(4,i),i = 1,itrk(4))
do 740 i = 1,itrk(4)
740 vr(4,i) = gestf(ntrk(4,i),nbw)
   write(18,1050) (vr(4,i),i = 1,itrk(4))
endif

if (itrack.eq.4) goto 810
if (iter.eq.1) write(19,2250) (ntrk(5,i),i = 1,itrk(5))
do 750 i = 1,itrk(5)
750 vr(5,i) = gestf(ntrk(5,i),nbw)
   write(19,1050) (vr(5,i),i = 1,itrk(5))
endif

if (itrack.eq.5) goto 810
if (iter.eq.1) write(20,2250) (ntrk(6,i),i = 1,itrk(6))
do 760 i = 1,itrk(6)

760 vtrk(i) = gstifntrk(6,i),nbw
    write(20,1050) (vtrk(i),i = 1,itrk(6))
c
    if (itrack.eq.6) goto 810
    if (iter.eq.1) write(21,2250) (ntrk(7,i),i = 1,itrk(7))
    do 770 i = 1,itrk(7)
770 vtrk(i) = gstifntrk(7,i),nbw
    write(21,1050) (vtrk(i),i = 1,itrk(7))
c
    if (itrack.eq.7) goto 810
    if (iter.eq.1) write(22,2250) (ntrk(8,i),i = 1,itrk(8))
    do 780 i = 1,itrk(8)
780 vtrk(i) = gstifntrk(8,i),nbw
    write(22,1050) (vtrk(i),i = 1,itrk(8))
c
    if (itrack.eq.8) goto 810
    if (iter.eq.1) write(23,2250) (ntrk(9,i),i = 1,itrk(9))
    do 790 i = 1,itrk(9)
790 vtrk(i) = gstifntrk(9,i),nbw
    write(23,1050) (vtrk(i),i = 1,itrk(9))

    if (itrack.eq.9) goto 810
    if (iter.eq.1) write(24,2250) (ntrk(10,i),i = 1,itrk(10))
    do 800 i = 1,itrk(10)
800 vtrk(i) = gstifntrk(10,i),nbw
    write(24,1050) (vtrk(i),i = 1,itrk(10))
c
810 continue
c
    if desired, generate linear solution plotting files
    if ((nprnt.eq.1).and.(iter.eq.1)) then
      do 345 iii = 1,nrm
      c
      if multi-fluid, avoid double values at the interface
      do 343 jji = 1,nnn
      if (iii.eq.ndn(jji,2)) goto 345
    343 continue
    jji = (iii-1)*ndf + 1
    kkk = (iii-1)*ndf + 4
    write(25,3060) x(iii),y(iii),gstif(jji,nbw)
    write(26,3060) x(iii),y(iii),gstif(kkk,nbw)
  345 continue
  endif
c
    if solving only Newtonian case go to post-processor
    if (itmax.eq.1) goto 400
c
    if first iteration: no check for error, re-iterate
    if (iter.eq.1) goto 310
c
    error calculation and convergence check for non-linear iterations
    err = 0.0
c
calculate accumulated error
    do 350 i = 1,neq
  350 err = err + (gf(i)-gstif(i,nbw))**2
write(7,3090) iter,err

if within tolerance, end non-linear iterations
if(err.lt.tol) goto 360

check for increasing error (divergence FATAL)
if ((err2.lt.err1).and.(err1.lt.err)) goto 980

if not within tolerance and max iterations not exceeded, re-iterate
if(iter.lt.itmax) go to 310

max iterations exceeded (FATAL)
goto 990

check time for solution of non-linear problem
360 continue
call timeck(itime(5))

if generating re-start file, output it
if ((istreq.1).or.(istreq.3)) then
write(14,1010) neq
write(14,1050) (stiff(j,nbw),j = 1,neq)
endif

------------------------------------------------------------------------
.
 postprocessor unit .
.
------------------------------------------------------------------------

400 continue

calculate the retardation terms and the elasticity parameters
call STRESS(nem,nmm,npe,ndf,nbw,t11,t12,t22,iwei,deb)

calculate We at appropriate nodes and write
write(7,3020)
do 410 i = 1,4
  we(i) = dabs((t11(i,iwei(i))-t22(iwei(i))))/2./t12(iwei(i))
do 410
write(7,3030) iwei(i),wei(i),deb(i)

write nodal solutions
write(7,3040)
write(7,3050) (x(i),y(i),stiff(5*i-4,nbw),stiff(5*i-3,nbw),
  t11(i),t12(i),t22(i),t11(i)-t22(i),i = 1,nmm)

write Ux to separate plotting file
do 430 i = 1,nmm

branch to avoid double values of Ux
do 420 j = 1,ndn
  if (i.eq.ndn(j,2)) goto 430
420 continue
write(8,3060) x(i),y(i),gstif(5*i-4,nbw)
430 continue

write Uy to separate plotting file
        do 450 i = 1,nmm

        branch to avoid double values of Uy
        do 440 j = 1,nndn
            if (i.eq.nndn(j,2)) goto 450
        440 continue
        write(9,3060) x(i),y(i),gstif(5*i-3,nbw)
450 continue

calculate and write pressure, shear rate etc.
call PRES(nem,npe,ndf,nbw,pp,xc,yc,shrr,extr,vort,p)
write(7,3070)
write(7,3080) (a,xc(n),yc(n),p(n),ahrr(n),extr(n),
        1 vort(n),n = 1,nem)

write pressure to separate file for plotting
write(10,3060) (xc(n),yc(n),p(n),n = 1,nem)

calc bifrfr and write to separate plotting files only
        do 460 i = 1,nmm
            itype = ndtype(i)
            bir = soc(itype)*sqrt(4.*t12(i)**2+(t11(i)-t22(i))**2)
            if (ndtype(i).eq.1) then
                write(11,3060) x(i),y(i),bir
            endif
            if (ndtype(i).eq.2) then
                write(12,3060) x(i),y(i),bir
            endif
460 continue

c end of post-processing

c streamlines calculation

if((nssf+naf).eq.0) goto 990

istr = 1
nndf = ndf
ndf = 1
b = 1.0
nh = nhbw/nndf

c rerun the program for the streamlines

c load non-linear solution into prev solution and initialize gstif

Appendix E. PTTFEM
do 500 i = 1, neq
   gfl(i) = gstdf(i, nbw)
do 500 j = 1, nbw
500 gstdf(i, j) = 0.0

loop over number of elements
do 510 n = 1, nem

set fluid constant set for current element
   itype = nod(n, rpt)

update non-linear terms
   call UPDATE(n, vel, b, gf, gp, elxy, nndf, npe)

evaluate element stiffness matrix
   call STIFFQ(npe, ndf, iel, itype, pp, vel, istr, iter)

assemble element stiffness matrix into global stiffness matrix
   call ASMBLE(n, nh, npe, ndf)

510 continue

incorporate streamline boundary conditions
   call bound(nsf, nsaf, ibs, vbl, vbs, nh)

constrain continuity of stream function at fluid/fluid interface
   if (nfl.eq.1) goto 520
   call DBLNOD(iter, ndn, ndm, nh, ndf)

solve matrix
520 call SOLVER(nmm, nh)

write stream function to plotting file only

if normalizing stream function to an input value (vpsi) goto 550
   if (dabs(vpsi) .gt. 1.0e-10) goto 550
write stream function as calculated
   do 540 i = 1, nmm
branch to avoid double values of the stream-function
   do 530 j = 1, ndn
   if (i.eq.nda(i, 2)) goto 540
530 continue
   write(13, 3060) x(i, y(i), gstdf(i, 2*nh)
   540 continue
   goto 9999

normalize stream function and write
550 continue
   do 570 i = 1, nmm

Appendix E. PTTFEM
psi = gstif(i,2*nh)/vpsi

branch to avoid double values of the stream-function
do 560 j = 1,ndn
  if (i.eq.ndn(j,2)) goto 570
560 continue
  write(13,3060) x(i),y(i),psi
570 continue
  goto 9999

FATAL ERROR TRAPS
900 write(7,9000) nnn
  goto 9999
910 write(7,9010) nem
  goto 9999
920 write(7,9020) npe
  goto 9999
930 write(7,9030)
  goto 9999
940 write(7,9040) nlf
  goto 9999
950 write(7,9050)
  goto 9999
960 write(7,9060) nbw
  goto 9999
970 write(7,9070)
  goto 9999
980 write(7,9080)
  goto 9999
990 write(7,9090)

write restart file after max iterations exceeded
write(14,1010) neq
write(14,1050) (gstif(i,nbw),i = 1,neq)

9999

formats

input file formats
1000 format(20a4)
1010 format(16.5)
1020 format(6e13.5)
1030 format(7e11.3)
1040 format(5e15.5)
1050 format(16e13.5)

complete output solution formats
2000 format(45x,20a4,//)
2010 format(5x,'number of nodes in the mesh .................',i4,20x,'no. of spec. primary degr. of freedom ........',i4)
2020 format(5x,'number of elements in the mesh .................',i4,20x,'no. of spec. non-zero secondary d.o.f. ........',i4)
Appendix E. PTTFEM
2 5x,'shear-rate',5x,' ext-rate ','5x,' vorticity',/,
3 19x,','------',',6(5x,','----------'))
3080 format(20x,i4,2x,6e15.4)
c
c  comment formats
8000 format('!/x, comments regarding input variables')
8010 format('5x,i3,' input tolerance unreasonable, tolerance set to',
1  ' 0.01')
8020 format('5x,i3,' input acceleration parameter unreasonable,,' 
1  ' set to 1.0')
8030 format('5x,i3,' ratio of eta1/eta2 greater than 8, set to 7.5')
c
    fatal error formats
9000 format('5x,***** FATAL ERROR! *****',5x,
1 ' number of nodes exceeds maximum of 661: nmm = ',i6)
9010 format('5x,***** FATAL ERROR! *****',5x,
1 ' number of elements exceeds maximum of 601: nem = ',i6)
9020 format('5x,***** FATAL ERROR! *****',5x,
1 ' bogus number of nodes per element (4,8,9) : npe = ',i6)
9030 format('5x,***** FATAL ERROR! *****',5x,
1 ' no boundary conditions supplied: nsdf+nsbf = 0')
9040 format('5x,***** FATAL ERROR! *****',5x,
1 ' multiple fluids indicated but no fluid/fluid interface nodes')
9050 format('5x,***** FATAL ERROR! *****',5x,
1 ' number of fluids exceeds maximum of 2: nfl = ',i6)
9060 format('5x,***** FATAL ERROR! *****',5x,
1 ' calculated bandwidth exceeds maximum of 301: nbw = ',i6)
9070 format('5x,***** FATAL ERROR! *****',5x,
1 ' constrained velocity not within half band-width')
9080 format('!/x,***** FATAL ERROR! *****',5x,
1 ' solution is diverging')
9090 format('!/x,***** FATAL ERROR! *****',5x,
1 ' maximum number of iterations exceeded')
c
c
c
9999 continue
c
c
ew

c"

subroutine ASMBLE(n,nhbw,npe,ndf)
c
c subroutine to assemble the elements in a global matrix of
c nbw x neq dimensions
c implicit real*8(a-h,o-z)
c common/msh/x(661),y(661),nod(601,10)
c common/stf/elst(f45,45),elxy(9,2),f(45)
c common/gstfm/gstf(3305,301)
c
    nbw = 2.*nhbw
do 30 i = 1,npe
    nr = (nod(i,1)-1)*ndf
do 30 ii = 1,ndf
    nr = nr + 1
    k = (i-1)*ndf + ii

Appendix E. PTTFEM 319

gstif(nr,nbw) = gstif(nr,nbw) + f(k)
do 20 j = 1,npe
   ncl = (nod(n,j)-1)*ndf
do 20 jj = 1,ndf
   m = (j-1)*ndf + jj
   nc = ncl + jj-nr + nhbw
   if (nc) 20,20,10
10 gstif(nr,nc) = gstif(nr,nc) + elstif(k,m)
20 continue
30 continue
return
e nd

subroutine bound(nsbf,nsdf,ibdf,vbsf,vbdf,nhbw)

c c subroutine to incorporate the boundary conditions

c implicit real*8(a-h,o-z)
dimension ibdf(351),vbdf(351),ibsf(199),vbsf(199)
common/gstfm/gstif(3305,301)
nbw = nhbw*2.
c
 impose specified non-zero secondary degrees of freedom
if (nsbf.eq.0) go to 20
 do 10 i = 1,nsbf
    ii = ibsf(i)
10 gstif(ii,nbw) = gstif(ii,nbw) + vbsf(i)
c
 impose specified primary degrees of freedom
20 continue
   do 40 i = 1,nsdf
      ib = ibdf(i)
vb = vbdf(i)
do 30 j = 1,nbw
30 gstif(ib,j) = 0.0
   gstif(ib,nhbw) = 1.0
40 gstif(ib,nbw) = vb
return
e nd

subroutine DBLNOD(iter,ndn,ndn,nhbw,ndf)

c c subroutine to impose variable continuity at fluid/fluid interface

implicit real*8(a-h,o-z)
dimension ndn(50,2)
common/gstfm/gstif(3305,301)
nbw = nhbw*2.
c
c set number of constrained nodal unknowns
ncnv = 2
 if (ndf.eq.1) ncnv = 1
c
c loop over constrained variables

Appendix E. PTTFEM
do 30 i = 1, ncnv
  
c  loop over number of double nodes
  do 20 j = 1, ndn
    
c  set equation number of current nodal value being constrained
    inode = ndn(j, 1)
    ii = (inode - 1) * ndf + i
    
c  set offset from nhbw where equivalent value resides
    jj = nhbw - (ndn(j, 1) - ndn(j, 2)) * ndf
    
c  set all entries in row (including force) to 0
  do 10 k = 1, nbw
    10 gstif(ii, k) = 0.0
    
c  set main diagonal to 1
    gstif(ii, nhbw) = 1.0
    
c  set constrained nodal value to -1
    20 gstif(ii, jj) = -1.0
  
c  30 continue
  
c  if not streamline calc, impose t12 continuity
  if (ndf.eq.1) goto 60
  
c  loop over number of double nodes
  do 50 j = 1, ndn
    
c  set equation number of current nodal value being constrained
    inode = ndn(j, 1)
    ii = (inode - 1) * ndf + 4
    
c  set offset from nhbw where equivalent value resides
    jj = nhbw - (ndn(j, 1) - ndn(j, 2)) * ndf
    
c  set all entries in row (including force) to 0
  do 40 k = 1, nbw
    40 gstif(ii, k) = 0.0
    
c  set main diagonal to 1
    gstif(ii, nhbw) = 1.0
    
c  set constrained nodal value to -1
    50 gstif(ii, jj) = -1.0
  
c  60 continue
  
c  return
end

******************************************************************************
subroutine PRESS(nem, npe, ndf, nbw, pp, xc, yc, shr, extr, vort, p)
  
c subroutine to calculate the pressure, the shear rate and

Appendix E. PTTFEM
the vorticity at the center of the elements

```
implicit real*8(a-h,o-z)
common/msh/x(661),y(661),nod(601,10)
common/gstifm/gstif(3305,301)
common/rheol/amul(2),rix(2),dkxi(2),epsi(2),den(2),retrat(2)
1.amun(2)
dimension sf(9),gdsf(2,9),elxy(9,2),extr(601),p(601),
& vort(601),sttr(601),xc(601),yc(601)
```

c
```
grad = 0
xij = 0.
yij = 0.
```

c
```
loop over number of elements
do 30 n = 1,nem
```

c
```
initialize coordinates and derivatives for each element
t = 0.
x(n) = 0.
y(n) = 0.
dux = 0.
duy = 0.
dvx = 0.
dvy = 0.
```

c
```
set required constants for appropriate set
itype = nod(n,npe + 1)
amu2 = amul(itype)/retrat(itype)
```

c
```
calculate the coordinates of the centroid of the element
do 10 i = 1,npe
nk = nod(n,i)
elxi(i,1) = x(nk)
elxi(i,2) = y(nk)
x(n) = x(n) + elxy(i,1)/dfloat(npe)
y(n) = y(n) + elxy(i,2)/dfloat(npe)
```

c
```
calculate the derivatives of the velocities at element center
call SHAPE (npe,xij,yij,sf,gdsf,det,elxy)
do 20 i = 1,npe
ni = nod(n,i)*ndf
dux = dux + gdsf(1,i)*gstif(ni-4,nbw)
duy = duy + gdsf(2,i)*gstif(ni-4,nbw)
dvx = dvx + gdsf(1,i)*gstif(ni-3,nbw)
dvy = dvy + gdsf(2,i)*gstif(ni-3,nbw)
```

c
```
evaluate the shear stress at the center of the element
20 t = t + sf(i)*gstif(ni-1,nbw)
```

c
```
combine non-linear and retardation shear-stress
  t = t + amu2*(duy + dvx)
```

c
```
evaluate pressure, shear-rate, extension-rate and vorticity
  grad = dux + dvy
  p(n) = -pp*grad
```
shrr(n) = dsqrt(2.0*dx**2 + 2.0*dy**2 + (dx + dy)**2)
extr(n) = 2.0*dsqrt(dx**2 + dy**2)
vort(n) = duy-dvx

30 continue

return

c***

c subroutine SHAPE(npe, xi, eta, sf, gdsf, det, elxy)

c c the subroutine evaluates the interpolation functions (sf(i)) and

c its derivatives with respect to natural coordinates (dsf(i,j)),

c and the derivatives of sf(i) with respect to global coordinates

c for four, eight, and nine node rectangular isoparametric elements.

c c sf(i)......interpolation function for node i of the element

c dsf(i,j).....derivative of sf(i) with respect to xi if i = 1 and

c with respect to eta if i = 2.

c gdsf(i,j)....derivative of sf(i) with respect to x if i = 1 and

c with respect to y if i = 2.

c xcode(i,j)....j-th (j = 1,2) coordinate of node i of the element

c np(i).......array of element nodes (used for defining sf and dsf)

c g(i,j)......jacobian matrix

c ginv(i,j)...inverse of the jacobian matrix

c det.........determinant of the jacobian matrix

c c

implicit real*8 (a-h,o-z)

dimension elxy(9,2),xcode(9,2),np(9),dsf(2,9),gj(2,2),gjinv(2,2),

1 sf(9),gdsf(2,9)

data xcode/1.0d0,2.0d0,-1.0d0,0.0d0,1.0d0,0.0d0,-1.0d0,0.0d0,
2 2*1.0d0,2*1.0d0,-1.0d0,0.0d0,1.0d0,2*0.0d0/

data np/1,2,3,4,5,6,8,9/

c

c fnc(a,b) = a*b

if (npe-8) 60,10,80

c quadratic interpolation functions (for the eight-node element)

c 10 do 40 i = 1, npe
	ni = np(i)

xp = xcode(ni,1)

yp = xcode(ni,2)

xi0 = 1.0 + xi*xp

eta0 = 1.0 + eta*yp

x1 = 1.0 - xi*xp

eta1 = 1.0 - eta*yp

if(i.gt.4) go to 20

sf(ni) = 0.25*fnc(xi0,eta0)*(xi*xp + eta*yp - 1.0)

dsf(1,ni) = 0.25*fnc(eta0,xp)*(2.0*xi*xp + eta*yp)

dsfl(ni) = 0.25*fnc(xi0,yp)*(2.0*eta*yp + xi*xp)

go to 40

20 if(i.gt.6) go to 30

sf(ni) = 0.5*fnc(xi1,eta0)

Appendix E. PTTFEM 323
dsf(1,ni) = -fnc(xi,eta0)
dsf(2,ni) = 0.5*fnc(yp,xi1)
goto 40
30 sf(ni) = 0.5*fnc(eta1,xi0)
dsf(1,ni) = 0.5*fnc(xp,eta1)
dsf(2,ni) = -fnc(eta,xi0)
40 continue
goto 130

linear interpolation functions (for four-node element)

60 do 70 i = 1, npe
xp = xcode(i,1)
yp = xcode(i,2)
xi0 = 1.0 + xi*xp
eta0 = 1.0 + eta*yp
sf(i) = 0.25*fnc(xi0,eta0)
dsf(1,1) = 0.25*fnc(xp,eta0)
dsf(2,1) = 0.25*fnc(yp,xi0)
goto 130
70 continue

quadratic interpolation functions (for the nine-node element)

80 do 120 i = 1, npe
ni = np(i)
xp = xcode(ni,1)
yp = xcode(ni,2)
xi0 = 1.0 + xi*xp
eta0 = 1.0 + eta*yp
xi1 = 1.0-xi*xi
eta1 = 1.0-eta*eta
xi2 = xp*xi
eta2 = yp*eta
if(i .gt. 4) goto 90
sf(ni) = 0.25*fnc(xi0,eta0)*xi2*eta2
dsf(1,ni) = 0.25*xp*fnc(eta2,eta0)*(1.0 + 2.0*xi2)
dsf(2,ni) = 0.25*yp*fnc(xi2,xi0)*(1.0 + 2.0*eta2)
goto 120
90 if(i .gt. 6) goto 100
sf(ni) = 0.5*fnc(xi1,eta0)*eta2
dsf(1,ni) = -xi*fnc(eta2,eta0)
dsf(2,ni) = 0.5*fnc(xi1,yp)*(1.0 + 2.0*eta2)
goto 120
100 if(i .gt. 8) goto 110
sf(ni) = 0.5*fnc(eta1,xi0)*xi2
dsf(2,ni) = -eta*fnc(xi2,xi0)
dsf(1,ni) = 0.5*fnc(eta1,xi)*eta2
goto 120
110 sf(ni) = fnc(xi1,eta1)
dsf(1,ni) = -2.0*xi*eta1
dsf(2,ni) = -2.0*eta*xi1
120 continue

calculate the determinant and the inverse of the jacobian

130 do 140 i = 1, 2

Appendix E. PTTFEM 324
do 140 j = 1,2
  gj(i,j) = 0.0
  do 140 k = 1,npe
  140 gj(i,j) = gj(i,j) + dsf(i,k)*elxy(k,j)
det = gj(1,1)*gj(2,2)-gj(1,2)*gj(2,1)
cif the determinant is not positive, there is an error in the data
cif(det.lt.1.d-12) go to 160
c  gjinv(1,1) = gj(2,2)/det
gjinv(2,2) = gj(1,1)/det
gjinv(1,2) = -gj(1,2)/det
gjinv(2,1) = -gj(2,1)/det
do 150 i = 1,2
do 150 j = 1,npe
gdsf(i,j) = 0.0
do 150 k = 1,2
150 gdsf(i,j) = gdsf(i,j) + gjinv(i,k)*dsf(k,j)
return
160 write(6,170) npe
do 165 i = 1,npe
165 write(6,180) (elxy(i,j),j = 1,2)
170 format(5x,*** error! the determinant of the jacobian is less & than or equal to zero. check input data./'.'/npe = '.i5')
180 format(2(5x,e15.6))
stop
cend
c******************************************************************************
subroutine SOLVER(n,iterm)
solver for non-symmetric system of linear banded equations.
solution is stored in gstiff(n,2*iterm).
cimplicit real*8 (a-h,o-z)
common/gstifm/gstiff(3305,301)
pare = 1.d-18
nbnd = 2*iterm
nbmu = nbnd-1
c..... begin elimination of the lower left
do 80 i = 1,n
ccheck if the diagonal term is zero
c10 if (dabs(gstiff(i,iterm)).lt.pare) go to 110
c20 i last = min0(i + iterm-1,n)
l = iterm + 1
do 40 j = 1,i last
  l = l-1
  if (dabs(gstiff(j,l)).lt.pare) go to 40
  b = gstiff(j,l)
do 30 k = 1,nbnd
30 gstiff(j,k) = gstiff(j,k)/b
if (i.eq.n) go to 90
40 continue
1 = 0
jfirst = i + 1
if (jlast.1e.i) go to 80
do 70 j = jfirst,jlast
1 = 1 + 1
if (dabs(gstiff(j,iterm-1)).lt.pare) go to 70
do 50 k = iter.n,nbm
50 gstiff(j,k-l) = gstiff(j-1,k-l) -
gstiff(j,nbnd) = gstiff(j-1,nbnd) -
gstiff(j,nbnd)
if (i.ge.n-iterm + 1) go to 70
do 60 k = 1,1
60 gstiff(j,nbnd-k) = -gstiff(j,nbnd-k)
70 continue
80 continue

90 l = iterm-1
do 100 i = 2,n
do 100 j = 1,l
if (n + 1-l+i.j.gt.n) go to 100
1 = 1 + 1 -
gstiff(n + 1-l,i,nbnd) = -
gstiff(n + 1-l,i,nbnd) -
gstiff(n + 1-l,i,iterm + j)
100 continue
return

if any term on the diagonal is zero print error message and stop

110 write (6,1000) i
111 write (6,1010) gstiff(i,iterm)
stop

1000 format (1x,*** FATAL ERROR! ***,/.5x,
1 zero on main diagonal for equation`,i5)
1010 format (1x,`gstiff(i,nbhw) = `.10.2
end

******************************************************************************
subroutine STIFFQ(npe,nrf,iel,itype,pp,vel,istr,iter)
******************************************************************************
stiffness matrix for isoparametric quadrilateral elements
******************************************************************************
in implicit real*8(a-h,o-z)
common/stf/elstif(45,45),elxy(9,2),fl(45)
common/rheol/amul(2),ril(2),dksil(2),epsil(2),den(2),retrat(2)
! _amul(2)
dimension sf(9),gsdf(2,9),gauss(4,4),wt(4,4),s(9,9),so(9,9)
1 ,s02(9,9),sv11(9,9),sv12(9,9),sv21(9,9),sv22(9,9),
2 s01v(9,9),s02v(9,9),vel(45),st11(9,9),st12(9,9)
3 ,st22(9,9),s11(9,9),s22(9,9),s12(9,9),a1(9)
data gauss=4*0.0d0,-.57735027d0,.57735027d0,.57735027d0,2*0.0d0,-.77459667d0,
2 0.0d0,.77459667d0,0.0d0,-.86113631d0,
3 -.33998104d0,.33998104d0,.86113631d0,
data wt/2.0d0,3*0.0d0,2*1.0d0,2*0.0d0,.55555555d0,.88888888d0,
c
nn = npe*ndf
ngp = iel + 1
ngps = ngp**2
if (ist.eq.1) go to 10

c
set calculation constants to appropriate fluid constants
amu = amul(iotype)
amu2 = amul(iotype)/retat(iotype)
ambda = rlx(iotype)
cksi = dksi(iotype)
ceps = epsil(iotype)
rho = den(iotype)

c
set constants to Newtonian values for first iteration
if (iter.eq.1) then
amu = amun(iotype)
amu2 = amun(iotype)/retat(iotype)
endif

c
initialize the arrays
10 continue
do 20 i = 1, npe
fi(i) = 0.
do 20 j = 1, npe
s(i,j)=0.0
so1(i,j) = 0.0
so2(i,j) = 0.0
s11(i,j) = 0.
s22(i,j) = 0.
s01v1(i,j) = 0.0
s02v2(i,j) = 0.0
st11(i,j) = 0.
st12(i,j) = 0.
st22(i,j) = 0.
sv11(i,j) = 0.0
sv12(i,j) = 0.0
sv21(i,j) = 0.0
20 sv22(i,j) = 0.0

c
do 30 i = 1,nn
fi(i) = 0.0
do 30 j = 1,nn
30 elstiff(i,j) = 0.0

c
dudx = 0.0
dudy = 0.0
dvdx = 0.0
dvdy = 0.0
gamadt = 0.0

c
do-loops on numerical (gauss) quadrature begin here

c
do 70 nni= 1,ngp
do 70 nnj = l,ngp

initialize non-linear terms
t11 = 0.
t12 = 0.
t22 = 0.
v1 = 0.0
v2 = 0.0
ux = 0.0
uy = 0.0
vx = 0.0
vy = 0.0

evaluate the shape functions at each gauss point
xi = gauss(nni,ngp)
eta = gauss(nnj,ngp)
call SHAPE(npe,xi,eta,sf,gdsf,det,elxy)

evaluate the non-linear terms
do 40 i = 1,npe
k = 5*i - 4
v1 = vel(k)*sf(i) + v1
v2 = vel(k+1)*sf(i) + v2
ux = ux + vel(k)*gdsf(1,i)
uy = uy + vel(k)*gdsf(2,i)
vx = vx + vel(k+1)*gdsf(1,i)
vvy = vy + vel(k+1)*gdsf(2,i)
if (istr.eq.1) goto 40
t11 = t11 + vel(k+2)*sf(i)
t12 = t12 + vel(k+3)*sf(i)
t22 = t22 + vel(k+4)*sf(i)
40 continue

dudx = ux/ngps + dudx
dudy = uy/ngps + dudy
dvdx = vx/ngps + dvdx
dvdy = vy/ngps + dvdy
const = det*wt(nni,ngp)*wt(nnj,ngp)
trt = t11 + t22
cptt = dexp(ceps*ambda/amu*trt)
calclulation of the galerkin integrals
do 60 i = 1,npe
do 50 j = 1,npe
s(i,j) = cptt*const*sf(i)*sf(j) + s(i,j)
s01(i,j) = const*sf(i)*gdsf(1,i) + s01(i,j)
s02(i,j) = const*sf(i)*gdsf(2,i) + s02(i,j)
s11(i,j) = const*gdsf(1,i)*gdsf(1,i) + s11(i,j)
s22(i,j) = const*gdsf(2,i)*gdsf(2,i) + s22(i,j)
if (istr.eq.1) go to 50
s01v1(i,j) = const*v1*sf(i)*gdsf(1,i) + s01v1(i,j)
s02v2(i,j) = const*v2*sf(i)*gdsf(2,i) + s02v2(i,j)
sv11(i,j) = const*ux*sf(i)*sf(j) + sv11(i,j)
sv12(i,j) = const*uy*sf(i)*sf(j) + sv12(i,j)
sv21(i,j) = const*vx*sf(i)*sf(j) + sv21(i,j)
sv22(i,j) = const*vy*sf(i)*sf(j) + sv22(i,j)
\begin{verbatim}
st11(i,j) = const*sf(i)*sf(j)*t11 + st11(i,j)
st12(i,j) = const*sf(i)*sf(j)*t12 + st12(i,j)
st22(i,j) = const*sf(i)*sf(j)*t22 + st22(i,j)
50 continue
60 fi(i) = const*sf(i)*(uy-vx) + fi(i)
70 continue

if (ist.eq.1) go to 130

evaluate element stiffness matrix
111 ii = 1
   do 90 i = 1,npe
      jj = 1
      do 80 j = 1,npe
         elast(i,j) = -rho*s01v1(i,j) - rho*s02v2(i,j) + amu2*s22(i,j)
         elast(i,j+2) = s01(j,i)
         elast(i,j+3) = s02(j,i)
         elast(i+j+1) = -rho*s01v1(i,j)-rho*s02v2(i,j)+amu2*s11(i,j)
         elast(i+j+3) = s01(j,i)
         elast(i+j+4) = s02(j,i)
         elast(i+j+2) = -2.0*amu*s01(i,j)
         elast(i+j+2) = s(i,j) + amu*a*s01v1(i,j)+amu*a*s02v2(i,j)
         l = 2.0*s11(i,j)*amu + 2.0*amu*amu*s11(i,j)
         elast(i+j+3) = -2.0*amu*a*s0v12(i,j)
         l = amu*a*amu*(s12(i,j)+s21(i,j))

         elast(i+j+3) = -amu*s02(i,j)

         elast(i+j+1) = -amu*s01(i,j)

         elast(i+j+2) = -amu*s02v12(i,j)

         elast(i+j+3) = s(i,j) + amu*a*s01v1(i,j)+amu*a*s02v2(i,j)
         l = -amu*a*(s11(i,j) - s22(i,j))

         elast(i+j+4) = -amu*a*s0v12(i,j)

         elast(i+j+4) = -2.0*amu*a*s02(i,j)

         elast(i+j+3) = -2.0*amu*a*s0v21(i,j)

         elast(i+j+4) = s(i,j) + amu*a*s01v1(i,j)+amu*a*s02v2(i,j)

         elast(i+j+4) = 2.0*amu*a*s0v22(i,j)

Appendix E. PTTFEM
\end{verbatim}
80 jj = ndf*i + 1
90 ii = ndf*j + 1

evaluate penalty parameter terms with reduced order of integration

DO 120 nni = 1, iel
   DO 120 nnj = 1, iel
      xi = gauss(nni, iel)
      eta = gauss(nnj, iel)
      CALL SHAPE(npe, xi, eta, sf, gdsf, det, elxy)
      const = det*wt(nni, iel)*wt(nnj, iel)
   III = 1
   DO 110 i = 1, npe
      jj = 1
      DO 100 j = 1, npe
         elstif(ii, jj) = pp*gdsf(1, i)*gdsf(1, j)*const + elstif(ii, jj)
         elstif(i + 1, jj) = pp*gdsf(2, i)*gdsf(2, j)*const + elstif(i + 1, jj)
         elstif(ii, j + 1) = pp*gdsf(1, i)*gdsf(1, j)*const + elstif(ii, j + 1)
         elstif(ii + 1, j + 1) = pp*gdsf(2, i)*gdsf(2, j)*const + elstif(ii + 1, j + 1)
      END DO
   END DO
110 CONTINUE
100 jj = ndf*j + 1
110 ii = ndf*i + 1
120 CONTINUE

RETURN

stream function calculation

130 CONTINUE
   DO 150 i = 1, npe
      DO 140 j = 1, npe
         st(l, i, j) = st1(l, i, j) + s2(l, i, j)
      END DO
   END DO
150 f(i) = -d(f(i)
RETURN

END

SUBROUTINE STRESS(nem, nnm, npe, ndf, nbw, t11, t12, t22, iwei, deb)

subroutine that calculates the stresses with the addition
of the retardation parameter. the stresses are computed at
the center of each element and for every global node they
are averaged over the elements they belong

IMPLICIT REAL*8 (A-H,O-Z)
DIMENSION t1(661,9), t2(661,9), t3(661,9), t11(661), t12(661),
    l22(661), sf(9), gdsf(2,9), elxy(9,2), xi(9), yi(9), deb(4),
    inode(661), iwen(4), g4(4)
COMMON/msh/x(661), y(661), nod(601,10)
COMMON/gstm/gstf(3305,301)
COMMON/rheo/amul(2), rix(2), dksi(2), epb(2), den(2), retrat(2)
   i_amun(2)
   DATA xi(-1.2*1.-1., 0., 1., 0., -1., 0., yi(-1., 1., 2*1.,-1.,
   0., 1., 2*0./

initialize shear-rates for calculation of De
   DO 10 i = 1, 4
      10 g(i) = 0.
   END

loop over number of elements

Appendix E. PTTFEM
do 60 n = 1,nem

set element x,y values
do 20 i = 1,npe
   nk = nod(n,i)
elxy(i,1) = x(nk)
elxy(i,2) = y(nk)
20

set fluid constants to appropriate set
itype = nod(n,npe+1)
dla = rlx(itype)
amu2 = amul(itype)/retrat(itype)

evaluate components of shear-rate at each node
do 50 i = 1,npe
   sr1 = 0.
sr2 = 0.
sr3 = 0.
50

sum nodal contributions at current node
do 30 j = 1,npe

set appropriate node numbers and evaluate shape functions
nnj = nod(n,j)*ndf
call SHAPE(npe,xi(i),yi(i),sf,gdsf,det,elxy)

calculate nodal contributions of shear-rate components
sr1 = sr1 + 2.*gdsf(1,j)*gstitf(nnj-4,nbw)
sr2 = sr2 + gdsf(2,j)*gstitf(nnj-4,nbw) +
    gdsf(1,j)*gstitf(nnj-3,nbw)
sr3 = sr3 + 2.*gdsf(2,j)*gstitf(nnj-3,nbw)

calculate retardation contributions
   t1(n,i) = amu2*sr1
   t2(n,i) = amu2*sr2
   t3(n,i) = amu2*sr3

check node for calculating De/We number
do 40 j = 1,4
   if (nod(n,i).eq.iwei(j)) then
      g(j) = g(j) + dla*dsqrt(sr1**2/2 + sr2**2 + sr3**2/2).
   endif
40 continue

50 continue
60 continue

find the contribution of each element to each global node
do 70 i = 1,nmm
   inode(i) = 0
   t11(i) = 0.
t12(i) = 0.
t22(i) = 0.
70
do 100 i = 1,nne
  do 90 j = 1,nem
    do 80 k = 1,npe
      if(nod(j,k),ne.i) goto 80
      inode(i) = inode(i) + 1
      t11(i) = t1(j,k) + t11(i)
      t12(i) = t2(j,k) + t12(i)
      t22(i) = t3(j,k) + t22(i)
    80 continue
  90 continue
  c average the retardation terms at the nodes over the elements
  c they belong and add the viscoelastic part
  if (inode(i),lt,1) go to 900
  ni = i*ndf
  t11(i) = t11(i)/dfloat(inode(i)) + gstiff(ni-2,nbw)
  t12(i) = t12(i)/dfloat(inode(i)) + gstiff(ni-1,nbw)
  t22(i) = t22(i)/dfloat(inode(i)) + gstiff(ni,nbw)
  100 continue
  c calculate the deborah number at node iwei(i)
  do 110 i = 1,4
    110 deb(i) = g(i)/dfloat(inode(iwei(i)))
  c return
  900 write(6,1000)
    stop
  c 1000 format(10x,'error!! in subroutine stress. check procedure for'
    '& inode')
  end
  c******************************************************************************
  subroutine UPDATE(n,vel,b,gf,elxy,ndf,npe)
  c subroutine that updates the solution vector for each element
  c implicit real*8(a-h,o-z)
  dimension gf(3305),gp(3305),vel(45),elxy(9,2)
  common/shb/x(661),y(661),nod(601,10)
  c do 20 i = 1,npe
  ni = nod(n,i)
  k = ndf*(i-1) + 1
  kk = ndf*(ni-1) + 1
  do 10 j = 0,ndf-1
    10 vel(k+j) = b*gf(kk+j) + (1.-b)*gp(kk+j)
  c elxy(1,1) = x(ni)
  c elxy(1,2) = y(ni)
  20 continue
  return
  end
Vita

The author was born in Kingston, New York on December 14, 1958. He moved to Pittsfield, Massachusetts in 1963, where he was unmercifully subjected to liberal propaganda throughout his formative years. Against the headmaster's better judgement and to his parent's relief, he was allowed to graduate from the Hall School in 1976. After mucking about for a few years, he attended college and graduated from Northeastern University in Boston in 1985. During his tenure at this fine academic institution, he worked as a cooperative education student at the General Electric Company in the plastics division. On an ill-fated day in November of 1983, he attended an in-house rheology course, where he met Professor D.G. Baird. The rest, as they say, is history.