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Viscoelastic Flow Effects in Multilayer Polymer Coextrusion

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Summary

Multilayer coextrusion is a process in which polymers are extruded and joined together in a feedblock or die to form a single structure with multiple layers. These layers normally should have uniform thicknesses throughout the structure for optimal performance. However, layer thickness non-uniformities have been observed in many commercial coextruded structures.

Previous work has shown that layer thickness variations can occur for many reasons. Several of the primary causes of layer thickness variations are interlayer instabilities, viscous encapsulation, and elastic layer rearrangement. Interlayer instabilities are similar to waves on the surface of the ocean, i.e. they are thickness changes at the interface between two fluids. Viscous encapsulation is a phenomenon in which a less viscous polymer will tend to encapsulate a more viscous polymer as they flow through a channel. Elastic layer rearrangement occurs when elastic polymers flow through non-radially symmetric geometries producing secondary flows which drive rearrangement of the layer thicknesses.

In this thesis experiments and numerical simulations were performed to determine the effect of viscoelasticity on the flow of multilayer coextruded polymer structures. Initial experiments were based on coextruding two layered polystyrene structures through a circular channel to determine viscous effects in coextrusion. The polystyrene resins had different viscosities in each layer and viscous encapsulation was observed. Encapsulation velocities were measured for layer viscosity ratios of up to 2.5:1. These measurements showed that the encapsulation velocity was at its maximum near the entry of the channel and decreased as a function of the distance down the channel. The maximum encapsulation velocity measured was approximately 15% of the average downstream velocity.

Coextrusion experiments were also conducted using identical materials in each layer of a multilayered structure. These experiments were used to determine elastic effects since there was no difference in viscosity between the layers. These experiments were conducted in channels with circular, square, rectangular, and teardrop shapes. Three resins (polyethylene, polystyrene, and polycarbonate) were chosen for this study since they had significantly different elastic responses based on Storage Modulus data. Polystyrene was the most elastic resin, polyethylene was intermediate in elasticity, and polycarbonate was the least elastic resin.

Two-layer experiments were conducted using all three resins in all four geometries. Elastic layer rearrangement was observed for the polystyrene and polyethylene resins but not for the polycarbonate resin which is the least elastic resin. Elastic layer rearrangement appears to be driven by secondary flows perpendicular the main flow direction that are driven by second normal stress differences. The flow patterns produced by elastic layer rearrangement and viscous encapsulation are distinctly different in shape and magnitude.

Elastic layer rearrangement was not observed for any of the resins when processed through a channel with a circular cross section. This is because there is no imbalance in second
normal stress difference in a radially symmetric channel and thus no driving force to produce secondary flows. The polystyrene and polyethylene resins both showed substantial layer rearrangement when processed through the square, rectangular and teardrop shaped channels. All of these geometries produced imbalanced second normal stress differences that generated secondary flows. The square channel produced the largest interface deformations while the rectangular channel produced the smallest deformations.

Experiments were also conducted using feedblocks that produced different coextruded structures to better understand the elastic layer rearrangement phenomenon. Feedblocks that produced 27, 165, and 5000 layers were evaluated and gave similar flow patterns to the two layered structures but also provided more details on the flow field. A feedblock that produced a 7 by 7 array of strands in a matrix was also developed and used to trace flow patterns through the various channel geometries. Feedblocks were also designed and built that produced 2 and 13 concentric rings of layers. All of these feedblocks were used to develop details on the secondary flow patterns that caused elastic layer rearrangement.

Measurements were made on the elastic layer rearrangement velocity just as they had been done for viscous encapsulation velocity. They showed that the elastic layer rearrangement velocities were constant down the channel length and did not decrease like the viscous encapsulation velocities. The magnitude of these velocities was approximately one third of one percent compared to the fifteen percent for the maximum viscous encapsulation velocity. These data show that the viscous encapsulation phenomenon will be the dominant mechanism to drive layer interface movement in a short channel (or narrow die) while the elastic layer rearrangement phenomenon will become more important as the channel length increases (or the die gets wider).

Numerical simulation of elastic layer rearrangement was done using finite element analysis. Several viscoelastic models were evaluated and the results were compared to the experimental results. The Giesekus and Phan Thien Tanner models produced results that were in good agreement with the experimental results while the White-Metzner model did not. This was to be expected since the Giesekus and Phan Thien Tanner models predict non-zero second normal stress differences while the White-Metzner model predicts a zero second normal stress difference. This shows that a non-zero second normal stress difference will produce elastic layer rearrangement in non-radially symmetric channels.

Numerical simulations were done on both two and a half and three dimensional models. The two and a half dimensional model solves the conservation equation in two planar dimensions and assumes the calculated fields remain constant in the main flow direction with the exception of the pressure. Both models gave similar results but the three dimensional model allowed enhanced capabilities in viewing the 3D change in the interface shape at the expense of increased computational requirements.

Numerical studies were also done using the Giesekus viscoelastic flow model with multiple relaxation times for the polyethylene resin. Studies were done using one, four and five relaxation times to describe the rheology of the system. The results of these studies showed that increasing the number of relaxation times increased the accuracy of the solution when comparing it to the experimental results.
In conclusion, this work has shown that both the viscous and elastic effects must be considered when processing multilayer polymer structures.

This thesis is based on a number of previous publications. The primary publications that relate to this work are listed below:

Chapter 1  Overview of different multilayer extrusion processes

1.1  Introduction

Multilayer coextrusion has developed into an important polymer fabrication process, providing large growth opportunities for the polymer industry. Coextruded multilayer polymers are challenging such traditional materials as metals, glass, paper, and textiles.

The attraction of coextrusion is both economic and technical. It is a single-step process starting with two or more polymer materials that are simultaneously extruded and shaped in a single die to form a multilayer sheet or film. Thus, coextrusion avoids the costs and complexities of conventional multistep lamination and coating processes, where individual plies must be made separately, primed, coated, and laminated. Coextrusion readily allows manufacture of products with layers thinner than can be made and handled as an individual ply. Consequently only the necessary thickness of a high performance polymer is used to meet a particular specification of the product. In fact, coextrusion has been used commercially to manufacture unique films consisting of hundreds of layers with individual layer thicknesses less than 100 nm (1). It is difficult to imagine another practical method of manufacturing these microlayer structures.

Layers may be used to place colors, bury recycle, screen ultraviolet radiation, provide barrier properties, minimize die-face buildup, and to control film-surface properties, for example. Additives, such as antiblock, antislip, and antistatic agents, can be placed in specific layer positions. High melt strength layers can carry low melt strength materials during fabrication.

The largest market for coextruded films and sheets is in packaging applications, e.g. two- or three-layer films for trash bags or five-to-nine-layer structures for flexible and semirigid packages. As many as five different polymers may be used to obtain heat sealability, barrier, chemical resistance, toughness, formability, and aesthetics. Coextrusion is also suitable for applying thin multilayer films as coatings on substrates (2). Growing applications for coextrusion are in automotive, construction, appliance, and food packaging markets.

Coextruded films are produced by a tubular-blown film process and a flat-die, chill-roll casting process. Capital and operating costs for blown-film vs. cast-film coextrusion lines are strongly dependent on product mix and utilization. Equipment suppliers provide comparative economic evaluations for specific products. Practical cast-film equipment has been discussed previously (3). Coextrusion dies are unique. Extruders used before the die and take-away equipment used afterwards are standard equipment for single-layer film manufacture of blown or cast film.

The choice of whether to use the blown or cast film process is normally dependent on the rate and final properties of the structure that are desired. Cast film lines can typically run at a higher rate than a blown film line because the cooling efficiency of a chill roll is higher than
using air to cool a bubble. However, the cast film process produces a product with uniaxial orientation rather than the biaxial orientation produced with the blown film process. In many cases, biaxial orientation is preferred to produce a film with more balanced physical properties.

Coextrusion die sizes vary and are dependent on the application. Blown film dies for the agricultural market can be as large as 2 meters in diameter that would produce a film approximately 8 meters wide. Cast film dies for agricultural films have been made with widths as large as 10 meters.

In addition to uses in bags and packaging, coextruded structures are also used in many other areas. Many coextruded sheets are made for use in thermoforming operations to form specific package or container shapes. Coextrusion is also used in the profile market. Pipes as well as window profiles have been made from coextruded structures.

1.2 Blown film, single versus multi manifolds

Tubular coextrusion dies were the earliest dies used to make multilayer polymer film. Successful design requires formation of uniform concentric layers in the annular die land formed by the mandrel and adjustable or nonadjustable outer die ring. Early designs included center-fed dies that had the mandrel supported by a spider (4). Feedports arranged a concentric melt stream that was pierced by the mandrel as it flowed to the die exit, forming annular layers. Limitations of this early design were discontinuity and nonuniformity caused by spider-induced weld lines in the layers.

Another early design used stacks of toroidal distribution manifolds, so that as flow proceeded to the die exit, concentric layers were extruded on one another sequentially (5). The number of layers could he varied by changing the number of toroidal manifolds in the stack. The crosshead design of this die eliminated the spider support of the mandrel with its attendant weld-line problem.

The design most commonly used today is the multimanifold spiral mandrel tubular-blown film die (Fig. 1-1). This die consists of several concentric manifolds, one within the other. The manifolds are supported and secured through the base of the die. Each manifold consists of a flow channel that spirals around the mandrel allowing polymer to flow down the channel or leak across a land area to the next channel. This flow pattern smoothes out the flow of the polymer and minimizes any weld lines in the final film. Whereas early designs were limited to two or three layers, dies containing seven or more layers are now offered commercially.
These dies must achieve uniform concentric flow of all layers because it is impractical to provide circumferential thickness adjustment for each layer. Most polymers are non-Newtonian, and polymer viscosity usually decreases with shear rate. Thus rheological data obtained at the intended extrusion temperature and shear rate are needed to size manifolds and channels for layer uniformity and minimum pressure drop. Frequently spiral mandrel manifolds, common in single-layer dies, are used to improve circumferential distribution. A well-designed spiral mandrel manifold can be helpful, but streamlining is necessary to minimize stagnation, residence time and purging. A manifold design is only optimum for a particular polymer. Employing a polymer with significantly different properties may require a different manifold insert in the die to obtain satisfactory layer distribution.

Most tubular-blown film lines are designed for oscillation of the die or winder to randomize film thickness variations at the windup and avoid buildup of gauge bands, which can cause problems with film flatness. More layers complicate bearing and sealing systems in an oscillating die, but designs have now been refined to employ new sealing materials that minimize polymer leakage. New designs incorporate temperature control of individual annular manifolds to permit coextrusion of thermally sensitive polymers.

Another style of tubular-blown film die is the stackable plate die (Figure 1-2). In this style of die, each layer is spread uniformly and formed into a tube in a single plate. Plates are then stacked on top of each other and the layers are added sequentially. This style of die is becoming popular for specific applications since the number of layers can be adjusted by simply
changing the number of plates in the die. The major disadvantage for this style of die is that there is a large separating force between the plates and so many die bolts are required to hold the plates together. This means that the plates must be rather large in diameter in order to maintain structural integrity and this can produce longer flow paths and temperature differentials that can be detrimental to thermally sensitive polymers. Depending on the types of polymers to be used and the number of layers, the prices of the spiral mandrel and stacked plate dies are comparable.

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Figure 1-2. A stackable blown film die

Tubular coextrusion dies are expensive, and care must be taken when disassembling and reassembling them to clean or change parts. Discussions of additional practical design, maintenance, and operating considerations have appeared (6-10).

1.3 Sheet extrusion, single versus multi manifolds

Flat dies, also called slit dies because the orifice is a wide rectangular opening, are used in chill-roll, cast film coextrusion. These dies are used almost exclusively for multilayer coextrusion with sheet thickness> 250 µm, as well as in coextrusion coating processes (2), where a multilayer web is extrusion-coated onto a substrate such as paperboard, aluminum foil, polymer foam, or textiles.

Another commercial application for flat-die coextrusion is biaxially oriented multilayer films (11) made with the tentering process to improve mechanical properties. Tentered film is biaxially oriented by stretching in the longitudinal and transverse direction, either sequentially or simultaneously, at uniform optimum temperature. In sequential stretching, the multilayer extrudate is cooled to a suitable orientation temperature on a first set of rolls then stretched in the machine direction between a second set of rolls which is driven faster than the first set. The uniaxially stretched film then enters a tentering frame, which has traveling clips that clamp the edge of the film. The clips are mounted on two tracks that diverge inside a temperature-
controlled oven increasing film width to provide transverse stretch. The film is then heat set and cooled. Simultaneous tentering frames are also used which feature accelerating clips that stretch the film longitudinally as they diverge transversely.

Two basic die types used in flat-die coextrusion systems are multimanifold dies and the feedblock/single-manifold die. A hybrid combines feedblocks with a multimanifold die.

1.4 Multimanifold dies

For each layer, these dies have individual manifolds that extend the full width of the die. Each manifold is designed to distribute its polymer layer uniformly before combining with other layers outside the die (external combining) or inside the die before the final die land (internal combining).

External-combining dies are typically limited to two-layer coextrusion because two slit orifices must be individually adjusted with die-lip adjusting bolts. The webs are combined at the roll nip.

The vast majority of multimanifold dies are internal combining rather than external combining. This is due to the fact that better adhesion between the layers is normally found with internally combined structures because they are in thermal contact for a longer period of time and can form products with better interfacial adhesion.

In principle, internal-combining dies are similar to multimanifold-tubular-coextrusion dies except that the manifolds are flat (Fig. 1-3). With these dies, it is possible to regulate flow across the width by profiling an adjustable restrictor bar in each manifold to help obtain uniform distribution. However, wide dies require numerous adjusting bolts on each layer manifold along with die-lip adjustment to control final thickness; this can make them difficult to operate. Multimanifold dies have been sold capable of coextruding five and six layers; they are expensive and require skilled operators. The principal advantage of multimanifold dies is the ability to coextrude polymers with very different viscosities since each layer is spread independently prior to combining.

Figure 1-3. Cross-sectional view of three-layer internal-combining multimanifold flat film or sheet die
A significant disadvantage of wide multimaniold dies is the difficulty in coextruding very thin layers, such as thin cap (surface) layers, or thin adhesive (tie) layers used to bond two dissimilar polymers. Frequently these thin layers represent only 1 or 2% of the total structure thickness and therefore are extruded at a relatively low rate. With wide dies it is difficult to obtain uniformity when extrusion rate per width is very low. Also, it is difficult to coextrude thermally sensitive polymers such as poly(vinyl chloride (PVC) and poly(vinylidene chloride) copolymers (PVDC) in wide dies because slow-moving material near the walls greatly increases residence time and thermal exposure.

1.5 Feedblock/single manifold dies

The feedblock method of flat-die coextrusion, originally developed and patented by The Dow Chemical Company, uses a feedblock before a conventional single manifold die as is shown in Figure 1-4 (12-13).

A layered melt stream, which is prearranged ahead of the die inlet by the feedblock, is extended the width of the die as it is reduced in thickness (Fig. 1-5). Polymer melts from each extruder can be subdivided into as many layers as desired in the final product. Feedports arrange metered layers in required sequence and thickness proportions. A commercial feedblock/single manifold die system is shown in Figure 1-6. Modular feedblock design similar to that illustrated can be used to change the number, sequence, or thickness distribution of layers by changing a flow-programming module in the feedblock. Programming modules consist of machined flow channels designed to subdivide and direct the flow of each material to specific locations and proportions required by the product. This technique can also be used to minimize edge waste through tuning of the feedblock to produce the desired structure across the desired width of the film or sheet.
Figure 1-5. The principle of the feedblock for coextruding multilayer film or sheet. Number of layers is equal to number of feedports.

The shape of the multilayer melt stream entering the die inlet can be round, square, or rectangular, as long as the feedblock is properly designed to deliver the layers to the die with constant composition (14). Some feedblock suppliers prefer round die entry design for ease of machining or retrofitting to old dies. Others prefer square or rectangular die entries for ease of design and minimization of shape change as the layer interfaces are extended to the rectangular die orifice. A thermally sensitive polymer can be encapsulated by stable polymers so it does not contact the die walls, thus reducing residence time.

The fact that the multilayer stream at the die inlet is narrow (~2.5-10 cm) compared to die width makes it relatively easy to meter thin surface or adhesive layers.

The versatility of the feedblock has made it the most popular flat-die coextrusion method. Large numbers of layers may be coextruded, layer structure may be readily altered with interchangeable modules, and thermally sensitive polymers may be protected by encapsulation. It is estimated that over 95% of flat-die coextrusion systems use a feedblock.

Figure 1-6. Exploded view of modular feedblock and single-manifold die for three polymers forming a five-layer coextrusion.
One limitation of feedblocks is that polymer viscosities must be matched fairly closely because the combined melt stream must spread uniformly within the die. Severe viscosity mismatch results in nonuniform layers; the lower viscosity material tends to flow to the die edges. A crude rule of thumb is that polymer viscosities must be matched within a factor of three or four, which is a reasonably broad range for many commercially important coextrusions. Layer uniformity may be adjusted by varying melt temperature within limits dictated by heat transfer. Increasing temperature decreases viscosity, and material moves from the center to the edges; decreasing temperature has the opposite effect. Typically, the individual polymer melt temperatures differ by as much as 30-60°C. Beyond that, heat transfer tends to nullify further adjustment by temperature variation.

Often polymers are intentionally selected with a mismatch in viscosities to avoid flow instabilities. Viscosity mismatch of a factor of ten or more may be necessary. Layer nonuniformity expected with the mismatch is compensated by using shaped feedport geometry; i.e., the layers are introduced into the die non-uniformly, so that uneven flow within the die results in a satisfactorily uniform final distribution. Considerable art has been developed to extend the range of viscosity mismatch that can be accommodated in a feedblock system by using compensating feedport geometry (15, 16).

Some feedblocks are reportedly capable of coextruding polymers with viscosity mismatch of 100 or more (17, 18, 19). This style of feedblock has movable vanes that partition individual layers prior to combining (Fig. 1-7). The vanes may be freely floating, automatically seeking their equilibrium position on the basis of flow rates and viscosities. This self-adjusting feature can accommodate wide ranges in relative flow rates and viscosities while maintaining layer uniformity in the final product. The vanes may be rotated manually and locked into a non-equilibrium position to adjust uniformity further.

Often distribution pins are used, or the vanes are profiled to compensate for nonuniform layers in this style of die. Distribution pins are cylinders that are placed across the flow stream that can be rotated during processing. These distribution pins are normally cut so that one side is shaped into a specific geometry (such as a deeper slot). By rotating these pins to expose a specific geometry during processing, more or less flow will occur in a specific channel thus allowing flexibility in determining the layer thickness in the individual layers.

Figure 1-7. Feedblock with movable vane partitions that adjust positions for different polymer viscosities and/or layer-flow rates
1.6 Combined feedblock/multimanifold dies

Combinations of feedblocks and a multimanifold die are also used commercially. The multimanifold die can incorporate the same design principles as the feedblock: i.e. vanes separating individual manifolds within the die. In a sense, the multimanifold die is a wide feedblock. A feedblock may be attached to one or more manifold inlets as shown in Figure 1-8. With this system polymers with widely different polymer viscosities and processing temperatures may be coextruded. A very viscous or high temperature polymer may be extruded through one or more die manifolds, while a thermally sensitive or much lower viscosity polymer is coextruded with adhesive layers through a feedblock feeding another manifold. Combining of all layers occurs prior to the final die land.

![Figure 1-8. Combination feedblock and multimanifold die system. Feedblock feeds center-die manifold](image)

1.7 Layer multiplication

1.7.1 Layer multiplication using a feedblock

The feedblocks and dies discussed in the previous section are suitable for producing multilayer structures with typically ten layers or less, which is adequate for many industrial applications. However, there have been multilayer structures developed in recent years that require hundreds or thousands of layers to produce unique properties (20-23). Among these properties are enhancements in mechanical and optical properties. For instance, films and sheet with tailored optical properties that reflect or transmit specific portions of the electromagnetic spectrum have been developed.

In order to produce film or sheet structures with hundreds or thousands of layers, new techniques had to be developed to produce those structures. Standard feedblock techniques would not allow this many layers because of mechanical difficulties in joining so many layers. One method to produce hundreds of layers was developed by Schrenk (24). This technique is shown schematically in Figures 1-9 and 1-10.
Figure 1-9. Feedblock for producing many layers – front view

Figure 1-10. Feedblock for producing many layers – side view
The technique shown in Figures 1-9 and 1-10 involves feeding two melt streams into opposite sides of a feedblock that distributes the melt streams radially to the edges of the feedblock. At this point, each of the two melt streams is subdivided into multiple substreams. These substreams are then interdigitated near the outer periphery of the feedblock in a manner similar to that shown in Figure 1-11. These interdigitated streams are then fed to a central cavity where they exit the feedblock as a multilayered structure.

![Figure 1-11. Feedblock layer distributor plate schematic](image1)

The feedblock described above is one solution to producing multilayer structures with many layers. However, this type of feedblock also offers other advantages. This type of feedblock was designed to produce structures with uniform layers but its geometry lends itself to easy manipulation of the layer thicknesses. For instance, Figure 1-12 shows a feedblock modification in which the feedblock has been subdivided into three sections labeled I, II, and III. In each of these sections, the joining slot geometry can be modified to vary the layer thickness gradient discretely in each section (25). This would be useful if by modifying the layer thicknesses in each section the mechanical or optical properties could be modified substantially.

![Figure 1-12. Feedblock for producing layer thickness gradients](image2)
Figure 1-13 shows a different modification of this feedblock. In this modification, the layer thickness gradient is continuous from one side to the other rather than changing discretely as it was in Figure 1-12. This is done by changing the land length, L, for each of the feed slots and thus changing the flow rate through each slot. This feedblock would produce a multilayer structure in which the layer thickness would progressively change from the top to the bottom of the structure.

![Figure 1-13. Schematic diagram of layer thickness gradient feedblock channels](image)

**1.7.2 Layer multiplication using Interfacial Surface Generators**

Another method that is discussed in the patent literature to increase the number of layers in a multilayer structure is the use of interfacial surface generators (26, 27). A schematic diagram of a simple interfacial surface generator is shown in Figure 1-14. This diagram shows how a melt stream can be split and stacked to increase the number of layers in a structure.

![Figure 1-14. Schematic diagram of a two-channel interfacial surface generator](image)

Figure 1-15 shows the operative segments and the final flow for a two channel interfacial surface generator being fed with a two-layer structure. The two-layer structure is represented as white on the top and gray on the bottom as is shown in the first segment. Note how the original two-layer structure is split, spread, and stacked to form a four-layer structure. This type of interfacial surface generator will always produce a structure with twice as many layers at the exit as at the beginning of the channel.
One method to further increase the number of layers in a multilayer structure would be to place a number of the two-channel interfacial surface generators in series. Each new two-channel interfacial surface generator would double the number of layers from the previous structure. This means that placing “n” number of two-channel interfacial surface generators in series would produce $2^{n+1}$ layers when starting with a two-layer structure.

Figure 1-15. Layer pattern produced in an interfacial surface generator

Numerical simulation of devices similar to interfacial surface generators has been discussed previously (28, 29). In these simulations, a device termed a multiflux static mixer was modeled. A finite element mesh of a multiflux static mixer is shown in Figure 1-16. Note how this device has all of the functional characteristics of the interfacial surface generator since the flow is split, spread, and stacked to increase the number of layers.

Figure 1-16. Finite element mesh for a multiflux static mixer

Figure 1-17 shows the velocity contours from one of the simulations. Note how the flow is not balanced between the different channels in the mixer, which would lead to non-uniform layers in the final structure. These simulation results show how crucial the design of interfacial surface generator is in order to achieve good layer uniformity.
Figure 1-17. Flow patterns in a multiflux static mixer

Figure 1-18 shows the finite element mesh for a new design for a multiflux static mixer (28) that was developed by van der Hoeven to improve flow uniformity in the mixer. The improvement that was introduced was adding extra straight elements before and after the mixing section. These sections add additional resistance to the flow and help balance out the flows in the different channels.

Figure 1-18. Finite element mesh for an improved multiflux static mixer

Figure 1-19 shows the numerical results from the improved multiflux static mixer design. These results show that the flow balance between the channels is significantly improved by the addition of the straight channels. This improvement in flow balance should relate directly to improved uniformity of layers in a multilayer system.

Figure 1-19. Flow patterns in an improved multiflux static mixer

The experimental device used by van der Hoeven had dimensions of 10 mm x 10 mm x 150 mm. This allowed the insertion of different geometrical sections to determine their effect on the final layer distribution.
Interfacial surface generators can be used to create a variety of structures. Figure 1-20 shows the steps involved when starting with a two-layer structure with equal thicknesses in each layer (shown in the figure as grey and white). Step A splits the structure in the center of the channel and then the two structures are stacked vertically in Step B. In steps C and D, the layers are spread horizontally and then rejoined to form a four-layer structure with equal layer thicknesses.

Figure 1-20. Layer multiplication starting with two layers of equal thickness

The process of layer multiplication with interfacial surface generators can also be used with other initial layered structures. Figure 1-21 shows an initial three layered structure with unequal layer thicknesses shown in black, white, and grey. The layer multiplication follows the same steps as before but the final structure now contains six layers with dissimilar layer thicknesses.

Figure 1-21. Layer multiplication starting with three layers of unequal thickness
A similar layer multiplication technique could be used on almost any layered structure. An example would be an asymmetric four-layered structure composed of three materials (A, B, and C) with individual layer thicknesses of 20% A / 5% B / 70% C / 5% B where resin B could be an adhesive material. This initial four-layer structure could flow through an interfacial surface generator and layer B would adhere the layers together keeping the structure intact after the layer multiplication.

Another way to increase the number of layers in a multilayer structure would be to use a four-channel interfacial surface generator rather than a two-channel interfacial surface generator. A four-channel interfacial surface generator is shown schematically in Figure 1-22. This device has been described (26) as a more efficient way to multiply the number of layers in a multilayer structure.

Figure 1-22 shows how a four-channel interfacial surface generator performs the same functions of splitting, spreading, and stacking that a two-channel interfacial surface generator does. However, this device will quadruple the number of layers at the exit compared to the number at the entry while the two-channel device will only double the number of layers.

Figure 1-22. Schematic diagram of a four-channel interfacial surface generator

The layer multiplication done in a four-channel interfacial surface generator is shown graphically in Figure 1-23. This diagram shows how a two-layer structure shown in the lower left hand corner of the diagram is split into four two-layer structures which are repositioned, spread horizontally, and stacked to form an eight-layered structure.
One improvement that has been developed for the four-channel interfacial surface generator is the addition of vanes to help control layer thickness (27). This addition is shown in the diagram in Figure 1-24. This diagram shows how the vanes have been added in the section at which the layers are rejoined. Movement of these vanes has similar results to those discussed earlier in conventional feedblocks and dies. The movement of these vanes will preferentially allow more or less material to flow through a specific channel that will change the final layer thickness in the structure.
In order to efficiently generate many layers in a multilayer structure, the style of feedblock described in Figure 1-9 could be combined with the interfacial surface generator concept described in Figure 1-24. This combination would allow the inherent advantages of each device (i.e., the many uniform layers from the feedblock and the quick multiplication of layers by the interfacial surface generator) to combine to produce many microlayers with good layer uniformity.

Another process that has been used in conjunction with the coextrusion of very thin layers is the injection molding process (30). This process has been called the Lamellar Injection Molding and uses multilayer generation in conjunction with layer multiplication prior to injection molding. Figure 1-25 shows the nozzle of an injection molding machine that can inject a multilayer structure into the mold shown that contains different geometric shapes within the molding cavity.

Figure 1-25. Lamellar Injection Molding process

Figure 1-26 shows the results of injecting a multilayer structure into the mold described above. Photographs were taken of the layered structure at each of the locations indicated in the figure. Note that the multilayer structure maintains its integrity throughout each of the geometries in the mold.
One difficulty that has been observed with processing polymeric structures containing very thin layers is instability in the layers near the walls of the processing equipment which can lead to the very thin layers fracturing and breaking up. If maintaining the layered structure is critical for the final properties (optical applications, for instance), even a small number of broken layers can significantly affect the final properties of the film or sheet.

One technique that has been used to minimize the amount of instability in multilayered structures with very thin layers is the use of protective boundary layers (31). Protective boundary layers are thicker layers that are added to the surface of microlayer stacks to move the very thin layers away from the wall of the processing equipment where the highest stresses on the layers occur. By moving the very thin layers into a region of the channel with lower stresses, the breaking up of the layers is avoided.

Figure 1-27 shows one method in which protective boundary layers have been added to an interfacial surface generator to minimize layer break up. This diagram shows a four-channel interfacial surface generator in which protective boundary layers are added at positions “A” and “B”. Position “A” is prior to the splitting of the multilayer structure while position “B” is near the point at which they are rejoined.
Figure 1-27. A four-channel interfacial surface generator with protective boundary layers

Figure 1-28 shows how a protective boundary layer can be added to the multilayer structure at point “A” in Figure 1-27 prior to splitting the channel into four sections. This allows the structures in each of the four channels to have protective boundary layers so that the layers will not breakup as they flow through the device. This diagram also shows how needle valves can be used to regulate the flow of the protective boundary layer material to each section of the multilayer structure before it is split into four sections.

Figure 1-28. Protective boundary layer addition in an interfacial surface generator

Figure 1-29 shows how the protective boundary layer material is added at point “B” in Figure 1-27 as the layers are rejoined together. This application of the protective boundary layer material at this point protects the very thin layers near the wall as they flow out of the interfacial
surface generator and into the die. Note in this figure how protective boundary layers are also present between the four stacks of multilayer structures. This is due to the splitting and stacking of the multilayer structure that already had protective boundary layers applied prior to the splitting.

Figure 1-29. Surface protective boundary layer addition in an interfacial surface generator

1.8 Conclusions

An overview has been given on how coextrusion can be used in many different processes to form multilayered polymer structures. Uniform layer thicknesses are normally required in a coextruded structure in order to produce an optimum product. The next chapter will deal with layer deformation in coextruded structures that may produce structures that are different than what is desired.
Chapter 2  Layer deformation in coextrusion processes

2.1 Introduction

Polymer rheology information is critical for designing coextrusion dies and feedblocks. The flow characteristics of the polymer must be considered when selecting materials for coextruded products.

Viscosities of non-Newtonian polymers are dependent on extrusion temperature and shear rate, both of which may vary within the coextrusion die. The shear rate dependence is further complicated in that it is determined by the position and thickness of a polymer layer in the melt stream. A polymer used as a thin surface layer in a coextruded product experiences higher shear rate than it would if it were positioned as a central core layer. There are several types of flow instabilities that have been observed in coextrusion.

The best designed die or feedblock does not necessarily ensure a commercially acceptable product. Layered melt streams flowing through a coextrusion die can spread nonuniformly or can become unstable leading to layer nonuniformities and even intermixing of layers under certain conditions. The causes of these instabilities are related to non-Newtonian flow properties of polymers and viscoelastic interactions.

2.2 Interfacial distortions in coextrusion

2.2.1 Interfacial distortion due to flow instability

Interfacial instability is an unsteady-state process in which the interface location between layers varies locally in a transient manner. Interface distortion due to flow instability can cause thickness nonuniformities in the individual layers while still maintaining a constant thickness product. These instabilities result in irregular interfaces and even layer intermixing in severe cases.

At very low flow rates, the interface is smooth as is shown in Figure 2-1(a). At moderate output rates, low amplitude waviness of the interface is observed (see Figure 2-1(b)), which is barely noticeable to the eye and may not interfere with the functionality of the multilayer film. At higher output rates, the layer distortion becomes more severe (Figure 2-1(c)). If a large amplitude waveform develops in the flowing multilayer stream within the die, the velocity gradient can carry the crest forward and convert it into a fold. Multiple folding results in an extremely jumbled, intermixed interface. This type of instability, commonly called zig-zag instability, has been observed in tubular-blown film dies, multimanifold dies, and feedblock/single manifold dies.
This instability develops in the die land, and its onset can be correlated with a critical interfacial shear stress for a particular polymer system (32). The most important variables influencing this instability are skin-layer viscosity, skin-to-core thickness ratio, total extrusion rate, and die gap. Although the interfacial shear stress does not cause instability, elasticity is related to shear stress, and interfacial stress is used to correlate variables for a particular system.

Interfacial instability in a number of coextruded polymer systems has been experimentally correlated with viscosity ratios and elasticity ratios (33), and a simplified rheology review has been given (34). Other studies have looked at viscosity differences (35-37), surface tension (38), critical stress levels (32, 39, 40), viscosity model parameters (41-43), and elasticity (44-52).

Other types of instabilities may exist: for example, a problem has been observed in feedblock coextrusion of axisymmetric sheet (53). A wavy interface is also characteristic of this instability but the wave pattern is more regular when viewed from the surface. The instability, commonly called wave instability, originates in the die, well ahead of the die land and internal die geometry influences both the severity and pattern. For a given die geometry the severity of instability increases with structure asymmetry and some polymers are more susceptible to
unstable flow than others. It has been suggested that this type of instability may be related to the extensional rheological properties of the polymers used in the coextruded structure (54). Examples of both zig-zag and wave instabilities are shown in Figure 2-2.

![Interlayer instability patterns](image)

(a) zig-zag
(b) wave

Figure 2-2. Interlayer instability patterns  (a) zig-zag   (b) wave

No complete predictive theory exists for these complicated rheological interactions, but the accumulated experience of polymer producers, equipment suppliers, and experienced fabricators provides guidance in polymer selection.

### 2.2.2 Interfacial distortion from viscosity mismatch

The importance of viscosity matching for layer uniformity was first studied in capillary flow of two polymers in bicomponent fiber production (55-58). Two polymers introduced side by side into a round tube experience interfacial distortion during flow if the viscosities are mismatched. The lower viscosity polymer migrates to regions of highest shear (at the wall) and tends to encapsulate the higher viscosity polymer. It is possible for the low viscosity polymer to
encapsulate the higher viscosity polymer totally. Nature seeks the path of least resistance. The degree of interfacial distortion due to viscosity mismatch depends on the extent of viscosity difference, shear rate, and residence time.

Layer nonuniformities in feedblock fed flat dies occur for the same reason when there is a large enough viscosity mismatch. Low viscosity polymer migrates to wet the die wall. For unencapsulated layers, this migration starts in the die manifold as the layered stream spreads, resulting in increased layer thickness for low viscosity polymer at the edges of the film or sheet. If unencapsulated low viscosity polymer is a core layer, it not only becomes thicker at the edges, but may even wrap around higher viscosity skin layers at the film edges.

Tubular blown-film dies are more tolerant of viscosity mismatch because the layers are arranged concentrically, i.e., there are no ends. Since streamlines cannot cross each other, further migration cannot occur. However, good die design is required to obtain concentric layers.

2.2.3 Interface distortion from viscoelasticity

While matching the viscosities of adjacent layers has proven to be very important, the effect of polymer viscoelasticity on layer thickness uniformity is also important (59-63).

It has been shown that polymers that are comparatively high in elasticity produce secondary flows normal to the primary flow direction in a die that can distort the layer interface. This effect becomes more pronounced as the width of a flat die increases. Appropriate shaping of the die channels can minimize the effect of layer interface distortion due to elastic effects.

Coextruding a structure that contains layers of polymers with low and high levels of elasticity can cause interface distortion due to the differences in elasticity between the layers in flat dies. The effect is typically not observed in tubular dies.

2.3 Solution methods for interfacial stability problems

The zig-zag type of interfacial instability can be reduced or eliminated by increasing skin-layer thickness, increasing die gap, reducing total rate, or decreasing skin polymer viscosity. These methods may be used singly or in combination. These remedies reduce interfacial shear stress, and stable flow results when it is below the critical stress for the polymer system being coextruded. Most often skin layer polymer viscosity is decreased. In feedblock coextrusion the resultant viscosity mismatch imposed by this remedy can cause variations in layer thickness as discussed earlier. Shaped skin layer feedslots are then typically used to compensate and produce a uniform product. A review of techniques used to minimize this type of instability has been given previously (64).

Care should also be taken when designing the joining geometry in a feedblock or die. In order to minimize instabilities, the layers should have similar velocities at the merging point. The joining of the layers should occur in a geometry that is as parallel as is realistically possible rather than joining in a perpendicular manner. The layers should also merge into a channel that is of an appropriate height that does not force one layer to flow into the other.
Wave instabilities are related to the extensional viscosities of the individual layers. This implies that all of the previously mentioned design criteria for layer joining are important for this type of instability as well. In addition, the spreading of the layers in a film or sheet die is also important. Since this type of instability is related to extensional viscosity, the rate at which the layers are stretched in the die will affect the forces in each layer. In structures containing materials with high extensional viscosities, the die should be designed to spread the layers slowly and at a uniform rate. This will help minimize wave pattern instabilities.

2.4 Conclusions

An overview of layer distortions in coextruded structures has been presented. The next chapter will deal specifically with layer distortions relating to structures with different viscosities in the coextruded layers.
Chapter 3  Layer uniformity in coextrusion for structures with layers with different viscosities

3.1 Introduction

Good layer uniformity in coextruded structures is usually a requirement for producing a structure with uniform properties. Many variables can affect layer uniformity. One of the most important variables is the viscosity of the polymer in each layer. This chapter will deal with coextrusion of structures with layers of different viscosities.

3.2 Visualization of viscous encapsulation

As was discussed earlier, when resins with significantly different viscosities are coextruded into a multilayer structure, a phenomenon known as “viscous encapsulation” can take place. This phenomenon is illustrated in Figure 3-1 that shows a two-layer structure flowing down a tube. In this figure, the viscosity of layer “A” is less than layer “B”. This figure shows how the less viscous layer (A) tends to move to the highest area of stress (near the tube walls) and so encapsulate the layer with the lower viscosity (B).

![Viscosity of A < Viscosity of B](image)

Figure 3-1. Viscous encapsulation in a tube

In this part of the study, two-layer coextruded structures were made using different polystyrene resins in each layer with different colored pigments added to each layer to determine the location of the interface. A series of experiments were conducted that showed that the addition of the pigments at the loadings used in these experiments did not affect the flow properties of the resins. These two-layer structures were extruded through a circular channel and the encapsulation velocity was measured experimentally.
The resins chosen for this study were four high impact polystyrene resins (STYRON* 482, 421, 484, and 495), all manufactured by The Dow Chemical Company. For simplicity, these resins will hereafter be referred to as Polystyrene A, B, C, and D, respectively. The rheological properties of these resins at 204°C are shown in Figure 3-2. This figure shows that these resins have significantly different viscosities over the shear rate range tested. These particular polystyrene resins were chosen for this part of the study in order to produce as large a viscosity ratio between the layers in the coextruded structures as possible.

![Figure 3-2. Viscosity comparison of polystyrene resins](image)

The primary coextrusion line used in this study for the two-layer experiments consisted of a 31.75 mm diameter, 24:1 length-to-diameter ratio (L/D) single screw extruder for the substrate resin and a 19.05 mm diameter, 24:1 L/D single screw extruder for the cap resin. These extruders were attached to a feedblock that was designed to produce a two-layer structure consisting of a 20% cap layer and 80% substrate layer. Attached to the exit of the feedblock was a die containing a circular cross-section. A schematic diagram of the primary coextrusion line used in these experiments is shown in Figure 3-3.

* Trademark of The Dow Chemical Company
The circular die channel was designed to have a cross-sectional area of approximately 0.91 cm$^2$ so as to be comparable to other channels that will be discussed later. The axial length of the die channel was 60.96 cm. This die channel was fabricated in two halves and bolted together so that it could easily be split apart for removal of the polymer sample.

For a typical experiment, the coextrusion line was run for a minimum of thirty minutes to ensure that steady-state conditions had been reached. The normal extrusion rate was approximately 3.4 kg/hr for the polystyrene resins that would give a wall shear rate in the range of 1 to 10 reciprocal seconds. The coextruded structures were extruded at approximately 204°C. Variable-depth thermocouples were placed in the melt streams from each extruder just prior to the entry into the feedblock to ensure that the temperatures of the materials were the same prior to being joined together. When steady state was reached, the extruders were stopped simultaneously and the coextruded material was cooled while still in the die channel. After it had cooled to room temperature, the frozen polymer "heel" was removed from the die and examined. This procedure allowed the major deformations of the interface to be analyzed.

Of the four polystyrene resins studied, Polystyrene D was the most viscous (see Figure 3-2). Since Polystyrene D resin was the most viscous, it was used as the substrate layer in a series of experiments in which each of the four polystyrene resins (A, B, C, and D) was coextruded in a two-layer structure as a 20% cap layer over a substrate of Polystyrene D. At a shear rate of 10 reciprocal seconds, the viscosity ratios of the structures were 2.5, 2, 1.4, and 1 for cap layers of polystyrene resins A, B, C, and D, respectively. These viscosity ratios are defined as the viscosity of the substrate divided by the viscosity of the cap layer. In these experiments, the substrate layer of Polystyrene D was always pigmented black while the cap layer containing the different polystyrene resins was always pigmented white.

Figure 3-4 shows the solidified samples removed from the circular die channel for the experiments described above. This figure shows all of the samples viewed from the bottom of the channel so that it is easier to observe the white cap layer flowing around and encapsulating the black substrate layer. The samples are labeled as the cap layer over the substrate layer, i.e., A/D represents a cap layer of Polystyrene A resin coextruded over a substrate layer of Polystyrene D resin.
The first observation to note from Figure 3-4 is that the D/D sample shows no sign of viscous encapsulation. This can be determined by the fact that only the black substrate layer is visible down the entire length of the die channel and so no white cap layer has moved to encapsulate the substrate. This is to be expected since each layer is composed of the same resin processed at the same temperature.

The second important observation from Figure 3-4 is that the higher the viscosity difference between the cap layer and substrate layer, the faster the viscous encapsulation occurs. The samples listed in order of the speed of encapsulation from fastest to slowest are A/D, B/D, C/D, and D/D which have viscosity ratios of 2.5, 2, 1.4, and 1, respectively. Note that complete encapsulation of the A/D sample takes place in approximately 13 cm (~ 5 inches) while the C/D sample is not completely encapsulated even after 20 cm (~ 8 inches).

Figure 3-4. Samples removed from the circular die channel. Viscosity ratio decreases from 2.5 to 1 from top to bottom

Figure 3-5 shows the A/D sample after it has been cut into 2.54 cm sections. This figure shows the sample cross sections beginning near the channel entry and then at distances of approximately 2.5, 5.1, 7.6, 10.2, and 15.2 cm from the channel entrance. Note how the white cap layer is flowing around and encapsulating the black substrate layer. This figure shows that the white cap layer completely encapsulates the black substrate somewhere between 10 and 15 cm downstream from the entry.
3.3 Determination of encapsulation velocity

The movement of the cap layer encapsulating the substrate layer can be measured in order to calculate an encapsulation velocity. The technique used to do this is illustrated in Figure 3-6. The image labeled (a) represents a cross-sectional cut near the entry of the channel while (b) is farther downstream. In the figure, the angle that is defined by the ends of the cap layer is indicated by $\theta_1$ and $\theta_2$. The angles $\theta_1$ and $\theta_2$ define arcs on the channel wall, which have a set length. Measurement of these arc lengths along the distance downstream at which the samples were cut can be used to calculate the encapsulation velocity.

Figure 3-6. Encapsulation measurement diagram

Figure 3-7 shows a plot of the arc length as a function of the channel distance for the various coextruded samples produced. This figure shows how the arc length gets smaller as the structure flows down the channel indicating the encapsulation of the substrate by the cap layer. Also notice how the order in which the structures reach zero arc distance (or full encapsulation) follows the trend of viscosity ratio. The structure with the largest viscosity ratio, A/D, is fully
encapsulated at the shortest distance while the structure with the smallest viscosity ratio, C/D, is fully encapsulated at the longest distance.

Figure 3-7. Encapsulation measurements

The information shown in Figure 3-7 can be used to calculate an encapsulation velocity. The encapsulation velocity is calculated by measuring the distance the cap layer moves divided by the time it takes for that movement. The cap layer movement distance can be taken directly from Figure 3-7. The time related to that distance is calculated based on the average downstream velocity of the cap layer. This can be approximated from the channel flow rate, the resin viscosity, and the channel geometry. Using these data, Figure 3-8 was developed.

Figure 3-8 shows the encapsulation velocity as a function of the distance down the channel. The curves in this figure were generated based on a least squares method to produce the best linear fit to the data. Note that the encapsulation velocities also follow the same trend of faster encapsulation with a larger viscosity ratio. For example, the A/D sample that has the largest viscosity ratio of 2.5 also has the highest initial encapsulation velocity and the shortest distance to full encapsulation.

This plot also shows that the encapsulation velocity is the highest near the entry of the channel and it decreases as the structure flows down the channel. This happens because the driving force for encapsulation is reduced as the structure flows down the channel since the less viscous material is occupying a larger percentage of the high stress region near the channel wall.

The other important point to note from Figure 3-8 is the magnitude of the encapsulation velocities near the entry of the channel, which is on the order of 0.15 cm/s. This compares to the average downstream velocity of the coextruded structure of 1 cm/s. This implies that the magnitude of the highest encapsulation velocity in this study is approximately 15% of the average downstream velocity.
3.4 Conclusions

This chapter has shown that a difference in viscosity between the layers in a coextruded structure can lead to a phenomenon known as viscous encapsulation. This is a very important effect that is encountered in many industrial processes. Designing coextruded structures with similar viscosities in the layers is a well understood technique to control layer deformation. The next chapter will deal with coextruded structures in which the layers have similar viscosities but layer deformation is still observed.
Chapter 4  Layer uniformity in coextrusion for structures with layers with similar viscosities: elastic layer rearrangement

4.1 Introduction

Viscous encapsulation occurs when layers of different viscosities are coextruded to form a multilayered structure. Designing the structure with similar viscosities in the layers, or “viscosity matching”, can minimize this effect. However, even with well matched viscosities, coextruded structures have been processed in which layer deformation still occurs. This chapter will discuss layer uniformity in coextruded structures with similar viscosities.

4.2 Materials

The resins chosen for this part of the study were a high impact polystyrene resin (STYRON 484), a low-density polyethylene resin (LDPE 6411), and a polycarbonate resin (CALIBRE* 300-22) all manufactured by The Dow Chemical Company. The rheological properties of these resins are shown in Figures 4-1 and 4-2. These figures show that these resins have significantly different viscous and elastic flow properties. Figure 4-1 shows that the polystyrene and polyethylene resins are both shear thinning while the polycarbonate resin is more Newtonian in viscosity. Figure 4-2 shows the differences in elasticity between the resins based on their storage moduli. The polystyrene resin appears to be the most elastic (based on storage modulus values) followed by the polyethylene resin and then the polycarbonate resin.

![Figure 4-1. Viscosity comparison of polystyrene, polyethylene, and polycarbonate resins](image)

* Trademark of The Dow Chemical Company
Two-layer coextruded structures were made using the same polymer in each layer with different colored pigments added to each layer to determine the location of the interface. A series of experiments were conducted that showed that the addition of the pigments at the loadings used in these experiments did not affect the flow properties of the resins.

4.3 Experimental set-up

The primary coextrusion line used in this study for the two-layer experiments again consisted of a 31.75 mm diameter, 24:1 length-to-diameter ratio (L/D) single screw extruder for the substrate resin and a 19.05 mm diameter, 24:1 L/D single screw extruder for the cap resin. These extruders were attached to a feedblock that was designed to produce a two-layer structure consisting of a 20% cap layer and 80% substrate layer. Attached to the exit of the feedblock was a die containing one of the different channel geometries studied. A schematic diagram of the primary coextrusion line used in these experiments was shown previously in Figure 3-3. Experiments were run on several different lines with different extruder sizes and feedblock designs, all producing similar results.

Five different die channel geometries were used in this study: a square channel, a rectangular channel, a teardrop shaped channel, a circular channel, and a rectangular manifold channel. The square, teardrop, circular, and rectangular geometries were chosen since they are common shapes used in the design of feedblocks, dies, and transfer lines while the rectangular manifold channel is meant to simulate half of a coat-hanger style die. The square channel had sides that were 0.95 cm long. The teardrop and circular channels were designed to have approximately the same cross-sectional area (0.91 cm$^2$) as the square channel. The rectangular channel had a 4:1 width to height aspect ratio with a width of 2.54 cm and height of 0.635 cm.
The rectangular manifold channel was designed based on principles used in the design of coat-hanger style die manifolds to produce uniform output across the die exit (65). The rectangular distribution manifold maintains a constant 4:1 aspect ratio down its entire length and starts with dimensions identical to those of the rectangular channel. This allows direct comparisons to be made between the two channels. The axial length of each of the die channels was 60.96 cm. These die channels were fabricated in two halves and bolted together so that they could easily be split apart for removal of the polymer sample.

For a typical experiment, the coextrusion line was run for a minimum of thirty minutes to ensure that steady-state conditions had been reached. The normal extrusion rate was approximately 8.6 kg/hr for the polystyrene resin that would give a wall shear rate in the range of 30 to 40 reciprocal seconds. Both the polystyrene and polyethylene resins were extruded at 204°C while the polycarbonate resin was extruded at 260°C. Variable-depth thermocouples were placed in the melt streams from each extruder just prior to the entry into the feedblock to ensure that the temperatures of the materials were the same prior to being joined together. When steady state was reached, the extruders were stopped simultaneously and the coextruded material was cooled while still in the die channel. After it had cooled to room temperature, the frozen polymer "heel" was removed from the die and examined. This procedure allowed the major deformations of the interface to be analyzed.

4.4 Elastic layer rearrangement in a square channel

The initial experiment conducted consisted of extruding the two-layer coextruded structure through the feedblock with no die channel attached. This experiment was run to ensure that the 80/20 (substrate layer thickness/cap layer thickness) structure was produced with a flat interface prior to introducing the coextruded structure into a die channel. The coextruded structure produced is shown in Figure 4-3. This figure shows that an 80/20 structure was indeed produced with a flat interface produced between the two layers.

![Figure 4-3. Two-layer structure at the exit of the feedblock](image)

The next experiment consisted of running an 80/20 structure of polycarbonate resin into the square channel geometry die. The structure produced near the exit of the channel is shown in
Figure 4-4. This figure shows that the interface in the polycarbonate structure is fairly flat and very little deformation has occurred.

![Figure 4-4. Two-layer polycarbonate structure near the exit of the square channel](image)

When a two-layered polystyrene structure was extruded through the square channel die, the resulting structure produced is shown in Figure 4-5. This structure is significantly different than that produced by the polycarbonate resin since there is extensive deformation of the interface. This structure shows that material has flowed up along the die walls to the corners and then it turns and flows toward the center of the channel. Simultaneously, material that was nearer the center of the channel is being pushed up towards the top of the channel.

![Figure 4-5. Two-layer polystyrene structure near the exit of the square channel](image)

This interface shape is obviously not due to viscous encapsulation since identical materials are present in each layer so there can be no difference in viscosity to drive the viscous encapsulation. It is hypothesized that this interface shape is the result of elastic forces that produce secondary flows in the square geometry. These secondary flows would be present in a direction perpendicular to the main flow direction and be driven by second normal stress differences. These secondary flows have been discussed previously for polymer flows by White.
They noted that second normal stress differences could influence the interface shape along with the viscosity difference. They showed experimentally that in bicomponent tube flow, the less viscous layer always encapsulated the more viscous layer regardless of the first and second normal stress differences between the two materials. It should be noted, however, that all of their studies were done in channels with circular cross-sections.

However, since the secondary normal forces are small and difficult to measure, it has been assumed in the past that they could be ignored (67). Studies have been done that show that these secondary flows are produced by differences in the normal forces (61-63) and are not due to viscous effects. These differences in normal forces are produced when a viscoelastic material flows through a non-radially symmetric channel.

Figure 4-6 shows the predicted secondary flows in a square channel for an elastic material. The predicted flow patterns for a viscoelastic fluid flowing through a square channel have been shown previously to contain eight recirculation zones or vortices, two each per quadrant (61-63, 68-72) as is shown in Figure 4-6. These flow patterns appear to correspond well with the interface deformation shown for the polystyrene resin in Figure 4-5. These flows would cause material to move up the walls and then turn back towards the center of the channel. These secondary flows would also cause the material near the center to be pushed upwards toward the top of the channel. Numerical predictions of these secondary flow patterns will be covered in a subsequent chapter.

Figure 4-6. Secondary flow patterns for an elastic material in a square channel

One interesting aspect of the elastic layer rearrangement shown in Figure 4-5 is that it is a phenomenon that continues indefinitely as an elastic material flows down a square channel. This is different from viscous encapsulation since the driving force for viscous encapsulation tends to decrease as the materials flow down the channel as the less viscous material encapsulates the more viscous material and an energetically preferred state is reached. The constant layer rearrangement in an elastic material is shown in Figure 4-7 for a two-layer polystyrene structure. This figure shows the progression of the elastic layer rearrangement as the structure flows down the channel by showing cuts at axial distances from the entry of 5, 20, 30, 40, 50, and 58 cm. The cut in the upper left hand corner was taken from the sample near the entry to the square
channel while the cut in the lower right hand corner was taken from the sample near the exit of the square channel. This figure shows the steady deformation of the layer interface as it progresses down the channel.

Figure 4-7. Progression of a two-layer polystyrene structure as it flows down a square channel

Figure 4-8 shows a sample taken near the exit of the square channel for a two-layered structure composed of polyethylene. This figure shows results similar to those obtained for the polystyrene resin. However, the layer rearrangement for the polyethylene sample is not quite as extensive as the deformation in the polystyrene sample. This is consistent with the results of the measurements of the resins’ storage moduli. If the storage modulus is used as an indication of elasticity of the resin, the polystyrene is the most elastic, the polyethylene is of intermediate elasticity, and the polycarbonate is the least elastic. This order of level of elasticity also corresponds to the amount of layer rearrangement observed in these samples with polystyrene showing the most rearrangement and the polycarbonate the least amount of layer rearrangement.

Figure 4-8. Two-layer polyethylene structure near the exit of the square channel
4.5 Elastic layer rearrangement in a circular channel

As described in the previous section, the layer rearrangements observed were hypothesized to be driven by secondary flows in the square channel caused by second normal stress differences. One way to test this hypothesis would be to extrude the materials through a channel with a radially symmetric geometry that would produce no secondary flows. This was done by extruding the three resins through a die with a circular cross-section. Very little layer rearrangement was observed in each case as compared to the rearrangement seen in the square and teardrop channels. Figure 4-9 shows a cross-sectional cut of the polyethylene resin near the exit of the die with the circular cross-section. The layer interface location in this sample is very similar to the interface location observed at the beginning of the channel implying that the interface did not move substantially as the structure flowed down the channel. Similar behavior was also observed for the polycarbonate and polystyrene resins even though they have substantially different viscoelastic properties compared to the polyethylene resin.

Figure 4-9. Two-layer polyethylene structure near the exit of the circular channel

4.6 Elastic layer rearrangement in a rectangular channel

One question that arises from the experimental studies in the square channel is what effect would changing from a square geometry to a rectangular geometry have on the location and magnitude of the secondary flows. This was studied experimentally by building a rectangular channel with an aspect ratio (width to height) of 4 to 1.

Figure 4-10 shows a sample cut from near the exit of the 4:1 rectangular channel when running polycarbonate resin in each layer. As can be seen in the figure, there is very little deformation in the layer interface over most of the width of the sample, as was true in the square channel as well.
Figure 4-10. Two-layer polycarbonate structure near the exit of the rectangular channel

Figure 4-11 shows the results when processing a polyethylene structure through the rectangular die channel. This figure shows that significant layer rearrangement has again occurred in the polyethylene sample, as was the case in the square channel. However, it can be seen that the interface remains fairly flat over a significant portion of the center of the rectangular channel, which did not occur in the square channel. The large layer rearrangement in the rectangular channel occurs primarily near the edges of the channel. The same general flow patterns that were observed in the square channel are also present in the rectangle as is evidenced by the flow up the walls along the edges of the channel and the upward movement on the substrate layer near the center of the channel.

Figure 4-11. Two-layer polyethylene structure near the exit of the rectangular channel

Figure 4-12 shows the progression of the layer rearrangement as the polystyrene structure flows down the rectangular channel from left (near the channel entrance) to right (near the channel exit). This progression is similar but not identical to the progression seen in the square channel for polystyrene (Figure 4-7). This figure shows that the elastic layer rearrangement continues as the structure flows down the channel. It also shows that the secondary flows are affected by the change in aspect ratio from 1 for the square to 4 for the rectangle. The recirculation zones (which produce layer rearrangement) observed in the rectangular channel for the polystyrene resin are similar to those seen in the square channel but are elongated along the major axis (channel width direction) of the rectangle. This channel geometry appears to cause the vortex associated with the major axis to increase in size while decreasing the minor axis vortex, correspondingly. Although the vortices in the rectangular channel have shifted positions compared to those in the square channel, they are still present in the polystyrene sample. The main difference between the square and rectangular samples appears to be the location and magnitude of the vortices found in each quadrant. However, a substantial portion of the rectangular channel near the center maintains a flat layer interface in contrast to the square channel. This implies that rectangular channels with higher aspect ratios will give more uniform
layers than channels with lower aspect ratios near unity. Like previous results, the polycarbonate resin did not show much movement of the layers.

Figure 4-12. Progression of a two-layer polystyrene structure as it flows down a rectangular channel

Figure 4-13 shows the progression of the layer rearrangement for a polyethylene structure flowing down the rectangular channel from left (near the channel entrance) to right (near the channel exit). This progression is similar to the flow seen in the rectangular channel for polystyrene resin (Figure 4-12). The polyethylene sample once again shows the secondary flows in the corner of the channel. This sample also shows how the white cap layer flows along the centerline back towards the center of the channel.

Figure 4-13. Progression of a two-layer polyethylene structure as it flows down a rectangular channel

4.7 Elastic layer rearrangement in a teardrop shaped channel

In experiments similar to those done with the square and rectangular channels, two layered structures were also processed through a teardrop shaped channel. The flow through a teardrop shaped channel is very important industrially because many commercial film and sheet dies use teardrop shaped channels in their distribution manifolds. Originally, many of the
manifolds in film and sheet dies were circular in cross-sectional shape, as is shown in Figure 4-14(a). However, the transition from a circular manifold to a rectangular shaped land region caused some flow difficulties because of the abrupt change in geometry, especially in coextruded structures. This difficulty was overcome by using a tapered transition from the circular manifold to the rectangular land area, thus producing a teardrop shaped channel. Figure 4-14(b) shows the area of the channel that would be removed to produce the shape in Figure 4-14(c). Because the cross-sectional shape shown in Figure 4-14(c) is difficult to cut across the entire width of a large distribution manifold, many times the back of the manifold (in the circular section) is cut with a flat section as is shown in Figure 4-14(d). This cross-sectional shape is the familiar teardrop shape that is used in many film and sheet die distribution manifolds.

Figure 4-14. Die manifold geometry development from circular to teardrop shape

Figure 4-15 shows the location of the interface near the exit of the teardrop channel for the polycarbonate resin. This sample shows a smooth interface with very little layer movement as the material flows down the channel. In contrast, a section cut from the two-layer polyethylene sample near the exit of the die is shown in Figure 4-16 while a cut from the two-layer polystyrene sample is shown in Figure 4-17.
Figure 4-15. Two-layer polycarbonate structure near the exit of the teardrop channel

Figure 4-16. Two-layer polyethylene structure near the exit of the teardrop channel

Figure 4-17. Two-layer polystyrene structure near the exit of the teardrop channel
The results obtained from the teardrop channel are consistent with those seen in the square channel in that the most layer movement was observed in the polystyrene sample, the least movement in the polycarbonate sample, and the polyethylene sample was intermediate. The teardrop geometry, however, produced only six vortices or recirculation zones (58, 59) as compared to the eight observed in the square channel. This produced a flow pattern that is symmetric only about the centerline of the channel and is therefore different than the flow patterns seen in the square and rectangular channels. It appears from these results that a teardrop shaped channel produces a coextruded structure with more layer thickness non-uniformity than a rectangular channel. These results are, however, dependent on the comparative aspect ratios of the rectangular and teardrop channels. These results are very significant from a commercial perspective since a large portion of the sheet and film dies produced in the industry contain teardrop shaped distribution manifolds.

4.8 Elastic layer rearrangement in a rectangular manifold channel

One half of the rectangular manifold channel is shown in Figure 4-18 along with a polymer sample that had been solidified in the channel. The rectangular manifold channel was designed based on principles used in the design of coat-hanger style die manifolds to produce uniform output across the die exit and is meant to simulate half of a coat-hanger style die. The rectangular distribution manifold maintains a constant 4:1 aspect ratio down its entire length and starts with dimensions identical to those of the rectangular channel. This allows direct comparisons to be made between the two channels. The axial length of the die channel was 60.96 cm. This die channel was fabricated in two halves and bolted together so that it could easily be split apart for removal of the polymer sample.

![Figure 4-18. Rectangular manifold die channel and solid polymer sample](image)

When the three resins were extruded through the rectangular manifold channel, a significant amount of layer rearrangement occurred for the polystyrene and polyethylene samples but not in the polycarbonate sample. Figures 4-19, 4-20, and 4-21 show a series of cross-sectional cuts made at 7.6 cm intervals down the rectangular manifold channel for the two-layer polycarbonate, polyethylene, and polystyrene samples, respectively. Again the cut on the left was made nearest to the feedblock while the cut on the right was near the end of the manifold.
Figure 4-19. Progression of a two-layer polycarbonate structure as it flows down a rectangular manifold channel

Figure 4-20. Progression of a two-layer polyethylene structure as it flows down a rectangular manifold channel

Figure 4-21. Progression of a two-layer polystyrene structure as it flows down a rectangular manifold channel
The flow patterns for the polycarbonate sample in the rectangular manifold channel are shown in Figure 4-19. Note that the layer ratio at the entrance of the manifold is 80/20 (substrate / cap layer) for the two-layer sample and remains constant down the channel. The layer interface between the cap and substrate layer does remain substantially parallel as the flow progresses down the channel. This can be contrasted with the results of the two-layer polyethylene and polystyrene samples as are shown in Figures 4-20 and 4-21. In both of these cases, the interfaces do not remain flat but are distorted at the top of the manifold channels. Note also that although the layer ratio at the entrance of the manifolds is 80/20 (substrate / cap layer) for the two-layer samples, the ratio is approximately 50/50 by the time the structures reach the ends of the manifolds.

It appears that the layer profiles seen in the polyethylene and polystyrene resins in the rectangular manifold channel are produced by a combination of the vortices formed in the corners of a rectangular channel and the leakage of the material out the bottom of the channel. The movements of layers in the top corners of the manifold are very similar to the movements seen in the rectangular channel shown in Figure 4-12. However, it appears that the further the material moves down the channel, the leakage flow becomes a larger percentage of the flow and pulls the layers that have moved in the upper section of the channel down towards the bottom of the channel. These figures show that the leakage flow comes primarily from the lower part of the channel. This combination of flow patterns will produce a fairly uniform layer pattern near the entry of the manifold but the layers near the end of the channel will be very distorted. This may be one mechanism to explain why coextruded structures of polymers with matching viscosities can sometimes produce products with non-uniform layer thicknesses. This could also explain why measurements of individual layer thicknesses in sheet products tend to show poorer distribution near the edges of the sheet compared to the center.

4.9 Conclusions

These results have many implications for the commercial coextrusion of viscoelastic polymers. Since the layer movements observed occurred without any differences in viscosity between the layers, this implies that layer rearrangement can occur in coextrusions in which the polymer viscosities are well matched. Also, since the layers continue to rearrange as the polymer flows down the channel, these results also imply that this effect will become more pronounced in sheet dies as they are scaled up to larger widths. The die used in these experiments would represent a die with a width of approximately 1.2 meters. As discussed earlier, commercial cast film dies have been built with widths up to 10 meters, or approximately eight times as wide as the die used in these experiments!
Chapter 5  Analyzing flow patterns during elastic layer rearrangement using different feedblock configurations

5.1 Introduction

The previous section dealt with the flow of two-layered structures through dies with different cross-sectional geometries. The two-layer structures show the interface deformations for the three materials studied but since there is only one interface, some interpretation of the experimental results is needed to understand how these results can be extended to cover the entire flow field in a particular geometry. For instance, Figure 4-7 shows the interface deformation for a two-layer polystyrene structure as it flows down the square channel. This figure certainly shows the layer movement in the upper half of the sample, but since the entire lower half is pigmented black, no additional information can be gained from this portion of the sample.

In order to address this lack of information about the rest of the flow area, samples were made in which a different feedblocks were developed that produced a variety of structures with alternating layers of the same material that were pigmented black and white. Among those feedblock designs were styles that produced multiple parallel layers (2, 27 and 165 layers), a matrix of 49 strands, and concentric rings with 2 or 13 layers. Schematic diagrams of the structures are shown in Figure 5-1.

![Figure 5-1. Schematic diagrams of structures produced by different feedblock designs](image-url)
5.2 Parallel layer feedblocks (27 and 165 layers)

Samples were made in a unique feedblock that produced a structure with 27 alternating layers of the same material that were pigmented black and white for comparison to the 2 layered samples. A schematic diagram of a feedblock that was similar in style to the one used to produce this structure was shown previously in Figure 1-9. These 27-layered structures contained many more interfaces that could be examined so that more insight could be obtained on the total flow field. Figure 5-2 shows just such a structure containing twenty-seven polystyrene layers near the entry and exit of the square channel. By increasing the number of layers, it is obvious that the flow patterns observed in the upper half of the two-layered structure also occur in the lower half. This many layers make it easier to observe the symmetry of the flow in the square channel.

![Figure 5-2. Twenty-seven-layer polystyrene structure near the entry and exit of the square channel](image)

Since increasing the number of layers from 2 to 27 gave more information on the flow field, the next logical step would be to increase the number of layers even further to gain even more information. This was done by developing a feedblock that could produce coextruded structures with 165 alternating layers of the same material that could be pigmented to show the interfaces. A schematic diagram of the style of feedblock used to produce this structure was shown previously in Figure 1-9. An example of this structure at the entry and exit of the square channel when processing polystyrene is shown in Figure 5-3. Note that at the entry of the channel, all 165 layers are fairly uniform and parallel. Near the exit of the channel, these layers have rearranged significantly and the symmetry of the flow perpendicular to the main flow direction is obvious. Comparing Figure 5-3 with the predicted secondary flows shown in Figure 4-6 makes it much easier to visualize the secondary flow patterns. When compared to a two-layer structure (Figure 4-7) the 165-layer structure gives much more detail on the flow pattern in the channel.
One important observation of the 165-layer structure is that some of the details of the flow in the channel are lost due to thinning of the layers. The thin, parallel layers at the beginning of the channel are thinned further and distorted by the layer rearrangement taking place in the channel and so some of the resolution of the layer interfaces is lost due to the extreme thinness of the layers. Structures with greater than 5000 alternating layers were produced to study the phenomenon of elastic layer rearrangement but details of the flow in the channel were not as easy to determine because of the extreme thinness of the individual layers. Figure 5-4 shows a photomicrograph of approximately 100 layers of a structure that contained over five thousand layers. Note how thin the layers are in this 100-layer section. If the entire structure was shown in this illustration, these layers would be approximately fifty times thinner than they appear in this photomicrograph. This illustrates how too many layers can reduce the optical resolution of the system when trying to trace layer movement.
It appears that 165 layers provide enough information on the entire flow field that structures with more layers are not necessary or desirable due to a lack of optical resolution. Since the 165-layer structures give more information than the 2 or 27-layer structures but do not suffer the optical resolution problems of the 5000-layer structures, they will be used in the remainder of this section to illustrate the flow patterns in the various channel geometries.

Figure 5-5 shows the 165-layer polystyrene structure near the exit of the rectangular channel. This figure shows a similar flow pattern as was seen in Figure 4-12 for the two-layer polystyrene sample but gives more detail. The 165 layers show the extreme symmetry of this flow field. Note that the recirculation zones near the ends of the channel are small in comparison to those seen in the square channel. It should also be seen that the layers near the center of the channel remain parallel over a substantial width of the channel. This implies that a rectangular channel would be superior to a square channel for maintaining parallel layers in a coextruded structure.

Figure 5-5. One hundred and sixty five-layer polystyrene structure near the exit of the rectangular channel

Figure 5-6 shows the 165-layer polystyrene structure near the exit of the teardrop channel. This figure also shows the symmetry that is evident in this flow field that could not be observed in the two-layer structure. It appears that there are only 6 recirculation zones in the teardrop channel compared to the 8 recirculation zones in the square channel. This figure also shows much more distortion in the layers than was present in the rectangular channel (Figure 5-5). This implies that the rectangular channel would produce more uniform coextruded structures than the teardrop channel. This is significant since many of the commercial scale coextrusion dies contain teardrop shaped distribution manifold channels.

Figure 5-6. One hundred and sixty five-layer polystyrene structure near the exit of the teardrop channel
Figure 5-7 shows a series of cross-sectional cuts made at 7.6 cm intervals down the rectangular manifold channel for the 165-layer polystyrene sample. The cut on the left was made nearest to the feedblock while the cut on the right was near the end of the manifold. This figure shows more of the details of the flow field than were seen in the two-layer structure (Figure 4-21). Figure 5-8 shows an expanded view of a few of the cross-sectional cuts shown in Figure 5-7.

Figure 5-7. Progression of a one hundred and sixty five-layer polystyrene structure as it flows down a rectangular manifold channel

Figure 5-8. Progression of a one hundred and sixty five-layer polystyrene structure as it flows down a rectangular manifold channel (expanded view)
As was observed in the two-layer structure, it appears that the layer profiles seen in the polystyrene structure in the rectangular manifold channel are produced by a combination of the vortices formed in the corners of a rectangular channel and the leakage of the material out the bottom of the channel. The movements of layers in the top corners of the manifold are very similar to the movements seen in the rectangular channel shown in Figure 4-21. However, it appears that the further the material moves down the channel, the leakage flow becomes a larger percentage of the flow and pulls the layers that have moved in the upper section of the channel down towards the bottom of the channel. This figure also shows that the leakage flow comes primarily from the lower part of the rectangular channel. This combination of flow patterns will produce a fairly uniform layer pattern near the entry of the manifold but the layers near the end of the channel will be very distorted.

5.3 Multi-strand feedblock

One disadvantage in using layered structures to analyze the elastic layer rearrangement in coextruded systems is the fact that the layers are all aligned in one direction. This means that movements of the layers in a horizontal or vertical direction will produce different visual patterns. One way to circumvent this problem would be to develop structures that are not composed of layers but of strands positioned in a specific array. This was done by designing a feedblock that produced a unique coextruded structure consisting of an array of individual strands of one polymer surrounded by a matrix of another polymer (see Figure 5-1 for an illustration of a 49-strand structure).

Figure 5-9 shows a schematic diagram of a multi-strand feedblock (73) that would produce a 6 row by 6 column array of strands. This diagram shows how the polymer “A” is distributed to the strands and then encapsulated by a matrix material “B”. It also shows how material from an additional flow stream “C” may be added to encapsulate the strands with another layer.

![Figure 5-9. Schematic diagram of a multi-strand feedblock](image-url)
Figure 5-10 shows an example of a cross section of a structure that could be produced by this type of feedblock in which an array of 9 rows by 9 columns of strands is produced in which each strand has been individually encapsulated.

![Cross section from a multi-strand feedblock](image)

Figure 5-10. Cross section from a multi-strand feedblock

This style of feedblock allows tracing of flow paths within a particular geometry by following the distortions of the individual strands from the entry to the exit of the channel which can be used to predict layer interface location(s) in coextruded layered structures. This feedblock not only allows tracing for interface locations for coextrusion flows, but also can be used to show the flow patterns in monolithic viscoelastic polymer flows.

The initial coextrusion experiment was conducted with a 49-strand feedblock (7 rows by 7 columns). This experiment consisted of operating the feedblock with no die attached to ensure that the strands were uniform in diameter and location at the exit of the feedblock before they entered the die. Figure 5-11 shows the strand profile for the polystyrene resin at the end of the feedblock. The 49 strands of polystyrene were pigmented black and the matrix polystyrene pigmented white so that the interfaces could be clearly distinguished between the two. Note that the positions of the strands at the exit of the feedblock clearly show the 7 row by 7 column pattern and the strands are fairly uniform.

![Forty-nine-strand polystyrene structure near the exit of the feedblock](image)

Figure 5-11. Forty-nine-strand polystyrene structure near the exit of the feedblock
The next experiment consisted of running the same polystyrene resin through the 49-strand feedblock system with an attached die with a square cross-sectional shape. Cross-sectional cuts of this sample taken at the entry to the channel (shown on the left in the figure) and at 50 cm downstream from the feedblock (on the right in the figure) are shown in Figure 5-12. This figure clearly shows that the strand movement is symmetrical within each quadrant as material moves along the channel walls into the corners and then toward the center of the channel along the 45 degree diagonal line. This figure also shows how each individual strand has moved and been deformed as it flowed down the channel. Note that the central point in the original matrix has remained essentially unchanged at the exit since it represents a neutral or stagnant point with respect to the secondary flows.

![Figure 5-12. Forty-nine-strand polystyrene structure near the entry and exit of the square channel](image)

The strands shown in Figure 5-11 rearrange progressively as the polymer flows down the channel producing a more distorted strand profile as the distance traveled increases. This phenomenon is shown in Figure 5-13. In this figure, the interfaces are shown at several intervals from the entry of the channel to near the exit (viewed from left to right, respectively). This figure allows the tracing of the flow path of any of the individual strands as it flows down the channel. This type of figure gives a much better understanding of the deformations occurring due to secondary flows compared to structures with parallel layers.

![Figure 5-13. Progression of a forty-nine-strand polystyrene structure as it flows down a square channel](image)
Figure 5-14 shows the forty-nine-strand polystyrene structure near the entry (top) and exit (bottom) of the rectangular channel. This figure gives an indication of just how much deformation actually occurs in the rectangular channel. Note the high degree of elongation of the strands near the top and bottom walls as well as the compression of the strands near the side walls. Even though the overall deformation in this geometry is less than the square geometry, there is still significant deformation for the elastic polystyrene resin.

Figure 5-15 shows the forty-nine-strand polystyrene structure near the entry (left) and exit (right) of the teardrop channel. These images show that the movement of the strands in the teardrop geometry is very large and the flow field appears to cause more deformation in the strands than was evident in the rectangular geometry. It is also interesting to note that the strand that started in the center of the 7 by 7 matrix at the entry of the channel has deformed at the exit of the channel. This differs from the results of the square and rectangular channels since the central strand was not deformed in those geometries. This deformation that occurs to the central strand in the teardrop channel is probably due to the fact that there is not a central neutral or stagnant point in the center of the channel due to the different symmetry that exists. This will be discussed in more detail in the numerical simulation section.
Figure 5-15. Forty-nine-strand polystyrene structure near the entry and exit of the teardrop channel

Figure 5-16 shows the forty-nine-strand polystyrene structure near the entry and exit of the circular channel. Note that the location and shape of the strands is essentially the same at the entry and exit of the channel. This once again confirms the fact that even with a very elastic material like polystyrene, no secondary flows are developed in a radially symmetric channel. This demonstrates the importance of understanding how channel geometry relates to viscoelastic flow effects when designing equipment for polymer processing.

Figure 5-16. Forty-nine-strand polystyrene structure near the entry and exit of the circular channel

Figures 5-17, 5-18, and 5-19 show a series of cross-sectional cuts made at 7.6 cm intervals down the rectangular manifold channel for the polycarbonate, polyethylene, and
polystyrene samples, respectively. Again the cut on the left was made nearest to the feedblock while the cut on the right was near the end of the manifold.

Figure 5-17 shows the progression of a forty-nine-strand polycarbonate structure as it flows down a rectangular manifold channel. Near the entry (on the left), all 49 strands in the matrix are still visible. As the flow progresses down the channel (left to right in the figure), the rows of 7 strands flow fairly uniformly out of the bottom of the channel leaving the rows above intact. This continues until the final cut where only the row that originated at the top of the channel at the entry remains. This is very important because it implies that any small defect in a coextruded structure that is introduced near the top of the die channel at the entry will be magnified by the time it reaches the end of the channel. Comparing the relative heights of the top row of strands near the entry and the exit of this manifold channel shows this phenomenon. Near the entry, the top row occupies approximately one seventh (~15%) of the channel height while near the end it occupies more than half (>50%) of the channel height.

Because the flow out of this manifold is so uniform for the polycarbonate material, this combination of manifold and material would be a good choice for an inverse design of the feedblock. The feedblock channels could be designed to produce a film or sheet with proper layer distribution in a fairly straightforward manner.

Figure 5-17 demonstrates how uniform the flow can be down a die manifold channel for an inelastic material like polycarbonate. However, the flow for more elastic materials like polyethylene and polystyrene are not so uniform in this type of manifold channel. Figure 5-18 shows the progression of a forty-nine-strand polyethylene structure as it flows down a rectangular manifold channel. As in the polycarbonate sample, all 49 strands are visible at the entry of the channel. However, as the strands flow down the channel there are significant differences in how the strands flow out of the bottom of the channel. In the polycarbonate
structure (Figure 5-17), the tops and bottoms of each row of strands remained essentially flat as they moved down the channel. In the polyethylene structure (Figure 5-18), the ends of the rows begin to exhibit curvature as they flow down the channel. This curvature means that each row will not flow out of the manifold as a group but will be spread out as the material near the center of the channel flows out sooner than that near the walls. This is shown in Figure 5-18 by the next to last sample from the end of the channel. In this cut, there are 9 strands across the width of the channel near the bottom as opposed to 7 strands at the beginning. This means that one row of strands has now overlapped with another showing that they are not flowing out in the same manner as the polycarbonate strands.

![Figure 5-18. Progression of a forty-nine-strand polyethylene structure as it flows down a rectangular manifold channel](image)

Figure 5-18 shows the progression of a forty-nine-strand polystyrene structure as it flows down a rectangular manifold channel. This figure shows very similar results to the polyethylene results shown in Figure 5-18. However, the distortions in the tops and bottoms of the rows of strands are even more exaggerated than in the polyethylene sample. The reason for the distortion of these strands can be determined by looking at the results shown in Figures 5-7 and 5-8 for the 165-layer polystyrene sample. These figures clearly show the secondary flows in the upper corners of the manifold channel that become magnified as the structure flows down the channel. These secondary flows would cause the material in the center of the channel to flow downwards more quickly while the material near the walls would be forced upwards and exit the channel later. This phenomenon produces the curvature of the tops and bottoms of the rows of strands in the more elastic materials.
5.4 Concentric ring feedblocks (2 and 13 rings)

As was described earlier, the two main styles of coextruded structures are parallel layers and concentric layers. While parallel layers are used in many film and sheet structures, many structures with concentric layers, such as in wire coating, are also used. This section will focus on the effect of secondary flows on coextruded structures composed of concentric layers.

Coextruded structures containing concentric layers were formed using a specially designed feedblock shown schematically in Figure 5-20 (74). This diagram shows the tip of the extruder screw on the right side where the polymer flows into the concentric ring feedblock. This feedblock will produce a core layer of polymer A encapsulated by a layer of polymer B. Adding more encapsulating sections to the feedblock will produce coextruded structures with more concentric layers. This concept is very similar to the one used in stackable plate blown film dies as was shown previously in Figure 1-2.
Figure 5-21 shows an example of a coextruded structure composed of concentric layers. This figure shows a two-layer concentric ring polystyrene structure near the entry of a circular channel. This polystyrene structure is composed of a 30% skin layer and a 70% core layer. The initial series of experiments were conducted with structures that contain two layers.

Figure 5-21. Two-layer concentric ring polystyrene structure near the entry of the circular channel

Figure 5-22 shows the same two-layer concentric ring polystyrene structure that was shown in Figure 5-21 but near the exit of the circular channel. As was observed in the parallel layered and strand structures, no significant deformation of the concentric layered structure occurs as it flows down the circular channel. This again demonstrates that no secondary flows occur in a radially symmetric structure.

Figure 5-22. Two-layer concentric ring polystyrene structure near the exit of the circular channel
Figure 5-23 shows the two-layer concentric ring polystyrene structure after it has been extruded into the entry of the square channel. This structure also contains a 30% skin layer of polystyrene that has been pigmented black to allow observation of the layer interface.

As the two-layer concentric ring polystyrene structure flows down the square channel, the layers rearrange and produce the structure shown in Figure 5-24. Note that the layers near the walls have become much thinner than in the original structure while material has flowed from the corners toward the center of the channel along the 45 degree diagonals. This result is very similar to that seen previously in the structures with initially parallel layers. This result also follows the trend predicted for secondary flows produced by elastic effects, as was shown in Figure 4-6. This figure demonstrates that the same elastic forces are present in structures composed of concentric rings as are present in structures composed of planar layers.
Figure 5-25 shows a two-layer concentric ring polystyrene structure near the entry of the teardrop channel. Note that the outer black layer is fairly thick and uniform around the structure. Compare that structure with Figure 5-26 which shows a two-layer concentric ring polystyrene structure near the exit of the teardrop channel. This figure shows that the black layer along the walls has thinned considerably while more material is flowing to the “corners” of the teardrop structure. The flow in these figures follows the trends shown earlier for flows of elastic materials in teardrop channels. The recirculation vortices should move material along the channel walls until it reaches the corners where it should move more toward the central axis of the teardrop.

Figure 5-25. Two-layer concentric ring polystyrene structure near the entry of the teardrop channel

Figure 5-26. Two-layer concentric ring polystyrene structure near the exit of the teardrop channel
Figure 5-27 shows a two-layer concentric ring polystyrene structure near the entry of the rectangular channel. This can be compared with Figure 5-28 which shows a two-layer concentric ring polystyrene structure near the exit of the rectangular channel. Note that the major deformation that has occurred near the exit of the channel is the movement of the black layer along the central axis toward the center of the channel. This demonstrates how much stronger the vortex near the center of the channel is compared to the vortex near the ends of the channel.

![Figure 5-27. Two-layer concentric ring polystyrene structure near the entry of the rectangular channel](image1)

Figure 5-28 also shows that there was less deformation in the rectangular channel than there was in the teardrop channel. This has been a consistent result in these experiments regardless of the type of layered structure used in the experiment.

Figure 5-29 shows the progression of a two-layer concentric ring polystyrene structure as it flows down a rectangular manifold channel (with the entry shown on the left and the end of the channel on the right). In the first cut on the left in the figure, the intact concentric ring structure can be seen. As the structure moves down the channel, the black and white layers can be observed to flow out of the bottom of the manifold channel. Note that by the time that the structure has flowed to near the end of the manifold channel, only black material that started near the top of the channel at the entry remains.
Figure 5-29. Progression of a two-layer concentric ring polystyrene structure as it flows down a rectangular manifold channel.

Figure 5-30 is similar to Figures 5-23 and 5-24 since it shows a two-layer concentric ring structure near the entry and exit of the square channel. However, this figure shows the results for the polycarbonate resin rather than the polystyrene resin. This figure shows how little deformation takes place in a less elastic material like polycarbonate when compared to polystyrene. Even though the square channel shows the largest layer deformations in the more elastic materials, very little layer deformation is seen in the polycarbonate material.

Figure 5-30. Two-layer concentric ring polycarbonate structure near the entry and exit of the square channel.

Figure 5-31 shows a two-layer concentric ring polycarbonate structure near the entry and exit of the teardrop channel. There seems to be very little difference in the structures at the entry and the exit of the teardrop channel when running polycarbonate. Once again, this figure shows how little deformation occurs when running a material like polycarbonate that is lower in elasticity than the other resins.
There are several advantages and disadvantages in using a structure with concentric rings compared to planar layers or a matrix of strands to examine secondary flows in different geometries. The main advantage of the ring structure compared to the planar structure is the symmetry of the concentric ring structure. The ring structure also has advantages over the multi-strand structure because it allows deformation very near the channel walls to be studied. The main disadvantage of the concentric ring structure is the inability to observe layer deformations near the center of the flow channel. This problem can be overcome by adding multiple concentric rings to the structure so that layer deformation can be studied from the center of the channel to the walls while still maintaining the symmetry of the ringed structure.

5.5 Thirteen concentric rings

To take advantage of the benefits provided by studying concentric ringed structures, a new feedblock was developed that produced a structure with 13 concentric rings. This style of feedblock used the same principles as those shown in Figure 5-20 but with more plates added to produce more concentric layers, similar to the technique shown in Figure 1-2. An example of the output from this feedblock is shown in Figure 5-32. This figure shows a thirteen-layer concentric ring polystyrene structure in which alternating layers have been pigmented black and white to allow observation of the interfaces. This structure will allow examination of elastic layer deformation across the entire channel while maintaining the symmetry of the structure.
Figure 5-33 shows a thirteen-layer concentric ring polystyrene structure near the entry and exit of the square channel. Note that the structure near the entrance of the square channel (on the left in Figure 5-33) has rings that extend across the entire structure so that layer deformations across the entire structure can be observed. The structure near the exit of the channel is shown on the right side of Figure 5-33. This shows in great detail the flow patterns that occur in each quadrant and how they follow the secondary flow patterns shown in Figure 4-6. This figure shows that the thirteen-layer concentric ring structure allows observation of the deformations that occur across the entire channel as well as showing the symmetry of the deformations in the channel.

![Figure 5-33. Thirteen-layer concentric ring polystyrene structure near the entry and exit of the square channel](image)

As was shown in the layered and strand structures, the layer deformation continues as the ringed structure flows down the channel. This is shown in Figure 5-34 for a thirteen-layer concentric ring polystyrene structure. These samples were taken at intervals of approximately 10.16 cm from the entry of the channel (shown in the upper left hand corner of the figure) to near the exit of the channel (shown in the lower right hand corner). These images show how uniformly the layers deform as they move down the channel.

![Figure 5-34. Progression of a thirteen-layer concentric ring polystyrene structure as it flows down a square channel](image)
Figure 5-35 shows a thirteen-layer concentric ring polystyrene structure near the entry and exit of the rectangular channel. Note that in the sample taken near the exit, the layers near the center of the structure are still parallel and have not been substantially deformed. However, there is more deformation of the layers near the edges of the channel. The layer deformations in the channel show that material is pushed from the edges towards the middle of the channel along the main axis which results in the oval layers that were present near the entry of the channel becoming more rectangular shaped near the end of the channel. This figure also shows, however, that the smaller secondary flow areas in the corners of the channel are also trying to move material from the center of the channel towards the edge of the channel along the main axis. This results in the small projections seen along the centerline of the main axis near the edges of the channel. The projections are formed from a portion of a ring near the wall that was stretched in opposite directions by the influence of the secondary flows near the wall. This flow pattern can also be observed in Figure 5-5, which shows a 165-layer structure in a rectangular channel, but it is less obvious in Figure 5-28 that shows the two-layer concentric ring structure. This once again shows the details that the multiple concentric ring structure can show compared to the other structures.

As was shown in Figure 5-35 for the rectangular channel, Figure 5-36 shows a thirteen-layer concentric ring polystyrene structure near the entry and exit of the teardrop channel. The structure near the entry is shown on the left and the structure near the exit is shown on the right. The structure near the entry of the teardrop channel shows uniform layers that change from a circular shape near the center of the channel to more teardrop shaped layers near the walls. These layers are deformed significantly in the structure shown near the exit of the channel on the right. The structure is very symmetric about the centerline.

This figure shows how the secondary flow fields can move material from near the center of the channel to near the walls, and vice versa. For example, the central white circular area
shown in the middle of the channel near the entry is stretched vertically into a line until the ends of the line are near the walls of the channel. In contrast, the outermost white ring near the entry has had the part that was near the tip of the teardrop deformed to the point that it is nearer the center of the channel near the exit.

Figure 5-36. Thirteen-layer concentric ring polystyrene structure near the entry and exit of the teardrop channel

Figure 5-37 shows the progression of a thirteen-layer concentric ring polystyrene structure as it flows down a rectangular manifold channel. This figure is very instructive in showing how the material flows down and out of this manifold channel. Recall that this channel was designed to mimic the flow in a coat-hanger style film or sheet die and so material not only flows down the manifold but also out the bottom of the channel. This can be seen by counting the number of black rings that are visible as the flow progresses down the channel from the entry (left) to the exit (right). Near the entry, all six black rings are visible since the flow out of the bottom of the channel has just begun. In the next cut, only the top halves of the six rings are visible since the bottom portions of the rings have all flowed out of the bottom of the channel. As the flow progresses down the channel, the number of rings decreases from six near the entry to only one near the exit. Once again as was seen with the layered and strand structures, the material that started at the top of the entry channel becomes the majority of the entire channel near the exit.

This figure also shows some similarities to the flow patterns seen in the rectangular channel with the thirteen-ring structure shown in Figure 5-35. The projections of the black layer observed near the ends of the rectangular channel also show up as a projection in the topmost black layer in the rectangular manifold channel. This is understandable since the aspect ratios of the rectangular channel and the rectangular manifold channel are both four to one. The only difference between the two channels is the addition of the flow out of the bottom of the rectangular manifold channel that affects the final flow pattern. This final flow pattern is a combination of the flow out of the bottom of the channel superimposed on the secondary flow patterns seen in the rectangular channel.
5.6 Concentric ring feedblock with tapered square channel

Since the thirteen-layer concentric ring structure proved to be very useful for showing layer deformations across an entire die channel, it was decided to maintain the use of this structure to look at the affect of modifying the square channel geometry. The square channel geometry used for all previous studies maintained constant dimensions (0.95 cm by 0.95 cm) down the entire length (61 cm) of the flow channel. In order to magnify the secondary flow effects, a new channel was designed and fabricated in which the square channel tapered from 0.95 cm on a side to 0.475 cm on a side, effectively reducing the flow area at the channel exit by a factor of four compared to the entry. This should cause an increase in the flow velocity and shear rate as the flow progresses down the channel, which should increase the secondary flow effects. This flow now becomes more three dimensional since the channel dimensions are now changing in all three dimensions.

Using the same processing conditions as were used previously for the straight square channel, an experiment was run using the tapered square channel. Figure 5-38 shows the results for the thirteen-layer concentric ring polystyrene structure near the exit of the straight and tapered square channels. The two images are shown in appropriate sizes to indicate difference in final dimensions between the straight and tapered channel. Note that more extensive deformation of the layers has occurred in the tapered channel as compared to the straight channel. This indicates that the secondary flows are stronger in the tapered channel than in the straight channel.
Figure 5-38. Thirteen-layer concentric ring polystyrene structure near the exit of the straight square channel and the tapered square channel

In order to show a better comparison, Figure 5-39 shows the thirteen-layer concentric ring polystyrene structure near the exit of the straight and tapered square channels with the tapered channel expanded to the same size as the straight channel. This figure makes it easier to observe how much more deformation occurs in the tapered square sample. For example, note how much farther the white material has moved down from the corners along the 45 degree diagonals toward the center of the channel in the tapered square sample compared to the straight square sample.

Figure 5-39. Thirteen-layer concentric ring polystyrene structure near the exit of the straight and tapered square channels with the tapered channel expanded to the same size as the straight channel

5.7 Conclusions

This chapter has discussed the phenomenon of elastic layer rearrangement. This effect is produced in non-radially symmetric geometries when processing elastic materials due to secondary motions that are produced by an imbalance in the second normal stress difference.
The next chapter will deal with measurement of the layer rearrangement velocities due to elastic as well as viscous forces.
Chapter 6  Comparison of layer rearrangement velocities

6.1  Introduction

Layer distortions due to viscous encapsulation and elastic layer rearrangement have been discussed. This chapter will seek to compare the layer rearrangement velocities produced by these two effects.

6.2  Experimental measurement of elastic layer rearrangement velocities

Figure 5-34 showed a series of samples taken at 10.2 centimeter intervals as the flow progresses down the square channel. It can be seen in this figure that the interface deforms more as the material flows farther down the channel.

The samples in Figure 5-34 were collected as is shown in Figure 6-1. Samples were cut from the die heel at 2.54 centimeter intervals down the channel (the “x” in Figure 6-1 represents the 2.54 cm distance downstream). These sequential samples could then be used to measure layer movement, which in turn could be used to calculate secondary flow velocities.

![Extrusion Direction](image)

Figure 6-1. Thirteen-layer concentric ring polystyrene structure sample collection

Figure 6-2 shows two representative polystyrene samples in which the layer movement could be determined by measuring the diagonal distances, L1 and L2. The layer movement distance (L2-L1) divided by the time (t) it took the material to flow “x” distance gives the secondary flow velocity (Vₛ).
There are two important points to consider when making these measurements to calculate the secondary flow velocity. The first point to consider is the symmetry of the thirteen-layer concentric ring structure in the square channel. Because of this symmetry, four measurements can be made on each sample since the flow along each diagonal should be equivalent. This allows for a better average layer movement distance measurement ($L_{\text{average}}$) for each sample. This is illustrated in Figure 6-3.

The second point to consider is using the correct time in the secondary flow velocity calculation. A very simplistic way to get the time would be to take the downstream distance between the samples (x) and divide it by the average downstream velocity (calculated from the volumetric flow rate and the channel cross-sectional area). However, since the downstream velocity is not constant across the channel but is more parabolic in shape, this type of calculation would be less accurate than one in which the cross-channel velocity gradient was taken into account.
An example of how this was done for the polystyrene sample is shown in Figure 6-4. This figure shows a plot of the velocity profile along a diagonal of the square channel starting in the corner and extending to the center of the channel. This plot was obtained by performing a three dimensional finite element analysis of the viscous flow of the polystyrene resin using the viscosity shown earlier. This velocity profile was used in conjunction with the downstream distance between samples to calculate a more accurate time for the secondary flow velocity calculation.

![Figure 6-4](image)

Figure 6-4. Downstream velocity along the square channel diagonal

Figure 6-5 shows the secondary flow velocity as a percentage of the downstream velocity versus the flow distance down the channel for the polystyrene resin used in this study. The data are plotted as percentages rather than absolute values to indicate their order of magnitude compared to the main flow direction velocity. As can be seen in the figure, the secondary flow velocity measurements range from 0.10 to 0.55% of the main flow direction velocity, with an average value of 0.32%.

![Figure 6-5](image)

Figure 6-5. Secondary velocity as a percentage of the downstream velocity for a polystyrene resin
Figure 6-6 shows similar results for the polyethylene resin as was shown in Figure 6-5 for the polystyrene resin. The experimentally measured secondary flow velocities ranged from 0.18 to 0.52% of the main flow direction velocity, with an average value of 0.33%. Even though these secondary velocities are small in magnitude compared to the main flow velocity, they can still produce substantial layer deformations as was demonstrated previously.

![Figure 6-6. Secondary velocity as a percentage of the downstream velocity for a polyethylene resin](image)

When comparing Figures 6-5 and 6-6, it can be seen that the secondary flow velocities for the polystyrene resin seem to increase with increasing channel distance while the polyethylene velocities seem to be less dependent on the distance down the channel. However, it should be remembered that the magnitudes of these velocities are very small and any errors in the layer deformation measurements could result in large changes in the calculated velocities. When the results for the polystyrene and polyethylene resins were analyzed statistically, it was found that the secondary flow velocities were independent of the flow distance down the channel and could be assumed to be constant.

6.3 Comparison of experimental measurements of viscous encapsulation and elastic layer rearrangement velocities

The next series of experiments consisted of structures in which both viscous encapsulation and elastic layer rearrangement could occur. These experiments consisted of extruding two-layer structures with different viscosities in each layer through a die channel with a square cross section. The differing viscosities (with the less viscous material always in the cap layer) would produce viscous encapsulation while the flow through a square channel would produce elastic layer rearrangement.

This combination of flows is illustrated in Figure 6-7. This figure shows a two-layer structure in which the white, less viscous cap layer flows around the black, more viscous...
substrate layer as is indicated by the arrows to the left of the figure. In addition, the elastically driven secondary flow pattern is shown in the black substrate layer. This is an interesting combination of flows since sometimes the two flows are aligned in the same direction while at other times they are opposed. For instance, when the cap layer first starts to move down the wall to begin encapsulating the substrate layer, the elastic secondary flows are trying to drive the flow upwards along the wall and so the two flow patterns are in opposition. Once the cap layer has crossed the centerline of the channel, the elastic secondary flows are now directed down the wall in the same direction as the encapsulating flow. However, once the encapsulating layer flows turn past the bottom corner, the elastic secondary flows are again moving in the opposite direction as the encapsulating flow. Of course, both of these flow patterns are also superimposed on the structure’s downstream velocity profile so the flow of a coextruded, viscoelastic polymer structure down a square channel is indeed a complex flow field.

![Figure 6-7. Combination of flow patterns produced by viscous encapsulation and elastic secondary flows in a square channel](image)

In order to make measurements of the velocities in these complex flow fields, experiments were carried out using the same four polystyrene resins (A, B, C, and D) that were described in section 3. These same combination structures were coextruded through a square channel rather than a round channel as was described previously. These experiments were conducted at flow rates of 3.4 kg/hr with the cap layer being 15 percent of the total channel flow rate and the substrate comprising the other 85 percent. As was described previously, the cap layer always contained the less viscous of the two polymers.

The experimental procedures used were similar to those described previously. The coextrusion line was run for a minimum of thirty minutes to ensure that steady-state conditions had been reached. The normal extrusion rate was approximately 3.4 kg/hr for the polystyrene resins that would give a wall shear rate in the range of 1 to 10 reciprocal seconds. The coextruded structures were extruded at approximately 204°C. Variable-depth thermocouples were placed in the melt streams from each extruder just prior to the entry into the feedblock to ensure that the temperatures of the materials were the same prior to being joined together. When steady state was reached, the extruders were stopped simultaneously and the coextruded material
was cooled while still in the die channel. After it had cooled to room temperature, the frozen polymer 'heel' was removed from the die and examined.

Figure 6-8 shows the interface locations for a coextruded structure with Polystyrene A in the cap layer and Polystyrene D in the substrate layer as it flows down a square channel. Recall that Polystyrenes A and D have the largest viscosity ratio of approximately 2.5. The figure shows the interface location as a function of the downstream distance from the entrance of the channel.

Note that near the entry of the channel the interface is fairly flat between the layers but some encapsulation flow has already begun near the edges of the channel. At a distance of approximately 5 centimeters, the cap layer has flowed down the channel walls to the point that it has reached the bottom corners of the channel. At this point the cap layer continues flowing on around the corner of the channel. Near the end of the channel, a cut is shown at approximately 50 centimeters in which the cap layer has fully encapsulated the substrate layer.

However, it should be noted that the shape of the interface at the 50-centimeter cut is not what would be expected from viscous encapsulation flows alone. Note that near the top of the channel, small layers of black substrate material are beginning to flow along the upper wall towards the corners. It can also be seen that the white cap layer interfaces in the lower corners have been somewhat distorted. These flows can be attributed to the elastic secondary flows that would drive flows along the upper channel walls towards the corners and also move material from the lower corners back towards the center of the channel. These flows are consistent with what is shown in Figure 6-8.

Figure 6-8. Viscoelastic flow in a square channel for A/D structure
Figure 6-9 is similar to Figure 6-8 but it shows the interface locations for a coextruded structure with Polystyrene B in the cap layer and Polystyrene D in the substrate layer as it flows down a square channel. The viscosity ratio between these resins is approximately 2:1. This difference in viscosity ratio gives rise to some differences in the layer interface locations as the structure flows down the channel.

This figure shows that the cap layer does not reach the bottom corners of the channel as soon as it did for the A/D structure, which has a higher viscosity ratio (2.5 vs. 2). There is also a significant difference in the shape of the interface at the last cut at approximately 50 centimeters. Note that the flow along the top wall and the distortion of the interface near the bottom corners is even more exaggerated in the B/D sample than in the A/D sample.

This is an important observation since it implies that as the viscosity ratio becomes smaller, the elastic forces become more important in determining the final interface shape. This makes sense since the lowest viscosity ratio achievable (1:1) would produce no viscous encapsulation since the layers would have identical viscosities. In this case, the elastic secondary flows would cause the interface deformation.

Figure 6-9. Viscoelastic flow in a square channel for B/D structure

Figure 6-10 is similar to Figures 6-8 and 6-9 but it shows the interface locations for a coextruded structure with Polystyrene C in the cap layer and Polystyrene D in the substrate layer as it flows down a square channel. This material combination produces a viscosity ratio of 1.4. Note that it takes even longer for the cap layer material to reach the bottom corners and the interface shape for the 50-centimeter cut is even more distorted than before. These observations follow the same trends produced by Figures 6-8 and 6-9.
Figure 6-10. Viscoelastic flow in a square channel for C/D structure

Figure 6-11 shows a comparison of the coextruded structures from near the exit of the square channel for the three samples described previously in Figures 6-8, 6-9, and 6-10 plus a sample from near the exit of the square channel for a structure composed of Polystyrene D in both layers. This figure also shows the viscosity ratios for the four structures, ranging from 2.5 down to 1. It should be noted that the sample with a viscosity ratio of 1 (same material in each layer) exhibits only elastic layer rearrangement since there is no viscosity difference between the layers. Note that as the viscosity ratio decreases from left to right, the interface deformation due to elastic layer rearrangement increases. As was discussed previously, this implies that as the viscosity ratio approaches unity, the elastic secondary flows become more important in affecting the interface deformation.

Figure 6-11. Comparison of final structures for viscoelastic flow in a square channel. The letters indicate the material combinations while the numbers indicate the viscosity ratios.
Data from images similar to those in Figures 6-8 through 6-10 were used to calculate a layer movement velocity for the three samples. This was done in a manner similar to the procedures described in previous sections. This technique is illustrated for these samples in Figure 6-12. This figure shows how the movement of the cap layer along the wall was measured. The difference in the two measured lengths, \( L_1 \) and \( L_2 \), is divided by the time it took for the layers to move downstream from the position where the first sample was located to the position of the second sample in order to calculate the layer movement velocity.

\[
V = \frac{(L_2 - L_1)}{t}
\]

Figure 6-12. Layer movement velocity measurement in a square channel

Using the technique shown in Figure 6-12, the three coextruded polystyrene samples with viscosity differences between the layers were analyzed for their layer movement velocities. Figure 6-13 shows the results for the three samples on one plot of velocity versus distance down the die channel. While it is known that there are differences in the samples, this plot allows an overall examination of the trends. This plot shows that the layer movement is at its maximum near the entry of the channel and then it drops off to a near constant value towards the end of the channel.

Figure 6-13. Layer movement velocity for polystyrene samples in a square channel
At this point, the data for each of the samples was examined individually. Figure 6-14 shows the layer movement velocity for just the A/D coextruded sample. In addition to the measured data from Figure 6-13, the results from section 3.3 for the viscous encapsulation results and the results from the elastic layer rearrangement studies in section 6.2 are also included.

In Figure 6-14, the points represent the data for the A/D coextruded sample taken from Figure 6-13. The solid line represents the viscous encapsulation results taken from section 3.3 for the flow of the A/D sample in a circular channel. The dashed line represents the elastic layer rearrangement results from section 6.2 for the flow of polystyrene in a square channel.

It can be seen in Figure 6-14 that the general trend observed in Figure 6-13 is followed by the combination of the solid and dashed lines. That trend was that the layer movement is at its maximum near the entry of the channel and then it drops off to a near constant value towards the end of the channel. This can be interpreted as the combination of the linear drop off in the viscous encapsulation velocity near the entry of the channel with the constant elastic layer rearrangement velocity near the end of the channel.

This plot implies that there is a mechanism change in what drives layer movement in a coextruded structure as it flows through a die channel. Near the entry to the channel, the viscous encapsulation forces are largest and they drive the interface deformation, if there is a large difference in the viscosity of the layers. Further down the channel when the driving force for viscous encapsulation is smaller, the effect of elastic layer rearrangement becomes more important and it continues to drive layer movement as the structure flows down the channel.

This mechanism also implies that in a small die, the viscous encapsulation effects will dominate and elastic effects can be ignored. However, as the die becomes larger, the flow path gets longer and then the elastic layer rearrangement forces are the dominant mechanism near the ends of the channel to cause layer deformation.

![Figure 6-14. Layer movement velocity for sample A/D in a square channel](image-url)
Figure 6-15 is similar to Figure 6-14 but it shows the velocity results comparison for the B/D coextruded sample rather than the A/D sample. Note that this figure also shows a drop in velocity near the beginning of the channel and then a region that is constant velocity further down the channel. The value of the constant velocity regime seems to agree more closely with the “elastic” line than did the A/D sample, but the transition point from the viscous to elastic seems to occur earlier than the lines would predict.

It should also be noted that the first measured data point is lower in velocity than the second point. This may be due to the fact that the elastic secondary flow velocity direction is in opposition to the viscous encapsulation flow velocity direction early in the channel and the final velocity of layer movement would be the vector sum of these two quantities. This phenomenon was not observed in the A/D sample but this may be because the relative difference in velocity magnitudes was so large because the viscosity difference between the two layers was large. This would imply that as the viscosity ratio approaches unity, both the magnitude and the direction of the individual velocity components become more important for calculating the layer movement velocity.

![Figure 6-15](image.png)

Figure 6-15. Layer movement velocity for sample B/D in a square channel

Figure 6-16 is similar to Figures 6-14 and 6-15 but it shows the velocity results comparison for the C/D coextruded sample. Once again this figure shows an initially higher layer movement velocity and then a constant value farther downstream which is the same trend observed in the other samples. However, since this sample has the lowest viscosity ratio (1.4) of the three samples, it appears that the elastic secondary flow velocity may have a larger impact in the magnitude of the layer movement velocity compared to the other samples early in the channel. This is shown by the fact that the first three velocity measurements are increasing slightly (rather than decreasing as the A/D sample did) before there is a drastic drop in the total velocity.
6.4 Conclusions

The data presented in this chapter show that the flow of a viscoelastic coextruded polymer structure through a non-radially symmetric channel is a very complex flow situation. The flow field is a complex function of the downstream velocity profile, the viscous encapsulation velocity, and the elastic secondary flow velocity. The data presented here show that the viscous encapsulation velocity is large early in the channel but decreases as the material flows down the channel. In contrast, the elastic secondary flow velocities were constant down the length of the flow channel. This implies that the viscous encapsulation forces will be dominant early in the channel but the elastic forces will become dominant in a longer channel. Once again this says that viscous encapsulation is the dominant mechanism in narrow dies while elastic layer rearrangement becomes more important as the die becomes wider.

The next chapter will discuss numerical simulation of these effects.
Chapter 7  Analysis of elastic layer rearrangement

7.1 Introduction

Experimental evidence of elastic layer rearrangement has been presented to show the effects of this phenomenon on the layer uniformity of coextruded structures. This chapter will deal with the numerical analysis and simulation of elastic layer rearrangement.

7.2 Numerical analysis

The momentum and continuity equations for the steady state flow of an incompressible viscoelastic fluid are given by

\[ -\nabla p + \nabla \cdot \mathbf{T} = 0 \]  \hspace{1cm} (7-1)
\[ \nabla \cdot \mathbf{v} = 0 \]  \hspace{1cm} (7-2)

where \( p \) is the pressure field, \( \mathbf{T} \) is the viscoelastic extra-stress tensor, and \( \mathbf{v} \) is the velocity field. Inertial and volume forces are assumed to be negligible.

The viscoelastic extra-stress tensor can be represented by \( \mathbf{T} \). If a discrete spectrum of \( N \) relaxation times is used then \( \mathbf{T} \) can be decomposed as follows:

\[ \mathbf{T} = \sum_{i=1}^{N} \mathbf{T}_i \]  \hspace{1cm} (7-3)

where \( \mathbf{T}_i \) is the contribution of the \( i \)-th relaxation time to the viscoelastic stress tensor. For the extra stress contributions \( \mathbf{T}_i \), a constitutive equation must be chosen. For example, the White-Metzner constitutive equation takes the form

\[ \mathbf{T}_i + \lambda_i \mathbf{v} \mathbf{D} = 2\eta_i \mathbf{D} \]  \hspace{1cm} (7-4)

where \( \lambda_i \) is the relaxation time, \( \eta_i \) is the partial viscosity factor, \( \mathbf{D} \) is rate of deformation tensor, and the symbol \( \nabla \) stands for the upper-convected time derivative operator. This constitutive equation predicts a zero second normal stress difference.

The rate of deformation tensor \( \mathbf{D} \) can be written as:

\[ \mathbf{D} = \frac{1}{2} \left( \nabla \mathbf{v} + \nabla \mathbf{v}^\top \right) \]  \hspace{1cm} (7-5)

A slightly more complex but more realistic viscoelastic equation is the Giesekus constitutive equation that has the form
where $\mathbf{I}$ is the unit tensor. In Equation 7-6, $\alpha_i$ are additional material parameters of the model, which control the ratio of the second to the first normal stress difference. In particular, for low shear rates, $\alpha_1 = -2N_2/N_1$, where $\alpha_1$ is associated with the highest relaxation times $\lambda_i$.

Another differential viscoelastic constitutive equation that can be used is the Phan Thien Tanner model, one form of which is given by

\[
\exp \left[ \frac{\epsilon \lambda_i \text{tr} (\mathbf{T}_i)}{\eta_i} \right] \mathbf{T}_i + \lambda_i \left[ (1 - \frac{\epsilon}{2}) \mathbf{T}_i + \frac{\epsilon}{2} \mathbf{T}_i \right] = 2\eta_i \mathbf{D}
\] (7-7)

where again $\lambda_i$ and $\eta_i$ are the relaxation time and the partial viscosity factor, respectively, while the symbol $\nabla$ stands for the upper-convected time derivative operator and $\dot{\mathbf{Q}}$ stands for the lower-convected time derivative operator. The parameters $\dot{\omega}_i$ and $\omega_i$ are material constants that relate to the extensional viscosity and second normal stress difference, respectively.

The finite element technique was used to solve the set of equations described. Depending on the number of relaxation times used, the computational resources required to solve the problem can be extremely large. When more than a single relaxation time was required, the elastic viscous split stress (EVSS) algorithm (75) was used. This method divides the viscoelastic extra stress tensor into elastic and viscous parts as follows:

\[
\mathbf{T}_i = \mathbf{S}_i + 2\eta_i \mathbf{D}
\] (7-8)

Using this algorithm, the equations expressed in terms of $\mathbf{S}_i$, $\mathbf{D}$, $\nu$, and $p$ can be solved. A postcalculation then gives the total viscoelastic extra-stress tensor $\mathbf{T}$. A quadratic interpolation is used for $\nu$, while a linear interpolation is used for $\mathbf{S}_i$, $\mathbf{D}$, and $p$.

### 7.3 Numerical results

In the initial simulations, a 2.5-D planar calculation was performed on a cross section normal to the main flow direction. In this technique, the pressure, the three velocity components and the components of the extra-stress tensor $\mathbf{T}_i$ depend on two spatial variables only. This implies that the calculated fields remain constant in the main flow direction with the exception of the pressure.

Typical finite element meshes for the square, rectangular, and teardrop channel geometries are shown in Figure 7-1. Note that in each geometry, the symmetry of the channel has been taken into consideration when developing the finite element mesh. For example, only one quadrant of the square, rectangular, and circular channels are meshed while one half of the teardrop channel is tessellated.
The set of partial differential equations for the flows in these channels requires boundary conditions along the channel walls and at the inlet. The boundary conditions used in these initial simulations were zero fluid velocity along the external walls (no slip condition) and axes of symmetry along the other edges. A constant flow rate was imposed at the entry to the channel. For the initial simulations, the constant flow rate produced an entry velocity of approximately 1 cm/s.

For the simulations, the viscometric properties of the three resins studied were used to develop the parameters for the viscoelastic constitutive equations. In the initial simulations, only a single relaxation time was used for each resin. These parameters were developed by fitting the viscometric data over a shear rate range of 10 to 100 s\(^{-1}\) since the average wall shear rate for a typical experiment was approximately 40 s\(^{-1}\). Table 7-1 shows the Giesekus parameters used for the polystyrene, polyethylene, and polycarbonate resins.

<table>
<thead>
<tr>
<th>Material</th>
<th>$\lambda$ (s)</th>
<th>$\eta$ (Poise)</th>
<th>$\alpha$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Polystyrene</td>
<td>0.06</td>
<td>80000</td>
<td>0.80</td>
</tr>
<tr>
<td>Polyethylene</td>
<td>0.16</td>
<td>26000</td>
<td>0.08</td>
</tr>
<tr>
<td>Polycarbonate</td>
<td>0.08</td>
<td>10000</td>
<td>0.001</td>
</tr>
</tbody>
</table>

Table 7-1. Material parameters for the Giesekus model with a single relaxation time
The numerical results for the simulation of a polystyrene melt flowing through a square channel are shown in Figure 7-2. These results show that there are two secondary flow vortices in each quadrant. These secondary flows circulate in a direction that would move material from the corners of the channel towards the center of the channel.

![Figure 7-2. Secondary flow patterns for an elastic material in a square channel](image)

These results were obtained using the Giesekus model. Very similar results were found when using the Phan Thien Tanner model. However, when using the White-Metzner model, no secondary flow patterns were predicted. Since the White-Metzner model predicts a zero second normal stress difference, this illustrates the importance of the second normal stress difference in producing secondary flows.

The simulations for the polystyrene resin using the Giesekus model produced a diagonal secondary flow velocity that was approximately 1% of the main flow direction velocity. This compares very favorably to the experimental measurements shown in Figure 6-5. When a similar simulation was run for the polycarbonate resin, a secondary flow velocity of approximately 0.001% of the main flow velocity was calculated. This demonstrates why no secondary flows were observed experimentally for the polycarbonate resin. These data imply that the die channel would have to be approximately 1000 times as long in order to produce similar interface deformation in a polycarbonate structure compared to a polystyrene structure.

When a simulation was performed using the polystyrene resin in the circular channel with the White-Metzner constitutive model, no secondary flows were seen. This was expected since the White-Metzner model predicts a zero second normal stress difference. However, when similar simulations were performed using the Giesekus and Phan Thien Tanner models, again no secondary flows were predicted even though these models predict non-zero second normal stress differences and the polystyrene resin modeled in the simulations is highly elastic. These simulations show that the secondary flows are only generated in non-radially symmetric geometries.
Figure 7-3 shows the simulation results for the polystyrene resin in a rectangular channel using a Giesekus model. Once again there are two secondary flow vortices in each quadrant similar to the square geometry but now they have different magnitudes and affect different portions of the channel. The strength of the vortices near the shorter walls is much smaller than those near the longer walls. While these vortices are smaller in magnitude, they also affect a smaller cross-sectional area than the larger vortices. It should also be noted that a large portion of the channel near the center is not substantially affected by the secondary flows. This implies that the overall layer interface deformation in a rectangular channel would be less than in a square channel.

![Secondary flow patterns for an elastic material in a rectangular channel](image)

Figure 7-3. Secondary flow patterns for an elastic material in a rectangular channel

Figure 7-4 shows the simulation results for the polystyrene resin in a teardrop channel using a Giesekus model. Recall that the teardrop shape channel approximates the geometry typically used in many die distribution manifolds. This figure shows that in the teardrop channel the number of secondary flow vortices has been reduced to six instead of the eight that were present in the square and rectangular channels. This figure also shows that the stronger secondary flows are along the longer channel dimension while the weaker secondary flows are along the shorter channel side, which is similar to the results from the rectangular channel.

![Secondary flow patterns for an elastic material in a teardrop channel](image)

Figure 7-4. Secondary flow patterns for an elastic material in a teardrop channel

Note that the direction of circulation of the secondary flows moves materials from the “corners” of the channel towards the center of the channel and the material from the center is moved towards the flat walls. This is similar to what was observed with both the square and rectangular channels. Since this geometry is one of the most used in die manifold designs, it is imperative to understand the effects secondary flows can produce when using this type of manifold design to process multilayer coextruded structures.
The simulation results from the four geometries studied (square, rectangle, teardrop, and circle) have shown that viscoelastic fluids produce secondary flows when they flow through non-radially symmetric geometries. This raises the question of what happens when a section of a channel contains a semicircular geometry but another section does not. This situation was investigated numerically by taking the square channel and adding a half circle to one side, as is shown in Figure 7-5. This geometry will be referred to as the “arch” geometry since it resembles an architectural archway that has been rotated ninety degrees. The dimensions of this arch geometry are similar to those of the square channel with an attached half circle.

![Figure 7-5. Development of the arch shaped geometry](image)

The finite element mesh used in the simulation of the arch geometry is shown in Figure 7-6. Once again the symmetry of the geometry has been considered and only the upper half of the geometry is actually modeled. The boundary conditions for this simulation are similar to those used previously, i.e., a no-slip condition is applied to the external walls while the centerline is considered to be an axis of symmetry allowing no flow across this boundary.

![Figure 7-6. Representative two-dimensional finite element mesh used for the arch shaped geometry](image)
Figure 7-7 shows the simulation results for the polystyrene resin in the arch channel using a Giesekus constitutive model. These results show that the arch channel also produces secondary flows in the channel. However, the simulation shows that this geometry produces six secondary flow vortices similar to the teardrop channel rather than the eight observed in the square channel. Comparison of these numerical predictions with experimental evidence will be presented in the next section.

Figure 7-7. Secondary flow patterns for an elastic material in an arch shaped channel

7.4 Comparison of experimental with numerical results

In this section, a comparison will be made between the numerical results shown in the previous section with the experimental results from the thirteen concentric ring studies. These comparisons will be qualitative in nature since the experiments produce actual final interface shapes while these numerical simulations only produce secondary flow patterns and not final interface shapes.

7.4.1 Numerical results using a single relaxation time

Figure 7-8 shows a comparison of numerical and experimental results for a polystyrene material in a square channel. This figure shows an excellent correlation between the predicted secondary flow patterns and the final interface shape for the thirteen concentric ring sample from the end of the square channel. Comparing the upper right quadrants, it can be seen that material flows along the diagonal from the corner towards the center of the channel. It can also be seen that material from near the center of the channel flows outwards towards the channel walls and then along the walls toward the corner. This flow pattern matches the secondary flow patterns predicted numerically.
Figure 7-8. Comparison of numerical and experimental results for flow patterns of a polystyrene material in a square channel

A comparison of the experimental and numerical results for the polystyrene resin in the rectangular channel is shown in Figure 7-9. Again the thirteen-layer concentric ring sample is used for the comparison. This figure shows that the final layer deformations agree well with the predicted secondary flow patterns. Note that the numerical prediction shows that material along the centerline of the channel will flow towards the center of the channel except when it is near the end walls in which case it flows away from the center of the channel. This flow pattern is indicated in the experimental sample as well by the black layer protrusions along the centerline near the end walls of the channel. This material is being stretched toward the center on one end and stretched toward the wall on the opposite end. The angle at which the material flows from the corners towards the channel centerline is also consistent between the experimental results and the numerical predictions.

Figure 7-9. Comparison of experimental and numerical results for flow patterns of a polystyrene material in a rectangular channel
Figure 7-10 shows a comparison between the numerical and experimental results for a polystyrene material in a teardrop channel. This comparison will also be done with the experimentally produced thirteen concentric ring polystyrene structure. This experimental sample clearly shows the six secondary flow vortices that were predicted by the numerical simulation. Starting with the tip of the teardrop (on the right in the figure), it can be seen that material has flowed along the walls towards the tip and is then redirected and flows along the centerline towards the center of the channel. Near the center of the channel, the central white ring has been elongated and stretched to very near the walls. The black ring that surrounds the central white ring shows how the material flows toward the walls and then splits and flows in opposite directions along the wall. On the left hand side of the channel, the material flows along the centerline towards the wall where it splits and flows in opposite directions towards the corners. When the flow reaches the corners, it then turns and flows back towards the centerline. Note that the angle at which the material flows back towards the centerline from the corner is in good agreement between the numerical and experimental results.

Figure 7-10. Comparison of numerical and experimental results for flow patterns of a polystyrene material in a teardrop channel

In the previous section, numerical simulations were performed on an arch shaped channel to determine the effect of rounding one side of a square channel. In order to determine how well these numerical simulations predicted the flow patterns, an experimental die channel was built containing an arch geometry. This die channel was run with a two-layer polystyrene structure and the results are shown in Figure 7-11. The image on the left side of the figure is from near the entry of the channel while the image on the right is from near the exit of the channel. This particular die channel was only half the length (~30 cm) of previous die channels so the sample from near the end of this channel would correspond to a position about half way down any of the previous channels.

Notice in Figure 7-11 that the interface between the layers near the entry is fairly flat and the layer ratio again corresponds to a twenty percent cap layer and an eighty percent substrate layer. By the time this structure reaches the end of the arch channel, a significant amount of layer interface deformation has occurred. In the sample near the end of the channel, a thin black layer is flowing up the wall in the square part of the channel (on the left). This same flow pattern is not seen in the portion of the channel with a rounded wall (on the right). This sample also
shows deformation of the interface near the center of the channel as it moves up towards the top of the channel.

Figure 7-11. Two-layer polystyrene structure near the entry and exit of the arch shaped channel

Figure 7-12 shows similar results for a thirteen concentric ring structure as was shown for the two-layer structure in Figure 7-11. This figure does, however, show more detail than the two-layer samples. Note specifically how the centermost black layer is deformed from a circular shape near the entry on the left to an almost rectangular shape near the end of the channel as is shown on the right. Also note how the layers are deformed in the corners of the channel due to the small secondary flows there.

Figure 7-12. Two-layer polystyrene structure near the entry and exit of the arch shaped channel

Figure 7-13 shows a comparison of the numerical and experimental results for a polystyrene material in an arch shaped channel. The numerical simulations predict that material should flow upwards from the center of the channel towards the top wall as is seen in the experimental results. The simulations also predict a small secondary flow near the left wall that should cause flow upward along that wall. This is the reason for the thin black layer observed in the experimental sample near the wall on the left. Near the rounded wall on the right, the simulations predict that material should flow down towards the centerline and then flow along the centerline towards the center of the channel. The experimental sample shows that material
has indeed flowed down to the centerline as was predicted. These samples and simulations show how strongly the flows are affected by the geometry. Just by rounding one side of a square channel, the flow changes significantly from side to side since material flows up the left side wall while it flows down the right side wall. Once again the simulations appear to predict the trends in the interface deformation very well.

Figure 7-13. Comparison of numerical and experimental results for flow patterns of a polystyrene material in an arch shaped channel

7.4.2 Numerical results using multiple relaxation times

In all of the previous simulations, only a single relaxation time was used in the simulations. At this point, it was decided to continue with the 2.5D simulations but use an increased number of relaxation times to see if the accuracy of the simulations could be improved.

For this part of the study, only the low-density polyethylene resin was simulated using the Giesekus constitutive equation. The polyethylene was chosen rather than the polystyrene resin because the polystyrene experimental results had shown very thin layers of polymer flowing along the channel walls while the polyethylene layers tended to be somewhat thicker. Trying to duplicate very thin layer flow near a wall for the polystyrene resin would have required very fine finite element meshes and made the computations more difficult.

Based on the dynamic rheological properties of the polyethylene resin at a temperature of 204°C, a discrete spectrum of five relaxation times \( \lambda_i \) ranging from \( 10^{-3} \) to 10 seconds was chosen. The corresponding partial viscosities \( \eta_i \) were fitted on the basis of the dynamic properties of the storage and loss moduli (\( G' \) and \( G'' \), respectively) while the \( \alpha_i \) parameters were selected based on the viscosity. The technique of identifying the material parameters is similar to that used by Laun for an integral viscoelastic model (76). Table 7-2 shows the values for the material properties used in the model.
Table 7-2. Material parameters for the Giesekus model with five relaxation times for low-density polyethylene

<table>
<thead>
<tr>
<th>i</th>
<th>$\lambda_i$ (s)</th>
<th>$\eta_i$ (Poise)</th>
<th>$\alpha_i$</th>
</tr>
</thead>
<tbody>
<tr>
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<td>$10^1$</td>
<td>17055</td>
<td>0.4</td>
</tr>
<tr>
<td>2</td>
<td>$10^9$</td>
<td>26680</td>
<td>0.5</td>
</tr>
<tr>
<td>3</td>
<td>$10^{-1}$</td>
<td>19011</td>
<td>0.6</td>
</tr>
<tr>
<td>4</td>
<td>$10^{-2}$</td>
<td>5242</td>
<td>0.7</td>
</tr>
<tr>
<td>5</td>
<td>$10^{-3}$</td>
<td>1790</td>
<td>0.8</td>
</tr>
</tbody>
</table>

The simulations performed were 2.5 D in nature, that is the calculation was performed in a cross section normal to the main flow direction and it was assumed that the calculated unknown fields did not vary along the main flow direction (except for the pressure gradient). In order to minimize the computational requirements, the elastic viscous split stress (EVSS) algorithm was used. Once again as in the previous simulations, boundary conditions of no slip at the walls and symmetry planes were used.

For these simulations, it was desired to be able to track the interface between the layers of a two-layer polyethylene melt rather than to just predict the secondary flow patterns. A two-layer system was chosen in order to simplify the problem since only one interface would have to be predicted.

Once the velocity field has been calculated for a cross section, it is possible to track the fluid particles and determine the fluid configuration at several cross sections along the main flow direction. In order to do this, the particle pathlines must be calculated. This was done using a technique similar to the one developed by Goublomme et. al. (77) for steady state flows that has been used for analysis of mixing. This technique uses a fourth order Runge-Kutta method to integrate the pathline in an element.

Pathlines are calculated for a large number of material points randomly distributed throughout the computational domain in the x-y plane at an initial downstream location of $z=0$. For these simulations, 1500 points were used. Each point was labeled with a color (black or white) depending on its location in the two-layer structure at $z=0$ and it maintains that color label as it follows the pathline. The pathlines of all of the particles are calculated until they reach a set downstream location, $z=z'$. Once these calculations have been performed, the fluid particle locations can be examined at any downstream distance between $z=0$ and $z=z'$.

When displaying the results of these simulations, only the white particles will be displayed against a black background. This will allow the interface between the white cap layer and the black substrate layer to be distinguished.

Figure 7-14 shows a comparison of experimental (top) and numerical (bottom) results for the flow of a two-layer polyethylene structure flowing down a square channel. In this figure, results are shown at cuts taken at 5.1, 17.8, 30.5, 43.2, and 50.8 centimeters (from left to right, respectively) downstream from the entry of the channel. The simulations were performed using
a total polymer flow rate of 8.6 kg/h, with the white cap layer representing 20% of the structure and the black substrate being 80% of the structure.

Note that the first experimental sample in the top row does not have a completely flat interface between the white cap layer and the black substrate layer. This is because it was taken at 5.1 centimeters downstream from the entry and so some interfacial distortion has already begun. In order to make a fair comparison between the experimental and numerical results, the location of the interface in the first simulation cut (on the left) was made to be identical to the experimental sample at 5.1 cm.

A comparison of the experimental and numerical results in Figure 7-14 shows very good agreement in the location of the layer interface. Both the experimental and numerical results show the distinctive secondary flow patterns described in the previous section for the flow of a viscoelastic material in a square channel. Both results show material flowing up the side walls of the channel and then turning back towards the center of the channel once they reach the corners of the channel. They also both show the characteristic upward flow of the material near the center of the interface. Considering the difficulty in producing these samples experimentally and the difficulty in numerically simulating this viscoelastic fluid, the agreement between the results is excellent.

Figure 7-14. Comparison of experimental and numerical results of the flow of a two-layer polyethylene structure flowing down a square channel

Figure 7-15 shows similar results as in Figure 7-14 but for the rectangular channel. The downstream distances in this figure are identical to those in Figure 7-14 (5.1, 17.8, 30.5, 43.2, and 50.8 cm from the entry with the sample taken nearest to the entry at the top of the figure while the sample taken nearest to the exit at the bottom of the figure). Once again the layer interface defined in the first numerical plane is matched to the experimental interface at 5.1 cm.

Figure 7-15 shows the excellent agreement between the experimental and numerical results for the flow of the polyethylene in a rectangular channel. The experimental results on the
left show how the interface deforms due to the secondary flow patterns described earlier. Note how the small secondary flow vortices in the corners cause material to flow up towards the corners while the larger recirculation vortices near the center of the channel cause the interface between the two layers to flatten out and be stretched horizontally while being moved upwards toward the top of the channel. The large secondary flow vortices also cause material to flow along the centerline towards the center of the channel, which pulls some of the white cap layer into the black substrate layer. All of these interface deformations are also seen in the numerical simulation results (on the right) as well.

There are some small differences between the experimental and numerical results for the flow in the rectangular channel. It appears that the numerical results predict more flow of the white cap layer along the centerline towards the center of the channel and more horizontal stretching of the black substrate layer near the upper wall. However, these differences are very small and subtle and overall the agreement between the experimental and numerical results appears to be excellent.

Figure 7-15. Comparison of experimental and numerical results of the flow of a two-layer polyethylene structure in a rectangular channel

Figure 7-16 shows a comparison of the experimental and numerical results of the flow of a two-layer polyethylene structure in a teardrop shaped channel. Recall that the teardrop shape was chosen because of its industrial importance in the design of die manifolds. Once again the cuts were taken at distances of 5.1, 17.8, 30.5, 43.2, and 50.8 cm from left to right. Also, the initial position of the interface in the numerical simulation was chosen to match the first experimental sample.
This figure shows that experimentally the interface undergoes significant deformation for this polyethylene resin. The black substrate material flows up along the left wall until it reaches the corner and then it turns back towards the centerline of the channel. The black substrate in the center of the channel is pushed upwards towards the top wall where it is stretched in both directions along that wall. The white cap layer near the tip of the teardrop flows down to the centerline and then it flows along the centerline to the left. All of these layer deformations match the secondary flow patterns predicted earlier.

A good correlation between the numerical simulation results and the experimental results is shown in Figure 7-16. This figure shows that the numerical simulations predict the flow of the substrate material into the corner and also up towards the top wall. Note also that the simulations predict the flow of the white cap material along the centerline in agreement with the experimental results. Overall, the agreement between the numerical and experimental results is very good.

![Figure 7-16. Comparison of experimental and numerical results of the flow of a two-layer polyethylene structure in a teardrop channel](image)

### 7.5 Full three dimensional calculations

In the previous simulations, a 2.5D formulation was used to predict the flow patterns. At this point, it was decided to develop a 3D simulation to see if the accuracy of the simulations could be improved even further.

For this part of the study, the low-density polyethylene resin was simulated using the Giesekus constitutive equation. Since a three dimensional formulation was used, only four relaxation times were used (compared the five relaxation times used in the 2.5D formulation) in order to use reasonable computational resources. Based on the dynamic rheological properties of this resin at a temperature of 204°C, a discrete spectrum of four relaxation times $\lambda_i$ ranging from $10^{-3}$ to 1 second was chosen. The corresponding partial viscosities $\eta_i$ were fitted on the basis of the dynamic properties of the storage and loss moduli ($G'$ and $G''$, respectively) while the $\alpha_i$ parameters were selected based on the viscosity. Table 7-3 shows the values for the material properties used in the model.
Table 7-3. Material parameters for the Giesekus model with four relaxation times for low-density polyethylene

<table>
<thead>
<tr>
<th>i</th>
<th>$\lambda_i$ (s)</th>
<th>$\eta_i$ (Poise)</th>
<th>$\alpha_i$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$10^4$</td>
<td>33711</td>
<td>0.5</td>
</tr>
<tr>
<td>2</td>
<td>$10^{-1}$</td>
<td>17345</td>
<td>0.6</td>
</tr>
<tr>
<td>3</td>
<td>$10^{-2}$</td>
<td>5467</td>
<td>0.7</td>
</tr>
<tr>
<td>4</td>
<td>$10^{-3}$</td>
<td>1752</td>
<td>0.8</td>
</tr>
</tbody>
</table>

The three dimensional finite element mesh developed for these simulations is shown in Figure 7-17. This mesh contains 1600 brick elements with 2106 vertices. Appropriate grading of the mesh is used near the channel walls to capture the flow gradients there.

Figure 7-17. Three-dimensional finite element mesh used for the square geometry

Since this is now a three-dimensional simulation, the boundary conditions must be applied to planes rather than lines. In view of the symmetry of this geometry, only one quarter of the flow domain has been used in the computations, as is shown in Figure 7-18. Boundary conditions of no slip at the walls along with suitable symmetry plane conditions were used. Fully developed viscoelastic extra stresses and velocity distributions were imposed at the inlet based on the specified flow rate. These were determined from the 2.5D simulations performed earlier. A fully developed velocity distribution was imposed at the outlet.

Figure 7-18. Boundary conditions for the three-dimensional finite element mesh used for the square geometry
In the 2.5D simulations, the interface location was determined by tracking the path of a large number of arbitrarily selected fluid particles. For the three dimensional simulations, a technique was used in which a pure advection equation was used for tracking the fluid motion. Let $c$ denote the advected variable that is constant along a given fluid trajectory. For a steady-state flow, the variable $c$ obeys the following equation:

$$\nu \cdot \nabla c = 0$$  \hspace{1cm} (7-9)

From a qualitative point of view, $c$ could be considered as the fluid color, white in the top layer and black in the bottom layer. The finite element integration of a pure advection equation is not trivial. An upwinding technique has been used along with the addition of an anisotropic diffusion of $c$ along the flow trajectories and no crosswind diffusion (78, 79). Knowledge of this variable allows tracking of any arbitrary material line or surface in the flow.

Figure 7-19 shows a comparison of experimental and numerical results of a two-layer polyethylene structure flowing down a square channel. The experimental results are shown in the upper half of the figure while the corresponding numerical results are shown in the lower half of the figure. The experimental results are similar to those shown previously in Figure 7-14 but the experiments were run at a slightly lower flowrate (7.9 kg/h vs. 8.6 kg/h). The numerical simulations are once again started with an interface location that matches the first experimental sample.

Comparison of the experimental and numerical results in Figure 7-19 shows that the numerical simulations predict the interface deformation trends well but there are significant differences. The simulations appear to predict the interface deformation in the early part of the channel fairly well but the differences between the two become more pronounced further down the channel. Near the exit of the channel, the simulations predict a greater thickness of material flowing along the vertical walls than is observed experimentally. Also the simulation underpredicts how far upwards the material near the center of the interface moves compared to the experimental results.

Figure 7-19. Comparison of experimental and numerical results of the flow of a two-layer polyethylene structure flowing down a square channel at a rate of 7.9 kg/h
Figure 7-20 also shows a comparison of experimental and numerical results of a two-layer polyethylene structure flowing down a square channel. The experimental results are similar to those shown previously in Figure 7-18 but the experiments were run at a lower flowrate (2.5 kg/h vs. 7.9 kg/h). The numerical simulations are once again started with an interface location that matches the first experimental sample.

Comparison of the experimental and numerical results in Figure 7-20 shows much the same results as were seen in Figure 7-19. Again the simulation tended to underpredict the amount of total interfacial deformation that took place. Comparing the experimental results in Figures 7-19 and 7-20 shows that a change in the total flow rate of more than a factor of three produced only small differences in the final interface location.

One of the strengths of the three-dimensional simulation is the ability to better view in three dimensions the actual deformation of the interface. This is shown graphically in Figure 7-21. This figure shows the deformation that the plane (or interface) between the top and bottom layer actually undergoes as the material flows down the channel.
It is interesting to note that the three-dimensional simulations appear to be less accurate in predicting the experimental results than the two and a half-dimensional simulations. This may be due to the fact that the number of relaxation times was reduced from five to four in order to make the computer simulation requirements more reasonable. It appears that using a more appropriate relaxation spectrum is more important than using a fully three-dimensional simulation in order to better predict experimental results for these particular simulations.

As was done with the straight square channel, simulations were also run for the tapered square channel. The tapered square channel starts out with the same dimensions as the straight square channel but it then tapers down to a square with one fourth the area of the entry, as was described earlier. The three-dimensional finite element mesh generated for the tapered square geometry is shown in Figure 7-22.

![Figure 7-22. Three-dimensional finite element mesh used for the tapered square geometry](image)

The boundary conditions used for the tapered square channel were similar to those used for the straight square channel. The total flow rate used in this simulation was 2.5 kg/h. The major difference in the boundary conditions between the tapered channel and the straight channel was at the outlet. The actual computational domain for the tapered channel does not allow the imposition of a developed velocity distribution at the exit because the flow is never fully developed. It is more convenient to relax the condition on the velocity at the exit by imposing a force that is derived from the developed flow conditions.

Figure 7-23 shows a comparison of experimental and numerical results of a two-layer polyethylene structure flowing down a tapered square channel. The experimental results are shown in the upper half of the figure while the corresponding numerical results are shown in the lower half of the figure. The experiments were run at a flowrate of 2.5 kg/h, which is the same as was run in some of the experiments in the straight square channel. In this figure, only samples at distances of 2.54, 27.9 and 53.3 centimeters downstream from the entry are shown. The numerical simulations are once again started with an interface location that matches the first experimental sample.

Comparing the experimental results in Figure 7-23 with those in Figure 7-20 shows that at the same total flow rate, the tapered channel produces a more distorted layer interface than does the straight square channel. This was expected since the tapered square channel produces
higher normal forces than does the straight square channel and so the forces driving the secondary flows are greater.

Comparing the results in Figure 7-23 shows good agreement between the experimental results and the numerical simulations. However, there are some differences in the trends that were observed in the tapered channel when compared to the straight channel. In the straight channel, the simulation underpredicted the upward movement of the interface near the middle of the channel while it overpredicted the thickness of the layers that moved up the channel walls. In the tapered channel, the simulation overpredicted both the upward movement of the interface near the middle of the channel and the thickness of the layers that moved up the channel walls. Overall, however, the agreement between the experimental results and the numerical simulations appears to be quite good.

As was shown previously in Figure 7-21, one of the strengths of the three-dimensional simulation is the ability to better view in three dimensions the actual deformation of the interface. This is shown graphically in Figure 7-24 for the tapered square channel. This figure shows the deformation that the plane (or interface) between the top and bottom layer actually undergoes as the material flows down the channel.
Figure 7-24. Numerical results showing the layer interface deformation of a two-layer polyethylene structure flowing down a square channel.

Comparison of the results shown in Figures 7-21 and 7-24 shows the real strength of the ability to model these flows in three dimensions. This comparison is shown in Figure 7-25. Note how much more deformation the interface undergoes in the tapered square channel compared to the straight square channel.

Figure 7-25. Comparison of numerical results showing the layer interface deformation of a two-layer polyethylene structure flowing down a straight and tapered square channel.
7.6 Conclusions

This chapter showed how the secondary flows in viscoelastic polymers could be simulated numerically. It was shown that viscoelastic models such as the Giesekus and Phan Thien Tanner models which predict non-zero second normal stress differences could simulate the secondary flows and thus layer movement while a model like the White-Metzner model could not predict the secondary flows because it predicts a zero second normal stress difference. Examples of single mode and multi-moded models were shown.

The next chapter will show how the secondary flows and layer movements can be modeled numerically using a mapping technique.
Chapter 8  Numerical analysis using a mapping method

8.1 Introduction

In the previous chapter, the finite element method was used to simulate the flow of viscoelastic polymers in different channel geometries and then different techniques were used to show the location of the coextruded interface. This chapter will describe an alternative technique that can be used to determine the interface locations, independently of the initial "color" distribution; the "mapping" method.

8.2 The mapping method

Several computational techniques can be used to track interfaces in fluid flows (80-84). These techniques can be separated into two broad categories: front capturing and front tracking. In the front capturing technique, massless markers are distributed within the fluid domain or a marker function is advected with the flow. The most difficult task within these approaches is to determine the location of the interface. Usually, it is recovered or “captured” using the calculated values of the marker function. These techniques were used and described in more detail in Chapter 7. The front tracking method on the other hand uses a separate moving mesh to describe the interface. Its location is accurately known at each time step. A disadvantage of both the front capturing and front tracking technique is that all computational work has to be repeated when the initial location of the interface changes. For passive interfaces, another method can be used to determine the evolution of the coextruded interface; the mapping method (85-88).

The mapping method is based on front tracking, but the main difference is that this technique does not track each material volume separately but instead creates a discretized mapping from a reference grid to a deformed grid. In the mapping method, the flow domain is divided into non-overlapping subdomains with boundaries. This subdivision is fully decoupled from any velocity field discretization. The boundaries of the subdomains are tracked (using front tracking) in a flow field from, for example \( t = t_0 \) to \( t = t_0 + \Delta t \) or from \( z = z_0 \) to \( z = z_0 + \Delta z \). Then the mapping from the initial grid to the deformed grid is constructed by determining the fraction of each cell in the deformed grid in the initial cells. The result is stored in a matrix \( \Phi \) with size \( n \times n \), where \( n \) is the number of cells in the grid. Using this technique, the interface location for coextruded structures with identical materials in each layer can easily be determined. The accuracy of the method depends on the accuracy of the velocity field and on \( n \) and \( \Delta t \).

One of the advantages of the mapping method over the front capturing and tracking method is the ability to deal with complex interfaces. Once the matrix \( \Phi \) is known, the advection of any initial color distribution, stored in a vector \( C_0 \) of length \( n \) is easily determined by the matrix vector multiplication providing the structure after \( \Delta z \), yielding \( C_1 \). The advection after \( 2 \Delta z \), \( C_2 \), is determined by multiplying \( \Phi \) with \( C_1 \), and so on for \( N \Delta z \). In this chapter the
mapping method is applied to determine the progression of the coextruded interface for the square and rectangular channel geometries.

8.3 Application of the mapping method

For this part of the study, only the polystyrene resin was simulated using the Giesekus constitutive equation. Based on the dynamic rheological properties of the polystyrene resin at a temperature of 204°C, a discrete spectrum of five relaxation times $\lambda_i$ ranging from $10^{-2}$ to $10^{2}$ seconds was chosen. The corresponding partial viscosities $\eta_i$ were fitted on the basis of the dynamic properties of the storage and loss moduli ($G'$ and $G''$, respectively) while the $\alpha_i$ parameters were selected based on the viscosity. Table 8-1 shows the values for the material properties used in the model. Two different volumetric flow rates were considered: a low flow rate of 0.72 cc/s and a high flow rate of 2.48 cc/s. The mapping technique was applied to the flow in the rectangular and square shaped channels.

<table>
<thead>
<tr>
<th>$i$</th>
<th>$\lambda_i$ (s)</th>
<th>$\eta_i$ (Poise)</th>
<th>$\alpha_i$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$10^{2}$</td>
<td>23740</td>
<td>0.86</td>
</tr>
<tr>
<td>2</td>
<td>$10^{1}$</td>
<td>97670</td>
<td>0.48</td>
</tr>
<tr>
<td>3</td>
<td>$10^{0}$</td>
<td>163500</td>
<td>0.56</td>
</tr>
<tr>
<td>4</td>
<td>$10^{-1}$</td>
<td>44640</td>
<td>0.53</td>
</tr>
<tr>
<td>5</td>
<td>$10^{-2}$</td>
<td>8540</td>
<td>0.51</td>
</tr>
</tbody>
</table>

Table 8-1. Material parameters for the Giesekus model with five relaxation times for polystyrene.

As was described in previous chapters, a finite element method was applied to determine the velocity field in the channels. For computational reasons a somewhat different method was used to determine the viscoelastic velocity field. The DEVSS/G-SUPG technique in combination with a theta-scheme was used to march in time to the steady state in the channel. Details of this method can be found in Bogaers (89). The finite element mesh consisted of 20 x 20 x 2 elements and the volumetric flow rate was prescribed via a LaGrange multiplier.

As an example of the mapping technique, Figure 8-1 shows the deformation of a coarse grid after 5 L/D and 10 L/D, where D is the width of the channel and L its length (see also Figure 8-2). The deformation of the grid follows the secondary flow field patterns described earlier in this thesis. Note that a comparison of a single cell in the three grids shows that the area of a cell is not preserved during deformation, but the flux through each cell is preserved. The actual computations are done on a much finer grid, i.e. 400 x 400 cells, and the mapping matrices created after 5 L/D and 10 L/D are combined to determine the evolution.
Figure 8-1. The figure on the left shows a coarse initial mapping grid consisting of 20 x 20 cells. The figures in the middle and the right show the deformation of the grid in (a) after 5 L/D and 10 L/D respectively. The actual mapping grid contains 400 x 400 cells.

Figure 8-2. Schematic figure showing the initial grid and the deformed grids after L/D = 5 and L/D = 10 in the channel.

8.3.1 Mapping results for the square channel

Results of the mapping operation on different initial layer configurations in the square channel are shown in Figures 8-3 (low flow rate) and 8-4 (high flow rate), respectively. These results were computed quickly (typically less than 1 CPU second on a standard PC) once the mapping matrix was determined, which is based on an accurate tracking of the velocity field. As was concluded from experimental results in previous chapters, the dependence of layer deformation on the flow rate is small. Furthermore, it can be concluded that by applying the mapping method, the details of the deformation patterns are accurately resolved.
Figure 8-3. Interface deformation evolution of six different initial (column wise) structures in a square channel. Each subsequent figure in a row shows the progression after a multiple of 5 L/D. The last column contains the extruded structure after 50 L/D. The flow rate was 0.72 cc/s.

Figure 8-4. Interface deformation evolution of six different initial (column wise) structures in a square channel. Each subsequent figure in a row shows the progression after a multiple of 5 L/D. The last column contains the extruded structure after 50 L/D. The flow rate was 2.48 cc/s.
8.3.2 Mapping results for the rectangular channel

Figure 8-5 shows the results of the layer interface deformation in the rectangular channel. The results can be compared with the experimental results shown in Figures 4-12, 5-28 and 5-35.

![Interface deformation evolution of three different initial (column wise) structures in the rectangular channel. Each subsequent figure shows the progression after a multiple of 5 L/D. The last column contains the extruded structure after 50 L/D. The flow rate was 2.48 cc/s.](image)

8.4 Comparison of experimental and computational results

Figure 8-6 shows experimental and numerical interface deformations for the progression of a 49-strand polystyrene structure as it flows down a square channel. Images (a) through (e) are experimental samples while images (f) through (j) are numerical predictions using the
mapping method. Images (a) and (f) are taken near the entry of the channel while images (e) and (j) are located near the exit of the square channel.

This figure shows excellent agreement between the experimental and numerical results. This is the most complex structure of the three examined since an interface location must be determined for each of the 49 strands as they flow down the channel. However, even though this is the most complex structure, it appears that the mapping method was able to predict the strand deformation very accurately at each location down the channel.

![Figure 8-6. Comparison of experimental and numerical results for the progression of a 49-strand polystyrene structure as it flows down a square channel. Images (a) through (e) are experimental samples while images (f) through (j) are numerical predictions using the mapping method. Images (a) and (f) are taken near the entry of the channel while images (e) and (j) are near the exit of the square channel.](image)

8.5 Influence of second normal stress difference

In order to determine whether the present experimental technique could, ultimately, be used as a measure of the second normal stress difference at high flow rates, a number of simulations were performed with different levels of viscoelasticity. The square channel geometry was chosen for these simulations since it produces strong secondary flow effects. For these simulations, the channel width and height were set to 1, the dimensionless flow rate was set to 1 as was the relaxation time. The level of viscoelasticity was changed by changing the non-linear $\alpha$ parameter in the single mode Giesekus model from 0.2 to 0.8 with steps of 0.2. The mapping matrices were computed for $L/D = 10$. Mapping was applied up to 20 times, yielding results up to $L/D = 200$. The initial (color) distribution was chosen such that the most reliable deformation patterns (from the corners to the center) were visualized.

The results of these simulations are shown in Figure 8-7. The pointed parts of the lines on the diagonals are numerical artifacts that could be solved by grid refinement. It can be concluded from numerical experiments like these that the relative level of viscoelasticity can
indeed be determined based on the amount of layer deformation caused by second normal stress difference driven flows. It is suggested by these results that (for this flow rate) a channel length of $50 < L/D < 100$ should be chosen to differentiate the level of viscoelasticity. Of course, quantification will require an iterative numerical and experimental approach.

Figure 8-7. Numerical simulations comparing layer rearrangement as a function of distance traveled down a square channel. The viscoelasticity was changed by changing the non-linear $\alpha$ parameter in the one mode Giesekus model from 0 to 0.8 with steps of 0.2

8.6 Conclusions

In this chapter, the mapping method was used to determine interface locations for coextruded polystyrene structures as they flowed through square and rectangular channels. Excellent agreement between the experimental and numerical results was obtained. These results show the power of the mapping technique in determining interface deformation in monolithic coextruded structures. Moreover, the simulation results suggest that the technique can be used to determine the second normal stress difference under realistic flow rates.
The next chapter will discuss the conclusions and make recommendations concerning this work on the viscoelastic flow of multilayer coextruded polymer structures.
Chapter 9  Conclusions and recommendations

9.1  Conclusions

There are several ways to join together layers of different polymers to produce a product with enhanced properties. The dominant method used to produce multilayer structures today is coextrusion. Coextrusion is the process of joining multiple layers of melt together in a die or feedblock to produce a multilayered structure. This process can be used to produce profiles, sheet, flat film, and blown film. By using specialized feedblocks and layer multiplication techniques, products with thousands of layers have been produced.

The coextrusion process has many advantages over other processes such as lamination because the joining of the layers and the formation of the final product is all done in one step. However, there are limitations. The two main difficulties that can arise when using the coextrusion process are interlayer instability and layer thickness non-uniformity.

Interlayer instability is a transient local layer thickness change that can be manifest in at least two ways: zig-zag and wave instabilities. Zig-zag instabilities are high frequency, random thickness changes that are typically correlated with a critical interfacial shear stress and occurs in the die lip region. Wave instabilities are characterized by parabolic waves that normally extend across the width of the film or sheet. It is believed to occur in the die prior to the lips and has been associated with polymer extensional viscosity properties.

Layer thickness non-uniformity generally refers to a more global process in which there is a general layer thickness gradient across the film or sheet. There are at least two processes that have been identified that can cause layer thickness non-uniformity: viscous encapsulation and elastic layer rearrangement. The first of the two processes, viscous encapsulation, occurs when a less viscous polymer encapsulates a more viscous polymer in a coextruded structure thereby producing a structure with a different layer thickness across the structure. The second process, elastic layer rearrangement, occurs when an elastic polymer flows through a non-radially symmetric channel producing secondary flows. These secondary flows will drive polymer flow in a prescribed motion across the flow channel that can lead to layer thickness non-uniformity in a coextruded structure.

Elastic layer rearrangement has been shown to occur in non-radially symmetric geometries but not to occur in a radially symmetric geometry. This phenomenon was observed in square, rectangular, and teardrop shaped channels as well as in a rectangular manifold channel. Elastic layer rearrangement has been produced using many styles of feedblocks that produce many different coextruded structures: planar layers, strands in a matrix, and concentric rings.

Experimental measurements of the velocities produced by viscous encapsulation and elastic layer rearrangement have been made and compared. Viscous encapsulation velocities were dependent on the viscosity difference between the two layers. These velocities are large at
the beginning of the channel but decrease toward zero farther down the channel as the less viscous material encapsulates the more viscous material. In contrast, the elastic layer rearrangement velocities measured were constant down the channel and were dependent on the channel geometry and the elasticity of the polymers. For the systems studied, the viscous encapsulation velocities were on the order of 10% of the downstream velocity at the channel entry while the elastic layer rearrangement velocities were on the order of 1% of the downstream velocity.

Experimental measurements were also made for elastic polymers with different viscosities in each layer flowing through a square channel. This would produce structures in which both viscous encapsulation and elastic layer rearrangement would occur. Since these two phenomena do not produce the same cross channel flows, the flow patterns can be aligned or opposed in different parts of the channel and thus produce a different final layer interface position. Another contributing factor to the final layer interface position is the fact that the viscous encapsulation velocity is high near the entry of the channel but decreases toward zero farther down the channel while the elastic layer rearrangement velocity is constant down the channel. This implies that viscous encapsulation is the dominant force early in the channel producing layer interface movement while the elastic layer rearrangement will be dominant further down the channel. This implies that viscous encapsulation will be the dominant mechanism in a short channel or narrow die while elastic layer rearrangement will become more important in longer channels or wider dies.

Numerical simulation of the elastic layer rearrangement flows has been done using finite element analysis. The polymers were modeled using differential viscoelastic constitutive equations. Only models that predicted second normal stress differences (such as the Giesekus and Phan Thien Tanner models) produced secondary flows that could lead to layer rearrangement. Models that did not predict a second normal stress difference (such as the White Metzner model) did not predict any secondary flows.

Simulations were done using single and multiple moded viscoelastic models in both 2.5 and 3 dimensional models for a polyethylene resin flowing through channels with different cross-sectional shapes. Different methods were evaluated and used to track the interface movement caused by the secondary motions produced by second normal stress differences. Comparison of the results of these numerical models with the experimental results showed good agreement. Using a higher number of time modes appeared to give better agreement with the experimental results at the cost of increased computational resources. The use of a three dimensional model gave an improved method for visualizing the flow in three dimensions. The mapping method appeared to give the best agreement with experimental results.

9.2 Recommendations

It has been shown that coextrusion is a powerful process for producing multilayered structures. However, layer deformation, whether from interfacial instability or layer non-uniformity, can be encountered if appropriate resins, equipment, and processing conditions are not used.
This work has shown that layer rearrangement due to viscoelasticity is a very important effect when working with large coextrusion dies. This type of layer rearrangement is manifested when coextruding elastic polymers through non-radially symmetric processing equipment. It is recommended, therefore, that when coextruding structures containing elastic materials, every effort be made to consider an appropriate geometry for the processing equipment flow channels to minimize this type of layer deformation. Since it is not practical to make all of the channels radially symmetric (round), it is recommended that die flow channels be rectangular in shape with a large aspect ratio (4 to 1 or greater for the ratio of height to thickness). It is also helpful to round the top of the rectangular channel so that there are no square corners in the channel. This type of geometry will not eliminate elastic layer rearrangement but it will help to minimize it.

It is also very important to keep the differences in viscosities between coextruded layers to a minimum. This will minimize the “viscous encapsulation” phenomenon and help maintain uniform layers. If this cannot be done, shaped feedslots can be used in the feedblock to attempt to overcome the viscosity mismatch and produce more uniform products.

Layer non-uniformity due to zig-zag layer instability can be controlled to a certain extent by controlling the process such that the critical interfacial shear stress is not exceeded. This can be done by increasing the die gap, lowering the total flow rate, decreasing the skin layer viscosity, increasing the skin layer thickness, or a combination of these parameters. Controlling wave type instabilities are more difficult but it appears that processing a polymer with a lower extensional viscosity minimizes this type of instability. It is also recommended that any spreading flows within the die or feedblock be gentle in order to minimize the extension rate and therefore the extensional viscosity.

The outlook for coextrusion technology is very encouraging. In the short time from the invention of this technology, coextrusion has spread into many markets and processes. From its beginnings in two layered structures through the development of microlayered structures with thousands of layers, this technology continues to expand in scope in both the commercial and scientific areas. Modeling of this process has reached a level of understanding that allows some inverse modeling techniques to be used to help design polymer processing equipment that will produce more uniform coextruded structures. The understanding of coextrusion technology and its capabilities and limitations is crucial for the polymer industry.
References


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Joe Dooley
April, 2002
Samenvatting

Meerlaagscoextrusie is een proces waarbij verschillende polymeren worden geextrudeerd en samengevoegd tot een meerlaagsproduct, typisch een geblazen of gegoten dunne film. Een uniforme laag dikte in het eindproduct is veelal vereist. In de praktijk worden vaak inhomogene laag diktes gevonden. Dit proefschrift beschrijft en onderzoekt een aantal oorzaken daarvan.

Inhomogene laag diktes kunnen worden veroorzaakt door grenslaaginstabiliteiten, door viskeuze incapsulatie en –verrassend genoeg- ook door viscoelastische effecten veroorzaakt door de secundaire stroming die wordt geïnduceerd door het tweede normaalspanningsverschil. Via een serie goedgeefundeerde experimenten en via numerieke analyses wordt vooral het laatste effect in dit proefschrift behandeld. Ter inleiding wordt echter eerst het tweede fenomeen van viskeuze incapsulatie nader onderzocht. Hier streeft het laag viskeuze polymer er naar het hoog viskeuze polymer in te bedden, te omhullen als het ware, omdat dit een voor transport gunstiger drukval geeft. Veelal gebeurt dit in de verdeelkanalen die zich bevinden in de uiteindelijk een dunne film opleverende vormgevende kop. Verschillende grades polystyreen met viscositeitverhoudingen tot maximaal 2.5 : 1 werden ge(co)extrudeerd door een cilindrisch kanaal. De gemeten incapsulatiesnelheid was maximaal bij binnenstroming van het kanaal en nam geleidelijk af, tegelijk met de daarvoor benodigde drijvende kracht. De gemeten maximale incapsulatiesnelheid bedroeg grofweg 15% van de stroomafwaartse snelheid.

Interessant genoeg bleek dat ook polymeren met dezelfde samenstelling en viscositeit inhomogene laag diktes voortbrachten. Dit fenomeen bleek te kunnen worden toegeschreven aan de secundaire stroming die in niet-cilindrische kanalen wordt gegenereerd door het tweede normaalspanningsverschil. Een typisch viscoelastisch effect dus, specifiek voor polymere smelten. Teneinde dit, vrij onbekende, effect te onderzoeken werden drie materialen, polystyreen, polyethyleen en polycarbonaat met afnemende viscoelasticiteit, geextrudeerd door een aantal geometrieën die gewoonlijk voorkomen in extrusiekoppen: een vierkant, een rechthoekig en een druppelvormig kanaal. Tenslotte werd ook nog een echt verdeelkanaal, waarbij zijdelingse lek plaatsvindt onderzocht. Ter visualisatie van de secundaire stroming werden ingangscondities aangeboden, waarin met ingekleurd materiaal verschillende stromen aan de kanalen werden aangevoerd. Deze werden gerealiseerd met speciaal ontworpen voedingsblokken. Voorbeelden van ingangsinformatie zijn (i) een tweelaagssysteem en een meerlaagsysteem bestaande uit 27, 165 en tenslotte 5000 in kleur alternerende (zwart-wit) horizontale lagen, (ii) een concentrisch systeem bestaande uit 2 en 13 in kleur alternerende concentrische lagen en (iii) een meerstrengensysteem waarin 49 zwarte strengen, verdeeld in een 7 bij 7 matrix, stromen in een verder witte matrix. Ook de snelheid van de viscoelastisch-geïnduceerde laag dikteverstoring kon worden bepaald. Hij bleek onafhankelijk van de positie in het kanaal en bedroeg ongeveer 0.3% van de stroomafwaartse snelheid (te vergelijken met de 15% van de maximale viskeuze incapsulatiesnelheid). Als conclusie blijkt dat viskeuze incapsulatie dominant is in korte
verdeelkanalen, terwijl viscoelastische secundaire stromingspatronen van belang worden bij langere kanalen.

Numerieke simulaties met Polyflow, met het Giesekusmodel voor de modellering van het tweede normaalspanningsverschil, kunnen in 2.5D en in 3D de secundaire stroming voorspellen en via particle tracking een beperkt aantal gevallen van gemeten grensvlakdeformatie nabootsen. Een kwantitatieve voorspelling van het gemeten gedrag in de onderzochte situaties bleek mogelijk.

Met een volledig 3D viscoelastische (isotherme) snelheidsberekening met een 5-modes Giesekusmodel in Sepran, in de drie gebruikte kanalen, vierkant, rechthoekig en druppelvormig, voor de twee in de experimenten gebruikte typische snelheden, konden voor een karakteristiek periodiek deel van de stationaire stroming deformatiematrixen worden bepaald, waarna de “Mapping Methode” kon worden toegepast. Deze voorspelt kwantitatief alle deformatiepatronen zoals in de experimenten gevonden.