

# Producing Microlayer Blown Film Structures Using Layer Multiplication and Unique Die Technology

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## Producing Microlayer Blown Film Structures Using Layer Multiplication and Unique Die Technology

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### Abstract

Many polymers are extruded through blown film dies to produce monolayer and multilayer films. The most popular style of die in use today to produce blown films is the spiral mandrel die. This type of die can be used effectively for many polymers in structures containing up to approximately 10 layers. This paper will discuss technology in which layer multiplication techniques are combined with unique die geometries to produce microlayer blown film structures with significantly greater numbers of layers.

### Introduction

Coextrusion is a common method used for producing multilayer blown films. Coextrusion is a process in which two or more polymers are extruded and joined together in a feedblock or die to form a single structure with multiple layers. This technique allows the processor to combine the desirable properties of multiple polymers into one structure with enhanced performance characteristics. The coextrusion process has been widely used to produce multilayer sheet, blown film, cast film, tubing, wire coating, and profiles [1-6].

Many polymers are extruded through blown film dies to produce monolayer and multilayer films. The most popular style of die in use today to produce blown films is the spiral mandrel die [7]. This type of die can be used effectively with many polymers to produce monolayer and multilayer blown films. However, this type of die normally cannot be used to produce microlayer blown films because the complex flow path in these dies can disrupt the continuity of the layers.

This paper will discuss a new, unique microlayer coextruded blown film technology in which a microlayer structure is formed using feedblock and layer multiplication technology and then processed through a unique blown film die to produce a microlayer blown film structure.

### Coextrusion

A breakthrough technology was originally developed by Schrenk and Chisholm to produce multilayer coextruded planar structures [8, 9]. In this technique, the layers are joined together in a device called a feedblock prior to the die. This layered structure is then processed through a single die manifold. This feedblock technique is shown conceptually in Figure 1.

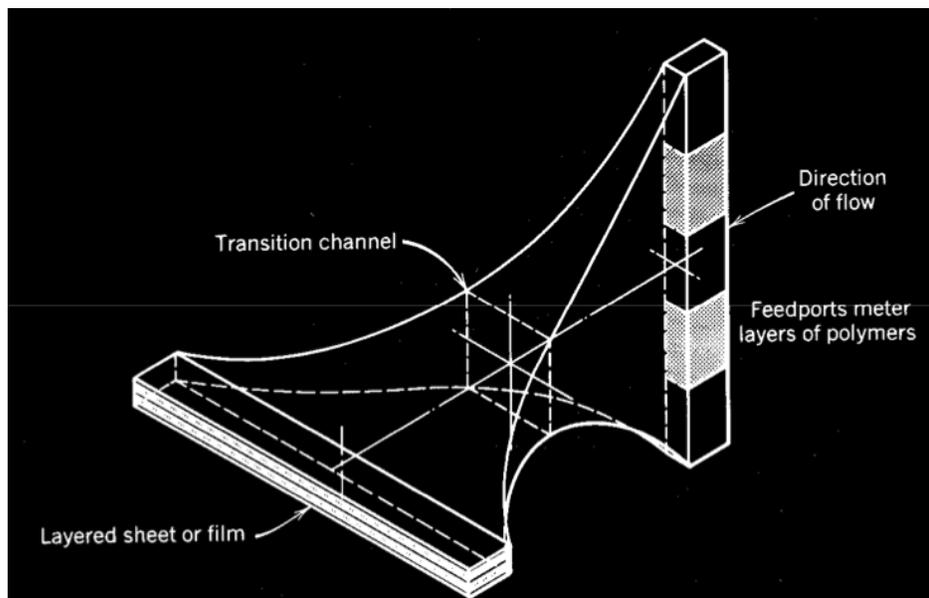


Figure 1. Feedblock coextrusion to make multilayer planar products.

Another method used to produce planar coextruded structures is with a multimanifold die. In this technology, planar layers are formed individually in separate die manifolds and then the layers are joined together before the exit of the die.

## Microlayer Coextrusion

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 that require hundreds or thousands of layers to produce unique properties [10-13]. Among these properties are enhancements in mechanical and optical properties.

In order to produce film or sheet structures with hundreds or thousands of layers, new techniques were developed to produce those structures. Standard feedblock techniques would not allow this many layers because of mechanical difficulties in joining so many layers. Methods to produce hundreds of layers were developed by Schrenk and co-workers [14-16]. A common technique is shown schematically in Figure 2.

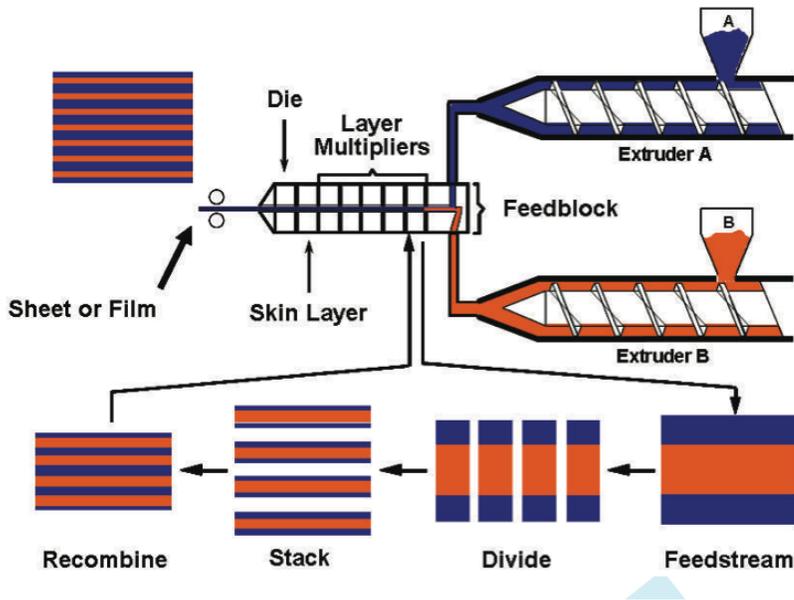


Figure 2. The microlayer coextrusion process.

Figure 2 shows how layers are joined together in a feedblock to form a three-layer structure. However, at the end of the feedblock, a layer multiplier is added that vertically divides the layers and then stacks and recombines the structure in order to increase the total number of layers. A series of layer multipliers can be added to significantly increase the number of layers in the final structure.

One important point to be made with this type of microlayer coextrusion is that this process was developed to produce flat sheet or cast film products. A large portion of the film industry uses the blown film process rather than the cast film process because of the biaxial orientation that is imparted to the film producing a product with more balanced properties. This process will be described in more detail in the following section.

## The Blown Film Process

A schematic diagram of the blown film process is shown in Figure 3. In this process, an extruder is used to melt and forward molten resin into an annular film die. Air is injected into the center of the annular die to inflate the polymer bubble. The bubble is cooled by an air ring that blows air on the surface of the bubble to lower its temperature until it becomes solidified. Above the die, a stabilizing cage may be used to minimize movement of the bubble as it is collapsed in the collapsing frame to make a flat film. This film is then pulled over idler rolls and fed into a film winder to make the finished film roll.

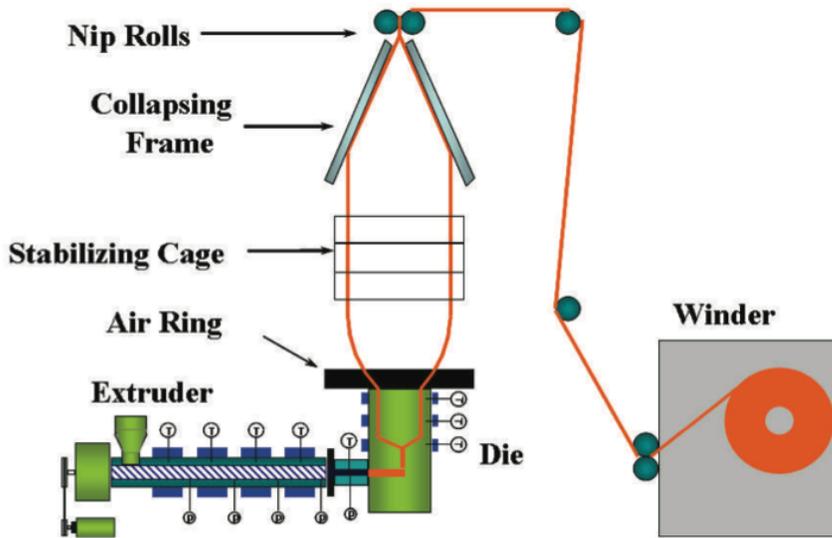


Figure 3. The blown film process.

A key part of this process is the blown film die. The blown film die takes the polymer melt from the extruder and shapes it into a tubular geometry to form the film bubble. This bubble must be uniform in thickness and temperature in order to form a uniform bubble. The different types of annular blown film dies that can be used in this process are described in the following section.

## Blown Film Dies

A schematic diagram of a spiral mandrel die is shown in Figure 4 [17]. In a spiral mandrel die, the cylindrical surface of the inner mandrel is spirally cut with grooves that become shallower as you progress down the channel. Since several spiral grooves are typically cut into a single manifold, there is flow down the channel grooves and also a leakage flow across the lands separating the grooves. This combination of flows produces a more uniform flow rate at the exit of the annular die and so a more uniform thickness in the resulting film. This type of design essentially eliminates the weld line difficulties seen in previous film dies. The application of this technology to the blown film industry was a significant breakthrough and spiral mandrel dies are now the dominant style of die used today in the blown film industry.

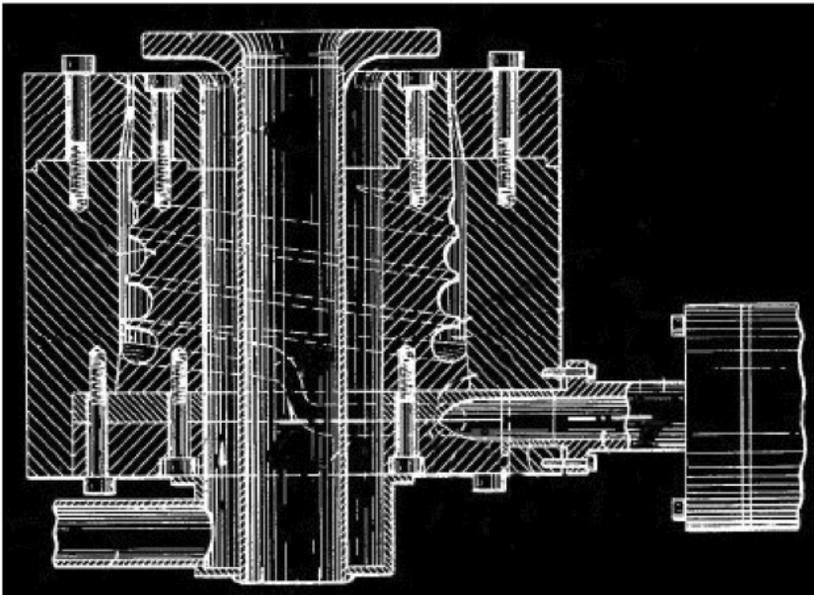


Figure 4. A single layer spiral mandrel die.

In order to make a multilayer blown film structure using spiral mandrel technology, a separate die manifold must be made for each layer. This concept is illustrated in Figure 5. Note in this figure how each layer is formed by a separate spiral mandrel manifold at a different radial distance from the center of the die. The individual annular flow streams are formed and then joined together near the exit of the die. If a multilayer feed stream was fed to the entry of this style of die, the layered structure could be destroyed as portions of the layers flowed down the spirals but other portions flowed across the land areas and over the next spiral. This combination of flows would produce an overall flow pattern that could

disrupt the continuity of the original layered melt fed to the die.

In this style of die, increasing the number of layers in the structure is accomplished by increasing the diameter of the die to make room for more spiral mandrel manifolds for each new layer. This tends to limit the number of layers that can practically be produced on this type of die because larger diameter dies have longer residence times and more surface area which can lead to degradation of the polymers being processed in the die.

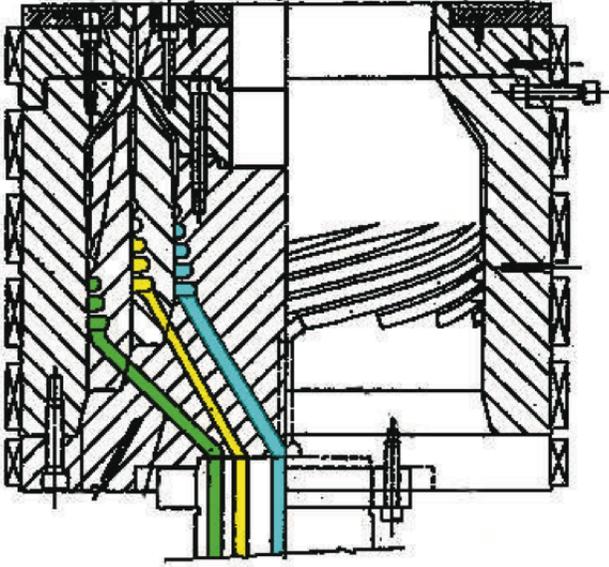


Figure 5. A multilayer spiral mandrel die.

Another style of spiral mandrel die has been developed in which the spiral channels are cut on the surface of a flat plate rather than on the surface of a cylinder. In this type of die, there can be multiple overlapping spirals cut into the same plate just as was done in the previous cylindrical spiral mandrel dies. By bolting two of these flat plates with matching grooves together, a spiral flow channel is created. Because of the dimensions of these large flat plates, these dies are sometimes referred to as "pancake" style dies.

One advantage of using a flat plate die for producing coextruded structures is the ability to stack plates on top of each other. This is shown in Figure 6. This figure shows a schematic diagram of multiple stacked plates bolted together in which each set of plates produces one layer. Each layer is added sequentially to the previous layer as the structure flows up the die towards the exit. This figure also shows a photograph of a commercial stacked plate die used to produce coextruded films.

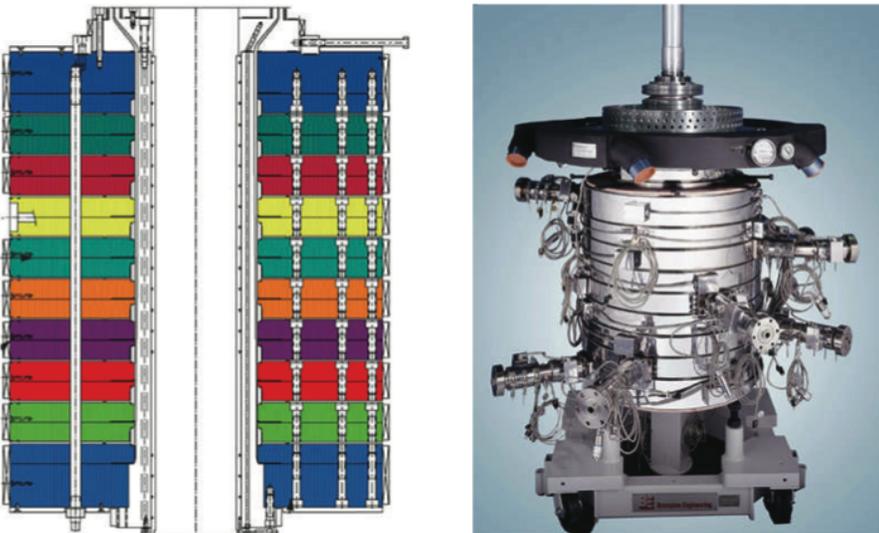


Figure 6. A multilayer stacked plate or "pancake" die.

This style of die is very versatile relative to changing the number of layers in a structure. By increasing or decreasing the number of plates stacked together, it is relatively easy to change the number of layers in the final film structure.

Structures containing up to 11 layers have been demonstrated using this type of die design. However, stacking more die plates to increase the number of layers beyond 11 layers becomes difficult due to pressure drop considerations and lack of space for more extruders.

## Microlayer Blown Film Concept

As discussed in the previous section, making multilayer blown films containing more than 11 layers is currently not feasible using existing technology. Technology has been developed by Schirmer [18, 19] in which thin annular disks are stacked together to form an annular structure with up to approximately 30 layers. This technology is similar to what is shown in Figure 6 but the plates are very thin and use a specific plate geometry and stacking to produce more layers. Making the plates thinner allows more layers to be added sequentially but a limit is reached when the pressure drop through all of the die plates becomes too large to process the structure. This process can also produce non-uniform layer thicknesses in the film.

The new concept for producing microlayers in a blown film is based on using a feedblock and layer multipliers in combination with a unique film die. This technique would overcome many of the pressure drop and residence time issues seen with the existing technology.

There are two main considerations to be taken into account when feeding a microlayer structure into a film die: (1) protecting the very thin microlayers as they flow from the feedblock to and through the die, and (2) the design of the geometry of the die to allow the layers to flow smoothly through the die while maintaining the microlayered structure.

Protecting the microlayered structure as it flows through the process can be done by encapsulating the microlayers with another layer. This is shown schematically in Figure 7. In this figure, an encapsulation die is shown schematically on the left producing a circular encapsulated structure on the right. This diagram shows a single core material being encapsulated by another layer but conceptually the single core material could be replaced by a microlayer structure.

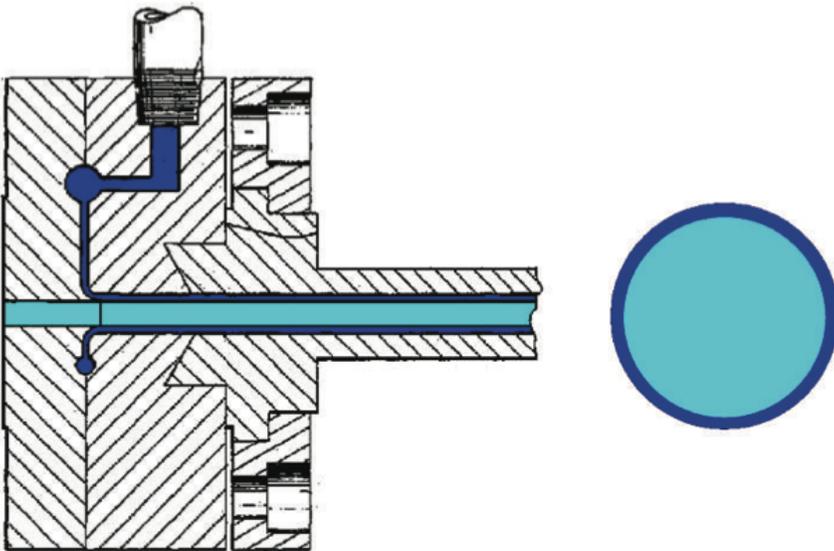


Figure 7. A schematic drawing showing an encapsulation die on the left producing an encapsulated structure on the right.

The next consideration is the geometry of the die to be used. As was discussed previously, a spiral mandrel type die would tend to disrupt the layered geometry fed to it and so this type of die geometry probably cannot be used for a uniformly microlayered structure.

Another type of die that could be used for this process is a crosshead style die. In this type of die, the flow is fed to the front side of the die where it is split such that half of the material flows around one side of the die while the other half flows around the other side of the die. These two flows then join together on the back side of the die forming a weld line region. Once rejoined, the material then flows annularly around the central mandrel.

Figure 8 shows two photographs of the front and back sides of a crosshead style die. The photograph on the left shows the front where the flow is split into two halves to flow around the central mandrel. The photograph on the right shows the back of the die where the two flow channels rejoin to form the annular structure. Note that at the point that the two flows join together, there is a place at which the two flows must "weld" back together to form a single flow stream. This area is sometimes referred to as the "weldline" area.



Figure 8. Photographs showing the front (left) and back (right) geometries of a crosshead style die.

The difficulty encountered with feeding a microlayer structure into a crosshead style die occurs in the weldline area. When the microlayer structure flows around the central mandrel and tries to rejoin in the weldline area, the microlayers do not form a continuous structure since there is a high probability that each individual microlayer will not align perfectly with the corresponding microlayer from the opposite side of the die which leads to the discontinuity of layers at the weldline. This difficulty can be overcome by using technology developed and patented for making coextruded blown barrier films [20-22].

The technology developed for making coextruded blown barrier film consisted of encapsulating the barrier polymer melt with a separate layer and then processing that through a crosshead style die in which the ends of the flow channels were overlapped. This overlapping of the layers in the weldline area produced a film structure with equivalent barrier properties in the weldline area compared to other areas of the film and eliminated the need to join the layers exactly together in the weldline area. Dies containing this type of technology are commercially available.

A diagram comparing an encapsulated melt in a standard crosshead die and a die in which the ends of the channels have been overlapped is shown in Figure 9. We have taken this technology and adapted it to produce microlayer blown films. This diagram shows a single core material being encapsulated by another layer but the single core material can be replaced by a microlayer structure and overlapped as well.

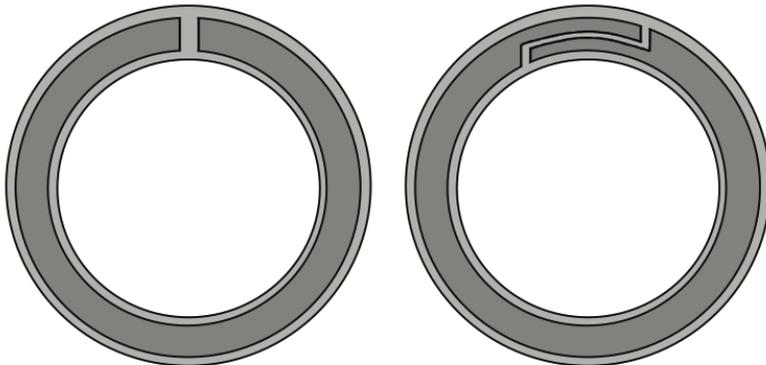


Figure 9. Comparison of an encapsulated melt in a standard crosshead die and a die in which the ends of the channels have been overlapped.

## Proof of Concept for Microlayer Blown Film

In order to validate the concept of using a feedblock, encapsulation, and a crosshead die with an overlap geometry to produce a microlayer blown film, a 7 inch diameter laboratory blown film line was modified.

### *Microlayer Blown Films Containing 30+ Layers*

As a first attempt to make a microlayer blown film, a laboratory blown film line was modified by (1) adding a feedblock to join together materials from two extruders for a core layer, (2) adding a layer multiplier to increase the number of core layers, and (3) adding an encapsulation extruder and feedblock to encapsulate the layered core structure. This equipment modification produced a microlayer core structure which was subsequently fed to a blown film die containing a crosshead style manifold with an overlap. With these line modifications in place, an experiment was carried out to try to make a microlayer blown film on this line. The first attempt consisted of processing a low density polyethylene (LDPE) resin, DOWTM LDPE 501 resin, manufactured by The Dow Chemical Company, in all the extruders. The final film structure would contain 27 layers from the feedblock/layer multiplier combination, 2 encapsulation layers (top and bottom), and 5

additional layers from the other 5 die manifolds for a total of 34 layers. A photograph of the film bubble with this structure as it is being produced is shown in Figure 10. This photograph is being taken from a vantage point that shows the overlap side of the die. Note that the film appears clear and uniform with no discernable optical difference between the main part of the bubble and the overlap area.

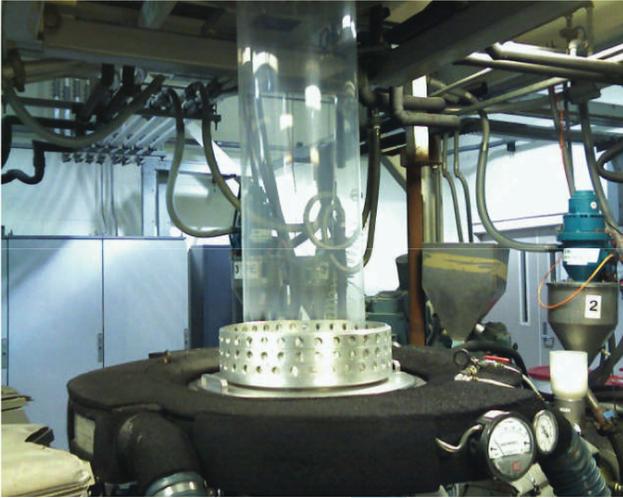


Figure 10. A microlayer blown film bubble containing 30+ layers.

Figure 11 shows a photomicrograph of a crosssection the microlayer blown film containing 30+ layers. The core microlayers are pigmented to make them more visible during microscopy. This sample showed that all of the microlayers were intact in the structure.

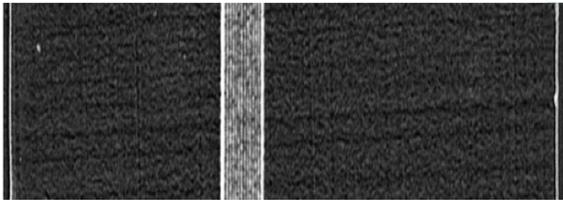


Figure 11. A photomicrograph of a microlayer blown film containing 30+ layers.

This photomicrograph shows that the microlayer core containing 27 layers was about 8% of the total thickness of the 2 mil film. This makes each microlayer approximately 0.006 mils or 0.15 microns thick.

A second set of microlayer blown films were produced with different materials in the core layers in order to improve our ability to differentiate between the layers during microscopy. These films had microlayer cores composed of alternating layers of DOW™ LDPE 501 resin and AFFINITY™ 1140 Polyolefin Plastomer resin, both manufactured by The Dow Chemical Company. The density difference between these resins allowed the use of Atomic Force Microscopy (AFM) to produce images showing the layered structure.

Figure 12 shows an AFM image of this microlayer blown film containing the LDPE and polyolefin plastomer resins with a thicker core. This core layer is approximately 25% of the total thickness compared to only 8% in the previous structure.

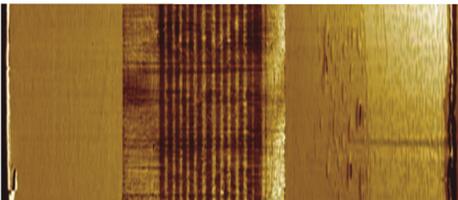


Figure 12. An AFM image of a microlayer blown film containing 30+ layers with a thicker microlayer core.

One important question that needed to be answered in these blown film trials was whether the microlayer structure would remain intact all the way around the film tube and in the overlap area. AFM images are shown in Figures 13 and 14 with expanded views of the core microlayers near the die entry region and near the die overlap for our microlayer film with a core composed of alternating layers of LDPE and the polyolefin plastomer resins. These images show the layers are intact near the die entry and remain so in the overlap area. Note in Figure 14 that the total number of core layers has doubled since we are now overlapping those layers on the back side of the die.

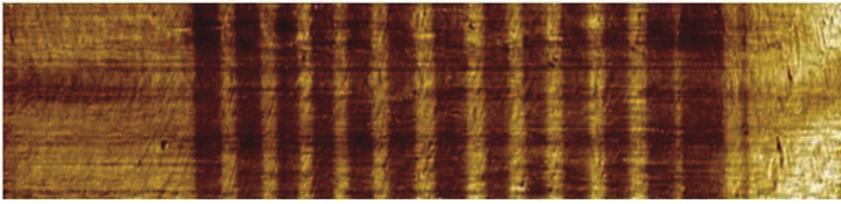


Figure 13. An AFM image showing an expanded view of the core microlayers near the die entry region in a microlayer blown film containing 30+ layers.

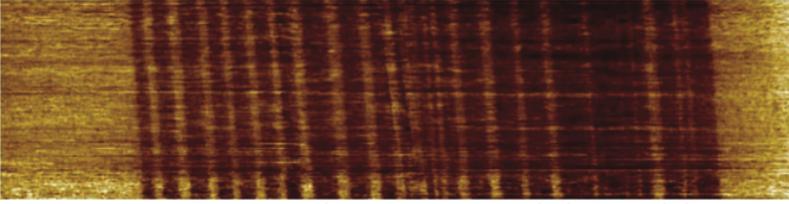


Figure 14. An AFM image showing an expanded view of the core microlayers near the die overlap region in a microlayer blown film containing 30+ layers.

Figure 15 plots the thickness of the 27 microlayers near the die entry in the core of the microlayer blown film described previously, but with a total overall thickness of 4 mils. Note that the thickness of the individual layers is fairly uniform and averages about 1 micron in thickness.

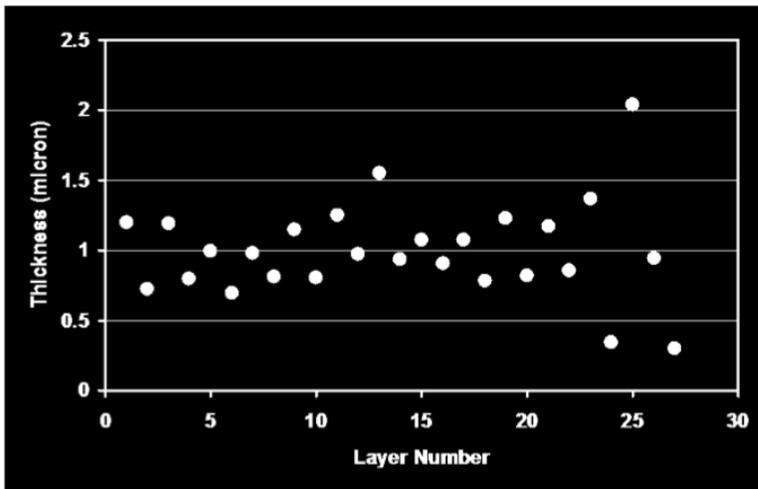


Figure 15. The layer thickness distribution for the core microlayers in a microlayer blown film containing 30+ layers.

*Microlayer Blown Films Containing 100+ Layers* Our next target in making microlayer blown films was to significantly increase the number of microlayers in the core of the film.

Using the modified line set-up, the 27 layers created previously were processed through a layer multiplier to make 108 core layers. These 108 core layers were then sent through the encapsulation die to coat the outside of the microlayer core structure. Figure 16 shows a sample taken from near the exit of the encapsulation feedblock showing the encapsulated 108 layer structure before it enters the die. This sample was taken to ensure that the layers were intact prior to their entry into the die. Note the four bands of black and white pigmented layers. These bands correspond to the channels in the layer multiplier. The surface layers in each band are white and so there are two white layers between each band producing a white layer with twice the thickness of the other layers.

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Figure 16. A photograph showing the encapsulated microlayer structure prior to the die entry.

With these new line modifications in place, an experiment was carried out to try to make a microlayer blown film on this line. Once again our first experiment consisted of processing low density polyethylene resin in all the extruders. The final film structure would contain 27 layers from the microlayer feedblock multiplied four times by the layer multiplier to produce 108 layers, 2 encapsulation layers (top and bottom), and 4 additional layers from the other 4 die manifolds for a total of 114 layers. A photograph of the film bubble with this structure as it is being produced is shown in Figure 17. This photograph is being taken from a vantage point that shows the overlap side of the die. Note that this 100+ layer film appears clear and uniform with no discernable optical difference between the main part of the bubble and the overlap area.

Figure 17. A microlayer blown film bubble containing 100+ layers.

Figure 18 shows an AFM image of a 100+ microlayer blown film containing LDPE and AFFINITY™ Polyolefin Plastomer resins. An important question that needed to be answered was whether the 100+ microlayer structure would remain intact in the film. The AFM image shown in Figure 18 is an expanded view of the core microlayers near the die entry region that shows the layers are intact near the die entry.

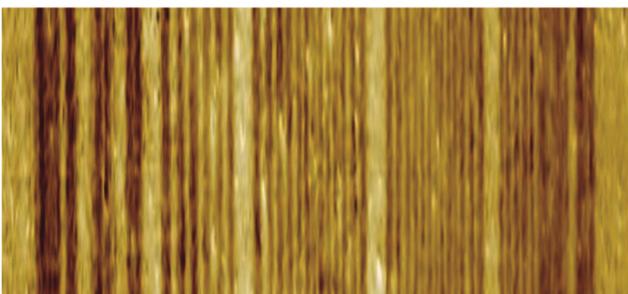


Figure 18. An AFM image showing an expanded view of the core microlayers near the die entry region in a microlayer blown film containing 100+ layers.

Just as Figure 18 shows an AFM image of the core microlayers near the entry region of our microlayer blown film containing 100+ layers, Figure 19 shows a Transmission Electron Microscopy (TEM) image of the core microlayers near the overlap region of the microlayer blown film containing 100+ layers.

Figure 19. A TEM image showing an expanded view of the core microlayers near the overlap region in a microlayer blown film containing 100+ layers.

Figures 18 and 19 show the 100+ layers in our microlayer blown film are intact near the die entry and remain so in the overlap area. Note in Figure 19 that the total number of core layers has doubled since we are now overlapping those layers on the back side of the die and so this figure actually shows more than 200 microlayers in the overlap area.

## Conclusions

A new, unique microlayer coextruded blown film technology has been developed in which a microlayer structure is formed using feedblock and layer multiplication technology and then processed through a unique blown film die to produce an annular blown film structure. This technology has been used to produce microlayer blown films with over 100 layers. A patent case has been filed covering this new processing technology.

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**Key Words:** Microlayer, Coextrusion, Blown Film, Dies, Spiral Mandrel, Crosshead, Feedblock, Layer Multiplication.

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