# 1997 Best Paper - Interfacial Instabilities during Coextrusion of LDPEs

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# Interfacial Instabilities during Coextrusion of LDPEs

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## Abstract

Coextrusion experiments were performed using well characterized LDPE resins in an effort to gain a better understanding of interfacial instability phenomena. The resins used were chosen carefully and included materials of high and low viscosity as well as broad and narrow molecular weight distributions (MWD). The experiments involved the coextrusion of either the same material in both layers or various combinations of the four materials and the focus of the work was to elucidate the effects of flow rates, molecular weight (MW) and MWD on interfacial instability. The effect of the geometry at the point where the materials merged was also investigated.

## Introduction

Interfacial instabilities occurring during coextrusion of polyethylene resins is a very crucial problem that has been considerably investigated over the years [1-8]. There are several theories as to the source of these interfacial instabilities. One possibility is that the interfacial instability begins at the point where the materials combine and the interface is created. Another theory is that the interfacial instability is created just prior to the die exit region where the interface usually experiences a maximum shear stress. Another possibility could be that the layer ratios of the materials is such that the flow is inherently unstable and given sufficient time, an interfacial instability can develop.

This paper presents the results of an experimental investigation that was performed in order to obtain a better understanding of the interfacial instability phenomena that may occur during coextrusion. The basic purpose of the experiment was to distinguish between the effects caused by different polymer characteristics and their relationship to the geometry of the flow field.

## Experimental

### Equipment

Experiments were performed on a Brampton Engineering coextrusion line that was equipped with two 45 mm diameter, 24:1 single screw extruders. The screws had a compression ratio of 2.8:1 and were equipped with fluted mixing sections. The extruders were equipped with gravimetric feeders to control the mass flow rate of material through each layer.

The experiments were performed using an 76 mm diameter annular coextrusion die with either a narrow or a wide exit gap. Figure 1 shows the narrow exit gap configuration in which layer ‘A’ flows upward to merge with layer B into a coextrusion area with a 6.35 mm gap and the coextrusion flow channel then converges to a 1.52 mm gap for the last 15 mm of the die exit. In the wide die gap configuration shown in Figure 2, the coextrusion flow channel gap remains at 6.35 mm right up to the die exit. Prior to combining, the polymer is distributed into a uniform annular flow using a flat spiral distribution system.

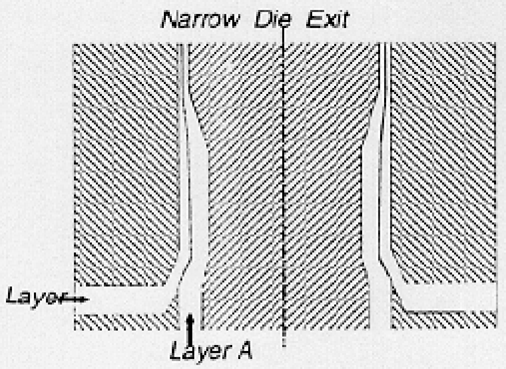


**Figure 1** Narrow die gap geometry

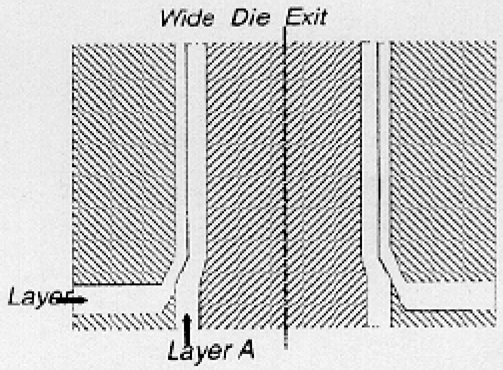


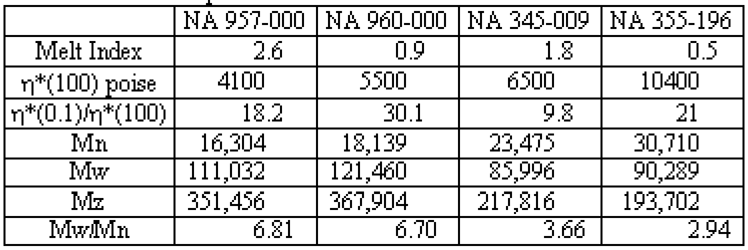
**Figure 2** Wide die gap geometry

Figures 1 and 2

**Materials**

The materials were judiciously chosen in an attempt to distinguish the difference between the effect s of MW and MWD. Four resins were chosen such that the a factorial type of experiment could be set-up. The commercially available materials used in this study were supplied by Quantum and their complex viscosity and molecular weight parameter values are summarized in Table 1. Resins NA957 and NA960 had a relatively broad MWD while NA345 and NA355 had a relatively narrow MWD. Each resin pair consisted of a relatively high viscosity polymer, NA 960 and NA 355 respectively, and a relative low viscosity polymer, NA 957 and NA 345.

Table 1 Material Properties



| | NA 957-000 | NA 960-000 | NA 345-009 | NA 355-196 |
|---|---|---|---|---|
| Melt Index | 2.6 | 0.9 | 1.8 | 0.5 |
| $\eta^*(100)$ poise | 4100 | 5500 | 6500 | 10400 |
| $\eta^*(0.1)/\eta^*(100)$ | 18.2 | 30.1 | 9.8 | 21 |
| Mn | 16,304 | 18,139 | 23,475 | 30,710 |
| Mw | 111,032 | 121,460 | 85,996 | 90,289 |
| Mz | 351,456 | 367,904 | 217,816 | 193,702 |
| Mw/Mn | 6.81 | 6.70 | 3.66 | 2.94 |

Table 1

**Procedures**

Using the narrow die gap geometry, the line was started- up and allowed to reach steady-state at a rate of 22.7 kg/hr and an A:B flow ratio of 20:80. The throughput rate was chosen such that the bubble stability could be maintained through all runs. The initial flow ratio of 20:80 was used because it showed no signs of interfacial instability. The two layers are designated as layers ‘A’ and ‘B’ as shown in Figure 1. Upon exiting the die, the coextruded annular sample was cooled with the air ring while it was pulled upward by the nips. This process necessitated finding conditions under which a stable ‘bubble’ could be maintained while making samples which best exhibited instabilities. It was determined that the interfacial instability was more readily observable in relatively thick (250 - 300 micron) film samples.

There were essentially 3 sets of experiments with 2 flow geometries in each set. In the first set of experiments, layer ‘A’ was the minor flow component while in the second and third set layer ‘A’ was the major flow component (i.e. A:B of 80:20). In addition, the first and second set of experiments always had the same material in both layers. Only in the third set of experiments, different materials were coextruded together. Once the line reached steady state, a sample was collected at this flow ratio. The flow ratio was then changed to 18/82 (still maintaining the 22.7 kg/hr output) and a sample was collected. This was repeated down to a layer thickness at which the interfacial instability was clearly visible. The processing temperature was then increased and the experiment was repeated for the same material. The above procedure was repeated for all of the materials. The die gap was then changed to 6.35mm and the experiment was repeated for each material but only at one of the temperatures used in the narrow die gap experiments.

The procedure for the second set of experiments was essentially identical to the first set but with the flows of the extruders reversed so that the B layer was the minor component. In addition, a small amount (about .5-1%) of carbon black was added to the minor component layer in order to facilitate the viewing of any instability. Finally, a similar procedure was followed for the third set of experiments in which combinations of materials were coextruded.

## Observations

The results of the above experiments are summarized in Tables 2 to 4 . In the tables, 'S' and 'U' are used to denote a stable and an unstable interface respectively. The 'SU' has been used to denote the indeterminate transition area from stable to unstable. The subscripts 'w' and 'z' indicate that a 'wave' and/or a 'zig-zag' instability is observed, respectively. In these experiments, two distinct type of instabilities are observed. These have been termed 'wave' and 'zig-zag' as introduced by some previous and parallel work performed by researchers at DOW Midland on some flat dies [9-11]. The 'zig-zag' instability is of relative high frequency (about 10-20 Hz in this experiment) and of low amplitude and has an appearance similar to the melt fracture phenomenon. The 'wave' instability has a much lower frequency (about 0.2 to 0.5 Hz in this experiment) than the 'zig-zag' instability and also has a higher amplitude. Of course both the 'wave' and the 'zig-zag' instability could appear concurrently in the same sample.

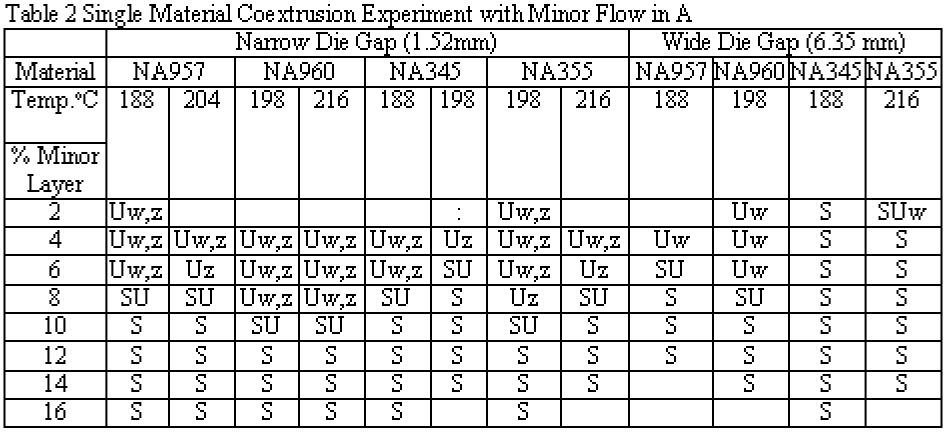


Table 2 Single Material Coextrusion Experiment with Minor Flow in A

| | Narrow Die Gap (1.52mm) | | | | | | | | Wide Die Gap (6.35 mm) | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Material | NA957 | | NA960 | | NA345 | | NA355 | | NA957 | NA960 | NA345 | NA355 |
| Temp.°C | 188 | 204 | 198 | 216 | 188 | 198 | 198 | 216 | 188 | 198 | 188 | 216 |
| % Minor Layer | | | | | | | | | | | | |
| 2 | Uw,z | | | | | : | Uw,z | | | Uw | S | SUw |
| 4 | Uw,z | Uw,z | Uw,z | Uw,z | Uw,z | Uz | Uw,z | Uw,z | Uw | Uw | S | S |
| 6 | Uw,z | Uz | Uw,z | Uw,z | Uw,z | SU | Uw,z | Uz | SU | Uw | S | S |
| 8 | SU | SU | Uw,z | Uw,z | SU | S | Uz | SU | S | SU | S | S |
| 10 | S | S | SU | SU | S | S | SU | S | S | S | S | S |
| 12 | S | S | S | S | S | S | S | S | S | S | S | S |
| 14 | S | S | S | S | S | S | S | S | | S | S | S |
| 16 | S | S | S | S | S | | S | | | | S | |

Table 2

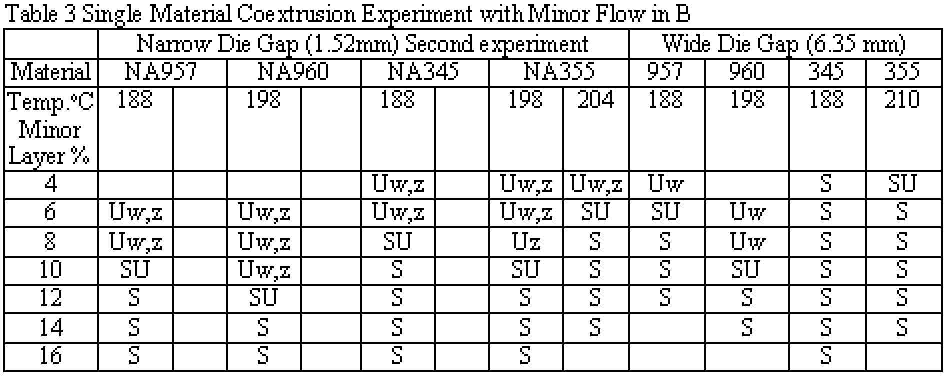


Table 3 Single Material Coextrusion Experiment with Minor Flow in B

| | Narrow Die Gap (1.52mm) Second experiment | | | | | | | | Wide Die Gap (6.35 mm) | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Material | NA957 | | NA960 | | NA345 | | NA355 | | 957 | 960 | 345 | 355 |
| Temp.°C Minor Layer % | 188 | | 198 | | 188 | | 198 | 204 | 188 | 198 | 188 | 210 |
| 4 | | | | | Uw,z | | Uw,z | Uw,z | Uw | | S | SU |
| 6 | Uw,z | | Uw,z | | Uw,z | | Uw,z | SU | SU | Uw | S | S |
| 8 | Uw,z | | Uw,z | | SU | | Uz | S | S | Uw | S | S |
| 10 | SU | | Uw,z | | S | | SU | S | S | SU | S | S |
| 12 | S | | SU | | S | | S | S | S | S | S | S |
| 14 | S | | S | | S | | S | S | | S | S | S |
| 16 | S | | S | | S | | S | | | | S | |

Table 3

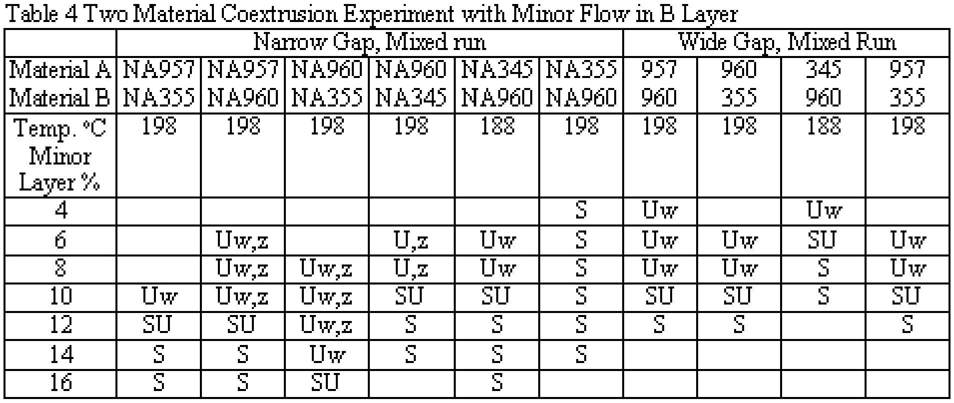


Table 4 Two Material Coextrusion Experiment with Minor Flow in B Layer

| | Narrow Gap, Mixed run | | | | | | Wide Gap, Mixed Run | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| Material A<br>Material B | NA957<br>NA355 | NA957<br>NA960 | NA960<br>NA355 | NA960<br>NA345 | NA345<br>NA960 | NA355<br>NA960 | 957<br>960 | 960<br>355 | 345<br>960 | 957<br>355 |
| Temp. °C Minor Layer % | 198 | 198 | 198 | 198 | 188 | 198 | 198 | 198 | 188 | 198 |
| 4 | | | | | | S | Uw | | Uw | |
| 6 | | Uw,z | | U,z | Uw | S | Uw | Uw | SU | Uw |
| 8 | | Uw,z | Uw,z | U,z | Uw | S | Uw | Uw | S | Uw |
| 10 | Uw | Uw,z | Uw,z | SU | SU | S | SU | SU | S | SU |
| 12 | SU | SU | Uw,z | S | S | S | S | S | | S |
| 14 | S | S | Uw | S | S | S | | | | |
| 16 | S | S | SU | | S | | | | | |

Table 4

# Discussion

Consider first the results in Tables 2 and 3 in which both layers are of the same material. All of the materials showed a 'zig-zag' interfacial instability when using the narrow die gap geometry although not at the same flow rate conditions. When the larger die gap was used, the 'zig-zag' interfacial instability was eliminated for all of the flow ratios used in this experiment. However, the wave instability was now easier to observe in some of the samples.

More specifically, all interfacial instabilities were essentially eliminated for the NA345 and NA355 materials with the exception of the condition when the minor, NA 355, layer is very thin. With the NA957 and NA960 materials, increasing the die gap had only a small effect on the layer ratio for which the interfacial instability appeared. Another interesting observation is that the interfacial instabilities (particularly the wave instability) occurred at approximately the same flow ratio in the first and second set of experiments. The geometry of the channels was originally designed for the B layer to be the minor component. In this mode of operation, the average velocities of the materials prior to and after combining were similar. It was rather surprising to see that operating the system in the reverse condition affected marginally the layer ratio at which the instability was observed.

For the third set of experiments (Table 4), in which different materials were coextruded together, similar results were obtained in that the 'zig-zag' instability was not observed with the wide die gap geometry. The layer ratio at which the interfacial instability was observed was dependent on the materials in each layer. For certain combinations, there was no observable interfacial instability for any flow ratio while other combinations showed flow instabilities at relatively high flows of the minor component when compared with the previous set of experiments. One of the most interesting observations in this set of experiments was made when the NA960 was coextruded with the NA345 (columns 5 and 6 in table 4. When the NA345 was the minor component, it exhibited a 'zig zag' instability at about 8 - 10 % but no 'wave' instability. However, when the NA960 was the minor component, only a 'wave' instability was observed starting at about the same flow ratio. This would support the theory that the two instabilities are somewhat independent of each other and should be investigated accordingly.

The observations made in this work indicate that the 'wave' type of interfacial instability occurs when the interface moves closer to the wall, however, there is still no explanation as to the reason for the interface to go unstable. What is the driving force? It stands to reason that the initial perturbation to the flow field must occur at the merge point of the two layers. Since the observations also indicate that the 'wave' type interfacial instability occurs more readily in broad MWD materials, it also stands to reason that the interfacial instability is influenced by the elastic properties of the materials only. However, since the extrusion of the same materials results in an instability, it is unlikely that the instability is due to elastic differences in the materials. It is more likely that the instability is due to the total deformation of the flow stream. This was investigated by comparing the average velocity of the individual layers before and after combining. A velocity ratio was calculated as follows:

Velocity ratio = Average Layer Velocity Prior to Combining / Average Layer Velocity After Combining

This was determined for the various flow rate ratios used in the experiments and plotted in Figures 3 and 4.

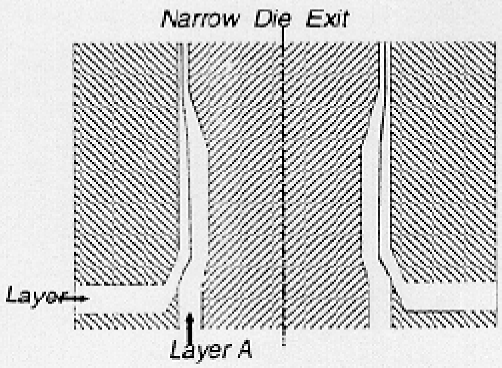


Figure 1 Narrow die gap geometry

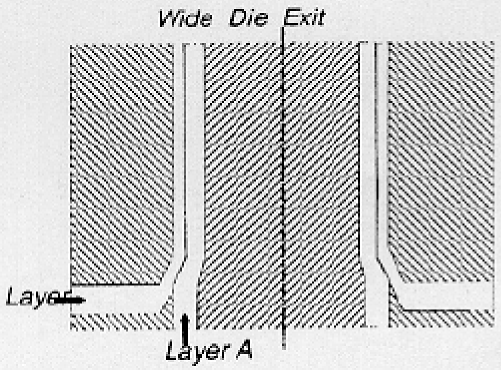


Figure 2 Wide die gap geometry

Figures 3 and 4

Figure 3 relates to the first set of experiments where the large channel (A) transported the minor component while Figure 4 represents the experiment in which the large channel (A) transported the major component. In both figures the same material, NA960, is used each layer. The graphs shows that the velocity ratio of the minor component increase dramatically as the percentage of the minor component gets smaller. In other words, the acceleration of the minor layer ,at the merge point, increases as the layer becomes thinner. However, there is a marked difference in velocity ratio at which the interfacial instability occurs for the first two sets of experiments. This difference is believed to be due to the orientation of the minor layer. In the first set of experiments the minor layer is always moving in the axial direction while in the second set of experiments, the minor layer is forced to turn from the radial direction to the axial direction. This results in an additional acceleration effect that is not accounted for in the simple calculation presented above. Regardless of the actual values, the minor layer does experience an increased acceleration or elongation as it becomes thinner. If the material is elongated too much at the merge point it will try and resist this deformation once it joins the other material. Whether it succeeds or not will depend on the relative viscoleastic properties of each stream. If the major layer has a higher resistance to deformation than the minor layer, than the flow field will be more stable than if the reverse situation were to exist. This of course is something that has been known for years by experienced coextrusion practitioners. What still remains to be determined is a method of quantifying the phenomenon. It is hoped that the data presented here will help all of us get closer to that goal.

## Conclusions

The experiments performed in this study indicate broad MWD materials have a greater tendency to exhibit interfacial instability and that these interfacial instabilities are more due to layer ratio than processing conditions or die geometries. In contrast, materials with narrower MWDs tend to exhibit an interfacial instability which appears to be related to the stress at the interface and hence can be affected by whatever affects the interfacial stress. It is believed that the origin of the ‘wave’ type of interfacial instability is due to an extreme deformation of the minor layer at the merge point and that the viscoelastic properties of adjacent layers determine the instability development.

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