The Influence of Using Fluoropolymer Processing Additives to Improve the Extrusion Characteristics of LDPE/LLDPE Resin Blends.

By John Horns, Senior Chemical Engineer, Dyneon
The Influence of Using Fluoropolymer Processing Additives to Improve the Extrusion Characteristics of LDPE/LLDPE Resin Blends.

ABSTRACT

The influence of using a fluoropolymer based processing additive (PPA) on the extrusion characteristics of LLDPE and LDPE/LLDPE blends has been investigated. Three extrusion process variables were identified and used for this comparison; material output, melt temperature, and die pressure. These experiments were performed on a series of LDPE/LLDPE blends with and without a PPA present. This work was performed on conventional extrusion equipment which had many characteristics of equipment designed and used for LDPE processing. The first investigation examined the reduction in die pressure and energy consumption when the output was held constant. The second investigation examined the decrease in melt temperature when the die pressure and output were held constant. The third investigation examined the increase in output when the die pressure and melt temperature were held constant.

INTRODUCTION

Polyethylene films have gained a wide acceptance in the packaging industry for a variety of applications. Low density polyethylene resins (LD) have been used to produce these films, but with the introduction of linear low density polyethylene resins (LL), LL and LD/LL blends are largely replacing LD and can provide specific advantages.[1] These new polymers and polymer blends offer definite advantages in physical and mechanical properties, but they are also more difficult to process.[2] Many older extrusion lines designed to process LD cannot process these new materials without changes to the extrusion process.

Equipment modifications have been identified which improve the capability of extrusion equipment designed for processing LD to process these LL and LD/LL blends.[3,4] These modifications include new and more complex screw designs to accommodate the increased melt viscosity of LL, and changing the die geometry to reduce the high die pressure associated with processing LL. Unfortunately this retrofitting also requires a significant capital investment.

Polymer processing additives (PPA) based on fluoropolymer chemistry are an attractive alternative. They are effective in reducing the apparent viscosity of high molecular weight LL and have been widely used to improve the processing characteristics of these resins.[5] The PPA is thought to function by forming a fluoropolymer coating on the die surface, which provides a low energy surface layer at the polymer melt-metal interface.[6] In fully developed pressure driven flow a zero velocity condition is defined at this interface. It is generally proposed that the presence of the PPA coating eliminates this zero velocity condition and a slip velocity is established, and the resistance of the polymer melt to flow from the extrusion die is reduced. The shear stress at the polymer melt-metal interface is also reduced. A primary benefit identified when using a PPA is the elimination of surface melt fracture. Secondary benefits are also observed which include reduction in die pressure and extrusion drive torque. These benefits expand the acceptable processing window for the resins and improve their extrusion characteristics.
EXPERIMENTAL PROCEDURE

These experiments were performed using commercially available LL (0.9 melt index, 0.920 density, C4 comonomer) and LD (0.8 melt index, 0.925 density). Both materials contained primary and secondary antioxidant stabilizers, and diatomaceous earth anti-block. Four resin ratios were evaluated, 100% LD, 100% LL and 30/70 and 70/30 blends of LD/LL. A 10/90 LD/LL blend was also included when testing the 0.4 mm and 0.6 mm die gaps.

The fluoropolymer based PPA used in these experiments is commercially available as DYNAMAR™ FX-5920A. This PPA was compounded into a 3% concentrate using a 2 MI LL carrier resin. All resin combinations were prepared by tumble blending prior to extrusion. The specific resin blends without a PPA present were extruded to establish a control condition. The extruded film was observed for melt fracture.

All PPA evaluations were performed using a 1000 ppm PPA concentration. This level exceeds the required minimum PPA level necessary for effective performance in these resins, but this high level does not have a negative effect on the properties of the film produced [7]. The required minimum PPA level necessary to eliminate melt fracture for the LD/LL resin combinations used in these experiments was not determined before performing these evaluations because the specific concentration of PPA may vary between the blends. To eliminate any question of having enough PPA present all experiments were performed at this 1000 ppm level.

The process variables were measured after a minimum of one hour extrusion time for the specific resin blends and extrusion conditions. These variables were monitored as a function of time, and data measurements were taken after an equilibrium was established. Table 1 and Figures 1 & 2 contain a description of the extrusion equipment and die used for this study. The target melt temperatures are included in the results summary, found in Tables 2-5. Flat temperature profiles were used in the extruder to achieve these values. Because of material shear heating in the extruder the set temperatures of the extruder were generally 20-25°C below the target melt temperature. The target outputs are also included in Tables 2-5.

The extrusion pressure was measured in the adapter connecting the die to the extruder. The melt temperature was measured in the same location using a thermocouple positioned 1.0 cm into the melt stream. The output was measured by weighing samples of the extrudate collected over specific time intervals.

Energy consumption calculations were made from measurements of the amperage draw and the operating rpm of the extruder motor. A 15 Hp DC drive motor, which draws 500 volts at 121 rpm maximum speed, was installed on the extruder. The specific formula used for this calculation can be represented by:

\[
\text{energy consumption} = \frac{\text{rpm actual}}{\text{rpm maximum}} \times \text{voltage maximum} \\
\times \frac{\text{amperage draw actual}}{\text{amperage reading (amps)}} \\
\frac{\text{amperage reading (amps)}}{\text{voltage maximum}}
\]

RESULTS and DISCUSSION

The first investigation examined the effect of the PPA on die pressure when the output was held constant. The results of these experiments have been summarized in Table 2 and are represented by graphing the die pressure versus LD/LL ratio and energy consumption versus % LLDPE in the blends. For all LD/LL blends tested the presence of the PPA reduces the die pressure. Graphs 1-4 summarize this data for the different die gaps and extrusion conditions which were tested. The reduction in die pressure as a result of the PPA presence was minimal for the 100% LD composition, but when LL was added to the blend the benefit of the PPA was observed as a reduction in die pressure. This influence of the PPA became more significant as the LL concentration was increased. For the 100% LL composition we observed die pressure reductions of: 16% for the 0.4 mm die gap, 18% for the 0.6 mm die gap, and 5% for the 1.0 mm die gap. This decrease at the larger die gap may be the result of the reduced apparent shear rate when maintaining constant output, which reduces the influence of the viscoelastic characteristics of the polymer melt. These three examples were generated using a flow rate of 0.60-0.75 Kg/(cm*hr). Additional data was generated by reducing the flow rate to 0.3 Kg/(cm*hr) in the 0.4 mm die gap. For this experiment a 19% reduction in die pressure was observed for the 100% LL composition.

When processing blends without a PPA present the addition of a small percentage of LL has a significant effect on the die pressure. The 0.4 mm and 0.6 mm die gaps displayed a significant increase in die pressure when 30% LL was added to the system. When the die gap was increased to 1.0 mm the system becomes less sensitive to addition of LL. A minimal increase in die pressure was observed for the 30% LL composition, but the 70% LL composition displayed a significant increase in die pressure. Raising the melt temperature helped control this increase in die pressure, but it did not control the increase enough to keep it from exceeding the die pressure with the 100% LDPE composition. Addition of PPA to the system provided a significant pressure reduction and allowed the processing of the LL and LD/LL blends at the same die pressure of the 100% LD composition. During this investigation the presence of the PPA eliminated melt fracture.
for all compositions and die gaps tested. Melt fracture was present when running the 30% LL control composition in all die gaps, and the melt fracture severity increased with higher LL concentrations.

The energy consumption of the extrusion motor can also be used to demonstrate the system was responding to the presence of the PPA. With PPA present, a 5% -15% reduction in energy consumption was recorded for all LD/LL blends and the LL composition tested in this study. Graphs 5-7 summarize this data for the different die gaps and extrusion conditions which were tested.

To quantify the benefit from using a PPA, an output comparison can be made between the control and PPA compositions while operating at the same die pressure. This was done after the equipment had been conditioned with the PPA by measuring the additional output obtained before the die pressure increased to the control composition operating die pressure. For all die gaps and the 100% LL composition tested in this study, with the PPA present a 50% increase in output was realized before reaching the control condition die pressure. The results of these experiments have been summarized in Table 3. This increase in output becomes significant and can be utilized when the extrusion process has the capability to cool, handle, and take off this additional output.

The second investigation examined the effect of the PPA from an alternative approach. These experiments determine how much the melt temperature needed to be increased to maintain the die pressure and output obtained with the 100% LD composition when processing the series of LD/LL blends with and without PPA present at a constant output. To maintain a constant die pressure and output when increasing the LL concentration in the resin blend the melt temperature had to be increased. Without a PPA present, as the concentration of LL is increased the required melt temperature goes beyond an acceptable melt temperature for stable processing. When the melt temperature was greater than 215°C the cooling capability of the equipment used for these experiments was exceeded. Bubble instability and cooling limitations are typical problems which can be observed when processing at high melt temperatures. The presence of the PPA reduces the requirement to increase the melt temperature as the LL concentration is increased in the blend. For all tested resin blends with a PPA present, including the 100% LL composition, a melt temperature below 215°C is maintained. The results of these experiments have been summarized in Table 4 and Graphs 8-10.

The third investigation examined the effect of the PPA on output when the die pressure and melt temperature were held constant. Melt temperature is a process variable which is generally changed to achieve specific extrusion properties. In this series of experiments the melt temperature and die pressure were held constant as the LL concentration was increased. Because of the higher melt viscosity of the LL resin, the output needs to be decreased as the LL concentration was increased to maintain these conditions. The presence of the PPA reduces the resistance to flow and resulted in a smaller decrease in output as the LL concentration was increased. Comparing the control with the PPA the most dramatic difference in output was observed with the 0.4 mm die gap. When comparing the control condition to the PPA condition a 67% difference in output was observed for the 100% LL composition at 21.4 MPa die pressure and Tmelt = 197°C. This difference decreases to 55%, 35%, and finally zero as the LL concentration was decreased. A similar trend was noted in the 0.6 mm and 1.0 mm die gap, but it became less pronounced as the die gap was increased. The results of these experiments have been summarized in Table 5 and Graphs 11-13.

**CONCLUSIONS**

The extrusion characteristics of high molecular weight LL, and LD/LL resin blends are improved by the presence of a fluoropolymer based processing additive. The resistance to flow is reduced and a reduction in die pressure is observed. These changes effectively expand the processing window for the polymer, and the extrusion process variables can be changed to more efficient operating conditions. This study demonstrated potential changes which can be made to melt temperature, material output, or die geometry.

The extrusion process is simultaneously controlled by many variables, but some general conclusions can be drawn:

- If the die pressure is limiting the extrusion process, adding a PPA will reduce the observed die pressure and allow higher outputs to be attained before exceeding the limiting die pressure.
- If die pressure is limiting the concentration of LL which can be added to a blend, adding a PPA will allow higher concentrations of LL to be added to the system before exceeding the limiting die pressure.
- To extrude specific LD/LL blends at constant die pressures, a lower melt temperature can be used when a PPA is present. This allows stable blown film extrusion conditions to be maintained as the LL concentration is increased.
If melt fracture is present when processing LL or LD/LL blends, adding a PPA will improve or eliminate this condition.

The presence of the PPA reduces the energy consumption of the extrusion motor. This energy reduction is also observed for the 100% LD composition, even though no reduction in die pressure is observed.

The presence of the PPA reduces the resistance to flow. This can be measured as an increase in output if the melt temperature and die pressure are held constant.

REFERENCES


### TABLE 1: EQUIPMENT SPECIFICATIONS

**EXTRUDER:**
- manufacturer: Filmmaster
- barrel: 3.8 cm diameter
- drive motor: DC drive, 15 horsepower

**SCREW DESIGN:**
- Equal length sections: feed, compression, metering
- Maddock mixing tip
- square pitch
- helix angle: 17°
- compression ratio: 3/1
- length/diameter ratio: 24/1
- flight-barrel gap: 0.06 mm

**DIE DESIGN:**
- flow design: minimal channeling, no spiral feed
- die diameter: 5.1 cm
- die gaps: 0.41/0.64/1.02 mm
- air ring: dual lip non impinging flow
### TABLE 2: VARIABLES MELT TEMPERATURE AND DIE PRESSURE AT CONSTANT OUTPUT

#### 0.4 mm die gap at 10 Kg/hr output

<table>
<thead>
<tr>
<th>% LL</th>
<th>Tmelt (°C)</th>
<th>Die pressure (Mpa)</th>
<th>Extruder screw speed (rpm)</th>
<th>Extruder motor amperage draw (A)</th>
<th>Extruder energy consumption (Kw)</th>
<th>Control</th>
<th>+PPA</th>
<th>Control</th>
<th>+PPA</th>
<th>Control</th>
<th>+PPA</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>208</td>
<td>34.93</td>
<td>29.30</td>
<td>60</td>
<td>57</td>
<td>7.7</td>
<td>7.5</td>
<td>1.909</td>
<td>1.766</td>
<td></td>
<td></td>
</tr>
<tr>
<td>90</td>
<td>207</td>
<td>33.51</td>
<td>28.99</td>
<td>60</td>
<td>59</td>
<td>7.4</td>
<td>7.1</td>
<td>1.835</td>
<td>1.731</td>
<td></td>
<td></td>
</tr>
<tr>
<td>70</td>
<td>196</td>
<td>32.47</td>
<td>28.55</td>
<td>63</td>
<td>61</td>
<td>7.0</td>
<td>6.7</td>
<td>1.822</td>
<td>1.689</td>
<td></td>
<td></td>
</tr>
<tr>
<td>30</td>
<td>168</td>
<td>30.85</td>
<td>28.68</td>
<td>64</td>
<td>63</td>
<td>6.8</td>
<td>6.5</td>
<td>1.798</td>
<td>1.692</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0</td>
<td>144</td>
<td>29.43</td>
<td>28.68</td>
<td>61</td>
<td>64</td>
<td>7.0</td>
<td>6.2</td>
<td>1.764</td>
<td>1.640</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

#### 0.4 mm die gap at 5 Kg/hr output

<table>
<thead>
<tr>
<th>% LL</th>
<th>Tmelt (°C)</th>
<th>Die pressure (Mpa)</th>
<th>Control</th>
<th>+PPA</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>195</td>
<td>39.30</td>
<td>31.85</td>
<td></td>
</tr>
<tr>
<td>70</td>
<td>191</td>
<td>37.71</td>
<td>31.72</td>
<td></td>
</tr>
<tr>
<td>30</td>
<td>174</td>
<td>35.30</td>
<td>32.06</td>
<td></td>
</tr>
<tr>
<td>0</td>
<td>152</td>
<td>32.41</td>
<td>31.78</td>
<td></td>
</tr>
</tbody>
</table>

#### 0.6 mm die gap at 10 Kg/hr output

<table>
<thead>
<tr>
<th>% LL</th>
<th>Tmelt (°C)</th>
<th>Die pressure (Mpa)</th>
<th>Extruder screw speed (rpm)</th>
<th>Extruder motor amperage draw (A)</th>
<th>Extruder energy consumption (Kw)</th>
<th>Control</th>
<th>+PPA</th>
<th>Control</th>
<th>+PPA</th>
<th>Control</th>
<th>+PPA</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>205</td>
<td>30.18</td>
<td>24.68</td>
<td>72</td>
<td>65</td>
<td>8.4</td>
<td>7.9</td>
<td>2.499</td>
<td>2.122</td>
<td></td>
<td></td>
</tr>
<tr>
<td>90</td>
<td>205</td>
<td>29.03</td>
<td>24.27</td>
<td>73</td>
<td>65</td>
<td>7.9</td>
<td>7.5</td>
<td>2.383</td>
<td>2.014</td>
<td></td>
<td></td>
</tr>
<tr>
<td>70</td>
<td>197</td>
<td>27.44</td>
<td>24.47</td>
<td>72</td>
<td>68</td>
<td>7.3</td>
<td>7.2</td>
<td>2.172</td>
<td>2.023</td>
<td></td>
<td></td>
</tr>
<tr>
<td>30</td>
<td>174</td>
<td>26.49</td>
<td>23.72</td>
<td>72</td>
<td>71</td>
<td>7.1</td>
<td>6.8</td>
<td>2.112</td>
<td>1.995</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0</td>
<td>153</td>
<td>24.30</td>
<td>23.85</td>
<td>72</td>
<td>75</td>
<td>7.0</td>
<td>6.6</td>
<td>2.083</td>
<td>2.045</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

#### 1.0 mm die gap at 10 Kg/hr output

<table>
<thead>
<tr>
<th>% LL</th>
<th>Tmelt (°C)</th>
<th>Die pressure (Mpa)</th>
<th>Extruder screw speed (rpm)</th>
<th>Extruder motor amperage draw (A)</th>
<th>Extruder energy consumption (Kw)</th>
<th>Control</th>
<th>+PPA</th>
<th>Control</th>
<th>+PPA</th>
<th>Control</th>
<th>+PPA</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>212</td>
<td>20.30</td>
<td>19.26</td>
<td>69</td>
<td>64</td>
<td>7.9</td>
<td>7.4</td>
<td>2.252</td>
<td>1.957</td>
<td></td>
<td></td>
</tr>
<tr>
<td>70</td>
<td>198</td>
<td>20.64</td>
<td>19.48</td>
<td>70</td>
<td>68</td>
<td>7.1</td>
<td>6.7</td>
<td>2.054</td>
<td>1.883</td>
<td></td>
<td></td>
</tr>
<tr>
<td>30</td>
<td>174</td>
<td>19.71</td>
<td>19.48</td>
<td>72</td>
<td>69</td>
<td>6.6</td>
<td>6.5</td>
<td>1.964</td>
<td>1.853</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0</td>
<td>150</td>
<td>19.44</td>
<td>19.39</td>
<td>68</td>
<td>71</td>
<td>7.2</td>
<td>6.7</td>
<td>2.023</td>
<td>1.966</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
### TABLE 3: VARIABLE OUTPUT AT CONSTANT DIE PRESSURE AND MELT TEMPERATURE

<table>
<thead>
<tr>
<th>%LL</th>
<th>output (Kg/hr)</th>
<th>output (Kg/hr)</th>
<th>output (Kg/hr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>control 5.0</td>
<td>+PPA 7.3</td>
<td></td>
</tr>
<tr>
<td>90</td>
<td>control 10.0</td>
<td>+PPA 15.0</td>
<td></td>
</tr>
<tr>
<td>80</td>
<td>control 11.3</td>
<td>+PPA 17.2</td>
<td></td>
</tr>
</tbody>
</table>

### TABLE 4: VARIABLE MELT TEMPERATURE AT CONSTANT OUTPUT AND DIE PRESSURE

<table>
<thead>
<tr>
<th>%LL</th>
<th>Tmelt (°C)</th>
<th>Tmelt (°C)</th>
<th>Tmelt (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>control 232</td>
<td>+PPA 208</td>
<td></td>
</tr>
<tr>
<td>70</td>
<td>control 238</td>
<td>+PPA 205</td>
<td></td>
</tr>
<tr>
<td>30</td>
<td>control 170</td>
<td>+PPA 197</td>
<td></td>
</tr>
<tr>
<td>0</td>
<td>control 144</td>
<td>+PPA 144</td>
<td></td>
</tr>
</tbody>
</table>

### TABLE 5: VARIABLE OUTPUT AT CONSTANT MELT TEMPERATURE AND DIE PRESSURE

<table>
<thead>
<tr>
<th>%LL</th>
<th>output (Kg/hr)</th>
<th>output (Kg/hr)</th>
<th>output (Kg/hr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>control 3.18</td>
<td>+PPA 5.31</td>
<td></td>
</tr>
<tr>
<td>70</td>
<td>control 3.67</td>
<td>+PPA 5.03</td>
<td></td>
</tr>
<tr>
<td>30</td>
<td>control 4.54</td>
<td>+PPA 5.67</td>
<td></td>
</tr>
<tr>
<td>0</td>
<td>control 14.06</td>
<td>+PPA 14.11</td>
<td></td>
</tr>
</tbody>
</table>
FIGURE #1: SCREW SCHEMATIC AND SPECIFICATIONS:

1-1/2” Extruder Screw Dimensions

Notes:
Material: 4140 Mill Hardened
Plate .001” Chrome
Dimensions shown after plating
Flame Hardened Flights
Flight Thickness = 0.25”
Runout on screw = ± 002”

Drawing No. 011492-SUL

FIGURE #2: DIE SCHEMATIC: CROSS-SECTIONED SIDE VIEW OF DIE

Air Inlet
GRAPH 1: EXTRUDER PRESSURE VERSUS %LLDPE

- Adaptor Pressure (MPa)
- Melt Temperature (°C)

0.4 mm die gap
output = 0.31 Kg/hr/cm
apparent shear rate = 330 s⁻¹

Pressure decrease 9-19% with PPA present

- control
- PPA
- Tmelt

GRAPH 2: DIE PRESSURE VERSUS %LLPDE

- Die Pressure (MPa)
- Melt Temperature (°C)

0.4 mm die gap
output = 0.63 Kg/hr/cm
apparent shear rate = 650 s⁻¹

Pressure decrease 10-20% with PPA present

- control
- PPA
- Tm
GRAPH 3: DIE PRESSURE VERSUS %LLDPE

0.6 mm die gap
output = 0.71 Kg/hr/cm
apparent shear rate = 330 s⁻¹

pressure decrease 10-18 % with PPA present

GRAPH 4: DIE PRESSURE VERSUS %LLDPE

1.0 mm die gap
output = 0.74 Kg/hr/cm
apparent shear rate = 120 s⁻¹

pressure decrease 1-5 % with PPA present
**GRAPH 5: ENERGY CONSUMPTION VERSUS %LLDPE**

0.4 mm die gap  
output = 0.63 Kg/hr/cm  
apparent shear rate = 650 s⁻¹  

energy consumption decrease 5-7 % with PPA present

**GRAPH 6: ENERGY CONSUMPTION VERSUS %LLDPE**

0.6 mm die gap  
output = 0.71 Kg/hr/cm  
apparent shear rate = 330 s⁻¹  

energy consumption decrease 5-15 % with PPA present

**GRAPH 7: ENERGY CONSUMPTION VERSUS %LLDPE**

1.0 mm die gap  
output = 0.74 Kg/hr/cm  
apparent shear rate = 120 s⁻¹  

energy consumption decrease 5-13 % with PPA present
GRAPH 11: OUTPUT VERSUS % LLDPE

GRAPH 12: OUTPUT VERSUS % LLDPE

GRAPH 13: OUTPUT VERSUS % LLDPE