A FORMULATION STUDY OF LONG FIBER THERMOPLASTIC POLYPROPYLENE (PART 4): THE EFFECT OF MOLDING CHANGES ON MECHANICAL PROPERTIES OF PP LFT PARTS

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Abstract

In part four of this multiyear study the relationship between the Polypropylene Long Fiber Thermoplastics (PP LFT) and the processes used to mold are presented. Parts are molded by injection molding, compression molding and plunger tool using the same formulation in the PP LFT part. The effects of glass fiber diameter, sizing and changes in molding conditions were explored. Prior work showed significant differences between injection molding and compression molding. The use of the plunger tool has given a unique opportunity to look at the effects of molding conditions and fiberglass products on the final part performance.

Background

Long Fiber Thermoplastics have enjoyed a long run of double digit growth and have found general acceptance as structural materials. Polypropylene (PP) LFT materials offer strength and stiffness and are recyclable having a long shelf life. Consequently, their introduction has led to an increased penetration of the automotive market for structural thermoplastic composites. These applications typically are metal replacement and facilitate both part weight reduction and part consolidation. PP LFTs are the largest segment of this market and are also experiencing the largest growth. Within the PP LFT segment there are three distinct methods of making and using these materials. The classic method is the pultrusion impregnation and/or cross head extrusion method of generating LFT pellets that are subsequently injection molded. For the purpose of this paper this type of material is labeled Granulate Long Fiber Thermoplastics (GLFT). The second main method of producing LFT is In Line Compounding(IC). Within the IC segment there are a number of distinct methods of producing parts. In one method, the IC material is compression molded into parts in a process similar to that used for the manufacture of Glass Mat Thermoplastics and thermoset parts. Typical compression molded parts are made by placing the hot charge in a mold and closing the press quickly to fill or stamp the part. A second method of compression molding uses a plunger tool where the hot charge is placed in a central cavity of the mold and the tool is closed. A plunger then pushes the charge out of the central cavity and into the mold. The parts produced have a different fiber dispersion pattern than what is achieved with the standard compression molding. In a third method the IC material is fed directly into an injection molding machine for either standard Injection molding or Injection compression molding. All three of these IC methods are Direct Long Fiber Thermoplastics (DLFT). The third distinct material within the LFT field is Continuous Long Fiber Thermoplastics (CLFT) and is not discussed here.

In this study, a comparison of parts produced with injection molding, standard compression molding and compression molding with a plunger tool are compared using identical resin formulations, fiber glass type and fiber glass loadings. The fiber glass sizing and the coupling agent loading were varied. In previous studies [1-2] the results of injection molded (IM) PP GLFT parts were studied. The coupling agent melt flow and maleic anhydride loading were found to be important to obtaining optimum properties.
Figure 1: Flexural Strength versus Coupling Agent MFI and MAH Loading In 60% PP GLFT with TufRov® 4575 Fiber Glass

Figure 1 shows the response of flexural strength in 60% glass filled PP GLFT versus the percent of MAH and the MFI of the coupling agent. The percent of MAH is reported as % MAH in the coupling agent. There are a number of trends that can be extracted from Figure 1. First: The maximum flexural strength obtained is almost 280 MPa. This is phenomenally high flex strength for a PP based composite. Second: The higher the MFI of the coupling agent, the higher the flex strength. Third: The higher the MAH loading, the higher the flex strength. The trends break down slightly at low coupling agent loadings but this may be because of an incomplete coverage of the fiber glass with coupling agent.

In the compression molding study [3], the composite properties were not easily affected by changing material variables. The study showed that the DLFT properties for compression molded parts are very stable and predictable even with significant changes in the formulation (Figure 2); even though the fiber glass sizing showed significant differences in adhesion performance in the same parts (Figure 3-4).
Figure 2: Coupling Agent Type and Loading versus Tensile Strength in Compression Molded DLFT. No real change in Tensile Strength observed.

Figure 3: Representative curve for TufRov® 4599 with 2% BondyRam 1001

Figure 3: The 4599 single fiber pull out curve follows a classic slip/stick failure mode and is further evidence that the primary mechanism of failure is fiber pull out. Friction is responsible for holding the fiber in the resin. When the load rises to a critical level the fiber slips until the load is alleviated. The cycle then starts again. There is good wetting of the resin onto the fiber surface, but little actual adhesion between the resin and glass.
Figure 4: The 4575 single fiber pull out test show excellent adhesion between the fiber and polypropylene matrix. The fiber does not slip from the resin but continues to hold until the load reaches a critical level and complete failure occurs. The interface between resin and fiber does not fail and is indicative of a high level of bonding between the fiber glass and resin. The maximum load is also significantly higher and indicative of better tensile and flexural strength in the composite.

Experimental

PPG TufRov 4575, TufRov 4599, TufRov 4588 fiber glass rovings were processed through a Coperion ZSK 40mm twin screw extruder to produce either DLFT compression molded plaques, DLFT plunger tool molded parts, or GLFT pellets for subsequent injection molding. A blended masterbatch containing a heat stabilizer package and carbon black remained constant at 5% (w/w). The amount of coupling agent was varied in the study from 0% to 5% using one coupling agent. The fiber glass type and fiber diameter were varied.

- Marlex® HGZ-1200 homopolymer (120MFI)
- Polyram BondyRam® 1001 100 MFI Maleic Anhydride grafted PP with 1% Maleic Anhydride (w/w)
- A heat stabilizer package
- Carbon Black

DLFT Sample Preparation

Both DLFT sample sets were produced by introducing a resin mixture into the twin screw extruder and allowing the resin to melt before directly introducing fiber glass rovings into the extruder at a later point. The heat profile of the twin screw extruder is listed in Table 1.
Table 1. Molding Thermal Profile of ZSK 40mm Twin Screw Extruder

<table>
<thead>
<tr>
<th>Molding Profile</th>
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<tbody>
<tr>
<td>Zone 1</td>
<td>170°C</td>
</tr>
<tr>
<td>Zone 2</td>
<td>190°C</td>
</tr>
<tr>
<td>Zone 3</td>
<td>190°C</td>
</tr>
<tr>
<td>Zone 4</td>
<td>220°C</td>
</tr>
<tr>
<td>Zone 5</td>
<td>240°C</td>
</tr>
</tbody>
</table>

The speed of the extruder varied from 60 rpm to 120 rpm while varying the amount of PP resin mixture being fed into the extruder to control the glass content of the samples. Figure 5 illustrates a curve produced to correlate rpm versus lb/hr PP resin to control the percent by weight of fiber glass in the samples.

![Screw Speed vs. Lbs/ Hr](image)

**Figure 5. Correlation of Screw Speed versus Resin Lbs/Hr to control glass content**

Table 2. Collection Time of Charge for Screw Speed of Extruder

<table>
<thead>
<tr>
<th>Screw Speed</th>
<th>Glass Content</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>40%</td>
</tr>
<tr>
<td>90 rpm</td>
<td>2min</td>
</tr>
<tr>
<td>60 rpm</td>
<td>2min 40sec</td>
</tr>
<tr>
<td>120rpm</td>
<td>1min 30sec</td>
</tr>
</tbody>
</table>
To ensure uniform thickness and consistency in the DLFT produced samples, the amount of extrudate was collected for equal amounts of time depending on the screw speed of the extruder. Table 2 lists the collection times for the different screw speeds.

Figure 6: Plunger Tool
To complete the sample for the compression molded parts the “charge” from the extruder was compression molded. A 40 cm by 40 cm plaque tool was installed in a St. Lawrence 150 ton hydraulic press. The charge was pressed at 450 psi for 40 seconds with a mold temperature of 140°F. The plaques produced were cut using an O-MAX 2626 water jet cutter to make the final test specimens for mechanical analysis.

To complete the sample for the plunger tool molded parts the “charge” from the extruder was rolled into a log and placed in the plunger tool central cavity (Figure 6-7). The St. Lawrence
150 ton hydraulic press was then closed and the hydraulic plunger activated to inject the extrudate into the mold cavities. The cycle time was 72 seconds with a long hold time of 55 seconds after plunger activation. Platen close time was as fast as possible with the press and mold temperature was set at 230 ⁰F.

**GLFT Sample Preparation**

GLFT pellets were produced on a cross head pultrusion die attached to the Coperion ZSK 40 mm twin screw extruder. The extruded strands were pelletized to an average length of 12 mm. Glass content was controlled in the cross head die to obtain the target of 40% glass by weight for these studies.

To complete the injection molded parts samples were dried at 120 °C for four hours and the moisture was measured for each sample prior to molding with maximum allowed moisture of 0.6% (w/w). A Van Dorn 55 injection molding machine was used for molding test specimens. A free flow check valve was installed on the screw tip prior to the initiation of this work. The mold used was standard ISO tensile test specimen mold with large diameter sprues and runners and large radius curves. The gate is a fan gate to the tensile specimens. The mold is a balanced design producing two tensile specimens per shot. Impact and flex specimens were cut from the tensile bars by removing the tab section of the specimen. All test specimens conformed to ISO requirements. The exact profile used for the molding is recorded in Table 3.

<table>
<thead>
<tr>
<th>Molding Profile</th>
<th>Temperature (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rear Zone</td>
<td>225</td>
</tr>
<tr>
<td>Center Zone</td>
<td>240</td>
</tr>
<tr>
<td>Front Zone</td>
<td>250</td>
</tr>
<tr>
<td>Nozzle</td>
<td>250</td>
</tr>
<tr>
<td>Mold</td>
<td>50</td>
</tr>
<tr>
<td>Shot Size, inch</td>
<td>2.25</td>
</tr>
<tr>
<td>Boost Pressure</td>
<td>1000</td>
</tr>
<tr>
<td>Injection Speed</td>
<td>1.5</td>
</tr>
<tr>
<td>Hold Pressure</td>
<td>925</td>
</tr>
<tr>
<td>Screw Speed</td>
<td>90</td>
</tr>
<tr>
<td>Back Pressure</td>
<td>50</td>
</tr>
</tbody>
</table>

**Table 3: Van Dorn 55 Injection Molding Profile for GLFT specimens**

Unless otherwise stated, all mechanical property testing was performed at 23°C and at a relative humidity of 50%. All testing was done in the PPG Fiber Glass Science and Technology Laboratory in Hoogezeand, The Netherlands. Tensile properties were tested according to ISO 527-2 using 10 specimens and a crosshead rate of 5 mm/min (0.2 in/min) with an extensometer gauge length of 50 mm (2 in). Flex properties were tested according to ISO 14125 using 10 specimens and a crosshead rate of 2.0 mm/min (0.8 in/min) and a span of 64 mm (2.56 in). Charpy notched impact properties were tested according to ISO 179-1 with 10 specimens and a Type A notch. Charpy UnNotched impact properties were tested according to ISO 179-1.
Results

GLFT Results

The GLFT results follow the trends of previous studies with a nice response to coupling agents loading and obvious differences in fiber performance in the composite (Figure 9-10). The previous trend of 4575 offering the best overall performance is confirmed. As more coupling agent is added the differences in fiber glass performance is reduced with 4588 offering equivalent performance at high loadings of coupling agent. 4575 still offers the best performance in properties and cost as it requires the least amount of the more expensive coupling agent. The 0% loading data does indicate some of the reasons for this improved performance. The 4575 sizing system includes some functional modalities that work as coupling agents. You also see a slight reduction in strengths with increasing fiber glass diameter in the 4575 product. This reduction is often observed in highly crystalline resin systems. The reduction is slight as would be expected in a semi-crystalline PP resin. Figure 11 is a compilation of Tensile and Flexural Strength properties of GLFT samples with 2% coupling agent. This shows some of the differences in fiber glass sizing performance.

![Image of Flexural Strength for various Fiber Glass Rovings](image-url)

Figure 9: GLFT Flexural Properties
Figure 10: GLFT Tensile Properties

Figure 11: Flexural and Tensile Strength in GLFT
DLFT Compression Molded Results

The DLFT results mimic those obtained in the previous study [3]. There is very little change in performance with either change in fiber glass or the amount of coupling agent added. Statistically the data is equivalent with the possible exception of some differences with no coupling agent added (Figure 12-13). In the case of 0% coupling agent 4588 does poorly and 4599 shows very high impact properties. Because we know the adhesion is not optimal with 4599 the mechanism of failure for the impact is probably changed to fiber pull out for this specific 0% coupling agent sample (Figure 14).

Flexural Strength for various Fiber Glass Rovings
40% PP LFT DLFT Compression Molded
Flow Direction Only

![Flexural Strength Chart](image)

Figure 12: DLFT Flexural Strength

Figures 15 and 16 show a compilation of properties for Tensile and Flexural strengths with a common 2% loading of coupling agent. There is a visual trend of different properties with the different fiber glass samples used. However, the trend is deceiving as there are no statistical differences between these fiber glass reinforcements in the composite.
Figure 13: Tensile Strength in Flow Direction for DLFT

Figure 14: Charpy UnNotched Impact of Fiber Glass Rovings in DLFT
Figure 15: Flexural Strength of Fiber Glass rovings in DLFT

Figure 16: Tensile Properties of Fiber Glass Rovings in DLFT
DLFT Plunger Tool Results:

The plunger tool LFT parts are made with a process that is similar to both the DLFT compression molding process and the GLFT injection molding process. The material is heated and mixed in the same system as the compression molded DLFT and is injected into the part cavity in the plunger tool similar to a standard injection molding machine. As such, you would expect that this methodology of part making would produce parts that have aspects of both.

The fiber burn offs shown in Figure 8 tell part of the story. The plunger tool is very gentle on the fiber glass with a very low shear process. The glass lengths obtained are longer on average than what you see in the injection molding process and the fiber alignment is very different from GLFT. In the GLFT parts you have a significant skin and core region of the part. The skin region is highly aligned with the flow while the core fiber glass alignment is more perpendicular to the flow. In the plunger tool parts the alignment appears to be more with the flow and the skin/core effect seen in injection molding was not discernible in the parts we produced with our very gentle system. The fiber lengths obtained in the plunger tool are similar to what was obtained in the compression molded parts. There seems to be a slight loss compared to compression molding, but it may not be significant. Comparison of the fiber alignment between plunger tool parts and compression molded parts indicate that they both have a high degree of alignment in the flow direction. In DLFT compression molding there are very different properties in the flow and the cross flow direction. This is why the flow and cross flow properties are always evaluated to give a better representation of the final part properties. No evaluation of the cross flow properties of the plunger tool parts were made as part of this initial study. If these fiber orientations prove correct, then an evaluation of the cross flow properties will need to be done to give a better representation of the actual composite properties.

![Flexural Strength of Various Fiber Glass Rovings](image)

**Figure 17: Flexural Strength in Plunger Tool Molded parts**
Charpy UnNotched Impact on Various Fiber Glass Rovings
40% PP LFT Plunger Tool

Figure 18: Charpy UnNotched Impact in Plunger Tool Parts

Tensile Strength of Various Fiber Glass Rovings
40% PP LFT Plunger Tool DLFT

Figure 19: Tensile Strength in Plunger Tool Parts
Figure 20: Flexural Strength in Plunger Tool parts

The plunger tool gives impressive mechanical properties that are comparable to those obtained in GLFT. The materials appear to be more sensitive to changes in the formulation of TufRov 4575 Flexural Strength in 40% PP LFT Comparison of Plunger Tool, Injection Molded and Compression Molded

Figure 21: Tensile Strength MPa

Comparison of Plunger Tool to GLFT Injection Molding to DLFT Compression Molding Flow Direction
TufRov 4575 Sizing System with Different Fiber Glass Diameters

Processing Method versus Fiber Diameter

The plunger tool gives impressive mechanical properties that are comparable to those obtained in GLFT. The materials appear to be more sensitive to changes in the formulation of...
the composite, as is seen in GLFT (Figure 17-19). A significantly higher mechanical performance is observed compared to DLFT compression molding (Figure 20-22). The higher fiber alignment in the flow direction is probably contributing to these better mechanical properties. The plunger tool properties are not as sensitive to changes in fiber glass sizing systems as observed in the GLFT data. The overall tensile and flexural properties are higher than for the compression molded samples. The Impact properties may indicate some degree of higher sensitivity to changes in coupling agent loading and fiber glass sizing, but the differences are minor and are not significant. Figures 20-22 show the comparison of properties of a set formulation with 2% coupling agent and with the 4575 fiber glass sizing. The variables evaluated are fiber glass diameter and the molding process for making the parts. The tensile properties in Figure 21 show that the plunger tool gives statistically equivalent performance to the injection molding. The lower flow tensile properties of the compression molded samples are telling. Figure 22 shows the impact performance comparison. The plunger tool impact properties are in between the injection molded and compression molded values. The injection molded Charpy UnNotched impact properties are statistically different from the compression molded parts. The flexural properties in Figure 23 indicate a higher value for the injection molded samples, but this may not be significant. The plunger tool performance is actually very good in tensile, flexural and impact performance.

The evaluation of fiber glass diameter to properties throughout this study indicate that there is little loss in properties with the use of the larger diameter fiber in a polypropylene composite.

A serious study of the plunger tool cross flow properties will need to be undertaken to determine if there is a similar flow/cross flow disparity in properties as is seen in the DLFT compression molded parts. The plunger tool is more expensive to manufacture than a compression molding tool, but the improved performance offered may make it the method of choice for certain LFT part production. In our study we did not optimize cycle time because of the constraints of our press. The long cycle time in this study is probably not indicative what can be obtained with a high speed press.

As a disclaimer: In this study all samples were targeted for a nominal 40% by weight glass content and the compression molded and plunger tool parts were very close to target. In the case of the GLFT parts the glass content on some samples was higher than expected. This will tend to favor the GLFT properties; giving somewhat artificially high numbers, when compared to the other 40% by weight samples. Table 4 shows the average glass content and the range in the samples produced for all three processes. The average values are well within normal variation with good standard deviations however, for the GLFT there were a couple of samples with high weight percent glass loadings that could skew some data. One must be careful in assuming that the results here will be completely indicative of what will be obtained in a production part where the glass content is better controlled. The purpose of this study was to show the potential for greatly improved mechanical properties with the plunger tool compared to standard compression molded DLFT. The GLFT data is included for reference only and was not the focus of the study.
Table 4: Glass content range in Data sets

<table>
<thead>
<tr>
<th></th>
<th>GLFT Injection</th>
<th>Plunger Tool</th>
<th>DLFT Compression</th>
</tr>
</thead>
<tbody>
<tr>
<td>Average Glass Content (% by weight)</td>
<td>42.89</td>
<td>39.37</td>
<td>38.90</td>
</tr>
<tr>
<td>Standard Deviation</td>
<td>1.46</td>
<td>1.55</td>
<td>1.81</td>
</tr>
<tr>
<td>Range High</td>
<td>46.56</td>
<td>42.24</td>
<td>43.43</td>
</tr>
<tr>
<td>Range Low</td>
<td>40.90</td>
<td>36.66</td>
<td>37.25</td>
</tr>
</tbody>
</table>

Comparison of Plunger Tool to GLFT Injection Molding to DLFT Compression Molding Flow Direction
TufRov 4575 Sizing System with Different Fiber Glass Diameters

Figure 22: Comparison of molding method to Charpy UnNotched Impact

Summary and Next Steps

The comparison of plunger tool molded, compression molded and Injection molded LFT parts all using the same formulations has allowed direct comparison of these three distinctly different methods of production. The GLFT parts are easy to mold and give high mechanical properties that are easy to predict. The DLFT parts are easy to mold with fast cycle times and give parts that are very stable in properties with little response to changes in formulation. The plunger tool molded parts are also easy to mold with high mechanical properties in the flow direction and are responsive to changes in the material formulation. It is yet to be seen if the properties observed in the flow direction of the molded parts will translate into higher or lower mechanical properties in the cross flow direction. It is important to know that the plunger tool parts are sensitive to formulation changes and that this can allow optimization of mechanical
properties as is seen in GLFT [1-2]. This is significantly different than what has been observed in the compression molded parts [3]. The lack of sensitivity to fiber glass sizing changes in this study with the plunger tool may be indicative of the lower shear process observed and the fiber orientation in the plunger tool. Further work will need to be done to determine how to optimize the fiber glass performance in the plunger tool process.

**Figure 23: Comparison of molding method to Charpy UnNotched Impact**

**Acknowledgements**

Many thanks to Rhonda Hopper and Frank Smith for running the numerous trials needed to generate this study. Also thank you to Piet Leegstra for his work in coordinating the testing.

**Bibliography**


5. Thomason, J.L., “The influence of fibre length and concentration on the properties of glass fiber


