

EXTERIOR LONG GLASS FIBER POLYPROPYLENE SYSTEM FOR AUTOMOTIVE APPLICATIONS

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Abstract

Long glass fiber technology is used to reinforce both polyolefins and engineering thermoplastics. There is a large potential for growth in polypropylene (PP) and opportunities for LGF-ABS automotive applications. There are two primary processes for introducing long glass fiber into composite systems which are used in the industry today: direct and pellet processes. The direct processes include new extrusion-compression and injection molding equipment whereas the pellets are formed via pultrusion type processes. These processes can be used in applications where design and material are optimized for metal replacement or other material substitution opportunities. Potential benefits include part consolidation and weight reduction, as well as improved economics.

In this work, a long glass fiber composite material was developed for use in the direct extrusion compression process, which met stringent part performance, needs including molded-in-color aesthetics, customer weathering specifications, mechanical properties, and impact performance. Application requirements were achieved by the use of functionalized masterbatches in conjunction with olefinic base resins in a materials system approach.

Introduction

Long glass fiber technology is used to reinforce both polyolefins and engineering thermoplastics. There is a large potential for growth in automotive applications for LGF-PP and LGF-ABS composite materials. Estimates are that about 30% of the 2 million metric tons of e-glass fiber consumed globally for reinforcement is used in thermoplastic composites. Glass filled thermoplastic composites have been growing at a very healthy pace of 15-20%/yr, largely fueled by automotive applications. Two key reasons that glass reinforced thermoplastics are so accepted are recyclability and ease of moldability. One of the most revolutionary technologies to come to the forefront is the long glass fiber reinforced (LGF) resins. Specifically, the use of long glass fiber reinforcement in polypropylenes has allowed the use of a lower cost polymer to be used in structural, engineered applications. Potential benefits include part consolidation and weight reduction, as well as improved economics. Long glass fiber thermoplastic composites can be used in applications where design and material are optimized for metal replacement or other material substitution opportunities.

There are two primary methods of delivering long glass fiber composite thermoplastics: direct compression/injection and pultruded pellet injection molding processes. The direct compression/injection processes include new extrusion-compression and extrusion-injection molding equipment whereas the pultruded pellet injection molding processes utilize pre-compounded pellets to deliver the long glass fiber to the application.

Pellet LGF-PP Process and Products

As a material solutions provider, Dow Automotive provides a portfolio of products and services in long glass fiber technology that include pultruded pellet grades for injection molding processes and DLGFPP systems for direct compression/ injection molding processes. For this paper, pultruded pellet injection molding processes are identified as processes needing a complete compounded formulation to supply the material requirements. Direct processes, however, use compounding technology integrated with compression or injection molding technology to manufacture long glass fiber composite parts.

For pultruded pellet injection molding processes, pultruded long glass fiber pellets are available in 20, 30, and 40 percent glass loading. These pultruded pellets start with 11mm fiber length before molding.

Table 1 – Injection Moldable LGF-PP Pellet Products

| LGF Pellet Materials¹ | Glass Loading @ 11 mm LGF Pellet | Automotive Applications |
|---|---|--|
| DLGF 9201.00Z | 20% | Claddings, Door Modules, Instrument Panel Substrate, Impact Applications |
| DLGF 9311.00Z | 30% | Front End Carriers, Structural Applications |
| DLGF 9411.00Z | 40% | Front End Carriers, Structural Applications |

Another long glass fiber solution for traditional molding is the use of highly loaded pultruded glass filled masterbatches. With these masterbatches, molders can utilize standard weigh blending equipment to add the long glass pellets to resins specifically selected to mix well with the engineered long glass fiber masterbatch. Again, the glass loading can be adjusted as needed for the application. Long glass fiber composites with glass loadings of 20, 30, and 40 percent can be accomplished.

¹ Supplier: Dow Automotive

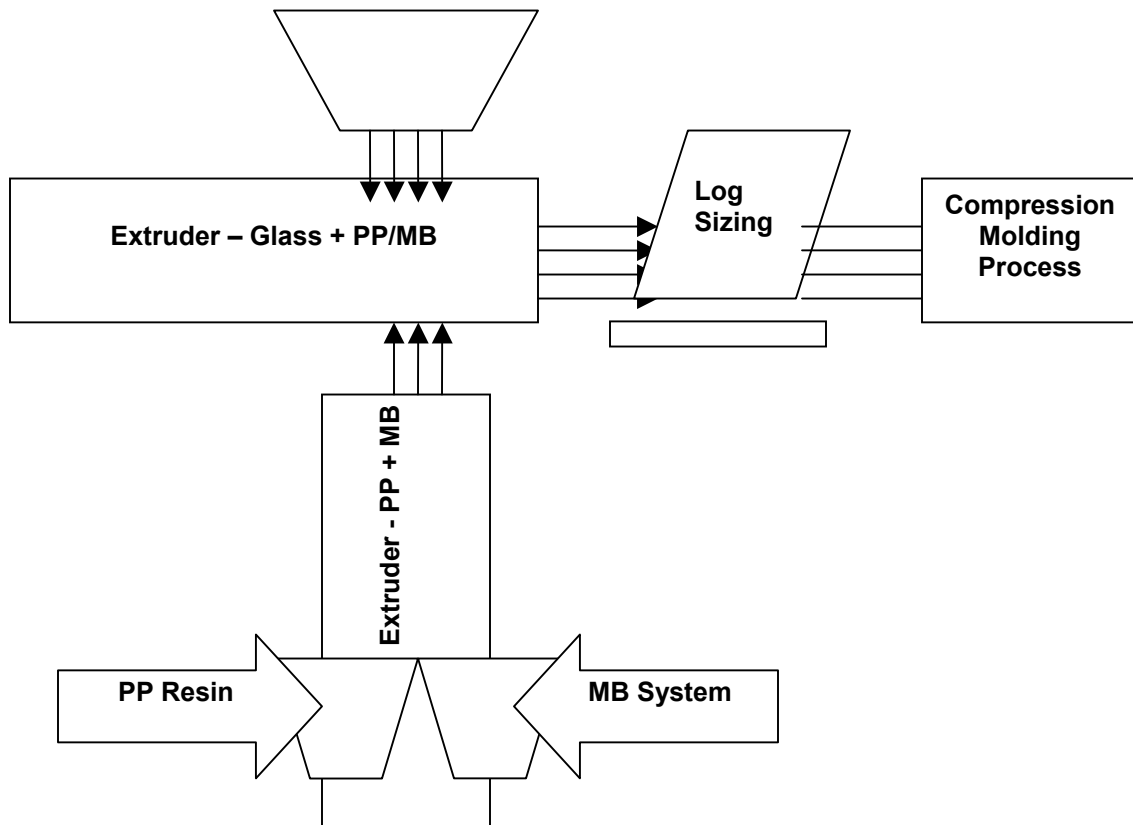
Table II – Injection Moldable LGF-PP Concentrate Pellet Solutions

| LGF Pellet Concentrate² | Glass Loading @ 11 mm LGF Pellet | Let Down Ratio % | Automotive Applications |
|---|---|-------------------------|--------------------------------|
| DLGF 9610.00Z | 60% | 33% | Equivalent to DLGF 9201 |
| DLGF 9610.00Z | 60% | 50% | Equivalent to DLGF 9311 |
| DLGF 9610.00Z | 60% | 67% | Equivalent to DLGF 9411 |

Direct LGF-PP Process and Products

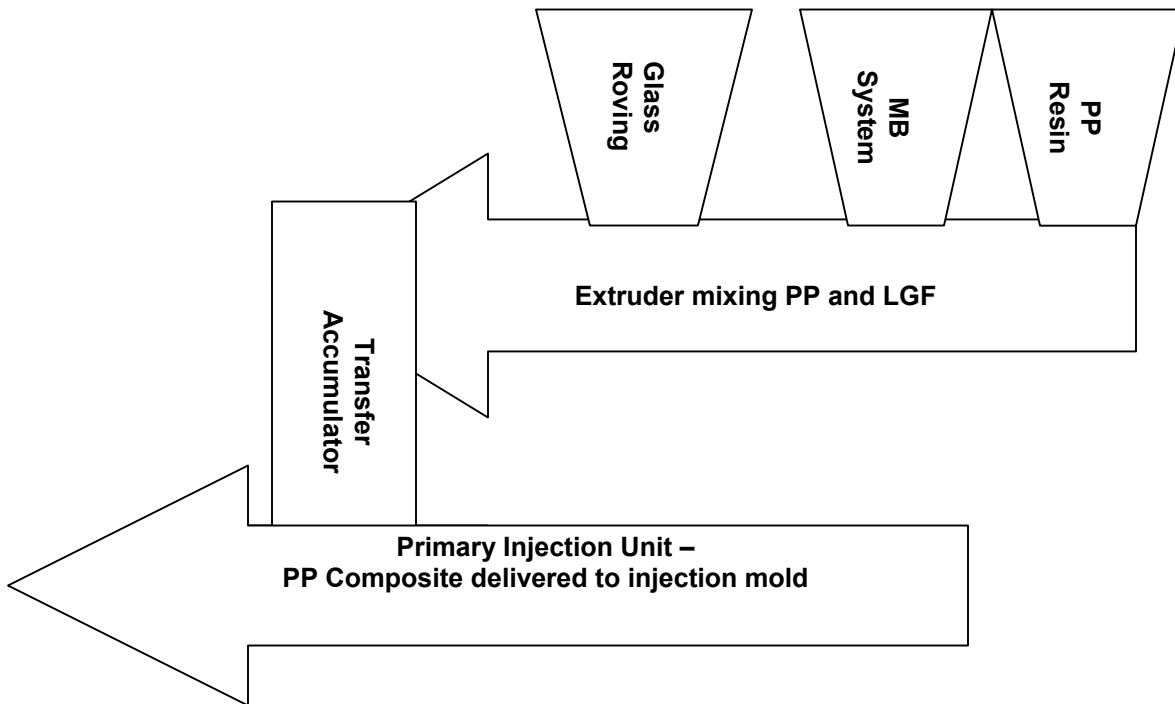
Direct processing of long glass fiber composite materials are unique because the compounding phase of the composite material is incorporated into the part molding process. The direct long glass fiber process incorporates chopped glass or continuous glass roving added to the resin and then delivered to the compression or injection mold.

Figure 1 – Example of Direct Compression Process



² Supplier: Dow Automotive

Figure 2 – Example of Direct Injection Process



As represented in Figures 1 & 2, direct long glass fiber composite processing enables the molder to also act as the material compounder. However, final part performance still requires that direct processes use engineered masterbatches to add the proper functionality to the final composite matrix. For example, engineered masterbatches provide critical functionality to the part such as glass coupling, heat resistance, impact resistance, color match and weathering resistance. The functionality contained in these masterbatches allow molders to use direct processing equipment to meet the application requirements of underhood, underbody, interior structural, exterior structural, and exterior structural molded in color parts. For direct long glass fiber processes, Dow offers system solutions for our customers. System solutions include a natural resin and an engineered masterbatch to meet the application requirements and are added during the direct LGF process.

Direct compression and direct injection processes provide a longer glass fiber length in the molded part compared to pellet injection. Comparing direct processes, injection does not result in as long of glass fiber as compression. System material solutions allow direct processing molders to meet the application requirements of underhood, underbody, interior structural, exterior structural, and exterior structural weatherable molded in color parts.

Table III – Direct Extrusion/Compression or Extrusion/Injection PP Material System Solutions

| Direct Processing System Solutions | MB Functionality | LGF Loading | Applications |
|---|---|--|---|
| DLGF 6500 | Thermal stability, impact modified, coupled, UV, color matched | Current 30% Can range as required (20,30,40) | Exterior Claddings, Running Boards, Step Assists |
| DLGF 7500 | Thermal stability, coupled, modulus | Current 40% Can range as required (20,30,40) | Front End Carriers, Structural Applications |
| DLFG 8500 | Thermal stability, coupled, toughened | Current 30% Can range as required (20,30,40) | Front End Carriers, Structural Applications |

Table above shows, there are currently three different material systems designed for direct LGF-PP processing. One can see the progression of functional requirements; the most advanced being the exterior molded in color system.

The focus of this paper is to discuss the development of UV stabilized, color matched, and molded-in-color long glass fiber polypropylene for exterior applications. Using Six Sigma methodology, Dow Automotive optimized the exterior DLGFPP composite system by understanding the application requirements and developing stabilization methods to meet those needs.

Definition of Performance Criteria

Working with our customers, performance requirements were benchmarked from exterior molded-in-color (MIC), weatherable part applications in both automotive and other durable markets. The target in the Table below represents a desirable balance of stiffness, impact resistance and weather resistance to meet a broad spectrum of applications.

Table IV – UV Stable, MIC, Impact Modified DLGFPP Performance Targets

| Application Requirements: | Criteria Targets: |
|-----------------------------------|--|
| Impact Resistance (IDI 3mm thick) | 13J peak energy |
| Tensile Modulus | 55 MPa |
| Flexural Modulus | 4260 MPa |
| Weatherability (SAE J1960) | 3 delta E after 2 yr equivalent exposure |
| Color Match | 1 delta E as molded |

Development of Weather Resistance Technology for LGF-PP

Typically, the pathway to failure via weathering for PP shows an initial discoloration followed by surface chalking then part failure due to PP degradation. Glass reinforced PP is known to be more difficult to stabilize against weathering than non-glass filled resins. Accordingly, the UV stabilization needs to be adjusted to compensate for the addition of long glass. Various accelerated weathering methods have been used to measure the weathering stability and develop UV stabilization packages for PP.

SAE J1960 is commonly used as a standard weathering test method for automotive exterior parts. This test utilizes quartz/borosilicate filters and a Xenon Arc light source. Customer specifications often require less than three delta E color change over a 2500 kJ/m² test cycle. Other commonly used accelerated weathering tests include an ASTM G155 Xenon Arc method and EMMAQUA. The ASTM G155 test utilizes borosilicate/borosilicate filters, while the Atlas EMMAQUA (ASTM 90-98) test uses mirrors to concentrate natural sunlight. Key conditions are summarized in Table I. Key criteria include measuring changes in color, surface chalking, and polymer molecular weight at the surface.

Table V – Key conditions and specifications for accelerated UV tests

| Conditions | EMMAQUA | SAE J1960 | ASTM G155 |
|-------------------------|---|---|---|
| Energy Requirement (kJ) | 576 000 (at 290-385 nm) or 6340 (at 340 nm) | 2500 (at 340 nm) | 2500 (at 340 nm) |
| Light source | Magnified Natural light (5 times) | Xenon Arc with quartz/boro filter (cut off at 270 nm) | Xenon Arc with boro/boro filter (cut off at 280 nm) |
| Light cycle | Day/night | Light (2 hr)/ Dark (1hr) | Light (2 hr)/ Dark (1hr) |
| Water spray cycle | 52 min dry/spray 8 min , 8cycle in day time and 3cycles/night | 40 min light followed by 20 min light with specimen spray followed by 60 min light, followed by 60 min dark with rack spray | Same as in SAE-J1960 |

It was noted that the performance of long glass fiber PP in these three tests show significant differences in failure rate. Figure 3 shows the comparison between the three tests with a black-pigmented sample. As noted in the figure, the severity of the methods increases in the following order: EMMAQUA < ASTM G155 < SAE J1960. Care must be taken in interpreting the results from each test method. The main difference is the light source and the filter cutoffs. Since the EMMAQUA light source is a magnification of natural light, it is reasonable to conclude that it will be closer to mimicking real time weathering than the artificial light from the Xenon Arc that includes higher amounts of short wavelength UV light. Further discussion comparing the performance of different UV packages will focus on the EMMAQUA test method.

Comparison Between Weathering Methods (Black Sample)

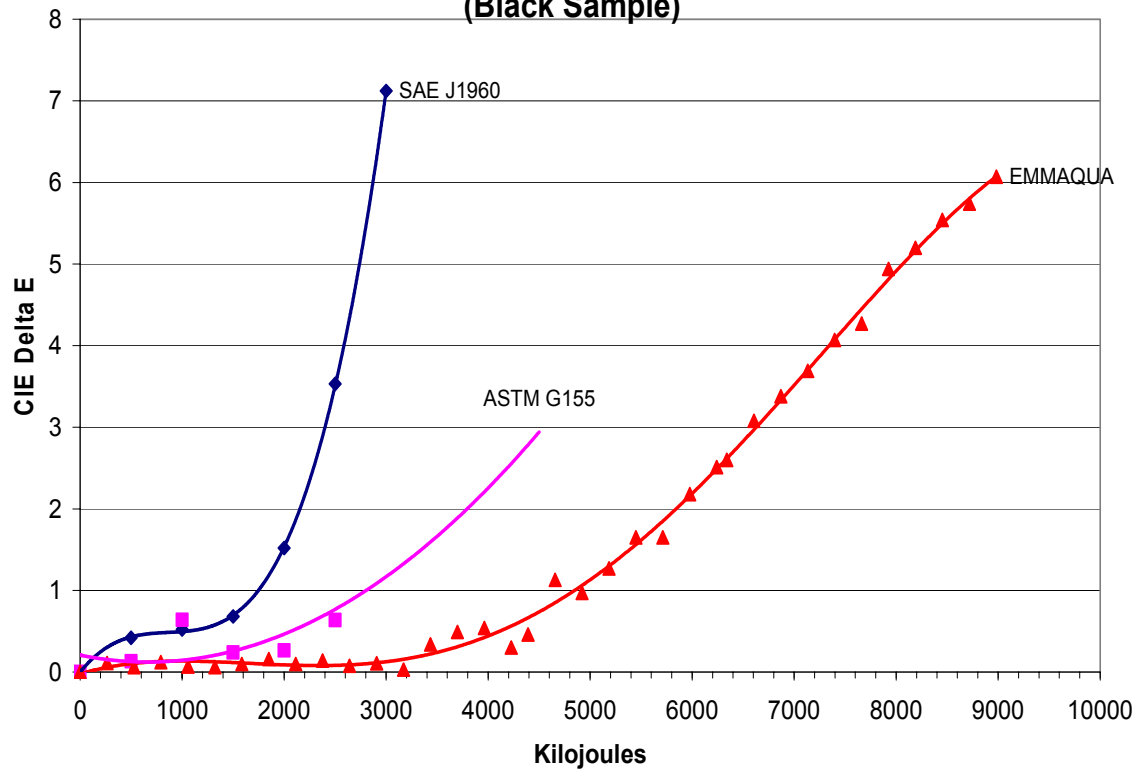


Figure 3 – Comparison of three methods and change in color with weathering. The EMMAQUA data was corrected to the energy at 340 nm to enable comparison to the Xenon Arc tests.

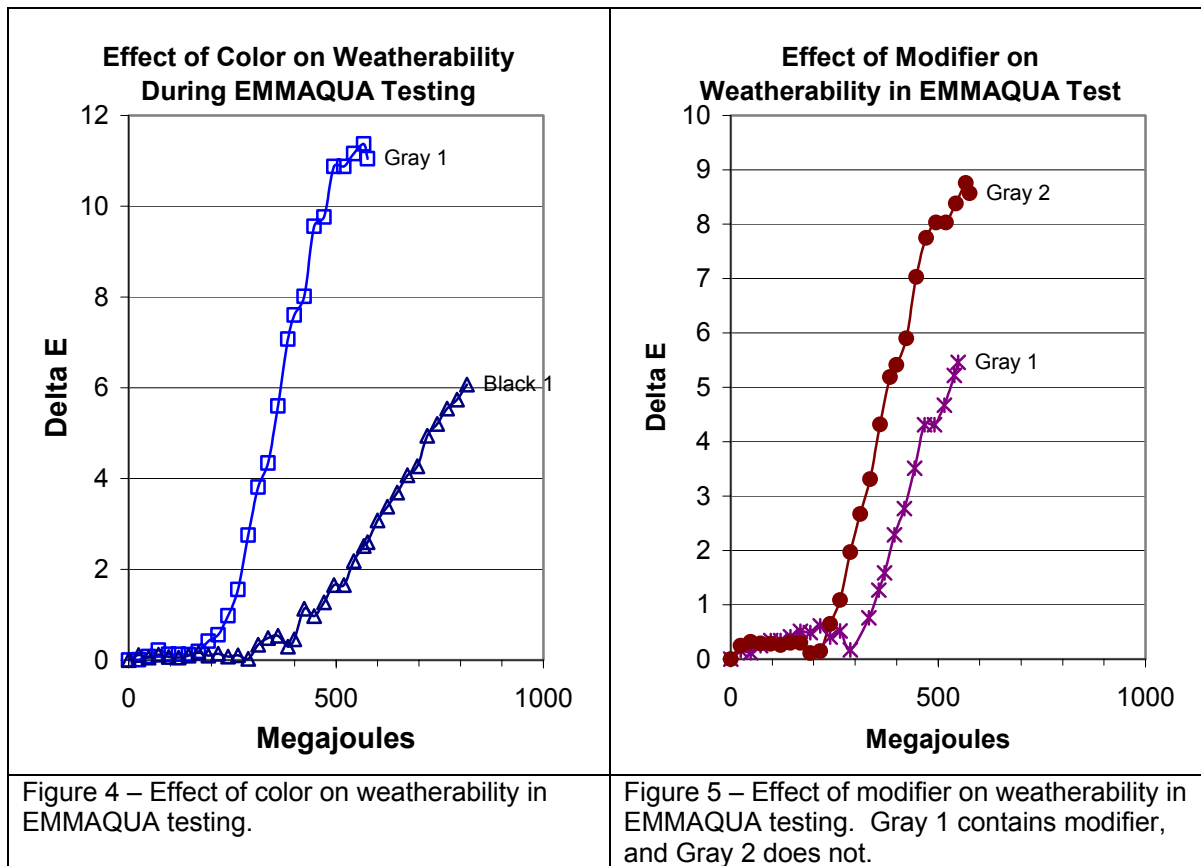


Figure 4 shows the effect of the pigment type on weatherability. It is well known in the industry that the addition of carbon black improves the weatherability of plastic parts. If white pigment is added, as in the Gray 1 sample, the weathering performance is significantly reduced. As mentioned previously, the addition of glass reduces the UV stability. Figure 5 shows that adding a modifier to the formulation improves the weatherability.

Sample Gray 3 in Figure 6 shows a typical UV stabilization formulation for PP used in automobile exterior applications which did not meet the performance discoloration criterion ($\Delta E < 3$ after 576 MJ). A special UV formulation was developed for the glass reinforced PP to meet the customer's criterion, Gray 4.

Through our understanding of the failure mechanism and the materials science of these compounds, we have developed a UV package that will meet the customer's criteria for pigmented, glass reinforced PP.

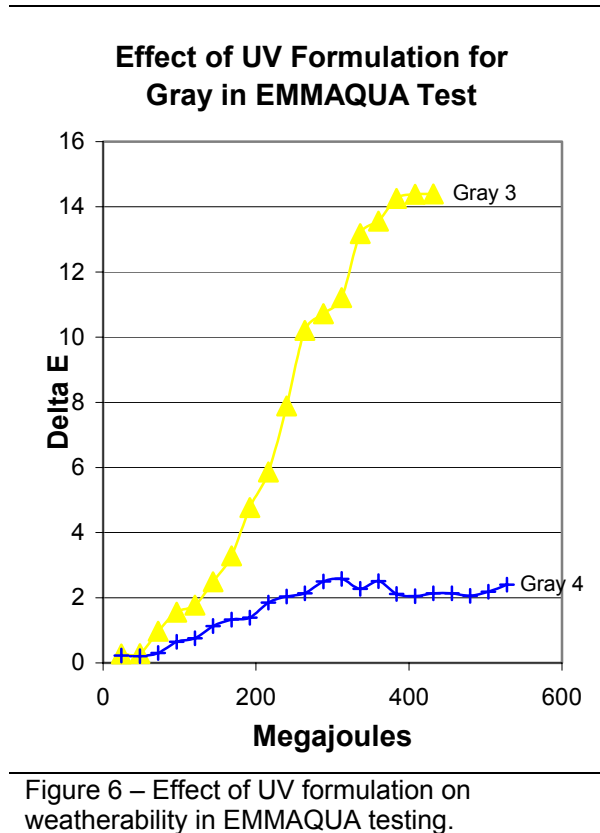


Figure 6 – Effect of UV formulation on weatherability in EMMAQUA testing.

Summary Results of Development Project

Table VI – Actual UV Stable, MIC, Impact Modified DLGFPP Performance

| Application Requirements | Targets | Actual Performance |
|-----------------------------------|--|--|
| Impact Resistance (IDI 3mm thick) | 13J peak energy | 16J peak energy |
| Tensile Modulus | 55 MPa | 57.9 MPa |
| Flexural Modulus | 4260 MPa | 4402 MPa |
| Weatherability (SAE J1960) | 3 delta E after 2 yr equivalent exposure | Weatherable Black & Gray Solutions Identified. See Table 4 & 6 |
| Color Match | <1 delta E as molded | past <1 delta E as molded requirement |

Conclusion

By working closely with our customers, and applying sound material fundamentals, Dow Automotive has successfully developed UV stabilization technology for molded-in-color long glass fiber polypropylene system. Furthermore, the masterbatch/PP system offering met strenuous impact, stiffness and color performance in addition to weatherability. We are confident that this technology can be leveraged to other long glass fiber polypropylene systems, direct and pellet, as required for new automotive applications. A broad portfolio of Pellet and DLFG material systems assures that Dow Automotive material solutions meet numerous long glass fiber polypropylene composite applications.

References

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2. BRG Townsend, Inc., "Long Fiber Reinforced Thermoplastic composites," Copyright 2001, Copy # 378.
3. Atlas Material Testing Solutions, www.atlas-mts.com .