VISCOSITY AND SCREW ASSEMBLY EFFECTS ON MECHANICAL PROPERTIES OF GLASS FIBER REINFORCED NYLON COMPOUNDS

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Abstract

Short glass fibers are widely employed in reinforcement of nylon compounds to significantly improve mechanical properties. These properties are dictated by fiber length and orientation distributions in the compounds. Fiber breakage takes place during the compounding process. When the fibers break into sub-critical length, the reinforcement of the compound is limited. This paper used a full factorial design investigating the effects of nylon’s viscosity and twin screw extruder’s screw configuration on glass fiber length retention and mechanical properties of the final compounds.

Introduction

Nylon is the generic designation of a family of polyamides. They are widely used in engineering plastics applications because of their toughness, excellent mechanical properties, resistance to chemicals, thermal stability and easy processability [1]. Among the commercial polyamides products, nylon 6 and 66 are the major types. Globally, 45% of total nylon compounds production is used in automotive industry and this consumption trend is increasing due to the driving of replacing metal parts with the lighter weight plastics [2]. Short glass fiber reinforced thermoplastics are a cost efficient solution which provide a large freedom of injection molded shapes as well as sufficient stiffness. High strength and stiffness glass fiber reinforced nylon (with 45-65 wt.% GF) are often required to substitute structural parts such as automotive body panels, levers, internal and external mirror housing, steering wheels, seat frames, etc. The strength and stiffness of these material depend on the fiber properties and the quality of fiber incorporated. This is determined by fiber length and orientation distributions and by the way in which stress is transferred into the fibers from the polymer matrix. The nature of the stress transfer interface is influenced by physicochemical and geometric factors, which are controlled by the compounding technology used to manufacture the material.

Co-rotating twin screw extruder is commonly used to compounding glass fiber reinforced thermoplastics due to the effectiveness of glass fiber dispersion. The configuration of shear input elements is a critical factor in the compounding process which directly related to the shear intensity and the residence time. The stronger shear ability and longer residence time will cause fiber break easily. This generates excessive fiber ends which nucleate the cracks and result in premature failure of the polymer matrix. Therefore, the screw assembly has a great impact on the final performance of the compound. McNally and co-workers [3] discussed a method to evaluate the effect of compounding technology on the stress transfer interface (STI) in short fiber reinforced thermoplastics. They investigated different polymer systems and found that the STI efficiency method was capable of evaluate the relationship of compounding processing variables and compound properties. Bader and co-workers [4] reported an improved method to produce high strength fiber reinforced nylon 66 involving continuous coating of fiber tows. Jiang [5] investigated effect of screw assembly on the properties of glass fiber reinforced nylon 66 composite. They concluded that using reverse combing mixer element ZME can increase fiber length and dispersion.

Another aspect of the compounding process is fiber wetting characteristics. The matrix cannot perform its duties in the compound unless the glass fibers have been well wetted and have good interfacial adhesion to form a continuous phase. The strength of the compound depends on the ability of the compound to effectively transfer the load from the matrix to the fiber via shear at the interface. Nylon resin has good wetting capability in the melt and the sizing chemistry on the glass fiber also promotes wetting. However, the viscosity of nylon is an important factor in wet-out. If the viscosity is too high, the resin cannot penetrate the fiber bundles. Low viscosity resin will have better fiber wetting due to easier penetration and lower surface tension.

Polyamides are condensation copolymers formed by reaction of dicarboxylic acid with diamines or by ring opening lactams. They are linear polymer and the viscosity depends on their molecular weight. Low viscosity resin will have good fiber wetting ability but have less strength and impact resistance because of lower molecular weight. This paper examined nylon’s viscosity (relatively high and low) and two different screw configurations using a factorial design. The compounds mechanical properties are the responsive variables. The main and interaction effects of the two factors were studied.

Materials
Commercial prime grades of polyamide 6 and 66 were used. Two different viscosity of each grades were chosen to compound, which were denoted as HV and LV. Their properties are summarized in Table 1. Chopped E-glass fiber with diameter of 10mm and length 4mm was used to reinforce both nylon 6 and 66.

Table 1. Characteristics of Nylon 6 and 66 resin.

<table>
<thead>
<tr>
<th></th>
<th>Nylon 6</th>
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<th>Nylon 66</th>
<th></th>
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<tbody>
<tr>
<td>Grade</td>
<td>HV</td>
<td>LV</td>
<td>HV</td>
<td>LV</td>
</tr>
<tr>
<td>Relative viscosity</td>
<td>3.75&lt;sup&gt;a&lt;/sup&gt;</td>
<td>2.6&lt;sup&gt;a&lt;/sup&gt;</td>
<td>190&lt;sup&gt;b&lt;/sup&gt;</td>
<td>117&lt;sup&gt;b&lt;/sup&gt;</td>
</tr>
<tr>
<td>Density (g/cm³)</td>
<td>1.13</td>
<td>1.13</td>
<td>1.14</td>
<td>1.14</td>
</tr>
<tr>
<td>Melt flow rate (g/10min)</td>
<td>1&lt;sup&gt;c&lt;/sup&gt;</td>
<td>10&lt;sup&gt;c&lt;/sup&gt;</td>
<td>20&lt;sup&gt;d&lt;/sup&gt;</td>
<td>100&lt;sup&gt;d&lt;/sup&gt;</td>
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a: 96% sulfuric acid  
b: viscosity number in sulfuric acid unit cm³/g  
c: 235°C/1Kg  
d: 275°C/2.16Kg

Experiments

The polymer-glass systems used were a 35wt.% glass fiber reinforced nylon 6 and nylon 66. This weight percentage corresponds to a volume percentage approximately 18.2%. Prior to processing, all the nylon 6 and 66 were dried under a vacuum oven at 80°C for 4 hours. The moisture content of resin was kept under 0.015%. The compounds were prepared using a ZSK-25mm Twin Screw Extruder (Coperion) with L/D ratio of 48/1. The additive package was kept same for all the compounds and glass fiber were fed downstream using a side feeder. Several different compounding technologies and varied operation conditions were employed. The extruded pellets samples were injection molded into standard test specimens using a Battenfeld 88 Ton injection molding machine.

After conditioning at room temperature (test specimens were kept in a foil bag) for 40 hours, the specimens were tested according to ISO standards. The tensile tests were conducted using an Instron model 4467 universal testing machine. The flexural moduli were measured with an Instron model 4465 universal testing machine with a 64mm span according to ISO 178 standard. Notched and unnotched Charpy impact strength were tested using Istron 9050. Glass fiber length was measured under an optical microscope after burning a handful of granules in a muffle furnace at 600°C for a few hours to remove resin. An image analysis software was used to analyze the length and distribution data on the replicate 200 fiber specimens needed for statistical reliability.

Results and Discussions

Factorial design

Two factors full factorial design was generated using Minitab as shown in Figure 1. Nylon’s viscosity is one factor, -1 indicates low viscosity and 1 indicates high viscosity. Compounding technology is the other factor, and similarly -1 and 1 represents a different processing set up respectively. 8 compounds were produced using random combination of the two factors with one replicates of each combination for both nylon 6 and 66 systems. This design allows for assessment of main and interaction effect of viscosity and compounding methods on final compounds’ mechanical properties (response variables), as well as estimation of experimental errors.

Glass reinforced nylon 6 mechanical properties

Figure 2 shows individual tensile strength and glass fiber length values of 35% glass fiber reinforced nylon 6 compounds produced with varying nylon’s viscosity and compounding methods. Tensile strength was in the range of 160-178Mpa in these Nylon 6 system. It was relatively higher in low viscosity system and achieved highest value in combination with (-1) compounding method. However, this property was not significantly affected in high viscosity system and the values were similar (~165MPa).

![Figure 1 Factorial design cube plot](image1)

![Figure 2 Tensile strength (Mpa) and Glass fiber length (μm) of Nylon 6 GF35 (average values with intervals)](image2)
Figure 3 plots main and interaction effects of the two factors on compounds’ tensile strength. The viscosity effect is statistical significantly (p-value=0.012<0.05) on this response. The compounding technology factor is not as significant on tensile strength (p=0.149). Similarly, the interaction between these two factors was not as significant either (p=0.099). Overall first order model fitted well (p=0.031) and linear relation is major (p=0.024). Tensile strength can be predicted using the following regression equation (in uncoded units):

\[
Tensile \text{ strength} = 169.493 - 4.225 \text{ Viscosity} - 1.745 \text{ Compounding} + 2.087 \text{ Viscosity} \times \text{ Compounding}
\]

(1)

Where \(\sigma_f\) is the ultimate tensile strength of the fiber, \(d\) is the fiber diameter, \(\tau\) is the interfacial shear strength between the fiber and matrix. This relation was defined under the assumption that both the matrix and fibers behave as linearly elastic materials [6]. The critical fiber length is the minimum length required for the maximum fiber stress to achieve the ultimate fiber strength at its mid-length. The calculated critical length for this Nylon 6 system is 263\(\mu\)m using a fiber strength of a fiber strength of 2.4Gpa and tensile strength of unfilled polymer matrix as interfacial shear strength [7]. The measured distribution showed about 70% of fiber is below this value. Although the fiber length plays an important role in final compound’s tensile strength, the orientation of fiber is also considerable. If all of the fibers aligned with the direction of service stresses in the injection molded tensile specimen, it can achieve as high as 430Mpa calculated using a simple law of mixture rule [8]. In this design of experiment, low viscosity system showed better retention of glass fiber length, however it is not statistically significant. In fact, both of the factors as well as the interaction between the factors are not significant (p>0.05).

The difference between the two compounding methods is the mean residence time, which was determined by screw elements assembly. -1 indicates shorter residence time and 1 is longer. Long residence time and high shear intensity will result in high amount of fiber breakage. To achieve desired mechanical properties of glass fiber reinforce thermoplastics, long fiber length retention (low fiber breakage) and good fiber wet out are critical aspects. However, good fiber wetting and high fiber breakage are both promoted by high shear intensity during compounding. In this study, the screw assembly was specially designed to balance these two opposing effects and varied the mean residence time. Although, the two assembly did not have a statistically significant effect on nylon 6 compounds’ tensile strength, the shorter residence time (-1) assembly did show slightly advantage.

The low viscosity resin system retained longer glass fiber length, which is due to better wet-out of glass. The longer glass fiber length results in higher tensile strength. The glass fiber length in both high and low viscosity system are in between 215 to 250\(\mu\)m, which are decent fiber length retention but still below the critical fiber length \((L_c)\) in Nylon 6 system. The critical fiber length can be estimated using Eq. (2)

\[
L_c = \frac{\sigma_f \times d}{2\tau}
\]

(2)

Figure 3 Main and interaction effect on tensile strength of PA6GF35

Figure 4 Unnotched charpy impact strength (Kj/m\(^2\)) of PA6GF35 (ambient temperature)
Other mechanical properties were analyzed in the same way such as flexural modulus, notched and unnotched charpy impact strength. Viscosity and compounding methods did not statistically significantly affect flexural modulus and notched charpy impact strength of the Nylon 6 system. The unnotched charpy impact was influenced by Nylon’s viscosity (p=0.029). Figure 4 shows the individual and mean values and Figure 5 shows main and interaction effects. High viscosity resin system has higher impact strength. This is due to high viscosity Nylon 6 has higher degree of chain entanglement, which provides impact resistance. The viscosity factor was more pronounced on unnotched charpy impact than the notched charpy impact response.

Glass reinforced Nylon 66 mechanical properties

The similar design of experiments were conducted on Nylon 66 systems. Figure 6 shows individual and mean tensile strength and glass fiber length values. Tensile strength is relatively higher than Nylon 6 system, which is in the range of 180-195Mpa. Similarly, the glass fiber length is longer than Nylon 6. The calculated critical fiber length for Nylon 66 system is 273μm and the distribution analysis showed that about 70% fibers are shorter than this value. Unlike the Nylon 6 system, both the viscosity and compounding method factors did not significantly influence tensile strength of the compound. The tensile strength of high viscosity resin system is slightly higher but not statistically different (p=0.832). It might be due to the viscosity difference between the two Nylon 66 reins are not as drastic as Nylon 6 resin. Glass fiber length was retained slightly better in high viscosity system. The shorter residence time showed less fiber breakage in both systems.

Compounding method affected flexural modulus in Nylon 66 system significantly independent of resin’s viscosity (p=0.025). Shorter residence time showed advantageous effect not only in length retention but also the dispersion of fiber in the polymer matrix. Especially when using low viscosity resin and in combination with short residence time screw design, the flexural modulus achieved highest value. Figure 7 shows the individual and mean values and Figure 8 shows the main and interaction effect plot. The regression model fit well (p=0.009) and the flexural modulus can be predicted by the following equation:

\[
\text{Flex} = 10330.8 - 45.7 \times \text{Viscosity} - 189.3 \times \text{Compounding} + 82.9 \times \text{Viscosity} \times \text{Compounding} \tag{3}
\]
Notched and unnotched charpy impact strength were measured. Notched charpy impact strength was not affected by the two factors but unnotched charpy impact strength was greatly influenced by resin’s viscosity (p=0.004). Higher impact resistance was obtained by using high viscosity resin. The same trend was also observed in Nylon 6 systems. The regression model fit better in Nylon 66 system (p=0.01), where linear relation is major (p=0.006). The interaction term has insignificant effect.

As shown in Figure 9 surface plot, the unnotched charpy impact value increases almost linearly with viscosity independent of compounding method. The unnotched charpy impact strength can be estimated using the following equation:

Unnotched charpy impact = 56.306 + 5.721 Viscosity – 2.592 Compounding + 0.509 Viscosity * Compounding (4)

Conclusions

The factorial design was adequate to evaluate the effects of resin’s viscosity and compounding method on glass fiber reinforced nylon compounds mechanical properties. Viscosity is a significant factor in determining the compound unnotched charpy impact resistance for both Nylon 6 and 66 systems. For Nylon 6, lower viscosity and shorter residence time screw assembly helped glass fiber wet out and length retention, leading to higher tensile strength. For Nylon 66, both viscosity and screw assembly did not affect compounds’ tensile strength but influenced significantly on flexural modulus. Depending on the injection molded part application requirements, appropriate combination can be selected to produce desired product.

Acknowledgement

The Author would like to thank Charles D. Park II, Process Engineer at A. Schulman, for screw design advice.

References

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