DEVELOPMENT OF NOVEL ONE-STEP HYBRID PROCESSING

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Abstract

The plastics processing industry typically must pre-compound using extrusion prior to part fabrication by injection molding (IM). The aim of this work is to implement a novel method that combines compounding and part fabrication into one processing step, thereby eliminating a costly, heat-intensive extra step. Poly (trimethylene terephthalate) (PTT) is blended with 10, 15, 20, and 30 % fiberglass (FG) by three methods, including standard IM, pre-compounding followed by standard IM, and a novel, one-step IM process using an innovative mixing screw design. The effect of processing method on the mechanical, impact, and thermal properties of a FG-PTT composite is presented.

Introduction

The plastics manufacturing industry is typically required to compound 1-5 % of a pelletized additives package into bulk polymer resin to fabricate plastic parts due to the poor mixing capability of single screw extrusion (SSE) and injection molding (IM) machines. The additives package is a pre-compounded concentrate of functional particles and base polymer resin that aids in mixing functional particles within the bulk resin. Although the additives package is the minor component, it is typically more costly than the bulk resin.

The base polymer in an additives package is often a low molecular weight polymer with poor mechanical properties. In addition, the base polymer is subject to two shear and heat histories, once during the pre-compounding step, as in extrusion, and secondly during the part fabrication step, as in IM. Subjecting the polymer to multiple processing steps has its disadvantages, including every time a polymer is subject to heat and shear forces there is potential for degradation, chemical or otherwise; multiple processing steps have been found to coarsen the morphology of a previously well mixed system[1)]; mechanical properties are dependent upon morphology; and particles may tend to agglomerate during extrusion. Additionally, multiple processing steps increase manufacturing costs and time.

The aim of this work is to eliminate the pre-compounding process step and the need for additives packages by developing a novel processing method to compound and fabricate parts in a one-step processing method. This novel processing method should achieve good dispersive and distributive mixedness and would allow powder or liquid pigments to be added directly into an IM machine with the bulk polymer resin. For comparison, the effect of three processing methods on mechanical, impact, and thermal properties of a hybrid materials system composed of a FG-PTT composite are investigated.

Background

Dispersing and distributing pigment, modifiers, filler, particles, reinforcing agents, and other various compounds within a polymer matrix are difficult. In most cases, twin screw extrusion (TSE) is commonly used for pre-compounding in order to achieve good mixing. However, single screw extrusion (SSE) offers several advantages, including low cost, rugged machinery more resistant to abuse, easy and inexpensive part replacement, widely available new or used, easy operation, no problems from high back pressures, and compounding and final product extrusion can be combined as a single operation [2)]. "It makes sense to compound using SSE whenever possible".

Industrial SSE use has lagged due to lacking multiple elongational flow fields as in multi-screw extruders, simple upstream axial mixing, and the ability to degas during mixing[3)]. To achieve good dispersion, surface treatments are employed to promote wetting by the polymer[4-6)] but have not been fully successful[7)] nor isolated the effect of mixing alone. Controlled feeding/melting mechanisms are used to decrease agglomerate formation to reduce the dispersion necessary for good mixing [2)]. To enhance distributive mixing, starve feeding may be used[5)], if the polymer is not subject to degradation[3)]. SSE is intrinsically limited in dispersive and distributive mixing but good dispersion can often be achieved by using specialized additives, whereas distributive mixing can equal any TSE compounder with retro-fitted mixing devices[2)]. The function of SSE has changed from only plasticating to both plasticating and mixing, achievable by adding a mixing element to the screw [8)].

There are several types of mixing elements suitable for SSE, each with their own advantages and disadvantages. For homogeneity, a combination of both dispersive and distributive mixing is optimal, specifically dispersion followed by distribution[9]. There are no standardized ways to evaluate the compounding ability of a mixer because it is difficult to quantitatively measure dispersion of filler particles in heavily filled thermoplastics[10]. Comparative studies have been performed in which different types of mixing elements are investigated to improve mixing of hybrid materials systems in SSE [2, 6, 8, 11-13]. And, there have been attempts to reduce manufacturing costs by improving the compounding role of SSE used in final product manufacture, specifically examining powders in polyolefins and typical liquid additives in various polymers[14]. However, it was determined that "SSEs are generally unsuitable for dispersive mixing powders into polymers" and several modifications are necessary to achieve distributive mixing of liquid additives in the polymer melt.

After years of experimental observation, the authors discovered a novel, high compounding mixer for use with SSE, termed an axial fluted extensional mixing element (AFEM)[15]. The AFEM promotes multiple elongational flow fields, upstream axial mixing, and thin film degassing. These attributes result in enhanced mixing of a variety of materials systems, including a Polystyrene/High-Density Polyethylene immiscible polymer blend, ceramic nano particle – Polymethylmethacryate (PMMA) composite, and discrete carbon nano tubes in PMMA[3].

The AFEM appears similar to the Maddock mixing element. However, the Maddock and AFEM perform differently and produce variant levels of mixedness. The key distinction is a subtle design difference. The Maddock entry flute dead ends (equivalent to a Union Carbide Mixer), while the flutes in the AFEM are open. The open flutes in the AFEM do not require high pressure and allow material flow to leave the mixer to continue down the length of the screw or to re-enter another flute and "recirculate" within the mixer again. This is a substantial difference regarding pressure and flow into and out of the compounding elements and has a profound influence on shear flow, degree of distributive mixing, and resulting mixedness and morphology.

Based on patented experimental success[16, 17] and collaboration[15], the authors incorporated the FEM into the screw of an IM machine in order to compound and fabricate parts in a one-step, novel IM process.

Materials

Two components were used for the experimental mixing study, including fiberglass (FG) and polytrimethylene terephthalate (PTT). The FG is typical micron-sized E Glass (d=20 microns, L = 4 mm). PTT is a unique thermoplastic polymer, manufactured by DuPont, based on 1,3 propanediol and contains 20-37 weight % renewably sourced material. Its beneficial properties, similar to high-performance polybutylene terephthalate, are derived from a unique, semi-crystalline molecular structure featuring a pronounced "kink". PTT has a melting temperature between 226-233 °C and a specific gravity of 1.3-1.5.[18]

Viscosity-shear rate for the PTT resin is shown in Figure 1 as a function of temperature. A frequency sweep from 100-0.01 Hz at 3.5 % strain and at temperatures of 240, 260, 280, and 300 °C was performed using a TA Instruments AR 2000. The viscosity-shear rate data was generated by performing a Cox-Merz transformation of the frequency sweep data at each temperature.

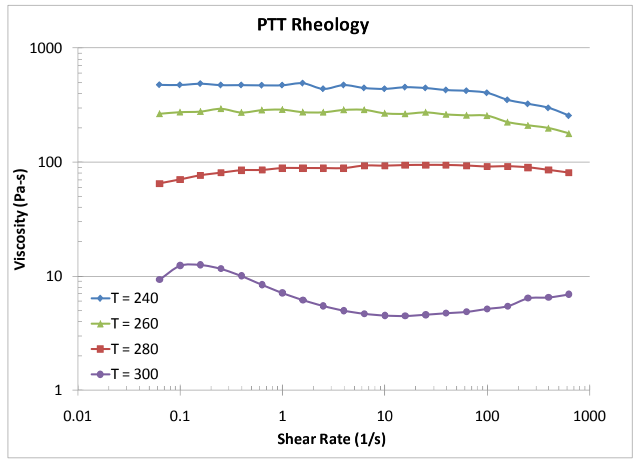

Figure 1. Shear rate vs viscosity for PTT at T = 240, 260, 280, and 300 °C.

Experimental Method

Three processing methods for producing a FG-PTT composite were compared and termed *Standard*, *2-Step*, and *Novel*. For each method, 0, 10, 15, 20, and 30 % FG in PTT were blended using a Negri Bossi V55-200 IM machine operated between 240-250 °C. The *Standard* method involved dry-blending FG and PTT in the selected composition ratios followed by melt blending using a standard IM screw in the IM machine. The *2-Step* method involved pre-compounding FG and PTT using a Randcastle Microtruder SSE fit with three AFEM elements, pelletizing, and a second processing step to achieve part fabrication using a standard IM screw in the IM machine. For the *Novel* method, the FG-PTT components were dry-blended followed by IM using a screw fit with one AFEM. The *Novel* method is a one-step processing method, in which compounding and part fabrication occurs in one processing step.

The FG-PTT composites produced by three processing methods were characterized by mechanical and impact properties. Tensile mechanical properties were determined using a MTS QTest/25 Elite Controller with a 5 kN load cell and extensometer, according to ASTM D 638. Modulus, ultimate tensile strength (UTS), load at UTS, percent strain at UTS, percent strain at fracture, and modulus were calculated. Izod impact properties were determined using an instrumented Instron Dynatup POE 2000 Impact Tester, according to ASTM D256.

Results

The mechanical properties in tension were determined and compared for the FG-PTT composite prepared by three different processing methods. The tensile modulus, ultimate tensile strength (UTS), % strain at fracture, and total energy absorbed are presented graphically as a function % FG in PTT in Figures 2 – 5, respectively. The *Standard*, *2-Step*, and *Novel* methods are represented by blue diamonds, red squares, and green triangles, respectively. The error bars indicate the standard deviation per sample. The 0 % FG samples did not fracture for all three processing methods therefore, the % strain at fracture is not shown in Figure 4. The total energy absorbed in Figure 5 corresponds to the energy absorbed up to the UTS.

The Izod impact properties were determined and compared for the FG-PTT composites prepared by three different processing methods. The impact energy and peak load as a function of % FG are shown graphically in Figures 6 and 7, respectively. The *Standard*, *2-Step*, and *Novel* methods are represented by blue diamonds, red squares, and green triangles, respectively. The error bars indicate the standard deviation per sample.

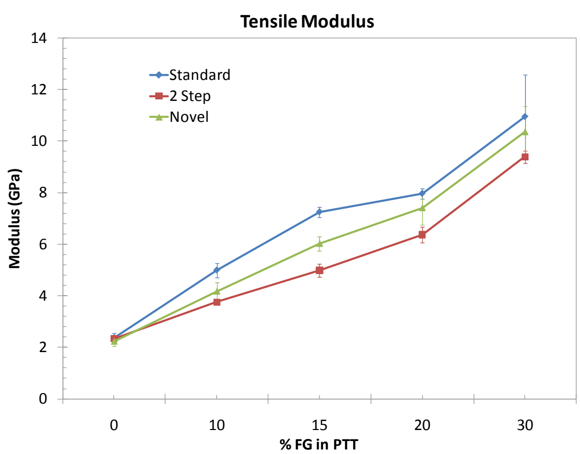

Figure 2. Tensile modulus comparison of FG-PTT processed by *Standard*, *2-Step*, and *Novel* methods

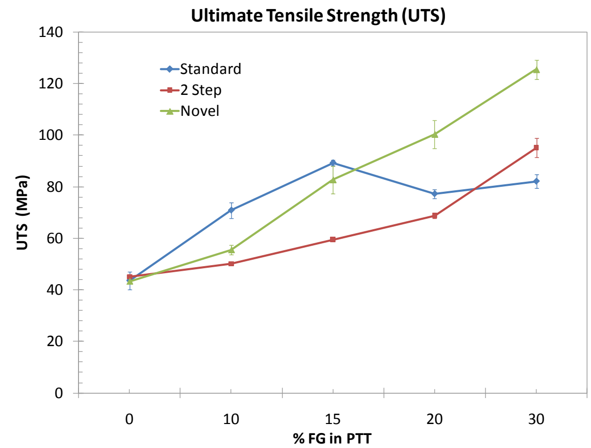

Figure 3. UTS comparison of FG-PTT processed by *Standard*, *2-Step*, and *Novel* methods

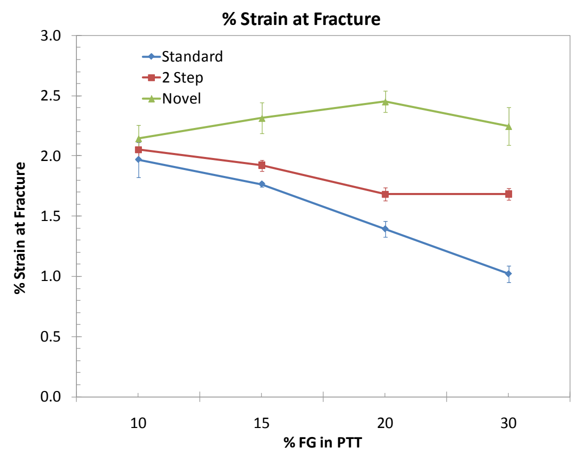

Figure 4. % Strain at fracture comparison of FG-PTT processed by *Standard*, *2-Step*, and *Novel* methods

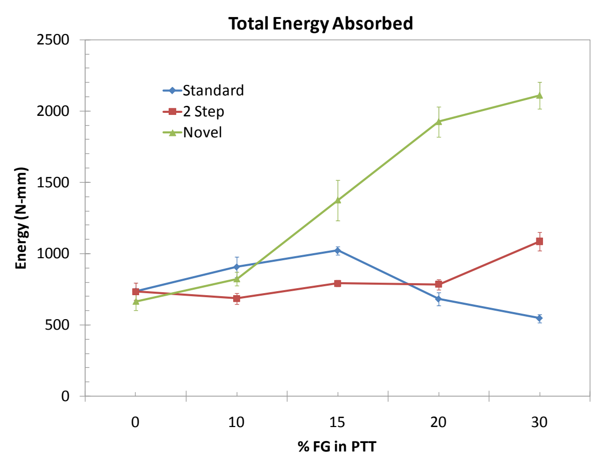

Figure 5. Energy absorption comparison of FG-PTT processed by *Standard*, *2-Step*, and *Novel* methods

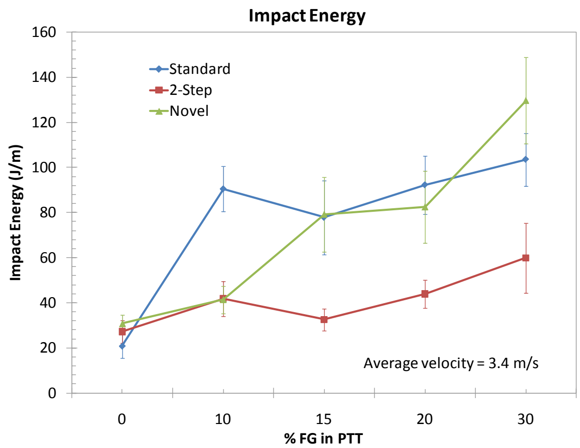

Figure 6. Izod impact energy comparison of FG-PTT processed by *Standard*, *2-Step*, and *Novel* methods.

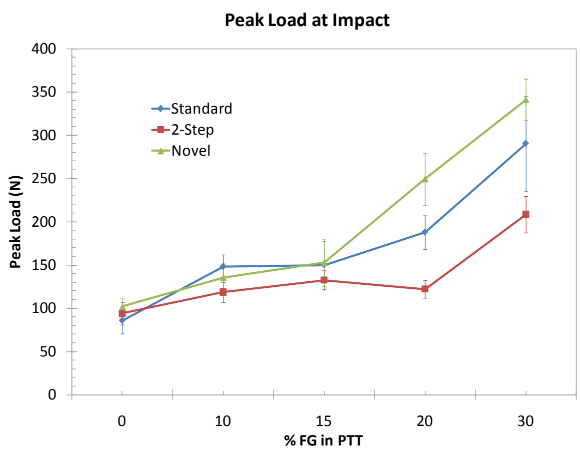

Figure 7. Peak load during impact comparison of FG-PTT processed by *Standard*, *2-Step*, and *Novel* methods.

Discussion

For all three processing methods, the tensile modulus increases with % FG in PTT from about 2.3 to 11 GPa (Figure 2). The *Standard* method produced a composite with the highest modulus for all compositions, followed by the *Novel* and *2-Step* methods. However, the differences at each % FG are not significant when noticing the standard deviation indicated by the error bars. The UTS increases with % FG in PTT for both the *Novel* (43-126 MPa) and *2-Step* (45-95 MPa) methods but only increases up to 15 % FG for the *Standard* method (44-89 MPa), as shown in Figure 3. The % strain at fracture decreases with % FG in PTT for both the *Standard* and *2-Step* methods (Figure 4). However for the *Novel* method, the % strain at fracture increases with % FG up to 20 % FG, remains above the 0 % FG value at 30 % FG, and is greater at all compositions than the % strain at fracture of the *Standard* and *2-Step* methods. The total energy absorbed increases slightly up to 15 % FG (740-1020 Nmm) for the *Standard* method and is below the 0 % FG value at 20 and 30 % FG (Figure 5). For the *2-Step* method, the energy absorbed is relatively constant from 0 to 20 % FG (averaging at 750 Nmm) and actually increases at 30 % FG (1090 Nmm). For the *Novel* method, the energy absorbed increases with % FG (665-2110 Nmm).

The *Novel* processing method produces a FG-PTT composite with enhanced ductility and toughness, as compared to the *Standard* and *2-Step* methods. Ductility is directly proportional to the % strain at fracture and toughness is related to the energy absorbed. Ductility and toughness are dependent upon the morphology and resulting mixedness. A fine morphology and good mixedness produces a composite with high ductility and toughness, while a coarse morphology or poor mixedness results in smaller % strain at fracture and less energy absorbed. This also applies to immiscible polymer blends when using the AFEM element.[16, 17)]

The AFEM incorporated into the IM screw for the *Novel* method produces very good dispersive and distributive mixing to impart enhanced mixedness. As the molton polymer enters the AFEM, the material is under little to no axial pressure. Material that enters the flute of the AFEM is elongated across the flute tip where it experiences almost completely pure shear with elongational flow, analogous to laminar plane flow. Uniform shear produces uniform, distributive mixing and high levels of mixedness. Once material exits the outlet flute, material may move axially downstream along the length of the screw or upstream and re-enter the AFEM for additional mixing.

The impact energy and peak load at impact increases with % FG for all three processing methods (Figure 6 and 7). The *Novel* method (31–130 J/m) incurs the most significant increase in impact energy, followed by the *Standard* method (21–104 J/m), and lastly, the *2-Step* method (27-60 J/m). The peak load at impact follows the same trend between all three processing methods, with the increase being most significant for the *Novel*, *Standard*, and then *2-Step* method. Upon observation of the fracture surfaces, it is evident that the fibers in the *Standard* samples de-bonded from the PTT matrix while the fibers in the *2-Step* and *Novel* samples sheared along with the PTT matrix.

Conclusions

A successful one-step processing method was developed and achieved a well mixed FG-PTT composite with enhanced ductility and toughness without sacrificing modulus and UTS. This method may be translated to polymer blends and other polymer-based composites to aid the polymer manufacturing industry to save costs and energy associated with traditional two-step pre-compounding followed by part fabrication manufacturing methods.

The *2-Step* method incorporated an AFEM in the screw of a SSE during the first processing step. However, morphology coarsening or de-mixing may have occurred after IM, resulting in lower modulus, UTS, and impact energy than the *Standard* and *Novel* methods. The *Standard* and *Novel* methods only use one processing step so de-mixing is not an issue. In addition, PTT is a condensation polymer and may be especially sensitive to two shear-heat histories as during the *2-Step* method.

Further, the FG and PTT matrix sheared during impact testing for the *2-Step* and *Novel* method samples, which may indicate that the FG was broken in the AFEM, since the materials tend to "recirculate" in the AFEM prior to continuing down the length of the screw. This would help to explain the enhanced energy absorbed, toughness, and ductility, particularly at 30 % FG. Future work will consist of characterizing the morphology of the blends by scanning electron microscopy methods to allow for a direct morphology comparison of the three processing methods and resulting FG length, distribution, and mixedness in the PTT matrix.

Key Words: Mixing, mixedness, novel processing, fiberglass, poly (trimethylene terephthalate), polymer composite, impact energy, tensile properties, AFEM

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