Optimized Extrusion Process for developing High Performance TPOs & Lightweight Polyolefin Composites.

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Abstract

Advanced technology in compounding thermoplastic polyolefin composites brings outstanding performance characteristics that make them highly valuable to automotive markets. Light weighting, high impact performance, low emissions are some of the key attributes that today's automotive market demands out of these TPOs and filled polyolefin compounds. However, it becomes highly challenging to make such a universal composite that shows a great balance of properties with equally high impact and high stiffness, low density and low emissions. The work presented here focusses on the development of a mineral filled polypropylene composite that exhibits high stiffness along with high impact properties and at the same time has a much lower density. This development included not only a balanced formulation approach but also an improved process design on twin screw extrusion (TSE) technology to achieve the desired balance of properties. The improved extrusion process was also utilized to convert certain batch process TPOs to a continuous TSE process while maintaining or enhancing the product properties.

Introduction

Light weighting has been one of the most important and hot topics in automotive industry over the last few decades. Especially with rising fuel costs, aggressive fuel efficiency targets and desires for larger vehicles such as SUV's, it has become imperative to incorporate advanced and engineered plastic resins as well as composites in automobiles. A 2018 report on Plastics and Polymer Composites in Light Vehicles, by American Chemistry Council [1], states that the average light vehicle today contains 342 pounds of plastics and polymer composites which accounts for 8.6% of the total vehicle weight. The usage of plastic components has increased from 20 pounds in 1960 to 190 pounds in 1990 and today there are more than 1000 plastic parts in a typical light vehicle. By volume, plastics make up almost 50% of the light vehicle.

Traditionally, metals like steel and aluminum have been used in cars, for structural components as they provide rigidity and impact resistance. In the initial time frame of the plastic revolution in automotive industry, polymer resins and composites were used to replace heavier metal parts, but more recently the focus has changed increasingly to replace existing plastic parts with even lighter polymer composites [2]. This trend is driven by the desire to have even more cost savings on part weight basis and with minimal risk of replacement of a plastic component with a further low density one. The focus of this research work also revolves around the development of a light polymer composite that would replace a heavier mineral filled polypropylene composite thereby accounting for a 7-10% density reduction by part weight. The development of this next generation light weight composite was mainly targeted towards an OEM specification which required an equal balance of high stiffness and high impact properties at room and cold temperatures and at the same time keeping the density below 1g/cm^3. The formulation strategy used in this development also helped in achieving low emissions as well as long term heat performance while creating a higher melt flow material for ease in molding large parts.

An important aspect of this development, in addition to the formulation strategy, was the choice of a unique process design on a twin-screw extruder (TSE) that is proprietary to LyondellBasell. Twin screw extruder's (TSE) are a standard choice in compounding industry for a wide variety of materials because of their continuous process capability, controlled residence times and throughputs with high stresses that enable substantial mixing, controlled temperature profiles over the entire barrel length and ease of feeding ingredients by advanced feeder systems [3,5]. However, the most important aspect in effectively using the shear dominated flow in TSE is by a careful choice of the type and geometry of screw elements and their positioning on the screw shaft to create a screw design that enables dispersive and distributive mixing. Such screw designs are very important especially in effectively dispersing small micron and Nano sized filler particles in high flow polymer resins. The development of such a unique screw profile not only helped in the development of this universal composite but also enabled the conversion of certain high performance TPO's from a batch to continuous process. Legacy A. Schulman TPO's (POLYTROPES) comprise of both filled and unfilled polyolefin compounds with low melt impact modifiers going in anywhere from 30 – 50 %. The large viscosity differentials between low melt impact modifiers and high flow polypropylene resins always created a challenge in dispersing the modifiers into the polymer matrix especially on a continuous TSE process with limited residence time. However, the development of the unique TSE profile and process enabled the effective dispersion and miscibility of the impact modifiers into polypropylene matrix, thereby improving the performance characteristics of the TPOs.

Materials

For the development of the high-performance light weight composite, a target OEM specification was provided which was typically designed for a 20-30% mineral filled polypropylene. A. Schulman already had a 20% talc filled PP composite approved to that spec with a density in the range of 1.05 g/cm^3. However, the OEM wanted a new development of a composite that would achieve the same physical properties or better and drop the density to around 0.98 g/cm^3. As such an array of light weight mineral fillers available in the market were trialed from nanoclay to fine micron talc to high aspect ratio (HAR) mineral as well. All these fillers were initially used to screen the best possible candidate in a polypropylene matrix.

Table 1. Fillers for DOE

	D50 (micron size)	**Bulk Density (Kg. / m^3)**
Filler 1	0.01	130
Filler 2	0.7	640
Filler 3	1.0	680
Filler 4	2.3	650

Given the target properties of high impact and stiffness, a combination of a copolymer and homopolymer polypropylene were used to form the polymer matrix. The copolymer (CoPP) was a No-Break, 25 – 30 melt (230ºC, 2.16 kg) resin with a density of 0.90 g/cm^3 available from Braskem. The homopolymer (HPP) was also a 35-melt (230ºC, 2.16 kg) resin with a density of 0.90 g/cm^3 available from Exxon. The usage of higher flow resins was consciously chosen as the OEM had a target melt flow spec of >20 g/10min (230ºC, 2.16 kg) on the final composite. The resins were varied in their ratios to achieve the most optimum combination in order to achieve the physical properties especially the impact resistance and modulus of elasticity in bending.

Table 2. Polypropylene Resins for DOE

	Melt Flow (230ºC, 2.16 kg)	**Izod Impact** (23 ºC)	**Flex Modulus** (1.3 mm /min)
CoPP	25 g/10min	No Break	1050 MPa
HPP	35 g/10min	26 J/m	1400 MPa

In addition to the development of this light weight high performance composite, as mentioned in the introduction, the development of the unique TSE process design also helped in converting impact modified TPOs from a batch (Banbury) to a continuous TSE process. The materials that were used in such TPOs were also homopolymer polypropylenes with high melt flow in range of 35 – 65 g/10min and had fractional to 1g/10 min (190ºC, 2.16 kg) melt impact modifiers at >30%.

Process

As mentioned earlier in the introduction, the success of this development relied heavily on the unique TSE screw design that was assembled for converting certain batch process TPOs to continuous TSE process. It is well documented that twin screw extrusion (TSE) is one of the most widely used technology by plastic compounders for blending of different polymers, fillers with polymers and even reactive extrusion [3]. The mixing in a TSE, mainly occurs via Distributive or Dispersive mixing. Figure 1 shows the effect and difference in distributive and dispersive mixing.

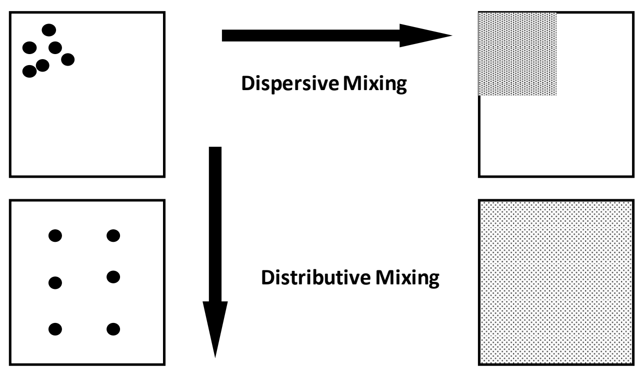

Figure 1. Distributive vs Dispersive Mixing

Both mixing aspects are equally important, especially when viscosity differentials are involved between the two components that are being mixed, be it a low melt resin in a high melt resin or a solid filler in a polymer matrix. Majority of the distributive and especially dispersive mixing occurs in the pressure driven flows generated via kneading blocks or elements. The choice of these mixing elements and their spatial arrangement is what allowed the creation of a unique screw design that enabed excellent dispersion of low melt (high viscosity) impact modifiers into higher flow (low viscosity) resins. Micrographs in figure 2. show a clear comparison in the dispersion of the low melt impact modifier into high flow PP when a general-purpose screw design was used versus the unique screw design that is proprietary to LyondellBasell. Micrographs on the left show that the low melt impact modifiers are not completely miscible in the high flow PP resin. These micrographs depict the usage of general-purpose screw design. Micrographs on the right show that by using the unique screw design, impact modifiers were completely miscible in the high flow PP resins.

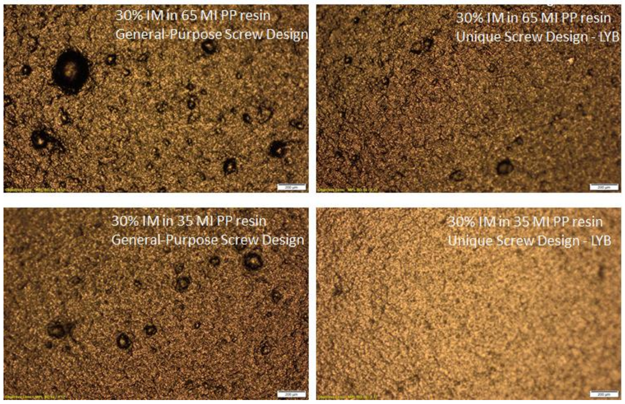

Figure 2. General Purpose vs Unique (LYB) Screw Design

Results and Discussion

For the development of the light weight composite, couple different DOE's were conducted to reach the optimal formulation that would hit all the target properties of the OEM. Some of the key properties are outlined in the specification table below.

Table 3. OEM specification requirements

Property	Units	Target
Density	g/ cm^3	<1.00
Melt Flow (230°C, 2.16 kg)	g/10min	>20
Tensile (30 mm/min)	MPa	>22
Flex Strength (30 mm/min)	MPa	>34
Flex Modulus (30 mm/min)	MPa	>2150
Izod Impact (23 °C)	J/m	>49
Izod Impact (-30 °C)	J/m	>20
Modified DuPont Impact (23 °C)	J	>9
Modified DuPont Impact (-30 °C)	J	>1
HDT (0.45 MPa)	°C	>130
HDT (1.80 MPa)	°C	>70
Rockwell Hardness	R	>85
Gloss (60°)	GU	<30

The two key properties that needed a balanced formulation approach were Izod impact and flexural modulus. It is well known, that these two properties can be antagonistic, in the sense that when trying to reach higher impact, impact modifiers are generally used which tend to reduce the stiffness or flexural modulus of the composite. Vice-versa, if the stiffness is increased then the impact generally is compensated. As such it was very important to find the right balance of CoPP, HPP and filler level to achieve both of those properties. In the first DOE of trials, only Filler 3 was chosen as that was a standard 1-micron particle mineral filler and the resin ratios were altered to see which combination gives the optimal result. Filler 3 was varied at 2 levels – (T1-T3) 10% and (T4-T6) 12% to keep the density of the composite below 1.00 g/ cm^3. It should be kept in mind that DOE 1 was started with a standard or general-purpose screw design on the TSE.The results from the first DOE showed that maxima was reached especially in stiffness with trial T6.

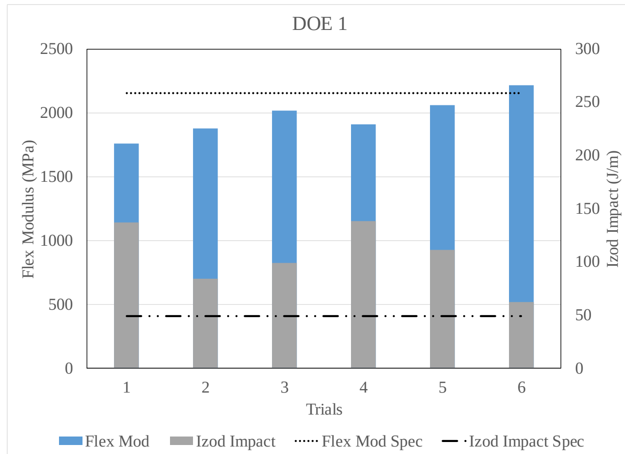

Figure 3. DOE 1– Flex vs Izod.

It can be seen clearly from figure 3 that the resin and filler combination of trial T6 was the optimum approach in balancing antagonistic properties of stiffness and impact. Even though the impact and stiffness from T6 passed the OEM specification, other properties like HDT and hardness failed. Figure 4 shows that HDT at both 0.45 MPa and 1.80 MPa are well below the specification level. It is well documented that the good dispersion of mineral filler is very important for creating nucleating sites which directly affect mechanical and thermal properties like tensile, flex and HDT. In DOE 1, as mentioned earlier, a general-purpose screw design was used, which may have not created efficient dispersion of the filler in the polymer matrix. This could have very well affected the HDT values as well the flex numbers were marginal to the spec.

In order to increase all properties substantially above the spec and to improve talc dispersion, the improvised screw design used for dispersing low melt impact modifiers into high flow resin was used in DOE 2. The concept in the dispersion of solid mineral filler into high flow PP remained the same as dispersing the low melt impact modifier. DOE 2 comprised of 9 trials which included all 4 fillers described in materials section with resin ratios revolving around + / - 5% of the trial T6 from DOE 1. Trial 1 in DOE 2 used Filler 1, which happened to be a high surfactant clay, with the smallest particle size. Given the high aspect ratio of nanoclay, only 5% of the filler was used, but this trial was later discarded because of the high yellowness imparted on a natural composite that was unacceptable.

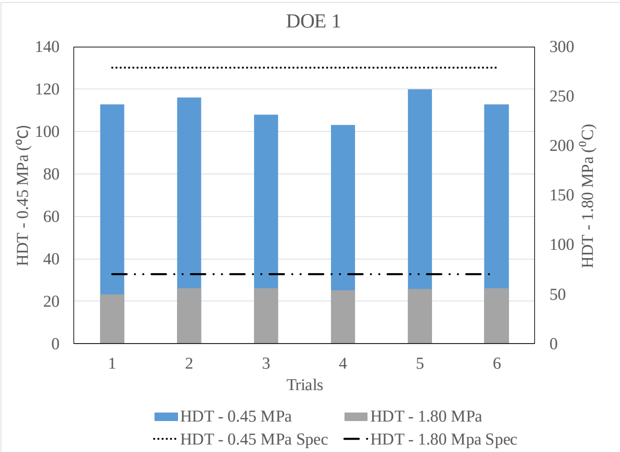

Figure 4. DOE 1 – HDT (0.45MPa & 1.80 MPa)

Trials 2 and 3 used Filler 2, trials 4,5 and 6 used Filler 3 and trials 7,8, and 9 used Filler 4. Even though filler 4 had the highest particle size of chosen fillers, it was also a high aspect ratio mineral. For this filler to be effective in the polymer matrix, it was imperative for it to be dispersed efficiently so that no agglomerates were formed. All trials from 2 – 9 used 12% of respective fillers.

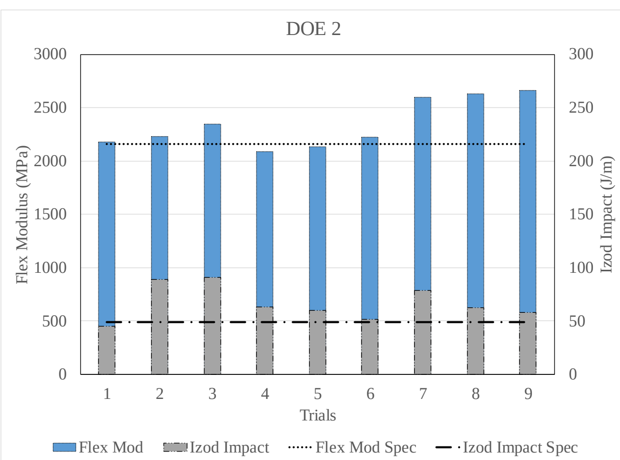

Figure 5. DOE 2 – Flex vs Izod.

Figure 5 shows that trials 7,8 and 9 which used Filler 4 have the highest stiffness and easily passed the Izod specification. Infact, Filler 2 (trials 2 and 3) also showed both stiffness and Izod pass the specification with impact being highest in both these cases. However, it was important that the chosen formulation should pass all properties in the table 3, well above the lower spec limit (LSL). In addition to the above two properties, just like DOE 1, HDT was plotted to identify the best possible candidate. Figure 6 shows the comparison of HDT at both 0.45 MPa and 1.80 MPa with respect to the specifications. Interestingly, T9 is the only formulation combination of resins and Filler 4 that passes the stringent HDT requirements. A complete set of properties from trial 9 are listed in table 4. It can be seen that the effective dispersion of just 12% of Filler 4 using the improvised TSE screw profile in the optimal resin combination helped in creating

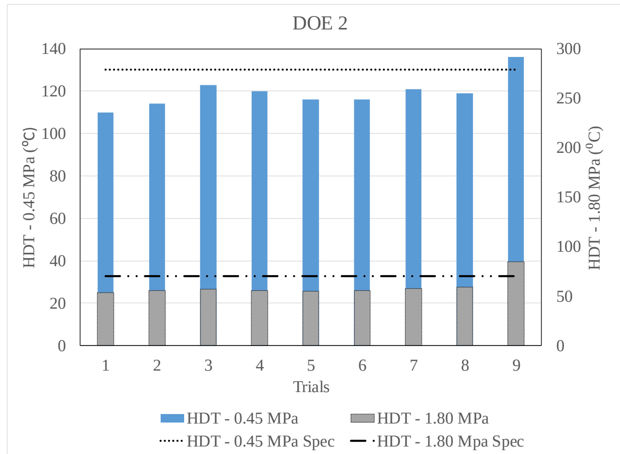

Figure 6. DOE 4 – HDT (0.45MPa & 1.80 MPa)

a perfect balance of properties that easily surpassed all the OEM specifications.

Table 4. DOE 2 – Trial 9 – Property comparison

Property	Units	Target	Data
Density	g/cm^3	<1.00	0.98
Melt Flow (230ºC, 2.16 kg)	g/10min	>20	26
Tensile (30 mm/min)	MPa	>22	29
Flex Strength (30 mm/min)	MPa	>34	48
Flex Modulus (30 mm/min)	MPa	>2150	2500
Izod Impact (23 ºC)	J/m	>49	84
Izod Impact (-30 ºC)	J/m	>20	38
Modified DuPont Impact (23 ºC)	J	>9	10.5
Modified DuPont Impact (-30 ºC)	J	>1	1.80
HDT (0.45 MPa)	ºC	>130	136
HDT (1.80 MPa)	ºC	>70	85
Rockwell Hardness	R	>85	99
Gloss (60º)	GU	<30	18

In addition to the mechanical and thermal properties, the correct choice of additives and process stabilizers also helped pass flammability, long-term light resistance (Xenon) and heat deterioration specifications.

Conclusions

The work presented in this study was targeted towards the development of a light-weight, high-performance, polypropylene composite that would show a balanced set of properties and exceed all the specifications requirements from the OEM. A careful choice of resins, filler and additives was necessary to achieve the target properties. However, more importantly, the improvised TSE screw profile was a critical factor in achieving the specification properties. It was clearly seen between the two DOE's (DOE 1 vs DOE 2), that the same filler when dispersed using the improvised screw profile showed better properties than a general-purpose screw design. The effect of dispersion was even more evident in high performance TPOs where a low melt impact modifier was dispersed efficiently in a high melt polypropylene resin. It was clearly seen in the micrographs, that the low melt impact modifier was completely miscible in the high flow PP making it a homogenous phase, when the improvised screw design was used. The usage of this TSE screw profile has been extended in the conversion of batch process commercial products needing a high residence time in a Banbury to more effective and efficient TSE process needing much lesser residence times.

References

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