Effect of length variation of flax/polypropylene pellets produced by multi-die pultrusion on the quality of injection molded product.

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Abstract

The aim of this study was to investigate the length variation effect of flax/polypropylene (flax/PP) pellets on the quality of injection molded final product. Pultruded rods of 4.76 mm in diameter were produced using multi-die pultrusion. The fiber volume content of the rods was 50% and the void content was around 3.5%. Rods were pelletized in three different lengths of 6, 9 and 15 mm. Three sets of dogbones coupons were injection-molded using the three different pellet sizes. The flax/PP pellets were blended with polypropylene in the injection molding machine to lower the fiber weight fraction to 25%. The injected dogbones were characterized by tensile and charpy impact tests to evaluate the mechanical strength. The impregnation quality and fiber distribution in pultruded rods were characterized by void content measurements, and optical microscopy. Results showed that pellets from the well consolidated pultruded rods were consistent in shape and size. The flax fiber distribution into the injected dogbones was uniform, whatever pellet length used. Finally, the usage of longer pellets significantly increased impact strength of the injected coupons. Tensile properties were also slightly improved when longer pellets were used.

Keywords: Biocomposites, thermoplastic, pultrusion, injection molding, flax, polypropylene

1.0. Background and Requirements

Inorganic fillers such as glass fibers, talc and mica are widely used in the in the automotive industry [1]. However, production and disposal of such materials have a negative impact on the environment. Due to the rising environmental awareness, new environmental regulations have been imposed to help reduce the inorganic waste problem [2]. The new environmental regulations are putting tremendous pressure on automotive industry to replace inorganic fillers with biodegradable alternatives. The automotive industry is actively trying to expand their use of natural fiber-based composites such as polypropylene/flax (PP/flax). In addition to being environmentally friendly, natural fibers are light. They are inexhaustible since they can be harvested every year in an environmentally friendly way. Among other natural fiber candidates, flax fibers are preferred for industrial applications since they have a low lignin content (2%) and a very high cellulose content (64%)[3]. Lignin starts degrading at a temperature around 160 ℃, whereas cellulose starts degrading in the temperature range of 260-350 °C. The high cellulose content allows flax fibers to be processed at higher temperature compared to other natural fibers [4]. Furthermore, flax fibers have higher tensile and stiffness properties compared to other natural fibers. This superiority in strength for flax fibers is due to the fact that flax fibers have the longest elementary fibers compared to other natural fibers [5]. In fact, specific Young's modulus for flax fibers is higher and the specific tensile strength is comparable with E-glass fibers [6]. Therefore, flax-based composites offer a great alternative to replace inorganic fillers from materials such as glass fibers and talc and mica particles.

PP resin is the most commonly used resin in automotive applications [7]. PP is preferred in automotive applications due to its low density, great processability, excellent dimensional stability, mechanical properties and impact strength [8]. PP is low cost and its melting temperature is below the degradation temperature of natural fibers [9]. PP compounds are being used in all kinds of parts from bumper facias to instrumental panels. Inorganic fillers are added to PP to improve its performance. The inorganic fillers can vary in length depending on the application. For example, short-length glass fibers are added to PP to increase the threshold of heat resistance to a temperature close to melting temperature of PP. Typically, short glass fiber reinforced PP are produced using a twin-screw extruder in which the matrix and chopped fibers are melt-mixed in the extruder. During extrusion, the high shear forces lead to major fiber breakage [10-12]. On the other hand, Long Fiber Thermoplastic (LFT) are molded using pellets where the reinforcing fibers are as long as the pellet length. LFT pellets are usually manufactured by pultrusion. Continuous glass fibers are pulled from glass-fiber rolls into an impregnation die. The molten resin is then supplied directly to the impregnation die. The glass fiber is impregnated by the molten resin. After that, the hot composite precursor passes through a cooler then cut into long pellets of 5-40 mm length [7].

When load is applied on a composite part, internal stresses are transferred to the fibers via shear stress at the fiber/resin interface. If the fiber length in the composite material is smaller than the critical length L_c, then the composite will fail by both fiber breakage and interface slippage simultaneously [13]. Conversely, if the fiber length in the composite material is larger than the L_c, then the fibers will fail by fiber breakage only [13]. Therefore, in order to fully utilize the strength of the fiber, the fiber length should be longer than critical length L_c. Using LFT as injection moldable pellets is proven to significantly improve impact/stiffness as well as creep, fatigue, high temperature properties and surface finish [14]. Feeding flax in the fiber-form into an injection molding hopper is problematic because it will cause fiber agglomeration and entanglements [5].Thus, a compounding method of flax and PP is necessary to manufacture LFT to be used as injection moldable pellets. Barkoula *et al.* [5] studied short & long flax/PP pellets to be used for injection molding. Combining flax and PP pellets was done by an extruder to produce short flax/PP pellets and by pultrusion to produce flax/PP LFT. The injection molding led to a fiber length reduction in all scenarios by about 30%. To estimate the critical length, L_c was characterized using Kelly-Tyson theory [15]. The fiber elementary strength was estimated using literature to be 1500 MPa. Interfacial bond strengths were also retrieved from literature as 8 MP for flax/PP and 16 MPa for flax/MAPP. The critical lengths of flax/PP and flax/MAPP were calculated to be 1.2 mm and 0.6 mm respectively. Moreover, Barkoula *et al.* observed that using pellet length that are higher than L_c may result in better impact strength as it may increase energy absorption via fiber fracture and debonding [5].

Another pultrusion technique recently developed is the multi-die pultrusion system. In this process, the resin and reinforcement fibers are pulled together through a cascade of dies [16]. The dies are heated to liquefy the resin fibers. To properly impregnate the reinforcement fibers, the yarns entering the heating dies should contain excess resin fibers. This excess resin is flushed through the die entrance in a so-called backflow. This excess backflow helps increase the impregnation pressure in each die [16-19]. Oswald *et al.* [18] used a double-die system to pultrude flax/Polylactic acid rods. A difference of 10% in die exit diameter from the first to second die, was used to guarantee a backflow in both dies. The void content of the beams pultruded with the double die system was 4%, compared to 12% when pultruded with a single die with the same conditions. Lapointe *et al.* [16] used four-die vacuum-assisted pultrusion system to pultrude Carbon/PEEK hybrid yarns rods. With this system, the lowest void content achieved was 1.7% [16].

The first objective of this study was to demonstrate the use the multi-die pultrusion system to produce flax/PP LFT injection moldable pellets. The second objective was to investigate the effect of different pellet lengths on mechanical properties of injection molded products.

2.0. Materials and method

2.1. Materials

Table 1 outlines the materials used in this study and their properties. Flax yarns having fineness of 200 tex were used (Safilin, France). PP-MAPP yarns of size 133.3 text were made of PP blended with 2 wt.% maleic anhydride polypropylene (MAPP) coupling agent (Guangzhou Chemical Fiber Co., China). PP pellets (PP 1043 N) as well as MAPP pellets (Guangzhou Chemical Fiber Co., China) were used for injection molding process.

Table 1 : Flax, MAPP and PP properties.

Material	Linear density (tex)	Solid density (g/cm^3)	Melting temperature
Flax Roving200	200	1.53	-
Polypropylene Filament (PP-MAPP YARN)	133.33	0.91	160-165 ℃
Polypropylene Pellets (PP 1043N)	-	0.90	160-165 ℃
Polypropylene-graft-maleic anhydride (MAPP)	-	0.89	160 ℃

2.2. Method

2.2.1. Thermoplastic pultrusion

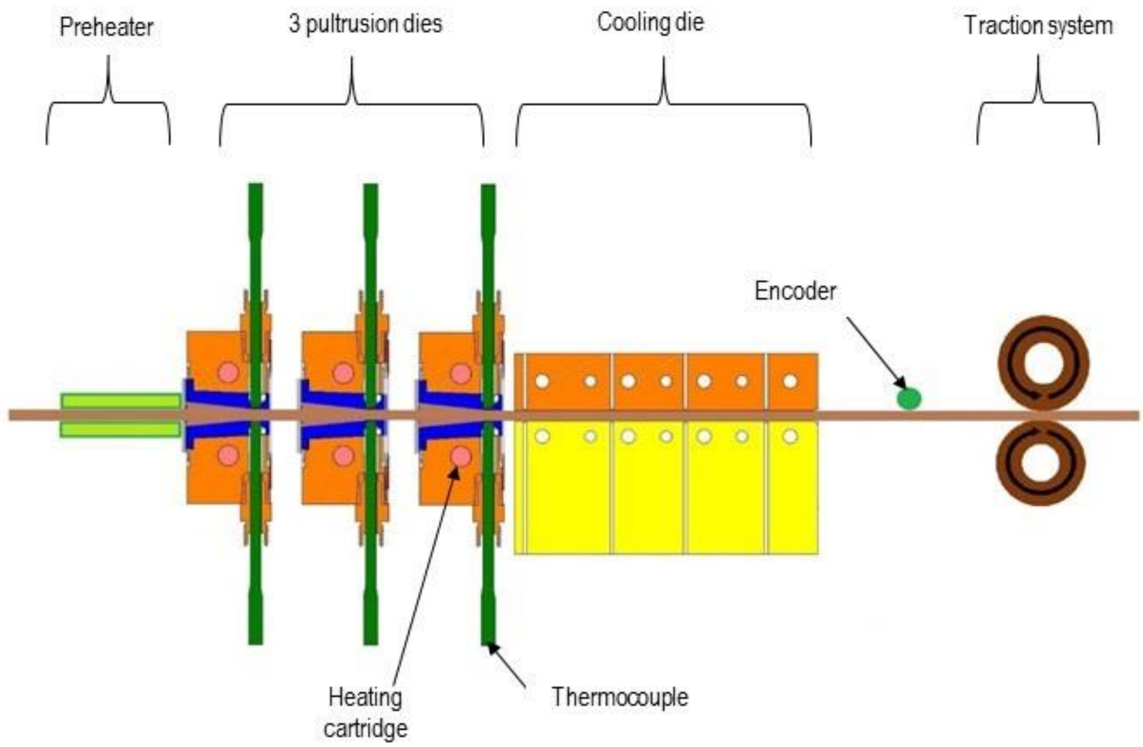

Figure 1 - Schematic view of a transversal cut of impregnation unit having three pultrusion dies and the cooling die.

Figure 1 shows the schematic for the thermoplastic pultrusion system used in this study. The pultrusion apparatus included a creel (not shown in Figure 1), pultrusion modules and a pulling system. The preheater used in this study was a 300-mm-long contactless preheater consisting of a heated pipe with an inner diameter of 12 mm. Yarn precursors were wound onto 34 bobbins containing 2 flax fibers and 3 PP-MAPP fibers in each bobbin. Note that all three pultrusion dies had a conical cavity with a 5° tapered angle followed by a 6.25 mm cylindrical cavity. The diameter of each die cylindrical exits were 5.13, 5.00 and 4.76 mm respectively. Before entering the pultrusion dies, the yarn precursors entered the preheater with fiber volume fraction $(V_f)\ of$ 42%. The excess resin in the yarn precursors generated an overfill of 14.8 %, 5.1% and 10.4% in the first, second and third die respectively. The overfill is calculated by the ratio of the area of excess material (resin and fiber) over the die exit area. The pultrusion dies were separated by a 25-mm air gap. The third pultrusion die was followed by a 50-mm-long cooling die where the composite mixture gave off the heat and consolidate to form a pultruded rod. Table 2 presents temperature set points and the production speed used for making flax/PP pultruded rods. The impregnation system temperature was set at 205 ℃. The cooling die temperature was set at 40 ℃. The production speed was controlled by a puller and measured by an encored. In addition, this speed was kept constant throughout the process at 50 mm/min. The nominal V_f of the 4.76 mm diameter-rods was 50%. This nominal Vf correspond to a flax weight fraction of 63%.

Table 2 - Temperature set points and the production speed used for making flax/PP pultruded rods

Specimen	Preheater (℃)	Pultrusion dies (℃)	Cooling die (℃)	Speed (mm/min)
Flax/PP	120	205	40	50

2.2.2. Injection molding

The rods were cut in 6 and 9 mm-long pellets using a pelletized (BT 25, SCHEER BAY Co., USA). The 15-mm-long pellets were cut using a circular saw (DW-250-N, DIAMANT RUBI, USA). Figure 2 shows the dry blend in the master mixture for injection molding. The flax/PP pellets were blended with PP and MAPP in the injection molding machine to lower the fiber weight fraction and improve interface properties between flax and PP. The final flax weight fraction of the injected dogbones can be calculated by multiplying the flax weight fraction in the pellets by the weight fraction of the pellets in the injected blend. Table 3 shows the composition of material in the master mix. According to that mix, the flax weight content in the injected coupons was 25%.

Figure 2 – The dry blend in the master mix for injection molding.

The master mix was then added directly to the injection molding hopper. Three sets of eight dogbone coupons were injection molded using these three pellet sizes. The size of the tensile dogbone specimen was 165 x 13 x 19 mm according to ASTM D638. The charpy impact test specimens were produced according to ASTM D6110. The injection molding machine used in this study was Sumitomo SE50S. Table 4 shows the process set points for compounding PP/flax in the injection molding machine. The maximum temperature in the injection process was kept at 210 ℃ to avoid any thermal degradation of flax fibers.

Table 3 – Composition of material in the master mix.

Specimen name	MAPP pellets (wt. %)	PP pellets (wt. %)	Flax/PP pellets (wt. %)
Master mixture	10	50	40

Table 4 : Overview of the process set points for compounding PP/flax (Injection molding)

Specimen name	T_1(℃)	T_2(℃)	T_3(℃)	T_4(℃)
Master mix	200	200	200	210

2.3. Characterization method

Cylindrical samples were produced using a precision saw. The cylindrical samples were polished and analyzed under a microscope (Metallovert, Leitz). Photos of 200 X magnification were taken and then stitched to form the whole cross-section of pultruded rod. For mechanical properties characterization after injection molding, the tensile test method was according to ASTM D638-44. The charpy impact test was done according to ASTM D6110-17. Void content was measured according to ASTM2734-09 Method C.

3.0. Results and discussions

3.1. Pultruded rod consolidation quality

The void content in the pultruded rods was calculated to be 3.5% ± 0.1%. Figure 3 shows the consolidation and microscopic qualities of the pultruded rod. Figure 3a) shows pultruded cylindrical rods of flax/PP produced using the multi-die pultrusion system. The rods look solid and straight. No fiber breakage was observed during pultrusion. Most probably, the flax fiber length in yarns was conserved in the pultruded yarns.

Figure 3b) presents a photo of a typical cross section taken at x 200 magnification showing good impregnation and a small amount of void can be observed. The void content appears low, confirming the measurement of 3.5% ± 0.1%. A small amount of void areas can be seen, showing as black spots. Figure 3c) shows a photo of a good impregnation region in the pultruded rod. The matrix appear to have well impregnated the fibers since most fibers are covered with resin. Based on these results, it can be stated that the overfilling of dies on to three of 14.8%, 5.1%, and 10.4% generated sufficient backflow and impregnation pressure. However, these overfill also create equal amount of resin loss during pultrusion. Further studies will investigate the effect of reducing the overfill on the impregnation quality.

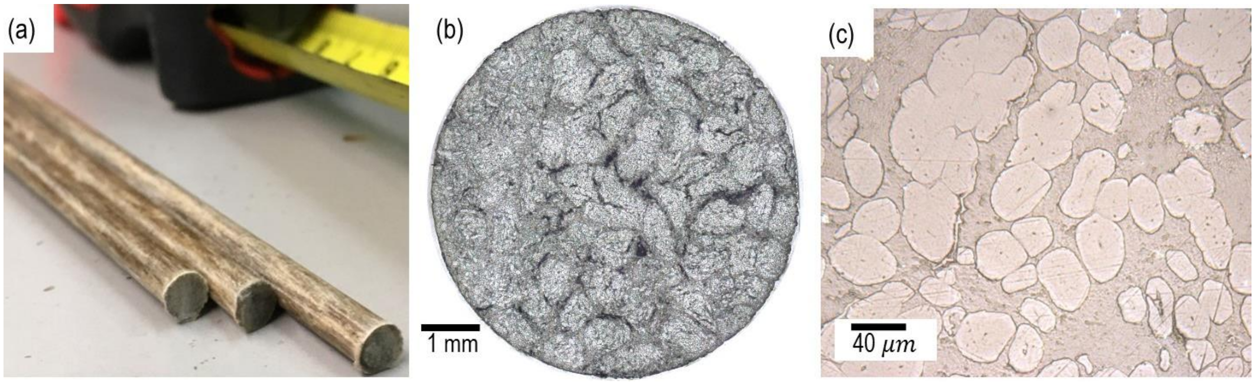

Figure 3: (a) Cylindrical rods of flax/PP produced using the multi-die pultrusion system. (b) Photo of a typical cross section taken at x 200 magnification showing good impregnation and a small amount of void. (c) Photo of a fully impregnated region.

3.2. Pelletizing quality

Figure 4 presents photos of pellets obtained with different lengths. The pelletizer was able to cut the rods onto two different pellet lengths (6 and 9 mm only). The pelletizer torque was inversely proportional to the selected pellet length. The pelletizer was incapable of cutting rods onto 15 mm long pellets. This pellet length was achieved using an automatic circular saw. In addition, the pelletizer's blade was dull. This caused a non-uniform cutting quality as one can notice in Figure 4a) & Figure 4b), the appearance of pellet size 6 & 9 mm looks exploded compared to pellet size 15 mm in Figure 4c).

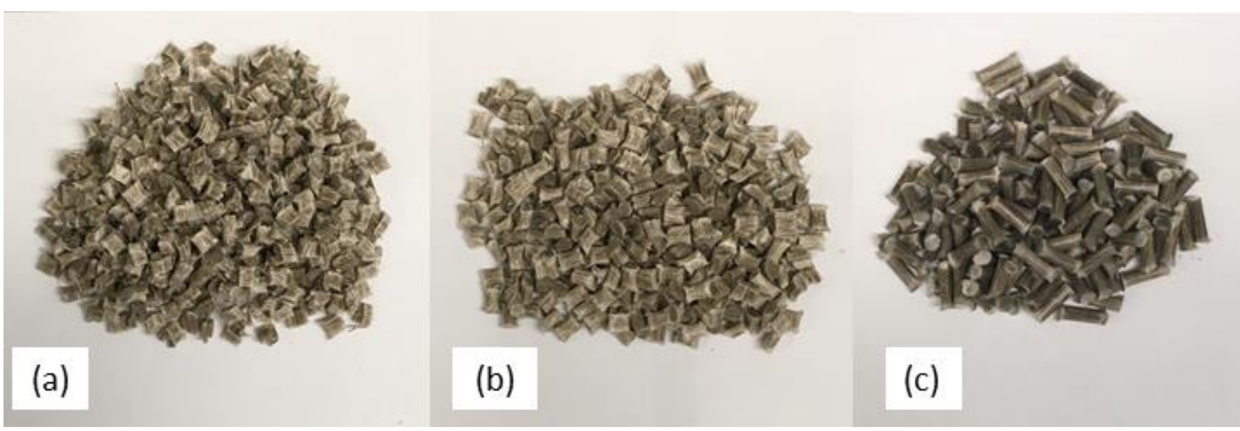

Figure 4: photos of pellets obtained with different lengths: (a) 6 mm, (b) 9 mm and (c) 15 mm.

3.3. Mechanical properties

3.3.1. Tensile strength

Table 5, Figure 5a) and Figure 5b) show tensile properties of injection molded products as a function pellet size. Note that the tensile modulus measured is the apparent tensile modulus since displacement was measured using cross-head displacement. Pellet size 9 and 15 mm are showing slightly superior tensile properties. More than 10% increase in tensile properties is observed when using pellet size 9 mm relative to pellet size 6 mm. However, the three values seem to be close to each other. The reason for this neutrality may be related to all pellets were higher than the estimated L_c of 1.2 mm by Barkoula *et al.* [5]. Using pellet size that are higher than L_c did not strongly affect the tensile strength.

Table 5 : Tensile properties of injection molded products as a function of pellet length.

Pellet size (mm)	Young`s modulus (MPa)	Max. Tensile (MPa)
6	1419 ± 26	39.3 ± 0.5
9	1708 ± 55	44.0 ± 0.7
15	1620 ± 37	42.5 ± 1.2

3.3.2. Impact strength

Table 6 and Figure 5c) present the impact strength of injection molded products as a function of pellet lengths. Using pellet size 15 mm in injection molding is showing more than 20% higher impact strength relative to pellet size 6 mm. This increase in impact strength confirms results found in literature on the effect of using longer pellets [5, 14]. Between pellet size 6 and pellet size 9 mm, there is a slight increase in impact strength but almost negligible in comparison to the strength increase observed when using pellet size 15 mm. The increase in impact strength when using pellet size 15 mm was not related to L_c since all pellets have lengths that are probably higher than L_c. Rather, it is possibly due to increased energy absorption via fiber fracture and debonding [5].

Table 6: Impact properties of injection molded products as a function of pellet length.

Pellet size (mm)	Impact strength (J/m^2)
6	3952 ± 287
9	4091 ± 132
15	4825 ± 189

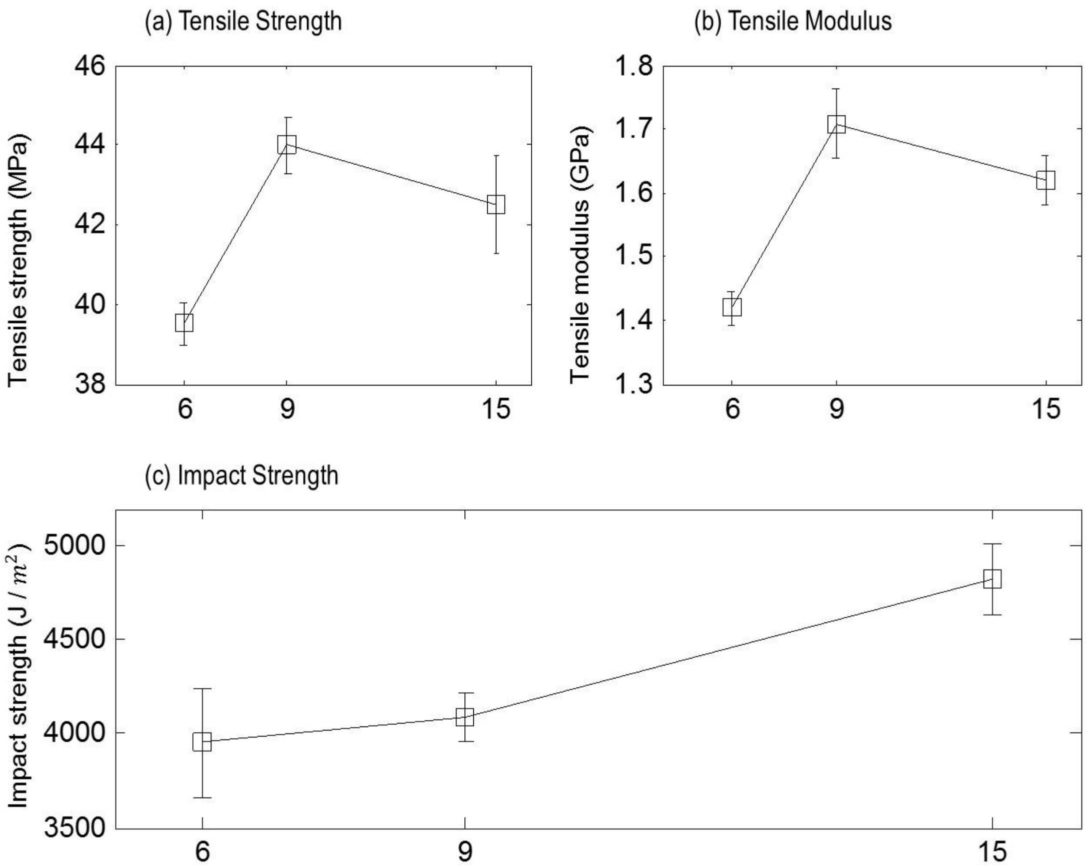

Figure 5: Mechanical properties of injection molded products as a function pellet length. (a) Tensile Strength. (b) Tensile Modulus. (c) Impact Strength.

4.0. Summary and Next Steps

A method to make flax/PP injection molding precursors using the multi-die pultrusion system was demonstrated. The effect of of polypropylene and flax (flax/PP) pellets length variation on the quality of injection molded product was characterized using mechanical tests. A noticeable increase in impact strength was observed when longer flax/PP pellets were used for injection molding. Further work will need to be conducted to confirm the influence of the pellet length on mechanical properties.

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