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VACUUM ASSISTED MULTI-DIE PULTRUSION OF GLASS/POLYAMIDE 6 AND GLASS/POLYPROPYLENE

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ABSTRACT

Pultrusion is a manufacturing method known for its efficiency in producing continuous composite structures with great mechanical properties. In thermoplastic pultrusion, multi-die systems and vacuum chambers between dies have proven effective in enhancing product quality. However, the effect that process parameters, including vacuum, have on their mechanical properties has not been researched extensively. Studies are particularly lacking for glass fiber/polyamide 6 composites, an important material used by the automotive industry that has a high moisture absorption rate that causes manufacturing challenges. This work studies the influence of pulling speed and vacuum pressure on pultruded glass fiber/polyamide 6 (GF/PA6) and glass fiber/polypropylene (GF/PP) flat bars by performing mechanical testing and micrography. Results show short beam strength was significantly improved and fewer voids were observed with the application of vacuum, for speeds up to 500 mm/min.

1 INTRODUCTION

Pultrusion is a highly efficient manufacturing technique for producing constant cross-section composite structures as it allows for high production rates, low costs and minimal manual intervention once the process is started [1]. Historically, pultrusion focused on thermosetting polymers. However, the use of thermoplastics has been gaining popularity due to their well-known advantages over thermosets. One key advantage of thermoplastics is their ability to be melted and reshaped, which improves recyclability but also allows the possibility of combining it with other manufacturing processes to create complex lightweight structures. This has captured the attention of industries like automotive, where thermoplastic pultruded profiles are now being used in complex parts such as body structures, bumpers, wheel rims and seat structures [1,2]. Polyamide 6 has seen increased use in the automotive industry for several lightweight components under the engine hood, due to its good mechanical properties and low density [3]. However, it is known to be a hygroscopic polymer, capable of absorbing moisture from the surroundings, up to 3.4% of its weight [4]. The presence of water in PA6 matrices can lead to degradation by hydrolysis and accelerate thermal oxidation at the high temperatures during processing, manifesting through a visible yellowing of the material [5]. Naturally, this poses challenges during processing and results in reduced mechanical performance. Therefore, a common industry practice to mitigate these effects is drying PA6 pellets prior to processing in extrusion and injection molding applications [4]. Once dried, the pellets are placed in injection machine hoppers having controlled environment that prevent moisture re-absorption. For a pultrusion system, this method would be more challenging to apply, as the polymer/reinforcement hybrid yarns are mounted in large creels. Thus, exploring in-line alternatives that can more effectively integrate with the pultrusion process is preferred.

In the case of thermoplastic pultrusion, multi-die systems have been proven to provide better results than single-die experiments [6,7]. It has also been suggested that the use of vacuum chambers between dies could have a positive impact on the quality of the pultrudates by lowering the void content [7,8]. Moreover, it is well documented that increasing the pulling speed yields a lower quality material as it reduces the time under pressure and translates into higher void contents [9,10]. However, the interaction between the simultaneous effects of speed and vacuum application has not yet been the subject of extensive research and to the best of the authors' knowledge, no study was published regarding GF/PA6 composites. This paper aims to provide insight into the influence of pulling speed and vacuum pressure on the impregnation and mechanical properties of pultruded glass fiber-reinforced polyamide 6 and glass fiber-reinforced polypropylene flat bars.

2 EXPERIMENTAL

Flat profiles of 3 mm × 15 mm were pultruded using GF/PA6 and GF/PP co-wound yarns (Concordia Fibers). The co-wound yarns are made by co-winding glass and polymer tows together. The term tow is used here to designate single material multifilament bundles. The reinforcement tows were 66 tex E-Glass fiber. The PA6 tows were 100 tex and the PP were 78 tex. Table 1 presents the co-wound yarns properties.

Table 1. Properties of the co-wound yarns.

	GF/PA6	GF/PP
[No. glass tows, No. polymer tows]	[21, 5]	[11, 4]
Vf (%)	55.4	45.5
T_g (°C)	52	-11
T_m (°C)	220	160

The schematic of the pultrusion apparatus used in this work is shown in Figure 1. It consists of a creel that holds the bobbins and applies tension to the yarns, which are then guided into the pre-heater box. Within the pre-heater, the polymer undergoes heating to a temperature slightly lower than its melting temperature. This is achieved by means of three heat guns and two radiative heating elements ensuring efficient heating through both convection and radiation. Afterwards, the yarns are passed through eight 31.75 mm long dies controlled by two 500 W cartridge heaters. The dies are separated by quartz vacuum chambers of 35 mm in length. The airtightness of the chambers is guaranteed with ConFlat® vacuum flanges and copper gaskets, along with the filling of the dies' cavities by the melted resin. These cavities have a rectangular cross section with rounded corners of 1 mm radius. The cavities are tapered with angles of 1.5° and 7° thicknesswise and widthwise, respectively, followed by a straight section of 6.35 mm. The exit cavities feature a reduction of 2.5 % in both thickness and width through the first four dies and 1.5 % in the remaining four dies, until reaching a cross-section of 3 mm x 15 mm in the last die. Forty-six co-wound yarns of GF/PA6 and seventy-two yarns of GF/PP were used in the pultrusion, with the aim of having the ratio of the total material area to the die exit area (filling ratio) of 103 % in the last die. Finally, the thin-walled tube protruding from the last die allows the material to be cooled and shaped to its final geometry and dimensions [11]. The cooling was done through a combination of multiple fans and heat sinks that allowed for an exit temperature between T_g and T_m across different speeds and prevented deconsolidation. A traction system is used to pull the pultruded material along the line and controlling the speed.

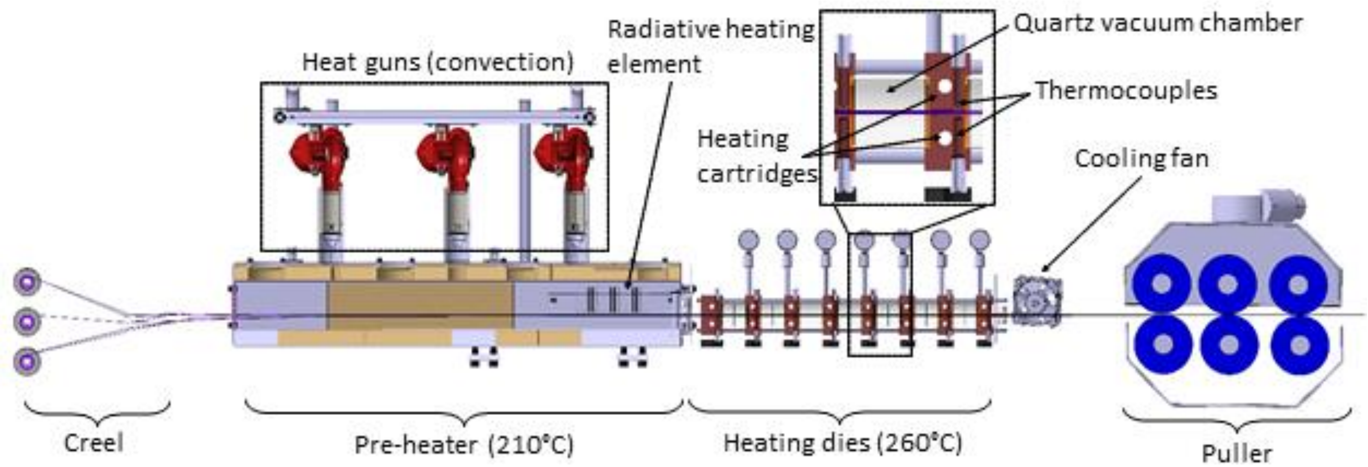

Figure 1. Schematic view of the multi-die pultrusion line.

Pultrusion experiments were conducted at different pulling speeds (50, 100, 200 and 500 mm/min). Firstly, experiments were performed at atmospheric pressure without the vacuum chambers installed; instead with a minimal gap between dies (5 mm) was maintained as a representation of a traditional multi-die experiment. Then, the vacuum chambers were set up to produce a 35 mm gap between dies and experiments were done both at atmospheric pressure and under partial vacuum, with an average absolute pressure of 7 kPa in the latter case.

Short-beam shear (SBS) tests were conducted according to ASTM D2344 on an MTS Insight 50 kN machine. The SBS is linked to interlaminar shear stresses and is a representative measure of the impregnation quality of the pultrudates. Additionally, samples were embedded in epoxy, ground and polished. Micrography was performed using an optical microscope (VHX7000, Keyence) to inspect the internal morphology and the presence of voids.

3 RESULTS AND DISCUSSION

Figure 2 shows the GF/PA6 pultrudate thickness between dies 6 and 7 at atmospheric pressure (a) compared to when vacuum is applied (b). With vacuum, the pultrudate was observed to be 4.95 mm thick compared to a thinner 4.32 mm when the pultrudate was under atmospheric pressure. Both thickness measures are higher than the Die 6's thickness of 3.08 mm. The increase in thickness is attributed to the deconsolidation of the pultrudate. Deconsolidation is driven by the elastic recovery of the compressed fiber bed and the expansion of compressed gases within the pultrudate. Therefore, the vacuum environment is considered to promote deconsolidation due to the further expansion of the compressed gases in the pultrudate. During the experiment, it was also observed that vacuum application led to degassing of the resin by bubbling being observed in the resin backflow. This can be seen at Figure 2 where the size of the resin backflow in the right side die entrance is larger in the vacuum case (b). Extraction of the gases from the pultrudate promotes an easier impregnation since the polymer flow is not hindered by a compressed gas bubble.

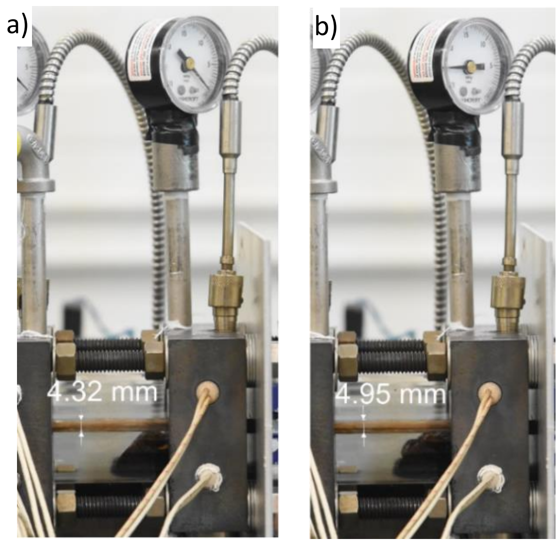

Figure 2. View of the pultrudate in between dies through the quartz vacuum chamber with a) P_{atm} and b) partial vacuum.

Figure 3 shows the cross-sections of GF/PA6 pultruded bars with and without application of vacuum at 200X magnification. Fewer porosities can be seen in the vacuum experiments at both speeds, as opposed to its atmospheric pressure counterparts. Regarding the effect of pulling speed, the sample pultruded at 100 mm/min exhibits a more uniform distribution of fibers and resin, indicating better impregnation, whereas at 500 mm/min it is still possible to identify the outline of individual yarns and resin rich areas in between. Finally, it is worth mentioning that the surface finish of the samples is irregular, for which further investigation is needed.

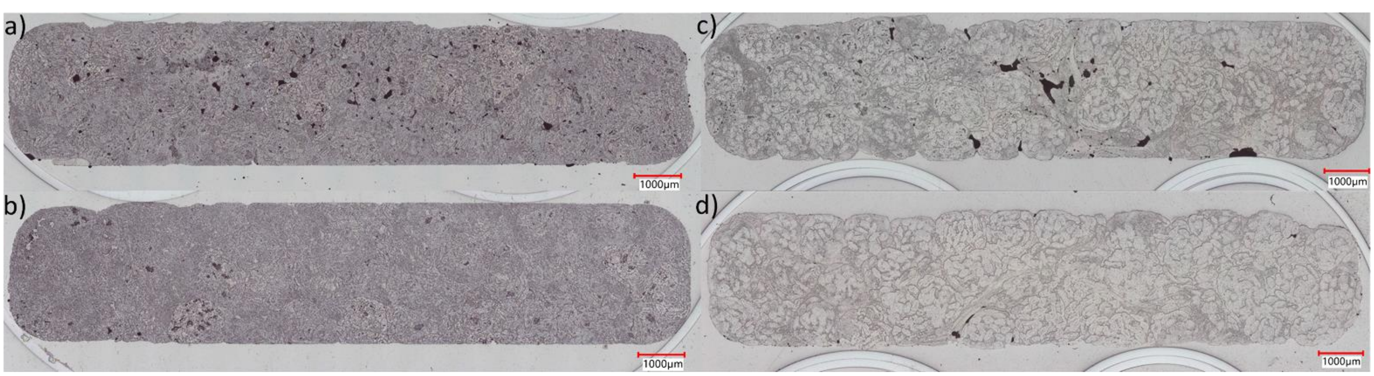

Figure 3. Micrographs of pultruded specimens at 100 mm/min a) P_{atm} b) vacuum, and at 500 mm/min c) P_{atm} d) vacuum. The fibers are shown in white, the matrix in gray, and the dark areas are voids.

Figure 4 presents the short-beam strength (or apparent shear strength) of specimens pultruded at different speeds and ambient pressure conditions. Figure 4a) and Figure 4b) show the results for GF/PA6 and G/PP pultrudates. Comparing the atmospheric pressure cases at 5 mm and 35 mm gap (vacuum chamber installed), it can be seen in Figure 4a) that the greater distance between dies yields lower shear strength. It is likely that the increased length resulted in decreased material's temperature as it exited the heated dies, reducing polymer flow and consequently hindering impregnation. A solution could be using shorter vacuum tubes or heating tape wraps to ensure uniform heating along the pultrusion line. Furthermore, as expected, the increase in pulling speed leads to a decrease in mechanical properties across all pressure conditions. This is caused by a shorter duration for polymer flow and glass fiber impregnation. However, the application of vacuum compensated in part the effect of the higher speed, making the average apparent shear strength at a higher speed under vacuum closer to that of an experiment with lower speed without vacuum. For example, at 500 mm/min with vacuum the apparent shear strength was 39.1 ± 3.1 MPa, comparable to the experiment at 200 mm/min without vacuum that yielded a strength of 38.7 ± 2.6 MPa.

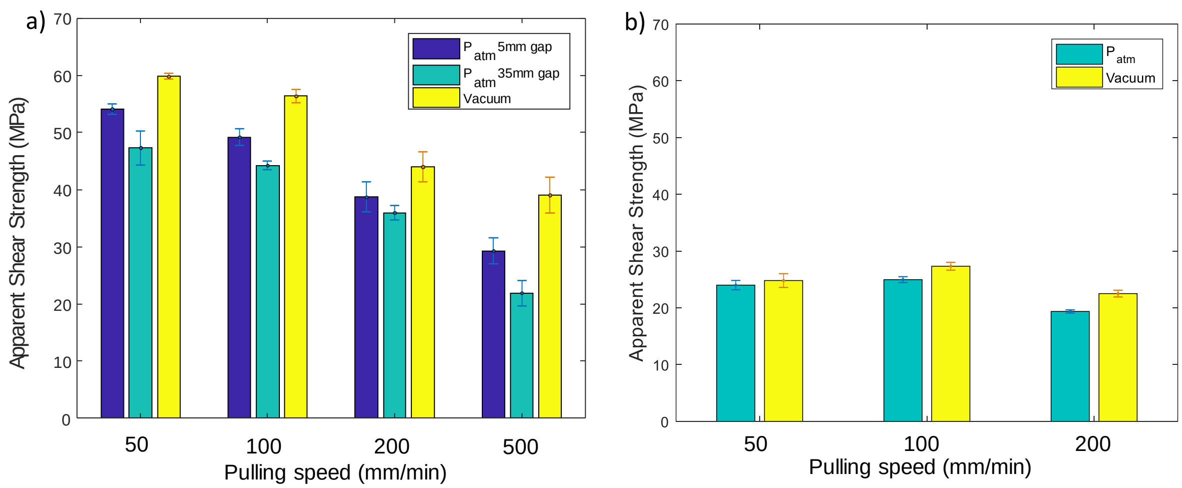

Figure 4. Short-beam strength of a) GF/PA6 specimens and b) GF/PP pultruded at different speeds and pressure conditions.

The average values of short-beam strength and respective relative increase for the P_{atm} 35 mm gap and vacuum cases are shown in Table 2. Regarding experiments performed with vacuum and GF/PA6, it is noticed that the apparent shear strength is improved for every pulling speed. The increase in SBS is situated between 20 % to 30 % at 50, 100 and 200 mm/min, and it reaches a maximum of 78.5 % at 500 mm/min. However, if the values for P_{atm} with 5 mm gap are considered, the increase in SBS is instead in the range of 10 % to 15 % at 50, 100 and 200 mm/min and 33 % at 500 mm/min. This indicates that the added complexity of installing a vacuum system when compared to a typical multi-die setup at P_{atm} seems to be justified by a non-negligible improvement of the properties.

Table 2. Short-beam strength of pultruded bars with and without vacuum for different speeds.

	GF/PA6			**GF/PP**		
Speed (mm/min)	P_{atm} (MPa)	Vacuum (MPa)	% Increase	P_{atm} (MPa)	Vacuum (MPa)	% Increase
50	47.4	59.8	26.2%	24.0	24.8	3.3%
100	44.3	56.4	27.3%	25.0	27.3	9.2%
200	36.0	44.0	22.2%	19.4	22.5	16.0%
500	21.9	39.1	78.5%		-	

In addition to the volatile organic compounds (VOC) that might be released at high temperatures, the absorbed moisture in the resin is deemed to be a concern, due to the evaporation of water that leads to the formation of bubbles within the material that might not be able to escape. Since PA6 has a higher moisture absorption rate compared to PP, the relation between water content and vacuum application can be studied by comparing both cases. The results shown in Figure 4b) for the SBS of GF/PP specimens indicate a slight improvement with vacuum usage. At 50 mm/min the difference is not statistically significant, while at 200 mm/min it is more accentuated. However, the maximum relative increase in apparent shear strength is not as high as with PA6. For instance, it increased by 9% for PP and 22% for PA6 at 100 mm/min, hence it can be inferred that one of the benefits of vacuum application during pultrusion is assumed to be closely linked to moisture removal.

4 CONCLUSIONS

In this work, GF/PA6 and GF/PP bars were pultruded in a multi-die setup with vacuum chambers. A comparison study of the effect of air pressure (vacuum or atmospheric) between dies was performed through short-beam strength tests and micrography. Without vacuum, voids can be seen in cross-sections of samples, while the samples subjected to vacuum present less porosity. Higher pulling speeds led to lower impregnation due to the reduction in the time for the polymer to flow, while the usage of vacuum improve quality. Finally, it was also observed that the benefit of vacuum was greater for the hygroscopic PA6 polymer.

5 REFERENCES

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