DEVELOPMENT OF A NEW PULTRUSION EQUIPMENT TO MANUFACTURE THERMOPLASTIC MATRIX COMPOSITE PROFILES

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Abstract
This paper describes the design and manufacture of a low-cost full scale pultrusion prototype equipment and discusses the production and obtained mechanical properties of polypropylene/glass (GF/PP) reinforced composite bars fabricated by using the prototype equipment. Three different GF/PP pre-impregnated raw-materials, a commercial GF/PP comingled system from Vetrotex, a GF/PP powder coated towpreg [1-3] and, a GF/PP pre-consolidated tape (PCT) produced in our laboratories, were used in the production of composite bars that were subsequently submitted to mechanical testing in order to determine the relevant mechanical properties and quantify the consolidation quality. Samples of the different composite profiles were also observed under SEM microscopy.

1 Introduction
Pultrusion is a versatile continuous high speed production technology allowing the production of fibre reinforced complex profiles. Thermosetting resins are normally used as matrices in the production of structural constant cross section profiles. In contrast, the use of thermoplastic matrices, that can allow higher production speeds and present higher toughness, chemical resistance, damping and are more ecological, are still in an early stage [4-11]. Nevertheless, it was only recently that new technologies allowed the production of low-cost continuous fibre reinforced pre-impregnated thermoplastic raw-materials. With these technologies it is possible to replace the former expensive thermoplastic matrix prepregs (obtained by melting and solvent based processes) by cheap dry commingled fibres [12-15] and powder coated pre-impregnated materials (towpregs) [9, 16, 17]. Work is currently in progress to consolidate and process these new promising materials into final composite parts by using existing high throughput technologies, such as heated compression moulding, filament winding and pultrusion [8, 18-25].
In this work, three different GF/PP pre-impregnated raw-materials, a commercial GF/PP comingled system from Vetrotex, a GF/PP powder coated towpreg and, a GF/PP pre-consolidated tape (PCT) produced in our laboratories, were used in the production of composite bars processed in a laboratorial pultrusion equipment developed for such purpose [26].

The obtained composite profiles were subsequently submitted to mechanical testing in order to determine the relevant mechanical properties and quantify the consolidation quality. Samples of the different composite profiles were also observed under SEM microscopy. Finally, the obtained results were compared with traditional thermosetting resins reinforced with glass fibres.

2 Processing

2.1 Raw-Materials

Table 1 summarises relevant properties of the glass fibres and polypropylene used in present work to produce pre-impregnated raw materials. In the GF/PP towpregs were used 2400 Tex type E fibre rovings from Owens Corning and a Icorene® 9184B P polypropylene from ICO Polymers France. On the other hand, the PCT tapes were manufactured with glass fibres (TufRov 4599) from PPG Industries and a polypropylene (Moplen RP348U) from Basell.

<table>
<thead>
<tr>
<th>Property</th>
<th>Units</th>
<th>Glass fibres</th>
<th>Polypropylene</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density</td>
<td>Mg/m³</td>
<td>2.56</td>
<td>0.91</td>
</tr>
<tr>
<td>Tensile strength</td>
<td>MPa</td>
<td>3500</td>
<td>30</td>
</tr>
<tr>
<td>Tensile modulus</td>
<td>GPa</td>
<td>76</td>
<td>1.3</td>
</tr>
<tr>
<td>Average powder particle size</td>
<td>µm</td>
<td>-</td>
<td>440</td>
</tr>
<tr>
<td>Linear roving weight</td>
<td>Tex</td>
<td>2400</td>
<td>-</td>
</tr>
</tbody>
</table>

Table 1. Properties of the raw materials used to produce the GF/PP towpregs.

Commercial commingled GF/PP fibres Twintex® R PP 60 B 1870 FU from Owens Corning were also used, as reference of a current commercially available pre-impregnated product, in the production of the pultrusion thermoplastic composite profiles.

2.2 Processing the pre-impregnated raw-materials

The prototype dry powder coating equipment used to produce fibre reinforced towpregs is schematically depicted in Figure 1 [8, 27]. It consists of six main parts: a wind-off system, a fibres spreader unit, a heating section, a coating section, a consolidation unit and a wind-up section. Initially, the reinforcing fibres are wound-off and pulled through a pneumatic spreader and then coated with polymer by heating in a convection oven and made to pass into a polymer powder vibrating bath. A gravity system allows maintaining the amount of polymer powder constant. The consolidation unit oven allows softening the polymer powder, promoting its adhesion to the fibre surface. Finally, the thermoplastic matrix towpreg is cooled down and wound-up on a spool.
On other hand, thermoplastic pre-consolidated tapes were manufactured by using a new specially developed cross-head extrusion hot melting process (Fig. 2).

![Figure 2. Cross-head extrusion.](image)

The core of this technology is a impregnation unit in which reinforcing fibres are introduced, spread and impregnated by the polymer melt. The impregnation is achieved through the build-up of pressure acting on the molten polymer trapped between the unit’s spreading elements and the fibre rovings. The apparatus consists of a creel system holding the fibre rovings, a guidance system to transport the fibre into the impregnation unit, an extruder to melt and feed the molten polymer into the impregnation unit, the impregnation unit itself, and subsequently a cooling unit, a puller, and a take-up device where the composite tape is collected. Such thermoplastic pre-consolidated tape is commercially available through the company Comp Tape Lda. [28]. An overview of such material data is given in Table 2.

<table>
<thead>
<tr>
<th>Property</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fibre type</td>
<td>E-Glass, 2400 Tex</td>
</tr>
<tr>
<td>Fibre diameter</td>
<td>17 µm</td>
</tr>
<tr>
<td>Fibre content</td>
<td>60 wt.%</td>
</tr>
<tr>
<td>Matrix type</td>
<td>Polypropylene (PP)</td>
</tr>
<tr>
<td>Tape width</td>
<td>25 mm</td>
</tr>
<tr>
<td>Tape linear density</td>
<td>16000 Tex</td>
</tr>
</tbody>
</table>

Table 2. PCT data.

2.3 Pultrusion
Figures 3 and 4 depict a schematic and a photograph of the pultrusion equipment developed in the present work. The 10 kN pultrusion line may be divided in five main parts: i) initial towpreg bobbins storing cabinet; ii) guiding system; iii) pultrusion head, that includes a pre-heating furnace and the pressurization/consolidation and cooling dies; iv) pulling system and, v) the final profile cutting system.

To produce the composite profiles, the pre-impregnated materials are guided into the pre-heating furnace to be heated up to the required temperature. Then, they enter in the pultrusion
die to be heated up and consolidated to the required size in its first zone and, after cooled down in order to solidify. The pultruded material is then cut into specified lengths. In this work, a die was designed and manufactured to allow producing a 20×2 mm tape-shaped profile.

![Figure 3. Schematic diagram of the proprietary pultrusion line.](image)

The pre-heating furnace may reach a temperature of 1000 °C and was designed to allow processing almost every type of fibre/thermoplastic-based pre-impregnated materials. Table 3 summarises the pultrusion conditions used to process the three different pre-impregnated raw-materials into final GF/PP composite profiles.

<table>
<thead>
<tr>
<th>Raw-material</th>
<th>Heated die temperature (°C)</th>
<th>Cooled die temperature (°C)</th>
<th>Pre-heating temperature (°C)</th>
<th>Pulling speed (m/min)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Twintex®</td>
<td>300</td>
<td>50</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Towpreg</td>
<td>275</td>
<td>25</td>
<td>170</td>
<td>0.2</td>
</tr>
<tr>
<td>PCT</td>
<td>230</td>
<td>50</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

![Figure 4. Overview of pultrusion equipment.](image)

As may be seen, it was already possible to produce from all GF/PP pre-impregnated raw-materials profiles at pull speeds of 0.2 m/min.

3 Tests and results

3.1 Microscopy analysis

To determine the impregnation quality of the pultruded thermoplastic composites, 2 cm-long samples were cut from the pultruded sections and were embedded in a thermoset EPIKOTE™ 04908 resin. After curing and polishing to a mirror like finish, using increasingly fine sanding paper, samples were ready for microscopy analysis. Observations were done using reflected light optical microscopy (Olympus BH-2). A digital camera (Leica DFC200) was used to image cross sectional views of the samples.
The microscopy images taken from the samples of the pultruded composites using PCT, Twintex® and towpreg, are given in Table 4.

<table>
<thead>
<tr>
<th>Material</th>
<th>250 ×</th>
<th>130 ×</th>
</tr>
</thead>
<tbody>
<tr>
<td>PCT</td>
<td><img src="image1.png" alt="Image" /></td>
<td><img src="image2.png" alt="Image" /></td>
</tr>
<tr>
<td>Towpreg</td>
<td><img src="image3.png" alt="Image" /></td>
<td><img src="image4.png" alt="Image" /></td>
</tr>
<tr>
<td>Twintex®</td>
<td><img src="image5.png" alt="Image" /></td>
<td><img src="image6.png" alt="Image" /></td>
</tr>
</tbody>
</table>

Table 4. Microscope images of the pultruded profiles.

As can be seen from the photos in table 4, all semi-finished lead to a reasonable distribution of the reinforcing fibres over the cross-section, although a small improvement in distribution can be observed going from PCT through Twintex® to towpreg composites. However, large differences in impregnation quality occur, between the different samples, that are likely directly related to the impregnation state of the semi-finished used during pultrusion. It may be seen that the impregnation quality of the PCT composite samples is excellent, presenting almost all fibres completely surrounded (‘wet-out’) by the polymer, hardly showing any dry spots in the pultruded samples. This is most likely due to the high degree of impregnation already achieved in the PCT raw-material tape prior to the pultrusion step. The Twintex-based samples also show a very good impregnation of the fibre. This is likely due to the intimate contact between the individual dry glass and PP fibres prior to the final pultrusion stage, making the effective remaining impregnation distance when melting the polymer very small, which leads to an easier consolidation. Although, some larger dry spots were observed between the glass fibres at larger magnifications, showing an overall impregnation quality poor when compared with the one observed in the PCT tape based pultruded composites. Finally, the depicted towpreg-based composite samples exhibit larger apparent dry zones. This is most likely due to the uneven distribution of the dry polymer powder in the towpreg, prior to pultrusion. Consequently, seems to be harder to bridge the large distances of dry glass
fibre during pultrusion, which results in bigger unimpregnated zones in the pultruded composites.

3.2 Mechanical testing
3.2.1 Short beam shear tests
In order to obtain an indication of the composite quality after pultrusion, short beam tests have been carried out according to ASTM 2344, at room temperature, using an Universal Instron 4505 testing machine fitted with a 50 kN load cell. Five 20 x 10 x 2 mm composite samples were cut from pultruded composites processed from each pre-impregnated raw-material to be tested. Figure 5 shows typical force-displacement curves of the tested specimens in the short beam shear test.

![Figure 5. Short beam test results.](image)

Those curves show that all specimens failed in plastic shear, and therefore, no breaking load could be obtained, which would allow the formal calculation of the interlaminar shear strength. Such results seem to show that a reasonable degree of adhesion between layers was obtained in the composites. Curves also show an obvious different behaviour between samples. While PCT tape and Twintex® based pultruded composites have similar load-bearing capacities, having similar shape and ultimate load curves, towpreg based samples showed lower performance. This mostly results from the already mentioned limited degree of impregnation of these samples.

3.2.2 Tensile and flexural tests
Table 5 presents the mechanical properties determined from flexural and tensile tests carried out on pultruded profiles. The fibre mass fraction and the flexural and tensile properties were determined in accordance with the ISO 1172, EN ISO 14125 and EN ISO 527-5 standards, respectively. For the fibre, specimens weighing approximately 20 g, cut from the profiles, were calcinated during 10 min at 600°C inside a Nabetertherm® S30 muffle furnace. For the second determination, rectangular, 100 mm x 20 mm x 2 mm samples were submitted to three-point bending tests using a 80 mm support span, a cross-head speed of 1 mm/min and a 100 kN load cell in a Shimadzu® universal testing machine. The tensile tests were conducted on 250 mm x 15 mm x 2 mm rectangular samples, at a 2 mm/min cross-head speed, using the same equipment and load cell. A 50 mm length strain-gauge was used up to 0.3% strain, to allow determining accurately the tensile modulus on each sample.
### Table 5. Results from tensile and flexural tests.

<table>
<thead>
<tr>
<th>Raw-material</th>
<th>Tensile strength (MPa)</th>
<th>Tensile modulus (GPa)</th>
<th>Flexural strength (MPa)</th>
<th>Flexural modulus (GPa)</th>
<th>Fibre mass fraction (%)</th>
<th>Fibre volume fraction (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Twintex®</td>
<td>&gt;416 24.9 24.9 1.1 24.9</td>
<td>595 24 26.2 2.0 26.2</td>
<td>62.5 37.1 62.5 37.1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Towpreg</td>
<td>&gt;305 29.9 29.9 3.5 29.9</td>
<td>125 11 27.1 1.2 27.1</td>
<td>78.8 56.8 78.8 56.8</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>PCT</td>
<td>&gt;424 21.4 21.4 1.5 21.4</td>
<td>329 30 16.8 1.5 16.8</td>
<td>54.8 30.0 54.8 30.0</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Av. – Average  
SD – Standard Deviation

As can be seen from Table 5, similar results that are compatible with the major common and structural applications were obtained in all pultruded composites materials from both tensile and flexural made tests. In any case, worse flexural strength and modulus results were found in towpreg and PCT tape based pultruded composites, respectively. These lower results obtained in the flexural tests are probably consequence of the inferior degree of impregnation observed in the towpreg based composites and, in the case of PCT tape based composites, result from the higher rich polymer regions exhibited by this material in its outside layers.

### 4 Conclusions

The tests made using a proprietary pultrusion equipment already allow producing in good conditions profiles from almost all commercial available thermoplastic matrix pre-impregnated raw-materials at pull speeds of 0.2 m/min. Currently, work is carried out to try increasing the pultrusion processing speed to values in the range from 2 to 6 m/min, which will equalizing the speed of the pultrusion line with that of the towpreg coating and PCT tape production lines. The use of similar operational speeds in both processes (equipments) will make possible, in future, assembling them in just one equipment.

The mechanical properties obtained in GF/PP pultruded composite profiles were also found to be adequate either for common or structural engineering applications. The experimental results also demonstrated that further work should be carried out to improve impregnation in towpreg based pultruded composites.

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### References


