PRODUCTION OF THERMOPLASTIC MATRIX TOWPREGS FOR HIGHLY DEMANDING AND COST-EFFECTIVE COMMERCIAL APPLICATIONS

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**Abstract.** This work reviews the work made in last years to produce thermoplastic matrix towpregs to highly demanding and more cost-effective commercial applications using a powder coating technology developed in Portugal by Minho and Porto Universities. Different thermoplastic matrices and continuous fibre reinforcements were used in the towpregs produced for highly demanding markets (e.g., carbon fibre reinforced Primospire® towpreg) and for more commercial applications (e.g., glass fibre reinforced polypropylene and polyvinyl chloride towpregs).

The relevant processing parameters, such as, fibre pull-speed and furnace temperature were varied to determine their influence on the polymer mass fraction obtained in the studied raw materials. Several technologies were also developed and used (compression moulding, pultrusion and filament winding) to process composite parts with adequate properties for the envisaged markets at compatible production rates.

The obtained results lead us to conclude that the studied thermoplastic matrix towpregs and their processed composite parts have very interesting conditions for being applied both in highly advanced and cost-effective markets.

**Introduction**

In recent years, due to their excellent properties continuous fibre thermoplastic matrix composites were successfully employed in the aircraft, military and aerospace industries [1,2]. In these and many other commercial engineering applications, they can replace with advantages thermosetting matrix composites. However, the high cost of the impregnation of continuous fibre thermoplastic composites, arising from the melting of the polymer or the use of solvents, still restricts their use in commercial applications. Hence, cost reduction largely depends on developing more efficient methods for impregnating fibres with high-viscosity thermoplastics and for processing final composite parts.

This paper summarises work carried out to develop new technologies to fabricate long and continuous fibre reinforced composite structures from thermoplastic matrix semi-products for commercial and highly demanding markets.

Continuous fibre reinforced thermoplastic matrix towpregs were produced using a recent developed coating line [3].

Using this prototype equipment, it was possible to produce glass fibre polypropylene (PP) and polyvinylchloride (PVC) towpregs for commercial markets and carbon fibre Primospire®, an amorphous highly aromatic material developed by Solvay Advanced Polymers, for application in advanced markets [2,4,5].

These thermoplastic pre-pregs were processed into composite structures by conventional equipments for thermosetting that were adapted to thermoplastic matrix composites fabrication, such as, filament winding, pultrusion and hot compression moulding.

As applications, filament wound pressure vessels prototypes for gas and incompressible fluids were produced from towpregs and submitted to internal pressure burst tests [6,7].
Experimental

Powder coating equipment. The prototype powder coating equipment used to produce glass and carbon fibre reinforced towpregs is schematically depicted in Figure 1. 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. In order to produce the towpregs, the reinforcing fibres are wound-off and pulled through a pneumatic spreader. After, they are heated in a convection oven and made to pass into a polymer powder vibrating bath to be coated. A gravity system allows maintaining constant the amount of polymer powder. The oven of the consolidation unit allows softening the polymer powder, promoting its adhesion to the fibre surface. Finally, the thermoplastic matrix towpreg is cooled down and wound-up on the final spool.

Fig. 1. Schematic diagram of the powder-coating line set-up

Raw materials. 2400 Tex type E glass fibre rovings, from Owens Corning, polypropylene, from ICO Polymers France (Icorene 9184B P), and polyvinyl chloride, supplied by CIRES (PVC - PREVINIL AG 736), powders were used to produce GF/PP and GF/PVC towpregs to be applied in common composite engineering parts. Table 1 summarises relevant properties of these materials.

<table>
<thead>
<tr>
<th>Property</th>
<th>Units</th>
<th>Glass fibres</th>
<th>Polypropylene</th>
<th>PVC</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density</td>
<td>Mg/m³</td>
<td>2.56</td>
<td>0.91</td>
<td>1.4</td>
</tr>
<tr>
<td>Tensile strength</td>
<td>MPa</td>
<td>3500</td>
<td>30</td>
<td>55</td>
</tr>
<tr>
<td>Tensile modulus</td>
<td>GPa</td>
<td>76</td>
<td>1.3</td>
<td>3.0</td>
</tr>
<tr>
<td>Average powder particle size</td>
<td>µm</td>
<td>-</td>
<td>440</td>
<td>150</td>
</tr>
<tr>
<td>Linear roving weight</td>
<td>Tex</td>
<td>2400</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

A new polymer developed by Solvay Advanced Polymers (Primospire® PR 120) and carbon fibre tows from TORAYCA (760 Tex M30SC) were used to produce towpregs for highly demanding markets. Table 2 presents the most relevant properties determined on these raw materials.

<table>
<thead>
<tr>
<th>Property</th>
<th>Units</th>
<th>Carbon fibres</th>
<th>Primospire® PR 120</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density</td>
<td>Mg/m³</td>
<td>1.73</td>
<td>1.21</td>
</tr>
<tr>
<td>Tensile strength</td>
<td>MPa</td>
<td>2833</td>
<td>104.3</td>
</tr>
<tr>
<td>Tensile modulus</td>
<td>GPa</td>
<td>200</td>
<td>8.0</td>
</tr>
<tr>
<td>Average powder particle size</td>
<td>µm</td>
<td>-</td>
<td>139.4</td>
</tr>
<tr>
<td>Linear roving weight</td>
<td>Tex</td>
<td>760</td>
<td>-</td>
</tr>
</tbody>
</table>

Optimising the towpregs processing conditions. Fig. 2 shows the polymer mass fraction experimentally determined in the commercial market glass fibre reinforced polypropylene (GF/PP) towpregs by varying the coating line oven temperature at different fibre pull-speeds. The polymer fractions were determined by cutting and weighting 1 m length of the towpreg strips produced in the coating line. As expected the polymer mass fraction decreased with increasing of fibre pull-speed
and maxima polymer depositions were obtained for oven temperatures range between 400 °C and 450 °C.

Fig. 2. Variation of the polymer mass fraction with oven temperature and fibre pull-speed

Fig. 3 presents the same type of results for the glass fibre reinforced polyvinyl chloride (GF/PVC) towpregs produced in the coating line using oven temperatures in the range between 260 °C and 315 °C. In this case, it was observed that only in such small temperature range was possible to produce enough good GF/PVC towpregs. A deep decrease in the amount of polymer was verified when lower oven temperatures were used and considerably polymer degradation was observed at higher oven temperatures (great changes in PVC colour). As it may be seen, a good and almost constant level of PVC mass content was obtained by using fibre pull-speeds between 2.0 and 6.0 m/min.

Fig. 3. Influence of production speed on the polymer content of the GF/PVC towpregs

Figures 4 to 6 show the variation of the polymer mass fraction in the Primospire®/Carbon towpregs with fibre pull speeds at three constant oven temperatures. It may be concluded that the polymer mass fraction decreases with the fibre pull speed at the lower oven temperature (600 °C). At the higher oven temperatures, the amount of polymer in the towpregs seems to keep an approximately constant value of 40% at all fibre pull speeds.

Fig. 4. Polymer mass fraction variation with fibre pull speed for 600 °C oven temperature.
Fig. 5. Polymer mass fraction variation with fibre pull speed for 647 °C oven temperature

T=647 °C

Fig. 6. Polymer mass fraction variation with fibre pull speed for 684 °C oven temperature

T=684 °C

Applications of Thermoplastic Matrix Towpregs

Figures 7 to 11 show different successfully developed applications for the thermoplastic towpregs produced in this work. Figure 7 and 8 show a GF/PP pressure vessel with capacity of 0.06 m³ for incompressible fluids able to withstand an internal burst pressure up to 40 bar and a GF/PVC pipe having an internal diameter of 80 mm, respectively.

Fig. 7. Filament wound GF/PP pressure vessel processed from towpregs

Fig. 8. Filament wound GF/PVC pipe
Figures 9 and 10 show a U-shaped $24 \times 4 \times 2$ (mm) GF/PP profile obtained by using the towpreg pultrusion and LFT compression moulded plates also processed from GF/PP towpregs. Such plates were stamped using cut towpregs mixed together at low shear stress to avoid fibre breakage.

Fig. 9. U-Shape GF/PP pultruded profile made from towpregs

Fig. 10. GF/PP LFT plates made from towpregs

Finally, a woven fabric manufactured from CF/Primospire® towpregs and suitable to be processed into a composite part by compression moulding is shown in Fig. 11.

Fig. 11. Primospire®/carbon woven fabric.

Conclusions

The new powder-coating equipment has shown to be suitable to produce towpregs adequate for common and advanced engineering markets. From the tests made, it was found that all of those different towpregs can be easily and continuously produced at industrial production speeds between 2 to 6 m/min.

For common engineering markets glass fibre reinforced polypropylene and polyvinyl chloride matrix were studied. For these materials the optimised processing oven temperatures were in the ranges of 400ºC to 450ºC and 260ºC to 315ºC, respectively.

Carbon fibre reinforced Primospire® towpregs were also studied envisaging possible applications in advanced composite structural markets. In such case, the optimised processing oven window was found in the much higher temperature range from 640 ºC to 690 ºC.

The mechanical properties of the composites processed from these towpregs by major different processing technologies were also found to be adequate either for structural as for common engineering applications.
This work also demonstrated the large potential of polymer powder deposition techniques to fabricate continuous fibre thermoplastic matrix towpregs that can be easily processed into composites with adequate engineering properties. By using efficient processing technologies different composite parts were already manufactured with success.

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References


