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Processing Thermoplastic Matrix Towpregs by Pultrusion

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ABSTRACT: In the work reported herein, glass fiber reinforced polypropylene towpregs, produced continuously at different processing conditions in a dry powder coating equipment (towpregger), were processed into final composites parts in a specially developed prototype pultrusion equipment. The influence of the towpregger fiber pull speed and the furnace temperature on the polymer content of the final towpregs was determined. The towpregs' quality was also assessed using optical microscopy and scanning electron microscopy. They were Q3 then processed into composite profiles in the pultrusion machine, and the influence of the pull speed and dies' temperatures on their mechanical and other relevant physical properties was studied. Finally, the best processing window and the optimization of the final composite profiles are discussed. © 2012 Wiley Periodicals, Inc. Adv Polym Techn 00: 1–7, 2012; View this article online at wileyonlinelibrary.com. DOI 10.1002/adv.21279

Q4 **KEY WORDS:** Composite mechanical properties, Pultrusion, Thermoplastic matrix composites, Towpregs

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Introduction

Nowadays, thermosetting matrices are being successfully replaced by thermoplastics in continuous fiber reinforced composites for advanced applications. This trend led to more durable and recyclable products, with higher thermal and mechanical properties, not involving hazardous chemical reactions during processing.[1–7] Nevertheless, it was only recently that new technologies allowed the production of low-cost continuous fiber- reinforced preimpregnated 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 fibers[8–11] and powder-coated preimpregnated materials (towpregs).[5,12,13] 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 molding, filament winding, and pultrusion.[4,14–21]

In this work, a pultrusion equipment was built to continuously produce composite profiles from glass fiber reinforced polypropylene (GF/PP) towpregs that had been made in a proprietary dry coating equipment (towpregger).[5,22,23] The towpregs processing window was optimized by varying the fiber pull speed and furnace temperature in the coating line and determining the influence of these parameters on the final polymer mass fraction. Two different pultrusion head tools (dies) were also designed to be used in the pultrusion equipment, allowing the production of profiles with two different shapes. Finally, the performance of the profiles was evaluated by mechanical testing. From the preliminary results obtained, it was concluded that they have adequate properties for application in common and structural engineering markets.

Experimental

POWDER COATING AND PULTRUSION EQUIPMENTS

The prototype powder coating equipment used to produce fiber-reinforced towpregs is schematically depicted in Fig. 1.[5,23] It consists of six main parts: a wind-off system, a fibers spreader unit, a heating section, a coating section, a consolidation unit, and a wind-up section. Initially, the reinforcing fibers 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 of the polymer powder, promoting its adhesion to the fiber surface. Finally, the thermoplastic matrix towpreg is cooled down and wound up on a spool.

Figure 2 shows a general overview of the existing powder coating equipment. Figures 3 and 4 depict a schematic and a photograph, respectively, of the pultrusion equipment developed in the present work.

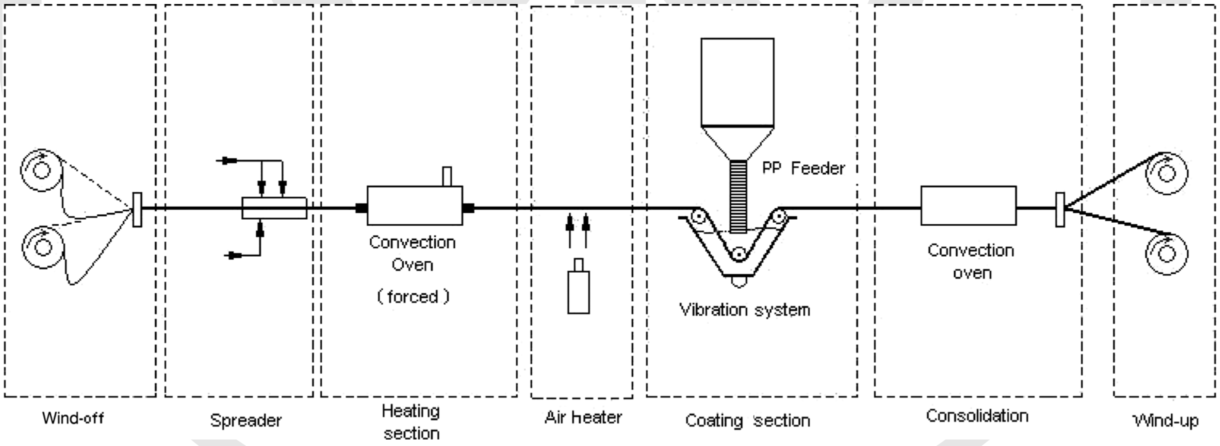

FIGURE 1. Schematic diagram of the powder coating line setup.

FIGURE 2. General overview of the powder coating equipment.

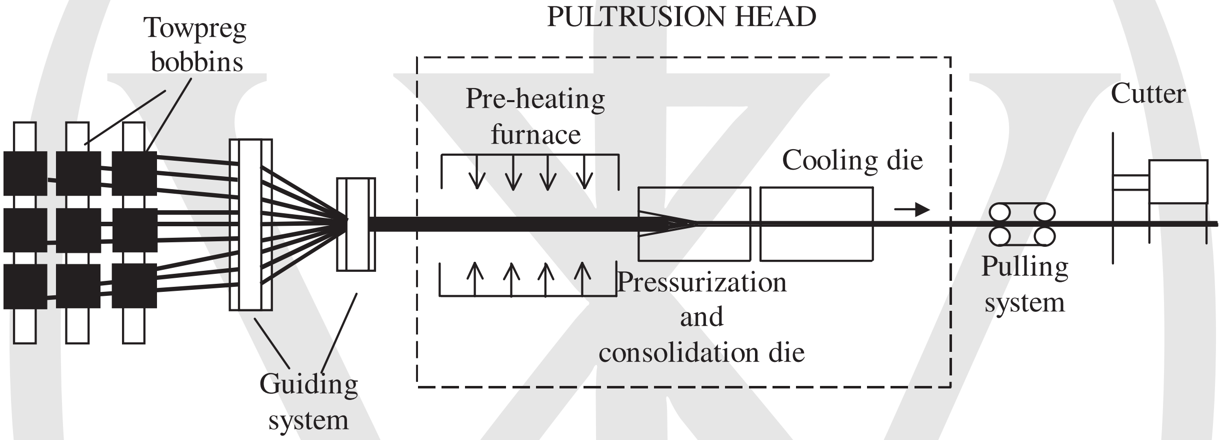

FIGURE 3. Schematic diagram of the proprietary pultrusion line.

FIGURE 4. Overview of the proprietary pultrusion equipment.

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 preheating furnace and the pressurization/consolidation and cooling dies,
- **iv.** pulling system, and
- **v.** the final profile cutting system.

To produce the composite profiles, the towpregs are guided into the preheating furnace, where the material is heated up to the required temperature. The towpregs then enter in the pultrusion die; in its first zone, the material is heated up and consolidated, and in the second it is cooled down to the required size. After solidification, the pultruded material is cut into specified lengths.

The preheating furnace, which may reach a temperature of 1000°C, was designed to allow processing almost every type of fiber/thermoplastic-based towpregs.

Two different groups of dies (pressurization/consolidation and cooling dies) were already designed. One allows to produce the Ushaped profile shown in Fig. 5 and the other a 20 × 2 mm tape-shaped profile.

RAW MATERIALS

The GF/PP towpregs used in the present work were produced by using 2400 Tex type E glass fiber rovings and a polypropylene from Owens Corning and ICO Polymers France (Icorene® 9184B P), respectively. The most relevant properties of both materials are summarized in Table I. Q5 Q6

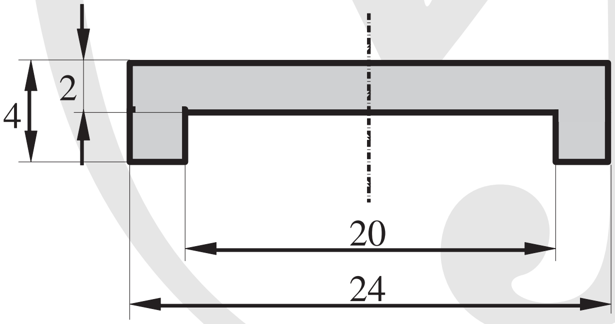

FIGURE 5. U-shaped profile considered in the die design.

TABLE I
Properties of the Raw Materials used to Produce the GF/PP Towpregs

Property	Units	Glass Fibers	Polypropylene
Density	Mg/m^3	2.56	0.91
Tensile strength	MPa	3500	30
Tensile modulus	GPa	76	1.3
Average powder particle size	μm	–	440
Linear roving weight	Tex	2400	–

GF/PP TOWPREGS PROCESSING CONDITIONS

Figure 6 shows the variation of the experimental polymer mass fraction determined in towpregs obtained with the coating line at different oven temperatures and fiber pull speeds. The determination was done by cutting and weighting 1-m length of towpreg strips and using the following expression:

$$\omega_{\mathrm{p}} = \left(1 - \frac{W_{\mathrm{f}}}{W_{\mathrm{T}}}\right) \times 100 \qquad (1)$$

where ω_{p} is the polymer mass fraction (in%), W_{f} is the fiber roving linear weight per meter (in kg; see Table I), and W_{T} is the total weight measured per meter of towpreg (in kg).

As it may be seen in Fig. 6, the polymer mass fraction decreases with increasing of fiber pull speed, as expected. Maxima polymer depositions were obtained when temperatures between 400 and 450°C were used in the convection oven. Such temperatures do not correspond to the actual towpreg

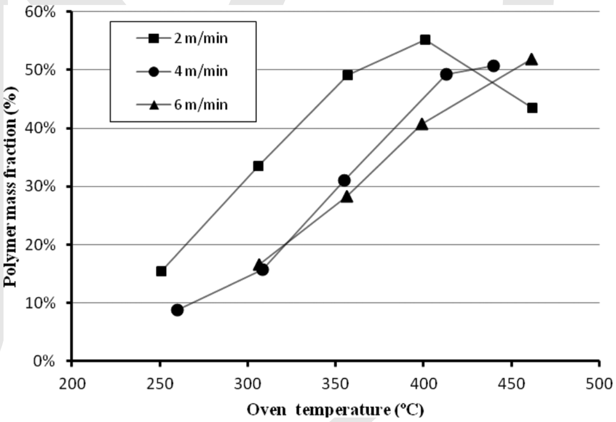

FIGURE 6. Variation of the polymer mass fraction with the oven temperature and fiber pull speed.

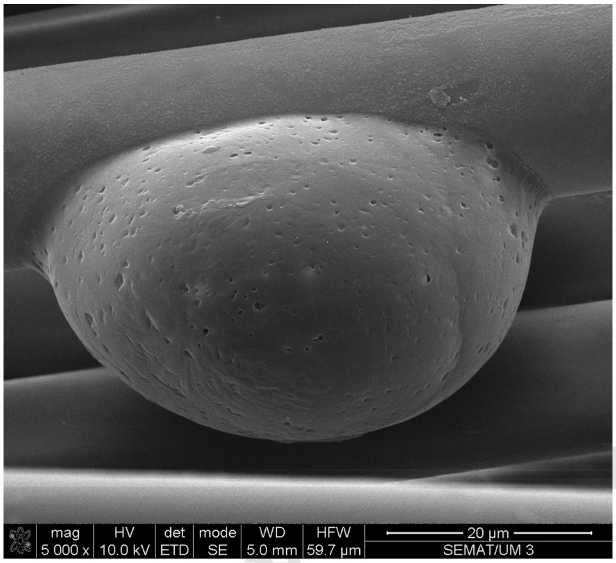

FIGURE 7. SEM micrograph of a polymer droplet adhered to a fiber in the towpreg (magnification of 5000×).

surface temperature, which is necessarily lower, but to that measured by the thermocouples inside the oven.

Several towpreg samples were analyzed under a Nova NanoSEM 200 scanning electron microscope (SEM) to evaluate the adhesion of the polymer powder to the fibers and its distribution. Figures 7 and 8 show SEM micrographs of towpreg samples produced using a temperature in the convection oven around 400°C and a fiber pull speed of 4 m/min. As may be seen, good polymer melting and adhesion (see Fig. 7) and a reasonable polymer powder distribution (see Fig. 8) on the glass fibers were achieved at these optimized operating conditions.

PULTRUSION

The GF/PP profiles were produced in the pultrusion equipment using the typical operating conditions presented in Table II.

TABLE II
Typical Pultrusion Operating Conditions

Variable	Units	Value
Pultrusion pull speed	m/min	0.2–0.8
Preheating furnace temperature	°C	160–250
Die temperature	°C	250–380
Cooling die temperature	°C	25–60

As may be seen, in the preliminary tests, it was already possible to produce GF/PP profiles in good conditions at pull speeds of 0.2 m/min. The optical micrograph in Fig. 9 shows that a quite homogeneous fiber/matrix distribution was obtained along the profile cross section.

Currently, experiments are been made to increase the pultrusion pull speed to values in the range between 2 and 6 m/min, which would allow producing the GF/PP profiles directly at the end of the dry coating towpreg line. In fact, the possibility of assembling a pultrusion head to the towpregger will be a major achievement of this future work. For this, it will be necessary to process the final GF/PP profiles at pulling speeds similar to those used in the towpreg equipment. This ultimate goal is yet to be achieved.

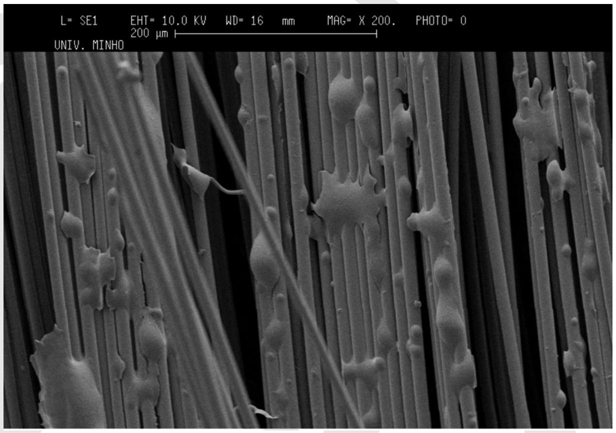

FIGURE 8. SEM micrograph showing the polymer powder distribution in the towpreg (magnification of 200×).

FIGURE 9. Optical micrograph of the pultruded profile cross section (magnification of 50×).

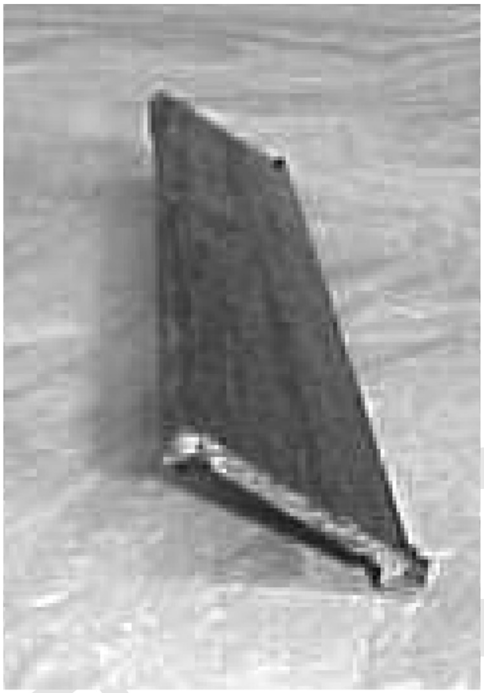

FIGURE 10. U-Shaped pultruded profile.

As it may be seen in Fig. 10, the pultruded rectangular profiles present well defined and smooth surfaces. However, some imperfections are still detectable (a few naked fibers can be observed on the surface), which seem to result from the nonuniform distribution of the polymer particles that had been observed in the initial towpregs.

MECHANICAL PROPERTIES OF THE PULTRUSION PROFILES

Table III presents the mechanical properties determined from flexural and tensile tests carried out on pultruded profiles with a fiber volume content around 55%. The fiber 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 former, specimens weighting approximately 20 g, cut from the profiles, were calcinated during 10 min at 600°C inside a Naberterherm® S30 muffle furnace (Naberterherm, Lilienthal, Germany). For the second determination, rectangular, 100 mm × 20 mm × 2 mm Q7 samples were submitted to three-point bending tests using an 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 Q8 on 250 mm × 15 mm × 2 mm rectangular samples, at a 2-mm/min cross-head speed, using the same equipment and load cell. A SG Shimadzu® 50–mm length strain gauge was used up to 0.3% strain to allow determining accurately the tensile modulus on each sample.

The tensile strength and modulus in the direction of the fibers of the GF/PP plates, σ_1 and E_1, respectively, were predicted from the fibers and polymer properties via the wellknown law of mixtures:

$$\sigma_1 = \sigma_f v_f + \sigma_m (1 - v_f) \tag{2}$$

and

$$E_1 = E_f v_f + E_m (1 - v_f) \tag{3}$$

where σ_f, E_f, and v_f are the glass fibers tensile strength, modulus, and volume fraction, respectively, and E_m and σ_m and are the matrix modulus and tensile strength at the fiber strain to break, respectively.

As can be seen from Table III, experimental strength results lower than the theoretical ones were obtained. In any case, such results seem to be compatible with the major commercial applications expected of GF/PP composites. On the contrary, the experimental moduli obtained are already in good agreement with the theoretical ones.

TABLE III
Mechanical Properties of GF/PP Composites

	Tensile Strength (MPa)		Tensile Modulus (GPa)		Flexural Strength (MPa)		Flexural Modulus (GPa)		Fiber Mass Fraction (%)		Fiber Volume Fraction (%)	
Kind of Data	Av.	SD	Av.	SD	Av.	SD	Av.	SD	Av.	SD	Av.	SD
Determined	>305	26	29.9	3.5	124.6	4.3	27.1	0.3	78.4	1.4	56.2	2.8
Theoretical	661.6	219	35.6	7.4	661.6	219	35.6	7.4				

Av.: average; SD: standard deviation.

Conclusions

An existing powder coating equipment was shown to be suitable to produce GF/PP towpregs that could be adequately processed into composite pultruded profiles. From the tests made, the towpregs can be easily and continuously produced at industrial production speeds between 2 and 6 m/min. An optimized oven temperature range between 400 and 450°C was determined for processing of these towpregs.

The preliminary tests made using a proprietary pultrusion equipment already allow producing profiles at pull speeds until 0.8 m/min. Currently, work is carried out to increase the pultrusion-processing speed in the range from 2 to 6 m/min, which will be equivalent to the speed of the pultrusion line with
Q9 that of the towpreg coating line. In the future, the use of similar operational speeds in both processes (equipments) will make possible to assemble them in just one equipment.

The mechanical properties of GF/PP profiles processed from these towpregs were also found to be adequate either for common or structural engineering applications. In particular, very good agreement was found between the experimental moduli and the theoretical ones.

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