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## **PROCESSING THERMOPLASTIC MATRIX TOWPREGS BY PULTRUSION**

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### **ABSTRACT**

In this work glass fibre reinforced polypropylene (GF/PP) towpregs, continuously produced at different processing conditions in a dry powder coating equipment, were processed into final composites by using a developed prototype pultrusion equipment.

The influence of the fibre pull-speed and furnace temperature used in the dry coating towpregger on the polymer content of final produced towpregs was determined. Optical microscopy and SEM were techniques also used to assess quality of the produced GF/PP towpregs.

The GF/PP towpregs were then processed into a composite bar by using a prototype pultrusion machine that will be described and presented in the workshop. Studies were carried out to determine the influence of the pull-speed and temperatures used in the pultrusion heated and cooled dies on the mechanical and other relevant physical properties of final composite profiles. Finally, the final composite profiles processing window and optimisation will be discussed and presented.

**Keywords:** Towpregs, Thermoplastic matrix composites; Pultrusion; composite mechanical properties

### **1. 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, and not involving hazardous chemical reactions during processing [1, 2]. Nevertheless, only recently newly developed dry-coating technologies allowed the substitution of expensive thermoplastic matrix prepregs (obtained by melting and solvent based processes to impregnate the fibers with the high viscosity thermoplastics) by cheap powder pre-impregnated materials, known as towpregs [2-4]. Today, work is in progress to consolidate and process these promising towpregs by using existing high volume rate technologies for composite parts [5-8].

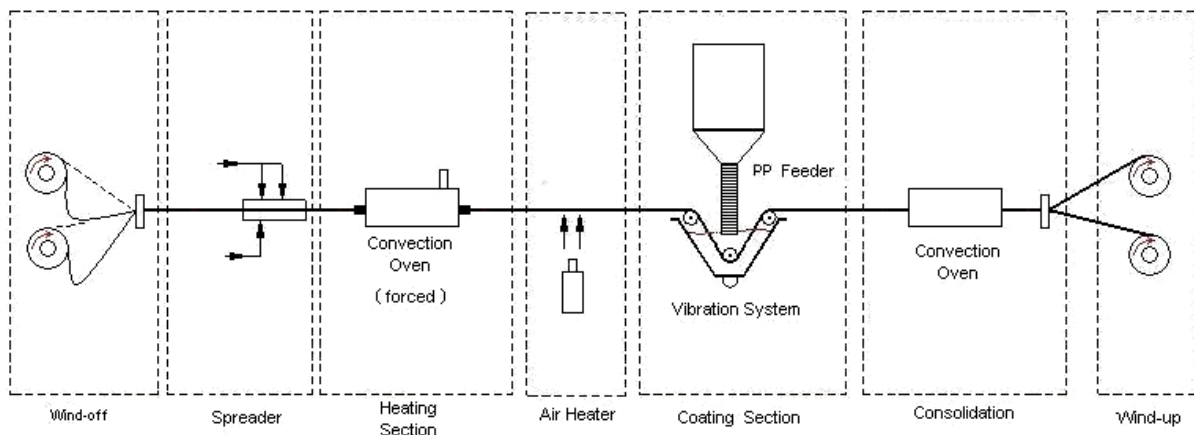
In this work, a pultrusion equipment was built to produce composite profiles from thermoplastic matrix towpregs continuously by using glass fibre reinforced polypropylene

(GF/PP) towpregs produced in a previously developed dry coating equipment (towpregger) [2, 9, 10]. The GF/PP towpregs processing window was optimised by varying the fibre pull-speed and furnace temperature in the coating-line and determining the influence of these parameters on their final polymer mass fraction. Two different pultrusion-head tools (dies) were also design for being used in the new pultrusion equipment for allow producing GF/PP composite profiles with two different shapes. Finally, the performance of the final GF/PP composite profiles produced was evaluated by mechanical testing. From the preliminary results obtained it was concluded that the produced GF/PP profiles have adequate properties for application in structural and common engineering markets.

## 2. EXPERIMENTAL

### 2.1 Powder coating and pultrusion equipments

The prototype powder coating equipment used to produce fibre reinforced towpregs is schematically depicted in Figure 1 [2, 10]. 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

The photograph depicted in Figure 2 shows a general overview of the developed powder coating equipment.

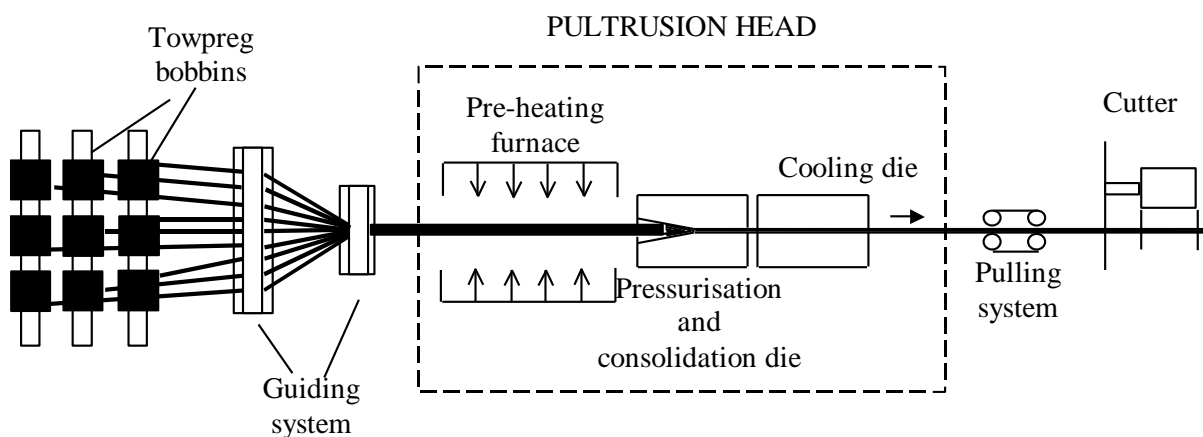


**Fig. 2.** General overview of the powder coating equipment.

The 10 kN pultrusion equipment (see Figures 2 and 3) developed in the present work to produce GF/PP composite profiles may be also 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 composite profiles, the thermoplastic matrix towpregs are guided into the pre-heating furnace where the material is heated up to a required temperature. Then, the towpregs enter in the pultrusion die; in its first zone, the material is heated up and consolidated, and in the second is cooled down to the required size. After reaching the solid state the pultruded material is cut into specified lengths.



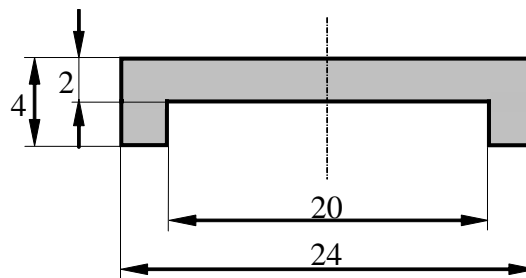
**Fig. 2 –** Schematic diagram of the developed pultrusion line

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



**Fig. 3** – Overview of the developed pultrusion equipment

Two different groups of dies (the pressurization/consolidation and cooling ones) were already designed. One to produce the U-shaped profile shown in Figure 4 and other to allow process a 30x2 mm tape-shaped profile.



**Fig. 4** – U-shaped profile considered in the die design

## 2.2 Raw materials

The GF/PP towpregs used in the present work were produced by using 2400 Tex type E glass fibre rovings and a polypropylene from Owens Corning and ICO Polymers France (Icorene 9184B P), respectively. The most relevant properties of both materials are summarised in Table 1.

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

Property	Units	Glass fibres	Polypropylene
Density	Mg/m <sup>3</sup>	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	-

### 2.3 GF/PP towpregs processing conditions

Figure 5 shows the polymer mass fraction experimentally determined in the produced glass fibre reinforced polypropylene (GF/PP) towpregs by varying the dry 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 it may be seen, the polymer mass fraction decreased with the increasing of fibre pull-speed as expected and, maxima polymer depositions were obtained for oven temperatures range between 400 °C and 450 °C.

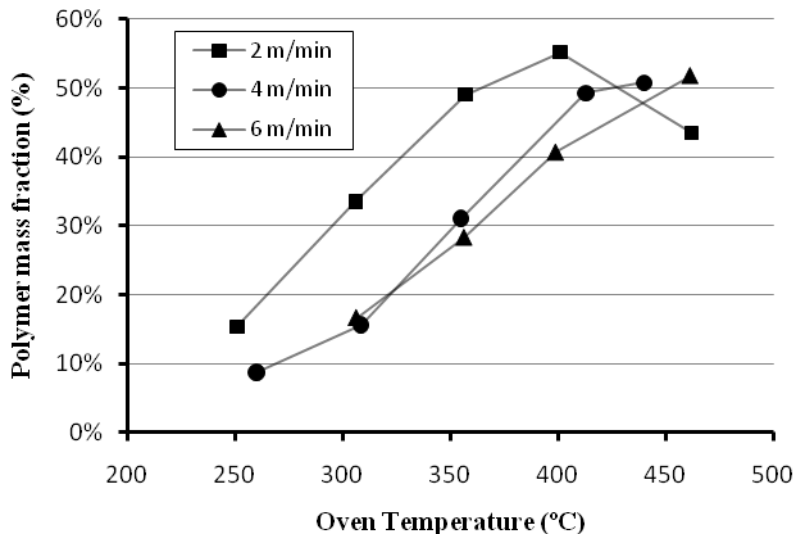


Fig. 5. Variation of the polymer mass fraction with oven temperature and fibre pull-speed

Several samples of the GF/PP produced towpregs were analysed under a Nova NanoSEM 200 scanning electron microscope to evaluate the polymer powder distribution and its adhesion to the fibres. Figure 6 shows a SEM micrograph of a towpreg sample produced in the dry coating line using an oven temperature around 400 °C and a fibre pull-speed of 4 m/min. As may be seen in this figure a good polymer melting and adhesion to glass fibres was achieved at those optimised coating line operating conditions.

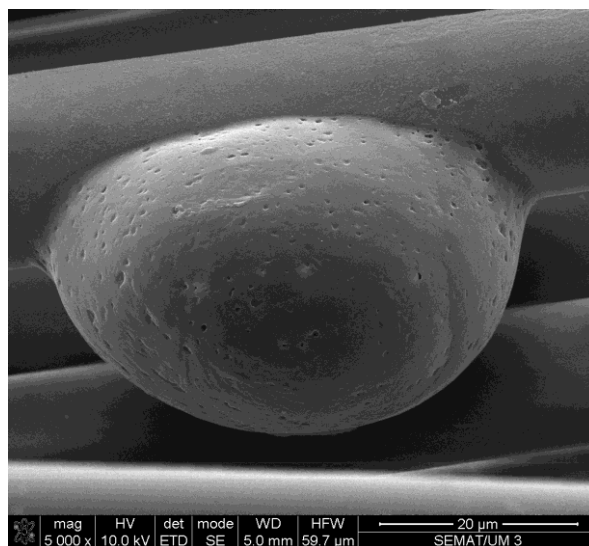


Fig. 6. SEM micrograph of GF/PP towpreg (magnification of 5000x).

## 2.4 Pultrusion

The GF/PP profiles were produced in the developed 10 kN pultrusion equipment using the typical operational conditions presented in Table 2.

As may be seen, it was already possible to produce in good conditions GF/PP profiles at pull speeds near 1 m/min in the preliminary tests. Now, we are still trying to increase the pultrusion pull speed for values in the range between 2 m/min to 6 m/min, which allow producing the GF/PP profiles directly at the end of the dry coating towpreg line. In fact, the possibility of assembling the pultrusion-head to the towpregger is a major achievement of this work in future. To do this it will be necessary to be able of processing the final GF/PP profiles at similar pulling speeds of those used in the towpreg production.

**Table 2.** Typical pultrusion operational conditions.

Variable	Units	Value
Pultrusion pull speed	m/min	0.5-0.8
Pre-heating furnace temperature	°C	200-250
Die temperature	°C	300-320
Cooling die temperature	°C	60

As it may be seen from the photo of a produced U-Shaped profile depicted in Figure 7, the pultruded profiles presented well-defined and smooth surfaces. However, some detected imperfections (few naked fibres shown at surface) seem to result from non-uniform distribution of polymer particles also observed in the initial towpregs.



**Fig. 7** – U-Shaped pultruded profile

## 2.5 Mechanical properties of the pultrusion profiles

Table 3 presents the mechanical properties obtained in the pultruded profiles processed from the initial produced GF/PP towpregs. They were obtained from flexural and tensile tests carried on samples presenting glass fibre mass content around 50%.

The fibre mass fraction, flexural and tensile properties of composite profiles fabricated were determined in accordance to ISO 1172, ISO 178 and EN 60, respectively.

**Table 3.** Mechanical properties of GF/PP composites

Kind of Data	Tensile strength (MPa)		Tensile modulus (GPa)		Flexural strength (MPa)		Flexural modulus (GPa)		Fibre mass fraction (%)		Fibre volume fraction (%)	
	Av.	SD	Av.	SD	Av.	SD	Av.	SD	Av.	SD	Av.	SD
Determined	305	26	29.9	3.5	>117	4.3	22.5	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				

As can be seen from Table 3, experimental strength results lower than the theoretical expected ones were obtained. In any case, such strength results seem to be compatible with the major commercial applications expected for GF/PP composites. Anyway, the experimentally modulli results obtained already present good agreement with the theoretical expected ones.

### 3. CONCLUSIONS

The powder-coating equipment has shown suitable to produce GF/PP towpregs adequate for being processed into composite pultruded profiles. From the tests made, it was found that the towpregs can be easily and continuously produced at industrial production speeds between 2 a 6 m/min. An optimised oven range temperature between 400°C and 450°C was determined for processing these towpregs.

The preliminary tests made by using the new developed pultrusion equipment already allow producing GF/PP profiles at pull speeds until 0.8 m/min. Currently, work is carried out to try increasing the pultrusion processing speed to values in the range from 2 m/min to 6 m/min, which make more compatible the processing speeds in both developed pultrusion and towpreg coating-line equipments. In future, the use of similar operational speeds in both processes (equipments) will turn possible assembling or joining them in just only one equipment.

The mechanical properties of GF/PP profiles processed from these towpregs were also found to be adequate either for structural as for common engineering applications. Very good agreement was especially found between the experimental determined modulli values and theoretical expected ones.

### References

- [1] Brandt, J., Drechsler, K. and Richter, H. The Use of High-Performance Thermoplastic Composites for Structural Aerospace Applications, 9th Int. Conf. on Composite Materials (ICCM-9), Vol. 6, pp. 143-150, Madrid, Spain, July 1993.
- [2] Nunes, J. P., van Hattum, F.W. J., Bernardo, C. A., Brito, A. M., Pouzada, A. S., Silva, J. F. e Marques, A. T.. Part III: Chapter 11- Production of Thermoplastic Towpregs and Towpreg-based Composites, In: Polymer Composites – From Nano to Macro-Scale, Eds K. Friedrich, S. Fakirov and Z. Zhang, Springer Science+Business Media, Inc., New York/USA, pp. 189-214, 2005.

- [3] Nunes, J. P., Silva, J. F., Velosa, J. C., Bernardo, C. A. and Marques, A. T. Production of New Thermoplastic Matrix Composites For High Demanding Applications, 13th European Conference on Composite Materials –ECCM 13, Stockholm, Sweden, June 2008.
- [4] Nunes, J. P., Amorim, L., Velosa, J. C. and Silva, J. F. Optimizing the Continuous Dry Impregnation of Thermoplastic Matrix Fibre Reinforced Materials, 14th European Conference on Composite Materials – ECCM 14, Budapest, Hungary, June 2010.
- [5] Silva, J. F., Nunes, J. P., Vieira. P. and Marques, A. T. GF/PP Towpregs Production Testing and Processing. *Int. Journal Mec. Maters Des.* 4, pp. 205-211, 2008.
- [6] Velosa, J. C., Nunes, J. P. , Antunes, P. J., Silva, J. F. and Marques, A. T. Development of a New Generation of Filament Wound Composites Pressure Cylinders. *Composites Science and Technology*, 69, pp. 1348-1353, 2009.
- [7] Silva, J. F., Nunes, J. P. and Bernardo, C. A. Determining the Final Properties of Thermoplastic Matrix Composites Produced from Towpregs, 14th European Conference on Composite Materials – ECCM 14, Budapest, Hungary, June 2010.
- [8] Nunes, J. P. and Silva, J. S. Production of Thermoplastic Matrix Towpregs for Highly Demanding and Cost-Effective Commercial Applications, VI International Materials Symposium - MATERIALS 2011, Guimaraes, Portugal, April 2011.
- [9] Nunes, J. P., Silva, J. F., Silva, L., Novo, P. J. and Marques, A. T., Equipment to Produce Continuously Powder Coated Thermoplastic Matrix Prepregs (Towpregs), *Internacional Patente WO 02/06027 A1*, July 2001.
- [10] Fazenda, R., Silva, J. F., Nunes, J. P., Bernardo, C. A., New Coating Equipment To Produce Long Fibre Thermoplastic Matrix Towpregs at Industrial Scale, *Proceedings of the ANTEC'07, Cincinnati, Ohio/EUA*, May 6-10, 2007.