PRODUCTION AND PROCESSING OF THERMOPLASTIC BASED TOWPREGS FOR COMMERCIAL AND ADVANCED MARKETS

J.P Nunes^{a*}, P. J. Novo^b, J. F. Silva^c, A. T. Marques^d

^a Institute of Polymers and Composites/I3N, Minho University, 4800-058 Guimarães, Portugal

^b School of Tech. and Management, Polytechnic Institute of Leiria, 2411-901 Leiria, Portugal

^c Dept. of Mechanical Engineering, ISEP, 4200-072 Porto, Portugal

^d DEMEGI / FEUP, 4200-465 Porto, Portugal

^{*}jpn@dep.uminho.pt

Keywords: Thermoplastic matrix composites, Towpregs, Processing, Mechanical properties

Abstract

In this work continuous fibre reinforced thermoplastic matrix towpregs were produced for commercial markets and advanced markets. The former based on glass fibre reinforced polypropylene matrix (GF/PP) and the latter on carbon fibre Primospire[®] matrix (CF/P). Primospire[®] is an amorphous highly aromatic material specifically developed by Solvay Advanced Polymers for application in advanced markets.

Both towpregs were processed into composite parts by different technologies. The mechanical properties determined on the final composites were compared with the theoretical predictions and have shown to be acceptable for the targeted markets.

1. Introduction

Continuous fibre reinforced thermoplastic matrix composites have been successfully employed in the aircraft, military and aerospace industries due to the excellent properties. In these and many other commercial engineering applications, they can replace other materials, such as 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 [1-3].

Two major technologies are used to allow wet reinforcing fibres with thermoplastic polymers [2, 3]: i) the direct melting of the polymer and, ii) the intimate fibre/matrix contact prior to final composite fabrication. Continuous fibre reinforced thermoplastic matrix pre-impregnated tapes are, for example, produced by direct melting processes. Alternatively, intimate contact processes allow producing cheap and promising pre-impregnated materials, such as, commingled fibres, co-woven fabrics and towpregs.

In this work, towpregs based on different fibres and thermoplastic matrices were produced and processed into composites for highly demanding and more commercial applications. Heated compression moulding and pultrusion were the processing methods used to obtain final composite parts. The processing parameters used both to produce the towpregs in our own developed coating line equipment [4, 5] as well as to process them, at industrial compatible production rates, into final parts composite having adequate properties were studied. Thus, the efficient processing windows allowing producing continuously the thermoplastic matrix towpregs, by dry deposition the thermoplastic matrix on the reinforcing fibres, and also to transform them into composites were established. Two different raw-materials were used in the production of the thermoplastic matrix towpregs, those to be used in parts for highly demanding markets were based on carbon fibres and Primospire[®] [6] and those for more commercial composites on glass fibre and polypropylene.

2. Experimental

2.1. Raw materials

The PP powder ICORENE 9184B P® from ICO Polymers and type E glass fiber direct rovings 305E-TYPE 30® from Owens Corning were, respectively, the raw materials used to produce the glass fibres reinforced polypropylene matrix towpregs studied in this work and intended for being used in more common composites for commercial markets. Table 1 shows the main properties of those materials.

Property	Units	Glass fibres	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
Melting temperature	°C	-	166
Linear roving weight	Tex	2400	-

Table 1. Properties of raw materials used in towpregs for common applications.

On other hand, composite parts for highly demanding advanced markets were processed from towpregs manufactured by using a highly aromatic amorphous thermoplastic polymer in powder form, the PRIMOSPIRE® PR 120 from Solvay Advanced Polymers, and 760 Tex M30SC carbon fibre tows TORAYCA. Table 2 presents the most relevant properties determined on both these raw materials.

Property	Units	Carbon fibres	Primospire®
Density	Mg/m ³	1.73	1.21
Tensile strength	MPa	2833	104.3
Tensile modulus	GPa	200	8.0
Average powder particle size	μm	-	139.4
Glass transition temperature (Tg)	°C	-	158
Linear roving weight	Tex	760	-

Table 2. - Properties of raw materials used in towpregs for advanced applications

2.2. Production of the towpregs

The towpregs were produced in a developed dry powder coating equipment schematically shown in Figure 1 and illustrated in the photo of Figure 2 [4, 7]. It consists of six main parts: i) wind-off system, ii) fiber spreader unit, iii) heating section, iv) polymer coating section, v) consolidation unit and vi) 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 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 1. Schematic drawing of the powder coating line



Figure 2. Powder coating line equipment used to produce towpregs

In order to optimise the conditions to produce towpregs, the powder coating equipment was operated at different woven temperatures and fibre linear pull speeds. From such work the best values of the operational variables depicted in Table 3, which allow simultaneously producing towpregs in good and stable circumstances and having the maximum polymer powder content were determined.

		V	Values	
Variable	Units	GF/PP towpregs	CF/Primospire [®] towpregs	
Convective oven temperature	°C	700	700	
Consolidation furnace temperature	°C	400	650	
Coating line pulling speed	m/min	4	5	

Table 3. Best conditions to produce towpregs used in composites for commercial and advanced markets.

2.3. Processing composites by compression moulding

A technique described elsewhere [8] was used to produce unidirectional fibre reinforced laminate plates with $100 \times 100 \times 4$ mm directly from the towpregs. First, the towpreg were wound over a plate with appropriate dimensions and the resultant pre-form then conveniently placed in the cavity of a heated mould. A 400 kN SATIM hot platen press was used to obtain the desired consolidation pressure. After heating the cavity, pressure was applied and, finally, the mould was cooled down to room temperature and the final composite laminate plate removed.

Table 4 shows the compression moulding conditions used to process composites by using the two kinds of towpregs produced in the present work

		Values	
Variable	Units	GF/PP towpregs	CF/Primospire [®] towpregs
Platen temperature	°C	250	320
Compression pressure	MPa	20	20
Compression time	min	15	20
Final cooling temperature (at press opening)	°C	30	30

Table 4. Conditions used to process composites by compression moulding by using the towpregs

2.4. Flexural properties of the composite plates obtained by compression moulding

The flexural properties in fibre direction were determined in the composite plates obtained by compression moulding, using three-point bending tests accordingly to ISO 178 standard. The tests were made in an universal INSTRON 4505 testing machine on five $100 \times 15 \times 4$ (mm) specimens cut from the processed composite plates and by using a cross-head speed of and a distance between supports of 2 mm/min and 80 mm, respectively. The fibre mass fraction was also determined according to EN 60. Table 5 summarizes the experimental results obtained on the two different studied composites.

As may be seen, flexural properties compatible with the applications envisaged for the composites processed from the produced towpregs were obtained in this work. Better

properties may be certainly obtained through the improvement of fibre/matrix adhesion, polymer powder distribution and fibre alignment.

Property	Units	GF/PP	CF/Primospire [®]
Flexural strength	MPa	66.3±9.4	124.3±15.0
Flexural modulus	GPa	24.7±2.6	30.0±5.0
Fibre mass fraction	%	85.6±1.6	59.7±0.3

Table 4. Properties of composite plates made from the towpregs

2.5. Processing composites by pultrusion

A 10 kN pultrusion equipment [9] was purposely designed and built to allow producing of continuous profiles made from thermoplastic matrix towpregs. Such equipment, schematically shown in Figure 3 and illustrated in Figure 4, includes three main parts: a pre-heating furnace, a pressurisation and consolidation die and a cooling die.



Figure 3. Schematic diagram of the developed pultrusion line

The towpregs are guided into the pre-heating furnace where the material is heated up to the required temperature. In the entering zone of the pultrusion die, the material is heated up and consolidated, and then cooled down to achieve the desired shape. After reaching the solid state the material is cut into specified lengths.



Figure 4. Developed prototype pultrusion line

		Values		
Variable	Units	GF/PP towpregs	CF/Primospire [®] towpregs	
Pultrusion pull speed	m/min	0.2	0.2	
Pre-heating furnace temperature	°C	160	400	
Pressurisation/consolidation die temperature	°C	260	475	
Cooling die temperature	°C	20	20	

Two 20×2 (mm) rectangular cross-section bars were pultruded from both types of towpregs produced in this work in operational conditions presented in Table 5.

Table 5. Conditions used to process the pultruded composite bars from the towpregs

As it may be seen and as expected, the CF/Primospire® towpregs required the use of much higher temperatures than the GF/PP ones in pre-heating furnace and pressurization/consolidation die. Due to such higher temperatures, tests still continue being done to optimise the operational conditions to be used in the pultrusion of the CF/Primospire® towpregs. We expect to be able to present the final results obtained from those tests in the conference.

Table 6 presents the mechanical properties obtained on the already pultruded profiles. As it was previously mentioned tests still are being made on pultruded profiles processed from the CF/ Primospire® towpregs we expect to present in the conference.

Property	Units	Value
Flexural strength	MPa	241.2±1.6
Flexural modulus	GPa	90.1±0.4
Fibre mass fraction	%	85.6±1.6

Table 6. Flexural test results on the pultruded composite profiles processed from the GF/PP towpregs

As results from above Table 6 show, composites pultruded from GF/PP towpregs presented much higher mechanical properties than those obtained on the compression moulding ones. This seems to be related with the much better consolidation and fibre alignment that pultrusion allows achieving.

3. Conclusions

Two different types of thermoplastic matrix towpregs were studied and produced in this work: one intended to be used in common commercial markets, made from glass-fibre reinforced polypropylene (GF/PP), and the other one using a carbon fibre reinforced Primospire[®] (CF/ Primospire[®]) for more high demanding advanced markets.

The production of both towpregs was optimised in order to maximise the polymer powder deposition and operating stable conditions.

The processing of both produced thermoplastic matrix towpregs into composite parts by compression moulding and pultrusion was also studied. The mechanical properties of the

processed composites were determined and evaluated. From obtained results it was possible to conclude that the mechanical properties were compatible with the requirements of the envisaged applications. As expected, composites processed by compression moulding from the CF/ Primospire[®] towpregs presented higher mechanical properties than those obtained from the GF/PP towpregs.

It was also possible to conclude that the pultruded profiles processed from the GF/PP towpregs presented much higher mechanical properties than those ones determined on compression moulding plates made with the same material. This seems to be mainly related with the better consolidation and more accurate fibre alignment that the pultrusion processing allows achieving.

In case of pultrusion, work still is ongoing in order to optimise the processing and determine the mechanical properties on pultruded profiles made from CF/ Primospire[®] towpregs.

References

- [1] Åström T., Carlsson A. Experimental investigation of pultrusion of glass fibre reinforced polypropylene composites. *Composites Part A: Applied Science and Manufacturing*, Elsevier, pp. 585-593, 1998.
- [2] Gibson A. G., Manson J. A. Impregnation technology for thermoplastic matrix composites. *Composites Manufacturing*, Vol 3 (4), pp. 223-233, 1992.
- [3] Bechtold G., Wiedmer S., Friedrich K. Pultrusion of Thermoplastic Composites New Developments and Modelling Studies. *Journal of Thermoplastic Composite Materials*, Vol. 15, pp. 443-465, 2002
- [4] Silva, R. F., Silva, J. F., Nunes, J. P., Bernardo, C. A., Marques, A. T. New Powder Coating Equipment to Produce Continuous Fibre Thermoplastic Matrix Towpregs. In "Materials Science Forum", *Trans Tech Publications*, Vol. 587-588, pp. 246-250, 2008
- [5] Silva, J. F., Nunes, J. P., Vieira, P., Marques, A. T. GF/PP Towpregs Production, Testing and Processing. *International Journal of Mechanics and Materials in Design*, Vol 4 (2), pp. 205-211, 2008.
- [6] Nunes, J. P.; Silva, J. F.; Velosa, J. C.; Bernardo, C. A.; Marques, A. T. New thermoplastic matrix composites for demanding applications. *Plastics, Rubber and Composites*, Vol.38 (2–4), pp.167–172, 2009.
- [7] 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 ANTEC'07*, Cincinnati, Ohio/USA, May 6-10, 2007.
- [8] Klett, J. W., Albiger, J., Edie D. D. and Lickfield, G.C. Production and Evaluation of a Polyimide/Carbon Fiber Powder-Coated Towpreg. *Proceedings of the Seventh Inter. Conference on Carbon, Carbon '92*, pp. 683-685, Essen, 1992.
- [9] Nunes, J. P., Silva, J. F. and Novo, P. J. Processing Thermoplastic Matrix Towpregs by Pultrusion. Advances in Polymer Technology, Vol. 32 (S1), pp. E306–E312, March 2013.