

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

P. J. Novo^{1*}, J. F. Silva^{2,3}, J. P. Nunes³, F.W.J. van Hattum³, A. T. Marques⁴

¹ School of Tech. and Management, Polytechnic Institute of Leiria, Morro do Lena-Alto do Vieiro, Apt. 4163, 2411-901, Leiria, Portugal

² ISEP, Dept. of Mechanical Engineering, R. Dr. Antonio B. Almeida 431, 4200-072 Porto, Portugal

³ Institute for Polymers and Composites/I3N, Minho University, Campus de Azurem, 4800-058 Guimaraes, Portugal

⁴ FEUP, Porto University, R. Dr. Roberto Frias, 4200-465 Porto, Portugal

* e-mail: paulo.novo@ipleiria.pt

Keywords: Towpreg, pultrusion, fibreglass composites, thermoplastic matrix composites

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 comingled 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.

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
Linear roving weight	Tex	2400	-

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.

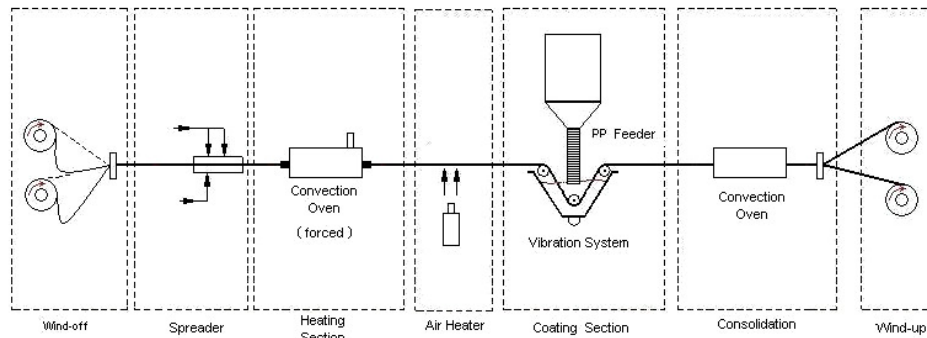


Figure 1. Powder coating line setup.

On other hand, thermoplastic pre-consolidated tapes were manufactured by using a new specially developed cross-head extrusion hot melting process (Fig. 2).

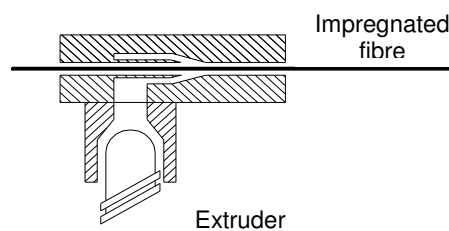


Figure 2. Cross-head extrusion.

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.

Property	Description
Fibre type	E-Glass, 2400 Tex
Fibre diameter	17 μm
Fibre content	60 wt. %
Matrix type	Polypropylene (PP)
Tape width	25 mm
Tape linear density	16000 Tex

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.

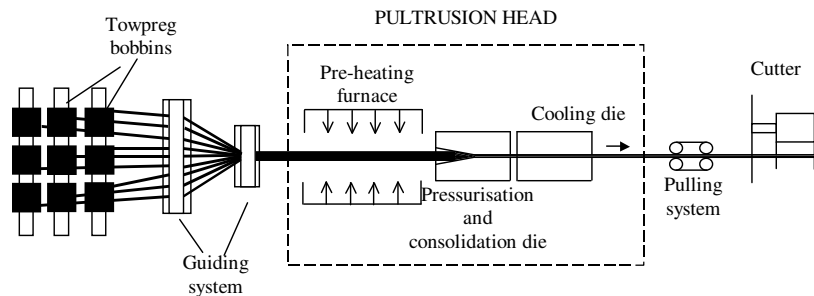


Figure 3. Schematic diagram of the proprietary pultrusion line.



Figure 4. Overview of pultrusion equipment.

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.

Raw-material	Heated die temperature (°C)	Cooled die temperature (°C)	Pre-heating temperature (°C)	Pulling speed (m/min)
Twintex®	300	50	170	0.2
Towpreg	275	25		
PCT	230	50		

Table 3. Pultrusion processing parameters.

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.

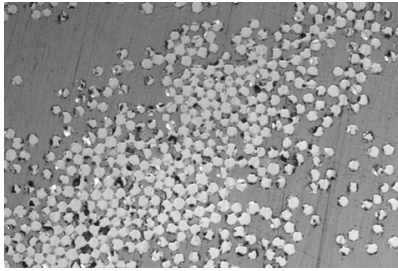
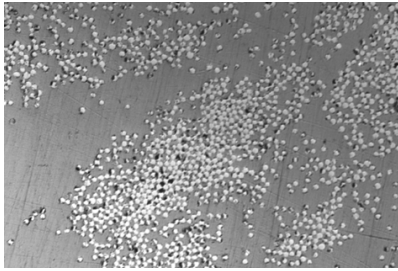
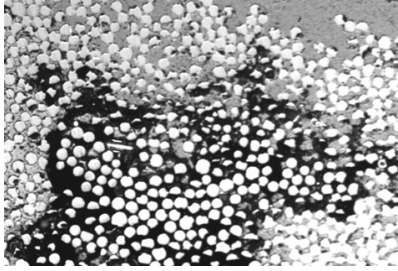
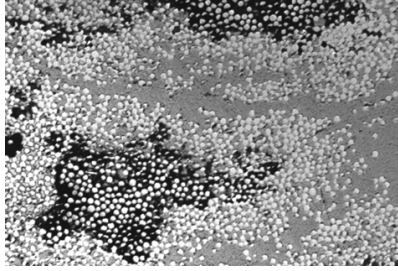
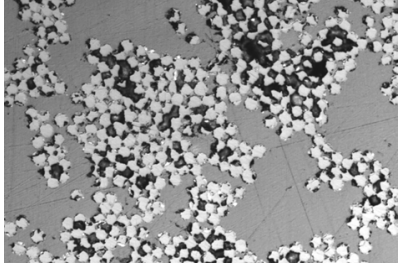
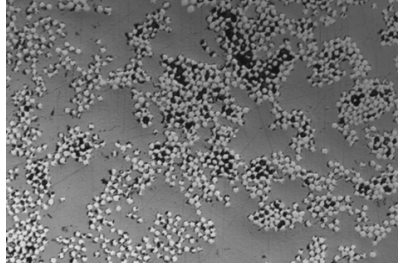
Material	Magnification	
	250 ×	130 ×
PCT		
Towpreg		
Twintex [®]		

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 on 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 show typical force-displacement curves of the tested specimens in the short beam shear test.

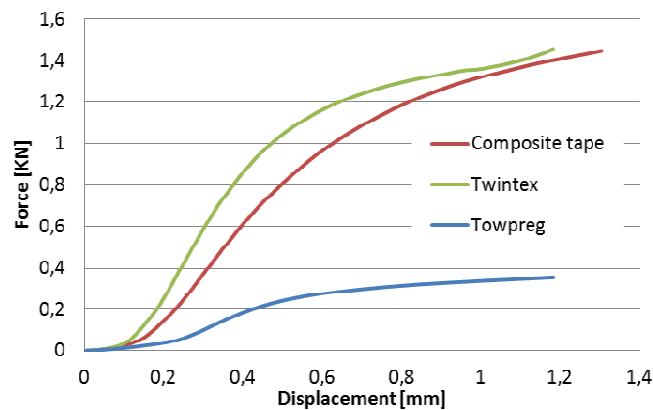


Figure 5. Short beam test results.

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 former, specimens weighting approximately 20 g, cut from the profiles, were calcinated during 10 min at 600° C inside a Nabertherm[®] S30 muffle furnace. For the second determination, rectangular, 100 mm×20 mm×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×15 mm×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.

Raw-material	Tensile strength (MPa) Av.	Tensile modulus (GPa)		Flexural strength (MPa)		Flexural modulus (GPa)		Fibre mass fraction (%) Av.	Fibre volume fraction (%) Av.
		Av.	SD	Av.	SD	Av.	SD		
Twintex®	>416	24.9	1.1	595	24	26.2	2.0	62.5	37.1
Towpreg	>305	29.9	3.5	125	11	27.1	1.2	78.8	56.8
PCT	>424	21.4	1.5	329	30	16.8	1.5	54.8	30.0

Av. – Average

SD – Standard Deviation

Table 5. Results from tensile and flexural tests.

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.

Acknowledgments

Authors would like to acknowledge the experimental work done by Ms. Anna Matveeva.

References

- [1] 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).
- [2] 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).
- [3] Silva, J. F., Nunes, J. P., Bernardo, C. A., and Marques, A.T. 'Thermoplastic Matrix Composites from Towpregs'. Advances in composite materials – analysis of natural and man-made materials, INTECH, ISBN 978-953-307-449-8, Rijeka, Croatia, pp. 307-324, (2011).
- [4] Guneri Arcovali, Handbook of composite fabrication, RAPRA Technology Ltd, U. K. (2001).
- [5] 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).
- [6] Miller, A. H., Dodds, N., Hale, J. M. and Gibson, A. G. High Speed Pultrusion of Thermoplastic Matrix Composites, Composites Part A, 29 A, pp. 773-782 (1998).

- [7] Bechtold G., Wiedmer S., Friedrich K. Pultrusion of Thermoplastic Composites – New Developments and Modelling Studies, in *Journal of Thermoplastic Composite Materials*, Vol. 15, pp. 443-465 (2002).
- [8] Nunes, J. P., van Hattum, F.W. J., Bernardo, C. A., Brito, A. M., Pouzada, A. S., Silva, J. F. and 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).
- [9] van Rijswijk, K. Thermoplastic Composite Wind Turbine Blades, PhD Thesis, TU Delft, Delft, Netherlands (2007).
- [10] Stewart, R. Thermoplastic Composites – Recyclability and Fast Processing Top List of Benefits, *Reinforced Plastics*, 55 (3), pp. 22-28, May/June (2011).
- [11] www.ocvreinforcements.com/solutions/Twintex.aspx, May (2012).
- [12] Ye, L., Friedrich, K., and Savadory, A. Manufacture of CF/PEEK Composites from Powder/Sheath Fibre Preforms, *Composites Manufacturing*, 5 (1), pp. 41-50 (1994)
- [13] Bechtold G., Kameo, K., Langler, F, Hamada, H and Friedrich K. Pultrusion of Braided Thermoplastic Commingled Yarn – Simulation of the Impregnation Process, 5th Int. Conf. On Flow Processes in Composite Materials, U. K., July (1999).
- [14] Alexis B. Processing and Benefits of Commingled Glass Fiber Reinforced Thermoplastic Composites. ANTEC'01, Dallas, TX, USA, May (2001).
- [15] 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).
- [16] 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).
- [17] Ramani, K., Borgaonkar, H. and Hoyle, C. Experiments on Compression Moulding and Pultrusion of Thermoplastic Power Impregnated Towpregs, *Composites Manufacturing*, Vol 6, pp. 35-43 (1995).
- [18] Astrom, B. T. and Pipes, R. B. 'Thermoplastic Filament Winding with On-line Impregnation'. *Journal of Thermoplastic Composite Materials*, Vol. 3, pp. 314-324 (1990).
- [19] Kim, D.-H., Lee, W. And Friedrich, K. A Model for a Thermoplastic Pultrusion Process using Commingled Yarns, *Composite Science and Technology*, Vol.61, pp. 1065-1077 (2001).
- [20] Wiedmer, S., Manolesos, M. An Experimental Study of the Pultrusion of Carbon Fiber-Polyamide 12 Yarn, *Journal of Thermoplastic Composite Materials*, Vol.19, pp. 97-112 (2006).
- [21] 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).
- [22] 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).
- [23] 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).
- [24] Nunes, J. P. and Silva, J. S. Production of Thermoplastic Matrix Towpregs for Highly Demanding and Cost-Effective Commercial Applications, VI International Materials Symposium - MATERIAIS 2011, Guimaraes, Portugal, April (2011).
- [25] 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).
- [26] Nunes, J. P., Silva, J. F., Novo, P. J. Processing Thermoplastic Matrix Towpregs by Pultrusion, *Advanced in Polymer Tech.*, in press (2012).
- [27] 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).
- [28] Website: www.compositetape.com, May (2012).