

## PULTRUSION OF FIBRE REINFORCED THERMOPLASTIC PRE-IMPREGNATED MATERIALS

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### Abstract

Fibre pre-impregnated thermoplastic materials produced by different methods and processing conditions were used to produce composites using pultrusion and compression moulding. The processing windows used to produce these materials and composite profiles were optimised by using method of Taguchi/ DOE (Design of Experiments). **Composites were manufactured by pultrusion and compressing moulding** and subsequently submitted to mechanical testing and microscopy analysis. The obtained results were compared with the expected theoretical ones predicted from the rule of mixtures and with those of similar conventional available materials. The results obtained shown that produced composites have adequate properties for common and structural engineering markets.

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## 1 Introduction

1 Although only recently thermoplastic matrices have been used in long and continuous fibre reinforced  
2 composites replacing **1**ith success thermosetting matrices, the number of their applications is increasing  
3 due to their better ecological and mechanical performance. Composites with thermoplastic matrices **2**fers  
4 increased fracture toughness, higher damage tolerance, short processing cycle times and excellent  
5 environmental stability. They are recyclable, post-formable and can be joined by welding. The use of  
6 long/continuous fibre reinforced thermoplastic matrix composites involves, however, great technological  
7 and scientific challenges since thermoplastics present much higher viscosity than thermosettings, which  
8 makes much difficult and complex the impregnation of reinforcements and consolidation tasks [1-6].  
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10 Today, two major technologies are being used to allow wet reinforcing fibres with thermoplastic  
11 polymers [1, 4-6]: i) the direct melting of the polymer and, ii) the intimate fibre/matrix contact prior to  
12 final composite fabrication. Continuous fibre reinforced thermoplastic matrix pre-impregnated tapes  
13 (PCT's) are, for example, produced by direct melting processes. Alternatively, intimate contact processes  
14 allow producing cheap and promising pre-impregnated materials, such as, commingled fibres and powder  
15 coated towpregs. Sometimes, thermoplastic compatibilizers are added to the matrices to improve their  
16 adhesion and facilitate impregnation to reinforcements [7].  
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18 This work studies and compares the processability of final composite parts by using three different  
19 pre-impregnated materials **3**roduced by each one of the above mentioned wetting techniques. All studied  
20 pre-impregnated materials were based on a continuous fibres (carbon and glass) reinforced polypropylene  
21 matrix (CF/PP and GF/PP) system. **4**ne is a pre-consolidate, **5**pe (Fig. 1 a)) that was produced by the  
22 melting process (cross-head extrusion) [8]. From the other two produced by fibre/matrix intimate contact  
23 methods, one is a commercially available commingled fibres product (Fig. 1 b) and the other a towpreg  
24 (Fig. 1 c)) produced by our own developed dry coating line [9]. **6**ltrusion and compression moulding  
25 were the selected manufacturing methods for processing all these pre-impregnated materials into  
26 composite parts.  
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28 The optimization of both processes was made by studying the influence of the most relevant  
29 processing parameters in the final properties of the produced pre-impregnated materials and composites.  
30 The method of Taguchi / DOE (Design of Experiments) was used to achieve this aim.  
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32 The possibility of using maleic anhydride as compatibilizer of carbon and glass fibre reinforced  
33 polypropylene composites (CF/PP and GF/PP) was also analysed in the present work.  
34

35 Towpregs were characterized by scanning electron microscopy (SEM), visual analysis and their  
36 polymer mass contents were determined. The final composite parts were also submitted to tensile,  
37 interlaminar and flexural tests, as well as calcination and optical microscopy tests and the results were  
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compared with theoretical ones that can be predicted by using the ROM (Rule Of Mixtures) and <sup>1</sup>her traditional materials.

## 2 Experimental

### 2.1 Raw Materials

The following raw materials were used to produce CF/PP pre-impregnated materials for this work: i) a PP powder ICORENE 9184B P<sup>®</sup> and carbon fibre roving M30 SC<sup>®</sup> from the ICO Polymers and TORAY, respectively, were used to produce the CF/PP towpregs, ii) PP powder Moplen RP348U<sup>®</sup> from Basell and the carbon fibre roving already mentioned were used to manufacture the CF/PP PCT tapes.

For the GF/PP towpregs, a 2400 Tex type E fibre rovings from Owens Corning and Icorene<sup>®</sup> 9184B P polypropylene from ICO Polymers France were used. Also, the GF/PP PCT tapes were manufactured with glass fibres (TufRov 4599) from PPG Industries and a polypropylene matrix (Moplen RP348U) from Basell.

Commercial commingled GF/PP fibres TWINTEX<sup>®</sup> R PP 60 B 1870 FU from Owens Corning were also used in the production of pultrusion thermoplastic composite profiles, as reference of a current commercially available pre-impregnated product.

Some batches of CF/PP and GF/PP towpregs were also produced using PP powder (ICORENE 9184B P<sup>®</sup>) blended with 1% in mass content of maleic anhydride S 47 29608 707<sup>®</sup> from Merck Schuchardt OHG, in order to assess the possible enhancement of fibre/matrix adhesion [10-14].

Tables 1 and 2 summarise relevant properties of the polypropylene, glass and carbon fibres used in present work to produce pre-impregnated raw materials (towpregs and PCT's). Table 3 shows the manufacturer datasheets properties of TWINTEX<sup>®</sup>.

### 2.2 Production of Thermoplastic Matrix Pre-Impregnated Products

The dry powder coating equipment used to produce fibre reinforced towpregs is schematically depicted in Figure 2 [9, 15].

The pre-consolidated tapes (PCT's) used in this work were produced in a cross-head extrusion equipment (see Fig.3) from our own laboratories [8]. Using this equipment, it was possible to produce the tapes (PCT's) pre-impregnated raw-materials. The overview of their main properties is given in Table 4.

#### 2.2.1. CF/PP towpregs production and optimization

In order to optimize the production of CF/PP powder coated towpregs, different processing variables combinations were experimented and the number of trials optimized using the Taguchi approach. The studied operational parameters were:

- heating oven temperature (600, 650 and 700 °C); consolidation oven temperature (350, 400 and 450 °C); linear pull speed (4, 6 and 8 m/min).



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The Taguchi approach was applied to the towpregs production process in order to obtain the condition that maximizes polymer powder content.

The polymer mass fraction in the towpregs,  $\omega_p$ , was determined by weighting towpreg strips produced in those different conditions, using equation 1:

$$\omega_p = \frac{W_t - W_f}{W_t} \quad (1)$$

where  $W_t$  and  $W_f$  are the measured unit length weights of the towpreg strip and fibre roving, respectively.

Table 5 shows the used processing conditions and obtained results, according to the established design of experiments. The average polymer mass content in towpregs, established by the design of experiences was 34.5% (Table 5).

The **1**ains and **2**ects of the processing variables on the results can be seen from Figure 4.

The optimal condition obtained from Taguchi method application led to the following operating parameters selection: heating oven temperature and consolidation oven temperatures of 700 °C and 400°C respectively, and a linear pulling speed of 4 m/min. Using this optimal operative condition, the amount of polymer should increase up to 45.6%. However, the operative condition that was chosen as optimal had a line pull speed of 6 m/min allowing a high rate of production, lower processing problems and sufficiently levels of polymer content **3**%, enough for the of use of towpregs in the pultrusion process). Also, the addition of 1% of maleic anhydride to the PP polymer had no influence on the towpreg polymer mass fraction. Table 6 summarizes the determined towpreg properties obtained with the combination of **4**ptimal parameters.

### 2.2.2 GF/PP towpregs production and optimization

To try maximizing the polymer powder content in the towpregs the following processing conditions were varied within the next ranges: i) convective oven temperature (°C): 650 - 700; ii) Consolidation furnace temperature (°C): 350 – 450; iii) Coating line pulling speed (m/min): 4 – 6.

From the polymer mass fractions obtained in produced towpreg strips it was possible to establish as optimal the following operating parameters: convective and consolidation oven temperatures of 700 °C and 400°C respectively, and a linear pulling speed of 6 m/min. In such operational conditions the GF/PP towpregs were continuously produced with polymer mass content of 30.7 %.

Unexpectedly, very low polymer mass fractions ( ~10 % - 16 %) were obtained when PP powder with 1% of maleic anhydride was used to produce GF/PP towpregs. Hence, the idea of using this additive to improve adhesion of the PP matrix to the glass fibres was abandoned. Table 6 summarizes the determined towpreg properties obtained with the combination of selected parameters.

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## 2.3 Pultrusion of pre-impregnated materials

The towpregs, PCT's and commingled fibres were processed into composite **1** profiles using the laboratorial pultrusion line, Figure 5 [10, 18].

To produce 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 heated die to be heated and consolidated to the required size and, after cooled down in the cooling die to solidify. **2** the die temperature distribution profile in order to obtain enhanced final properties can be seen in Figure 6.

In this work, it was designed and manufactured a die to allow producing a 20×2 mm<sup>2</sup> bar-shaped profile.

### 2.3.1 Towpreg processing

CF/PP towpregs were manufactured by pultrusion into composite bar profiles using the most relevant operating conditions. The Taguchi's/DOE method was applied, maintaining the cooling die at 25 °C, in order to optimize the processing parameters:

i) furnace temperature (160 or 180 °C); ii) heating die temperature (240 or 260 °C); iii) linear pull-speed (0.2 or 0.3 m/min).

**3** results have shown that was not possible to produce, in steady, conditions pultruded profiles from towpregs at pultrusion speeds and consolidation die temperatures higher than 0.4 m/min and 260 °C, respectively. By using higher values of these two parameters, the process became unsteady, mainly due to reflux and accumulation of the thermoplastic polymer at the entrances of the consolidation and cooling dies.

Table 7 summarizes the flexural test results, as described in **4** next paragraph 2.5.2, obtained with the studied processing conditions according to established in the design of experiments. The variation of the flexural modulus and strength with the selected processing parameters can be seen in Figures 7 and 8.

The optimal condition concerning flexural stiffness maximization obtained from Taguchi method application led to the following operating parameters selection: furnace and heated die oven temperatures of 160 °C and 260°C respectively, and a linear pulling speed of 0,2 m/min. For optimizing the flexural strength the obtained **5** parameters combination was: furnace and heated die oven temperatures of 160 °C and 240°C respectively, and a linear pulling speed of 0.3 m/min.

It is possible to observe that the furnace temperature of 160°C lead to the better results. That could be explained by the lower polymer reflux on the entrance of the heated die. The optimal operating conditions to maximize both flexural properties were: furnace and heated die oven temperatures of 160 °C and 260°C respectively, and a linear pulling speed of 0.2 m/min.

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How did the authors determine the heating die temperatures in Figure 6? Using a thermal camera? Please clarify this **Text clarified (Highlighted in the attached file).**

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1nally, towpregs with additive were also pultruded into composite bars using the condition that optimizes both flexural properties and two more conditions (Table 8), being the results from flexural tests using towpreg pultruded bars with and without maleic anhydride additive shown in Table 9. The use of such additive had no significant influence on the flexural properties.

GF/PP pre-impregnated towpreg rovings were processed by pultrusion into rectangular 20×2 (mm<sup>2</sup>) profiles. To determine the best processing window, the main processing conditions were varied, maintaining the cooling die at 25 °C: i) pre-heating temperature (°C): 170 – 180; ii) pressurization/consolidation die temperature (°C): 240 – 300; iii) linear pultrusion speed (m/min): 0.2 - 0.4.

Results have shown 2at was not possible to produce, in steady conditions, profiles from towpregs at pultrusion speeds and pressurization/consolidation die temperatures higher than 0.3 m/min and 280 °C, respectively. By using higher 3values the process became unsteady as it was already found for CF/PP towpregs processing. 4problems also occurred for temperatures below 270° C in the pressurization/consolidation die. By maintaining constant the temperatures in the cooling and pressurization/consolidation dies at 25 °C and 280 °C, respectively, profiles pultruded in different processing conditions were submitted to the flexural tests (see 2.5.2). 5s may be seen (Table 10), very similar values of flexural moduli and strengths were found by using pre-heating furnace temperatures and linear pultrusion pulling speeds in the ranges of 170 - 180 (°C) and 0.2 - 0.3 m/min, respectively. While the slower pultrusion pulling speed of 0.2 m/min seemed to generate profiles with higher absolute values, this was not confirmed by the flexural properties divided by the determined fiber volume fraction depicted in two last the columns of Table 10. On the other hand, higher flexural strength values, both in 6bsolute and relative terms, were obtained at the higher temperature of 180 °C in pre-heating furnace. 7ence, it was concluded to use as optimal pultrusion operating window the following one: i) pre-heating temperature (°C): 170 – 180; ii) pressurization/consolidation die temperature (°C): 280; iii) cooling die temperature (°C): 25; iv) linear pultrusion speed (m/min): 0.2 - 0.3.

### 2.3.2 Pre-consolidate tapes(PCT's) and Twintex® processing

PCT's and Twintex® were processed into rectangular 20×2 (mm<sup>2</sup>) bar using the already mentioned pultrusion equipment being operating conditions shown in Table 11.

## 2.4 8eated Compression Moulding

Three different GF/PP fibre reinforced pre-impregnated products were studied and also processed into rectangular 180 × 180 × 2 (mm<sup>3</sup>) plates. All 9e-impregnates were cut, weighted and introduced in a

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Why heated compression molding is used? This section (Section 2.4) is confusing with the rest of the manuscript which is focusing with the pultrusion process.

Which material is produced using which manufacturing technique. This is not clear in the manuscript:

CF/PP and GF/PP are both manufacturing using pultrusion. Waht is the difference for compression moulding. Please make the manuscript easy to understand and follow.

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1 cavity placed between the heated plates of a 400 kN press. 1 After a 10 min delay, the press was closed up  
2 to a compression force of 200 kN. One minute after that, the press plates were cooled down maintaining  
3 constant the press closing force. At 30 °C, the press plates were opened and the final composite plate  
4 removed from the mould. The unidirectional fibre reinforced composite plates were produced with the  
5 following processing conditions: 2 press platen temperature (°C): 250; ii) pre-heating time (min): 10; iii)  
6 press closing force (kN): 200; iv) delay at maximum closing force (min): 1.0; v) opening platen  
7 temperature (°C): 30.  
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## 11 2.5 Testing

### 12 2.5.1 Microscopy analysis

13 CF/PP and GF/PP towpreg 3 mples were characterized by scanning electron microscopy (SEM) to  
14 evaluate the adhesion of the polymer powder to the fibres and its distribution. Those samples were coated  
15 with a layer of 4 lld with approximately 6.5 µm thick, before the observation.  
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21 To determine the impregnation quality and to evaluate the fibre distribution and fibre/matrix adhesion  
22 of the thermoplastic composites, their cross-sections were studied under optical microscopy. Observations  
23 were done using reflected light optical microscopy (Olympus BH-2). A digital camera (Leica DFC200)  
24 was used to get the image cross sectional views of the samples.  
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### 30 2.5.2 Mechanical testing

31 Bar samples were submitted to flexural, tensile and interlaminar testing according to the ISO  
32 standards 14125, 527 and 14130, respectively. The mechanical properties obtained were compared to the  
33 theoretical ones predicted by using the Rule of Mixtures (ROM).  
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36 Tensile tests were conducted, according to ISO 527, in a 100 kN universal testing machine at the  
37 crosshead speed of 2 mm/min using 180×20×2 mm<sup>3</sup> rectangular samples. The tensile modulus was  
38 determined from the slope of the initial linear portion of the experimental stress/strain curve. A SG  
39 Shimadzu<sup>®</sup> 50 mm length strain-gauge was used up to 0.3% strain, for accurate determination of the  
40 tensile modulus. Regarding the determination of tensile strength, it was not possible to proceed with the  
41 test until specimen failure due to grip slippage. Hence, new specimen geometry was designed and tested  
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50 5 with good results (Figure 9).  
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52 Three-point flexural tests were also conducted on five 100 × 20 × 2 (mm<sup>3</sup>) composite specimens,  
53 using 100 kN universal testing machine and a distance between supports of 80 mm, according to ISO  
54 14125, at a crosshead speed of 1 mm/min.  
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57 Samples with dimensions of 20 × 20 × 2 (mm<sup>3</sup>), cut from composites processed from each pre-  
58 impregnated raw material, were submitted to interlaminar shear tests according to ISO 14130. The tests  
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What does good results mean? How did the authors justify the "good" result?

Explained in the text. (Highlighted in the attached file).

were conducted in a 50 kN universal testing machine by using an initial pre-load of 1 N at the crosshead speed of 1 mm/min and a 10 mm span between supports.

### 2.5.3 Calcination testing

Carbon and glass fibre composites mass content was determined by using calcination tests according to the EN ISO 1172. Composite samples, weighting approximately 2 g, were submitted to calcination inside a crucible in a muffle furnace during 10 min at 620° C.

## 3 Results and Discussion

Figure 10<sup>1</sup> SEM micrographs of CF/PP and GF/PP towpregs samples. A good degree of adhesion between both carbon and glass fibres and the polymer powder particles was obtained. Also, a reasonable polymer powder distribution on the fibres was achieved at the optimised operating conditions.

The cross-sections of the pultruded composites were studied under optical Microscopy. As can be seen from Figure 11, all CF/PP composite profiles from towpregs (with and without additive) and PCT's have a reasonable distribution of the reinforcing fibres over the cross-sections. However, large differences in impregnation quality occur between the different samples that are likely to be related, directly, to the impregnation state of pre-impregnated materials used in pultrusion. It may be seen that the impregnation quality of the PCT composite samples is good, presenting almost all fibres completely surrounded ('wet-out') by the polymer. Only a few large dry spots were observed. This is most likely due to the good degree of impregnation already achieved in the PCT raw-material tape prior to the pultrusion step.

<sup>2</sup>he samples from CF/PP towpreg with additive show a higher quantity of dry zones than the ones without additive.

As Figure 12 shows, it was still possible to distinguish discontinuities on the cross section of pultruded composites from GF/PP towpregs, <sup>3</sup>here it may be seen zones very rich in polymer contrasting with others with much higher quantity of fibres.

The microscopy images taken from the samples of the pultruded composites using GF/PP PCT, TWINTEX<sup>®</sup> and GF/PP towpreg, are given in Table 12. <sup>4</sup>l semi-finished products lead to a reasonable distribution of the reinforcing fibres over the cross-sections, <sup>5</sup>though small improvements in fibre distribution can be observed going from PCT through TWINTEX<sup>®</sup> to towpreg composites.

It may be seen that the impregnation quality of the PCT composite samples is excellent, <sup>6</sup>esenting almost all fibres completely surrounded ('wet-out') by the polymer, hardly showing any dry spots in the pultruded samples.

The TWINTEX<sup>®</sup> -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 <sup>7</sup>hen melting the polymer very small, which leads

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please make this sentence clearer. It is difficult to understand its meaning.  
**Clarified in figure 12.**
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What does it mean? Isn't the samples pultruded completely?  
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to an easier consolidation. However, [1]me 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. It 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.

Tables 13 and 14 summarize all experimentally results obtained from the CF/PP and GF/PP unidirectional composites processed by pultrusion and compression moulding, respectively, from the pre-impregnated products under study. To better evaluate and compare the mechanical properties obtained on the composites processed from the different pre-impregnated products studied, Tables 13 and 14 also present theoretical expected values and relative values of specific properties.

The theoretical values of moduli,  $E$ , [2]as directly obtained from the rule of mixtures using the raw-material properties presented in Tables 1, 2 and 3, following the Eq. 2:

$$E = E_f \cdot v_f + E_p \cdot (1 - v_f) \quad (2)$$

where,  $E_f$ ,  $E_p$  and  $v_p$  are the fibre modulus, polymer modulus and fibre volume fraction, respectively.

As can be seen from Tables 13 and 14, the experimental moduli obtained from de CF/PP and GF/PP composites are in good agreement with the predicted theoretical ones. Some experimental values are even higher than the theoretical expected ones. This can be explained considering that the volume fraction content of some samples can be higher than the determined by the calcination tests.

Using the proposed new geometry (Figure 9) for the tensile test specimens, it was possible to reach [3]eaking loads and therefore determine their tensile strengths.

Analysing [4]ble 13, one can conclude that composites processed from the CF/PP PCT's demonstrated [5]have better flexural and interlaminar shear strengths than those produced from CF/PP towpregs. From Table 14, it is possible to conclude that commingled fibres (TWINTEX®) presented, in general, better properties and had also shown to be more adjusted to commercial application demands and to be easily processed into final composites by the currently used manufacturing methods, probably [6]cause their easy consolidation.

In any case, worse flexural strength and modulus results were found in GF/PP towpreg and PCT pultruded composites, respectively. These [7]wer results obtained in the flexural tests are probably consequence of the inferior degree of impregnation observed in the towpreg based composites and, in the [8]se of PCT tape based composites, result from the higher rich polymer regions exhibited by this material in its outside layers.

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In flexure, if in the sample thickness the top and bottom layers were rich in polymer (lack of fibres), the flexure stiffness and strength will decrease significantly comparatively to samples where the impregnated fibres show a good distribution through the thickness

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Nevertheless, the GF/PP pre-impregnated products produced in **1**ur laboratories (towpregs and PCTs) have already demonstrated very good mechanical behaviour, namely, in terms of stiffness. In fact, the composites manufactured from these products presented experimental moduli values very closed to the theoretical expected ones. **2**hile composites processed from the PCTs demonstrated to have better mechanical strength, those produced from towpregs presented higher moduli. As mechanical strength values are more affected by small defects than those from moduli, the composites manufactured from PCTs seem to profit from the pre-consolidate state already presented by this product before final processing. Finally, it may be noted that any of composites made from pre-impregnated materials **3**nder study reached failure **4** the interlaminar shear tests. This fact reveals the high degree of ductility exhibited by these materials which may be relevant for many applications. Thus, the interlaminar shear strength results shown in Tables 13 and 14 correspond to maximum force applied in the test.

Figure 13 **5**ow typical force-displacement curves of the GF/PP composites tested specimens in the short beam shear test, from towpregs, PCT's and TWINTEX<sup>®</sup>. Those curves show that all specimens failed in plastic shear. 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<sup>®</sup> pultruded composites have similar force-displacement behaviour, towpreg based samples showed lower performance. This **6**ostly results from the already mentioned limited degree of impregnation of these samples.

The flexural properties of composite materials under study were compared with those of common use and technical materials, in particular metals, polymers and polymer matrix composites and as **7**u can see in Table 15 [17-22] the results of produced composites bars are higher than traditional materials, revealing a possible growing interest in its application.

#### 4 Conclusions

The tests made using a proprietary pultrusion equipment already allow to conclude that is possible to produce, in **8**ood conditions, profiles from almost all available thermoplastic matrix pre-impregnated raw materials using pull speeds of 0.3 m/min. Currently, work is carried out to increase the pultrusion processing speed to values in the range from 2 to 6 m/min. This will equalize 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) **9**ill make possible, in future, **10**embling them in one.

Existing powder-coating equipment was shown to be suitable to produce CF/PP and GF/PP towpregs that could be adequately processed into pultruded profiles. From the tests made, the towpregs can be easily and continuously produced at industrial production speeds between 2 a 6 m/min.

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The authors could show their powder coating line as well as their pultrusion line in the manuscript.  
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It is also worth mentioning that it was possible to produce GF/PP plates by compression moulding from pre-impregnated materials with similar and sometimes **1** better properties than those obtained from pultruded profiles.

It was possible to optimize the production of CF/PP pultruded profiles and CF/PP towpregs, through the use of Taguchi/DOE method, achieving optimal conditions. The addition of the compatibilizing agent (1% maleic anhydride) did not improve the polymer mass content in towpregs neither the mechanical properties on the final composites.

Three different commercial promising glass fibre reinforced thermoplastic matrix pre-impregnated materials were easily processed by pultrusion and compression moulding: a commercial available GF/PP commingled fibres product and also GF/PP towpregs and tapes manufactured in our own laboratories.

A process window was established for the production of PCT's and towpregs and for the processing of towpregs, PCT's and commingled fibres.

The mechanical properties of the composites processed from all those **2** free GF/PP pre-impregnated were determined and evaluated. All of them demonstrated to have mechanical properties compatible with the requirements of the major current structural engineering applications. In general, commingled fibres TWINTEX<sup>®</sup> presented **3** light better mechanical properties and have shown to be more **4** justified for composite processing than the other pre-impregnated products.

In particular, very good agreement was found between the experimental moduli values of all composites produced and the theoretical ones.

More research must be done in order to increase the processing speeds of CF/PP and GF/PP towpregs as well as PCT's and to improve the impregnation, uniformity and dispersion of raw-materials in the composites.

The mechanical properties obtained in pultruded and **5** impressed moulded composites allow predicting their adequate use either in general or structural engineering applications.

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There is no mechanical property information for compression molded composites?

According to the suggestion, compression molding was removed from the final paper text.

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As suggested by the reviewer, three additional relevant papers concerning the pultrusion process were added.

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**Tables**

Table 1. Properties of Towpregs and PCT PP raw-materials

| Property                                       | PP powder<br>(ICORENE 9184B P <sup>®</sup> ) |                   | PP granules<br>(Moplen RP348U <sup>®</sup> ) |
|--|--|-------------------|--|
|  | Manufacturer datasheet                       | Experimental      | Manufacturer datasheet                       |
| Specific gravity (Mg/m <sup>3</sup> )          | 0.91   | 0.91              | 0.90   |
| Tensile strength (MPa)                         | Yield Strength 30                            | Yield Strength 19 | Yield Strength 30                            |
| Young Modulus (GPa)                            | 1.3  | 0.98              | 1.1  |
| Poisson's ratio                                | -  | 0.21              | -  |
| Average powder particle size (µm)              | 440  | 163               | -  |
| Glass transition temperature (T <sub>g</sub> ) | Typical value 0-20                           | -                 | Typical value 0-20                           |
| Melting temperature (T <sub>m</sub> )          | Typical value 170                            | 166               | Typical value 170                            |

Table 2. Properties of Towpregs and PCT fibres raw-materials

| Property                              | Glass fibre<br>(305E-TYPE 30 <sup>®</sup> ) |              |                        | Carbon fibre<br>(TORAY M30 SC <sup>®</sup> ) |              |
|---------------------------------------|---|--------------|------------------------|--|--------------|
|                                       | Manufacturer datasheet                      | Experimental | Manufacturer datasheet | Manufacturer datasheet                       | Experimental |
| Linear density (Tex)                  | 2400  | -            | 2400                   | 760  | -            |
| Specific gravity (Mg/m <sup>3</sup> ) | 2,65  | -            | 2,54-2,6               | 1,73   | -            |
| Tensile strength (MPa)                | 3500  | 1657         | 1900-2400              | 5490   | 2731         |
| Young Modulus (GPa)                   | 76  | 62.5         | 69-76                  | 294  | 194.5        |
| Average fibre diameter                | 17  | 13.7         | 17                     | 5  | 7.37         |

Table 3. TWINTEX<sup>®</sup> R PP 60 B 1870 FU from Owens Corning

| Property               | Values |
|------------------------|--------|
| Linear density (Tex)   | 1870   |
| Tensile strength (MPa) | 760    |
| Young Modulus (GPa)    | 29.5   |
| Fibre mass content (%) | 60     |

Table 4. Overview of the main properties of the produced pre-consolidated tapes (PCT's)

| Property            | CF/PP              | GF/PP              |
|---------------------|--------------------|--------------------|
| Fibre type          | Carbon, 760 Tex    | E-Glass, 2400 Tex  |
| Filament diameter   | 7 µm               | 17 µm              |
| Fibre content       | 45 wt. %           | 60 wt. %           |
| Matrix type         | Polypropylene (PP) | Polypropylene (PP) |
| Tape width          | 25 mm              | 25 mm              |
| Tape linear density | 14000 Tex          | 16000 Tex          |

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Table 5. Taguchi approach applied to towpregs manufacturing process

| Experiments | Processing variables          |  |                              | Results                   |
|-------------|-------------------------------|--|------------------------------|---------------------------|
|             | Heating oven temperature (°C) | Consolidation furnace temperature (°C) | Linear pulling speed (m/min) | Polymer mass fraction (%) |
| 1           | 600                           | 350                                    | 4                            | 32.2                      |
| 2           | 600                           | 400                                    | 6                            | 31.4                      |
| 3           | 600                           | 420                                    | 8                            | 20.6                      |
| 4           | 650                           | 350                                    | 8                            | 27.9                      |
| 5           | 650                           | 400                                    | 4                            | 39.9                      |
| 6           | 650                           | 420                                    | 6                            | 40.7                      |
| 7           | 700                           | 350                                    | 6                            | 35.6                      |
| 8           | 700                           | 400                                    | 8                            | 40.6                      |
| 9           | 700                           | 420                                    | 4                            | 40.4                      |
| Average     |                               |  |                              | 34.5                      |

Table 6. Overview of the main properties of the produced Towpregs

| Property            | CF/PP              | GF/PP              |
|---------------------|--------------------|--------------------|
| Fibre type          | Carbon, 760 Tex    | E-Glass, 2400 Tex  |
| Filament diameter   | 7 µm               | 17 µm              |
| Fibre content       | 64,5 wt.%          | 30,7 wt.%          |
| Matrix type         | Polypropylene (PP) | Polypropylene (PP) |
| Tape width          | 5 mm               | 5 mm               |
| Tape linear density | 1170 Tex           | 3465 Tex           |

Table 7. Taguchi approach applied to towpreg processing by pultrusion

| Experiments | Processing variables     |                              |                              | Bending properties     |                         |
|-------------|--------------------------|------------------------------|------------------------------|------------------------|-------------------------|
|             | Furnace temperature (°C) | Heating die temperature (°C) | Linear pulling speed (m/min) | Flexural modulus (GPa) | Flexural strength (MPa) |
| 1           | 160                      | 240                          | 0.2                          | 86.7 ± 1.3             | 229.0 ± 7.3             |
| 2           | 180                      | 240                          | 0.2                          | 79.5 ± 2.0             | 212.4 ± 12.6            |
| 3           | 160                      | 260                          | 0.2                          | 91.0 ± 0.4             | 241.2 ± 1.6             |
| 4           | 180                      | 260                          | 0.2                          | 85.1 ± 1.7             | 218.2 ± 9.1             |
| 5           | 160                      | 240                          | 0.3                          | 82.1 ± 2.8             | 241.7 ± 13.1            |
| 6           | 180                      | 240                          | 0.3                          | 87.5 ± 1.9             | 239.6 ± 13.3            |
| 7           | 160                      | 260                          | 0.3                          | 85.0 ± 4.4             | 234.5 ± 11.5            |
| 8           | 180                      | 260                          | 0.3                          | 83.7 ± 2.8             | 221.3 ± 7.1             |

Table 8. Taguchi approach applied to towpregs with additive processed

| Experiments | Processing variables     |                              |                              | Flexural properties    |                         |
|-------------|--------------------------|------------------------------|------------------------------|------------------------|-------------------------|
|             | Furnace temperature (°C) | Heating die temperature (°C) | Linear pulling speed (m/min) | Flexural modulus (GPa) | Flexural strength (MPa) |
| 1           | 160                      | 260                          | 0.2                          | 89.0 ± 7.3             | 87.6 ± 1.3              |
| 2           | 160                      | 240                          | 0.2                          | 81.7 ± 7.8             | 70.4 ± 2.8              |
| 3           | 160                      | 240                          | 0.3                          | 87.4 ± 11.8            | 80.5 ± 2.6              |

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Table 9. Flexural test results on composite bars with and without additive

| Processing parameters        |     | Flexural modulus (GPa) |               | Flexural strength (MPa) Corrected |               |
|------------------------------|-----|------------------------|---------------|-----------------------------------|---------------|
|                              |     | Without                | With additive | Without additive                  | With additive |
| Furnace temperature (°C)     | 160 |                        |               |                                   |               |
| Heating die temperature (°C) | 260 | 90.1 ± 0.4             | 87.6 ± 1.3    | 241.2 ± 1.6                       | 229.0 ± 7.3   |
| Linear pulling speed (m/min) | 0.2 |                        |               |                                   |               |

Table 10. Influence of pultrusion conditions on the flexural properties of profiles made from GF/PP towpregs

| Pultrusion conditions        |                          | Flexural properties |                | Fibre content |            | Flexural properties / fibre volume fraction |                                   |
|------------------------------|--------------------------|---------------------|----------------|---------------|------------|---|-----------------------------------|
| Pre-heating temperature (°C) | Pultrusion speed (m/min) | Modulus (GPa)       | Strength (MPa) | Mass (%)      | Volume (%) | Relative modulus (GPa)                      | Relative strength (MPa) Corrected |
| 170                          | 0.2                      | 29.1 ± 0.6          | 149.2 ± 14.4   | 76.1          | 52.2       | 55.7 ± 1.2                                  | 285.2 ± 27.6                      |
| 170                          | 0.3                      | 28.6 ± 1.2          | 142.3 ± 16.2   | 75.4          | 51.3       | 55.8 ± 2.3                                  | 255.0 ± 31.6                      |
| 180                          | 0.2                      | 29.5 ± 0.1          | 156.1 ± 5.1    | 76.5          | 52.8       | 55.9 ± 0.2                                  | 295.6 ± 9.7                       |
| 180                          | 0.3                      | 28.6 ± 0.9          | 157.7 ± 12.3   | 76.0          | 52.1       | 54.9 ± 1.7                                  | 302.7 ± 23.6                      |

Table 11. Pultrusion processing parameters for PCT's and Twintex®

| Raw-material | Heated die temperature (°C) | Cooled die temperature (°C) | Pre-heating temperature (°C) | Pulling speed (m/min) |
|--------------|-----------------------------|-----------------------------|------------------------------|-----------------------|
| CF/PP PCT    | 230                         |                             | 160                          |                       |
| GF/PP PCT    | 230                         | 50                          | 170                          | 0.2                   |
| Twintex®     | 300                         |                             | 170                          |                       |

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Table 12. Microscope images of GF/PP pultruded <sup>1</sup>ofiles.

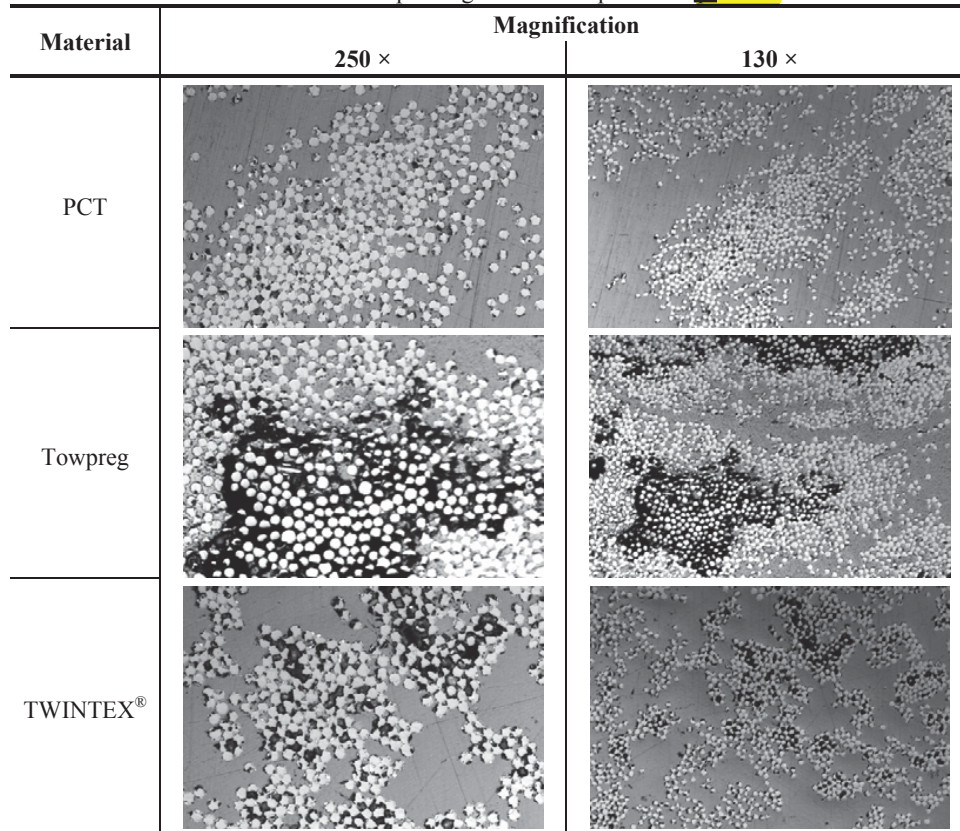


Table 13. CF/PP composite mechanical test results

| Test Type                 | Property                                      |  | Pultrusion  |                       |              |
|---------------------------|---|--|-------------|-----------------------|--------------|
|                           |   |  | Towpreg     | Towpreg with additive | PCT          |
| Flexural                  | Flexure Modulus (GPa)                         | Experimental                                   | 90.1±0.4    | 87.6±1.3              | 37.7±2.2     |
|                           |   | Theoretical                                    | 98.9        | 98.7                  | 62.7         |
|                           | Flexure Modulus / Fibre volume fraction (GPa) |  | 178.1±0.8   | 173.5±2.6             | 118.2±6.9    |
|                           | Flexure Strength (MPa)                        | Experimental                                   | 241.2±1.6   | 229.0±7.3             | 158.7±4.2    |
|                           |   | Flexure Strength / Fibre volume fraction (MPa) | 476.7±3.2   | 453.5±14.5            | 497.5±13.2   |
| Tensile                   | Tensile Modulus (GPa)                         | Experimental                                   | 110.6±5.9   | 106.1±6.3             | 63.5±4.3     |
|                           |   | Theoretical                                    | 98.9        | 98.7                  | 62.7         |
|                           | Tensile Modulus / Fibre volume fraction (GPa) |  | 218.6±11.7  | 210.1±12.5            | 199.1±13.5   |
|                           | Tensile Strength (MPa)                        | Experimental                                   | 1060.8±43.1 | 1129.3±34.6           | 636.9±38.4   |
|                           |   | Tensile Strength / Fibre volume fraction (MPa) | 2096.4±85.2 | 2236.2±68.5           | 1996.6±120.4 |
| Inter-laminar Shear       | Interlaminar Shear Strength (MPa)             |  | 12.3±0.3    | 13.0±0.4              | 14.0±0.2     |
| Fibre volume fraction (%) |   |  | 50.6        | 50.5                  | 31.9         |

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Also indicate what is black, what is gray and white standing for? Is there any defect, void, dry spot in the figures?



Table 14. Test results on the processed GF/PP composites

| Test Type                                      | Property                                      |              | Pultrusion        |              |            |
|--|---|--------------|-------------------|--------------|------------|
|  |   |              | Commingled fibres | Towpregs     | PCT        |
| Flexural                                       | Flexure Modulus (GPa)                         | Experimental | 26.2±2.0          | 28.6±0.9     | 16.8±1.5   |
|  |   | Theoretical  | 23.8              | 33.1         | 19.1       |
|  | Flexure Modulus / Fibre volume fraction (GPa) |              | 70.6±5.4          | 54.9±1.7     | 56.0±5.0   |
|  | Flexure Strength (MPa)                        | Experimental | 595.0±24          | 158.0±12.3   | 329.0±30   |
| Flexure Strength / Fibre volume fraction (MPa) |   | 1603.8±64.7  | 303.3±23.6        | 1096.7±100   |            |
| Tensile  | Tensile Modulus (GPa)                         | Experimental | 24.9±1.1          | 33.9±1.5     | 21.4±1.5   |
|  |   | Theoretical  | 23.8              | 33.1         | 19.1       |
|  | Tensile Modulus / Fibre volume fraction (GPa) |              | 67.1±3.0          | 63.5±2.9     | 71.3±5.0   |
|  | Tensile Strength (MPa)                        | Experimental | 545.9±31.7        | >336.3±22.3  | 355.8±53.2 |
| Tensile Strength / Fibre volume fraction (MPa) |   | 1471.4±85.4  | >645.5±42.8       | 1186.0±177.3 |            |
| Inter-laminar Shear                            | Interlaminar Shear Strength (MPa)             |              | 26.8±1.7          | 7.5±0.1      | 27.8±0.6   |
| Fibre volume fraction (%)                      |   |              | 37.1              | 52.1         | 30.0       |

Table 15. Compared the flexural properties of composite materials under study with those of common use and technical materials

| Material  | Density (kg/m <sup>3</sup> ) | Corrected                 | Corrected                     | Flexural properties     |                            |  |
|---|------------------------------|---------------------------|-------------------------------|-------------------------|----------------------------|--|
|   |                              | 1 <sup>length</sup> (MPa) | 2 <sup>length</sup> (kN*m/kg) | Modulus (GPa)           | Specific modulus (MN*m/Kg) | Modulus <sup>1/2</sup> /Density (GPa) <sup>1/2</sup> /(Mg/m <sup>3</sup> ) |
| CF/PP (from towpreg)[66%] <sup>a)</sup>   | 1322.5                       | 241.2                     | 182.4                         | 90.1                    | 68.1                       | 7.2  |
| GF/PP (from towpreg)[76%] <sup>a)</sup>   | 1767.3                       | 158.0                     | 89.4                          | 28.6                    | 16.1                       | 3.0  |
| GF/PP (from Twintex <sup>®</sup> )[63%] <sup>a)</sup>                                 | 1519.0                       | 595.0                     | 391.7                         | 26.2                    | 17.2                       | 3.4  |
| CF/Epoxy laminate (from prepreg hand layup – AS4 continuous tows) [74%] <sup>a)</sup> | 1560.0                       | 1724.0                    | 1105.1                        | 134.0                   | 85.9                       | 7.4  |
| Kevlar/Epoxy UD   | 1360.0                       | 655.0                     | 481.6                         | 67.0                    | 49.2                       | 6.0  |
| GF/PP compression molded (from UD melt-impregnated prepegs) [60%] <sup>a)</sup>       | 1480.0                       | 570.0                     | 385.1                         | 22.0                    | 14.9                       | 3.2  |
| GF Pultrusion [60-80%] <sup>a)</sup>  | 2016.0-2288.0                | 345.0-552.0               | 171.1-241.3                   | 31.0-41.0 <sup>e)</sup> | 15.3-17.9                  | 2.8-3.2  |
| Stainless steel (304) <sup>b)</sup>   | 8000.0                       | 517.0 <sup>d)</sup>       | 64.6                          | 193.0 <sup>e)</sup>     | 24.1                       | 1.7  |
| Carbon Steel (1020) <sup>b)</sup>   | 7870.0                       | 400.0 <sup>d)</sup>       | 50.8                          | 206.0 <sup>e)</sup>     | 26.2                       | 1.8  |
| Aluminium alloy (5052-H34) <sup>c)</sup>  | 2680.0                       | 262.0 <sup>d)</sup>       | 97.8                          | 70.0 <sup>e)</sup>      | 26.1                       | 3.1  |
| Aluminium alloy (3003-H14) <sup>c)</sup>  | 2730.0                       | 152.0 <sup>d)</sup>       | 55.6                          | 69.0 <sup>e)</sup>      | 25.2                       | 3.0  |
| Aluminium (1100) <sup>b)</sup>  | 2710.0                       | 90.0 <sup>d)</sup>        | 33.2                          | 69.0 <sup>e)</sup>      | 25.5                       | 3.1  |
| Epoxy   | 1150.0                       | 110.0-115.0               | 95.7-100                      | 3.0-3.2                 | 2.6-2.8                    | 1.5-1.6  |
| Polyester   | 1200.0                       | 80.0-123.0                | 66.7-102.5                    | 3.0-3.5                 | 2.9                        | 1.5  |
| PEEK  | 1292.0                       | 110.2                     | 85.2                          | 3.9-2.8                 | 2.2-3.0                    | 1.4  |
| PPS   | 1350.0                       | 96.0-151.0                | 71.1-111.9                    | 3.4-4.1                 | 2.5-3.0                    | 1.4-1.5  |
| PA-6  | 1115.0                       | 69.0-117.3                | 61.8-105.2                    | 1.9-2.8                 | 1.7-2.5                    | 1.2-1.5  |
| PC  | 1215.0                       | 81.4-93.2                 | 66.9-76.7                     | 2.1-2.4                 | 1.8-2.0                    | 1.2-1.3  |
| PP  | 909.5                        | 44.8-55.2                 | 49.3-60.7                     | 0.8-1.7                 | 0.9-1.7                    | 1.0-1.4  |

a) Fibre mass content; b) Annealed; c) At temper; d)Tensile Strength; e) Tensile Modulus

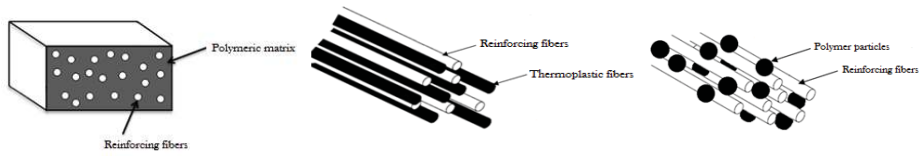
**T** Number: 1  
strength

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**T** Number: 2  
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## Figures



a) Pre-consolidate tapes (PCTs) b) Commingled fibres c) towpregs  
Figure 1. Pre-impregnated products under study

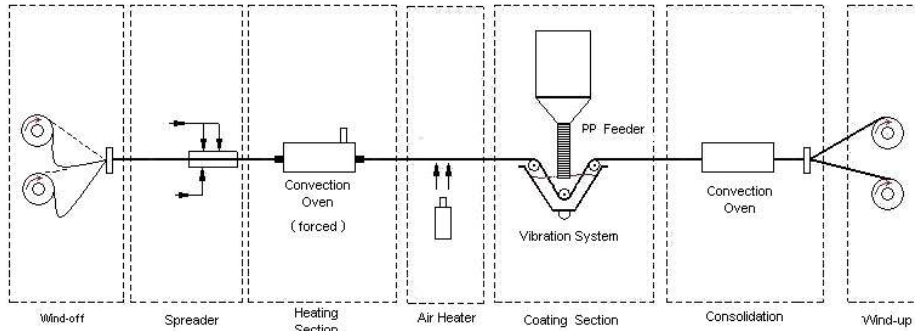


Figure 2. Powder coating line setup.

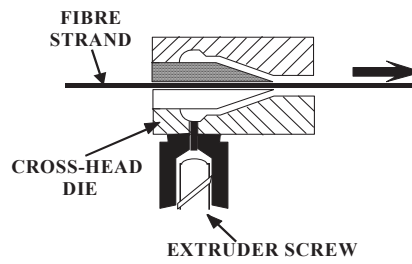


Figure 3. Cross-head extrusion die

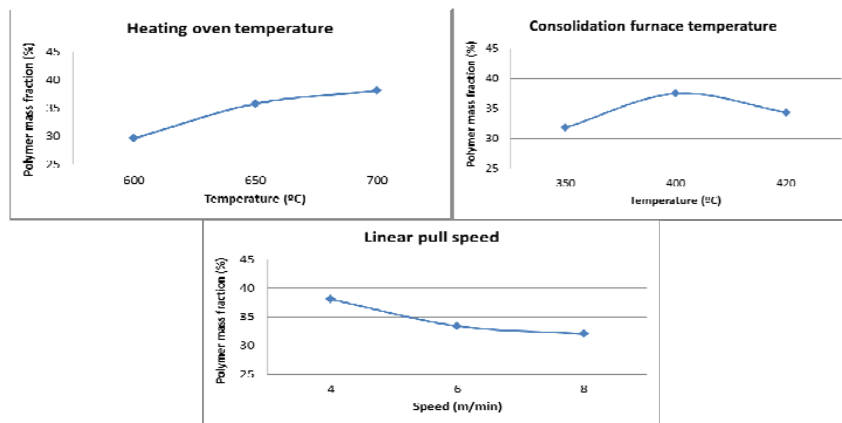


Figure 4. Variation of towpreg polymer content with processing parameters

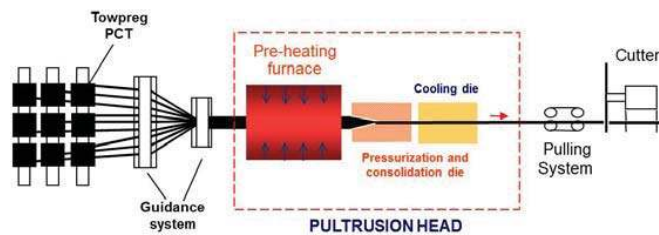


Figure 5. Schematic diagram of the pultrusion line

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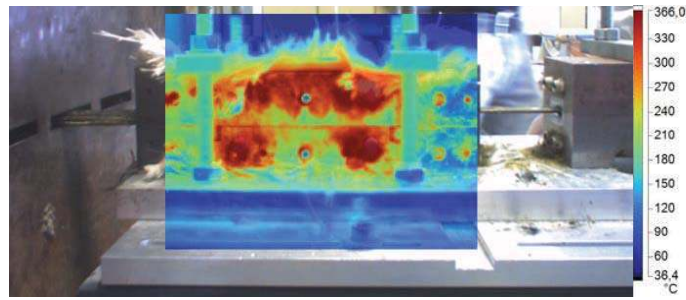


Figure 6. Heated die temperature distribution profile

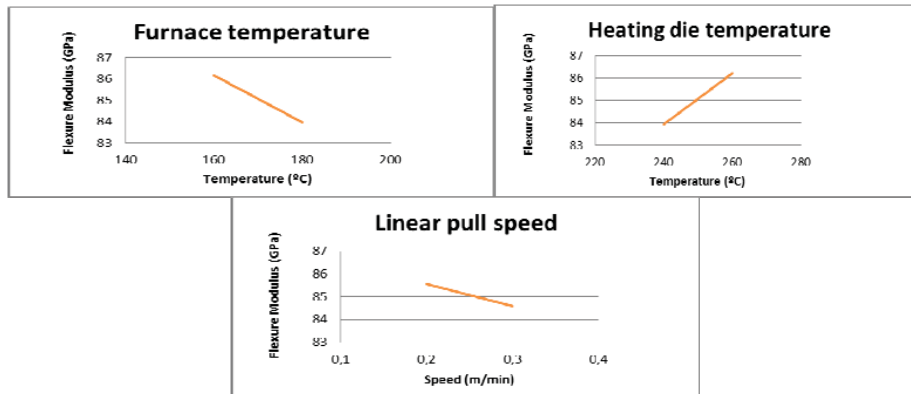


Figure 7. Variation of the flexural modulus with the selected processing parameters

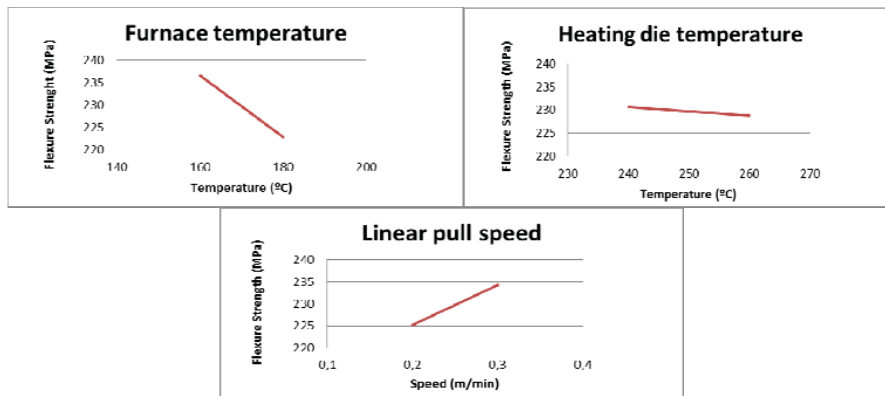


Figure 8. Variation of the flexural strength with the selected processing parameters

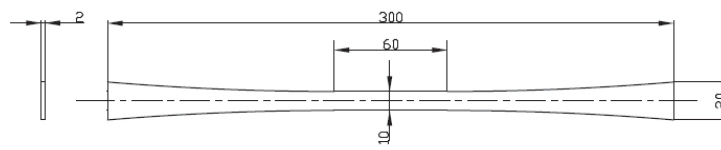
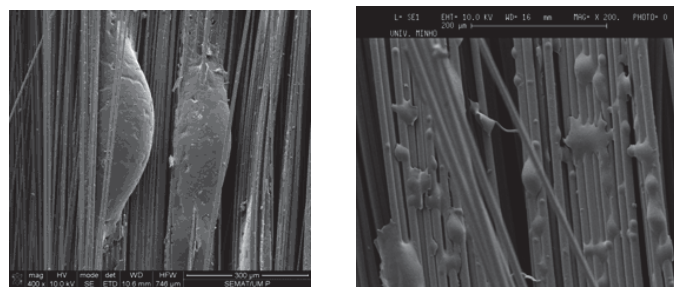


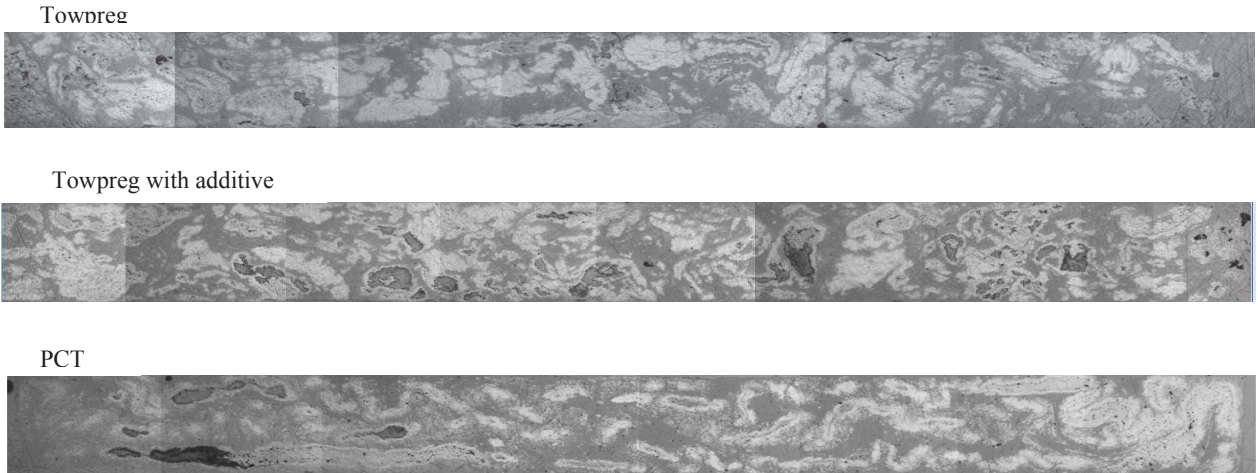
Figure 9. Geometry of the tensile test specimens



a) CF/PP towpreg (magnification 400×) b) GF/PP towpreg (magnification 200×)

Figure 10. Typical CF/PP and GF/PP towpreg SEM micrographs

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Carbon fibres impregnated with polypropylene;  
 Polypropylene matrix;  
 Dry carbon fibres;  
 Defects. Corrected

Figure 11. Optical micrographs of the CF/PP pultruded profiles cross-sections (magnification of 8.75x)

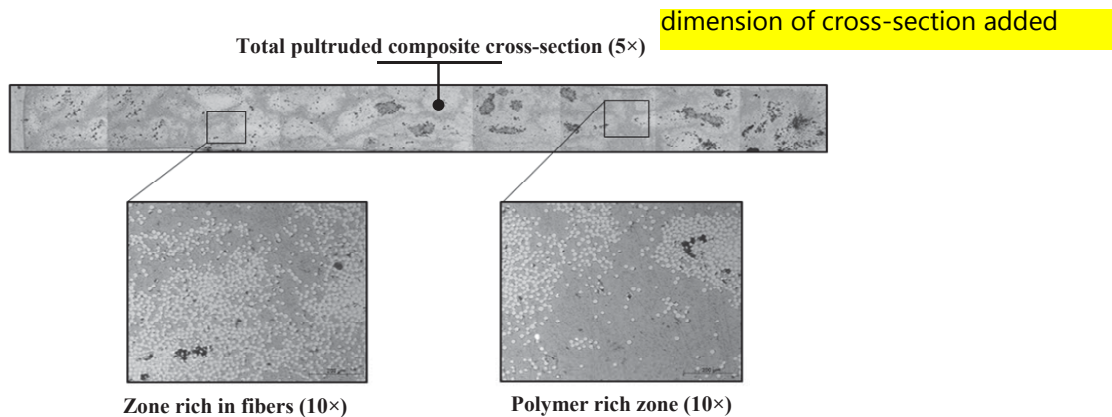


Figure 12. Cross-section of pultruded profiles from GF/PP towpregs observed under optical microscopy

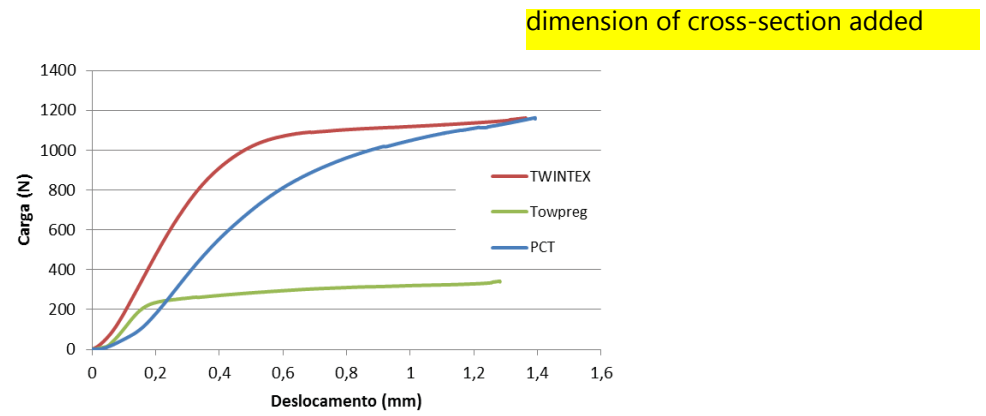


Figure 13. Short beam test results of pultruded composites from GF/PP pre-impregnated materials

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**T** Number: 1  
what kind of defect? void, porosity? please specify.

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Please indicate the scale of the graphs on the figure.

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Please show the dimensions on the cross section. I presume it is 20x2 mm.

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**T** Number: 4  
Please use the axis titles in English!

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# PULTRUSION OF FIBRE REINFORCED THERMOPLASTIC PRE-IMPREGNATED MATERIALS

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**Keywords:** *towpreg; PCT; pre-impregnated materials; pultrusion; thermoplastic composite; mechanical properties*

## Abstract

Fibre reinforced thermoplastic pre-impregnated materials produced continuously by diverse methods and processing conditions were used to produce composites using pultrusion. The processing windows used to produce these materials and composites profiles were optimized by using the Taguchi / DOE (Design of Experiments) methods. Those composites were then submitted to mechanical testing and microscopy analysis. The obtained results were compared with the expected theoretical ones predicted from the Rule Of Mixtures (ROM) and with those of similar engineering conventional available materials. The results obtained shown that produced composites have adequate properties for applications in common and structural engineering markets.

## 1 Introduction

1 Although only recently thermoplastic matrices have been used in long and continuous fibre reinforced  
2 composites as alternative to thermosetting matrices, the number of their applications is increasing due to  
3 their better ecological and mechanical performance [1]. Composites with thermoplastic matrices offer  
4 increased fracture toughness, higher damage tolerance, short processing cycle times and excellent  
5 environmental stability. They are recyclable, post-formable and can be joined by welding. The use of  
6 long/continuous fibre reinforced thermoplastic matrix composites involves, however, great technological  
7 and scientific challenges since thermoplastics present much higher viscosity than thermosettings, which  
8 makes much difficult and complex the impregnation of reinforcements and consolidation tasks [1-6].

9 Today, two major technologies are being used to allow wet reinforcing fibres with thermoplastic  
10 polymers [1, 4-6]: i) the direct melting of the polymer and, ii) the intimate fibre/matrix contact prior to  
11 final composite fabrication. Continuous fibre reinforced thermoplastic matrix pre-impregnated tapes  
12 (PCT's) are, for example, produced by direct melting processes. Alternatively, intimate contact processes  
13 allow producing cheap and promising pre-impregnated materials, such as, commingled fibres and powder  
14 coated towpregs. Sometimes, thermoplastic compatibilizers are added to the matrices to improve their  
15 adhesion and facilitate impregnation to reinforcements [7].

16 This work studies and compares the processability of final composite parts by using three different  
17 pre-impregnated materials produced by each one of the above mentioned wetting techniques. One is a  
18 pre-consolidated tape (Fig. 1 a)) that was produced by the melting process (cross-head extrusion) [8].  
19 From the other two produced by fibre/matrix intimate contact methods, one is a commercially available  
20 commingled fibres product (Fig. 1 b) and the other a towpreg (Fig. 1 c)) produced by our own developed  
21 dry coating line [9]. All studied pre-impregnated materials were based on a continuous carbon and glass  
22 fibres reinforced polypropylene matrix system. Pultrusion was the selected manufacturing method for  
23 processing all these pre-impregnated materials into composite parts. It is a versatile continuous high speed  
24 production technology, allowing the production of fibre reinforced complex profiles. Nowadays crucial  
25 challenges in pultrusion such as the residual stresses in the product which may induce damage or  
26 premature cracks and delamination are being studied [10-12].

27 The optimization of the pultrusion process was made by studying the influence of the most relevant  
28 processing parameters in the final properties of the produced pre-impregnated materials and composites.  
29 The method of Taguchi / DOE (Design of Experiments) was used to achieve this aim.

30 The possibility of using maleic anhydride as compatibilizer of carbon and glass fibre reinforced  
31 polypropylene composites was also analysed in the present work.

1 Towpregs were characterized by scanning electron microscopy (SEM), visual analysis and their  
2 polymer mass contents were determined. The final composite parts were also submitted to tensile,  
3 interlaminar and flexural tests, as well as calcination and optical microscopy tests and the results were  
4 compared with theoretical ones that can be predicted by using the ROM (Rule Of Mixtures) and other  
5 engineering traditional materials (steel, aluminium and several polymers).  
6

## 7 **2 Experimental**

### 8 **2.1 Raw Materials**

9 The following raw materials were used to produce carbon reinforced polypropylene (CF/PP)  
10 pre-impregnated materials for this work: i) a PP powder ICORENE 9184B P<sup>®</sup> and carbon fibre roving  
11 M30 SC<sup>®</sup> from the ICO Polymers and TORAY, respectively, were used to produce the CF/PP towpregs,  
12 ii) PP powder Moplen RP348U<sup>®</sup> from Basell and the carbon fibre roving already mentioned were used to  
13 manufacture the CF/PP PCT tapes.  
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16 For the glass reinforced polypropylene (GF/PP) towpregs, a 2400 Tex type E fibre rovings from Owens  
17 Corning and Icorene<sup>®</sup> 9184B P polypropylene from ICO Polymers France were used. Also, the GF/PP  
18 PCT tapes were manufactured with glass fibres (TufRov 4599) from PPG Industries and a polypropylene  
19 matrix (Moplen RP348U) from Basell.  
20

21 Commercial commingled GF/PP fibres TWINTEX<sup>®</sup> R PP 60 B 1870 FU from Owens Corning were also  
22 used in the production of pultrusion thermoplastic composite profiles, as reference of a current  
23 commercially available pre-impregnated product.  
24  
25

26 Some batches of CF/PP and GF/PP towpregs were also produced using PP powder  
27 (ICORENE 9184B P<sup>®</sup>) blended with 1% in mass content of maleic anhydride S 47 29608 707<sup>®</sup> from  
28 Merck Schuchardt OHG, in order to assess the possible enhancement of fibre/matrix adhesion [13-16].  
29

30 Tables 1 and 2 summarise relevant properties of the polypropylene, glass and carbon fibres used in  
31 present work to produce pre-impregnated raw materials (towpregs and PCT's). Table 3 shows the  
32 manufacturer datasheets properties of TWINTEX<sup>®</sup>.  
33  
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### 35 **2.2 Production of Thermoplastic Matrix Pre-Impregnated Products**

36 The dry powder coating equipment used to produce fibre reinforced towpregs is depicted in Figure 2  
37 [9, 17].  
38

39 The pre-consolidated tapes (PCT's) used in this work were produced in a cross-head extrusion equipment  
40 (see Fig.3) from our own laboratories [8]. Using this equipment, it was possible to produce the tapes  
41 (PCT's) pre-impregnated raw-materials. The overview of their main properties is given in Table 4.  
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### 2.2.1. CF/PP towpregs production and optimization

In order to optimize the production of CF/PP powder coated towpregs, different processing variables combinations were experimented and the number of trials optimized using the Taguchi approach. The studied operational parameters were:

- heating oven temperature (600, 650 and 700 °C); consolidation oven temperature (350, 400 and 450 °C); linear pull speed (4, 6 and 8 m/min).

The Taguchi approach was applied to the towpregs production process in order to obtain the condition that maximizes polymer powder content.

The polymer mass fraction in the towpregs,  $\omega_p$ , was determined by weighting towpreg strips produced in those different conditions, using equation 1:

$$\omega_p = \frac{W_t - W_f}{W_t} \quad (1)$$

where  $W_t$  and  $W_f$  are the measured unit length weights of the towpreg strip and fibre roving, respectively.

Table 5 shows the used processing conditions and obtained results, according to the established design of experiments. The average polymer mass content in towpregs, established by the design of experiences was 34.5% (Table 5).

In Figure 4 an average plot is obtained by plotting the average polymer mass fraction effect against the corresponding main processing variables (temperatures/linear pull speed) levels.

The optimal condition obtained from Taguchi method application led to the following operating parameters selection: heating oven temperature and consolidation oven temperatures of 700 °C and 400°C respectively, and a linear pulling speed of 6 m/min. Using this optimal operative condition, the amount of polymer should increase up to 45.6%. However, it was found that the average polymer content in continuous towpreg production was only 40.0% (enough for the use of towpregs in the pultrusion process, where a limit value of polymer mass content of 22% should be achieved as minimum, if only rovings are used [18]). Also, the addition of 1% of maleic anhydride to the PP polymer had no influence on the towpreg polymer mass fraction. Table 6 summarizes the determined properties on towpregs produced using the optimal combination of parameters.

### 2.2.2 GF/PP towpregs production and optimization

To try maximizing the polymer powder content in the towpregs the following processing conditions were varied within the next ranges: i) convective oven temperature (°C): 650 - 700; ii) Consolidation furnace temperature (°C): 350 – 450; iii) Coating line pulling speed (m/min): 4 – 6.

From the polymer mass fractions obtained in produced towpreg strips it was possible to establish as optimal the following operating parameters: convective and consolidation oven temperatures of 700 °C

and 400°C respectively, and a linear pulling speed of 6 m/min. In such operational conditions the GF/PP towpregs were continuously produced with polymer mass content of 30.7 %.

Unexpectedly, very low polymer mass fractions ( ~10 % - 16 %) were obtained when PP powder with 1% of maleic anhydride was used to produce GF/PP towpregs. Hence, the idea of using this additive to improve adhesion of the PP matrix to the glass fibres was abandoned. Table 6 summarizes the determined towpreg properties obtained with the combination of selected parameters.

### 2.3 Pultrusion of pre-impregnated materials

The towpregs, PCT's and commingled fibres were processed into composite rectangular bar profiles using the laboratorial pultrusion line, Figure 5 [8, 19].

To produce 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 heated die to be heated and consolidated to the required size and, after cooled down in the cooling die to solidify. The control of the die temperatures was made using two PT 100 sensors, one in each half-die. The die temperature distribution profile, obtained with a FLUKE TI45FT infrared camera, can be seen in Figure 6.

In this work, it was designed and manufactured a die to allow producing a 20×2 mm<sup>2</sup> bar-shaped profile.

#### 2.3.1 Towpreg processing

CF/PP towpregs were manufactured by pultrusion into composite bar profiles using the most relevant operating conditions. The Taguchi's/DOE method was applied, maintaining the cooling die at 25 °C, in order to optimize the processing parameters:

i) furnace temperature (160 or 180 °C); ii) heating die temperature (240 or 260 °C); iii) linear pull-speed (0.2 or 0.3 m/min).

Results have shown that it was not possible to produce pultruded profiles from towpregs in steady conditions at linear pull-speeds and consolidation die temperatures higher than 0.4 m/min and 260 °C, respectively. By using higher values for those two parameters, the process became unsteady, mainly due to the thermoplastic polymer reflux at the entrance of the consolidation die and also to their accumulation at the entrance of the cooling die.

Table 7 summarizes the processing conditions according to established in the design of experiments. The variations of the flexural modulus and strength (determined according to the procedure described in paragraph 2.4.2) with the selected processing parameters, needed for achieving the optimal condition, can be seen in Figures 7 and 8.

1 The optimal condition concerning flexural stiffness maximization obtained from Taguchi method  
2 application led to the following operating parameters selection: furnace and heated die oven temperatures  
3 of 160 °C and 260°C respectively, and a linear pulling speed of 0,2 m/min. For optimizing the flexural  
4 strength the obtained parameters combination were: furnace and heated die oven temperatures of 160 °C  
5 and 240°C respectively, and a linear pulling speed of 0.3 m/min.  
6

7 It is possible to observe that the furnace temperature of 160°C lead to the better results. That could be  
8 explained by the lower polymer reflux on the entrance of the heated die. The optimal operating conditions  
9 to maximize both flexural properties were: furnace and heated die oven temperatures of 160 °C and 260°C  
10 respectively, and a linear pulling speed of 0.2 m/min.  
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15 Finally, towpregs with additive were also pultruded into composite bars using the condition that  
16 optimizes both flexural properties (Table 8, experiment 1) and two other experiments (Table 8,  
17 experiments 2 and 3). The results from flexural testing shown in Table 9 allow concluding that the use of  
18 such additive had no significant influence on the flexural properties.  
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23 GF/PP pre-impregnated towpreg rovings were processed by pultrusion into rectangular 20×2 (mm<sup>2</sup>)  
24 profiles. To determine the best processing window, the main processing conditions were varied,  
25 maintaining the cooling die at 25 °C: i) pre-heating temperature (°C): 170 – 180; ii)  
26 pressurization/consolidation die temperature (°C): 240 – 300; iii) linear pultrusion speed (m/min): 0.2 -  
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34 Results have shown that it was not possible to produce, in steady conditions, profiles from towpregs at  
35 pultrusion speeds and pressurization/consolidation die temperatures higher than 0.3 m/min and 280 °C,  
36 respectively. By using higher, values the process became unsteady as it was already found for CF/PP  
37 towpregs processing. The same problems also occurred for temperatures below 270° C in the  
38 pressurization/consolidation die. By maintaining constant the temperatures in the cooling and  
39 pressurization/consolidation dies at 25 °C and 280 °C, respectively, profiles pultruded in different  
40 processing conditions were submitted to the flexural tests (see 2.5.2). As it is seen in Table 10, very  
41 similar values of flexural moduli and strengths were found by using pre-heating furnace temperatures and  
42 linear pultrusion pulling speeds in the ranges of 170 - 180 (°C) and 0.2 - 0.3 m/min, respectively. While  
43 the slower pultrusion pulling speed of 0.2 m/min seemed to generate profiles with higher absolute values,  
44 this was not confirmed by the flexural properties divided by the determined fibre volume fraction  
45 depicted in two last the columns of Table 10. On the other hand, higher flexural strength values were  
46 obtained at the higher temperature of 180 °C in pre-heating furnace, even if flexural strength relative  
47 values were considered (ratio between the flexural strength and the fibre volume fraction). Hence, it was  
48 concluded that the GF/PP towpreg pultrusion operating window should be the following one: i) pre-  
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heating temperature (°C): 170 – 180; ii) pressurization/consolidation die temperature (°C): 280; iii) cooling die temperature (°C): 25; iv) linear pultrusion speed (m/min): 0.2 - 0.3.

### 2.3.2 Pre-consolidate tapes(PCT's) and Twintex® processing

PCT's and Twintex® were processed into rectangular 20×2 (mm<sup>2</sup>) bar using the already mentioned pultrusion equipment being operating conditions shown in Table 11.

## 2.4 Testing

### 2.4.1 Microscopy analysis

CF/PP and GF/PP towpreg samples were characterized by scanning electron microscopy (SEM) to evaluate the adhesion of the polymer powder to the fibres and its distribution. In order to prevent charge build-up by electron absorbed by the samples, those were coated with a layer of gold with approximately 6.5 µm thick, before the observation.

To determine the impregnation quality and to evaluate the fibre distribution and fibre/matrix adhesion of the thermoplastic composites, their cross-sections were studied under optical microscopy. Observations were done using reflected light optical microscopy (Olympus BH-2). A digital camera (Leica DFC200) was used to get the image cross sectional views of the samples.

### 2.4.2 Mechanical testing

Bar samples were submitted to flexural, tensile and interlaminar testing according to the ISO standards 14125, 527 and 14130, respectively. The mechanical properties obtained were compared to the theoretical ones predicted by using the Rule of Mixtures (ROM).

Tensile tests were conducted, according to ISO 527, in a 100 kN universal testing machine at the crosshead speed of 2 mm/min using 180×20×2 mm<sup>3</sup> rectangular samples. The tensile modulus was determined from the slope of the initial linear portion of the experimental stress/strain curve. A SG Shimadzu® 50 mm length strain-gauge was used up to 0.3% strain, for accurate determination of the tensile modulus. Regarding the determination of tensile strength, it was not possible to proceed with the test until specimen failure due to grip slippage. Hence, new specimen geometry (see Figure 9) was designed and tested with good results, allowing determining the samples tensile strengths.

Three-point flexural tests were also conducted on five 100 × 20 × 2 (mm<sup>3</sup>) composite specimens, using 100 kN universal testing machine and a distance between supports of 80 mm, according to ISO 14125, at a crosshead speed of 1 mm/min.

Samples with dimensions of 20 × 20 × 2 (mm<sup>3</sup>), cut from composites processed from each pre-impregnated raw material, were submitted to interlaminar shear tests according to ISO 14130. These three point bending tests were conducted in a 50 kN universal testing machine by using an initial pre-load of 1 N at the crosshead speed of 1 mm/min and a 10 mm span between supports.

### 2.4.3 Calcination testing

Carbon and glass fibre composites mass content was determined by using calcination tests according to the EN ISO 1172. Composite samples, weighting approximately 2 g, were submitted to calcination inside a crucible in a muffle furnace during 10 min at 620° C

## 3 Results and Discussion

Figure 10 shows SEM micrographs of CF/PP and GF/PP towpregs samples. A good degree of adhesion between both carbon and glass fibres and the polymer powder particles was obtained. Also, a reasonable polymer powder distribution on the fibres was achieved at the optimised operating conditions.

The cross-sections of the pultruded composites were studied under optical Microscopy. As can be seen from Figure 11, all CF/PP composite profiles from towpregs (with and without additive) and PCT's have a reasonable distribution of the reinforcing fibres over the cross-sections. However, large differences in impregnation quality occur between the different samples that are likely to be related, directly, to the impregnation state of pre-impregnated materials used in pultrusion. It may be seen that the impregnation quality of the PCT composite samples is good, presenting almost all fibres completely surrounded ('wet-out') by the polymer. Only a few large dry spots were observed. This is most likely due to the good degree of impregnation already achieved in the PCT raw-material tape prior to the pultrusion step.

The samples from CF/PP towpreg with additive show a higher quantity of dry zones than the ones without additive. This could be due to some lack of compatibility between the fibre sizing and the used maleic anhydride coupling agent.

As Figure 12 shows, it was still possible to distinguish discontinuities on the cross section of pultruded composites from GF/PP towpregs, where it may be seen zones very rich in polymer contrasting with others with much higher quantity of fibres.

The microscopy images taken from the samples of the pultruded composites using GF/PP PCT, TWINTEX® and GF/PP towpreg, are given in Table 12. All pre-impregnated materials lead to a reasonable distribution of the reinforcing fibres over the cross-sections, although small improvements in fibre distribution can be observed going from PCT through TWINTEX® to towpreg composites. A good fibre distribution means that the fibres are uniformly spread on the polymeric matrix, without significant agglomeration areas.

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.

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 very small the effective remaining impregnation distance between polypropylene and glass fibres

1 which leads to an easier consolidation. However, some larger dry spots were observed between the glass  
2 fibres at larger magnifications (see Table 12), showing an overall impregnation quality poor when  
3 compared with the one observed in the PCT tape based pultruded composites. Finally, the depicted  
4 towpreg-based composite samples exhibit larger apparent dry zones. This is most likely due to the uneven  
5 distribution of the dry polymer powder in the towpreg, prior to pultrusion. It seems to be harder to bridge  
6 the large distances of dry glass fibre during pultrusion, which results on bigger unimpregnated zones in  
7 the pultruded composites.  
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13 Tables 13 and 14 summarize all experimentally results obtained from the CF/PP and GF/PP  
14 unidirectional composites processed by pultrusion, from the pre-impregnated products under study. To  
15 better evaluate and compare the mechanical properties obtained on the composites processed from the  
16 different pre-impregnated products studied, Tables 13 and 14 also present theoretical expected values and  
17 relative values of specific properties.  
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23 The theoretical values of moduli,  $E$ , were directly obtained from the rule of mixtures using the raw-  
24 material properties presented in Tables 1, 2 and 3, following the Eq. 2:  
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$$E = E_f \cdot v_f + E_p \cdot (1 - v_f) \quad (2)$$

27 where,  $E_f$ ,  $E_p$  and  $v_p$  are the fibre modulus, polymer modulus and fibre volume fraction, respectively.  
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30 As can be seen from Tables 13 and 14, the experimental moduli obtained from de CF/PP and GF/PP  
31 composites are in good agreement with the predicted theoretical ones. Some experimental values are even  
32 higher than the theoretical expected ones. This can be explained considering that the volume fraction  
33 content of some samples can be higher than the determined by the calcination tests.  
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40 Using the proposed new geometry (Figure 9) for the tensile test specimens, it was possible to reach  
41 failure loads and therefore determine their tensile strengths.  
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44 Analysing Table 13, one can conclude that composites processed from the CF/PP PCT's demonstrated  
45 to have better flexural and interlaminar shear strengths than those produced from CF/PP towpregs. From  
46 Table 14, it is possible to conclude that commingled fibres (TWINTEX<sup>®</sup>) presented, in general, better  
47 properties and had also shown to be more adjusted to commercial application demands and to be easily  
48 processed into final composites by the currently used manufacturing methods, probably due to their easy  
49 consolidation.  
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56 In any case, worse flexural strength and modulus results were found in GF/PP towpreg and GF/PP  
57 PCT's pultruded composites, respectively, when compared to TWINTEX<sup>®</sup>. These lower results obtained  
58 in the flexural tests are probably consequence of the inferior degree of impregnation observed in the  
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towpreg based composites and, in the case of GF/PP PCT's based composites, result from the higher rich polymer regions exhibited by this material in its outside layers, decreasing the flexural properties.

Nevertheless, the GF/PP pre-impregnated products produced in our laboratories (towpregs and PCTs) have already demonstrated very good mechanical behaviour, namely, in terms of stiffness. In fact, the composites manufactured from these products presented experimental moduli values very closed to the theoretical expected ones. While composites processed from the PCTs demonstrated to have better mechanical strength, those produced from towpregs presented higher moduli. However, if the fibre volume fractions were considered, then the relative moduli (flexure modulus/fibre volume fraction) of PCT's and towpregs exhibit similar values. As mechanical strength values are more affected by small defects than those from moduli, the composites manufactured from PCTs seem to profit from the pre-consolidate state already presented by this product before final processing. Finally, it may be noted that none of the composites made from pre-impregnated materials reached failure in the three point bending interlaminar shear tests. This fact reveals the high degree of ductility exhibited by these materials which may be relevant for many applications. Thus, the interlaminar shear strength results shown in Tables 13 and 14 correspond to maximum force applied in the test.

Figure 13 shows a typical force-displacement curves of the GF/PP composites tested specimens in the short beam shear test, from towpregs, PCT's and TWINTEX<sup>®</sup>. Those curves show that all specimens failed in plastic shear. 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<sup>®</sup> pultruded composites have similar force-displacement behaviour, towpreg based samples showed lower performance. This is due to the results from the already mentioned limited degree of impregnation of these samples.

The flexural properties of composite materials under study were compared with those of common use and technical materials, in particular metals, polymers and polymer matrix composites and as it can be seen in Table 15 [20-25] the results of produced composites bars are higher than traditional materials, revealing a possible growing interest in its application.

#### 4 Conclusions

The tests made using a proprietary pultrusion equipment already allow to conclude that is possible to produce, without major production problems, profiles from almost all available thermoplastic matrix pre-impregnated raw-materials using pull speeds of 0.3 m/min. Currently, work is carried out to increase the pultrusion processing speed to values in the range from 2 to 6 m/min. This will equalize the speed of

the pultrusion line with that of the towpreg coating and PCT tape production lines. With the use of similar operational speeds in both processes (equipments) it will be possible, in future, combining them in one.

Existing powder-coating equipment was shown to be suitable to produce CF/PP and GF/PP towpregs that could be adequately processed into pultruded profiles. From the tests made, the towpregs can be easily and continuously produced at industrial production speeds between 2 a 6 m/min.

It was possible to optimize the production of CF/PP pultruded profiles and CF/PP towpregs, through the use of Taguchi/DOE method, achieving optimal conditions. The addition of the compatibilizing agent (1% maleic anhydride) did not improve the polymer mass content in towpregs neither the mechanical properties on the final composites.

Three different commercial promising glass fibre reinforced thermoplastic matrix pre-impregnated materials were easily processed by pultrusion: a commercial available GF/PP commingled fibres product and also GF/PP towpregs and tapes manufactured in our own laboratories.

A process window was established for the production of PCT's and towpregs and for the processing of towpregs, PCT's and commingled fibres.

The mechanical properties of the composites processed from all those three GF/PP pre-impregnated (TWINTEX<sup>®</sup>, towpregs and PCT's) were determined and evaluated. All of them demonstrated to have mechanical properties compatible with the requirements of the major current structural engineering applications. In general, commingled fibres TWINTEX<sup>®</sup> presented slightly better mechanical properties and have shown to be more suitable for composite processing than the other pre-impregnated products.

In particular, very good agreement was found between the experimental moduli values of all composites produced and the theoretical ones.

More research must be done in order to increase the processing speeds of CF/PP and GF/PP towpregs as well as PCT's and to improve the impregnation, uniformity and dispersion of raw-materials in the composites.

The mechanical properties obtained in the pultruded composites allow predicting their adequate use either in general or structural engineering applications.

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## Tables

Table 1. Properties of Towpregs and PCT PP raw-materials

| Property                                       | PP powder<br>(ICORENE 9184B P <sup>®</sup> ) |                   | PP granules<br>(Moplen RP348U <sup>®</sup> ) |
|--|--|-------------------|--|
|  | Manufacturer datasheet                       | Experimental      | Manufacturer datasheet                       |
| Specific gravity (Mg/m <sup>3</sup> )          | 0.91   | 0.91              | 0.90   |
| Tensile strength (MPa)                         | Yield Strength 30                            | Yield Strength 19 | Yield Strength 30                            |
| Young Modulus (GPa)                            | 1.3  | 0.98              | 1.1  |
| Poisson's ratio                                | -  | 0.21              | -  |
| Average powder particle size (µm)              | 440  | 163               | -  |
| Glass transition temperature (T <sub>g</sub> ) | Typical value 0-20                           | -                 | Typical value 0-20                           |
| Melting temperature (T <sub>m</sub> )          | Typical value 170                            | 166               | Typical value 170                            |

Table 2. Properties of Towpregs and PCT fibres raw-materials

| Property                              | Glass fibre<br>(305E-TYPE 30 <sup>®</sup> ) |              |                        | Carbon fibre<br>(TORAY M30 SC <sup>®</sup> ) |              |
|---------------------------------------|---|--------------|------------------------|--|--------------|
|                                       | Manufacturer datasheet                      | Experimental | Manufacturer datasheet | Manufacturer datasheet                       | Experimental |
| Linear density (Tex)                  | 2400  | -            | 2400                   | 760  | -            |
| Specific gravity (Mg/m <sup>3</sup> ) | 2,65  | -            | 2,54-2,6               | 1,73   | -            |
| Tensile strength (MPa)                | 3500  | 1657         | 1900-2400              | 5490   | 2731         |
| Young Modulus (GPa)                   | 76  | 62.5         | 69-76                  | 294  | 194.5        |
| Average fibre diameter                | 17  | 13.7         | 17                     | 5  | 7.37         |

Table 3. TWINTEX<sup>®</sup> R PP 60 B 1870 FU from Owens Corning

| Property               | Values |
|------------------------|--------|
| Linear density (Tex)   | 1870   |
| Tensile strength (MPa) | 760    |
| Young Modulus (GPa)    | 29.5   |
| Fibre mass content (%) | 60     |

Table 4. Overview of the main properties of the produced pre-consolidated tapes (PCT's)

| Property            | CF/PP              | GF/PP              |
|---------------------|--------------------|--------------------|
| Fibre type          | Carbon, 760 Tex    | E-Glass, 2400 Tex  |
| Filament diameter   | 7 µm               | 17 µm              |
| Fibre content       | 45 wt.%            | 60 wt.%            |
| Matrix type         | Polypropylene (PP) | Polypropylene (PP) |
| Tape width          | 25 mm              | 25 mm              |
| Tape linear density | 14000 Tex          | 16000 Tex          |

Table 5. Taguchi approach applied to towpregs manufacturing process

| Experiments | Processing variables          |  |                              | Results                   |
|-------------|-------------------------------|--|------------------------------|---------------------------|
|             | Heating oven temperature (°C) | Consolidation furnace temperature (°C) | Linear pulling speed (m/min) | Polymer mass fraction (%) |
| 1           | 600                           | 350                                    | 4                            | 32.2                      |
| 2           | 600                           | 400                                    | 6                            | 31.4                      |
| 3           | 600                           | 420                                    | 8                            | 20.6                      |
| 4           | 650                           | 350                                    | 8                            | 27.9                      |
| 5           | 650                           | 400                                    | 4                            | 39.9                      |
| 6           | 650                           | 420                                    | 6                            | 40.7                      |
| 7           | 700                           | 350                                    | 6                            | 35.6                      |
| 8           | 700                           | 400                                    | 8                            | 40.6                      |
| 9           | 700                           | 420                                    | 4                            | 40.4                      |
| Average     |                               |  |                              | 34.5                      |

Table 6. Overview of the main properties of towpregs produced using the optimal condition

| Property            | CF/PP              | GF/PP              |
|---------------------|--------------------|--------------------|
| Fibre type          | Carbon, 760 Tex    | E-Glass, 2400 Tex  |
| Filament diameter   | 7 µm               | 17 µm              |
| Fibre content       | 60.0 wt. %         | 69.3 wt. %         |
| Matrix type         | Polypropylene (PP) | Polypropylene (PP) |
| Tape width          | 5 mm               | 5 mm               |
| Tape linear density | 1270 Tex           | 3460 Tex           |

Table 7. Taguchi approach applied to towpreg processing by pultrusion

| Experiments | Processing variables     |                              |                              | Bending properties     |                         |
|-------------|--------------------------|------------------------------|------------------------------|------------------------|-------------------------|
|             | Furnace temperature (°C) | Heating die temperature (°C) | Linear pulling speed (m/min) | Flexural modulus (GPa) | Flexural strength (MPa) |
| 1           | 160                      | 240                          | 0.2                          | 86.7 ± 1.3             | 229.0 ± 7.3             |
| 2           | 180                      | 240                          | 0.2                          | 79.5 ± 2.0             | 212.4 ± 12.6            |
| 3           | 160                      | 260                          | 0.2                          | 91.0 ± 0.4             | 241.2 ± 1.6             |
| 4           | 180                      | 260                          | 0.2                          | 85.1 ± 1.7             | 218.2 ± 9.1             |
| 5           | 160                      | 240                          | 0.3                          | 82.1 ± 2.8             | 241.7 ± 13.1            |
| 6           | 180                      | 240                          | 0.3                          | 87.5 ± 1.9             | 239.6 ± 13.3            |
| 7           | 160                      | 260                          | 0.3                          | 85.0 ± 4.4             | 234.5 ± 11.5            |
| 8           | 180                      | 260                          | 0.3                          | 83.7 ± 2.8             | 221.3 ± 7.1             |

Table 8. Taguchi approach applied to towpregs processed with additive

| Experiments | Processing variables     |                              |                              | Flexural properties     |                        |
|-------------|--------------------------|------------------------------|------------------------------|-------------------------|------------------------|
|             | Furnace temperature (°C) | Heating die temperature (°C) | Linear pulling speed (m/min) | Flexural strength (MPa) | Flexural modulus (GPa) |
| 1           | 160                      | 260                          | 0.2                          | 229.0 ± 7.3             | 87.6 ± 1.3             |
| 2           | 160                      | 240                          | 0.2                          | 191.7 ± 7.8             | 70.4 ± 2.8             |
| 3           | 160                      | 240                          | 0.3                          | 237.4 ± 11.8            | 80.5 ± 2.6             |

Table 9. Flexural test results on composite bars with and without additive from CF/PP towpregs

| Processing parameters        |     | Flexural modulus (GPa) |               | Flexural strenght (MPa) |               |
|------------------------------|-----|------------------------|---------------|-------------------------|---------------|
|                              |     | Without additive       | With additive | Without additive        | With additive |
| Furnace temperature (°C)     | 160 |                        |               |                         |               |
| Heating die temperature (°C) | 260 | 90.1 ± 0.4             | 87.6 ± 1.3    | 241.2 ± 1.6             | 229.0 ± 7.3   |
| Linear pulling speed (m/min) | 0.2 |                        |               |                         |               |

Table 10. Influence of pultrusion conditions on the flexural properties of profiles made from GF/PP towpregs

| Pultrusion conditions        |                          | Flexural properties |                | Fibre content |            | Flexural properties / fibre volume fraction |                         |
|------------------------------|--------------------------|---------------------|----------------|---------------|------------|---|-------------------------|
| Pre-heating temperature (°C) | Pultrusion speed (m/min) | Modulus (GPa)       | Strength (MPa) | Mass (%)      | Volume (%) | Relative modulus (GPa)                      | Relative strength (MPa) |
| 170                          | 0.2                      | 29.1 ± 0.6          | 149.2 ± 14.4   | 76.1          | 52.2       | 55.7 ± 1.2                                  | 285.2 ± 27.6            |
| 170                          | 0.3                      | 28.6 ± 1.2          | 142.3 ± 16.2   | 75.4          | 51.3       | 55.8 ± 2.3                                  | 255.0 ± 31.6            |
| 180                          | 0.2                      | 29.5 ± 0.1          | 156.1 ± 5.1    | 76.5          | 52.8       | 55.9 ± 0.2                                  | 295.6 ± 9.7             |
| 180                          | 0.3                      | 28.6 ± 0.9          | 157.7 ± 12.3   | 76.0          | 52.1       | 54.9 ± 1.7                                  | 302.7 ± 23.6            |

Table 11. Pultrusion processing parameters for PCT's and Twintex®

| Raw-material | Heated die temperature (°C) | Cooled die temperature (°C) | Pre-heating temperature (°C) | Pulling speed (m/min) |
|--------------|-----------------------------|-----------------------------|------------------------------|-----------------------|
| CF/PP PCT    | 230                         |                             | 160                          |                       |
| GF/PP PCT    | 230                         | 50                          | 170                          | 0.2                   |
| Twintex®     | 300                         |                             | 170                          |                       |

Table 12. Microscope images of GF/PP pultruded profiles.

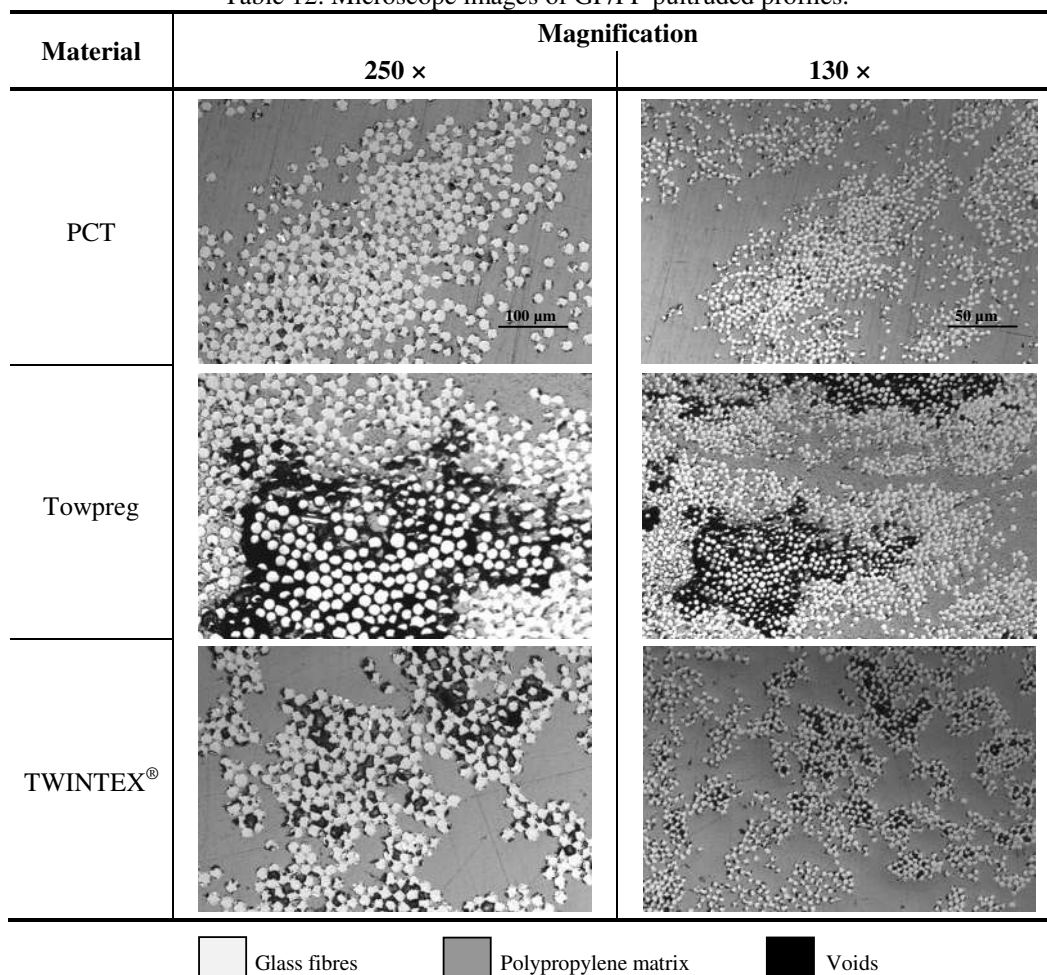


Table 13. CF/PP composite mechanical test results

| Test Type                  | Property                                      |  | <i>Pultrusion</i> |                              |              |
|----------------------------|---|--|-------------------|------------------------------|--------------|
|                            |   |  | <i>Towpreg</i>    | <i>Towpreg with additive</i> | <i>PCT</i>   |
| <b>Flexural</b>            | Flexure Modulus (GPa)                         | Experimental                                   | 90.1±0.4          | 87.6±1.3                     | 37.7±2.2     |
|                            |   | Theoretical                                    | 98.9              | 98.7                         | 62.7         |
|                            | Flexure Modulus / Fibre volume fraction (GPa) |  | 178.1±0.8         | 173.5±2.6                    | 118.2±6.9    |
|                            | Flexure Strength (MPa)                        | Experimental                                   | 241.2±1.6         | 229.0±7.3                    | 158.7±4.2    |
|                            |   | Flexure Strength / Fibre volume fraction (MPa) | 476.7±3.2         | 453.5±14.5                   | 497.5±13.2   |
| <b>Tensile</b>             | Tensile Modulus (GPa)                         | Experimental                                   | 110.6±5.9         | 106.1±6.3                    | 63.5±4.3     |
|                            |   | Theoretical                                    | 98.9              | 98.7                         | 62.7         |
|                            | Tensile Modulus / Fibre volume fraction (GPa) |  | 218.6±11.7        | 210.1±12.5                   | 199.1±13.5   |
|                            | Tensile Strength (MPa)                        | Experimental                                   | 1060.8±43.1       | 1129.3±34.6                  | 636.9±38.4   |
|                            |   | Tensile Strength / Fibre volume fraction (MPa) | 2096.4±85.2       | 2236.2±68.5                  | 1996.6±120.4 |
| <b>Inter-laminar Shear</b> | Interlaminar Shear Strength (MPa)             |  | 12.3±0.3          | 13.0±0.4                     | 14.0±0.2     |
| Fibre volume fraction (%)  |   |  | 50.6              | 50.5                         | 31.9         |



Table 14. Test results on the processed GF/PP composites

| Test Type                                      | Property                                      |              | Pultrusion        |              |            |
|--|---|--------------|-------------------|--------------|------------|
|  |   |              | Commingled fibres | Towpregs     | PCT        |
| Flexural                                       | Flexure Modulus (GPa)                         | Experimental | 26.2±2.0          | 28.6±0.9     | 16.8±1.5   |
|  |   | Theoretical  | 23.8              | 33.1         | 19.1       |
|  | Flexure Modulus / Fibre volume fraction (GPa) |              | 70.6±5.4          | 54.9±1.7     | 56.0±5.0   |
|  | Flexure Strength (MPa)                        | Experimental | 595.0±24          | 158.0±12.3   | 329.0±30   |
| Flexure Strength / Fibre volume fraction (MPa) |   | 1603.8±64.7  | 303.3±23.6        | 1096.7±100   |            |
| Tensile  | Tensile Modulus (GPa)                         | Experimental | 24.9±1.1          | 33.9±1.5     | 21.4±1.5   |
|  |   | Theoretical  | 23.8              | 33.1         | 19.1       |
|  | Tensile Modulus / Fibre volume fraction (GPa) |              | 67.1±3.0          | 63.5±2.9     | 71.3±5.0   |
|  | Tensile Strength (MPa)                        | Experimental | 545.9±31.7        | >336.3±22.3  | 355.8±53.2 |
| Tensile Strength / Fibre volume fraction (MPa) |   | 1471.4±85.4  | >645.5±42.8       | 1186.0±177.3 |            |
| Inter-laminar Shear                            | Interlaminar Shear Strength (MPa)             |              | 26.8±1.7          | 7.5±0.1      | 27.8±0.6   |
| Fibre volume fraction (%)                      |   |              | 37.1              | 52.1         | 30.0       |

Table 15. Compared the flexural properties of composite materials under study with those of common use and technical materials

| Material  | Density (kg/m <sup>3</sup> ) | Flexural properties |                             |                         |                            |  |
|---|------------------------------|---------------------|-----------------------------|-------------------------|----------------------------|--|
|   |                              | Strength (MPa)      | Specific strength (kN*m/Kg) | Modulus (GPa)           | Specific modulus (MN*m/Kg) | Modulus <sup>1/2</sup> /Density (GPa) <sup>1/2</sup> /(Mg/m <sup>3</sup> ) |
| CF/PP (from towpreg)[66%] <sup>a)</sup>   | 1322.5                       | 241.2               | 182.4                       | 90.1                    | 68.1                       | 7.2  |
| GF/PP (from towpreg)[76%] <sup>a)</sup>   | 1767.3                       | 158.0               | 89.4                        | 28.6                    | 16.1                       | 3.0  |
| GF/PP (from Twintex <sup>®</sup> )[63%] <sup>a)</sup>                                 | 1519.0                       | 595.0               | 391.7                       | 26.2                    | 17.2                       | 3.4  |
| CF/Epoxy laminate (from prepreg hand layup – AS4 continuous tows) [74%] <sup>a)</sup> | 1560.0                       | 1724.0              | 1105.1                      | 134.0                   | 85.9                       | 7.4  |
| Kevlar/Epoxy UD   | 1360.0                       | 655.0               | 481.6                       | 67.0                    | 49.2                       | 6.0  |
| GF/PP compression molded (from UD melt-impregnated preregs) [60%] <sup>a)</sup>       | 1480.0                       | 570.0               | 385.1                       | 22.0                    | 14.9                       | 3.2  |
| GF Pultrusion [60-80%] <sup>a)</sup>  | 2016.0-2288.0                | 345.0-552.0         | 171.1-241.3                 | 31.0-41.0 <sup>e)</sup> | 15.3-17.9                  | 2.8-3.2  |
| Stainless steel (304) <sup>b)</sup>   | 8000.0                       | 517.0 <sup>d)</sup> | 64.6                        | 193.0 <sup>e)</sup>     | 24.1                       | 1.7  |
| Carbon Steel (1020) <sup>b)</sup>   | 7870.0                       | 400.0 <sup>d)</sup> | 50.8                        | 206.0 <sup>e)</sup>     | 26.2                       | 1.8  |
| Aluminium alloy (5052-H34) <sup>c)</sup>  | 2680.0                       | 262.0 <sup>d)</sup> | 97.8                        | 70.0 <sup>e)</sup>      | 26.1                       | 3.1  |
| Aluminium alloy (3003-H14) <sup>c)</sup>  | 2730.0                       | 152.0 <sup>d)</sup> | 55.6                        | 69.0 <sup>e)</sup>      | 25.2                       | 3.0  |
| Aluminium (1100) <sup>b)</sup>  | 2710.0                       | 90.0 <sup>d)</sup>  | 33.2                        | 69.0 <sup>e)</sup>      | 25.5                       | 3.1  |
| Epoxy   | 1150.0                       | 110.0-115.0         | 95.7-100                    | 3.0-3.2                 | 2.6-2.8                    | 1.5-1.6  |
| Polyester   | 1200.0                       | 80.0-123.0          | 66.7-102.5                  | 3.0-3.5                 | 2.9                        | 1.5  |
| PEEK  | 1292.0                       | 110.2               | 85.2                        | 3.9-2.8                 | 2.2-3.0                    | 1.4  |
| PPS   | 1350.0                       | 96.0-151.0          | 71.1-111.9                  | 3.4-4.1                 | 2.5-3.0                    | 1.4-1.5  |
| PA-6  | 1115.0                       | 69.0-117.3          | 61.8-105.2                  | 1.9-2.8                 | 1.7-2.5                    | 1.2-1.5  |
| PC  | 1215.0                       | 81.4-93.2           | 66.9-76.7                   | 2.1-2.4                 | 1.8-2.0                    | 1.2-1.3  |
| PP  | 909.5                        | 44.8-55.2           | 49.3-60.7                   | 0.8-1.7                 | 0.9-1.7                    | 1.0-1.4  |

a) Fibre mass content; b) Annealed; c) At temper; d) Tensile Strength; e) Tensile Modulus

**Figures**

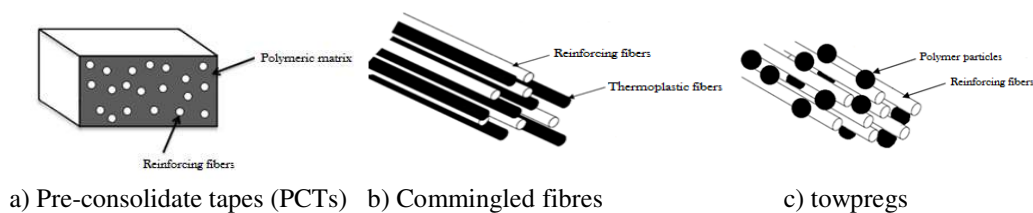


Figure 1. Pre-impregnated products under study

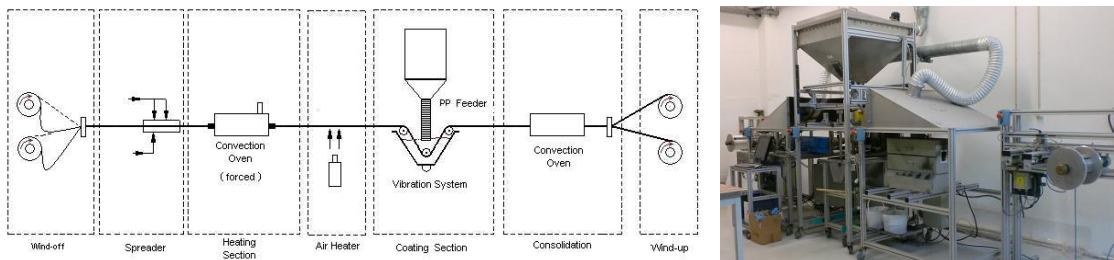


Figure 2. Powder coating line setup.

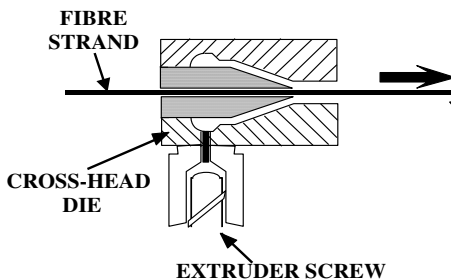


Figure 3. Cross-head extrusion die

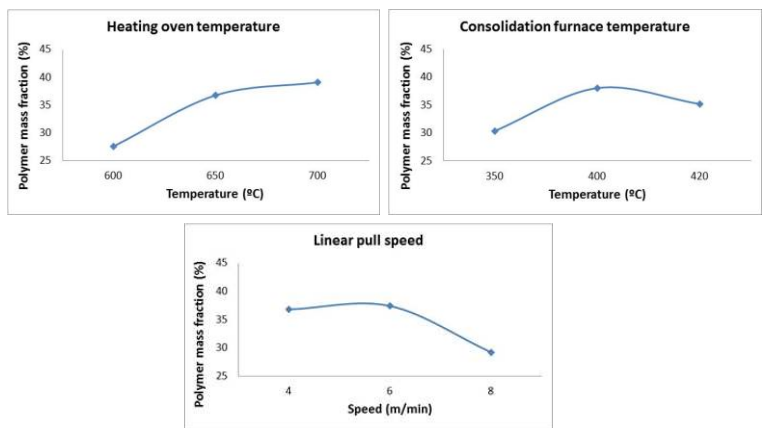


Figure 4. Variation of towpreg polymer content with processing parameters

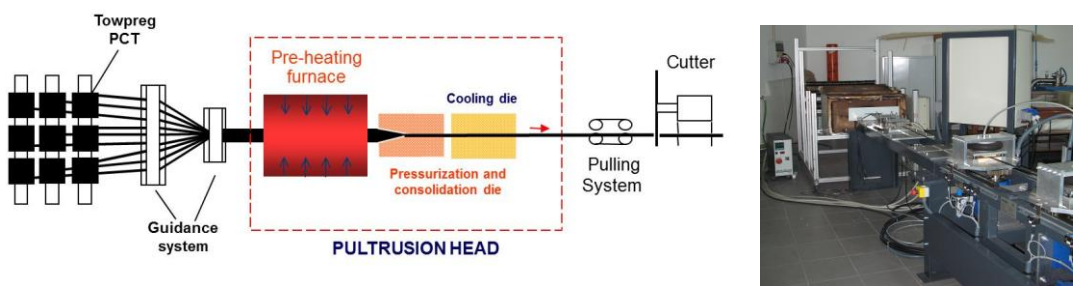


Figure 5. Schematic diagram and overview of the pultrusion line

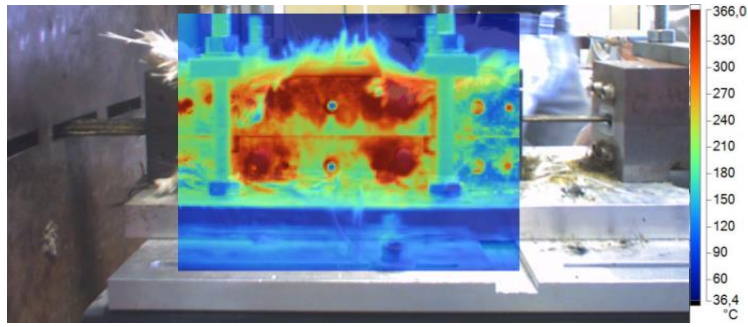


Figure 6. Heated die temperature distribution profile

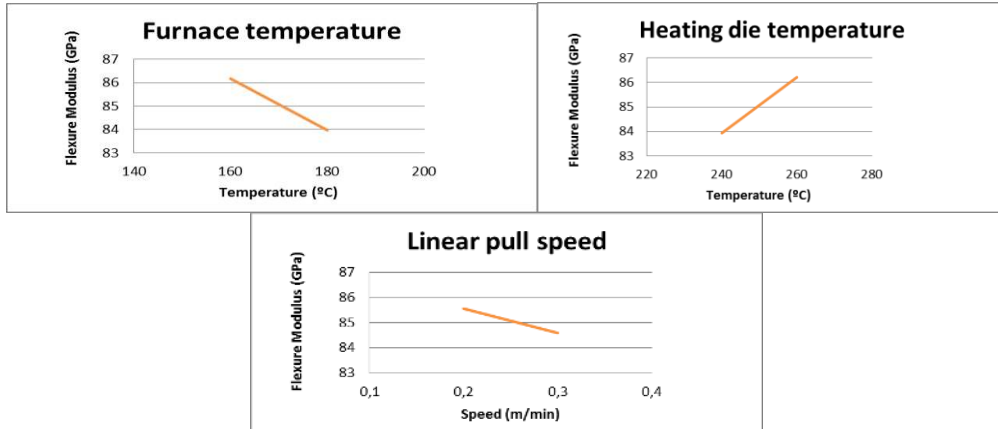


Figure 7. Variation of the flexural modulus with the selected processing parameters

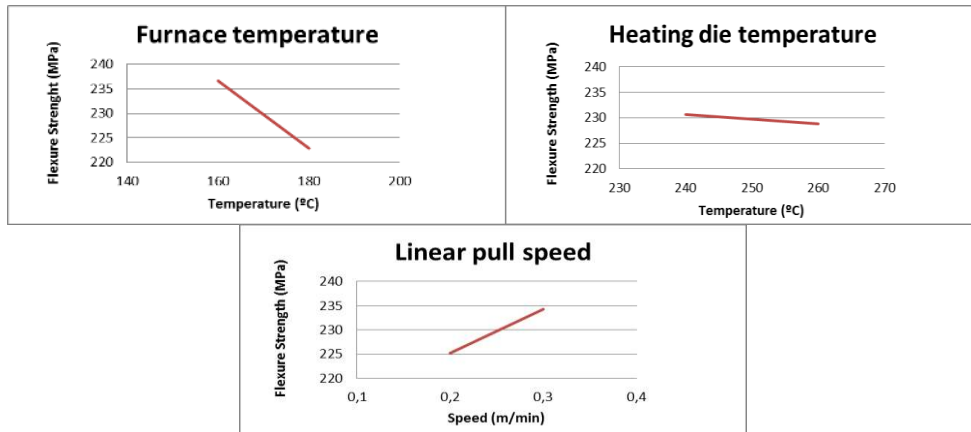


Figure 8. Variation of the flexural strength with the selected processing parameters

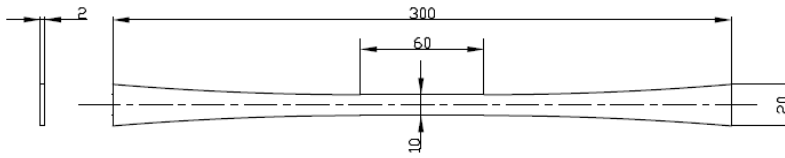
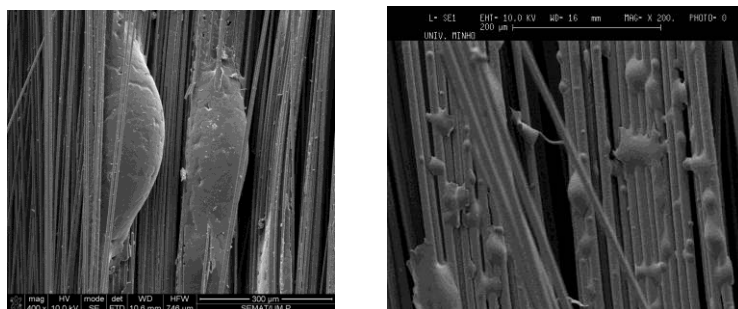


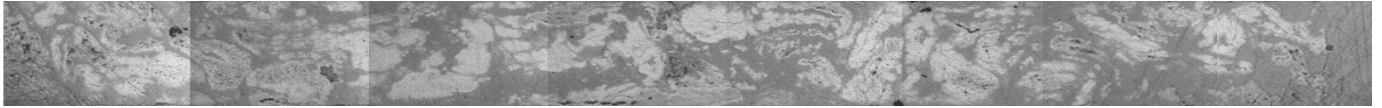
Figure 9. Geometry of the tensile test specimens



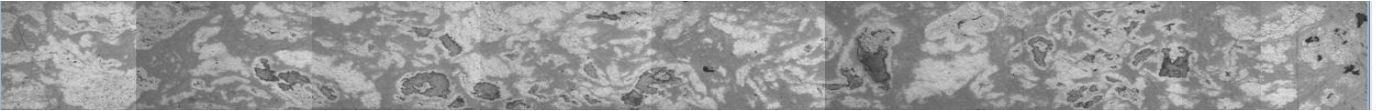
a) CF/PP towpreg (magnification 400×)      b) GF/PP towpreg (magnification 200×)

Figure 10. Typical CF/PP and GF/PP towpreg SEM micrographs

Towpreg



Towpreg with additive



PCT

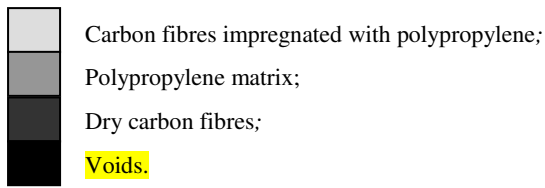
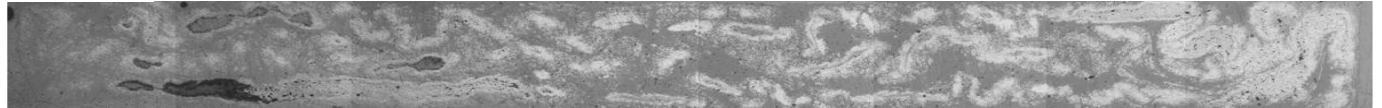


Figure 11. Optical micrographs of the CF/PP pultruded profiles cross-sections (20x2 mm<sup>2</sup>) (magnification of 6.25x)

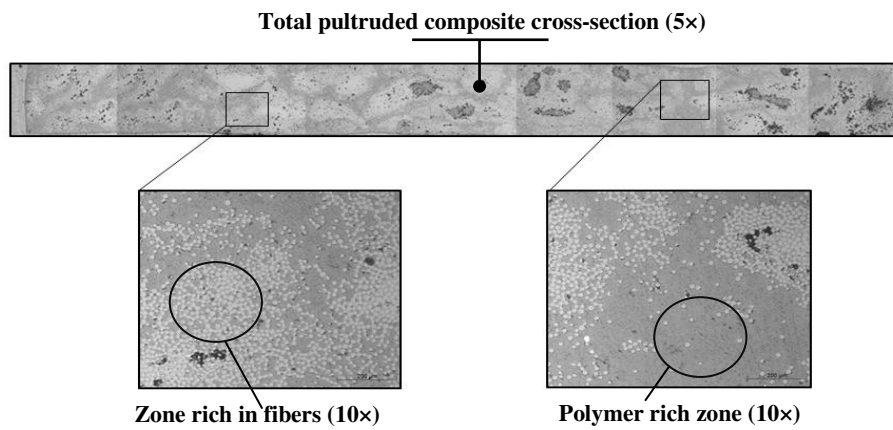


Figure 12. Cross-section (20x2 mm<sup>2</sup>) of pultruded profiles from GF/PP towpregs observed under optical microscopy

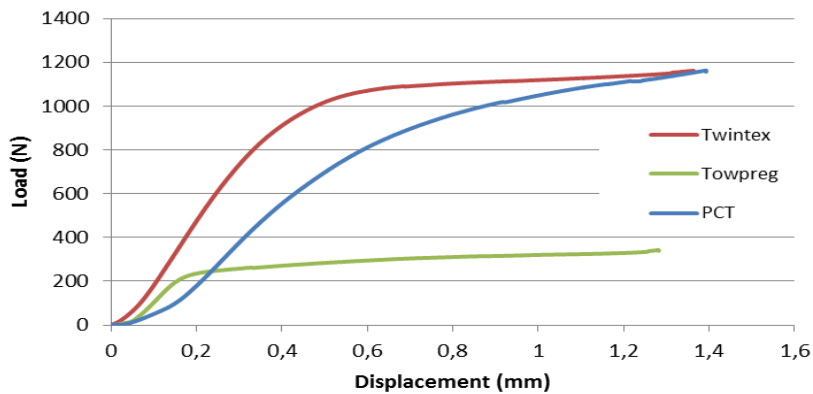


Figure 13. Short beam test results of pultruded composites from GF/PP pre-impregnated materials