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Producing LFT composite parts for large consumption markets from thermoplastic powder-coated towpregs

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Abstract

Thermoplastic matrix composites are receiving increasing interest in last years. This is due to several advantageous properties and speed of processing of these materials as compared to their thermoset counterparts. Among thermoplastic composites, Long Fibre Thermoplastics (LFTs) have seen the fastest growth, mainly due to developments in the automotive sector [0, 2]. LFTs combine the (semi-)structural material properties of long (>1 cm) fibres, with the ease and speed of thermoplastic processing.

This paper reports a study of a novel low-cost LFT technology and resulting composites. A patented powder-coating machine [3, 4] able to produce continuously pre-impregnated materials directly from fibre rovings and polymer powders was used to process glass-fibre reinforced polypropylene (GF/PP) towpregs. Such pre-impregnated materials were then chopped and used to make LFTs in a patented low-cost piston-blender developed by the Centre of Lightweight Structures, TUD-TNO, the Netherlands [5, 6]. The work allowed studying the most relevant towpreg production parameters and establishing the processing window needed to obtain a good quality GF/PP powder coated material. Finally, the processing window that allows producing LFTs of good quality in the piston-blender and the mechanical properties of final stamped GF/PP composite parts were also determined.

Keywords: long fibre thermoplastics; LFT; towpreg; mechanical properties; glass-fibre; polypropylene

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1. Introduction

The demands of the automotive industry for light, high performing materials generated an increasing interest in thermoplastic composites, especially Long Fibre Thermoplastics (LFTs). Currently, however, the high investments required for processing these materials limit their further application in other industries. A typical installation comprise of a specially designed extruder, where either dry fibres with thermoplastic granules or composite pellets can be mixed with minimised fibre breakage. The resulting composite melt can then be either injection or compression moulded. The high cost of the equipment involved, however, has limited the use of LFT-technology mainly to the automotive industry, thus not exploiting the many opportunities that lie outside this sector.

The present work reports the experimental and theoretical study of a new cost-effective technology for making composites from powder-coated GF/PP LFTs [3, 4], by mixing in a piston-blender and compression moulding [5, 6]. It is shown that the process can be carried out without major problems, and that the properties of the composites produced are

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comparable to those of other LFT-materials. Overall, the current work shows the feasibility of this novel technology as a low-cost alternative to current LFT-processes. The blender produces a composite melt that can be directly compression moulded into composite plaques. Due to its simplicity, this process can generate LFT-moulded products with competitive properties, directly from fibre and matrix materials, at a fraction of the cost of commercially available equipment.

2. Experimental

2.1. Raw Materials

LFTs are almost only focused in highly competitive commercial markets. Thus, a polypropylene (PP) and glass fibres (GF) were the raw materials chosen for being used in this work due to the excellent properties/cost ratios they presented. The PP powder ICORENE® 4014 from ICO Polymers France, with particle size between 60 and 850µm, and the 2400 Tex glass fibres, 305E-Type 30 Direct Roving® from Owens Corning's, were selected as matrix and reinforcement material, respectively, to produce the towpregs by using the abovementioned powder-coating technology. Table 1 presents the properties of the glass fibre. Experimental results obtained in an earlier work [7] have shown that fibres present lower tensile strength than the one depicted in the manufacturer datasheet.

Table 1. Properties of glass fibres

Property	Units	Experimental results	Manufacturer datasheet
Density	Mg/m^3	-	2.56
Tensile strength	MPa	1657	3500
Tensile modulus	GPa	62,5	-
Linear weight	Tex	-	2400
Av. filament diameter	µm	13.7	17
Poisson's ratio		-	0,26

To determine the density and tensile properties of the ICORENE® 4014 PP, compression moulded samples of this material were submitted to tests according to the standards ISO 1183-1:2004 and ASTM D638 – 14, respectively. Table 2 shows the results obtained from those tests and compares them with those ones depicted in the manufacturer datasheets.

As it may be seen from table 2 only small differences were detected between experimental and manufacturer datasheet properties.

Table 2. Properties of ICORENE® 4014 PP

Property	Units	Manufacturer data sheet	Experimental results
Density (ISO 1183)	Mg/m^3	0,9	0.89± 0.06
Tensile strength (ASTM D 638)	MPa	24	21.2 ± 0.5
Tensile modulus	GPa	-	1.05 ± 0.006
particle powder dimensions	µm	400	-

The PP ICORENE® 4014 was submitted to further experimental tests to study its nature and determine its thermal properties, which allowed better defining its processing conditions.

Figure 1 shows a curve obtained from a test made in a differential scanning calorimeter (DSC) TA Instruments, model DSC Q20, by using a temperature range between 40ºC and 260ºC and a 10ºC/min heating rate.

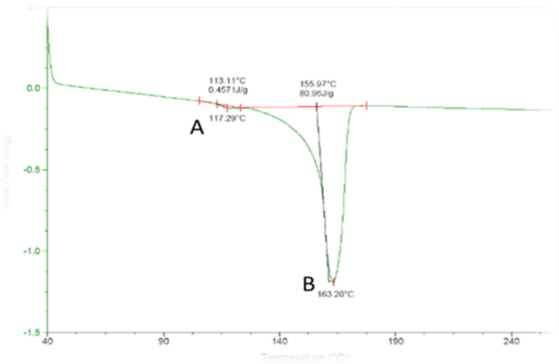


Fig. 1. Result from the DSC test on the PP

As it may be seen from figure 1, two deflection points were with surprise found on the curves obtained from DSC: the first one approximately at 117.3ºC and another one around 163.2ºC. Such behaviour allowed to conclude that PP ICORENE® 4014 may perhaps be a Polypropylene-Polyethylene (PP/PE) copolymer because such temperatures correspond to the possible melting points of these two polymers.

To analyse the above peculiarity with more detail ICORENE® 4014 PP samples were submitted to the Fourier transform infrared spectroscopy (FTIR) tests on a Perkin-Elmer Spectrum BX. Figure 2 presents a spectrum obtained from these tests.

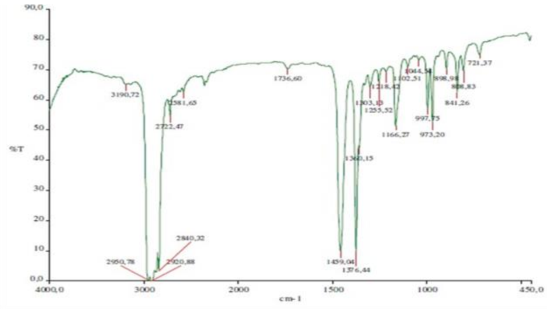


Fig. 2. PP spectrum obtained from the FTIR tests

As the spectrum depicted in figure 2 shows, the bands in the range of 2950-2840 cm^{-1} observed correspond to aliphatic liaisons characteristic of both PP and PE, but bands visible around 1300 -720 cm^{-1} correspond only to deformations of liaisons characteristic of PP. This allowed us to conclude that ICORENE® 4014 is in fact a PP/PE copolymer.

Finally, figure 3 shows the results of the thermal gravimetric analysis (TGA) to which the ICORENE® 4014 was submitted, in a inert nitrogen atmosphere, by using a TA Instruments TGA, Q500 model, and heating and a flux rates of 10ºC/min and 50 ml/min, respectively.

As can be observed in the figure at the temperature of 421.7 ºC the polymer lost around 99.8% of its mass and is totally degraded.

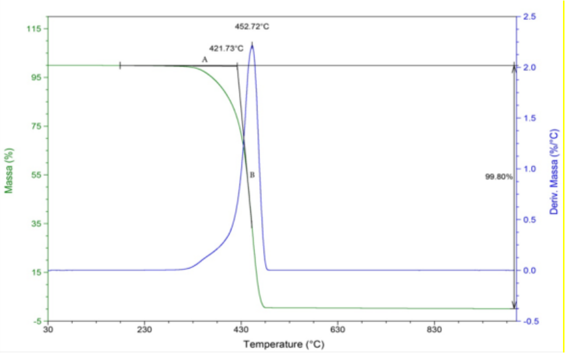


Fig. 3. Result of the TGA analysis made on the ICORENE® 4014

2.2. Production of GF/PP towpregs

The GF/PP towpregs were produced in a developed dry powder coating equipment schematically shown in Fig. 4 and illustrated in the photo of Fig. 5 [3, 4]. It consists of six main parts: wind-off system, fiber spreader unit, heating section, coating section, consolidation unit and a wind-up section. Initially, the reinforcing fibers are wound-off and pulled through a pneumatic spreader and then coated with polymer by heating in a convection oven and made to pass into a polymer powder vibrating bath. A gravity system allows maintaining the amount of polymer powder constant. The consolidation unit oven allows softening the polymer powder, promoting its adhesion to the fiber surface. Finally, the thermoplastic matrix towpreg is cooled down and wound-up on a spool.

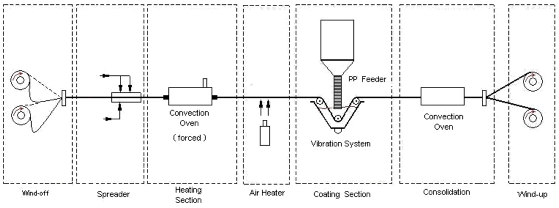


Fig. 4. Powder coating line setup

Fig. 5. Prototype coating line used to produce towpregs

To try maximizing the polymer powder content in the towpregs the coating line pull speed and the temperatures in the convection and consolidation ovens were varied. Table 3 presents the different operational conditions used and the corresponding polymer content achieved in the processed GF/PP towpregs.

The polymer mass fraction in the towpregs, ω_p, was determined by weighting towpreg strips produced in those different conditions and using the following equation:

$$\omega_p = \frac{w_t - w_f}{w_t} \qquad (1)$$

where w_t and w_f are the measured unit length weights of the towpreg strip and fiber roving, respectively.

Table 3. Operational conditions used to optimise the GF/PP towpregs production

Condition	Pull speed (m/min.)	Convection oven temperature (°C)	Consolidation oven temperature (°C)	Polymer mass fraction on towpregs (%)
1	3.5	700	400	27.4
2	4	700	400	30.5
3	4	700	420	19.8
4	5	700	400	29.4
5	6	700	400	30.1
6	6	700	420	24.2

From values of polymer mass fraction obtained shown in table 3 it was possible to conclude that the conditions 1 and 5 were those where maxima polymer mass content was obtained in the produced towpregs. However, as much higher pull speed is used in condition 5, it was chosen as the best to produce the towpregs because represents the most costly-effective one. Thus, the towpregs applied to produce the LFT composites were process by using the parameters shown in table3 for condition 5.

2.3. Piston-blender and stamping

The towpregs produced in the conditions defined in the previous paragraph were subsequently cut into 25 and 50 mm lengths to be introduced in the low shear piston-blender. The idea of using LFTs with two different fibre lengths was to analyse the importance of this parameter on the mechanical properties of the final LFT composite parts.

Then, the chopped towpregs were mixed with polymer material in the piston-blender, which was specifically developed to melt LFTs while maintaining fibre length [5,6]. In this machine, the traditional concept of using an extrusion screw to attain the required levels of mixing and melting, has been replaced by a simple mechanism of rotating heated rods. Due to the very low shear induced on the melt, fibre breakage is limited to a minimum, while accomplishing a sufficient level of mixing. Due to the machine's simple construction, its costs are only a fraction of those of the commercially available LFT-extruders. Therefore, it offers the possibility of processing LFTs with only minor investments.

A schematic representation and photograph of the piston-blender developed by the Centre of Lightweight Structures are shown in figures 6 and 7, respectively. The LFTs and polymer powder are fed to the hopper and the heated rods start rotating to mix the material. At the same time the heat of the cylinder wall (heated by elements from the outside) and the rods melts the material. After adequate melting and mixing, a piston moving forward ejects the mixture that is cut by a cutting device. The material is then ready for stamping.

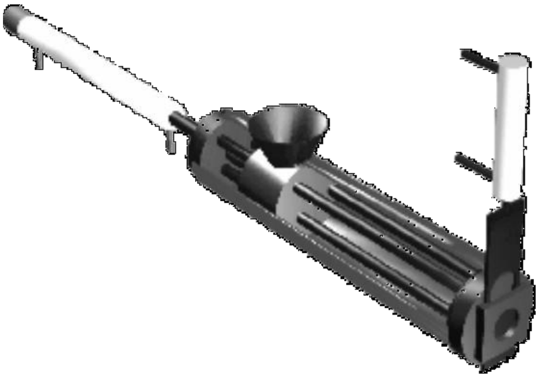


Fig. 6. Schematic representation of the piston-blender

Fig. 7. Photograph of the piston-blender used in the present work

By adding different proportions of virgin PP powder and cut glass fibres in the hopper, composites with varying fibre content could be obtained. In this way, it was possible to test the effect of fibre length, fibre mass fraction and hopper temperature in the final properties and content on processing and properties. The molten materials produced by the piston-blender were final stamped in a hydraulic press using a cooled mould with dimensions of 170 mm×120 mm×2.5 mm to produce LFT composite plates. Table 4 shows the different conditions used to produce LFT composite plates. As can be seen, the stamping conditions were maintain constant in 100 bar compression pressure, 1 min compression time and 10ºC as mould temperature.

To assess the influence of all those above mentioned operational conditions on the final stamped GF/PP composite plates, they were submitted to tensile and flexural testing accordingly to the ISO 527-1:2012 and ISO 14125:1998 standards, respectively. Density tests were also used to determine the void content on the final stamped LFT plates and their sections observed under microscopy.

Table 4. Operational conditions used to produce the final GF/PP LFT composite plates

Case	Piston-blender temperature (ºC)	Fibre mass fraction (%)	Fibre length (mm)	Stamping pressure (bar)	Stamping time (min)
1	220	30	25	100	1
2	230	30	25	100	1
3	240	30	25	100	1
4	240	40	25	100	1
5	240	50	25	100	1
6	240	30	50	100	1

Figures 8 and 9 show the hydraulic press belonging to the Pole for Innovation in Polymer Engineering (PIEP) used to stamp the GF/PP produced LFTs and a final stamped LFT composite plate, respectively.

Fig. 8. Hydraulic press used to stamp the final GF/PP LFT plates

Fig. 9. Final stamped GF/PP LFT composite plate

2.4. Testing the final stamped LFT composite plates

The void contents and properties obtained from the tensile and flexural tests made on GF/PP LFT plates produced in the conditions defined for the cases shown in table 4 are depicted in next three tables 5, 6 and 7, respectively.

Table 5. Void content determined in the final stamped LFT plates

Case	Fibre mass fraction (%)	Fibre Volume fraction (%)	Theoretical density (Mg/m^3)	Experimental density (Mg/m^3)	Void content (%)
1	29,3	12,6	1,10	1,04	5,18
2	29,3	12,6	1,10	1,06	3.90
3	29,3	12,6	1,10	1,06	3,18
4	43,9	21,4	1,24	1.16	6.53
5	54,0	27,2	1,34	1.22	8.95
6	34,3	15,3	1,14	1.11	2,54

Results in table 5 show that only processing conditions corresponding to the cases 1, 4 and 5 defined in table 4 increased slighter the level of void content in the stamped GF/PP LFT plates.

Table 6. Tensile properties obtained in the final stamped LFT plates

Case	Tensile strength (MPa)		Tensile Modulus (GPa)	
	Average	Stand. Dev.	Average	Stand Dev.
1	18,50	8,31	3.7	0.34
2	24,12	1,95	3.3	0.41
3	22,80	2,26	3.3	0.72
4	27,27	4,97	4.8	0.63
5	26,49	3,39	4.5	1.2
6	22,14	7,14	4.7	0.54

On other hand, the mechanical properties obtained, both in the tensile and flexural tests (see tables 6 and 7), seem to show that the LFT stamped plates produced in the conditions corresponding to the case 4 (see table 4) presented the best mechanical properties.

The processing conditions in that case (4) were: 240 ºC in the piston-blender hopper, a fibre length of 40 mm and 40% as fibre mass fraction.

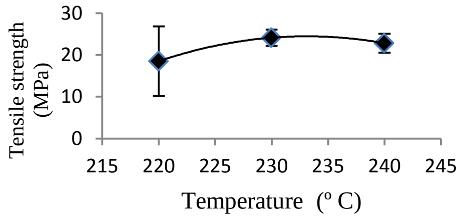
Table 7. Flexural properties obtained in final stamped LFT plates

Case	Flexure strength (MPa)		Flexure Modulus (GPa)	
	Average	Stand. Dev.	Average	Stand. Dev.
1	58,73	23,34	2.9	0.97
2	65,79	11,54	3.5	0.65
3	55,24	5,8	2.8	0.38
4	88,45	14,75	4.2	0.80
5	71,41	12,24	3.7	0.57
6	54,30	10,35	3.3	0.75

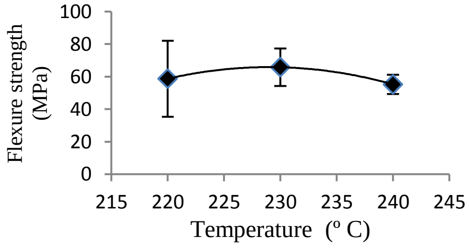
Other conclusion that made be taken from the tensile and flexural results obtained is that while bigger strengths were obtained in the bending tests, higher elastic moduli were obtained in tensile ones. This may be caused by differences in the distribution of the fibre and matrix along the sample thickness cross-section and also by lack of adhesion between fibre and polymer.

A deeper analysis of the influence of each processing parameter on the mechanical properties of the final stamped LFT composite plates was made by plotting the different mechanical properties against the variation of each one.

Figures 10 and 11 depicted the variation of the strength and moduli with the temperature in piston-blender hopper.

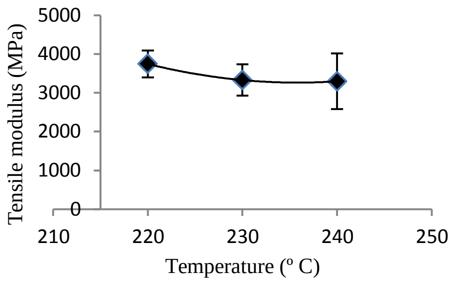


a) Tensile strength

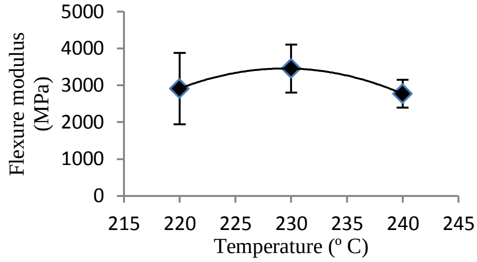


b) Flexure strength

Fig. 10. Influence of piston-blender hopper temperature on : a) tensile strength; b) flexural strength



a) tensile modulus



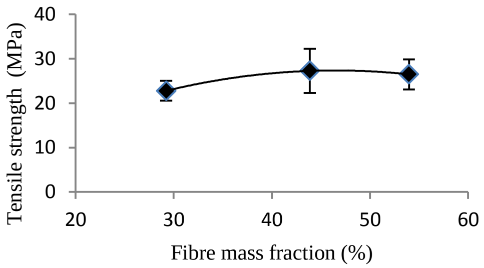
b) flexural modulus

Fig. 11. Influence of piston-blender hopper temperature on : a) tensile modulus; b) flexural modulus

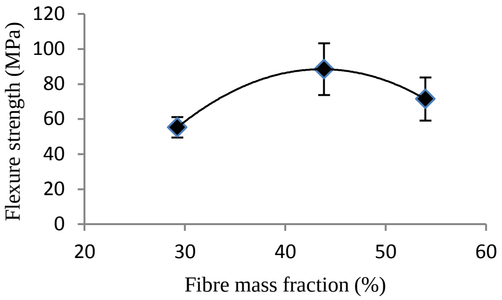
From the above figures 10 and 11 it seems possible to conclude that while the temperature in piston-blender hopper didn´t influence clearly the tensile flexural modulus (see fig. 11 a)), selecting a temperature around 230ºC in the piston-blender hopper may be considered one good contribute to maximise the mechanical properties on the final stamped LFT plates.

Figures 12 and 13 show the influence of the fibre mass fraction on the strength and moduli of the stamped LFT plates, respectively. As can be seen from those graphs, the use of fibre mass fraction around 45% seems to maximise both the strength and moduli of the stamped LFT plates in flexure and tension.

Finally, figures 14 and 15 show the effect of the fibre length in the same mechanical properties. In this case, the effect was not clearly. The increase of fibre length seems to cause slight decrease on strengths and, on the contrary, a small increase on the moduli of the final stamped LFT composite plates. However, it seems to see wiser using fibre lengths in the range from 30 mm to 40 mm to produce stamped LFT plates.

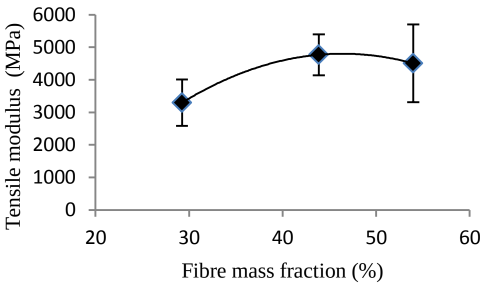


a) Tensile strength

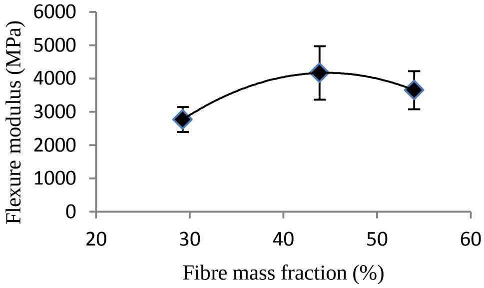


b) Flexure strength

Fig. 12. Influence of the fibre mass fraction on : a) tensile strength; b) flexural strength

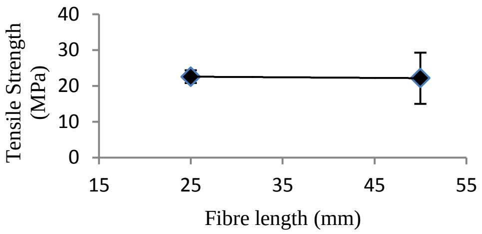


a) tensile modulus

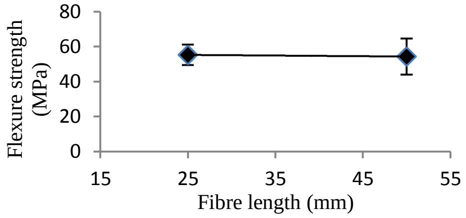


b) flexural modulus

Fig. 13. Influence of the fibre mass fraction on : a) tensile modulus; b) flexural modulus

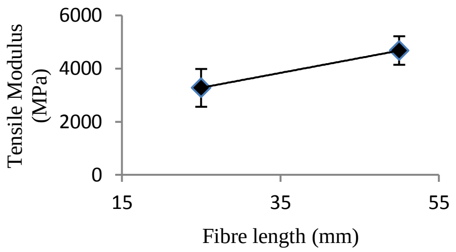


a) Tensile strength

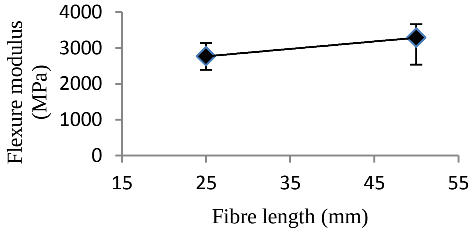


b) Flexure strength

Fig. 14. Influence of the fibre length on : a) tensile strength; b) flexural strength



a) tensile modulus



b) flexural modulus

Fig. 15. Influence of the fibre length on : a) tensile modulus; b) flexural modulus

In summary, from the results obtained it may be conclude that the following parameters seem to maximise the mechanical behaviour of the stamped LFT plates: a temperature in the piston-blender hopper around 230ºC, a fibre mass fraction of 45% and fibre lengths between 30 mm to 40 mm.

However, a deeper study already made using the statistical analysis of variance (ANOVA) which is out of the scope of the present paper allowed to conclude that the fibre mass fraction was the only parameter that had a really influence the final mechanical properties of the mechanical behaviour of the stamped LFT plates.

2.5. Assessing the properties of the stamped LFT plates

Table 8 compares the mechanical properties obtained on the GF/PP LFT plates stamped in the present work is compared in able to the theoretical ones that may be predicted from the Classical Lamination Theory (CLT) by using the rule of mixtures [8].

Table 8. Comparison between experimental and theoretical mechanical properties of final stamped LFT plates

Case	Tensile strength			Young Modulus		
	Experi-mental (MPa)	Theo-retical (MPa)	Rela-tive error (%)	Experi-mental (GPa)	Theo-retical (GPa)	Rela-tive error (%)
1	18.5	96.7	80.9	3.7	3.9	3.0
2	24.12	96.7	75.1	3.3	3.9	13.8
3	22.80	96.7	76.4	3.3	3.8	14.6
4	27.27	149.51	81.8	4.8	5.8	18.2
5	26.49	184.66	85.7	4.5	7.1	36.9
6	22.14	113.33	80.5	4.7	4.5	4.3

As results in table 8 show that te LFT plates stamped in this work even presenting much lower strength values than the theoretical expected ones revealed to have values of stiffness (modulus) nearby the theoretically predicted ones. This may be explained by the fact that the strength to be much more related with the presence of defects than the modulus, which is a much more intrinsic characteristic of a material.

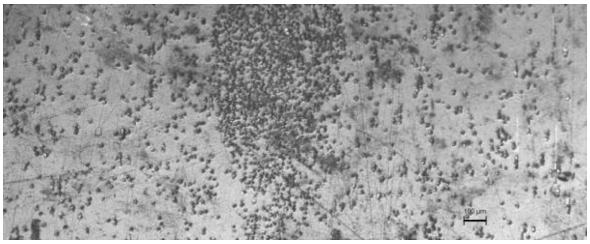
The mechanical properties obtained in the tests made on the LFT stamped plates are also compared with those presented by similar materials already available in the market in table 9. As can be seen the LFTs plates stamped in the work in general still have lower mechanical properties than other similar materials already available in the market especially in terms of tensile strength. The values of bending strength and moduli in spite of being lower are already very similar to the other competitors in the market. However, it may be said that the GF/PP LFT parts stamped from have already enough good properties for being used in the major structural engineering commercial applications, namely, on the automotive industry.

Further studies that could be made in future in order to decrease the level of defects and improve fibre/polymer adhesion in LFT parts stamped from towpregs may allow these materials presenting similar or even much better mechanical properties than their material competitors in the commercial market.

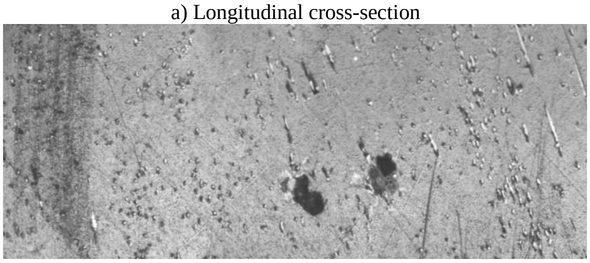
Table 9. Comparing of mechanical properties obtained on the stamped LFT plates with those of other materials

Material	Tensile strength (MPa)	Tensile modulus (GPa)	Flexure strength (MPa)	Flexure modulus (GPa)	Density (Mg/m^3)	Fibre mass fraction (%)
Stamped LFT plates	27.3	4.8	88.5	4.2	1.20	43.9
Azdel (GMT)	65.0	4.5	97.7	4.3	1.12	31
Verton (LFT)	103.0	7.8	153.0	6.5	1.13	30
RTP (LFT)	76.0	6.2	112.0	4.8	1.12	30
Celstran (LFT)	95.0	6.0	150.0	6.0	1.13	30

Finally, samples cut from the stamped LFT plates were also observed under the optical microscope. Figure 16 depicts two typical captions of the longitudinal (Fig. 16a)) and transversal (Fig. 16b)) of the stamped LFT plates taken under the optical microscope.



a) Longitudinal cross-section



b)Transversal cross-section

Fig. 16. Cross-sections of stamped LFT plate observed under the optical microscope (magnification: ×25): a) Longitudinal cross-section ; b) Transversal cross-section

As figure 16 shows, an expected roughly fibre distribution and some voids were well observed under microscopy. Such observations reinforced the idea that further work will be need in future to remove those defects and improve significantly the already good properties of the LFT plates studied in the present work.

3. Conclusions

The present work demonstrated that the production and processing of a new type of Long Fibre Thermoplastic composites can be carried out without major problems. The properties of the composites obtained are at a level comparable to other LFT-materials. Whenever differences were found, improvement of the fibre-matrix adhesion of the current system should lead to better properties. Overall, the system (material and technology) has proven its feasibility as a low-cost alternative to current LFT-processes. Furthermore, it was possible to optimise and found the best processing windows to produce the towpregs and the LFT material to be adequately stamped.

Acknowledgements

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