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Rapid manufacturing of woven comingled flax/polypropylene composite structures

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Abstract
The use of natural fibre reinforced composites such as flax fibre / polypropylene is in a constant expansion particularly in automotive and marine industries due to their good mechanical properties, low density and thus, lightness, low environmental impact, low cost, recyclability, renewable properties of flax fibres and a minimum energy intake during their process. One of the major challenges of thermoplastic composites for the automotive industry is to manufacture finished parts in a single processing step within a minimum amount of time. For this purpose, a stamping airflow device was specifically developed. It is designed to produce woven comingled composite parts from comingled woven fabrics such as flax/polypropylene in only 200 s. Firstly, preliminary tests and the optimization of processing parameters were performed. Then, a quasi-static mechanical characterization of the formed parts was realized. By using criteria based on mechanical properties, the optimal process parameters such as the level of pressure, temperature, holding time and cooling rate so that to obtain the lowest voids rate were determined. Finally, a comparison of the mechanical properties of parts obtained from using the new manufacturing process and a conventional thermo-compression process is presented to demonstrate the interest and the level of performance achieved by this original and fast manufacturing device.

Keywords Flax/polypropylene comingled fabric · Rapid manufacturing · Thermo-compression · Void effect · Mechanical characterization

Introduction

The use of fibre reinforced composites is constantly expanding. It is particularly the case in the aerospace, automotive and marine industries. The reason for this choice is due to the lightness, high specific strength and stiffness, good corrosion resistance and superior fatigue characteristics of such fibre reinforced composites [1]. However, the use of synthetic reinforced fibre composites may become a problem from a health and environmental point of view. Therefore, one of the typical challenges is to replace the synthetic fibres by renewable natural fibres extracted from plants [2, 3]. The matrix should also be eco-friendly. Apart from using natural renewable and recyclable resources of thermoplastic matrices, the plant fibre composites, or biobased-composites, possess noticeable properties such as high toughness, high shear and compressive strength, low density, low cost, non-abrasiveness and low impact on environment. These composites can also be tailored to provide other attractive properties for the automotive and aerospace industries, such as a good forming ability to process required large and complex structural parts [4, 5].

Among all plant fibres reinforced composites, the flax fibres offer interesting mechanical properties and appear as a green technological solution [6]. Usually, the unidirectional laminates are used for uniaxial loading. For more complex loading, woven fabrics are generally considered as the best choice due to their multi-axial performance [7, 8].

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In the past decades, the automotive industry had to deal with two major issues concerning these biobased-composites: i) the ability to process the composite parts both in one step process and reduced production time, ii) by keeping a good quality of obtained structures (resistance to mechanical loading, for example). Nowadays, only few manufacturing processes such as light resin transfer molding (light RTM) or thermal-compression are considered by this industry [9–11]. When using conventional heating equipment such as a hot press Picher-Martel et al. [12] showed that short carbon fibre/PEEK structures were successfully obtained by thermo-compression in about 40 min at 400 °C which is the melting temperature of the resin [12]. This processing time is far beyond the objectives of the automotive requirements and therefore the use of a conventional hot press equipped by electrical resistances for heating the assembly (mould and material) and the compressed air for their cooling cannot be satisfactory. Another review shows that RocTool company has successfully developed a moulding process by thermal compression in a short time and one operation step applied to large parts based on thermoplastic composites [13]. This technique consists in heating both mould (punch and die) and eventually the material by using an electromagnetic induction current and by cooling the mould by injecting cold water through specific channels in the mould.

The quality and the quantity of the products that can be produced in a range of time are the main parameters considered by industry. If one wants to accelerate the manufacturing process of composite parts, the level of property of the obtained part needs to be equivalent to the one manufactured using a "classical" process. As a consequence, the process parameters of the accelerated technique should be well adjusted because using un-optimized thermal compression process parameters may lead to major manufacturing defects such as voids, delamination and poor or rich regions of resin in the composite.

Natural fibres have distinct characteristics from synthetic fibres. Different authors [14–16] reported that the vegetal fibres have multi-scale and porous structures, whereas the transversal section of artificial fibres are round, regular (except carbon fibres) and usually have good mechanical properties. It is also reported that debonding at the fibres-matrix interface may be generated due to low adhesion of the polymer resin to natural fibres [17, 18]. All the defects mentioned above depend on the microstructure and the thermomechanical manufacturing history and contribute to decrease the mechanical properties of the composite material and consequently those of the composite structures [19, 20].

In order to manufacture homogenous flax fibre/thermoplastic matrix composites with low content of voids and high mechanical properties, answering industrial and financial needs, several manufacturing conditions must be met: These ones can be found in the literature and for example in the following references for hot press manufacturing [21–23] and in [24] for rapid manufacturing process using the RocTool system [13]. The main considerations to manufacture a high performance natural fibre composite consists in optimising the compression stress, the processing temperature and time and the heating and cooling temperature ramps so that to obtain parts with a minimum amount of voids and an adequate level of crystallinity.

Nowadays, automotive production rates (few minutes per part) together with good quality of complex parts are generally not performed as a one step process (one step of heating, one step of thermo-compression) or do not use comingled woven fabric as a semi-product material as it is the case for processes using pre-impregnated tapes [12].

In this study, an experimental forming thermal-compression device using temperature controlled air flow was developed at the LAMPA laboratory. It allows a one-step realization of V-shaped thermoplastic composite parts in less than 200 s. It consists of shaping a comingled flax / polypropylene (PP) fabric by using the thermal-compression process together with hot airflows used to reach temperatures above the melting point of the resin and then cold airflows to quickly cool the part.

The objectives of this study are:

(i) to present the specific fast manufacturing process;
(ii) to define the optimal process parameters;
(iii) to assess the reliability of this process by comparing the mechanical properties of composites obtained by the fast manufacturing process developed in this work with conventional manufacturing technique.

Materials and methods

Materials

In this work, two composite materials were used, based on flax/PP comingled fabric, with different weave architectures. These materials were kindly provided by DEPESTELE group (France). A 2 × 2 twill weave fabric (310 g/m^2, areal weight) and a satin weave fabric (280 g/m^2, areal weight) are considered. 300 tex rovings constitute the warp and weft flat yarns. For both fabrics, the mass fraction depends on direction: 39% and 61% for the warp and weft yarns respectively. For both materials, the volume fraction of fibers is equal to 0.5. The stacking sequences of individual layers follow the arrangements of $[0/90]_9$,. The number of layers was defined so that to obtain a 2 mm thickness for tensile tests.

Manufacturing processes

In this study, an experimental compression device with thermal airflow, shown in Fig. 1a, was developed at the LAMPA

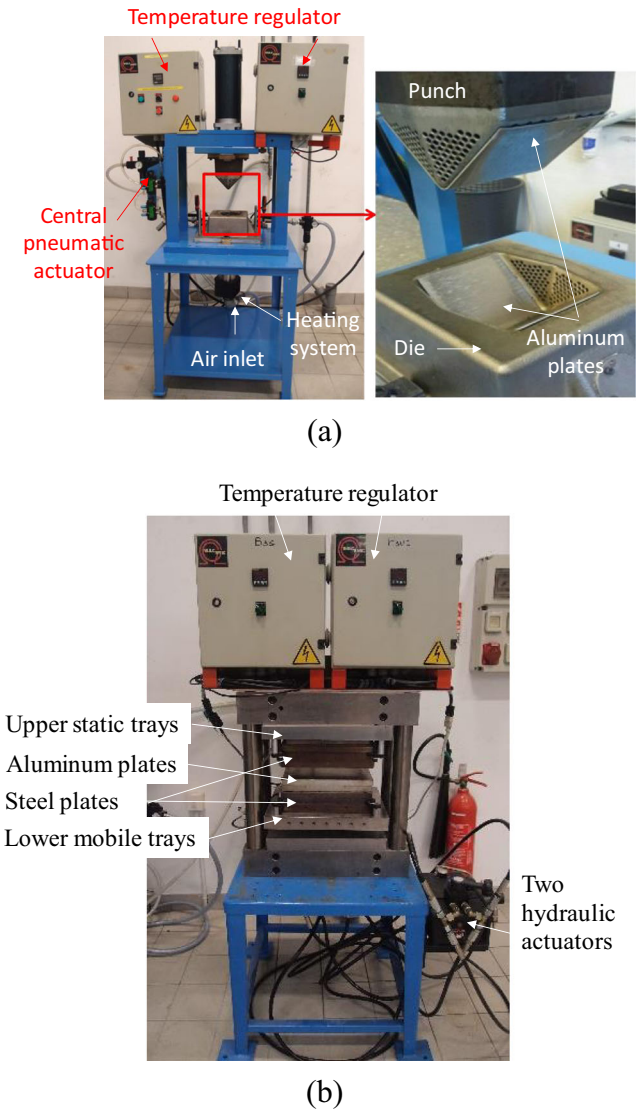


Fig. 1 Thermal compression process: (**a**) stamping airflow device and (**b**) conventional hot press

laboratory. The main objective of this device is to form and perform the thermo-compression process in a single and fast step. The idea is to form the desired number of comingled flax/polypropylene fabric layers (180×90 mm^2) above the melting point of the resin by holding the temperature at 200 °C for 30 s under a pressure of 6 bars. This pressure is not the one applied on the material but the pressure of the compressed air required by the hydraulic unit of the actuator. Hot and cold airflows for rapid heating and cooling are applied respectively. A mold with a V-shape was considered and parts were manufactured as presented in Fig. 1a.

The stamping system used in this study is composed of a punch/die commutable couple. The device was originally devoted to the pre-forming of powdered woven fabrics in the frame of the first step of the RTM process prior to injection and not for the composite part manufacturing. For that the system had a lot of holes allowing the hot air transit between the punch and die through the fabric in order to obtain homogeneous temperature. A central pneumatic actuator controls the movement of the punch and is used for varying the applied pressure. For composite manufacturing, in order to block the holes, aluminum plates are used to control both the homogeneous distribution of pressure and temperature.

The airflow is heated by electrical resistances and is maintained under a constant pressure for rapid heating (about 2 min) of fabrics located in the mold. This system is equipped with two temperature controllers. The first one controls the temperature of the material in the mold thanks to the thermo-couple located in the fabric during the process. The second one displays the air temperature at the outlet of the heating system.

Composite parts were manufactured by using the new developed device, described above. In order to compare the mechanical performances of these parts, composite plates were also manufactured, using a traditional thermal-compression hot press (Fig. 1b). The same comingled fabrics with the same stacking sequences were used to manufacture the composite plates ($250 \times 200 \times 2$ mm^3). The press is made of steel and aluminum plates. The dimensions are 300×250 mm^2, and they are located between lower mobile and upper static trays. These trays are heated using Joule effect. The temperature is controlled by thermocouples connected to the plates with upper and lower temperature regulators respectively (Fig. 1b). The lower mobile trays are guided by two hydraulic actuators which ensure the application of a sufficient pressure on the material placed between the two aluminum plates.

The thermal-compression cycle consists of holding the sample under a given pressure at 200 °C for 5 min (Fig. 1b). The pressure is not specified because the device only gives the pressure of the compressed air required by the hydraulic unit empowering the actuators. It can be noticed that the whole "classical" process takes a much longer time (about 140 min at total) than the one described as the innovative process. This is due to a longer heating (about 5 °C/min) and cooling (about 1 °C/min) cycles.

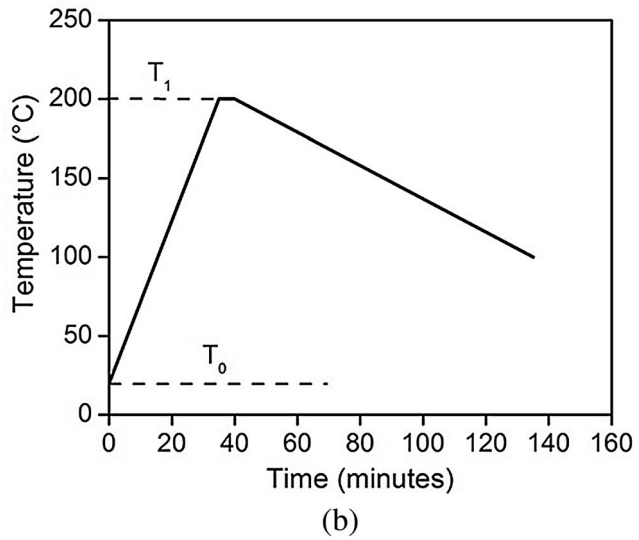
A graph of the conventional thermal cycle for a composite flax/PP plate is presented in Fig. 2.

Experimental methods

Tensile tests

To evaluate the quality of parts manufactured using the innovative fast process, the mechanical properties of $[0/90]_9$ flax/PP composite parts were measured and compared to the ones of composites manufactured by using a conventional hot press. Different process conditions were tested. A Zwick testing machine with a capacity of 100 kN was used. The principal strains ε_I and ε_{II}, used for determination of Young's Modulus and Poisson's ratio, were measured by both the

(a)



(b)

Fig. 2 **a** Composite plate manufactured by conventional hot press and (**b**) Conventional thermal cycle

biaxial strain gage glued on the samples and an extensometer. Each mechanical test was conducted using a cross-head speed of 2 mm/min. Six samples were tested for each manufacturing condition. The tensile test dimensions of samples manufactured by conventional thermal-compression follow the recommendations of the ISO 527-4 and ISO 527-5 standards. In contrast, the samples made using the forming airflow device were extracted from V-shaped parts and their dimensions ($90 \times 25 \times 2$ mm^3) could not follow the requirements described by the standards because the V-shaped parts are too small to allow extraction of standard size samples.

Microstructural observations

Micro structural observations were realized in order to evaluate the microstructure of the composites. An Axio Zeiss Optical Microscope (OM) was used for the optical observations in the cross section of the manufactured materials. Additionally, samples were cut in the cross section and coated with a thin layer of gold in order to be examined using a Zeiss Supra 25 Scanning Electron Microscope (SEM).

Density measurements

Measurements of bulk density and porosity were conducted to define the quality of the manufactured biocomposites. The density of the samples was determined using a milligram very accurate scale. Each sample was measured three times. The void content or porosity was determined using the equation below, following the Archimedes' principle in accordance with ISO 1183-1 standard:

$$v_p = 1 - \left(\frac{(1-\tau_f)\rho_c}{\rho_m} + \frac{\tau_f \rho_c}{\rho_f} \right) \quad (1)$$

where:

v_p is the void content, $\rho_{composite}$ is the volumic mass of the composite, ρ_f is the volumic mass of the flax fibers, ρ_m is the volumic mass of the PP matrix, τ_f is the fiber mass fraction.

Results and discussion

Optimization of the newly-developed thermal compression process

A preliminary work was realized on the newly developed thermal-compression device described in previous paragraph. Because the device was first designed for another application, the airflow is free to circulate through both the holes of the die and the side walls of the punch. A first batch of manufacturing tests was carried out. The variation of the stacking sequence from $[0/90]_6$ to $[0/90]_{10}$ was investigated in order to determine the limits of this process to obtain the desired composite shape. Figure 3 shows some geometric defects such as the prints of the mold holes, on the surface of the composite, due to the airflow conducting system or the heterogeneous

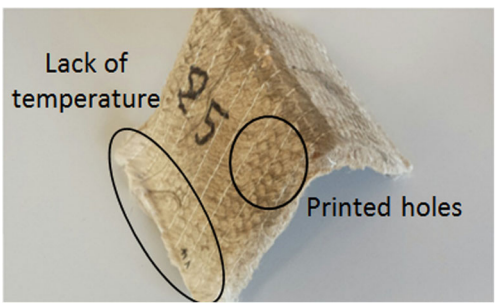


Fig. 3 Result of the preliminary tests

color of the composite due to non-homogeneous temperature and pressure on the surface of the parts.

Two thermocouples were then placed at the bottom and at the edge of the part during the process. A great difference in temperature was observed. To better understand this phenomenon, the mold was equipped with eight thermocouples at various positions (Fig. 4a). Once again, a maximum gradient of temperature about $\Delta T \approx 80$ °C was recorded between the bottom and the edge of the die of two different sides. This is explained by a non-homogeneous airflow: it is too hot at the bottom and too cold at the edges of the die. Fig. 4b shows, as an example, the airflow temperature evolution at 4 positions in the die over the 5 min of the heating phase. The temperature values measured at the bottom (positions 2, 4, 6 and 8) are higher than those measured at the edges (positions 1, 3, 5 and 7). Moreover, the gradient of temperature at each side is also different, as also the values measured at the bottom of the die. For this reason, positions 2 and 6, for example, show higher temperatures compared to those found at positions 4 and 8. This great difference of temperature is due to a poor distribution of hot airflow into the die since the output of the heating system is located just below the die. For this reason, the bottom of the die is hotter than the other positions. Fig. 4b also shows irregular temperature curves due to an inefficient temperature regulation system.

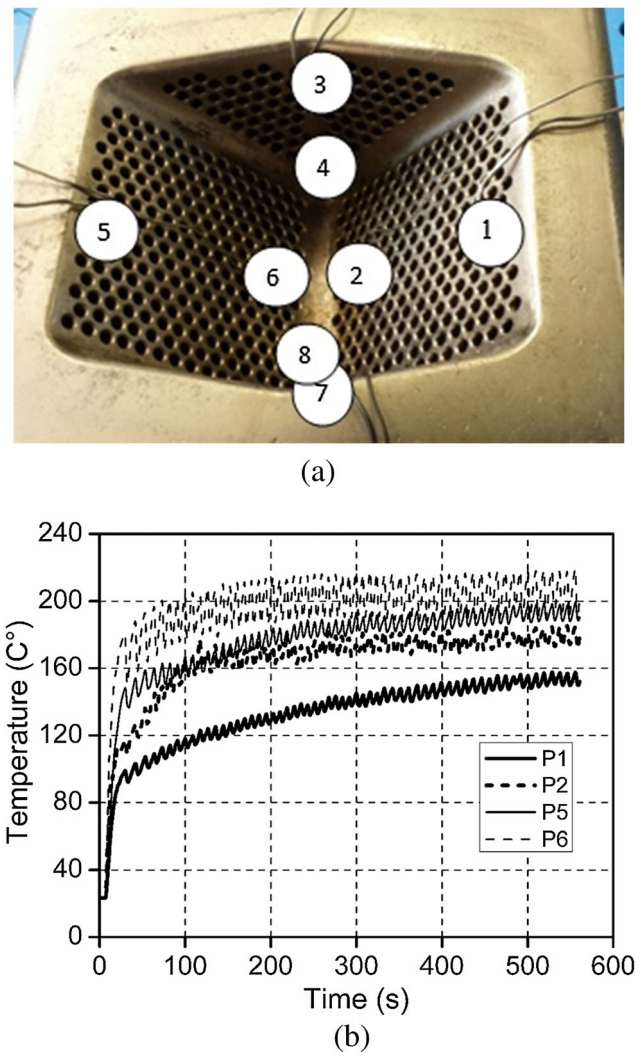


Fig. 4 Temperature distribution in the die: (**a**) points measured and (**b**) evolution of the temperature as a function of time for a few points

To solve the temperature heterogeneity issue, a more reliable temperature system was set up: aluminum plates of 1 mm thickness were placed on the surface of the mold to control the temperature distribution and prevent the holes prints on the specimen surface (Fig. 5a). Table 1 shows data of the optimization of the process parameters used after the die modification (addition of aluminium plates).

As shown in Table 1, the temperature of the airflow is the first parameter that was optimized. For the first tests, the air pressure at the inlet of the heater system was set at 2 bar for both thermal cycles. With this parameter, a good V-shaped composite part was obtained in less than 7 min. Then, in the second series of tests, the air pressure during the heating and cooling cycles was differently set. The rise of the air pressure, up to 3 bar, during the cooling phase has for consequence to increase the cooling rate from 1.1°Cs^{-1} up to 1.6°Cs^{-1}, whereas a decrease of the heating airflow pressure permits to reach the processing temperature faster during the heating stage. A parabolic decrease of the heating time is observed as a function of the decreasing air flow pressure (Fig. 5b). Figure 5b shows that heating the air under lower pressure (and lower flow rate) allows to reach the targeted temperature much faster. However, it implies a smaller quantity of air which is not sufficient to melt the resin throughout the entire mold. The low air flow travels at a low speed, and therefore the air cools before passing to the second part of the mold (punch side). At a higher flow rate, the air does not have the time to heat up very much and this leads to a slow heating rate of the composite. Tests at an air flow in the middle of the interval at about 2 bar allows the air heating rate to be increased from 1.0 °Cs^{-1} to 1.4 °Cs^{-1}. The heat is successfully brought up to the upper part of the tool without having much loss. This air flow rate and hence air pressure was therefore chosen as an optimum process parameter.

Finally, the time duration for which the plateau of temperature and pressure are applied was also optimized (Fig. 6a). This time was reduced to 30 s. Therefore, by taking into account all the optimization steps, the same V-shaped composite part was obtained in 200 s, twice faster than before optimization.

Figure 6b shows a 2 × 2 twill weave based V-shaped composite part manufactured using the newly developed thermal compression device associated to the optimized thermal and compression parameters. The overall shape analysis does not show any geometrical defects, and the surface color of composite is homogeneous. No apparent dry zones are visible on the surface of the part.

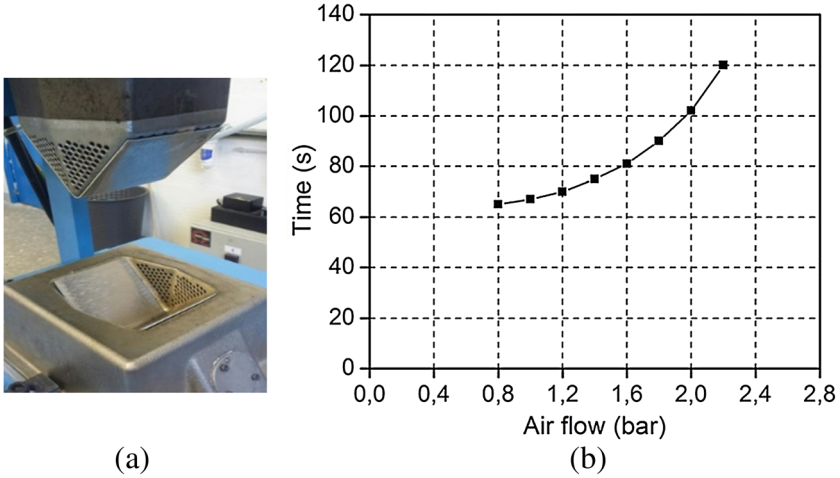


Fig. 5 **a** Optimized die configuration and (**b**) Heating time as a function of the air flow pressure

Tensile properties

Preliminary mechanical tests

As the samples manufactured by the newly-developed thermal-compression device do not have standardized lengths for realizing the tensile tests, special sample holders were designed and prepared for allowing the use of the length extensometer as shown in Fig. 7. This set-up permits to extend the sample length.

To validate the reliability of this set-up, to investigate the volume effect and to determine the tensile mechanical properties, a comparison was performed between two sample lengths (90 and 250 mm) of satin flax/PP composite with stack of $[0/90]_9$ manufactured by the traditional thermal-compression press. The section was the same: 25×2 mm^2. The short samples were tested using the extension holders (Fig. 7) and compared to the long standard samples. The standard initial length of long samples is 250 mm. The Fig. 8 shows the stress-strain behavior of both samples and it can be noticed that the behaviors are very similar. Table 2 summarizes the tensile properties of the tests performed. It can be noticed that no statistical difference (by performing Student's tests) were observed between properties measured from short and standard size samples. The size of the samples does not have any influence on the measured properties in this case.

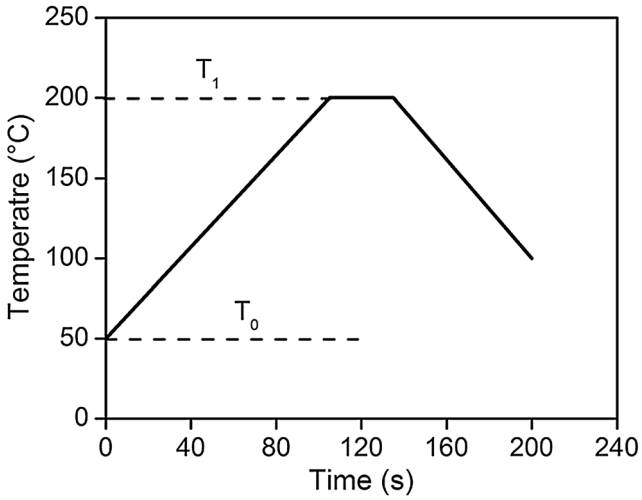
The results presented in Table 2 can be considered as our reference for comparison to the ones obtained for composites manufactured by using the newly developed equipment. The performance level of our reference material is located in the upper value of the ones reported in the literature [21–33] (Table 3) despite the differences in polymer matrices and fibers volume ratio.

Influence of the process and material parameters on the tensile properties

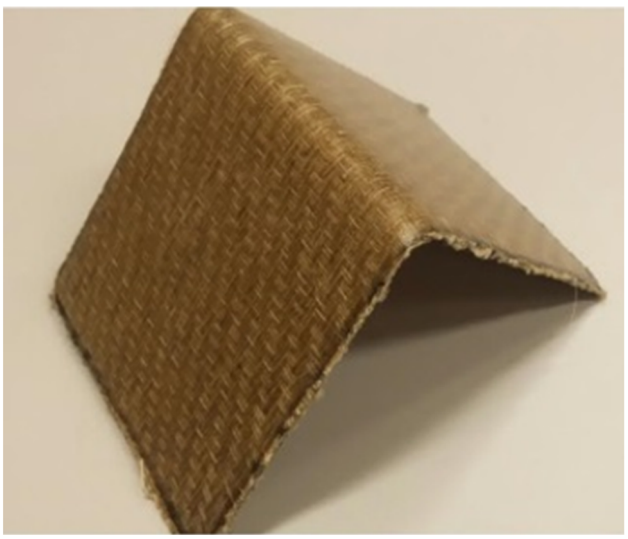
The main objective of the tensile test is to investigate the performance and the influence of process parameters on the tensile properties of the composites manufactured by using the newly-developed thermal-compression process in comparison to those obtained by using the conventional thermal-compression process.

Table 1 Evolutions of process parameters during the optimization

Test	N° of layer	Temperature target (°C)	Time (s)			Air pressure (bar)		Applied mechanical pressure (bar)	Manufacturing time (s)
			Heating	Holding	Cooling	Heating	Cooling		
1	9	200	130	240	120	2	2	6	490
2	9	200	130	240	65	2	3	6	435
3	9	200	120	240	65	1.8	3	6	425
4	9	200	115	240	65	1.6	3	6	420
5	9	200	105	240	65	1.4	3	6	410
6	9	200	105	120	65	1.4	3	6	290
7	9	200	105	60	65	1.4	3	6	230
8	9	200	105	30	65	1.4	3	6	200



(a)



(b)

Fig. 6 **a** Optimized thermal cycle and (**b**) Composite part manufactured by using the newly optimized process

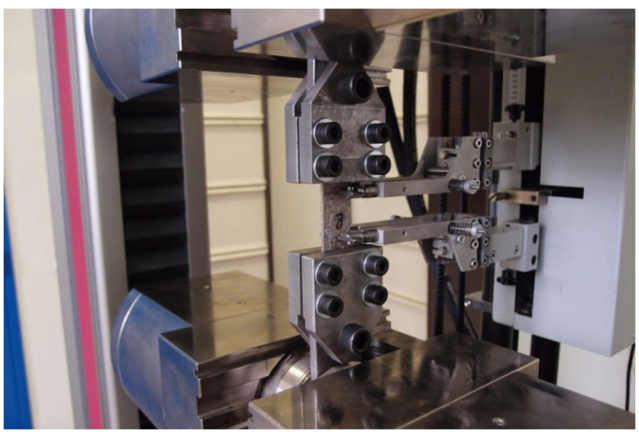


Fig. 7 Specially developed tensile test holders for non-standard samples

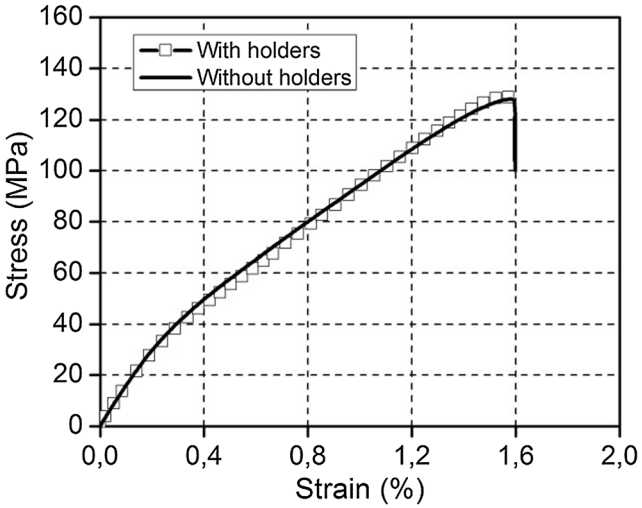


Fig. 8 Stress-strain curves of flax/PP composite with stack of $[0/90]_9$ made by thermo-compression press: influence of length's sample

Time, temperature and pressure are the main three parameters to take into consideration for the optimization of the thermoplastic based composite parts. The values of these parameters depend on fiber and polymer nature. It is a combination of the melting temperature and viscosity of the polymer and maximum temperature undergone by the fibers. Thus, to optimize the composite parts, each of the previously mentioned parameters was varied individually and separately, by keeping constant the two other ones as described below:

- 2, 4 and 6 bar of pressure at 200 °C for 30 s;
- 0, 30,60, 120, 240 s of holding time at 200 °C and 6 bar;
- 180, 200, 210, 230 and 250 °C under 6 bar for 30 s.

For each level of pressure and holding time, measurements of the material density and the amount of voids were considered as outcome parameters.

Density and void content The Fig. 9a–d and Table 4 show the results of composite density measurements and the ratio of voids as a function of the different pressure levels and holding times chosen. It can be observed that when the applied pressure rises, the density increases too, as expected. However, there is no observed difference in density and porosity amounts for each holding times considered at 200 °C. A slight

Table 2 Tensile mechanical properties of bio-composites manufactured by using a traditional hot press

Type of samples	Loading direction	E (GPa)	σ_{ult} (MPa)	ε_{ult}(%)
Standard samples	Weft	16.9 ± 0.3	136 ± 1	1.46 ± 0.02
Short samples	Weft	16.9 ± 0.3	133 ± 2	1.5 ± 0.10
Statistical difference (t-test)		No	No	No

Table 3 Tensile properties of composites from the literature similar to the ones studied in this work

Materials	Fiber content (%)	Tensile strength (MPa)	Young modulus (GPa)	Elongation at failure(%)	Reference
Flax/epoxy UD	50	311 ± 14	31 ± 1	1.5 ± 0.1	[25]
Flax/epoxy (laminate[0/90])	43.1	170	14.5	1.72 (0.13)	[26]
Flax/epoxy(UD)	45	218 ± 8	23.52 ± 0.30	–	[27]
Flax/epoxy(UD)	32	132 ± 4	15.0 ± 0.6	1.2	[28]
Flax/PLA(UD)	19.5	99 ± 5	16.0 ± 0.80	0.07	[29]
Flax/PP(MAT)	36.5	73	7.9	1.6–1.7	[30]
Flax/PLA(UD)	44.4	339 ± 22	20.1 ± 1	–	[31]
Flax/PP(UD)	72	321	29	–	[32]
Flax/PP(UD)	30	89/70	7/6	–	[33]
Flax/PP (UD)	50	40	7	–	[34]
Flax/PLA (twill 2*2)	40	72.2 ± 2.0	13.0 ± 0.9	1.5 0 ± 0.08	[35]
Flax/vinylester (2D fabric)	34	76.7	9.1	–	[36]
Flax/PP (twill 2*2)	50	92.42	10.71	2.08	[37]

difference was observed for the case without plateau (holding time 0 s). This means that a minimum holding time is required, but it is not a predominant parameter which could influence the density of the material compared to the pressure.

A good correlation between the density and the pressure evolution was observed. The composite manufactured with the lowest applied pressure exhibits the lowest density (1.05 g/cm^3). A significant increase in density about 3.8% and 9.5% respectively for 4 bar and 6 bar of applied pressure,

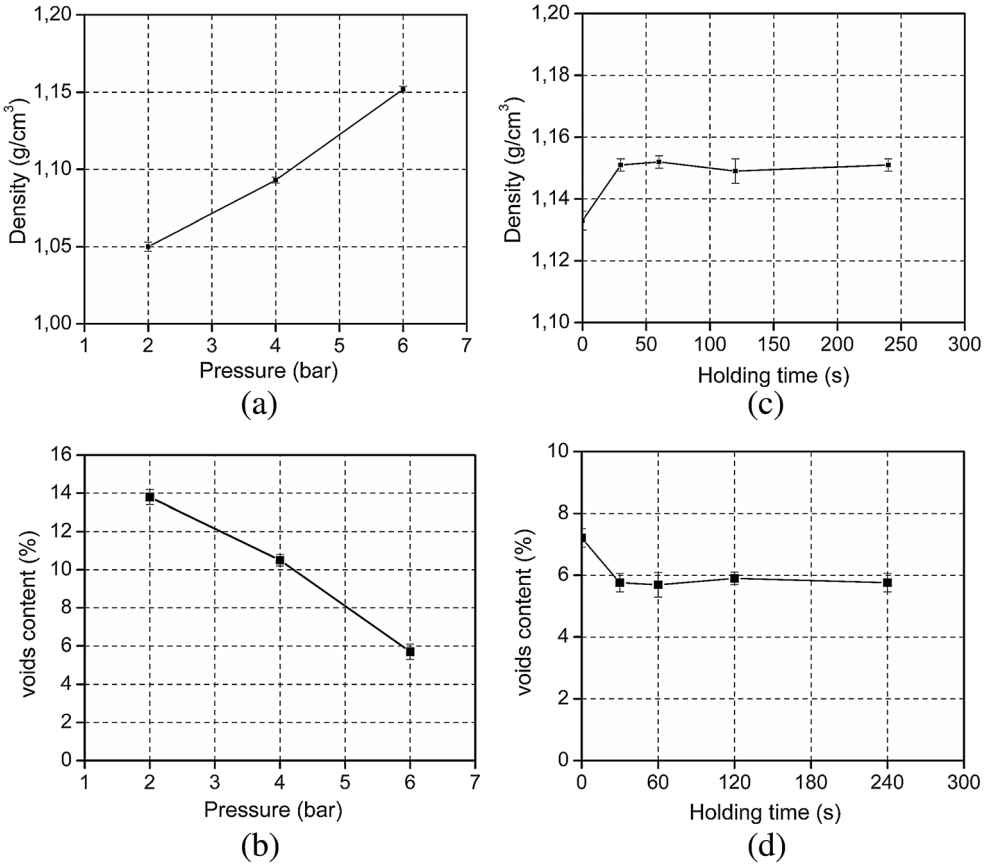


Fig. 9 **a** Density vs pressure and (**b**) voids content vs pressure at 200 °C and 30 s of holding time; (**c**) density vs holding time and (**d**) voids content vs holding time at 200 °C and 6 bar of pressure for flax/PP composite satin based fabric. Six samples were tested for each manufacturing condition

Table 4 Density and voids content of composites as a function of pressure and holding time levels

Parameter	Level	Density g/cm^3	Voids content %
Pressure (bar)	2	1.051 ± 0.003	13.85 ± 0.4
	4	1.093 ± 0.002	10.51 ± 0.3
	6	1.152 ± 0.002	5.70 ± 0.4
Holding time (s)	0	1.133 ± 0.003	7.21 ± 0.3
	30	1.151 ± 0.002	5.76 ± 0.3
	60	1.152 ± 0.002	5.69 ± 0.4
	120	1.149 ± 0.004	5.9 ± 0.2
	240	1.151 ± 0.002	5.76 ± 0.3

was measured. Fig. 9b presents the evolution of the voids content as a function of the applied pressure. A linear relationship between pressure and voids content was obtained. With each increase of pressure, a drop of the voids content takes place. The density evolution as a function of the applied pressure is explained by the presence of voids (porosity) due to debonding at the fibers-matrix interface, between fibers in the yarn and finally by the porosity due to air trapped within the material during the manufacturing phase of composite. According to [38–43], the voids formation is mainly due to the air trapped between the layers during the manufacturing process, but also due to the moisture absorbed during the material storage, moisture dissolved in the resin and volatiles released by chemical reactions.

To better understand the density changes observed in the composite parts, representative sections of composites were examined using optical microscopy. Fig. 10 shows the debonding phenomenon at the fibers-matrix interfaces (Fig. 10a) and within the fibrous yarns in the composite structure obtained by different pressures (Fig. 10b). At low applied pressures the debonding mainly takes place between the flax fibers within the yarns and at the interface between the yarn and the matrix (Fig. 10a–c). However, when the applied pressure is increased, very few debonding is observed inside the flax yarns and at the yarn-matrix interface. It is reasonable to think that the trapped air is expelled at high applied pressures and then filled by resin. It is also easier for the resin to flow into the flax yarns, but due to the fabrics structures, small interfacial debonding may persist into the yarns.

In addition, the residual thermal stresses may also participate to the creation of debonding mechanism in thermoplastic composites. The difference between the thermal expansion coefficients of the constituents of the composite could possibly create defects at the fibers-matrix interfaces and in the fibers yarns. The contraction of the matrix, larger than the one of flax fibers, during the cooling phase can lead to the debonding initiation or growth. As shown in Fig. 10 the porosity is almost suppressed at a higher pressure of 6 bar, but interfacial debonding in the fibers yarns are always present within the material with a level of about 5.7%.

Fig. 11 also shows SEM images of porosity of different sizes and shapes within the samples. A significant reduction in their level, size and shape, as well as the same interfacial debonding is observed when the pressure is increased. The porosity shape also changes from elongated to spherical due to the increase of this pressure (Fig. 11a–c). For similar investigations [37, 43], observed that the porosity was flat and elongated at low applied pressure and spherical for higher ones. This should have for consequence to increase the level of the mechanical properties as stress concentration is not as high in spherical porosity than in elongated ones.

The Fig. 12 shows the optical micrographs of PP/flax composites for different holding times using the newly-developed manufacturing process. Once again, the interfacial debonding within the fibrous yarn is observed when no pressure holding time is considered (Fig. 12a). A better quality of the interface cohesion is observed from 30 s holding time. It should also be mentioned that the optical micrographs emphasize the debonding interface at fiber bark and matrix, which is not observed with SEM analysis (Fig. 12e). Some residual pieces of bark that were not removed during the preparation of the yarns may be observed between technical fibers (fiber bundles). The debondings observed at the interface between remaining barks and the polymer matrix suggest that the preparation of flax fiber polymer composite should lead to bark free technical fibers. This point should be investigated into more details in future works so that to quantify the impact of residual barks (for different levels of bark quantities) on the quality of the fiber/ matrix interface and therefore, on the mechanical properties.

Stress-strain curves Figure 13 shows the influence of pressure and holding time on the tensile behavior of flax/PP satin based fabric composites manufactured using a plateau temperature of 200 °C at different holding times. All these composite samples were loaded in the direction of the weft yarns.

A common similar stress-strain evolution for the different applied pressures is observed with two linear portions (Fig. 13a). Visual differences between the stress-strain curves due to the pressure levels applied during the manufacturing process can be noticed. As summarized in Table 5, the mechanical properties of the manufactured composites rise by increasing the applied pressure. The best mechanical properties were obtained with the highest pressure plateau of about 6 bar. Low porosities (5.7%) are observed. Lower tensile Young's modulus and strength were obtained with the lowest pressure plateau (2 bar). A void content of about 13.8% is observed in this case. An increase in mechanical properties was then observed with a medium range pressure of 4 bar. This is directly linked to a reduction of the void content (0.5%). In summary, an insufficient holding pressure leads

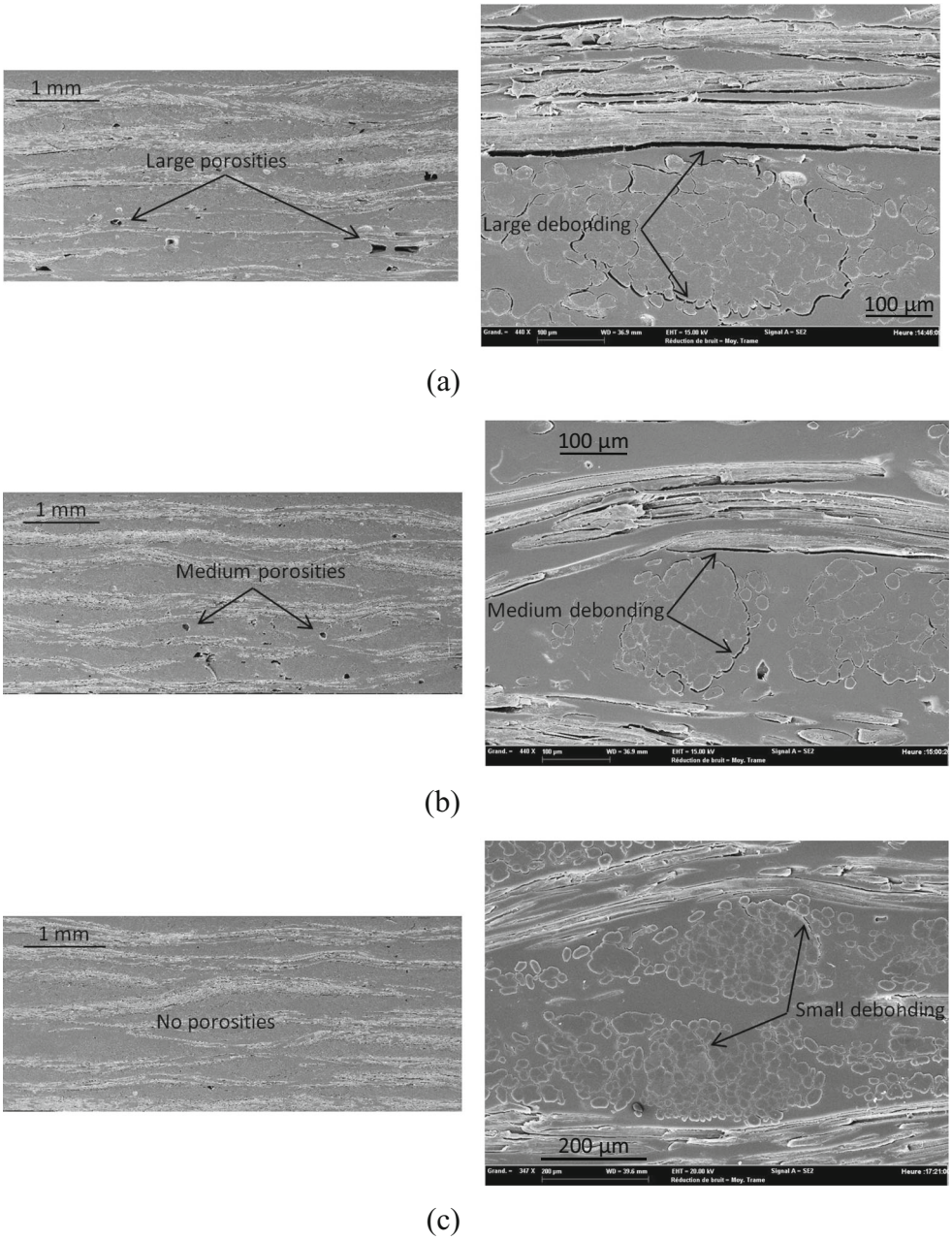


Fig. 10 Microstructures for different applied pressures at 200 °C and 30s of holding time: (**a**) 2 bar; (**b**) 4 bar and (**c**) 6 bar for flax/PP composites satin based fabric

to a very porous material with relatively poor mechanicals properties, as expected.

However, in the case of temperature holding time at 200 °C and under an applied pressure of 6 bar, no significant behavior difference was observed for each holding time from 30 s to 240 s (Fig. 13b and Table 5). This could be explained by the fact that 30 s are sufficient to melt the resin at 200 °C under 6 bar of pressure. Moreover, when no temperature and pressure plateau are applied (0 s), a small decrease of the mechanical proprieties was confirmed.

Temperature also has a great influence on the quality of the materials produced. It requires a precise identification of its optimum value in order to obtain the lowest resin viscosity without degrading the lignocellulosic fibers. In general, vegetal fibers do not support high processing temperatures (generally not higher than around 200 °C). It is possible that the degradation of natural fibers may occur before the resin melts when the processing temperature is too high. More specifically for flax fibers and polypropylene resin, there is a low difference between the resin melting temperature and the temperature at which fibers degradation could occur. In general, a compromise needs to be found in order to get a good impregnation without fibers degradation.

Temperature is the third main parameter that is considered in our study. As presented in Fig. 14, five different holding temperatures were applied for manufacturing the parts with

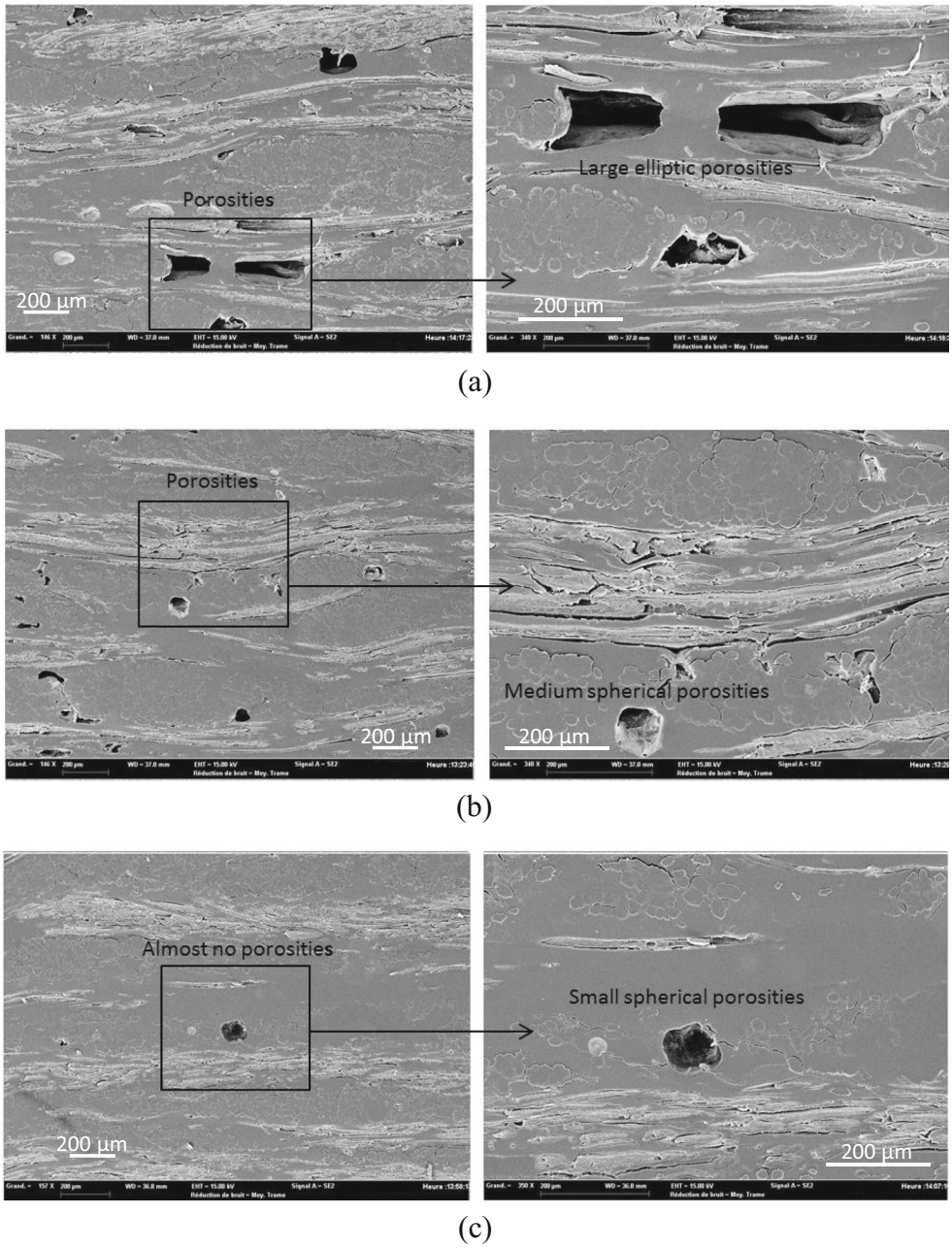


Fig. 11 Evolution of porosity shape and size for different applied pressures **at 200 °C and 30s of holding time** for flax/PP composite satin based fabric: (**a**) 2 bar; (**b**) 4 bar and (**d**) 6 bar

the newly developed device. The tensile properties of these parts are also shown in Table 5. For temperatures higher than 200 °C, a clear modification of the tensile behavior was observed with lower maximum stress and strain. It probably underlines a modification of the composite nature. Unlike Young's modulus, a significant progressive decrease of the ultimate strength is observed at manufacturing temperatures from 200 °C to 250 °C. This could be explained by the degradation of the vegetal fibers as explained previously [40]. Identically, the resistance of the elementary fibers of ramie is reduced by 40% after holding them for one hour at 200 °C [44]. Gourier et al. [45] found a low variation in Young's modulus and ultimate strength of flax fibers at temperatures lower or equal to 190 °C and a strong degradation of mechanical properties at a temperature of 250 °C. A number of authors explained this significant decrease of mechanical properties at high temperatures by the evolution of the fiber cell wall structure and organization of plant fibers [45]. These modifications may be caused by transition phenomena within the polymers which constitute the cell walls of the plant (cellulose, hemicellulose and pectin). A thermal cycle of 250 °C involves modification of the parameters responsible for the fibers stiffness such as the biochemical composition and macromolecular arrangement. In addition, for temperatures lower than 200 °C, a reduction in the ultimate strength was observed (Fig. 14). For example, the composite manufactured at 180 °C

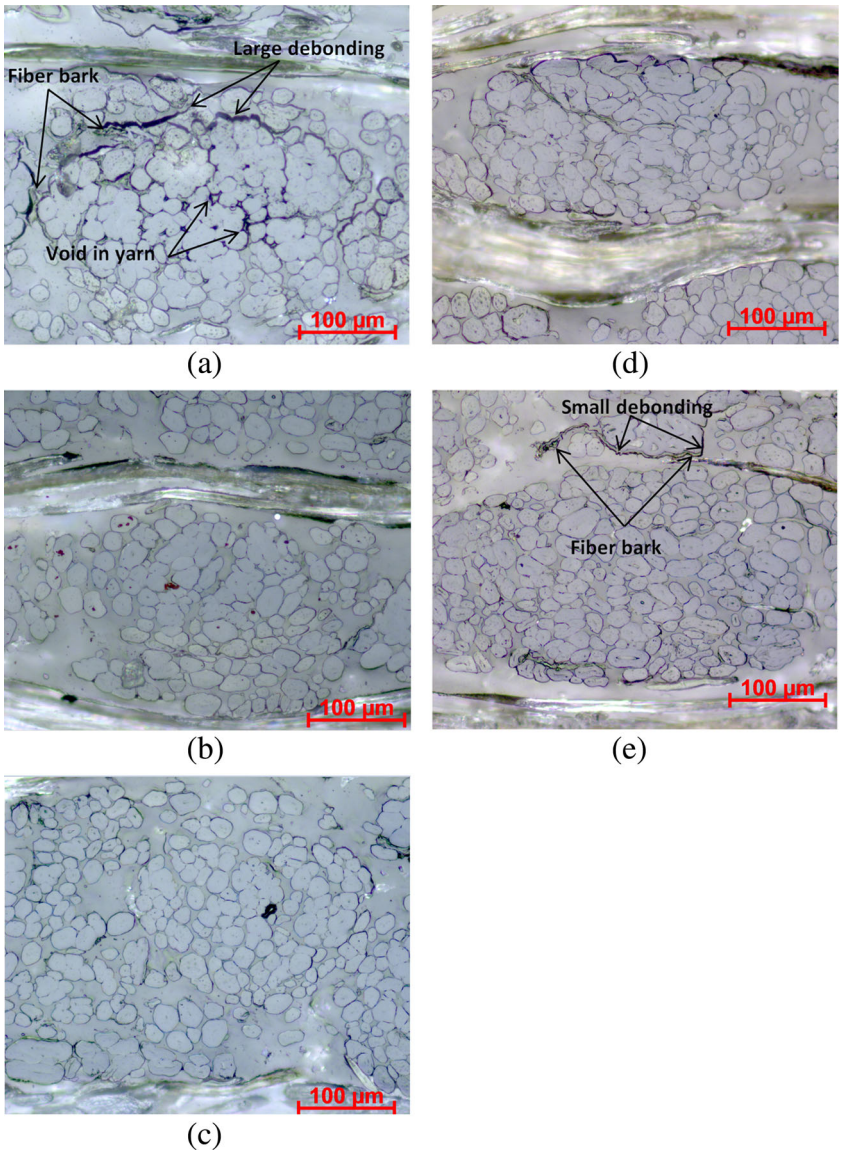
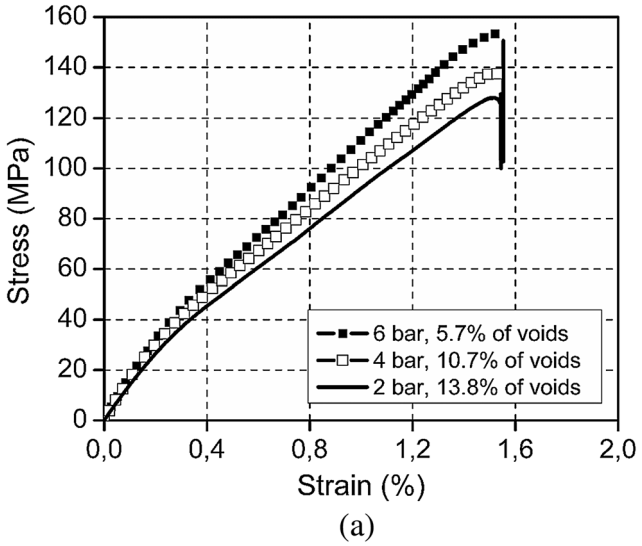


Fig. 12 Microstructures for different holding times at 200 °C and 6 bar of pressure for flax/PP composite satin based fabric: (**a**) 0 s; (**b**) 30s; (**c**) 60; (**d**) 120 s and (**e**) 240 s

for 30 s loses about 9% of its ultimate strength in comparison to the one manufactured at 200 °C for 30 s. This shows the lower limit of the optimum temperature. Actually for our process this temperature is not sufficient for a very short holding time (30 s) and a large material thickness. This decrease of mechanical properties is a consequence of the difficulty for the resin to well impregnate the fibers within the yarn in a fast time. To decrease the viscosity of the polypropylene a higher temperature than the strict melting temperature is required to enable the resin to flow in the interfacial voids. The best mechanical properties were obtained for a short holding time (30 s) with an optimal temperature of 200 °C. An increase in the compaction temperature (from 180 °C to 200 °C) leads to good mechanical properties by increasing the interface resistance of the composite. For similar materials, the best strength value was encountered for a processing temperature of 180 °C for a jute/copolyester Biopol composite [46] or 200 °C for a flax/polypropylene composite [47]. A previous study [48] showed that the melting temperature of all-PP unidirectional laminates was exceeded by 20 °C without any significant effect on the tensile modulus or the strength of the laminates. In this study, our results confirm that a temperature higher than that of polypropylene melting point and lower than the one leading to severe degradation of the flax fibers (200 °C) is recommended. As it was showed by [24], the degradation of flax fibers occurring during high temperature manufacturing depends on combination of temperature and holding temperature time. In our case, a temperature of 200 °C combined to a fast holding temperature and pressure was found to be a good compromise to manufacture flax fiber based bio-composite parts with the minimum amount of defects and the best mechanical properties.

Finally, as our process is not conventional, the influence of the fast cooling on the mechanical properties was also investigated. Fig. 15a–c respectively show the tensile behavior, the



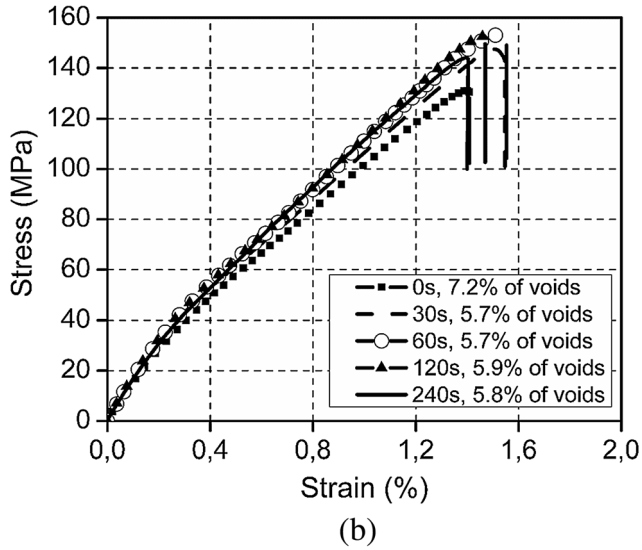


Fig. 13 **a** Pressure at 200 °C and 30s of holding time, and (**b**) holding time effect at 200 °C and 6 bar of pressure for the satin weave composite tested on the weft direction. The composite was made using the newly-developed thermal-compression process

evolution of the cooling cycles and the voids content obtained for each cooling cycle.

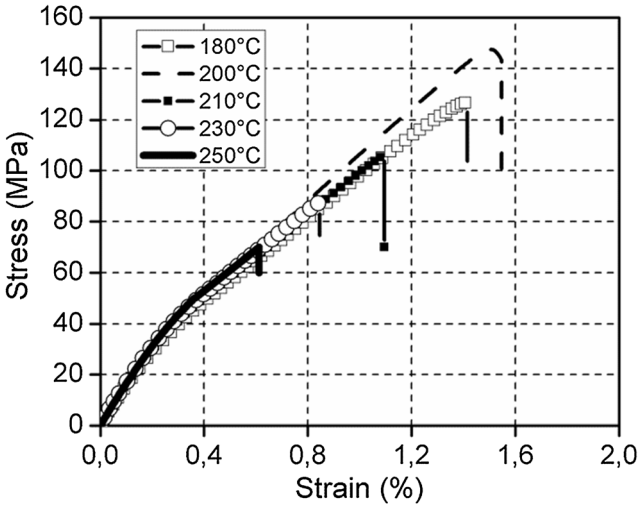


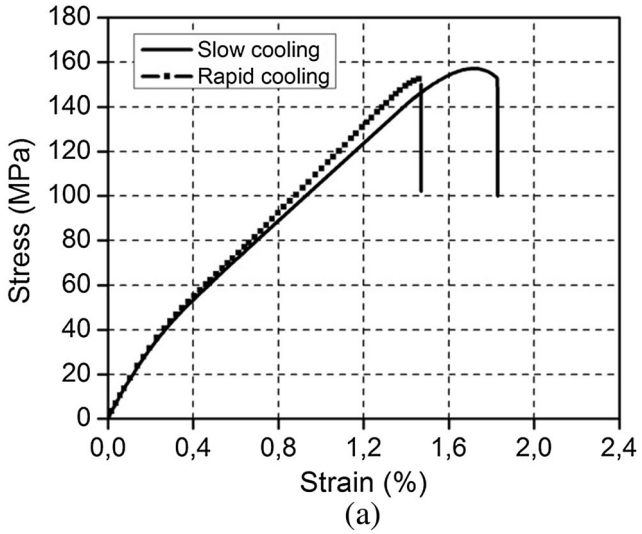
Fig. 14 Influence of processing temperature on the mechanical behavior (6 bars of pressure, 30s of holding time)

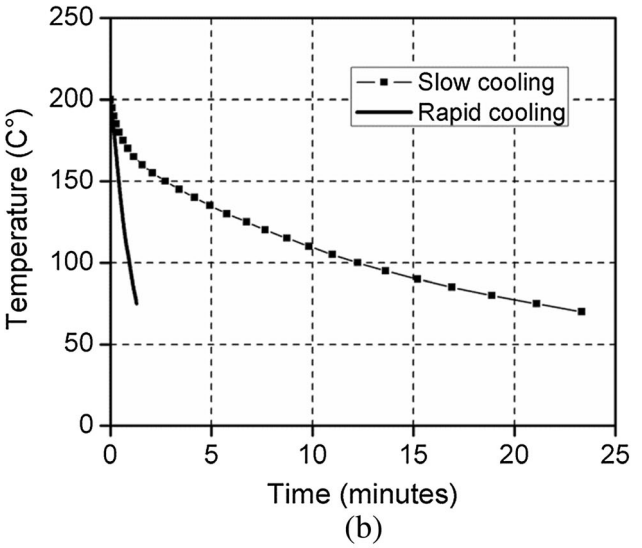
A slight decrease in tensile mechanical properties of 0.8 GPa, 5 MPa and 0.27% with an evolution of 4.8%, 3.4% and 18.2% respectively for Young modulus, ultimate strength and maximum strain was observed when the fastest cooling rate was used. Fig. 15b shows measured values of the temperature evolution in the material during the cooling phase for both open air and fast cooling. An almost linear cooling rate for faster cooling was obtained. For the second cooling cycle, a hyperbolic evolution of the temperature begins with a rapid drop of 30 °C followed by a slower drop to ambient temperature. These two completely different cooling cycles could explain the slight difference between the mechanical properties found as the levels of crystallinity are probably different in the PP semi-crystalline matrix. Moreover, Fig. 15c shows that the cooling rate has a low effect on the voids content. Therefore, the possibility to realize the V-shaped parts at the fast cooling rate without any significant decrease of the mechanical properties is of particular relevance and interest for the automotive industry, always seeking to reduce the manufacturing cycle times.

Finally, to conclude on the level of properties of the composites obtained by using the optimized fast process, Fig. 16 shows the tensile behavior of composites manufactured by the

Table 5 Tensile properties of composites for different process parameters

Loading direction	Holding temperature (°C)	Holding time (s)	Pressure (bar)	E (GPa)	σ_{ult} (MPa)	ε_{ult} (%)
Weft	200	0	6	15.4 ± 0.8	128 ± 8	1.46 ± 0.07
	200	30	6	16.6 ± 0.1	146 ± 2	1.48 ± 0.07
	200	60	6	16.9 ± 0.4	141 ± 8	1.43 ± 0.08
	200	120	6	16.5 ± 0.6	144 ± 5	1.45 ± 0.05
	200	240	6	16.4 ± 0.4	145 ± 4	1.43 ± 0.06
	200	30	4	16.2 ± 0.3	138 ± 5	1.5 ± 0.1
	200	30	2	13.7 ± 0.7	126 ± 4	1.5 ± 0.1
	180	30	6	15.4 ± 0.8	127 ± 9	1.46 ± 0.07
	210	30	6	15.9 ± 0.3	102 ± 12	1.1 ± 0.1
	230	30	6	16.6 ± 0.3	95 ± 13	1.0 ± 0.2
	250	30	6	16.5 ± 0.3	71 ± 16	0.62 ± 0.02





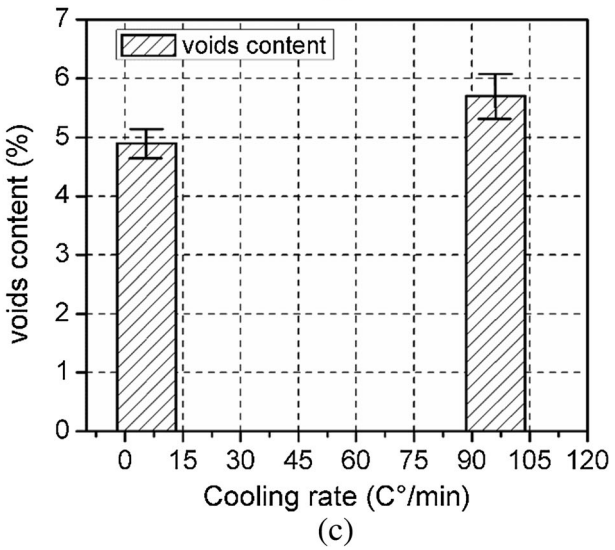


Fig. 15 (**a**) Tensile behavior; (**b**) cooling cycles and (**c**) Voids content

newly optimized process and the conventional one using a hot press. The composites manufactured by the newly developed device for which the whole processing time takes only 200 s exhibit higher strength and strain than the ones manufactured by using a conventional hot press. In that case the processing time was 140 min but it could be lower using specific devices to cool down the mold and the part. This is probably due to the fact that the fibers exposition to temperatures above 180 °C is longer than with the newly developed process (Table 6).

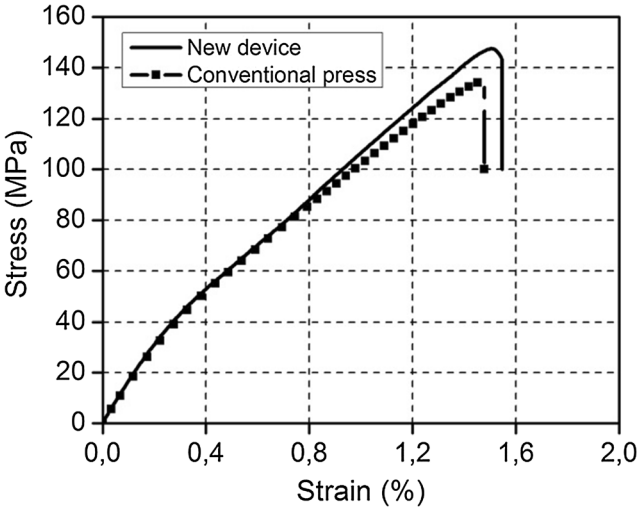


Fig. 16 Comparison of mechanical properties of satin PP/flax composites in the direction of weft strand obtained by two considered processes

These last results therefore indicate that the newly developed process also permits to increase the mechanical properties of the manufactured composites even for a fast processing time.

Conclusions

The main aim of this study is to reduce the manufacturing time of composite parts based on a comingled flax/polypropylene fabric. A single processing step is considered. To reach this goal a new rapid thermal compression device was developed. V-shaped parts were manufactured in less than 200 s. This is a huge improvement if one compares this time to classical thermal-compression cycles which take about 140 min. The process parameters were individually optimized through parametric studies. The best compromise for temperature, holding time and pneumatic pressure are respectively 200 °C, 30s and

Table 6 Tensile properties of composite parts made by the newly optimized and conventional thermo-compression processes

Process	Material	Loading direction	E (MPa)	σ_{ult} (MPa)	ε_{ult} (%)	V
New device	Satin	Weft	16.6 ± 0.1	146 ± 2	1.48 ± 0.08	0.12
Conventional press	Satin	Weft	16.9 ± 0.3	133 ± 2	1.5 ± 0.1	–

6 bar in order to favor a good impregnation with the lowest amount of porosity (even for short holding time of the temperature and pressure plateau) and this without thermally degrading fibers, hence the mechanical properties of the flax fibers. The cooling rate was also assessed and low differences where observed between the fast and slow cooling rates applied in this study. The mechanical properties from samples extracted from composites manufactured using the fast device were very close to the ones of equivalent parts obtained using classical hot press compression molding, therefore validating the new fast process.

In the future, the fast manufacturing device may be improved by heating and cooling both sides of the tool separately (punch/die). This would allow the manufacture of much more complex parts and also to decrease even more the manufacturing time. Subsequently, this technique could be used in the automotive industry at a larger scale and for bigger parts.

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Compliance with ethical standards We confirm that this work is original and has not been published elsewhere nor is currently under consideration for publication elsewhere.

Conflict of interest The authors declare that they have no conflict of interest.

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