Improvement of Natural Fiber Composite Materials by Carbon Fibers

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ABSTRACT: The purpose of this work is the improvement of flax fiber-reinforced composites obtained by vacuum molding in order to encourage their insertion into industrial products. The relatively high degree of porosity in these kinds of composites, due to the lack of compatibility between epoxy matrix and flax fibers and the hydrophilicity of flax fiber, remains a major constraint to their use in the industrial world. Hence, we have used a combination of carbon fibers with those of flax in order to optimize the properties of the assembly. Several stacking sequences have been tested in order to analyze the influence of the addition of carbon fibers on the water recovery behavior and the mechanical tensile behavior according to their position inside the laminate. It has been shown that adding surface carbon plies presents a barrier effect for the water sorption by limiting the creation and the percolation of internal porosities. The same effect is observed on tensile behavior where stacking sequences with external carbon fiber plies are more resistant than stacking sequences with internal carbon plies.

KEYWORDS: Hybrid composite, flax fiber, natural fiber, reinforced composite

1 INTRODUCTION

Natural fiber-reinforced composites (NFRCs) are attractive to the scientific community as well as the industrial world. Due to their relatively low densities, biobased origins, low environmental impact and availability as co-products of agricultural production, natural fibers can be serious candidates for strengthening polymeric matrices [1–4]. Numerous scientific and technological barriers must be removed before considering their widespread use. Natural fibers are known to be relatively difficult to properly impregnate with polymeric matrix. Vegetal fibers generally have a hydrophilic behavior whereas thermoplastic or thermoset polymeric matrixes are generally hydrophobic [5, 6]. Consequently, poor impregnation of natural fiber fabrics results in a high porosity rate, which could be detrimental to NFRC properties. Furthermore, in order to enhance an industrial tool already in place, most natural fiber fabrics come from the textile sector. These textile fabrics are made with twisted yarns of short natural fibers instead of continuous fiber strands as

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for artificial fibers (glass, carbon aramid, etc.) [7]. The advantage of this approach is the use of existing industrial weaving tools. The disadvantages are a low compressibility of textile fabrics and a low fabric fill rate, which is defined as the orthogonal projection of the fabric divided by the total area. The fabric fill rate plays a crucial role in the composite porosity appearance [8, 9]. A high porosity rate in NFRC will have several harmful consequences for the properties used in structures incorporating this kind of material. First of all, the mechanical properties are below expectations [10]. Indeed, internal cohesion between the reinforcement elements is poorly provided by the porous matrix and, moreover, porosities can be the initiation source of defects propagating all over the composite structure [9]. Secondly, resistance of the composite structure to the external environment is reduced with porosities [11]. Fluids, gas or liquids can easily penetrate the material and thus lead to accelerated aging or loss of structure sealing [8, 12–16]. Nowadays, several ways are being explored to solve the porosity problem in NFRC. Some new polymeric matrixes, biobased or not, having a chemical affinity with natural fibers are being developed [17]. In order to increase fiber/matrix computability, another explored possibility is the chemical treatment of flax reinforcement fabrics during



yarn spinning or fabric weaving [8, 18, 19]. Finally, some well-chosen composite operating conditions can address this problem and reduce the porosity formation [9]. The purpose of the research work presented here is to investigate the effect of the hybridization of NFRC by incorporation of carbon fiber fabrics into the stacking sequence on the final composite properties. There has been a remarkable review of the concept of hybrid composite by Swolfs *et al.* [20], and Sanjay et al. compiled a fairly exhaustive review focused on hybridization of natural and artificial fibers, especially glass fibers [21]. Hybrid composite is defined as a matrix containing at least two different types of reinforcing fibers. Hybridization can be made at yarn or strand level by mixing fibers of different origins, at layer level with ply made of different yarns or strands, and at stacking sequence level by alternating different plies. Until now, hybridization of natural fibers has been tested and studied with another natural fiber or with glass fiber, and hybridization of natural fiber with carbon fiber is less common.

All the reviewed papers have concluded that hybridization of natural fibers with glass fibers is useful to improve NFRC properties. As an example, hemp- and sisal-reinforced polypropylene hybridized with glass fibers has higher flexural, impact and water resistance, compared to pure hemp or sisal fiber composites [22, 23]. Zhang et al. studied the role of the stacking sequence of different natural fibers/glass fiber-reinforced hybrid composites [24]. They concluded that the stacking sequences have no effect on the tensile modulus, but are preponderant for the failure of mechanical properties such as fracture toughness, tensile and interlaminar shear strengths. The best results are obtained with composite with alternate stacking sequence [Glass-Flax]_{4s}, compared to [Glass2-Flax2], and [Glass4-Flax4]. Similar results have been obtained by Sabeel et al. [25], Amico et al. [26] and Khalil et al. [27] for various hybrid composites.

Concerning hybridization of carbon and natural fibers, there are fewer articles in the literature. Assarar *et al.* used the concept of hybridization of carbon and flax hybrid fibers to obtain composite material involving structural rigidity and vibration damping [28]. Bagheri *et al.* showed that incorporation of flax plies improves the fatigue strength of carbon composite used for bone fracture plate applications [29]. Dhakal *et al.* studied the effects of external carbon plies on flax composites [30]. They demonstrated that external carbon plies have a beneficial effect on water sorption and mechanical properties.

It appears that hybridization of natural fiber layer with synthetic fiber layer is an effective and powerful method to combine their respective properties. Hybridization of carbon and flax fibers can be

considered from two aspects. First of all, flax fiber can be added to carbon fiber-reinforced composite (CFRC). The goal is then for the flax fibers to bring some new added value as vibration damping, partially biosourced material, without degradation of the technical performance of CFRC. The second way is to consider the addition of carbon fibers on NFRC in order to improve the NFRC's technical properties. In this work, we were rather oriented to studying the hybridization with carbon fibers which could be efficient to limit the sensibility of flax fiber-reinforced composite (FFRC) to moisture and environmental stresses and to reinforce FFRC mechanical properties. Composites with various stacking sequences made of flax and carbon fiber fabrics were manufactured. Influence of the carbon plies position in the stacking sequence on the hybrid composite performance was studied, focusing on the microstructures, water sorption, and mechanical behavior under tensile solicitations.

2 STUDIED MATERIALS AND PROCESSING METHODS

2.1 Carbon Fiber Fabric and Flax Fiber Fabric

The flax cloth used is marketed by Flax Technic under the trade name Twinflax 2D 235. It is a flax fiber woven fabric with a basis weight of 235 g/m², using noncoated twisted strands (Figure 1), while the carbon cloth is a 2/2 twill, with a 200 g/m² basis weight.

2.2 Epoxy Matrix

The thermosetting polymer matrix is a mixture of an epoxy resin with a hardener. The epoxy resin, marketed under the name of Araldite LY 1564, was developed by the Huntsman company especially for impregnated structures and has a viscosity between 10 to 20 mPa.s at the temperature 25 °C and a density of 1.1 to 1.2 g/cm³. The amine hardener developed by the same company



Figure 1 Optical photographs of the microstructure of flax fabric before impregnation.

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under the name Aradur 3487 has a viscosity between 30 to 70 mPa.s at the temperature 25 °C and a density of 0.98 to 1 g/cm³. The optimal mix proposed by the manufacturer is 100 parts by weight of Araldite LY 1564 to 34 parts by weight of Aradur 3487 hardener.

2.3 Composite Manufacturing

A composite plate is made of a total of eight plies of tissue. Plates of 8 flax plies, called 100% flax, were first carried out and served as a reference to determine the intrinsic properties of a laminate ply. Different stacking sequences have been tested mixing 4 plies of flax and 4 plies of carbon; all stacking sequences comply with mirror symmetry ([C C F F], [C F C F], [F C F] C].). For each stacking sequence, several plates have been manufactured. We have selected the plates that have the same fiber volume fraction in order to focus our analysis on the effect of the stacking sequence. Unfortunately the porosity volume fraction and consequently the matrix volume fraction are then not equal. However, we consider that the porosity formation depends on the stacking sequence (formation of porosity network in the flax plies). Once the plates have been selected, in each plate one specimen for sorption test and seven specimens for mechanical tests are extracted.

The process used to manufacture the composite plates is the manual impre gnation of plies associated with the vacuum bagging technique. After pressurization under 0.9 bar for 24 hours and at room temperature, post-curing in an oven is required to obtain a complete polymerization of the epoxy matrix, which consists of two bearings; the first at 80 °C is for decreasing the viscosity of the resin and promoting its dispersion, the second bearing at 130 °C allows for a complete crosslinking (Figure 2).

3 EXPERIMENTAL METHODS

3.1 Sorption Test

Measurement of water sorption using gravimetric analysis is relatively simple to implement. Consequently, it is currently used to characterize the hygroscopic behavior of a composite material.

Prior to immersion, the samples are carefully prepared. Following ASTM D5229 norm, 120 mm square samples are cut in the composite plates. They are oven-dried at 103 °C for 24 hours to remove any moisture. In order to consider only transversal water sorption through the sample surface, the four borders were covered with silicone paste to prevent lateral edge sorption. Then, the sample was immersed into a water bath at a controlled temperature of 20 °C. It has been verified that silicone sorption is negligible compared to composite sorption. Plates are weighed regularly using an electronic weigh scale of 0.1 g precision. Before being weighed, the sample surface was wiped with an absorbent cloth. The water sorption rate M_t at time *t* can be expressed as follows:

$$M_t = \frac{W_t - W_0}{W_0} \tag{1}$$

where W_0 is the weight of dried specimen and W_t is the weight of the wet specimen at time *t*.

In order to determine the sorption mechanism and the parameters' characteristics, such as saturation water content and diffusion coefficient, several tests were conducted to compare the results, verify



Figure 2 Post-curing cycle.

and validate their consistency and draw a conclusion thereafter.

3.2 Tensile Test

A tensile test was used to determine the mechanical properties of the composites, in particular the Young's modulus and ultimate tensile stress. The test stand used a servo-hydraulic Instron 8801 tensile machine equipped with a 100kN force cell and self-tightening mechanical jaws. The tests were carried out at room temperature with a travel speed of 5 mm/min on specimens with a width of 20 mm \pm 0.5 and a length of 250 mm. In accordance with ASTM D3029/D3039M norm, specimens were cut using a water-cooled diamond saw. To prevent slippage problems and crushing in the clamping jaws, aluminium stubs were bonded with a structural adhesive epoxy on the two faces of the two ends of the specimen. Measuring deformation was made locally on the specimen using an extensometer in contact with the surface of the sample on a gauge length of 25 mm. So, the estimation of the material's mechanical properties, such as Young's modulus or strain at break, was not disturbed by the problems of deformation or sliding in the jaws of the tensile machine.

4 **RESULTS**

4.1 Microstructural Analysis

The plates have an average dimension of 270 mm long and 230 mm wide. The thickness of the different plates is a function of the laminate stacking sequence and the composition of the boards obtained. In all cases, are substantially thicker than hybrid plates. The rates of porosity, V_p , of matrix V_m and of fiber V_f for each plate are determined from the dimensions of the plates (length L, width w and thickness h) and its mass $M_{composite}$; the mass M_{carbon} and M_{flax} the number of ply n_{carbon} and n_{flax} and the basis weight Gr_{carbon} and Gr_{flax} of each type of fabric (flax or carbon), the densities of epoxy matrix ρ_{matrix} , carbon fiber ρ_{carbon} and flax fiber ρ_{flax} .

$$V_{f} = \frac{\text{Volume}_{\text{fibre}}}{\text{Volume}_{\text{composite}}} = \frac{\text{Volume}_{\text{carbon}} + \text{Volume}_{\text{flax}}}{\text{Volume}_{\text{composite}}}$$
$$= \frac{\frac{M_{\text{carbon}}}{\rho_{\text{carbon}}} + \frac{M_{\text{flax}}}{\rho_{\text{flax}}}}{L.w.h} = \frac{\left(\frac{Gr_{\text{carbon}}}{\rho_{\text{carbon}}} \cdot n_{\text{carbon}} + \frac{Gr_{\text{flax}}}{\rho_{\text{flax}}} \cdot n_{\text{flax}}\right) \cdot L.w}{L.w.h} (2)$$
$$V_{m} = \frac{M_{\text{matrix}}}{\rho_{\text{matrix}}} = \frac{\left(M_{\text{composite}} - M_{\text{fibre}}\right)}{\rho_{\text{matrix}}}}{V_{p} = 1 - \left(V_{f} + V_{m}\right)}$$

With natural fiber composite, the porosity rates obtained in these plates are very important; they are superior to 30% for 100% flax composite. For high rate of porosity, mechanical properties are affected and degraded significantly. Replacing 4 flax plies by 4 carbon plies reduces the porosity rate to an average value of 25.5%.

During manual composite manufacturing, mechanical causes responsible for the creation of porosity are basically the mechanical trapping of air in the dry fabric plies during the draping and in the resin during its mixing phase or during the impregnation of the fibrous reinforcement. More specifically, during the step of draping, gaseous microcavities are formed when a fiber is broken or between overlapping plies. Pressure applied with vacuum bagging technique is then useful to increase compaction of the laminate and consequently to reduce the porosity volume fraction. However, excessive depression could play the reverse role and wring a significant amount of matrix from the fabric lay-up and subsequently increase the porosity. In addition, a rapid and local temperature increase of the resin or a rapid decompression can generate the formation of gas bubbles within the resin. Despite the use of the same composite manufacturing process (amount of matrix, experimental conditions [pressure and temperature], impregnation operating process, etc.), a fairly clear dispersion in the porosity rate between the various plates is found (Figure 3). In view of these results, the stacking sequence is highlighted. It is clear that the more the carbon plies are on the outside of the composite plate, the lower the porosity rate is. This can be explained by the architecture and structure of the flax fabric which is insufficiently filled and which has large spaces between the warp and weft yarns (Figure 1). Optical microscopic observations of the surface porosity reveals that some porosities formed in an internal flax ply are opening out on the surface through a carbon ply. Thus, open porosities composed of intercommunicating voids connected to the outer portion of the material (white spots in Figure 4) are clearly observable. The porosity network, formed mainly in flax plies, is percolating and forms diffusion channels for water through the composite material. The use of more dense carbon fiber plies may limit the connection between the porosities.

Due to the low compatibility of flax fiber with epoxy matrix, with external flax ply placed on the laminate surface a considerable amount of matrix is absorbed by the breather/absorption fabric. The laminate external surface is then dried, with too low a matrix volume fraction and consequently a high porosity volume fraction.



Figure 3 Volume rate of each component (matrix, fiber and porosity) of the different composite plates.



Figure 4 Porosities formed in an internal ply of flax and opening out on the surface through a carbon ply.

4.2 Water Absorption

Sorption test were carried out on four stacking sequences $[C C F F]_{s'}$ $[C F C F]_{s'}$ $[F C F C]_{s}$ and 100% flax. The experimental results for water sorption are plotted in Figure 5 and the characteristic values associated with the porosity rate for each composite plate are presented in Table 2.

4.2.1 Validation of the Fickian Behavior of Water Recovery

Diffusion behavior is distinguished into three categories [31, 32]

• Case I: diffusion-controlled water recovery, called Fickian diffusion, in which the penetrating mobility is much lower than the mobility of polymer segments.

- Case II (and Super Case II): relaxationcontrolled water recovery, in which the penetrating mobility is much higher than the relaxation process.
- Non-Fickian or anomalous diffusion, in which the mobility of the penetrant and the relaxation of the polymer segments are comparable.

To study the mechanism of absorption of water by the composites, the experimental data are adjusted with Equation 3, which can be expressed as Equation 4, where is water content at instant t and is water content at saturation and are constants.

$$\frac{M_t}{M_{\infty}} = kt^n \tag{3}$$

$$\log\left(\frac{M_t}{M_{\infty}}\right) = \log k + n\log t \tag{4}$$

Constants and are used to identify the mechanism of water absorption by the composite and are identified by linear regression on experimental values. The constant is a constant characteristic of the studied material and characterizes the interaction between the material and the water. The constant is used to identify the sorption mechanism in the following cases [33]:

- n < 0.5: Pseudo-Fickien,
- n = 0.5: Case I (Fickien),
- 0.5 < n < 1: anomalous,
- n > 1: Case II.

Thus, stacking sequences 100% flax, $[F C F C]_s$ and $[C F C F]_s$ have a pseudo-Fickian diffusion behavior





Figure 5 Adjustment of the classical Fickian diffusion model on the experimental results of water sorption by the laminates: (a) 100% Flax, (b) [F C F C]s, (c) [C F C F]s and (d) [C C F F]s.

Plate stacking sequence	Fickian mechanism constant	Porosity rate (%)	Diffusion coefficient (mm ² .s ⁻¹)	Water content at saturation (%)
100% flax	0.453	32.4	4.72	32.9
[F C F C] _s	0.232	27.50	4.05	21.6
[C F C F] _s	0.380	26.34	1.94	20.1
[C C F F] _s	0.500	22.52	1.80	20.0

 Table 1 Characteristics of the sorption test.

while the plate $[C C F F]_s$ has a Fickian diffusion behavior (Table 1)

4.2.2 Modeling the Diffusion Kinetics

The experimental data were used to draw the water content curve as a function of the root of time. This data is compared with the theoretical expression of Fickian diffusion for a large plate thickness h. The water absorption over time is given by Equation 5 for the short time and Equation 6 for the long time [34].

$$\frac{M_t}{M_{\infty}} = \frac{4}{h} \sqrt{\frac{D.t}{\pi}} \tag{5}$$

$$\frac{M_t}{M_{\infty}} = 1 - \frac{8}{\pi^2} \exp(\frac{-D.\pi^2.t}{h^2})$$
(6)

Figure 5(a–d) is used to check the fit of the Fickian diffusion model on experimental data. The fit of the experimental values with the Fickian model shows an overlay more or less perfect; the fit is almost perfect for stacking sequences $[F C F C]_s$ and $[C C F F]_s$, less perfect for 100% flax, while the gap increases more for the $[C F C F]_s$. The water recovery cannot only be modeled with a unique Fickian law. Several parameters influence the sorption process. Mainly cited are the relative humidity and the ambient temperature of the environment in which the plates are placed at which the water

diffusion coefficient is very sensitive. However, these two factors cannot be considered as influential in our case since all the plates were placed in the same moisture and temperature conditions. Consequently, other factors must be taken into consideration. Contrary to composites reinforced by conventional reinforcements where the water sorption phenomenon is controlled by the matrix, in NFRC sorption is also associated with flax fibers because of their hydrophilic character. Moreover, the flax ply location plays a major role in the sorption kinetics by its contribution to increasing the degree of porosity, as detailed in the previous section on microstructural analysis.

For hybrid carbon-flax composite, the location of flax plies on the surface of the structure exposes them to more moisture and increases the absorption of water. According to Table 1, the diffusion coefficient increases when flax plies are placed near the composite external surface instead of being placed in the heart. Therefore the diffusion coefficient is nearly doubled when a flax ply is placed at the surface. In addition, placing the carbon plies on the surface reduces the diffusion coefficient of water but does not stop the water recovery. Indeed, the water content at saturation is nearly constant whatever the hybrid stacking sequence.

It seems obvious that the positioning of the flax plies influences the diffusion coefficient through the porosity rate parameter. The diffusion coefficient is directly linked to the creation of a percolating porosity network through the material. Positioning flax plies on the surface of the plate makes it more susceptible to the creation of porosity and cavity network that represent potential sites for water sorption, while, by limiting the formation of surface cavity, external carbon ply limits the diffusion coefficient.

Stacking sequence with alternating carbon and flax plies $[C F C F]_s$ is not more efficient in limiting water diffusion speed than stacking sequence with all the carbon plies on the surface $[C C F F]_s$. The barrier made by the internal carbon ply is not enough to stop the percolation of porosity network and slow down water diffusion.

4.3 Mechanical Properties in Tension

Conventional tensile tests were performed on samples extracted from the different composite plates made with various hybrid stacking sequences. Each material was tested 5 times. The tensile test results are illustrated in Figure 6, where each stacking sequence is plotted for the most representative behavior curve. The stress-strain curves of the three hybrid composites are broadly similar to each other and very different from the 100% flax composite one. After a short linear stage, the 100% flax composite behavior is



Figure 6 Example of experimental tensile stress-strain curves for the different hybrid stacking sequences and 100% flax composites.

nonlinear with a strong softening. Rupture occurs at a loading level much lower than hybrid composites. As for carbon fiber-reinforced composite, the stress-strain curves of the laminates [C F C F], and [C C F F], are linear up to the breakage of the sample, whereas the curve [F C F C]_e is no longer linear from a stress level around 50% of the ultimate strength. For the stacking sequence [C C F F], it should be specified that the short horizontal bearing observed in the stress-strain curve corresponds to a slip of the extensometer sensor during the experimental tensile test. Depending on the flax ply position in the stacking sequence, the hybrid composite fiber behavior may tend towards the brittle behavior of carbon fiber-reinforced composite or towards the ductile behavior of the flax-reinforced composite.

According to Figure 7a, hybridization with carbon fibers significantly increases the Young's modulus by almost 6 times; subsequently, the location of carbon plies in the structure maximizes this result. Despite a fiber rate almost equivalent (39.2%), the stacking sequence [F C F C], [C C F F] and [C F C F] do not have the same Young's modulus and a clear difference is observed in the results that even reaches 23% between the [FCFC] and [CCFF] sequences. Putting only one carbon ply on the surface, [C F C F] stacking sequence, increases the Young's modulus about 18%. We deduce that what drives the optimization of the Young's modulus is essentially the nature of the surface ply and, thereafter, the alternation of carbon and flax internal plies aims to improve the results but not as significantly.

According to the classical laminate theory, in the case of a symmetric laminate composed of orthotropic layers of different nature that have material directions aligned with the laminate directions and that is submitted to a uniform and longitudinal tensile loading, the strain field is constant all over the laminate and is equal whatever the laminate layer. Moreover, given the





Figure 7 Mean values and standard deviation of (a) Young's modulus, (b) ultimate tensile stress and (c) failure tensile strain for the different hybrid stacking sequences and 100% flax composites.

same global composition for the laminate, the stacking sequence has no influence on the value of the strain field [35]. Consequently, the Young's modulus should be the same regardless of the stacking sequence, which is not observed experimentally. However, the classical laminate theory, based on the Love-Kirchhoff scheme for plates, assumes, among other things, that interlaminar transversal shear stress between laminate layers is negligible. The interface and the mechanical load transfer between two successive layers must then be perfect. In their study on hybrid glass/flax-reinforced composite, Zhang *et al.* did not observe some influence of the stacking sequence on the Young's modulus [24]. They concluded that the interface between layers did not play a crucial role in the elastic modulus. But they also observed, with SEM micrographs, that the hybrid glass/flax interfaces were efficient for transfer loading because of the morphology of flax yarns.

Considering our hybrid laminate structure and the relatively high porosity volume fraction of the tested composite materials, this assumption of perfect interface and uniform displacement field needs to be checked. The first clue is the nonlinear behavior of the $[F C F C]_s$ laminate, whereas the other hybrid stacking sequences with external carbon ply $[C C F F]_s$ and $[C F C F]_s$ present a linear behavior. This nonlinear behavior could only be representative of the external flax ply. In a future test campaign, we plan to monitor the displacement field by digital image correlation on the specimen edge, which should provide us with interesting information on this point.

Regarding the ultimate tensile stress and failure tensile strain (Figure 7b,c), the differences between sequences $[F C F C]_{s'}$, $[C C F F]_{s}$ and $[C F C F]_{s'}$, having the same fiber content, is around 10%. Variations of the same order of magnitude were observed by Zhang *et al.* on hybrid flax/glass fiber-reinforced composite [24]. It has been concluded that the carbon ply location within the structure will not have a

great influence on the failure strength of the hybrid composite. The failure of hybrid composite is mainly governed by the failure of the carbon plies, whose maximal strain at break is dramatically lower than that of flax ply.

5 CONCLUSIONS

Measure of porosity rate, sorption properties and tensile properties of different stacking sequences of hybrid carbon-flax laminate composites were carried out to produce a global result. We deduced that the more carbon plies are near the surface, the less the rate of porosity, which can be explained by the architecture and structure of the flax fabric which is insufficiently filled. In addition, some experimental parameters were highlighted, like manual draping, temperature test and application pressure. Thereafter, sorption tests were conducted to determine suitable sorption mechanism and adjust it with Fickian modeling to determine the water content at saturation and the diffusion coefficient of each stacking sequence. The main result obtained shows that the diffusion coefficient is decreased to half when just a carbon ply is placed on the surface and the water content at saturation is decreased by 40% between 100% flax and carbon/flax hybrid plates, which can be explained by the hydrophilic character of natural fiber and the high porosity rate in plates with flax surface ply. We ended with tensile tests to determine the mechanical properties of the various stacking sequences. As a result, we deduced that for the optimization of the Young's modulus of carbon-flax/epoxy laminate, what matters is the nature of surface ply (carbon or flax) in the structure. The failure properties are governed by the carbon ply. Indeed, ultimate tensile stress is highly affected by the insertion of carbon fibers in the structure, regardless of their position, and increases more

than 3 times with hybrid stacking sequences. Failure tensile strain is affected negatively and its value is dropped almost to half by the inclusion of carbon ply inside the structure.

It is obvious that the volume fraction of carbon fiber of 50% used in this work strongly limits the environmental benefits induced by the utilization of natural fiber to reinforce composite material. However, this first step has been useful to validate the concept of carbon-flax hybrid composite and to identify future lines of investigation. We plan to continue the work on thicker laminated structures with a larger total number of plies, but always with 2 carbon plies on each surface, that will limit the carbon fiber volume fraction.

REFERENCES

- 1. K. Van de Velde and P. Kiekens, Wettability of natural fibres used as reinforcement for composites. *Die Angew. Makromol. Chem.* **272**, 87–93 (1999).
- 2. A.K. Bledzki and J. Gassan, Composites reinforced with cellulose based fibres. *Prog. Polym. Sci.* 24, 271–274 (1999).
- 3. C. Baley, Analysis of the flax fibres tensile behaviour and analysis of the tensile stiffness increase. *Compos. Part A* **33**, 939–948 (2002).
- M.I. Misnon, M.M. Islam, J.A. Epaarachchi, and K.T. Lau, Potentiality of utilising natural textile materials for engineering composites applications. *Mater. Des.* 59, 359–368 (2014).
- 5. B.V. Kokta, R.G. Raj, and C. Daneault, Use of wood flour as a filler in polypropylene: Studies on mechanical properties. *Polym. Plast. Technol. Eng.* **28**, 247–259 (1989).
- 6. J.M. Felix and P. Gatenholm, The nature of adhesion in composites of modified cellulose fibres and polypropylene. J. Appl. Polym. Sci. **42**, 609–620 (1991).
- 7. H. Ma, Y. Li, and D. Wang, Investigations of fibre twist on the mechanical properties of sisal fibre yarns and their composites. *J. Reinf. Plast. Compos.* **33**, 687–696 (2014).
- Z.E. Cherif, C. Poilâne, T. Falher, A. Vivet, N. Ouail, B. Ben Doudou, and J. Chen, Influence of textile treatment on mechanical and sorption properties of flax/ epoxy composites. *Polym. Compos.* 34, 1761–1773 (2013).
- 9. Y. Li, Q. Li, and H. Mao, The voids formation mechanisms and their effects on the mechanical properties of flax fibre reinforced epoxy composites. *Compos. Part A* **72**, 40–48 (2015).
- B. Madsen and H. Lilholt, Physical and mechanical properties of unidirectional plant fibre composites— An evaluation of the influence of porosity. <u>Compos. Sci.</u> Technol. 63, 1265–1272 (2003).
- 11. J.L. Thomason, The interface region in glass fibrereinforced epoxy resin composites: 2. Water absorption, voids and interface. *Compos.* **26**, 477–485 (1995).
- Q. Lin, X. Zhou, and G. Dai, Effect of hydrothermal environment on moisture absorption and mechanical properties of wood four-filled polypropylene composites. *J. Appl. Polym. Sci.* 85, 2824–2832 (2002).

- A. Espert, F. Vilaplana, and S. Karlsson, Comparison of water absorption in natural cellulosic fibres from wood and one-year crops in polypropylene composites and its influence on their mechanical properties. *Compos. Part A* 35, 1267–1276 (2004).
- 14. M. Assarar, D. Scida, A. El Mahi, C. Poilâne, and R. Ayad, Influence of water ageing on mechanical properties and damage events of two reinforced composite materials: Flax-fibres and glass-fibres. *Mater. Des.* **32**, 788–795 (2011).
- 15. D. Scida, M. Assarar, C. Poilâne, and R. Ayad, Influence of hygrothermal ageing on the damage mechanisms of flax-fibre reinforced epoxy composite. *Compos. Part B* **48**, 51–58 (2013).
- A. Le Duigou, A. Bourmaud, and C. Baley, In-situ evaluation of flax fibre degradation during water ageing. *Ind. Crop. Prod.* 70, 204–210 (2015).
- A.K. Mohanty, M. Misra, and G. Hinrichsen, Biofibres, biodegradable polymers and biocomposites: An overview. *Macromol. Mater. Eng.* 276, 1–24 (2000).
- C.A.S. Hill and H.P.S. Abdul Khalil, Effect of fibre treatments on mechanical properties of coir or oil palm fibre reinforced polyester composites. *J. Appl. Polym. Sci.* 78, 1685–1697 (2000).
- I. Van de Weyenberg, J. Ivens, A. De Coster, B. Kino, E. Baetens, and I. Verpoest, Influence of processing and chemical treatment of flax fibres on their composites. *Compos. Sci. Technol.* 63, 1241–1246 (2003).
- Y. Swolfs, L. Gorbatikh, and I. Verpoest, Fibre hybridisation in polymer composites: A review. *Compos. Part A* 67, 181–200 (2014).
- M.R. Sanjay, G.R. Arpitha, and B. Yogesha, Study on mechanical properties of natural–glass fibre reinforced polymer hybrid composites: A review. *Mater. Today* 2, 2959–2967 (2015).
- 22. B. Panthapulakkal and M. Sain, Injection-molded short hemp fibre/glass fibre reinforced polypropylene hydrid compostes—Mechanical, water absorption and thermal properties. *J. Applied Poly. Sci.* **103**, 2432–2441 (2007).
- 23. K. Jarukumjorn and N. Suppakarn, Effect of glass fibre hybridization on properties of sisal fibre polypropylene composites. *Compos. Part B* **40**, 623–627 (2009).
- 24. Y. Zhang, Y. Li, H. Ma, and T. Yu, Tensile and interfacial properties of unidirectional flax/glass fibre reinforced hybrid composites. *Compos. Sci. Technol.* **40**, 172–177 (2013).
- 25. A.K. Sabeel and S. Vijayarangan, Tensile, flexural and interlaminar shear properties of woven jute and jute-glass fabric reinforced polyester composites. *J. Mater. Process. Technol.* **207**, 330–335 (2008).
- 26. S.C. Amico, C.C. Angrizani, and M.L. Drummond, Influence of the stacking sequence on the mechanical properties of glass/sisal hybrid composites. *J. Reinf. Plast. Compos.* 29, 179–189 (2010)
- H.P.S.A. Khalil, C.W. Kang, A. Khairul, R. Ridzuan, and T.O. Adawi, The effect of different laminations on mechanical and physical properties of hybrid composites. *J. Reinf. Plast. Compos.* 28, 1123–1137 (2009).
- 28. M. Assarar, W. Zouari, H. Sabhi, R. Ayad, and J.M. Berthelot, Evaluation of the damping of hybrid



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carbon-flax reinforced composites. *Compos. Struct.* **132**, 148–154 (2015).

- 29. Z.S. Bagheri, I. El Sawi, H. Bougherara, and R. Zdero, Biomechanical fatigue analysis of an advanced new carbon fibre/flax/epoxy plate for bone fracture repair using conventional fatigue tests and thermography. *J. Mech. Behav. Biomed. Mater.* **35**, 27–38 (2014).
- H.N. Dhakal, Z.Y. Zhang, R. Guthrie, J. MacMullen, and N. Bennett, Development of flax/carbon fibre hybrid composites for enhanced properties. *Carbohydr. Polym.* 96, 1–8 (2013).
- 31. J. Comyn (Ed.), *Polymer Permeability*, pp. 77–85, Chapman & Hall, London (1985).

- 32. L.H. Sperling, *Introduction to Physical Polymer Science*, 4th ed., pp. 145–164, Wiley-Interscience Publications, New Jersey (2006).
- C. Salome, O. Godswill, C. Ikechukwu, and O. Ikechukwu, Kinetics and mechanisms of drug release from swellable and non swellable matrices. *RJPBCS* 4, 97–103 (2013).
- C.H. Shen and G.S. Springer, Moisture absorption and desorption of composite materials. <u>J. Compos. Mater.</u> 10, 2–20 (1976).
- 35. J.M. Berthelot, *Composite Materials: Mechanical Behaviour and Structural Analysis*, Mechanical Engineering Series, Springer Editions, New York, Verlag (1999).