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Abstract

The aim of this work was to study the effect of free surface energy modification of flax fibres by a thermal treatment on the mechanical behaviour of bio-based composites. It has been proved that this modification enhances the wettability of flax fibres by liquid epoxy resin and results in a lower porosity amount in composites. Tests to evaluate mechanical properties of elementary fibres, yarns and composites have been performed. The main outcome of this multiscale study, even if elementary fibres and yarns have been embrittled and interface properties have been lowered after thermal treatment, is that the mechanical behaviour of composites manufactured by Liquid Composite Molding (LCM) is better with treated fibres.

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1. Introduction

Composite materials are, by design, characterised by the quality of the interface between their constituents. One of the main focus in developing innovative composite materials is to ensure a proper bonding between reinforcements and matrix. This bonding can result from covalent bonds or physico-chemical interactions. The strongest bonding results from covalent bonds and a modification of the chemistry of the fibre surface is often used in petro-sourced composites to induce chemical reactions with resin, such as epoxy. However, those kinds of surface treatment induce an increase of the carbon dioxide footprint of the overall process of composite manufacturing. Focusing on minimizing this footprint by the use of bio-based reinforcement [1, 2], the addition of a chemical treatment can counterbalance the gain from the environmental point of view. Low emission treatments have thus to be found.

Flax fibres are usually considered as adequate bio-reinforcements for composites [3], but their hydrophilic character makes them sensitive to moisture sorption and difficult to wet by hydrophobic resins [4, 5]. Several ways have been explored to improve wettability and bonding of bio-based reinforcements by resins. In this work, a thermal treatment aiming at modifying the surface properties of flax fibres [6] has been applied to enhance their wettabillity, and thus modifying the impregnation of fibres by resins, decreasing at the same

time their sensitivity to water sorption. The wettability of untreated and treated fibres has been assessed in a previous work [7], by measuring contact angles between the fibres and two test liquids: water and diiodomethane. Contact angles have been determined via the Wilhelmy relation, once the wetted length and the weight of the meniscus formed by the test liquid in contact with the fibre are known. Modification of wetted length during immersion in test liquids (due to fibres swelling or shrinking) was also taken into account [7, 8].

During manufacturing of biobased composites by Liquid Composite Moulding (*LCM*) processes, such as Vacuum Assisted Resin Transfer Molding (*VARTM*) impregnation of fibres by the liquid resin can induce an imperfect interface and micro-porosity depending on the physicochemical affinity of liquid and solid phases. This family of composite manufacturing processes, being effective and low cost, is increasingly employed. It is then mandatory to understand the effect of the impregnation phenomena in order to match high quality requirements, especially for parts with a complex shape. However, a large amount of voids in bio-based composites manufactured by LCM processes have been observed [9] at different scales [10, 11].

The aim of the present study is to evaluate the effect of the modification of the free surface energy (by modifying one of its components) of flax fibres by a thermal treatment on the mechanical behaviour of flax reinforced biobased composites. It has already been proved [7] that this modification has the effect of enhancing the wettability of flax fibres by liquid epoxy resin,

resulting in a lower porosity amount in composites reinforced by treated flax fibres.

It is well established that the surface chemical composition of fibres has an impact on wetting and thus on the impregnation dynamic. Polar and dispersive components of fiber surface energy, causing hydrophilic or hydrophobic behaviour of the reinforcement, have an effect on the interface formation during manufacturing but also on its toughness in the composite under service conditions. However, in literature a controversy still exists about the effective nature of surface energy and wetting behaviour of flax fibres [5, 12, 13]. Controlling the chemical surface state of such sustainable reinforcements is thus an issue of paramount importance, that has to be tackled with an approach that neither harms the environemental impact of the reinforcement processing and nor induces the production of unhealthy wastes.

Numerous studies [1, 14, 15, 16, 17, 18, 19, 20, 21, 22] have been devoted to the experimental methods dedicated to the measurements of mechanical properties of fibres and their interface with the matrix, without considering the formation of interfaces during manufacturing that affect the resulting properties.

The present paper thus details tests on treated and untreated fibres to evaluate mechanical properties of elementary fibres, yarns and composites. Tensile tests on elementary fibres, but also on yarns with different gauge length, have be performed. Composites mechanical properties have been investigated through Short Beam Shear Tests (SBST) and four points bending

tests. In order to assess the effect of the surface energy modification on the interface toughness, microdroplets debonding tests on fibres have been performed with the same resin used for bio-based composites manufacturing.

The originality of this study is to give another point of view on studies that have proved the effects of the thermal treatment on swelling [8] and on void formation during manufacturing by LCM processes [7]. Indeed, the focus is here to understand the effect of treatment on the mechanical behaviour of elementary fibres, yarns, interface and then to extend these results at the scale of service conditions. These results will address the main issue of the impregnation quality of fibres and its effects at the scale of the composite and its application.

2. Materials and methods

2.1. Materials

Quasi-unidirectional flax fibres were provided by Libeco (FLAXDRY UD 180®). The areal weight of the fabric is 180 g/m^2. Some fabrics were subjected to a thermal treatment at 220°C for 2h under inert atmosphere [6, 7, 8]. Elementary fibres were all extracted from untreated and treated twisted yarns belonging to fabrics (Fig. 1).

The epoxy resin used for manufacturing composites by LCM processes and microdroplets was SP106® provided by Gurit which cures rapidly at room temperature.

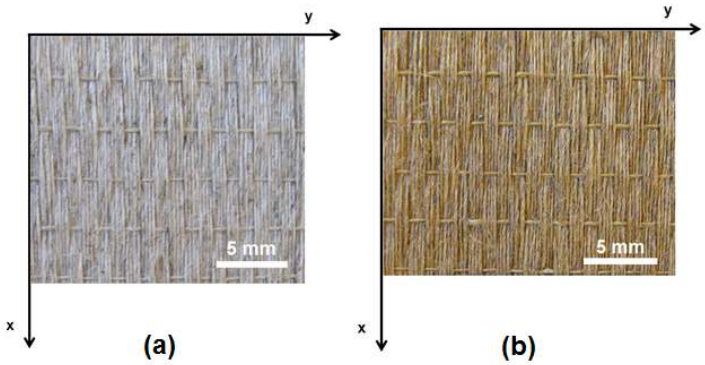

Figure 1: Untreated (a) and treated (b) flax fiber reinforcements

2.2. Methods

2.2.1. At fibres scale

2.2.1.1 Tensile tests on single fibres

Tensile tests were performed on 25 individual fibres for each batch. A gauge length of 10 mm was chosen. Samples were clamped on MTS type tensile testing machine equipped with a 2 N load cell. These tests were performed in a temperature and relative humidity controlled laboratory (23 $\pm$ 0.5 °C, 48 $\pm$ 2 % RH). Longitudinal mechanical properties under traction (apparent tensile modulus, strain and stress at break) of the different kind of elementary fibres were determined in accordance with the NFT 25-501-2 [23] standard. Compliance of the loading frame was taken into account. The considered mean diameter value of each fibre, required to calculate apparent modulus and strength, was taken as an average value from five measurements along the fibre with an optical microscope.

2.2.1.2 Debonding of micro-droplets

Resin droplets were placed on flax elementary fibres using a single glass fibre which had been dipped in the epoxy resin. Microbond specimens were then checked under the microscope to control the droplet geometry, length and height. Samples with defects (kink bands on the fibre or lack of symmetry of the droplet) were systematically rejected. Besides being symmetrical, microdroplets need to be smaller than 150 μm, the length to prevent fibre break. At least 25 specimens were tested for each test condition. Then the flax fibre with the epoxy microdroplet was mounted in the shearing device and continuously observed with a microscope. The fibre was pulled out of the droplet while the latter was constrained by knife edges. The loading rate during debonding was 0.1 mm.min^{-1}. The apparent shear strength at debonding (τ_{app}) was calculated using the equation 1, from Miller et al. [24].

$$\tau = \frac{F}{S_{emb}} \tag{1}$$

where τ corresponds to either the apparent shear stress at debonding τ_{app} or the friction stress after debonding friction. F is either the debonding force or the friction force. S_{emb}, the embedded surface area, corresponds to the bonded area between the fibre and the matrix.

2.2.2. Mechanical tests on flax yarns

When dealing with composite reinforcement, yarn characteristics need to be investigated. In the present work flax yarns have been characterized in terms of density, structure and mechanical properties. The density of un-

treated and thermally treated flax yarns was measured by helium pycnometry (AccuPyc 1330) [25]. Yarn structure, in terms of twisting angle of flax fibres and yarn diameter, was determined by scanning electron microscopy (Philips XL40). In particular, five flax yarn samples for untreated and treated fibres (sputter coated with gold prior to examination), each of 15 mm in length, were observed, and within each sample, the fiber twisting angle and diameter were measured at 10 locations along the yarn. With regard to mechanical properties, the single yarns were tested using the same recommendations of the method described in ASTM C1557 [26]. Single yarns were carefully separated by hand from the fabric. Tensile tests were carried out at room temperature by means of a Zwick/Roell Z010 equipped with a 200 N load cell. Three different gauge lengths were tested, namely 20, 30, and 40 mm, in order to measure the system compliance. Individual yarns were glued onto card tabs with a central window cut out to match the desired gauge length for the test. Tests were performed in displacement control at a crosshead speed of 2 mm.min^{-1}. For each gauge length and fabric treatment, fifty yarns were tested. The mechanical properties of the flax quasi-UD fabric before and after thermal exposure was assessed according to ASTM D5035 [27]. Tensile tests were carried out at room temperature by means of a Zwick/Roell Z010 equipped with a 10 kN load cell. One gauge length was used, i.e. 75 mm, for specimens with a width equal to 25 mm. Tests were performed in displacement control at a crosshead speed of 100 mm.min^{-1} to ensure failure within 20$\pm$3 s.

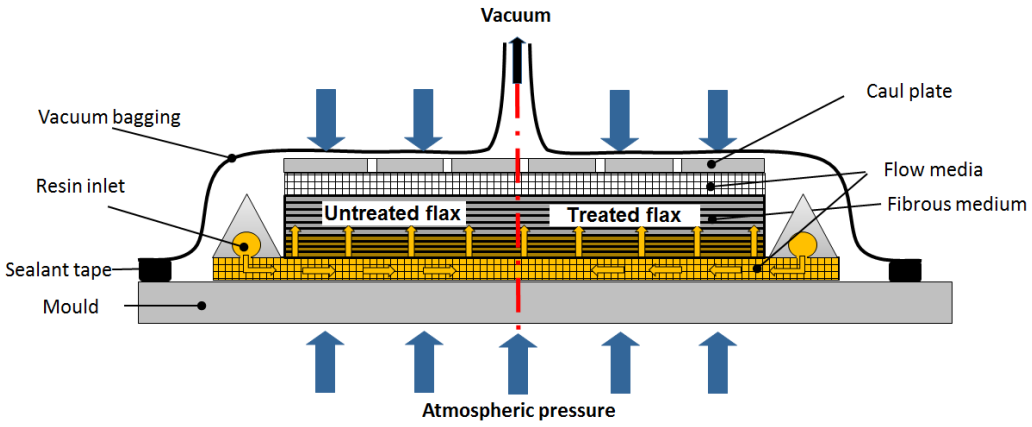

Figure 2: Infusion strategy with the two reinforcements [7]

2.2.3. Bio-based composites manufacturing

A composite plate was manufactured by Vacuum Assisted Resin Transfer Molding (VARTM). The infusion configuration used for this test is described in Fig. 2. The aim was to manufacture plates using both reinforcements, untreated and treated flax fabrics, in a single infusion ($[0]_{10}$) with the same processing conditions for the two types of reinforcements (Fig. 3). A distribution medium was placed under the preform to ensure the resin feeding. Another distribution medium was placed under the vent, which is located at the center of the top of the plate, to ensure the quality of the vacuum on the upper part of the preform. Infusion was performed at room temperature with the SP106® resin. A 1.4 mbars vacuum was pulled, and resin was left entering until it could be visible in the outlet silicone pipe.

Figure 3: Preform with both reinforcements: untreated (left) and treated (right).

2.2.4. At composites scale

2.2.4.1 Four points bending on flax reinforced composites

To evaluate the flexural strength of manufactured composites, four points bending tests (Fig. 4) have been performed on samples reinforced by both treated and untreated flax fabrics. The distance between two consecutive supports is 20 mm for a total length between the external supports of 60 mm, following ASTM D7264 [28]. Dimensions of samples were 75x25x3 mm^3. A preload of 50 N and a travel speed of 1 mm/min have been used. Six samples for each type of composites were tested.

2.2.4.2 Beam Shear tests on flax reinforced composites

Short Beam Shear Tests (SBST) have been conducted following ASTM

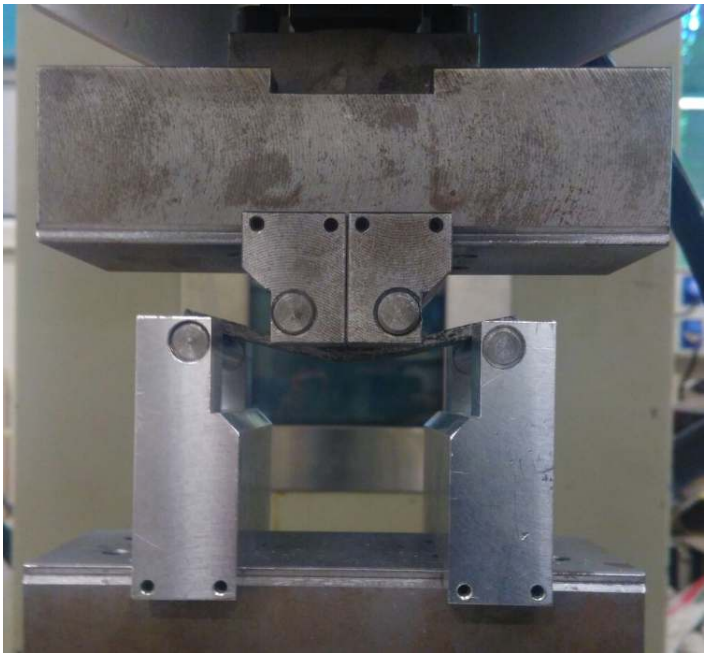

Figure 4: 4-point bending on a composite reinforced by untreated flax fabrics.

D2344 [29] in order to evaluate the interlaminar shear strength of both composites reinforced by treated and untreated flax fibres. Five samples for each type of composites were tested. Surface dimensions of the samples were 5x30 mm^2. Thicknesses were measured as 3 mm for untreated flax reinforcements and 3.2 mm for treated reinforcements. This led to a small difference in the fiber volume ratio (41.4 % for flax fibres and 38.8 % for treated flax fibres). To induce a failure in agreement with the standard (shear at the neutral axis) the spacing of the two bottom loading points has to be five times the thickness of the sample. It was thus set to 16 mm (Fig. 5).

Figure 5: a) Short Beam Shear bench; b) Sample after failure during test.

3. Results

3.1. Tensile properties of flax fibres

Evaluation of the tensile modulus and strength requires the measurement of fibre diameters. Measurements performed in the present study confirm the ones made on fibres from the same fabric in a previous work [8]. It also confirms that the thermal treatment has no effect on the fibres morphology. Results obtained after tensile tests are summarized in Table 1.

Materials	**Diameter** (μm)	$\mathbf{E}_{fibre}$ (GPa)	**Strength** (MPa)	**Strain at failure** (%)
Untreated fibres	16.4±4.1	39±11	1009±392	2.50±1.40
Treated fibres	16.0±4.0	38±10	447±216	1.50±0.60

Table 1: Tensile properties obtained on both untreated and treated elementary flax fibres

Results obtained on untreated fibres can be compared to literature [30]. Values of tensile modulus and tensile strength are relatively moderate. It can be due to the quality of the fibres itself or to the fabric manufacturing

processes. Indeed, the values of literature [30] have been obtained from elementary fibres that have been directly extracted from flax slivers.

The main result here is that the tensile modulus is not modified by the thermal treatment. On the other hand, a strength loss of about 50% was recorded after thermal treatment. This effect has been observed in the literature [31] but, again, on elementary fibres that were not extracted from a fabric. This loss of tensile strength can suggest a hornification mechanism or a degradation of cellulosic constituents [31]. This could be due to a progressive and irreversible modification of biochemical structure of fibres, like pectins [32], hemicellulose [33] or even non cristalline fibrillar cellulose [34]. Thus, Gassan and Bledzki [35] evidenced that the structural properties of cellulose, hemicelluloses and pectines may be altered by high temperature. Additional and specific investigations are needed to get insight into this degradation of tensile strength.

3.1.1. Interfacial bond strength

The micro-droplet debonding test on both treated and untreated fibres allowed to calculate an InterFacial Shear Strength (IFSS) for the fibre/matrix interface but also the friction stress after debonding. Values obtained from those tests are summarized in Table 2.

Again, results on untreated fibres appear to be lower than those for elementary fibres before weaving process [36, 37]. The same conclusion can be drawn from the friction but the scatter makes this statement unsure. This

Materials	**IFSS (MPa)**	**Friction stress (MPa)**
Untreated fibres	10.7±2.2	2.7±2.8
Treated fibres	7.0±2.0	3.1±1.0

Table 2: Values obtained from micro-droplet debonding tests

may be due to the nature of the fibre itself but most likely the twisting and weaving processes to make yarns and fabrics are harmful for fibre surfaces.

It is clear that the treatment has a significant effect on the IFSS estimated by these tests. The reduction is of approximatly 30%. As for the friction stress, it is not significantly modified by thermal treatment, even if the scatter seems to be reduced on the treated fibres. The interfacial bonding is thus significantly weakened after treatment. This result was indeed predictable since the surface free energy was lowered by a weakening of its polar component as it was proved in previous studies [7, 38] by tensiometric methods [39, 40]. Indeed, considering the bonding energy calculated with the surface energies of both constituents, the interfacial strength should be lowered as one of the surface energies have been lowered by treatment. However, it is not the only parameter that should be taken into account for interfacial interactions since the fibres roughness could have an effect on mechanical interlocking. The effect of the thermal treatment on the fibres surface roughness will be investigated in further studies using some specific techniques such as the Atomic Force Microscopy (AFM) [40].

3.2. Effective modulus and tensile strength of treated and untreated flax yarns

The density of flax yarns was found to be only slightly affected by the thermal treatment. Measured values for untreated and thermally treated flax fibres are 1.6098±0.0001 $g.cm^{-3}$ and 1.6113±0.0003 $g.cm^{-3}$, respectively. This slight difference can be ascribed to a relative higher cellulose content in the thermally treated fibres due to the thermal degradation of the less thermally stable constituents (for instance, hemicellulose) [41, 35]. Both values are higher than those usually reported for lignocellulosic fibres, that lie in the range 1.2-1.5 $g.cm^{-3}$ [42]. This is an indirect evidence of a high cellulose content with a high degree of crystallinity in the flax fibres under investigation, as the density of crystalline cellulose estimated from diffraction data is reported to be around 1.64 $g.cm^{-3}$ [43]. SEM micrographs of the two flax yarns types at different magnifications (Fig. 6 and 7) show that the yarn structure consists of many individual fibres twisted with a right-handed angle with respect to the yarn axis (Z-twist). From the images at higher magnification, it is evident that fibres cannot be considered as homogeneous entities, but exhibit variations in cross-section area and twisting angle which governs the fibre orientation in aligned natural yarn composites and therefore the resulting mechanical properties.

Both flax yarns showed similar twisting angles, equal to 14.05±0.87° and 15.01±1.83° for untreated and thermally treated fibres, respectively, while the apparent yarn diameter was found to exhibit a negative relationship with regard to the twisting angle: the apparent yarn diameter is reduced when the

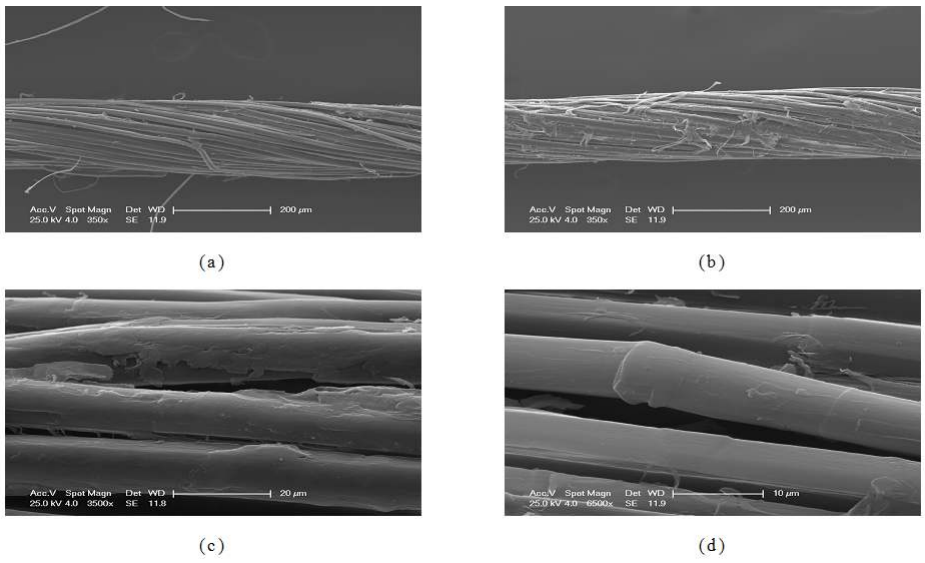

Figure 6: SEM micrographs of untreated flax yarn

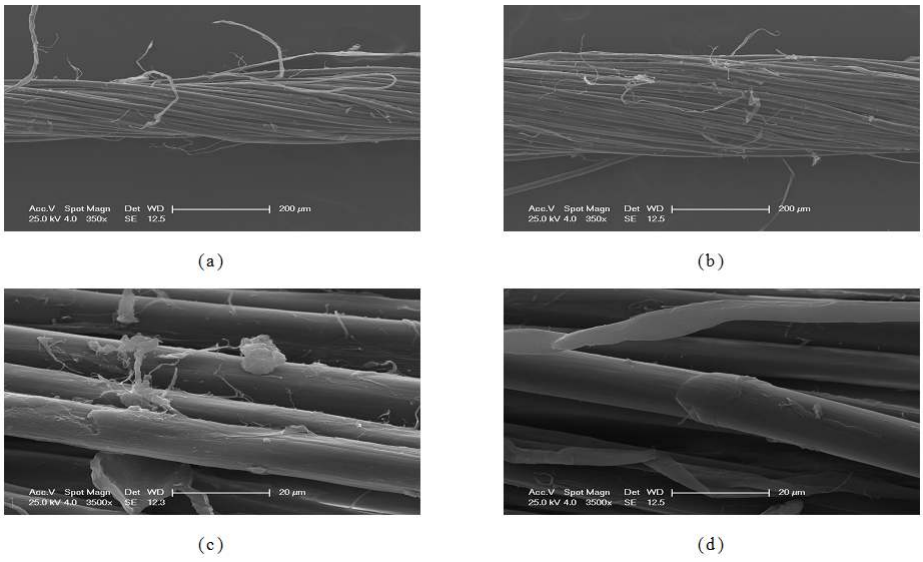

Figure 7: SEM micrographs of thermally treated flax yarn

fibre twisting angle is increased, as already observed for hemp yarns [44]. In particular, the yarn diameter was 250.92±41.01 μm and 235.35±41.80 μm for untreated and thermally treated flax yarns, respectively. The thermal treatment did not seem to alter in a significant way the surface features of the flax yarns. Despite the lower fibre twisting angle of untreated fibres, the mechanical properties were found to be markedly reduced as a result of the thermal exposure, as can be noted from the results summarized in table 3. It is to be noted that no clear effect of gauge length can be observed for breaking stress and strain at failure, while the values for untreated flax yarns compare quite favourably with those reported by Madsen et al. [44] for hemp yarns (19.4-19.5 N). In this work the authors reported also data for hemp yarns after a short thermal treatment (15 min) in the range 180-220 °C and a decrease in ultimate stress was detected. This behaviour is ascribed to the thermal degradation of cellulosic fibres, which is mainly due to the scission of cellulose chains and depolymerization [35, 45].

Specimen type	**Breaking force (N)**	**Strain-at-failure (%)**
UF 20mm	18.43±3.15	2.61±0.29
UF 30mm	17.04±3.18	2.33±0.26
UF 40mm	18.01±3.25	2.49±0.24
TF 20mm	11.14±1.99	1.66±0.17
TF 30mm	9.74±2.06	1.47±0.21
TF 40mm	10.88±1.94	1.52±0.14

Table 3: Mechanical properties of untreated (UF) and thermally treated (TF) flax yarns

The enhanced brittle behaviour and cellulose degradation caused by ther-

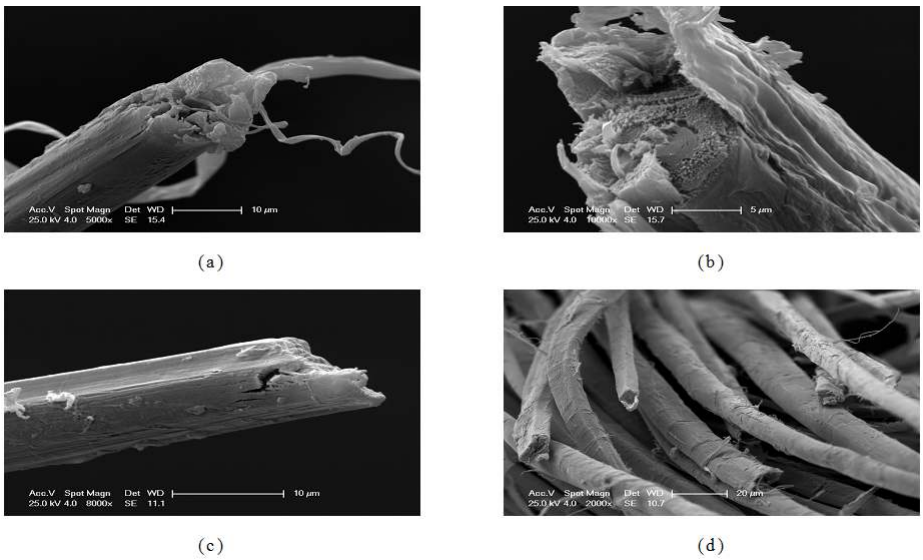

Figure 8: SEM micrographs of untreated flax yarns after tensile failure

mal exposure did not result in a change in the fracture modes of flax yarns, as confirmed by SEM images reported in Fig. 8 and 9, where a brittle behaviour with the absence of significant fibril splitting can be observed.

The reduction of mechanical properties at the yarn scale due to the thermal treatment resulted also in a significant strength loss of the whole quasi-UD fabric, which dropped from 1427.88±61.07 N for untreated flax, to 742.54±48.75 N after thermal treatment, thus confirming some degradation effect of the cellulose and of the cell wall materials.

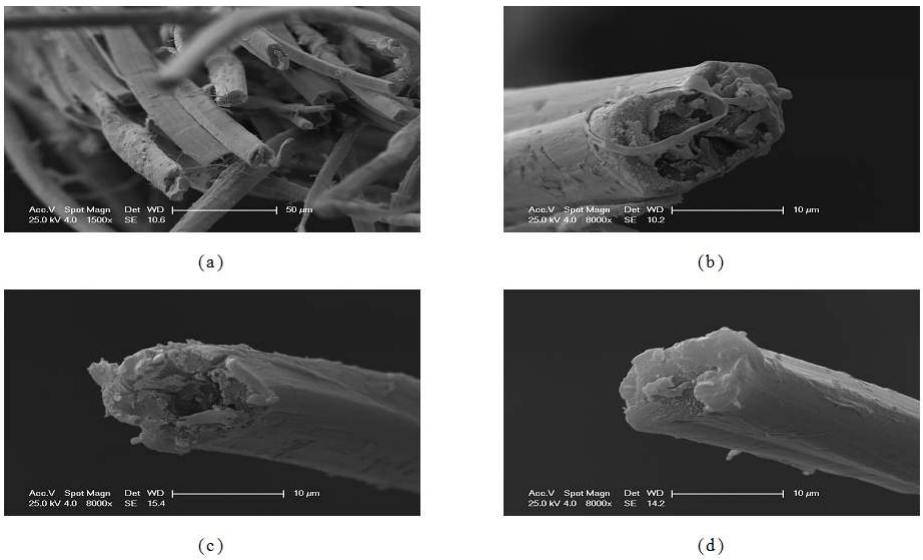

Figure 9: SEM micrographs of thermally treated flax yarns after tensile failure

3.3. Shear strength of treated and untreated flax reinforced composites

Five samples of composites reinforced by treated and untreated flax fabrics have been tested, and force-displacement graphs are shown in Fig. 10. It appears that the trend is not linear for the composite reinforced with untreated flax fibres. This could be due to gradual damage growth reflected in a loss of linear behaviour (the curve is linear up to a displacement of about 0.1 mm) [46].

On the contrary, for the composite reinforced with treated flax fibres, the trend is linear up to rupture, which is similar to composites reinforced with synthetic fibres. In addition, samples examined after rupture (Fig. 11) show a difference. Untreated flax reinforced composites show a non-conform rupture to SBST standard but closer to a tensile failure due to

bending. Composites reinforced with treated flax fibres exhibit a failure due to interlaminar shear, as expected in SBST tests for Inter Laminar Shear Strength (ILSS) characterisation.

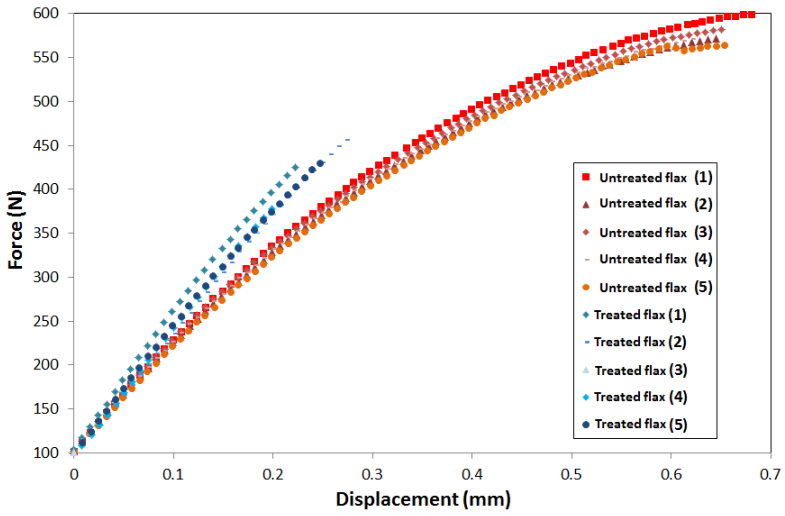

Figure 10: SBST experimental curves for composites reinforced by treated and untreated flax.

The shape of the force-displacement curves obtained with untreated flax is not common for this kind of test, and thus it is not possible to reach a direct conclusion. Therefore, a direct comparison between strengths of composites reinforced with both treated and untreated flax cannot be made. It can be seen that composites reinforced with untreated flax show a failure on the tensile side when the deflection becomes significant (Fig. 11 - left). Composites reinforced with treated flax show a linear behaviour under SBST and a breakage that complies with shear at neutral axis (Fig. 11 - right). The shear stress calculated can thus be considered as an ILSS at neutral axis

for composites reinforced by treated fibres only. These stresses for untreated fibre reinforced composites are given for information, keeping in mind the non-linear response.

Sample	τ_{xy}^{s} (MPa)	τ_{xy}^{r} (MPa)
Untreated flax (1)	16.5	29.9
Untreated flax (2)	16.5	28.5
Untreated flax (3)	16.2	28.2
Untreated flax (4)	16.6	28.2
Untreated flax (5)	16.7	28.2
Treated flax (1)	19.6	19.6
Treated flax (2)	21.2	21.2
Treated flax (3)	18.7	18.7
Treated flax (4)	16.8	16.8
Treated flax (5)	19.7	19.7

Table 4: Stress calculated at first damage (τ_{xy}^{s}) and at break (τ_{xy}^{r}), for each short beam shear test.

A maximum shear stress at break τ_{xy}^{r} has been calculated for treated flax. For untreated flax, the stress corresponding to the loss of linearity τ_{xy}^{s}, has also been calculated. Results for each test are presented in Tab 4. For treated flax, stress at break can be estimated at $\tau_{xy}^{r} = 19.2 \pm 1.6\ MPa$. For untreated flax, the first damage can be estimated at $\tau_{xy}^{s} = 16.3 \pm 0.4\ MPa$, but the stress at break is $\tau_{xy}^{r} = 28.8 \pm 0.7\ MPa$ due to the non-linear response. Considering the first value, it can be supposed that the better impregnation of the resin into the fibre network leads to an increase of the interface tougthness, but considering values at break, the conclusion would be the opposite. On the contrary, the non-linear response for untreated fibres can suggest a dif-

ferent mechanism of damaging such as, for example, slipping of fibres in the composites, leading to bending rather than shear failure. These results have to be commented along with other tests in order to draw a conclusion, that is why four-point bending tests have been performed on the same composites.

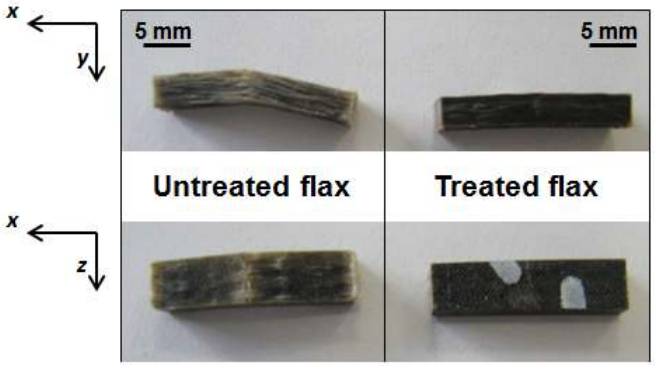

Figure 11: Samples after short beam shear tests

3.4. Flexural strength and modulus

Six tests for each type of reinforcements have been conducted on composite samples extracted from half plates manufactured by LCM. Fig. 12 shows the force-displacement curves, which compare quite favourably with data from literature [47]. Both types of composite have a non-linear behaviour, but for lower loads for untreated fibres. According to SBST results, composites reinforced with treated flax show a higher stiffness. And again, composites reinforced with treated flax have a more brittle behaviour compared to samples reinforced with untreated flax (Fig. 13).

It can be also seen in Fig. 12 that some samples reinforced with untreated flax show uncommon behaviour with a failure occurring out of the hinging points. This is likely due to severe impregnation problems. This kind of behaviour has not been observed in composites reinforced with treated flax, which appear to show more reproducible results.

Considering that the beam theory can be applied on those samples, it is possible to calculate an effective stiffness for each test. Fig. 14 summarizes the mean values of effective stiffness and the dispersion depicted as an envelope for both composites reinforced with untreated and treated flax. Samples that exhibited severe impregnation problems leading to a non conform mechanical behaviour have been removed from the calculated mean and dispersion.

It can be seen in Fig. 14 that the behaviour is neither constant nor linear, as it would be expected with conventionnal composites. But it is clearly

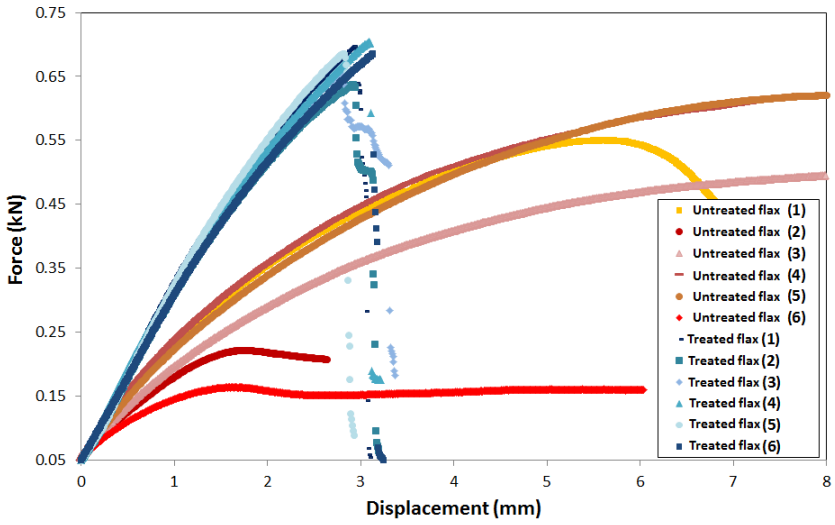

Figure 12: Experimental curves for composites reinforced by treated and untreated flax samples under 4 point bending.

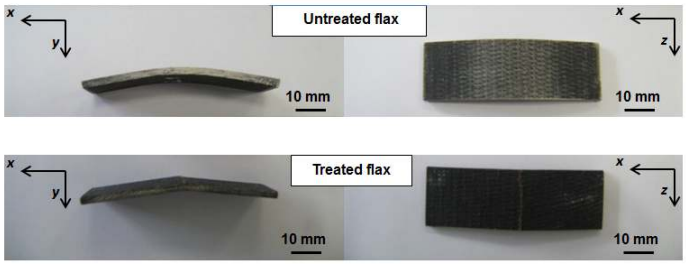

Figure 13: Samples after 4 points bending tests.

shown that composites reinforced with treated flax have a higher stiffness than the ones reinforced with untreated flax. Most important, maximal forces at failure that can be seen in Fig. 12 are reached for samples reinforced by treated flax, even if the strain at failure is lower. Corresponding stress at failure is more repeatable and higher compared to the same values calculated

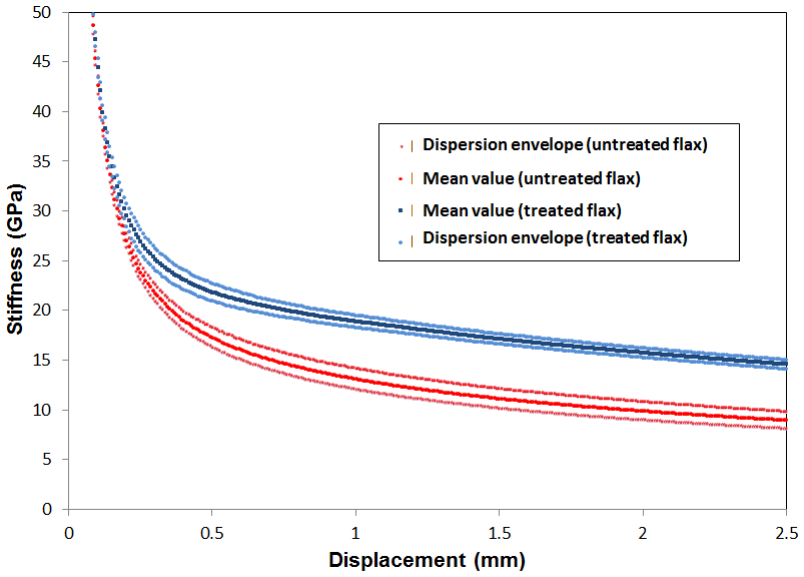

Figure 14: Mean stiffness against displacement during four-point bending test.

for composites reinforced with untreated flax fibres.

Following hypotheses of the beam theory for the entire duration of tests, stresses at failure σ_{xx}^{r} have been calculated (Tab. 5). Mean values for stresses at failure for untreated (excluding untreated samples with impregnation problem) and treated flax fibres are 153.7 ± 16.5 and 180.9 ± 6.7 MPa, respectively.

4. Discussions

It has been proved by all results obtained at the scale of fibres and yarns that they have been embrittled by the thermal treatment : the stiffness has not been modified but the strain and stress at failure have been severely

Sample	σ_{xx}^{r} (MPa)	**Sample**	σ_{xx}^{r} (MPa)
Untreated flax (1)	146.7	Treated flax (1)	186.1
Untreated flax (2)	~~59.2~~	Treated flax (2)	170.1
Untreated flax (3)	133.7	Treated flax (3)	175.5
Untreated flax (4)	168.2	Treated flax (4)	187.7
Untreated flax (5)	166.2	Treated flax (5)	183.0
Untreated flax (6)	~~43.7~~	Treated flax (6)	182.8

Table 5: Stresses at failure σ_{xx}^{r} calculated for each 4-point bending test.

lowered. Previous studies [7] have proved that the thermal treatment tends to reduce the polar component of the surface energy of flax fibres. This has also the effect of lowering the total surface energy of fibres. Following that statement, the associated bonding energy should also be lowered considering an interface between treated fibres and a polymer matrix (epoxy in this case) compared to a similar interface with untreated flax fibres. This assumption was verified with the interfacial bond strength that was measured in the present study. The apparent shear strength for treated flax fibres and epoxy resin has been lowered by 30% compared to untreated flax fibres with the same resin. According to a basic rule of mixture with similar fibre volume fractions, properties of the composites should be lowered too. However, previous studies [7] have shown that manufacturing composites by LCM processes like VARTM, both overall void content and micro-voids content in a composite reinforced with treated flax are lower than in a composite reinforced with untreated flax manufactured in the same conditions. Mechanical tests, performed on a plate manufactured in the same condition

than for the void content characterization from Pucci et al. [7], have shown that mechanical properties are enhanced in terms of stiffness and strength for composites reinforced with treated flax. This tends to prove that the impregnation process is more important than the intrinsic mechanical properties of both elementary fibres and interfaces with polymers. Thus, the quality of impregnation and the micro-void content in bio-based composites can not be neglected compared to the choice of the fibres themselves. This should lead to rethink the selection of fibres for bio-based composites which is, for now, only focused on the fibres mechanical properties, ignoring the manufacturing of the resulting composites.

5. Conclusion

Effects of the thermal treatment developed and used in previous studies on flax fibres [6, 7, 8] were investigated from the mechanical point of view. Previous results have proved [7] that the thermal treatment modifies the overall surface energy of elementary fibres. This has an impact on wetting dynamics [6, 8], thus modifying the void content of composites manufactuered by LCM processes both at the macroscopic and microscopic scale.

It was demonstrated during the present study that the thermal treatment has a negative impact on elementary fibres and yarns strength. The effective stiffness of fibres and yarns does not seem to be impacted by the thermal treatment.

It was also proved that the interfacial strength between treated fibres and

resin has been lowered compared to interfacial strength between untreated fibres and the same resin. This result was predictable because of the reduction in the overall surface energy of treated fibres, but it has been confirmed by mechanical tests.

Mechanical tests at the scale of composites have also been conducted. Composite plates, from which samples were taken, were manufactured by LCM processes, following the same methodology used for void content characterisation purpose in a previous work [7]. Results indicate that the mechanical behaviour of composites manufactured with both types of reinforcements is different, and the flexural strength was found to be higher for composites based on thermally treated fibres.

It can be concluded that even if elementary fibres and yarns have been embrittled and interface properties have been lowered after thermal treatment, since the wettability of fibres has been improved, the overall mechanical behaviour of composites manufactured by LCM is better with treated fibres. Thus, the impregnation during LCM, related to microvoids formation, is more critical than the mechanical behaviour of fibres, since the interface appears to be preponderant in the bio-composite mechanical properties. Rule of mixtures is thus not sufficient for describing composites mechanical behaviour if the fiber/matrix interface is not perfect. Further studies will focus on the description of the degree of imperfection of interfaces, including this parameter in simulations and models to correct the interfacial strength estimation and predict the behaviour of interfaces in a composite under service

conditions. A full characterisation of the environmental impact of the treatment compared to composites behaviour enhancement has also to be carried out.

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