



HAL
open science

Elaboration by Wrapping Process and Multiscale Characterisation of Thermoplastic Bio-Composite Based on Hemp/PA11 Constituents

Chaimae Laqraa, Manuela Ferreira, Ahmad Rashed Labanieh, Damien Soulat

► **To cite this version:**

Chaimae Laqraa, Manuela Ferreira, Ahmad Rashed Labanieh, Damien Soulat. Elaboration by Wrapping Process and Multiscale Characterisation of Thermoplastic Bio-Composite Based on Hemp/PA11 Constituents. *Coatings*, 2021, *Coatings*, 11, 10.3390/coatings11070770 . hal-04519817

HAL Id: hal-04519817

<https://hal.univ-lille.fr/hal-04519817v1>

Submitted on 25 Mar 2024

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.





L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Distributed under a Creative Commons Attribution 4.0 International License

Article

Elaboration by Wrapping Process and Multiscale Characterisation of Thermoplastic Bio-Composite Based on Hemp/PA11 Constituents

Chaimae Laqraa ^{*}, Manuela Ferreira , Ahmad Rashed Labanieh  and Damien Soulat 

Centrale Lille, Laboratoire de Génie et Matériaux, Gemtex, Ensait, F-59000 Roubaix, France; manuela.ferreira@ensait.fr (M.F.); ahmad.labanieh@ensait.fr (A.R.L.); damien.soulat@ensait.fr (D.S.)

* Correspondence: chaimae.laqraa@ensait.fr; Tel.: +33-7-67-49-99-22

Abstract: The present work investigates the potential of developing bio-composites based on thermoplastic polymers reinforced with natural fibres by using hybrid yarns. The hybrid yarns were produced by the wrapping technique, in which a multifilament of polyamide 11 (PA11) was wrapped around an untreated low-twisted hemp roving to produce a yarn with sufficient tenacity and stiffness for the next step of weaving. The tensile behaviour of the wrapped yarns was identified both in the dry- and thermo-state. Then, two different fabrics were woven and tested to study the influence of yarn densities and weave diagrams on the tensile and flexural properties. At this fabric scale, properties of fabrics made from hybrid yarns were compared with those of fabrics from a previous study made from 100% hemp roving. Composites made from these fabrics, with stacking of two cross-ply, were produced by thermocompression and characterised regarding mechanical strength.



Citation: Laqraa, C.; Ferreira, M.; Rashed Labanieh, A.; Soulat, D. Elaboration by Wrapping Process and Multiscale Characterisation of Thermoplastic Bio-Composite Based on Hemp/PA11 Constituents. *Coatings* **2021**, *11*, 770. <https://doi.org/10.3390/coatings11070770>

Academic Editor:
Fengwei (David) Xie

Received: 1 June 2021
Accepted: 24 June 2021
Published: 26 June 2021

Publisher's Note: MDPI stays neutral with regard to jurisdictional claims in published maps and institutional affiliations.



Copyright: © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<https://creativecommons.org/licenses/by/4.0/>).

Keywords: hybrid yarns; hemp; PA11; woven fabric; bio-based composite; mechanical characterisation

1. Introduction

Among plant fibres, flax (*Linum Usitatissimum* L.) and hemp (*Cannabis Sativa*) fibres are now the two most-produced bast fibres in Europe [1]. Due to their properties, such as important environmental advantages, good specific mechanical properties, and often a viable cost [1], these fibres have emerged as an alternative to synthetic fibres, and the use of plant fibre composites (PFCs) has become a market reality [2–4]. Despite numerous similarities (cell wall, thicknesses, and numbers of layers/sub-layers, biochemical composition, cellulose microfibril angle, MFA), these two bast fibres generally exhibit differences in their tensile properties and their global tensile behaviour. Flax fibres generally have slightly better tensile properties than hemp fibres, especially in terms of tensile strength and stiffness; on the other hand, they reach a lower tensile strain at failure than hemp [5]. If a lot of studies are conducted at the scale of fibres to explain these differences [1,5–7], few papers are dedicated to the development of hemp-continuous fabrics used as reinforcement of composite samples [4,8,9]. This deficit for hemp could be attributed to technological barriers, such as fibre separation and the alignment of fibres throughout the transformation process and consequently the unavailability of these products on an industrial scale [10]. Hemp fibres are naturally discontinuous; therefore, hemp reinforcements have so far been based on twisted yarns of staple fibres by means of long-staple spinning techniques, mainly ring spinning. However, the high twist level in the yarns leads to fibre misalignment in composite materials and thus reduced stiffness. Furthermore, the high twist level compacts the yarn section and reduces the inter-fibre gaps, making it very difficult for the resin to penetrate inside the yarn structure [11]. Therefore, the use of very low twisted yarns is advised for composite application [12,13]. However, a low twist implies poor inter-fibre cohesion, and the yarn loses its otherwise good weaveability properties which are due to good tenacity and low hairiness.

Along with this fibrous reinforcement, the challenge is to identify new combinations of raw materials for the production of green composites whose performance is good enough to propose their use in suitable applications. Among the various bio-based thermoplastic resins, polyamide 11 (PA11) is a semi-crystalline bio-polyamide produced using 11-aminoundecanoic acid derived from castor oil and has gained a special industrial interest due to a good combination of mechanical properties and chemical resistance [14]. In particular, PA11 exhibits good toughness, compared to other bio-based thermoplastic resins, such as, polylactic acid (PLA), which is often proposed as a matrix for bio-composites [15,16]. The natural fibre/PA11 combination has been used to study the performance of bio-composites. Haddou et al. [17] associated long bamboo fibres with PA11 films to analyse the tensile behaviour. Gourrier et al. [18] studied the tensile, impact, and thermal properties of unidirectional flax tape with PA11 films. In these studies, composites were made by film stacking, but other processes offer a route for efficient manufacturing of thermoplastic composites due to the reduced flow distance of resin in reinforcement to optimise impregnation. Awais et al. [19] compared tensile, flexural and impact behaviour of commingled fabrics (woven and knitted) based on jute/flax/hemp fibres with PP yarns. To improve the impregnation of thermoplastic resin into fibre yarns, Kobayashi et al. [20] used the micro-braiding method to mix hemp roving and PLA multifilament in the yarns. These micro-braided yarns were placed in a pre-heated moulding die for consolidation by compression moulding to produce composite specimens. Zhai et al. [21] compared yarn morphologies, structures, mechanical tensile properties, and braidabilities of commingled flax/PP yarns obtained by micro-braiding or wrapping methods. In the wrapping process [22,23], a thermoplastic multifilament is wrapped around hemp roving, resulting in increased inter-fibre friction and improved yarn cohesion. This manufacturing process was successfully used by Corbin et al. [8] to produce a commingled yarn based on hemp roving and PA12 multifilament and associated woven fabrics and composite samples. In all of these studies, although the mechanical and thermal properties of thermoplastic polymers are described, as in Di Lorenzo et al. [24], few papers [25] deal with the identification of the thermomechanical properties of commingled yarns, which can be essential to improve parameters of impregnation during the thermocompression process. This paper deals with hemp roving and PA11 multifilament and describes the wrapping process, and how the hybrid yarns are used to weave fabrics. Composite samples reinforced by these fabrics were manufactured by thermocompression. The tensile properties of these commingled yarns were studied according to temperature and strain rates, and the mechanical properties of the woven fabrics and the composite samples were identified.

2. Materials and Methods

2.1. Materials

An extruded PA11 multifilament yarn produced at GEMTEX Laboratory was used as the matrix material and as the wrapping material for the production of the hybrid yarn. The PA11 yarn was made from Rilsan[®] PA11 pellets supplied by Arkema, Colombes, France. This thermoplastic matrix has a density of 1.03 g/cm³, a melt temperature of 190–195 °C, and a glass transition temperature of 55–60 °C. Hemp roving with a twist of 37 turns per meter (TPM) was used as the core yarn of the hybrid yarns. This untreated roving was supplied by the Italian company Linificio e Canapificio Nazionale, Villa d'Almè, Italy. The main properties of these yarns are shown in Table 1.

Table 1. The main properties of the raw materials.

| Yarns | Density (g/cm ³) | Linear Density (Tex) | Twist Level (tpm) | Tenacity (cN/Text) | Deformation at Break (%) |
|-------------|------------------------------|----------------------|-------------------|--------------------|--------------------------|
| Hemp roving | 1.50 | 312 ± 19 | 37 ± 2 | 10.09 ± 2.48 | 3.67 ± 0.42 |
| PA11 | 1.03 | 111 ± 3 | – | 27.02 ± 0.95 | 35.02 ± 5.43 |

Their linear density was measured according to the NF G07-316 standard [26], the twist level according to the NF G07-079 standard [27], and tenacity at break according to the NF EN ISO 2062 standard [28].

2.2. Methods

2.2.1. Manufacturing of Hemp/PA11 Hybrid Yarns

The hemp/PA11 hybrid yarns were produced by the wrapping process, on a hollow spindle machine, Gualchieri e Gualchieri shown in Figure 1. In the wrapping process, the thermoplastic multifilament yarns (PA11) are wrapped around low twisted and untreated hemp roving. These wrapping yarns will melt during the thermocompression process, thus forming the matrix part of the composite material. The wrapping process is mainly used to increase the inter-fibre cohesion of the core yarn to sustain the tension loads applied during the transformation into reinforcement. Furthermore, the outer wrapped yarn protects the core yarn from rubbing aggression when passing machinery surfaces during these processes. These manufacturing loads represent the basic required weaveability conditions.

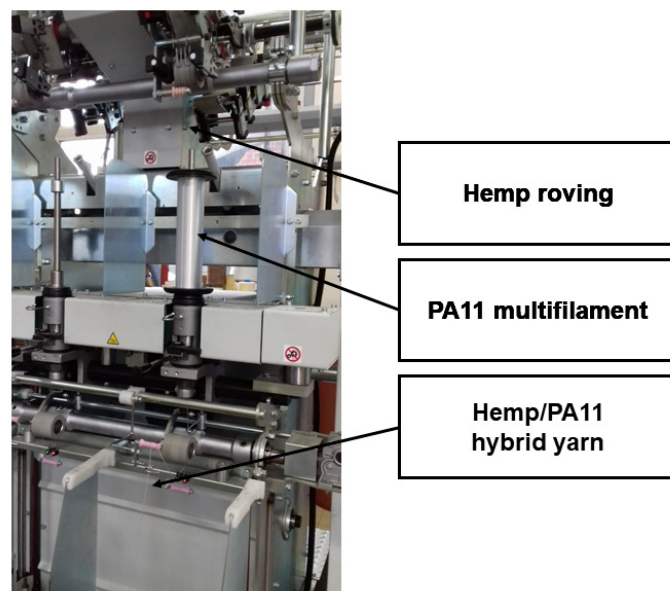


Figure 1. The wrapping process.

During production, the hemp roving passes through a roving condenser and drafting rollers and then is guided inside the hollow spindle together with the PA11. The PA11 is wrapped around the core roving by rotational movement of the hollow spindle to obtain the wrapped hemp/PA11 yarn. During the wrapping process, the ratio between hollow spindle rotational speed and yarn delivery speed determines the PA11 wrapping turns per meter and the mass proportion of two yarns for a given linear density. As output, to obtain the desired hybrid yarns with a mass ratio of hemp between 43% and 60% and a tenacity allowing its weaveability, the wrapping turn number and spindle speed are fixed at 500 tpm and 5000 turns/min, respectively. The linear densities of the hybrid yarns were measured according to the NF G07-316 standard [26], and the twist level according to the NF G07-079 standard [27].

2.2.2. Weaving Process with Hemp/PA11 Yarns

The hybrid yarns were woven on a Leclerc Weavebird manual loom in the GEMTEX laboratory (Roubaix, France). After the different preparation steps of the weaving process (warping and drawing-in), two woven fabrics with different weaving diagrams were produced, twill 6 weft effect and satin 6 weft effect. The two fabrics were woven with the

same hybrid yarns in both directions and have the same warp density at 6 yarns/cm but different weft densities, namely, 11 yarns/cm for twill 6 and 6 yarns/cm for satin 6.

The textile properties of these fabrics in terms of yarn densities, areal density, thickness, and air permeability were identified and will be presented later. Areal density was measured according to the NF EN 12127 standard [29], thickness according to NF EN ISO 5084 standard [30], and air permeability according to the NF EN ISO 9237 [31]. The crimp level of warp and weft yarns after weaving was measured for both fabrics according to the NF ISO 7211-3 standard [32].

2.2.3. Composite Manufacturing

Composite plates based on hemp/PA11 satin 6 fabric and hemp/PA11 twill 6 fabric were manufactured by thermocompression moulding on an Agila Press 100 kN hot-press (Menen, Belgium). The fabrics were cut into $300 \times 300 \text{ mm}^2$ squares and conditioned at a temperature of $23 \text{ }^\circ\text{C}$ and relative humidity of 50% for at least 24 h prior to the composite manufacturing. For the two types of fabrics, two cross-ply ($0/90^\circ$) were stacked, as shown in Figure 2a. Then, they were placed between two Teflon-coated plates. The melting temperature of PA11 is $190 \text{ }^\circ\text{C}$ and the degradation temperature of hemp roving is $276 \text{ }^\circ\text{C}$ [8]; therefore, to produce the composite plates, the process temperature was fixed at $200 \text{ }^\circ\text{C}$ to preserve the hemp fibre properties and avoid its degradation. The composites were then prepared by pressing the layers at a temperature of $200 \text{ }^\circ\text{C}$ and according to the cycle presented in Figure 2b. These temperature and pressure cycles are specific to the combination of constituents involved (hemp/PA11) in the commingled yarn and to the stacking chosen in this study (two cross-ply of woven fabrics). Specimens for the mechanical testing were cut according to the appropriate standard, given later, and before the testing, they were conditioned for at least 24 h at $23 \text{ }^\circ\text{C}$ and 50% relative humidity.

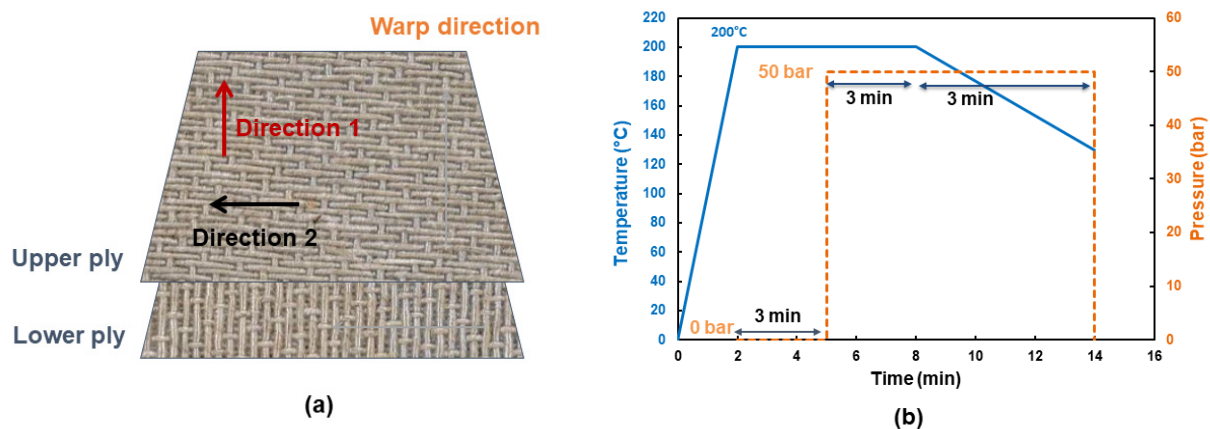


Figure 2. (a) The stacking of fabrics, adapt from: [8]; (b) the thermocompression moulding cycle.

2.3. Characterisation Steps

2.3.1. Properties of the Hybrid Yarns

First, dry-state tensile tests were performed on the hybrid yarns at different test speeds in order to investigate the influence of this parameter on the behaviour of the hybrid yarns. The tests were performed on an MTS Criterion 45 universal tensile apparatus (Eden Prairie, MN, USA) according to the NF EN ISO 2062 standard [28] with a gauge length of 200 mm, without pretension, and with three different test speeds: 20, 100, and 200 mm/min. For each crosshead speed, the test was repeated 20 times. In addition, tensile tests were also conducted on the hemp roving, but only at 200 mm/min test speed, to compare its behaviour with the hybrid yarns.

Then, thermo-state tensile tests were performed on these yarns in order to study the effect of temperature on the yarn properties and to understand the behaviour of these hybrid yarns at the chosen temperature. Tensile tests were then conducted on the same

machine as the dry-state tests, using an isothermal oven. Due to the size of the oven, the gauge length of the yarn was 100 mm, the crosshead speed was 100 mm/min, and the temperature was set at 50, 70, 100, 120, and 150 °C. The single yarn was first inserted in the oven and clamped between the two jaws, and then the desired temperatures were set. Once the temperature was reached and stabilised, the test started.

2.3.2. Properties of the Produced Fabrics

Tensile tests were performed on the woven fabrics according to the NF EN ISO 13934-1 standard [33] and were carried out on an MTS Criterion 45 machine at ambient temperature. The two main directions of the woven fabrics were tested and for each direction, five samples were used, with a length of 300 mm and a width of 50 mm. For the tensile test, the gauge length was 200 mm, the test speed was 20 mm/min, and the preload was 5 N. To present the properties of the two woven fabrics, we studied the maximum load/yarn and strain at maximum load for each fabric in order to eliminate the effects of the density of yarns. The bending rigidity of the fabrics was also identified by using a cantilever apparatus according to the ISO 4604 (05) standard [34], with the same samples used for the tensile tests (Figure 3). As described in the literature [35,36], the fabric sample is progressively advanced until the end, under its own weight, is in contact with the inclined plane at 41.5°. Then, the overhang length of the fabric is measured and the bending stiffness is computed according to Equation (1) as follows:

$$G = 9.81 \times m_s \times \left(\frac{l_m}{2}\right)^3 \quad (1)$$

where G is the bending stiffness coefficient (N·mm), m_s is the areal density of the fabric (g/m^2), and l_m is the overhanging length (m).



Figure 3. Cantilever test bench.

2.3.3. Properties of the Composite Plates

The tensile and flexural behaviours of hemp/PA11 composites were tested in an MTS Criterion 45 universal testing machine. The tensile properties of the composite specimens were performed according to the ASTM D3039-00 standard [37], while the flexural properties were tested according to the NF EN ISO 14125 standard [38]. The tensile tests were carried out over a gauge length of 150 mm and a test speed of 1 mm/min. The reported modulus was calculated (between 0% and 1% strain) for all composite samples. Thus, the strength and strain at break were calculated from the recorded force–displacement curves. For flexural behaviour, tests were performed at a test speed of 1 mm/min, with specimens of 25 mm width and 64 mm span length. For both tensile and flexural tests, five samples were tested in the two main directions of the composite plates which are

direction 1 (associated with the warp direction of the upper ply) and direction 2 (associated with the weft direction of the upper ply).

3. Results and Discussion

3.1. Yarn Properties

3.1.1. Textile Properties of the Hybrid Yarns

The measured textile properties of the hybrid hemp/PA11 yarns and hemp roving are listed in Table 2. The hybrid yarn has a higher linear density than hemp roving, as a result of the addition of the thermoplastic multifilament PA11 (111 ± 3 Tex). Moreover, adding the wrapped PA11 multifilament leads to a decrease in the hybrid yarn hairiness. This decrease is due to the removal of most impurities during the wrapping process. Figure 4 presents the visual aspect of the core untreated hemp roving and the wrapped yarn. There are no more defects on the surface of the hybrid yarn in comparison to the hemp roving. After wrapping, the yarn structure becomes more compact and uniform, and its section is more circular. In addition, the wrapping process preserves the structure of the core roving and its properties by creating a mechanical bond between the two materials, unlike conventional methods used to improve the interfacial bond with polymer matrices, which use chemical, physical or biological treatments instead [39]. This hybrid yarn has, in weight, 50% of hemp fibre and 50% of PA11.

Table 2. The textile properties of the hybrid yarns and hemp roving.

| Yarns | Linear Density (Tex) | Twist Level (tpm) | Hairiness ($H \pm sh$) | Hemp Fibre Mass Fraction (%) |
|--------------|----------------------|-------------------|--------------------------|------------------------------|
| Hybrid yarns | 486 ± 9 | 400 | 10.05 ± 2.86 | 50 |
| Hemp roving | 313 ± 19 | 37 ± 2 | 18.96 ± 3.31 | 100 |



Figure 4. The visual aspect of a hybrid yarn and a hemp roving.

3.1.2. Dry-State Tensile Behaviour

• Comparison of Hemp Roving and Hemp/PA11 Hybrid Yarn Behaviour

Figure 5 shows the tenacity–strain curve of the hybrid yarn and the hemp roving at ambient temperature and at a speed of 200 mm/min. The behaviour of the hybrid yarn has the first peak of tenacity at a strain of around 3%, corresponding to the roving breakdown. At this first peak, tenacity reaches 8 cN/Tex. This peak is followed by a high deformation phase before the full breakdown of the hybrid yarn at a strain of 35% (Figure 6a), the same

as that of the PA11 multifilament (Table 1). This part of the curve can be attributed to the elongation of the PA11 multifilament, with compacting of the hemp roving around which it is wrapped, as the load increases. By increasing the elongation, occasional slippage can occur between the PA11 filaments and the broken hemp roving, and the filaments can be stretched unevenly, leading to their rupture that appears on the tenacity–strain curve as a non-smooth part. In this part of the curve, some small breaks are detected by the load cell and involve a high standard deviation between samples on the measured load.

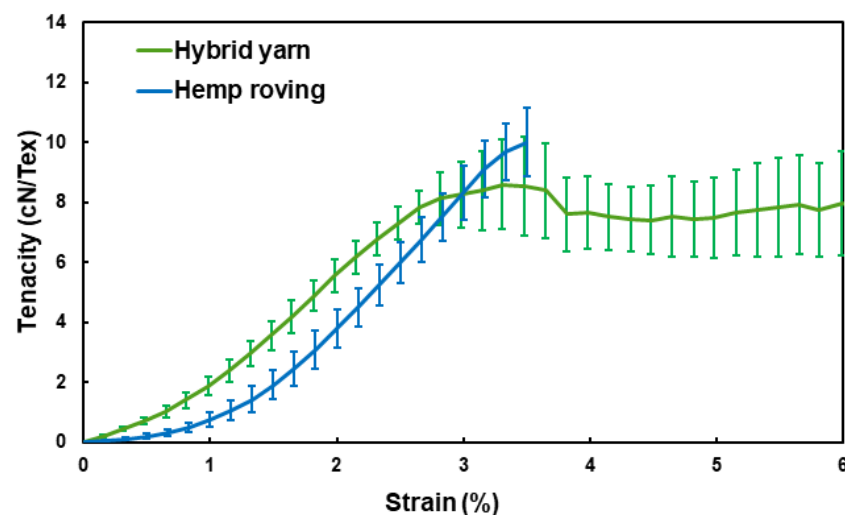


Figure 5. The tensile behaviour of the hybrid yarn and hemp roving alone.

From these results, it can be concluded that the wrapped multifilament PA11 does not induce a significant increase in the roving tenacity and rigidity. During the first phase (low tenacity), the roving curve corresponds to the fibre redressing in the axial load direction. This results from the roving tension applied during the wrapping process. Thus, even if the hybrid yarn did not reach the required tenacity for weaving, which is 15 cN/Text, it can be woven because adding PA11 during the wrapping process increases the inter-fibre cohesion and protects them from damage during the weaving process. Weaveability of natural fibre reinforcement can be also improved by other methods, as described in [8,40,41].

- Tensile Behaviour of the Hybrid Yarns at Different Test Speeds

Figure 6a shows the tensile behaviour of the produced hybrid yarns for different test speeds (20, 100, and 200 mm/min) at ambient temperature. For these different speed tests, the shape of the tenacity–strain curve remains the same. High rigidity is observed in the first phase up to a peak, followed by a non-smooth part characterised by a high elongation and fluctuation of the tenacity around the tenacity of the first peak. However, a dependence of the tenacity at the first peak on the test speed can be seen in Figure 6b. At the 200 mm/min speed, the tenacity is 70% higher than at 20 mm/min. In addition, the behaviour of yarns at 100 and 200 mm/min is almost the same: in the range of 0 to 5% strain with a difference of 10% for the tenacity. These results match the results obtained in previous studies conducted on commingled glass/PP and on flax/PA12 yarns at different test speeds and in which the tenacity increases with increasing the test speed [25,35].

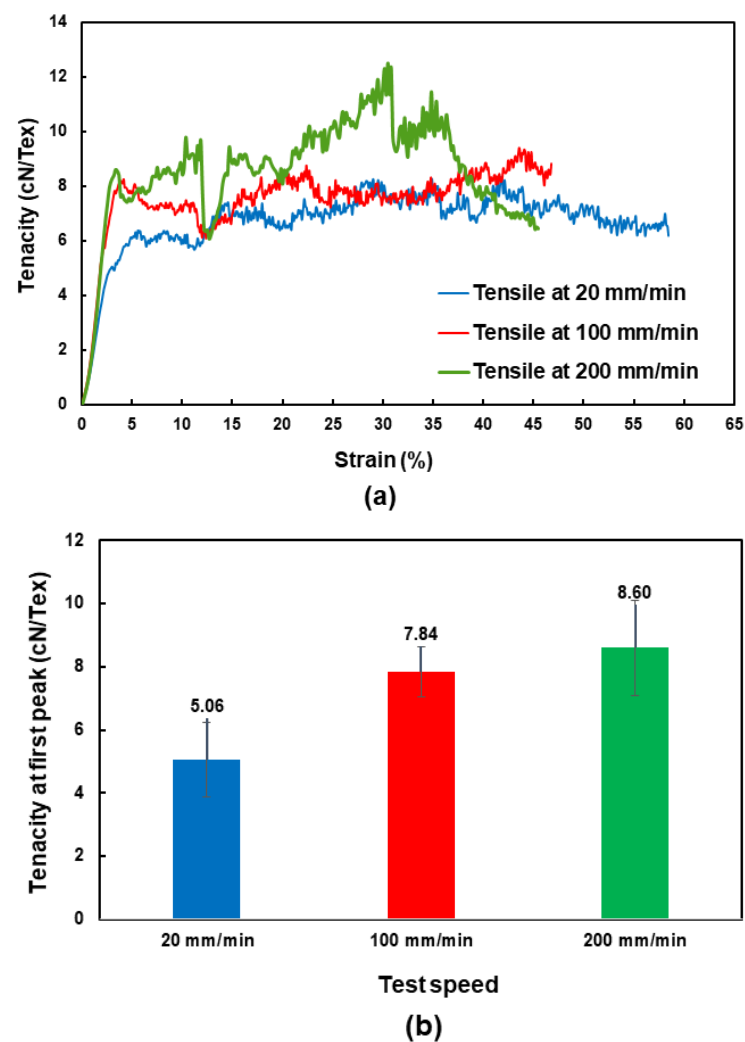


Figure 6. (a) Tenacity–strain curve of hybrid yarns at different test speeds; (b) tenacity at first peak at different test speeds.

3.1.3. Thermo-State Tensile Behaviour

The tensile behaviour of the hemp/PA11 yarns at different temperatures is shown in Figure 7a. The tensile tests for the different temperatures were performed at the same test speed of 100 mm/min. Depending on the temperature, the results obtained can be divided into three groups, namely, around the glass transition of PA11 ($T = 50\text{ }^{\circ}\text{C}$), above the glass transition ($T = 70, 100,$ and $120\text{ }^{\circ}\text{C}$), and below the melting temperature ($T = 150\text{ }^{\circ}\text{C}$). The first phase of the different curves at the studied temperatures differs from the one at room temperature, as does the second phase. In contrast, the curves obtained at $T = 100$ and $120\text{ }^{\circ}\text{C}$ show almost similar trends in the first and second phases. Hence, the tensile behaviour of these yarns depends strongly on the temperature setting. When the temperature is increased to $50\text{ }^{\circ}\text{C}$ (upper glass transition), the tenacity of the wrapped yarn decreases as a result of the modification in the multifilament around the hemp roving, which becomes softer by passing the transition temperature and the interaction between hemp fibres and PA11 filaments changes. As the temperature increases (but is still far from the melting temperature), the PA11 filaments keep slipping over the core hemp roving. This is because adhesion between the filaments and fibres increases with increasing temperature. This increased adhesion strengthens the yarn and increases its tenacity, Figure 7b. Then, at $T = 150\text{ }^{\circ}\text{C}$, the maximum tenacity of the first phase decreases significantly because the temperature nears the melting temperature, which strongly affects the structure of the hybrid yarn: the PA11 begins to stick locally to hemp fibres without creating a continuous

medium to distribute the efforts between the hemp fibres. This was observed on the structure of the yarn once extracted from the climatic chamber after the tests. In the sections where the yarn had become solid in comparison to the original wrapped yarn, it accordingly displayed decreased extensibility.

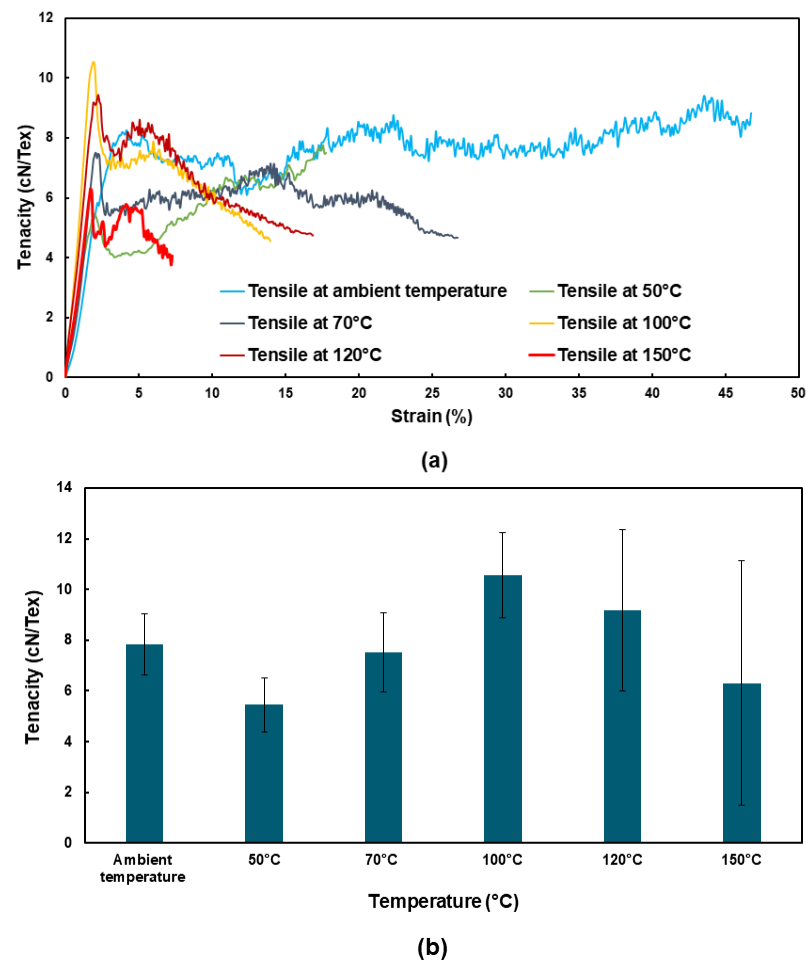


Figure 7. (a) Tenacity–strain curve of the hybrid yarns at different temperatures; (b) tenacity at first peak at different temperatures.

According to previous results obtained on flax/PA12 hybrid yarns [25], when the temperature increases and nears the melting value, the strain at break decreases, compared to the strain obtained at ambient temperature, whereas the deformation of hybrid yarns increases with increasing temperature above the melting temperature because below this temperature, the PA12 is fluid enough to increase the slippage between broken fibres. The same trend is observed in the presented results for tests below the melting temperature (Figure 7a). This can be attributed to the helical path of the wrapped PA11 filaments and the change in the interaction properties with the hemp fibres. At the ambient temperature, the spiral filaments stretch until they become aligned with the longitudinal axis of the yarn (unfolding). However, as the temperature increases, the PA11 filaments stick more locally to the hemp fibres, and that leads to concentrating the deformation at weak places on the wrapped yarn instead of having complete unfolding. Figure 7b shows the evolution of the tenacity at the first peak at different temperatures. As explained before, this tenacity increases initially with increasing temperature up to 100 °C and then decreases, but at higher temperatures (120 and 150 °C), the standard deviation is greater than at lower temperatures (50° and 70 °C), which is mainly due to the irregular behaviour of these yarns at high temperature.

3.2. Fabric Properties

3.2.1. Textile Properties

At this scale, the properties of the fabrics developed in this study will be compared to the properties of fabrics made in a previous study from 100% hemp roving. Table 3 shows the textile properties of these produced woven fabrics. For fabrics based on hybrid yarns, the twill 6 fabric has the higher weft density, its areal density and thickness are greater than satin 6. The areal density of this woven fabric depends mainly on the weft density, which, in turn, depends on how the fabric is packed during weaving and on the weave diagram. In addition, these fabrics are made of hybrid yarns that are heavier than conventional 100% natural fibre yarns. However, the air permeability of satin 6 is five times greater than that of twill 6, which highlights the higher inter-yarns gap, and it is correlated with a lower areal density, in comparison with twill 6, since it contains higher weft density. On the other hand, the fabrics in this study are heavier and thicker than the satin 6 and twill 6 of the previous study [42] manufactured with only hemp roving. This difference could be explained by the difference in the linear density of yarns which is higher for hybrid yarns (486 Tex) than for 100% hemp roving (259 Tex). In addition, even if the areal density of commingled fabrics is higher than that of 100% hemp fabrics, the air permeability is still higher. This could be due to the structure of the yarn used. In the case of hybrid yarns, the structure is more compact and the surface of the yarn contains less fibrils, compared to the structure of the roving, which is hairier, flat, and contains more fibrils.

Table 3. The textiles properties of the woven fabrics of this study and Corbin et al. [42] study.

| Fabric Pattern | Twill 6 from Hybrid Yarns | Satin 6 from Hybrid Yarns | Satin 6 with 100% Hemp Roving [42] | Twill 6 with 100% Hemp Roving [42] |
|--|---------------------------|---------------------------|------------------------------------|------------------------------------|
| Warp density (yarns/cm) | 6 | 6.4 | 6 | 6 |
| Weft density (yarns/cm) | 10 | 6 | 9.5 | 9.5 |
| Areal density (g/m ²) | 827 ± 15 | 583 ± 16 | 426 ± 8 | 402 ± 3 |
| Thickness (mm) | 2.19 ± 0.05 | 2.13 ± 0.04 | 1.55 ± 0.05 | 1.59 ± 0.06 |
| Air permeability (L/m ² /s) | 605 ± 91 | 3485 ± 239 | 401 ± 46 | 670 ± 115 |
| Warp crimp (%) | 0.67 ± 0.16 | 2.38 ± 1.11 | 2.24 ± 0.20 | 2.56 ± 0.34 |
| Weft crimp (%) | 2.65 ± 0.47 | 1.20 ± 0.30 | 1.90 ± 0.30 | 1.58 ± 0.22 |

3.2.2. Mechanical Properties

- Tensile Behaviour

Figure 8a shows the tensile properties of the two commingled fabrics and the two fabrics made from 100% hemp roving both in warp and weft direction. The tensile load is given in cN/yarn/Tex to remove the effects of the density and linear density of yarns and concentrate on the effect of the weave pattern and tenacity at the break of the roving. For the commingled satin 6 and twill 6 fabrics, a small difference in maximum tensile load is noted between the two directions. In terms of breaking strain, the weft direction of twill 6 is 55% higher than the warp direction, whereas the strain of satin 6 is similar for the two directions even if the crimp level is higher in the warp direction (Figure 8b). The high strain at break of twill 6 weft direction results from the higher crimp level of weft yarns as the warp yarns are under higher tension than weft yarns during the weaving process. Thus, the twill 6 fabric exhibits better tensile properties, both in warp and weft direction, than satin 6, and this difference can be explained by the difference of weave diagram between the structure and the arrangement of yarns inside the structure. In comparison to the previous study [42], it can be seen that twill 6 and satin 6 structures made from 100% hemp roving exhibit better maximum loads than twill 6 and satin 6 made from hybrid yarns, in the two directions for satin 6 structure and only in weft direction for twill 6 structure. That is mainly attributed to the low tenacity of these hybrid yarns (8 cN/Tex), compared to the

tenacity of 100% hemp roving (24 cN/Text) [42]. Furthermore, the higher hairiness level of hemp roving conduces higher inter-roving friction, leading to higher maximum loads. Strain at maximum load of the woven fabrics is balanced between the two directions for these structures except for commingled twill 6 structure, which has a high strain at break in the weft direction in comparison with the other structure.

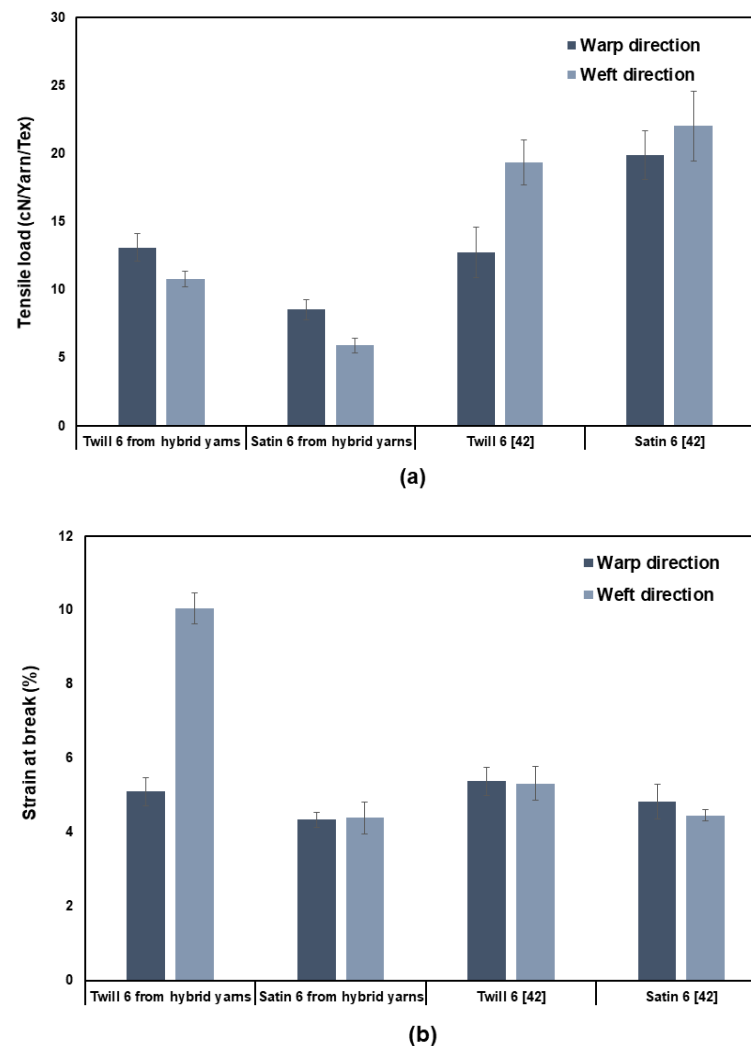


Figure 8. (a) The tensile load (in cN/yarn/Text) of the woven fabrics; (b) the tensile strain at break of the woven fabrics.

- Flexural Behaviour

Figure 9 shows the flexural rigidity of the two fabrics. The two fabrics have the same warp density and differ only by their weft density and structure. Twill 6 exhibited better rigidity than satin 6 in the two directions, and this difference is mainly due to the high areal density of the fabric (42% higher than that of satin 6) and its yarn density in the weft direction. The flexural rigidity depends strongly on those two parameters. However, a high weft density and linear density result in a heavy fabric. The flexural rigidity also depends on the crimp level of yarns inside the structure. In the case of twill 6 fabric, the shrinkage of weft yarns is greater than warp yarns, and that led to higher rigidity in the weft direction of the fabric than in the warp direction. By contrast, for satin 6 fabric, the warp yarns exhibit higher crimp, which results in a higher rigidity in this direction than in the weft direction.

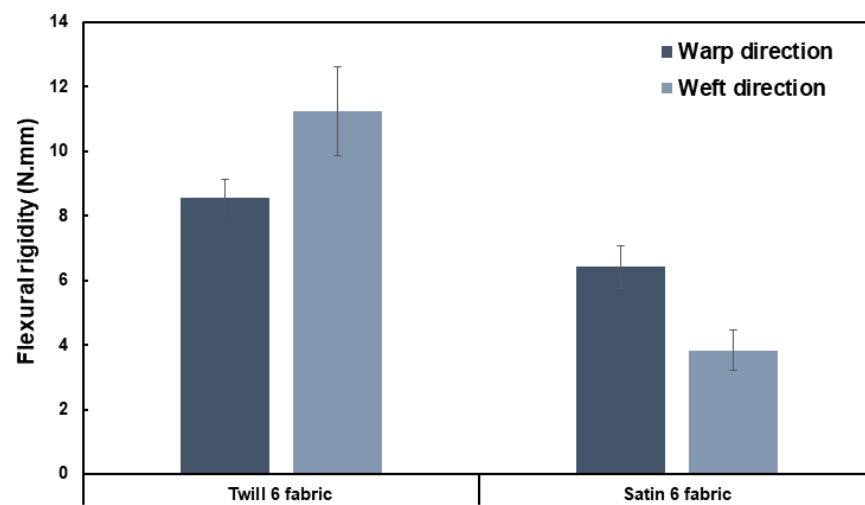


Figure 9. The flexural rigidity of the two fabrics.

3.3. Composite Properties

3.3.1. Composition of the Composite Plates

The obtained physical compositions of the two composite plates are summarised in Table 4. The two types of composites were produced by stacking plates two cross-ply. As a result, the thickness of twill 6 hemp/PA11 composite (C2) is 28% higher than composite made from satin 6 (C1). The weight of the two types of composites is the same and differs from that in the original (50% of hemp and 50% of PA11), which is explained by the loss of PA11 during the compression process. By contrast, the volume of fibre in the C2 is 22% greater than C1.

Table 4. The physical properties of the two composites.

| Composite Plates | Thickness (mm) | Density (g/cm ³) | Fibre Mass Fraction (%) | Fibre Volume Fraction (%) |
|--|----------------|------------------------------|-------------------------|---------------------------|
| Hemp/PA11 Satin 6 composite plate (C1) | 1.01 ± 0.01 | 1.01 ± 0.01 | 52 ± 1 | 35.1 ± 0.1 |
| Hemp/PA11 Twill 6 composite plate (C2) | 1.39 ± 0.01 | 1.24 ± 0.03 | 52 ± 0.1 | 42.8 ± 1.2 |

3.3.2. Tensile Properties of the Composite Plates

The tensile strength, strain at break, and modulus of the two types of composites are shown in Figure 10. For each structure, the properties are almost the same for both directions of the composite plates, while at the same time, the tensile properties differ slightly per composite. The tensile stress and modulus of C1 exceed those of C2, and the opposite is the case for strain. Even if the fibre content of C1 composites is lower than that of C2, their tensile properties are higher. At the fabric scale, the twill 6 fabric exhibited better properties, both in tensile and flexural rigidity, whereas this is no larger than the case at the composite scale.

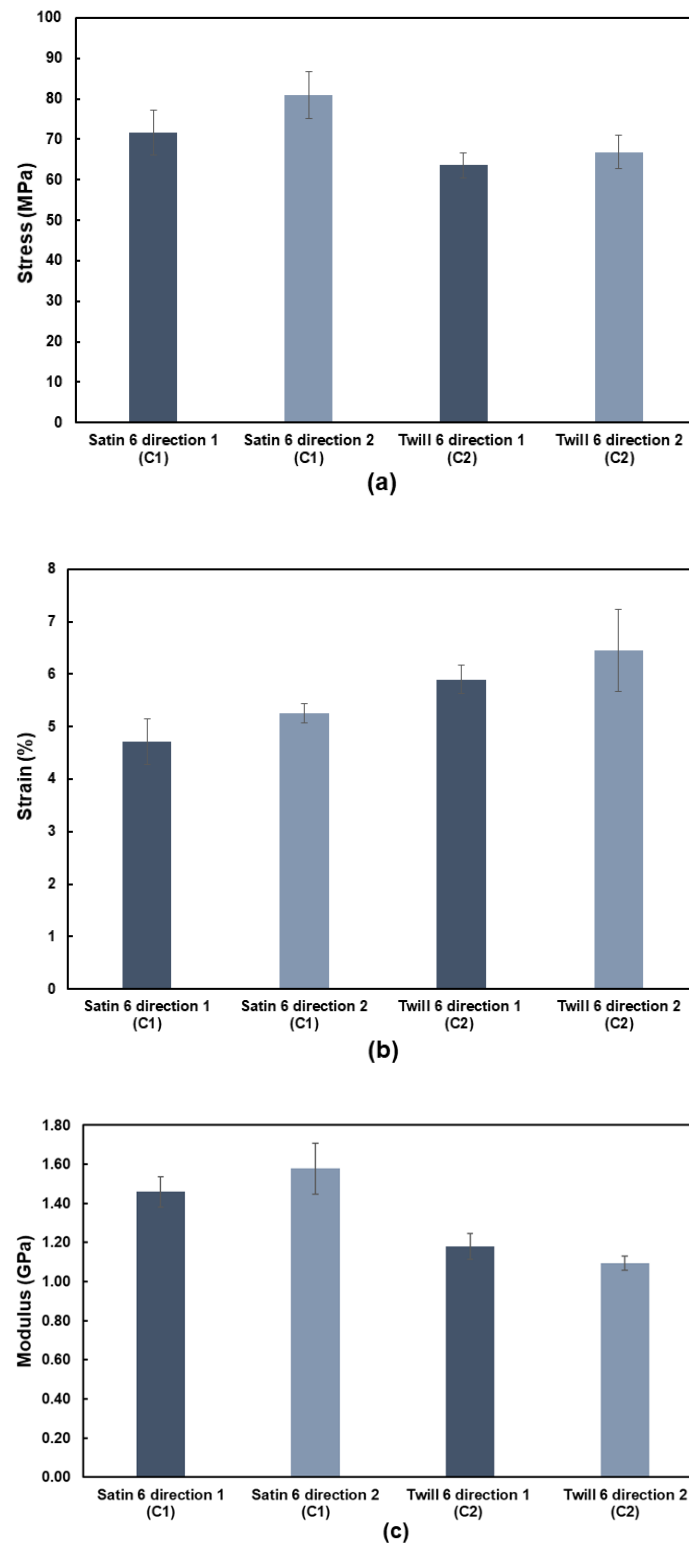


Figure 10. (a) Tensile strength of composite materials; (b) strain at the maximum strength; (c) modulus.

3.3.3. Flexural Behaviour of the Composite Plates

The results of the flexural testing are shown in Figure 11. For the satin 6 composites (C1), direction 2 presents better properties than direction 1, while for twill 6 (C2) the opposite is the case. The flexural strength is not balanced between the two directions of the composite plates even if the stacking is balanced. This difference is explained by the

arrangement and the orientation of the yarns inside the structure of the composite. In the case of satin 6 composite plates (C1), the float yarns of direction 2 are located outside the specimen, and that provides additional rigidity to this direction. The same phenomenon happens to direction 1 of the twill 6 composite plates (C2). This behaviour has been identified in previous work within the same project [8] and confirmed in this study. The arrangement of yarns inside the composite plates depends strongly on the nature of the fabric structure used and on the way of stacking the different layers.

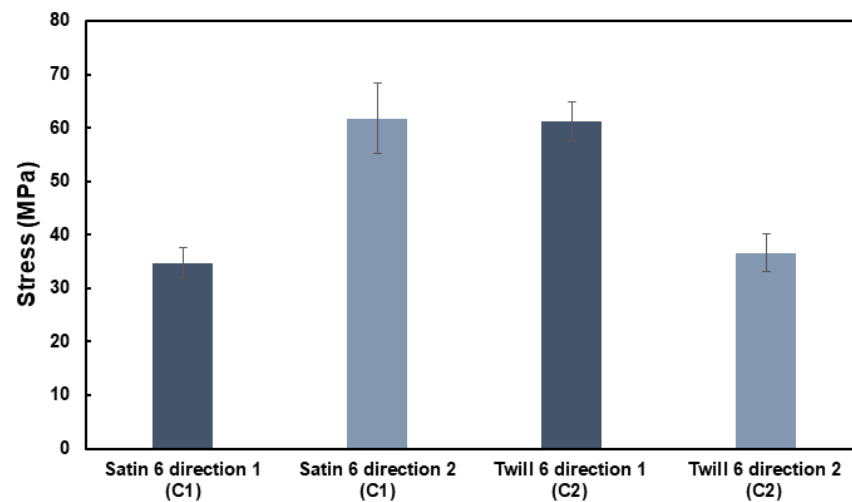


Figure 11. The flexural strength of the two composite materials.

4. Conclusions

This study investigated the multiscale analysis of bio-based composite materials made of hemp/PA11 commingled yarns. Understanding the different manufacturing stages and the influence of process parameters on yarn, fabric, and composite properties allows materials to be produced which better fit the final application requirements.

In this work, the hybrid yarns were produced by the wrapping process on a hollow spindle machine by wrapping a thermoplastic PA11 multifilament around an untreated hemp roving in order to produce yarns with sufficient tenacity (allowing them to be woven) and with a fibre content of no less than 40%. This process improves the hemp roving structure, which becomes more compact and less hairy.

At this yarn scale, tensile tests were initially conducted at different test speeds including the speed involved during fabric testing in order to investigate its influence on the mechanical properties, then at different temperatures including a temperature in the range of the glass transition temperature of the multifilament. Results from this test show the dependence of yarn properties on test speed and temperature, which is mainly due to the nature of the multifilament used.

Then, these yarns were used in weaving to produce two different fabrics with approximately the same warp density and different weft densities and weave diagrams. The textile and mechanical properties of these fabrics were determined, and the results show the dependence of these properties on the production parameters: preform properties in terms of maximum load and strain are either balanced or unbalanced between the warp and weft directions.

At the composite scale, tensile strength and stiffness for each structure are almost balanced between the two main directions of the composite plates. In addition, composite made from a satin 6 fabric shows an improvement of its mechanical properties even though at the fabric scale, the twill 6 fabric presented better properties.

The aim of future work will be the testing of other parameters at each scale in order to produce a light structure adapted to the end-use application. Moreover, the thermomechanical behaviour of hybrid yarns and commingled fabrics will be tested across a range of

temperatures, including the melting temperature of PA11, in order to better understand the behaviour of these materials at high temperatures. Thus, the hydrophilic behaviour of these hybrid yarns will be characterised and compared with that of hemp roving [43].

Author Contributions: Investigation: C.L.; writing—original draft preparation: C.L.; validation; supervision: M.F., A.R.L. and D.S.; writing—review and editing: M.F., A.R.L. and D.S.; All authors have read and agreed to the published version of the manuscript.

Funding: This project was funded by “Bio-Based Industries Joint Undertaking” under the “European Union’s Horizon 2020” Research and Innovation Program with Grant Agreement No. 744349—SSUCHY Project and Hauts de France Region.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Data are contained within the article.

Acknowledgments: The authors gratefully acknowledge the Italian company Linificio and Canapificio Nazionale (LCN) for providing the hemp roving used in this study.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Bourmaud, A.; Beaugrand, J.; Shah, D.U.; Placet, V.; Baley, C. Towards the design of high-performance plant fibre composites. *Prog. Mater. Sci.* **2018**, *97*, 347–408. [[CrossRef](#)]
2. CELC. *Flax and Hemp Fiber Composites, a Market. Reality: The Biobased Solutions for the Industry*; JEC Group: Paris, France, 2018; ISBN 978-2-490-26300-4.
3. Mohanty, A.K.; Vivekanandhan, S.; Pin, J.-M.; Misra, M. Composites from renewable and sustainable resources: Challenges and innovations. *Science* **2018**, *362*, 536–542. [[CrossRef](#)] [[PubMed](#)]
4. Sala, B.; Gabrion, X.; Trivaudey, F.; Guicheret-Retel, V.; Placet, V. Influence of the stress level and hygrothermal conditions on the creep/recovery behaviour of high-grade flax and hemp fibre reinforced green epoxy matrix composites. *Compos. Part A Appl. Sci. Manuf.* **2021**, *141*, 106204. [[CrossRef](#)]
5. Del Mastro, A.; Trivaudey, F.; Guicheret-Retel, V.; Placet, V.; Boubakar, L. Investigation of the possible origins of the differences in mechanical properties of hemp and flax fibres: A numerical study based on sensitivity analysis. *Compos. Part A. Appl. Sci. Manuf.* **2019**, *124*, 105488. [[CrossRef](#)]
6. Baley, C.; Gomina, M.; Breard, J.; Bourmaud, A.; Davies, P. Variability of mechanical properties of flax fibres for composite reinforcement. A review. *Ind. Crop. Prod.* **2020**, *145*, 111984. [[CrossRef](#)]
7. Réquillé, S.; Mazian, B.; Grégoire, M.; Musio, S.; Gautreau, M.; Nuez, L.; Day, A.; Thiébeau, P.; Philippe, F.; Chabbert, B.; et al. Exploring the dew retting feasibility of hemp in very contrasting European environments: Influence on the tensile mechanical properties of fibres and composites. *Ind. Crop. Prod.* **2021**, *164*, 113337. [[CrossRef](#)]
8. Corbin, A.-C.; Ferreira, M.; Labanieh, A.R.; Soulat, D. Natural fiber composite manufacture using wrapped hemp roving with PA12. *Mater. Today Proc.* **2020**, *31*, S329–S334. [[CrossRef](#)]
9. Corbin, A.-C.; Sala, B.; Soulat, D.; Ferreira, M.; Labanieh, A.-R.; Placet, V. Development of quasi-unidirectional fabrics with hemp fiber: A competitive reinforcement for composite materials. *J. Compos. Mater.* **2021**, *55*, 551–564. [[CrossRef](#)]
10. Placet, V.; François, C.; Day, A.; Beaugrand, J.; Ouagne, P. Industrial hemp transformation for composite applications: Influence of processing parameters on the fibre properties. In *Advances in Natural Fibre Composites: Raw Materials, Processing and Analysis*; Springer: Berlin/Heidelberg, Germany, 2018; pp. 13–25. ISBN 978-3-319-64640-4.
11. Shah, D.U.; Schubel, P.J.; Clifford, M.J. Modelling the effect of yarn twist on the tensile strength of unidirectional plant fibre yarn composites. *J. Compos. Mater.* **2013**, *47*, 425–436. [[CrossRef](#)]
12. Goutianos, S.; Peijs, T.; Nystrom, B.; Skrifvars, M. Development of flax fibre based textile reinforcements for composite applications. *Appl. Compos. Mater.* **2006**, *13*, 199–215. [[CrossRef](#)]
13. Omrani, F.; Wang, P.; Soulat, D.; Ferreira, M. Mechanical properties of flax-fibre-reinforced preforms and composites: Influence of the type of yarns on multi-scale characterisations. *Compos. Part A Appl. Sci. Manuf.* **2017**, *93*, 72–81. [[CrossRef](#)]
14. Russo, P.; Simeoli, G.; Vitiello, L.; Filippone, G. Bio-polyamide 11 hybrid composites reinforced with basalt/flax interwoven fibers: A tough green composite for semi-structural applications. *Fibers* **2019**, *7*, 41. [[CrossRef](#)]
15. Lebaupin, Y.; Chauvin, M.; Hoang, T.-Q.T.; Touchard, F.; Beigbeder, A. Influence of constituents and process parameters on mechanical properties of flax fibre-reinforced polyamide 11 composite. *J. Thermoplast. Compos. Mater.* **2017**, *30*, 1503–1521. [[CrossRef](#)]
16. Oliver-Ortega, H.; Méndez, J.A.; Reixach, R.; Espinach, F.X.; Ardanuy, M.; Mutjé, P. Towards more sustainable material formulations: A comparative assessment of PA11-SGW flexural performance versus oil-based composites. *Polymers* **2018**, *10*, 440. [[CrossRef](#)]

17. Haddou, G.; Dandurand, J.; Dantras, E.; Maiduc, H.; Thai, H.; Giang, N.V.; Trung, T.H.; Pontains, P.; Lacabanne, C. Mechanical properties of continuous bamboo fiber-reinforced biobased polyamide 11 composites. *J. Appl. Polym. Sci.* **2019**, *136*, 47623. [[CrossRef](#)]
18. Gourier, C.; Bourmaud, A.; Le Duigou, A.; Baley, C. Influence of PA11 and PP Thermoplastic polymers on recycling stability of unidirectional flax fibre reinforced biocomposites. *Polym. Degrad. Stab.* **2017**, *136*, 1–9. [[CrossRef](#)]
19. Awais, H.; Nawab, Y.; Amjad, A.; Anjang, A.; Md Akil, H.; Zainol Abidin, M.S. Effect of comingling techniques on mechanical properties of natural fibre reinforced cross-ply thermoplastic composites. *Compos. Part B Eng.* **2019**, *177*, 107279. [[CrossRef](#)]
20. Kobayashi, S.; Takada, K. Processing of unidirectional hemp fiber reinforced composites with micro-braiding technique. *Compos. Part A Appl. Sci. Manuf.* **2013**, *46*, 173–179. [[CrossRef](#)]
21. Zhai, W.; Wang, P.; Legrand, X.; Soulat, D.; Ferreira, M. Effects of micro-braiding and co-wrapping techniques on characteristics of flax/polypropylene-based hybrid yarn: A comparative study. *Polymers* **2020**, *12*, 2559. [[CrossRef](#)]
22. Alagirusamy, R.; Fangueiro, R.; Ogale, V.; Padaki, N. Hybrid yarns and textile preforming for thermoplastic composites. *Text. Prog.* **2006**, *38*, 1–71. [[CrossRef](#)]
23. Asghar, A.; Imad, A.; Nawab, Y.; Hussain, M.; Saouab, A. Effect of yarn singeing and comingling on the mechanical properties of jute/polypropylene composites. *Polym. Compos.* **2020**, *42*. [[CrossRef](#)]
24. Di Lorenzo, M.L.; Longo, A.; Androsch, R. Polyamide 11/poly(butylene succinate) bio-based polymer blends. *Materials* **2019**, *12*, 2833. [[CrossRef](#)] [[PubMed](#)]
25. Xiao, S.; Wang, P.; Soulat, D.; Gao, H. Thermo-mechanical characterisations of flax fibre and thermoplastic resin composites during manufacturing. *Polymers* **2018**, *10*, 1139. [[CrossRef](#)]
26. NF G07-316. *Textiles—Tests of Yarns—Determination of Linear Density*; AFNOR: La Plaine Saint-Denis, France, 1988.
27. NF G07-079. *Textiles—Testing Threads—Determining the Twisting of Threads by Untwisting/Retwisting with a Double Re-Test*; AFNOR: La Plaine Saint-Denis, France, 2011.
28. NF EN ISO 2062. *Textiles—Yarns from Packages—Determination of Single-End Breaking Force and Elongation at Break Using Constant Rate of Extension (CRE) Tester*; AFNOR: La Plaine Saint-Denis, France, 2010.
29. NF EN 12127. *Textiles—Fabrics—Determination of Mass per Unit Area Using Small Samples*; AFNOR: La Plaine Saint-Denis, France, 1998.
30. NF EN ISO 5084. *Textiles—Determination of Thickness of Textiles and Textile Products*; AFNOR: La Plaine Saint-Denis, France, 1996.
31. NF EN ISO 9237. *Textiles—Determination of Permeability of Fabrics to Air*; AFNOR: La Plaine Saint-Denis, France, 1995.
32. NF ISO 7211-3. *Textiles—Woven Fabrics—Construction—Methods of Analysis—Part 3: Determination of Crimp of Yarn in Fabric*; AFNOR: La Plaine Saint-Denis, France, 2017.
33. NF EN ISO 13934-1. *Tensile Properties of Fabrics—Part 1: Determination of Maximum Force and Elongation at Maximum Force Using the Strip Method*; AFNOR: La Plaine Saint-Denis, France, 2013.
34. ISO 4604. *Reinforcement Fabrics—Determination of Conventional Flexural Stiffness—Fixed-Angle Flexometer Method*; AFNOR: La Plaine Saint-Denis, France, 2011.
35. De Bilbao, E.; Soulat, D.; Hivet, G.; Gasser, A. Experimental study of bending behaviour of composite reinforcements. *Int. J. Mater. Form.* **2009**, *2*, 205–208. [[CrossRef](#)]
36. Bilisik, K. Bending behavior of multilayered and multidirectional stitched aramid woven fabric structures. *Text. Res. J.* **2011**, *81*, 1748–1761. [[CrossRef](#)]
37. ASTM D3039/D3039M-17. *Standard Test. Method for Tensile Properties of Polymer Matrix Composite Materials*; ASTM International: West Conshohocken, PA, USA, 2017.
38. NF EN ISO 14125. *Fibre-Reinforced Plastic Composites—Determination of Flexural Properties*; AFNOR: La Plaine Saint-Denis, France, 1998.
39. Tanasa, F.; Zănoagă, M.; Teacă, C.; Nechifor, M.; Shahzad, A. Modified hemp fibers intended for fiber-reinforced polymer composites used in structural applications—A review. I. Methods of modification. *Polym. Compos.* **2019**, *41*. [[CrossRef](#)]
40. Baley, C.; Gomina, M.; Breard, J.; Bourmaud, A.; Drapier, S.; Ferreira, M.; Le Duigou, A.; Liotier, P.J.; Ouagne, P.; Soulat, D.; et al. Specific features of flax fibres used to manufacture composite materials. *Int. J. Mater.* **2019**, *12*, 1023–1052. [[CrossRef](#)]
41. Corbin, A.-C.; Soulat, D.; Ferreira, M.; Labanieh, A.; Gabrion, X.; Placet, V. Improvement of the weavability of natural-fiber reinforcement for composite materials manufacture. *RCMA* **2019**, *29*, 201–208. [[CrossRef](#)]
42. Corbin, A.-C.; Soulat, D.; Ferreira, M.; Labanieh, A.-R. Influence of process parameters on properties of hemp woven reinforcements for composite applications: Mechanical properties, bias-extension tests and fabric forming. *J. Nat. Fibers* **2020**, 1–13. [[CrossRef](#)]
43. Bismarck, A.; Aranberri, I.; Springer, J.; Lampke, T.; Wielage, B.; Stamboulis, A.; Shenderovich, I.; Limbach, H.-H. Surface characterization of flax, hemp and cellulose fibers; Surface properties and the water uptake behavior. *Polym. Compos.* **2002**, *23*, 872–894. [[CrossRef](#)]