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Mechanical Properties of a PLA/Nettle Agro-Composite with 10% Oriented Fibers

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Featured Application: The possible applications of this eco-friendly material seem to be very wide, as long as they do not have to undergo too important structural loads, as for example the sector of interior furniture, packaging, or even interior elements in the automotive sector.

Abstract: Within the framework of the environmental policies which tend toward new ecological materials, without the use of petroleum-based materials, the objective of this work is to develop a composite 100% natural associating fibers of European nettles (*Urtica dioica*) and PLA (Poly-Lactic Acid). After having determined the properties of both components of the new composite, a methodology was implemented to maximize the PLA/nettle association. Then, tensile tests with standardized specimens were carried out to assess the mechanical characteristics of this new composite, which are ultimately very dependent on its manufacturing process. Experimental results demonstrated that the manufacturing process results in a material with enhanced overall mechanical properties, compared to those found in the literature (Young's modulus: 5.41 ± 0.23GPa vs. 1.2 to 4.37 ± 0.14 GPa). Our findings suggest widespread use of this PLA/nettle composite, although its properties clearly stem from the quality of the nettle fiber, which is itself highly dependent on variable parameters linked to the growing conditions of the plant. The possible applications of this eco-friendly material seem to be very wide, as long as they do not have to undergo too important structural loads, as for example the sector of interior furniture, packaging, or even interior elements in the automotive sector.

Keywords: fiber nettle; PLA; process; mechanical properties

1. Introduction

The use of composite materials is becoming more and more widespread each year in various sectors of the industry [1], due to the development of technical applications with ever-increasing performance requirements and ever-stronger constraints on raw material consumption. Constraints to which composite materials are particularly well adapted, given their combination of reinforcements of different natures and a matrix allowing fine-tuning for each specific application. In the frame of agro-composites, many types of fibers can be considered usable [1–5], but the present study will focus on the nettle fibers only [6] (Figure 1).

Nettle is part of the *Urticaceae* family and belongs to the genus *Urtica* [7], which comprises more than 70 species. In Europe, eleven species of nettles are listed, five of which are common in France: it is, therefore, a locally available resource, although little exploited at present. Long used for its medicinal properties [8,9], it was gradually forgotten during the 19th century before a revival of interest during the First World War in the textile industry for the manufacturing of tents, bags, and clothing due to supply shortages of conventional fibers such as cotton, hemp, and linen [10,11]. Nevertheless, the increasing industrialization of agricultural processes that followed the war did not apply to nettle, as labor costs increased sharply. Consequently, the plant fell into oblivion and only recently came back into the limelight due to current environmental and societal concerns. The quality of natural nettle fibers depends on the growing conditions, extraction, treatment, and processing, with high variability in mechanical properties due to the chemical composition of the fibers [12–14] and the difficulty in obtaining an accurate estimation of the cross-section of these fibers [15].

It varies from one species to another and even often from one plant to another within the same species and the same crop [10,16,17]. The main constituents of lignocellulosic fibers are cellulose, hemicellulose, pectin, and lignin [18]. Cellulose is a natural polymer consisting of long glucose chains in crystalline (80%) or amorphous (20%) form [19]. Hemicelluloses and pectins are the second major polysaccharidic constituents of natural fibers and constitute the interface between cellulose fibers. Lignin is a phenyl propane-based polymer combined with polysaccharides. More resilient, it is one of the most abundant biopolymer constituents on Earth after polysaccharides; it provides structural rigidity to the cell walls and limits microbiological attacks [20].

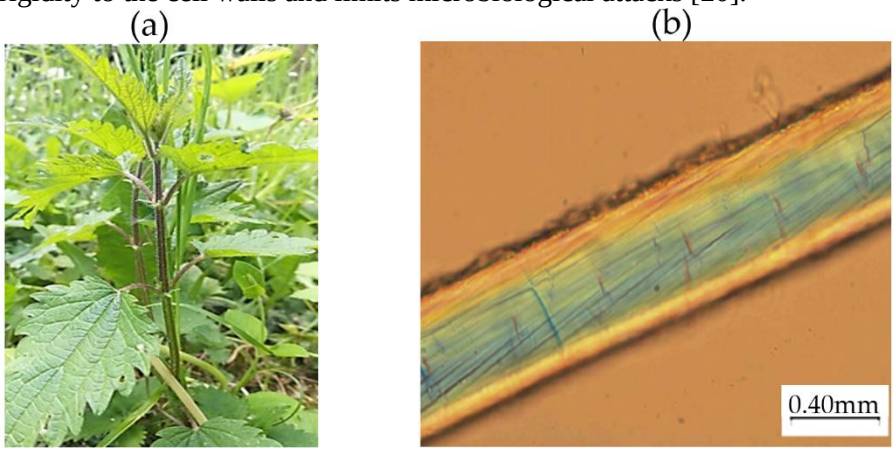

Figure 1. (**a**) Nettle stem (*Urtica dioica*) [6], (**b**) fibrillar structure of a nettle fiber cell in polarized light, the bundles of microfibrils can be seen as colored strings bordered by darker outlines [21].

Although these different chemical elements are the base components of natural fiber, the overall organization of the fiber also plays a major role in its macroscopic properties [10,21–23] and explains the craze for nettle fiber.

Our choice to use PLA (poly-lactic acid) as the matrix for this agro-composite is explained by the desire to achieve an entirely natural and therefore completely biodegradable material. Among all thermoplastics, PLA has undoubtedly experienced the fastest growth in its use in recent years, not to

mention that it is also one of the latest to be widely commercially available, in 2002 [24]. Its main advantage is that it is completely biobased and biodegradable (or at least compostable under industrial conditions). This category only comprises a few other materials, such as starch, PHA (poly-hydroxy-alkanoates), cellulose acetate, and regenerated cellulose [25]. PLA is obtained from the extraction of starch from potatoes, corn, or sugar from beet or cane. After a fermentation step, lactic acid is obtained to form lactides, which can then be distilled and polymerized to form PLA. One of the major properties of PLA is its molecular weight, the higher the molecular weight, the longer the polymer chains and the better its temperature and mechanical resistance. However, the temperature remains a parameter to be monitored when using PLA. Polylactic acid is also sensitive to humidity, which acts on it as a plasticizer by breaking the polymer chains through hydrolysis reactions [26]. This affection for moisture reflects the strong presence of the hydroxyl group, which makes this thermoplastic a polar material, just like ligno-cellulosic fibers. As a result, a good cohesion with fibers is achieved and explains the common association between PLA and natural fibers without the need for any chemical treatments, contrary to other thermoplastics [27,28]. Historically, flax and hemp have been among the most widely used fibers because of their ease of cultivation and the high demand from the textile industry. However, few studies focusing on these fibers relate lower mechanical properties compared to nettle fibers. The local production also reinforces the interest in nettle since its exploitation is likely to boost the economy of this sector. Similarly, the use of a recyclable, biodegradable polymer with promising mechanical properties compared to petro-sourced polymers is an undeniable asset. In view of these multiple benefits, the nettle fiber, in association with a PLA matrix, seems to be able to become a major player in the bio-composites sector.

After having given the mechanical characteristics of the components (obtained with tensile tests); nettle and PLA, a methodology for the shaping of the PLA/nettle composite is presented. It combines the technique of film staking and compression molding to obtain composite plates. They are then analyzed to validate the cohesion of PLA-nettle and then tested mechanically by the means of standardized specimens.

2. Materials and Methods

2.1. Nettle Fibers

The nettle fibers are supplied by Fibres Recherche Développement (FRD, Troyes, France), a local supplier. In the literature, the natural fibers used are usually very clean and free of any bark or plant residue. The fiber stock was received as a genuine bulk product in the form of a disordered straw. The fibers were manually sorted in three batches as shown in Figure 2. In the following, we will refer to sorted fibers (SF) (i.e., long fibers between 10 and 70 mm, oriented in the same direction), raw fibers (RF) (i.e., a mixture of fibers of different lengths (between 5 and 50 mm) that can be oriented if necessary), and chopped fibers (CF) (mixture of short and medium fibers).

Figure 2. Sorted fiber (SF), raw fiber (RF), chopped fiber (CF).

Long fibers are normally preferred, but they also make the composite orthotropic, which may be problematic depending on the targeted application. Short fiber composites, while less able to withstand high stresses, result in a more isotropic material due to the random distribution of their orientations; hence the idea of testing all three fiber configurations above. Moreover, chemical analysis was performed by the Van Soest method (NF EN ISO 13906 and NF V18-122) to detail the composition of the fiber; it is composed of 48.0% of cellulose, 5.2% of hemicellulose, 2.8% of lignin, 43.6% of soluble element and 0.3% of inorganic material.

2.1.1. Mechanical Properties of the Nettle Fibers

The values of the Young's modulus and tensile strength mechanical properties of our fibers (Table 1) were found to match the lower range of those usually given in the literature, while elongation at break and density are consistent [29–32]. One possible cause of these discrepancies is the drought that occurred early in the year before harvest, which severely restricted crop development and may also partially explain the high presence of plant growth defects.

Table 1. Mechanical properties of the nettle fiber.

Supplier Data	**Young's Modulus (GPa)**	**Tensile Strength (MPa)**	**Elongation at Break (%)**	**SF Density (g/cm^3)**
	13.93 ± 6.83	299.38 ± 182.08	2.33 ± 0.62	1.18 ± 0.12

2.1.2. Optical Analysis of the Fibers

Optical microscopy (performed with an OPTIKA B-150 optical microscope with magnifications of 4, 10, 40, and 100 times coupled to the digital acquisition software LITEVIEW) allows the analysis of the anatomy of nettle fibers. It highlights the growth defects of the plant also called "kink-band" which can induce a local fragility (Figure 3). These defects exist naturally on all types of lignocellulosic fibers [21]. We can also see from these figures that the number of dislocations is quite large compared to the number of lighter bands that stripe the fibers.

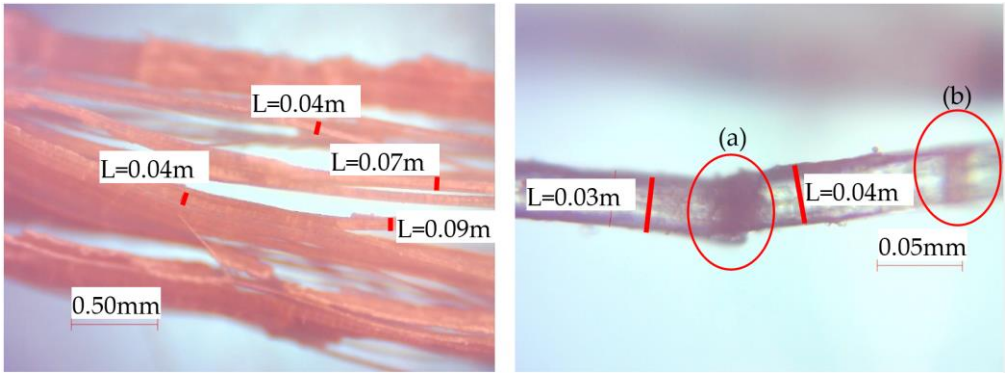

Figure 3. Bundle of nettle fibers under the light microscope. Detail of a growth defect (a) and a dislocation (b).

Figure 3 allows us to visualize diameter measurements; it fluctuates between 30 and 90 µm for our sample in agreement with the measurement carried out by the supplier FRD (distribution of the diameters in a sample of about thirty fibers included between 40 and 100 µm).

The use of scanning electron microscopy (performed with the JEOL JSM 6460LA and samples coated with a nanometric deposit of carbon black or gold/palladium) will allow us to see that the fibers are generally grouped in bundles of about 100 µm wide (Figure 4).

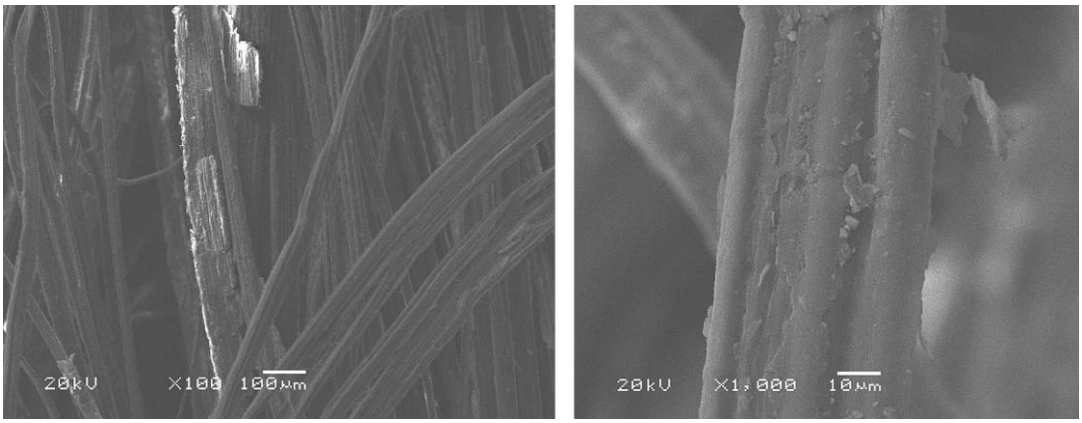

Figure 4. Bundle of nettle fibers under the electron microscope.

Observations made are classic for non-chemically treated lignocellulosic fibers with an overall smooth surface appearance (similar images for flax [33], ramie (Asian nettle) [28,34,35] and nettle [21,29,30]). Many wax or sap residues are visible on the surface. These residues resisted the various retting and mechanical sorting steps

2.2. Polylactic Acid

There are many references to PLA; the one used here is the 4043D available at NatureWorks in pellets form.

2.2.1. Mechanical Properties of the PLA

Table 2 lists the mechanical properties given by the manufacturer for this 4043D PLA.

Table 2. Mechanical properties of PLA (manufacturer data).

Supplier Data	Young Modulus (GPa)	Tensile Strength (MPa)	Elongation at Break (MPa)	Density (g/cm^3)
	3.6	60	6	1.24

2.2.2. Differential Scanning Calorimetry (DSC) of the PLA

A DSC analysis was carried out on a DSC Q100 to characterize the PLA granules (samples placed 48 h at 50° in a BINDER KBF115 oven, analysis carried out under a nitrogen environment of 50 mL/min). The protocol used consists of a first heating step at a rate of 10 °C/min up to 210 °C, keeping the final temperature for 5 min before cooling at 10 °C/min to the ambient temperature. After 5 min, a second run was carried out at 10 °C/min to characterize the material.

The first heating aims at resetting the history of the material, including possible ageing, whereas the second is the actual DSC measurement of the material (Figure 5).

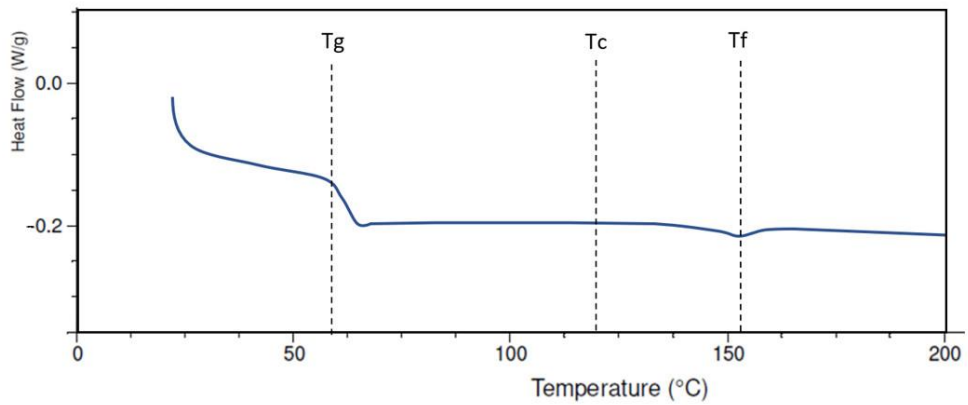

Figure 5. DSC analysis of the PLA.

The PLA used has a glass transition temperature of 61 °C. Melting occurs around 153 °C, with a very tenuous crystallization peak. The crystallinity rate is evaluated at 4.5%.

2.3. Constitution of PLA/Nettle Plates; Operating Mode

The optimal procedure for shaping the composite plates required a certain amount of skill and many steps. A manual FONTIJNE heating press was used, featuring two 32x32 cm platens able to reach a temperature of 300 °C. The lower plate of the press is mobile and compresses the mold of the part to be produced, with a maximum applicable force of 300 kN.

2.3.1. Agro-Composite Plate Manufacturing

The selected manufacturing technique consists in making elementary plates, which are then piled up and pressed to form the final plate. To this end, a 200 × 200 mm mold was cut in a 0.8 mm thick aluminum sheet. It will was used for both the PLA granule plates and PLA + fiber plates (called intermediate plates). Stacking and pressing the resulting elementary plates will form 4 mm thick plates in which the standardized specimens for the tensile test were cut (Figure 6).

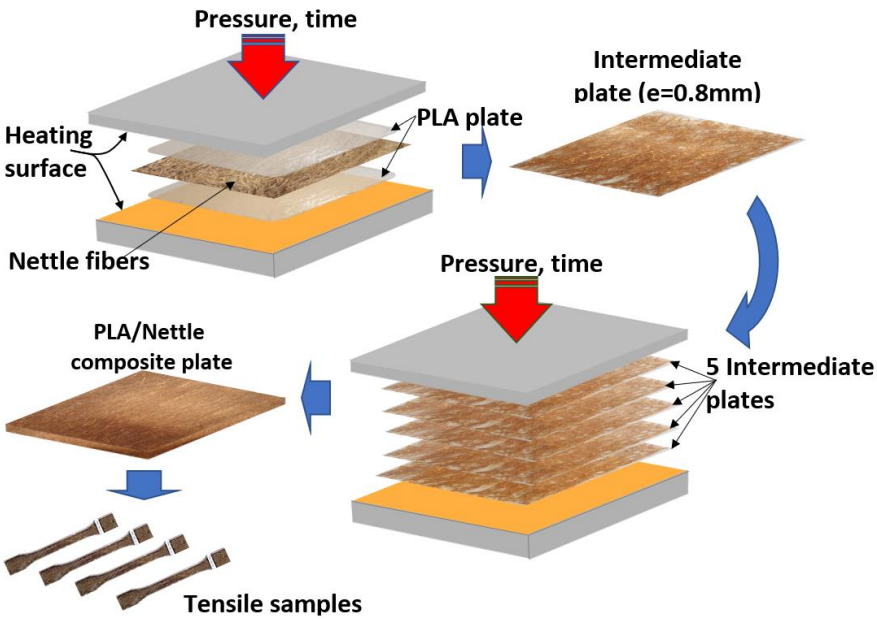

Figure 6. Manufacturing techniques used for agro-composite plates. PLA alone, intermediate plates, and final assembly of intermediate plates.

The fraction of fibers per elementary film was checked out to ensure a 10% mass ratio in the final plate.

2.3.2. Fibers' Orientation in the Plate

Different SF fiber positioning tests were performed (10 < L < 70 mm), RF (a mix of long and short fibers between 5 and 50 mm), and short fibers only (between 5 and 10 mm) (Figure 7).

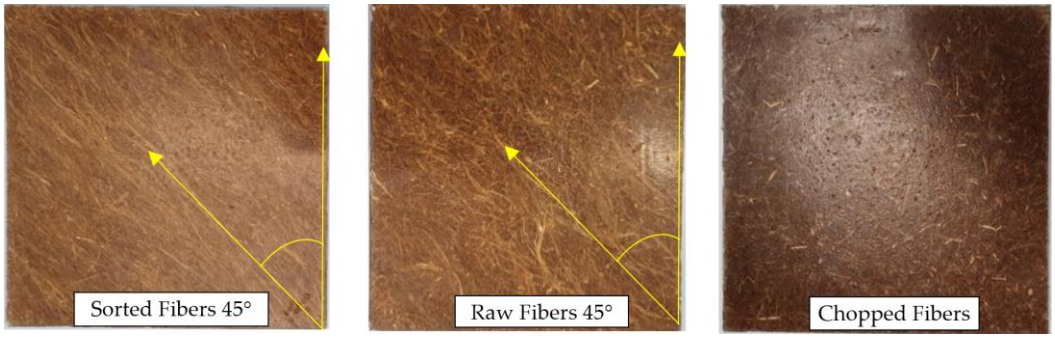

Figure 7. Examples of plates obtained with SF (oriented at 45°), RF (oriented at 45°), and CF (short ones).

It should be noted that the coloration of the plate containing CF is darker than the other two although they were made under the same conditions (temperature, pressure, and holding time); either the fibers may have undergone degradation, or the CF fraction might contain some remaining leaf or sap-colored components that could have leached into the PLA matrix. This plate presents orientation variations that will potentially prevent any interpretation of the results of future mechanical tests. Therefore, this plate was discarded, and tensile tests were performed only on RF and SF with orientations at 0°, 45°, and 90°.

2.3.3. Physico-Chemical Analysis of a PLA/Nettle Plate, Choice of Fiber Arrangement

Performing DSC analysis is key to monitoring the nucleation effect of the lignocellulosic fibers on the PLA; several PLA/nettle plate samples were taken from the center of a plate to perform this DSC analysis, the same protocol as for the study of PLA alone was used. The numerous microfibrils that separate from the main beam should facilitate this nucleation [24,36] resulting in a larger crystallization on the composites compared to PLA alone (Figure 8). The fiber nucleation effect results in a shift of the cold crystallization peak to lower temperatures due to greater ease of crystallization at low temperatures; these results were observed on a PLA/ramie composite [37]. Regarding our composite, a crystallinity of 14.1% is found for the 0° SF versus 9% for the RF. For the 45° oriented sorted fibers, the crystallinity reaches 26.1% versus 19.6% for RF. It thus seems that the SF participates much more significantly in the nucleation of PLA. This strong increase in the crystallinity is moreover directly related to the manufacturing process that we used: increased fiber distribution within the matrix leads to a much more significant nucleation effect.

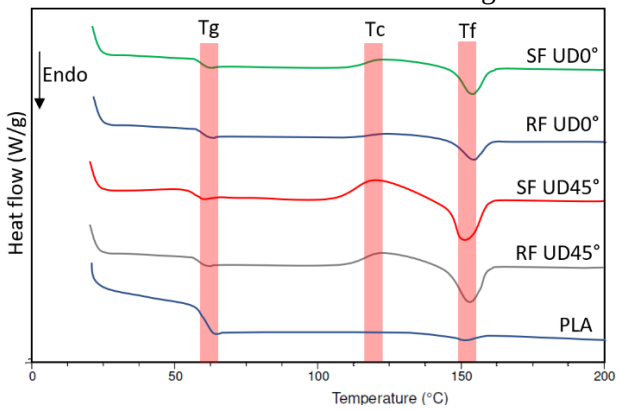

Figure 8. DSC analysis on the different fiber organizations in the composite (Tg: glass transition temperature, Tc: crystallinity temperature, Tf: fusion temperature).

These results (for glass transition temperature Tg, crystallinity temperature Tc, fusion temperature Tf and crystallinity rate R) allow us to validate the chosen manufacturing concept based on the use of oriented fibers rather than non-oriented short fibers (Figure 7) and enable the realization of the tensile specimens used in the next step for mechanical test (Table 3).

Table 3. Condensed results of DSC tests on the different composite configurations investigated (SF: sorted fibers, RF: raw fibers).

	SF UD0°	**RF UD0°**	**SF UD45°**	**RF UD45°**	**PLA**
Tg (°C)	59.06	59.38	57.16	59.07	61.16
Tc (°C)	126.44	129.13	121.48	124.99	126.52
Tf (°C)	153.23	152.92	150.74	152.23	153.35
R (%)	14.1	9	26.1	19.6	0.7

2.3.4. SEM Analysis of the Plate

The SEM analysis provides actual visualization of the PLA–fiber association in this shaping (samples were obtained by cryofracturing, i.e., broken after immersion in liquid nitrogen).

Very satisfying impregnation of the fibers in the PLA is observed (Figure 9 on the right) compared to other manufacturing tests performed (Figure 9 on the left) where the PLA does not cover the entire fiber.

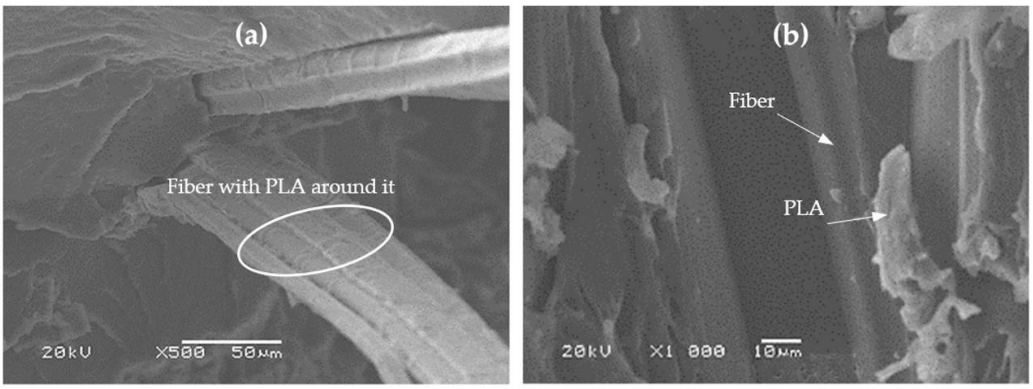

Figure 9. Nettle fiber imbedded in PLA (**a**) compared to a smoother fiber, where PLA did not cling well (**b**).

3. Sample Preparation and Mechanical Tests

PLA has a low glass transition temperature and tends to heat up and melt quickly with conventional machining processes. In accordance with the ISO 527 standard and the dimensions of the specimens, the cutting was carried out on a MAXIEM 1515 numerical control machine using the water jet cutting process enriched with abrasive sand. This allowed for a clean cut with precise dimensions. To avoid excess water effects, the samples were placed in a controlled atmosphere just after the cutting, before the mechanical tests were carried out. The nettle fibers and PLA were prepared at 60 °C and 2%

humidity for 48 h before manufacturing and equilibrated under 50% humidity for 48 h at 25 °C before the mechanical tests. The measurements were carried out on five samples for each plate (Figure 10).

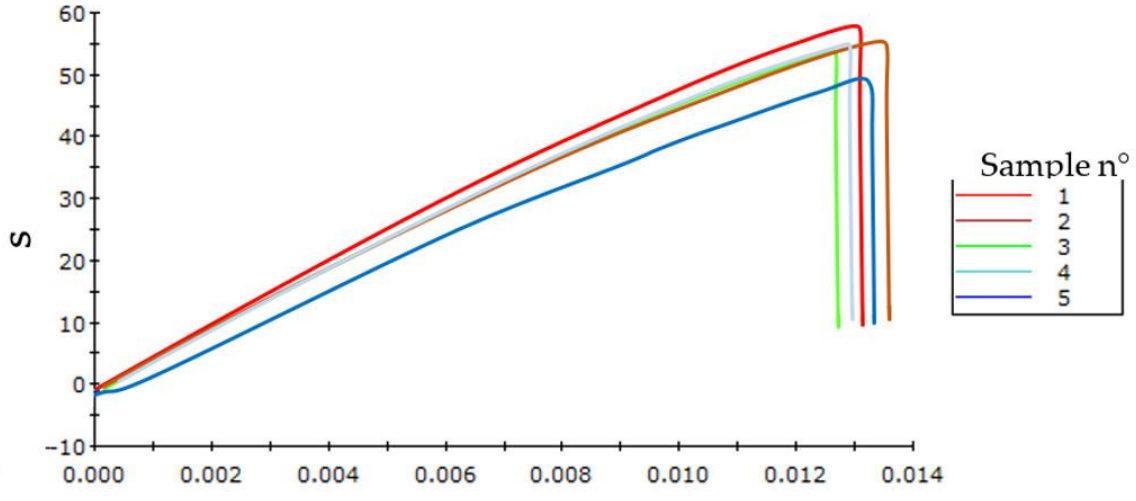

Figure 10. Tensile curves obtained for five sorted fibers specimens with a fiber orientation of 0° and for a test performed at a speed of 1 mm/min.

Table 4 presents the mean values of Young's modulus, tensile strength, and elongation at break obtained for a sample of five specimens.

Table 4. Summary of tensile tests (1 mm/min) on unidirectional SF and RF.

Samples from Plate (with Sorted Fibers and Raw Fibers)	**Young's Modulus (GPa)**		**Tensile Strength (MPa)**		**Elongation at Break (%)**	
	Average	**SD**	**Average**	**SD**	**Average**	**SD**
SF UD0°	5.41	±0.23	53.98	±3.11	1.24	±0.05
RF UD0°	4.51	±0.19	44.70	±3.36	1.21	±0.07
SF UD45°	3.85	±0.08	35.89	±2.58	1.19	±0.10
RF UD45°	3.99	±0.07	38.21	±3.64	1.24	±0.12

The sorting of fibers appears to be beneficial to the strength of the specimen, it increases from 4.51 for RF to 5.41 GPa (20% gain) for SF. Similar observations can be made regarding Young's modulus, with a value increasing from 44.07 for RF to 53.99 MPa on average (22.5% gain) for SF.

Figure 11 shows that the specimen broke cleanly at both 0 and 45° (no delamination), which validates our intermediate plate technique used for specimen plate production. Similarly, no warping deformation (curvature of the plate after demolding) that may affect the outcome of the tensile test was noted.

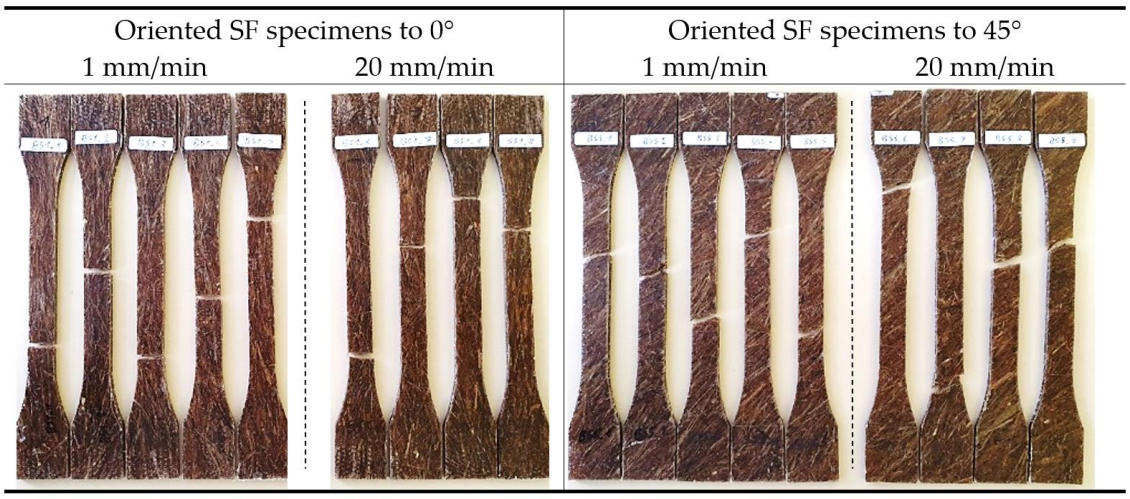

Figure 11. Tensile test on sorted fibers oriented 0° (**right**) and 45° (**left**); for a traverse speed of 1 mm/min and 20 mm/min.

As seen in Figure 11, the breakage occurred at different locations on each of the specimens, which indicates that the fibers are distributed sufficiently homogeneously so as not to create defects at the scale of the plate.

4. Results and Discussion

The addition of fibers to the PLA matrix leads to an increase in longitudinal elastic modulus, accompanied by a slight decrease in tensile strength and elongation at break (Table 4). The fracture occurs perpendicular to the fibers so that the fracture profiles in the specimens with 45° fibers are also inclined at 45°. In addition, a two-step fracture can be shown, firstly with the matrix and then on the fibers.

Moreover, it has also been shown that fiber alignment plays a minor role in the mechanical performance of the specimens, with the 45° oriented sorted fibers participating less in the strengthening than the 45° oriented raw fibers. Although the standard deviations do not really allow for differentiation of the two plate types, it seems that the use of raw fibers at 45° brings a greater resistance compared to the use of sorted fibers. This result could be due to the fact that the greater disorganization creates reinforcements in a wider angular range, which takes part in the load transport. If we add to this the fact that a large part of the mechanical strength comes from the matrix for a 45° UD composite, it is normal to see better results for the raw fibers. We can already assume that the same will be true for a 90° fiber orientation. Since the tensile modulus of the fibers oriented at 45° can also be seen as the shear modulus of the composite, we can retain a value of 3.99 GPa for this shear modulus.

Furthermore, failure occurs around a 1.24% strain at best (Table 4), whereas the elongation at break of both PLA and fibers lie above 2%. This loss of strain of the composite compared to the two components alone may be caused by the heating process that weakened the structure of the fibers or caused by poor point adhesion between the fibers and the matrix creating local de-cohesions with excessive load transfer to the PLA matrix. As a matter of fact, we noted (Figure 4) the presence of wax or sap residues visible on the surface of some fibers used. These residues can affect the proper transmission of forces between the fibers and the matrix because the mechanical fiber/matrix anchoring depends on the roughness, the wettability of the fibers [22] as well as the chemical affinity (polarity) with the matrix. To improve this bonding, chemical treatments (NaOH, etc.), have been used [14,34,35,38] to clean the fiber and can, in some cases, significantly improve the performance. The

choice to use chemical treatment can also, in some cases weaken the fiber. However, above all, the biocomposite thus treated loses its 100% bio-sourced aspect, which we do not want here.

The fiber ratio used in this work was 10%. It constitutes a preliminary approach to visualizing the properties of the PLA/nettle composite. Tests at 20, 30, and 40% (by mass) are in progress. The presence of a higher fibers ratio does not necessarily seem to be advantageous for the composite mechanical properties because the fibers' embedding in PLA is not optimal; the fibers tend to agglomerate together in bundles. This creates porosities that lead to a non-linear evolution of the mechanical properties and progressive embrittlement of the composite [39,40,41].

This porosity phenomenon is also the reason for our change in the manufacturing process of the plates. Indeed, prior to the procedure depicted in Figure 6, a selected mass ratio of fibers and PLA pellets were placed on different layers (sandwich method), directly in a mold to produce the plates for the tensile specimens. Subsequent tests showed an average Young's modulus of 4.48 ± 0.13 MPa compared to 5.41 ± 0.23 MPa obtained following manufacturing with the intermediate plates. Similarly, the tensile strength rose from 43.41 to 53.86 MPa. This improvement in the manufacturing process is also visible in the rupture mode of the specimen, which switches from delamination in the fibers' layer for the sandwich method to clean rupture for the stratified method (Figure 12). It should also be noted that no bulging deformation was observed during the manufacture with the intermediate plates, in contrast to the sandwich method, thanks to a better distribution of the fibers in the thickness of the plate.

Figure 12. Visualization of fracture defects (delamination) on sandwich tensile specimens.

Figure 13 below shows the composition of the plates in their thickness by each of the two assembly methods. We can easily discern strata in the sandwich composite, whereas with the intermediate plate method, the composite is visually homogeneous with fibers well distributed across the width.

Mechanical performance tests confirm that this method provides better resistance. However, it is also more time-consuming in terms of preparation since it requires about thrice the time required for the sandwich method.

Figure 13. Comparison of specimens (side view) obtained by the sandwich method (**a**) and intermediate plate method (**b**) after testing.

Table 5 compares the main mechanical characteristics of the PLA/nettle composite obtained by the intermediate plate method to results from the literature.

Table 5. Comparison of the mechanical properties of the obtained PLA/nettle composite with literature results.

Composite	Obtention Process	Fiber Length (mm)	Treatment	Fiber (Mass Ratio)	Young's Modulus (GPa)	Tensile strength (MPa)	Elongation at Break (%)
Present studyPLA/nettle	Intermediate plate	10 < L < 70	-	10	5.41 ± 0.23	53.98 ± 3.11	1.24 ± 0.05
nettle/PLA [39]	Nettle and PLA fibers carded and molded	55 ± 1	NaOH	10	2.37 ± 0.05	15.54 ± 0.45	1.42 ± 0.06
				25	3.11 ± 0.05	36.70 ± 1.30	1.80 ± 0.08
				50	3.34 ± 0.04	50.82 ± 0.82	2.45 ± 0.10
nettle/PLA [42]	Nettle and PLA fibers carded and molded	Woven fabric	-	20	4.8/3.5	45/52	1.2/1.8
				30	5.6/3.5	59/52	1.5/1.8
				40	4.8/3.5	40/52	1.3/1.8
nettle/PS [43]	Hand layup technique	fabric	-	5	-	~19.8 ± 0.67	-
				10		~23.22 ± 0.85	
				15		~30.69 ± 0.57	
				20		~29.08 ± 0.81	
ramie/PLA [36]	Ramie fabric and PLA molded	fabric	-	24	1.64	66.33	4
				38	1.2	29.37	3
ramie/PLA [38]	two-roll mill heated	10	NaOH-silane	30	2/4	52/66.8	3.2/4.8
nettle/PLA [44]	PLA fiberwebs molded	fabric	-	20	3.35 ± 0.12	35.56 ± 1.16	1.59 ± 0.12
				35	3.97 ± 0.08	38.35 ± 0.30	1.76 ± 0.05
				50	4.37 ± 0.14	39.87 ± 0.60	2.18 ± 0.07
nettle/PP [32]	Film stacking	Woven mat	-	-	-	32.56	-

We notice that our results with 10% mass ratio fibers are better than those mentioned in the literature for identical compositions. The results are even better than those with higher fiber content. The chemical treatment seems to be beneficial for the elongation properties of tensile strength, and fiber content on the improvement of mechanical properties up to different percentages according to the authors. The obtention process, for these authors, also has a great influence. Moreover, the structural form of reinforcement, for example, fabric, yarn, fiberweb, etc. played a decisive role in determining the mechanical properties of biocomposite. We believe that our results are mainly due to the manufacturing technique used for our specimens. The technique of combining film stacking and hot pressing, but with thicker plates than the films and by refining the pressure, the temperature, and the holding time, has resulted in not only good mechanical properties but also low waste, reduced part cost, uniform pressure distribution on the composite and good surface finish [45].

Additional tests were also performed at a velocity of 20 mm/min (Figure 11) to evaluate the viscoplastic behavior of the composite. For each plate with fibers oriented in the tensile direction (0°), left part of Figure 11, it appears that increasing the test velocity improves the elongation at break as

well as the tensile strength of the composites. In contrast, the effects on the modulus are negligible due to the strong scattering of the values obtained. It can be stated that the composite exhibits viscoelastic behavior, of elastic type for high strain rates and viscous type for low strain rates. Similar behaviors have already been observed in polypropylene/glass fiber composites [46].

5. Conclusions

This study aimed to characterize a 100% natural composite material combining European nettle fibers (*Urtica dioica L.*) and PLA (poly-lactic acid). The first step of this work was to determine the properties of the two components of the new composite and to implement a methodology to maximize the PLA/nettle association. Next, tensile tests were performed on standardized specimens to evaluate the mechanical characteristics of this new composite, which are ultimately very dependent on its manufacturing process. The results of our measurements revealed improved mechanical properties (Young's modulus, tensile strength) compared to those reported in the literature for equivalent composites. These positive results were obtained thanks to the use of an intermediate plate, i.e., a combination of film stacking and compression molding techniques, which proved to be largely beneficial. The other interesting and innovative feature of this agro composite is that it is manufactured without any additional chemical treatment or additives, our goal is to create a 100% biodegradable composite. We are currently testing higher percentages of fibers while optimizing the manufacturing technique with the objective of achieving a PLA/nettle agro composite for industrial use. Ultimately, this environmentally friendly material could find applications in various sectors of activity such as interior furniture, packaging, or the automotive sector. The only limit to its industrial use could be its resistance to structural load.

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References

1. JEC Observer. Overview of the Global Composites Market. 2021-2026. Available online: https://www.jeccomposites.com/press/jec-observerbroverview-of-the-globalbrcomposites-market-2021-2026-eco/ (accessed on 03/03/2022).
2. Manu, T.; Reza Nazmi, A.; Shahri, B.; Emerson, N.; Huber, T. Biocomposites: A review of materials and perception, *Mater. Today Commun.* **2022**, *31*, 103308. https://doi.org/10.1016/j.mtcomm.2022.103308.
3. Chang, B.P.; Rodriguez-Uribe, A.; Mohanty, A.K.; Misra, M. A comprehensive review of renewable and sustainable biosourced carbon through pyrolysis in biocomposites uses: Current development and future opportunity. *Renew. Sustain. Energy Rev.* **2021**, *152*, 111666. https://doi.org/10.1016/j.rser.2021.111666.
4. Andrew, J.J.; Dhakal, H.N. Sustainable biobased composites for advanced applications: Recent trends and future opportunities—A critical review. *Compos. Part C Open Access* **2022**, *7*,100220. https://doi.org/10.1016/j.jcomc.2021.100220.
5. Mahmud, S.; Hasan, K.M.F.; Jahid, M.A.; Mohiuddin, K.; Zhang, R.; Zhu, J. Comprehensive review on plant fiber-reinforced polymeric biocomposites. *J. Mater. Sci.* **2021**, *56*, 7231–7264. https://doi.org/10.1007/s10853-021-05774-9.
6. Bogard, F.; Bach, T.; Abbes, B.; Bliard, C.; Maalouf, C.; Bogard, V.; Beaumont, F.; Polidori, G. A comparative review of Nettle and Ramie fiber and their use in biocomposites, particularly with a PLA matrix. *J. Nat. Fibers* **2021.** https://doi.org/10.1080/15440478.2021.1961341.
7. Hassler, M. World Plants: Synonymic Checklists of the Vascular Plants of the World. *Cat. Life* **2020**. https://doi.org/10.15468/7ppkhn.
8. Allais, D. L'ortie dioïque. *L'Imag. Méd.* **2009**, *48*, 53–55.
9. Draghy, F. L'ortie dioïque (Urtica dioica): Étude bibliographique. *Sci. Pharm.* **2005**.
10. Bodros, E.; Baley, C. Study of the tensile properties of stinging Nettle fibres (Urtica dioica). *Mater. Lett.* **2008**, *62*, 2143–2145. https://doi.org/10.1016/j.matlet.2007.11.034.
11. Koronis, G.; Silva, A.; Fontul, M. Green Composites: A review of adequate materials for automotive applications. *Compos. Part B* **2013**, *44*, 120–127. https://doi.org/10.1016/j.compositesb.2012.07.004.
12. Torres, J.P.; Vandi, L.J.; Veidt, M.; Heitzmann, M.T. The mechanical properties of natural fibre composite laminates: A statistical study. *Compos. Part A Appl. Sci. Manuf.* **2017**, *98*, 99–104. https://doi.org/10.1016/j.compositesa.2017.03.010.
13. Summerscales, J.D.; Dissanayake, N.P.J.; Virk, A.S.; Hall, W. A review of bast fibres and their composites. Part 1—Fibres as reinforcements. *Compos. Part A* **2010**, *41*, 1329–1335. https://doi.org/10.1016/j.compositesa.2010.06.001.
14. Shahzad, A. Mechanical properties of lignocellulosic fiber composites. In *Lignocellulosic Fibre and Biomass-Based Composite Materials*; Jawaid, M., Tahir, P., Saba, N., Eds.; Woodhead Publishing Series in Composites Science and Engineering: Sawston, UK, 2017; pp. 193–223. https://doi.org/10.1016/B978-0-08-100959-8.00011-1.
15. Scida, D.B.; Bourmaud, A.; Baley, C. Influence of the scattering of flax fibres properties on flax-epoxy woven ply stiffness. *Mater. Des.* **2017**, *122*, 136–145. https://doi.org/10.1016/j.matdes.2017.02.094.
16. Bledzki, A.K.; Gassan, L. Composites reinforced with cellulose based fibres. *Prog. Polym. Sci.* **1999**, *24*, 221–274. https://doi.org/10.1016/S0079-6700(98)00018-5.
17. Rowell, R.M.; Han, J.; Rowell, J. Characterization and factors effecting fiber propertie. *Nat. Polym. Agrofibers Based Compos.* **2000**, 115–134. Available online: https://www.researchgate.net/publication/237255433_Characterization_and_Factors_Effecting_Fiber_Properties (accessed on 27 September 2022).
18. Eichhorn, S.J.; Baillie, C.; Zafeiropoulos, N.; Mwaikambo, L.Y.; Ansell, M.P.; Dufresne, A.; Entswistle, K.M.; Herrera-Franco, P.; Canché-Escamilla, G.; Groom, H.; et al. Current international research into cellulosic fibres and composites. *J. Mater. Sci.* **2001**, *36*, 2107–2131. https://doi.org/10.1023/A:1017512029696.
19. Frollini E., Leao A., Mattoso L.H.C., Rowell R., Han J., Rowell J. Cellulose nanocrystals: Pretreatments, preparation strategies, and surface functionalization *Int. J. Bio. Macromol.* **2021**, 182, 1554-1581.
20. Do Thi, V. Matériaux Composites à Fibres Naturelles/Polymère Biodégradables ou Non. Ph.D. Thesis, Université de Grenoble, Grenoble, France, 2011.
21. Bergfjord, C.; Holst, B. A procedure for identifying textile bast fibres using microscopy: Flax, Nettle/Ramie, hemp and jute. *Ultramicroscopy* **2010**, *110*, 1192–1197. https://doi.org/10.1016/j.ultramic.2010.04.014.
22. Gourier, C. Contribution à L'étude de Matériaux Biocomposites à Matrice Thermoplastique Polyamide-11 et Renforcés Par des Fibres de Lin. Ph.D. Thesis, Université de Bretagne Sud, Lorient, France, 2016.
23. Baley, C. Analysis of the flax fibres tensile behaviour and analysis of the tensile stiffness increase. *Comp. Part A Appl. Sci. Manuf.* **2002**, *33*, 939–948. https://doi.org/10.1016/S1359-835X(02)00040-4.
24. Murariu, M.; Dubois, P. PLA composites: From production to properties. *Adv. Drug Deliv. Rev.* **2016**, *107*, 17–46. https://doi.org/10.1016/j.addr.2016.04.003.
25. Kueny, R. Biocomposites: Composites de Hautes Technologies à Renfort de Fibres Naturelles et Matrice de Résines Naturelles. Ph.D. Thesis, Université de Lorraine, Nancy, France, 2018.
26. Yorseng, K.; Rangappa, S.M.; Pulikkalparambil, H.; Siengchin, S.; Parameswaranpillai, J. Accelerated weathering studies of kenaf/sisal fiber fabric reinforced fully biobased hybrid bioepoxy composites for semi-structural applications: Morphology, thermo-mechanical, water absorption behavior and surface hydrophobicity. *Construct. Build. Mater.* **2020**, *235*, 117464. https://doi.org/10.1016/j.conbuildmat.2019.117464.
27. Lee, C.H.; Khalina, A.; Lee, S.H. Importance of Interfacial Adhesion Condition on Characterization of Plant-Fiber-Reinforced Polymer Composites: A Review. *Polymers* **2021**, *13*, 438. https://doi.org/10.3390/polym13030438.
28. Du, Y.; Yan, N.; Kortschot, M.T. The use of Ramie fibers as reinforcements in composites. In *Biofiber Reinforcements in Composite Materials*; Faruk, O., Sain, M., Eds.; Woodhead Publishing: Sawston, UK, 2015; pp. 104–137. https://doi.org/10.1016/C2013-0-16470-7.

29. Kumar, N.; Das, D. Alkali treatment on nettle fibers Part I: Investigation of chemical, structural, physical, and mechanical characteristics of alkali-treated nettle fibers. *J. Text. Inst.* **2016**, *108*, 1461–1467. https://doi.org/10.1080/00405000.2016.1257346.
30. Thilgavathi, G.; Karan, C.P. Investigations on oil sorptions capacity of Nettle fibrous assembly and 100% Nettle and Nettle/kapok blended needle punched nonwovens. *J. Indus. Text.* **2019**, *49*, 415–430. https://doi.org/10.1177%2F1528083718787532.
31. Davies, G.C.; Bruce, D.M. Effect of relative humidity and damage on the tensile properties of flax and Nettle fibers. *Text. Res. J.* **1998**, *68*, 623–629. https://doi.org/10.1177%2F004051759806800901.
32. Bajpai, P.K.; Meena, D.; Vasta, S.; Singh, I. Tensile behavior of Nettle fiber composites exposed to various environments. *J. Nat. Fibers* **2015**, *10*, 244–256. https://doi.org/10.1080/15440478.2013.791912.
33. Bar, M.; Alagirusamy, R.; Das, A. Properties of flax-polypropylene composites made through hybrid yarn and film stacking methods. *Compos. Struc.* **2018**, *197*, 63–71. https://doi.org/10.1016/j.compstruct.2018.04.078.
34. Ramdani, N.; Derradji, M.; Mokhnach, E.O. Natural fiber reinforced polybenzoxazine composites: A review. *Mater. Today Commun.* **2022**, *31*, 103645. https://doi.org/10.1016/j.mtcomm.2022.103645.
35. Zhan, J.; Wang, G.; Li, J.; Guan, Y.; Zhao, G.; Naceur, H.; Coutellier, D.; Lin, J. Effect of the compatilizer and chemical treatments on the performance of poly (lactic acid)/ramie fiber composites. *Compos. Commun.* **2021**, *27*, 100843. https://doi.org/10.1016/j.coco.2021.100843.
36. Chen, X.; Ren, J.; Zhang, N.; Gu, S.; Li, J. Effects of heat treatment on the thermal and mechanical properties of fabric-reinforced PLA bio composite. *J. Reinf. Plast. Compos.* **2015**, *34*, 28–36. http://dx.doi.org/10.1177/0731684414562222.
37. Xu, H.; Liu, C.; Chen, C.; Hsiao, B.S.; Zhong, G.; Li, Z. Easy alignment and effective nucleation activity of Ramie fibers in injection-molded poly (lactic acid) biocomposite. *Biopolymers* **2012**, *97*, 825–839. https://doi.org/10.1002/bip.22079.
38. Yu, T.; Ren, J.; Li, S.; Yuan, H.; Li, Y. Effect of fiber surface-treatments on the properties of poly (lactic acid)/Ramie composites. *Compos. Part A* **2010**, 41, 499–505. https://doi.org/10.1016/j.compositesa.2009.12.006.
39. Kumar, N.; Das, D. Fibrous biocomposites from Nettle (Girardinia diversifolia) and poly (lactic acid) fibers for automotive dashboard panel application. *Compos. Part B* **2017**, *130*, 54–63. https://doi.org/10.1016/j.compositesb.2017.07.059.
40. Moonart, U.; Utara, S. Effect of surface treatments and filler loading on the properties of hemp fiber/natural rubber composites. *Cellulose* **2019**, *26*, 7271–7295. https://doi.org/10.1007/s10570-019-02611-w.
41. Kumar, N.; Das, D.; Neckar, B. Effect of fiber orientation on tensile behavior of biocomposites prepared from Nettle and poly (lactic acid) fibers: Modeling & experiment. *Compos. Part B* **2018**, *138*, 113–121. https://doi.org/10.1016/j.compositesb.2017.11.019.
42. Fischer, H.; Werwein, E.; Graupner, N. Nettle fibre (Urtica dioica L.) reinforced poly (lactic acid): A first approach. *J. Compos. Mater.* **2012**, *46*, 3077–3087. https://doi.org/10.1177%2F0021998311435676.
43. Pokhriyal, M.; Prasad, L.; Rakesh, P.K.; Raturi, H.P. Influence of fiber loading on physical and mechanical properties of Himalayan Nettle fabric reinforced polyester composite. *Mater. Today Proc.* **2018**, *5*, 16973–16982. https://doi.org/10.1016/j.matpr.2018.04.101.
44. Nandi, P.; Das, D. Mechanical, thermo-mechanical and biodegradation behaviors of green-composites prepared from woven structural nettle (Girardinia diversifolia) reinforcement and poly (lactic acid) fibers. *Ind. Crops Prod.* **2022**, *175*, 114247. https://doi.org/10.1016/j.indcrop.2021.114247.
45. Rajeshkumar, G.; Arvindh Seshadri, S.; Devnani, G.L.; Sanjay, M.R.; Siengchin, S.; Prakash Maran, J.; Al-Dhabi, N.A.; Karuppiah, P.; Mariadhas, V.A.; Sivarajasekar, N.; et al. Environment friendly, renewable and sustainable poly lactic acid (PLA) based natural fiber reinforced composites—A comprehensive review. J. Clean. Prod. 2021, 310, 127483. https://doi.org/10.1016/j.jclepro.2021.127483.
46. Ségard, E.; Benmedakhene, S.; Laksimi, A.; Laï, D. Damage analysis and the fibre–matrix effect in polypropylene reinforced by short glass fibres above glass transition temperature. Compos. Struct. 2003, 60, 67–72. https://doi.org/10.1016/S0263-8223(02)00294-5.