



HAL
open science

Physicochemical and Mechanical Performances of Technical Flax Fibers and Biobased Composite Material: Effects of Flax Transformation Process

M Khennache, A Mahieu, M Ragoubi, S Taibi, C Poilâne, N Leblanc

► To cite this version:

M Khennache, A Mahieu, M Ragoubi, S Taibi, C Poilâne, et al.. Physicochemical and Mechanical Performances of Technical Flax Fibers and Biobased Composite Material: Effects of Flax Transformation Process. *Journal of Renewable Materials*, 2019, 7, pp.821 - 838. 10.32604/jrm.2019.06772 . hal-03347106

HAL Id: hal-03347106

<https://hal.science/hal-03347106>

Submitted on 16 Sep 2021

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.

Physicochemical and Mechanical Performances of Technical Flax Fibers and Biobased Composite Material: Effects of Flax Transformation Process

M. Khennache^{1,*}, A. Mahieu¹, M. Ragoubi¹, S. Taibi¹, C. Poilâne² and N. Leblanc¹

¹UniLaSalle, Unité de recherche Transformations & Agro-Ressources, VAM²IN (EA 7519 UniLaSalle-Université d'Artois), Mont Saint Aignan, France.

²Normandie University, 14032 Caen, France; UNICAEN, CIMAP, 14032 Caen, France; ENSICAEN, 14050 Caen, France; CEA, UMR6252, 14070 Caen, France; and CNRS UMR6252, 14050 Caen, France.

*Corresponding Author: M. Khennache. Email: Mehdi.KHENNACHE@unilasalle.fr.

Abstract: In France, the use of flax fibers as reinforcement in composite materials is growing exponentially in the automotive sector, thanks to their good physicochemical properties, environmental reasons, health neutrality and due to the European Council Directives on the reuse, recycling and valorization of car components and materials. The aim of our study is to investigate biochemical, physicochemical, and mechanical properties of technical flax fibers to evaluate the impact of transformation processes (scutching, hackling, and homogenization) on final properties of associated composite materials. Different chemical analysis such as Van Soest (biochemical fraction measurement), FTIR (Fourier Transform InfraRed spectroscopy), and XRD (X-ray diffraction) were carried out on different process modalities and show that there is no significant difference in terms of biochemical fraction and crystallinity index. By the same token, mechanical behavior shows that Young's modulus is not affected by the transformation process. This result is also observed for thermal behavior. The results highlight the fact that the transformation processes of technical fibers do not really affect their physicochemical and mechanical performances.

Keywords: Technical flax; physicochemical properties; processes; composite

1 Introduction

The use of flax fibers in composite materials retains great attention and is growing in industrial sectors such as automotive, sports and leisure [1]. This interest is due to their physical properties (density and specific mechanical properties) which can be competitive with glass fibers [2]. In addition, flax fibers are renewable resources [3], unharmed to health unlike glass fibers [4] and supposedly low cost [5]. They mainly consist of cellulose, hemicellulose, pectin and lignin which are the most widespread polymers on earth [6]. Other important advantages compared to glass fibers concern the production which requires less energy [7] and biodegradability facilities of flax fibers [8].

Many studies [2,7,9,10] have been carried out on elementary flax fiber but there is a lack of knowledge about the potential of technical flax fibers as reinforcement [11,12]. Furthermore, in real conditions, plant fiber composites are not made with elementary fibers only but also with technical fibers. Technical flax fibers consist of a combination of elementary fibers interconnected at their interface by pectin [11]. They are obtained by scutching flax stem and hackling flax bundles. When the stems are scutched, flax shives and flax tows that have weak bond between fibers are recovered. After that, once this flax tow has been scutched and hackled, the fibers are separated through these weak interfaces and consequently, the fibers are thinner and contain fewer cells in their bundles [13].

Given the complex morphology of the technical flax fibers, their mechanical performances are governed by certain physical properties (density, section size...). Several studies have focused on the determination of fiber section. Ruan et al. [14] have determined the technical flax section using an optical microscope and obtained an average of 4-6 different points throughout gage length. The authors show that for twenty bundles of flax fibers, the diameter value varies from 84.75 to 106.79 μm with a standard deviation ranging from 4.60 to 35.69 μm . Garat et al. [15] have used an automated laser scanning method, it can take for each scan position 600 apparent diameters on the rotating fiber bundle with 40 μm pitch along the fiber, which allows obtaining 45 000 values of apparent diameter for technical flax fibers of 3 mm length. The authors show that for twelve bundles of flax fibers, the apparent diameter was between 49 and 140 μm with a median noted at 91 μm . Masseteau et al. [16] have used a weighing method on yarns which consists of knowing the weight, the density and the length of the yarns. With these values we can easily calculate the section. However, Ilcyszyn et al. [17] developed a method of digital image processing which consists of taking several pictures at different angles (0° , 36° , 72° , 108° and 144°) with specific mounting. After that, they treated the results using software to calculate the area of the fiber. They showed the average diameter for elementary hemp fibers is estimated at 42 μm with a standard deviation of 10 μm . According to the authors, this method can be used also for bundles of fibers.

These methods are limited because the section is not absolutely circular, the morphology is very heterogeneous, and they do not consider fiber lumen and real conditions occurring during the tensile tests (temperature and relative humidity). That is why it is very important to improve the section measurement which is necessary to determine mechanical properties. To determine the section fibers, Masseteau et al. [16] proposed a method based on the weighing of flax specimens. This involves a knowledge of technical flax fibers density which is the most important parameter in order to conduct these measurements accurately. A comparative study between five methods (linear density and diameter calculation, Archimedes, helium pycnometry, gradient column and liquid pycnometry) showed that helium pycnometry gives results with very small standard deviation [18].

The influence of processing of flax fibers and their composites has been analyzed by Van de Weyenberg et al. [19]. They showed that the technical flax fibers with different levels of retting (half, normal and a mixture of green-half retted) are finer after hackling process compared to scutching process. It was suspected that the mechanical properties of composites would decrease due to the damaging of fibers during the process, but the results did not confirm this assumption. This study concluded that refining outweighs the fiber damage induced by the transformation process. It would have been interesting to investigate the impact of the transformation process into the physicochemical properties of technical flax fibers in order to understand more what happens at the level of the technical flax fibers and composites. Unfortunately, we don't have this kind of information in this paper. Regarding the time needed for transformation flax fibers, authors presented a detailed schema of the transformation stages of flax fibers and shows that an optimization of process steps could be reduce the time of preparation and manufacturing of materials (hackling stage comes after the scutching). Concerning the price of flax fibers according to the evolution of transformation process, Van de Weyenberg [20] reported the evolution of the price for 2005. he shows that hackled flax fibers are more expensive than scutched ones, with an average price respectively of 2.5 €/kg and 1.5 €/kg. Moreover, it is important to remind that the price of flax fibers varies from one culture to another. However, the tendency of increasing processing costs remains the same.

Another study focused on the mechanical properties of composites reinforced with flax fibers which come from different positions along the stem (Top, Middle, Bottom) [21]. They show that the composite reinforced with flax from the middle exhibits the best mechanical properties. This observation is also valid for the properties of single flax fibers.

Behind the methodology effects, it is important to analyze genotypic and phenotypic effects. A study on composites reinforced with different varieties of flax fibers (Hermes, Andrea and Marilyn) showed that Marilyn exhibits the best rigidity [22]. Similar findings were also observed for the mechanical properties of single flax fibers. A study [23] carried out on the mechanical properties of different flax varieties (Bolchoï, Eden, Ariane, Liral Prince) showed that the Young's modulus is lower for the older varieties

(Ariane, Liral Prince). The mechanical properties of the newer varieties (Bolchoï, Eden) seem to be better (Young moduli approximately about 52 ± 12 GPa and 40.9 ± 10 GPa respectively for new and old flax varieties). Regarding, the effect of climatic conditions, Lefeuvre et al. [2] have investigated mechanical performances of flax fibers that were grown in the same geographical area (30 km radius) over 4 years. The authors showed that there is no significant effect of climatic conditions on their mechanical behavior.

The aim of our study is to investigate biochemical, physicochemical, and mechanical properties of technical flax fibers from six modalities to evaluate the impact of transformation processes (scutching, hackling and homogenization) on final properties of the associated composite materials. Chemical analysis, Van Soest (biochemical fraction measurement) [24,25], FTIR (Fourier Transform InfraRed spectroscopy), XRD (X-ray diffraction) [15], physical analysis, TGA (Thermo Gravimetric Analysis) and density measurements with Helium pycnometry, and mechanical analysis on technical fibers and composite materials were carried out on all the flax modalities. The results highlight the fact that the transformation processes of technical fibers do not really affect their physicochemical and mechanical performances.

2 Materials and Methods

2.1 Materials

The matrix used in this study is an Epoxy based Prepreg system based on Resin XB 3515/Aradur® 5021. XB 3515 is a hot melt epoxy resin and Aradur® 5021 is a hardener based on polyamine. This matrix system is an HUNSTAMN advanced material used in industrial composites. It has been provided by VITECH COMPOSITES.

The flax was cultivated and retted on the fields in the region of Normandy in France. After the harvesting, the flax stems are transferred to the companies for the scutching and the hackling steps. The scutching enables the separation of the flax fibers from the stem and the hackling permit the refining and homogenization of the flax fibers scutched on the ribbon of flax fibers which will be used to elaborate the UD flax fibers.

Different flax modalities used as reinforcements in composites presented on Tab. 1 have been studied to determine the influence of transformation processes on the physico-chemical properties of technical flax fibers and their influence on the mechanical properties of their composites. All the modalities were conditioned in a conditioning room at 20°C with 65% relative humidity (RH), as mentioned in ASTM D1776-04 [26]. The flax modalities differ by hackling and preparation.

- (i) Hackling: 0 corresponds to scutched flax and 1 corresponds to hackled flax.
- (ii) Preparation: this comprises the number of passages through an Auto-Spreader which allows the homogenization of flax fibers. 0 corresponds to the 1st passage through the Auto-Spreader which is linked to the hackling line. On the other hand, 1 and 2 correspond respectively to the 2nd and 3rd passage in Auto-Spreader which are independent of the hackling line.

The modality number 6 mentioned in Tab. 1 is the reference which is currently marketed by the industry. It is also the most expensive.

Table 1: The six studied modalities

Modalities	Hackled	Preparation
1	0	0
2	0	1
3	0	2
4	1	0
5	1	1
6	1	2

2.2 Experimental Protocol for Composite Materials

Fig. 1 shows the elaboration process of composite materials. The composite materials are constituted by one layer only (Epoxy-FlaxTape-Epoxy) prepared by thermocompression with dimensions of 30×30 cm, then cut with laser machine (280×25 mm) and stuck on the ends of the composite heels (Lin/Epoxy) to minimize composite damage during tensile tests. The dimensions of our specimens were inspired by the international standard organization ISO 527-4 [27]. The weight fractions of flax fibers and those of epoxy for all the modalities are 50%.

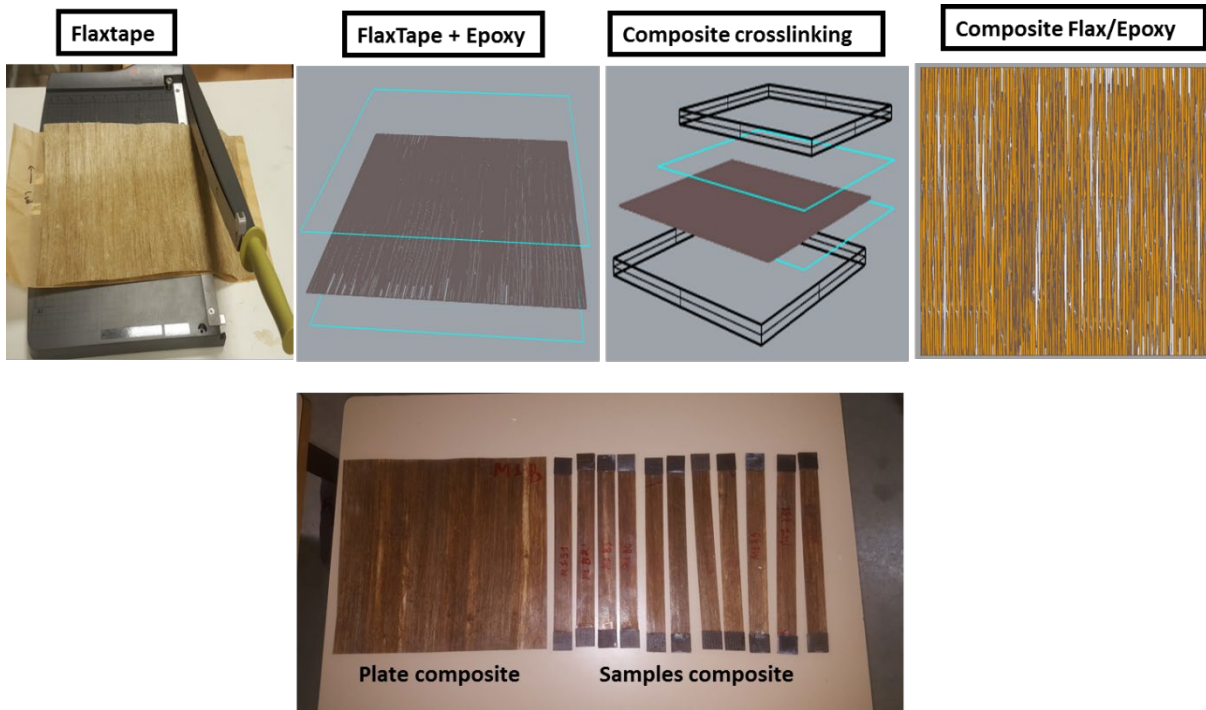


Figure 1: The elaboration steps of composites materials

The curing cycle of composite materials is illustrated in Fig. 2 and declined on four steps:

- i) the first step allows the adhesion between the reinforcement and the matrix,
- ii) the second step consists of the pre-crosslinking of the composites,
- iii) the third step lets the total crosslinking of the composites,
- iv) at the end the cooling step allows recovering the composite.

The increase of the pressure improves the impregnation of the reinforcement with the resin, and the adhesion between matrix and reinforcement.

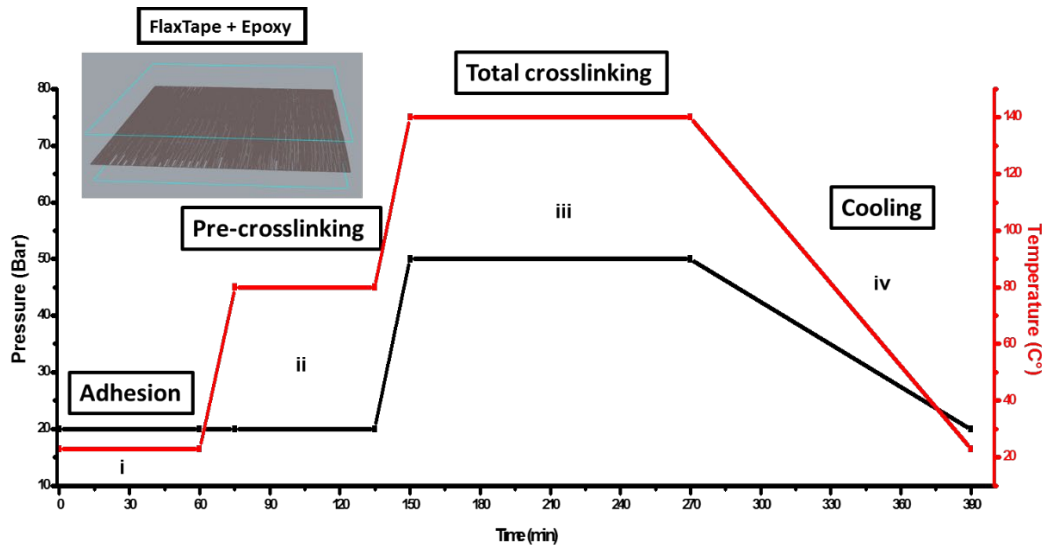


Figure 2: The curing cycle of composite materials

2.3 Methods Analysis

We describe here the method used to determine the physico-chemical properties of technical flax fibers.

2.3.1 Determination of Biochemical Composition

The Van Soest method allows us to determine the biochemical composition (fraction of soluble compounds, hemicellulose, cellulose and lignin). The tests were carried out on 1 gram of material with three repetitions for each modality, using a FOSS manufactured raw fiber extractor device. This method allows to determine the biochemical content by fractionation of plant matter using different solvents.

2.3.2 Fourier Transform InfraRed Spectroscopy (FTIR Analysis)

The infrared analyzes were carried out with three repetitions to complete the results of the Van Soest method by seeing if there are variations of the biochemical composition at the surface (results not presented in this paper). The flax fibers were characterized with FTIR, ATR mode (Attenuated Total Reflectance) using spectrometer (Thermo Scientific Nicolet iS10) in the frequency range $4000-900\text{ cm}^{-1}$ with a resolution of 4 cm^{-1} , 64 scans have been carried out for each sample. The recorded ATR-FTIR spectrums are not presented in this paper but discussed to complete the results of the Van Soest method.

2.3.3 X Ray Diffraction Analysis

The XRD technique was used to estimate the crystallinity index according to the Segal method [28]. This study was carried out with three tests, the measurements were made on disks fibers having a diameter of 30 mm and a thickness of 3 mm obtained by compression. X-ray diffractograms were recorded from $2\theta = 3$ to 60° , with a scan rate of $0.04\text{ }^\circ\text{s}^{-1}$. The crystallinity index must not be confused with the crystallinity rate because it is not the same method and the results obtained are not comparable. We consider that the crystallinity index allows detecting some variation in crystallinity between different characterized samples. This value is calculated from the following equation:

$$CrI (\%) = \frac{I_{002} - I_{am}}{I_{002}} \times 100$$

where $CrI (\%)$ is the relative degree of crystallinity, I_{002} is the maximum intensity of the 002 lattice diffraction and I_{am} is the intensity of diffraction of the amorphous material at $2\theta = 18^\circ$.

2.3.4 Thermogravimetric Analysis (TGA)

TGA analysis was carried out to study the thermal stability of the fibers. Three tests were carried out under air from 25 to 800°C with a heating rate of 3°C. This analysis was performed using Netzsch TG 209 F1 equipment.

2.3.5 The Density Measurement

The density was determined with helium pycnometer (AccuPyc 1330 Pycnometer from Micromeritics). Three tests were carried out on different samples for each modality, each result is an average of three measurements carried out on the same sample.

2.3.6 Tensile Tests for the Technical Flax Fibers and Composites

For fiber samples, tensile tests have been carried out using MTS Criterion 43 machine with a 0.5 kN load cell and a crosshead displacement fixed at 1 mm.min⁻¹. Sample fibers were glued on paper frames cut with Trotec laser machine to have perfect gauge length. Gauge lengths were chosen from 10 mm to 100 mm by step of 2 mm. Concerning composites samples, tensile tests were carried out on an Instron 8801 machine fitted with 100 kN load cell and crosshead displacement fixed at 2 mm.min⁻¹. The gauge length was fixed at 230 mm. For each modality, nine tests were realized.

2.3.7 The Proposed Method of Determining the Technical Fiber Section

Fig. 3 shows the heterogeneity which is seen in the shape of the technical flax fibers. We cannot use the classical method of determination of the diameter for the technical flax fibers under microscope which is used currently for the single fiber, because compared to the single flax fibers, the technical flax fibers present high heterogeneity in the dimension depending on how they are positioned on the frame (front or profile view). Also, there is another problem with this method which is the under estimation of the section because the size of the lumen is not taken into account. For all these reasons we developed a new method in our laboratory.

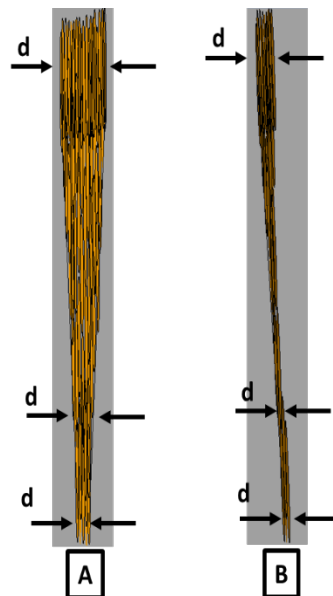


Figure 3: Technical Flax fiber **A.** front, **B.** profile view

This method allows to determine the true section of the technical flax fibers. It consists of weighing the technical flax fibers on a microbalance with a precision of 1 µg in which temperature and relative

humidity are controlled, on the same condition of their tensile tests and to determine their densities with the helium pycnometry. Thanks to this data, the sections are easily calculated using the following equation:

$$S_t = \frac{m_f}{\rho_f \times L_f}$$

In this equation, where S_t expresses the true section of the technical flax fiber, m_f is the mass of the technical flax fiber, ρ_f is the density of the technical flax fiber and L_f is the length of the technical flax fiber. The weighing process of the technical flax fibers follows two steps.

- (i) Firstly, we realize the tensile tests of the technical flax fibers. Testing is monitored to eliminate specimens where fibers are lost, and the relative humidity and temperature conditions are noted for every specimen. Through the tests, a range of temperatures from 17 to 23°C is observed and the relative humidity was between 35 to 51%.
- (ii) Secondly, we carefully recover each specimen in aluminum paper, and we note their designation. Finally, we weigh on the microbalance each specimen with and without the technical fibers three times. The difference between these values give the mass of the technical flax fibers:

$$m_f = m_{with} - m_{without}$$

where m_f is the mass of the technical flax fiber, m_{with} is the mass of the frame with technical flax fiber and $m_{without}$ is the mass of the frame without technical flax fiber.

2.3.8 Analyses of Variance (ANOVA)

Statistical analysis was assessed by mean comparison with an ANOVA test. Significant differences are revealed by a P -value inferior to 0.05.

3 Results and Discussion

Fig. 4 displays the biochemical fraction of fibers components obtained by the Van Soest method. As can be shown, flax fibers are mostly composed of cellulose (81% to 82%). However, for the hemicellulose rate, it varies from 6.5% to 9%. The lignin content is less than 5%. The remainder corresponds to the soluble compounds which vary between 6.6% and 8.5% in the studied fibers. In view of the small differences observed between the results and the values of the standard deviations, statistical analyses were carried out. These showed that P -values are 0.64, 0.32, 0.93, 0.08 respectively for soluble, hemicellulose, cellulose and lignin fractions. The ANOVA results showed that the P -values are superior to 0.05 for all components, which allows us to conclude that there is no significant difference in biochemical composition between all flax modalities.

Tab. 2 shows the biochemical compositions of some plant fibers found in the literature with different methods, sometimes the used method is reported in the article, but it is not the case for all articles.

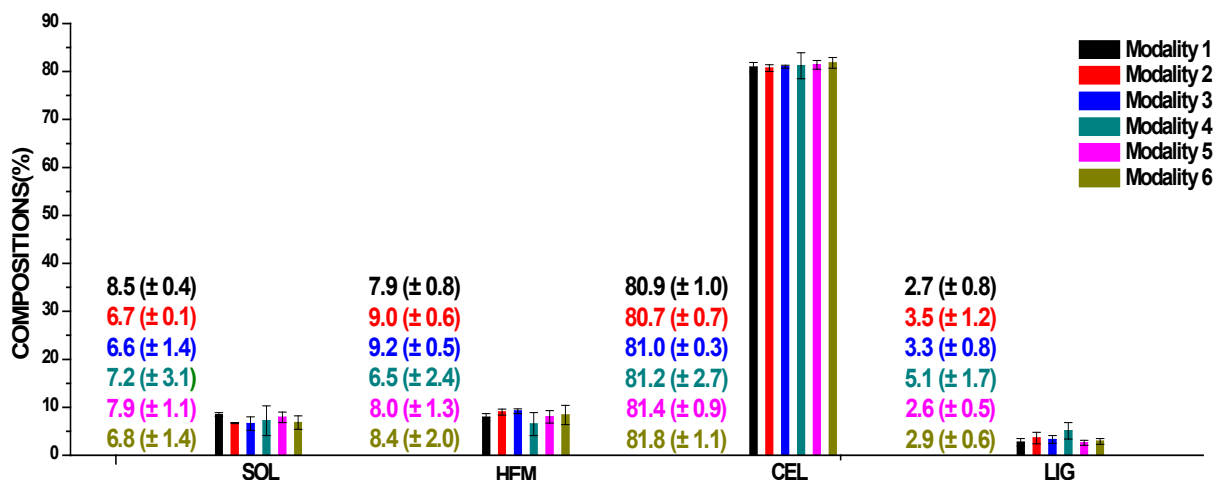


Figure 4: Biochemical compositions (SOL: soluble, HEM: hemicellulose, CEL: cellulose, LIG: lignin)

Table 2: Biochemical composition of plants fibers

Natural fibers	Cellulose (%)	Hemicelluloses (%)	Lignin (%)	Others (%)	Wax/Fat (%)	Pectines (%)	Moisture (%)	Methods	Ref.
Flax	79	11	3	7	-	-	-	Van Soest and Wine	[29]
Flax	71	18.6-20.6	2.2	-	1.7	2.3	-	-	[30]
Flax (unretted)	56.5	15.4	2.5	10.5	1.3	3.8	10	-	[31]
Flax (retted)	64.1	16.7	2.0	3.9	1.5	1.8	10	-	[31]
Flax	85	9	4	-	-	-	-	-	[32]
Flax Stem	49	29	18	-	-	3	-	-	[32]
Flax shives	53	13	24	>3.5	-	-	-	-	[33]
Wood	48	15	24	13	-	-	-	Van Soest and Wine	[29]
Hemp	72	10	3	15	-	-	-	Van Soest and Wine	[29]
Hemp (Raw)	-	18.42	6.77	-	2.3	6.17	-	GB 5881-86	[34]
Hemp (1week retting)	-	17.16	6	-	0.44	4.8	-	GB 5881-86	[34]
Hemp (2weeks retting)	-	16.5	4.23	-	0.23	3.2	-	GB 5881-86	[34]
Jute	64.4	12.0	11.8	1.1	0.5	0.2	10	-	[35]
Sisal	65.8	12.0	9.9	1.2	0.3	0.8	10	-	[35]
Abaca	63.2	19.6	5.1	1.4	0.2	0.5	10	-	[35]
Coir	35-45	1.25-2.5	30-46	-	1.3-1.80	-	20	-	[35]

When we compare these results with our results, we can see clearly that there are significant differences with some natural fibers. For example, the fraction of cellulose in our flax fibers is about 81.5% and that of wood, sisal and coir are respectively 48%, 65.8% and 35-45%. The part of hemicellulose in our fibers is around 8.2% and that of the plant fibers mentioned earlier correspond to 15%, 12% and 1.25-2.5%.

The portion of lignin in our fibers is around 3.5% and that of the other natural fiber cited previously are 24%, 9.9%, 30-46% [29,35].

It is very important not to confuse and compare the biochemical content of the flax stem with that of flax fibers, because it is not the same thing. In the flax stem there are the flax shives with the flax fibers and their compositions (the fraction of the components) are very different [32]. As we can see, this difference is clearly observed in the data reported from the literature. The fractions of cellulose, hemicellulose and lignin for the flax stems are 49%, 29% and 18 % and those of flax fibers are very much larger: 56.5-85% for the cellulose, 9-20.6% for hemicellulose and 2.2-4% for lignin. Despite the large data variations between the different flax fibers, the difference is significant between the flax stems and the flax fibers. This high variation is due to the composition of the flax shives which is estimated to 53% for the cellulose, 13% hemicellulose and 24% for the lignin. There is more lignin and less cellulose in the flax shives than in the flax fibers [29-33]. This can be due to the variety of the fiber, the weather conditions during the growth phase, the nature of the soil, the retting time, the dew during the retting and the fiber turning in field during the retting step. We can also see that the natural fibers which are composed with a majority of cellulose are the flax and the hemp, which can explain that these fibers have the best mechanical properties.

To complete and confirm the results of the biochemical composition obtained by Van Soest method, ATR-FTIR analysis were performed. This method gives a representation of the chemical bonds present on the surface of the analyzed samples. The comparison between the different spectra obtained by ATR-FTIR analysis shows no variation between the six modalities. This confirms the absence of variation in the chemical composition between the flax fibers of the different modalities as seen before with the Van Soest method.

The crystallinity index is calculated by the Segal method [28] for each batch of flax fibers. The results are presented in Tab. 3. The crystallinity indexes obtained are between 85.1% and 86.1%. Given the *P*-value equal to 0.86 for the six modalities, we can deduce that there is no significant variation. These results are similar to those found in the literature [14,29] and estimated between 83.53% and 86.1%.

Tab. 3 shows the crystallinity index of various plant fibers which have been studied by different researchers. We can see clearly that the hemp and flax fibers have the highest crystallinity index which is estimated respectively around (66-85%) and (85.53-86.1%). In comparison those of jute, sisal, wood, luffa and kapok fibers have a lower index that is estimated respectively at 78.47, 70.9, 65.1, 50.00 and 45.75%. We can see also that there is high variation in the ratio of crystallinity index for the same hemp fibers having different degree of retting which increases from 66 to 85%. This observation has been confirmed by other authors who concluded that the increase of the retting times evolves the augmentation of the crystallinity index which can be related to the degradation of non-cellulosic compounds during the retting [36].

Table 3: Crystallinity index of our technical flax fibers and those of plants fibers found in the literature

Plant fiber	Crystallinity index (%)	Ref.
Modality 1	85.3 (\pm 0.4)	
Modality 2	85.1 (\pm 0.6)	
Modality 3	85.2 (\pm 1.7)	
Modality 4	85.8 (\pm 0.5)	
Modality 5	85.2 (\pm 0.4)	
Modality 6	86.1 (\pm 0.2)	
Flax	86.1	[29]
Flax 2days water-reeted	83.53 (\pm 0.31)	[14]
Flax 10days water-reeted	83.65 (\pm 2.84)	[14]
Hemp	79.9	[29]
Hemp (green)	66	[34]

Hemp 1-week reeting	84	[34]
Hemp 2-weeks reeting	85	[34]
Jute	78.47	[37]
Sisal	70.9	[37]
Wood	65.1	[29]
Luffa	50.00	[38]
Kapok	45.75	[37]

Fig. 5 shows the derivative thermogravimetric curves. According to these curves the flax fibers are degraded into three distinct mass losses. The first loss of mass occurs between 25 and 100°C. It corresponds to the evaporation of water [39]. The second takes place around 180-350°C, it corresponds to the degradation of pectins, waxes, hemicelluloses and celluloses [7,40]. The third loss of mass occurs around 350-450°C, it corresponds to the degradation of non-polysaccharide substances such as phenols [7] and may be some oxidized residues and residues of the 2nd loss which are not fully degraded.

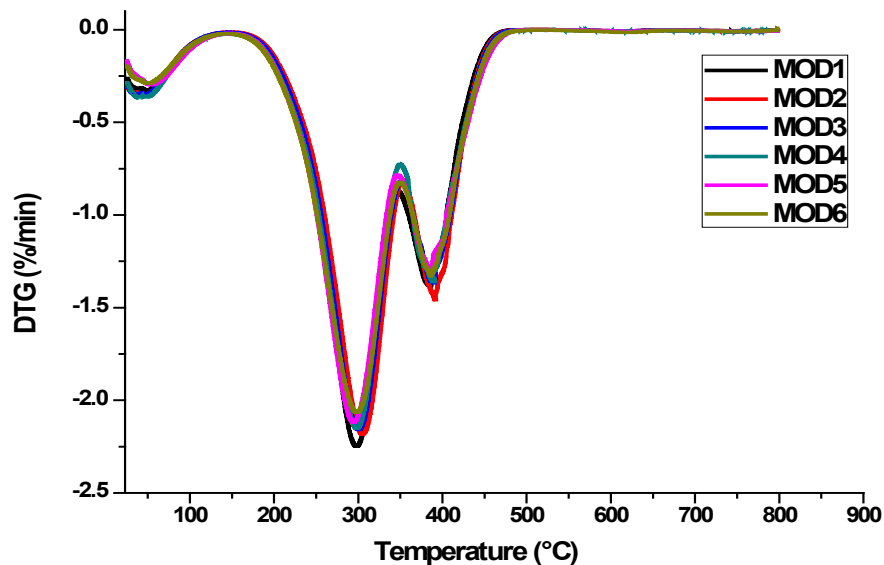


Figure 5: Derivative thermogravimetric (DTG) on the six modalities

The results of thermogravimetric analysis show that the transformation process does not affect the thermal properties of the different studied modalities.

Fig. 6 shows the variation of the fiber density measurements. All modalities display values in the range of 1.52 and 1.53. More precisely, results show a slight increase with the passage number (homogenization process of the fibers) for both series of fibers: scutched (modalities 1 to 3) or hackled (modalities 4 to 6). Statistical analysis displays P -value equal to 5.46×10^{-4} and confirms that this slight increase of the density is significant. This trend could be attributed to the removal of lightweight components during the homogenization steps.

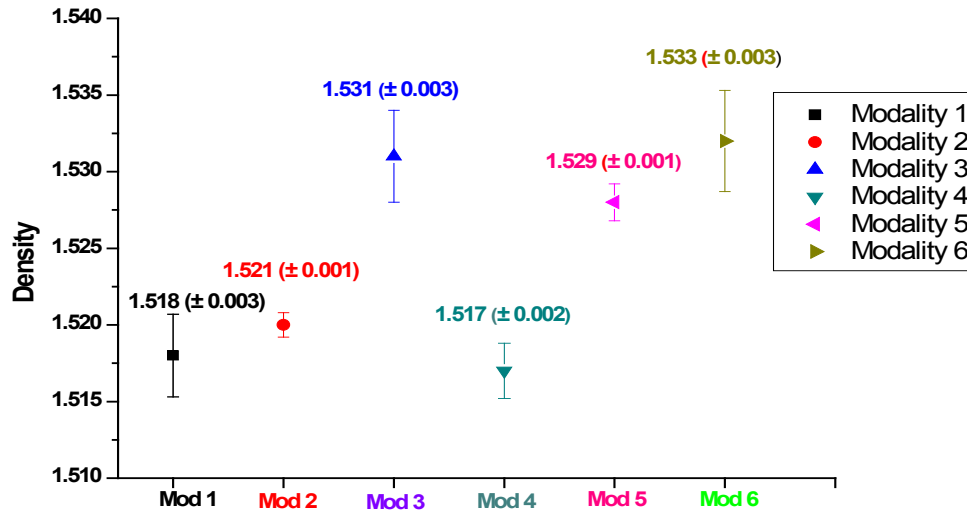


Figure 6: The Density of the 6 flax categories

The density reported in the literature is estimated at 1.55 ± 0.18 for flax fibers [41]. The density determined from the immersion in different oils is between 1.41-1.47 and the one determined with helium pycnometry for the same fibers is 1.4900 ± 0.0022 [42]. The results of the density by immersion methods with different oils and helium pycnometry are different even if the studied materials are the same. This is a good example to show the importance of the chosen method and also the comparison of the results obtained. Relating the standard deviations obtained by the linear density method to those by the helium pycnometry, a ratio of $0.18/0.0022 = 82$ is found. It clearly shows the good precision of the later.

During tensile test, three types of fracture mechanisms were noted for technical flax fibers (total, partial and sequential fracture). They are illustrated in Fig. 7. The first mechanism is the total fracture (Fig. 7(a)) which means that the technical fiber breaks in two pieces once. The second one is the partial fracture (Fig. 7(b)), it corresponds to the break of the technical fibers in two steps or more. The major break being followed by others. It means that the technical fiber was constituted by two or more equivalent bundles of elementary fibers. The last type of fracture is the sequential one (Fig. 7(c)) which occurs in multiple steps before the main fracture. In fact, several thin fibers, elementary or not, break and move away from the main sample step by step.

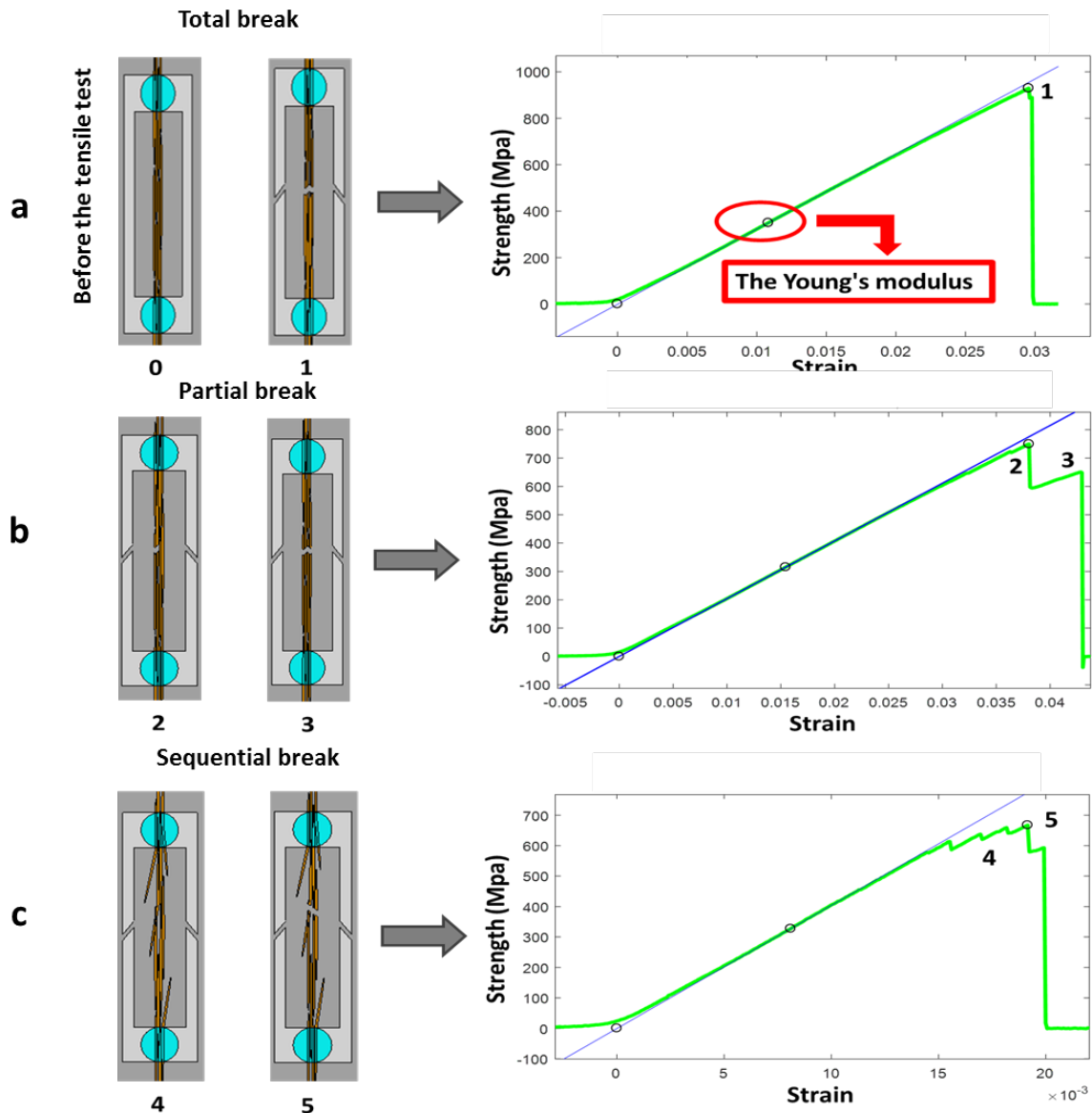


Figure 7: Description of breaking mechanisms for technical flax fibers. **a).** Total break, **b).** Partial break, **c).** Sequential break

Results of mechanical properties for the six modalities are regrouped in Tab. 4. The Young's modulus is calculated where the slope is maximal because we consider that the rigidity of the fiber is maximal at this point. It is estimated between 43.1 GPa and 46.4 GPa for all the modalities. In view of the high variation of the standard deviations and results of ANOVA (P -value = 0.95), we can conclude that the variation of the rigidity is not significant. The same observation was observed for the tensile strength which varies from 518 MPa to 618 MPa and for the failure strain which is estimated between 1.4% and 1.6% with P -values respectively of 0.51 and 0.36. These results confirm that the variation of the mechanical performances between the different technical flax fibers were insignificant.

Tab. 4 also shows mechanical property values from literature. The values of Young's modulus are dispersed from 18 to 70 GPa [13,14,43-46]. This wide range of results can be explained by treatment [45], time of reeting [7], variety of flax [23] and method of measuring their section, particularly. We cannot

really compare our results with those found in the literature because we do not use the same method to define the section, but we can have an idea of the reliability of our method because our values are included in the range defined before.

Table 4: Mechanical properties of technical flax fibers

Technical flax fibers	Young's modulus (GPa)	Tensile strength (MPa)	Failure strain (%)	Ref.
Modality 1	43.1 (\pm 15.6)	558 (\pm 231)	1.6 (\pm 0.7)	
Modality 2	45.5 (\pm 14.1)	545 (\pm 234)	1.5 (\pm 0.6)	
Modality 3	45.3 (\pm 16.1)	549 (\pm 228)	1.5 (\pm 0.7)	
Modality 4	43.8 (\pm 13.8)	518 (\pm 190)	1.4 (\pm 0.4)	
Modality 5	46.4 (\pm 20.4)	618 (\pm 236)	1.6 (\pm 0.6)	
Modality 6	44.5 (\pm 18.0)	541 (\pm 279)	1.4 (\pm 0.5)	
Technical fibers	30-70	-	-	[13]
Technical fibers	25.0-33.0	458-648	1.73-3.54	[14]
Technical fibers (Raw)	-	750 (\pm 131)	-	[43]
Technical fibers (dewaxed)	-	820 (\pm 52)	-	[43]
Technical fibers 10 mm	38.4 (\pm 2.2)	613 (\pm 76)	0.95 (\pm 0.02)	[44]
Technical fibers 15 mm	45.9 (\pm 2.6)	724 (\pm 150)	1.1 (\pm 0.3)	[44]
Technical fibers 20 mm	54.4 (\pm 2.0)	812 (\pm 176)	1.25 (\pm 0.33)	[44]
Technical fibers 25 mm	56.5 (\pm 3.0)	641 (\pm 369)	1.01 (\pm 0.51)	[44]
Technical fibers 30 mm	57.5 (\pm 5.1)	650 (\pm 286)	1.07 (\pm 0.40)	[44]
Technical fibers (Raw)	30 (\pm 11)	300 (\pm 100)	1.1 (\pm 0.4)	[45]
Technical fibers treated with maleic anhydride	18 (\pm 5)	185 (\pm 60)	1.2 (\pm 0.3)	[45]
Technical fibers treated with acetic anhydride	24 (\pm 10)	185 (\pm 85)	0.8 (\pm 0.2)	[45]
Technical fibers treated with silane	40 (\pm 13)	555 (\pm 210)	1.6 (\pm 0.6)	[45]
Technical fibers treated with styrene	28 (\pm 9)	245 (\pm 95)	1.1 (\pm 0.4)	[45]
Technical fibers 20 mm	-	613 (\pm 442)	-	[46]
Technical fibers 40 mm	-	454 (\pm 231)	-	[46]
Technical fibers 80 mm	-	264 (\pm 127)	-	[46]

To extend the investigations on Young's modulus for each modality, we studied the linear regression according to the cross-section. The intercept values show that the Young's moduli are estimated from 58.9 to 79.2 GPa when the fiber cross-section tends to zero. These results tend to increase according to the scutching/hackling and homogenization steps (Fig. 8).

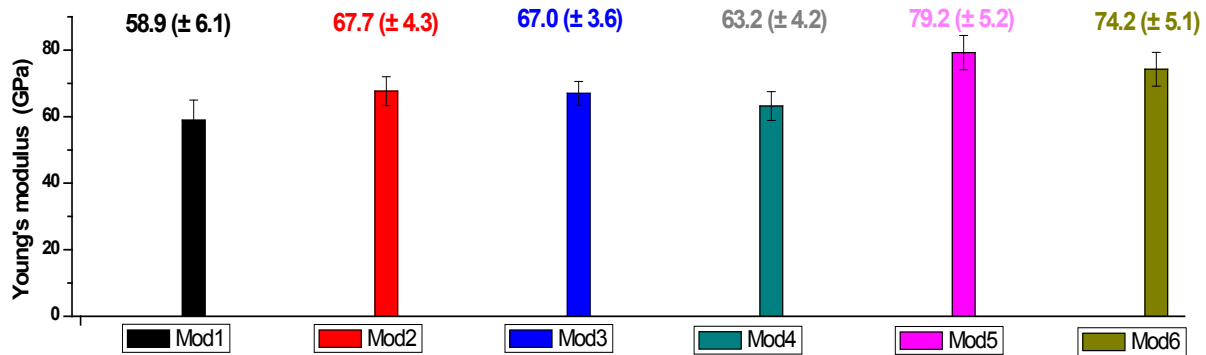


Figure 8: Young's modulus originally of technical flax fibers when the section tends to 0

We thought that the removal of lightweight components (confirmed with the results of density measurement) favors the stiffness of flax fiber which improves the mechanical behavior of flax fibers. Furthermore, note that when the cross-section tends to zero, Young's modulus are rather similar to those of elementary flax fibers extracted from the top and middle of the flax stem reported by Charlet et al. [21] (68.6 ± 21.3 GPa or 76.7 ± 40.8 GPa depending on the diameter of fiber). From this correlation we assume that the performances of technical fibers tend to those of elementary flax fibers when the cross-section of the technical fiber is extrapolated to zero. In fact, the thinner a technical fiber, the closer to elementary fiber.

The evolution of the Young's modulus of associated composites is finally represented in Fig. 9. The average modulus of composite materials is estimated around 22.3 ± 1.3 GPa. In view of the mechanical results obtained on the composites and considering the result of ANOVA (P -value = 0.71), the tensile tests revealed that there is no significant variation between the different modalities. This conclusion is consistent with the work of Van de Weyenberg et al. [19]. The synergistic effect of the reinforcement with the epoxy matrix in the composite material outweighs the variation of the mechanical properties observed on technical fiber alone.

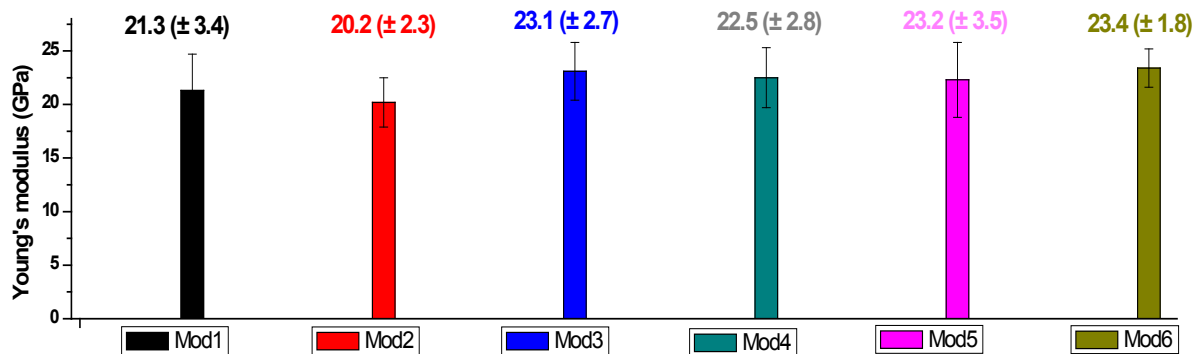


Figure 9: Young modulus of different composites materials (one ply)

Tab. 5 shows the mechanical properties of similar composites elaborated with epoxy and flax fibers found in the literature. We can see clearly that the range of the results is very wide, the Young's modulus going from 11.1 GPa to 39 GPa. This high variation is due to the weight or the volume fraction of flax fibers, the positions of the fibers along the stem (Top, Middle, Bottom) [21], the flax variety [22,23] and the process of composite elaboration [47]. For example, Oksman et al. reported a Young's modulus for 46% fraction of fibers at 35 GPa which is manufactured using the resin transfer molding (RTM). Compared to our results on composite materials with a weight fiber fraction of 50% and prepared by thermocompression, we see that the Young's modulus reported in the literature could be higher, which expresses superior

efficiency of the reinforcement [47]. Our results are rather close to those of Coroller et al. for composites elaborated with fibers of the varieties Hermes and Andera by the same process. They reported a Young's modulus for both varieties respectively at 26 ± 2.0 GPa and 28 ± 3.6 GPa [22].

Table 5: Mechanical properties of composites (Flax/Epoxy)

Composites with Flax/Epoxy	Weight fraction (%)	Volume fraction (%)	Tensile strength (MPa)	Young's modulus (MPa)	Elongation at break (%)	Ref.
UD	26	21	193 (± 30)	22 (± 4)	0.9	[47]
UD	46	42	280 (± 15)	35 (± 3)	0.9	[47]
UD	56	47	279 (± 14)	39 (± 6)	0.8	[47]
UD	37	32	132 (± 4.5)	15 (± 0.6)	1.2	[48]
UD (Top)	-	19.7	126 (± 14)	12.4 (± 1.3)	1.3 (± 0.2)	[21]
UD (Middle)	-	20.1	127 (± 14)	16.7 (± 3.7)	0.9 (± 0.2)	[21]
UD (Bottom)	-	19.8	113 (± 11)	11.1 (± 1.4)	1.5 (± 0.1)	[21]
UD	-	40	133	28	-	[19]
UD	-	44	259	26.3	1.4	[49]
UD	-	47 (± 2)	296 (± 0.5)	27.2 (± 0.5)	1.65 (± 0.06)	[50]
UD (Hermes)	-	51 (± 2)	408 (± 36)	26 (± 2.0)	1.3 (± 0.05)	[22]
UD (Andrea)	-	51 (± 4)	290 (± 22)	28 (± 3.6)	1.1 (± 0.15)	[22]
UD (Marylin)	-	54 (± 3)	364 (± 14)	34 (± 3.0)	1.3 (± 0.01)	[22]

4 Conclusion

The aim of our study is to investigate biochemical, physicochemical, and mechanical properties of technical flax fibers to evaluate the impact of transformation processes (scutching, hackling, and homogenization) on final properties of the associated composite materials. The results obtained both with the Van Soest method and FTIR analysis show that there is no significant difference in terms of biochemical composition and nature of chemical bonds present on the fiber surface between fibers having undergone different levels of physical treatment. This may confirm the absence of chemical variation between the different flax modalities. This result was also confirmed by the TGA and XRD analysis which show no significant change in terms of thermal stability and microstructural properties. Therefore, there is no substantial impact of transformation process on the physicochemical properties of flax modalities. The only property which seems to vary a little according to the transformation process is the flax fibers density. It could be due to the removal of lightweight components in the surface of the technical flax fibers during the homogenization steps. These results highlight the fact that the transformation processes of the technical fibers do not really affect their physicochemical and mechanical performance on composites.

According to these results, the industrial partners have estimated that time and money could be saved thanks to the suppression of the hackling step which follows the scutching. It has been shown that the technical flax fibers properties would not be affected by this suppression. Modality 3 has been chosen by the firm to continue this project.

Acknowledgement: This study is funded by FEDER and it was carried out in collaboration with LSM and LINEO Company. Thanks to Nicolas COUV RAT and Céline BENS AKOUN from the laboratory of Separative Sciences and Methods for the DRX analyzes.

References

1. Mathijssen, D. (2018). The renaissance of flax fibers. *Reinforced Plastics*, 63, 138-144.
2. Lefeuvre, A., Bourmaud, A., Morvan, C., Baley, C. (2014). Tensile properties of elementary fibres of flax and glass: Analysis of reproducibility and scattering. *Materials Letters*, 130, 289-291.
3. Baley, C. (2004). Influence of kink bands on the tensile strength of flax fibers. *Journal of Materials Science*, 39, 331-334.
4. Le Duigou, A., Davies, P., Baley, C. (2012) Interfacial bonding of Flax fibre/Poly(L-lactide) bio-composites. *Composites Science and Technology*, 70, 231-239.
5. Bourmaud, A., Le Duigou, A., Gourier, C., Baley, C. (2016). Influence of processing temperature on mechanical performance of unidirectional polyamide 11-flax fibre composites. *Industrial Crops and Products*, 84, 151-165.
6. Baley, C. (2002). Analysis of the flax fibres tensile behaviour and analysis of the tensile stiffness increase. *Composites: Part A*, 33, 939-948.
7. Martin, N., Mouret, N., Davies, P., Baley, C. (2013). Influence of the degree of retting of flax fibers on the tensile properties of single fibers and short fiber/polypropylene composites. *Industrial Crops and Products*, 49, 755-767.
8. Baley, C., Busnel, F., Grohens, Y., Sire, O. (2006). Influence of chemical treatments on surface properties and adhesion of flax fibre-polyester resin. *Composites: Part A*, 37, 1626-1637.
9. Martin, N., Davies, P., Baley, C. (2016). Evaluation of the potential of three non-woven flax fiber reinforcements: Spunlaced, needlepunched and paper process mats. *Industrial Crops and Products*, 83, 194-205.
10. Bourmaud, A., Morvan, C., Bouali, A., Placet, V., Perré, P. et al. (2013). Relationships between micro-fibrillar angle, mechanical properties and biochemical composition of flax fibers. *Industrial Crops and Products*, 44, 343-351.
11. Bos, H. L., Van Den Oever, M. J. A., Peters, O. C. J. J. (2002). Tensile and compressive properties of flax fibres for natural fibre reinforced composites. *Journal of Materials Science*, 37, 1683-1692.
12. Charlet, K., Jernot, J., Gomina, M., Bizet, L., Bréard, J. (2010). Mechanical properties of flax fibers and of the derived unidirectional composites. *Journal of Composite Materials*, 44, 2887-2896.
13. Bos, H. L. (2004). *The potential of flax fibres as reinforcement for composite materials (Ph.D. Thesis)*. Eindhoven: Technische Universiteit Eindhoven.
14. Ruan, P., Du, J., Gariépy, Y., Raghavan, V. (2015). Characterization of radio frequency assisted water retting and flax fibers obtained. *Industrial Crops and Products*, 69, 228-237.
15. Garat, W., Corn, S., Le Moigne, N., Beaugrand, J., Bergeret, A. (2018). Analysis of the morphometric variations in natural fibres by automated laser scanning: Towards an efficient and reliable assessment of the cross-sectional area. *Composites Part A*, 108, 114-123.
16. Masseteau, B., Michaud, F., Irle, M., Roy, A., Alise, G. (2014). An evaluation of the effects of moisture content on the modulus of elasticity of a unidirectional flax fiber composite. *Composites: Part A*, 60, 32-37.
17. Ilczyszyn, F., Cherouat, A., Montay, G. (2014). Effect of hemp fibre morphology on the mechanical properties of vegetal fibre composite material. *Advanced Materials Research*, 875-877, 485-489.
18. Truong, M., Zhong, W., Boyko, S., Alcock, M. (2009). A comparative study on natural fibre density measurement. *Journal of the Textile Institute*, 100, 525-529.
19. Van de Weyenberg, I., Ivens, J., De Coster, A., Kino, B., Baetens, E. et al. (2003). Influence of processing and chemical treatment of flax fibres on their composites. *Composites Science and Technology*, 63, 1241-1246.
20. Van de Weyenberg, I. (2005). *Flax fibers as a reinforcement for epoxy composites (Ph.D. Thesis)*. Katholieke Universiteit Leuven.
21. Charlet, K., Baley, C., Morvan, C., Jernot, J. P., Gomina, M. et al. (2007). Characteristics of Hermès flax fibres as a function of their location in the stem and properties of the derived unidirectional composites. *Composites: Part A*, 38, 1912-1921.
22. Coroller, G., Lefeuvre, A., Le Duigou, A., Bourmaud, A., Ausias, G. et al. (2013). Effect of flax fibres individualisation on tensile failure of flax/epoxy unidirectional composite. *Composites: Part A*, 51, 62-70.
23. Baley, C., Goudenhoofft, C., Perré, P., Lu, P., Pierre, F. et al. (2019). Compressive strength of flax fibre bundles within the stem and comparison with unidirectional flax/epoxy composites. *Industrial Crops & Products*, 130, 25-33.

24. Goering, H. K., Van Soest, P. J. (1970). *Forage fiber analyses (apparatus, reagents, procedures, and some applications)*. US Agricultural Research Service, Washington, D.C.
25. Van Soest, P. J. (1963). Use of detergents in the analysis of fibrous feeds. II. A rapid method for the determination of fiber and lignin. *Journal of the Association of Official Analytical Chemists*, 46, 829-835.
26. ASTM D1776-04 (2004). Standard practice for conditioning and testing textiles. *American Society for Testing and Materials*.
27. ISO 527-4 (1997). Test conditions for isotropic and orthotropic fibre-reinforced plastic composites. *International Organization for Standardization*.
28. Segal, L., Creely, J. J., Martin Jr, A. E., Conrad, C. M. (1959). An empirical method for estimating the degree of crystallinity of native cellulose using the X-Ray diffractometer. *Textile Research Journal*, 29, 786-794.
29. Tserki, V., Zafeiropoulos, N. E., Simon, F., Panayiotou, C. (2005). A study of the effect of acetylation and propionylation surface treatments on natural fibres. *Composites: Part A*, 36, 1110-1118.
30. Mohanty, A. K., Misra, M., Hinrichsen, G. (2000). Biofibres, Biodegradable polymers and biocomposites: an overview. *Macromolecular Materials & Engineering*, 1(24), 276-277.
31. Batra, S. K. (2007). *Other Long Vegetable Fibers*: Abaca, Banana, Sisal, Henequen, Flax, Ramie, Hemp, Sunn, and Coir. 453-520*, Taylor & Francis Group. LLC, Boca Raton, FL.
32. Salmon-Minotte, J., Franck, R. R. (2005). *Flax, 94-174*. Woodhead Publishing Limited and CRC Press LLC, Boca Raton, FL.
33. Sain, M., Fortier, D. (2005). Flax shives refining, chemical modification and hydrophobisation for paper production. *Industrial Crops and Products*, 15, 1-13.
34. Li, Y., Pickering, K. L., Farrell, R. L. (2009). Analysis of green hemp fibre reinforced composites using bag retting and white rot fungal treatments. *Industrial Crops and Product*, 29, 420-426.
35. Pritchard, M., Sarsby, R. W., Anand, S. C. (2000). *Textiles in civil engineering. Part 2-natural fibre geotextiles. 372-406*. Woodhead Publishing Ltd and CRC Press LLC, Cambridge, CB.
36. Mazian, B., Bergeret, A., Benezet, J., Malhautier, L. (2018). Influence of field retting duration on the biochemical, microstructural, thermal and mechanical properties of hemp fibres harvested at the beginning of flowering. *Industrial Crops & Products*, 116, 170-181.
37. Mwaikambo, L. Y., Ansell, M. P. (1999). The effect of chemical treatment on the properties of hemp, sisal, jute and kapok for composite reinforcement. *Angewandte Makromolekulare Chemie*, 272, 108-116.
38. Ghali, L., Msahli, S., Zidi, M., Sakli, F. (2009). Effect of pre-treatment of Luffa fibres on the structural properties. *Materials Letters*, 63, 61-63.
39. Kim S. H., Park, C. H. (2017). Direct impregnation of thermoplastic melt into flax textile reinforcement for semi-structural composite parts. *Industrial Crops and Products*, 95, 651-663.
40. Khalfallah, M., Abbès, B., Abbès, F., Guo, Y. Q., Marcel, V. et al. (2014). Innovative flax tapes reinforced Acrodur biocomposites: a new alternative for automotive applications. *Materials and Design*, 64, 116-126.
41. Poilâne, C., Cherif, Z. E., Richard, F., Vivet, A., Ben Doudou, B. et al. (2014). Polymer reinforced by flax fibres as a viscoelastoplastic material. *Composite Structures*, 112, 100-112.
42. Amiri, A., Triplett, Z., Moreira, A., Brezinka, N., Alcock, M. et al. (2017). Standard density measurement method development for flax fiber. *Industrial Crops and Products*, 96, 196-202.
43. Bos, H. L., Molenveld, K., Teunissen, W. (2004). Compressive behaviour of unidirectional flax fibre reinforced composites, *Journal of Materials Science*, 39, 2159-2168.
44. Thomason, J. L., Carruthers, J., Kelly, J., Johnson, G. (2011). Fibre cross-section determination and variability in sisal and flax and its effects on fibre performance characterisation. *Composites Science and Technology*, 71, 1008-1015.
45. Alix, S., Philippe, E., Bessadok A., Lebrun, L., Morvan, C. et al. (2009). Effect of chemical treatments on water sorption and mechanical properties of flax fibres. *Bioresource Technology*, 100, 4742-4749.
46. Romhány, G., Kocsis, J. K., Czigány, T. (2003). Tensile fracture and failure behavior of technical flax fibers. *Journal of Applied Polymer Science*, 90, 3638-3645.
47. Oksman, K. (2001). High quality flax fibre composites manufactured by the resin transfer moulding process. *Journal of Reinforced Plastics and Composite*, 20, 621-627.

48. Oksman, K., wallström, L., Berglund, L. A., Filho, R. D. T. (2002). Morphology and mechanical properties of unidirectional sisal-epoxy composites. *Journal of Applied Polymer Science*, 84, 2358-2365.
49. Cherif, Z. E., Poilâne, C., Momayez, L., Chen, J. (2012). Pré-imprégnés lin/époxy: influence des paramètres d'élaboration sur les propriétés mécaniques. *Matériaux & Techniques*, 100, 459-466.
50. Kersani, M., Lomov, S. V., Vuure, A. W. V., Bouabdallah, A., Verpoest, I. (2015). Damage in flax/epoxy quasi-unidirectional woven laminates under quasi-static tension. *Journal of Composite Materials*, 49, 403-413.