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1 **Can we predict the microstructure of a non-woven flax/PLA composite through**  
2 **assessment of anisotropy in tensile properties?**

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12 **Abstract**

13 Flax/Poly-(lactide) non-woven composites are an alternative to conventional  
14 glass/poly-(propylene), offering a potentially lower environmental impact solution for  
15 the automotive industry. To understand this complex material, its detailed architecture  
16 and void distribution are examined through 3D microtomography. Anisotropy in fibre  
17 orientation is also observed, further verified through off-axis tensile tests. Then micro-  
18 mechanics and laminate theory are used, taking into account fibre orientation  
19 distribution, to predict mechanical properties and compare with experimental  
20 measurements. Even though some deviation is observed at off-axis angles greater than  
21 45°, we report that off-axis tensile tests (property) can be used to predict fibre  
22 orientation (structure) **in industrial production facilities as a simple means of quality**  
23 **control and tailored design of new non-woven preforms.**

24 **Keywords**

25 A. Natural fibre composites ; B. Mechanical properties ; C. Anisotropy ; D. X-ray  
26 tomography ; B. Porosity/void

## 27 **1. Introduction**

28 Due to their lower environmental impact [1] and competitive specific mechanical  
29 properties [2], flax fibres have replaced glass fibres in some automotive parts, such as  
30 interior panels [3]. Manufactured from non-woven preforms, these parts are thermo-  
31 compressed to a near-net shape. Several processes are available to manufacture non-  
32 woven preforms, among which spunlacing and needle-punching are common [4]. In  
33 addition, thermoplastic polyolefins, such as poly-(propylene) (PP), are currently used  
34 as a matrix, leading to the recyclability of any scraps. However, with the emergence of  
35 biodegradable thermoplastics, the automotive industry has started to look at  
36 alternatives, such as poly-(lactide), which offer industrial composting as an alternative  
37 end-of-life scenario [5].

38 Furthermore, PLA appears to have the advantage to be stiffer than PP with a tangent  
39 modulus of 3.8 GPa against 1.4 GPa [6] and to have a quasi-linear tensile behaviour. As  
40 matrix properties influence the mechanical properties of the composite, non-woven  
41 flax/PLA composites are observed to have higher stiffness at all volume fractions than  
42 non-woven flax/PP composites (Figure 1).

43 While matrix properties are relevant, it is mainly the fibres responsible for composite  
44 mechanical behaviour, with their content directly affecting composite stiffness. The  
45 non-woven preform manufacturing process will induce fibre orientation anisotropy  
46 resulting in different properties in the machine- and cross- directions of a needle-  
47 punching line [4]. Neckar and Das [7] studied the orientation of the fibres in non-

48 woven preforms and derived analytical laws related to the manufacturing process  
49 used. The fibre orientation in the non-woven preforms could be obtained simply by  
50 transparency observation or optical microscopy [8]. Such methods are relevant for  
51 mono-constituent non-woven preforms. For composites manufacturing, the non-  
52 woven preforms may comprise two types of fibres, the reinforcement and the matrix.  
53 Other methods have to be used as only the reinforcement fibre orientation needs  
54 measurement (as the matrix fibres will melt) to investigate the effect of fibre  
55 architecture on composite mechanical properties. Graupner et al. [9] used a  
56 synchrotron radiation-based micro-computer tomography approach to obtain cellulose  
57 fibre orientation in PLA composites. Another non-destructive method is to use  
58 ultrasound scanning, leading to the measurement of fibre orientation and ply spacing,  
59 fibre volume fraction and porosity distribution [10]. In both cases, computation is  
60 required to extract fibre orientation from the raw data, leading orientation  
61 information on the composite volume and not only on the surface.

62 The orientation of the flax in the composite (or the non-woven preform) is of great  
63 industrial interest. It can be used as a quality controller for industrial production or for  
64 pilot tests to validate machinery set-up to obtain specific fibre orientations and  
65 tailored architectures. The latter is of interest as highly aligned non-wovens offer a  
66 viable route, technically and economically speaking, to obtain anisotropic preforms  
67 with mechanical properties intermediate to quasi-isotropic non-wovens and  
68 unidirectional mats.

69 Once orientation frequency distribution is known, several theories exist to predict non-  
70 woven mechanical properties. The first approach is to modify the rule-of-mixtures (1)

71 by including a corrective factor  $\eta_o$  (2) [11], where  $E$  and  $V$  denote stiffness and volume  
 72 fraction respectively, and subscripts  $NW$ ,  $f$  and  $m$  denote non-woven composite, fibre  
 73 constituent and matrix constituent, respectively. The parameter  $p_n$  is the appearance  
 74 frequency of the angle represented by  $\theta_n$ .

$$E_{NW} = \eta_o \cdot E_f \cdot V_f + E_m \cdot V_m \quad (1)$$

$$\eta_o = \sum_n p_n \cdot \cos^4(\theta_n) \quad (2)$$

75 In the above approach, even though the fibre orientation is taken into account, the  
 76 shear contribution is not taken into account, nor are the anisotropic properties of the  
 77 reinforcement. Halpin and Pagano [12] developed a more precise method by  
 78 assimilating randomly oriented fibrous composites to a laminate. By considering  
 79 symmetric laminate with a thickness weighted with the fibre orientation frequency,  
 80 they showed a good mechanical prediction.

81 A third constituent important to tackle is the porosity value. The compaction of the  
 82 non-woven could be controlled, leading to low or higher porosity content, to tailor  
 83 mechanical or acoustic properties [13]. Focussing on fully-compressed composites,  
 84 porosity is still present due to manufacturing and should not be neglected as it  
 85 influences mechanical properties. Their distribution and shape are also reported to be  
 86 important [14]. Looking specifically at natural fibres, Madsen et al. discussed the  
 87 porosity distribution inside hemp thermoplastic composites [15] and its influence on  
 88 composite stiffness [16]. Thus, fibre orientation, fibre anisotropy and porosity appear  
 89 to be key structural parameters required in precisely describing a non-woven  
 90 composite, all of them impacting non-woven composite mechanical properties.

91 In this paper, the microstructure (porosity/reinforcement content) and the  
92 architecture (fibre orientation) of a flax/PLA non-woven are characterised thanks to X-  
93 ray computed microtomographic (X-ray micro-CT) analysis. Off-axis tensile tests are  
94 used to characterise the observed anisotropy. Micro-mechanics and laminate theory  
95 are used to approximate experimental data. Matching calculated and measured values  
96 allows proposing the off-axis mechanical properties as an efficient and simple  
97 validation tool to address the fibre orientation in a non-woven composite.

## 98 **2. Materials and Methods**

### 99 **2.1. Reinforcements**

100 Flax/PLA non-woven preform was provided by Ecotechnilin (Yvetot, France). It was  
101 made from 50%/50% weight of scutched flax tows and INGEO™ PLA fibres. Due to the  
102 utilisation of flax tow, shives were still present in the non-woven preform. Our non-  
103 woven presents a larger proportion of preferential fibres orientations than needle-  
104 punched non-wovens due to their manufacturing process: carding and calendaring.  
105 The preferential fibres direction is observed on the machine direction (direction of the  
106 non-woven preform production). Cross direction refers to the direction perpendicular  
107 to the machine direction. Non-wovens with 10% and 30% fibre weight content were  
108 also manufactured for comparison. Following, if nothing is indicated, non-woven  
109 composite refers to the 50%/50% flax/PLA non-woven.

110 Unidirectional (UD) and bi-axial (BX) composites were made of 50 g/m<sup>2</sup> Flax-tape®  
111 (Ecotechnilin) and PLA films. This film was made in our laboratory with a process  
112 developed in a previous study [6].

## 113        **2.2. Composite manufacturing**

114        Non-woven composites were made of several preform plies, UD and BX lay-ups were  
115        prepared via the film stacking method. Pure PLA plates of both references were also  
116        prepared to obtain matrix properties. These lay-ups were dried in an oven at 40°C for  
117        24h under vacuum as flax and PLA are moisture-containing. They were then hot  
118        compressed at 200°C with a hydraulic press LabTech Scientific 50T (Labtech,  
119        Samutprakarn, Thailand), yielding laminate plates of 20 cm x 20 cm x 2mm. The  
120        optimised pressure cycle used is presented in a previous study [6]. A milling machine  
121        was used to cut samples for tensile property characterisation.

## 122        **2.3. Tensile tests**

123        An Instron universal testing machine was used to achieve static tensile tests based on  
124        ISO 527-4. A displacement rate of 1mm/min was applied, and the elongation was  
125        recorded with a unidirectional extensometer, gauge length taken equal to 25mm. For  
126        the pure PLA samples as well as for the UD, a bi-axial extensometer was preferred. For  
127        each formulation, at least five samples were tested to obtain mean values and  
128        standard deviation. The ultimate strength and strain are recorded as well as tangent  
129        modulus. The latter was calculated over a strain of 0.02% to 0.1% as recommended for  
130        flax composites [17]. The Poisson's ratio is obtained using the NF EN 2561 standard  
131        when the bi-axial extensometer is used.

132        The same set-up is used, using a bi-axial extensometer and speed of 2mm/min, to do  
133        some in-plane shear testing on BX according to ISO 14129. As reported previously [18],  
134        the shear modulus is measured between a shear strain of 0.1% and 0.5%.

135 Pure PLA, UD and BX characterisation is only performed for back-calculation to obtain  
 136 flax fibres properties.

#### 137 **2.4. Density**

138 The density of our non-woven composite was obtained through a hydrostatic **scale**  
 139 using ethanol as the immersion liquid. This method was chosen as suggested by  
 140 Kergariou et al. [19] for flax/PLA composite. It was used to obtain the density of the  
 141 composites. Knowing the apparent density of the composite ( $\rho_c$ ), the volume fraction  
 142 of porosity ( $V_p$ ) of each formulation is obtained using equation (3), where  $W_f$  is the  
 143 weight fraction of fibres and  $\rho_{PLA}$ ,  $\rho_{flax}$ , are the density of PLA and flax taken  
 144 respectively as 1.24 [6] and 1.5 [20]. Porosity results are given as mean values of five  
 145 samples.

$$V_p = 1 - \left( \frac{1 - W_f}{\rho_{PLA}} + \frac{W_f}{\rho_{flax}} \right) \cdot \rho_c \quad (3)$$

#### 146 **2.1. X-ray microtomography (X-ray micro-CT)**

147 The microstructure of the non-woven composites was investigated with a Brucker<sup>®</sup>  
 148 SkyScan 1272 high-resolution scanner. The samples were scanned at a nominal  
 149 resolution of 4,5  $\mu\text{m}$ . The current (50 eV), the intensity (200  $\mu\text{A}$ ) of the X-ray beam, and  
 150 the nature and thickness of filters were selected to obtain a constant signal  
 151 transmission of 30%. The X-Ray power source ( $P=U.I$ ) is kept constant at 10W. A  
 152 camera pixel binning of 4032 $\times$ 2688 was applied. The scanned orbit was 180 degrees  
 153 with a rotation step of 0.2 $^\circ$  adapted to the magnification. Bruker's NRecon<sup>®</sup> software  
 154 was used to reconstruct the scan projections into 2D images using the Feldkamp  
 155 algorithm. Gaussian smoothing, ring artefact reduction and beam hardening correction



156 were applied. Volume rendered 3D images were generated using an RGBA transfer  
157 function in SkyScan CTVOX® software. Image analysis was performed using SkyScan  
158 CTAn® software. A specific task list analysis was developed to characterise the porosity  
159 and the pore size distribution within composites. In that way, two image  
160 segmentations were successively carried out on the original image: the first one to  
161 define the sample volume of interest (VOI) and the second one, using an automated  
162 Otsu algorithm, to define the object volume within this VOI. After image binarisation,  
163 structure separation (=pore size) is preceded by skeletonisation in which the two  
164 medial pore axes are identified. Then the "sphere-fitting" local thickness measurement  
165 is made for all the voxels lying along this axis. In order to determine fibre orientation, a  
166 sequence of 12 in-plane cuts on face direction on the 3D render is used to generate 2D  
167 images. This method is based on granulometry analysis and was the subject of a  
168 previous study [21]. Due to the low contrast – a result of the comparable material  
169 densities– between flax and PLA, the 2D images had to be pre-treated with Fiji to  
170 obtain binary images before orientation analysis. The pre-treatment process is  
171 illustrated in S.I. Figure 1 and validated by comparing the analysis of the orientation of  
172 the fibre, with and without pre-treatment, of an optical micrograph, S.I Figure 2.

## 173 **2.2. SEM**

174 Complementary microstructure observations were performed using a JEOL SEM (JSM-  
175 IT500HRSEM) at an acceleration voltage of 3 kV. Before the observation step, gold  
176 sputter coating was applied to the samples using an Edward sputter coater  
177 (Scancoat6).

## 178 **3. Results and discussion**

### 179        **3.1. Fine-scale composite volumetric analysis**

180        The behaviour of a composite is predominantly influenced by the matrix, the  
181        reinforcement properties and their content. In the case of flax reinforcements, things  
182        are more complex as the matrix is not reinforced by one single type of constituent but  
183        by at least two: elementary fibres and fibre bundles. Figure 2 presents SEM  
184        observations of our non-woven composite. The presence of elementary flax fibres and  
185        bundles is confirmed, as well as any preferential orientation. Indeed, non-woven  
186        composites are anisotropic as they present higher properties in one direction than  
187        another, depending on the preferential orientation of the fibres [4]. However, the  
188        microstructure of this non-woven composite is more complex as it also includes  
189        porosity and shives. In addition, quantification of fibre orientation is required to  
190        describe the non-woven composite architecture, the aim being to understand better  
191        and estimate the composite properties.

#### 192                *3.1.1. Porosity analysis*

193        Porosity content of  $5.6 \pm 3.2$  % was measured for the non-woven composite thanks to  
194        a hydrostatic balance using ethanol as the immersion liquid. To investigate pore  
195        localisation and size, a fine-scale analysis was done by micro-CT. A porosity of 4.5 %  
196        was obtained, which is comparable to the immersion method value. It could be  
197        observed in Figure 3.c) thanks to the microtomographic 3D view of porosity. Via a  
198        quantitative analysis presented in Figure 3.b) a few meso-porosity are highlighted  
199        inside the matrix.  
200        On the other hand, there is an important amount of micro-porosity. The latter appears  
201        to be mainly located inside bundles and/or in shives. Indeed, shives come mainly from

202 the xylem tissue, where its main function is to conduct sap [22]. Thus, inside the stem,  
203 these cells exhibit a large lumen of several micrometres, potentially visible through  
204 micro-CT investigations.

### 205 *3.1.2. Shives quantification*

206 Regarding the porosity analysis, it appears that shives are a predominant host of  
207 micro-porosity. It is observed in Figure 2 and confirmed in Figure 4 that shives inside  
208 the non-woven composite had collapsed cells, probably due to the compaction  
209 pressure applied during the manufacturing process. Indeed, shives contain less  
210 cellulose than elementary flax fibres, 45% of dry matter against 80% respectively [23].  
211 As cellulose is responsible for plant fibres' mechanical properties, it is necessary to  
212 quantify this third reinforcement type as it should have mechanical behaviour lower  
213 than that of the bundles and elementary fibres. Following manual extraction, 5wt% of  
214 shives were extracted for the total non-woven reinforcement weight. Considering a  
215 density of  $1,430 \text{ kg.m}^{-3}$  [24], the volume fraction of shives inside the composite is  
216 assessed to be 2.2%, against 43.1% for flax fibres (elementary fibres and fibre bundles).  
217 In the following analysis, shives are **taken** into account by assimilating them into the  
218 total flax fibre content at 45.3% by volume.

### 219 *3.1.3. In-plane orientation of fibres*

220 Finally, a fine-scale description of the non-woven composite architecture should  
221 include fibre orientation. Thanks to micro-CT, the orientation was obtained from 12  
222 images, and the mean orientation was calculated. The analysis methods for one image  
223 is detailed in Figure 5 b), c), and d). All results are summarised in Figure 5 e). A  
224 preferential orientation appears with a maximum relative frequency (calculated

225 degree by degree) of 0.82% and a minimum of 0.38%. It illustrates the expected  
 226 anisotropy of non-woven materials at the composite scale. This orientation anisotropy  
 227 appears to be lower than the pure flax fibres non-woven preform analysed with the  
 228 same technique by Gager et al. [21]. This can be explained by the presence of fine PLA  
 229 fibres in our non-woven preform, modifying the flax fibre alignment during the  
 230 calendaring process. The mean experimental curve, equation (4) developed by Neckar  
 231 and Das [7], is used to fit experimental data (fig. 5.e).

$$f(\theta) = \frac{1}{\pi} \frac{C}{C^2 - (C^2 - 1)\cos^2(\theta)} + q \quad (4)$$

232 where  $f(\theta)$  is the fibre frequency at an angle  $\theta$  taken between  $-90^\circ$  and  $+90^\circ$ ,  $C$  is the  
 233 fitting parameter corresponding to the anisotropy of the non-woven, and the offset is  
 234 corrected via the parameter  $q$ . The fit was done by lowering the sum of the difference  
 235 between experimental and model at each orientation, leading to  $C = 1.84$  and  $q =$   
 236  $0.235\%$ . As the trend curve is symmetric, it is preferred over the experimental curve  
 237 for the micro-mechanical development in section 3.2.3.

### 238 **3.2. Mechanical characterisation and prediction of non-woven composite**

239 Fibre orientation is a key parameter affecting mechanical properties; its direct analysis  
 240 is possible through expensive and time-consuming 3D investigations such as micro-CT.  
 241 In contrast, using mechanical property measurements as an indirect method to  
 242 ascertain fibre orientation may be faster, easier, and lower-cost. To check the  
 243 relevancy of this alternative method, off-axis tensile experiments are compared to a  
 244 theoretical prediction using laminate theory as described by Halpin and Pagano [12].  
 245 The orientation data for the non-woven composites analysed in the preceding sections

246 are matched with a theoretical model, which is used to implement a laminate with 181  
247 distinct laminae (from -90 to +90) with a thickness weighted by the relative orientation  
248 frequency. As symmetric lay-up is required, each ply is made of an angle and its  
249 symmetric counterpart. The laminate creation is summarised in S.I. Figure 3.

### 250 *3.2.1. Angle influence on the tensile response of non-woven composite*

251 To study anisotropy in tensile properties arising from fibre orientation, experimental  
252 measurements are presented in Table I and Figure 6. As expected, the loading angle  
253 influences the material's response. A decrease in stiffness and strength of 40.5% and  
254 47.3%, respectively, are observed between the orientations 0° and 90°, see Figure 7  
255 and Table I. This confirms the anisotropy of this material. Furthermore, the mechanical  
256 properties of the non-woven composite stay unchanged after a critical angle of 67.5°,  
257 being equal to the transverse properties.

### 258 *3.2.2. Flax fibres properties for laminate theory*

259 Input values greatly impact micro-mechanical models, especially for plant fibre whose  
260 values have a larger scatter. Indeed, regarding the longitudinal Young's modulus of  
261 elementary flax fibres, it is possible to find literature values ranging from 36 GPa to 75  
262 GPa [25,26]. Noting this scatter in data, two sets of values are considered here. The  
263 first one is directly extracted from literature, presented in  
264 Table II. The second one is obtained experimentally through back-calculation of UD  
265 flax/PLA composite.

266 Table II presents the mechanical properties of these UD composites and the back-  
267 calculated flax fibre properties for each UD composite. Comparing this data with the  
268 literature, flax fibres longitudinal modulus is similar, whereas its transverse and shear

269 modulus obtained are lower. Thus, the thermal history of the fibres and the  
270 compaction during the process may have impacted the structure of the flax fibres as  
271 well as their mechanical properties.

272 Furthermore, both bundles and shives have been considered as 'fibres' during the  
273 back-calculation. They generally have lower transverse and shear properties than  
274 elementary flax fibres.

275 The reinforcement distribution inside the composite is also irregular, leading to some  
276 regions of higher stress distribution than the theoretical approach. Additionally, as a  
277 high volume fraction is achieved, some contact between fibres are present. These  
278 contact points will be damaged during processing, damaging the fibres.

### 279 *3.2.3. Influence of non-woven structure on its mechanical properties*

280 Comparing the laminate theory models with the experimental data will inform us  
281 whether off-axis tensile tests can be used to predict the fibre orientation of non-woven  
282 flax composite. For each set of values, the mechanical properties of a ply are obtained  
283 using the rule of mixture for the longitudinal direction and the Poisson's ratio and  
284 using Halpin-Tsai [27] for the transverse and shear moduli. Furthermore, as discussed  
285 in section 3.1.1, porosity is present and has to be taken into account. We use the  
286 hydrostatic balance-obtained single porosity value and assume that it is concentrated  
287 in the matrix. This induces that the porosity inside shives is assimilated to be matrix  
288 porosity. Equation (5), developed by Madsen et al. [16], is used to account for the  
289 effect of matrix porosity on mechanical properties,

$$P_{m,real} = P_{m,bulk} \cdot (1 - V_p)^2 \quad (5)$$

290 with  $P$  representing any mechanical property,  $V_p$  the volume fraction of porosity and  
291 subscripts  $m, \text{bulk}$  and  $m, \text{real}$  denoting the bulk matrix and the matrix with porosity. Once  
292 the ply properties are obtained, the laminate is built as explained previously, and the  
293 mechanical properties of the equivalent composite are found. For each set of values,  
294 the model generated a non-woven composite's tangent modulus versus orientation  
295 curve. The range between both is represented in Figure 7 (grey area). The modulus in  
296 the machine direction ( $0^\circ$ ) is well-predicted, falling between both models. A small  
297 deviation appears for  $22.5^\circ$  and  $45^\circ$ , but the models stay close to the experimental  
298 value.

299 However, increasing the orientation further to  $45^\circ$  leads to an overestimation. This  
300 deviation at high orientations is principally due to the assumptions and simplifications  
301 used for the model compared to reality. First of all, the model considers an in-plane  
302 orientation. It is relevant for machine direction due to a stretching induced by the  
303 manufacturing process. However, this stretching is less or not present in other  
304 directions. That is why a slight out-of-plane orientation exists, needed for handling the  
305 non-woven preform.

306 Additionally, the model does not consider the reinforcement effect of bundles. A  
307 bundle present a higher diameter than elementary flax fibres and an irregular  
308 geometry (fluctuating circularity, section and length), increasing the heterogeneity of  
309 the composite structure. Bundles behaviour depends on its cohesion, meaning the  
310 middle lamella properties. The latter impacts predominantly the composite properties  
311 reliant on matrix and interface (such as transverse tensile properties). Furthermore, a  
312 small number of shives were detected, adding heterogeneity in the structure too.

313 What is more, the manufacturing process smashes the shives. This transversal  
314 smashing creates damages in this porous structure, decreasing the transverse  
315 properties of shives. Furthermore, the model used here is based on an assumption of  
316 linear behaviour. However, it is known that elementary flax fibres do not have a linear  
317 longitudinal behaviour [28]. The transverse tensile behaviour of elementary flax fibres  
318 is still obscure as it is harsh to make some relevant tests at this micro-scale. In the non-  
319 woven composite, fibres are oriented but are also curved. This curvature is not  
320 considered in our orientation analysis, but it is known to have a non-negligible impact  
321 on the composite behaviour.

322 Finally, it appears that the anisotropy increases with fibre volume fraction, as observed  
323 in Figure 8. This new manufacturing process of non-woven preform allows overtaking  
324 in machine direction literature value without decreasing the stiffness significantly in  
325 cross-direction.

#### 326 **4. Conclusion**

327 The complex microarchitecture of a non-woven flax/PLA composite with 50%wt of flax  
328 fibres was described thanks to micro-CT analysis. The microstructural observations  
329 show porosity and reveal the preferential orientation of the fibres inside the  
330 composite in the machine direction. This anisotropy was also measured through off-  
331 axis tensile tests. The mechanical properties highlight a clear anisotropy where  
332 transversal properties are 60% of the longitudinal properties. Both anisotropy  
333 characterisations are compared using micro-mechanics and laminate theory to predict  
334 the mechanical properties from the micro-CT orientation analysis.



335 It is found that off-axis tensile tests could be used to predict the fibre orientation  
336 distribution in a non-woven composite indirectly. This is interesting as tensile tests are  
337 much cheaper, faster and convenient to perform than micro-CT analysis. **Industrially, it**  
338 **could be used for quality control of a production line or as a tool to develop new non-**  
339 **woven preforms with specific flax fibres orientation. The comparison of several set-up**  
340 **machinery or even manufacturing processes could also be handled.**

341 However, some deviation is apparent between experimental data and micro-  
342 mechanical prediction because of the complexity of describing a flax composite due to  
343 the presence of flax bundles, shives, curved fibres, potential out-of-plane orientation  
344 of fibres, and also compounding effects from processing/thermal history. **It may be of**  
345 **interest to further develop the models and ascertain their consistency with**  
346 **experimental data by examining the specific influence of shives and degree of flax fibre**  
347 **individualisation on the mechanical properties of non-woven flax composites.**  
348 **However, this may be experimentally challenging as flax preform and fibre quality,**  
349 **degree of fibre individualisation and quantity of shives are inherently interlinked.**

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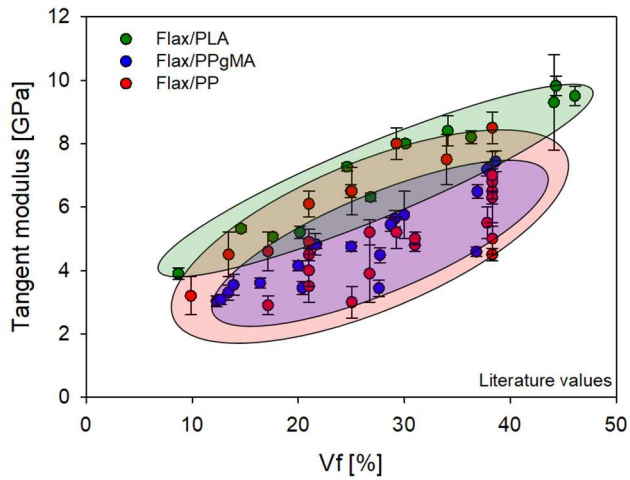
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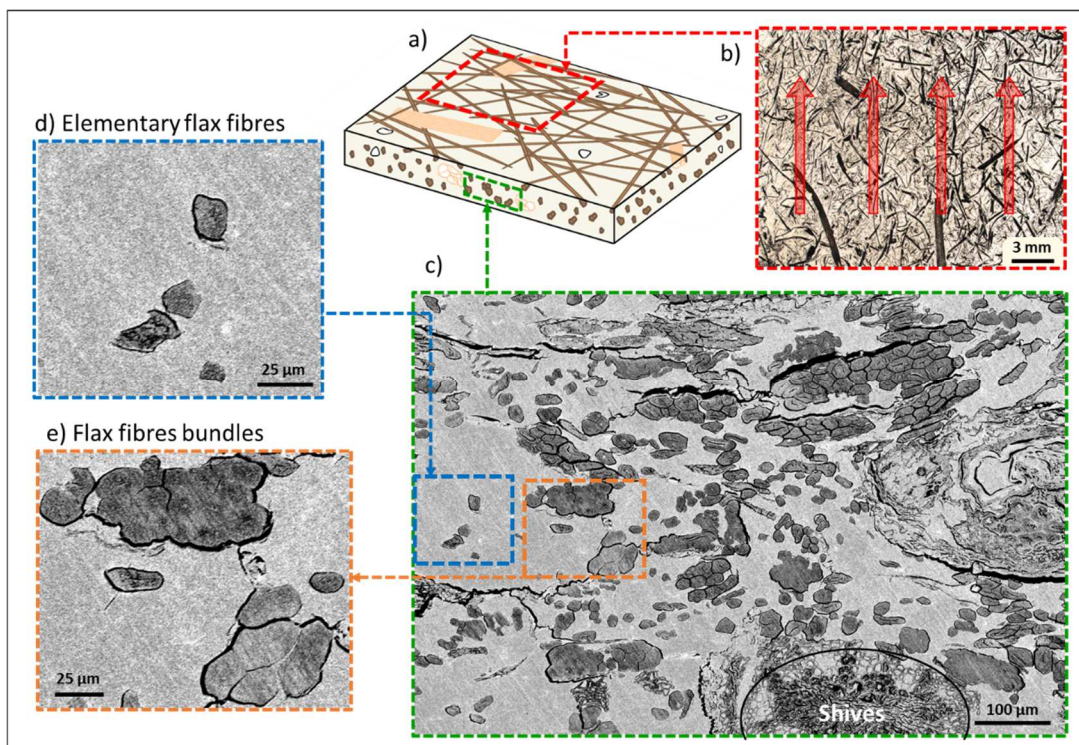
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487 **Figures:**

488

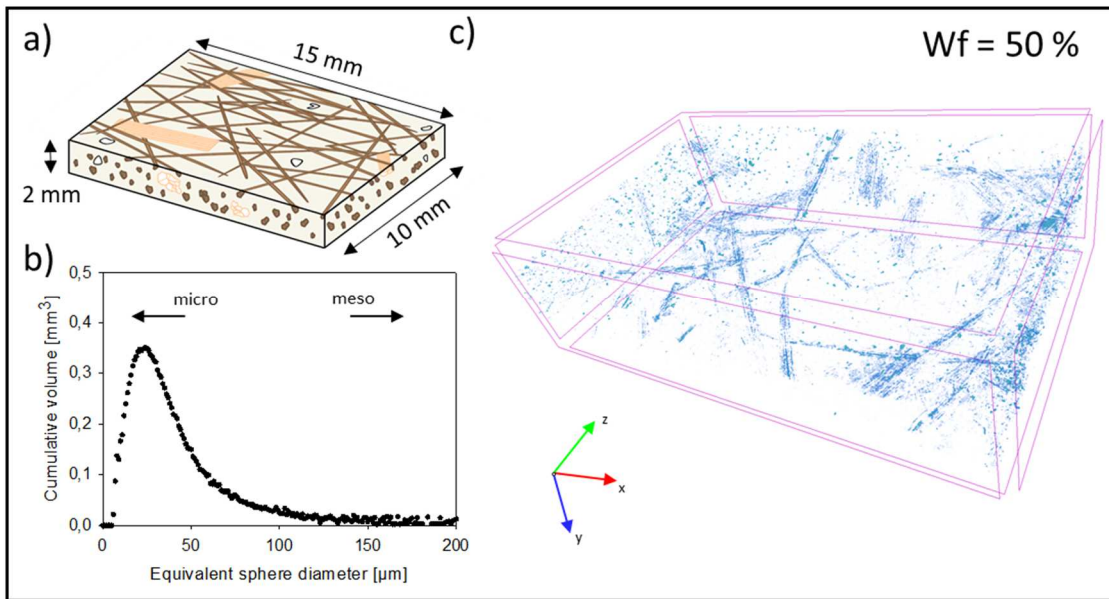
489 Figure 1: Graphical presentation of literature review of the mechanical properties of  
 490 non-woven flax composites reinforcing PLA [29–33] ; PP [13,16,34–36] ; MAPP [4,29]



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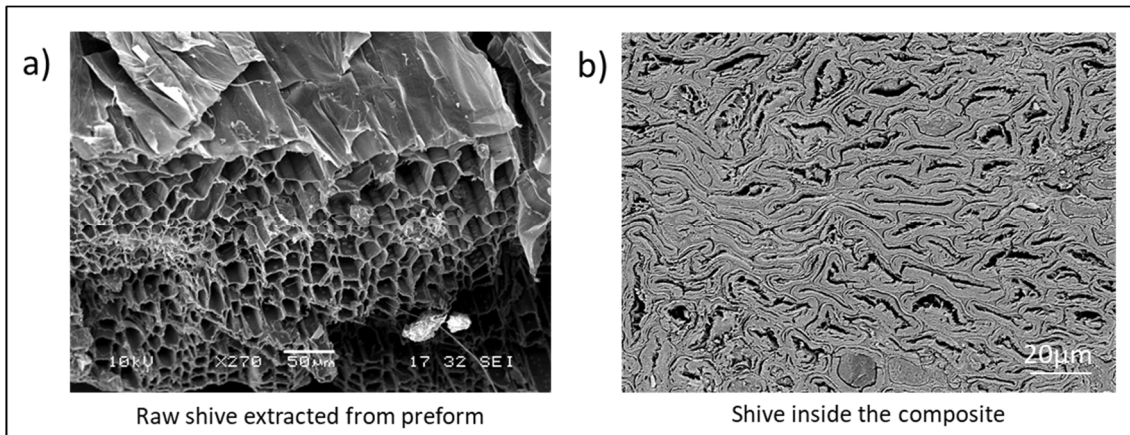
492 Figure 2: SEM observation of the flax/PLA non-woven investigated, a) a **schematic**  
 493 **representation** of the analysed sample, b) upper observation, showing preferential  
 494 fibre orientation (red arrows), c) transverse observation, **cracks** might be due to water  
 495 polishing, d) zoom on elementary flax fibres, e) zoom on flax bundles.





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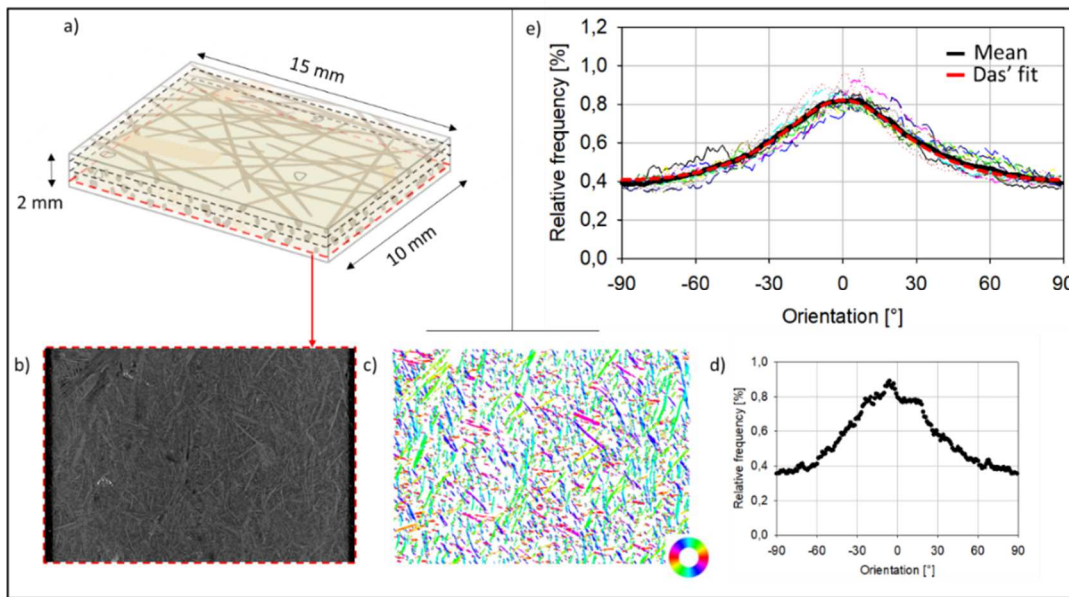
497 Figure 3: Porosity analysis by micro-CT a) schema of the analysed sample, b) analysis of  
 498 porosity size, c) the micro-CT 3D view of porosity.



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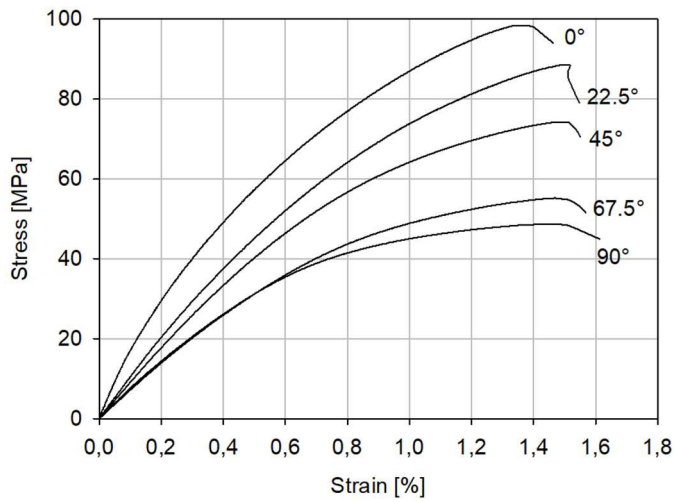
500 Figure 4: SEM observation of a shive a) before the composite manufacturing step with  
 501 open cells, b) after composite manufacturing, showing collapsed cells.





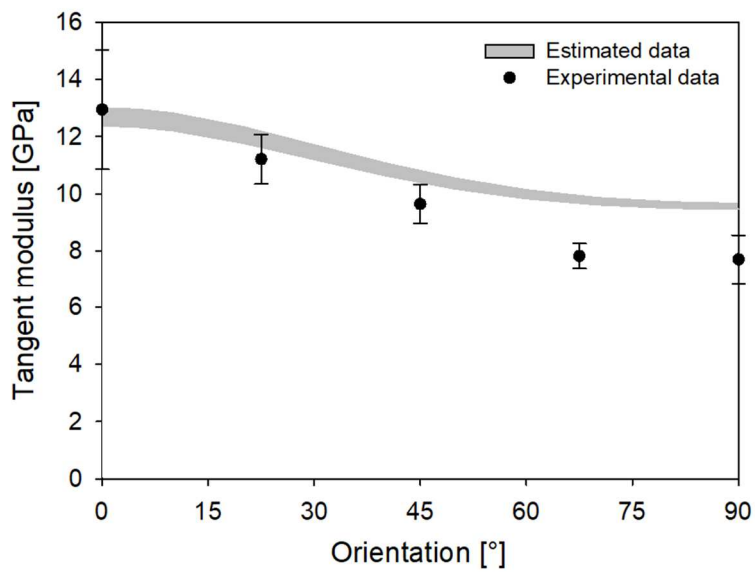
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503 Figure 5: Orientation analysis. a) sample schema with slices (marked by dotted line)  
 504 investigated, b) micro-CT image analysed for local fibre orientation, c) orientation  
 505 analysis representation of micro-CT image (each colour represents an orientation), d)  
 506 Orientation frequency histogram of one micro-CT slice, e) global orientation analysis of  
 507 the composite, based on the mean orientation (black) of twelve micro-CT slices (dotted  
 508 coloured lines). An interpolation [7], based on the mean value, is shown in red dotted  
 509 lines.



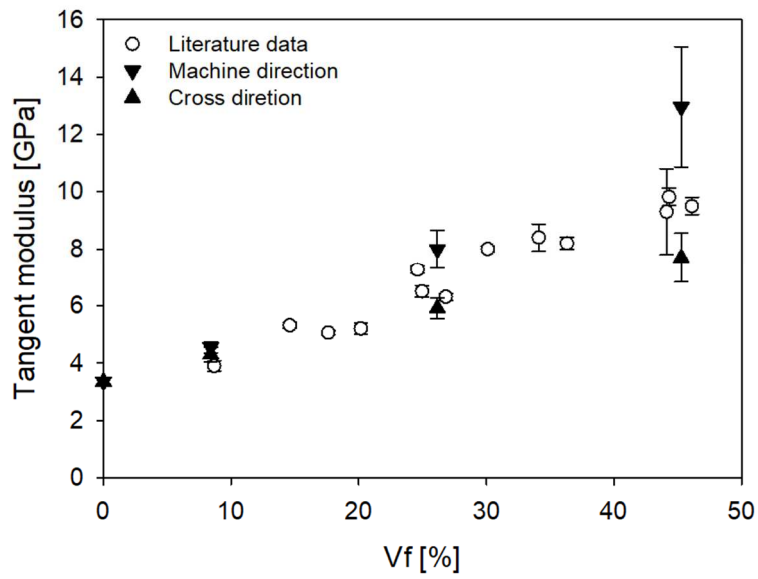
510

511 Figure 6: Tensile behaviour of non-woven flax/PLA with 50 wt% of fibres at several  
 512 loading angles. The 0° refer to the machine direction and the 90° to the cross direction.



513

514 Figure 7: Evolution of the tangent modulus with loading orientation for non-woven  
 515 flax/PLA with 50wt% of fibres tested in tension.



516

517 Figure 8: Evolution of tangent modulus with fibre volume fraction, with measurements

518 in the orthogonal machine and cross directions presenting anisotropy. A comparison

519 with literature data is also presented.

520 **Tables:**

521 Table I: Experimental values of longitudinal, transversal and off-axis tensile tests on  
 522 non-woven flax/PLA with 50 wt% of fibres.

Angle [°]	Tangent modulus [GPa]	Strength [MPa]	Strain at rupture [%]
0	12.95 ± 2.09	90.0 ± 6.1	1.44 ± 0.27
22.5	11.21 ± 0.87	84.9 ± 4.3	1.48 ± 0.27
45	9.64 ± 0.67	69.6 ± 6.5	1.30 ± 0.31
67.5	7.82 ± 0.44	53.2 ± 4.1	1.38 ± 0.25
90	7.70 ± 0.85	47.4 ± 4.4	1.25 ± 0.26

523

524 Table II: Flax/PLA UD composite mechanical properties used to obtain mechanical  
 525 properties of flax via a back-calculation method : rule of mixture for longitudinal  
 526 modulus ( $E_l$ ) and Poisson's ratio ( $\nu_{lt}$ ), Halpin Tsai for shear modulus ( $G_{lt}$ ) and transverse  
 527 modulus ( $E_t$ ).

	$V_f$ [%]	$E_l$ [GPa]	$E_t$ [GPa]	$G_{lt}$ [GPa]	$\nu_{lt}$ [-]
Experimental values for Unidirectional Composites	0	3.73	3.73	1.31	0.41
	30	20.09	4.17	1.76	-
	40	25.98	4.29	1.88	0.39
	50	27.47	4.13	2.04	0.37
Flax fibres properties by back-calculation	30	58.26	5.35	3.76	-
	40	59.34	5.24	3.42	0.35
	50	51.21	4.56	3.33	0.33
Mean back calculated flax fibres value		56.2 ± 3.6	5.05 ± 0.35	3.50 ± 0.18	0.34 ± 0.01
Literature value for flax fibres		52.5 ± 8.6 [26]	8 ± 3 [37]	2.5 ± 0.2 [38]	0.48 [39]

528

