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## **Understanding the effect of moisture variation on the hygromechanical properties of porosity-controlled nonwoven biocomposites**

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### **Abstract**

This study investigates the evolution of hygromechanical properties of flax/PP nonwoven composites in a wide range of environmental Relative Humidity (RH) conditions from 10 to 98% RH. The influence of microstructure with various porosity content ( $\Phi = 5, 30, 50\%$ ) on the mechanical and hygroscopic behaviours is studied and compared to glass/PP composites as a baseline. No significant changes in the moisture content and mechanical properties of glass-fibre reinforced composites are observed. For flax fibre nonwoven composites, the porosity greatly impacts the kinetic of sorption with moisture saturation varying from 9 hours to 15 days with decreasing voids. Hygroscopic expansion is hardly modified by the porosity content while anisotropic expansion is always observed. Tensile behaviour and properties are slightly changed over a range of 10-75%RH but negatively impacted between 75% and 98% RH. Interestingly, unlike the tangent tensile modulus and strain at rupture of flax/PP composites, the yield strength exhibits a non-monotonic trend with an optimal value over 50% RH; compressive stresses at the fibre/matrix interface induced by flax fibres hygroexpansion are proposed to explain this trend.

**Keywords:** Biocomposite; Flax; Nonwoven; Porosity; Moisture; Mechanical properties

### **1. Introduction**

Due to environmental awareness and policies, nonwoven biocomposites have attracted a growing interest in the last few decades in the automotive industry [1]. Indeed, to comply with state regulations concerning standards [2], car manufacturers need to **minimise** the fuel consumption of vehicles by all possible means. This involves the improvement of powertrain capacities, automotive aerodynamics as well as reducing the vehicle weight and the rolling resistance. To reduce the weight of vehicles and hence lower the overall fuel consumption [3], much industrial research and development has been undertaken on lighter materials and especially plant fibres. Due to their low cost, low density, high specific stiffness, sound and vibration

absorption capacity and recyclability, plant fibres represent a promising reinforcement that can be used to replace synthetic fibres, such as glass ones, in composite materials [4–7].

Among the composites used in the automotive industry, nonwovens represent a significant market. Other materials may also be used such as extruded or injection-moulded plant fibre-reinforced composites [8,9].

Commingled nonwovens are made of bast (mainly flax, jute, kenaf and hemp etc.) and polypropylene fibres processed by compression moulding and are mainly used to manufacture vehicle interior parts. These materials offer good deformability during processing, short process cycles, limited raw material cost, allow a good recyclability and can provide good sound absorption and mechanical properties [10–12]. From the same semi-finished product, it is possible to control the porosity within the material (from 5 to 60%) by modifying the compression rate during the hot moulding process and hence cover a large range of applications. The geometry and connections of the pores are modified by the compression with more opened and interconnected voids with increased porosity. High porosity (60%) is ideal to ensure good acoustic properties due to the tortuous porosity network which tends to disperse and attenuate the acoustic waves [13]. In terms of mechanical evolution, a drop of properties occurs with increasing amount of porosity, associated to composite higher strains. For a given architecture, a composite material is defined by the combination of three factors: reinforcement, matrix and fibre/matrix interfacial properties [14].

Mechanically speaking, additional parameters such as fibre volume ratio, fibre architecture and individualisation, porosity content as well as fibre orientation are of great importance. As discussed by Merotte et al. [15], reinforcement, matrix and fibre/matrix interfacial properties contribute differently to the mechanical properties accordingly to the porosity of the material. Indeed, at low porosities, the improvement of interfacial properties has little influence on the mechanical properties of the composite, while, with the increasing void content, the fibre/matrix interface has a greater impact.

In the automotive industry, materials not only need to confer acoustical and mechanical advantages but also need to be durable over the vehicle's lifetime (i.e. thermal and moisture variations). Research has highlighted temperature variations in vehicle interiors under different meteorological conditions and at different locations. In Athens (Greece), the maximum temperature in vehicle interiors measured during the period from April to August 2007 varied from 41°C to 76°C [16], while, during summer in Western Australia, the interior temperature in parked cars ranged from 15°C to 70°C depending on the time of the day, as well as the vehicle's colour and orientation [17]. The variation of temperature is generally coupled with moisture variation inside vehicles. Currently, even if nonwoven composites reinforced by vegetal fibres have a sufficiently high performance to compete with glass fibre composites at room temperature and

humidity, their sensitivity to high temperature and moisture variations limit their use, especially when mechanical functions are required. This phenomenon is a major bottleneck in the development of innovative products and should be investigated and understood. Actually, this problem is complex due to the coupling of thermal, hygroscopic and mechanical phenomena. To improve the understanding of the phenomena involved, the thermal and hygroscopic behaviours of plant fibre composites need to be decoupled.

This paper focuses on the effect of moisture content variations on the hygromechanical behaviour of nonwoven composites at constant temperature. Few studies [18–20] investigated the hygromechanical behaviour of plant fibre composites but no information is currently available for nonwoven composites with or without a porosity-controlled microstructure. Further understandings of the influence of moisture variations on the behaviour and properties of nonwoven composites are required. This study investigates the effect of a large range of relative humidity (from 10 to 98% RH) on the structure and static mechanical behaviour of flax nonwoven biocomposites with different porosity levels. The results are discussed and combined with SEM observations, in relation to the sorption and hygroexpansion behaviour of these materials.

## 2. Materials

- 1.1 Long scutched technical fibres of flax (*Linum usitatissimum L.*) of the Eden variety were used in this study. The plants were cultivated in the year 2017 in Normandy (France) under normal meteorological conditions [5], dew-retted in fields and mechanically scutched. E-glass fibres were also used to compare with raw flax fibres and composites. Flax fibres were used as reinforcement in the nonwoven material and combined with raw polypropylene (PP). Commingled nonwovens were manufactured according to the carding/over-lapping/needle punching technology [21] by Eco-technilin SAS (Normandy, France). Products have a mass per unit area of  $300 \pm 30$  g/m<sup>2</sup> and a fibre/matrix ratio of 50-50% wt. in compliance with industrial requirements. Due to the difference between their nature (density, stiffness, fineness), fibres react differently to the successive steps of the nonwoven manufacturing process. This results in differences between the nonwoven characteristics reported in the description of composite microstructure and properties.

## 3. Experimental

### 3.1 Composite manufacturing

Commingled nonwovens were processed by hot compression moulding on a LabTech Engineering 50T laboratory hydraulic press to obtain 20 x 20 cm<sup>2</sup> plates. To be consistent with the industrial process,

preforms were not oven-dried before transformation and were stored at 23°C and RH=50%. The plies were then hot pressed at 200°C and 20 bar for 4 minutes before being placed between cold plates at 50 bar for another 3 minutes. The press was briefly opened (less than 10 seconds) between the melting and cooling steps allowing a slight degassing of the material which is representative to the industrial process. To obtain different porosities ( $\Phi$ ), the quantity of nonwoven plies in the fixed cavity mould (2mm) was adjusted (from 2 to 8 plies for  $\Phi = 50\%$  and  $\Phi = 5\%$ , respectively).

### 3.2 Porosity measurement

Three porosity values were targeted: 5, 30 and 50%. The void content was controlled according to the ASTM D2734-09 density method. The weight and dimensions of the composite plates were measured and the percentage porosity was calculated using the following equation:

$$\Phi = 1 - \rho_c((1 - w_f)/\rho_m + w_f/\rho_f) \quad (1)$$

Where  $\Phi$  is the porosity,  $\rho_c$ ,  $\rho_m$  and  $\rho_f$  are the densities of the composite, matrix and fibres, respectively, and  $w_f$  is the fibre weight ratio. In a prior step, flax and PP fibres densities were determined from the weight difference of samples in air and ethanol according to ISO 1183-1 standard. Weight and dimensions were measured using a Mettler Toledo XS205 high precision scale with  $10^{-4}$ g accuracy and a Mitutoyo IP65 0-25 mm digimatic micrometre with an accuracy of  $10^{-3}$  mm, respectively.

### 3.3 Conditioning of composites

The composites were dried in a Memmert convection dryer ULP500 at 105°C until constant weight to remove as many molecules of free water and bound water as possible from the material [22]. Samples were then placed in controlled humidity chambers at 23°C with different relative humidity levels (10, 33, 50, 75 and 98% RH). Different binary saturated aqueous solutions were used to maintain constant relative humidity in each chamber [23].

### 3.4 Water sorption and swelling measurements

Flat test specimens (20 mm x 20 mm) were cut from the composite plates with a Computed Numerical Control (CNC) machine ICP 4030 iMC6P supplied by Isel. The thickness of the samples was set at 2 mm. All specimens were marked with three points for each dimension measurement: thickness, width and length. Directly after drying, nonwoven composites specimens were weighed, dimensions were measured and then placed in controlled humidity chambers (10, 33, 50, 75 and 98% RH). Weight and dimension variations were checked until constant values were obtained. Gravimetric measurements were performed to monitor the

water uptake of flax/PP biocomposites over time for one humidity. For this purpose, five samples of each porosity were placed in a climatic chamber (Mettler) at 90% RH and 23°C. Measurements were first carried out every hour, and the interval was then increased progressively up to 1 day depending on the rate of water uptake of the samples.

The percentage weight gain  $M_f$  is calculated as follows:

$$M_f = \frac{W_f - W_0}{W_0} \times 100 \quad (2)$$

Where  $W_f$  and  $W_0$  are the weight of the sample after **stabilisation** and the weight of the dry material, respectively. A Fisherbrand™ scientific high precision scale with 10<sup>-4</sup>g accuracy was used for weight measurements.

Hygroexpansion was calculated in the same way as weight gain, by determining the ratio between the dimensions in the wet and dry state. The longitudinal, transverse and out-of-plane hygroexpansion coefficients were determined by applying a standard compression force during measurement, using a Mitutoyo IP65 0-25 mm digimatic micrometre with an accuracy of 10<sup>-3</sup> mm. **For the sorption/desorption tests, samples were placed at 50% RH/23°C after being dried as described in the conditioning of composite section, then placed at 98% RH/ 23°C and finally at 50% RH/ 23°C. Samples were stored up to three weeks until the stabilisation of moisture content was reached in all the conditioning steps. Between each step, the samples were withdrawn to be tested, using the same materials and methods as described above.**

### 3.5 **Characterisation** of mechanical properties

Monotonic tensile tests were conducted on the nonwoven composites using an electromechanical INSTRON 5566A testing machine equipped with a 10 KN load cell and an INSTRON extensometer with a nominal length of 25 mm. Tensile tests were performed on dumbbell geometry specimens in a controlled atmosphere (23°C and RH = 50%), according to the ISO 527 standard. At least seven samples were tested at a cross-head speed of 1 mm.min<sup>-1</sup> and the results were arithmetically averaged. The tensile modulus was calculated on the linear part of the stress-strain curve, at the very outset of loading between 0.05% and 0.1% of strain according to previous studies focused on properties of nonwoven composites [15].

### 3.6 Microscopy

SEM observations were performed on cross-sections of nonwoven composites. Samples were then sputter-coated with a thin layer of gold in an Edwards Sputter Coater and analysed with a Jeol JSM 6460LV electron scanning microscope at 20 kV.

## 4. Results and discussion

### 4.1 Description of composite microstructure and properties

Table 1 presents the main characteristics of the manufactured composite plates. For automotive applications, nonwovens usually have the same fibre weight fraction whatever the type of reinforcement fibre used, providing materials that are economically and mechanically viable. Due to the difference in fibre density, fibre volume ratios are lower for glass/PP nonwoven composites than flax/PP. For flax/PP nonwoven composites, fibre volume ratios vary between 34% to 17.4%, with lower values of 23.6% to 10.4% for glass fibre/PP composites according to the porosity.

It should be noted that nonwovens manufactured by the carding/over-lapping/needle punching technology are not fully isotropic materials since the fibres exhibit a preferred orientation due to the process [21]. Indeed, the anisotropy ratio is equal to 1 in isotropic materials, while for the materials studied here, a ratio of about 1.2 is obtained for flax/PP. Glass/PP composites evidence higher anisotropic ratio (1.9) due to difference of production settings.

### Figure 1

The levels of porosity were set before manufacturing of the composites at 5, 30 and 50% by adjusting the quantity of nonwoven plies for a given thickness. Figure 1 shows SEM images of cross-sections of the materials. It is noteworthy that, at a low porosity level ( $\Phi = 5\%$ ), voids are mostly located in the matrix and at the fibre/matrix interface. Interface porosity is created during processing of the composite due to poor fibre wetting with polypropylene and differential swelling/shrinkage of the composite components, while matrix porosity might be caused by the trapping of air [24]. At  $\Phi = 30\%$ , voids are larger and located around fibre bundles, even though they do not seem to be interconnected. At  $\Phi = 50\%$ , most of the fibres are still embedded by the matrix, but the pores are larger and start to become interconnected, creating a tortuous porosity network [13]. In brief, by increasing the material compression rate, the matrix volume ratio is also increased, which helps the matrix to impregnate the fibrous network. Broadly the same observations can be made regarding the microstructure of glass/PP nonwoven composites. However, since glass fibres are **unitary fibres** and are sized, the interface porosity in glass/PP composites is less well developed and voids are mostly observed within the matrix.

### Table 1

### 4.2 Sorption measurement

Figure 2 shows the water sorption isotherm curves obtained from moisture sorption measurements on different materials at saturation. All the curves of both plant fibres alone and biocomposites exhibit a sigmoidal shape, typical of cellulose based materials [25]. Three sorption domains can be identified [26,27]: below 0.15 water activity (the relative vapour pressure of water,  $a_w$ ), monolayer adsorption takes place according to Langmuir's mode in which water is absorbed onto the cell wall and linked with OH groups. The second domain, with water activity ranging from 0.15 to 0.7, is defined by the formation of polylayer water in microcapillaries, corresponding to Henry's mode. Finally, above 0.7  $a_w$ , water clustering starts to occur and capillary condensation becomes dominant. Water concentration leads to a relaxation of existing voids and results in swelling of the material, thus opening new sites available for water sorption [28,29].

### Figure 2

From Fig. 2.a, it can be seen that the moisture uptake of raw flax fibres increases progressively with water activity up to a maximum value of  $23.2 \pm 2.2\%$  at  $0.98 a_w$ , which is in concordance with results found in the literature [30]. As expected, glass fibres are far less sensitive to moisture and only show a slight increase of uptake ( $0.72 \pm 0.08\%$ ) at high water activity due to the sizing components on glass fibres [31]. Interestingly, the moisture uptake is not exactly related to the fibre weight ratio of the composite but slightly lower, indicating that the constraining effect of the matrix, due to its apolar nature, might limit fibres sorption [32]. Also, one can notice from Fig. 2.b, that despite a large change of microstructure with different porosity content, sorption behaviour is only slightly modified. Indeed, at 0.98 water activity, for low porosity level ( $\Phi = 5\%$ ) the saturated moisture uptake is around  $8.0 \pm 0.1\%$  and increase with the porosity reaching values of  $9.4 \pm 0.2$  and  $9.5 \pm 0.1\%$  for composites with 30 and 50% porosity contents, respectively. Comparatively, glass/PP composites absorb a very limited amount of moisture whatever the water activity and the porosity content.

Beyond steady-state regime with sorption isotherm, sorption under unsteady-state condition, is represented on Fig.3 with the evolution of moisture uptake versus the square root of time at 90% RH and 23°C. Here, porosity has a significant contribution, since equilibrium is definitely more rapidly reached for materials with higher porosity content.

Flax/PP nonwoven composites with  $\Phi = 5\%$  only reach moisture saturation after 15 days while it only takes 18 and 9 hours to reach equilibrium for  $\Phi = 30$  and 50%, respectively.

### Figure 3

## 4.3 Hygroexpansion

A first consequence of moisture uptake is the dimensional variation of biocomposite. Figures 4 a and b present hygroscopic variations in the steady-state regime as a function of moisture content. Figure 4.a shows the hygroexpansion in three dimensions as a function of moisture content, but only for  $\Phi = 5\%$  flax/PP nonwoven composites because other types of microstructure (flax/PP) exhibit the same behaviour. Again, microstructural difference of nonwoven biocomposite does not lead to a significant change of hygro-expansion during the steady-state regime. This suggests that the excess moisture for high porosity content is located in voids as free water and, consequently, does not lead to swelling of the material.

Interestingly, out-of-plane hygroexpansion is much higher than longitudinal or transverse hygroexpansion. Indeed, the anisotropic swelling of vegetal fibres combined with quasi-isotropic orientation of fibres in the material reduces the in-plane swelling. For the same reason, Almgren et al. [33] pointed out that, for wood polymer composites (WPC) with randomly dispersed short fibres, no in-plane hygroexpansion is observed. On the contrary, for flax/MAPP unidirectional composites ( $V_f = 60\%$ ), transverse swelling occurs when the material is submitted to immersion tests leading to hygroexpansion values up to 3.3% at water uptake equilibrium [34]. This demonstrates that fibre orientation distribution has an impact on dimensional stability when the composite is submitted to moisture variations. Recent work of Requile et al. [18] have proved that orthotropic hygroexpansion in a unidirectional hemp/epoxy biocomposite with high width/thickness ratio is due to difference of fibre volume fraction in both y and z directions. Indeed, changing geometry from beam to cube reduces the orthotropic swelling.

#### Figure 4

A sigmoidal regression proposed by Péron et al [35] and Le Duigou et al [36] enable to fit properly the experimental data. This means that hygroscopic coefficient  $\beta_{z,t}$  depends on the moisture content, probably due to the evolution of the free volume fraction, the effects of the fibre/fibre and fibre/matrix interfaces and the hygroscopic stress state. Glass/PP composites do not exhibit significant hygroexpansion.

#### 4.4 Hygromechanical behaviour and properties

Figure 5 shows typical tensile stress-strain curves (solid lines) obtained during tensile tests (23°C, 50% RH) and their corresponding tangent modulus-strain curves (dashed lines) for  $\Phi = 5$  and 50% flax/PP (Fig. 5.a) and glass/PP (Fig. 5.b) composites.

#### Figure 5

Flax composites exhibit a non-linear relationship between stress and strain which is illustrated by a two-stage tangent modulus-strain curve. The first part of the curve (A on Fig. 5.a) is assumed to represent rupture caused by breakage of fibres, fibre bundles and matrix as well as fibre/matrix debonding, while the second part (B) corresponds to post-debonding fibre/matrix friction and fibre network slippage. The overall behaviour is slightly modified by the change of microstructure with a loss of dissipated energy (area under the curve) as well as earlier transition between rupture and fibre slippage (shifting of A to lower strain values) with increased porosity [13].

It is often considered that dimensioning structures based with these materials are complex due to their tensile behaviour. However, glass/PP nonwoven composites evidence a nearly monotonic decrease of tangent modulus (Fig. 5.b). Differences could be due not only to the nature of the flax fibres, which show a non-linear behaviour [37], but also the poor **individualisation** of flax fibres potentially leading to breakage and slippage of the fibre/fibre interface [38]. It can be noted from Fig. 5.a and b that properties are equivalent for flax/PP and glass/PP at  $\Phi = 5\%$ , but not at  $\Phi = 50\%$ , suggesting that the effect of microstructure is more drastic for flax composites due to the nature of the fibre.

Figure 6 illustrates the influence of relative humidity on the tensile behaviour of nonwoven composites. As shown on Fig. 6.d, the variation of relative humidity does not affect the behaviour of glass/PP composites whatever the relative humidity due to a very low moisture uptake.

Conversely, the behaviour of flax/PP composites evidence some visible change at high relative humidity ( $>75\%RH$ ) and is **emphasised** with the modification of microstructure (higher porosity amount) (Fig. 6.a, b and c). Indeed, the mechanical behaviour of flax/PP composites with  $\Phi = 5\%$ ,  $30\%$  and  $50\%$ , remains similar up to  $75\% RH$ , with only the end of the curves being slightly modified due to the greater degree of elongation reflecting a less breakable behaviour. Whatever the porosity, the global behaviour only begins to be modified at  $98\% RH$ . Indeed, the transition between the two parts of the curve appears at lower strain and then **stabilises** before the onset of progressive rupture. The rapid fall of tangent modulus represented by the first part of the slope could be due to variations of flax fibre properties as well as the rupture of fibre/fibre (bundle) interfacial debonding. A plasticising effect can be observed in vegetal fibres submitted to high relative humidity, caused by water molecules entering cellulose network via the hemicellulose-lignin matrix which is hydrophilic. This allows cellulose molecules to move more freely, inducing a loss of stiffness of the cellulose structure, which also affects the plant fibre stiffness [39].

**Figure 6**

The plateau of the curve reflects a quasi-linear relation between tangent modulus and strain, highlighting a stress transfer mechanism that occurs essentially by friction. Indeed, as the matrix in flax/PP composites is apolar, no chemical bonding occurs and the adhesive pressure is induced solely by thermal and hygroscopic stresses [40]. For  $\Phi = 5\%$ , the plateau reaches a value of 2000 MPa. It should be kept in mind that tensile modulus of polypropylene (at 23°C and 50% RH) is  $1464 \pm 121$  MPa, which suggests that flax fibres still contribute to the composite's performances at this stage. As can be seen on Fig. 6.a, b and c), the plateau is lowered from 2000 MPa ( $\Phi = 5\%$ ) to 800 MPa ( $\Phi = 50\%$ ). This fall is mainly attributed to the increase in porosity and, consequently, the decrease of fibre volume ratio in the material (Table 1).

Figure 7. a, b and c presents the variation of tensile properties with moisture content in nonwoven composites. Glass fibre composites do not show any change of mechanical properties, which is in line with the results described in the previous sections for sorption measurements. Flax/PP composites, on the other hand, exhibit large variations of tangent modulus and yield strain which could be detrimental for their use but only for high relative humidity above 75% RH. For the three porosity levels tested in this study, trends are similar for all microstructures.

Since the tangent modulus of a composite material is highly dependent on its component properties, and polypropylene is very insensitive to moisture, the variation observed here is due to the evolution of flax properties due to their softening. Similar variations have been described in the literature with regard to raw flax fibres [41] as well as unidirectional composites reinforced with flax fibres [42]. Consequently, yield strain increases with decreasing tangent modulus due to the **plasticisation** of flax fibres and network slippage.

Interestingly, yield stress is not negatively impacted by the variation of moisture content in the material. Indeed, the yield stress initially increases with rising moisture content (MC), reaching a maximum at around 3% MC, and tends to **stabilise** beyond that point. Some previous studies on fibres such as flax and hemp show that individual fibres have a maximum tensile strength between 50 and 70% RH, but then decreases at higher RH [43–45]. However, in this study, no drop in yield stress is observed even if severely high RH is reached. **This could be explained by compressive stresses at the fibre/matrix interface**. Indeed, when associated with a polymeric matrix, the swelling of fibres is reduced since the matrix plays a constraining role which creates radial compressive stresses [32]. Le Duigou et al [40] showed that increasing humidity increases radial stress of flax/MAPP composites. This was also proposed by Almgren et al. [46] who claimed that theoretically, increase of moisture content in biocomposites could lead to higher IFSS.

As shown in Fig. 7.c, porosity only affects the initial mechanical properties of the nonwoven composites and does not modify their evolution with variations in moisture content.

### Figure 7

#### 4.5 Behaviour during a single environmental cycle

A sorption/desorption cycle was carried out to evaluate the reversibility of moisture induced degradation. Table 2 reports the variations in dimensions and mechanical properties during the moisture cycle. Careful attention should be paid as results were obtained when moisture content equilibrium was reached which might not be the case for real life applications as environmental conditions are not constant overtime.

**Table 2**

As regards moisture content and hygroexpansion, reversibility is not fully achieved after one cycle. Similar behaviour has been reported in the literature regarding the out-of-plane hygroexpansion of wood polymer composites humidified at 97% RH and redried at 40°C for 72 hours [47]. These latter authors claim that the swelling of the composite is non-reversible, which suggests that the weak fibre-matrix interphase is debonded by moisture induced swelling. Moreover, the composition and structure of the fibres might be modified at high moisture contents along with variations in parietal constituents and microfibril angle [44]. Flax fibres are gelatinous fibres that are rich in matrix polymers such as pectins and hemicelluloses [48]. These latter constituents are known to be hydrolabile [49] and might not return to their original form after a cycle.

Since the difference between initial and final phases is not necessarily due to a degradation of the material, another hypothesis is proposed. Indeed, a hysteresis loop between the adsorption and desorption isotherm curves is commonly observed in studies of the sorption properties of vegetal fibres [30], and this could also explain the only partial recovery of dimensional variations. Derome et al [50] showed that the swelling/shrinkage hysteresis of wood tissue is strongly linked to the moisture sorption hysteresis. Guyer et al. [51] suggested that the difference of forces induced by swelling and shrinkage is reflected by the difference of chemical potential required for adsorption and desorption, which is actually the difference in relative humidity required to obtain the same moisture content in a sorption loop. Hence, after being stored at 98% and 50% RH until attainment of equilibrium, nonwoven composites are in a desorption state. This could explain the difference between the initial and final values of moisture content and out-of-plane swelling.

Tensile properties also show differences between the two 50% RH conditioning steps. Tangent modulus shows a decrease, while yield strain increases. Yield stress shows a slight decrease, which could suggest a degradation of the material, with stress transfer becoming less efficient after a cycle. Similar changes are observed with the variation in porosity.

These variations are not surprising because the moisture contents for the second storage step are higher than in the initial steps, which means that the materials being compared are not in the same state. As discussed in paragraph 4.4, any increase of moisture content necessarily affects the mechanical properties of flax/PP nonwoven composites. With  $\Phi = 5\%$ , it is possible to compare materials with equivalent moisture contents. Indeed, the material in a desorption state at 50% (RH 50%(d\*)) RH has a moisture content of 3.55%, which is similar to the moisture content in a sorption state at 75% RH represented on Fig.7 (MC=3.65%). However, the tensile properties are degraded, with a tangent modulus of  $5719 \pm 423$  MPa for 50%<sub>d</sub> compared to  $6309 \pm 700$  MPa for 75%<sub>s</sub>. This difference of tangent modulus, added to the drop of yield stress described above, could imply degradation of the material and thus, **plasticisation**.

## 5. Conclusions

Nonwoven composites are mostly used in the automotive industry to manufacture vehicle interior parts. These items are submitted to a complex environment that combine temperature, humidity and mechanical stresses during a vehicle's life cycle.

The present study focuses the hygromechanical behaviour of microstructured nonwoven PP/flax biocomposites having various porosity content (5, 30 and 50%). Materials were stored at five different relative humidity levels (RH = 10, 33, 50, 75 and 98%) and at a constant temperature of 23°C until equilibrium moisture content was reached. Glass fibre/PP nonwoven composites were evaluated as reference as they are non sensitive to moisture; whatever the considered microstructure.

Results are quite different when using flax as reinforcement compared to glass. Water uptake kinetic is found to be dependent on porosity. The time required to reach a **stabilised** moisture content varies from 9 hours to 15 days, for high and low porosities, respectively. As consequence to moisture sorption, hygroexpansion appears due to the quasi-isotropic distribution of fibres in the material but only out-of-plane swelling is observed. This type of hygroexpansion is found to be similar for the three porosity levels, suggesting that the proportion of bound and free water depends on the composite microstructure.

Tensile behaviour is also affected by the increase in moisture content, with a more progressive breakage suggesting a structural **reorganisation** with increasing fibre network slippage. A dramatic change of stiffness

and strain at break appears at high RH (98%); it is attributed to variations in the mechanical properties of the flax fibres themselves as well as fibre network slippage. It underlines that nonwoven flax biocomposite could be use in an encouraging range of RH. Interestingly, yield stress is found to be almost constant over the whole range of relative humidity tested. Although porosity has a direct impact on the tensile properties and behaviour of the composites, it has no influence on their evolution with the increase in moisture content. Finally, composites were also submitted to environmental cycling tests (50/98/50% RH) showing that the hygromechanical properties are only partially reversible and do not return to their original state due to the desorption state of the materials.

Further work will be dedicated to evaluate the effect of temperature on these microstructured materials to evaluate their design envelop.

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### **Declaration of interests**

None

### **References**

- [1] Y. Chen, O. Chiparus, L. Sun, I. Negulescu, D. V. Parikh, T.A. Calamari, Natural Fibers for Automotive Nonwoven Composites, *J. Ind. Text.* 35 (2005) 47–62. doi:10.1177/1528083705053392.
- [2] European Parliament, Council of the European Union, Regulation (EC) no. 443/2009, *Off. J. Eur. Union.* 140 (2009) 1–15. doi:10.1524/zkri.2009.1105.
- [3] Lotus Engineering. Inc, An Assessment of Mass Reduction Opportunities for a 2017 – 2020 Model Year Vehicle Program, 2010.
- [4] A. Bourmaud, J. Beaugrand, D.U. Shah, V. Placet, C. Baley, Towards the design of high-performance plant fibre composites: How can we best define the diversity and specificities of plant cell walls?, *Prog. Mater. Sci.* 97 (2018) 347–408. doi:10.1016/j.pmatsci.2018.05.005.
- [5] A. Lefeuvre, A. Bourmaud, C. Morvan, C. Baley, Tensile properties of elementary fibres of flax and glass: Analysis of reproducibility and scattering, *Mater. Lett.* 130 (2014) 289–291. doi:10.1016/j.matlet.2014.05.115.

- [6] U. Berardi, G. Iannace, Acoustic characterization of natural fibers for sound absorption applications, *Build. Environ.* 94 (2015) 840–852. doi:10.1016/j.buildenv.2015.05.029.
- [7] A. Bourmaud, C. Baley, Rigidity analysis of polypropylene/vegetal fibre composites after recycling, *Polym. Degrad. Stab.* 94 (2009) 297–305. doi:10.1016/j.polymdegradstab.2008.12.010.
- [8] O. Faruk, Cars from Jute and Other Bio-Fibers, *Macromol. Mater. Eng.* 291 (2006) 449–457. doi:DOI:10.1002/mame.200600113.
- [9] G. Koronis, A. Silva, M. Fontul, Green composites: A review of adequate materials for automotive applications, *Compos. Part B Eng.* 44 (2013) 120–127. doi:10.1016/j.compositesb.2012.07.004.
- [10] A. Hao, H. Zhao, J.Y. Chen, Kenaf/polypropylene nonwoven composites: The influence of manufacturing conditions on mechanical, thermal, and acoustical performance, *Compos. Part B Eng.* 54 (2013) 44–51. doi:10.1016/j.compositesb.2013.04.065.
- [11] H.L. Bos, J. Müssig, M.J.A. van den Oever, Mechanical properties of short-flax-fibre reinforced compounds, *Compos. Part A Appl. Sci. Manuf.* 37 (2006) 1591–1604. doi:10.1016/j.compositesa.2005.10.011.
- [12] N. Renouard, J. Merotte, A. Kervoelen, K. Behlouli, C. Baley, A. Bourmaud, Exploring two innovative recycling ways for poly-(propylene)-flax non wovens wastes, *Polym. Degrad. Stab.* (2017). doi:10.1016/j.polymdegradstab.2017.05.031.
- [13] J. Merotte, A. Le Duigou, A. Bourmaud, K. Behlouli, C. Baley, Mechanical and acoustic behaviour of porosity controlled randomly dispersed flax/PP biocomposite, *Polym. Test.* 51 (2016) 174–180. doi:10.1016/j.polymertesting.2016.03.002.
- [14] R.M. Jones, Mechanics of composite materials, *Mech. Compos. Mater.* (1999) 519. doi:10.1007/BF00611782.
- [15] J. Merotte, A. Le Duigou, A. Kervoelen, A. Bourmaud, K. Behlouli, O. Sire, C. Baley, Flax and hemp nonwoven composites: The contribution of interfacial bonding to improving tensile properties, *Polym. Test.* 66 (2018) 303–311. doi:10.1016/j.polymertesting.2018.01.019.
- [16] A. Grundstein, V. Meentemeyer, J. Dowd, Maximum vehicle cabin temperatures under different meteorological conditions, *Int. J. Biometeorol.* 53 (2009) 255–261. doi:10.1007/s00484-009-0211-x.
- [17] I.R. Dadour, I. Almanjahie, N.D. Fowkes, G. Keady, K. Vijayan, Temperature variations in a parked

- vehicle, *Forensic Sci. Int.* 207 (2011) 205–211. doi:10.1016/j.forsciint.2010.10.009.
- [18] S. Réquillé, A. Le Duigou, A. Bourmaud, C. Baley, Deeper insights into the moisture-induced hygroscopic and mechanical properties of hemp reinforced biocomposites, *Compos. Part A.* (2019). doi:10.1016/j.compositesa.2019.05.006.
- [19] A. Stamboulis, C.A. Baillie, S.K. Garkhail, H.G.H. Van Melick, T. Peijs, Environmental durability of flax fibres and their composites based on polypropylene matrix, *Appl. Compos. Mater.* 7 (2000) 273–294. doi:10.1023/A:1026581922221.
- [20] A. Moudood, A. Rahman, H. Mohammad, W. Hall, A. Ochsner, G. Francucci, Environmental effects on the durability and the mechanical performance of flax fiber / bio-epoxy composites, 171 (2019) 284–293. doi:10.1016/j.compositesb.2019.05.032.
- [21] S.J. (Stephen J.. Russell, *Handbook of nonwovens*, CRC Press, 2007.
- [22] C. Baley, A. Le Duigou, A. Bourmaud, P. Davies, Influence of drying on the mechanical behaviour of flax fibres and their unidirectional composites, *Compos. Part A Appl. Sci. Manuf.* 43 (2012) 1226–1233. doi:10.1016/j.compositesa.2012.03.005.
- [23] L. Greenspan, Humidity Fixed Points of Binary Saturated Aqueous Solutions, *J. Res. Natl. Bur. Stand. -A. Phys. Chem.* 81 (1977). [https://nvlpubs.nist.gov/nistpubs/jres/81A/jresv81An1p89\\_A1b.pdf](https://nvlpubs.nist.gov/nistpubs/jres/81A/jresv81An1p89_A1b.pdf).
- [24] B. Madsen, A. Thygesen, H. Lilholt, Plant fibre composites - porosity and volumetric interaction, *Compos. Sci. Technol.* 67 (2007) 1584–1600. doi:10.1016/j.compscitech.2006.07.009.
- [25] S. Brunauer, L.S. Deming, W.E. Deming, E. Teller, On a Theory of the van der Waals Adsorption of Gases, *J. Am. Chem. Soc.* 62 (1940) 1723–1732. doi:10.1021/ja01864a025.
- [26] S. Alix, E. Philippe, A. Bessadok, L. Lebrun, C. Morvan, S. Marais, Effect of chemical treatments on water sorption and mechanical properties of flax fibres, *Bioresour. Technol.* 100 (2009) 4742–4749. doi:10.1016/j.biortech.2009.04.067.
- [27] G.S. Park, Transport Principles---Solution, Diffusion and Permeation in Polymer Membranes, in: P.M. Bungay, H.K. Lonsdale, M.N. de Pinho (Eds.), *Synth. Membr. Sci. Eng. Appl.*, Springer Netherlands, Dordrecht, 1986: pp. 57–107. doi:10.1007/978-94-009-4712-2\_3.
- [28] A. Bessadok, S. Marais, F. Gouanvé, L. Colasse, I. Zimmerlin, S. Roudesli, M. Métayer, Effect of chemical treatments of Alfa (*Stipa tenacissima*) fibres on water-sorption properties, *Compos. Sci.*

Technol. 67 (2007) 685–697. doi:10.1016/j.compscitech.2006.04.013.

- [29] I.D. Hartley, F.A. Kamke, H. Peemoeller, Cluster theory for water sorption in wood, *Wood Sci. Technol.* 26 (1992) 83–99. doi:10.1007/BF00194465.
- [30] C.A.S. Hill, A. Norton, G. Newman, The water vapor sorption behavior of natural fibers, *J. Appl. Polym. Sci.* 112 (2009) 1524–1537. doi:10.1002/app.29725.
- [31] L. Peters, Influence of Glass Fibre Sizing and Storage Conditions on Composite Properties, in: P. Davies, Y.D.S. Rajapakse (Eds.), *Durab. Compos. a Mar. Environ. 2*, Springer International Publishing, Cham, 2018: pp. 19–31. doi:10.1007/978-3-319-65145-3\_2.
- [32] T. Joffe, E.L.G. Wernersson, A. Miettinen, C.L. Luengo Hendriks, E.K. Gamstedt, Swelling of cellulose fibres in composite materials: Constraint effects of the surrounding matrix, *Compos. Sci. Technol.* 74 (2013) 52–59. doi:10.1016/J.COMPSCITECH.2012.10.006.
- [33] K.M. Almgren, E.K. Gamstedt, F. Berthold, M. Lindström, Moisture uptake and hygroexpansion of wood fiber composite materials with polylactide and polypropylene matrix materials, *Polym. Compos.* 30 (2009) 1809–1816. doi:10.1002/pc.20753.
- [34] A. Le Duigou, S. Requile, J. Beaugrand, F. Scarpa, M. Castro, Natural fibres actuators for smart bio-inspired hygromorph biocomposites, *Smart Mater. Struct.* 26 (2017) 125009. doi:10.1088/1361-665X/aa9410.
- [35] M. Péron, A. Céline, M. Castro, F. Jacquemin, A. Le Duigou, Study of hygroscopic stresses in asymmetric biocomposite laminates, *Compos. Sci. Technol.* 169 (2019) 7–15. doi:10.1016/j.compscitech.2018.10.027.
- [36] A. Le Duigou, V. Keryvin, J. Beaugrand, M. Pernes, F. Scarpa, M. Castro, Humidity responsive actuation of bioinspired hygromorph biocomposites (HBC) for adaptive structures, *Compos. Part A Appl. Sci. Manuf.* 116 (2019) 36–45. doi:10.1016/j.compositesa.2018.10.018.
- [37] D.U. Shah, Damage in biocomposites: Stiffness evolution of aligned plant fibre composites during monotonic and cyclic fatigue loading, *Compos. Part A Appl. Sci. Manuf.* 83 (2016) 160–168. doi:10.1016/j.compositesa.2015.09.008.
- [38] G. Coroller, A. Lefeuvre, A. Le Duigou, A. Bourmaud, G. Ausias, T. Gaudry, C. Baley, Effect of flax fibres individualisation on tensile failure of flax/epoxy unidirectional composite, *Compos. Part A Appl. Sci. Manuf.* 51 (2013) 62–70. doi:10.1016/j.compositesa.2013.03.018.

- [39] A. Stamboulis, C.A. Baillie, T. Peijs, Effects of environmental conditions on mechanical and physical properties of flax fibers, *Compos. Part A Appl. Sci. Manuf.* 32 (2001) 1105–1115. doi:10.1016/S1359-835X(01)00032-X.
- [40] A. Le Duigou, J. Merotte, A. Bourmaud, P. Davies, K. Belhouli, C. Baley, Hygroscopic expansion: A key point to describe natural fibre/polymer matrix interface bond strength, *Compos. Sci. Technol.* (2017). doi:10.1016/j.compscitech.2017.08.028.
- [41] G.C. Davies, D.M. Bruce, Effect of Environmental Relative Humidity and Damage on the Tensile Properties of Flax and Nettle Fibers, *Text. Res. J.* 68 (1998) 623–629. doi:10.1177/004051759806800901.
- [42] A. Moudood, W. Hall, A. Öchsner, H. Li, A. Rahman, G. Francucci, Effect of Moisture in Flax Fibres on the Quality of their Composites, *J. Nat. Fibers.* 00 (2017) 1–16. doi:10.1080/15440478.2017.1414651.
- [43] B. Van Voorn, H.H.G. Smit, R.J. Sinke, B. De Klerk, Natural fibre reinforced sheet moulding compound, *Compos. - Part A Appl. Sci. Manuf.* 32 (2001) 1271–1279. doi:10.1016/S1359-835X(01)00085-9.
- [44] V. Placet, O. Cisse, M.L. Boubakar, Influence of environmental relative humidity on the tensile and rotational behaviour of hemp fibres, *J. Mater. Sci.* 47 (2011) 3435–3446. doi:10.1007/s10853-011-6191-3.
- [45] A. Thuault, S. Eve, D. Blond, J. Bréard, M. Gomina, Effects of the hygrothermal environment on the mechanical properties of flax fibres, *J. Compos. Mater.* 48 (2014) 1699–1707. doi:10.1177/0021998313490217.
- [46] K.M. Almgren, E.K. Gamstedt, Characterization of interfacial stress transfer ability by dynamic mechanical analysis of cellulose fiber based composite materials, *Compos. Interfaces.* 17 (2010) 845–861. doi:10.1163/092764410X539235.
- [47] K.M. Almgren, E.K. Gamstedt, J. Varna, Contribution of wood fiber hygroexpansion to moisture induced thickness swelling of composite plates, *Polym. Compos.* 31 (2009) 762–771. doi:10.1002/pc.20858.
- [48] P. Mikshina, T. Chernova, S. Chemikosova, N. Ibragimova, N. Mokshina, T. Gorshkova, Cellulosic Fibres: Role of Matrix Polysaccharides in Structure and Function., *Cellul. - Fundam. Asp.* (2013) 91–

- [49] M. Lindner, Factors affecting the hygroexpansion of paper, *J. Mater. Sci.* 53 (2017) 1–26. doi:10.1007/s10853-017-1358-1.
- [50] D. Derome, M. Griffa, M. Koebel, J. Carmeliet, Hysteretic swelling of wood at cellular scale probed by phase-contrast X-ray tomography, *J. Struct. Biol.* 173 (2011) 180–190. doi:10.1016/j.jsb.2010.08.011.
- [51] R.A. Guyer, H.A. Kim, D. Derome, J. Carmeliet, J. TenCate, Hysteresis in modeling of poroelastic systems: Quasistatic equilibrium, *Phys. Rev. E.* 83 (2011) 61408. doi:10.1103/PhysRevE.83.061408.

## **Figure captions**

**Figure 1.** SEM images of the manufactured composites cross-sections with different porosities- a) flax/PP  $\Phi = 5\%$ ; b) flax/PP  $\Phi = 30\%$ ; c) flax/PP  $\Phi = 50\%$ ; d) glass/PP  $\Phi = 5\%$ ; e) glass/PP  $\Phi = 30\%$ ; f) glass/PP ( $= 50\%$ ). Red circles represent matrix porosity and green circles fibre/matrix interface porosity. (For an explanation of the colour coding on this figure, the reader is referred to the web version of this article.)

**Figure 2.** Water sorption isotherms for a) flax and glass raw fibres as well as b) flax/PP and glass/PP nonwoven composites with 5, 30 and 50% porosity ( $\Phi$ ). Curves are obtained by fitting experimental values with the sigmoidal model. (For an explanation of the colour coding on this figure, the reader is referred to the web version of this article.)

**Figure 3.** Moisture uptake of flax/PP nonwoven composites with 5, 30 and 50% porosity as a function of the square root of time.

**Figure 4.** Dimensional variations as a function of moisture content of a) flax/PP composite  $\phi = 5\%$  in longitudinal (x), transverse (y) and out-of-plane (z) directions. **X and y regression curves are overlapping as the swelling is not significant.** b) Comparison of out-of-plane swelling as a function of moisture content for composites reinforced with glass and flax fibres with different porosities ( $\phi = 5, 30, 50\%$ ). Continuous lines represent sigmoidal fits (For an interpretation of the references to colour coding on this figure, consult the web version of this article.)

**Figure 5.** Tensile behaviours of a) flax/PP and b) glass/PP nonwoven composites at 5% and 50% porosity contents. Solid lines represent stress-strain curves and dotted lines the evolution of the tangent modulus during testing at 23°C and 50% RH. Red dashed lines 1 and 2 in figure a) are used to highlight damage mechanisms in the material. (For an explanation of the colour coding on this figure, consult the web version of this article.)

**Figure 6.** Influence of relative humidity on the evolution of the tangent modulus of nonwoven composites. Different RH levels are represented by the colour coding (10% RH: purple; 33% RH: blue; 50% RH: green; 75% RH: orange; 98% RH: red). Figures 6 a), b) and c) represent the evolution of tangent modulus of flax/PP nonwoven composites with porosities of 5, 30 and 50%, respectively. Figures 6 d), e) and f) represent the evolution of tangent modulus of glass/PP nonwoven composites with porosities of 5, 30 and 50%, respectively. (For an explanation of the colour coding on this figure, consult the web version of this article.)

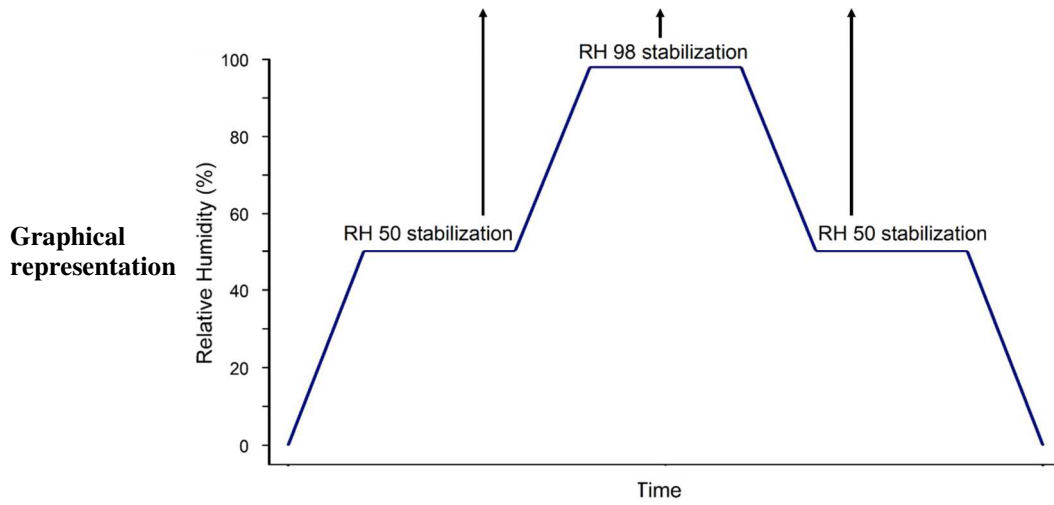
**Figure 7.** Evolution of tensile properties with moisture content in the materials for flax/PP and glass/PP nonwoven composites with 5, 30 and 50% porosity contents. Evolution of tangent modulus a), yield strain b) and yield stress c) as a function of moisture content. (For an explanation of the colour coding on this figure, consult the web version of this article.)

**Tables:**

Material	$W_f$ (%)	$V_f$ (%)	Density [g/cm <sup>3</sup> ]	Porosity (%)	Anisotropy ratio
Flax/PP $\Phi = 5\%$	48.1 $\pm$ 0.1	34 $\pm$ 0.1	1.01 $\pm$ 0.01	6.9 $\pm$ 1.3	1.2 $\pm$ 0.1
Flax/PP $\Phi = 30\%$		23.5 $\pm$ 0.1	0.74 $\pm$ 0.01	32.3 $\pm$ 1.2	1.2 $\pm$ 0.1
Flax/PP $\Phi = 50\%$		17.4 $\pm$ 0.1	0.56 $\pm$ 0.03	48.3 $\pm$ 2.8	1.2 $\pm$ 0.1
Glass/PP $\Phi = 5\%$	49.6 $\pm$ 0.1	23.6 $\pm$ 0.1	1.24 $\pm$ 0.01	5.4 $\pm$ 1.0	1.9 $\pm$ 0.3
Glass/PP $\Phi = 30\%$		15.7 $\pm$ 0.1	1.01 $\pm$ 0.01	29.8 $\pm$ 2.6	1.8 $\pm$ 0.2
Glass/PP $\Phi = 50\%$		10.4 $\pm$ 0.1	0.67 $\pm$ 0.03	49.5 $\pm$ 2.1	2.0 $\pm$ 0.3

**Table 1.** Characteristics of the manufactured composite plates with fibre weight ( $W_f$ ) and volume ( $V_f$ ) ratios, measured density, porosity ( $\Phi$ ) and anisotropy ratios of composites.

Properties	Porosity (%)	RH 50 (s*)	RH 98 (s*)	RH 50 (d*)	RH50 Variation (%)
<b>Tensile properties</b>					
<b>E<sub>0</sub> (MPa)</b>	5	7550 ± 809	3695 ± 611	5719 ± 423	-24
	30	4518 ± 809	2161 ± 336	3597 ± 809	-20
	50	2329 ± 809	1001 ± 235	1741 ± 809	-25
<b>σ (MPa)</b>	5	62 ± 4.3	61.2 ± 4	57.3 ± 4.3	-8
	30	48.1 ± 3.6	45.8 ± 4.5	38.0 ± 3.3	-21
	50	22.4 ± 1.5	21.9 ± 2.5	20.7 ± 3.4	-7
<b>ε (%)</b>	5	1.98 ± 0.28	3.28 ± 0.12	2.46 ± 0.13	+24
	30	2.30 ± 0.12	3.51 ± 0.27	2.39 ± 0.17	+4
	50	2.08 ± 0.07	3.60 ± 0.22	2.62 ± 0.34	+26
<b>Dimensional variations</b>					
<b>Δm (%)</b>	5	2.56 ± 0.10	7.98 ± 0.08	3.55 ± 0.11	+39
	30	2.79 ± 0.03	8.38 ± 0.07	3.35 ± 0.04	+20
	50	2.90 ± 0.13	8.98 ± 0.09	3.31 ± 0.06	+14
<b>Δz (%)</b>	5	1.76 ± 0.08	6.76 ± 0.28	3.48 ± 0.23	+98
	30	1.67 ± 0.23	6.82 ± 0.09	3.74 ± 0.07	+123
	50	1.70 ± 0.28	6.78 ± 0.21	3.90 ± 0.16	+130



**Table 2.** Evolution of hygromechanical properties of flax/PP nonwoven composites with 5, 30 and 50% porosity during a sorption/desorption cycle. Properties (tangent modulus ( $E_0$ ), yield stress ( $\sigma$ ), yield strain ( $\epsilon$ ), moisture uptake ( $\Delta m$ ) and out-of-plane hygroexpansion ( $\Delta z$ )) are assessed after being **stabilised** at each relative humidity level. s\* and d\* denotes the sorption or desorption state of the material, respectively. RH50 Variation represents the evolution of properties between the two 50% RH conditioning steps (sorption and desorption) with negative values in red and positive values in green.

**Figure 1.**

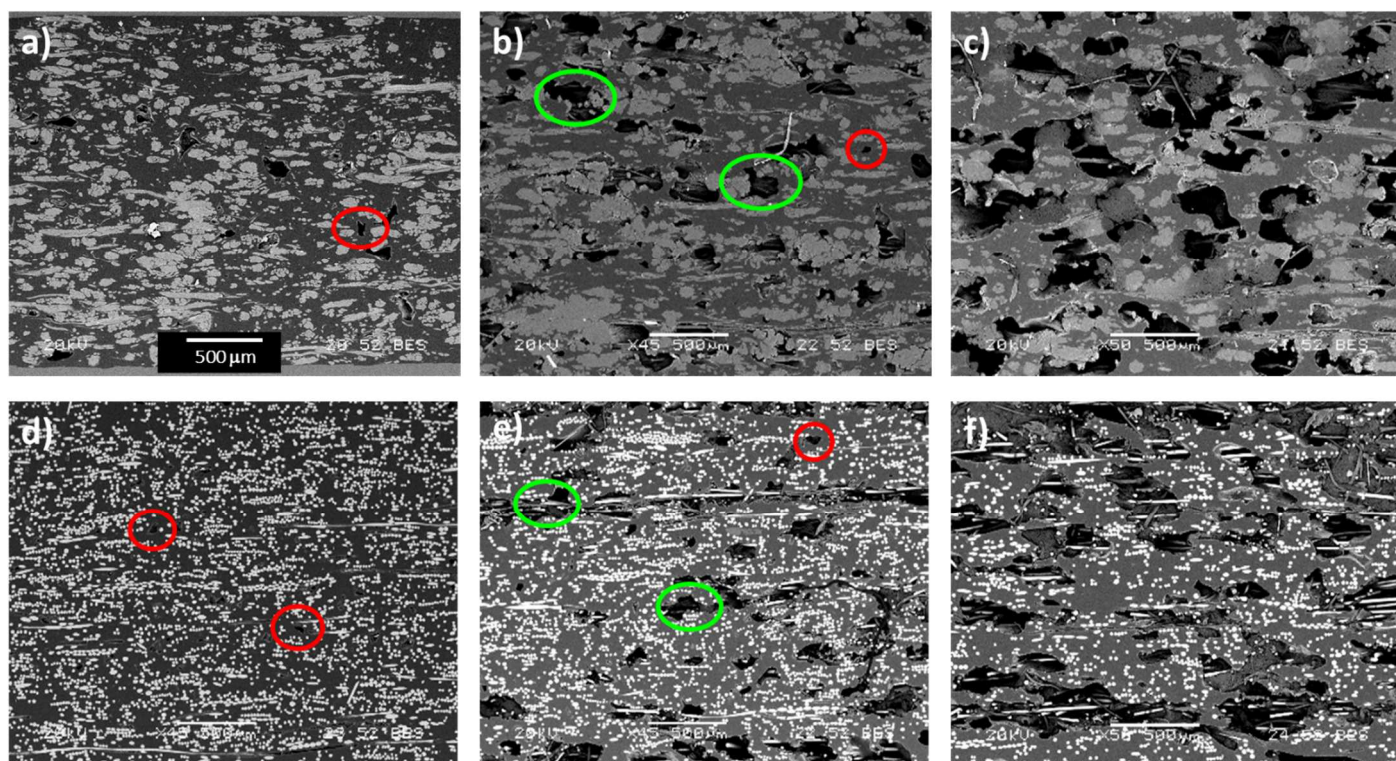
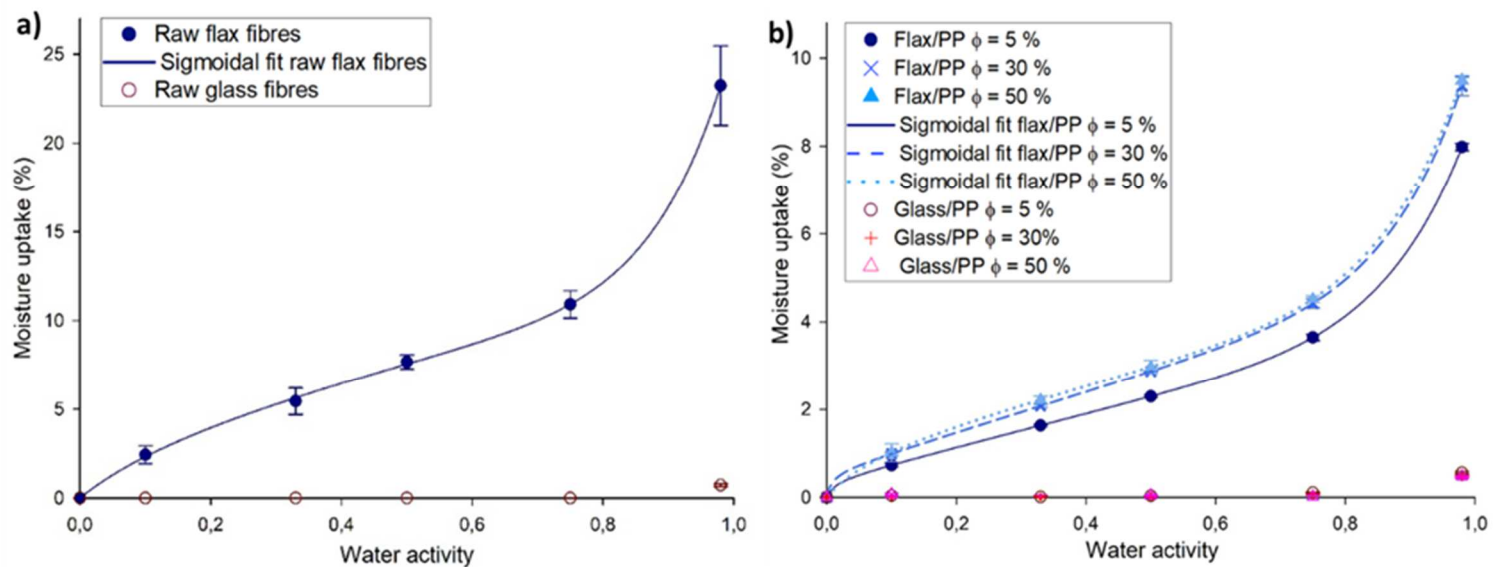
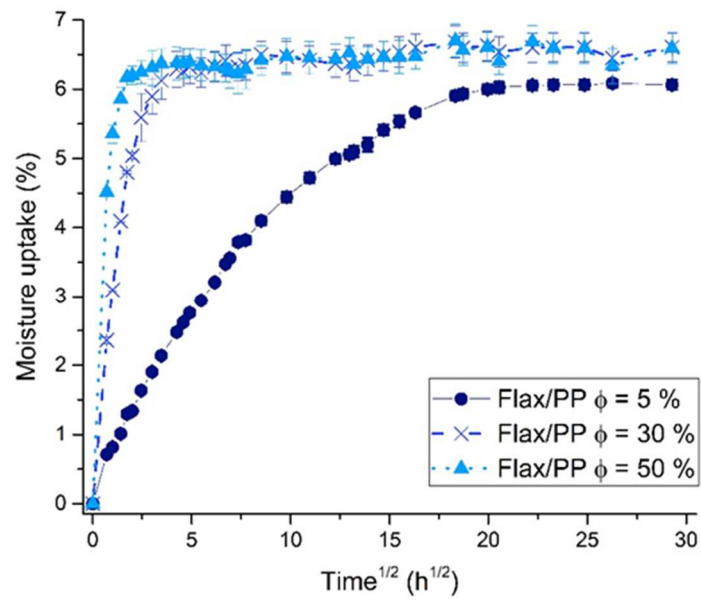


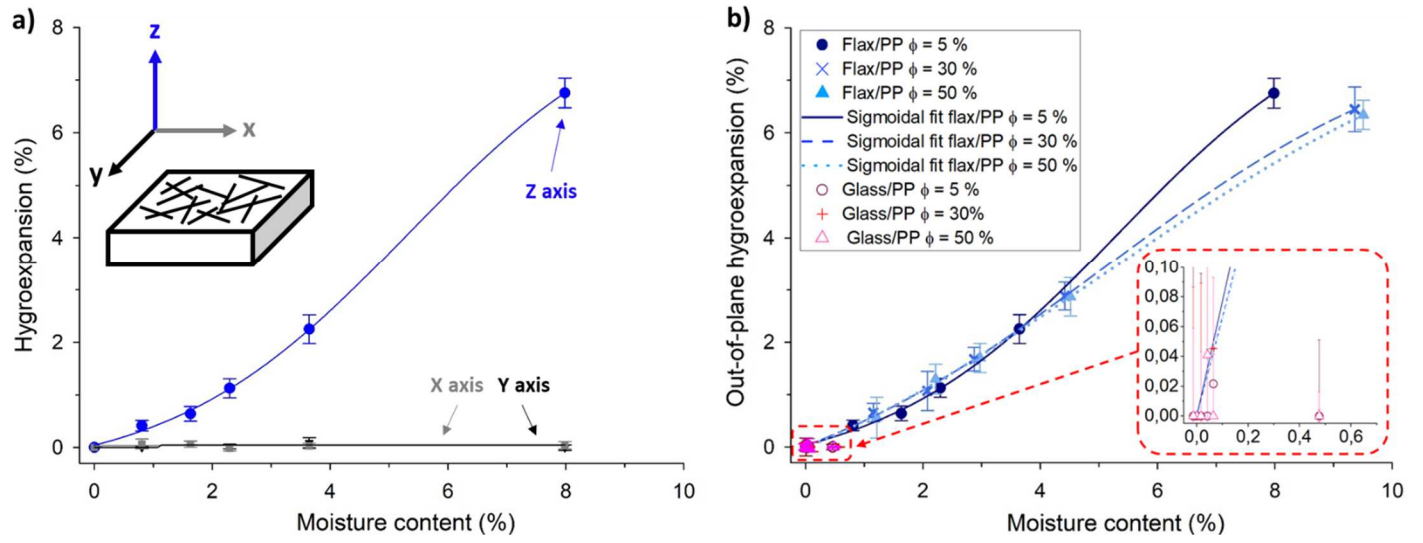
Figure 2.



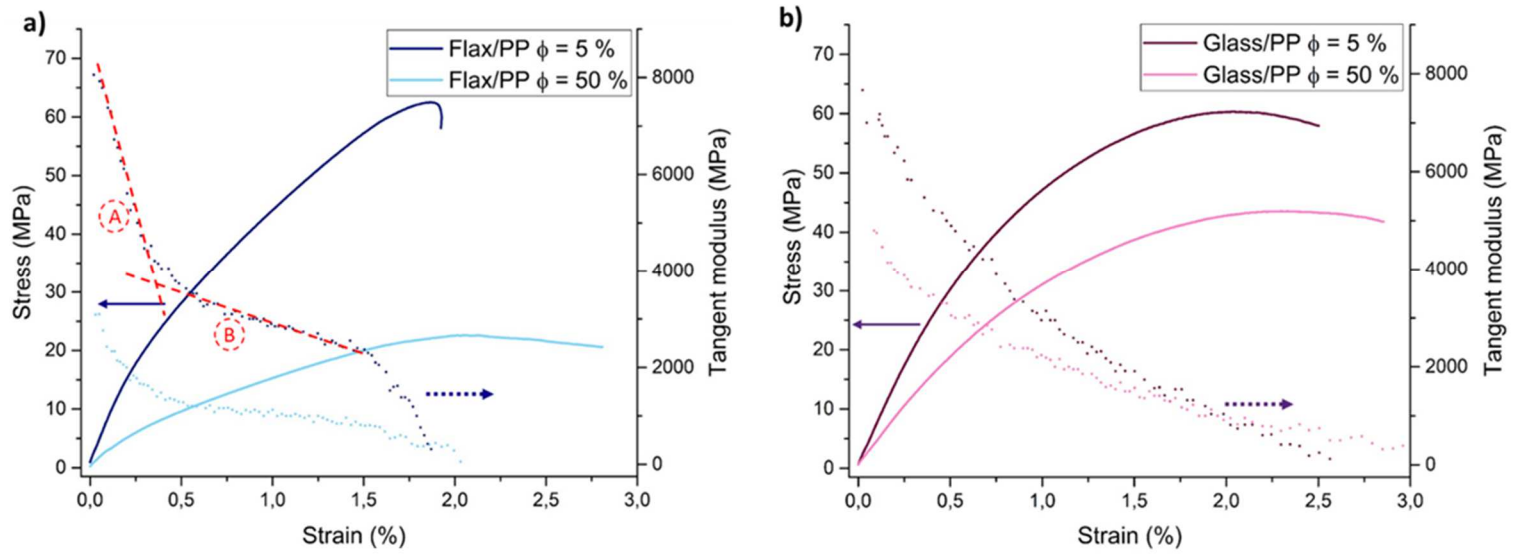
**Figure 3.**



**Figure 4.**

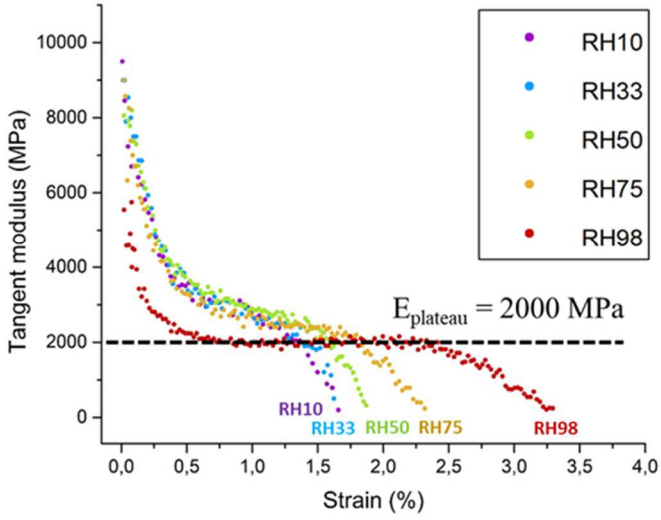


**Figure 5.**

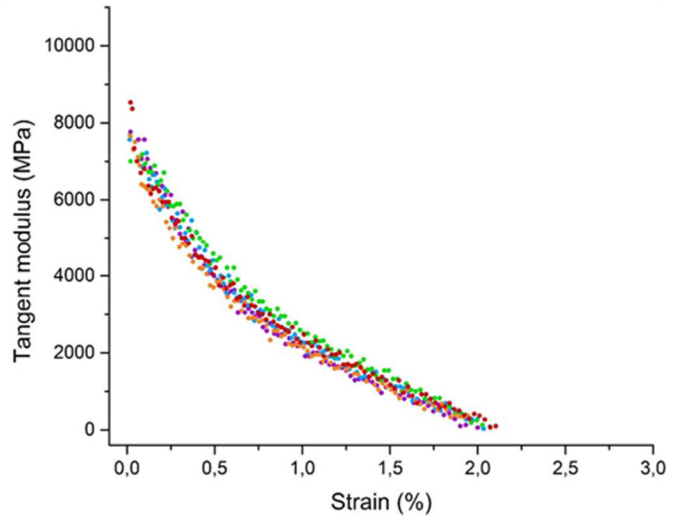


**Figure 6.**

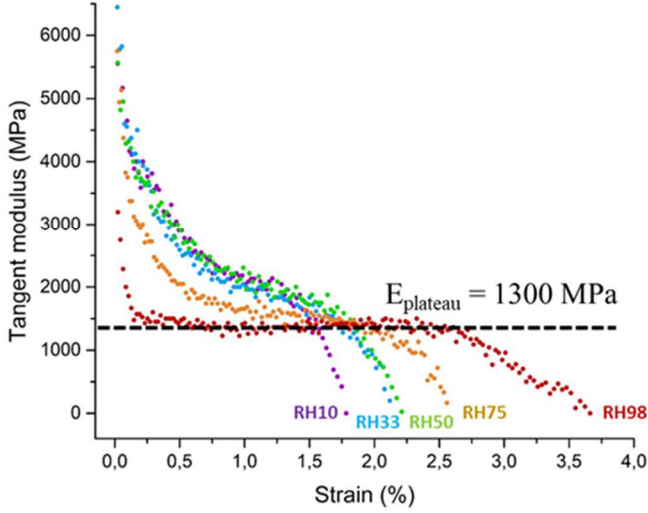
**a) Flax/PP  $\phi = 5\%$**



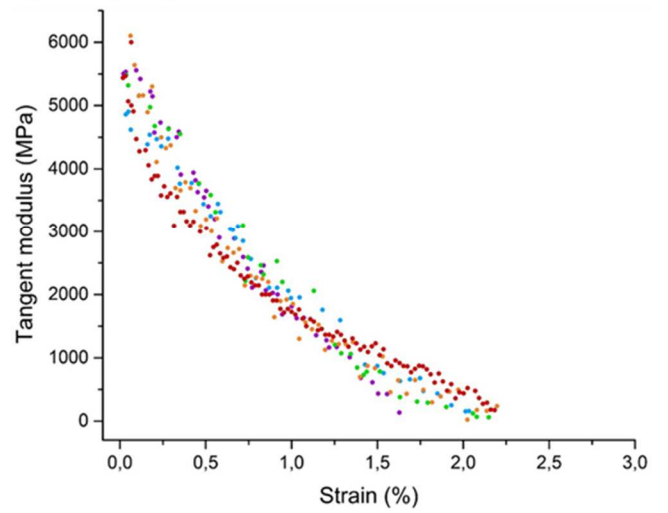
**d) Glass/PP  $\phi = 5\%$**



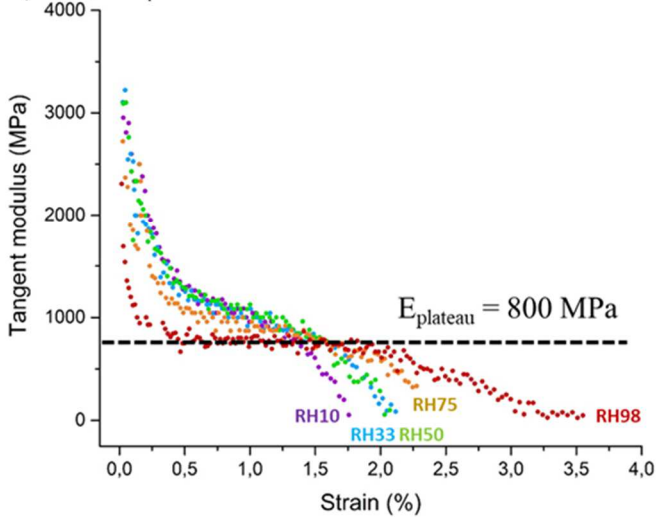
**b) Flax/PP  $\phi = 30\%$**



**e) Glass/PP  $\phi = 30\%$**



**c) Flax/PP  $\phi = 50\%$**



**f) Glass/PP  $\phi = 50\%$**

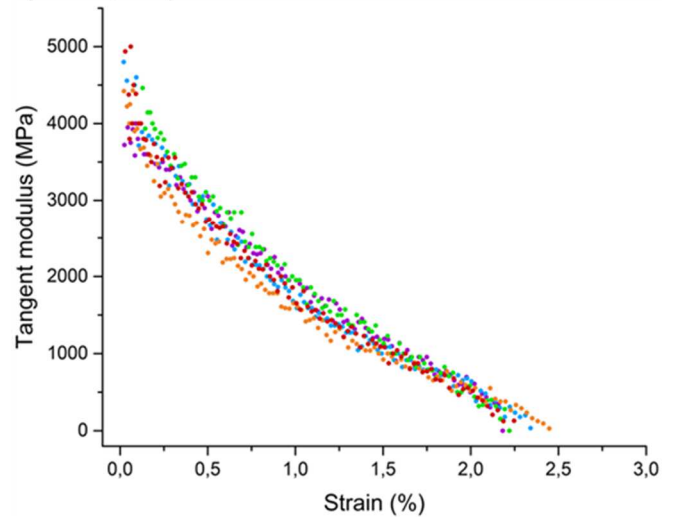


Figure 7.

