EFFECT OF MOISTURE UPTAKE ON FLAX FIBER-REINFORCED COMPOSITE LAMINATES: INFLUENCE ON DYNAMIC AND QUASI-STATIC PROPERTIES

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Abstract

This study proposes to investigate the effect of moisture uptake on a wide spectrum of mechanical behaviours of unidirectional flax fibre-reinforced epoxy laminates. It includes tensile monotonic and fatigue testing. Results show that UD flax-epoxy composites, when exposed to hygrothermal conditioning at 70°C and 85% RH, exhibit a diffusion kinetic which follows a one dimensional Fickian behaviour. The mass uptake at equilibrium is approximately 3.3 % and the diffusion coefficient 6.5 10-6 m^2.s^{-1}. Water vapour sorption is shown to induce a significant change in the shape of the tensile stress-strain curve. Contrary to all expectations, water saturation does not degrade the monotonic tensile strength of such a flax-epoxy composites and induces an increase in the fatigue strength for a high number of cycles.

1. Introduction

Because of environmental and sustainable issues, new rules and regulations, many industrial sectors have more and more interest in bio-based materials [1]. Thanks to outstanding research and recent developments of the last ten years, plant fibres reinforced bio-composites are usually dedicated to non structural applications, e.g. where high loading levels are not required. For example, these applications take places in automotive, aerospace, packaging and building industries. High mechanical properties of fibres extracted from plants, such as flax, hemp or sisal make them good candidates to be used as composite reinforcements for lightweight structural applications. Up-to-date critical reviews of literature [1-7] show that several technological and scientific barriers still remain to be knocked down in order to obtain fully optimised plant fibre composites (PFCs) dedicated to structural applications. One of the main question concerns the long-term durability of such composites.

Indeed, for some intended applications, Plant Fibre Composites (PFC) will be heavily exposed to moisture, various temperatures and UV radiation. These environmental loadings can strongly affect their mechanical properties. Their durability will be thus drastically affected.

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Most of the polymer composites absorb moisture in humid atmosphere and when immersed in water. Water absorption can lead to a change in the physical, chemical, mechanical and dimensional properties of the matrix (plasticization, hydrolysis, swelling). Water can also degrade fibre-matrix interface. This induces a decrease in the stress transfer efficiency and in the mechanical properties of the composite materials. Contrary to glass or carbon fibres, plant fibres have a high affinity with water. This property lead composite material to high water uptake capacity. Vapour or liquid water can thus be easily absorbed by PFCs through fibres. Consequences are (i) water adsorption into the cell wall through hydrogen bonds (ii) degradation of cellulose macromolecules, , (iii) swelling and (iv) risk of drastic decrease of the mechanical properties of the PFCs [8-11]. It is therefore of paramount importance to characterize moisture absorption and its influence on the long-term behaviour of PFCs. Despite recent works on fatigue, the influence of moisture absorption and environmental loading has not been yet considered.

Thus, the aims of this paper is to quantify and show the influence of water vapour sorption on a large spectrum of mechanical behaviours of flax fibres reinforced epoxy laminates. Here, targets are quasi-static, fatigue.

This work was conducted as part of the scientific collaboration between three French laboratories (DRIVE, FEMTO-ST and C2MA). The laboratories are members of a research consortium called BIOCOMP. This consortium groups several laboratories. These partners have expertise in the field of mechanical and fiber reinforcements, methods for preparation and characterization of fibrous reinforcements, polymers and composites, biochemistry as well as chemistry of interfaces.

2. Material and methods

2.1 Composite materials

Composite bio-based-materials have been fabricated from prepregs flax fibres and epoxy resin. FlaxPreg UD 150 was supplied by Lineo company (France). The flax fabric has an areal weight of 154 g/m^2. It contains 95% of warp yarns and 5% of weft yarns in weight. The epoxy resin is an Epoxy Huntsman LY 5150. The fibre fraction is approximately 50% in weight before curing.

2.2 Composites fabrication

Five plates (400 mm x 400 mm x 2.2 mm) were fabricated using a thermo-compression process under specific pressure and temperature cycle (curing under 130°C/4,5 Bar during 90 minutes). They are numerated P1 to P5. These plates consist in 10 elementary unidirectional plies. Each plate has been characterized in terms of porosity and fibre contents, according to ASTM D 3171-99 standard (table 1).

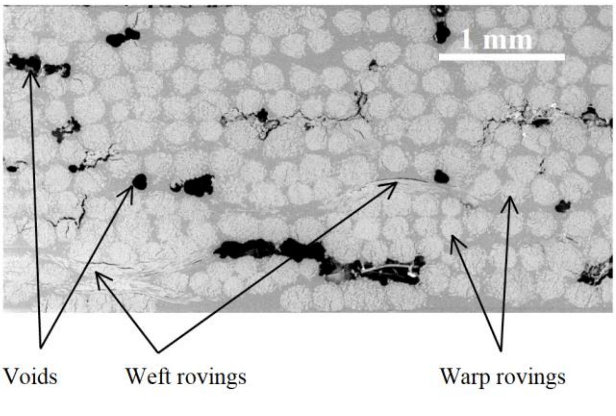

Figure 1. Scanning Electron Microscopy (SEM) cliché / cross-section of the produced material

A scanning electron microscope (Hitachi S-3000N) has also been used to observe the cross-section areas of the produced materials. Images have been recorded under partial vacuum with back-scattered electrons. A specific attention was dedicated to the study and the distribution of the voids (figure 1). On such image, the warp yarns (white areas) and the voids (black areas) can be easily identified. The presence of weft yarns into the reinforcement structure is also easily observed.

The Table 1 summarize principal characteristics of the produced plates and the way they have been aged and mechanically tested.

Table 1. Plate thickness, porosity and fibre/matrix volume fractions

Plates	Thickness (mm)	Fibre vol. ratio (%)	Matrix vol. ratio (%)	Void vol. ratio (%)	Water uptake	Monotonic tests	Fatigue tests
P1	2.19	45.0	46.4	8.6			X
P2	2.18	45.4	47.4	7.2	X		X
P3	2.16	45.6	47.7	6.7	X	X	
P4	2.17	45.7	48.0	6.3	X	X	
P5	2.17	45.8	46.1	8.1	X	X	

2.3 Sample preparation

Rectangular specimens (250 mm long and 25 mm large) were sawn with a high-speed diamond cutting machine. This operation allowed to produce unidirectional laminates, first in the fibres direction (UD 0°) and second perpendicularly to the fibres direction (UD 90°). After cleaning and preparing the surface (sanding and cleaning with acetone), aluminum tabs (50 mm long, 25 mm large and 3 mm thick) have been bonded (with Loctite super glue 3 adhesive) to the test specimens dedicated to quasi-static and fatigue tests. Samples were speckled in order to follow strain plane tensor by digital image correlation [12].

2.4 Hygrothermal conditioning (P2, P3, P4, P5)

After fabrication, samples were stored in a regulated chamber at 21°C and 50% RH before mechanical testing. This hygroscopic state was named “DRY”. Samples were also aged in a hygrothermally regulated chamber at 70°C and 85% RH for more than 2 weeks. During this period, relative weight uptake, dimensions, dynamic tensile modulus were monitored. A Mettler-Toledo AT200 balance was used for weight monitoring (precision of 0.001 g) and a caliper with a precision of 10^{-2} mm was used to evaluate the swelling. At the end of the two-week monitoring period, an equilibrium state named WVS (Water Vapour Saturated state) has been achieved.

2.5 Monotonic tensile tests (P3, P4, P5)

Young modulus, ultimate tensile stress and strain were determined by uniaxial tensile tests on a MTS testing machine (model Criterion C45.105), equipped with a 100 kN load cell. Samples were loaded at a crosshead speed of 1 mm/min. Each set of samples (DRY and WVS) have been tested.

2.6 Tensile-tensile fatigue tests (P1, P2)

The tensile-tensile fatigue tests were performed under a triangular waveform loading, at a frequency of 5 Hz. Using a load amplitude control mode, the ratio between minimum and maximum stress (R) was fixed to 0.1. The experimental configuration (loading machine, specimen shape and preparation) was similar to monotonic tensile tests. Fatigue tests were performed in the fibre direction for DRY and WVS specimens. Four levels of maximum stress were applied. The levels were 80%, 65%, 50% and 40% of the quasi-static tensile failure stress (at 2 $mm.min^{-1}$) for the DRY specimens and approximately 75%, 60%, 55% and 45 % for the VWS specimens. At least three replicates were tested at each stress level. Tests were stopped at failure.

Tests were carried out under controlled hygrothermal conditions (T = 21 °C ±3 °C, RH = 50% ±5%). For WVS specimens, it was necessary to keep the water-saturated state during fatigue loading. Thus, the specimens were covered, after the hygrothermal treatment, with a flexible coating film. This film was made with an aqueous mixture of iron oxide-filled and acrylic resin. The mass of each specimen was measured after hygrothermal treatment but also before and after fatigue loading. The mass variation was checked to be negligible and the water-saturation state has been assumed to be constant during tests. Peak-to-peak and average amplitudes of load and strain sensors were recorded for each cycle.

3. Results and discussion

3.1 Water Vapour absorption behaviour

Figure 2 displays the water uptake of samples from P2, P4 and P5 under humid environment described before. The diffusion kinetic follows a one dimensional Fickian behaviour (solid lines in figure 2) as presented by Scida et al. [13]. After 6 days, the final water uptake reaches an equilibrium between 3,0 and 3.3 ± 0.2%. The coefficient of diffusion D is estimated at $6.47\text{x}10^{-6} \pm 0.14\text{x}10^{-6}$ $m^2.s^{-1}$. It is interesting to notice the different kinetic in the case of P4 specimen. This plate shows a lower maximal mass uptake at 3,0 %. This phenomenon can be explained by its lower void ratio (6.3%) in comparison to other specimens from P2 and P5 (respectively 7.2% and 8.1% - see table 1).

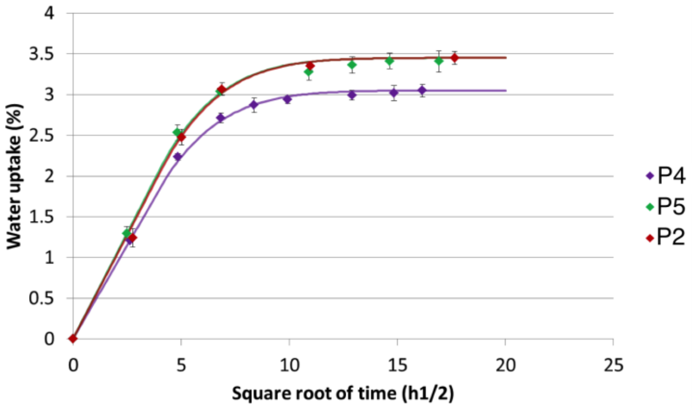

Figure 2. Water uptake of composite samples and 1D-Fickian kinetic prediction (solid lines) when exposed to a hot humid environment (70°C/85%rh)

3.2 Monotonic tensile tests

Results are presented in figure 3 and completed by values presented in table 2. Moduli were evaluated in the strain interval 0.01 to 0.15%, using linear regression. Concerning DRY specimens, the mean values of tensile moduli (approximately 30 GPa in the fibre direction and 4.7 GPa in the transverse direction), tensile strength (approximately 320 MPa for UD 0° and 30 MPa for UD 90° direction) and strain at failure (approximately 1.5% in the fibre direction and 1% in the transverse direction) are in good agreement with data of literature collected on the same type of flax-epoxy laminates [14, 15].

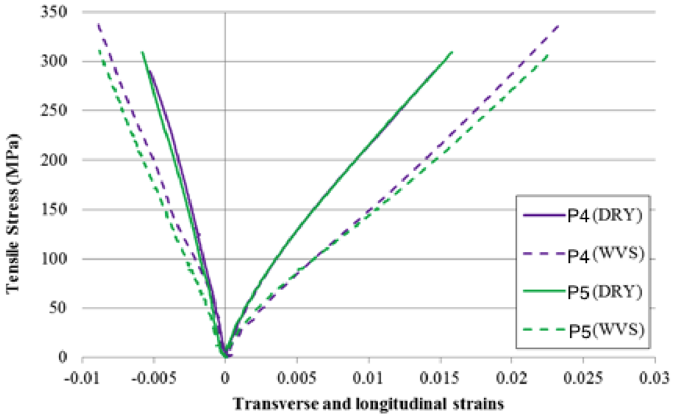

Figure 3. Tensile stress-strain curves for 0°UD specimens before and after aging

Moreover, results show that water absorption induces a change in the stress-strain curves (UD 0°). An increase of strains, a decrease of the Young modulus as well as a slight increase of the Poisson's ratio are observed. These evolutions can be attributed to the plasticization of both matrix and fibres by water molecules. WVS specimens exhibit slightly superior tensile strengths to DRY specimens. Munoz and Garcia-Manrique [16] also observed a higher tensile strength of flax-epoxy composites after aging. They attributed this behaviour to the swelling of flax fibres in the composite material (as a result of water absorption). This phenomenon was assumed to induce a stronger interfacial bonding between the fibre and the matrix.

Table 2. Properties obtained in monotonic tensile tests (mean value ±standard deviation)

Specimens	**E_L (GPa)**	**σ_L (MPa)**	**σ_T (MPa)**	**ε_L (%)**	**ε_T (%)**	**ν_{LT} (-)**
DRY (P4, P5)	30.4 ± 2.7	319 ± 20	30.0 ± 1.3	1.59 ± 0.14	0.99 ± 0.16	0.34 ± 0.02
WVS(P4, P5)	19.6 ± 2.0	320 ± 20	25.9 ± 1.5	2.32 ± 0.14	1.80 ± 0.21	0.37 ± 0.02

3.3 Tensile-tensile fatigue tests

S-N curves for tensile-tensile fatigue tests on DRY and WVS specimens are presented in figure 4. For each studied load level, and particularly for WVS specimens, there is a slight dispersion in the maximum tensile stress. This is due to the variation in cross-section of the specimens. Results show that both set of specimens (DRY and WVS) present a gradual decline in fatigue strength when the number of fatigue cycles increases. For a fixed loading level, dispersion in fatigue life duration is observed. The order of magnitude of this dispersion is in agreement with data collected on flax-epoxy laminates [14, 15, 18]. The fatigue strength can be fitted by the Wöhler law, as a function of number of cycles. The fatigue strength for a high number of cycles (1 million) is approximatively 140 MPa for DRY specimens. This value represents 42 % of the monotonic tensile strength. This range of value corresponds to the observations produced by El Sawi et al. [14] on the same type of unidirectional flax-epoxy laminates.

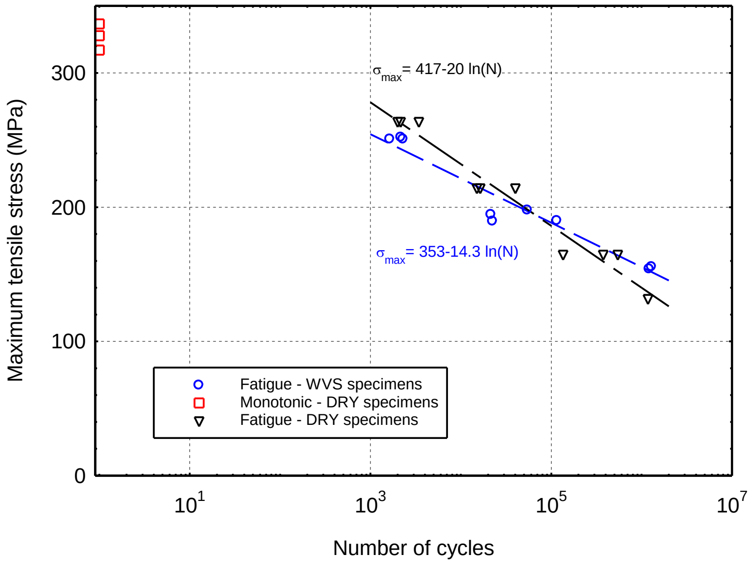

Figure 4. S-N curves of DRY (P1) and WVS (P2) specimens

Results also show that the fatigue strength is significantly influenced by water sorption. Concerning WVS specimens, the initial dynamic modulus is 22 GPa, against 33GPa for DRY specimens. Contrary to all expectations, the fatigue strength for a high number of cycles is higher for WVS specimens than for DRY ones. In the case of WVS, the fatigue strength reach 155 MPa (47% of the monotonic tensile strength). This phenomenon is assumed to be due to the plasticization of the laminate.

4. Conclusions

This study focused on the consequences of water vapour sorption of unidirectional flax-epoxy laminates. Mechanical investigations concerned monotonic and fatigue behaviours. The motivations of this work was to evaluate the durability of bio-based laminates for structural applications.

Results show that, when exposed to hygrothermal conditioning at 70°C and 85% RH, the diffusion kinetic of UD flax-epoxy composite follows a one dimensional Fickian behaviour. Water vapour sorption is showed to induce a significant change in the shape of the tensile stress-strain curve and a decrease in dynamic elastic modulus. Contrary to all expectations, water saturation does not degrade the monotonic tensile strength of these flax-epoxy composites. Moreover, an increase in the fatigue strength is clearly observed on wet specimens. Water sorption seems to activate some hardening phenomena. This is a benefit for both static and fatigue strength. This result is of paramount importance for bio-based composites end-users. Future of this work will help to quantify the influence of the exposure time to such hygrothermal conditions (i.e. ageing) on the mechanical behaviours of PFCs. The final purpose is to understand how microstructural mechanisms helps to harden these bio-based composites composite when thy are submitted to moisture.

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