Influence of hydrothermal ageing on the fatigue behaviour of a unidirectional flax-epoxy laminate

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ABSTRACT

Although the research and development in plant fibre composites is growing rapidly, these materials still require specific considerations before being adopted by industry in structural applications. One of the main issue is related to their durability. The objective of this study is to investigate the influence of hydrothermal ageing on the fatigue behaviour of an unidirectional flax-epoxy laminated composite by implementing the fatigue tests in a water bath. Results show that while the quasi-static strength and rigidity are significantly affected by the ageing, the fatigue strength is however improved. The fatigue strength coefficient is more than two times lower after ageing, indicating a lower decrease of the maximum stress with the increasing number of cycles. After ageing, the maximum stress level for an expected

lifetime of $5.3 \ 10^6$ cycles is approximately 90 MPa, which is comparable to the one of unaged specimens. It is also pointed out that the remaining quasi-static tensile properties and behaviour of the aged specimen are not significantly affected after $1.3 \ 10^6$ cycles at a stress level of 90 MPa.

1. INTRODUCTION

Research and innovation in plant fibre composites (PFCs) is growing rapidly [1, 2], in particular in well-established application sectors such as automotive and plastic industries, building and construction and emerging market of consumer goods and end-use industries [3]. Until now, the most attention has been devoted to the development of short-fibre and non-woven based composites, which are not suitable for structural applications. The development of structural PFCs is a result expected from the scientific community but it is not yet fully achieved. Anyway, promising progress has been recently achieved, in particular in the development of plant-based preforms with low areal weight and perfectly aligned fibres [4] and their composites. For some of them, based on flax, hemp and sisal fibres mainly, the reached static and fatigue mechanical performances pave the way for load-bearing applications [5-9]. The potential of PFCs for this type of structural application is also increasingly demonstrated at the scale of prototypes such as wind turbine [10] and boats [11] for examples. However, PFCs still require specific considerations before being adopted by industry in structural applications [12]. One of the main issue is related to their durability and, in particular, to the effect of environment and complex loading path on their long-term

mechanical behaviour. Indeed, for some intended applications, PFCs will be heavily exposed to moisture, liquid water, various temperatures and UV radiations in addition to static, cyclic and dynamic mechanical loadings. These environmental exposures can strongly affect their mechanical properties and therefore play a critical role in the service life of PFCs.

Wetting/drying cyclic effects has been recently studied. Sodoke et al. [13] showed on a quasi-isotropic flax/epoxy composite that despite the induced structural degradation, a good retention of the mechanical properties is observed for the considering cycling times.

The fatigue behaviour of PFCs has been recently critically reviewed by Mahboob and Bougherara [12]. Their review of the state-of-the-art points out the main limitations of the existing studies. They are mainly related to loading path parameters, the influence of fibre type and the understanding of internal physical mechanisms and damage during fatigue loading. One additional point that can be raised is related to the influence of moisture content. To the best of the authors knowledge, this issue has been only poorly considered in literature. We proposed in a previous paper [14] an investigation of the influence of hygrothermal ageing on the fatigue behaviour of a quasi-unidirectional flax-epoxy composite. In the present study, we investigate the influence of hydrothermal ageing on the fatigue behaviour of a purely unidirectional flax epoxy composite. Indeed, the influence of hydrothermal ageing has been poorly studied whether on the static properties [15-19] or cyclic behaviour [20-22]. Assarar et al. [23] observed on a flax/epoxy laminate after 40 days of immersion at room temperature a decrease in tensile strength and modulus of 25% and 39% respectively and an increase in strain at failure of approximately 63%. Moudood et al. [18] reported a decrease by 9% and 57% for the tensile strength and modulus, respectively, for a unidirectional flax/bio-epoxy composite after immersion until saturation. Li and Xue [15] studied the

influence of water immersion time and temperature on the tensile properties of an unidirectional flax/epoxy composite. They pointed out the influence of hydrothermal ageing, as well as drying after ageing, on the nonlinear shape of the tensile curve. After 17 weeks of immersion at 60°C, their results show a decrease in the tensile strength of approximately 15% for the water-saturated material and of approximately 37.5% for the aged and then dried specimen. Chilali et al. [21] highlighted for flax fibre-reinforced thermoplastic and thermoset composites that the stiffness loss during load-unload tensile tests is accentuated with ageing due to the microstructural change and the plasticization phenomena caused by water absorption. Sodoke et al. [20] showed that the fatigue strength of a quasi-isotropic flax/epoxy composite drops with hydrothermal ageing. These results were collected on specimens which were dried after ageing. Habibi et al. [22] showed that the water uptake has also a significant effect on the fatigue properties on short flax fiber-reinforced epoxy composites.

Therefore, for all these studies, the mechanical tests were performed either on specimens dried after ageing or in ambient air after having taken the specimens out of the immersion bath. In the first case, it is difficult to discriminate the origins of damage and microstructural changes between water ageing and drying. In the latter, it may be assumed that the moisture content remains at its equilibrium and homogeneous in the specimen volume only for short times, which may be valid for monotonic tests up to failure but not for fatigue tests.

So, in this work, we propose to study the influence of hydrothermal ageing on the fatigue behaviour of a UD flax-epoxy composite by implementing the fatigue tests in a water bath.

2. MATERIAL AND METHODS

2.1 Composite material and specimen preparation

Unidirectional flax/epoxy composites were manufactured by thermos-*compression. The manufacturing process is fully detailed in [24]. In short, a tape of purely unidirectional flax fibres supplied by LINEO© (FlaxTape^{TM} 110) was used. Its areal weight is 110 g/m². The DGEBA epoxy resin SR8500 and its hardener SZ8525 were supplied by Sicomin©. 20 layers of flax fibres were manually impregnated with the liquid resin and cured in a Fontjine Grotnes TPC 321 thermo-compression press under a pressure of 1 bar at 80°C for 1h, followed by a post-curing at 80°C for 2h. The glass transition temperature of the epoxy resin is around 110°C.

Samples were cut using a water lubricated circular saw at the following dimensions: 250 mm x 25 mm and 80 mm x 25 mm for the longitudinal and transverse tests, respectively. The thickness was around 4 mm for all samples. Their volumetric fibre and void contents were respectively 37.7% and 1.7%. They were determined by SEM image analysis of the transverse cross-section of composites. Approximately 200 SEM images, each one representing an area of 1.2 mm², were analysed for this determination.

2.2 Conditioning and hydrothermal ageing

After manufacturing and cutting, and prior to any test or immersion, all the samples were stored under controlled conditions at 50 % RH and 23 °C until their mass equilibrium is reached. This state corresponds to the unaged state.

Some of the specimens were then immersed in distilled water at 70°C during 26 days, representing approximately the time necessary to reach the moisture content equilibrium.

This state corresponds to the aged-saturated state. The temperature was used to fulfil the NF EN 2823 standard requirements. The samples were periodically taken out and weighted using an electronic balance with a 0.01 g precision.

Finally, some of the aged specimens were air-dried and then again conditioned at 23°C and 50% RH until their mass equilibrium is reached. This last state is called aged-dried state.

2.3 Mechanical testing

2.3.1 Monotonic tensile tests

Monotonic tensile tests were performed accordingly to the ISO 527-4 standard on a MTS Criterion 50 tensile machine equipped with a 100 kN load cell. The crosshead speed was 2 mm/min. The deformations in the longitudinal and transverse directions were measured using clip-on extensometers. Aluminium tabs were glued on the specimens. The gauge lengths of the samples were 150 mm for the tests performed in the longitudinal direction. For each condition, at least 3 specimens were tested.

Tests were performed in the ambient atmosphere for the unaged and aged specimen. For aged specimens, the total time outside the immersion bath before testing was kept within 10 minutes to limit water desorption.

Similar tests were also done after ageing and drying (aged-dried specimens).

Since the unidirectional flax fiber-reinforced composites exhibited a non-linear behaviour, two Young's moduli were determined with a linear regression on the first part of the curve (below the inflexion point) and with a linear regression on the second part of the curve (above the inflexion point). More details can be found in a previous work on the same composites [24].

2.3.2 Tensile-tensile fatigue tests

Tensile-tensile uniaxial fatigue tests were performed in the ambient atmosphere for the unaged specimens and in water immersed conditions for the aged specimens. An Instron 8501 hydraulic press equipped with a 100 kN load cell was used. A triangular waveform loading was imposed at 5 Hz, using a load amplitude control mode, with a loading ratio R of 0.1. The longitudinal deformation was measured using an extensometer with a 50 mm gauge length.

Fatigue tests were performed in the fibre direction. Five and six levels of maximum stress were applied on the aged and unaged specimens, respectively. For the unaged specimens, the levels were 80, 72.5, 65, 57.5 and 50% of their mean quasi-static tensile failure stress (measured on specimen equilibrated at 50% RH and 23°C), i.e. 235, 213, 191, 169 and 147 MPa. For the aged specimens, levels were 70, 65, 60, 55, 50 and 40% of their mean quasi-static tensile failure stress (measured on water-saturated specimens), i.e. 147, 137, 126, 116, 105 and 84 MPa. At least three replicates were tested at each stress level. Tests were stopped at failure or at a maximum of approximately $1.5 \ 10^6$ cycles.

Tabs were used only for unaged specimens. Indeed, for aged specimens, no significant difference was observed on elastic and fracture properties by using tabs or not. For unaged specimens, tabs were made in the same composite material and with a fibre orientation tilted of 90° when compared to the fibre orientation in the specimen (Figure 1).

For the aged samples, a specific in-house set-up was designed and manufactured to perform the fatigue tests under immersed conditions, and to ensure that the specimens remain at their moisture content during the fatigue test. The scheme and some pictures of the immersion set

up are shown is Figure 2. It consists of a PVC tank, fixed on the shaft on the bottom part of the machine. The specimen and its clamps are fully immersed in the bath during testing. Specific mechanical wedge-action grips were designed and machined in stainless steel. A specific clip-on extensometer was also designed and manufactured specifically to operate under water-immersed conditions. The gauge part of the extensometer was made in copper beryllium alloy and the strain gages covered with a gel coat (Nitrile Rubber) to seal them after gluing.

3. RESULTS AND DISCUSSION

3.1 Water absorption

Figure 3 shows the evolution of the moisture uptake in composite and neat resin as a function of the square root of immersion time divided by the thickness of the sample. The experimental results are fitted using a one dimensional Fickian law as suggested by several authors [21, 23] for flax-epoxy composites (solid line in the plot). This type of model allows the description of the two-steps behaviour generally observed experimentally for this material. The identified diffusion parameters are summarized in Table 1.

Results show that the maximum water absorption is significantly larger than those of epoxy resin. This change in the absorption behaviour is attributed to the flax fibres as well as to the interfaces between individual fibres (pectic-rich region) and between fibres and resin.

The diffusion coefficient of the composite material is equal to approximately $2.85 \ 10^{-6} \ mm^2.s^{-1}$ in the thickness direction. This value is higher than some values reported in literature for woven flax-epoxy composites immersed in tap or salt water at ambient temperature [16, 25]

and in the same order of magnitude when compared to flax-epoxy composites immersed at 60°C [15] (see Tab. 1). The value of the water uptake at saturation, approximately 11.9%, is in agreement with the literature. The reported values are comprised between 7.45% and 13.5% [16, 25]. This scattering is attributed to many parameters, as the nature of the epoxy matrix, the first state before immersion, and the form of the fibres. The figure 3 shows that after 23 days of immersion, the equilibrium plateau is reached meaning that the composite is completely saturated. Table 1 reports also the dimensional variation induced by the water sorption. Average increases of approximately 10% and 3% are measured in the thickness and width of the composite specimens, respectively. It represents a swelling of approximately 5.7 and 2 times higher than for the neat resin, in the two considered material directions, respectively.

3.2 Influence of the hydrothermal ageing on the monotonic tensile properties

The monotonic tensile properties were determined for the three conditioning states (unaged, aged-saturated, aged-dried). Data are synthetized in Table 2. For the unaged specimens, the mean values of the elastic modulus E_L^{m1}, tensile strength and strain at failure, respectively 23.7 GPa, 288 MPa and 1.6%, are in agreement with the data from literature [2, 6, 7], in particular when considering the fibre volume fraction (37%). The ageing induces a significant decrease in the initial stiffness of the specimens (E_L^{m1}) approximately -64% and -25% for the aged-saturated and aged-dried specimens. These results show that a part of this decrease is due to the plasticizing effect of water and the other part to the ageing induced damage. Anyway, considering the highly non-linear behaviour of this type of composite, the comparative study needs to be undertaken with care. Figure 4 (a) represents the typical stress-

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strain curves obtained for the three different conditioning. It can be observed that all the curves present a non-linear behaviour with a yield point located near 0.2% of longitudinal strain. This is particularly obvious in Figure 4 (b) where the tangent apparent modulus is plotted as a function of the longitudinal strain. This result is globally in agreement with the one reported by Li and Xue [15]. But, it has also to be underlined that significant differences also exist in the shape of the tensile curves. Interestingly, curves for the aged specimens exhibit a three-phasic behaviour. Contrary to unaged specimens, after ageing a tensile strain hardening is observed above the yield point. A maximum stiffness increase of even so 57% when compared to the stiffness at the yield point is measured, meaning that the specimen recovers just before failure its stiffness measured for the very low strains. This strain hardening could result from a hydro-activation of time-dependent phenomena such as the cellulose microfibrils re-orientation.

For the tensile strength, a decrease is also observed after ageing, with a reduction of 26.8% in water-saturated condition and 30.3% after drying. The drying, following hydrothermal ageing, certainly produces extra damage which explains this reduction in strength. It can be observed also that the coefficient of variation (CoV) for this condition is significantly higher, reaching 17.4%, when compared to the unaged and aged-saturated conditions for which the CoV is lower than 4%. This increase in the scattering could be attributed to a heterogeneous damage state in specimens after drying.

Regarding the strain at failure in the longitudinal direction, an increase of 35.4% is observed after ageing in water up to saturation. This is consistent with the plasticizing effect of water on the amorphous carbohydrates constituting the flax fibre. Contrarily, the strain at failure after drying is 20% lower than before ageing. This trend is also observed for the strain at

failure in the transverse direction. The unaged material presents a strain at failure in the transverse direction of 1.61% and after ageing the value reach 2.18%.

3.3 Influence of the hydrothermal ageing on the tensile-tensile fatigue properties

The S-N curves for tensile-tensile fatigue tests for aged-saturated and unaged specimens are presented in Fig. 5. To the best of the authors' knowledge, it is the first time that fatigue results collected in immersion for PFCs are presented in the open literature.

For the two states (unaged and aged-saturated), results show a gradual decline in fatigue strength with increasing number of fatigue cycles. For unaged specimens, the order of magnitude is in agreement with data from literature collected on flax-epoxy specimens [9, 26-30]. The fatigue strength as a function of number of cycles is accurately fitted by a logarithmic law (Eq. 1).

$$S_{max} = \alpha - \beta \ln(N) \tag{1}$$

where N is the number of cycles at failure, S_{max} the maximum tensile stress, α the single cycle ultimate strength and β the material fatigue strength coefficient.

For the unaged specimens the β parameter is equal to 18.4. This value is also in agreement with data from literature [14, 29].

For the aged-saturated specimens, it can be observed, that the maximum stress in the low – cycle range is lower when compared to the unaged specimens. As an example, for 20 000 cycles, the maximum stress after ageing is approximately 148 MPa against 196 MPa before ageing.

Interestingly, it can be observed (Figure 5) that this difference decreases with increasing number of cycles. Indeed, the β parameter is significantly lower for the aged-saturated

specimens (9.4 vs. 18.9). For a lifetime of approximately 5.3 10^6 cycles, the two fitted S-N curves converge toward the same maximum stress value, approximately 90 MPa. It was demonstrated recently for this material that the maximum stress continues to decrease as a function of increasing number of cycles up to at least 10^8 cycles [29]. So, the fitted S-N curves can be extrapolated reliably in the high cycle fatigue range.

Based on this result, it can be concluded that the hydrothermal ageing, while inducing a decrease in the static rigidity and strength, leads to an improvement of the fatigue resistance (reflected by the decrease in the β parameter). This could be attributed to a better damage-tolerance induced by the swelling of the fibres and the potential increase in strength at the interface between the fibres and the matrix.

The typical stress-strain hysteresis loops are presented in Figure 6 for different life fraction values (5, 50 and 90%), for a given dynamic stress level (147 MPa), for aged-saturated and unaged specimens. For both conditioning, the mean strain increases equally as a function of the increasing number of cycles. This figure shows that the ageing induced three main effects: an increase in the mean strain, a decrease in the initial dynamic modulus and an increase in the dissipated energy, whatever the considered life fraction. These effects are consistent with the plasticizing of the material.

Figure 7 provides more details by representing, for the different loading levels, the evolution of the mean strain and of the dynamic modulus as a function of the normalized number of cycles. For the unaged specimens Figures 7a and 7c), the trends are in agreement with previous works [29]. The mean strain increases with the increasing number of cycles, reflecting the time-dependent behaviour of this type of PFCs and the fatigue-creep coupling

observed in the fibre direction in tension-tension fatigue tests. In the same way, the dynamic modulus increases as the number of cycles increases. This stiffening effect is dependent on the maximum stress level. As for the strain-hardening observed under monotonic testing, this stiffening under cyclic loading is attributed to the cellulose microfibrils reorientation in the flax fibre wall. The stiffening reaches a maximum value of 8%. Again, this result is in agreement with previous works [14, 29]. For these two quantities (mean strain and dynamic modulus), the evolution as a function of the number of cycles can be divided into three stages, for most of the loading levels. First, they significantly increase until a life fraction comprised between 0.05 and 0.1. Following this increasing step, they reach a second stage where they still increase but with a lower rate. The rate is all the more important that the stress is high. Lastly, just before failure (final fatigue stage), a sudden change is observed.

The increase in the mean strain could also be attributed to damage. If it does exist at the early stages of the lifetime, its effect is negligible in the fibre direction or compensated by the stiffening phenomena. In the latter stages, its effect is sometimes noticeable, traduced by a sudden drop in dynamic modulus.

For the aged-saturated specimens, the same trends are observed with a maximum stiffening of about 12%. Contrary to all expectations, it can be observed that despite the severe ageing conditions, and whatever the maximum stress level, the apparent rigidity of the aged-saturated specimens never decreases below its initial value during the whole fatigue tests. This result reflects the very good fatigue and damage resistance of this composite material (in the water-saturated state) even after a severe hydrothermal ageing.

Some of the aged-saturated specimens tested at the lower stress levels survived the maximum number of cycles pre-determined for the test. These specimens were then submitted to a

monotonic tensile test up to failure to determine their remaining properties after hydrothermal ageing and fatigue loading. Results are presented in Figure 4. For the considered specimens, the tensile properties remaining after 26 days of hydrothermal ageing at 70°C followed by $1.3 \, 10^6$ cycles at a maximum stress of 90 MPa in a water bath at ambient temperature are still high. A moderate decrease in the stress and strain at failure is induced by the fatigue loading. The reduction in these two quantities is approximately 18% and 27% respectively, when compared to aged-saturated only specimens. It can also be observed that the non-linear behaviour is more prominent when compared to the aged-saturated state. A very strong strain-hardening is observed with an apparent rigidity which varies from approximately 5 to 15 GPa during the monotonic tensile loading. The shape of the non-linearity is comparable to the one observed for the aged-dried specimens. So, these results highlight the existence of couplings between hydrothermal ageing, cyclic tensile loading and the non-linear tensile behaviour of UD flax-epoxy composites. These couplings and their origin at the microscale deserve to be investigated in further works.

4. CONCLUSIONS

There is a lack of data on the mechanical behaviour, and particularly fatigue behaviour, of PFCs after accelerated ageing. The objective of this manuscript was to provide fatigue data for a hydrothermal aged UD flax-epoxy composite tested in water-saturated conditions. For the first time, this type of material was fatigue tested in a water bath.

Results show that the hydrothermal ageing while inducing a decrease in the static rigidity and strength, leads to an improvement of the fatigue resistance. The material fatigue strength coefficient is almost divided by two after hydrothermal ageing when compared to unaged

specimens. It implies that, despite the difference in static strength, the S-N curve of the aged-saturated specimens converge toward the same maximum stress value, approximately 90 MPa, than for the unaged specimen, for a lifetime of approximately 5.3 10^6 cycles. Interestingly, it is also demonstrated that for this maximum stress level, the remaining properties after 1.3 10^6 fatigue cycles is still very high, with a tensile strength and an average apparent modulus of approximately 175 MPa and 10 GPa, respectively. So, despite the high sensitivity of this PFC to water, its resistance to ageing in quite severe conditions is good. Nevertheless, it could be improved by using coatings to prevent or limit water absorption when immersed or exposed to liquid water. The results of this study should be taken into consideration when designing bio-based composite structures and when predicting their lifetime and remaining properties.

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Table 1: Diffusion properties and cross- sectional areas variation (immersion at 70°C)

	Flax/epoxy composite	Pure epoxy
Water uptake at equilibrium (%)	11.9 ± 0.7	2.5 ± 0.1
Diffusion coefficient ($mm^2.s^{-1}$)	2.85 ± 0.37 10^{-6}	$2.78 \ 10^{-6}$
Thickness variation (%)	10.3 ± 0.9	1.8 ± 1.4
Width variation (%)	3 ± 0.2	1.4 ± 0.3

Table 2: Monotonic tensile properties measured before and after hydrothermal ageing (mean value ±standard deviation) at 2mm/min crosshead speed

Conditions	Tensile strength in fibre direction σ_L (MPa)	Tensile strain at failure in fibre direction ε_L (%)	Tensile strain at failure in transverse direction ε_T (%)	E_L^{m1} (GPa)	E_L^{m2} (GPa)	Poisson ratio ν_{LT} (-)
Unaged	288.1 ± 11.7	1.61 ± 0.08	0.77 ± 0.10	23.7 ± 0.2	17.3 ± 1.6	0.44 ± 0.01
Aged-saturated	210.8 ± 3.5	2.18 ± 0.05	1.02 ± 0.07	8.6 ± 0.2	10.5 ± 0.3	0.51 ± 0.03
Aged-dried	200.7 ± 35	1.29 ± 0.09	0.62 ± 0.05	17.7 ± 4.6	16.2 ± 4.9	0.64 ± 0.12

Figure 1: Aged and unaged specimens after fatigue tensile failure.
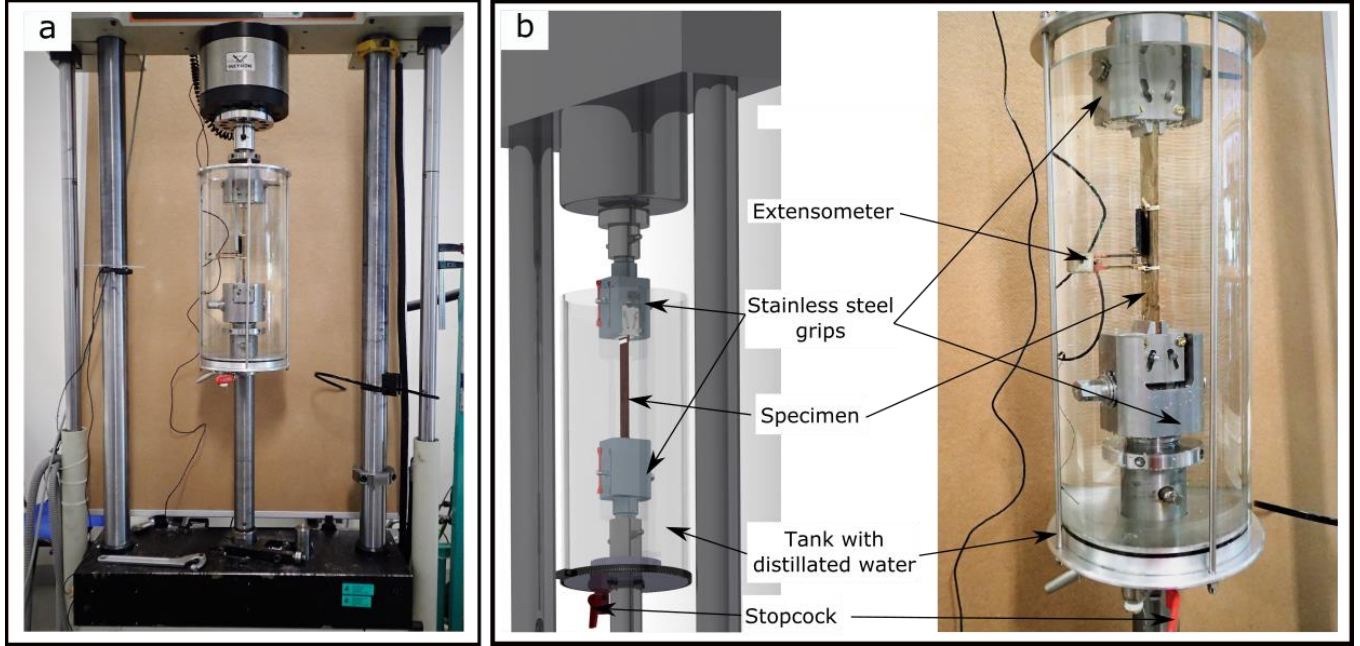
Figure 2: In-house test set-up for immersed monotonic and cyclic tensile loading of composite specimens. a: global view of the tensile test machine with the set-up, b: 3D view and zoom on the tank and grips.
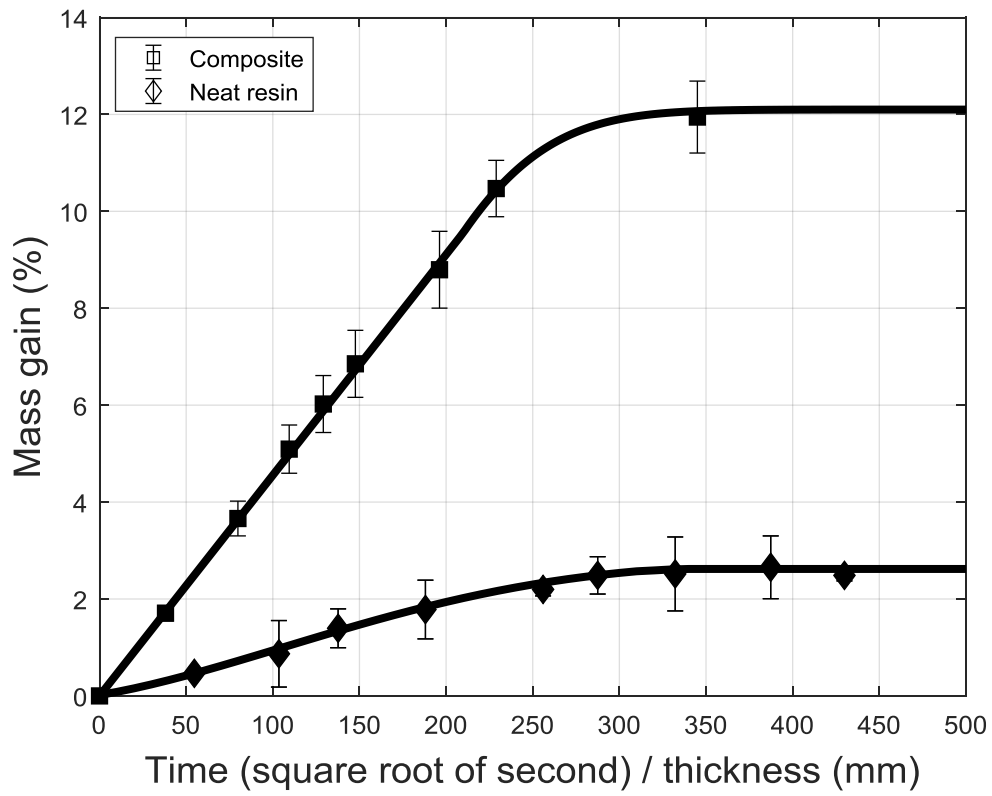
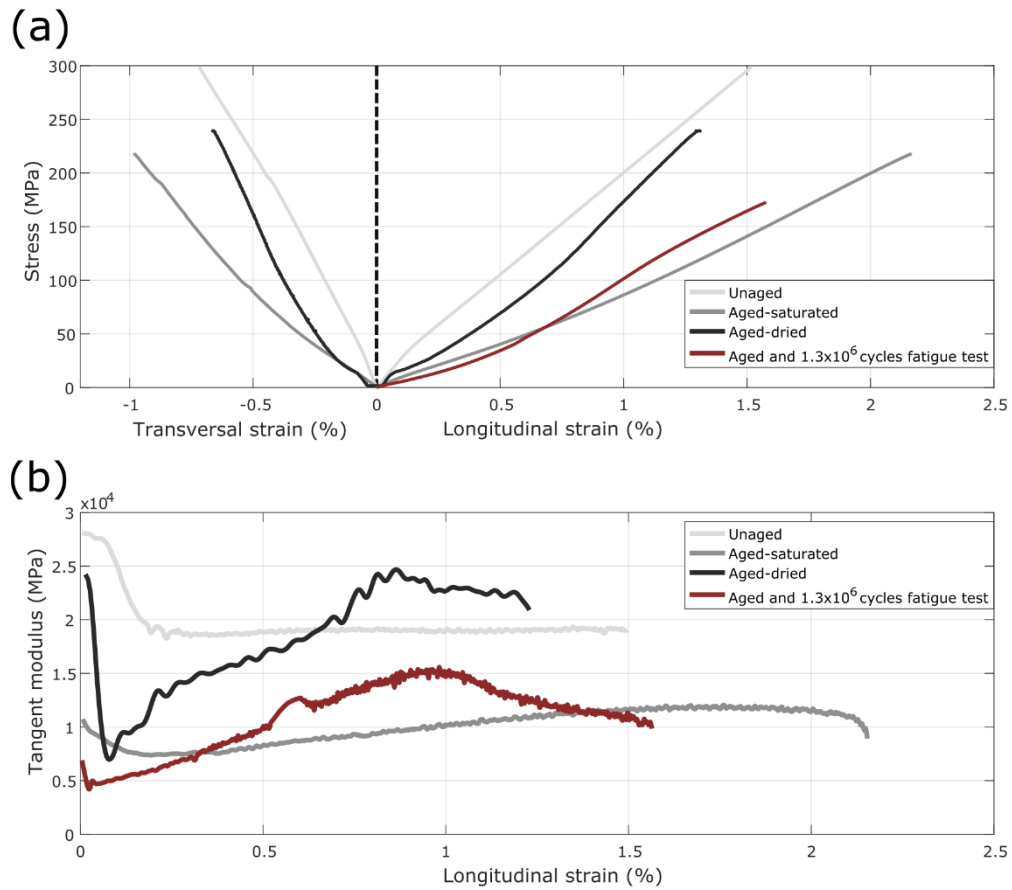**Figure 3: Evolution of water uptake of composite and neat resin samples when immersed in distilled water at 70°C**

Figure 4: Typical stress-strain and tangent modulus-strain curves of UD laminates tensile loaded in the fibre direction, before ageing (unaged specimen), after ageing (aged-saturated and aged-dried specimens) and after ageing and fatigue testing (1.3×10⁶ cycles).
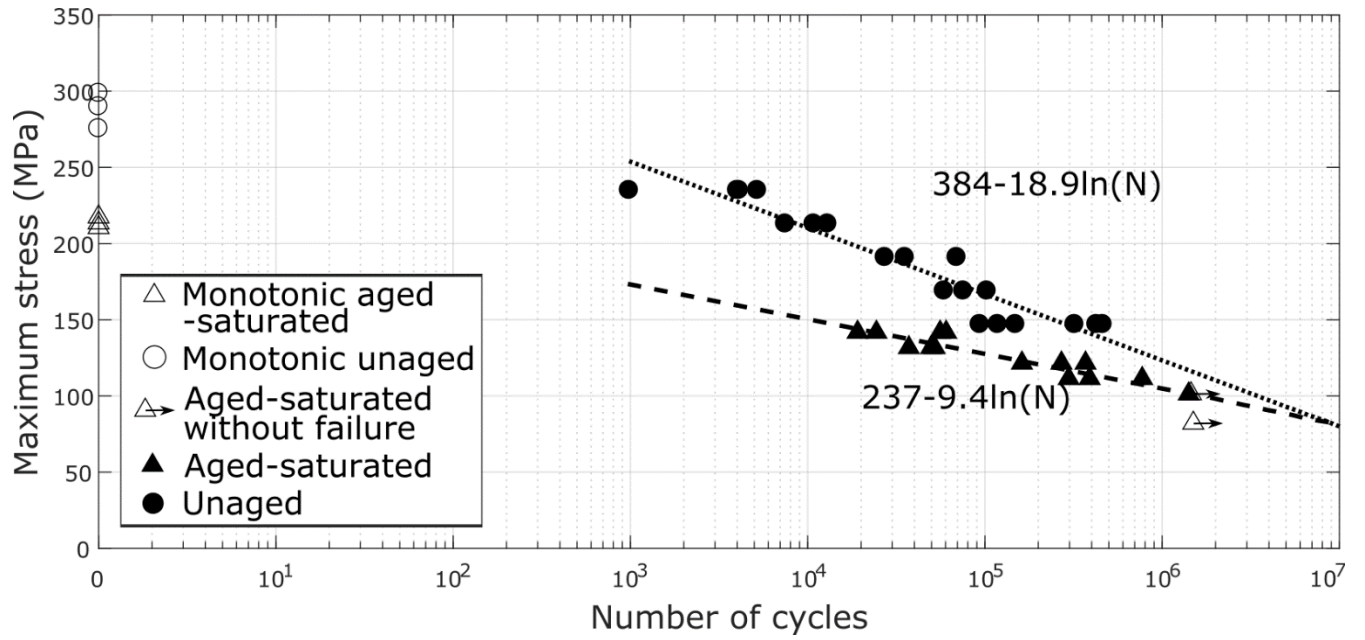
Figure 5: S-N curves collected before ageing (unaged) and after ageing (aged-saturated)
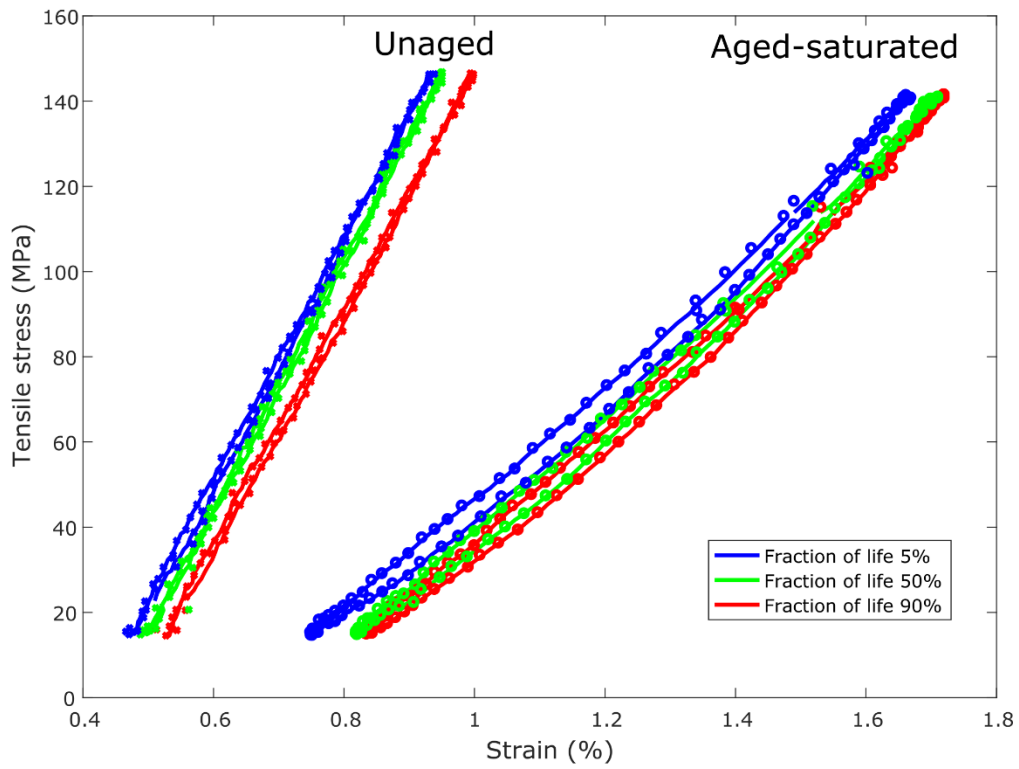
Figure 6: Typical hysteresis loops before and after hydrothermal ageing at different lifetime fractions
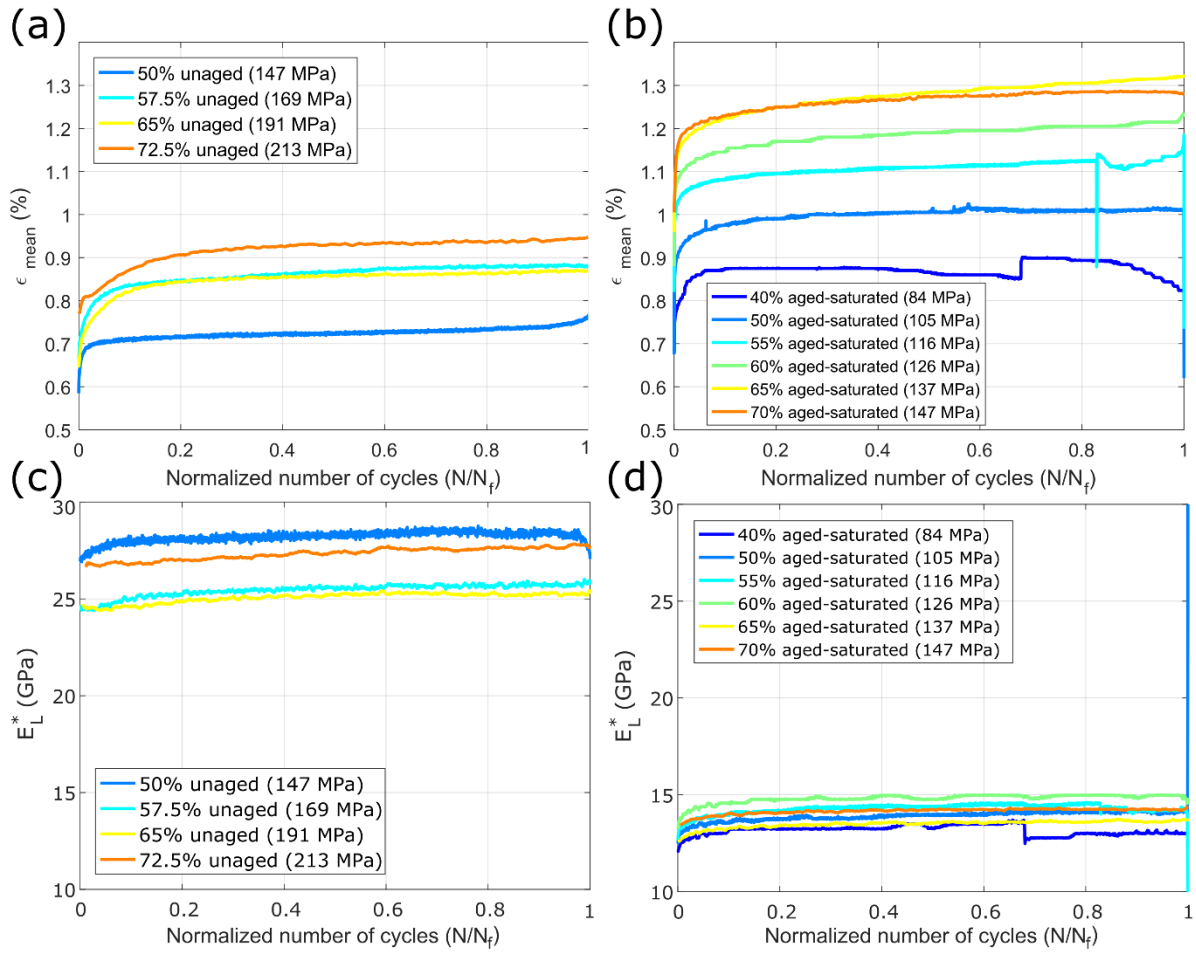
Figure 7: Typical curves of axial mean strain, and dynamic elastic modulus recorded during fatigue tests as a function of normalized number of cycles, for different maximum stress levels, before (a, c) and after (b, d) hydrothermal ageing.