Lifetime prediction of flax fibre reinforced composites

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ABSTRACT: Flax fibre reinforced composites are demonstrating promising outcomes which makes them potential candidates to replace synthetic composites in various industrial applications. However, there is limited information regarding their long-term performance, and it is usually acknowledged that natural fibres are less resistant than their synthetic counterparts. In this context, it is crucial to study their durability before considering their use for structural rehabilitation and strengthening in construction. This study, lying within the framework of the National Research Agency project MICRO, aims to study and predict the performance of flax fibre reinforced polymer (FFRP) composites with a bio-based epoxy matrix. The test program consists in exposing FFRP laminates and FFRP strengthened concrete blocks to different accelerated ageing conditions over a total period of 2 years, and with various combinations of temperature and relative humidity in the ranges 20°C-60°C and 50%-100% RH, respectively. Then a series of tensile, short beam and pull-out tests are performed periodically on aged samples to evaluate property evolutions over ageing time. Finally, collected experimental data are analysed using statistical tools, in view of developing a performance evolution model and evaluate the service lifetime performance of this new bio-based composite.

1 INTRODUCTION

For the time being, flax fibre reinforced polymer (FFRP) composites are still in a research and development phase but are considered a highly promising solution for the future. With their low carbon footprint and high mechanical properties to weight ratio, FFRPs are poised to be a viable replacement for the traditional synthetic composites in specific industrial applications. This transition is due to the shift in international interest towards recyclable and bio-sourced materials and lowering carbon emissions (Yan et al., 2014). However, a major challenge in using natural fibres, remains their sensitivity to environmental conditions, and more specifically in the presence of water (Le Duigou et al., 2010; Scida et al., 2013). Numerous studies have investigated the effect of humidity on the mechanical behaviour of FFRPs, but until now, there are still unanswered questions related to the lifetime performance prediction of FFRP and the coupling effects of temperature and humidity on the ageing behaviour of these materials.

In civil engineering applications, it is crucial for engineers to get knowledge about the long-term performance of materials used and not just their short-term characteristics. This extended performance is highly dependent on service conditions in the outdoor environment such as temperature, humidity, uv light exposure, alcalinity of concrete… This study focuses on the effects of temperature and humidity, considered as major factors influencing the long-term durability of FRPs and more specifically bio-sourced composites given the hydrophilic nature of their fibers (Benzarti et al. 2009, 2011; Karbhari & Abanilla, 2007; Quiertant et al., 2017). An emphasis is made on the influence of these two factors on the mechanical properties of FFRP laminates and their adhesive bond with concrete, and on the development of a performance evolution model to predict its delayed performances.

2 MATERIALS AND METHODS

2.1 Materials and ageing conditions

Unidirectional flax/bio epoxy composites, consisting of 2 layers of 200g/m^2 flax fibre fabrics, were manually prepared using the hand layup technique. The flax fibre fabrics used in this study were produced by Groupe Depestele, a French natural textile company. The epoxy resin (CHS-EPOXY G520) was supplied by Spolchemie, a Czech chemical company known for its environmentally-friendly products. This resin (30% bio-sourced) was mixed with a 100% bio-sourced amine hardener in stoichiometric proportions. The final FFRP laminates had a fibre volume fraction of around 16% and were cured in the laboratory conditions {20°C/35-50% RH} for 3 weeks until stabilization of the polymerization process.
In addition, FFRP reinforced concrete blocks were also prepared for adhesive bond tests. Concrete blocks were first cast using a ready-to-mix commercial mixture of compressive strength 50 MPa at 28 days. The blocks were stored for 90 days before strengthening of the upper face with a single ply of UD flax fabric impregnated by the bio-epoxy matrix. As previously, a 3-week cure was achieved before starting exposure tests.
All these specimens (laminates and reinforced blocks) were then divided into 7 series that were placed in various environments, corresponding to either accelerated ageing conditions or outdoor weathering conditions (see Table 1). During the test program, specimens were periodically removed from the ageing environments in order to be mechanically tested.

Table 1. Ageing conditions selected for the design of experiments

Name	Temperature	Humidity	Test Schedule
V1	20°C	50% RH (climatic chamber)	3, 6, and 12 months
V2	20°C	100% (immersion in water)	3, 6, and 12 months
V3	60°C	50% RH (climatic chamber)	3, 6, and 12 months
V4	40°C	100% (immersion in water)	3, 6, and 12 months
V5	60°C	75% RH (climatic chamber)	3, 6, and 12 months
V6	60°C	100% (immersion in water)	3, 6, and 12 months
VN	Outdoor exposure in Lyon, France		12 months

2.2 Tensile testing procedure

Direct tensile tests were carried out according to NF EN ISO 527 standard and French AFGC guidelines in the longitudinal direction of the FFRP laminates. Glass fibre composite tabs were glued to each extremity of the specimens with an epoxy adhesive, in order to improve the grip during tensile tests. An Instron 5969 universal testing machine, equipped with a contact-free Advanced Video Extensometer (AVE), was used to apply the loading speed of 1 mm/min, as advised in the standard method. Instead of calculating a tensile strength considering the actual section of sample (which is very dependent on the quality of impregnation of the laminate), one determines a maximum tensile force (or tensile capacity) per unit width of fabrics according to the following equation (ASTM D7565/D7565M):

$$\text{Maximum tensile force per unit width (kN/m)} = \frac{\text{Force at failure}}{\text{sample's width} \times \text{number of layers}}$$

For FFRP, the tensile stiffness is the slope of the curve tensile force per unit width – strain when the strain ranges between 0.3 and 0.5%.

$$\text{Tensile stiffness (in kN/m)} = \frac{\Delta\ \text{Applied tensile force}}{\Delta\ \text{strain}}$$

2.3 Short beam testing procedure

Short beam 3-point-bending tests were carried out according to NF EN ISO 14130 standard. Samples were cut into small plates of FFRP laminates (2 layers) with base dimensions of 10mm x 20mm. An Instron 5969 universal testing machine equipped with a 3 point-bending setup was used to apply the compressive loading speed of 1 mm/min as advised in the standard method. To calculate the interlaminar shear stress τ, the following equation was used:

$$\text{Interlaminar shear stress } \tau = \frac{3}{4} \times \frac{\text{Failure load}}{\text{width} \times \text{thickness}}$$

2.4 Pull-off testing procedure

Pull-off tests were performed according to EN 1542 / AFGC standards. The single FFRP layer reinforcing each concrete block was first drilled using a cylindrical core drill of diameter 50 mm, until reaching a depth of 4 mm within the concrete substrate. A cylindrical steel disc of diameter 50 mm was then glued to the drilled zone using an epoxy adhesive. Finally, a tensile load was applied to the disc at constant speed of 0.05 MPa/sec using a Proceq DY-216 dynamometer until failure occurred, leading to the determination of the bond strength and failure mode.

3 EXPERIMENTAL RESULTS

Mechanical tests were first carried out on unaged specimens, providing the initial reference performances before ageing. Samples were then tested after 3, 6, and 12 months ageing in the various conditions. Exposures up to 24 months are still underway and are not reported here.

3.1 Tensile properties

Tensile tests performed on unaged FFRP samples have established reference values of 87 kN/m for the maximum tensile force per unit width of fabrics and 7350 kN/m for the stiffness per unit width. These reference values were used afterwards to normalize the residual performances of the aged specimens. Figure 1 presents the normalized residual tensile properties of the FFRP laminates up to 12 months of ageing.

FFRP composites did not show any significant degradation of their tensile capacity per unit width for the non-immersed conditions after 12 months ageing (Fig. 1.a). An increase was even observed for specimens exposed to the temperature of 60°C, which can be explained by the post-curing process of the bio epoxy matrix (confirmed by complementary DSC analyses). Differently, specimens immersed in water show a 20% reduction of their residual tensile capacity per unit width after 12 months, with a kinetics that is dependent on the ageing temperature (the higher the temperature, the higher the degradation kinetics). Specimens exposed to outdoor conditions (VN) for 12 months exhibit a 13% increase in tensile capacity per unit width compared to the reference, also attributed to the post-cure effect.

However, in the case of immersed specimens, the tensile stiffness per unit width was significantly affected after 12 months ageing, with decreases up to 40% at 60°C-100% RH (Fig 1.b). This phenomenon was assigned to a plasticization of the polymer matrix induced by water.
Unlike the immersed samples, specimens exposed to outdoor conditions for 12 months did not exhibit significant decrease in tensile stiffness compared to the reference (variation of 4%). This result suggests that immersion conditions are very severe compared to natural ageing.

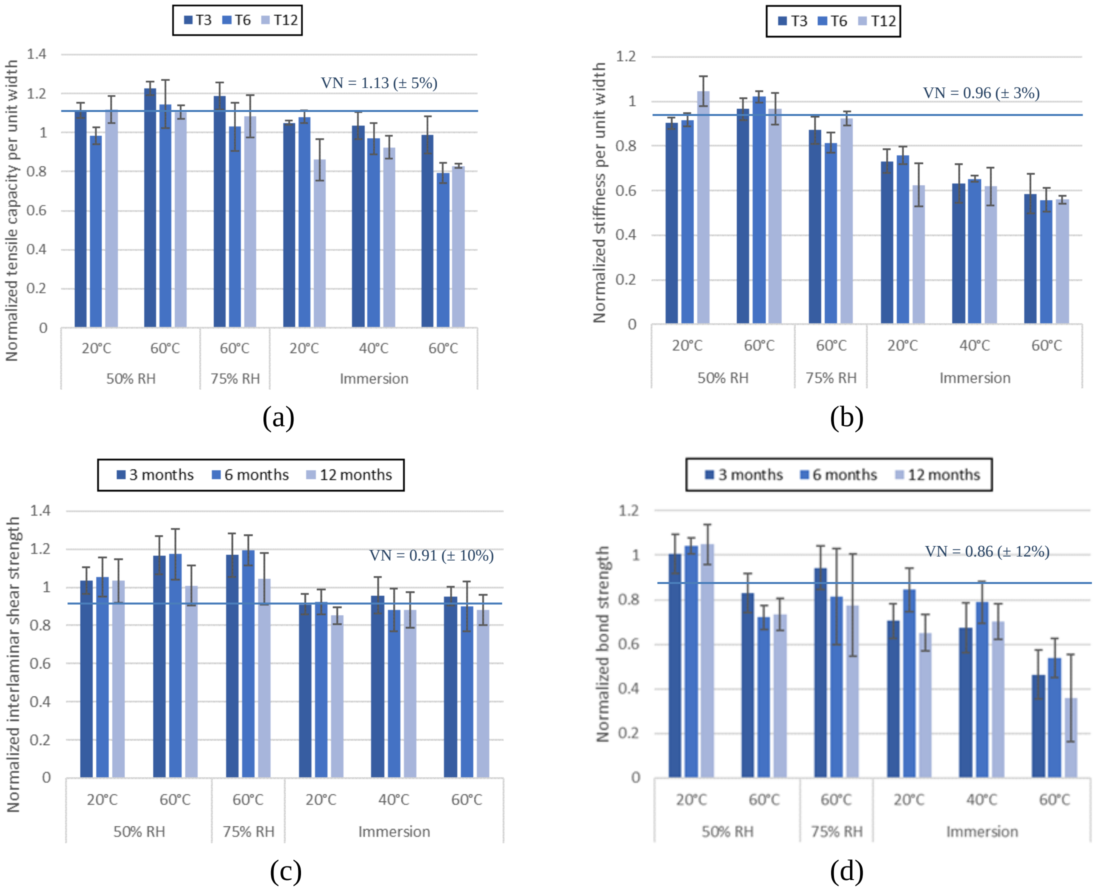

Figure 1. Normalized results of mechanical tests (a) Maximum tensile force per width unit (b) Tensile stiffness per width unit (c) Interlaminar shear strength (d) Pull-off bond strength, for specimens exposed up to 12 months.

3.2 Interlaminar properties

Short beam tests were also carried out first on unaged FFRP coupons, providing reference values of 8.5 ± 0.8 MPa for the initial interlaminar shear strength (ISS). Tests were then performed on aged specimens and their residual properties were normalized by the reference value. Figure 1.c shows that, like for the tensile capacity, no significant decrease in ISS was observed over time. An increase in ISS can be detected again for specimens exposed to the temperature of 60°C at 50% and 75% RH, which was linked to the post-curing effect.
Specimens exposed to outdoor conditions for 12 months exhibit a slight decrease in ISS (about 9%) compared to the reference samples.

3.3 Adhesive bond properties

Pull-off tests performed on unaged specimens gave a reference value of the pull-off strength equal to 4.1 ± 0.3 MPa with a typical cohesive concrete failure (Figure 2.a). After ageing, a

significant decrease in bond strength was observed for all specimens, except for those subjected to 20°C-50% RH (Figure 1.d). Samples directly immersed in water were the most affected, with a reduction up to 50% in the case of samples immersed at 60°C. Such a degradation of the bond strength under wet conditions was accompanied by a change in failure mode, from an initial cohesive concrete failure towards a mixed (Figure 2.b) or FRP debonding mode (Figure 2.c). This change in failure mode and the reduction in bond strength can both be attributed to a weakening of physico-chemical bonds at the concrete/composite interface in presence of water.
Specimens exposed to outdoor conditions for 12 months show only limited reduction of their pull-off strength (around 14%), together with a mixed failure mode similar to Figure 2-b. It suggests that outdoor exposure is less severe than accelerated ageing by immersion in water.


(a) (b) (c)

Figure 2. Failure modes (a) Cohesive concrete failure (b) Mixed failure- debonding and cohesive concrete failure (c) Debonding of the laminate

4 LIFETIME PREDICTION

Previous researchers have proved that the evolution of FRP performance can be expressed in the form of P(t) = A + B x ln(t) where P is the mechanical performance, A and B are constants, t is the time and ln refers to Napierian logarithm (Karbhari & Abanilla, 2007; Silva et al., 2014). However, in this study the objective is to introduce the temperature and the humidity in the model, and to take into account their coupling effect on the long-term performance of the FFRP composites as well. Therefore, a specific performance evolution model was constructed as a function of temperature, humidity and time:

$$P(t, T, H) = a_1 + a_2 \times \ln(AF(T, H) \times t^{a_3}) \quad (1)$$

Where P(t, T, H) is the performance at the considered time t, temperature T, and humidity H; a_1, a_2, and a_3 are constants; and AF is the accelerating factor written as:

$$AF(T, H) = b + b_T T + b_H H + b_{T-H} TH + b_{T-T} T^2 + b_{H-H} H^2 \quad (2)$$

The primary and quadratic influence of temperature and humidity as well as their coupling are implemented in the accelerating factor AF(T, H). The coefficients (a_1, a_2, a_3, b, b_T, b_H, b_{T-H}, b_{T-T}, b_{H-H}) are calculated from experimental data using nonlinear regressions, and their values determined for the various performance indicators are reported in Table 2.

Table 2 Coefficients of the performance evolution model identified for the considered mechanical properties

	Tensile capacity per width unit	Tensile stiffness per width unit	Interlaminar shear strength	Bond Strength
a_1	1.143728	0.933409	1.436329	0.923282
a_2	0.801560	0.529093	0.592745	0.751270
a_3	-0.090025	-0.004122	-0.100433	-0.050636
b	0.226989	0.519393	-0.357632	-0.622291
b_T	0.021020	0.005961	0.004402	0.013295
b_H	0.019563	0.017691	0.028683	0.055331
b_{T-H}	-0.000127	-0.000100	-0.000038	0.000058
b_{T-T}	-0.000146	0.000006	-0.000004	-0.000318
b_{H-H}	-0.000133	-0.000157	-0.000204	-0.000432

Considering the expression of AF, the performance evolution model P(t, T, H) can also be written in the form of P(t, T, H) = A(T, H) + B x ln(t):

$$P(t,T,H) = a_1 + a_2 \times \ln(AF(T,H) \times t^{a_3})$$
$$P(t,T,H) = a_1 + a_2 \times a_3 \times \ln(AF(T,H)^{1/a_3} \times t)$$
$$P(t,T,H) = a_1 + a_2 \times a_3 \ln(AF(T,H)^{1/a_3}) + a_2 \times a_3 \times \ln(t) \tag{3}$$

Where A(T, H) = $a_1 + a_2 \times a_3 \ln(AF(T,H)^{1/a_3})$ and B = $a_2 \times a_3$.

It is crucial to note that, by modelling the mechanical performance as a function of ln(t), we force the model to strictly decrease with time. But in reality, experiments show that there is an initial increase in performance due to the post-curing of the polymer matrix, especially in high temperature environments. As the model is unable to describe this initial improvement, it predicts normalized performances superior to 1 at the early ageing times. To overcome this unexpected artefact, it is thus proposed to assign the value of 1 to all theoretical data greater than 1, while retaining the values inferior or equal to 1. This method is illustrated in Figure 3.a (dashed line) for the tensile capacity of specimens subjected to ageing conditions V4. The proposed degradation model is then applied to predict the evolutions of the tensile properties and ISS for FFRP laminates subjected to the various ageing conditions, and the evolution of the pull-off strength of strengthened slabs as well (Figs. 3b to 3.c)

Error percentages between theoretical and experimental values of performance indicators are presented in Table 3 for aged specimens. This comparison shows globally a fair agreement and suggests the degradation model catches well the coupled effects of temperature and humidity.

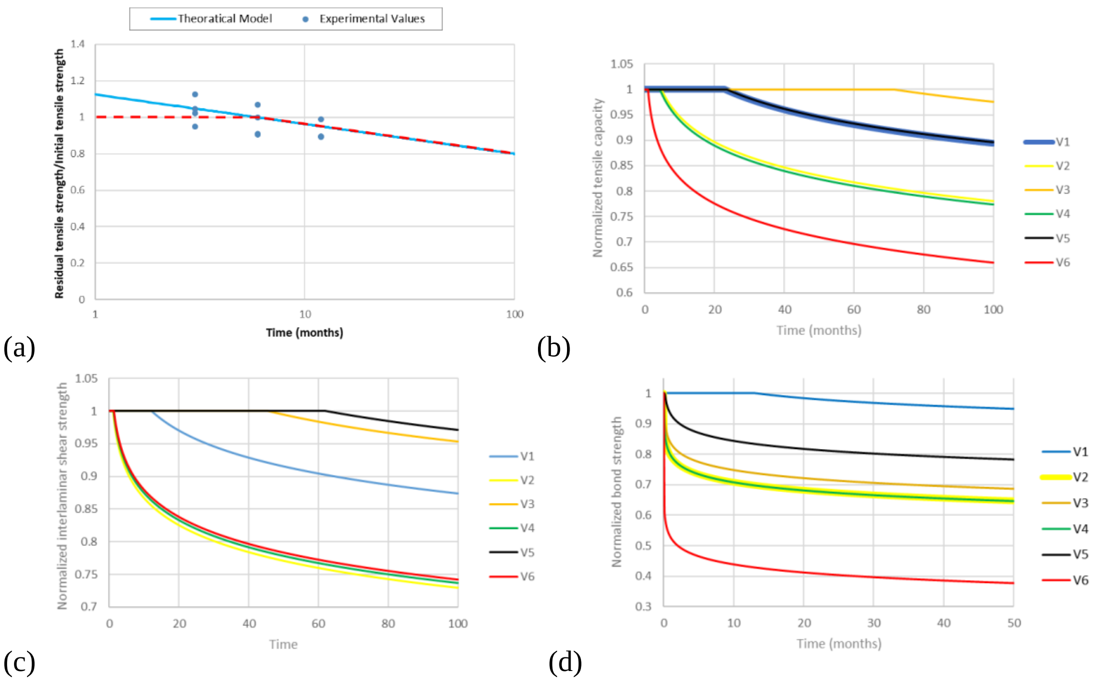

Figure 3. Predicted evolutions of mechanical performances (a) Example of model correction due to post curing (for tensile strength under V4 conditions) (b) Tensile strength (c) Interlaminar shear strength (d) Bond strength

To go further and predict the lifetime of FFRP laminates and their adhesive bond with concrete, it is then necessary to state an end-of-life criterion that should be considered to ensure safe functioning of the FRP. In the present approach, this criterion was defined as the maximum acceptable value of the degradation rate of mechanical performances. Considering that the

purpose of this paper in not to suggest the value of such threshold but rather to illustrate how to use the model when an end-of-life criteria is specified, a maximum reduction of the mechanical performances of 15% was arbitrarily chosen as example in the following.
Minimum allowable performances were then calculated according to Equation (4), considering the mean experimental values determined on unaged specimens and their standard deviation:

$$\text{Minimum allowable performance} = 85\% \times (\text{Initial mean performance} - 3 \times \text{standard deviation}) \qquad (4)$$

After calculation of the minimum allowable performances, the previous degradation model was used to estimate the time needed to reach these lower limits. Calculated lifetimes are reported in Table 4, based on the simulated evolutions of the various performance indicators.

As expected, FFRP laminates immersed in water show much shorter lifetimes compared to non-immersed samples. In addition, the bond strength seems to be the most critical performance indicator, as it provides the smallest lifetimes in the various environments.

Table 3 Error percentage between theoretical and experimental values

	Time (months)	V1	V2	V3	V4	V5	V6
Tensile capacity per width unit	3	2.9%	-1.6%	0.2%	-0.8%	-3.1%	-7.5%
	6	11.5%	-8.9%	3.0%	0.7%	6.8%	8.9%
	12	-6.3%	8.5%	2.2%	0.3%	-3.2%	-2.0%
Tensile stiffness per width unit	3	5.1%	-5.2%	1.0%	-0.5%	-0.2%	-4.8%
	6	3.2%	-8.7%	-4.6%	-4.0%	6.5%	-0.3%
	12	-9.6%	10.7%	0.7%	1.2%	-6.3%	-0.7%
Interlaminar shear strength	3	4.7%	3.0%	-0.5%	-1.2%	1.0%	-0.2%
	6	-1.2%	-2.8%	-4.6%	2.7%	-4.6%	1.1%
	12	-3.2%	0.4%	7.0%	-1.9%	5.2%	-1.5%
Bond strength	3	5.0%	7.0%	-4.5%	12.1%	-5.8%	4.6%
	6	-1.1%	-13.9%	6.6%	-7.8%	6.0%	-14.7%
	12	-4.4%	7.5%	0.8%	-0.1%	7.9%	21.0%

Table 4 Lifetime prediction under various ageing conditions

		Performance Indicators			
		Tensile capacity	**Tensile stiffness**	**ISS**	**Bond strength**
Mean reference value		87 kN/m	7350 kN/m	8.5 MPa	4.1 MPa
Standard deviation		6.6 kN/m	645 kN/m	0.8 MPa	0.25 MPa
Minimum allowable performances (= end-of-life criterion)		57 kN/m	4603 kN/m	5.2 MPa	2.84 MPa
Time to reach the end-of-life criterion (in years)	V1	>100	>100	>100	>100
	V2	47.7	>100	61.6	1.2
	V3	>100	>100	>100	3.5
	V4	47.7	0.7	70.7	1.2
	V5	>100	>100	>100	42.1
	V6	8.9	<0.25	76.5	<0.25

5 CONCLUSIONS

This paper has presented a lifetime prediction approach based on a performance evolution model, which was calibrated from a comprehensive experimental test program. FFRP laminates and FFRP strengthened concrete blocks were subjected to various accelerated ageing conditions and their mechanical performances (tensile properties, ISS and bond strength with concrete) were monitored over a period of 12 months.

A plasticization phenomenon was observed for samples immersed in water, leading to a reduction in mechanical performances of the FFRP. The tensile capacity showed a decrease reaching 20% whereas the tensile stiffness was more severely affected with decreases up to 40% compared to unaged reference specimens. FFRP/concrete interface was also negatively affected by the presence of water. A decrease in bond stress was observed as well as a change in failure mode from a cohesive concrete failure to a debonding of the FFRP laminate.

A post-curing effect was observed for non-immersed specimens exposed to high temperatures with an increase in their tensile capacity and interlaminar shear strength.

Concerning samples subjected to natural ageing in outdoor environment limited variation of mechanical performances was observed after 1 year of exposure apart from the bond stress where a decrease of 14 % was detected. Globally, outdoor exposure condition was found less severe than the considered accelerated ageing environments.

Finally, a performance evolution model was constructed to describe the performance evolutions of the laminates, based the treatment of collected experimental data. It takes into account the effects of temperature and humidity, as well as their coupling. Finally, this degradation model was applied to predict lifetimes of FFRP composites under the different ageing conditions, by assessing the time necessary to reach the imposed criterion of 15% maximum degradation on the mechanical performances. Results showed that the adhesive bond strength between the FFRP and concrete is the most affected property and governs the lifetime of the composite.

6 ACKNOWLEDGMENTS

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7 REFERENCES

Benzarti, K., Argoul, P., Freddi, F., Frémond, M., & Nguyen, T. H. T. (2009). Durability of bonded assemblies. European Journal of Environmental and Civil Engineering, 13(9), 1141–1151.

Benzarti, K., Chataigner, S., Quiertant, M., Marty, C., & Aubagnac, C. (2011). Accelerated ageing behaviour of the adhesive bond between concrete specimens and CFRP overlays. Construction and Building Materials, 25(2), 523–538.

Karbhari, V. M., & Abanilla, M. A. (2007). Design factors, reliability, and durability prediction of wet layup carbon/epoxy used in external strengthening. Composites Part B: Engineering, 38(1), 10–23.

Le Duigou, A., Davies, P., & Baley, C. (2010). Interfacial bonding of Flax fibre/Poly(l-lactide) bio-composites. Composites Science and Technology, 70(2), 231–239.

Quiertant, M., Benzarti, K., Schneider, J., Landrin, F., Landrin, M., & Boinski, F. (2017). Effects of Ageing on the Bond Properties of Carbon Fiber Reinforced Polymer/Concrete Adhesive Joints: Investigation Using a Modified Double Shear Test. Journal of Testing and Evaluation, 45(6).

Scida, D., Assarar, M., Poilâne, C., & Ayad, R. (2013). Influence of hygrothermal ageing on the damage mechanisms of flax-fibre reinforced epoxy composite. Composites Part B: Engineering, 48, 51–58.

Silva, M. A. G., da Fonseca, B. S., & Biscaia, H. (2014). On estimates of durability of FRP based on accelerated tests. Composite Structures, 116(1), 377–387.

Yan, L., Chouw, N., & Jayaraman, K. (2014). Flax fibre and its composites - A review. Composites Part B: Engineering.