EFFECT OF UPTAKE BEHAVIOUR ON TENSILE PROPERTIES OF FLAX FIBRE REINFORCED COMPOSITES

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Abstract

This study concerns the investigation of a biocomposite solution made with flax fibres to reduce the weight of structural parts used in medical environment. Besides the mechanical specifications, this composite solution is requested to particular environmental conditions, and distinctively its response to some specific cleaning solutions (Surfanios® and Terralin®) *must be characterized. Vacuum moulded Flax/PLA and Flax/PP samples were made, and samples of each material has been subjected to immersion test for different concentration of cleaning solutions and distilled water. Weight measurements as well as tensile tests were performed in order to follow the evolution of the physical and mechanical behaviour of the composites.*

1 Introduction

Due to environmental and ecological issues, natural fibre composites are being more and more investigated as a potential solution to replace some existing glass fibre composites application. Indeed, their mechanical properties have been demonstrated to be comparable to glass reinforced composites [1]. For instance, their applications in different industries are expanding, notably in the automotive industry. Furthermore, natural fibres have the intrinsic advantage of being renewable and their use in combination with thermoplastic biopolymers result in a recyclable or biodegradable biocomposite [2]. However, in spite of these promising properties, their response to some specific environmental conditions needs to be investigated.

In a medical environment, the parts are subjected to various cleaning products such as germicides and fungicides. Few written studies focus on the ageing of natural fibre composites with such products. Research has been done on several biocomposites based on sisal, flax, hemp fibres associated to either PLA, PP or epoxy matrix and exposed to distilled water or seawater [3-6]. The absorption of water at room temperature conditions was shown to be Fickian, [3-5] and otherwise not Fickian if the damage in the material is particularly important [7]. Non-Fickian behaviour was also observed on woven reinforced composite [17] and other models have been developed to characterise these behaviour [18]. It has also been shown that the nature (hydrophobic or hydrophilic) of the used matrix affects the uptake behaviour and then the damage of the composite [8]. A hydrophilic matrix will enhance the uptake through the capillarity of flax fibres and results in a higher damage in the interfacial fibre/matrix zone.

Undeniably, the product uptake has an effect on the mechanical behaviour of the composite and thus very often the immersion tests are coupled with tensile tests in order to show the effects on the Young's modulus, the tensile stress and impact resistance [2-4,7,9,10]. Indeed, the Young's Modulus and the tensile stress are affected with water absorption and tend to decrease with immersion time [2,3,5,7,11]. Although, the drying of the composite shows a recover of these mechanical properties [5] the tendency to decrease remains.

In this current study, a biocomposite solution is being investigated for an application in a medical environment. The part to be designed has to fulfil requirements which include a loading case and the compatibility with the usual cleaning products used in the medical environment (Water, Surfanios® and Terralin®). Our selection of renewable resources based components are woven flax fibre as reinforcement and PLA (poly-lactic-acid) as matrix. A secondary choice of petroleum based matrix, the PP (polypropelene), has been made in order to compare its performance with the PLA matrix. How the mechanical properties are influenced by the environment needs to be addressed and the present work focuses on this aspect. In order to understand the influence of the environment on the composites behaviour, immersion tests and tensile tests were performed.

2 Materials and testing methods

2.1 Materials

Flax woven reinforcements used were commingled with either PLA or PP. The weaving architecture for both reinforcements was a 2*2 twill with a fibre fraction of 40%. The fabric weight is 420 g/m² for the flax/PLA and of 430 g/m² for the flax/PP.

2.2 Samples manufacturing

The woven reinforcements were cut in squares of 360x360 mm² and stacked up in the same direction in 6 layers. The fabric layers were placed between two steel plates and sealed for standard vacuum moulding. The whole set-up was then placed in an oven and connected to a vacuum pump (-0.9bar) and undergone a temperature cycle; first a heating rate of 5°C/min until 200°C, then a holding temperature of 200°C during 40min and a cooling rate of 5°C/min down to room temperature. The measured thickness of the manufactured plates is 2.07mm for the flax/PLA composite and 2.66mm for the flax/PP composite.

Two types of specimens were machined after the manufacturing of the composite plates. Samples for product uptake and physical properties evolution were square in shape. For the mechanical tensile tests, specimens were prepared according to the ISO 527-4 standard [13].

2.3 Immersion tests

Three cleaning solutions were used; distilled water, Surfanios® and Terralin®. The concentrations for Surfanios® and Terralin® were of 0.25% and of 0.5% in volume respectively and the products were renewed weekly during immersion tests. All specimens prepared for immersion test were characterised in their dry state before undergoing any immersion. The dry state was taken after 24h at 105°C in the oven.

Once the specimens were dried, their initial anhydrous mass, M_0 and thickness were measured. They were then immersed in the cleaning solutions. They were removed at different time intervals and their mass M^i measured in order to monitor the apparent weight gain (Eq. 1), and then replaced in solutions. Other specimens were taken out, then dried and their anhydrous mass M_0^i measured to monitor the mass loss (Eq 2.), at different time and were not replaced in the solutions. The precision of the balance used was of 0.1 mg.

$$\text{Apparent weight gain (\%)},\ M_t = \frac{M^i - M_0}{M_0} * 100 \qquad (1)$$

$$\text{Mass loss (\%)} = \frac{M_0^i - M_0}{M_0} * 100 \qquad (2)$$

2.4 Tensile tests

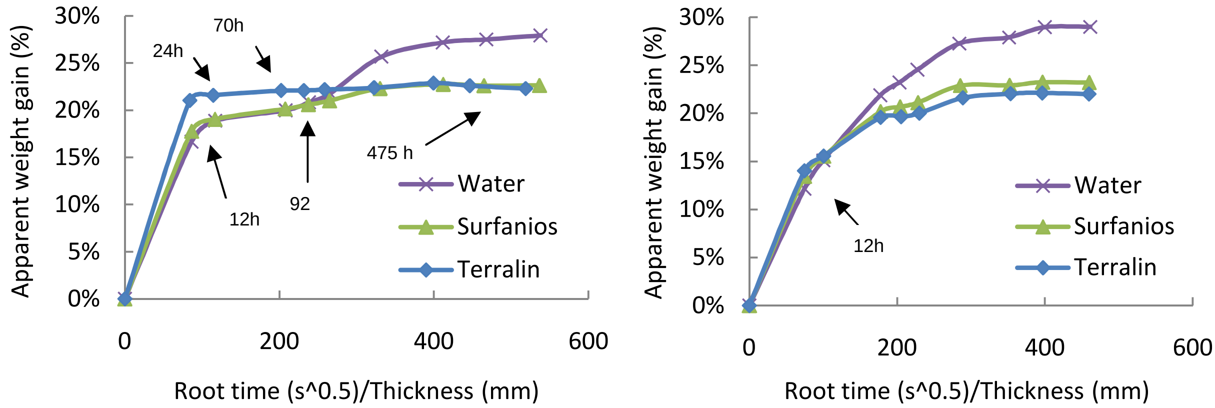
Tensile tests were performed on an INSTRON universal tensile machine 4507, with a 10kN load capacity. The samples were loaded at a displacement rate of 2mm/min according to the ISO 527-1 standard [12]. The strain measurements were carried out using a marker tracking method [14-15]. Specimens were characterised in the wet state and in the dry state. The dry state was taken at equilibrium with the room standard conditions after a drying cycle in the oven. Tests on dry specimen were done to investigate the permanent changes occurring during ageing.

3 Results and discussion

3.1 Uptake and physical measurement

Figure 1 shows the evolution of the apparent weight gain, M_t, of the two composites. The uptake rate of the flax/PP composite seems to be Fickian. On the other hand, the uptake of the flax/PLA composite is more complex. Indeed, the flax/PLA samples immersed in Terralin® almost reached equilibrium after 12 hours of immersion whereas the samples immersed in water and Surfanios® reached pseudo-equilibrium at the same time before rising (after 92 hours of immersion time) to reach the final equilibrium state after 475 hours of immersion time. Keeping in mind that the apparent weight gain measures two simultaneous mechanisms occurring: the product uptake and the mass loss, the presence of this pseudo-equilibrium for the flax/PLA sample can be explained by the evolution of the mass loss curve (figure 2). Most of the absorption is occurring during the first 12 hours while the mass loss is relatively small [8]. After that period, the absorption rate is lower. Between 24 and 92 hours of immersion is when the most of the mass loss occurs. During that time interval, the absorption of the product (in mass) must be slightly higher than the mass loss resulting in the pseudo-equilibrium state on the apparent weight gain curve. After 92 hours, the mass loss is almost constant and the apparent weight gain rises again as diffusion is still taking place. With relatively small mass loss (figure 2), this phenomenon is not observed for the flax/PP composites and for the flax/PLA sample immersed in Terralin®.

During the first 12 hours of immersion, the uptake rate of Terralin® is higher and that of water is lower for both composite (expressed by *k* in table 1 which is the slope of the linear part of the weight gain curve). This tendency is then inverted as immersion time increases. For both composites, the uptake at equilibrium with water clearly distinguishes itself as compared to the two other cleaning products. This underlines the effect of the addition of the cleaning products on the diffusion within the composites.



(a)Flax/PLA (b)Flax/PP

Figure 1. Product uptake of the composites.

The influence of the matrix seems to play a role only in the first period of immersion since that at equilibrium the values are close for both composite (Table 1). Studies made on pure PLA and PP matrices lead to absorptions of around 1% and 0.5% respectively for PLA and PP [8]. Therefore, the high retention capacity observed for the flax/PLA and flax/PP composites are mainly due to the presence of flax fibres and their hydrophilic nature.

Composite	**Product**	**$k*10^3$ (mm/√s)**	**M_m (%)**
Flax/PLA	***Water***	1.77	27.51%
	Surfanios®	1.85	22.63%
	Terralin®	2.50	22.61%
Flax/PP	***Water***	1.46	28.30%
	Surfanios®	1.67	23.06%
	Terralin®	1.72	21.94%

Table 1. Slope of linear part of the weight gain curve k , Weight gain at equilibrium M_m.

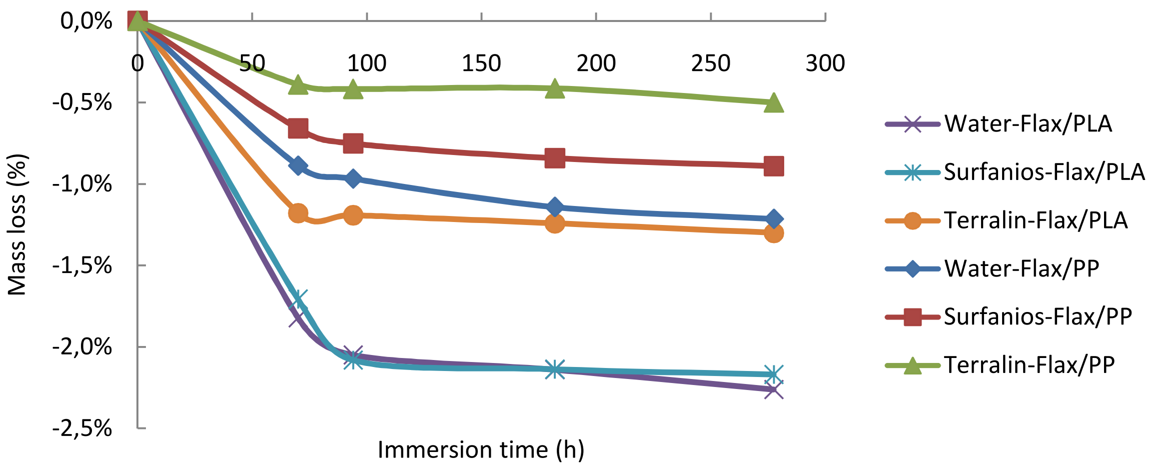
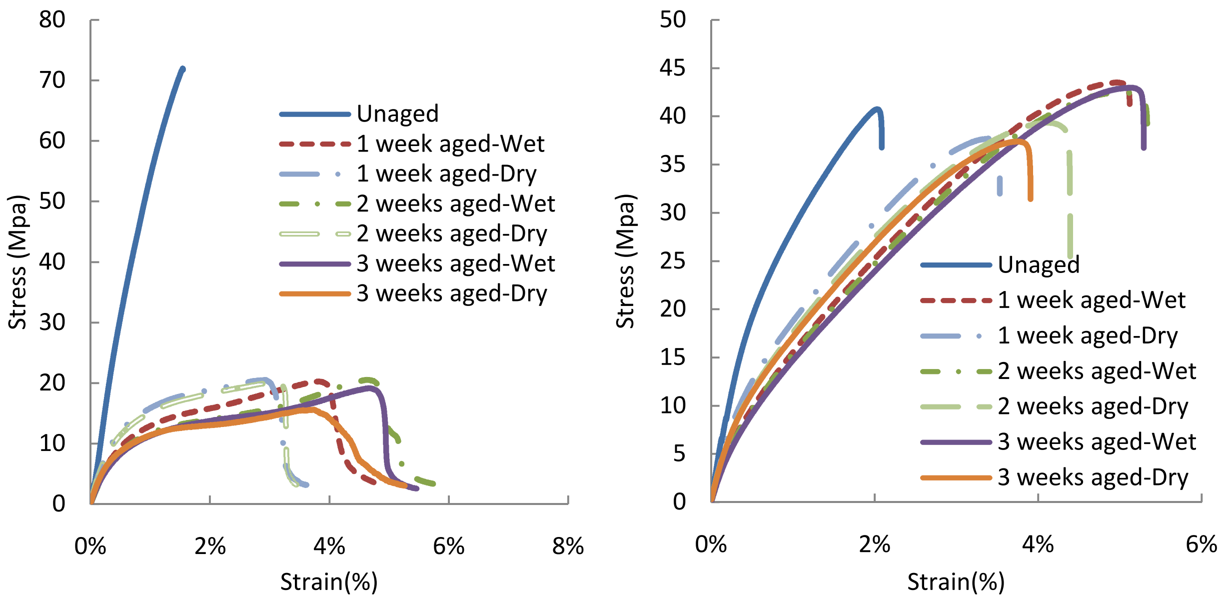


Figure 2. Mass loss of the composites for immersion tests.

On the figure 2, we can observe that the mass loss is more important for the flax/PLA composite. We can expect that despite showing very close values of weight gain, the flax/PLA is more damaged than the flax/PP and this should result in more important loss of mechanical properties for the flax/PLA composite.

3.2 *Mechanical properties*

The evolution of mechanical behaviour and properties with ageing was studied for water and Surfanios®. Figure 3 shows the evolution of the mechanical behaviour of the composites with water characterised in the wet and dry state. Tables 2 & 3 summarise the tensile properties. Ageing has an irreversible effect on the flax/PLA mechanical behaviour. The unaged flax/PLA composite is brittle and ageing results in a ductile behaviour of the composite even after drying: a short linear evolution at the beginning and a non-linear evolution until fracture. As the Young's modulus and maximum stress decreases drastically, the failure strain almost doubles the initial values (figure 3-a, and table 2). This is not the case for the flax/PP composite, the initial behaviour of the flax/PP composite has two linear zones (figure 3-b). With ageing, the same behaviour is observed, with a shorter elastic zone followed by a longer linear zone with the gentler slopes.



(a)Flax/PLA water aged (b)Flax/PP water aged

Figure 3. Tensile behaviour of water aged composites for dry and wet samples.

Table 2 shows a significant decreasing, (≈-72%) of the maximum stress of the flax/PLA composite within the first week of immersion in the products. Both water and Surfanios® produces the same effect on the maximum stress and the influence of drying is insignificant. This may be caused by matrix and fibre/matrix interface damage due to the hydrophilic nature of both components: the cleaning product is soaked by the fibre by capillarity and propagated through the matrix at the interface. Indeed figure 2 shows important mass loss for the PLA reinforced composite compared to the PP one implying that damage occurs in PLA. Furthermore, the tensile flax/PLA aged samples were not completely broken apart at failure; the reinforcement still maintains the tensile specimen in the failure zone. This is typical of fibre/matrix decohesion and matrix damage. On the other hand, the tensile specimens of flax/PP showed a neat fracture at failure and it can be seen on figure 3 (b).

	Water- wet state			Water-dry state		
	σ_{max} (MPa)	E (GPa)	ε_{max} (%)	σ_{max} (MPa)	E (GPa)	ε_{max} (%)
Unaged	67.48 ± 6.31	6.56 ± 0.30	1.44 ± 0.14	67.48 ± 6.31	6.56 ± 0.30	1.44 ± 0.14
1 week	18.42 ± 2.57	2.55 ± 0.43	4.22 ± 0.59	17.12 ± 3.00	3.25 ± 0.25	2.59 ± 0.34
2 weeks	19.58 ± 1.29	2.37 ± 0.04	4.51 ± 0.18	18.85 ± 1.82	3.07 ± 0.18	3.00 ± 0.07
3 weeks	19.15 ± 0.78	2.37 ± 0.03	4.69 ± 0.32	19.30 ± 2.08	3.22 ± 0.28	2.89 ± 0.20
	Surfanios®-wet state			Surfanios® -dry state		
	σ_{max} (MPa)	E (GPa)	ε_{max} (%)	σ_{max} (MPa)	E (GPa)	ε_{max} (%)
Unaged	67.48 ± 6.31	6.56 ± 0.30	1.44 ± 0.14	67.48 ± 6.31	6.56 ± 0.30	1.44 ± 0.14
1 week	18.71 ± 3.51	2.43 ± 0.37	4.05 ± 0.63	22.09 ± 2.82	3.65 ± 0.34	1.84 ± 0.13
2 weeks	17.86 ± 0.23	2.34 ± 0.02	4.16 ± 0.01	18.42 ± 2.82	2.70 ± 0.32	3.52 ± 0.36
3 weeks	16.00 ± 0.57	2.14 ± 0.01	4.60 ± 0.23	16.04 ± 3.00	2.80 ± 0.25	3.71 ± 0.34

Table 2. Evolution of mechanical properties for the flax/PLA composite in the wet and dry states.

Table 3 shows a very different evolution of the maximum tensile stress at break of the flax/PP composite compared to the flax/PLA composite. For the wet samples, the maximum stress increases with product uptake (≈+7.5%) before stabilizing. Whereas for the dry sample, a relatively small decrease (≈-5.5%) is observed. The relatively good conservation of the maximum stress for both wet and dry samples shows that the matrix and fibre/matrix interface is not much damaged and can be related to the hydrophobic nature of the PP matrix. The increase in strength observed for the wet samples could be due to the combination fibre swelling and the conservation of the fibre/matrix interface due to the hydrophobic PP. However this variation remains in the natural dispersion of the tensile properties obtained for this composite (table 3).

Both composites follow the same decreasing trend of their Young's modulus in the wet state: an important decrease (≈-62% for the flax/PLA and ≈-50% for the flax/PP composite) within the first week and a steady slight decrease afterwards. This confirms the sensitivity of the fibres modulus with uptake [5] and the steadiness observed afterwards could be explained to the fact that uptake equilibrium has almost been reached [11]. The effect of drying resulted in a partial recovery of the modulus. The loss of modulus for the flax/PLA composite is greater than the flax/PP composite just as it was for mass loss. Flax/PLA samples immersed in Surfanios® showed more lost of modulus at week 3 but the difference remains rather small. This can be due to the fact that the fibre stiffness could be more sensible to irreversible damage with Surfanios® at long-lasting exposure. In all cases, the initial properties are never recovered after drying. This underlines irreversible damage of the fibre, matrix and interface [16]. Ageing causes an important increase in the strain at failure [11] and the drying of specimens reduces its value [5].

	Water- wet state			**Water-dry state**		
	σ_{max} (MPa)	**E (GPa)**	**ε_{max} (%)**	**σ_{max} (MPa)**	**E (GPa)**	**ε_{max} (%)**
Unaged	39.82 ± 1.63	5.07 ± 0.26	2.82 ± 0.19	39.82 ± 1.63	5.07 ± 0.26	2.82 ± 0.19
1 week	43.60 ± 0.57	2.60 ± 0.03	4.95 ± 0.01	37.71 ± 0.01	3.47 ± 0.06	3.47 ± 0.08
2 weeks	42.85 ± 0.48	2.62 ± 0.01	4.87 ± 0.24	38.63 ± 1.02	3.00 ± 0.01	3.93 ± 0.27
3 weeks	43.00 ± 0.05	2.49 ± 0.05	5.26 ± 0.15	36.08 ± 1.84	3.08 ± 0.07	3.85 ± 0.13
	Surfanios®-wet state			**Surfanios® -dry state**		
	σ_{max} (MPa)	**E (GPa)**	**ε_{max} (%)**	**σ_{max} (MPa)**	**E (GPa)**	**ε_{max} (%)**
Unaged	39.82 ± 1.63	5.07 ± 0.26	2.82 ± 0.19	39.82 ± 1.63	5.07 ± 0.26	2.82 ± 0.19
1 week	41.77 ± 0.83	2.56 ± 0.08	4.41 ± 0.20	38.41 ± 1.28	3.47 ± 0.16	3.33 ± 0.08
2 weeks	43.29 ± 1.85	2.43 ± 0.06	5.38 ± 0.54	35.93 ± 0.11	2.80 ± 0.06	4.15 ± 0.10
3 weeks	42.62 ± 2.64	2.47 ± 0.02	5.22 ± 0.60	36.51 ± 0.45	2.88 ± 0.12	4.04 ± 0.35

Table 3. Evolution of mechanical properties for the flax/PP composite in the wet and dry states.

The results show that the product uptake has an important effect on the tensile properties of the biocomposites, particularly by decreasing the modulus and increasing the strain at break. The matrix also plays a role in the rate of absorption.

4 Conclusion

Two composites, flax/PLA and flax/PP were studied to investigate the effect of product uptake on their mechanical properties. It was shown that the uptake rate was dependant of the matrix and somehow related to the mass loss. The tensile tests performed showed that for both composites, the mechanical properties vary, mainly in the first week of immersion. The Young's Modulus decreases drastically and the failure strain increases. The flax/PLA showed a more important decrease of its failure stress and its mechanical behaviour is modified with ageing. These changes in behaviour were linked to the nature of the matrix of the composite. Although, the retention of water was superior in both composites, the damage caused by the uptake of Surfanios® was of the same order or higher at longer exposure times. The recovery of the mechanical properties with drying was less than expected for both composites.

Among the prospects with ongoing studies, the analysis of the influence of the cleaning products concentrations and also the effect of the manufacturing process and its parameters on the uptake behavior.

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