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Visco-elastoplastic Characterization of a Flax-fiber Reinforced Biocomposite

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Abstract: *In the presented study, the load induced long-term behavior of a biocomposite material is analyzed. The studied material is a unidirectional flax fiber reinforced epoxy resin, material, whose quasi-static mechanical properties can compare with those of glass fiber composites. Samples with a fiber direction of 0° were subjected to two types of multi-level creep-recovery tests, one with a varying creep duration, and the other with a varying creep stress, with the purpose of discriminating the viscoplastic and viscoelastic behavior of the composite. Results show a significant viscous response in time, dependent on both creep duration and creep stress, up to 20% of the elastic one. Sample damage is absent, leading to the conclusion that the viscoplastic response is caused by the permanent reorganization of the fiber's internal structure.*

Keywords: *biocomposite, creep, flax fiber, experimental*

1. Introduction

The demand for low cost, high quality materials is increasing continuously [1, 2]. Due to this fact, one of the disadvantages regarding composite materials, their impact on the environment, is becoming an issue. These materials require significant amounts of energy to fabricate and tend to have a poor end of life cycle, as they are difficult and costly to recycle [3]. A solution to the reduction of the environmental footprint of these materials is to implement bio-based solutions [4]. By using vegetable reinforcements, there is a significant reduction in the environmental impact of the overall composite material [5–7]. Vegetable reinforcements are being studied due, mainly, to this advantage they possess over their synthetic counterparts. They also possess other strongpoints, such as a lower density [8], which results in higher specific properties (specific strength, rigidity etc.) and vibration damping properties [9, 10]. Several steps towards large scale use of bio-based materials still need to be made, however, as their long-term behavior requires further study. Due to the organic nature, the plant fiber reinforcement, is itself considered a composite material, consisting mainly of a hemicellulose matrix reinforced by cellulose microfibrils [11]. The structure provides a more complex time dependent behavior, which in turn, influences the overall composite mechanical properties.

Presently, focus is on the study of fiber reinforcements which can compete with the most widely used synthetic one, glass fiber [12]. These are, mainly, flax, jute, hemp and kenaf [13]. In Europe, the most available is flax fiber, around which an industry is already developed, due to its wide use in textile applications [14]. Thus, focus of the present study is on flax fiber reinforced composites.

It has been shown, mainly through tensile tests [15–17], that the elementary fibers present a viscous component in their response, which is inherited by the composites they reinforce [18]. This behavior, while still under study, has been attributed to the elementary fiber reorganization under stress, mainly the cellulose microfibrils [17]. In the case of synthetic fiber composites, the time dependent behavior on the fiber direction is insignificant [19, 20], leading to the conclusion that the reinforcement is responsible for the viscous response exhibited by FFRPs.

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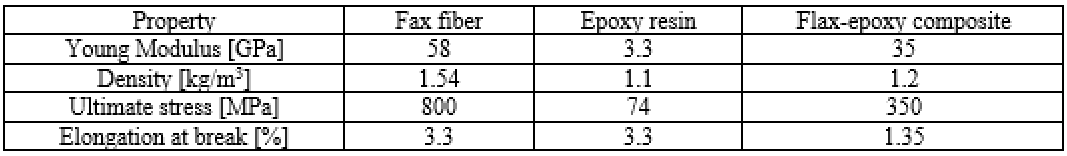
The present works is aimed at evaluating the time dependent response of a FFRP. While taking a purely experimental approach to the analysis, with the objective of quantifying the time dependent behavior, interpretations are as well presented for the obtained results. The required experimental procedures for characterization, while varied, tend to keep constant one of the testing parameters, such as the strain (relaxation test), strain rate (ramp loading test) or the load (creep test) [21]. The measured response is, for the first two examples, the stress, while for the third, the strain. Here, repeated constant load tests are chosen. Two procedures were designed, consisting in multi-level creep-recovery tests in which the creep duration and creep stress are varied, respectively, in order to determine their influence on the material response.

2. Materials and methods

The composite material to be studied is composed of a unidirectional flax fiber reinforcement in an epoxy resin matrix, commercially known as Lineo FlaxPreg T-UD 110. It was supplied as a roll of impregnated fibers, with a fiber surface density of 110g/m^2 [22]. From the rolls, 16 layers of 280x280 mm rectangular shape were cut and stacked in a mold, for a final composite thickness of 2 mm. The stacked layers were subjected to a thermocompression cycle, at 130°C and 3 bars for one hour, followed by a posturing cycle of 2 h at the same temperature and no pressure. From the resulting square plate, samples were cut, 250x25 mm in size. In Table 1, mechanical properties, reported in the literature, are presented for the fiber, the epoxy resin and the composite. The composite of this study has been measured to contain, neglecting void content, approximately 60% fiber volume fraction and 40% matrix volume fraction. Considering the rule of mixtures and the properties presented in the table, this results in a Young modulus of 36.1 GPa, similar to the one reported by the fiber manufacturer.

Table 1. Mechanical properties for the composite and its constituents [18, 20, 22]



Property	Fax fiber	Epoxy resin	Flax-epoxy composite
Young Modulus [GPa]	58	3.3	35
Density [kg/m^3]	1.54	1.1	1.2
Ultimate stress [MPa]	800	74	350
Elongation at break [%]	3.3	3.3	1.35

Two of the fabricated specimens were chosen for the experimental procedure and were equipped with aluminum tabs and a pair of strain gauges, mounted back-to-back (Figure 1). The strain gauges were connected in a half bridge configuration to eliminate bending induced deformations. Before testing, the samples were conditioned for seven days, at a 23°C and 50% humidity, in order to reduce these factors possible influence. Load was applied with a Shimadzu AGX-X electromechanical testing machine.

Previous work concerning this flax fiber - epoxy resin composite material [23] has shown the existence of a damage threshold, at approximately 130 MPa and, thus, the appearance of damage induced deformations. To avoid it, stress levels were chosen below this limit. Avoiding damage is advantageous in the experimental procedure, as a structurally intact sample can be tested multiple times. Consequently, multiple creep tests on the same sample can be conducted, with a recovery period in-between during which the viscoelastic deformations can dissipate. Since there are two parameters whose influence is analyzed, the choice has been to subject two samples to the creep-recovery tests, one with varying creep duration and the other with varying creep stress. The loading phase was made with a constant crosshead speed of 2 mm/min, recommended in tensile tests by ASTM 3039 [24]. The selected parameters for creep-recovery procedures are presented in Table 2. The recovery was chosen to be constant and at least six times more than the longest creep period.

The sample response to the applied load cycle consists of a deformation, variable in time. During the creep phase, the material responds as a cumulation of viscoelastic and viscoplastic deformation. In the recovery phase, the viscoplastic response remains constant, at the value prior to unloading and the viscoelastic component tends to recover, completely if sufficient time is allowed between two consecutive loadings. Thus, at the end of the recovery phase, the viscoplastic deformation can be determined.

Figure 1. Sample used for the creep-recovery tests

Table 2. Creep-recovery parameters for: variable creep duration (left); variable creep stress (right)

Stress [MPa]	Creep period [h]	Recovery period [h]
80	1	24
	2	
	3	
	4	

Stress [MPa]	Creep period [h]	Recovery period [h]
30	1	24
60		
80		
100		

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3.Results and discussions

The deformation response signal for the varied creep duration test is presented in Figure 2 and the one for the varied creep stress, Figure 3.

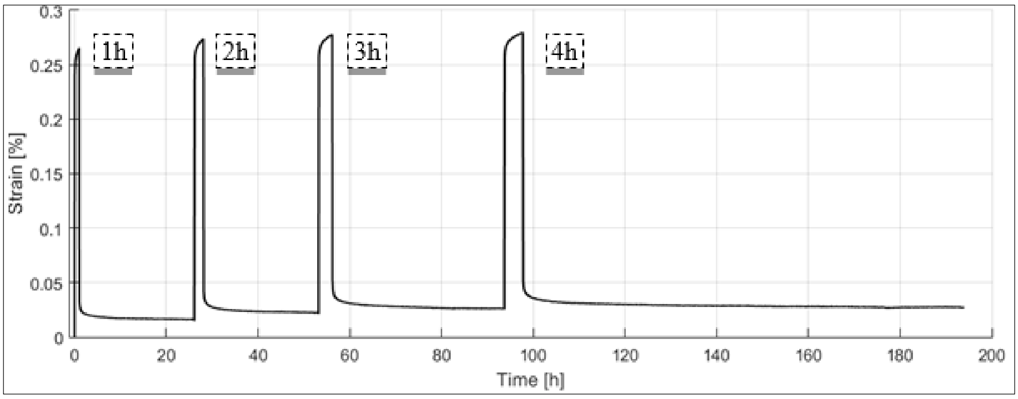

Figure 2. Experimental results for the creep-recovery tests with variable creep duration

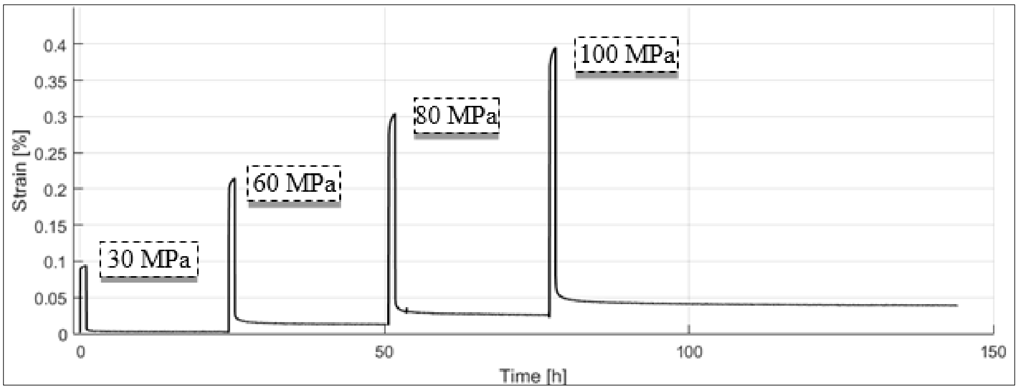

Figure 3. Experimental results for the creep-recovery tests with variable creep stress

It can be noticed that there is a significant evolution in the material response during both the creep and the recovery phases. The recovery shows that it is composed by both viscoelastic deformations (which dissipate) and viscoplastic deformations. They both increase with respects to time and stress level. The variable creep duration tests show the first load level as having the highest viscoplastic response. While clear that there is a pronounced time dependent behavior, it needs to be analyzed from the experimental data. For this, several parts of the creep-recovery curves are extracted, as presented in Figure 4: the beginning of the creep stage, from which the instantaneous response, ε_0, is identified, the duration of the creep phase, from which the transient response in creep, $\Delta\varepsilon_{fl}$ can be extracted, the recovery phase, which gives the transient response in recovery, $\Delta\varepsilon_{rec}$, and the end of the recovery phase, from where the cumulated viscoplastic response, ε_{pl}, can be determined.

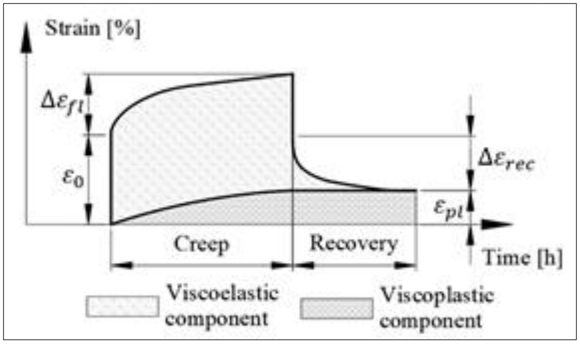

Figure 4. Data of interest from the creep-recovery curves

The information is presented in Table 3 for the variable creep duration tests and in Table 4 for the variable creep stress tests. By analyzing the deformation response in the loading phase, ε_o, it can be noticed that it tends to slightly decrease for the variable creep duration tests, suggesting hardening of the sample. An analysis of the secant modulus, E, shows that it tends to increase, from 35.6 GPa to 36.9 GPa (3.6%). The hardening is theorized to be a consequence of the irreversible component in the reorganization of the elementary fibers which occurs during the loading phase. The theory is supported by the results of Pitarresi [25], who obtained a similar result through repeated traction tests. The calculated modulus, close to that reported in the literature (Table 1) and its evolution indicate the absence of damage during the creep phases.

In the variable creep stress results, a decrease in modulus is observed, from 35.2 GPa to 31.6 GPa (or 10%), showing a softening of the sample with stress level. This behavior has been observed in all traction tests conducted on composites reinforced by flax fibers [11], long or short, suggesting that the nonlinear behavior of the fiber leads to the composite response. The theory is supported by tensile tests with variable loading speeds, where an increase in modulus was observed with the increase of loading speed [18]. This suggests that raising the load speed could reduce the softening effect. A too elevated loading speed, however, presents the risk of overcharging the sample and producing a shock, detrimental to the testing procedure as they are sources of errors.

Regarding the creep and recovery phases, Tables 3 and 4 show the proportion of time dependent response with respect to being between 10 and 20%, increasing with load level. The viscoelastic component, as resulted from the recovery response, 8 - 16% of the elastic response and the plastic one, 1.5 - 7%. It can also be noticed that the sum of recovery and viscoplastic deformation is slightly higher than the response during creep. It has been shown that, during the loading phase, the material tends to creep as well, due to the impossibility of an instantaneous loading [26], caused by the testing setup and the risk of appearance of shock.

The viscoplastic deformations are a result of the entire loading history and are, thus, analyzed as cumulated, ε_{pl}. While, in Tables 3 and 4, an increase in time is obvious, the dependance on creep duration and creep stress is not linear. However, by passing in a logarithmic scale, a linear evolution is noticed, in Figure 5, showing an exponential dependance with respect to the two parameters. The type of response is similar to that observed for polymer materials [27], for short fiber composites [28] or off axis unidirectional composites with synthetic reinforcements [29].

The two reported reasons are micro-damage, which has been infirmed in the present work, and the permanent reorganization of the polymer molecular structure (epoxy matrix and fiber hemicellulose). It can be concluded, thus, that the latter is the mechanism for viscoplastic deformations observed.

Table 3. Data obtained from the variable creep duration tests

Level	Time [h]	σ [MPa]	ε_0 [%]	E [MPa]	$\Delta\varepsilon_{fl}$ [%]	$\Delta\varepsilon_{rec}$ [%]	ε_{pl} [%]	ε_{pl} (cumulated) [%]
1	1	80	0.2247	35.597	0.0406 (18.08%)	0.0314 (13.97%)	0.0161 (7.16%)	0.0161
2	2		0.2212	36.174	0.0365 (16.48%)	0.0327 (14.79%)	0.0064 (2.90%)	0.0225
3	3		0.2185	36.616	0.0366 (16.73%)	0.0344 (15.73%)	0.0041 (1.86%)	0.0266
4	4		0.2169	36.876	0.0354 (16.30%)	0.0348 (16.04%)	0.0032 (1.48%)	0.0298

Table 4. Data obtained from the variable creep stress tests

Level	Time [h]	σ [MPa]	ε_0 [%]	E [MPa]	$\Delta\varepsilon_{fl}$ [%]	$\Delta\varepsilon_{rec}$ [%]	ε_{pl} [%]	ε_{pl} (cumulated) [%]
1	1	30	0.085	35.277	0.009 (10.60%)	0.007 (8.27%)	0.0025 (2.93%)	0.0025
2		60	0.184	32.556	0.028 (15.04%)	0.023 (12.72%)	0.0104 (5.61%)	0.0128
3		80	0.240	33.226	0.050 (21.00%)	0.035 (14.76%)	0.0128 (5.43%)	0.0257
4		100	0.315	31.689	0.054 (17.18%)	0.049 (15.46%)	0.0153 (4.84%)	0.0410

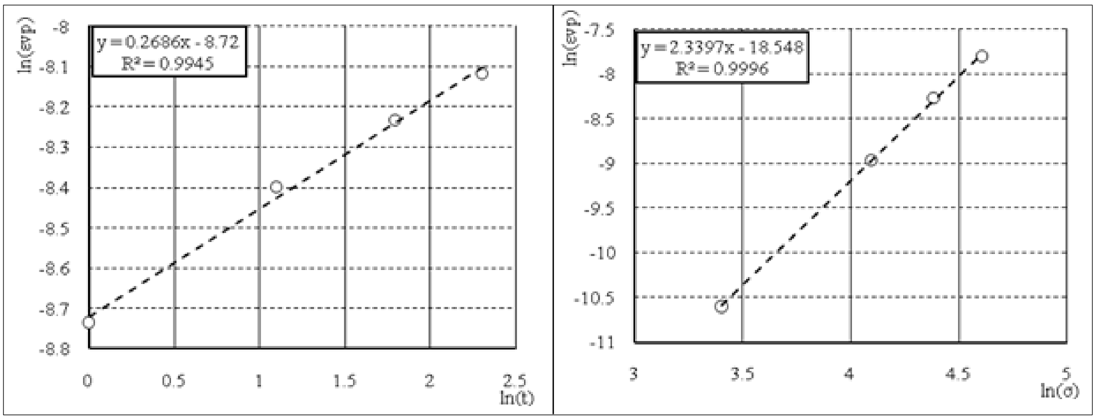

Figure 5. Cumulated viscoplastic deformations with respect to: total creep duration (left); creep stress (right)

4.Conclusions

A flax fiber - epoxy resin composite material with the reinforcement direction of 0° has been studied for its time dependent response. For the experimental procedure, samples have been fabricated and equipped with strain gauges, which allowed for long term testing.

It has been noticed, through creep-recovery tests, that there is a pronounced response, composed of a viscoelastic and viscoplastic component. Both tend to be influenced by creep duration and creep stress, the latter having the higher influence, when referring to percentage of instantaneous response.

It has also been noticed that the sample tends to slightly harden in the tests with variable creep duration and that it tends to reduce in rigidity with respects to stress, similarly to what has been noticed in traction tests. This proves that damage induced deformations are absent and that the stress-strain response is non-linear, similar to what has been observed in tensile tests. A possible improvement of the experimental procedure would be to increase the loading speed, form the 2 mm/min to higher values, which could reduce the nonlinearity of the instantaneous response.

The cumulated viscoplastic deformation, has been observed to have an exponential evolution with respect to both time and stress, similar to what has been reported for polymeric materials.

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