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Mechanical behavior of a cementitious composite incorporating flax fibers treated with citric acid

L. Freitas Dutra ¹, S. Chafei ², A.D. Tran Le¹, T. Langlet¹

¹ Innovative Technologies Laboratory (LTI), University of Picardie Jules Verne, Avenue des Facultés, 80025 Amiens Cedex 1, France

² Civil and Geo-Environmental Engineering Laboratory (LGCgE), University of Artois, 62400 Béthune, France

lorena.dutra-freitas@u-picardie.fr

Abstract. The building and construction sector is one of the sectors most concerned by the challenges of sustainable development. Indeed, building (construction) represents about 40% of CO₂ emissions in developed countries, 37% of energy consumption and 40% of waste produced. In this perspective, the development of building materials and devices that emit less CO₂, consume less non-renewable resources and are as widely recyclable as possible has been the subject of numerous studies. This work relates to the treatment of flax fibers with citric acid in order to improve the behavior of a cementitious mortar. The effect of the treatment on the kinetics and the water absorption rate of the fibers was first evaluated, then its influence on the properties of the fibrous mortar was studied. The treatment reduced the setting time of the cementitious mixtures and significantly increased the flexural strength compared to the raw fiber composites. SEM and wettability test confirmed the treatment efficiency to reduce the hydrophilic nature of the treated fibers.

Keywords: Cementitious composite, flax fibers, bio-based materials, mechanical performance

1 Introduction

The use of flax fiber has a very ancient origin for the creation of objects and clothing, but today there is a renewed interest in this material that has developed due to its good mechanical properties. This aspect opens new frontiers for the use of flax fiber as a low density, ecological and cost-effective material that can be used as reinforcements in composite materials, improving the mechanical properties and also the life cycle of the final product.

However, attempts to incorporate plant fibers into cementitious materials have yielded mixed results because they are accompanied by a decrease in the workability of the mixture, a delay in setting and hardening, and a reduction in mechanical performance after 28 days. These shortcomings reflect the physicochemical incompatibility between the cementitious matrix and the plant fibers, which motivates many works aiming at stripping the fibers by chemical or physical treatments, or covering them with a protective coating with respect to the cementitious medium [1].

The adaptation of the reinforcement/cement matrix couple was also sought by modifying the cement matrix [1–3].

A chemical treatment of flax fibers with a citric acid solution is proposed in order to partially clean the fiber from its surface impurities. Its impact was evaluated on the properties of the fibers and those of the cementitious composite in the fresh and hardened states.

2 Flax fibers

2.1 Fibers treatment

Flax was produced in Normandy, France in 2018. The physical and mechanical properties of the raw fibers are given in Table 1. Fibers of length 24 mm were used in the preparation of the mixtures. Citric acid powder was used for the preparation of the solution for the treatment of the fibers. The fibers were immersed in a 5% citric acid solution for 2 hours, rinsed with tap water and air dried for 24 hours.

Table 1: Physical and mechanical properties of flax fibers

Diameter (μm)	$22,75 \pm 6$
Density	1,52
Thermal conductivity (W.m-1.K-1)	0,041
Modulus of elasticity (GPa)	44 ± 21
Tensile strength (MPa)	849 ± 482
Ultimate deformation (%)	$1,78 \pm 0,6$

2.2 Kinetics and the rate of saturation in water

The water uptake kinetics of FB and FA fibers were followed up to 24 h. The test consists of immersing 5 samples of 1 g of each type of fiber in water using tea tongs for 5, 20, 30 minutes and 24 h. The fibers are then superficially wiped with absorbent paper and their masses are then measured. The water absorption rate at these times is calculated with the equation 1.

$$Abs\% = \frac{\text{damp mass} - \text{dry mass}}{\text{dry mass}} \quad (1)$$

Figure 1 shows the water absorption kinetics of raw and treated fibers. It can be seen that raw fibers can absorb up to 133% of their dry mass. Their saturation is reached after 20 minutes of immersion. Following the citric acid treatment, a considerable decrease of 41% of the water saturation rate of the treated fibers after 24 h of immersion is noted.

The time required for water saturation remained unchanged. This could be explained by the fact that the treatment partially cleaned the surface of the fibers by reducing the level of hydrophilic compounds. As hemicellulose and other non-

cellulosic impurities are removed from the fibers after the acid treatment, it is expected that absorption decreases.

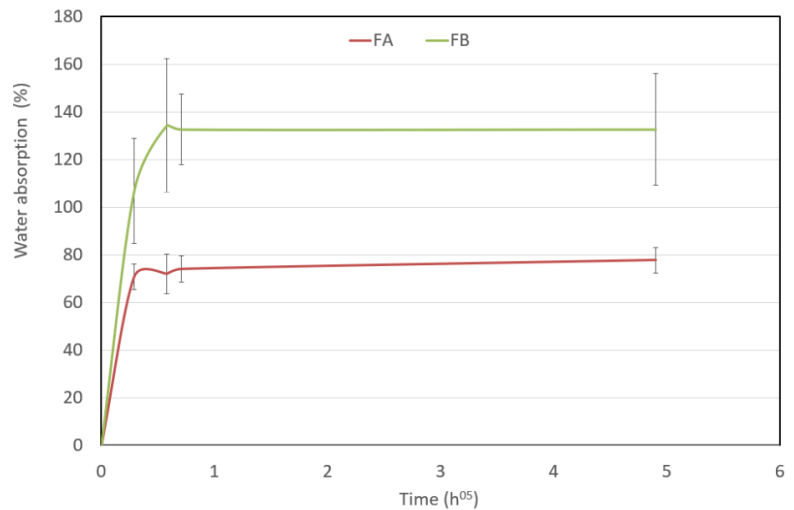


Figure 1: Water absorption kinetics of raw and treated flax fibers

2.3 Scanning electron microscopy (SEM)

Images of untreated and treated fibers were acquired using a Quanta 200 field emission gun environmental scanning electron microscope (ESEM, FEI Company, USA) in "low vacuum" mode at the pressure of 0.5 torr and an accelerating voltage of 5kV.

Figure 2 shows SEM images of untreated (a) and treated fibers (b). It can be seen from Figure 2a that raw flax fibers are formed by elementary fibers held together. They are mainly composed by wax, pectin, lignin, and hemicellulose. After treatment with citric acid (Figure 2b), in addition to the modification of the fiber surface, it can be seen the individualization of the fibers which indicates that substances related to cellulose were removed. This is congruous with results presented by [4] who used alkaline treatment on flax fibers.

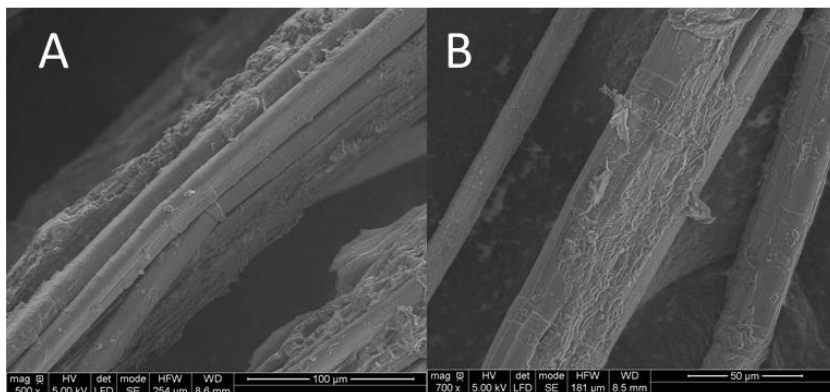


Figure 2 : Scanning electron microscopy images of flax fiber bundles untreated (A) and treated with citric acid (B). The magnification is 500× on the left column and 700× on the right column.

2.4 Contact angle

Contact angle can be used to characterize the wettability of materials: the smaller the contact angle between a solid and a liquid, the more the liquid wets the solid, which reflects a certain affinity between the two elements. Thus, an angle between 0° and 90° characterizes a hydrophilic surface. Conversely, an angle between 90° and 180° means that the surface is hydrophobic.

The contact angle between the fiber and a water has been performed by using a drop shape analyzer. A water droplet was deposited on the fiber with a syringe. The profile of the droplet was filmed against the light and the contact angle between the fiber and the droplet can be measured using an image processing software.

Images of testing samples untreated and treated can be found in Figure 3. From these results, it can be concluded that the use of citric acid significantly increased the contact angle (from 44° to 90°) providing a more hydrophobic fiber surface. These findings corroborate the decrease in water absorption described in the previous sections.

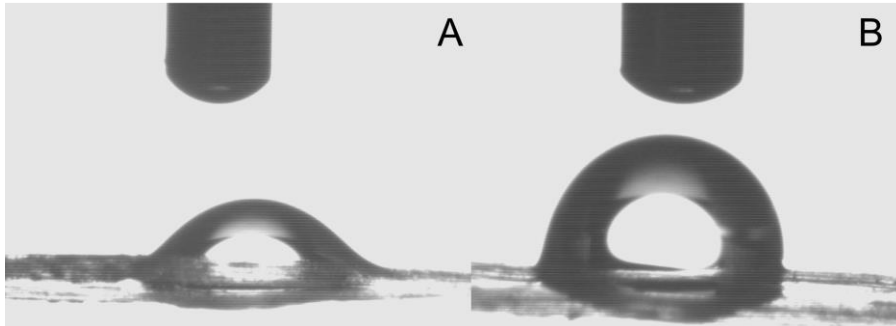


Figure 3: Contact angle for raw (A) and treated flax fiber (B).

2.5 Preparation of mortars

The binder used is a portland cement CEM I 52,5 N in compliance with the NF EN 196-1 standard [5]. The sand is a 0/4 compliant with the NF P 18-545 standard [6]. The flax was produced in Normandy in 2018.

Three formulations were made: mortar incorporating raw fibers, mortar incorporating treated fibers and control mortar, called MFB, MFA and MT respectively.

Twelve $4 \times 4 \times 16$ cm³ specimens were prepared for each formulation for mechanical tests at 7 and 28 days. They were demolded after 48 hours and stored in a storage room at 20°C and 50% humidity until the mechanical tests.

2.6 Setting time

The setting time was measured with a Vicat device according to EN 196-3 [7].

The presence of fibers in the mortar hinders the penetration of the Vicat needle which could lead to incorrect results. Therefore, another protocol was followed:

- The raw or treated fibers were first immersed in water for 48 hours with the same fiber/water ratio of the mortar formulations;
- Then, the water containing the FB and FA extractables was recovered and used to make cementitious pastes called PFB and PFA respectively;
- A control cementitious paste (CP) was also prepared with tap prepared with tap water.

2.7 Mechanical tests

The three-point bending test was carried out on the mortar specimens at 7 and 28 days of curing in accordance with NF EN 196-1 [5], using an electromechanical press of 50 kN capacity operating at a speed of 50 N/s.

The compression test was performed with another press of higher capacity. The loading speed was 2400 N/s.

3 Results and discussion

3.1 Setting time

The setting times of cementitious pastes made with fiber extractables (PCFB and PCFA) or with tap water (PC) are shown in Figure 4. For the PC paste, it can be noted a time of about 2.5 hours. A significant delay of about 60% was noted for PCFB compared to PC. Indeed, the non-cellulosic polysaccharides present on the surface of the fibers, in particular the pectins, are known for their role as chelators of calcium [9], an ion necessary for the advancement of the cement hydration reaction [10]; this could explain the delay noted. For the PCFA paste, there is a decrease in the delay of about 24% compared to PCFB. This also confirms the partial elimination of the polysaccharide treatment responsible for this delay.

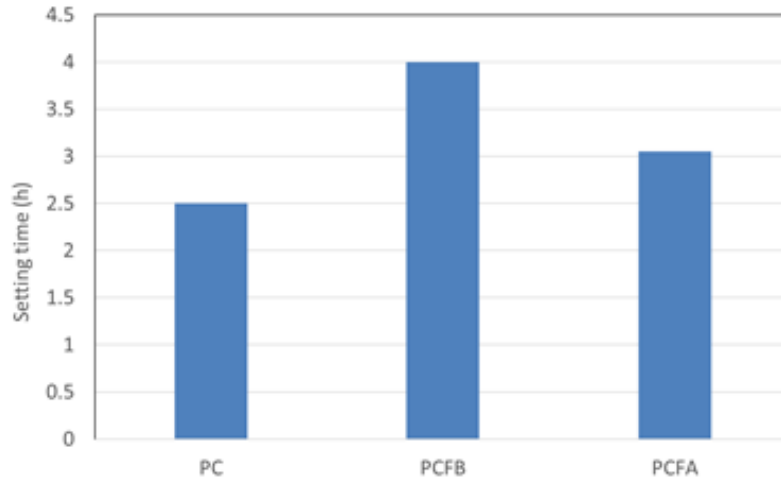


Figure 4: Setting time of cementitious materials.

3.2 Mechanical tests

Figure 5 shows the three-point bending behavior of the cementitious composites and the control mortar. Comparing the curves of the composite incorporating raw fibers MFB and that of the control mortar MT, it can be noted very different behaviors. In the case of MT, the behavioral domain of the material ends with an abrupt failure which corresponds to the abrupt propagation of the first matrix crack. In the case of MFB, the first matrix crack (point $(\delta P1, P1)$) appears for the same value of displacement as MT, the force and displacement continue to grow after this first damage until a maximum load value $P2$ (associated displacement $\delta P2$). The range of the curve between $\delta P1$ and $\delta P2$ is the range of extension of the damage by microcracks in the matrix and the resumption of the loading by the fibers.

From the point $(\delta P2, P2)$, the cracks propagate in the composite in a controlled manner due to the presence of the fibers bridging the two lips of the macrocrack (if the loading rate is slow enough). A somewhat different behavior is observed for MFA: the post-peak phase is much larger although the maximum load is similar to that of MFB. The larger deformation is explained by a better load transfer from the matrix to the fibers due to a better adhesion between the fibers and the matrix

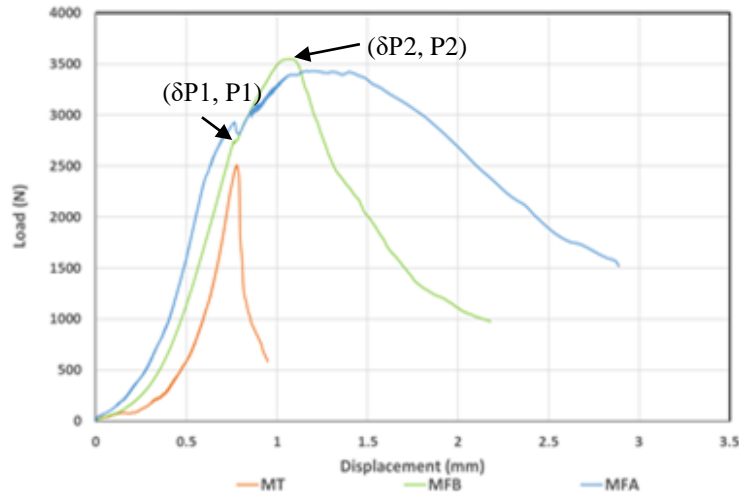


Figure 5: Three-point bending behaviour of the different formulations.

The bending stresses obtained for these different formulations are presented in **Erreur ! Source du renvoi introuvable.** A considerable increase was noted for MFB in comparison with the control mortar MT. It is more important at 7 days (about 29%) than at 28 days (10.5%).

The treatment of the fibers with citric acid resulted in even greater increases compared to MT which could be due to a better surface quality of these fibers leading to a good fiber-matrix adhesion as early as the seventh day of curing.

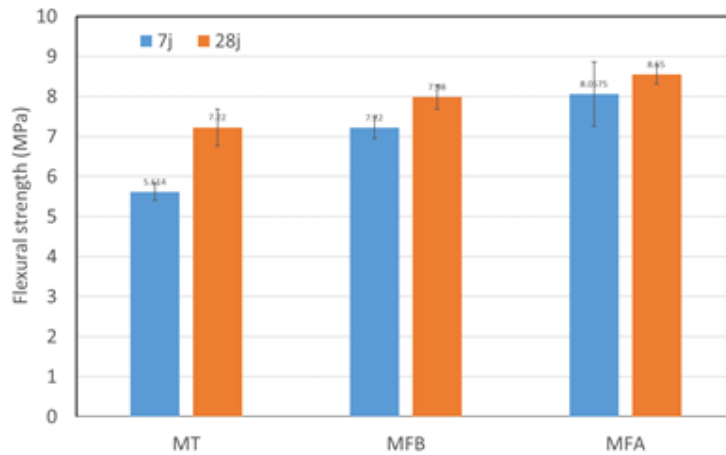


Figure 6: Flexural stresses of mortars

4 Conclusion

Pure cementitious matrices that exhibit brittle behavior change to more ductile behavior when added with plant fibers. In this work, the influence of a treatment based on citric acid on the mechanical properties of a mortar incorporating these fibers was studied.

The treatment significantly reduced the water saturation rate of the fibers as well as the setting time of the cementitious mixtures. SEM and wettability test confirmed that the treatment was effective in changing the surface of the fiber from hydrophilic to hydrophobic. Mortar with treated fibers (MFA) exhibited better flexural and compressive strengths compared to the mortar with untreated fibers (MFB).

Therefore, the surface modification of plant fibers could improve the composite in complementary ways: increasing their mechanical properties, improving the adhesion of the fibers to the matrix to optimize the resistance of the fiber/matrix transition zone and finally limiting the problems related to the high water absorption of the fibers. Considering that this treatment slows down the degradation of the fibers within the composite without stopping it, other ways to improve the durability of these plant fiber-reinforced cementitious composites are being explored.

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