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Dependence of the properties of cementitious composites on the nature of the hydraulic binder coating the reinforcing flax fibers

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Abstract: *This work aims to improve the formulation of short flax fiber-reinforced mortar through coating the raw fibers with hydraulic binders like cement or lime, or cleaning them in boiling water prior to the coating. These treatments are chosen for their ability to reduce the water intake by the fibers and to minimize the migration of their extractible compounds toward the cement matrix. Formulations incorporating raw or treated fibers were characterized in the fresh state. The results showed a drastic reduction of water intake, which correlates with an improvement of the properties of the fiber-mortar mixtures in the fresh state, particularly in terms of the heat of hydration, initial setting time and consistency. Mechanical behavior of the different formulations is also evaluated by flexural tests.*

Keywords: *Cement matrix; flax fiber; treatments; fresh state; mechanical behavior.*

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I. Introduction

For ten years plant fibers have entered in the field of polymer matrix composites for weight saving needs. Their valuation now extends to the field of civil engineering because of their good specific mechanical properties [1], renewability, biodegradability and low environmental impact. Thus, their use in construction opens new opportunities for producers.

On the other side, the highly hydrophilic nature of plant fibers [2, 3] creates some problems among which the most crucial are the low workability and lack of homogeneity of the cement-fiber mixture [4]. Furthermore, cement hydration and setting are disrupted because of the presence of polysaccharides at the fiber's surface [5, 6], which is incompatible with the desired applications. These two major drawbacks could influence the mechanical properties of the composite [7]. It is therefore necessary to apply treatments to the fibers to improve their hydric behavior and reduce exchanges with the cement matrix.

The objective of this study is the investigation of treatments aimed at enhancing the compatibility of the fibers with the cement matrix. They consist mainly in the coating of the fibers with cement or lime, optionally after pretreatment in boiling water. The effect of these treatments on the water intake of the fibers is evaluated. Their effects on the properties of the composites in the fresh and hardened states are also evaluated by measuring the consistency, the heat of hydration and the initial setting time and by flexural tests correlated with scanning electron microscopy (SEM) observations.

II. Experimental Study

2.1 Materials

Flax used in this study were cultivated in Normandy in 2010 and provided by Depestele group in the form of short fibers 2.5 cm long. An ordinary Portland cement CEM I 52.5 and a rolled sand were used in accordance with European standard NF EN 197-1 [8] and with French Standard XP P 18-545 [9], respectively.

The fibers were added to the cement/sand/water mixture as partial replacement of sand at a level of 2 vol. %. A mixture of the sand, cement and water (1350 ± 5 g/l, 450 ± 2 g/l and 225 ± 1 g/l respectively) was also prepared to provide the control mortar [10]. Standard prismatic specimens of 4x4x16 cm in size were used for the measurements in the hardened state. They were demolded after 48h and were kept in a cure condition at 20 ± 2 °C and 50 % RH. We could not immerse mortar samples including fibers in water due to the hydrophilic behavior of flax fibers.

2.2 Treatments applied to the flax fibers

2.2.1 Coating with hydraulic binders

Flax fibers were coated with two hydraulic binders which are CEM I 52.5 R cement grout or hydraulic lime grout prepared with the following mass ratio: water/hydraulic binder = 1 and fiber/hydraulic binder = 2/3 [11]. The obtained fibers are called respectively CF and LF. These treatments are chosen due to their low hydrophilicity and their chemical resistance in cement environment, with the aim to reduce water intake of the fibers.

The fibers were mixed with the chosen grout in a mortar mixer for 2 minutes and stored for 28 days in a storage room at 20 °C and 98% RH.

2.2.2. Pretreatment with boiling water

We have also implemented the pretreatment of flax fibers with boiling water [3] with the aim to clean them prior to coating with hydraulic binders. We expect a synergistic effect between the two treatments. Practically the fibers are first cleaned with boiling water, then coated with cement or lime grout. The obtained fibers are respectively called WCF and WLF.

2.3. Testing methods

2.3.1. Water absorption coefficient

In order to evaluate the effect of fibers treatments on their water intake, we have measured the water absorption coefficient of raw and treated fibers. [2]. We have first dried 5 specimens of 1g of flax fibers at 60°C until constant weight. Fibers were then immersed in water for 5, 15, 30 minutes and 24 hours. We have then superficially dried them with paper towels and measured their masses. The water absorption coefficient was calculated with formulation (1):

$$\%\,\text{Absorption} = \frac{\text{saturated mass} - \text{dried mass}}{\text{dried mass}} \times 100 \quad (1)$$

2.3.2. Vebe test

Vebe time was measured for all mortar formulations, namely the control mortar (CM), the reference composite (composite with raw fibers, called RFM), the mortar with cement coated fibers (CFM), the mortar with lime coated fibers (LFM), the mortar with boiling water pretreated fibers and coated with cement or lime (WCFM and WLFM, respectively), using a consistometer in accordance with NF EN 12350-3 standard [12]. Unlike the mortar maniabilimiter test, this test is suitable for low-workability cementitious materials. However, we have also measured vebe time of the CM for comparison to other formulations.

2.3.3. Hydration and initial setting time

The influence of raw and treated fibers on the cement hydration was assessed using a semi-adiabatic calorimeter in accordance with NF EN 196-9 standard [13]. This test was implemented for 48h on plain mortar samples or mortar filled with raw or treated fibers. Hydration heat of the cement contained in the sample (expressed in J/g) is the sum of the heat accumulated in the calorimeter and the one dissipated to the ambient environment along the test duration t.

The initial setting time of each formulation was measured with a Vicat apparatus in accordance with the French standard NF P 15-431 [14].

2.3.4. Flexural tests

The hardened materials were tested when aged 7, 14, 28 or 90 days. Flexural tests were run on an Instron 5566, equipped with a 50 kN capacity load cell in accordance with NF EN 196 1 standard [10]. The loading rate was 50 N/s.

III. Results And Discussion

3.1. Water absorption coefficient of raw and treated fibers

Water absorption coefficient of raw and treated fibers is presented in Fig. 1. A significant decrease is noted for all treated fibers compared to raw fibers. It is -35 % for cement coated fibers, -45 % for those coated with lime and -50 % for those pretreated in boiling water and then coated with cement or lime grout. Hydraulic binders have thus acted as to create a barrier which isolates the fiber and reduces its water affinity. This effect is more pronounced when the fibers have already been cleaned up in boiling water; fibers free of impurities are likely to react better under the effect of hydraulic binders.

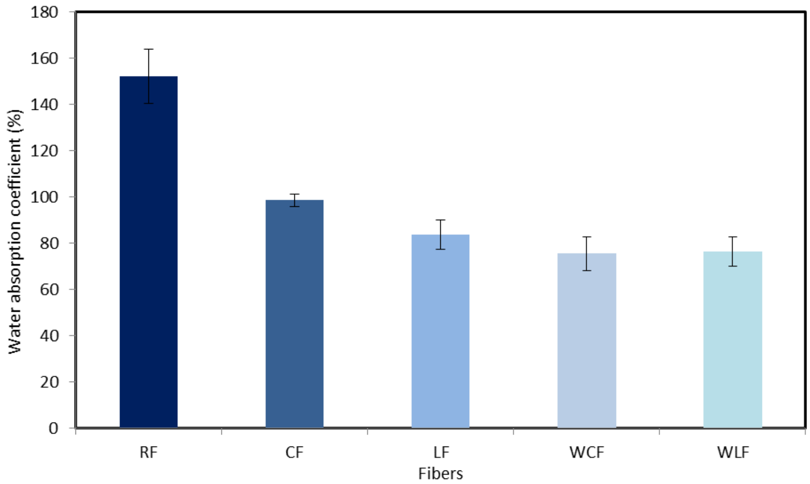


Fig.1. Water absorption coefficient of raw and treated fibers

3.2. Consistency of the cement mixtures

Vebe time measurements obtained for the cement mixtures are recapitulated in Fig. 2. Table 1 shows consistency classes according to the NF EN 206-1 / CN [15].

A Vebe time of 35 s was obtained for RFM, thus this mixture cannot be classified in the frame of this measurement method [15]. However, mixtures including coated fibers have classifiable consistency. The improvement is 77 % for CFM and LFM, and 80 % for WCFM and WLFM. The four formulations are in V3 class. The obtained consistency results are in accordance with the trend of water saturation rate of the coated fibers.

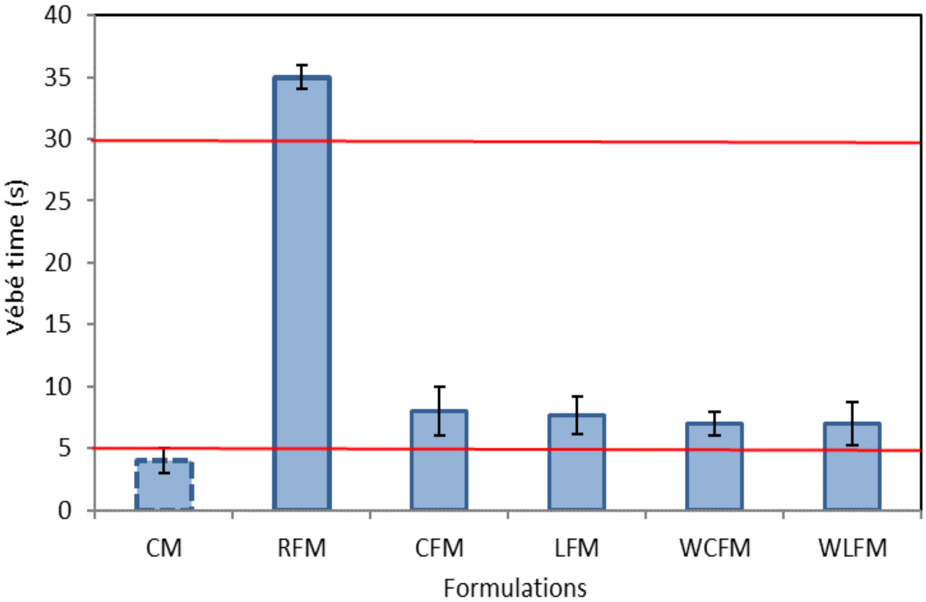


Fig.2. Consistency of cement formulations

Table1. Consistency classes defined from Vébé time measures [15]

Consistency class	Vébé time (s)
V0*	>= 31
V1	30 - 21
V2	20 - 11
V3	10 - 6
V4*	5 - 3

*The consistency cannot be characterized in the frame of this measurement method.

3.3 Hydration and initial setting time

Initial setting times of the different formulations are presented in fig. 3. Compared to CM, the initial setting time of the reference composite RFM is increased by about four times. This could be explained by the trapping of calcium ions by the non-cellulosic polysaccharides, in particular by pectins [16]. These ions are important for the cement hydration and for the formation of hydrated calcium silicate gel (C-S-H) [17].

Compared to RFM, initial setting time of mortars with coated fibers is significantly decreased. This can be explained by the fact that the coated fibers are previously saturated with calcium ions, which reduces their tendency to fix those of the fresh cement paste, hence the acceleration of the setting of CFM, LFM, WCFM and WLFM. This effect is slightly greater when the fibers are first treated in boiling water (case of WCFM and WLFM compared to CFM and LFM, respectively): boiling water allows partial elimination of non-cellulosic polysaccharides [18].

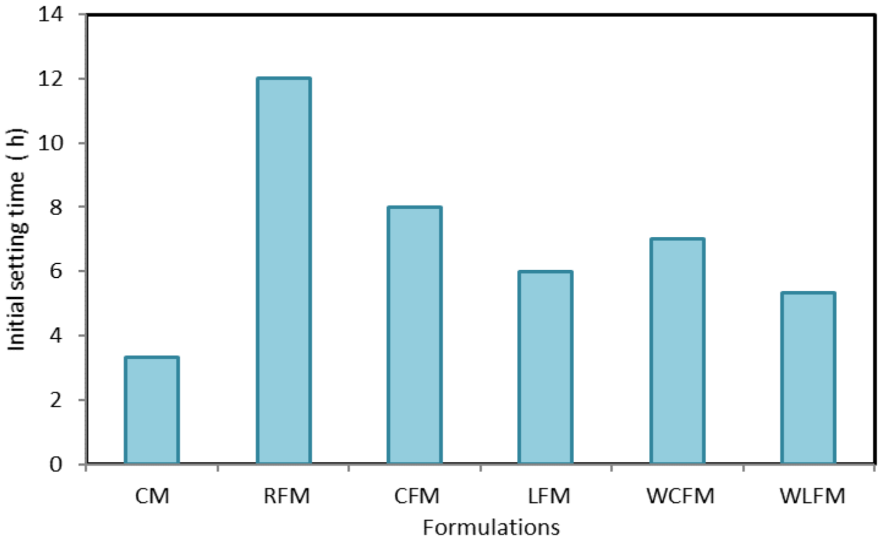


Fig.3. Initial setting time of the different formulations

In order to further investigate the impact of raw and treated fibers on the process of cement hydration, calorimetric study was performed on the control mortar and the fiber composites. Fig. 4 shows the evolution of heat of hydration of the different formulations for 48 hours. The heat of hydration of the cement paste is drastically reduced when raw flax fibers are added (-28 %). For all other formulations the hydration curve is between the control and the RFM material: -20 % for CFM, -13 % for LFM, -23 % for WCFM and only -2 % for WLFM. These results confirm on the one hand the disruption of the cement hydration and its retardation when raw flax fibers are added, and on the other hand the improved behavior in the fresh state when these experimental treatments are applied.

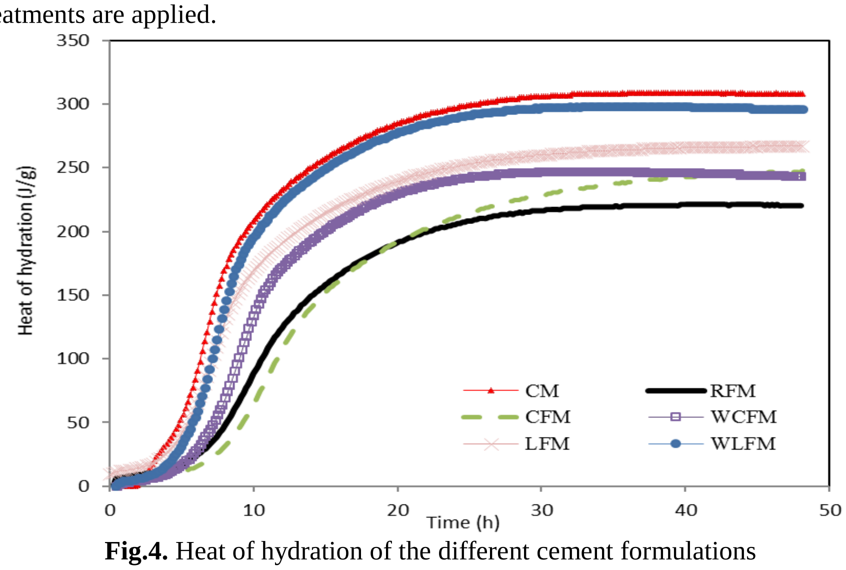


Fig.4. Heat of hydration of the different cement formulations

3.4 Flexural strength

The flexural strength of the different composites increases with the maturation time except for formulations including lime (Fig. 5). For CFM maturity times over 28 days the strength gain is 30 % compared to CM and 14.3 % compared to RFM. SEM images reveal that the fibers are fairly good coated by the cement grout (Fig. 6), which would create a rigid layer around the fibers. On the other hand, the water saturation rate of cement-coated fibers (CF) is decreased by -35 % compared to raw fibers; this could reduce the occurrence of cavities between the fiber and the matrix after water release and thus hinder fiber debonding. SEM images of CFM compared to RFM (Fig. 7) seem to confirm the good fiber-matrix adhesion.

However, flexural strength of LFM was greatly decreased compared to the control mortar CM (-38 % over 28 days and reaches -60 % over 90 days) and to RFM. These results are comparable with those obtained in previous studies [6, 19] and cannot be assigned only to the fibers given the results discussed above. This suggests a negative interaction between the lime and the cement [6]. Despite the positive role of lime on the properties of fresh mixtures, the cured materials have poor mechanical performance, this point is discussed further.

Regarding WCFM and WLFM composites, WCFM exhibits the highest strength whatever the maturation. At 28 days, the improvement is 6.3 % compared to CFM, 21.6 % compared to RFM and 38 % compared to the control mortar CM. However the difference is very low compared to CFM over 90 days of maturation. Thus, the fiber cleaned in boiling water shows a better compatibility with mortar (synergistic effect).

Comparing LFM and WLFM, it appears that the potential beneficial effect of pretreatment of the flax fiber in boiling water is inoperative. This result can be explained in the context of a study of the nucleation and growth of calcium hydrosilicate (CSH) formed during the hydration of tricalcium silicates (C_3S) and dicalcium (C_2S) [20]. This study showed in particular that the calcium hydroxide $Ca(OH)_2$ concentration in solution governs the kinetic of hydration of C_3S and C_2S (which depends on the number of initial nuclei precipitated at the early beginning of hydration) and the mode of growth of the hydrates. The number of germs decreases when the calcium hydroxide concentration increases, while it grows in the presence of sodium chloride or potassium hydroxide. Especially, direct observation by atomic force microscopy showed that the growth of CSH occurs by aggregation of anisotropic nanoparticles laterally and perpendicularly to the surface of the anhydrous silicate [20]. Numerical simulations of experimental hydration kinetics revealed that aggregation is less and less parallel to the surface of calcium silicate grains as the calcium hydroxide concentration in solution increases, whereas in the presence of calcium chloride the growth velocity perpendicular to the surface of the grains increases. The particle aggregation is oriented until the surface of the anhydrous grains is completely covered by the CSH layer. Once the surface is totally covered, CSH formation is governed by a reactive diffusion regime through the hydrate layer and the assembly becomes less organized. Another study showed that the dissolution of C_3S slows progressively as the concentration of $Ca(OH)_2$ increases [21]. From these results, it can be inferred that the initial high concentration of lime in mixtures LFM and WLFM could disrupt the reactivity of C_3S and the kinetic factors governing the formation of CSH, hence the poor mechanical properties of the composites.

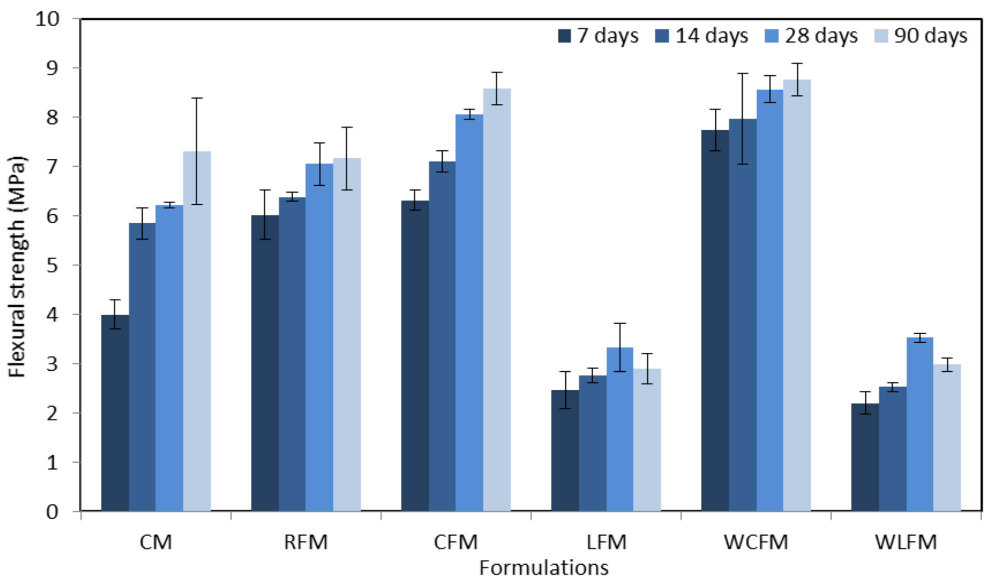


Fig 5. Flexural strength of the different formulations

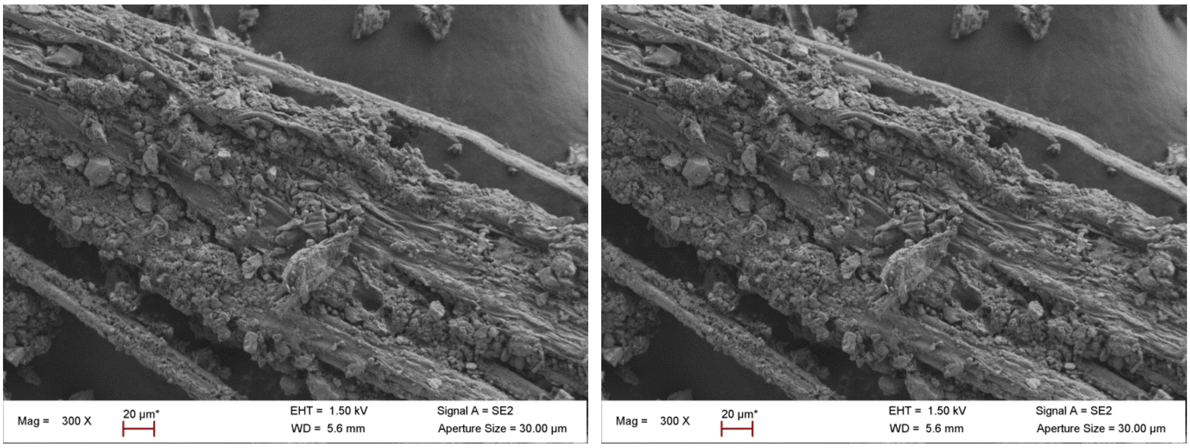
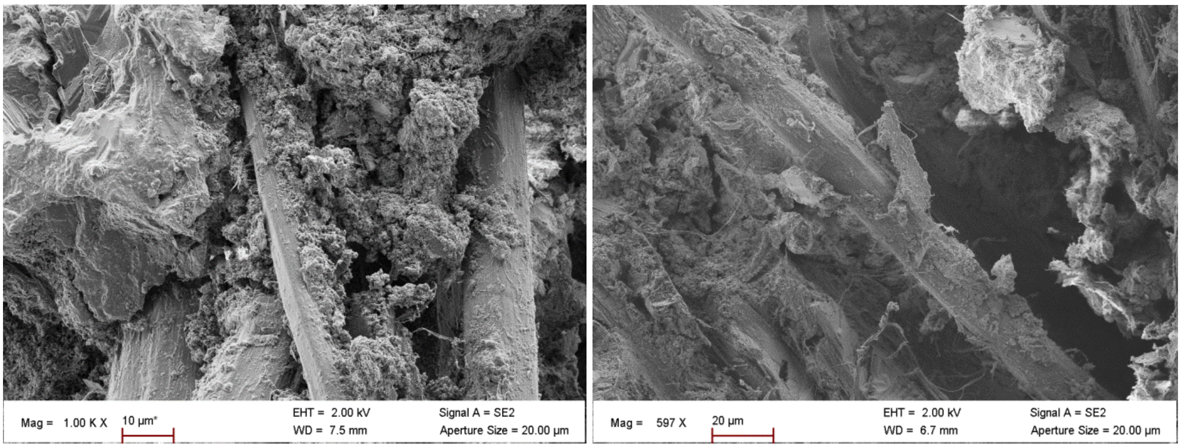


Fig 6. SEM images for CF fibers



(a) (b)

Fig 7. SEM images for (a) CFM (b) RFM

IV. Conclusion

In this work, we have investigated the influence of the nature of hydraulic binder (lime or cement) used as coating of flax fibers (pretreated in boiling water or not) on the properties of fiber-cement mixtures in the fresh state and the mechanical behavior of the obtained composites. The main results are:

- Significant decrease of the water absorption coefficient for all treated fibers was noted: 50 % for the boiling water-treated and cement coated fibers (WCF) and lime-coated fibers (WLF). This correlates with a huge decay of the Vébé time from 35 seconds for the reference mixture RFM up to 7 seconds for mixture WCFM (with WCF fibers) and WLFM (mixture with WLF fibers).
- Important improvement in the cement hydration correlates with a reduction of the initial setting time for all cement mixtures incorporating treated fibers compared to raw fiber mixture (RFM). Coating the fibers with hydraulic binders avoids consuming the calcium ions of the fresh paste, and therefore contributes to improve its hydration. Cement mixtures incorporating cleaned fibers by prior treatment in boiling water and then coated with hydraulic binders (WCFM and WLFM) show the shortest initial setting time.
- Appreciable increase in the flexural strength of composites incorporating cement treated fibers (CFM and WCFM) is noted for all maturation times, and that is corroborated by SEM images. However, despite the improved behavior in the fresh state of the mixtures incorporating lime treated fibers (LFM and WLFM), low flexural strengths are obtained for the composites due to poor hydration that we assigned to the lime content.

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