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Improvement of the Behavior of Cement Mortar by Vinegar Treatment of the Flax Fibers Reinforcement

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Abstract

The association between plant fibers and a cement matrix generates different problems, in the fresh state and in the hardened state of the material. Several studies propose to treat the plant fibers with chemical, physical or thermal treatments. The aim of this work is to ameliorate the behavior of a cement mortar incorporating short flax fibers by treating them with white vinegar which is a non-polluting product. This natural treatment was chosen in order to clean the fiber's surface by partial removal of the non-cellulosic compounds known to deeply disturb the behavior of the material in the fresh state especially in terms of consistency and setting time. Two treatment's immersion time were tested, 2 and 24 hours. To evaluate the efficiency of this treatment, tensile, thermal and water absorption tests were conducted on treated and raw fibers. Different mortar formulations (Control mortar, Mortar with raw fibers and mortar with treated fibers) were prepared and characterised in the fresh and hardened states. Results show a significant reduction of the water absorption of the fibers and an increase of their average tensile strength. The properties of the cement composites have also been improved in terms of consistency, initial setting time and mechanical performances.

Keywords

Cement Mortar, Flax Fibers, Vinegar Treatment, Fresh State, Flexural Behavior

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1 Introduction

The use of plant fibers in building materials is the subject of several studies. Indeed, plant fibers are characterized by several qualities such as their natural and renewable character, their lightness and their specific high mechanical properties which are comparable to those of glass fibers [1, 2]. However, the development of plant fibers reinforced cement materials requires the use of fibers treatments and/or cement matrix additives to mitigate their incompatibility points. These latter mainly lead to two issues. First, the mixture's workability is reduced due to the hydrophilic nature of the fibers [3, 4]: they absorb a large amount of the mixing water. Second, the cement hydration reaction is disturbed, leading to a considerable delay of the initial setting time [5]. The objective of the treatments is therefore to ameliorate the fresh state properties of the cement mixtures in order to ameliorate their flexural behavior in the hardened state.

Different plant fiber treatments have been studied in previous works. NaOH treatment has been probably the most experimented [6, 7], and its ability to partially remove impurities and non-cellulosic polysaccharides known to degrade the properties of cementitious materials in the fresh state were evidenced. Besides, it has been shown that after

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treatment the fibers are chemically more homogeneous and stiffer, which improves their adhesion with the cement matrix. However, the treatment's effect is strongly related to NaOH concentration and to the application temperature [8]. Tolêdo Filho et al. [9] have investigated the effects of coating sisal and coconut fibers with a slurry of silica fume. They reported an improvement in the cement hydration and the mechanical properties of the cement composite. In the study of Francis et al [10], wood pieces were treated with sodium silicate solution which makes them inert and prevent their interaction with the cement matrix.

In this work, white vinegar is used to clean flax fibers for cementitious mortar reinforcement. The efficiency of this treatment is evaluated by its action on the properties of the fibers alone and on those of the cementitious mixtures and the composites in the hardened state.

2. Materials and Tests

2.1. Raw Materials

Flax plants were cultivated in Normandy (France). 24 mm long fibers were provided by Depestele Company. An amount of 2 vol. % was added to the mortar as partial replacement of the sand. Portland cement CEM I 52.5 N was selected in accordance with the European standard NF EN 197-1 [11] as well as a dry sand of particle size 0/4 mm. Chryso Fluid Premia 205 superplasticizer was used in all formulations. Mixing water was adjusted for the composites to consider the water absorption of the fibers. Standard prismatic specimens of sizes 4×4×16 cm^3 were used for the mechanical tests. They were demolded after 48 h because of the hardening delay due to the presence of the fibers and cured in a climate chamber at 20 ± 2°C and 50% H_R until mechanical testing.

2.2. Fibers Treatment

White vinegar is a natural product, industrially manufactured by acetification of beet alcohol or maize: transformation of ethanol into acetic acid. It could be also obtained by simply diluting pure acetic acid until obtaining of a concentration between 6 and 8%. It is mainly used as a cleaning and descaling product thanks to its performances. Unlike NaOH which is one of the most popular fibers surface treatment, white vinegar is non-polluting to the environment. It's used here with the aim to clean the fibers surfaces from its impurities (fats, waxes…).

White vinegar solution of 7% acidity was used for the fibres treatment. This treatment has the advantage of being natural. It is chosen with the aim to clean the fiber surface from organic impurities. Fibers were first immersed for different times in the white vinegar solution, then rinsed with tap water and dried in an oven at 60°C for 48 h. Two immersion times (2 and 24 hours) were chosen and the fibres are referenced FT2 and FT24, respectively, while the raw fibers are noted RF.

2.3. Fibers Characterization

2.3.1. Tensile Test

The tensile tests were carried out on 22 unit fibers using the protocol defined in the standard NF T25-501-2 [12]. They were performed using a universal testing machine (Instron 5566) equipped with a 10 N capacity load cell, at a crosshead displacement rate of 1 mm/min. The tensile test was conducted only on FT2 fibers.

2.3.2. Thermal Properties

The measurement of the thermal conductivity and the heat capacity of flax fibers was conducted in accordance with the French Standard NF EN 12664 [13]. The fibers sample was placed in a $20 \times 20 \times 6$ cm^3 polystyrene mold to minimize heat exchanges with the outside. The test was carried out on raw and treated fibers.

2.3.3. Water Absorption Test

Water absorption rate was determined for raw and treated fibers. For each type of fibers, 5 samples of mass 1g were immersed in water for 5, 15, 30 minutes or 24 hours. At the end of the immersion time, the fibers were superficially dried with paper towels and weighted for the wet mass. The water absorption rate was calculated using relation (1):

$$\%\,Water\ Absorption = \frac{\text{wet mass} - \text{dried mass}}{\text{dried mass}} \times 100 \qquad (1)$$

2.4. Composites Characterization

2.4.1. Consistency Tests

The consistency of a mortar refers to its ability to flow on a horizontal plane when subjected to a vibratory stress. Consistency of the cement mixtures made with raw fibers (RFM), treated fibers for 2 hours (FT2M) or 24 hours (FT24M) and the control mortar (CM) was measured in accordance with the French standard NF EN 1015-3 [14]. A truncated conical mold is placed on the flow table with internal diameter of 100 mm at the bottom and 70 mm at the top. The standard recommends the filling of the mold in 2 equal layers with the mortar. After spreading each layer, 10 taps are made in the dough to ensure uniform filling of the mold. The mold is then removed gently and the flow table is jolting 15 times at a constant frequency of approximately one per second. The average spreading diameter is calculated using two measured values at the base of the dough in two orthogonal directions.

2.4.2. Initial Setting Time

To evaluate the effect of the fibers treatment on the initial setting time of the cement mixtures, treated or raw fibers were immersed in tap water for 48 hours with the same water/fiber ratio used during the preparation of the cement mixtures. The water containing water-soluble compounds issued from the fibers was then used to prepare cement pastes. A control cement paste (CP) was prepared with tap water. Initial setting time is then measured with a Vicat apparatus in accordance with Afnor standard NF EN 196-3 [15]. This protocol is followed because it is difficult to make measurements of setting time on cement fiber mixtures: the presence of fibers prevents the penetration of the Vicat needle and could lead to false results. The paste made with raw fibers and their water-soluble compounds is called RFP, those made with two hours or 24 hours vinegar treated fibers and their water-soluble compounds are referenced FT2P and FT24P, respectively.

2.4.3. Three-Point Bending Tests

Three-point bending tests were implemented on mortar samples cured for 7, 14 and 28 days using a Shimadzu testing machine equipped with a 50 kN capacity load cell. The loading rate was 50 N/s.

2.4.4. Thermal Properties

The thermal conductivity of the composites and the control mortar were measured with the same method used for the fibers. The tests were carried out on specimens with dimensions 20 × 20 × 6 cm^3 after 28 days of cure.

3. Results and Discussion

3.1. Effect of White Vinegar Treatment on the Fibers Properties

3.1.1. Mechanical and Thermal Properties

The tensile strength was investigated on RF and FT2 fibers. The results are displayed in Table 1. We have noted significant increases in the average tensile strength and Young's modulus accompanied by significant reduction of ultimate deformation following two hours vinegar treatment. Moreover, for all these parameters the standard deviations are substantially reduced. The partial removal of the amorphous compounds from between the microfibrils in the fiber would have increased the crystallinity and thus ameliorated the load transfer between them. This results in lower variability. The decrease in the diameter of the fibers and their rigidification can contribute significantly to the reinforcement of the mortar if the fiber / matrix interface is not degraded by the vinegar treatment.

Table 2 presents the thermal characteristics of the fibers. There is a slight increase in the thermal conductivity of the fibers following the vinegar treatment, which is attributed to their densification.

Table 1. Mechanical properties of raw fibers and 2 hours treated fibers.

	Diameter (µm)	Young's modulus (GPa)	Ultimate strain (%)	Tensile strength
RF	22.75 ± 6	44 ± 21	1.78 ± 0.6	849 ± 482
FT2	21.6 ± 4	69 ± 12	1.2 ± 0.2	969 ± 255

Table 2. Thermal properties of raw fibers and vinegar treated flax fibers.

	Thermal conductivity (W/m °C)	Heat capacity (J/Kg °C)	Apparent density (kg/m^3)
RF	0.041	1661	1045.2
FT2	0.049	1676	1177.2
FT24	0.044	1659	1122.1

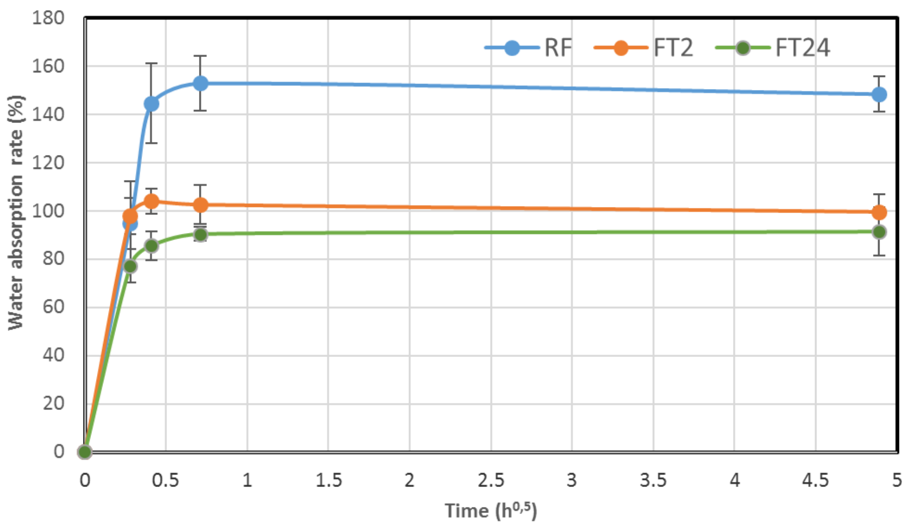

Figure 1. Water absorption rate of raw fibers and vinegar treated fibers.

3.1.2. Water Absorption Rate

The water absorption kinetics were investigated for the raw and the treated fibers. When immersed in water, raw fibers can absorb up to 150% of their dry mass in less than one hour [16]. The kinetics of water absorption are identical for RF and FT2 fibers, but for the latter the water absorption saturates at much lower rate (99%) i.e. 33% reduction compared to RF. After a more prolonged vinegar treatment (for FT24), the hydration kinetics is lower and the saturation occurs for an absorption rate of 91% i.e. 39% reduction relatively to RF. The vinegar treatment reduced the affinity of the fibers with water by partial removal of hydrophilic compounds, the water saturation time is the same for all fibers (30 minutes).

3.2. Effect of White Vinegar Treatment on the Composite Properties

3.2.1. Consistency

Relative flow of the different formulations are presented in figure 2. The control mortar CM showed a relative flow of 107.2% which is an important value due to the use of a superplasticizer. For RFM, the relative flow was almost non-existent. Thus the flow table test did not allow spreading RFM. Mixtures incorporating vinegar treated fibers spread out much better than RFM: the improvement is about 21% for FT2M and 35% for FT24M. This is correlated with the reduction of the water saturation rate of the vinegar treated fibers.

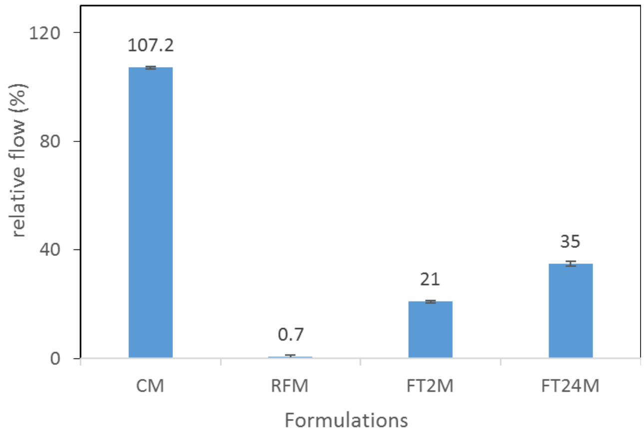

Figure 2. Consistency of the different formulations.

3.2.2. Initial Setting Time

Figure 3 presents initial setting times of the different cement pastes. The initial setting time obtained for the paste with raw fibers was almost twice that of the control paste. This delaying effect of the setting is explained by the chelating of calcium ions by non-cellulosic polysaccharides, especially the pectins [17]. Indeed, calcium is needed for the progress of the cement hydration reaction. The delay is reduced after the white vinegar treatment (almost 10% for FT2P and about 22% for FT24P. These results confirm the effectiveness of vinegar treatment to reduce the rate of non-cellulosic polysaccharides responsible for the delay.

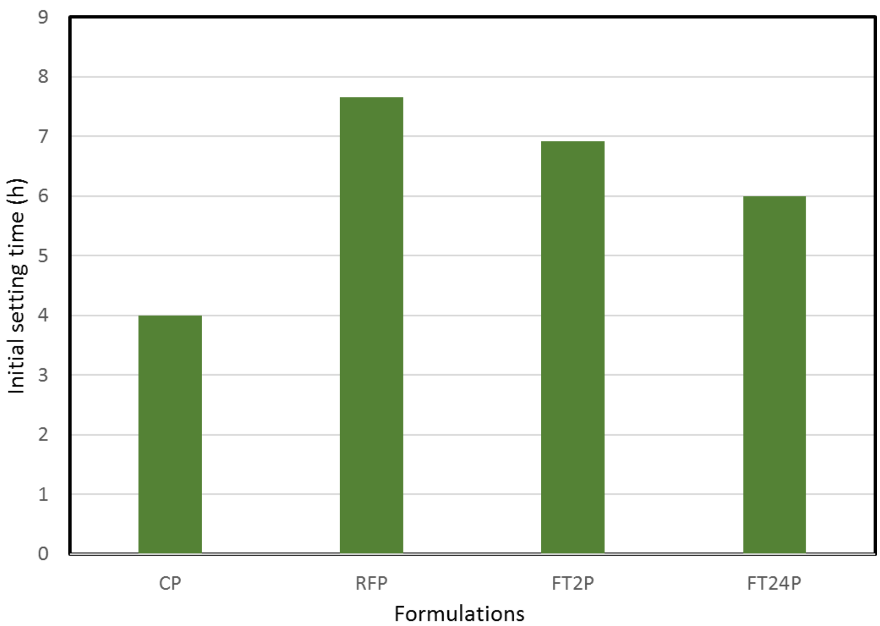

Figure 3. Initial setting times of the different cement pastes.

3.2.3. Flexural Behavior

The comparison of CM and RFM bending loading curves (Figure 4) shows the same linear behavior up to the catastrophic failure point of CM. Beyond this point there is a non-linear behavior persists for RFM up to the maximum load. The ensuing controlled failure (post-peak behavior) reflects fiber bridging of the macrocrack.

The three-point bending curves of the fibered mortars are reported in figure 5. The implementation of the white vinegar treatment modifies even more the composites behaviors:

1) The composite made with 2 hours vinegar treated fibers (FT2M) shows the same stiffness as RFM but the first matrix crack appears for a lower displacement while the post-peak behavior is larger (Figure 4). These observations could be explained by a more efficient mechanical adhesion of the cement to the fibers following the vinegar treatment (the fibers are rougher).

2) The increase in the vinegar treatment time induces a higher strength with no noticeable increase in rigidity, but a more pronounced post-peak behavior (Figure 5). This reflects a more efficient load transfer capability from the matrix to the fibers due to a more efficient interface. Thus the increase of the treatment time from 2 hours to 24 hours seems to make it more efficient.

3) The strength of the different mortar formulations normally increases as a function of the curing time (Figure 6). At a given time, the strength is the same for RFM (untreated fibers) as FT2M (treatment for 2 hours). The beneficial effect of the vinegar treatment is perceptible for the formulation FT24M (treatment for 24 hours). After 28 days of cure, the increase in strength is 12% compared to RFM and 22% compared to the control mortar CM.

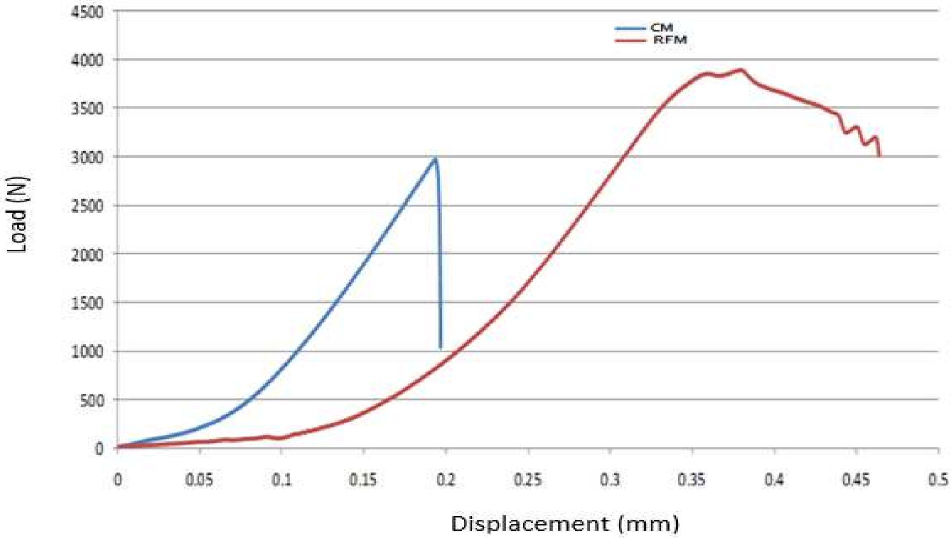

Figure 4. Three-point bending loading curves of the control mortar and the RFM reference composite.

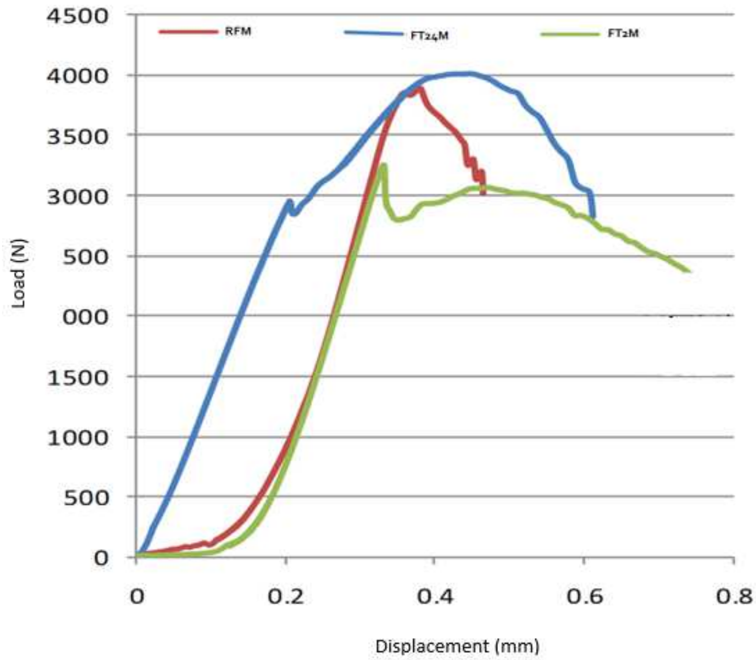

Figure 5. Three-point bending loading curves of the fibered mortars highlighting the effect of vinegar treatment.

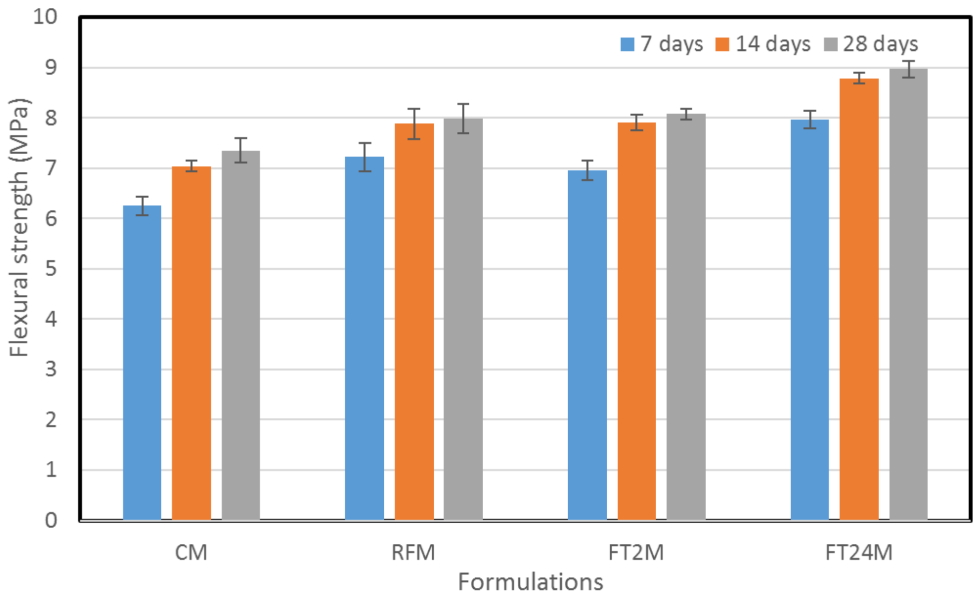

Figure 6. Effect of the white vinegar treatment on the flexural strength.

3.2.4. Thermal Properties

The thermal conductivity of the different formulations is shown in figure 7. Globally, the addition of flax fibers significantly reduces the thermal conductivity of the mortar. The reduction rates relatively to CM are 55%, 63% and 65% for RFM, FT2M and FT24M, respectively. Decreases noted are correlated to the first order at lower densities of the composites compared to CM. The higher conductivity for RFM when its density is lower than FT2M and FT24M would be explained by a higher porosity of the fibers after the vinegar cleaning.

Table 3. Thermal conductivity and density of the composites and the control mortar.

Formulation	Thermal conductivity (W/m °C)	Density (kg/m^3)
CM	1.261	2213.0
RFM	0.568 (-55%)	2077.54
FT2M	0.471 (-63%)	2137.77
FT24M	0.446 (-65%)	2146.26

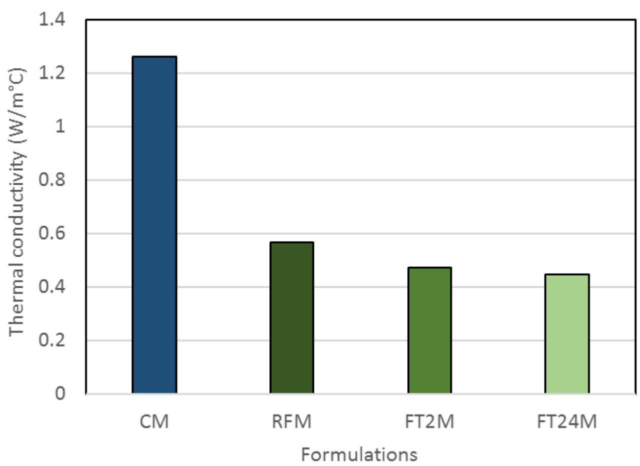

Figure 7. Conductivity of the composites compared to CM.

4. Conclusion

In this work, white vinegar treatment was implemented with the aim to clean the flax fibers from their surface impurities and thus to ameliorate the behavior of the cement composites. The efficiency of the treatment for two immersion time was evaluated on the fibers properties, on fresh state properties of the cement composites and on their flexural behaviour in the hardened state.

The partial elimination of the hydrophilic compounds of the flax fibers following the vinegar treatment made it possible on the one hand to considerably reduce their affinity to water, on the other hand to raise their strength and Young's modulus. Thus the consistency and initial setting time of fibred mortars made with these treated fibers were ameliorated. In addition, this treatment improves fiber/matrix adhesion and promotes efficient load transfer between the fibers and the matrix. Three-point bending test results showed that the beneficial impact of the vinegar treatment on the strength is noticeable only when applied for at least 24 hours, but the post-peak behavior is pronounced even after only 2 hours of treatment. Addition of 2 vol.% of fibers to the cement mortar reduces thermal conductivity up to 65%.

The properties in the fresh state are little improved when the duration of vinegar treatment is increased from 2 to 24 hours (11% for the consistency and 13% for the initial setting time), but the two behaviors in 3-point bending are significantly different. Natural ageing tests will be performed on these composites in order to evaluate the merits of these vinegar treatments.

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