Natural fiber reinforced cement mortar composite physico-mechanical properties: from cellulose microfibers to nanocellulose

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Abstract

The use of asbestos in building materials is a risk to human health and this fact has driven the interest in utilizing other natural fibers, such as cellulosic fibers, in cement based building materials. In the literature, some authors studied cement based composites reinforced with cellulose microfibers, other authors studied cement based composites reinforced with nanocellulose. However, to the best of our knowledge, in the literature, there is not any study where the effect of cellulose fiber reinforcement, starting from raw natural fiber to nanocellulose, was studied. On the other hand, the comparison of literature data of cellulose reinforced cement composites is difficult since there are many variables (matrix, cement, sand, the dosification, w/c, superplasticizer, the fabrication method) that affect the final composite performance. In the current work, the effect of cellulose fiber reinforcement with different scale (micro and nano) on cement based composites properties was studied starting from raw natural fiber to nanocellulose using the same variables as well as fabrication method. After the addition of microfibers, the strength values of mortar decreased with respect to plain mortar, the reduction being higher as the fiber content was increased. On the other hand, after the addition of nanocellulose fiber, the density value hardly changed respect to unreinforced mortar. Moreover, contrarily to microfibers addition, the presence of 0.25 wt% nanocellulose in mortar slightly increased the flexural strength. On the other hand, mechanical properties obtained in the current study were compared with literature data for similar systems.

Keywords: cement, mortar, natural fiber, nanocellulose, mechanical properties

Introduction

The use of asbestos in building materials is prohibited due to its carcinogenic properties (Ruers and Schouten 2005) and consequently it has been increased the interest in utilizing natural fibers in cement composites. Roma, Martello, and Savastano (2008) concluded that the roofing tiles reinforced with vegetable fiber were acceptable as substitutes of asbestos-cement sheets that are still in use in several developing countries. Composites with vegetable fibers are important for construction of inexpensive buildings in developing regions of the world (Tonoli et al., 2011). Some advantages of natural fibers are that they are energy efficient, economical and ecofriendly (Dawood and Ramli, 2012) materials. The production of vegetable fibres requires little energy and plant absorb CO_2 from the air for photosynthesis process, and oxygen is given back to the environment, during plant grow. Regarding the end life of natural fibers, in contrast to the most synthetic fibers, they are biodegradable materials. In the current work, even though the amount of natural fibers incorporated to the mortar composites is very low, between 0.9-2.7% respect with the all composite weight, the sum of little changes like this contribute to a more sustainable world. It has been demonstrated that the addition of fique fiber into Portland cement, is appropriate for low cost housing applications (Delvasto et al., 2010). Silva et al. (2010) demonstrated the potential of long aligned sisal fibers as reinforcement in cement based laminates for semi-structural and structural applications. They observed that the material reinforced with sisal fibers presented a multiple cracking process with a strain hardening behavior. However, the industrial production of cement-based composites reinforced with vegetable fibers is currently limited by the lack of durability of these materials (MacVicar, Matuana, and Balatinecz, 1999; Toledo Filho et al., 2003). The reduction of mechanical properties of cement composites with lignocellulosic fibers is attributed, mainly, to the damage caused by the basic medium of cements on the fibers and fiber/matrix adhesion (Ardanuy et al., 2011; Savastano et al., 2009). In the alkaline medium of ordinary Portland cement, the lignocellulosic fiber components such as lignin and hemicellulose are degraded (Toledo Filho et al., 2003; Savastano, Warden, and Coutts, 2003a). One way to improve the durability of vegetable fiber reinforced cement composites could be the removing of the hemicellulose and lignin compounds from fibers. In the literature, it was observed that after pulping process the fiber resistance to alkaline attack was improved (Savastano, Warden, and Coutts, 2003a) and it was suggested that cementitious products reinforced with short fibers or pulp were more suitable for non-structural applications (Silva et al., 2010).

The research about composite materials reinforced with nanoscale reinforcements has gained increasing attention due to nanoscale reinforcement outstanding properties. One advantage of incorporating nanocelluloses to cement based systems is that they are easily dispersible in water (Claramunt et al., 2019). In the literature there are several studies where nanocellulose was added to cement based materials (Ardanuy et al., 2012; Cao et al., 2016; Cengiz, Kaya, and Bayramgil, 2017; Claramunt et al., 2019; Hisseine et al., 2019; Parveen et al., 2017). The incorporation of nanomaterials to cementitious materials could retard the growth of cracks at nanoscale, resulting in improvements in fracture performance (Parveen et al., 2017).

In the current work microfibers, raw sisal fiber and cellulose pulp, and nanocellulose, isolated from pulp, are used as reinforcements in cement based materials. The micro and nano cellulose were added to cement mortars with the aim to study the effect on physico-mechanical properties of cement mortar.

Experimental part

Materials

Sisal fiber bundles (*Agave sisalana*) and bleached sisal pulp were kindly supplied by Celulosa de Levante S.A. (Tortosa, Spain). Sisal fibers were chopped by the cutting mill SM200 (RETSCH, Hann, Germany) using a mesh size of 8 mm (Figure 1). Sisal fiber tensile strength varied from 325 to 366 MPa depending on the fiber length (Orue et al., 2015, 2016).

Bleached sisal pulp was obtained by cooking sisal fibers using NaOH-anthraquinone and the obtained product was bleached with a totally chlorine free process. Bleached sisal pulp was chopped by the cutting mill SM200 (RETSCH, Hann, Germany) using a mesh size of 8 mm.

Nanocellulose fibers were isolated from bleached pulp using the following chemo-mechanical procedure. Firstly, chopped bleached pulp was treated with a solution of 7.5 % NaOH for 1.5 h at boiling temperature and the resultant pulp was filtered and washed adding distilled water. After this, pulp was treated with a mix of nitric acid and acetic acid (9:1 volume ratio) for 90 min under vigorous stirring with a solid-to-liquid ratio of 0.03 g/ml. After diluting the acid solution with water, pulp suspension was submitted to vigorous stirring with a dispermat to individualize nanofibers. The consistency of nanocellulose suspension obtained after the isolation procedure was of 0.25%. In Figure 1c and 1d, a photo of the nanocellulose suspension in water and an atomic force microscopy image of nanocellulose are shown, respectively. Nanofibers have around 30 nm in diameter and the length could be of several micrometres, being the aspect ratio value high.

Here insert Figure 1

Materials preparation and characterization techniques

UNE-EN 197-1:2000 CEM II/A-L type cement supplied by FYM italcementi group was used in the current research. The cement is composed by Portland 80-94%, limestome 6-20% and minor components 0-5%. The commercial AF-T-0/1-C sand was supplied by Canteras de Alaiz, S.A. company. The sand used is adequate for mortar according to UNE-EN 13139 Spanish standard, being fine aggregate sizes 0/1 mm. The sand is composed by 98% of calcium carbonate.

Preparation and mechanical characterization of mortar composites was carried out based on UNE-EN 196-1 Spanish standard. Prismatic specimens of 40x40x160 mm^3 volume of mortar composites were prepared using the mould type UNE-EN-1:2005. The cement:sand:water mass ratio used for cement mortar composites was 1:3:0.5. Different raw sisal fiber and pulp fiber contents, 4, 8 and 12 wt%, respect to cement content, were incorporated to cement mortar. On the other hand, nanocellulose content varied from 0.1 to 0.5 wt%, respect to cement amount. To use the same water grams and maintain w/c ratio of 0.5 in all mixes, the consistency of starting nanocellulose suspension was modified for different nanocellulose contents, adding or evaporating water. The consistency values used were in the range of 0.2-1%.

The cement mortar composite preparation procedure differs when micro and nano fibers were used. When micro fibers were used, the necessary cement and water amount was put in the mixing bowl and the mixing started at low speed for 30 s. Afterwards, the sand was incorporated and the blending speed was up and the blend was mixed for 30 s. The dough mixer was stopped for 90 s and during this step the raw sisal fibers or pulp fibers were incorporated, further mixing was carried out at high speed for 60 s. On the other hand, when nanocellulose fibers were used, they were incorporated in the first step, i.e. when the cement and water were put in the bowl. After mixing process, formulations were cast in moulds and they were compacted on an impact compactor. All the specimens were hold in moulds for 24 h at room temperature, after this, they were demoulded and dipped into water to cure at 22 °C for 27 days. Physical and mechanical properties were characterized after 28 days age.

A three-point bend configuration was employed in the determination of maximum load. A span of 100 mm and a rate of 50 ± 10 N/s were used. Prismatic specimens of 40x40x160 mm^3 volume of mortar composites were prepared using the mould type UNE-EN-1:2005 and six specimens were tested using a three-point bend configuration.

On the other hand, compressive strength was determined using 6 semiprismatic specimens of around 40x40x80 mm^3 volume obtained after the flexural test and a rate of 2400 ± 200 N/s was used. For all tests an Ibertest model C18.200.MDA universal testing machine was used and the flexural (R_f) and compressive (R_c) strengths were calculated using the equations 1 and 2, respectively. In the literature the flexural strength is also called as modulus of rupture (MOR).

$$R_f = \frac{1{,}5 \text{ x } F_{max} \text{ x } l}{b^3} \qquad (1)$$

$$R_c = \frac{F_{max}}{b^2} \qquad (2)$$

Where b is the height as well as width of the specimen in millimeters, l is the span length in millimeters and F_{max} is the maximum force in N unit. A minimum of six specimens were tested and the average values were reported.

Apparent density of cured specimens was determined after drying the specimens in an oven at 100 ºC and assuming that demoulded specimen have 256 cm^3 volume, the values were expressed in kilograms per cubic meter.

$$\text{Density } (\text{kg/m}^3) = \frac{\text{Dried specimen weight}}{\text{Volume of specimen}}$$

Ultrasonic Pulse Velocity Tester model 58-E0048 (Controls™, Cernusco, Italy) was used to measure the ultrasonic propagation through the prepared mortar specimens. To ensure good acoustical contact between ultrasonic pulse velocity tester and specimens, a coupling medium was used and direct transmission measurements were performed by the contact method. First, calibration rod was used to ensure the correct functioning of ultrasonic pulse velocity tester, thereafter specimens after 28 day that were dried for one day in an oven, were used for ultrasonic pulse velocity determination. Three different specimens were used for each system and the average values were reported. The temperature of specimens was around 20 ºC and transmitter and receiver head are 54kHz type.

Results and discussion

Mortar reinforced with cellulosic microfibers

Fractured surface morphology:

Figure 2 shows the fracture surface morphologies of composites reinforced with cellulosic microfibers after flexural test. In red color circles, pulp agglomerations can be observed indicating that the distribution of pulp fibers was poorer in the cement mortar than raw sisal counterpart.

Insert here Figure 2

The starting material of pulp was in form of sheets where fibers created an interconnected network trough different mechanisms such as, interdiffusion, mechanical interlocking, capillary forces, Coulomb forces, hydrogen bonding and Van der Waals forces (Hirn and Schennach, 2015). Based on fractured surface images, it seemed that the separation of single pulp fiber was not achieved and consequently many pulp fiber agglomeration zones were observed. These agglomerations are local fiber concentrations where air could be entrapped and can result in poor crack bridging (Akkaya, Picka, and Shah 2000). On the other hand, even though the raw sisal fibers are homogeneously distributed in the mortar matrix, the fact that many pulled out fibers are observed, indicate that the fiber/matrix adhesion is poor. Similarly, Tonoli et al. (2010b) observed many fibers pulled out from the matrix when they studied fractured surfaces of cement-based composites reinforced with both eucalyptus and pinus fibers. They mentioned that the pull out process contributed to its frictional energy, resulting in the higher toughness of the composite. Savastano, Warden, and Coutts (2003b) examined the fracture surfaces of weathered composites. They observed fiber pullout rather than fiber fracture in all composites suggesting that much energy was dissipated due to fiber pullout mechanism, improving the toughness of the composite.

After the addition of raw sisal fiber and pulp fiber the density value decreased, the reduction being higher as the fiber content was increased (Figure 3a). Similar trend was observed by other authors for cement composites reinforced with cellulosic fibers (Dawood and Ramli, 2012, Savastano, Warden, and Coutts 2003a). Dawood and Ramli (2012) determined physical and mechanical properties of mortar reinforced with different percentages of palm fiber. They concluded that the addition of palm fiber reduced the density of mortar. On the other hand, Savastano Warden, and Coutts (2003a) investigated the performance as reinforcement in ordinary Portland cement (OPC) and chemically activated blast furnace slag (BFS) matrices of fibers obtained from commercial and by-product sisal by thermomechanical pulping and chemi-thermomechanical pulping (CTMP) processes. From density data reported by Savastano, it can be concluded that as the fiber content was increased in the composite then the density value decreased.

Insert here Figure 3

The density of raw sisal fiber is lower than bleached pulp one, since during pulping process amorphous components, hemicellulose and lignin, are removed. However, mortar reinforced with raw fiber showed higher density than mortar reinforced with bleached pulp one. In fractured surface morphology, many pulp fiber agglomerations were observed, these agglomerated fibers were not wetted with cement mortar matrix and probably air was trapped between these fibers during specimen preparation. This fact could be a possible reason for lower density values for pulp fiber reinforced systems than raw fiber counterparts.

Ultrasonic is a non-destructive testing technology that has been widely used to evaluate damage and cracking. In the current study, ultrasonic testing has been used to characterize the change of the material properties caused by the addition of cellulosic fibers to cement mortar matrix. The velocity to propagate of ultrasonic pulse signal in mortar specimens as a function of reinforcement type and loading is shown in figure 3b. After the addition of 4 wt % of reinforcement, the velocity to propagate ultrasonic pulse signal in mortar reinforced with raw fiber is similar to unreinforced system. However, for mortar reinforced with pulp fiber the velocity reduced drastically. For both type of reinforcements, the pulse velocity reduced as reinforcement loading was increased in mortar. In all reinforcement loadings, the pulse velocity was higher for mortar systems with raw fiber than pulp counterparts. It should be mentioned that mechanical properties and ultrasonic wave propagation are influenced by the elastic properties of materials as well as air voids in mortar (Wang et al., 2017). Air voids are defects in mortar that can increase the ultrasonic wave propagation time in mortar. Taking into account density and ultrasonic wave propagation velocity results, it seems that the addition of reinforcements would increase the number of voids in mortar, being this increment more accused in pulp reinforced system than in raw sisal fiber counterpart.

Figure 3c and 3d shows the flexural and compression strength data as a function reinforcement type and loading. After the incorporation of both type of reinforcements the strength values decreased indicating that raw fiber and pulp are not reinforcing the mortar. Contrarily, De Pellegrin, Acordi, and Montedo (2021) highlighted that cellulose fiber addition increased the flexural strength and the modulus of elasticity of mortar composites. Omoniyi and Olorunnisola (2020) suggested that the manufacturing method, fiber content and pre-treatment, and the interaction of these variables had significant effects on the strength properties of the cement-bonded bagasse fiber composites.

As can be observed, the strength values decreased as the reinforcing content was increased. As observed in fracture surface, the fiber/matrix adhesion is poor and fibers are not adhered to cement strongly, probably, close to fiber surface, in the interface, voids or defects are formed. In the interphase region there are porous that can create a gap between the cellulosic fiber and cement based matrix due to shrinkage suffered by fibers during the drying (Savastano and Agopyan, 1999). As fiber loading was increases in the mortar, then higher numbers of voids/defects are formed and consequently the strength value of systems reduced with increasing reinforcement loading. Petrella et al. (2019) added wheat straw to cement mortars and they characterized the prepared composites by means of thermal, acoustic, mechanical, and microstructural analysis. They suggested that the results were strongly dependent on the porosity of the composites. The porosity was ascribed to the straw features and to the voids at the cellulose fibers/cement matrix interface. Therefore, cellulose fibers/cement matrix interface is crucial to develop composites with improved mechanical properties.

In raw sisal fiber, individual fibers are linked to each other by hemicelluloses and lignin. These amorphous compounds are decomposed in an alkali media such as cement mortar and consequently the mechanical properties of cement-based composites with lignocellulosic fibers reduced in a relatively short lifetime (Savastano, Warden, and Coutts, 2003a). Pulped fibers can resist more the alkali media of cement based materials than raw fiber one since non-cellulosic compounds were removed during pulping process. However, comparing both type of reinforcements, mortar reinforced with raw fiber showed higher strength values than pulp reinforced systems. The higher strength values of composites reinforced with raw sisal fibers than pulp reinforced one, can be explained due to a better distribution of vegetable fibers in the cement matrix, which is in agreement with fractured surface morphology observations. The poor dispersion of the cellulose pulp disturbed the efficiency of the matrix reinforcement (Tonoli et al., 2010a). Tonoli et al. (2010b) observed that cement composites reinforced with eucaliptus fibers showed higher mechanical performance than pinus reinforced ones. They suggested that the distribution of eucaliptus fibers in the cement matrix was better than pinus fiber ones and consequently showed improved mechanical performance.

The capacity to reinforce is function of fiber length, being the reinforcement capacity higher when longer fibers were used. Based on fractured surface images, pulp fibers were shorten than raw sisal counterparts, this could be another reason for lower strength values of composites reinforced with pulp fibers than raw sisal fiber counterpart. Savastano et al. (2009) compared the mechanical performance of cement composites reinforced with different lignocellulosic fibers. They observed that when longer fibers were incorporated to the composite then the system showed a more stable fracture behavior.

Savastano, Warden, and Coutts (2003a) prepared cement composites with different cellulosic fiber loadings. They used as reinforcements thermomechanical pulping or chemi-thermomechanical pulping fibers obtained from commercial and by-product sisal. Ordinary Portland cement and chemically activated blast furnace slag were examined as binders. The three-point bending test was carried out and at the fiber content about 8% they observed a maximum flexural strength between 18 and 20 MPa. Contrary to the mechanical results obtained in the current work, Savastano, Warden, and Coutts (2003a) observed strength improvements of at least 58% over that of the neat ordinary Portland cement matrix. They indicated that the combination of the vacuum de-watering and pressing procedures contributed to the composites mechanical improvement. They mentioned that the flexural strength values less than 4 MPa were obtained when the specimens were prepared using a dough-mixing machine followed by the compaction by vibration method.

In figure 3e is shown a photograph of prepared prismatic specimens of mortar composites. In the current study, even though the properties of fresh state were not characterized, the figure 3e suggests a not-too cohesive, and dry mix. In the cementitious mixture preparation it was not added superplasticizer and cellulosic fibers, that have many hydroxyl groups, can retain mixing water reducing its workability. For example Sawsen et al. (2015) observed that flax fibers can absorb water up to 150% of their dry mass. Page et al (2021) observed that as the vegetable fibre content was increased in the cementitious mixture, the flow decreases leading to a reduction of workability. The influence of cellulosic fibers on the flowability of fresh cement mixture has been studied by Chakraborty et al. (2013). They prepared mortar specimens using different mixing sequences. They observed that when jute fibres were either added directly into the cement slurry or into the water, the workability of the mortars was reduced significantly. They suggested that a significant portion of water required for cement hydration was absorbed by jute fibers and consequently the workability of the mortars was reduced. In the current work, as the fibers are added directly into the cement slurry, Figure 3e suggests that a dry mix was obtained. On the other hand, Chakraborty et al (2013) observed that when the fibres were saturated with water prior to the mixing process the workability obtained was similar to the plain mortar.

In addition to fiber/matrix adhesion and fiber dispersion, the water-retaining implication of these cellulosic fibers could be another factor that affect on the mechanical properties of prepared mortar systems. The cement hydration reaction could be limited since a significant portion of water required for cement hydration was absorbed by fibers and consequently the mortar strength could be reduced respect with plain mortar.
As shown in the flexural stress-strain curves (Figure 4), after incorporating the cellulosic microfibers, the maximum stress value decreased significantly respect with plain mortar. After the maximum, the failure for plain mortar is catastrophic and down the stress value to zero. On the other hand, in systems with microfibers, after the maximum value the stress does not go to zero, can withstand some stress which could be mainly attributed to the fiber pullout process. Regarding the toughness, the value of fiber reinforced systems is similar or slightly lower than the plain mortar. In some cases, the deformation capability was increased respect with plain mortar, however, this increment was not enough to compensate the strength reduction effect. It should be mentioned that the flexural test carried out was an open-loop test system, however, a closed-loop system would provide a stable deformation rate, and thereby, more precise results than an open-loop test system.

Insert here Figure 4

Mortar reinforced with nanocellulose fiber

Figure 5 shows the fractured surface after flexural test of mortar composite reinforced with 0.5 wt% of nanocellulose content. In the fractured surface, sand particles were homogeneously dispersed within the cement matrix.

Insert here Figure 5

Figure 6 shows the effect of nanocellulose content on prepared mortar system properties. After the addition of nanocellulose the density value hardly changed respect with unreinforced mortar (Figure 6a), the density values being around 2100 kg/m^3. However, the pulse velocity value increased as the nanocellulose content was increased, the velocity being slowest for unreinforced mortar (Figure 6b). The addition of nanocellulose increased the ultrasonic wave propagation velocity, being higher as increasing nanocellulose loading. This trend is contrary to the trend observed in microfiber reinforced mortar. The addition of nanocellulose did not increase the number of voids in mortar, in contrast, as observed previously, microfiber addition led to the decrease of density and the reduction of pulse propagation velocity. It should be mentioned that in addition of nanosize dimension, the reinforcement loading are considerable lower for nanocellulose reinforced systems than systems reinforced with microfiber counterparts. Cao et al. (2016) found that the sonication of CNCs avoided the formation of agglomerates and consequently reduced the probability to entrapment air or the formation of pores and voids.

Insert here Figure 6

In Figure 6c and 6d the effect of nanocellulose content on compression and flexural strength values is shown. It should be mentioned that the density and pulse velocity values of unreinforced mortar are different to the values reported in figure 3. Even the cement used was the same; the reason of differences in properties could be due to aging effects during cement storage. After the addition of nanocellulose to cement mortar, it was not observed improvements in compression strength. On the other hand, the incorporation of nanocellulose led to a maximum value of flexural strength at 0.25 wt% content followed by a slight decrease for higher content. The explanation of micromechanics responsible for this improvement is complex. There are different mechanisms that can act simultaneously being difficult to explain the main reason for flexural strength improvement at 0.25 wt% nanocellulose content. The size of nanocellulose leads to a high surface area in contact with matrix that could improve fiber-matrix interactions respect to microfiber counterparts. On the other hand, a minimum amount of nanocellulose is necessary to obtain a good dispersion within the matrix, however, at high concentrations nanocellulose agglomerations could happen, being these points stress concentration zones that reduce the strength value. If stresses can transfer from the matrix to the nanocellulose, i.e. a strong adhesion between nanocellulose and matrix is created, then nanocellulose fibers can act as bridging of microcracks, and consequently, flexural strength can be improved. In addition to the mentioned mechanisms, another additional mechanism could happen simultaneously.

Cao et al. (2015) found that at CNC concentrations larger than 0.2 vol%, CNCs created agglomerates that induce stress concentrators limiting the strength of the cement pastes. Onuaguluchi, Panesar, and Sain (2014) observed that the addition of CNC led to a maximum value of MOR at 0.2 wt% content followed by a slight decrease for higher contents. Cengiz, Kaya, and Bayramgil (2017) used algal mats from nature to produce nanofiber that were added as a reinforcement material to concrete. They observed that plain concrete showed a flexural strength of 2.21 MPa, when algal cellulose nanofiber was incorporated to concrete, the maximum flexural strength increased until 5.96 MPa. Hisseine et al (2019) observed that after adding 0.20 % of CF to the reference paste, the flexural strength enhanced around 21% respect to unreinforced system.

Contrarily to the results reported in the current study, Mejdoub et al. (2017) observed that after the addition of nanofibrillated cellulose, the porosity of Portland cement was reduced and the compressive strength after 28 days increased about 40 % respect to Portland cement without nanofibrillated cellulose. A possible explanation of this strength increase could be the different chemical method used for the isolation of nanocellulose. Mejdoub et al. (2017) used TEMPO-mediated oxidation to facilitate the defibrillation process of nanofibers. Probably, the generated carboxylic groups in nanofibrilated cellulose could create strong bond with the Portland matrix and consequently strengthened the composite.

Mechanical properties comparison with similar systems

The flexural and compressive strength values reported in the literature range from 3.81 to 19 MPa and 20.5 to 62.1, respectively (Table 1). Even though the values reported in the current work for composites reinforced with raw sisal fiber are in the same range, the strength values for bleached sisal pulp reinforced composites are slightly lower than the values reported in the literature for similar systems. Regarding the density values, all data reported in the current study are in the density values range observed in the literature. When nanocellulose reinforced systems are compared, table 2, the reported data in the current work are in the same range values observed for similar nanocellulose reinforced systems. However, it must highlighted that the direct data comparison is not easy for different cement based composites since there are many variables that affect on the final composite mechanical performance. For example the type of cement influences mechanical strength value, even for ordinary Portland cement, which is the most widely used cement, there are different cement grades that show different compressive strength after 28 days of curing. When talking about cement based materials, matrix could be just hydrated cement or mortar (hydrated cement and sand). In mortar based composites, in addition to hydrated cement, sand is also present and the chemical composition of the sand depends on the source of rock. Even though using the same cement and sand, the proportion of these components is critical for final mechanical properties. Another variable can be the water amount used for cement hydration, the water-cement (w/c) ratio is very important since the final mechanical properties depends on this ratio. Sometimes, minor components, such as a superplasticizer, are used for cement based composites. Furthermore, depending on the fabrication method used, the mechanical properties can be different for the same cement based composites. So the direct comparison of mechanical properties summarized in the table is complicated.

Conclusions

Cement mortar composites reinforced with cellulose micro and nano fibers were prepared and characterized. Based on the physico-mechanical properties obtained it can conclude that the dispersion of fiber within cement matrix is critical since the strength values reduced drastically when high amount of fiber aglomerations are present, as observed for pulp reinforced systems. Obtained results suggested that the interconnected network of pulp fibers were not broken during cement mortar specimen preparation and consequently many pulp fiber agglomeration zones were observed. On the other hand, even though a good fiber dispersion within cement based matrix is obtained, as observed in raw fiber reinforced systems, the fiber/matrix adhesion is also critical on mechanical performance of prepared systems. Raw sisal fiber showed a poor adhesion with cement based matrix and consequently the stresses can not transfer from the matrix to the fiber and the strength was not improved respect to plain mortar. In some cases, after the addition of microfibers, the deformation capability was increased respect with plain mortar, however, this increment was not enough to compensate the strength reduction effect to improve the toughness. After the addition of nanocellulose, the ultrasonic wave propagation velocity was increased respect with plain mortar and a slight improvement in flexural strength was observed at nanocellulose content of 0.25 wt%. The size of nanocellulose led to a high surface area in contact with matrix that could improve fiber-matrix interactions respect to microfiber counterparts. However, the strength values obtained in the current study suggested that the cellulose fiber/matrix adhesion is poor and to enhance significantly the strength of cement based composites, the surface of fibers should be modified.

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Data Availability Statement

Some or all data, models, or code that support the findings of this study are available from the corresponding author upon reasonable request.

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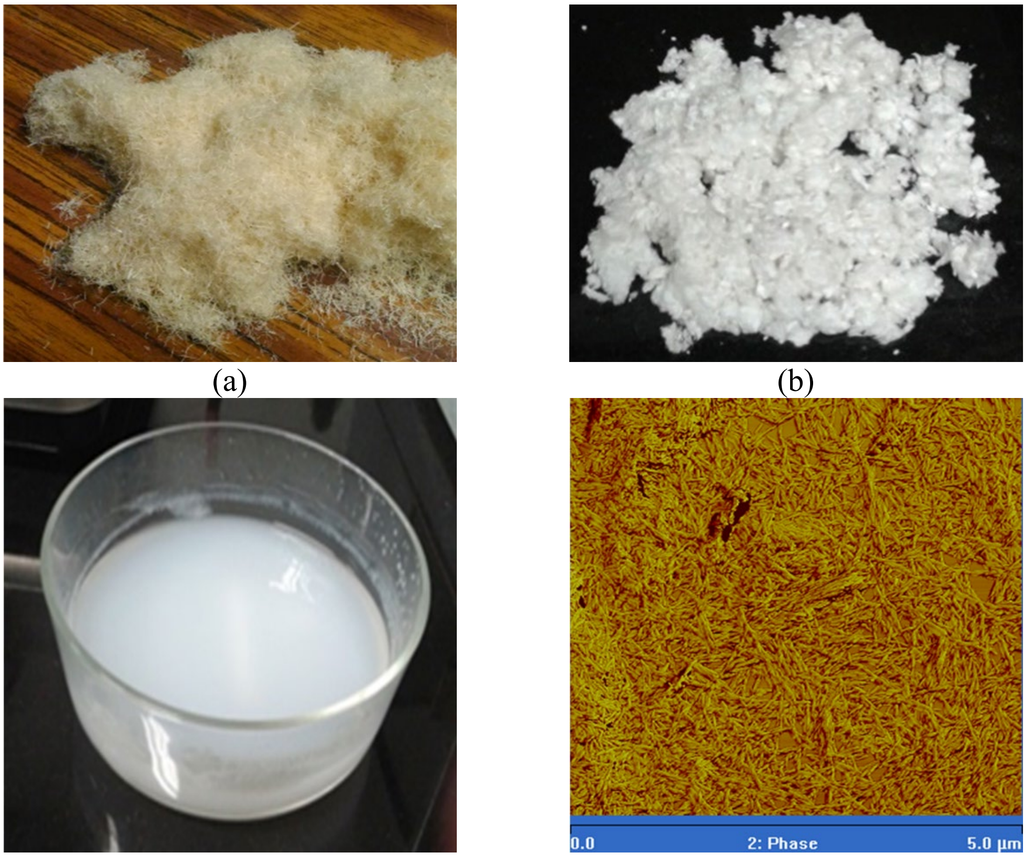
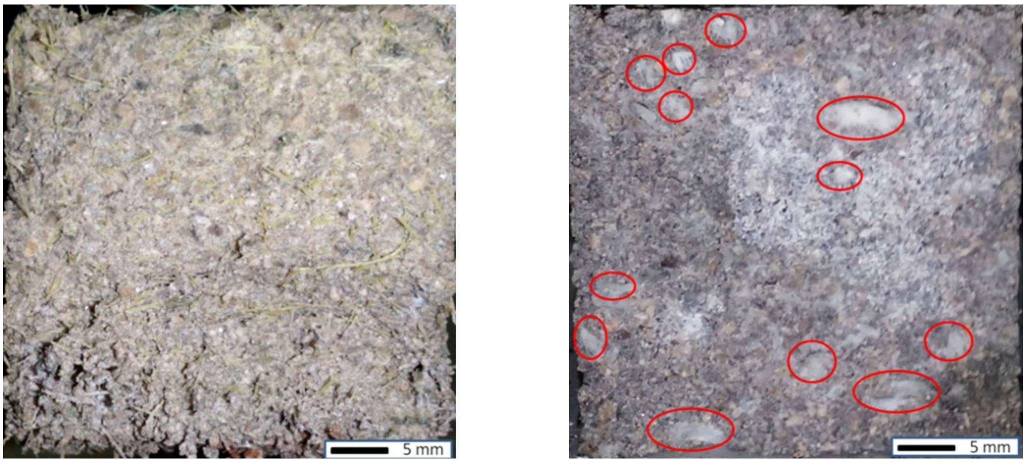


(a) (b)

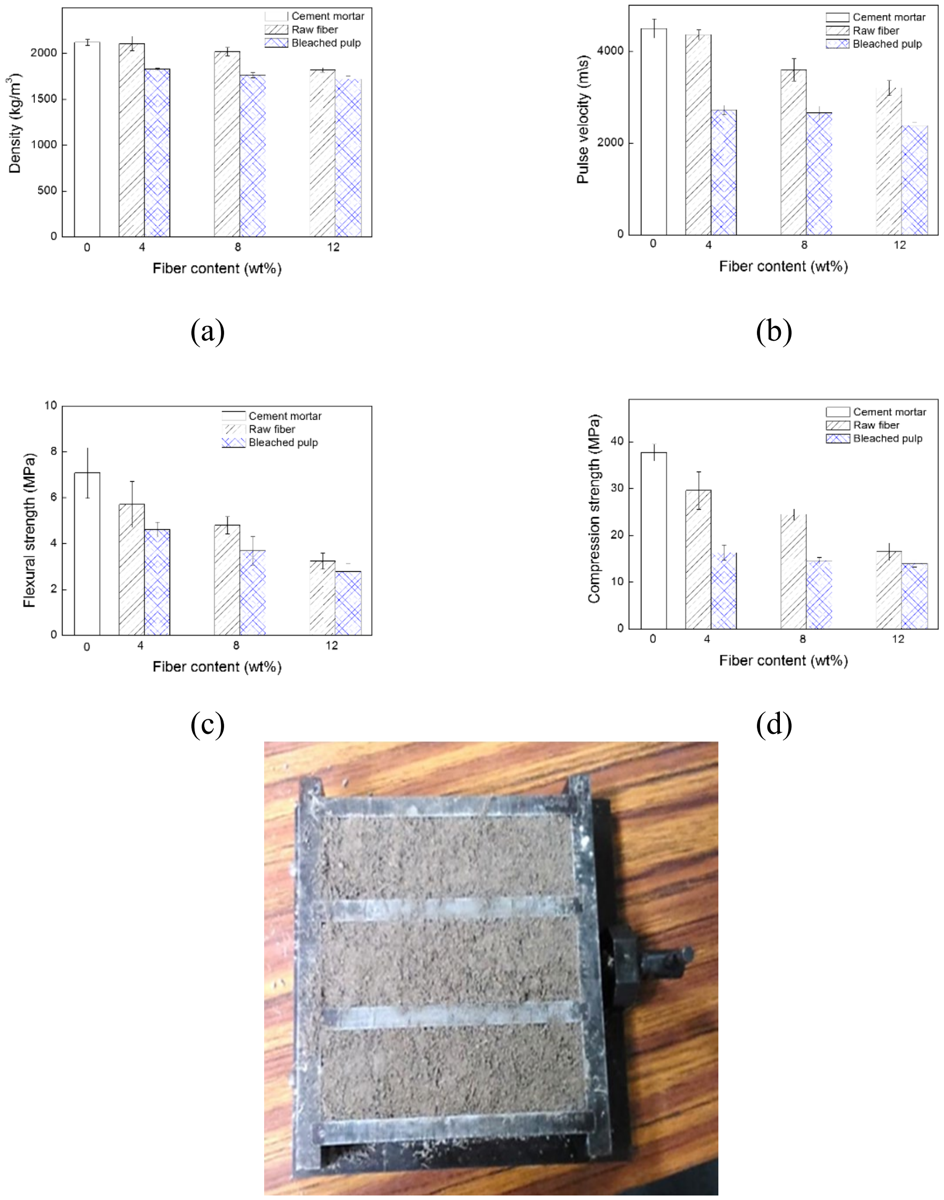
(c) (d)

Figure 1. (a) Chopped raw sisal fiber, (b) Chopped pulp, (c) Nanocellulose suspension in water, (d) Atomic force microscopy image of nanocellulose.



(a) (b)

Figure 2. Fractured surface morphologies of composites reinforced with (a) raw sisal fibers and (b) pulp fibers after flexural test.



(a)

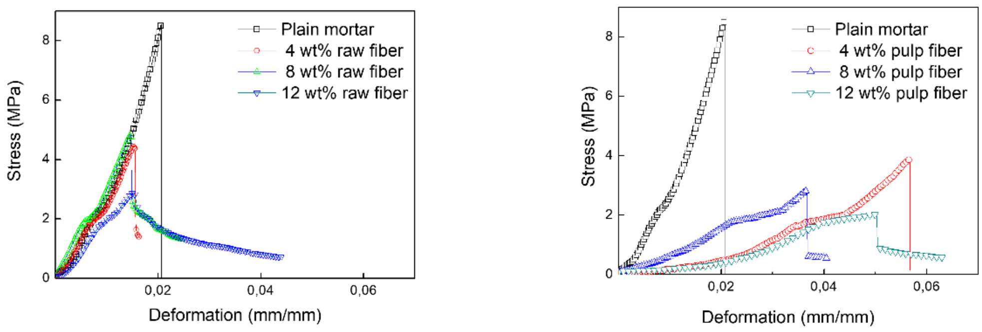
(b)

(c)

(d)

(e)

Figure 3. The effect of fiber content and fiber type on: (a) density, (b) the velocity to propagate of ultrasonic pulse signal, (c) flexural strength, and (d) compression strength, and (e) Prismatic specimens preparation using the mould type UNE-EN-1:2005



(a) (b)

Figure 4. Flexural stress-strain curves of prepared mortar systems after incorporating the cellulosic microfibers: (a) raw sisal fibers and (b) pulp fibers.

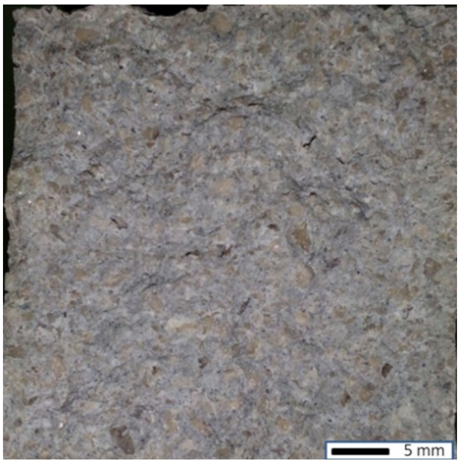
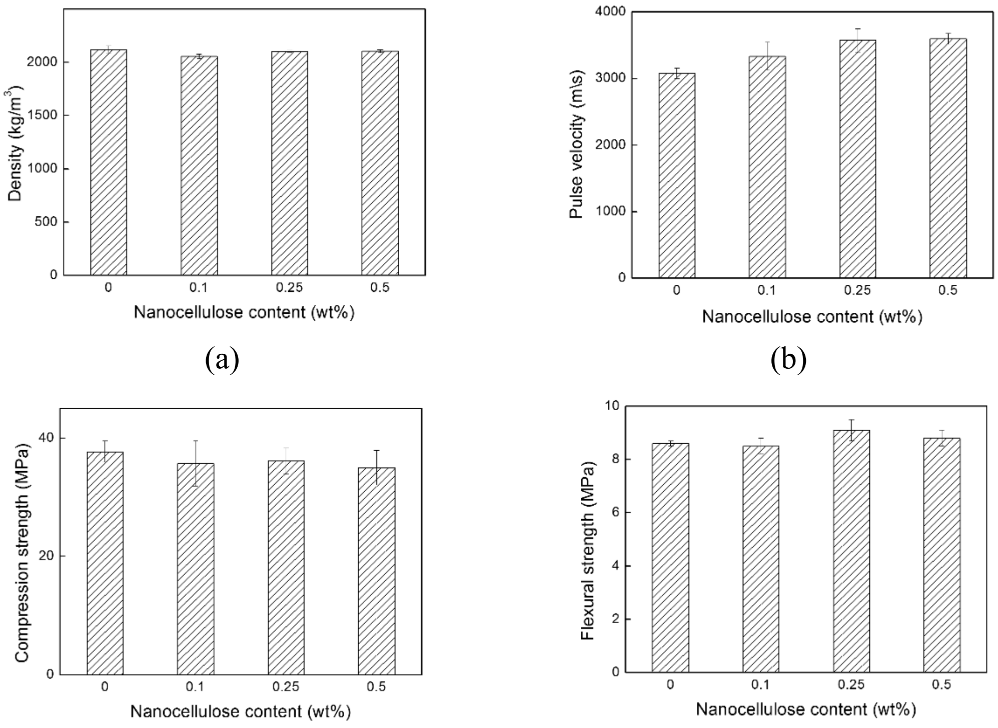


Figure 5. Fractured surface after flexural test of mortar composite reinforced with 0.5 wt% of nanocellulose content.



(a) (b)

(c) (d)

Figure 6. The effect of nanocellulose content on: (a) density, (b) the velocity of propagation of ultrasonic pulse signal, (c) compression strength, and (d) flexural strength.

Reinforcement	Composition	Fiber mass content respect binder (%)	Flexural strength (MPa)	Compressive strength (MPa)	Tensile strength (MPa)	Density (kg/m^3)	Reference
Sisal fiber bundles	Portland cement: Sand: Water (1:3:0.5) Fiber Water-cement ratio 0.5	4, 8 and 12	3.3-5.7 (28 days)	16.3-29 (28 days)	--	1830-2120	Current work
Bleached sisal pulp	Cement:sand:water (1:3:0.5) Fiber Water-cement ratio 0.5	4, 8 and 12	2.8-4.6 (28 days)	14-16.3 (28 days)	--	1720-1850	Current work
Kraft sisal pulp	Cement (78.8 wt%) Ground carbonate (16.5 wt%) Fiber (4.7 wt%)	6	3.81- 6.61 (aged at different conditions)	---	--	1400-1590	Tonoli et al. 2010a
Thermomechanical pulp	Ordinary Portland cement Water/cement ratio 0.40	4, 8 and 12	17-19 (28 days)	--	--	1440-1680	Savastano et al., 2003a
Chemi-thermomechanical pulp	Chemically activated blast furnace slag as matrices Water/cement ratio 0.40	4, 8 and 12	11-18 (28 days)	--	--	1260-1520	Savastano et al., 2003a
Palm fiber	Cement (~24.1 wt%) Silica fume (~1.8 wt%) Water (~10.7 wt%) Superplasticizer (~0.5 wt%) Sand (~62.3 wt%) Fiber (0.1-0.9 wt%)	0.45, 0.90, 1.36, 1.82, 2.30, 2.75 and 3.67	5.9-8.7 (28 days)	42.2-62.1 (28 days)		2180-2300	Dawood and Ramli, 2012
Malva fiber	Portland cement matrix Different Water/Cement ratios (0.30,0.38 and 0.46) Fiber (4 vol%)	--	--	--	2-2.6 (28 days)	--	Savastano and Agopyan, 1999
Sisal fiber		--	--	--	1.4-2.2 (28 days)	--	
Coir fiber		--	--	--	2-2.8 (28 days)	--	
Fique fibres	Portland cement, hydrated lime and river sand mass proportion 1:0.125:0.33 Water-cement ratio 0.35 Fiber 3wt% of the total mass of solids	4.2	--	--	--	1970	Tonoli et al. 2011
Cellulose sisal fibres	Cement:sand:water (1:1:0.46) Reinforcement amount was fixed at 3.3wt.%	~7.5 wt%	10.3 (28 days)	--	--	--	Ardanuy et al., 2012
Bagasse fiber	Portland cement Sand Water Fiber (2, 3 and 4%)	--	3.3-6.2 (28 days)	--	--	--	Omoniyi and Olorunnisola, 2020
Bagasse cellulose fibers	Cement CP-II F Sand Water Water-cement ratio 0.48	0.25, 0.375 and 0.50	6.6-7.5 (28 days)	20.5-25.5 (28 days)	--	--	De Pellegrin, et al., 2021

Reinforcement	**Composition**	**Fiber mass content respect binder (%)**	**Flexural strength (MPa)**	**Compressive strength (MPa)**	**Density (kg/m^3)**	**Reference**
Nanocellulose	1:3:0.5	0.1, 0.25 and 0.5	8.5-9.1 (28 days)	35.0-36.2 (28 days)	2060-2100	Current work
Nanocellulose	Portland cement (80.3-97.2 wt%) Water-cement ratio 0.30 Cellulose filaments (0.04-0.3 wt%) Polycarboxylate-based admixture (0.1-0.2)	0.05, 0.10, 0.20 and 0.30	5.6-5.8 (28 days)	75-90 (28 days)	--	Hisseine et al., 2019
Nanocellulose	Cement (73.6-74.1 wt%) Water (25.8-25.9 wt%) Cellulose nanocrystals (0.015-0.567 wt%) Water-cement ratio 0.35	0.02, 0.05, 0.10, 0.26, 0.51,0.77	17-19 (28 days)	--	--	Cao et al., 2015
Nanocellulose	Limestone cement Water Nanocellulose fibers Water-cement ratio 0.5	0.05, 0.1, 0.2 and 0.4	3-6.5 (28 days)	--	--	Onuaguluchi et al., 2014
Nanocellulose	Portland cement (~16.5 wt%) Fine sand (~66.5 wt%) Water (~16.5 wt%) Nanocellulose fibers (0.08-0.83 wt%) Water-cement ratio 1	0.5, 1.25, 2.5, 3.75 and 5.0	1.41-5.96 (7 days)	--	--	A. Cengiz et al., 2017
Nanocellulose	Portland cement Water Nanocellulose fibers Water-cement ratio 0.26	0.01, 0.05, 0.1, 0.2, 0.3 and 0.5		13-43 (28 days)	1780-1820	Mejdoub et al., 2017
Micro crystalline cellulose	Portland cement Standardized sand Water Nanocellulose fibers (0.25-1.5% on the weight of cement mix) Surfactant 0.25-1.5% on the weight of cement mix) Superplasticizer (0-3 wt%) Water-cement ratio (0.5-0.6)	--	5-9 (28 days)	27-59 (28 days)	2020-2189	Parveen et al., 2017