MECHANICAL PERFORMANCE OF CEMENT MORTAR COMPOSITES REINFORCED WITH NANO AND MICRO-SCALE CELLULOSE FIBERS

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ABSTRACT

The combination of the reinforcements at different levels can lead to a synergetic effect on the mechanical properties. In this work, the preparation and characterization of new cement mortars reinforced with conventional pulps at the micro-scale level and nanofibrillated cellulose fibres at the nano-scale level has been carried out. The conventional pulps have been obtained by subjecting sisal fibres to a soft mechanical treatment and the nanofibrillated cellulose have been prepared by the application of a high intensity refining process. Based on the preliminary results of this paper, it can be concluded that to obtain cement mortar composites with high modulus and resistance and high toughness the combination of both, cellulose nanofibers and microfibers could be interesting.

KEYWORDS:

Nanofibrillated cellulose, sisal, cement mortar composites, mechanical performance

INTRODUCTION

The use of vegetable fibres to reinforce brittle matrices such as cement mortar or concrete constitutes an interesting possibility that offers many advantages with respect to the utilization of other fibres or reinforcements. On the one hand, due to their mechanical properties, vegetable fibres can improve the ductility, flexibility and crack resistance of the resulting material. On the other hand, due to their low cost, the use of vegetable fibres in fibre–cement materials constitutes a very interesting option for the building industry, mainly in less developed countries or countries that need low cost constructions (Agopyan, 2005; Roma 2008; Savastano, 2003a; Savastano, 2000; Saxena, 1992).

From the environmental point of view, it is worthy of mention the great effort that has been made in the civil engineering field with the aim of improving the materials and techniques commonly used. With regard to materials, the use of waste and renewable sources as raw materials together with the use of more environmentally friendly materials has notably increased over the last few years (Coutts, 2005).

So far, most of the published works on the field of natural fibre reinforced cement mortar composites, describe the use of natural fibres of several millimetres in length (2-10mm) and diameters ranging between 10-30 µm, such as wood pulps, sisal and abaca fibres, cotton linters, etc (Ardanuy, 2011; Asasutjarit, 2007; Claramunt, 2011; Eichhorn 2010, Mohr, 2007; Savastano 2003b; Silva, 2010; Toledo Filho, 2010; Toledo Filho, 2003; Tonoli, 2009).

It is well known that the reinforcing capability of the fibres can be increased by reducing their size to the nanometre scale. Nanofibres can be obtained from natural fibres by subjecting them to mechanical, chemical or enzymatic treatments (Eichhorn, 2010; Sio, 2010). The application of nanofibres in polymer composites is widespread, however, to our knowledge, research related to the use of these fibres as reinforcements in cement mortar matrices is scarcely known (Ardanuy et al. 2012; Claramunt et al., 2011).

In this work, the performance new cement mortar composites reinforced with cellulose fibres from conventional pulps of sisal and cellulose nanofibres prepared by the application of a high intensity refining process has been evaluated.

MATERIALS AND EXPERIMENTAL PROCEDURES

Materials

UNE-EN 197-1:2000 Type I cement supplied by Ciments Molins (Spain) was used for the present research work. Silica fume has been used to replace a 10 wt. % of the cement. The sand was supplied by Sibelco and a part was subjected to grinding with a Ball mill in order to analyse the effect of the particle reduction in the performance of the composites. Figure 1 show the particle size distribution of the milled sand and the as received sand compared to the size of cement particles. As seen after the milling process, the sand particles have similar size to the cement particles allowing the formation of a more homogeneous matrix.

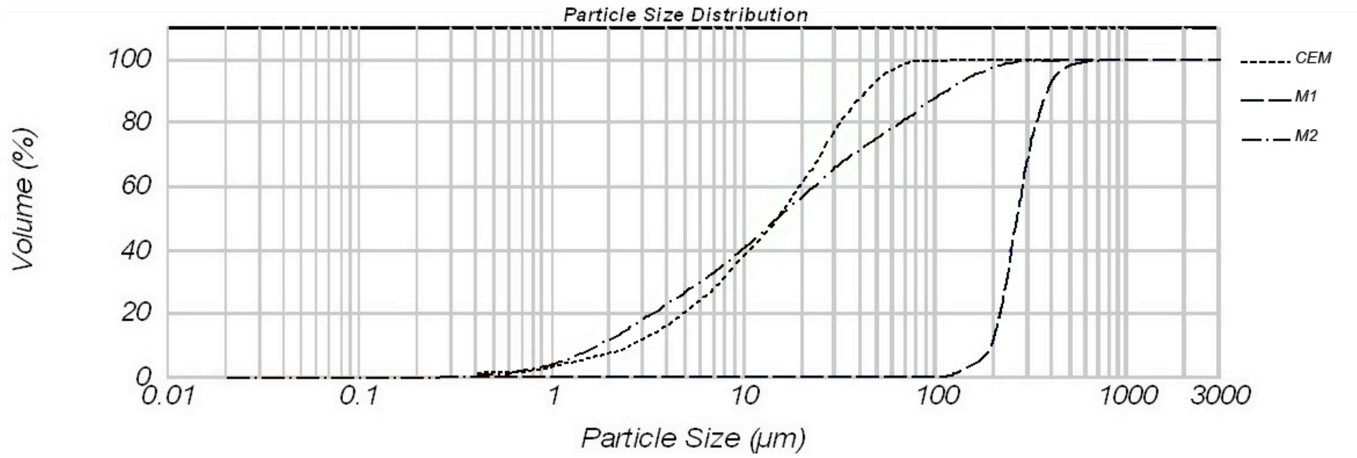

Figure 1 – Particle size distribution of the milled sand (M2) and as received sand (M1) compared with the one of cement particles.

Sika Viscocrete-3425 fluidizer, obtained from Sika S.A.U., was used at a maximum dosage rate of 40 g/1000 g of cement to aid workability. Sisal (Agave sisalana) pulp from a soda-anthraquinone cooking process was kindly supplied by CELESA (Spain).

Preparation of nanofibrillated cellulose

Cellulose nanofibres have been prepared by the application of a high intensity refining process in a Valley Beater. Following the ISO 5264/ 1-1979 (E), 360 grams of oven-dried sisal pulp were added to deionised water, in such a way as to give a final volume of 23 litres, corresponding to a consistency of 1.57 % (m/m). The mixture was placed at the Valley Beater device, where fibrillation of the sisal fibres took place due to the mechanical action. Different refining times of 1, 2, 3, 4, 5 and 6 hours were studied.

An initial characterisation of the sisal pulp was performed. The fibre dimensions such as length and width were measured according to TAPPI T271 om-02 by using a Kajaani FS300 Analyzer. Measurements were taken from □5000 fibres. In order to study the intrinsic resistance of the sisal fibres, the zero-span tensile index (ISO 15361:2000) was also determined by using a Pulmac tester. According to ISO 15361:2000 (E), the zero-span tensile index (ZI) was calculated from equation 1.

$$Z_I \; 8 \; \frac{Z_T}{G} \qquad (1)$$

Where ZI is the zero-span tensile index (KNm/g); ZT is the zero-span tensile strength (Kilonewton/meter) and G is the oven-dry weight (g/m2). To perform the zero-span test, homogenous isotropic sheets of sisal pulp with a weight of about 60g/m2 were produced.

The microstructure and morphology of the sisal pulps obtained at different refining times were analysed by scanning electron microscopy (SEM), using a JEOL JSM-S610 microscope at an accelerating voltage of 10 kV. Prior to examination, a little amount of pulp was diluted in deionised water and an aliquot of this suspension was dropped on a metallic support surface and dried in an oven overnight at 80°C. Finally, the dry pulp surface was sputtered with a thin layer of gold to make it conductive.

Composite preparation and characterization

In order to study and compare both the reinforcing effect provided by the incorporation of the sisal microfibres and nanofibres and the effect of their combination, 3 series of composites were prepared following the same procedure described previously (Claramunt, 2011). Table 1 shows the composition of the samples studied.

Table 3 – Reference and composition of the cement mortar composites prepared.

Sample reference	Fibre treatment	Fibre content (wt. %)	Cement/Silica Fume/sand proportions (by weight)	Water/Cement ratio
M1SC	Conventional pulp	4	0.9:0.1:1	0.5
M1SNSC	Conventional pulp / Nanofibrillated pulp	2 / 2	0.9:0.1:1	0.7
M1SN	Nanofibrillated pulp	4	0.9:0.1:1	0.6

Prismatic specimens were prepared for the flexural tests. The mould used was UNE-EN 196-1:2005 type with internal dimensions of 40x40x160mm modified to allow the compression of the specimens to 20mm of thickness. The specimens were cured for 7 days at 20 ± 1 °C and 95% relative humidity. Three point bending tests were performed using an Incotecnic press equipped with a maximum load cell of 30 kN at a load speed of 50 ± 10 N/s.

RESULTS AND DISCUSSION

Characterization of the nanofibrillated cellulose

Table 2 shows the results of the physical and morphological characterization of the sisal pulp as received.

Table 2 – Physical characterization of sisal fibers

Length (mm)	Width (μm)	Aspect ratio (length/width)	Z_I (KNm/Kg)
1.14	15.9	71.4	130±12

Figure 2 shows the microstructure of the initial sisal fibres (first row), and of the pulps obtained after 3 (second row) and 6 (third row) hours of refining at low magnifications (left) and high magnifications (right).

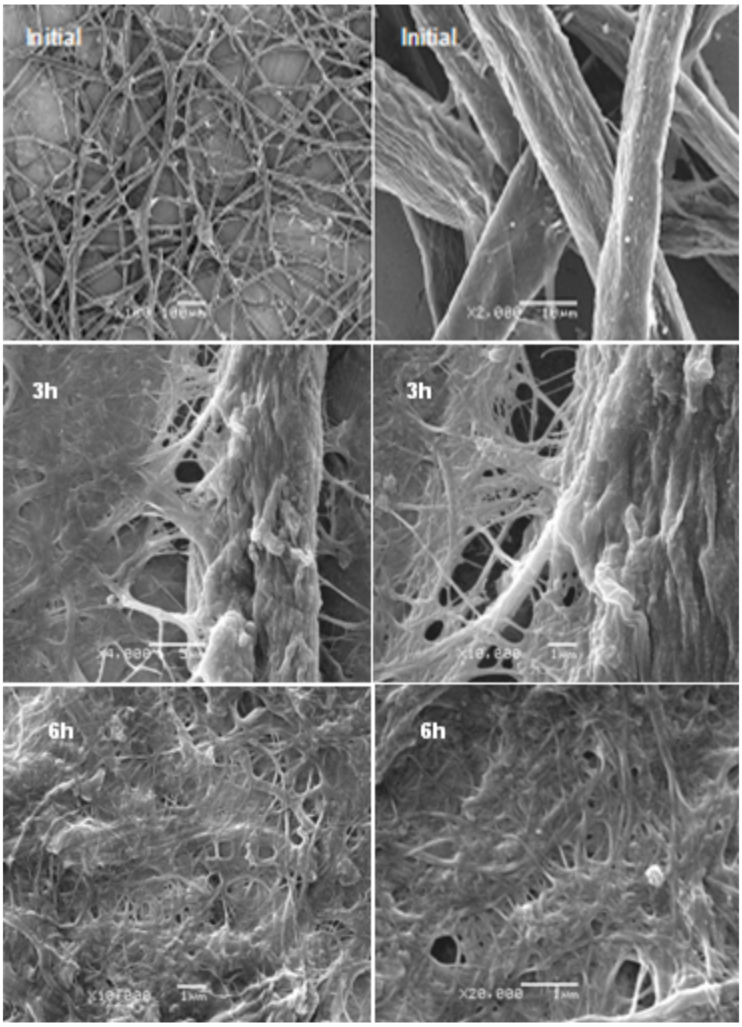

Figure 2 – SEM micrographs of the initial sisal pulp and after different refinement times.

As shown in Figure 2, initially, the sisal fibres have a diameter ranging from 10 to 20 □m, which confirms the sisal average width measured by Kajaani (Table 1). After 1 h of refinement, the fibres remain almost intact, although an initial external fibrillation starts to appear, after which a progressive fibrillation of the fibres can be observed. After 3 hours of refinement, the external fibrillation of the fibres can be clearly observed. An increase of the refining time to 4 hours involves an enhancement of the fibrillation degree of the fibres. The initial fibre diameter is reduced to □5 □m as a consequence of the fibrillation of the outer layers that creates branches in the fibre, leading to the formation of nanofibrils, increasing in this way the fibre specific surface area. Further refinement (6 hours, bottom) yields highly branched fibres in the nanometer scale, between 25 and 250 nm. This increase of the aspect-ratio could enhance the reinforcing capabilities of these pulps, making them potentially suitable for the production of composites. Moreover, their high specific surface area would potentially favour the interaction with the matrix, giving rise to a better stress transfer. Taking into account all these issues, the optimum refining time was found at 6 hours and this pulp was selected for the preparation of the cement composites.

Characterization of the cement mortar composites

The main parameters obtained from the flexural tests are compiled in Table 3 and the typical bending curves for the specimens analyzed after 7 days of curing are presented in Figure 3.

Table 2 – Mechanical properties of the cement composites prepared.

Reference	Fibre treatment	Flexural Strength (MPa)	Fracture energy (J)
M1SC	Conventional pulp	9.0±1.3	308±30
M1SNSC	Conventional pulp / Nanofibrillated pulp	9.6±0.9	157±15
M1SN	Nanofibrillated pulp	10.6±0.2	49±9

As shown, although a slight increase of the maximum flexural strength could be observed when the nanofibrillated pulp is used, the composites exhibit brittle behaviour, as can be easily deduced by the small plastic area in the stress-displacement curve and from the fracture energy value. This brittle failure can be attributed to the low crack bridging capacity of the nanofibrils, since their short length is not enough to prevent the growth of matrix microcracks. Unlike the composites reinforced with nanofibrillated cellulose, those reinforced with the conventional sisal pulp show a more plastic behaviour since long fibres are more effective in bridging the crack faces. In addition, the interfacial properties of these composites are weaker as a consequence of the low fibre specific surface area, favouring toughening by debonding and fibre pullout. Finally, for the composites prepared with a combination of the nanofibrillated and conventional pulp an improvement of flexural strength as well as a strain hardening behaviour could be observed. In this composite, the combination of these fibres at micro-scale and nano-scale levels leads to a combination of interesting properties provided by both reinforcements. Although more research must be done, it could be concluded from these results that an interesting reinforcement of brittle matrices such as cement mortars would be a combination of both types of cellulose fibres, at the nano- and micrometric level, to match the good flexural properties provided by the nanofibrils (modulus and strength) with the high toughness rendered by the cellulose fibres at the micrometric level.

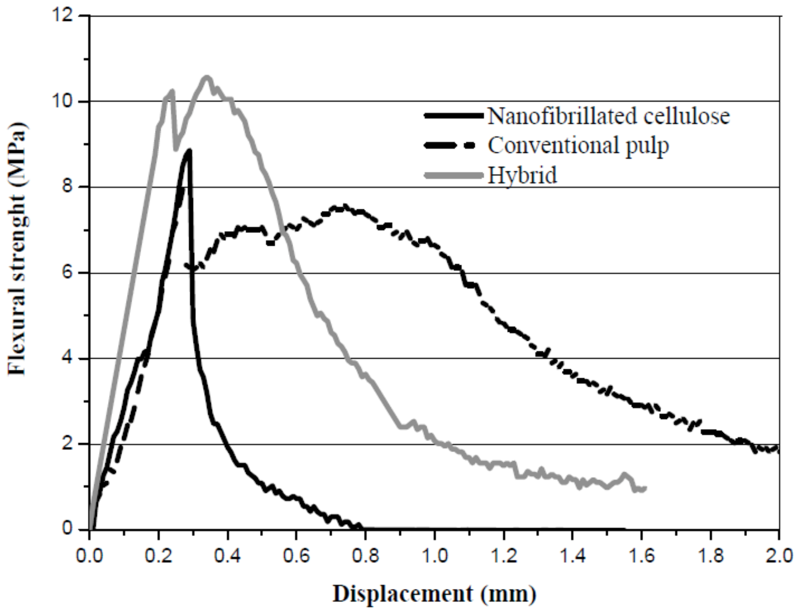

Figure 3 – Typical stress versus displacement curves of the composites prepared with the nanofibrillated cellulose, the conventional pulp and a combination of both reinforcements (hybrid).

CONCLUSION

In this study, cement mortar composite materials reinforced with nanofibrillated cellulose fibers and a combination of nanofibrillated cellulose and conventional pulp have been prepared and characterized in order to analyse the potential reinforcement of these fibres compared with conventional fibres. Composites prepared with nanofibrillated fibres had significantly lower toughness than those prepared with conventional pulps. Nevertheless, composites prepared with a combination of nanofibrillated cellulose and conventional pulp possesses greater flexural modulus and flexural resistance and strain-hardening behaviour.

Based on these preliminary results, it can be concluded that to obtain cement mortar composites with high modulus and resistance and strain-hardening behaviour the combination of both, cellulose nanofibers and microfibers could be interesting.

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