NANOFIBRILLATED CELLULOSE AS REINFORCEMENT FOR HIGH PERFORMANCE CEMENT MORTAR COMPOSITES

M. Ardanuy[1*], J. Claramunt[2], F. Pares[1], E. Aracri[1] and T. Vidal[1]

[1]Departament d'Enginyeria Tèxtil i Paperera, EET. Universitat Politècnica de Catalunya. C/Colom 1, 08222, Terrassa (Barcelona) Spain
[2]Departament d'Enginyeria Agroalimentària i Biotecnologia. Universitat Politècnica de Catalunya. Avinguda del Canal Olímpic, 15, 08860 Castelldefels (Barcelona), Spain
**monica.ardanuy@upc.edu*

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Abstract

In this work, the preparation and characterization of new cement mortars reinforced with nanofibrillated cellulose fibres has been carried out. The effect of the fibre fibrillation degree on the mechanical performance of the obtained composites is presented and analyzed, taking into account the reinforcement final size depending on the intensity of the mechanical treatment applied. On the one hand, conventional pulps have been obtained by subjecting sisal fibres to a soft mechanical treatment and, on the other hand, nanofibrillated cellulose pulps have been prepared by the application of a high intensity refining process. These pulps were incorporated and homogenously dispersed in cement mortars, and subsequently, the resulting pastes were cured for 28 days. The mechanical performance of the different composites was determined by flexure tests.

1 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. By one hand, due to their mechanical properties, vegetable fibres can improve the ductility, flexibility and crack resistance of the resulting material [1-2]. 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 [3].

So far, most of the published works on the field of natural fibre reinforced cement mortar composites, describe the use of natural fibres with several millimetres in length (2-10mm) and diameters ranging between 10-30 µm, such as wood pulps, sisal and abaca fibres, cotton linters, etc. [2] [4-6]. It is well-known that the reinforcing capability of these fibres can be increased by reducing their size into the nanometre scale. The obtaining of nanofibres from natural fibres can be achieved by subjecting them to mechanical, chemical or enzymatic treatments. [7]

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 has not been published yet.

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

2 Materials and testing methods

UNE-EN 197-1:2000 Type I cement was used in this research. The sand, provided by Sibelco, was subjected to grinding with a Jar mill (1000 cm^3 of capacity and spheres of 12 mm) at 400 rpm during 5 min to achieve a maximum particle size of 0.1 mm. 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-anthraquinona cooking process was kindly supplied by a CELESA (Spain).

Nanofibrillated cellulose was prepared by the application of a high intensity refining process in a Valley Beater. Following the ISO 5264-1:1979, 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 the cut and fibrillation of the sisal fibres took place due to the mechanical action. The fibrillation degree of sisal fibres was studied for different refining times of 1, 2, 3, 4, 5 and 6 hours.

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 (Z_I) was calculated from equation 1.

$$Z_I = \frac{Z_T}{G} \tag{1}$$

where Z_I is the zero-span tensile index (KNm/g); Z_T is the zero-span tensile strength (Kilonewton/meter) and G is the oven-dry grammage (g/m^2). To perform the zero-span test, homogenous isotropic sheets of sisal pulp with a grammage of about 60g/m^2 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 them conductive.

In order to study and compare the reinforcing effect provided by the incorporation of the cellulose fibres and nanofibrillated cellulose in the cementitious matrix, 3 composites of each type of reinforcement were prepared following the same procedure described previously [1-2].

Prismatic specimens were prepared for the flexural tests. The mould used was UNE-EN 196-1:2005 type with internal dimensions of 40x40x160 mm3 modified to allow the compression of the specimens to 20 mm of thickness. The specimens were cured for 28 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.

3 Results and discussion

3.1 Initial characterization of the sisal fibres

Table 1 shows the results of the physical and morphological characterization of the untreated sisal pulp and Figure 1 illustrates the fibre length distribution of the fibres obtained from Kajaani analyser.

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

Table 1. Physical characterization of sisal fibers.

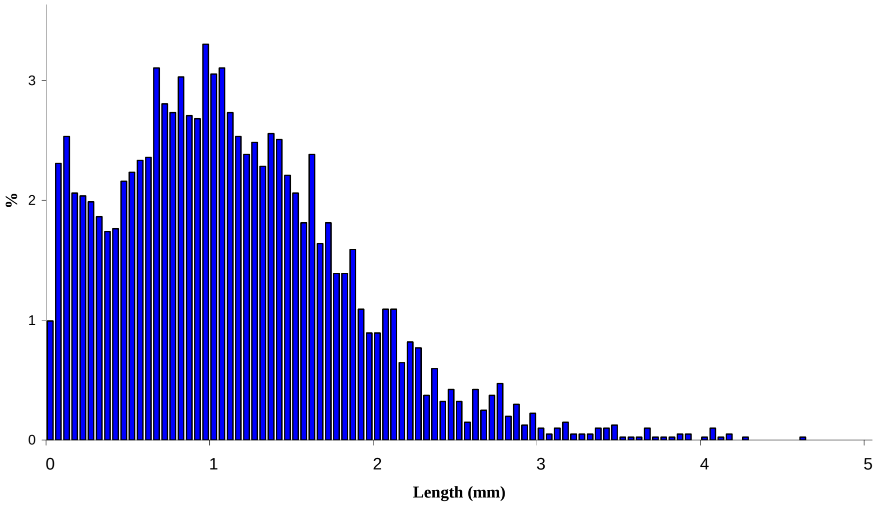

Figure 1. Fibre length – weigthed distribution of sisal fibres.

As shown, the majority of the fibres have lengths less than 3 mm (average of 1.14 mm) and diameters of around 16 microns. These fibres have a tensile index around 130 kNm / kg, in agreement with the mechanical properties of sisal found in the literature [10].

3.2 Effect of the refining process on the fibre microstructure

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). As shown, initially, the sisal fibres have a diameter ranging from 10 to 20 µm, 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 µm 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. These nanofibres present a higher intrinsic strength than the initial fibres. This fact, together with the 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 place 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.

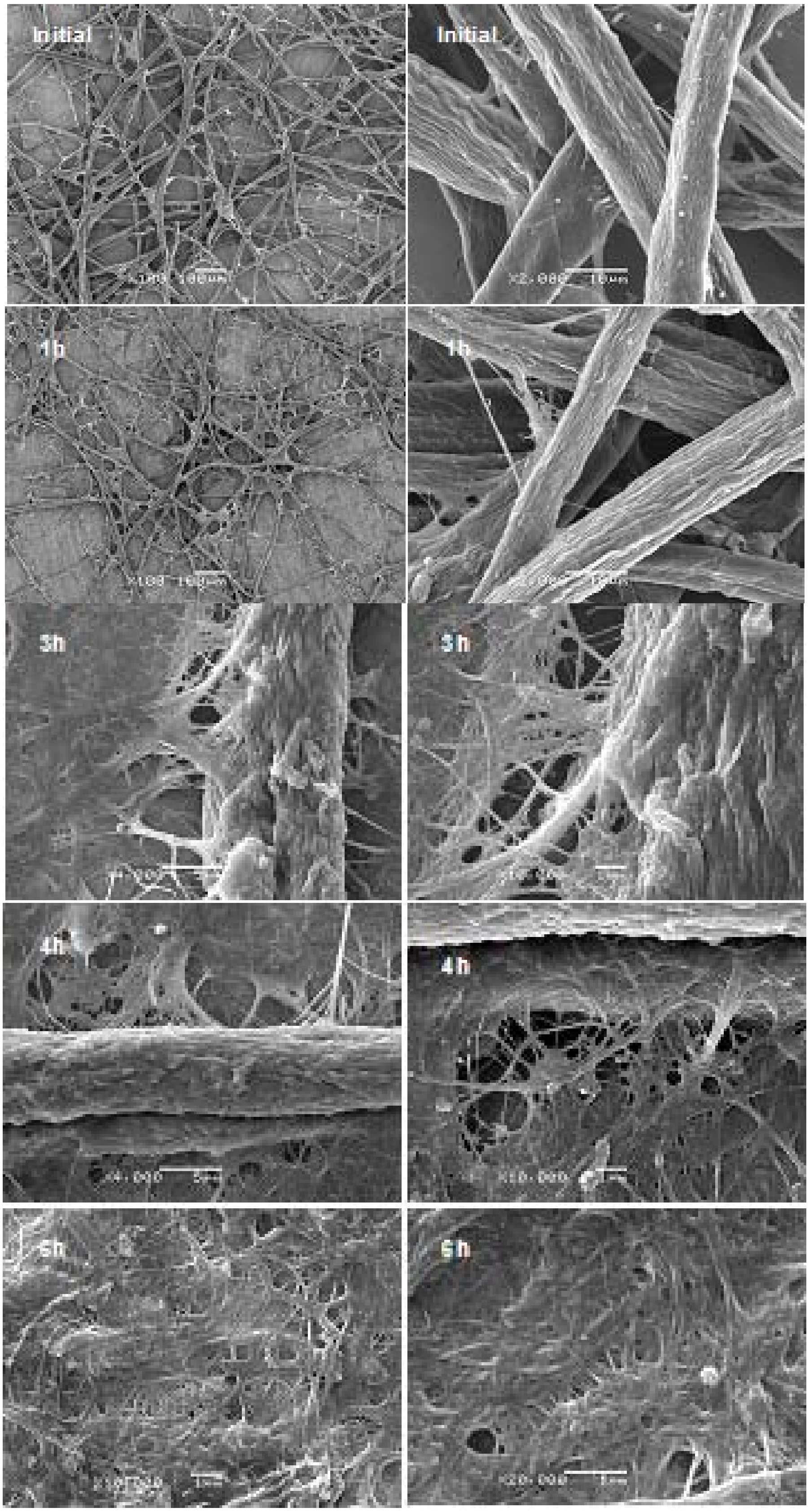

Figure 2. SEM micrographs of the initial sisal pulp and after different refinement times.

3.3 Effect of the use non-refined and highly refined sisal fibres as reinforcement on mechanical properties of the cement composites

The influence of the conventional sisal pulp and the nanofibrillated one as reinforcement on the mechanical properties of the cement mortar composites is shown in Table 2. A clear improvement of both the flexural modulus and strength can be observed for the composites reinforced with the nanofibrillated pulp compared to those reinforced with the conventional one. As previously mentioned, it is expected a higher reinforcing effect of the nanofibrillated cellulose due to the higher intrinsic strength of the cellulose crystals. Moreover, the high specific surface area of the nanofibrillated cellulose results in an enhanced of fibre-matrix interaction which involves a better stress transfer from the matrix to the reinforcement. Nonetheless, a significant reduction of the fracture energy is also observed. Probably due the finesse of the cellulose nanofibres with respect to the matrix particles the toughness of the composite is not improved.

Cement mortar composite	Flexural Modulus (GPa)	Flexural Strength (MPa)	Fracture Energy (J)
Reinforced with conventional sisal pulp	2.4±0.1	10.3±0.6	759±78
Reinforced with nanofibrillated sisal pulp	4.1±0.1	14.0±0.6	357±48

Table 2. Results of flexural tests.

4 Conclusions

New nanofibrillated cellulose reinforced cement mortar composites have been successfully prepared. These composites showed a significantly higher modulus and strength compared to the ones prepared with conventional sisal pulps. The high nanofibrillated cellulose specific surface area results in an enhanced of fibre-matrix interaction leading to an strong fibre-matrix bonding, which involves a better stress transfer from the matrix to the nanofibrils but, in turn, an embrittlement of the composite.

These results confirm the higher potential reinforcement of nanofibrillated cellulose with respect to the microfibers. Moreover, based on the results it can be concluded that to obtain cement mortar composites with high modulus and resistance and high toughness could be interesting the combination of both, nanofibrillated cellulose and microfibers.

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