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Autoclaved cellulose fibre reinforced cement: Effects of silica fume

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HIGHLIGHTS

- The effect of silica fume and fibres on cement hydration is explored.
- Image analysis method is used to determine the degree of hydration of cellulose cement composites.
- Hydration products and microstructure were also analysed with FTIR and BSE–SEM.

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ABSTRACT

With the aim of developing vegetable-fibre cement composites free of portlandite and with short curing process, this study analyses the influence of the curing conditions and the addition of pozzolanic material on the hydration of Portland cement–fibre matrices. Different specimens of cement composites with and without cellulose fibres and with and without silica fume were cured using autoclaving steam and normal curing. The hydration products and the microstructure of the resulting pastes were analysed by means of FTIR and BSE–SEM and the degree of hydration was quantified with Image analysis. The results indicate that the hydration products and the hardness of the pastes depend on the three analysed factors: curing method, silica fume, and presence of fibres.

1. Introduction

The use of vegetable fibres to reinforce brittle matrices such as cement mortar or concrete offers some advantages with respect to the utilisation of other fibres or reinforcements. Due to their mechanical properties, vegetable fibres can improve the ductility, flexibility, and crack resistance of the resulting material [1–4]. On the other hand, due to their low cost, availability, and renewability, the use of these fibres constitutes a very interesting option for the development of more sustainable construction materials [5–9].

Concerning the manufacturing techniques for cement based products, the curing time supposes a limitation because of the long periods used (usually 28 days). That is why enterprises that work with pre-casting products usually expose their materials to special curing conditions to decrease the hardening time and to improve the resistance of the resulting materials. The curing procedures generally used to accelerate the cement hydration are steam curing at high pressure – the autoclaving process – or steam curing at atmospheric pressure [10–12]. Autoclaving shortens the curing process considerably – from 28 days to 1 day or hours – with respect to the normal conditions (20 °C and 100% RH). Some examples of pre-casting products manufactured with this technique are flat and corrugated sheets for roofing and cladding, fire insulation boards, sand–lime bricks, lightweight cellular concrete, and small pre-cast concrete products, among others [13,14]. However, the material properties and microstructure can be substantially affected by the conditions of curing.

Nonetheless, the industrial production of cement matrix composites reinforced with vegetable fibres is currently limited by the lack of durability of these materials. Many studies have related the presence of portlandite (calcium hydroxide) in the cementitious matrix with the degradation of the vegetable fibres and thus with the loss of durability of these composites [4,15–17]. One related possibility for improving the durability is to modify the composition of the matrix in order to reduce or remove the portlandite compound. Tolêdo Filho et al. [18] used pozzolanic additions, like silica fume, to precipitate the calcium hydroxide

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as calcium silicate hydrate. However, it is important to optimise the dosages to ensure the disappearance of portlandite with the minimum addition of pozzolanic compounds. On the other hand, it must be taken into account that the curing conditions as well as the presence of the fibres can alter the cement hydration mechanisms as well as the pozzolanic reactions.

Given that the main applications of the vegetable fibre cement composites are for pre-cast products, it is of crucial importance to know the effects of the fibres and the additions on the autoclave curing of these materials. Toutanji and Bayasi [19] reported the effects of the curing conditions (steam, moist, and air curing) and silica fume contents on the properties of silica fume concrete finding that steam curing was found to enhance the properties of silica fume concrete, whereas air curing exhibited adverse effects as compared to moist curing. However, as far as we know, the combined effects of the fibres and silica fume addition and the curing conditions have not yet been reported.

So, in this paper, with the aim of developing cement composites reinforced with vegetable fibres that could exhibit higher durability (i.e. free of portlandite) and that could be cured in a short time, we analyse the influence of the curing conditions – autoclaving or a standard curing chamber – and the addition of fumed silica on the hydration degree of Portland cement composites reinforced with sisal fibres.

2. Materials for testing

UNE-EN 197-1:2011 Type I cement supplied by CEMEX (Spain) was used for this work. Following previous research, 10 wt.% of silica fume sourced from Sika was used as the pozzolanic addition to ensure the total conversion of portlandite in hydrated calcium silicate [20]. The mineralogical composition of the cement and silica fume was determined using X-ray diffraction. The main detected crystalline phases present in the Portland cement (PC) were alite (C_3S), belite (C_2S), C_3A, and brownmillerite. The silica fume (SF) had an amorphous halo in the 2θ range of 23.8°, indicating amorphous silica, but also the diffraction lines at $2\theta = 26°$ and 35.6° indicate the presence of quartz and silicon carbide, respectively.

Sisal (*Agave sisalana*) fibres were used as pulps prepared from a soda–anthraquinone cooking process kindly supplied by CELESA (Spain). These fibres presented an average length of 1.14 mm and average width of 13.5 μm as previously determined [21].

3. Experimental procedures

3.1. Preparation of pastes: mix proportions and curing process

Four series of pastes were prepared with Portland cement (PC) with and without silica fume (SF) and with and without fibres (F). The mix proportions of the pastes are shown in Table 1.

Table 2 presents a summary of the curing conditions studied for the four series of pastes. The samples were put into the autoclave or the curing chamber after casting, respectively. The autoclave cured pastes (A) were put into the autoclave at a temperature of 120 °C and pressure of 1.5 atm. The autoclave process was performed in a SELECTA MED 20, with a heating ramp temperature of 10 °C/min until 120 °C. The curing chamber process (C) was performed at ambient temperature (20 °C) in an atmosphere with high relative humidity (approximately 95%).

The samples incorporating the sisal pulp fibre were produced using the following procedure. Firstly, dispersion of 12 g of dried fibres in 2 L of water followed by filtration to give 100 g of mixture (12 g of fibres and 88 g of water) was carried out. In a second step, the dispersed fibres were mixed with the cement paste (or the cement paste with silica fume) and water until a homogeneous paste was obtained.

Table 2
Curing conditions of the samples and references used in the text.

Reference	Autoclave (h)	Curing chamber (days)
A1	1	0
A2	2	0
A2 + C7	2	7
A3	3	0
A4	4	0
A4 + C7	4	7
C7	0	7
C28	0	28

With this paste, eight specimens of 100 g each were prepared and subjected to vacuum using a Kitasato flask in order to eliminate the excess water. The final water/cement ratio depended on the amount of water eliminated from each specimen, with an average of 0.42 for the specimens with silica fume and 0.53 for those without silica fume. The specimens were cast in a silicon mould and cured following the conditions shown in Table 2.

3.2. Characterisation

To characterise the pastes cured under different conditions, the reactions were frozen with the following methodology:

(a) Specimens for FTIR: grinding of the pastes during 5 min using a ball mill, followed by drying with acetone during 45 s. Afterwards the samples were washed with ethanol and stored in a full automatic dessicator (DRY-KEEPER AUTO A-type) maintained at 15%RH and at ambient temperature and atmospheric pressure until the analysis.
(b) Specimens for hardness and BSE–SEM: once demoulded the samples were immersed in isopropyl alcohol for 7 days and afterwards polished in order to obtain specimens with parallel faces.

The hardness was determined using microindentation tests. The tests were performed using a Vickers indenter (Duramin Microhardness Tester), applying a weight of 4 g during 5 s. For each sample, 20 indentations were performed in a paper grid of 5 × 4 mm with separations of 0.5 mm.

The Fourier Transformed Infrared Spectroscopy (FTIR) analysis was conducted in a NICOLET 6700 Thermo Scientific spectrometer with a DGTS CsI detector, and 64 scans were recorded to register each sample. The samples were prepared by mixing the samples with KBr. The scans were taken in the mid-infrared region at frequencies of 4000–400 cm^{-1}, with a spectral resolution of 4 cm^{-1}.

In order to study the microstructure of the samples and the morphology of the developed phases backscattered electron (BSE) images were obtained using a Scanning electron microscopy (SEM) JEOL JSM-6300 attached to a LINK ISIS-200 for energy dispersive X-ray analysis (EDS).

The degree of hydration (i.e. the volume fractions of unreacted cement and hydration products) was measured by image analysis on BSE–SEM images following the methodology proposed by Wong and Buenfeld [22]. As proposed by these authors, the degree of hydration (m) can be estimated with the following equation:

$$m = \frac{V_{HP}}{\delta_v \cdot V_{AH} + V_{HP}} \quad (1)$$

where V_{HP} and V_{AH} represent absolute volumes of hydration products and unreacted (anhydrous) cement respectively and δ_v the volumetric ratio of hydration products to the reacted cement. The volumes of hydrated products and anhydrous particles were obtained from the areas of the different grey levels of the BSE–SEM images. The fraction of hydration products was obtained with a segmentation of the grey levels whereby the lower threshold value is taken as the inflection point of the cumulative brightness (porous zone and fibres) and the upper from the minimum grey value between the peaks of hydration products and the unreacted cement [22].

Table 1
References of group of samples, mix proportions and water/cement ratio.

Group of samples	Cement (g)	Silica fume (g)	Fibre (g)	Initial water/cement ratio	Final water/cement ratio
PC	300	0	0	0.53	0.53
PC-SF	270	30	0	0.42	0.42
PC_F	300	0	12	0.80[a]	0.53[b]
PC-SF_F	270	30	12	0.80[a]	0.42[b]

[a] These pastes were prepared with higher content of initial water to allow the mixture with the sisal fibres.
[b] The final ratio water/cement was calculated measuring the water eliminated during the preparation of the pastes.