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Preliminary Study of the Mechanical and Hygrothermal Performance of Concrete Reinforced with Fibrillated Cellulose

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Abstract. Cement, being the most widely used building material, is the responsible for a large share of greenhouse gas emissions. To reduce the environmental impact of its production, natural fibres can be used as eco-friendly additives. Moreover, their potential use in traditional lime-based mortars makes them an ideal choice for green buildings as well as for the retrofit of historical buildings. An innovative cementitious composite reinforced with fibrillated cellulose (hereafter called «green concrete») was tested to assess its mechanical and physical properties. Samples were casted using Portland cement and natural hydraulic lime and varying the ratios among the constituents. Viscosity and setting time of the fresh pastes were determined with a viscosimeter and a Vicat apparatus, while their hydration was studied by thermal analysis. The influence of the fibres on the flexural strength of the final composite was determined through mechanical tests. The expected hygrothermal performance of the «green concrete» was explored through dynamic hygrothermal simulation to investigate its potential use as a retrofit material. A sensitivity analysis (SA), based on the hygrothermal properties of natural-based building materials similar to the «green concrete», was conducted to identify the parameters influencing more the simulation of annual internal temperature and moisture variations. The preliminary assessment of the mechanical properties of the «green concrete» showed that at higher percentages the cellulose fibres can negatively affect the workability/setting time of the fresh pastes and the flexural strength. The most promising samples were identified and will undergo further investigation. The SA results outlined that the «green concrete» might not be effective for thermal insulation, although it might be used as a moisture-buffering layer by adjusting the values of the free water saturation moisture content, the equilibrium moisture content at RH=80% and the dry vapour diffusion resistance factor of the final composite.

INTRODUCTION

Cement is the most widely used building material and its production is the responsible for a large share of greenhouse gas emissions. For this reason, several efforts have been directed towards the development of the concept of a «green concrete», e.g. by using eco-friendly additives to reduce the environmental impact of concrete, thus benefiting the sustainability. Natural fibres, commonly used during ancient times to reinforce and to reduce the shrinkage in mortars and concrete, are nowadays receiving a growing interest since they represent a renewable, economical and abundant resource [1].

Nanocellulose materials can overall enhance the mechanical properties of modern concrete due to their high aspect ratio and high Young's modulus [2], [3], [4]. Moreover, due to their high hygroscopicity they can act as an internal curing agent of cement, preventing self-desiccation and promoting hydration [5]. Their use as viscosity modifiers in self compacting concrete (SCC) allows to stabilise the fresh concrete, inhibiting bleeding and segregation phenomena [6]. On the other hand, some disadvantages must be considered: for example, being highly hydrophilic, their incorporation can greatly decrease the workability of the fresh concrete, making the casting process more difficult. The use of natural fibres in concrete can also influence the composite porosity and its hygrothermal behaviour [7], [8]. In particular, cellulose fibres enhance the moisture transfer and the storage capacities of mortars and have been classified as excellent hygric regulators. To the extent of our knowledge, no studies have been reported so far on the influence of nanocellulose on the physical and hygrothermal properties of lime-based mortars.

The use of nanocellulose in traditional lime-based mortars is a promising solution for green buildings as well as for the retrofit of historical buildings [9]. The fibres can increase the flexural strength and avoid crack propagation in mortars and plasters, delaying the need for maintenance interventions. Moreover, they could be employed as a compatible and sustainable retrofit alternative for historic buildings to improve the energy efficiency and thermal comfort without using active systems for the indoor climate control/management [10]. Passive solutions -such as the thermal and moisture insulation of the building envelope- are preferable in the case of historical buildings, being less invasive and economically advantageous.

A versatile tool to assess retrofitting solutions is the whole-building dynamic simulation, which allows to non-invasively study the expected hygrothermal performance of materials according to their physical and hygrothermal properties. For this reason, it has been increasingly exploited to optimise energy efficiency, human comfort and conservation in historical buildings [11]. The European projects 3ENCULT (2007-2013) and EFFESUS (2012-2016) focused on the study through dynamic simulation of passive and active retrofit solutions for historical buildings. For this kind of buildings, it is crucial to reliably assess their impact in terms of climate-induced deterioration risks [12] before any intervention. Moreover, pro and cons of the use of newly-developed materials need to be thoroughly studied before their application [13], since they have never been tested in complex structures.

Although simplifications can be introduced in many cases, due to the lack of sensibility of the most common simulation models to moisture-related properties of retrofit materials [14], the whole-building dynamic simulation integrating HAM (Heat, Air and Moisture) models is considered adequate to reproduce the heat and moisture exchanges between the historical envelope components and the indoor climate [15]. In general, to obtain accurate results in the simulation of heat and moisture transport across the walls, it is necessary to reliably know the stratigraphy and the properties of the constituent materials, which however are not fully known - particularly in the case of newly-developed materials. The physical and hygrothermal properties of innovative natural-based building materials available in literature are measured following various procedures [8], [16], [17]; moreover, as they depend on both the constituent materials and the manufacturing process, they can have rather different hygrothermal properties [18]. Sensitivity Analysis (SA) can be employed to determine the most influential parameters on the simulation results and to quantify their effect on the variability of the outputs. In particular, being SA results strongly related to the specific configuration to be tested, the analysis must be performed in relation with the modelling goals (e.g. the evaluation of the effectiveness of insulation solutions), carefully choosing the variability range of the input parameters [19].

The present paper is a preliminary study about the characterisation of an innovative «green concrete» reinforced with fibrillated cellulose and for evaluating its behaviour as hygrothermal layer for the retrofit. The work is divided in two sections: 1) preliminary assessment of the physical and mechanical properties of the fresh and hardened

samples of the «green concrete» reinforced with fibrillated cellulose; 2) identification of the parameters influencing more the hygrothermal performance of the innovative composite through a sensitivity analysis.

MATERIALS AND METHODS

Samples of an innovative «green concrete» made of a cementitious composite reinforced with two different commercial fibrillated cellulose (FC), here called type I and type II, were casted using Portland cement (CEM II/B 32.5 R) and natural hydraulic lime (NHL 3.5) and varying the ratio among the constituents. No superplasticiser was added in order to focus the study on the effect of the fibres on the pastes and the mortars.

The influence of the fibrillated cellulose on lime and cement pastes was evaluated using a Haake VT500 viscosimeter. The apparent viscosity (η) versus time (t) was registered at different fibre contents keeping the water to binder ratio (w/b) constant. A w/b of 0.35 and 0.5 was chosen for Portland cement and natural hydraulic lime pastes respectively. The premixing *iter* proposed by Nehdi et al. [20] was then used to calculate the plastic viscosity (μ_p) and the yield stress (τ_0) of the pastes based on the experimental shear stress (τ) and shear rate ($\dot{\gamma}$). μ_p and τ_0 were calculated by two different modified Bingham equations: equation (1) was used to approximate the curve from 0 to 10 s^{-1} shear rate [21], while equation (2) was used to fit the data from 10 to 50 s^{-1} shear rate.

$$\tau = \tau_0 \left[1 - \exp\left(-3 \frac{\dot{\gamma}}{\dot{\gamma}_{crit}}\right) \right] + \mu_p \dot{\gamma} \quad (1)$$

$$\tau = \tau_0 + \mu_p \dot{\gamma} + c \dot{\gamma}^2 \quad (2)$$

With $\dot{\gamma}_{crit}$ and c being critical shear rate and regression constant, respectively.

Mortars with 1% of fibres by weight of binder were prepared (Table 1) on the basis of the literature survey [4], [7],[22]. The water to binder ratio was adjusted for each mix, according to the rheological findings, in order to contrast the loss of workability induced by the higher FC amount. The cellulose fibres were magnetically stirred with the mixing water for 10 minutes and then sonicated for 15 minutes in an ultrasonic bath. The obtained gel was added to the binder and mixed for 5 minutes. The aggregate (0/4 siliceous sand) was then slowly added over 1 minute while mixing. The mortar was finally mixed for other 5 minutes to get a homogeneous product.

TABLE 1. Samples series of the «green concrete» made with Portland cement (P series) and Natural hydraulic lime (N series) with 1% of two different commercial FC (Type I, Type II), mixed using different water/binder ratios (w/b).

Sample	Binder	Fibre	w/b
P0	CEM II/B 32.5 R	-	0.57
P1	CEM II/B 32.5 R	Type I	0.87
P2	CEM II/B 32.5 R	Type II	0.87
N0	NHL3.5	-	0.62
N1	NHL3.5	Type I	1.00
N2	NHL3.5	Type II	0.96

A Vicat apparatus was used to study the influence of the fibres on the setting time (t_0) of the designed mortar mix containing 1% of fibres. Samples of 20 × 20 × 80 mm were casted to investigate the flexural strength of the composite at 28 days by three-point bending test (Fig. 1). For curing, fresh samples were covered with plastic sheets (to control evaporation) and kept at laboratory temperature 22±2°C for 24 h before demoulding. After demoulding, the samples were transferred in a climatic box and kept at 100% RH and temperature 22±2°C until the age of testing. For each mix, three different samples were casted.



FIGURE 1. Samples of the innovative «green concrete» used for the flexural strength test.

The software IDA ICE (Indoor Climate and Energy) 4.8 extended with the HMWall (Heat and Moisture transport) model was chosen for the whole-building modelling of the hygrothermal performance of the composite [23]. An exploratory literature survey was conducted to collect the hygrothermal properties measured on various types of natural-based construction materials similar to the «green concrete»: three hemp concretes [8], a cement mortar filled with date palm fibres [16], clayey plasters with olive fibres [17], and two corn stalk concretes [18]. Then, a Sensitivity Analysis (SA) was carried out to identify the hygrothermal parameters in the model influencing more the behaviour of the considered natural-based construction materials. Table 2 summarises the intervals of values used in the SA based on the hygrothermal properties of the referred natural-based building materials.

TABLE 2. Ranges of the selected hygrothermal properties tested in the Sensitivity Analysis based on the hygrothermal properties measured on the referred innovative natural-based construction materials.

Sensitivity analysis inputs	Range
Density (ρ)	400–950 kg/m ³
Specific heat capacity (C_p)	850–1500 J/(kg·K)
Dry thermal conductivity (λ_0)	0.06–0.20 W/(m·K)
Thermal conductivity supplement (b)	1–5
Free water saturation moisture content (w_f)	65–430 kg/m ³
Equilibrium moisture content at RH=80% (w_{80})	20–60 kg/m ³
Dry vapour diffusion resistance factor (R_f)	6–25

The SA was conducted by using the Elementary Effects (EE) method based on the Morris random sampling method [24]. The Morris random sampling considered 10 EE for each parameter and 4 discretised levels to span within the ranges of the selected hygrothermal parameters in Table 2. In Fig. 2 is shown a synthetic scheme of the simplified configuration used in the Sensitivity Analysis. A 50-cm thick wall made of traditional Belgian bricks and covered with a 3 cm thick internal layer of «green concrete» was considered. The brick wall was connected, on the external side, to the average climate in Rome [25] and, on the internal side, to indoor constant conditions of T=20°C and RH=50%. The values of the hygrothermal properties of the bricks were taken from the MASEA database [26]. The presence of internal and external plasters was neglected as its effects are usually low for thick masonry walls [27]. The maximum annual variations of temperature (ΔT) and relative humidity (ΔRH) in the middle of the «green concrete» layer were calculated from the hourly simulations run over an entire year and chosen as the target outputs in the SA.

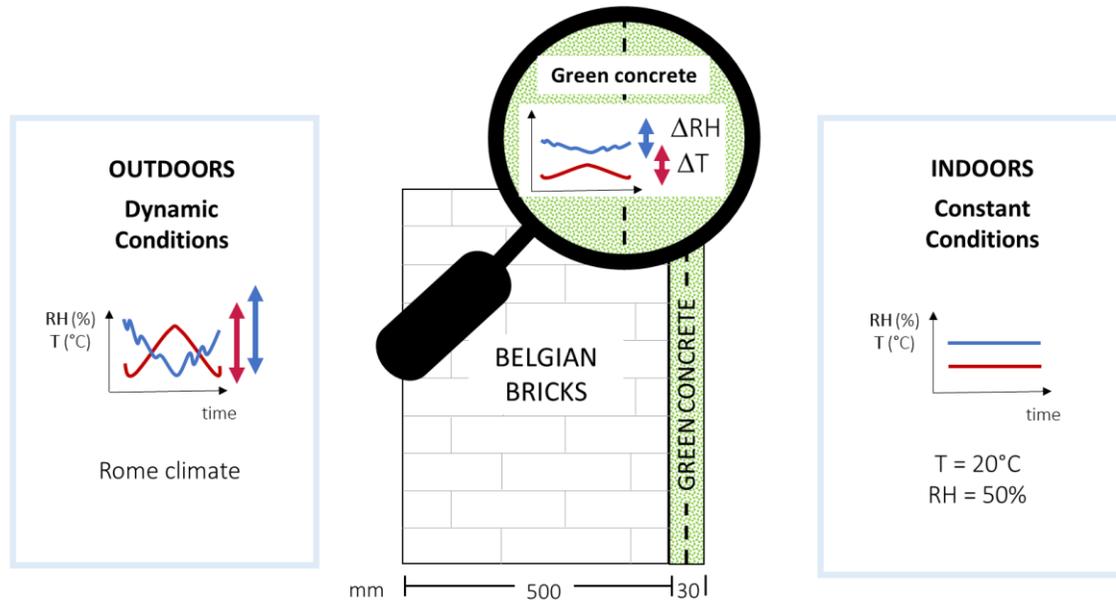


FIGURE 2. Scheme of the configuration of the wall model used for the Sensitivity Analysis based on the maximum annual variations of temperature (ΔT) and relative humidity (ΔRH) in the middle of the «green concrete» layer over a year.

RESULTS AND DISCUSSION

The plastic viscosity and the yield stress could be calculated only for the lime pastes, due to the viscosimeter limit. The four samples exhibit a thixotropic behaviour and after 150s all the curves reach a plateau (Fig. 3a). The fibre content (FC) clearly influences the workability of the fresh pastes and the apparent viscosity strongly rises in pastes with higher FC at early t. Figure 3b shows how the shear stress proportionally increases with the FC content. The yield stress (τ_0), after which the paste can flow, also shows a similar behaviour, with τ_0 increasing with FC; on the other hand, no remarkable differences were found in the average μ_p (Table 2). The adopted premixing procedure may have affected the results due to the shear thinning effect of the paste at higher shear rates [6], which is more pronounced in pastes with FC. Indeed, at high shear rates the network formed by the fibres in the paste can be broken, so that the fibrils start to align around the rotor along the direction of flow, reducing their influence on the fluid rheology [28].

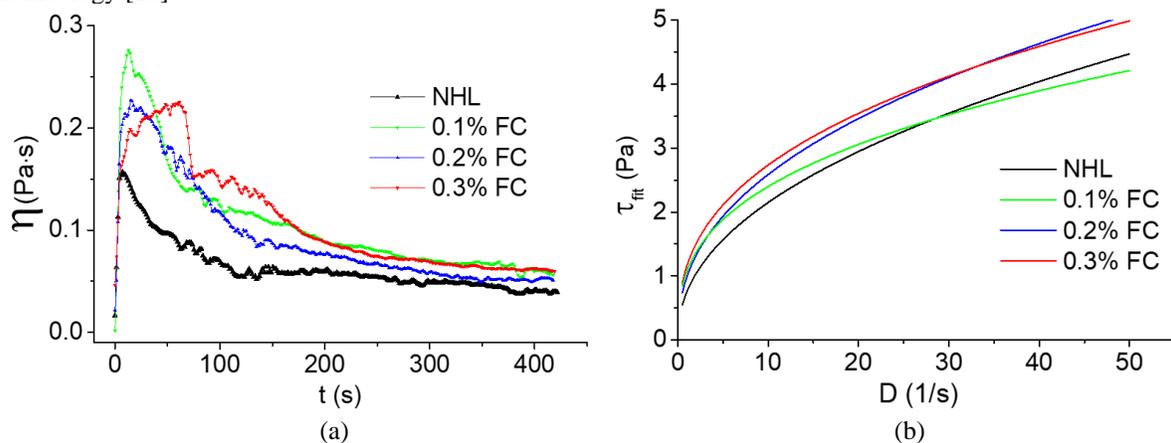


FIGURE 3. Apparent viscosity of the tested lime pastes as a function of time at 50 s^{-1} shear rate (a); shear stress obtained after the premixing *iter* calculated through the fitting eq. (1) and (2)(b)

TABLE 3. Average yield stress (τ_0) and plastic viscosity (μ_p) calculated from fit equations (1) and (2) for lime pastes at different fibres content.

Sample	τ_0 (Pa)	μ_p (Pa·s)
NHL	1.09 ± 0.20	0.09 ± 0.02
0.1%FC	1.55 ± 0.05	0.11 ± 0.01
0.2%FC	1.72 ± 0.02	0.10 ± 0.01
0.3%FC	1.89 ± 0.10	0.09 ± 0.01

For mixing 1% of fibres, the water to binder ratio (w/b) was adjusted to obtain a workable paste and then to prepare the mortar. A delay in the initial setting time was observed for both the Portland cement (P) and the Natural hydraulic lime mortars (N) with the two different FC (Fig. 4); indeed, the stronger delay was obtained with Type II FC (2) than Type I FC (1), both higher than blank reference (0), despite a similar w/b. Furthermore, the flexural strength (f_n) of the «green concrete» is negatively affected by FC (Table 4). However, also for this analysis, the Type II FC shows a worse impact on the properties of the mixtures, making Type I FC a preferable choice for future investigations. Since recent studies have demonstrated that lower amounts of fibres (0.05%-0.3%) can positively affect the properties of mortars and concretes [3], [29], a rethinking of the ratios among the constituents may help reducing the difficulties experienced in the incorporation in the pastes.

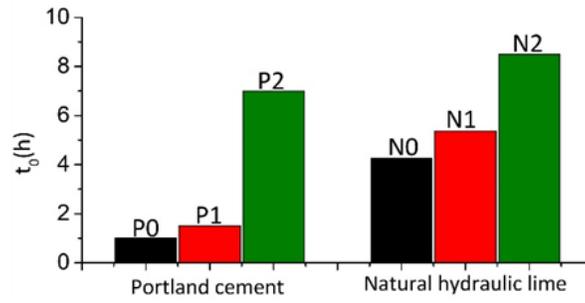


FIGURE 4. Initial setting time (t_0) for Portland cement and Natural hydraulic lime mortars calculated with a Vicat apparatus.

TABLE 4. Flexural strength (f_n) and flexural strength variation (Δf_n) at 28 days with 1% load of FC for Portland cement and lime mortars.

Sample	f_n (MPa)	Δf_n
P0	6.81 ± 0.10	-
P1	5.19 ± 0.17	-24%
P2	4.49 ± 0.30	-34%
N0	0.88 ± 0.18	-
N1	0.71 ± 0.07	-19%
N2	0.65 ± 0.05	-26%

The obtained results were a useful starting point to organise a new series of tests and to perform a Sensitivity Analysis on the physical and hygrothermal properties of the referred innovative natural-based construction materials. The wall stratigraphy used in the SA was chosen according to the preliminary assessment of the mechanical properties, which made the «green concrete» more suitable to be used as an indoor retrofit layer. The results of the Sensitivity Analysis are shown in Fig. 5 in terms of the mean (mi^*) and the standard deviation (σ) of the Elementary Effects (EEs) calculated on the maximum annual variations at the middle of the «green concrete» layer, as schematised in Fig. 2. In particular, Fig. 5(a) describes the EEs calculated on the annual gradients of temperature (ΔT), while Fig. 5(b) describes the EEs calculated on the annual gradients of relative humidity (ΔRH). The estimated relevance of a parameter is given by the value of mi^* , while the ratio σ/mi^* is an indicator of the linearity of the EEs [30].

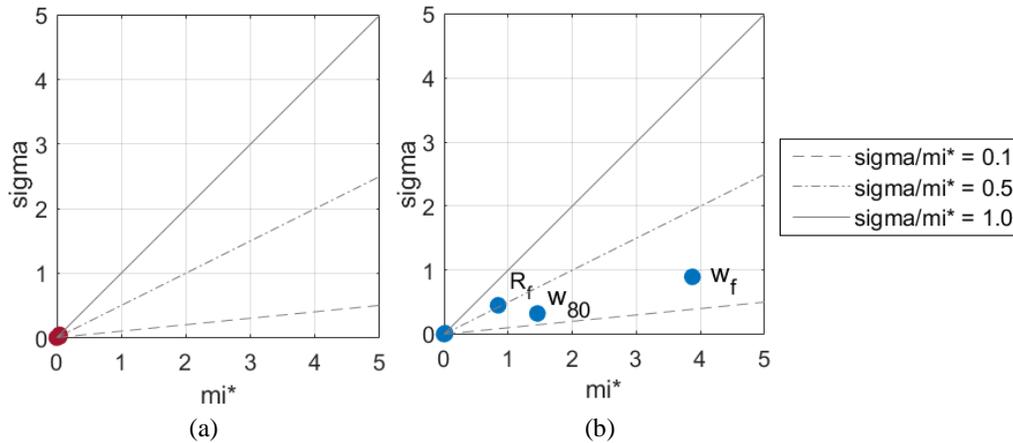


FIGURE 5. Results of the Sensitivity Analysis (SA) performed as described in Fig. 2. The SA results are expressed in terms of the mean (mi^*) and the standard deviation (σ) of the elementary effects calculated on the annual variations of temperature (a) and relative humidity (b).

As evident in Fig. 5(a), none of the considered parameters can significantly affect the simulation of the annual thermal variations ($mi^* \ll 1$), meaning that the composite might not be effectively used for thermal insulation. On the contrary, Fig. 5(b) highlights that some of the moisture-related parameters can have an influence on the simulation of the annual moisture variations. In particular, the free water saturation moisture content (w_f) has a monotonic influence on the ΔRH , with EEs having a value of $mi^* \gg 1$ and low variability. As discussed in [30], when $\sigma/mi^* < 0.5$, then 95% of the EEs is concordant: if the value of the tested parameter increases, the same does the chosen output and *vice versa*. This effect is explained by the predominant role of w_f over the definition of the moisture storage capacity curve of the material and is physically consistent with the fact that the flatter is the curve, the more the variability of the material moisture content is reduced. The low mi^* and σ resulting from the EEs associated to the equilibrium moisture content at $RH=80\%$ (w_{80}) and to the dry vapour diffusion resistance factor (R_f) suggest that their influence on the simulated ΔRH is limited.

CONCLUSIONS

The investigation provided some interesting results about the mechanical and hygrothermal performance of a newly-developed «green concrete» reinforced with fibrillated cellulose.

The preliminary analyses showed that higher percentages of fibres can negatively affect the workability and setting time of the fresh mortars and the flexural strength of the «green concrete». The samples based on natural hydraulic lime and Type I FC resulted to be a preferable choice for future investigations as sustainable lime-based mortars. However, a redefinition of the mix design could help obtaining more workable and easy to cast mortars (e.g. using admixtures or lowering the amount of FC).

The Sensitivity Analysis outlined that natural-based building materials similar to the «green concrete», used as an indoor thin surface layer, cannot significantly affect the simulation of annual thermal variations, meaning that the composite might not be effective for thermal insulation. Nevertheless, as moisture-related properties can have a monotonic influence on the simulation of annual moisture variations, these parameters should be adjusted in order to use the composite as a moisture-buffering layer.

An experimental campaign is planned in 2021 to measure the hygrothermal properties of the «green concrete». Once the hygrothermal properties of the final composite will be available, whole-building dynamic hygrothermal simulations will be performed to investigate its use as a retrofit material.

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