Results in Engineering 7 (2020) 100150

Contents lists available at ScienceDirect

Results in Engineering

journal homepage: www.editorialmanager.com/rineng/Default.aspx

Study of modifications on the chemical and mechanical compatibility between cement matrix and oil palm fibres

P.-A. Bonnet-Masimbert [a,b], F. Gauvin [a,*], H.J.H. Brouwers [a], S. Amziane [b]

[a] Department of the Built Environment, Eindhoven University of Technology, P. O. Box 513, 5600, MB Eindhoven, the Netherlands
[b] Université Clermont Auvergne, Institut Pascal, BP 20206, Clermont-Ferrand, France

ARTICLE INFO

Keywords:
Cementitious matrix
Oil palm fibres
Alkaline treatment
Rheology
Compatibility
Interface

ABSTRACT

Nowadays, the substitution of the traditional reinforcements of cement-based composite materials by natural fibres is more and more common. However, some problems remain, mostly because the addition of natural fibres to cement can delay its hydration due to the high concentration of polysaccharide in the fibres' leachates. Many studies have shown the effectiveness of alkaline treatments in order to improve the natural fibre/cement compatibility but the mechanical compatibility, i.e. the bonding strength between the cement and the fibres has never been characterized. Therefore, this study aims to fully understand the natural fibre/cement interface. Oil palm fibre is chosen as reference fibre because of its very low compatibility with cement. NaOH treatments and superplasticizer are used, in order to modify the surface of the fibre and the viscosity of the matrix. Both chemical and mechanical compatibilities are characterized, by various methods such as isothermal calorimetry, FTIR, tensile tests on single fibres and pull-out tests. Results show that alkali-treatments are very effective since it can improve both compatibilities, while a more fluid matrix allows to further enhance the properties of the interface, by limiting the amount of sugar at the fibre's surface without hindering the cement hydration.

1. Introduction

Nowadays, the concepts of the circular economy are becoming a key research question regarding the increasing demand for infrastructure and the necessity to address the current environmental issues, like the increasing greenhouse gas emissions, the raw resource consumption and the increasing energy consumption. Many fields are responsible, but as it can be seen in Fig. 1, the building sector is one of the most problematic, with 11% of global CO_2 produced [1,2]. The cement industry itself is responsible for the consumption of large quantities of raw materials, important energy demand and generate a massive amount of solid waste materials and hazardous gases such as NOx and SOx [3]. Thus, since the last few years, the field of civil engineering has been trying to adopt an environmentally friendly approach in order to solve these issues [4]. Many processes and innovations have emerged, such as the use of green roofs and solar or geothermal energy [5,6].

However, another way to reduce the energy footprint of buildings is to use natural or recycled materials for construction or insulation [8]. In this context, industries are trying to substitute conventional materials by so-called "green materials" with a much lower environmental impact. Thus, in recent years, extensive research has been performed in order to replace steel in concrete. Many types of fibre, such as glass or carbon, have been developed and tested. They have shown good performances combining low density and high mechanical properties [9,10]. However, for a better ecological approach, synthetic fibres can be substituted by natural fibres (NF) like flax, hemp, coir, pineapple or bagasse. All of them present different properties, but numerous studies have been shown their great potential as building material [11–20].

NF is a complex natural composite, made of cellulose, hemicellulose and lignin in several cell walls as shown in Fig. 2. The secondary wall is mainly composed by helically cellulose microfibrils, assuring the mechanical strength of the fibre, bonded together by both the hemicellulose and the lignin which act as a matrix [21–23]. Furthermore, NFs are also composed of a small number of extractable components such as wax, tyloses and pectin. Besides, cellulose and hemicelluloses are constituted of mono and polysaccharides such as glucose, sucrose or mannose, which can be easily solubilized in any aqueous medium [22,24,25].

Nevertheless, although the use of organic fibres as reinforcement in composite materials has increased, the association of natural fibres with cementitious matrices is still problematic. Indeed, the compatibility between lignocellulosic materials and cement is not always good and depends on several factors. For example, Na et al. [26] concluded that the

* Corresponding author.
E-mail address: f.gauvin@tue.nl (F. Gauvin).

https://doi.org/10.1016/j.rineng.2020.100150
Received 23 March 2020; Received in revised form 24 June 2020; Accepted 24 June 2020

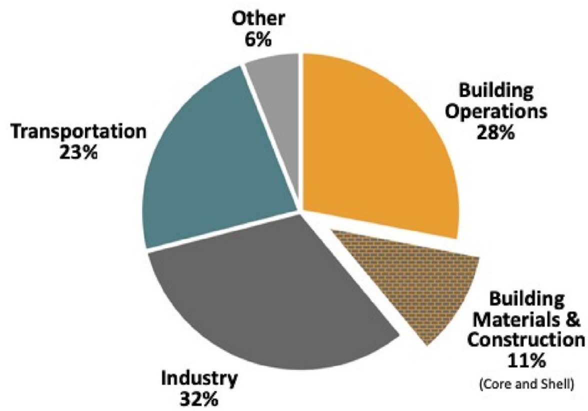


Fig. 1. Global CO_2 emissions by sector, from the Global Alliance for Buildings and Construction [7].

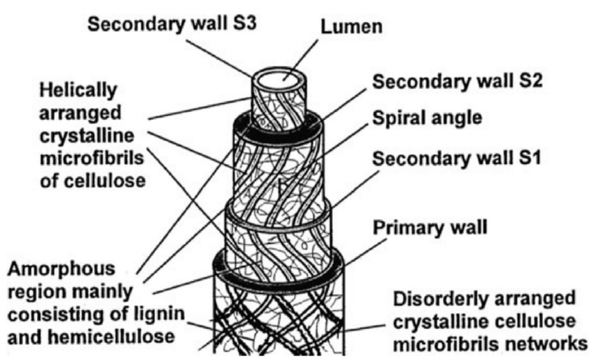


Fig. 2. Structure of a natural fibre cell, from S. Thomas et al. [22].

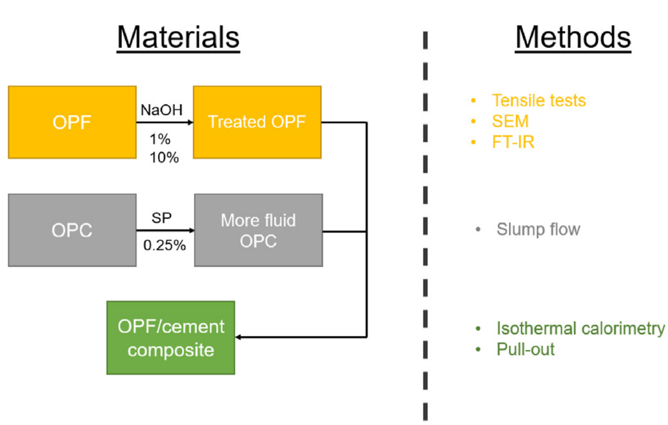


Fig. 3. Process of the study, with the materials and the test methods.

compatibility between wood and cement depends on the type of wood, its growing location, the part of the tree used, the season during wood-cutting, the storage conditions or also the type of cement. All these factors point out that the main issue for NF-cement composite is the chemical incompatibility which can exist between the two components. Indeed, organic fibres contain organic substances which can leach out, having inhibitory effects on the hydration of cement and therefore negatively affect the properties of the composite. This phenomenon is due to a large number of polysaccharides, which leach out during the hydration phases of the cementitious matrix thereby affecting its hydration reaction [26]. However, not all types of polysaccharides have the same inhibitory effect. Firstly, Na et al. [26] related that glucose and sucrose are the main issue. In a second time, Ko et al. [27] confirmed this study and demonstrated that the sucrose has the highest impact while glucose is a strong retarded, which was confirmed by Do et al. [28].

In order to reduce the negative impact of fibres on the cement matrix and to improve the compatibility between them, different solutions can be used. One of the most studied and effective solutions is the chemical treatment of NF in order to remove unwanted constituents [21,26,29,30]. Indeed, many studies were performed on fibres having strong incompatibility with cement, and pre-treatments have been very efficient in order to make them compatible. Among the existing solution, sodium hydroxide treatments are the most effective solution [21,26,31–34]. Indeed, Sa et al. [30] have reported that NaOH treatment is able to make many wood species and NF to be fully compatible with Portland cement (OPC). This alkali treatment acts on the surface of the fibre and allows to remove the main inhibitory components that can be in contact with cement. Moreover, hemicelluloses, lignin and surface impurities such as pectin, wax, tyloses and fatty substances are removed thereby increasing the surface roughness of the fibre which is needed to achieve good bonding with matrix [25,33–38]. According to Su et al. [21], a large variety of pre-treatments can be done on natural fibres in order to increase their mechanical, chemical, hydrophilic and thermal properties. However, in this study, only pre-treatments used to improve the compatibility between cement and fibres are evaluated. Besides, alkaline treatments can damage the fibre by decreasing significantly its properties. Indeed, above a certain concentration, the alkali treatment can degrade and damage fibres by removing the hemicellulose and the lignin that act as a matrix, but also the cellulose, which lead to a drop in their mechanical properties [29,39,40]. Yet, another way to improve the cement-fibres compatibility would be to modify the viscosity of the matrix in order to change the sugars diffusion. Indeed, sugars diffusion is affected by the matrix viscosity, and a more fluid matrix could mitigate this issue [41].

The main goal of this study is to understand the mechanical and chemical behaviour of natural fibres when mixed with cement, in order to ultimately solve this issue to pave the way to the utilization of natural fibres in the building sector. This study aims to compare and combine the effect of the treatment on fibres and the modification of the matrix in order to improve the compatibility between NF and cement, but also to have a better understanding of these phenomena. The natural fibres chosen in this study are oil palm fibres (OPF), a lignocellulosic fibre extracted from the empty fruit bunches which have already been used in many applications [42,43]. Besides, a recent study has shown that OPFs are not compatible with cement, because their leachates are too rich in glucose, galactose and arabinose, which hinder the cement hydration [27]. Therefore, OPF is the perfect case study for this research. In order to improve its compatibility with cement, an alkali-treatment is done on OPF [44]. The influence of the cement viscosity is investigated by adding superplasticizer into cement paste in order to study a more fluid matrix. For each condition, the compatibility is studied by isothermal calorimetry. Then, the mechanical properties of the interface, between cement and fibres, are examined by single fibre pull-out tests.

2. Materials and methods

The whole methodology is summarized in Fig. 3

2.1. Materials

Oil palm fibres are provided by WUR: Wageningen University and Research, the Netherlands. The chemical composition of these fibres has been studied by different researchers and thanks to Sh et al. [45], these data have been summarized in Table 1. It can be observed that OPFs are mainly composed of cellulose, but also with an amount of hemicellulose and a high concentration of sugars (such as glucose and xylose) in its leachate. Mechanical properties of OPFs are summarized in Table 2 [46]. It can be observed that despite the high heterogeneity of these values, these fibres are often quite light, brittle and strong. These fibres are

Table 1
Chemical composition of OPF from the literature [45].

Constituents	%
Cellulose	42.7–65
Lignin	13.2–25.31
Hemicellulose	17.1–33.5
Ash content	1.3–6.04
Alcohol	2.7–12
Pentosan	17.8–20.3
Arabinose	2.5
Xylose	33.1
Mannose	1.3
Galactose	1
Glucose	66.4
Silica	1.8

Table 2
Mechanical properties of OPF from the literature [46].

Properties	Value
Density (g/cm^3)	0.7–1.51
Tensile strength (MPa)	25–550
Young's modulus (GPa)	0.5–9
Elongation at break (%)	4–18

stored in plastic bags at room temperature until use. For this study, CEM I 52.5R (OPC) provided by ENCI, the Netherlands, is used as a binder and Table 3 gives its chemical composition. A superplasticizer (SP) called MasterGlenium 51 con 35% from BASF, the Netherlands, is also used. Finally, Sodium hydroxide (NaOH) is produced by VWR CHEMICALS, Belgium.

2.2. Alkali treatment

The most common treatment of natural fibres is based on alkaline solutions such as NaOH, NaOCl or $Na(OH)_2$ [47,44]. In this study, sodium hydroxide has been chosen because of its effectiveness on the removal of unwanted constituents such as lignin or hemicellulose. OPF is treated with 2 different concentration of NaOH (1% and 10%) and 3 different soaking times (2 h, 6 h, 24 h). For each solution, pH is measured by a pH Meter (Metrohm 780) and is always above 12. OPFs are soaked in these two NaOH solutions with the same conditions, namely 6 h and 24 h at room temperature (20 °C). Then, they are washed several times with water and dried at 60 °C for more than 24 h.

2.3. Tensile tests

The tensile properties of oil palm fibres are measured using an INSTRON 5957 universal testing machine with a 100N load cell, following the ASTM D3822 standard. The measurements are conducted with a crosshead speed of 5 mm/min. For each condition, about 20 specimens are tested and the average strength of the fibres is determined. Fibres are mounted between the grips where each of the upper and lower jaws covers 2 cm of the sample. Due to the OPF macrostructure (i.e. short fibre with an average length of 6 cm), the distance between the jaws is set at 1–2 mm. The tensile stress is measured as a function of the linear density of the fibre (in tex) in order to minimize the impact on stress riser, quite common in heterogeneous natural fibres.

Table 3
Chemical composition of OPC used in this study.

Chemical composition	%
CaO	64.6
SiO_2	20.08
Al_2O_3	4.98
Fe_2O_3	3.24
K_2O	0.53
Na_2O	0.27
SO_3	3.13
MgO	1.98
TiO_2	0.30
Mn_2O_4	0.10
P_2O_5	0.74
Cl-	0.05

2.4. SEM

The characterization of untreated and treated fibres surfaces is performed by a Scanning Electron Microscope (SEM) (Phenom ProX), with BSE detector at an accelerating voltage of 10 kV. Before the scanning, each fibre is coated by a thin layer of gold to become more conductive and suitable for SEM analysis. Pictures are taken at two magnifications:×400 and ×1800.

2.5. FT-IR spectroscopy

In order to characterize the effect of the treatments on components at the surface of the fibres, Fourier Transform Infrared Spectroscopy (FT-IR) spectra are recorded. For this, a PerkinElmer Frontier FTIR with a GladiATR diffuse reflectance device is used. Fibres treated with a 1% NaOH treatment and different times of soaking (0 h, 2 h, 6 h and 24 h) are studied. Hence, six scans are acquired with optical retardation of 0.25 cm and a resolution of 4 cm^{-1} from 4000 to 400 cm^{-1}. Characterization of the peaks corresponding to the different constituents of the fibres (e.g. lignin, wax, hemicellulose …) has been done in accordance with previous studies [48].

2.6. Processing of the cement pastes and slump (flow) tests

In order to study the influence of superplasticizer on the viscosity of cementitious matrix, for the w/c ratio set at 0.5, slump flow tests are performed following the EN 196-1 [49] standard. An optimum amount of SP is set at 0.25% (m/m binder). Table 4 shows the viscosity difference between the OPC and the OPC + SP samples.

2.7. Isothermal calorimetry

Isothermal calorimetry measurements are performed with a TAM Air Isothermal calorimeter at a constant temperature of 20 °C during 3–6 days. Untreated and treated fibres are mixed with OPC and water. For all samples, the water/cement ratio and the fibres content are kept constant at 0.5 (w/c = 0.5) and at 7.5% (f/b = 0.075) respectively. For some samples, 0.25% of SP is added to the mix. Firstly, cement, water and SP are mixed during 30 s. Then fibres, with a size of 1–3 cm, are added to the paste and is mixed during 90 s.

2.8. Cement-fibres compatibility

Following the work of Na et al. [26] on the compatibility between wood and cement, the equation of Pasca et al. [50], based on isothermal calorimetry results is used in this study. As Kochova et al. [44] have reported in their work, this equation is developed for cement-wood compatibility but can be used for other types of lignocellulosic materials such as OPF, due to the assumption that hydrated cement/wood/water mixtures and cement/fibres/water mixtures tend to have the same

Table 4
Summary of the viscosity characterization by mini-slump test.

W/C ratio	SP concentration (%)	Diameter (mm)	Yield stress (Pa)	Viscosity (Pa.s)
0.5	0	150	72.73	2.19
	0.25	440	0.33	0.04

cement hydration behaviour as they have similar chemical composition. Thus, the compatibility factor CX', between fibres and cement, can be determined using the following equation [26,50]:

$$CX' = \sqrt[3]{\left(\frac{HRmax*HR3.5-24*t'max}{HR'maxHR'3.5-24*tmax}\right)}*100 \tag{1}$$

where HR_{max} is the maximum heat rate of the fibres-cement mixture (J/hg), HR'_{max} is maximum heat rate of neat cement paste (J/hg), $H_{3.5-24}$ is the total heat released by fibres-cement mixture in 3.5–24 h interval (J), $H'_{3.5\text{-}24}$ is the total heat released by neat cement paste within 3.5–24 h interval (J), t_{max} is the time to reach maximum heat rate of the fibres-cement mixture (h), t'_{max} is the time to reach maximum heat rate of neat cement paste (h). To classify the compatibility of fibres with OPC, three criteria are determined. Thus, fibres are considered incompatible if CX' < 40, moderate compatible if $40 <$ CX' < 80 and compatible if CX' $>$ 80.

2.9. Pull-out tests

In this study, single fibre pull-out tests are performed by using a universal testing machine (INSTRON 5967) and specific grips that can hold the cement block as well as a single fibre. Different samples with different conditions are studied in this work. For each condition, 8 samples are made and tested after 7 and 28 days. After mixing, the OPC mixture is poured into one-half of the dog-bone shaped moulds, in which a single OPF fibre is carefully held in place by EPS foams. The cast specimens are covered with plastic films and stored at room temperature prior testing. During the sample preparation, special care has been taken to keep the fibre straight while pouring the cement. The pull-out load is recorded with a 100N load cell under displacement control at a rate of 5 mm/min.

According to recent studies [51–53], the characteristic single fibre pull-out curve is composed of three different stages, partial debonding (OA), complete debonding (AB) and interfacial debonding (BC). The final stage, also called the pull-out stage, is characteristic of the nature of the frictional interface. Three main regimes can occur, which are the constant friction, the slip-softening and the slip-hardening (Fig. 4).

During the constant friction, the load decreases linearly while the slip increases. This can be attributed to the fact that stress across the interface and the coefficient of friction remains the same during this process [52]. During the slip-softening, there is a rapid decrease of the load while the slip increases. This can be attributed to the fact that the fibre hardness is higher than that of the surrounding matrix [51,52], which means rather a bad interface. During the slip hardening, the load increases while the slip increases. This can be attributed to the fact that fibres can get damaged causing jamming effect and thereby resisting pull-out load [51,52], which corresponds to a rather good interface.

3. Results and discussion

3.1. Chemical characterization of fibres

As described in the literature, hemicellulose, waxes, pectin and other surface impurities, which are present in OPF, are known to delay the hydration of cement [27,35,38]. Therefore, the FT-IR characterization is focusing on these constituents. The FT-IR spectra of untreated and treated fibres are given in Fig. 5. Three main differences are observed between untreated and treated OPF FT-IR spectra. The first one is the removal of the peak at 1710 cm^{-1}, attributed to the C=O stretching of the acetyl groups of hemicellulose, for treated fibres [35,37,54]. Indeed, even for a 2 h of soaking, this peak is not present, which mean that the hemicellulose is mostly removed from the fibre surfaces with any type of alkali-treatment. Secondly, by increasing the soaking time, two peaks at 2920 cm^{-1} and 2850 cm^{-1}, corresponding to the $-CH_2$ bonds of the long aliphatic hydrocarbon chain of waxes [24,44], become weaker and weaker. According to the literature, NaOH treatment is quite effective as removing wax from natural fibres, as it has been characterized with sisal fibres by Mishra et al. [55]. Finally, the last interesting peak visible in the spectra is characterized at 1240 cm^{-1} and it corresponds to the C–O aryl group of lignin (i.e. phenolic C–OH) [56,57], which also almost disappear with alkali-treatments. This indicates that the longer the treatment is, the more organic constituents are eliminated from the surfaces of the fibres since these peaks are disappearing after 24 h. Indeed, an increase of the concentration and/or time of the alkaline treatments remove more constituents such as wax, lignin and hemicellulose as it has been shown numerous times in the literature [58].

3.2. Surface characterization of fibres

The surfaces of untreated and treated OPF are shown in Fig. 5. According to the literature, it is known that OPFs are covered by waxes and other natural impurities [45,59,60]. Comparing the untreated fibres (Fig. 6-a and Fig. 6-b) to the treated fibres (Fig. 6-c to Fig. 6-h), it appears that the sodium hydroxide treatment removes a layer from the OPF surface. According to Ib et al. [38] and to Ma et al. [61], NaOH breaks the hydrogen bonds of the cellulose structure resulting in the removal of impurities. Thus, the higher the sodium hydroxide concentration, the more hydrogen bonds are broken and the more impurities are being removed.

After a weak treatment, it is noticed that a large number of micrometric compounds are still attached to pits on the OPF surface (Fig. 6-d). These particles are silica bodies which are commonly found at the surface of oil palm and coir fibre. The longer the soaking time (2 h: Fig. 6-d; 6 h: Fig. 6-f and 24 h: Fig. 6-h), the greater the number of pits are opened due to the removal of silica. Indeed, by analyzing the SEM pictures, it can be found that the 2 h and 6 h treatments remove 6% and 8% of the silica bodies, respectively, and the 24 h treatment is significantly more effective as it removes 55% of the silica bodies. Sr et al. [60] and Ib et al. [38] have shown that the pores formed due to the removal of silica become more prominent with stronger alkali treatment, with an average diameter growing from 0.07 μm to 0.15 μm. Therefore, the dissolution of natural impurities, the removal of silica bodies and the increase of the size of the pores leading to a more porous and a rougher fibre surface [61–64].

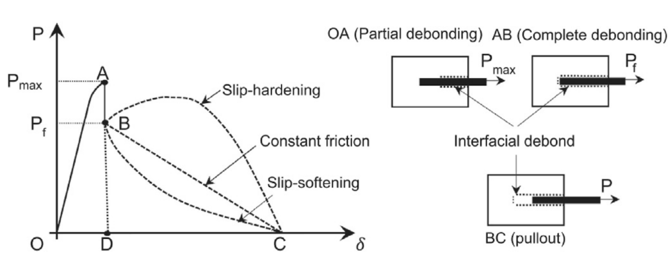


Fig. 4. Typical single fibre pull-out curve and pull-out mechanisms [44].

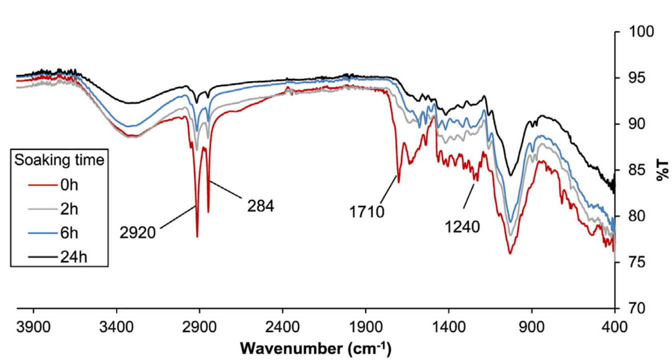


Fig. 5. FT-IR spectra of different soaking times of 1% NaOH treatment.

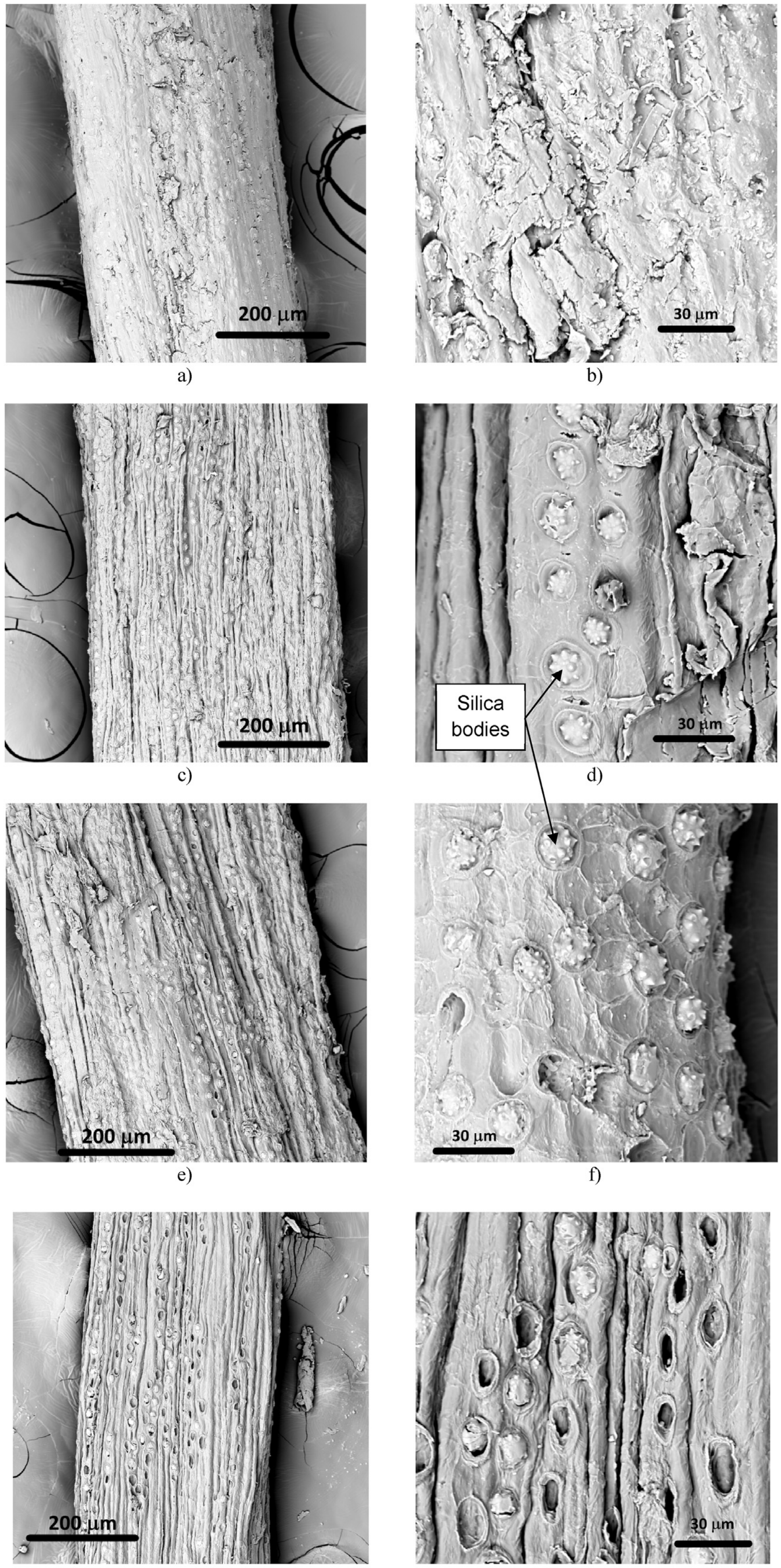


Fig. 6. a) Untreated OPF; b) Untreated OPF; c) 2 h Treated OPF; d) 2 h Treated OPF; e) 6 h Treated OPF; f) 6 h Treated OPF; g) 24 h Treated OPF; h) 24 h Treated OPF.

3.3. Mechanical properties of fibres

In order to evaluate the effect of different NaOH treatments on the mechanical properties of oil palm fibres, the tensile stress of untreated and treated fibres is measured and compared. Results are shown in Fig. 7. The first observable result is that weak treatments (1% NaOH) do not affect the tensile stress of the OPF, regardless of the soaking time, whereas stronger treatments have a significant effect on the OPF. Indeed, for a soaking time of 6 h, a significant increase is noticed (50% for 10% NaOH), while for a 24 h of soaking, a reduction of the strength is observed (20% for 10% NaOH).

From the literature, oil palm fibres can be considered as a fibre-reinforced composite, with cellulose fibrils as reinforcement and a hemicellulose-lignin network as a matrix [21,22,24]. Thus, cellulose mainly ensures the mechanical properties of fibres, while hemicellulose and lignin act as a bonding agent. As characterized by FT-IR and SEM, the alkali-treatment removes from the OPF surface different impurities, such as waxes and silica bodies, but also removes a part of the hemicellulose and lignin. Hence, as stated in the literature, the increase of treated OPF tensile stress can be explained by this removal of these components because according to Cs et al. [36], Sy et al. [39] and Iz et al. [64], with less binding hemicellulose and lignin in the interfibrillar regions, the fibre becomes less rigid and allows cellulose microfibrils to re-arrange themselves along the direction of the load applied. This promotes a more effective load transfer and an increase in tensile properties. Moreover, another explanation of this phenomena can be that the re-arrangement of the cellulose microfibrils which reduces their spiral angle and leads to better resistance and better mechanical properties of the fibres [64]. However, for a strong concentration and/or a too long soaking time, there is a decrease of the tensile stress of treated OPF, because too many constituents (i.e. hemicellulose, lignin) are removed creating defects in the fibre.

From these results, it can be concluded that the tensile properties of oil palm fibres can be improved by treating them with a short alkali-treatment using sodium hydroxide. However, in this study, where the improvement of the compatibility between cement and fibres is sought, the short hydrolysis time treatment may not remove enough inhibitory constituents which delay the cement hydration. Thus, weak and strong treatments with a soaking time of 24 h are chosen for the rest of the work.

3.4. Effect of modifications on the chemical compatibility of the oil palm fibres and portland cement

Untreated and treated oil palm fibres are added to cement paste containing or not 0.25% of superplasticizer. This experiment shows the influence of treatments and/or the viscosity of cementitious matrix on the hydration of cement. Fig. 8 shows the heat flow during the exothermic reaction of OPC while Fig. 9 shows the total heat released during that reaction. The chemical compatibility is calculated by using the Pasca's equation (1) and the isothermal calorimetry measurements and the compatibility factor (CX′) results are shown in Table 5.

Firstly, the untreated oil palm fibres, with a compatibility factor of 9.4, appear to be incompatible with cement. Results confirm the results found by Ko et al. with the hydration peak that begins to appear nearly 1 day after the end of the chemical reaction of OPC [44]. Moreover, this peak reaches its maximum 4 days after the one of Portland cement. Thus, in addition to delaying the start of the hydration, untreated oil palm fibres also slow it down considerably. This phenomenon is explained from the literature due to the high amount of sugars present in oil palm fibres (Table 3) which prevent and delay the formation of hydrated cement products by forming a thin permeable layer around the cement grains [27,65–67].

Alkali-treatments greatly reduce the inhibitory effect of OPF, with a significant increase of the compatibility factor, with values over 85 for every single tested condition. For both treatments, the retardation is very low as compared to the reference and the heat released is just a little bit lower than the normal OPC reaction. It has to be noticed that the weak treatment is surprisingly good, with a CX' = 91.5 which means that the stronger treatment removes more constituents from the surface, but is not needed to solve the chemical compatibility issue of OPF with OPC.

Addition of 0.25% of superplasticizer to the cement paste causes the hydration peak to be delayed for less than half a day and the heat released is also delayed at the beginning of the reaction, but after 2 days, it nearly has the same value than the reference. As seen by the analysis of the rheological properties of the cementitious matrix, the addition of 0.25% of SP strongly decreases the viscosity of cement paste which becomes more fluid. Therefore, according to Zahia et al. [68], as the fluidity increases, it is normal that the hydration reaction is delayed and takes a longer time. When OPFs are added, the addition of a small amount of superplasticizer only decreases the compatibility factor by 4 for each treatment.

From these results, it can be concluded that NaOH alkali-treatments seem very effective in order to avoid the retarding effect of the unwanted constituents of OPF. Even though OPF has a high sugar content, a weak treatment is very effective, showing that high concentration is not needed in order to solubilize and remove the sugar from the fibre. Moreover, a more fluid matrix leads to a better diffusion of sugars in the cement paste and therefore, increase the retarding effect. Nonetheless, this chemical phenomenon is not obvious because the hydration peaks of cement with SP and with or without treated fibres are almost identical.

3.5. Effect of modifications on the mechanical compatibility of the oil palm fibres and portland cement

Besides the chemical compatibility, the quality of the interface

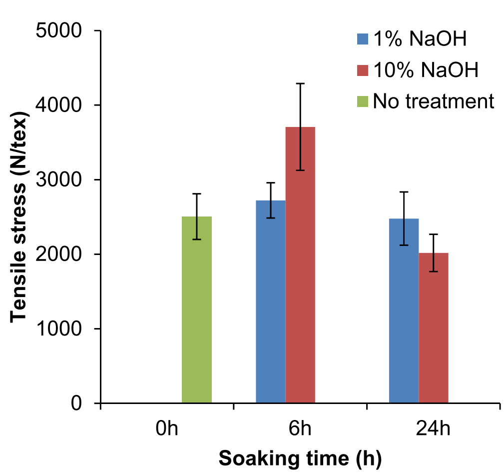


Fig. 7. Tensile stresses of oil palm fibres.

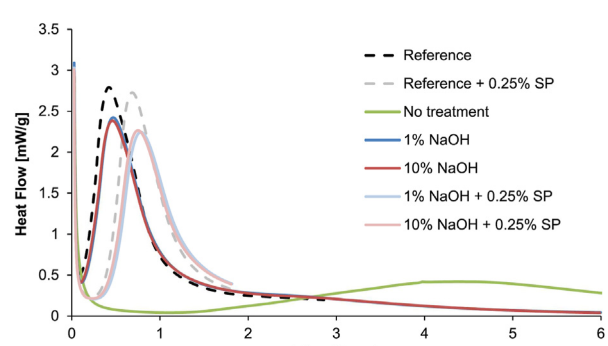


Fig. 8. Effect of different modifications on the hydration of Portland cement (Heat Flow).

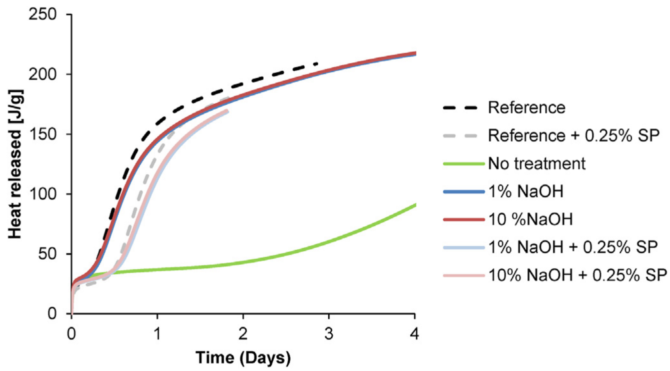


Fig. 9. Effect of different modifications on the hydration of Portland cement (Heat Released).

Table 5
Chemical compatibility of ordinary Portland cement I 52.5R with or without SP and with untreated or treated OPF.

Sample	No treatment	1% NaOH	10% NaOH	1% NaOH +0.25% SP	10% NaOH +0.25% SP
CX′	9.4	91.5	93.2	86.4	89.9

between NFs and cement is also a critical factor in fibre-reinforced materials. Therefore, pull-out tests are carried out in order to measure the strength of the interface. As shown in Fig. 4, three different regimes can occur during the final stage of a single pull-out test. It can be constant friction, slip-softening and slip-hardening. According to this theory, the sliding pull-out mechanisms and the maximum load of the different samples for each condition are summarized in Fig. 10.

With untreated fibres, the pull-out is mostly following slip-softening (more than 50% of the time) and constant friction mechanisms (40–50%). Therefore, the interface between untreated fibres and cementitious matrix can be characterized as bad, especially for samples tested at 7 days. For samples made with treated fibres, the three mechanisms are also observed but as compared to untreated fibre samples, less slip-softening behaviours rule the pull-out stage (28–33%). Hence, it can be concluded that the treatment of fibres and the addition of SP in the cementitious matrix improve the mechanical properties at the interface between the two components. The study of the debonding load confirms previous observations. Indeed, by treating fibres, the load significantly increases, and this effect is even more pronounced at 28 days (+250 and + 530%, respectively). Moreover, it appears that the addition of SP also allows improving this characteristic, with the highest load average recorded at 6.3 N, with more slip-hardening behaviour.

Ultimately, among all the conditions, samples with treated oil palm fibres and containing superplasticizer have better mechanical properties at the interface and therefore the better bonding stress. It can be explained by the fact that alkali-treatment removes from OPF the sugars which delay the hydration of cement close to the fibre, thereby creating a bad interface around the fibre in the matrix (Fig. 11-a). However, for treated OPF, Fig. 11-b shows that the interface is clearly better because the alkaline treatment removes the sugars at the OPF's surface, but also natural impurities which increases the diameter of the pores. Thus, this leads to a more porous and rougher surface morphology which improves the mechanical interlocking with the matrix [38,63,64].

Changing the viscosity of the matrix has a significant influence on the interfacial properties. Indeed, because the OPC is more fluid, there is a better diffusion of sugars in the paste and their inhibitory effect is not concentrated around the fibre, but it is distributed throughout the matrix, and consequently allowing a better interface. However, this effect is mainly observable for samples with treated OPF because, without treatment, the amount of sugar in OPF is too high which cause a major inhibitory effect on the whole matrix.

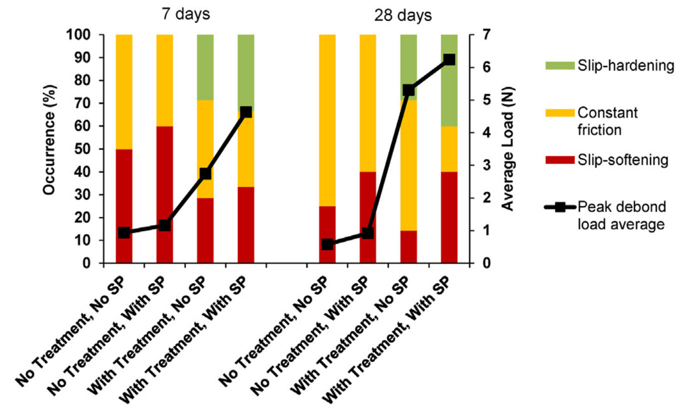


Fig. 10. Sliding pull-out mechanisms and peak debond load average of different modifications made on the composite.

4. Conclusions

This work aims to characterize the chemical and mechanical compatibility between oil palm fibres and OPC. Two different methods are studied, namely alkali-treatments (i.e. surface treatment of the fibres) and addition of SP (i.e. increase of the matrix fluidity). Based on the results, the following conclusions can be drawn:

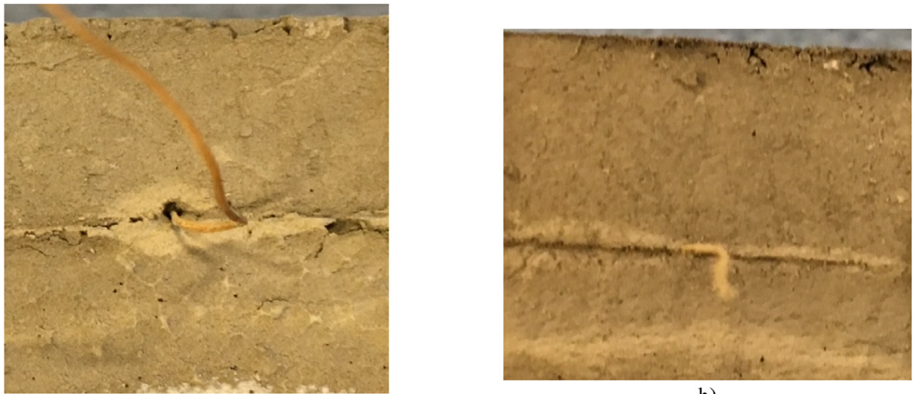
- Alkali-treatment removes from the OPF some constituents known to delay the hydration of cement (e.g. hemicellulose and waxes). Moreover, natural impurities and silica bodies are being removed from the fibre's surface, leading to bigger pore size and rougher surface.
- Mechanical properties of the OPF are significantly affected by the alkaline hydrolysis but these treatments have a very good effect on the chemical compatibility of untreated OPF. However, it appears that a strong treatment does not have a significant effect as compared to a weaker treatment, and thus should be avoided.
- Pull-out tests show that treated oil palm fibres have a better interface and a better mechanical interlocking with OPC than the untreated fibres. This improvement is mainly due to the removal of sugars and the rougher and more porous fibre's surface.
- A more fluid matrix does not have a major impact on the cement hydration. However, treated OPF has a much better interface when mixed with a less viscous matrix, due to the diffusion of sugars in the paste being facilitated.

In overall, the modifications made to the matrix and the fibres lead to much better compatibility between OPF and OPC, both mechanically and chemically. Moreover, even though the effectiveness of the alkali-treatment is much more obvious, a more fluid matrix can further improve the interface, proving the importance of removing the hydrolysis by-products from the surface of the fibres.

Author statement

All persons who meet authorship criteria are listed as authors, and all authors certify that they have participated sufficiently in the work to take public responsibility for the content, including participation in the concept, design, analysis, writing, or revision of the manuscript.

Furthermore, each author certifies that this material or similar material has not been and will not be submitted to or published in any other publication before its appearance in Results in Engineering.



a) b)

Fig. 11. a) Interface between untreated fibre and matrix without SP; b) Interface between treated fibre and matrix with SP.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgement

The authors would like to acknowledge the financial support provided by NWO (Nederlandse Organisatie voor Wetenschappelijk Onderzoek), under the project 10013077 "Development of sustainable and functionalizes inorganic binder-biofibres composites" and the Erasmus+ "Traineeship mobilities between programme countries" program. For the material support, the authors would like to acknowledge Dr. J. E.G. van Dam (Wageningen Food & Biobased Research, the Netherlands) and ENCI (the Netherlands).

References

[1] M.J. González, J. García Navarro, Assessment of the decrease of CO2 emissions in the construction field through the selection of materials: practical case study of three houses of low environmental impact, Build. Environ. 41 (2006) 902–909, https://doi.org/10.1016/j.buildenv.2005.04.006.

[2] H.J. Wu, Z.W. Yuan, L. Zhang, J. Bi, Life Cycle Energy Consumption and CO 2 Emission of an Office Building in China, (n.d.). https://doi.org/10.1007/s11367-011-0342-2.

[3] M. Stajanca, A. Estokova, Environmental Impacts of Cement Production, Tech. Univ. Kosice, Civ. Eng. Fac. Inst. Archit. Eng. (2012) 296–302. http://ena.lp.edu.ua. (Accessed 24 October 2018). Environmental Impacts of Cement Production.

[4] S.H. Ghaffar, M. Burman, N. Braimah, Pathways to circular construction: an integrated management of construction and demolition waste for resource recovery, J. Clean. Prod. (2020), https://doi.org/10.1016/j.jclepro.2019.118710.

[5] A. Lenoir Etienne Wurtz, F. Garde, Zero Energy Buildings in France: Overview and Feedback, n.d.

[6] S. Lou, E.K.W. Tsang, D.H.W. Li, E.W.M. Lee, J.C. Lam, Towards zero energy school building designs in Hong Kong, Energy Procedia 105 (2017) 182–187, https://doi.org/10.1016/j.egypro.2017.03.299.

[7] Global Alliance for Buildings and Construction, No Title, (n.d.).

[8] L.H. Maccarini Vefago, J. Avellaneda, Recycling concepts and the index of recyclability for building materials, Resour. Conserv. Recycl. 72 (2013) 127–135, https://doi.org/10.1016/j.resconrec.2012.12.015.

[9] K.I.M. Ibrahim, Mechanical properties of glass fiber reinforced concrete (GFRC), IOSR J. Mech. Civ. Eng. 13 (2016) 47–50, https://doi.org/10.9790/1684-1304054750.

[10] H.A.T., E-Korchi, Behaviour of carbon fiber reinforced cement composites in direct tension, Cement Concr. Res. 23 (1993) 618–626, https://doi.org/10.1016/0008-8846(93)90013-Y.

[11] H. Danso, Properties of coconut, oil palm and bagasse fibres: as potential building materials, Procedia Eng. 200 (2017) 1–9, https://doi.org/10.1016/j.proeng.2017.07.002.

[12] D.B. Dittenber, H.V.S. GangaRao, Critical review of recent publications on use of natural composites in infrastructure, Compos. Part A Appl. Sci. Manuf. 43 (2012) 1419–1429, https://doi.org/10.1016/j.compositesa.2011.11.019.

[13] N. Benmansour, B. Agoudjil, A. Gherabli, A. Kareche, A. Boudenne, Thermal and mechanical performance of natural mortar reinforced with date palm fibers for use as insulating materials in building, Energy Build. 81 (2014) 98–104, https://doi.org/10.1016/j.enbuild.2014.05.032.

[14] W. Ma, Y. Qin, Y. Li, J. Chai, X. Zhang, Y. Ma, H. Liu, Mechanical properties and engineering application of cellulose fiber-reinforced concrete, Mater. Today Commun. 22 (2020) 100818, https://doi.org/10.1016/j.mtcomm.2019.100818.

[15] C. Niyigena, S. Amziane, A. Chateauneuf, L. Arnaud, L. Bessette, F. Collet, C. Lanos, G. Escadeillas, M. Lawrence, C. Magniont, S. Marceau, S. Pavia, U. Peter, V. Picandet, M. Sonebi, P. Walker, Variability of the mechanical properties of hemp concrete, Mater. Today Commun. 7 (2016) 122–133, https://doi.org/10.1016/j.mtcomm.2016.03.003.

[16] S.O. Odeyemi, R. Abdulwahab, A.G. Adeniyi, O.D. Atoyebi, Physical and mechanical properties of cement-bonded particle board produced from African balsam tree (Populous Balsamifera) and periwinkle shell residues, Results Eng. (2020), https://doi.org/10.1016/j.rineng.2020.100126.

[17] S.H. Ghaffar, M. Al-Kheetan, P. Ewens, T. Wang, J. Zhuang, Investigation of the interfacial bonding between flax/wool twine and various cementitious matrices in mortar composites, Construct. Build. Mater. 239 (2020) 117833, https://doi.org/10.1016/j.conbuildmat.2019.117833.

[18] A.R. Azevedo, M.T. Marvila, E.B. Zanelato, J. Alexandre, G.C. Xavier, D. Cecchin, Development of mortar for laying and coating with pineapple fiber, Rev. Bras. Eng. Agrícola Ambient. (2020), https://doi.org/10.1590/1807-1929/agriambi.v24n3p187-193.

[19] M.T. Marvila, A.R.G. Azevedo, J. Alexandre, E.B. Zanelato, S.N. Monteiro, D. Cecchin, L.F. Amaral, Mortars with pineapple fibers for use in structural reinforcement, Miner. Met. Mater. Ser. (2019), https://doi.org/10.1007/978-3-030-05749-7_72.

[20] K.H. Mo, C.S. Bong, U.J. Alengaram, M.Z. Jumaat, S.P. Yap, Thermal conductivity, compressive and residual strength evaluation of polymer fibre-reinforced high volume palm oil fuel ash blended mortar, Construct. Build. Mater. (2017), https://doi.org/10.1016/j.conbuildmat.2016.11.005.

[21] S. Kalia, B.S. Kaith, I. Kaur, Pretreatments of natural fibers and their application as reinforcing material in polymer composites-a review, Polym. Eng. Sci. (2009), https://doi.org/10.1002/pen.21328.

[22] S. Thomas, S.A. Paul, L.A. Pothan, B. Deepa, Natural Fibres: Structure, Properties and Applications, (n.d.). https://doi.org/10.1007/978-3-642-17370-7_1.

[23] A. Célino, S. Fréour, F. Jacquemin, P. Casari, The hygroscopic behavior of plant fibers: a review, Front. Chem. 1 (2014) 1–12, https://doi.org/10.3389/fchem.2013.00043.

[24] B.D. Prasad, N.S. Kumar, Chemical composition of natural fibers and its influence on their mechanical properties, Mech. Compos. Mater. 50 (2014) 359–376, https://doi.org/10.1007/s11029-014-9422-2.

[25] D. Jones, G.O. Ormondroyd, S.F. Curling, C.-M. Popescu, M.-C. Popescu, 2 - chemical compositions of natural fibres. https://doi.org/10.1016/B978-0-08-100411-1.00002-9, 2017.

[26] B. Na, Z.Z. Wang, H.H. Wang, X. Lu, Wood-cement compatibility review, Wood Res. 59 (2014) 813–826, https://doi.org/10.1023/B:BREA.0000004377.12288.3c.

[27] K. Kochova, K. Schollbach, F. Gauvin, H.J.H. Brouwers, Effect of saccharides on the hydration of ordinary Portland cement, Construct. Build. Mater. 150 (2017) 268–275, https://doi.org/10.1016/j.conbuildmat.2017.05.149.

[28] G.C.H. Doudart De La Grée, Q.L. Yu, H.J.H. Brouwers, The effect of glucose on the hydration kinetics of ordinary portland cement, in: First Int. Conf. Bio-Based Build. Mater, Clermont-Ferrand, 2015, pp. 126–132.

[29] N. Venkatachalam, P. Navaneethakrishnan, R. Rajsekar, S. Shankar, Effect of pretreatment methods on properties of natural fiber composites: a review, Polym. Polym. Compos. 24 (2016) 555–566.

[30] N. Aderinsola Sadiku, A. Sanusi, Wood pre-treatment influence on the hydration of portland cement in combination with some tropical wood species, Pro Ligno 10 (2014) 3–10.

[31] A.O. Olorunnisola, Effects of pre-treatment of rattan (Laccosperma secundiflorum) on the hydration of Portland cement and the development of a new compatibility index, Cement Concr. Compos. 30 (2008) 37–43, https://doi.org/10.1016/j.cemconcomp.2007.08.002.

[32] J.N. Putro, F.E. Soetaredjo, S.-Y.Y. Lin, Y.-H.H. Ju, S. Ismadji, Pretreatment and Conversion of Lignocellulose Biomass into Valuable Chemicals, Royal Society of Chemistry, 2016, https://doi.org/10.1039/c6ra09851g.

[33] X. Li, A.E. Lope, G.T. Ae, S. Panigrahi, Chemical Treatments of Natural Fiber for Use in Natural Fiber-Reinforced Composites: A Review, (n.d.). https://doi.org/10.1007/s10924-006-0042-3.

[34] J.M. Ferraz, C.H.S. del Menezzi, D.E. Teixeira, S.a. Martins, Effects of treatment of coir fiber and cement/fiber ratio on properties of cement-bonded composites, BioResources 6 (2011) 3481–3492.

[35] N. Sgriccia, M.C. Hawley, M. Misra, Characterization of Natural Fiber Surfaces and Natural Fiber Composites, (n.d.). https://doi.org/10.1016/j.compositesa.2008.07.007.

[36] C.S. Hassan, S.M. Sapuan, A.A. Nuraini, M.Z.M. Yuoff, Effect of chemical treatment on the tensile properties of single oil palm empty fruit bunch (OPEFB) fibre, Trends Text. Eng. Fash. Technol. 3 (2018), https://doi.org/10.31031/tteft.2018.03.000560.

[37] J. Rout, S.S. Tripathy, S.K. Nayak, M. Misra, A.K. Mohanty, Scanning electron microscopy study of chemically modified coir fibers, J. Appl. Polym. Sci. 79 (2001) 1169–1177, https://doi.org/10.1002/1097-4628(20010214)79:7<1169::AID-APP30>3.0.CO;2-Q.

[38] Z. Ibrahim, A.A. Aziz, R. Ramli, K. Jusoff, M. Ahmad, M.A. Jamaludin, Effect of treatment on the oil content and surface morphology of oil palm (Elaeis guineensis) empty fruit bunches (EFB) fibres, Wood Res. 60 (2015) 157–166.

[39] M.C. Symington, O.S. David-West, W.M. Banks, J.L. Thomason, R.A. Pethrick, The effect of alkalisation on the mechanical properties of natural fibres, J. Chem. Inf. Model. 53 (1989) 160, https://doi.org/10.1017/CBO9781107415324.004.

[40] M.K. Faizi, A.B. Shahriman, M.S.A. Majid, B.M.T. Shamsul, I. Zunaidi, Y.G. Ng, Z.M. Razlan, W.K. Wan, The effect of alkaline treatments with various concentrations on oil palm empty fruit bunch (OPEFB) fibre structure, IOP Conf. Ser. Mater. Sci. Eng. 429 (2018) 6–11, https://doi.org/10.1088/1757-899X/429/1/012006.

[41] D.P. Bentz, M.A. Peltz, K.A. Snyder, J.M. Davis, VERDiCT: viscosity enhancers reducing diffusion in concrete technology, Conc. Int. 31 (2009) 31–36.

[42] M. Baskaran, R. Hashim, O. Sulaiman, S. Hiziroglu, M. Sato, T. Sugimoto, Optimization of press temperature and time for binderless particleboard manufactured from oil palm trunk biomass at different thickness levels, Mater. Today Commun. (2015), https://doi.org/10.1016/j.mtcomm.2015.04.005.

[43] S. Shinoj, R. Visvanathan, S. Panigrahi, M. Kochubabu, Oil palm fiber (OPF) and its composites: a review, Ind. Crop. Prod. (2011), https://doi.org/10.1016/j.indcrop.2010.09.009.

[44] K. Kochova, F. Gauvin, K. Schollbach, H.J.H. Brouwers, Using alternative waste coir fibres as a reinforcement in cement-fibre composites, Construct. Build. Mater. 231 (2020) 117121, https://doi.org/10.1016/j.conbuildmat.2019.117121.

[45] S. Shinoj, R. Visvanathan, S. Panigrahi, M. Kochubabu, Oil palm fiber (OPF) and its composites: a review, Ind. Crop. Prod. 33 (2010) 7–22, https://doi.org/10.1016/j.indcrop.2010.09.009.

[46] R. Mahjoub, J. Bin Mohamad Yatim, A.R. Mohd Sam, A review of structural performance of oil palm empty fruit bunch fiber in polymer composites, Ann. Mater. Sci. Eng. (2013), https://doi.org/10.1155/2013/415359.

[47] A.E.O. Ben Sghaier, Y. Chaabouni, S. Msahli, F. Sakli, Morphological and crystalline characterization of NaOH and NaOCl treated Agave americana L. fiber, Ind. Crop. Prod. (2012), https://doi.org/10.1016/j.indcrop.2011.09.012.

[48] F. Xu, J. Yu, T. Tesso, F. Dowell, D. Wang, Qualitative and quantitative analysis of lignocellulosic biomass using infrared techniques: a mini-review, Appl. Energy 104 (2013) 801–809, https://doi.org/10.1016/j.apenergy.2012.12.019.

[49] F. Suomen Standardisoimisliito, Methods of testing cement, Chem. Anal. Cem. 3 (1995).

[50] S.A. Pasca, I.D. Hartley, M.E. Reid, R.W. Thring, Evaluation of compatibility between beetle-killed lodgepole pine (pinus contorta var. Latifolia) wood with portland cement, Materials (Basel) 3 (2010) 5311–5319, https://doi.org/10.3390/ma3125311.

[51] R. Jd, Effect of single fiber pull out test result on flexural performance of ECC, J. Civ. Environ. Eng. 4 (2014) 140, https://doi.org/10.4172/2165-784X.1000140.

[52] D.L. Naik, A. Sharma, R.R. Chada, R. Kiran, T. Sirotiak, Modified pullout test for indirect characterization of natural fiber and cementitious matrix interface properties, Construct. Build. Mater. 208 (2019) 381–393, https://doi.org/10.1016/j.conbuildmat.2019.03.021.

[53] C. Wang, L.F. Friedrich, Continuous Modeling Technique of Fiber Pullout from a Cement Matrix with Different Interface Mechanical Properties Using Finite Continuous Modeling Technique of Fiber Pullout from a Cement Matrix with Different Interface Mechanical Properties Using Finite, 2016, https://doi.org/10.1590/1679-78252575.

[54] C. Sawsen, K. Fouzia, B. Mohamed, G. Moussa, Effect of flax fibers treatments on the rheological and the mechanical behavior of a cement composite, Construct. Build. Mater. 79 (2015) 229–235, https://doi.org/10.1016/j.conbuildmat.2014.12.091.

[55] S. Mishra, A.K. Mohanty, L.T. Drzal, M. Misra, G. Hinrichsen, A review on pineapple leaf fibers, sisal fibers and their biocomposites, Macromol. Mater. Eng. (2004), https://doi.org/10.1002/mame.200400132.

[56] L. Risanto, E. Hermiati, Y. Sudiyani, Properties of lignin from oil palm empty fruit bunch and its application for plywood adhesive, Makara J. Technol. (2014), https://doi.org/10.7454/mst.v18i2.2944.

[57] M.S. Nazir, B.A. Wahjoedi, A.W. Yussof, M.A. Abdullah, Eco-friendly extraction and characterization of cellulose from oil palm empty fruit bunches, BioResources (2013), https://doi.org/10.15376/biores.8.2.2161-2172.

[58] A. Oushabi, S. Sair, F. Oudrhiri Hassani, Y. Abboud, O. Tanane, A. El Bouari, The effect of alkali treatment on mechanical, morphological and thermal properties of date palm fibers (DPFs): study of the interface of DPF–Polyurethane composite, S. Afr. J. Chem. Eng. 23 (2017) 116–123, https://doi.org/10.1016/j.sajce.2017.04.005.

[59] K. Law, W.R.W. Daud, A. Ghazali, Morphological and chemical nature of fiber, Bioresource 2 (2007) 351–362.

[60] M.S. Sreekala, M.G. Kumaran, S. Thomas, Oil palm fibers: morphology, chemical composition, surface modification, and mechanical properties, J. Appl. Polym. Sci. 66 (2002) 821–835, https://doi.org/10.1002/(sici)1097-4628(19971031)66:5<821::aid-app2>3.3.co;2-l.

[61] H. Ma, G. Wu, Z. Liu, Z. Li, H. Zhou, Effect of alkali treatment on the tensile properties of cashmere fibers, Wool Text. J. 45 (2017) 48–51, https://doi.org/10.19333/j.mfkj.2016090120904.

[62] T.H. Nam, S. Ogihara, N. Huy Tung, S. Kobayashi, Effect of Alkali Treatment on Interfacial and Mechanical Properties of Coir Fiber Reinforced Poly(butylene Succinate) Biodegradable Composites, 2011 https://doi.org/10.1016/j.compositesb.2011.04.001.

[63] M. Mizanur Rahman, M. Ahmad Khan, M. Mizanur Rahman, M.A. Khan, Surface Treatment of Coir (Cocos Nucifera) Fibers and its Influence on the Fibers' Physico-Mechanical Properties, 2007, https://doi.org/10.1016/j.compscitech.2007.01.009.

[64] M.A.N. Izani, M.T. Paridah, U.M.K. Anwar, M.Y. Mohd Nor, P.S. H'ng, Effects of fiber treatment on morphology, tensile and thermogravimetric analysis of oil palm empty fruit bunches fibers, Compos. Part B. 45 (2013) 1251–1257, https://doi.org/10.1016/j.compositesb.2012.07.027.

[65] M. Bishop, A.R. Barron, Cement hydration inhibition with sucrose, tartaric acid, and lignosulfonate: analytical and spectroscopic study, Ind. Eng. Chem. Res. 45 (2006) 7042–7049, https://doi.org/10.1021/ie060806t.

[66] J.F. Young, A review of the mechanisms of set-retardation in portland cement pastes containing organic admixtures, Cement Concr. Res. 2 (1972) 415–433, https://doi.org/10.1016/0008-8846(72)90057-9.

[67] N.L. Thomas, J.D. Birchall, The retarding action of sugars on cement hydration, Cement Concr. Res. 13 (1983) 830–842, https://doi.org/10.1016/0008-8846(83)90084-4.

[68] D. Zahia, E. Karim, M. Fatiha, S. Fatima, Cinetique d'hydratation d'un ciment en presence des adjuvants, n.d. https://pdfs.semanticscholar.org/e5e7/0ff0b6c2e156bcf9639de27ed35a9c3abf5c.pdf.