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Moroccan heated clay-based geopolymer reinforced with date palm cellulose: microstructure characterization and mechanical/physical properties

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Abstract

The objective of this research was to study the cellulose addition effect on the geopolymerization of heated clay. The clay, composed of illite, plagioclase and kaolinite, was heated at 700°C for 2 h and mixed with cellulose (up to 10 mass%). The mixtures were NaOH-activated, and shaped samples were aged at 83°C for 30 days. The cured samples were investigated by using X-ray diffraction, Fourier-transform infrared spectroscopy and scanning electron microscopy. The influence of the cellulose addition on the mechanical/physical properties was also evaluated. The results showed that zeolite ZK-14, hydrosodalite, sodium carbonate and a geopolymer composed of poly(sialate) units were formed in all cured samples. The relative amounts of zeolite and metakaolin evolved antagonistically, whereas that of illite slightly decreased with increasing cellulose content. Metakaolin and illite were involved in the geopolymerization process. Cellulose addition led to the improvement of the flexural strength of the samples and to porosity reduction. By contrast, water absorption was increased. The positive effect of cellulose on sample performance is explained on the basis of hydrogen bonding between the functional moieties of cellulose and the active sites of sample constituents, namely zeolites, metakaolin, illite and the geopolymer. As a filler, zeolite probably contributed to sample strengthening, and the detrimental impact of Na-carbonate was insignificant. Based on the results obtained, the composites could be used as binders for brick manufacturing or as lightweight mortars.

Keywords: Cellulose; geopolymer; heated clay; microstructure; sample performances

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Recently, major concern has been expressed regarding ordinary Portland cement (OPC) manufacturing because of the huge amounts of carbon dioxide it releases (Zhang *et al.*, 2023), the energy required and the scarcity of the geomaterials needed. Therefore, research has been undertaken worldwide to find eco-friendly cementitious materials with good physical and mechanical properties (Mohamad *et al.*, 2022; da Silva Rego *et al.*, 2023; Shi *et al.*, 2023). Geopolymers, which are low-temperature synthesized aluminosilicates, have been considered as potential alternatives to OPC (Ranjbar *et al.*, 2020; Giacobello *et al.*, 2022; Tanu *et al.*, 2022; Ayub & Khan, 2023; Jwaida *et al.*, 2023). Fly ash, calcined clays, volcanic ash and blast furnace slag are amongst the used aluminosilicate sources for geopolymer synthesis. For the latter purpose, these materials are dissolved using moderate to strong alkaline solutions (4–12 M). NaOH is by far the most used alkali-hydroxide activator. Mixtures of NaOH and Na₂SiO₃ are also used. In the latter condition, OH⁻ ions (NaOH-derivative species) are involved in the dissolution of the aluminosilicate, whilst sodium silica plays the role of a binder (Mijarsh *et al.*, 2015). Na⁺ ions contribute to the charge balance as well as to zeolite formation. The excess of Na⁺ ions

induces the formation of carbonate by reaction with atmospheric carbon dioxide. Such a phenomenon occurs particularly in the case of the NaOH-activated metakaolin system (Valentini, 2018).

Considering metakaolin, for instance, which is considered to be a high-grade source material for geopolymer synthesis, alkali activation resulted in the formation of Al(OH)₄⁻, H₃SiO₄⁻ and H₂SiO₄²⁻ species (Valentini, 2018; Weng & Sagoe-Crentsil, 2007) together with a sodium aluminosilicate hydrate (Na₂O–Al₂O₃–SiO₂–H₂O gel; Valentini, 2018). According to Weng & Sagoe-Crentsil (2007), the association of Al(OH)₄⁻ and H₂SiO₄²⁻ results in the formation of small oligomers (dimers and trimers). However, the condensation between Al(OH)₄⁻ and H₃SiO₄⁻ leads to the formation of polymers and larger oligomers. The latter products consist of linked silica ([SiO₄]⁴⁻) and alumina ([AlO₄]⁴⁻) tetrahedra. The three-dimensional structure obtained is called a geopolymer (Van Deventer *et al.*, 2007; Sotelo-Piña *et al.*, 2019; Almutairi *et al.*, 2021).

Cellulose is one of the main constituents of plants. It exists as fibres and has a polymeric structure. Cellulose chains are composed of β-D-glucose units, and their organization into sheets is achieved due to its methylol (–CH₂OH) and the hydroxyl moieties (Bashline *et al.*, 2014; Etale *et al.*, 2023). Cellulose fibres have high strength/density ratios (Chen *et al.*, 2021; Emenike *et al.*, 2023). Therefore, they have been proven to be suitable for reinforcing some matrix materials (Ferreira *et al.*, 2021; Mourak *et al.*, 2021; Lazorenko *et al.*, 2022; Zheng *et al.*, 2023). Cellulose's reinforcing capability is mainly influenced by its

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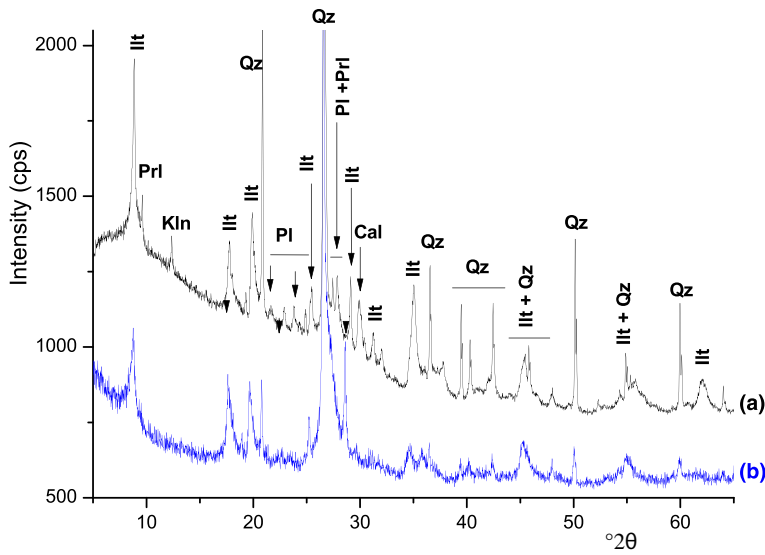


Figure 1. X-ray diffraction traces of the (a) raw and (b) heated clays. Cal = calcite (PDF#83-1762); cps = counts per second; Ill = illite (PDF# 43-0685); Kln = kaolinite (PDF# 83-0971); Pl = plagioclase (PDF# 41-1486); Pri = pyrophyllite (PDF# 96-0277); Qz = quartz (PDF# 05-0490).

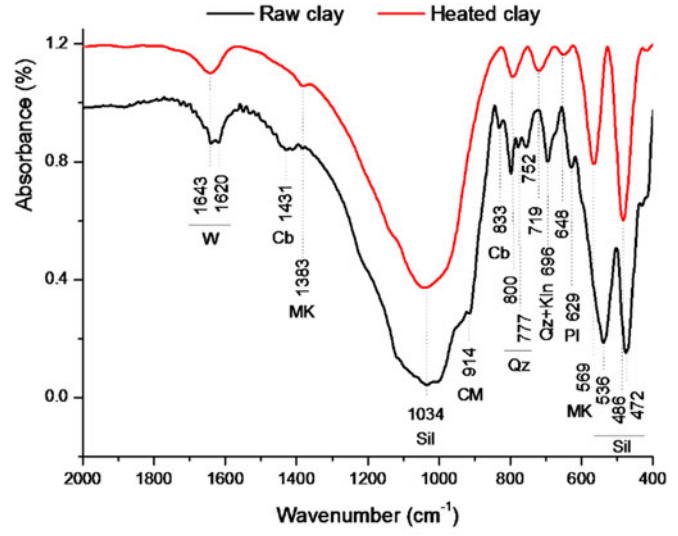
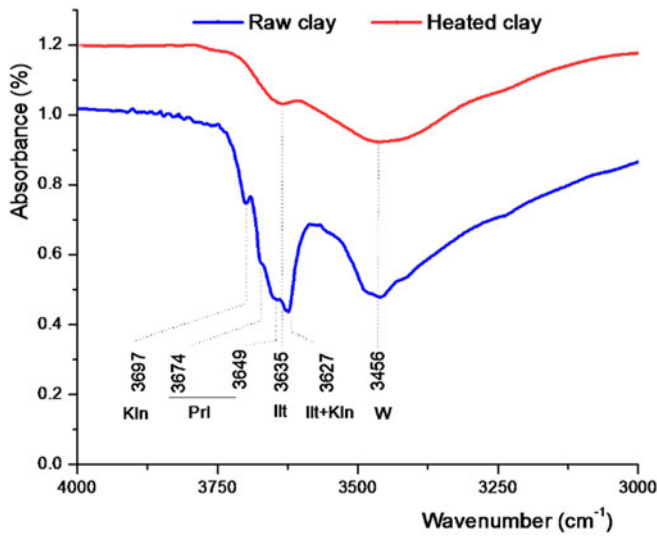


Figure 2. Fourier-transform infrared spectra of the raw and heated clays. cb = carbonates; CM = clay minerals; Ill = illite; Kln = kaolinite; MK = metakaolin; Pl = plagioclase; Pri = pyrophyllite; Qz = quartz; Sil = silicates; W = water.

fibre dimensions, surface properties and moisture retention (Farhan *et al.*, 2021; Santana *et al.*, 2021). According to some studies, the addition of cellulose to cementing materials should be limited (0.2–2.0% v/v; Jamshaid *et al.*, 2022; Raghunath *et al.*, 2023; de Souza *et al.*, 2023; dos Reis *et al.*, 2023). The addition of large amounts of cellulose resulted in increased water adsorption, and consequently the mechanical strength of the cellulose-containing cementing materials decreased (Singh & Gupta, 2020; Jaberizadeh *et al.*, 2023).

The reinforcing effect of the cellulose fibres can be improved by alkali activation using NaOH solutions (18–32 mass%) at 25–40°C. Due to this chemical treatment, fibrils with large surface areas and high aspect ratios form, and consequently adhesion between the reinforcing material and the matrix is enhanced (Aravindh *et al.*, 2022; Asyraf *et al.*, 2022).

Considering the above literature data, this work of research, which aimed to investigate the microstructure and determine the mechanical/physical properties of heated clay-based geopolymer samples, has fundamental and application interests.

Regarding the latter interest, this work is a contribution to the development of eco-friendly clay-based building materials demonstrating acceptable performance. The raw materials (clay and palm date rachis) used are found globally, and the product preparation process does not require sophisticated plants or as much energy as for OPC manufacturing (Semugaza *et al.*, 2023). Fundamentally, this study is a contribution to the understanding of the chemical interactions that could occur between cellulose’s functional moieties and the active sites of geopolymers and associated aluminosilicates (zeolite, illite and metakaolin).

Materials and methods

Clay material and cellulose

The raw clay material was from Kharrou Mountain (Rehamna, Morocco). It was essentially composed of mica/illite, pyrophyllite, kaolinite, plagioclase and calcite (Figs 1 & 2). As an aluminosilicate-rich material (Table 1), the clay seemed to be

Table 1. Chemical composition (mass%) of the used clay, the relative amounts of the date palm rachis constituents and the physical characteristics of the extracted cellulose.

Clay									
SiO ₂	Al ₂ O ₃	TiO ₂	Fe ₂ O ₃	MnO	MgO	K ₂ O	CaO	Na ₂ O	
56.75	33.74	1.80	1.24	0.01	0.58	2.50	0.27	1.76	
Date palm rachis constituents									
Lignin	Hemicellulose	Cellulose	Balance						
20	30.5	47.5	2						
Physical characteristics of the extracted cellulose									
Crystallinity index (%)	Average molecular weight (g mol ⁻¹)	Polymerization degree							
67	144 622	892							

suitable as a precursor for geopolymer preparation. Therefore, the clay was the object of mechanical and heating treatments, as detailed below. As a result of heating, the kaolinite disappeared and metakaolin was formed (Figs 1 & 2), whereas the illite resisted heating as its structure breaks down at >900°C (Gualtieri & Ferrari, 2006). As can be deduced from the thermograms given in Fig. 3, kaolinite underwent dehydroxylation at ~523°C. In this respect, it should be noted that the differential thermal analysis (DTA) curve showed endothermic phenomena at ~88°C and 272°C, which were assigned to the losses of hygroscopic and strongly retained waters. The weak endothermic peak at 812°C was associated with calcite decomposition.

The cellulose used as reinforcement was extracted from date palm (*Phoenix dactylifera*) rachis following the steps shown in Fig. 4. More details regarding the cellulose extraction are given below. The physical characteristics of the isolated cellulose, which manifested as shown in Fig. 5, are provided in Table 1.

Sample preparation

For geopolymer synthesis, dried portions of the clay heated at 700°C for 2 h and cellulose (up to 10 mass%) were introduced

in a glass container and mixed for 15 min. The blend was transferred to a plastic bag and mixed with NaOH solution (6 M). The optimal (NaOH solution volume)/(blend weight) ratio was of 12.5 mL 100 g⁻¹. The paste was kneaded for 15 min (to obtain a plastic paste), and the shaping operation was manually accomplished in ~20 min. The shaped samples (3.8 × 9.2 × 40 mm) were placed in a Memmert oven and aged at 83°C for 30 days. The oven used was functioning in the open air. Regarding the adopted curing conditions, it can be noted that, according to Mo *et al.* (2014), increasing the curing temperature (up to 100°C) accelerates the dissolution, polymerization and reprecipitation processes of the geopolymerization reaction. Through this, the physical/mechanical properties of the product are enhanced. The effect of curing time on geopolymerization is still under debate (Heah *et al.*, 2011; Van Jaarsveld *et al.*, 2022). The NaOH activation solution was prepared by dissolution of NaOH pellets (Sigma-Adrich product, purity: >97.0%) in distilled water.

Experimental procedures and instruments

The raw clay was crushed and ground using an agate mortar then passed through a 100 µm sieve. The sieved fraction was heated in a Nabertherm furnace. The heating conditions were as follows: heating rate: 7°C min⁻¹; maximum temperature: 700°C; soaking time: 2 h; atmosphere: air.

For cellulose extraction, the pigments and lipids associated with cellulose were removed using an azeotropic mixture of ethanol and toluene. The solvent extraction was performed using a conventional Soxhlet extractor. The hemicelluloses and lignin were eliminated using a solution of sodium hydroxide (2 mass%) and *via* heating at 80°C for 2 h. Fibre bleaching was achieved with a solution of sodium chlorite (1.7% w/v) at 70°C (Mourak *et al.*, 2021).

The polymerization degree (PD) of the extracted cellulose was calculated using Equation 1:

$$PD = (AMW)/M_C \quad (1)$$

where AMW is the average molecular weight (144 622 g mol⁻¹) and M_C is the molecular weight of cellulose (162.14 g mol⁻¹). AMW

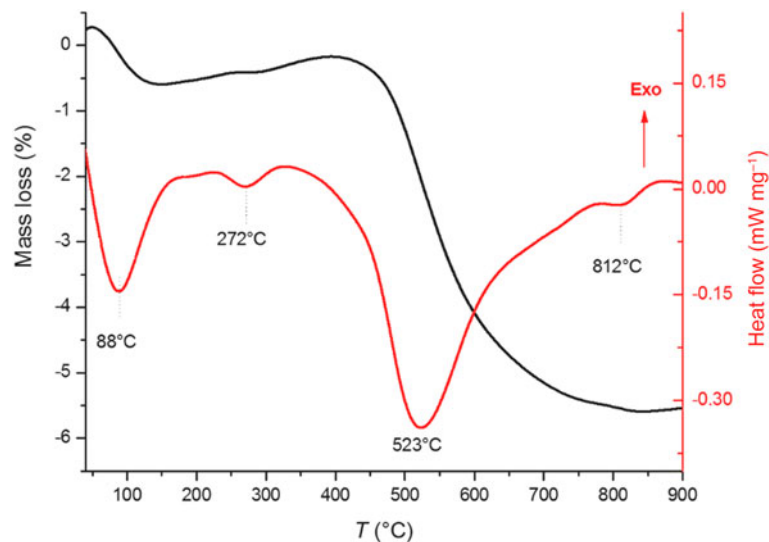


Figure 3. Thermal curves (TG, DTA) of the raw clay.

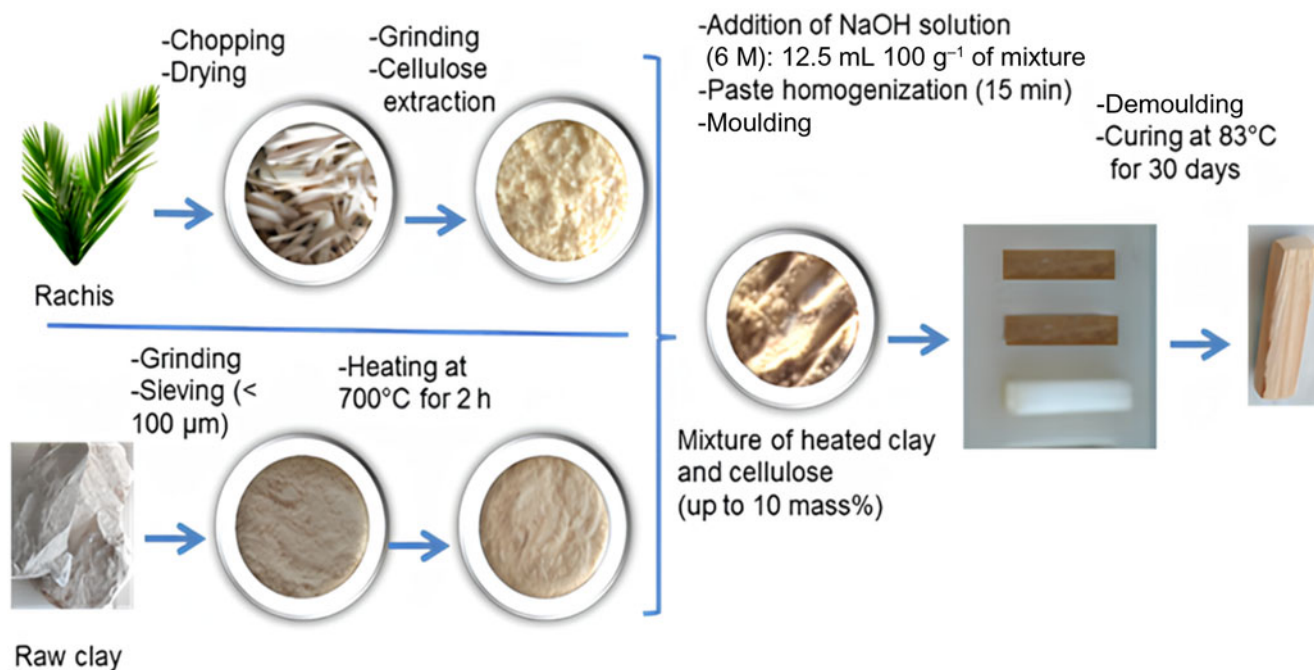


Figure 4. Main steps followed for date palm rachis and raw clay treatments and experimental operations for heated clay–cellulose sample preparation.

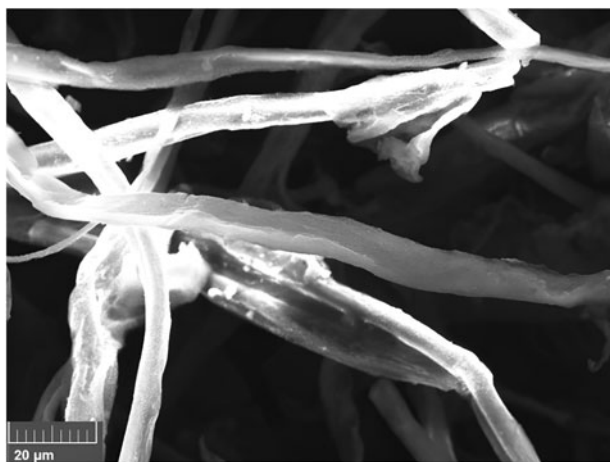


Figure 5. Scanning electron microscopy images showing the cellulose fibre morphology.

was measured following a procedure detailed elsewhere (Evans *et al.*, 1989; Malešič *et al.*, 2021).

As compared with the literature data (Hallac & Ragauskas, 2011), the PD obtained was of low to moderate value. It is believed that cellulose extracted with such a PD value can be used without significant agglomeration tendency.

The crystallinity index (CI) was determined by using data obtained from the X-ray diffraction (XRD) trace of cellulose and Equation 2 (Segal, 1957):

$$CI = \frac{I_{200} - I_{am}}{I_{200}} \quad (2)$$

where I_{200} is the maximum intensity of the (200) lattice diffraction and I_{am} is the intensity diffraction at $18^\circ 2\theta$.

The XRD analysis was performed on powder samples using a Rigaku Smart Lab diffractometer operating with a copper anode ($\lambda_{K\alpha} = 1.5418 \text{ \AA}$). The XRD traces were recorded under the following conditions: generator voltage: 40 kV; tube current: 30 mA; scan step size: 0.05° ; time step: 2s.

For the Fourier-transform infrared (FTIR) spectroscopy investigation, thin discs (study sample: 1 mg; KBr: 99 mg) were shaped and analysed using a Perkin Elmer 1725-x spectrophotometer functioning in the range of $4000\text{--}400 \text{ cm}^{-1}$. *PeakFit* v4.12 software was used for infrared band deconvolution. The best curve fit was evaluated on the basis of the following statistical parameters: correlation coefficient (r^2), standard error (SE) and the Fisher ratio (F-statistic). The relative quantities of metakaolin, zeolite and illite were determined on the basis the areas of their FTIR bands. The areas were evaluated using *PeakFit* software.

For thermal analysis, a SetaramSetsys 24 apparatus was used. The operating conditions were as follows: atmosphere: air; heating rate: $10^\circ\text{C min}^{-1}$; reference material and crucible: alumina; sample mass: 78 mg.

The microstructure of carbon-coated cured samples was investigated using a TESCAN VEGA3 electron microscope, which was equipped with 20 mm^2 X-Max diffusion silicon detector.

The flexural strength (σ_{FS}) of the samples was evaluated using an EX 150 DeltaLab apparatus (loading rate: $12 \times 10^{-3} \text{ kN s}^{-1}$) and Equation 3:

$$\sigma_{FS} = \frac{3FL}{2bh^2} \quad (3)$$

where F is the load at failure, L is the distance between the two supports (2.8 cm) of the apparatus and b and h are the width and the thickness of the tested samples, respectively. The measurements were carried out in triplicate, and the standard deviations were in the range of 0.1–0.7 MPa.

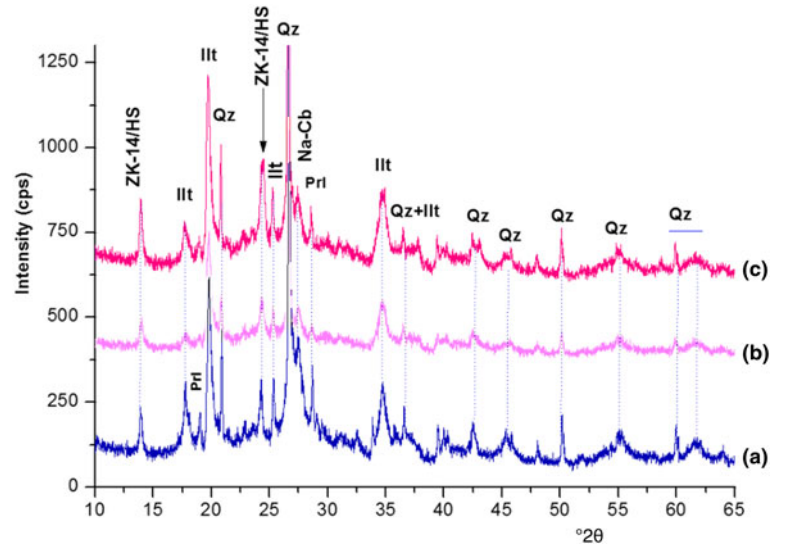


Figure 6. XRD traces of the cured samples: (a) cured cellulose-free samples; (b): cured heated clay-5 mass% cellulose; (c): cured heated clay-10 mass% cellulose. Cb = carbonate (PDF# 86-0298); HS = hydrosodalite (PDF# 76-1639); Ilt = illite (PDF# 43-0685); Prl = pyrophyllite (PDF# 96-0277); Qz = quartz (PDF# 05-0490); ZK-14 = zeolite ZK-14 (PDF# 84-0698).

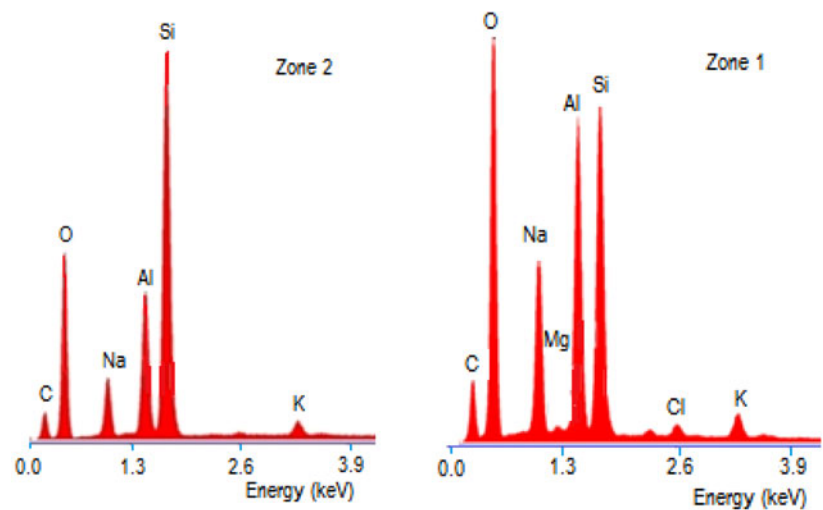
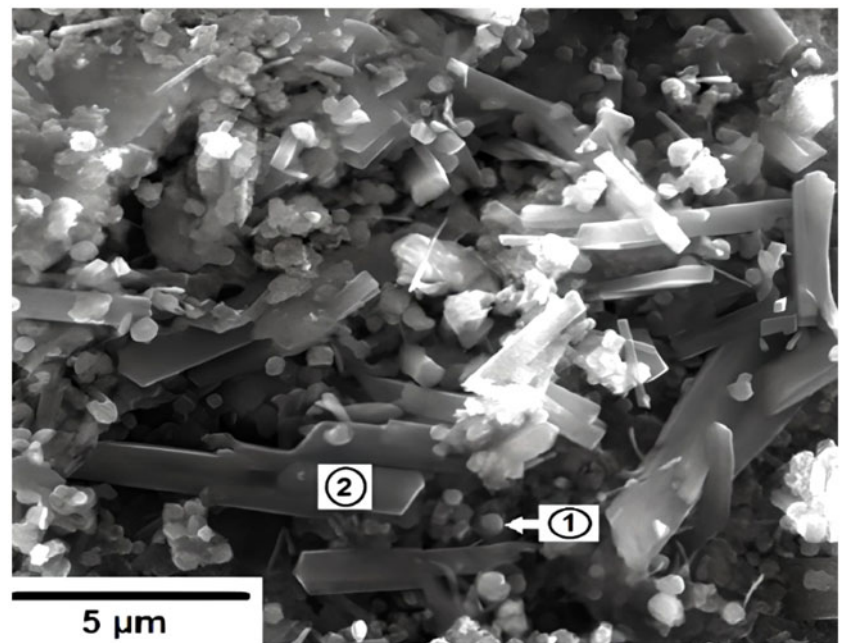


Figure 7. SEM image showing the typical microstructure of the cured sample (10 mass% cellulose) and energy-dispersive spectroscopy spectra associated with hydrosodalite (zone 1) and zeolite ZK-14 (zone 2).

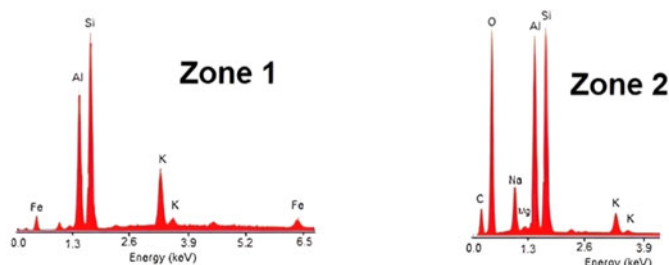
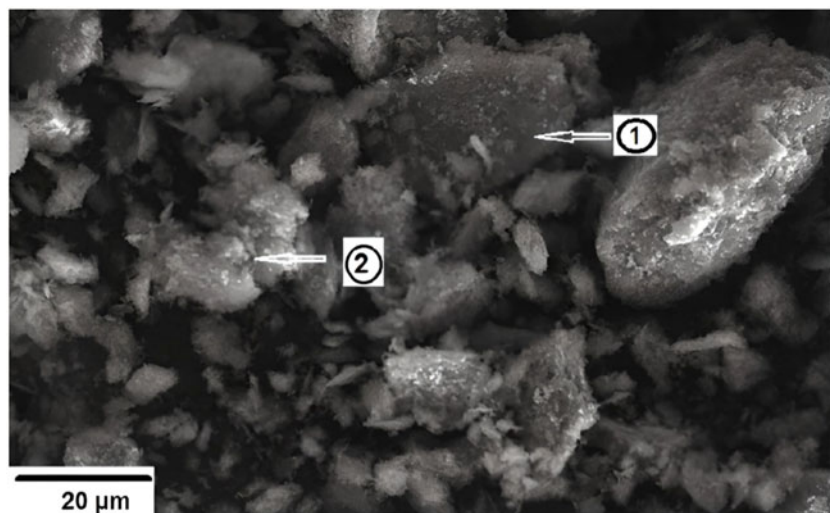


Figure 8. SEM image showing unreacted illite particles (zone 1) and the geopolymer-rich domain (zone 2) and their energy-dispersive spectroscopy spectra.

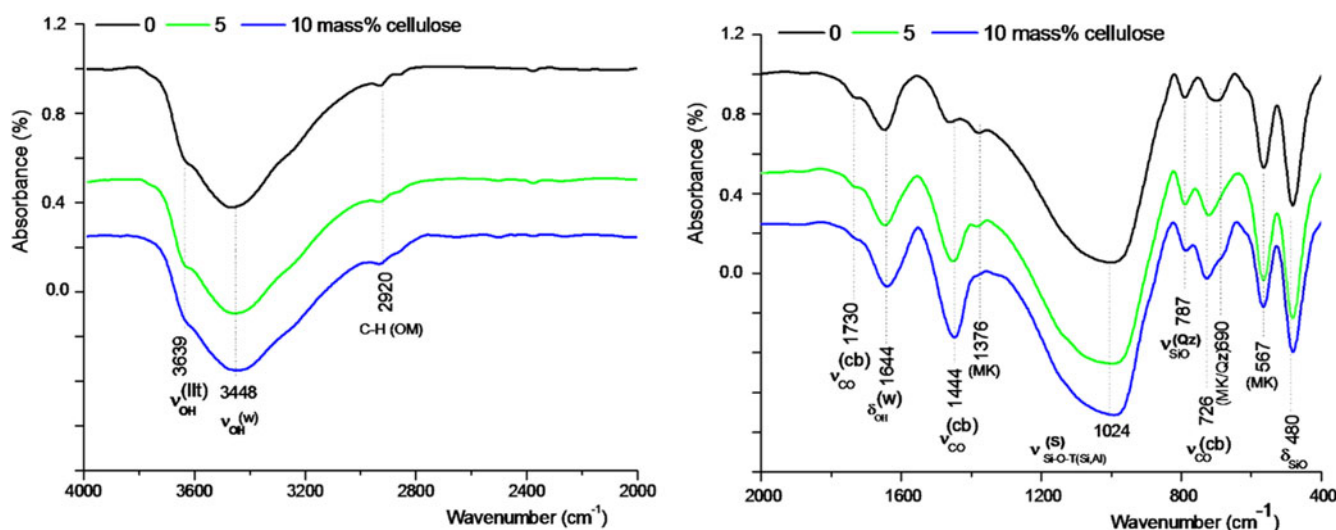


Figure 9. FTIR spectra of the cured cellulose-free sample (0% mass cellulose) and cured cellulose-containing samples (5 and 10 mass% cellulose). For clarification, the O–H stretching vibration zone and the fingerprint region are separated. cb = carbonates; Ill = illites; MK = metakaolin; OM = organic matter; Qz = quartz; S = silicates; W = water.

The total porosity (P_T) of the samples was calculated according to Equation 4:

$$P_T = 100 \left(1 - \frac{\rho_a}{\rho_t} \right) \quad (4)$$

where ρ_a and ρ_t are the apparent and true densities, which were measured using the pycnometry method.

The maximum amount of water retained by the samples (saturation time: 5 min) was determined by weighing the samples before and after contact with water. A water-submerged sponge was used as a source of water.

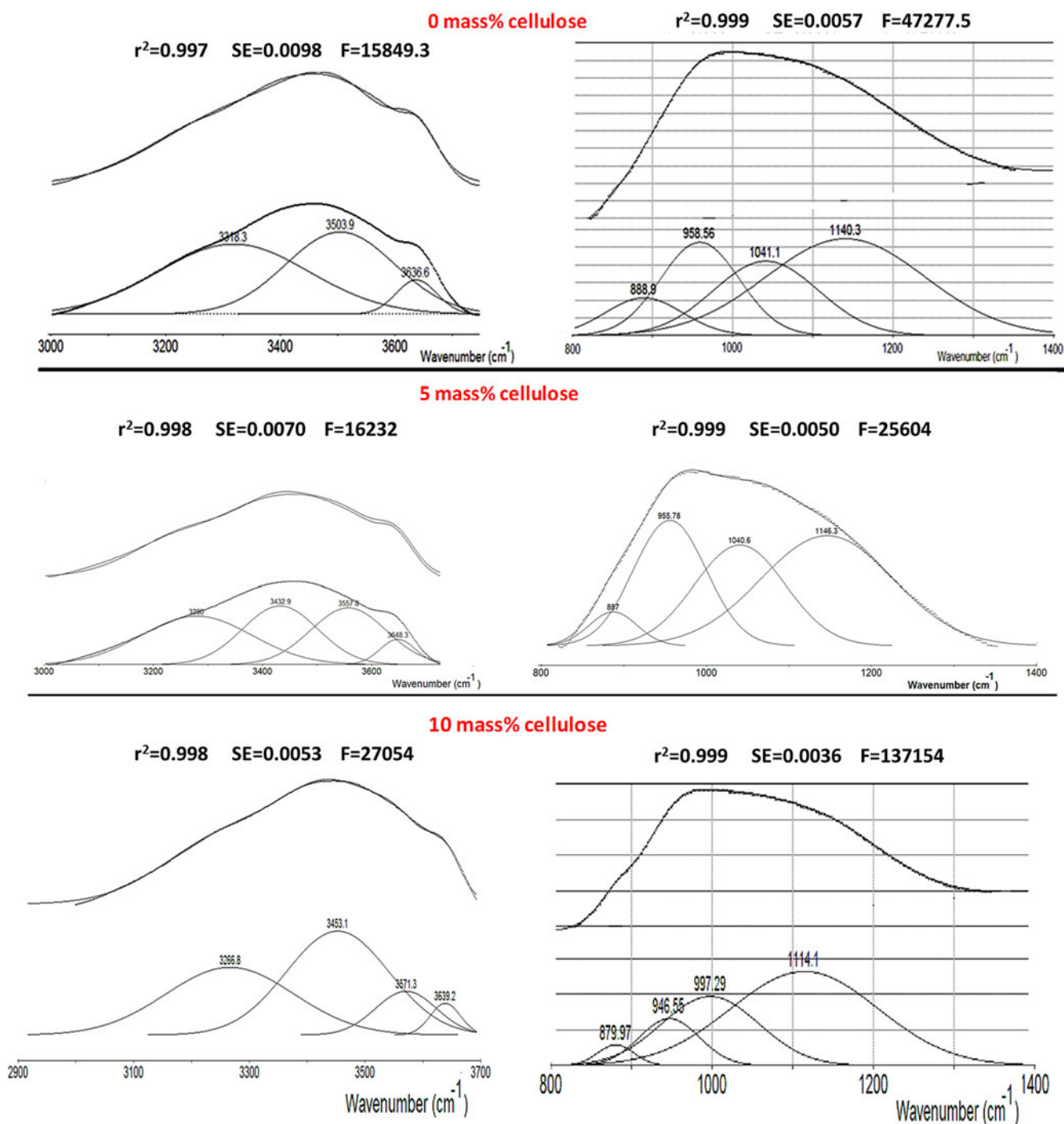


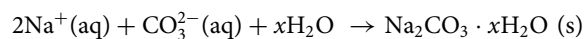
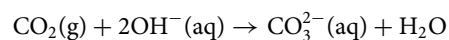
Figure 10. Deconvolution plots of the broad FTIR bands in the ranges of 3000–3700 and 800–1400 cm⁻¹.

Results and discussion

Microstructure characterization

Based on the XRD traces shown in Fig. 6, zeolite ZK-14 (PDF #84-0693) and sodium carbonate (PDF #86-0298) were formed in all cured alkali-activated samples. In line with the results of Valentini (2018), carbonation occurred in the alkali-activated metakaolin samples. According to Valentini (2018) and Zhang *et al.* (2014), such carbonation is the result of the presence of excess Na ions in the pore solution, and it occurred according

to the following reactions:



Referring once again to Fig. 6, the formation of hydrosodalite (PDF#76-1639) was suspected seeing as its main X-ray reflections

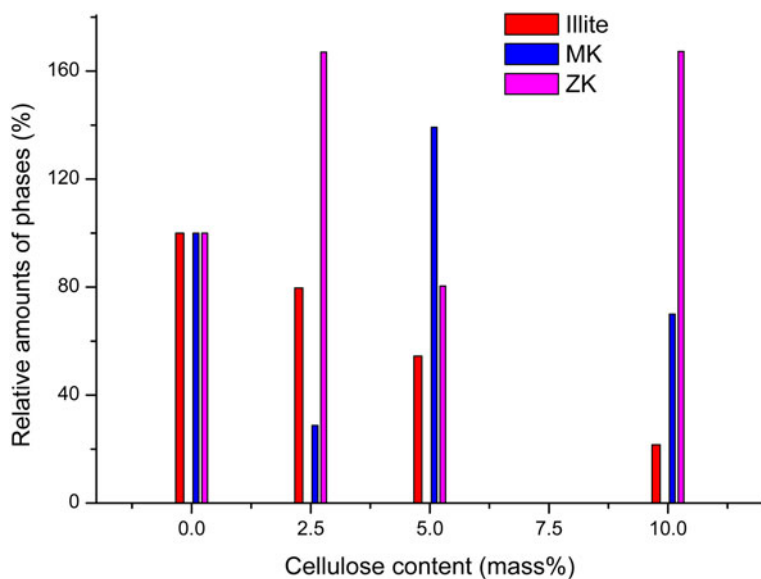


Figure 11. Variation of the relative amounts of zeolite ZK-14 (ZK), metakaolin (MK) and illite as a function of cellulose content.

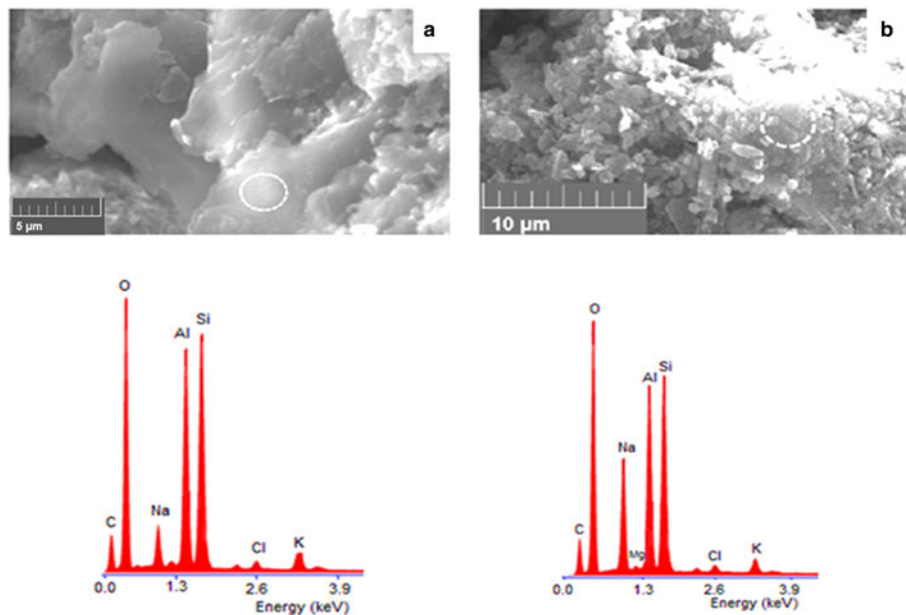


Figure 12. SEM images showing geopolymer-rich zones and their corresponding energy-dispersive spectroscopy spectra: (a) 0 mass% cellulose; (b) 10 mass% cellulose.

($2\theta = 14.09$ (6.286 Å), 24.53 (3.629 Å)) are overlapped with those of zeolite ZK-14 ($2\theta = 14.02$ (6.314 Å), 24.41 (3.646 Å)). According to Król *et al.* (2017), the formation of sodalite can be explained thermodynamically due to the higher stability of its denser lattice.

As expected, scanning electron microscopy (SEM) examination (Figs 7 & 8) revealed pseudo-granular particles identified as hydrosodalite together with elongated particles associated with zeolite ZK-14 particles. A careful examination of the microstructure displayed hydrosodalite-free zones in the vicinity of zeolite ZK-14 particles. This could be related to the differences in the nucleation and the coalescence processes of these zeolites or to the elemental heterogeneity across the matrix. Discussion of zeolite formation is provided below.

FTIR spectroscopy analysis of the cured samples indicated bands associated with metakaolin, illite, quartz and

carbonate (Fig. 9). The zeolite/geopolymer bands could not be identified with certainty because of the bands overlapping. Therefore, the broad bands in the ranges of 3000–3700 and 800–1400 cm^{-1} were deconvoluted (Fig. 10). The bands at 3504–3571 and 958–946 cm^{-1} were associated with zeolites (Hajjaji, 2014; El Hafid & Hajjaji, 2015, 2018; Mourak *et al.*, 2021). Based on the areas of the FTIR spectroscopy bands of the zeolites, metakaolin and illite, the amounts of the two former minerals evolved antagonistically, whereas that of illite slightly declined as the cellulose content increased (Fig. 11). Due to the fact that metakaolin is a precursor of zeolite (Khaled *et al.*, 2023; Maruoka *et al.*, 2023), the increase in the zeolite content should be accompanied by decreased metakaolin contents and vice versa. The unexpected presence of metakaolin in the cured samples could be due to the limited access of OH^- ions to the embedded metakaolin

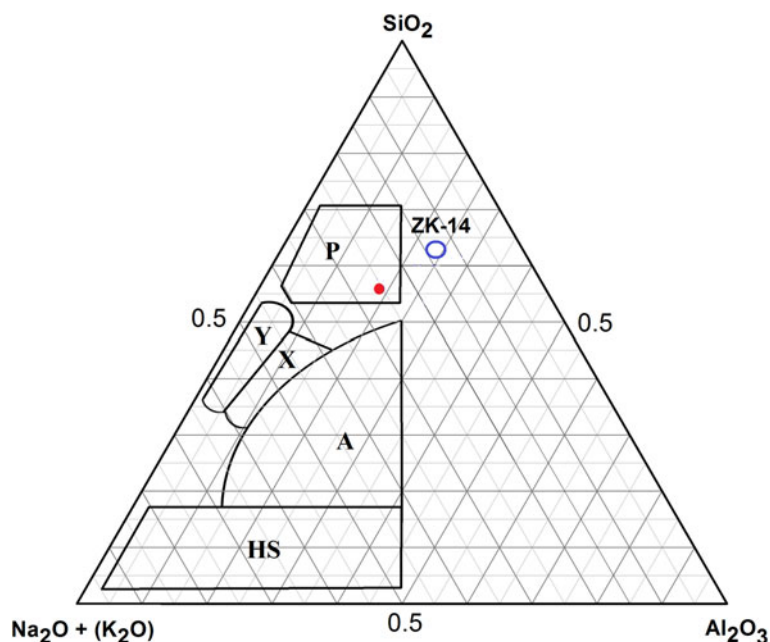


Figure 13. Ternary diagram showing the stability domains of zeolites (P, Y, X, A and HS; Xu *et al.*, 2009). The location of the zeolite ZK-14 (ZK-14) is introduced. The representative point of the cured sample is indicated in red.

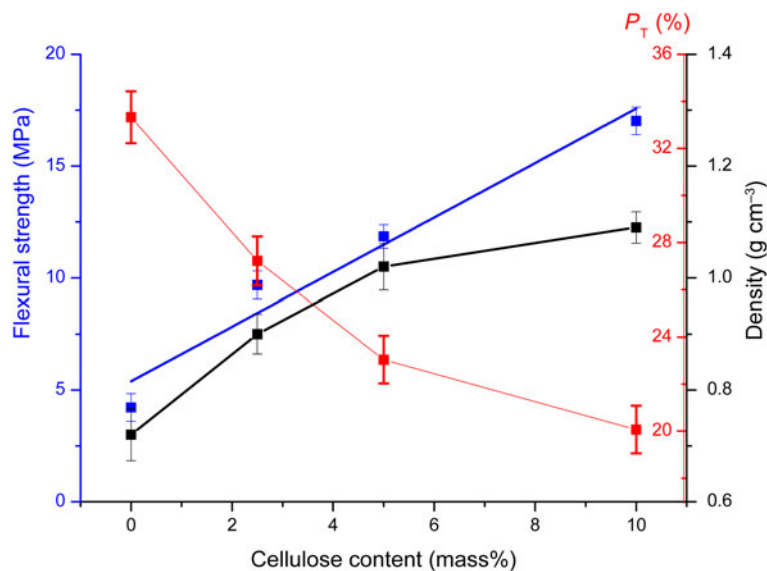


Figure 14. Variations in the mechanical and physical properties of the cured samples against cellulose content.

particles and to the interaction between OH⁻ ions and cellulose chains (mercerization reaction; Okano & Sarko, 1985; Nishimura *et al.*, 1991).

SEM examination showed that the microstructure of the cured samples was composed of zones such as those shown in Fig. 12. The features of such domains did not resemble those of the above-identified zones, and their Al/Si atomic ratios were almost constant (~1). By contrast, their (Na,K)/Si atomic ratios varied between 1.25 and 0.60. In this respect, it is worth noting that the atomic ratios of Si/Al, Si/Na and Al/Na of the used material were 1.4, 1.0 and 0.74, respectively. It was believed that the aforementioned domains corresponded to the areas of the geopolymer that was consisted of poly(sialate) units (Al/Si = 1; Zibouche *et al.*, 2009; Chen *et al.*, 2022). However, it should be noted that the geopolymer and hydrosodalite chemical compositions were quite

similar. Therefore, it could be assumed that geopolymer was a precursor of hydrosodalite formation.

Regarding the ternary diagram (SiO₂-Al₂O₃-Na₂O) that delimits the stability domains of zeolites and the chemical composition of the cured cellulose-free sample, the elemental representative point of the sample fell within the zeolite P domain (Fig. 13). The mismatch between the detected zeolites (zeolite ZK-14 and hydrosodalite) and the expected one could be essentially due to sample heterogeneity and consequently to the difference in the spatial distribution of oxides. In fact, the formation of zeolite ZK-14 required a large amount of silica (~63 mass%) and relatively low contents of Al₂O₃ (~22.9 mass%) and Na₂O (~14.1 mass%). By contrast, hydrosodalite is supposed to occur in matrix locations where the contents of silica, alumina and sodium oxide are below 19, 39 and 42 mass%, respectively.

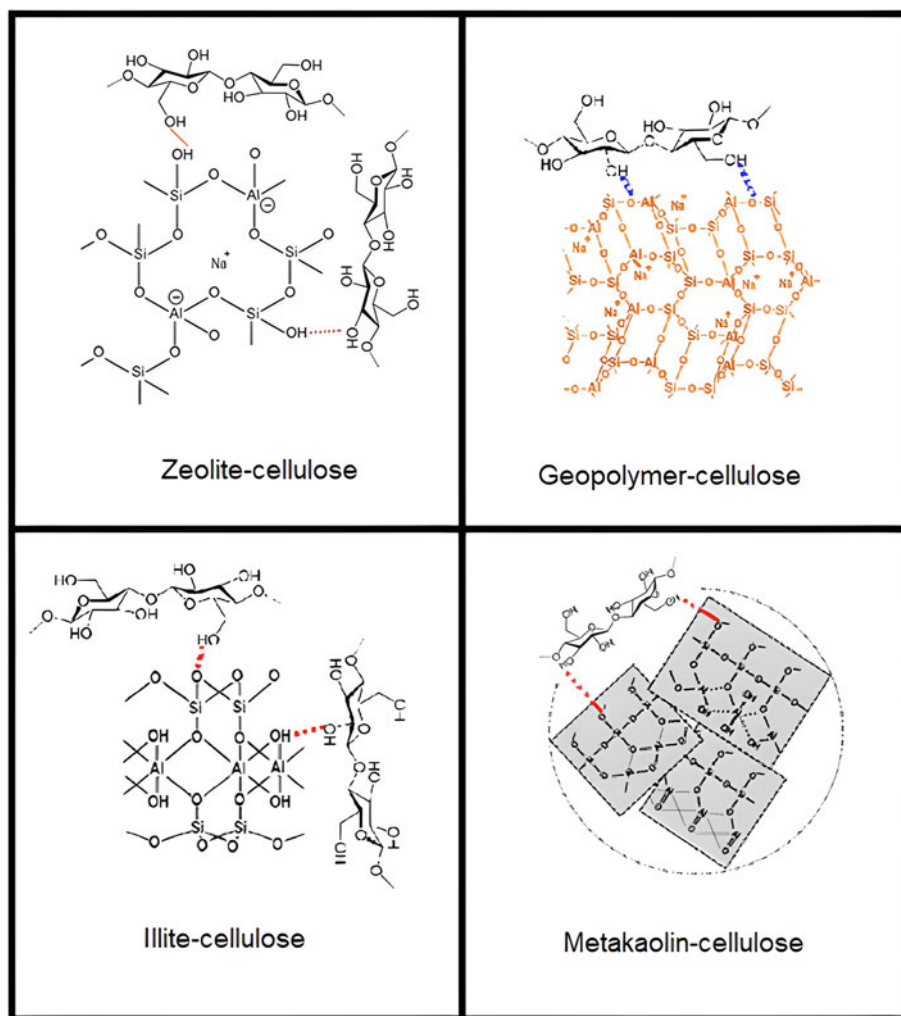


Figure 15. Schematic representations of hydrogen bonds that occurred between cellulose and the cured sample constituents (zeolite, geopolymer, illite and metakaolin).

Mechanical and physical properties

The flexural strength of the cured samples increased almost linearly with increasing cellulose content (Fig. 14). Amongst the mechanisms that might be involved in the strengthening process are the formation and the intensification of bonds between the cellulose fibres and sample constituents, the homogeneous distribution of the fibres and pores and porosity reduction (Ye *et al.*, 2018; Mourak *et al.*, 2021; Zheng *et al.*, 2023).

Regarding the influence of porosity on the mechanical strength of the cured samples, the results indicated that porosity decreased with increasing cellulose content (Fig. 14), and its evolution affected the flexural strength as follows:

$$\sigma_{FS} \text{ (MPa)} = 5.39 - 5.47 \ln \left(\frac{P_T - 18.3}{15} \right) \quad (r_{adj}^2 = 0.94)$$

The addition of cellulose also resulted in sample densification (Fig. 14). Therefore, the compaction process played a key role in sample strengthening. Moreover, the co-presence of zeolite and geopolymers induced an increase in the mechanical strength, as zeolite particles act as fillers (Andrejkovičová *et al.*, 2016). It was believed that the sample strengthening was essentially linked to the formation of hydrogen bonds between the functional moieties of cellulose and the active sites of the remaining sample

constituents. To this point, it was observed that the FTIR spectroscopy band at 3504 cm^{-1} , which was associated with the stretching vibrations of the OH of zeolite attached to the Na^+ ions (Szostak, 1992), shifted as the addition of cellulose increased

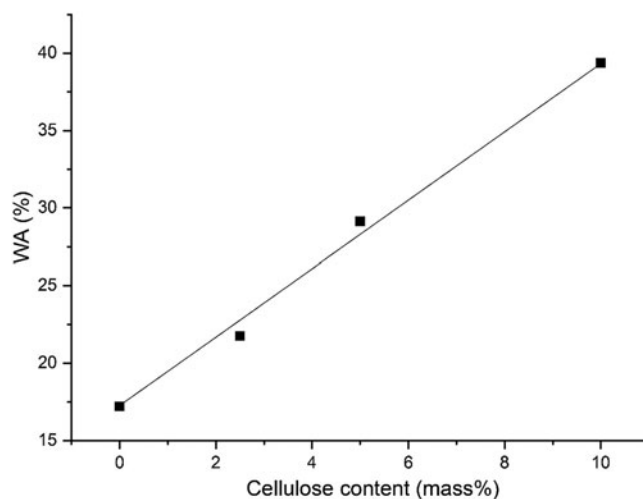


Figure 16. Evolution of water absorption (WA) by the cured samples as a function of cellulose content.

Table 2. Flexural strengths of some fibre-reinforced geopolymer composites.

Fibre	Flexural strength (MPa)	Reference
Cotton	5.85	Korniejenko <i>et al.</i> (2016)
Sisal	5.90	
Jute	5.40	Silva <i>et al.</i> (2019)
Kenaf	6.08	Abbas <i>et al.</i> (2023)
Sawdust	10.20	Duan <i>et al.</i> (2016)
Oil palm trunk	4.12	Kavipriya <i>et al.</i> (2021)
Carbon	3.90–5.10	Nuaklong <i>et al.</i> (2021)
Cellulose	Up to 17	This study

(54 and 67 cm⁻¹ for samples with 5 and 10 mass% cellulose, respectively; Fig. 10). Schematic representations of such a bond and the hydrogen bonds between the cellulose and the other constituents are provided in Fig. 15.

The increase in cellulose content led to increased water absorption, as shown in Fig. 16. This cannot be associated with sample porosity because it was shown that porosity continuously decreased with increasing cellulose content. In this context, it was believed that the measured amount of adsorbed water was an average value of the water amounts retained in the open and connected pores and by zeolites, geopolymers and cellulose fibres.

The products obtained could be used as lightweight mortars as well as binders for brick manufacturing. As compared with some natural fibre-reinforced geopolymer composites, the studied products had high flexural strength (Table 2).

Conclusion

The results of this study yielded the following conclusions:

- (1) A Moroccan clay that consisted of kaolinite, pyrophyllite and illite (the main clay mineral) was thermally activated by heating at 700°C for 2 h for geopolymer synthesis. Due to the heating, kaolinite underwent dehydroxylation at ~523°C, whereas illite and pyrophyllite resisted heating up to ~900°C. The clay heating resulted in the formation of metakaolin, which is considered to be a high-grade aluminosilicate, for the geopolymer preparation. The heated clay was mixed with cellulose (up to 10 mass%), which was extracted from date palm (*P. dactylifera*) rachis, and alkali-activated with NaOH solution (6 M). The samples prepared were cured at 83°C for 30 days under an open atmosphere.
- (2) As a result of curing, hydrosodalite, zeolite ZK-14, Na-carbonate and a geopolymer composed of poly(sialate) units (Al/Si atomic ratio = 1) were formed. The formation of these compounds, which was in line with some previous studies dealing with the geopolymerization of fly ash- or metakaolin-based source materials, was discussed in relation to the literature data.
- (3) Metakaolin as well as illite were found in all of the studied samples. The relative amounts of metakaolin and zeolites evolved in opposite directions with the addition of cellulose, whereas that of illite slightly decreased. Zeolites were relatively abundant in the cured samples containing 2.5 and 10 mass% cellulose. However, it was believed that because of the low to moderate PD (892) of the cellulose, the fibres were well dispersed throughout the matrix because no bundles were observed at the SEM magnification (20 000×) used.
- (4) Cellulose addition improved the performances of the cured samples as it increased their mechanical strength and reduced

their porosity. In addition, it resulted in their densification. Taking, for instance, the sample containing 10 mass% cellulose, the strength/density ratio was enhanced by ~17%. This performance improvement was mainly attributed to hydrogen bonds between the functional groups of cellulose and the active sites of the formed aluminosilicate minerals as well as metakaolin.

- (5) As compared with some natural fibre-reinforced geopolymer composites, the studied cured mixtures had high mechanical strength. They could be used as lightweight mortars or as binders for brick manufacturing.

Conflicts of interest. The authors declare none.

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