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Pozzolanic activity of kaolinite material rich in gibbsite calcined at low temperature and its effect on physical and mechanical properties of Portland cement mortars

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Abstract: Gibbsite in tropical kaolinite material greatly influences its reactivity as pozzolanic material. The aim of this research is to study the effect of gibbsite, $\text{Al}(\text{OH})_3$, (15.9 mass%) on the pozzolanic activity of a kaolinite material calcined at low temperature. To this effect, the as-received raw kaolinite material was calcined at 600°C and the output product was used to partially replace Portland cement by 0, 10 and 20 mass% to produce mortars. The calcined kaolinite material was firstly subjected to the modified Chappelle test and strength activity index in order to evaluate its pozzolanic activity and physical and mechanical properties of the as-produced specimens were assessed. The results show that the calcined kaolinite material presents a high pozzolanic activity (Chappelle test = 1665 mg of $\text{Ca}(\text{OH})_2/\text{g}$) compared to the reference un-calcined kaolinite material (535.0 mg of $\text{Ca}(\text{OH})_2/\text{g}$). After 28 days of curing, mortars obtained from partial replacement with 10 mass% of the calcined kaolinite material show higher compressive strength (54.5 MPa) compared to those obtained with 20 mass% (51.3 MPa). Indeed, these values are higher compared to those of mortars produced without replacement. In fact, aluminium compound promotes the formation of metastable hydrated phases ($\text{CAH}_{10}/\text{C}_2\text{AH}_8$) at early age which temporarily hinder cement hydration. Conversely, these phases are progressively transformed into stable hydrated phases of C_3AH_6 with time, thereby favoring the hardening of specimens. Thus, partial replacement of Portland Cement by 10 mass% of the calcined kaolinite material is suitable to obtain mortars endowed with enhanced compressive strength for construction purpose.

1. Introduction

Nowadays, civil construction projects are becoming more important for development of infrastructures that will sustain the population growth (Tiffo *et al.*, 2021). However, Ordinary Portland Cement (OPC) remains the main binder used to produce mortars and concretes although its production is energy consuming and it releases about 5-8% of worldwide carbon dioxide (CO_2) along with other anthropogenic gases, which are responsible of global warming and acid rain (Tiffo *et al.*, 2021). Due to the huge amount of cement necessary to carry out these projects, the use of Supplementary

Cementitious Materials (SCM) is therefore required in order to reduce the overall amount of cement to be used along the process. This will contribute to limit the use of main raw cement material (CaCO_3) as well as the energy linked to its processing (Raheem *et al.*, 2023; Verma *et al.*, 2023). However, pozzolanic materials appear as the most widely used SCM known. Yet, these materials include naturally occurring minerals, industrial wastes or by-products or those requiring less energy of processing (Alaa, 2015; Ahmed *et al.*, 2022). Among others, the most commonly used are Fly Ash (FA), Silica Fume (SF), Ground Granulated Blast Furnace Slag (GGBS), Rice Husk Ash (RHA) and Metakaolin (MK) (Sabir *et al.*, 2001; Ahmed *et al.*, 2022). Moreover, there is an ongoing interest of using clay minerals such as kaolinites due to their low energy of thermal activation in construction industries especially in areas where cement industries are in need of new raw materials to manufacture cements (Fabbriet *et al.*, 2013). In the past, the output product of thermal treatment of kaolin obtained in between 600-800°C known as metakaolin (Elimbi *et al.*, 2011) and commonly used as pozzolanic material in mortars and concretes had shown that it greatly influences strength development and durability of as-obtained products (Shvarzman *et al.*, 2003; Douamba *et al.*, 2018; Piotr, 2018), (Kwame *et al.*, 2020). In addition, several studies had revealed that metakaolin obtained at 800°C exhibits high reactivity while others had emphasized on 700°C (Elimbi *et al.*, 2011; Ndigui *et al.*, 2013; Nasir *et al.*, 2015; Magdalena and Piotr, 2018; Haw *et al.*, 2020). Indeed, the reactivity is the ability of metakaolin to react with Portlandite ($\text{Ca}(\text{OH})_2$) released during the hydration of Portland cement, with causes great improvement in pore structure and hence increasing the resistance of concretes mainly monitored by their chemical and mineralogical compositions (Rebiai *et al.*, 2010; Cassagnabère *et al.*, 2010; Hassan *et al.*, 2012; Mechti *et al.*, 2012; Alaa, 2015; Yuanyuan *et al.*, 2017; Mayuri and Pawade, 2018; Tchamo *et al.*, 2020). Concerning kaolinite mineral, several other minerals such as quartz, muscovite and gibbsite influence its reactivity (Fabbri *et al.*, 2013; Njijomou *et al.*, 2015; Yao *et al.*, 2019; Elbagermi *et al.*, 2019). Focusing on gibbsite, previous studies had shown that the presence of this mineral in the raw kaolinite material can lead to high pozzolanic activity along with simultaneous formation of both C-S-H (Calcium Silicate Hydrate) and C-A-S-H (Calcium Aluminium Silicate Hydrate) phases, which promote good strength development in the resulting Portland cement mortars (Xu and Stark, 2008; Tchamo *et al.*, 2020). Additionally, the highest pozzolanic activity is achieved when gibbsite is totally dehydroxylated and transformed into its amorphous state (Brykov *et al.*, 2012). So, the increasing amount of gibbsite or aluminium hydroxide either in its natural or synthetic form in kaolinite material, increase the mechanical strength of the resulting Portland cement products when used as SCM up to a certain threshold level before decreasing (Njijomou *et al.*, 2015; Tchamo *et al.*, 2020). Therefore, it is important to better understand the role played by the dehydroxylated product of gibbsite on the pozzolanic properties of kaolinite material calcined at low temperature when used as SCM to produce Portland cement mortars.

Based on these previous studies, the aim of the current research is to assess the pozzolanic activity of kaolinite rich in gibbsite, $\text{Al}(\text{OH})_3$, calcined at low temperature (600°C) and its effect on physical and mechanical properties of Portland cement mortars when used as partial replacement. This will contribute to valorize local mineral resources such as kaolinite materials as Supplementary Cementitious Materials in construction industries due to their low energy of thermal activation. Depending on the initial raw materials (Portland cement, kaolinite material and sand) and the synthesized products (mortars), chemical and mineralogical compositions, physical (pozzolanic activity, activity index and microstructure) and mechanical properties (compressive strength and strength activity index) were assessed.

2. Materials and Experimental Methods

2.1 Materials and characterization

The raw kaolinite material used in this study was collected in the South Region of Cameroon. It was crushed, sieved through a 20 µm mesh-sifter in accordance with the ISO 3310-1 Standard and the resulting clay fraction denoted as Kc was oven-dried (Heraeus, type VT 5042 EK) at 105°C. Its specific surface area was measured thanks to BET method while its chemical composition was determined by Inductively Coupled Plasma Optical Emission Spectroscopy using a PerkinElmer Spectrometer (Optima™ 7000 DV ICP-OES). In addition, thermogravimetric (TG) analysis was obtained by using a NETZSCH STA 409 brand instrument operating at 75 L.min⁻¹ under a self-generated flux of air at the heating rate of 5°C.min⁻¹ in order to assess its quantitative analysis. The raw kaolinite fraction was then calcined at 600°C at the heating rate of 10°C / min for 6 hours at the highest peak in a programmable electric furnace (Nabertherm, model LH, 60/14) and the output product was denoted as MKc. The mineralogical changes of Kc and MKc were followed by X-ray Diffraction analysis using a Siemens D-5000 brand diffractometer featuring with a Bragg-Brentano assembly with a back graphite monochromator using radiation from the Kα1 of cobalt ray (~1.17 Å). Also, the Infrared spectroscopy analysis was done using a Cary 630 FTIR Spectrometer and all spectra were recorded with a resolution of 4 cm⁻¹ within the range of 600-4000 cm⁻¹. The Portland cement used was CEM I 52.5 N CP2, composed of 97 mass% of clinker and 3 mass% of gypsum. The silica sand with a 0 / 4 particle size distribution obtained after sieving was also used. Its mass density measured thanks to a pycnometer was 2.61 g.cm⁻³ and its fineness modulus acquired in accordance with ASTM C33 test (1999) was 2.18. Its sand equivalent value was determined in accordance with the ASTM D2419-95 (1998). The pozzolanic activity of both Kc and MKc was assessed by the modified Chapelle test according to NF P18-513 Standard.

2.2 Mixture design and characterization of mortars

Mix proportions in mortars were done by partially replacing Portland cement by 0, 10 and 20 mass% by the calcined kaolinite (**Table 1**). Based on preliminary considerations, MC was referenced as the control mortars where the binder (B) is Portland cement. The water / binder (W / B) mass ratio in mortars were obtained by adding to each paste 1350 g of sand (ratio of binder / sand = 1 / 3). Indeed, fresh mortars were poured into parallelepipedic metallic moulds (4x4x16 cm³) and vibrated for 10 minutes using an electrical vibrating table (M & O, type 202, N° 106) in order to remove entrapped air bubbles. The resulting specimens were conserved in a thin film plastic and then placed in a chamber having 100% of humidity at 20°C prior to be characterized respectively at 3, 7 and 28 days of curing. Compressive strength was assessed thanks to an electro-hydraulic press, type ENERPAC, 3R according to NF EN 1015-11 Standard. The strength activity index was evaluated according to the ASTM C618 Standard (**Eqn. 1**).

$$I_c = \frac{R_c}{R_{c(\text{control})}} \times 100 \quad \text{Eqn. 1}$$

Where R_c is the compressive strength of mortars, and $R_{c(\text{control})}$ is the compressive strength of the control mortars (MC).

After compressive strength test, fragments of specimens aged 28 days were crushed, sieved at 20 µm mesh-sifter and the resulting powders were used to carry out both FT-IR and X-Ray Diffraction (XRD) analyses. In addition, the polished and etched surfaces of the broken specimens were analyzed by Scanning Electron Microscopy (SEM / EDX) with JSM 5900 LV mode apparatus.

Table 1. Formulation of mortars

Formulations	Cement (g)	B / S	MKc (g)	W/B
MC	450	1/3	00	0.55
M(MKc) ₁₀	405	1/3	45	0.55
M(MKc) ₂₀	360	1/3	90	0.55

W/B = Water / Binder mass ratio; B/S = Binder / Sand mass ratio

3. Results and Discussion

3.1 Characterization of starting materials

Results of chemical, physical and mineralogical compositions of kaolinite material (Kc), calcined kaolinite material (MKc) and Portland cement are reported in **Table 2**. It can be observed that the mineral C₂S responsible of the long-term durability does not appear in the mineralogical composition of the cement. Therefore, this absence can significantly influence the mechanical strength of the resulting cement mortars (Dupain *et al.*, 2004).

Table 2. Chemical, mineralogical and physical characteristics of starting materials

Oxides (mass%)	Portland Cement CEM I 52.5 N (C)	Kaolinite (Kc)	Calcined kaolinite (MKc)
Chemical compositions			
Al ₂ O ₃	3.30	40.10	48.40
SiO ₂	20.10	38.00	45.10
K ₂ O	3.00	0.24	0.25
Fe ₂ O ₃	5.20	0.70	0.86
CaO	64.10	0.05	0.63
TiO ₂	-	1.10	1.23
Na ₂ O	0.28	0.04	0.03
MgO	0.80	0.13	0.17
SO ₃	3.00	0.01	0.01
LOI	1.80	18.75	2.81
Mineralogical compositions (mass%)			
Kaolinite	-	75.30	
Gibbsite	-	15.90	
C ₃ A	8,60	-	
C ₃ S	61.00	-	
C ₄ AF	11,10	-	
Physical characteristics			
Specific surface (m ² /g)	-	16.99	
Density	3.09	2.38	2.61

LOI: Loss On Ignition

Regarding the kaolinite, it is apparent that it contains silica (SiO₂) and alumina (Al₂O₃) as major oxides associated among others with iron oxide (Fe₂O₃), potassium oxide (K₂O) and titanium oxide (TiO₂) as minor oxides. The SiO₂ / Al₂O₃ mass ratio is 0.94 instead of 1.17 generally encountered for a pure kaolinite (Njiomou *et al.*, 2015). This difference confirms the presence of free alumina, deriving

from the gibbsite fraction of the kaolinite material. The amount of oxides (% Al₂O₃ +% SiO₂ +% Fe₂O₃) is 78.88 mass% which confers to Kc a first attribute to be a pozzolanic material (ASTM C618) (Ndigui *et al.*, 2013). Indeed, Al₂O₃ and SiO₂ especially in their amorphous state appear to be the basic ingredients for a good pozzolanic reaction (Alaa, 2015). In fact, these oxides exhibit the ability to react with the CaO of cement or lime to give cementitious phases such as C-S-H or C-A-S-H gels, responsible of the compactness of as-produced material and thus making it to be more resistant to chemical attack (Worrall, 1986; Sabir *et al.*, 2001; Jaward *et al.*, 2022). Furthermore, the presence of gibbsite in the kaolinite is responsible for the high amount of Al₂O₃ (40.10 mass%), and it is confirmed by its TG curve in Figure 3 which exhibits a mass loss of 5.5 mass% due to the thermal decomposition of gibbsite in between 200-400°C, and a characteristic mass loss connected to kaolinite in between 400-700°C which is 10.5 mass%. The presence of kaolinite is also confirmed by IR spectrum in Figure 2 with the absorption bands of a disorder kaolinite at 3620, 3649 and 3690 cm⁻¹ with the lack of the absorption band at 3665 cm⁻¹ (Worrall, 1986). Other absorption bands at 1113, 1027 and 1006 cm⁻¹ in the IR spectrum of Kc correspond respectively to the bending vibrations of symmetric Si-O et Si-O-Si, and asymmetric Si-O-Al (Kakali *et al.*, 2001), while others at 911 cm⁻¹ as well as those at 786, 750 and 677 cm⁻¹ are attributed to bending vibrations of Al-O-Si and Al-OH of kaolinite mineral (Sujeet *et al.*, 2020). The disorder in Figure 2 in Kc enables it to be a good reactive material after thermal activation (Kakali *et al.*, 2001).

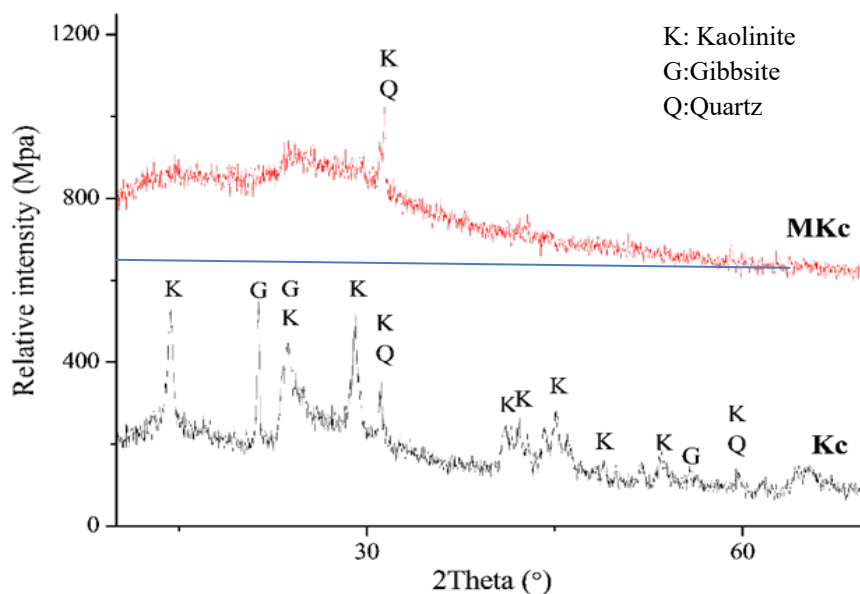


Figure 1. X-ray patterns of the raw (Kc) and calcined kaolinite (MKc).

The thermal treatment creates an amorphous phase within MKc evidenced in the X-ray pattern by a large dome observed in between 0 and 30° 2θ on the X-ray pattern of MKc in Figure 1 (Tchakoute *et al.*, 2018). Asymmetric and symmetric broad vibrations at 1053 and 753 cm⁻¹ of Si-O-Al and Si-O-Si bending vibrations and the presence of amorphous silica on the absorption bands of IR in Figure 2 (Farmer, 1998; Tchakoute *et al.*, 2018) confirm these results. The X-ray pattern of MKc also shows the presence of two peaks at around 30° 2θ characteristic of kaolinite and quartz, suggesting that the calcination had not converted MKc into a totally amorphous compound (Tchakoute *et al.*, 2018). This result is confirmed by the TG curve of Kc which shows that the dehydroxylation of the kaolinite is still ongoing above 600°C in Figure 3.

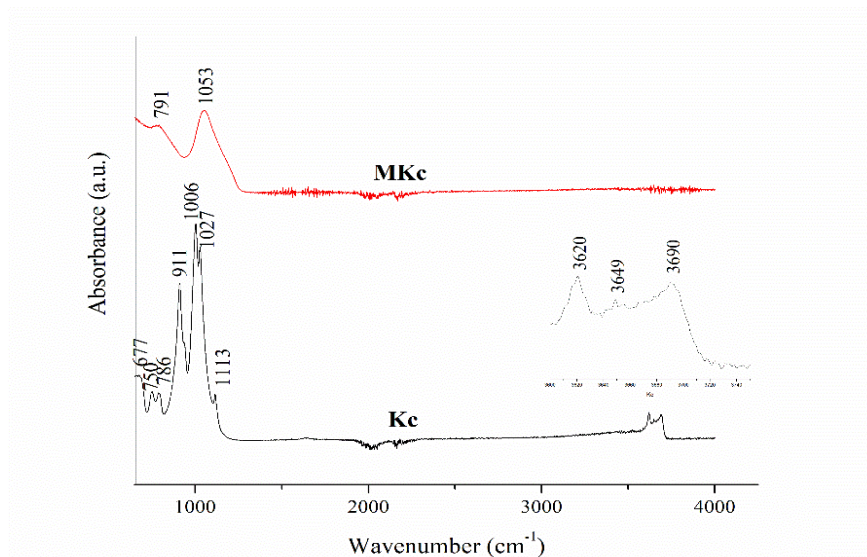


Figure 2. Absorption bands of the raw (Kc) and calcined kaolinite (MKc).

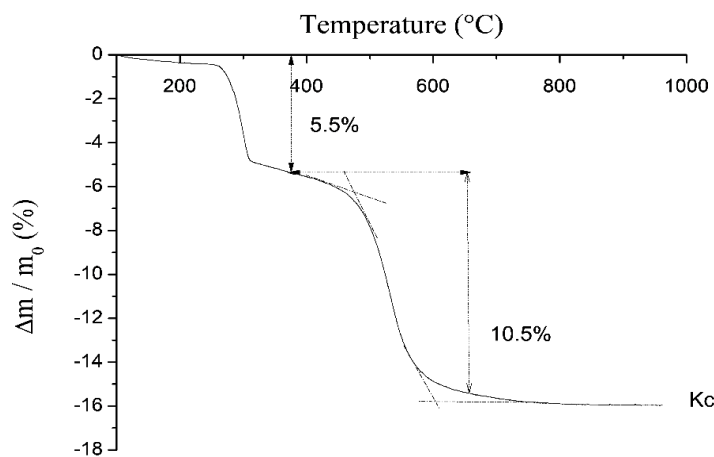


Figure 3. TG curve of the raw kaolinite material (Kc).

3.2 Pozzolanic activity and strength activity index

The pozzolanic activities of both raw and calcined kaolinite powders are presented in **Table 3**. It can be observed that thermal treatment increases the pozzolanic activity of the raw kaolinite from the value of non-pozzolanic material (535.0 mg Ca(OH)₂/g) to the value of pozzolanic material (1665.8 mg of Ca(OH)₂/g) according to the NF P18-513 Standard. Indeed, that standard states that the pozzolanic activity of a pozzolanic material should not be less than 700 mg Ca(OH)₂ per gram of this material (Antoni *et al.*, 2012; Njiomou *et al.*, 2015; Edwin and Dunstan, 2017; Tironi *et al.*, 2017). This material can be classified as highly reactive calcined kaolinite in cementitious systems (Antoni *et al.*, 2012). These results are in accordance with the strength activity index of the specimens obtained by partially replacing the cement by 10 and 20 mass% of calcined kaolinite in **Figure 4**. Strength activity index results show that at early age, specimens obtained from partial replacement of cement by 10 mass% of MKc presents a greater strength activity than those obtained with 20 mass%. Indeed, the highest strength activity index observed with the lower replacement level (10 mass%) can be attributed to the high concentration of CH (Portlandite) available to react with the pozzolanic material to yield a greater amount of cementitious phases (Poon *et al.*, 2001). In addition, this highest strength

activity index can also be ascribed to the lower presence of dehydroxylated gibbsite in 10 mass% substitutions to cement which inhibits cement hydration at young age less than in 20 mass% substitutions (Brykov *et al.*, 2012).

Table 3. Pozzolanic activity of starting materials

	Raw kaolinite (Kc)	Calcined kaolinite (MKc)
Pozzolanic activity (mg Ca(OH) ₂ /g)	535.00	1665.80

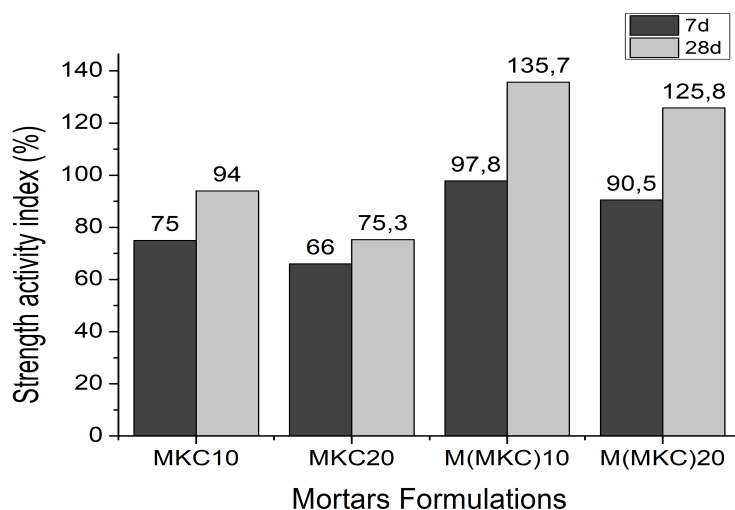


Figure 4. Strength activity index of mortars

3.3 Mineralogical and microstructural changes

The FT-IR absorption bands of the mortars denoted as M(MKc)₁₀ and M(MKc)₂₀ are presented in **Figure 5**. It can be observed that the absorption bands are the same except the intensities of peaks on M(MKc)₂₀ which are lower than those on M(MKc)₁₀. This difference can be observed on the broad absorption bands of M(MKc)₂₀ at 1081 and 998 cm⁻¹ and those of M(MKc)₁₀ at 963 cm⁻¹ which are characteristics of stretching vibration of strong Si-O group of crystallized C-S-H (Yilmaz and Olgun, 2008; Sujeet *et al.*, 2020). The absorption bands are more pronounced on M(MKc)₁₀ as compared to those observed on M(MKc)₂₀ which could indicate a lower content of silicate phases in the latter than on M(MKc)₁₀. The Al-O vibrations from silico-aluminate hydrates can also be responsible for the strong bands at around 1000 cm⁻¹ and 963 cm⁻¹ for M(MKc)₂₀ and M(MKc)₁₀ respectively (Tchakoute *et al.*, 2018; Sujeet *et al.*, 2020). The stretching vibration of SO₄²⁻ which is responsible of the presence of gypsum in the clinker appear around 1075 and 1165 cm⁻¹ respectively in M(MKc)₂₀ and M(MKc)₁₀ (Sujeet *et al.*, 2020). C-O stretching vibrations are respectively observed around 1420 and 873 cm⁻¹ for CO₃²⁻ of carbonate phases (Gopalakrishnan and Govindarajan, 2011; Kwame *et al.*, 2020). As compared to M(MKc)₁₀, M(MKc)₂₀ has low carbonate contents (weak bending and stretching vibrations). The absorption bands at 651 cm⁻¹ can be assigned to S-O vibration band of ettringite 3CaO•Al₂O₃•3CaSO₄•32H₂O (Yilmaz and Olgun, 2008; Sujeet *et al.*, 2020). The broad absorption band center at 3400 cm⁻¹ can be attributed to the symmetric and the asymmetric stretching vibrations of O-H group of bound water in mortars (Sujeet *et al.*, 2020).

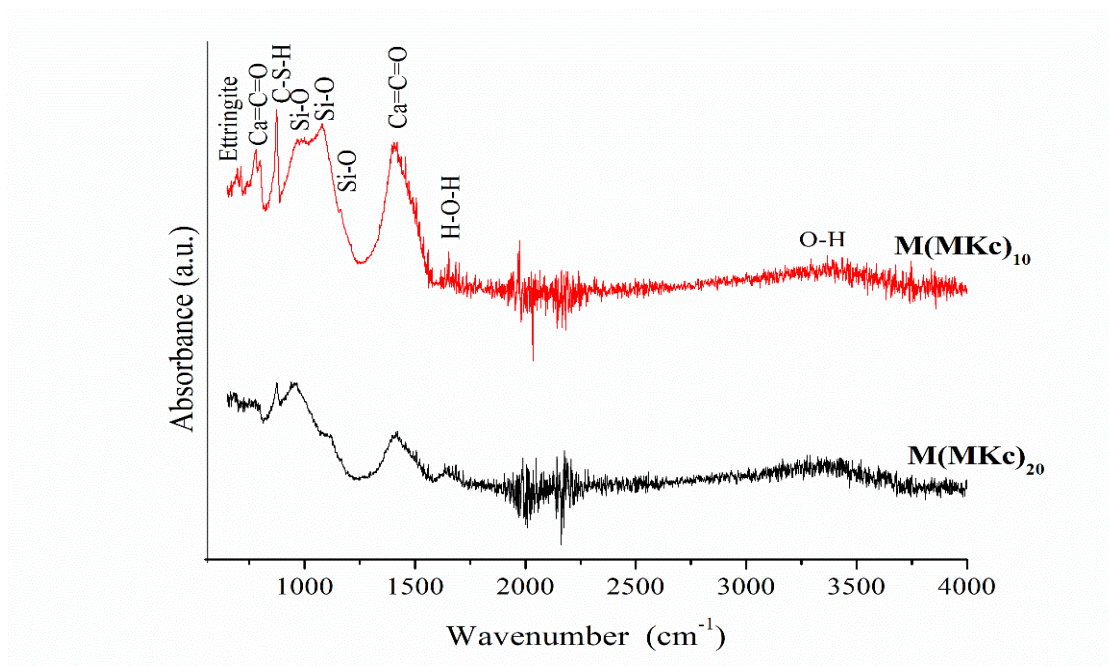


Figure 5. FT-IR adsorption bands of the mortars M(MKc)₁₀ and M(MKc)₂₀.

The absorption bands at 1653 cm⁻¹ correspond to the bending vibration of H-O-H bonds of adsorbed water molecules (Çelik *et al.*, 2008; Sujeet *et al.*, 2020). A strong and reduced intensities of the peak at 870 cm⁻¹ assigned to Si-O of C-S-H is observed on M(MKc)₂₀. This could suggest that C-S-H is more observed on M(MKc)₁₀ specimens than on those of M(MKc)₂₀. This shows that partial replacement of cement by 10 mass% with MKc reacts more than those obtained with 20 mass% after 28 days of curing. The diffractograms of mortars obtained after 28 days of curing are shown in **Figure 6**.

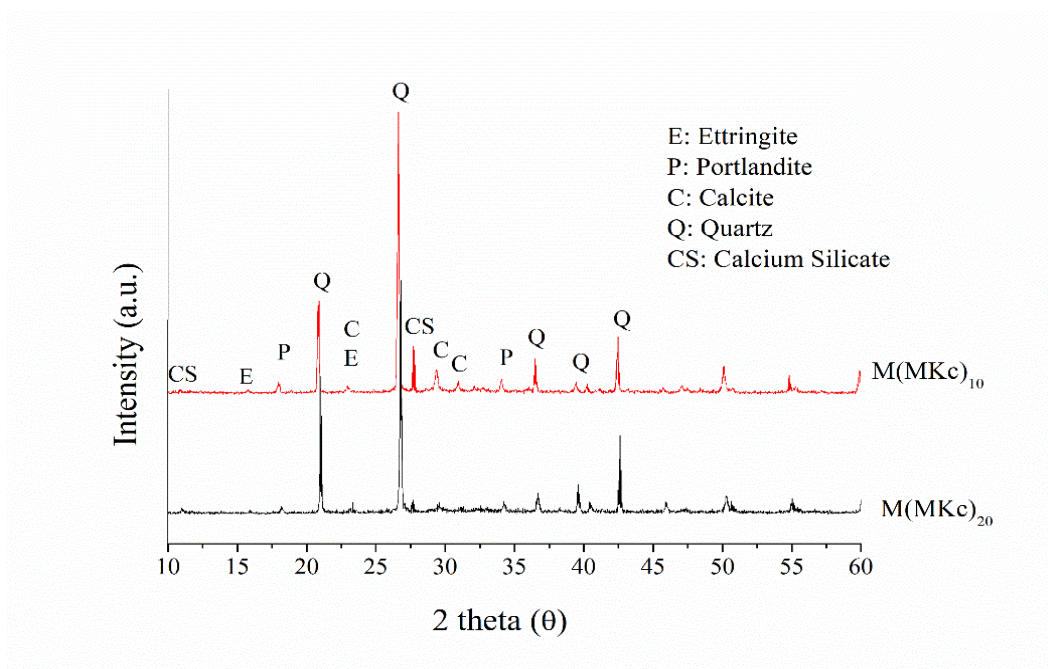


Figure 6. Diffractograms of the mortars M(MKc)₁₀ and M(MKc)₂₀.

There are no major differences on their diffractograms unless around 25-30° 2θ. This difference suggests that C-S-H phases formed in the mortars M(MKc)₁₀ are in greater amount as result of the high intensity of the main peak as compared to its counterpart M(MKc)₂₀. This is in accordance with the IR

results of mortars. It can also be noticed that on various micrographs of the mortars M(MKc)₁₀ and M(MKc)₂₀ presented in **Figure 7**, the sample M(MKc)₁₀ exhibits high compactness (in **Figure 7a**) as compared to M(MKc)₂₀ one (in **Figure 7c**). In addition, EDX results also confirm the presence of calcium silicate phases on both features.

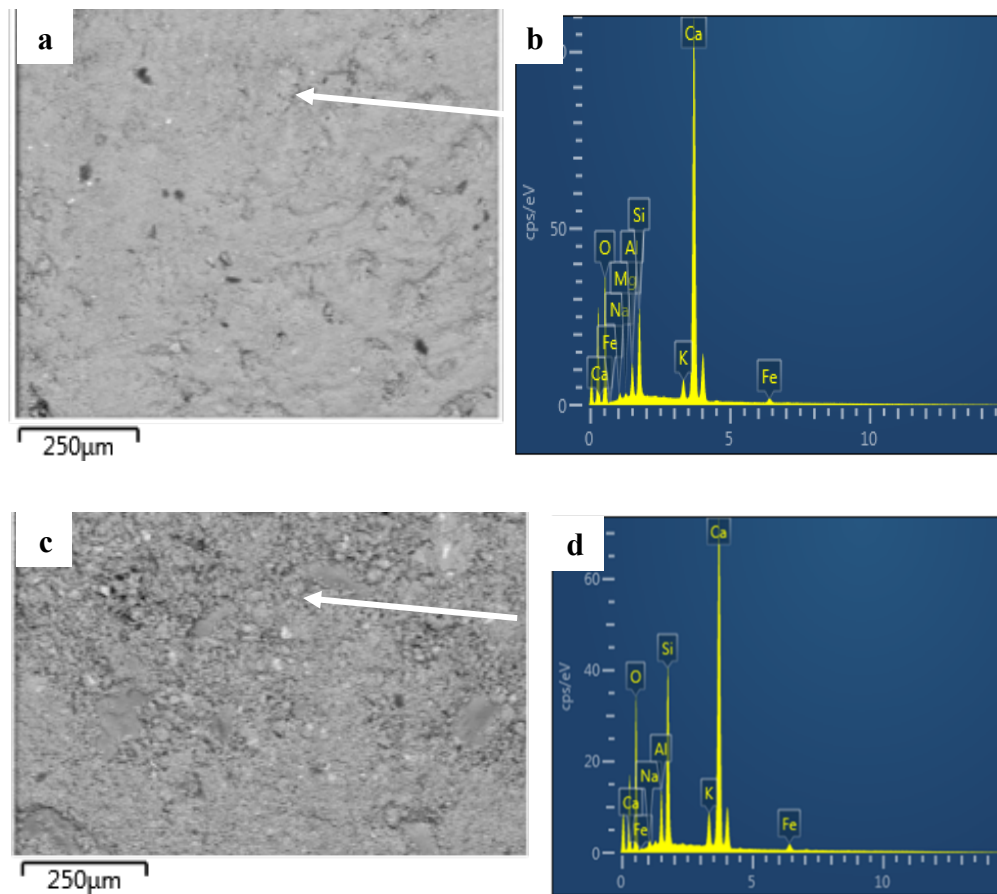
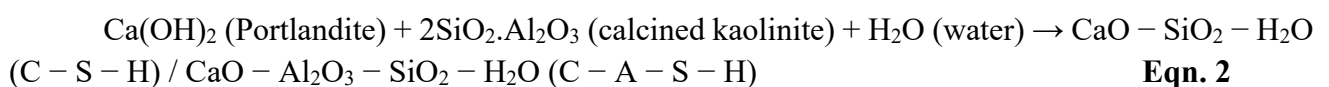


Figure 7. Micrographs of M(MKc)₁₀ (a,b) and M(MKc)₂₀ (c,d)

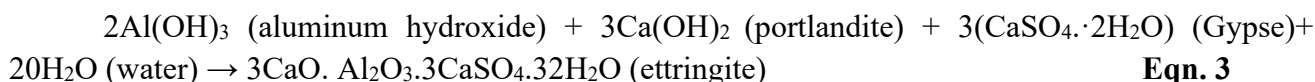
3.3 Physical and mechanical properties of mortars

Figure 8 presents the compressive strengths of the reference mortars (MC) along with those obtained by partially replacing cement by 10 and 20 mass% of calcined kaolinite material and cured respectively at 3, 7 and 28 days. It can be observed that compressive strength of all mortars increases with the curing time. In fact, mortars obtained from partial replacement with calcined kaolinite material present the highest compressive strength at all ages. This is probably due to the pozzolanic reaction occurring between reactive calcined kaolinite and calcium hydroxide (Portlandite) released during cement hydration which forms additional calcium silicate hydrate gels (C-S-H) (**Eqn. 2**) responsible of strength development (**Poon et al., 2001**), (**da Silva et al., 2023**).



In addition, the highest compressive strengths were observed in the specimens obtained with 10 mass% substitution of cement by calcined kaolinite at all ages, with an increase of about 33% at 28 days as compared to the control mortars. This is probably due to the presence of aluminium in the

structure which inhibits cement hydration by forming metastable hydrated phases of CAH_{10}/C_2AH_8 at young age, and which are transformed with time into more stable phases of C_3AH_6 (Eqn. 3-6) which favored the hardening of resulting mortars (Xu and Stark, 2008; Tchamo *et al.*, 2020).



This suggests that three reactions occur in the cement system: the cement hydration reaction, the pozzolanic reaction and the aluminium hydration reaction. The pozzolanic and aluminium hydration reactions could reveal that aluminium reactions are preponderant than pozzolanic reactions in Portland cement mortars. Then, to obtain high mechanical performance of cement mortars, it is very important to control the amount or dosage of aluminium compound to be used in mortars (Jabri *et al.*, 2012).

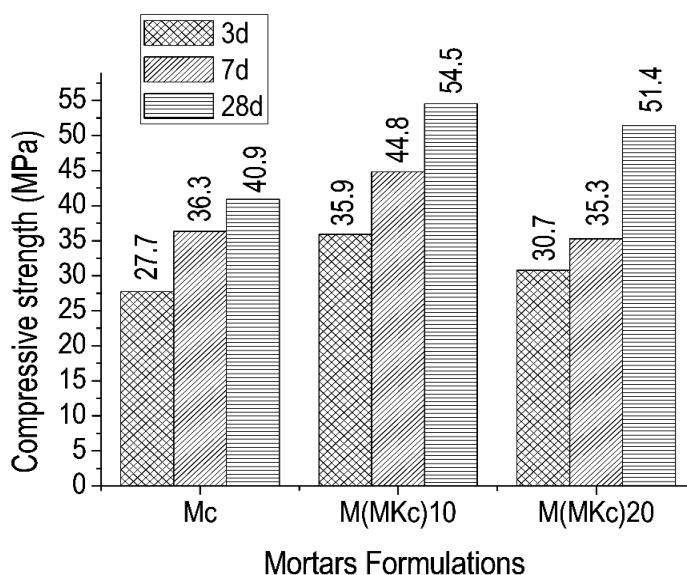


Figure 8. Compressive strength of mortars at various ages.

Conclusion

This study has investigated the pozzolanic activity of kaolinite material rich in gibbsite calcined at low temperature and its effect on physical and mechanical properties of Portland cement mortars when used as partial replacement. The following conclusion can be drawn.

- The thermal treatment at 600°C converts the kaolinite rich in gibbsite into a high reactive pozzolanic material as compared to the raw material according to the NF P18-513 Standard.
- The calcined kaolinite used as supplementary cementitious material increases considerably the compressive strength of resulting Portland cement mortars as compared to those obtained without replacement.
- The aluminium hydrate reactions in cement system are preponderant to pozzolanic reactions.

Hence, it is very important to control the amount or the dosage of aluminium compound to be used in Portland cement mortars in order to enhance considerably the performance of resulting mortars. Therefore, it can be concluded that 600°C is the convenient temperature to be used to convert kaolinite rich in gibbsite into a reactive supplementary cementitious material.

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Compliance with Ethical Standards: This article does not contain any studies involving human or animal subjects.

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