Insight into the geopolymerization of earth-based materials: Effect of clay/silt precursor and Na_2SiO_3/NaOH activator on geopolymerization

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Abstract. The geopolymerization of earth-based materials is a promising alternative for low-carbon building materials. This study explores the synthesis of geopolymers with clay, silt, and a clay-silt mixture as precursors activated by NaOH and NaOH+Na_2SiO_3 (Meta-Si), with an activator/precursor mass ratio A/P=0.4. The results showed that the use of Meta-Si increases compressive strength compared to samples activated only with NaOH. The increase in strength is linked to the resulting microstructure. Infrared spectroscopy (FTIR) shows the presence of O-H groups characteristic of N-A-S-H and C-A-S-H gels, which agrees with thermogravimetric analysis (TGA). The synergy between the two gels contributes to the densification of the matrix and the mechanical consolidation of the materials. These results highlight the key role of silicate activators in the development of high-performance earth-based geopolymers, paving the way for the use of local resources and industrial by-products in the manufacture of environmentally friendly and sustainable building materials.

Keywords: Geopolymerization, Clay, Silt, Mechanical Strength, Microstructure.

1 Introduction

Earthen construction has been employed for ages in many parts of the world due to the abundance, low cost, and thermal performance of soils such as clay and silt [1]. However, raw earth is often criticized for its low mechanical strength, high sensitivity to water, and shrinkage during drying, which limits its widespread use in modern construction [2]. Meanwhile, the environmental impact of conventional cementitious materials, responsible for approximately 7-8% of global CO_2 emissions, has motivated the search for sustainable, low-carbon solutions in the building sector [3]. In this context, improving the performance of earth-based construction while maintaining its ecological advantages has become a major scientific challenge [4].

Several reinforcement strategies have been proposed in the literature to improve the durability and mechanical performance of earth-based materials. The most common approach remains stabilization with cement or lime, which increases compressive strength and water resistance [5]. However, the use of Portland cement raises environmental concerns due to its high carbon footprint [6]. More sustainable solutions involve the use of mineral additives such as fly ash, granulated blast furnace slag (GGBS), silica fume, or metakaolin [7]. These additions act as precursors and participate in geopolymeric reactions with activators, thus generating gels (C-A-S-H, N-A-S-H) which densify the matrix and improve strength and durability. In the case of simple additions without activator, pozzolanic reactions occur in the presence of water, forming C-S-H gels [7].

More recently, focus has shifted to alkaline activation and geopolymerization as innovative methods of soil stabilization [8]. Geopolymers result from the reaction between aluminosilicate-rich precursors (clay, silt, fly ash) and alkaline activators (NaOH, Na_2SiO_3), producing amorphous to semi-crystalline gels such as N-A-S-H and C-A-S-H. These binders offer higher mechanical strength than soil alone (approximately 5 MPa), resulting in a 30% improvement in mechanical strength and good durability, while also allowing for the reuse of local soil and industrial by-products [8, 9]. Several studies have shown that combining clays with fly ash or other reactive additives promotes matrix densification and increased strength, highlighting the synergistic role of hybrid gels in improving performance [7].

Based on these findings, earth-based geopolymerization is a promising method for developing sustainable building materials. Indeed, the use of excavated soil, which may be contaminated, under optimized alkaline activation conditions makes it possible to design eco-efficient, high-performance binders that can serve as a credible alternative to traditional cementitious materials. This work aims to study the geopolymerization of clay, silt, and mixtures of clay/silt, which are considered precursors activated by sodium hydroxide NaOH (10M) or a mixture of sodium hydroxide and Meta-Silicate (Na_2SiO_3/NaOH=2).

2 materials and mixtures

2.1 Raw materials

2.1.1 Soils preparation

The clay and silt used in this study were obtained from the DEPAUL platform (Environment and Renewable Natural Resources) based in Annet-sur-Marne (France). These materials are excavation products sourced from the Île-de-France area. Firstly, the clay and silt were dried at 105 °C for 24 hours to remove moisture and ensure consistent conditions before processing (Step 1, Fig.1). After drying, the materials were subjected to mechanical grinding using a Netzsch ball mill TM 300. The milling process aimed to reduce the particle size to below 100 µm, ensuring adequate reactivity during subsequent geopolymerization. The milling parameters were carefully controlled. A total of

18.75 kg of steel balls was used, consisting of a mixture of 40% with a diameter of 30 mm, 30% with a diameter of 20 mm, and 30% with a diameter of 10 mm. This mass corresponded to 80% of the drum capacity (10 L). In addition, 6 kg of soil (clay or silt) was added, representing approximately one-third of the drum volume. The milling operation was carried out at a speed of 80 rpm for 1 hour. According to the crusher constructor, these parameters are optimal to obtain more fines [10]. After milling, the powders were sieved, and only the fine fraction with particle sizes below 100 µm was retained for subsequent experiments.

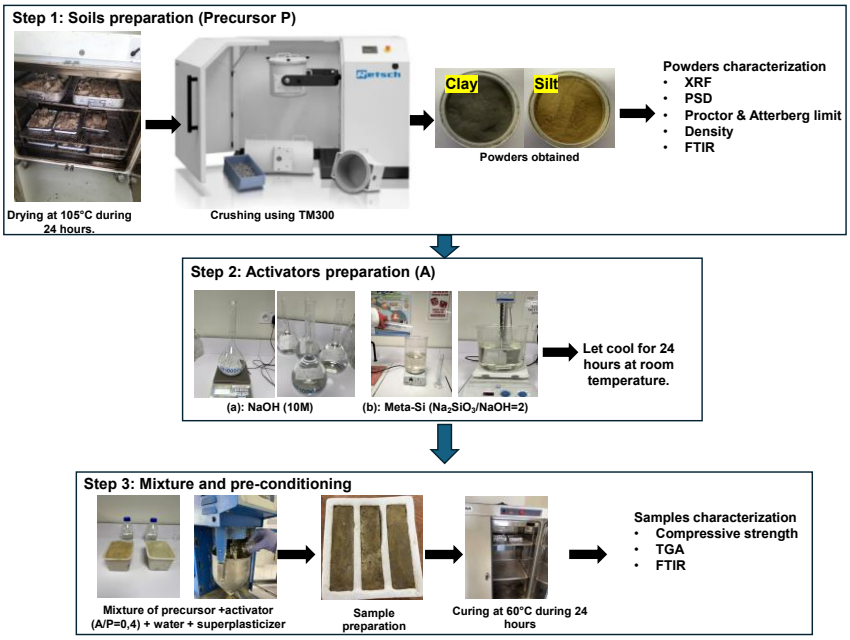

Fig.1. The approach taken during this study. Step 1: Preparation of precursors. Step 2: Preparation of the activation solution. Step 3: Mixing, curing, and preconditioning of samples.

2.1.2 Activators preparation

In this study, two activator solutions were used (Step 2, Fig.1). The first solution was prepared using only sodium hydroxide (NaOH), a caustic soda with 98% purity, supplied by Fisher®. It was obtained by dissolving NaOH pellets in distilled water until reaching a concentration of 10 M and then allowed to cool for 24 hours before use. The second solution noted Meta-Si consisted of a mixture of sodium silicate (Na_2SiO_3) and sodium hydroxide (NaOH), both supplied by Fisher®, with a mass ratio of Na_2SiO_3/NaOH = 2. The selection of these concentrations is based on the literature [9]. The composition of Na_2SiO_3 was 27.53% SiO_2, 11.47% Na_2O, and 61% H_2O by mass according to its technical datasheet. Sodium hydroxide was prepared separately by dissolving NaOH pellets of 98% purity in distilled water to achieve a 10 M concentration. The NaOH solution was then mixed with Na_2SiO_3 and allowed to cool for 24 hours before use.

2.2 Mixtures

The optimization of geopolymer formulations based on two types of soil was carried out. First, precursors consisting of 100% clay, 100% silt, and a binary mixture (50% clay + 50% silt) were investigated using two types of activators: (i) NaOH and (ii) Meta-Si. Three different precursor compositions and two alkaline treatments were used in six different mix designs (Table 1). The objective was to identify the most suitable activator/precursor (A/P) combination. It should be noted that the A/P mass ratio is taken equal to 0.4 according to the literature [9, 11, 12]. The precursors were introduced into the mixer and mixed at low speed for 30 seconds, followed by the addition of the activator while continuing to mix for 30 seconds. After that, the superplasticizer and water were added while mixing at low speed for 30 seconds. Then, mixing was stopped for 60 seconds in order to scrape the tank. The mixing was restarted at high speed for 60 minutes. The 4 × 4 × 16 cm³ samples were filled in two layers while vibrating using shock table equipment with 60 shocks for 60 seconds. The samples were wrapped in plastic film and then cured in an oven at 65°C for 24 hours (Step 3, Fig.1). Subsequently, the samples were conditioned in a climate-controlled room at 22 °C and 60% of relative humidity until the scheduled mechanical testing at 7 and 28 days. At 28 days, the microstructure of the samples was characterized by mineralogical analysis using thermogravimetric analysis (TGA), and chemical analysis using Fourier-transform infrared spectroscopy (FTIR).

Table 1. Mix design of earth-based geopolymer (grammes).

	Clay	Silt	NaOH	Meta-Si	Water	Superplasticizer
Clay_NaOH	1000	0	400	0	100	25
Clay_ Meta-Si	1000	0	0	400	100	25
Silt_NaOH	0	1000	400	0	100	25
Silt _ Meta-Si	0	1000	0	400	100	25
50%Clay+50%Silt_NaOH	500	500	400	0	100	25
50%Clay+50%Silt_ Meta-Si	500	500	0	400	100	25

3 Characterization

3.1 Physicochemical and physical analysis of raw materials

The particle size distribution (PSD) was determined using a dry laser particle analyzer (LS 13 320 XR, Beckman Coulter). The elemental composition was determined from X-ray fluorescence spectrometry (XRF) using a S2 Ranger (Bruker) instrument in pellet mode. The density of the raw powders was measured using a ULTRAPYC 5000 helium pycnometer, in compliance with ISO 12154. This method provided accurate values of true density, essential for assessing the compactness and reactivity of the materials. Proctor tests and Casagrande consistency limit tests were performed to

determine the optimum moisture content, maximum dry density, and Atterberg limits of the raw soils in accordance with the French standards NF P94-051 and NF P94-093, respectively. These parameters provided baseline information on soil workability and plasticity before alkali activation.

3.2 Compressive strength

The mechanical properties of specimens were investigated according to the French standard EN 196–1. The corresponding loading rates of the compressive strength were 2400 ± 200 N/s. The maximum load sustained by each specimen was recorded, and the compressive strength was calculated. Three specimens were tested, and the results were averaged to ensure accuracy and consistency.

3.3 Thermogravimetric analysis (TGA) and Fourier transform infrared spectroscopy (FTIR)

TGA was performed using a STA 449 F5 Jupiter instrument from NETZSCH to evaluate the physically and chemically bound water content as well as the different phases present in the tested powder samples. The experiments were conducted at a heating rate of 10 °C/min over a temperature range from 25 to 1100 °C. Approximately 100 mg of each sample was placed in an alumina crucible. The purge atmosphere was regulated with a nitrogen flow of 40 ml/min at the sample position and 70 ml/min in the furnace chamber, to avoid contamination of the sample environment.

FTIR analysis was conducted using a Spectrum Two FTIR spectrometer (PerkinElmer) to assess the impact of chemical treatments on the chemical composition of earth-based geopolymer. The analysis was performed in attenuated total reflection (ATR) mode, which eliminates the need for extensive sample preparation. The powders were gently pressed onto a diamond crystal, ensuring good contact for effective signal acquisition. The spectra were recorded over a wavenumber range of 4000 to 400 cm^{-1}, with a resolution of 4 cm^{-1} and an accumulation of 64 scans to enhance the signal-to-noise ratio. This method allowed for the detection of functional groups and chemical changes resulting from the geopolymerization of earth-based materials.

4 Results and discussion

4.1 Raw materials characterization

PSD result reveals very different particle size distributions between clay and silt, despite their prior sieving to 100 µm (Fig.2). For clay, the curve reveals a multimodal distribution, with several peaks between 2 µm and 20 µm and a significant fraction of fine particles smaller than 10 µm. The presence of submicron to micrometric particles results in a high specific surface area, which is favorable for reactivity during geopolymerization. These fines contribute to good dispersion and facilitate the dissolution of the aluminosilicates necessary for the formation of N-A-S-H and C-A-S-H gels. Comparatively, silt has a rather unimodal distribution, centered around 40-50 µm, with most

particles close to the cut-off limit (100 µm). This particle size indicates a coarser and less reactive texture, dominated by siliceous particles (mainly quartz), which explains its low contribution to the formation of reactive gels. Nevertheless, the predominance of these relatively large particles gives silt a granular skeleton role, improving the compactness and dimensional stability of mixtures.

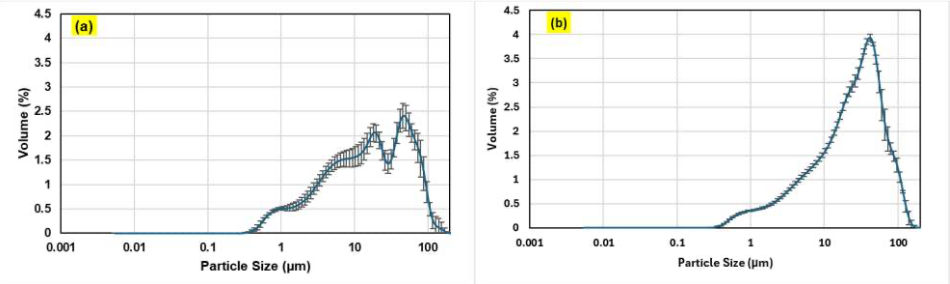

Fig.2. PSD of (a) clay and (b) silt precursors.

The elemental composition of the powders (Table 1) reveals significant differences between clay and silt. Clay is characterized by a larger Ca content than silt and a Si/Al ratio of 4.2, while silt is richer in Al and Fe, with a more moderate Ca/Si ratio of 40%. In fact, XRF analysis allows to quantify the chemical composition in terms of oxides. In order to determine the elemental composition, calculations based on molar mass were performed. From a geotechnical point of view, the Atterberg limits (ω_P and ω_L) reveal a significantly higher plasticity and water demand for clay than for silt. This high plasticity translates into larger water retention capacity. Also, the Proctor test ($\omega_{optimal}$) reveals a higher optimal water content for clay compared to silt. In addition, the absolute density measured by a helium pycnometer is 2.9 g/cm³ for clay and 2.7 g/cm³ for silt, indicating a composition richer in heavy phases in the former.

Table 2. Chemical composition, density, plastic and liquid limits of clay and silt precursors.

	Element composition (%)												
	Na_2O	MgO	Al_2O_3	SiO_2	P_2O_5	SO_3	K_2O	CaO	TiO_2	MnO	Fe_2O_3	Ca/Si	Si/Al
Clay	4.5	1.53	11.22	53.79	0.44	0.41	2.41	17.85	1.14	0.18	6.03	0.5	4.2
Silt	-	3.53	15.38	51.57	0.65	0.47	5.14	13	1.16	0.14	8.45	0.4	3.6

	Physical properties			
	ω_P(%)	ω_L(%)	$\omega_{optimal}$(%)	Density (g/cm³)
Clay	50	70	39.4	2.9
Silt	25	60	22.5	2.7

4.2 Compressive strength

The compressive strength at 7 and 28 days of the earth-based geopolymer highlights the combined effects of the precursor type (clay, silt, or a mixture of both) and the

activator composition (NaOH vs. Meta-Si). Clay exhibits a larger reactivity than silt, with Clay_NaOH reaching 2.6 MPa at 28 days compared to only 2.2 MPa for Silt_NaOH. Also, the use of soluble silicate Meta-Si significantly improved compression performances: Clay_Meta-Si achieved the largest compressive strength (5.1 MPa at 28 days), more than twice the one of Clay_NaOH, while Silt_Meta-Si increased to 3.7 MPa, confirming the positive role of additional Meta-Si in promoting N-A-S-H and C-A-S-H gel formation. Mixing precursors gave rise to intermediate compressive behavior, with 50%Clay+50%Silt_NaOH reaching 3.0 MPa and 50%Clay+50%Silt_Meta-Si improving to 4.6 MPa at 28 days, suggesting a beneficial synergy between the reactivity of clay and the contribution of silt to compaction. In general, the results demonstrate that clay is the most suitable precursor and that the Meta-Si activator significantly improves the development of mechanical strength.

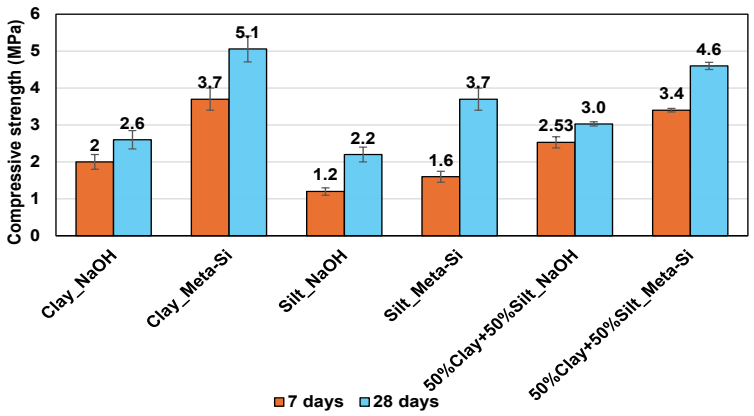

Fig.3. Compressive strength at 7 and 28 days of the geopolymers.

4.3 TGA and FTIR analysis

TGA results (Fig.4-a) highlight three main stages for all earth-based geopolymers. The first one is observed from about 65 °C to 300 °C and is attributed to the evaporation of bound water adsorbed on the surface of mineral particles and geopolymer gels (until 105°C) and to the dehydration of chemically bound water (between 105°C to 300°C) which is associated with the microstructure of geopolymer gels (N-A-S-H and C-A-S-H) [13], which is consistent with the FTIR analysis (Section 4.4). The second stage, between 400 and 550 ° C, is attributed to the decomposition of hydroxides and oxyhydroxides (goethite, vermiculite, illite, montmorillonite …) [14]. The final mass loss stage, between 600 and 800°C, is due mainly to the (i) decomposition of calcium carbonates ($CaCO_3$) during high-temperature calcination [15] and (ii) the dihydroxylation of kaolinite [14]. The mixtures based on Meta-Si activator, particularly those containing clay, exhibit greater mass losses in these temperature ranges, confirming a more extensive geopolymerization and significant formation of hydrated geopolymer products. Table 3 summarizes the mass losses of the geopolymer soils. The Clay_NaOH and Clay_Meta-Si geopolymers have a higher mass loss than Silt_NaOH and Silt_Meta-Si between 65 and 105°C, reflecting the amount of water absorbed and the higher water

demand of clay compared to silt. The two mixtures 50%Clay+50%Silt_NaOH and 50%Clay+50%Silt_Meta-Si show intermediate values due to the dilution effect of clay by silt. The second mass loss between 105-300°C reveals that Clay_Meta-Si presents the highest values, reflecting more N-A-S-H and C-A-S-H gels formed. Regarding the mass loss between 400 and 550°C, clay has more goethite, vermiculite, illite, and montmorillonite than silt. As for the final loss between 600 and 800°C, silt shows more mass loss, generally related to the decarbonation of $CaCO_3$.

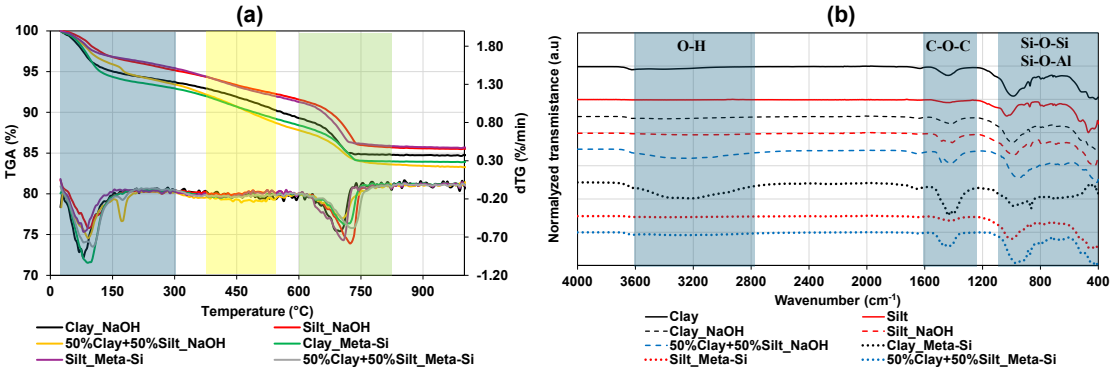

Fig.4. (a) TGA results of earth-based geopolymers and (b) FTIR result of clay, silt, and earth-based geopolymers.

Table 3. Mass losses ΔM (%) of the earth-based geopolymers studied at different temperature ranges (65-105°C, 105-300°C, 400-550°C, and 600-800°C).

	ΔM (%) 25-105°C	**ΔM (%) 105-300°C**	**ΔM (%) 400-550°C**	**ΔM (%) 600-800°C**
Clay_NaOH	3.9	2.3	4.0	4.5
Silt_NaOH	2.0	2.1	2.5	5.8
50%Clay+50%Silt_NaOH	3.0	2.8	3.1	4.1
Clay_Meta-Si	4.1	3.1	3.3	4.4
Silt_Meta-Si	2.5	2.1	2.9	5.3
50%Clay+50%Silt_Meta-Si	3.4	2.8	3.2	5.1

FTIR analysis reveals significant characteristic bands that provide a better understanding of their chemical structure (Fig.4-b). The broad band between 2800 and 3600 cm^{-1} corresponds to the stretching vibrations of the O–H hydroxyl groups, which is pronounced for Clay_Meta-Si and less significant to even absent for the other earth-based geopolymers, indicating the presence of physically adsorbed water as well as chemically bound water in the crystalline and amorphous phases, particularly within N-A-S-H and C-A-S-H gels [15, 16]. The band observed around 1500 cm^{-1} is attributable to the vibrations of carbonyl groups (O-C-O), indicating the possible presence of carbonates formed by partial carbonation of the materials or organic compounds bound to the surface [17]. The bands between 1100 cm^{-1} and 400 cm^{-1} correspond to the vibrations of the Si–O–Si and Si–O–Al bonds, characteristic of the aluminosilicate network of geopolymers. More specifically, the band around 1100 cm^{-1} reflects the asymmetric

stretching of Si–O–Si bonds, a sign of effective condensation of the polysilicate network, while the band near 400 cm^{-1} corresponds to the bending vibrations associated with these bonds. Mixtures based on Meta-Si activator show sharper and more intense bands in this region, indicating more advanced geopolymerization and better structural organization compared to samples activated only by NaOH [15]. These observations confirm the formation of complex hydrated gels and the coexistence of N-A-S-H and C-A-S-H gels in agreement with thermogravimetric data.

5 Conclusion

This study shows that clay is a significantly more reactive precursor than silt, while the mixed solution of Meta-Si seems to be the most effective activator, offering the best mechanical strength. Microstructural analyses confirm that the clay-Meta-Si combination promotes the formation of C-A-S-H and N-A-S-H gels which are responsible for improving the mechanical performance of Meta-Si-activated clay, which achieves a 96% increase compared to NaOH-activated clay and a 38% increase compared to silt activated by Meta-Si. These observations highlight the potential of developing geopolymer binders based on clay activated by soluble silicates. For the future, the addition such as fly ash or blast furnace slag appears to be a promising strategy for further enhancing performances while promoting industrial by-products in line with sustainability and the circular economy.

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