



FLY ASH-BASED GEOPOLYMER MORTARS USING SEAWATER AND SEA SAND

Ceramics-Silikáty 69 (1), 9-18 (2025)

THAI THANH TRUC*, **, [#]HUYNH NGOC MINH*, **, [#]DO QUANG MINH*, **

*Department of Silicate Materials, Faculty of Materials Technology, Ho Chi Minh City University of Technology (HCMUT), 268 Ly Thuong Kiet Street, District 10, Ho Chi Minh City, Vietnam **Vietnam National University Ho Chi Minh City, Linh Trung Ward, Thu Duc City, Ho Chi Minh City, Vietnam

[#]E-mail: hnminh@hcmut.edu.vn, mnh_doquang@hcmut.edu.vn

Submitted July 8, 2024, accepted September 24, 2024

Keywords: 35 ‰ salinity Seawater, Alkaline activator solution, Hydrothermal treatment, Drying treatment, Phillipsite-Na

Geopolymers are a commonly researched binder material due to their properties equivalent to Portland cement (PC). Unlike PC, the geopolymer can be used in seawater environments without the risk of sulfate and chloride attacks. Due to the scarcity of river sand and freshwater sources, it is necessary to use sea sand and seawater in construction, especially in coastal areas and islands. In this study, seawater with a salinity of 35 ‰ (representing the highest seawater salinity in Vietnam) and sea sand were used to produce fly ash-based geopolymer mortars with the NaOH concentration of the alkaline activator solution (AAS) ranging from 4 M to 12 M. The fly ash (FA) used was sourced from the Duyen Hai 1 thermal power plant, the natural sea sand was collected from the coast of Vung Tau in southern Vietnam. The mortar specimens were cured under different conditions, including drying and hydrothermal treatment at a pressure of about 9 atmospheres for 16 hours. The experimental and detected to the formation of phillipsite-Na crystals, commonly observed in hydrothermal-treated geopolymer materials. The FTIR and SEM analyses also identified the characteristic bonds and microstructure of the geopolymer in the samples. The 28-day compressive strength of the 12 M hydrothermal-treated sample reached up to 49.6 MPa, demonstrating the potential of a fly ash-based geopolymer mortar using seawater and sea sand. Additionally, it is noteworthy that the setting time of geopolymer mortars lasts up to 60 hours.

INTRODUCTION

In the 1970s, the term "geopolymer" was first introduced by Joseph Davidovits [1]. It refers to a solid material created through the reaction of an aluminosilicate powder with an alkaline activator solution (AAS) [2]. The geopolymer structure can be chained in the form of polysialate (-Si-O-Al-), polysialate-siloxo (-Si-O-Al-O-Si-O), polysialate-disiloxo (-Si-O-Al-O-Si-O-Si-O) or other sialate frameworks (silicon-oxo-aluminate) [3]. The sialate framework has a three-dimensional structure of $[SiO_4]^{4-}$ and $[AlO_4]^{5-}$ tetrahedrons linked together by shared oxygen atoms [4]. The alkali metal cations, such as Na⁺ and K⁺, provided by the activator solution balance the charge. In the case of a fly ash-based geopolymer, the fly ash (FA) initially dissolves into the AAS, forming an alkaline-silicate solution containing various silicate structural units (Q₀, Q₁, Q₂, and Q₃). The symbols Q₀, Q1, Q2, Q3, and Q4 denote the oxygen indices shared in the silicate unit structure. During the subsequent condensation process, the geopolymer material forms three-dimensional network structures (Q₃, Q₄), contributing to its hardening and structural integrity [5]. A geopolymer is considered an environmentally friendly material due to its utilisation of precursors derived from solid waste, such as FA and bottom ash, combined with its low energy requirements [6]. A life cycle assessment (LCA) study estimated that the unit CO_2 emission over a 100-year period for geopolymer concrete (GPC) is 285.1 v, while for Portland cement concrete (PCC) it is up to 454.6 kg·cm⁻³ [7]. The strength and durability of GPC are comparable to, and in some cases exceed, those of conventional concrete when produced under favourable conditions [8]. Furthermore, GPC exhibits a strong bond with steel reinforcing bars, surpassing that of PCC [9]. These characteristics make geopolymer materials a promising replacement for cement.

Another notable point is that GPC may exhibit greater durability in seawater compared to PCC. The principal inorganic constituents of seawater are Cl⁻, Na⁺, Mg^{2+} , SO_4^{2-} , Ca^{2+} etc. When seawater reacts with the hydrated Portland cement products, the PCC expands, cracks, and peels. Typically, the reaction of SO₄²⁻ ions with portlandite (Ca(OH)₂), monosulfate, and unreacted C_3A leads to the formation of gypsum (CaO·SO₃·2H₂O) and ettringite ((CaO)6(Al₂O₃)(SO₃)₃·32H₂O), resulting in an increased volume [10]. This volume expansion creates internal stress that breaks the cement concrete structure. Additionally, chloride ions penetrate the PCC, leading to portlandite $(Ca(OH)_2)$ leakage and a reduced compressive strength. Conversely, GPC, which does not use Portland cement and contains minimal CaO in class F fly ash (FFA), is not susceptible to sulfate attack. However, sulfate can still penetrate the GPC, potentially causing reinforcement corrosion. The corrosion of steel reinforcement in GPC has yet to be fully clarified. According to the Pourbaix diagram, the geopolymer paste is in the passivity region, allowing the formation of a passive layer that protects the reinforcement bar against corrosion [11]. This passive layer is iron-silicate, formed due to the interaction between the silicate and an alkaline environment [12].

Many studies have focused on the strength and durability of geopolymer binders under seawater conditions, especially in coastal and island countries where freshwater and river sand are scarce. GPC made from FA, sand, coarse aggregates with a sodium silicate solution, and seawater showed excellent performance, achieving a high compressive strength of 58 MPa after 180 days [13]. GPC combinations using sea sand and distilled water, or river sand and seawater, exhibited higher compressive and flexural strengths compared to those made with regular river sand and distilled water [14]. The compressive strength of geopolymer composites with seawater does not change significantly when the amount of NaCl utilised reaches up to 3 %. However, the compressive strength decreases when NaCl content exceeds 4.5 %. This reduction in the compressive strength at a high NaCl content may be due to increased salt crystallisation in the pores of the geopolymer [15].

During the geopolymerisation process, Na^+ ions are essential in neutralising the product charge at the oligomeric condensation stage and guiding the converting stage from the oligomeric to the high polymer state. If the concentration of Na^+ ions is too low, the process will be interrupted due to the failure to balance the charge. The polymer's degree of polymerisation will be low, causing a shortened polymerisation chain and a lack of structure. On the other hand, exceeding the Na^+ concentration will accelerate these stages, rapidly forming polymers that encapsulate the raw material, preventing the dissolution of the raw material and reducing the degree of hydration [16]. Furthermore, excess Na^+ ions can also combine with CO_2 in the atmosphere to form Na_2CO_3 on the surface of geopolymer material, interrupting the polymerisation reactions. The source of Na⁺ generation also needs to be distinguished. AAS is most commonly employed either as a sodium hydroxide solution (NaOH) alone or in combination with a sodium silicate solution [17]. The Na⁺ content in the geopolymer can be increased due to the increase in the content of the alkali activator. The compressive strength of the geopolymer binder increases with a high NaOH concentration of the AAS. However, when the NaOH concentration exceeds a certain level, it can cause a reduction in the compressive strength [17, 18]. Seawater and sea sand are not the AAS, so the amount of Na⁺ ions from these sources do not enhance the bond strength, but instead negatively impact the mechanical strength [19].

The durability of concrete in various environments is affected by its permeability, with denser concrete being more resistant to destructive agents entering and flowing through its pores [20]. The texture and structure of geopolymer materials also play a crucial role in their environmental durability. When GPC encounters water or another solution environment, Na^+ ions in the Si(Q₁) and Si(Q₂) structures can migrate and be leached out. However, in Si(Q₃) and Si(Q₄) structures, Na⁺ ions are fixed within the structural framework, making them less likely to leach out [5]. The hydrothermal treatment of geopolymers has been shown to form a higher proportion of Q4 structures compared to drying or microwave treatments [21]. The Q4 structure helps maintain the stability of the alumino-silicate network. In the hydrothermal-steam method, geopolymer materials are cured under high temperature and pressure conditions in an autoclave. Water molecules provide hydroxide ions (OH⁻) that permeate the structures of Na₂O, SiO₂, and Al₂O₃. Under these conditions, the AAS dissolve SiO₂ and Al₂O₃ more rapidly, enhancing the efficiency of the geopolymerisation reaction. The high amount of Al³⁺ replacing Si⁴⁺ in the structure results in the formation of many Q₄(4Al) structures.

The potential of fly ash-based geopolymer materials using seawater and sea sand as replacements for commonly used freshwater and river sand was investigated in this study. Due to the variability in the salinity and composition of seawater over time and between locations, a sea salt solution with a salinity of 35 ‰ was used to simulate typical seawater. The geopolymer samples were cured under different conditions, including drying at 60 °C and a hydrothermal treatment. The compressive strength after 7 and 28 days and the setting time of geopolymer mortar were determined at different NaOH concentrations of the AAS (4-12 M). The microstructure, mineral composition, and bonds formed in the geopolymer were detected using SEM, XRD, and FTIR to explain the strength of the geopolymer. The results demonstrated the influence of the microstructure (geo-mineral formation) on the compressive strength.

EXPERIMENTAL

Materials

FA was obtained from the Duyen Hai 1 Thermal Power Plant in Duyen Hai Town, Tra Vinh Province, Vietnam. The chemical composition of the FA is given in Table 1.

Table 1. Chemical composition of the FA raw material.

Component	SiO_2	Fe_2O_3	Al_2O_3	CaO	MgO	SO_3	K ₂ O	Na ₂ O	TiO ₂	Others	LOI*
wt. %	55.25	7.15	23.6	1.76	1.07	0.08	3.05	0.24	1.01	1.64	6.16
* LOI: loss o	n ignition.										

According to the ASTM C618 standard [22], Duyen Hai FA belongs to type F (CaO < 18 wt. %). The sea sand, originating from Vung Tau province, Vietnam, has a fineness modulus of 1.01 and a specific gravity of 2.698 g·cm⁻³. The AAS consisted of 35 ‰ salinity seawater and 98 % industrial-grade sodium hydroxide (NaOH) flakes. Sea salt (NaCl > 97 %) was used to prepare a 35 ‰ salinity seawater, representing the highest salinity of Vietnam's natural seawater.

Geopolymer sample preparation

To prepare the AASs, 35 ‰ salinity seawater was formed by mixing 35 g of sea salt with 1 litre of water. The AASs with five different concentrations of NaOH (4, 6, 8, 10, and 12 M) were prepared by dissolving NaOH flakes in the 35 ‰ salinity seawater, using the corresponding masses to achieve the appropriate molar concentrations. The mixture proportions of the mortar samples are presented in Table 2.

Table 2. Mixture proportions of the geopolymer samples.

Raw materials	FA	Sea Sand	AAS (4-12 M NaOH)
Weight for one mould casting (g)	675	800	240

First, FA and AASs (4-12 M) were low-speed-mixed in a mortar mixer (Matest E094, Italy) for 2 mins. The sea sand was added slowly while all the mixtures were mixed for an additional 6 min. Next, the mixed mortar was poured into a three-gang mould measuring $40 \times 40 \times 160$ mm. Each sample component was moulded in 2 batches. After 24 hours, the sample batches were de-moulded and treated under two conditions. A batch of samples was treated at 60 °C in a laboratory oven for 24 hours, with the samples wrapped in plastic film to prevent rapid water evaporation. The remaining sample batch was treated with hydrothermal steam in an autoclave at a pressure of about 9 atmospheres for 16 hours, consistent with the typical autoclaving conditions used in calcium silicate using a 300/15 kN compression and flexural testing machine (Matest E183N, Italy). The samples with the highest compressive strength in each group of treated geopolymers were selected for the analysis of the minerals and bonds in the material. These samples were ground into a powder with a particle size of less than 63 µm. X-ray diffraction (XRD) was performed using an Empyrean X-ray diffractometer (PANalytical, France) with Cu-Kal radiation ($\lambda = 1.540598$ Å) to identify the minerals formed. Based on the XRD pattern of the geopolymer samples, the phase ratios were calculated semi-quantitatively. Fourier transform infrared spectroscopy (FTIR) was used to study the association bonded in the geopolymer using a Nicolet 6700 FTIR spectrometer (Thermo, USA). The hydrothermal-treated sample with the highest compressive strength was also selected for the microstructure observation via a scanning electron microscope (SEM).

board manufacturing plants in Vietnam. Following these treatments, the samples were exposed to air at room

temperature. After curing, white hairs appeared on the

surface of the drying-treated samples, while this did not

samples was tested after 7 and 28 days of curing

The compressive strength of the geopolymer

occur in the hydrothermal-treated samples.

Moreover, the setting time of the geopolymer mortar was measured. Since there is no standard for geopolymer materials, the TCVN 6017:2015 standard (compliant with ISO 9597:2008) (Cements - Test methods - Determination of setting time and soundness) was applied to investigate the setting time of the geopolymer mortar using a Vicat apparatus. Specifically, the setting begins when the distance between the needle and the top of the base plate reaches (6 ± 3) mm, and it ends when the distance reaches 0.5 mm.

The schematic diagram summarises the implementation process, as shown in Figure 1.

RESULTS AND DISCUSSION

Setting time of the geopolymer

The data presented in Figure 2 illustrate the influence of the NaOH concentration of the AAS on the setting time of the geopolymer mortar. The setting times of the geopolymer samples using freshwater or seawater exhibited no significant difference, indicating that the ion content in the seawater does not notably affect the geopolymer setting process. In both cases, the initial and



Figure 1. The schematic diagram of the implementation process

final setting times increased as the NaOH concentration of the AAS increased. A higher NaOH concentration of the AAS facilitates the more effective dissolution of the FA in the early stages, but extends the solidification process of the geopolymer structure. Conversely, when the NaOH concentration of the AAS is low, the FA only partially dissolves, and the dissolution process halts when the AAS is no longer active. At this stage, the polycondensation process of dissolved FA becomes dominant, expediting the solidification stage of the geopolymer [23].

The initial and final setting times of the geopolymer were up to 16.5 hours and 60.5 hours, respectively (sample at 12 M). Meanwhile, the setting time of the Portland cement mortars ranges from approximately 1.5 to 7.25 hours [24]. Due to the extended setting time, geopolymer materials are suitable for applications requiring prolonged transportation and storage, which are not feasible with traditional cementitious materials. Furthermore, according to the results, the setting time of the geopolymer can also be easily modified by adjusting the concentration of the AAS, provided that the durability remains acceptable. As stated by publications, raw materials containing CaO can be added to geopolymer



Figure 2. . The effect of the NaOH concentration and types of mixed-water in the AAS on the setting times of the geopolymer mortars.

materials to adjust the appropriate setting time [10, 25]. With only a 10 % addition of blast furnace slag, the setting time of the geopolymer can be shortened to 5 hours [26]. Gradually replacing FFA with class C fly ash (CFA) can also accelerate the setting time; 70 % CFA can achieve a setting time faster than cement, ranging from 20.6 to 36.9 min [27]. Moreover, small percentages of chemicals such as CaCl₂, CaSO₄, Na₂SO₄, and sucrose can be added to flexibly adjust the setting time of the geopolymer. Shortening the setting time using these raw materials or chemicals does not compromise the strength of the geopolymer material, unlike cutting the concentration of the activator solution [28].

The compressive strength of the geopolymer

Figure 3 presents the compressive strength of the geopolymer samples as the NaOH concentration increases under different curing conditions. The results indicate that the compressive strength of the geopolymer samples increased as the NaOH concentration in the activator solution increased. This phenomenon is attributed to the higher NaOH concentration, which enhances the dissolution of aluminosilicates, thereby generating more Si and Al ions. Consequently, the geopolymerisation process is accelerated resulting in the formation of additional N-A-S-H gel.

Curing conditions play a crucial role in developing the mechanical properties of geopolymer materials. Since geopolymer materials take a long time to set, it is essential to cure them during solidification. Curing at this stage aids in enhancing the gelation process and transforming the gel into crystals in the geopolymer structure. Compared to the drying treatment, the hydrothermal treatment of the geopolymer samples demonstrated a more effective improvement in the strength of the geopolymer materials. The compressive strength of the hydrothermal-treated sample was more than double



Figure 3. The effect of the NaOH concentration and curing condition on the compressive strength of the geopolymer samples.

that of the drying-treated sample. Under hydrothermal conditions, the 28-day sample achieved the lowest compressive strength of 15.3 MPa at 4 M concentration and the highest of 49.6 MPa at 12 M concentration. For both curing conditions, the compressive strength of the samples continued to develop post-curing, as evidenced by the increase in the compressive strength at 28 days compared to 7 days.

Almost all the generated geopolymer samples met the requirements for unburnt bricks according to the ASTM C129 standard, which requires a minimum compressive strength of 3.45 MPa [29]. However, the drying-treated 4 M sample after 28 days only achieved a compressive strength of 2.5 MPa, which is insufficient for use as unburnt bricks.

Microstructure analysis XRD patterns

The XRD patterns of the geopolymer samples mixed with 12 M AAS under different curing conditions is shown in Figure 4.



Figure 4. The XRD patterns of the 28-day, 12 M hydrothermaltreated, drying-treated geopolymer samples, and FA powder.

Some typical peaks for quartz remained on the XRD diffraction, indicating that the dissolution and reaction of FA were not complete.

Specifically, the characteristic peaks of phillipsite-Na mineral (Na₆(Si₁₀Al₆)O₃₂·12H₂O, ICDD 73-1419) were detected in the hydrothermal-treated samples, while a hydro-sodalite mineral (Na₈Al₆Si₆O₂₄(OH)₂·2H₂O, ICDD 01-076-1639) was identified in the dryingtreated samples. The high compressive strength of the geopolymer samples is related to the formation of zeolite within the structure. Both phillipsite and hydrosodalite minerals are characteristic minerals for the geopolymer. Phillipsite is a type of zeolite with a singly connected 4-ring structure, while hydro-sodalite belongs to a singly connected 6-ring structure [30]. The 4-ring structure in zeolites generally leads to higher stability and mechanical strength due to the smaller ring size, resulting in a more rigid framework. This may explain why the compressive strength of the hydrothermaltreated sample was significantly higher than that of the drying-treated sample.

Furthermore, the proportion (wt. %) of the crystalline phase produced in the two groups of samples differed. Origin software was used to estimate the phase ratio in the samples. Accordingly, the proportion of the hydrogen-sodalite crystalline phase was only about 0.3 %, while the amorphous portion was 86.3 % in the drying-treated sample. For the hydrothermal-treated sample, the phillipsite-Na crystalline phase was about 4.3 %, and the amorphous phase was 78.5 %. The formation of more zeolite crystals, or the conversion of the gel to more crystals, enhances the compressive strength of the geopolymer material. However, it should also be noted that excessive formation of zeolite crystals can damage the pore structure of the geopolymer matrix, thereby reducing the compressive strength [31].

FTIR spectra

Figure 5 shows the FTIR spectra of the FA powder and geopolymer samples mixed with AAS NaOH 12 M under different curing conditions. On the FTIR spectra of the FA raw material, the typical bonds associated with quartz and mullite crystals were identified, as previously determined in the XRD analysis. Specifically, vibrational modes were observed at about 439-462 and 777 cm⁻¹, corresponding to the symmetric stretching vibrations of the Si-O-Si and O-Si-O bonds belonging to quartz minerals [32, 33]. The presence of the mullite phase was demonstrated by two bands corresponding to symmetric and asymmetric stretching vibrations of Al-O-Si type bonds related to octahedral aluminium (AIVI) at 550 and 875 cm⁻¹ [32]. In the FTIR spectra of the geopolymer



Figure 5. The FTIR spectra of 28-day, 12 M hydrothermaltreated, drying-treated geopolymer samples, and FA powder. Note: v: stretching vibration; δ : bending vibrations; s: symmetric; as: asymmetric.

material samples, the typical bands of quartz crystal remained prominent, while the mullite bands at 550 cm⁻¹ wave number had almost disappeared.

All the FTIR spectra exhibited absorption bands at about 3440 and 1645 cm⁻¹, corresponding to the O-H stretching and H-O-H bending vibrations of water molecules [34, 35]. The absorption frequency around the wavenumber of 1415 cm⁻¹ is typical for the C-O bond stretching vibration in carbonate, formed by the reaction of alkalis with atmospheric CO₂ [36, 37]. Larger bands on the FTIR spectrum of the drying-treated geopolymer sample indicate high water and carbonate content. "White hairs" appeared on the surface of these samples, but did not grow on the hydrothermal-treated samples. This phenomenon is attributed to the drying process conducted in a CO₂-rich environment rather than the hydrothermal steaming.

The band observed between 1200 and 800 cm⁻¹ is attributed to the asymmetric stretching vibration of the Si-O-T bonds (where T represents Si or Al) in the aluminosilicate structural networks [32]. The positional shift of this band towards lower frequencies in the geopolymer material sample indicates the occurrence of the geopolymerisation process. Specifically, the band at 1078 cm⁻¹ of the FA shifted to 1042 cm⁻¹ and 1001 cm⁻¹ in the hydrothermal-treated and drying-treated samples, respectively. This shift is associated with a higher substitution of $[SiO_4]^{4-}$ groups by $[AIO_4]^{3-}$ groups in the geopolymer gel [38]. Increased substitutions will result in weaker Al-O bonds, which vibrate at lower frequencies than the Si-O bond [32]. The extent of the wavenumber shift depends on the decrease in the Si/Al ratio, with an estimated reduction of approximately 19 cm⁻¹ per 0.1 atomic part of the Al ion substitution [39]. Furthermore, the shift to lower frequencies and the reduced intensity of the Si-O-T band also indicate the formation of crystalline phases from the geopolymer gels [40].

The positioning of the prominent bands in the geopolymer samples is correlated with the resulting $Si(Q_n)$ structure. The band positions for $Si(Q_n)$ units are approximately 1150-1200, 1100, 1000-1030, 900-920, and 820-876 cm⁻¹, for n = 4, 3, 2, 1, and 0, respectively [41]. Consequently, the band position of the hydrothermal-treated sample (1042 cm⁻¹) is determined to be associated with units Q₃ and Q₄, while the drying-treated sample is suggested to have a unit structure corresponding to Q_2 and Q_3 . This reinforces the advantages of the hydrothermal treatment over the drying treatment, resulting in the formation of a higher Q₄ structure [21]. Apart from this primary band, the geopolymer material samples also exhibited several small shoulders around the wavenumbers of 875 cm⁻¹ (coincident with mullite) and 1080-1760 cm⁻¹, indicating the existence of other structural units.

Along with the positional shift, the appearance of new bands in the geopolymer material samples revealed the formation of amorphous and zeolite phases. The generation of an amorphous polymer phase was predicted by discovering a band at about 695 cm⁻¹. This band belongs to the stretching vibration of the Si-O-Si(Al) bond, with its intensity indicating a high level of aluminium substitution in the Si-rich gel [32]. Some further bands associated with zeolite structure were found in the 600-800 cm⁻¹ wavenumber region. The type of zeolite crystals obtained depends on the chemical composition of the FA material, the curing temperature, and the concentration of the activation solution. In the drying-treated sample, the bands at 735 and 662 cm⁻¹ represent symmetric stretching vibration of external linkage and internal tetrahedra of T-O(T), respectively, characteristic of hydrosodalite [42, 30].

About the hydrothermal-treated sample, the band at about 610 cm⁻¹ was identified as the T-O-T bending vibration-external linkage for the zeolites in ring structure group 4, which includes phillipsite-Na [30]. The FTIR analysis confirmed the presence of the phillipsite-Na mineral, which is consistent with the XRD analysis. This aligns with other research on the curing conditions and the FA's composition. When subject to a hydrothermal treatment at temperatures above 140 °C for an extended period, FA with a SiO₂/Al₂O₃ ratio lower than 1.7 predominantly develops analcime crystals in the geopolymer material. Conversely, a SiO₂/Al₂O₃ ratio higher than 1.7 is expected to result in the formation of phillipsite or sodalite crystals [43]. Typically, phillipsite commonly combines with types of extra framework cations, such as Na⁺, K⁺, Ca²⁺, or Ba²⁺ [30]. The FA-based geopolymer samples in this study have a SiO₂/Al₂O₃ ratio of approximately 4; with the high NaOH concentration of the AAS, seawater, and sea sand, the considerable amount of Na⁺ enables the formation of phillipsite with a Na structural framework in the geopolymer material, surpassing other frameworks.

SEM images

With the advantages in the properties of the hydrothermal-treated geopolymer samples presented in previous analyses, the highest compressive strength (12 M) in this group was selected for the SEM analysis. The SEM images of FA and 28-day, 12 M hydrothermal-treated geopolymer samples at different magnifications are shown in Figure 6.

In the first image, most FA particles look spherical (Figure 6a), with an average size of about 28.66 μ m (result obtained from the Laser particle size distribution analysis). The SEM images of the geopolymer material indicated that not all the FA particles were wholly dissolved. There were still unreacted or partially reacted FA particles (Figure 6b). The existence of unreacted FA particles suggested that further increasing the AAS concentration would enhance the geopolymerisation process. Gels of undetermined shape were formed within the structure, indicating that geopolymerisation had

occurred (Figure 6c). It can also be observed that there were needle crystals around some FA particles (Figure 6d). The SEM images of the geopolymer products show that the continuous geopolymer matrix resulting from the geopolymerisation of fly ash and ASS has bonded unreacted or partially reacted fly ash particles, tiny needle-shaped crystals (phillipsite-Na) and gel-like zone, which aids adhesion and develops strength in the resulting material.

CONCLUSIONS

By using sea sand and 35 ‰ salinity seawater to create an FA-based geopolymer, the setting time of this material was extremely extended to 60.5 hours. This prolonged setting time significantly impacts construction schedules and the usability of geopolymer materials in practical applications. Compared to the drying-treatment method, geopolymer samples that underwent hydrothermal treatment showed impressive compressive strength, and no "white hair" phenomenon occurred on the surface, highlighting the potential for producing high-quality materials through this alternative approach. The XRD, FTIR, and SEM analyses also revealed phase bonding properties comparable to those of freshwater-based geopolymer materials. Using sea sand and seawater instead of river sand and fresh water, most of the research samples' compressive strengths met the standard of unburned bricks as per ASTM C129, particularly reaching approximately 50 MPa when treated using the hydrothermal method. Additionally, the inherently low CO₂ emissions of geopolymer materials demonstrate their environmental potential.

Acknowledgments

This research is funded by Vietnam National University Ho Chi Minh City under grant number B2024-20-27. We acknowledge the Ho Chi Minh City University of Technology (HCMUT), VNU-HCM for supporting this study.

REFERENCES

- Perná I., Hanzlícek T., Zaloudková M. (2020): Microscopic study of the concrete / geopolymer coating interface. *Ceramics - Silikaty*, 64, 68-74. doi:10.13168/cs.2019.0050
- Provis J. L., Van Deventer J.S.J. (Eds). (2009). *Geopolymers-Structure, Processing, Properties and Industrial Applications.* 1st ed. Woodhead Publishing Limited.
- 3. Davidovits J. (2005): Geopolymer chemistry and sustainable Development. The Poly(sialate) terminology : a very useful and simple model for the promot ion and understanding of green-chemistry. *Geopolymer Institute*, 1, 9-17.



Figure 6. The SEM images of the (a) FA powder and 28-day, 12 M hydrothermal-treated geopolymer sample at magnifications of (b) \times 1000, (c) \times 2000, and (d) \times 10,000.

- 4. Mostafa N.Y. (2010): Investigating the Possibility of Utilizing Low. *Ceramics Silikaty*, 54, 160-168.
- Davidovits J. (2017): Geopolymers: Ceramic-like inorganic polymers. *Journal of Ceramic Science and Technology*, 8, 335-350. doi:10.4416/JCST2017-00038
- Soe P.S., Sornlar W., Wannagon A., Chaysuwan D. (2023): Mechanical and thermal properties of bottom ashbased porous geopolymer as thermal insulation material for construction. *Journal of Material Cycles and Waste Management, 25*, 2964-2975. doi:10.1007/s10163-023-01732-6
- Garces J.I.T., Dollente I.J., Beltran A.B., Tan R.R., Promentilla M.A.B. (2021): Life cycle assessment of selfhealing geopolymer concrete. *Cleaner Engineering and Technology, 4.* doi:10.1016/j.clet.2021.100147
- Varuthaiya M., Palanisamy C., Sivakumar V., Pushpanathan G. (2022): Concrete with sisal fibered geopolymer: a behavioral study. *Journal of Ceramic Processing Research*, 23, 912-918. doi:10.36410/jcpr.2022.23.6.912
- Cui Y., Qu S., Bao J., Zhang P. (2022): Bond Performance of Steel Bar and Fly Ash-Based Geopolymer Concrete in Beam End Tests. *Polymers*, 14. doi:10.3390/polym14102012
- 10. Bassuoni M.T., Nehdi M.L. (2009): Durability of self-

consolidating concrete to sulfate attack under combined cyclic environments and flexural loading. *Cement and Concrete Research, 39,* 206-226. doi:10.1016/j. cemconres.2008.12.003

- Zainal F.F., Amli S.F.M., Hussin K., Rahmat A., Abdullah M.M.A.B. (2017): Corrosion Studies of Fly Ash and Fly Ash-Slag Based Geopolymer. IOP Conference Series: *Materials Science and Engineering*, 209. doi:10.1088/1757-899X/209/1/012026
- Pasupathy K., Singh Cheema D., Sanjayan J. (2021): Durability performance of fly ash-based geopolymer concrete buried in saline environment for 10 years. *Construction and Building Materials, 281.* doi:10.1016/j. conbuildmat.2021.122596
- Do Q.M., Bui T.H., Nguyen H.T. (2019): Effects of seawater content in alkaline activators to engineering properties of fly ash-based geopolymer concrete. *Solid State Phenomena*, 296, 105-111. doi:10.4028/www.scientific.net/SSP.296.105
- Manjunath M.K., Shivasidda Reddy S., Rashmi K., Shreedhar K.P., Dhanush H.R. (2021): Geopolymer Pavers by using Sea Sand and Sea Water. *International Research Journal of Engineering and Technology (IRJET)*, 8, 1734-1741.

- Mani S., Pradhan B. (2020): Investigation on effect of fly ash content on strength and microstructure of geopolymer concrete in chloride-rich environment. *Materials Today: Proceedings*, 32, 865-870. doi:10.1016/j. matpr.2020.04.216
- Zhao Y., Wang H., He Y., Yang L., Wu H. (2022): Effect of Na⁺ on hydration degree of alkali activated metakaolin polymer. *Magazine of Civil Engineering*, 112. doi:10.34910/ MCE.112.8
- Torgal F.P., Gomes J.C., Jalali S. (2008): Alkali-activated binders: A review. Part 1. Historical background, terminology, reaction mechanisms and hydration products. *Construction and Building Materials*, 22, 1305-1314. doi:10.1016/j.conbuildmat.2007.10.015
- Shukla A., Chaurasia A.K., Mumtaz Y., Pandey G. (2020): Effect of Sodium Oxide on Physical and Mechanical properties of Fly-Ash based geopolymer composites. *Indian Journal of Science and Technology*, 13, 3994-4002. doi:10.17485/ijst/v13i38.1663
- Rashad A.M., Gharieb M. (2021): An investigation on the effect of sea sand on the properties of fly ash geopolymer mortars. Innovative Infrastructure Solutions, 6, 1-9. doi:10.1007/s41062-020-00421-9
- Reddy D. V, Edouard J.B., Sobhan K. (2013): Durability of Fly Ash–Based Geopolymer Structural Concrete in the Marine Environment. *Journal of Materials in Civil Engineering*, 25, 781-787. doi:10.1061/(asce)mt.1943-5533.0000632
- Thang N.H., Thach B.K., Minh D.Q. (2021): Influence of Curing Regimes on Engineering and Microstructural Properties of Geopolymer-Based Materials from Water Treatment Residue and Fly Ash. *International Journal of Technology*, 12, 700-710. doi:10.14716/ijtech.v12i4.4626
- 22. Suraneni P., Burris L., Shearer C.R., Hooton R.D. (2021): ASTM C618 fly ash specification: Comparison with other specifications, shortcomings, and solutions. *ACI Materials Journal*, 118, 157-167. doi:10.14359/51725994
- 23. Yomthong K., Wattanasiriwech D., Aungkavattana P., Wattanasiriwech S. (2021): Effect of NaOH Concentration and Curing Regimes on Compressive Strength of Fly Ash-Based Geopolymer. *Materials Today: Proceedings, 43*, 2647-2654. doi:10.1016/j.matpr.2020.04.630
- Verma N.K., Rao M.C., Kumar S. (2022): Effect of Curing Regime on Compressive Strength of Geopolymer Concrete. *IOP Conference Series: Earth and Environmental Science*, 982. doi:10.1088/1755-1315/982/1/012031
- 25. Jiao Z., Li X., Yu Q., Yao Q., Guan X., Hu P. (2023): Chloride resistance of class C/class F fly ash-based geopolymer mortars with different strength grades. *Case Studies in Construction Materials*, 18. doi:10.1016/j. cscm.2022.e01811
- 26. Prasanna K.M., Sharath B.P., Choukade H., Shivaprasad K.N., Das B.B., Mahesh G. (2023): Research on Setting Time, Compressive Strength and Microstructure of Fly Ash-Based Geopolymer Mixture Containing Slag. *Iranian Journal of Science and Technology Transactions of Civil Engineering*, 47, 1503-1517. doi:10.1007/s40996-022-01010-9
- Jiao Z., Li X., Yu Q., Yao Q., Hu P. (2023): Sulfate resistance of class C/class F fly ash geopolymers. Journal of Materials Research and Technology, 23, 1767-1780. doi:10.1016/j. jmrt.2023.01.131

- Rattanasak U., Pankhet K., Chindaprasirt P. (2011): Effect of chemical admixtures on properties of high-calcium fly ash geopolymer. *International Journal of Minerals, Metallurgy and Materials, 18*, 364-369. doi:10.1007/ s12613-011-0448-3
- 29. C129-01 A. (2003): Standard Specification for Nonloadbearing Concrete Masonry Units. *Annual Book of ASTM Standards*, ASTM International, West Conshohocken, PA.
- Ma B., Lothenbach B. (2020): Synthesis, characterization, and thermodynamic study of selected Na-based zeolites. *Cement and Concrete Research*, 135. doi:10.1016/j. cemconres.2020.106111
- 31. Zheng Z., Ma X., Zhang Z., Li Y. (2019): In-situ transition of amorphous gels to Na-P1 zeolite in geopolymer: Mechanical and adsorption properties. *Construction and Building Materials*, 202, 851-860. doi:10.1016/j. conbuildmat.2019.01.067
- 32. Villaquirán-Caicedo M.A., Mejía-de Gutiérrez R. (2015): Synthesis of ternary geopolymers based on metakaolin, boiler slag and rice husk ash. *DYNA (Colombia)*, 82, 104-110. doi:10.15446/dyna.v82n194.46352
- Patcharin W., Sriamporn K., Kanokkan A. (2012): Utilization biomass from bagasse ash for phillipsite zeolite synthesis. *Advanced Materials Research*, 383-390, 4038-4042. doi:10.4028/www.scientific.net/AMR.383-390.4038
- 34. Yu X., Ren C., Xu W., Xu L., Tian Q. (2024): Effect of kaolin pretreatment on the structure and properties of metakaolin phosphate-based geopolymers. *Journal of Ceramic Processing Research*, 25, 138-143. doi:10.36410/ jcpr.2024.25.1.138
- 35. Vlček J., Drongová L., Topinková M., et al. (2014): Identification of phase composition of binders from alkaliactivated mixtures of granulated blast furnace slag and fly ash. *Ceramics - Silikaty*, 58, 79-88.
- 36. Demir S., Yaşın S. (2022): Geopolymerisation of uncalcined kaolin by the novel heat curing method. *Journal* of Ceramic Processing Research, 23, 41-47. doi:10.36410/ jcpr.2022.23.1.41
- Heikal M., Ibrahim S.M. (2015): Characteristics and durability of alkali activated slag-microsilica pastes subjected to sulphate and chloride ions attack. *Ceramics -Silikaty*, 59, 81-89.
- Asadi M., Naghizadeh R., Nemati A., Arzani K., Nassiri R. (2012): Synthesis and microstructural properties of a geopolymer cement with high silica materials. *Journal of Ceramic Processing Research*, 13, 425-428.
- Szymanski H. A. (1974). Infrared Structural Studies of Zeolite Frameworks. In: Flanigen M, Khatami H. (Ed.): *Molecular Sieve Zeolites-I*. Advances in Chemistry. pp. 380-404. doi:10.1021/ba-1971-0101
- Onutai S., Sato J., Osugi T. (2023): Possible pathway of zeolite formation through alkali activation chemistry of metakaolin for geopolymer–zeolite composite materials: ATR-FTIR study. *Journal of Solid State Chemistry*, 319. doi:10.1016/j.jssc.2022.123808
- El Felss N., Gharzouni A., Colas M., Cornette J., Sobrados I., Rossignol S. (2020): Structural study of the effect of mineral additives on the transparency, stability, and aging of silicate gels. *Journal of Sol-Gel Science and Technology*, 96, 265-275. doi:10.1007/s10971-020-05385-x
- 42. Sasnauskas V., Palubinskaite D. (2005): The synthesis of hydrosodalite and its use in mortar technology. *Materials*

Science Poland, 23, 793-801.

43. Zgureva D., Stoyanova V., Shoumkova A., Boycheva S., Avdeev G. (2020): Quasi natural approach for crystallization of zeolites from different fly ashes and their application as adsorbent media for malachite green removal from polluted waters. *Crystals, 10*, 1-16. doi:10.3390/cryst10111064