



Mechanical and microstructural properties of high calcium fly ash one-part geopolymer cement made with granular activator



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ABSTRACT

In this present experimental study, geopolymer cement is developed using high calcium fly ash and used in the production of one-part alkali-activated binders. At 8–16 percent of the total precursor materials, the HCFA was activated with anhydrous sodium metasilicate powder and cured in ambient condition. Five mixtures of one-part geopolymer paste were intended at a steady w/b proportion. Density, flowability, setting time, compressive strength, splitting tensile strength and molar ratio impact were envisaged. It was observed that the setting time of the designed one-part geopolymer paste decreases with higher activator content. The experimental findings showed that the resistance of one-part geopolymer cement paste increases with comparatively greater activator content. However, raising the granular activator beyond 12 percent by fly ash weight decreases the strength and workability of the established one-part geopolymer cement. The optimum mix by weight of the fly ash was discovered to be 12 percent (i.e. 6 percent Na₂O). At 28 days of curing, one-part alkali-activated paste recorded the greatest compressive strength of almost 50 MPa. The density of the one-part geopolymer paste is nearly the same regardless of the mixes. Microstructural assessment by FESEM, FTIR and XRD has shown that the established geopolymer paste includes quartz, pyrrhotite, aluminosilicate sodium and hydrate gels of calcium aluminosilicate. Based on the experimental information acquired, it can be deduced that the strength growth of one-part geopolymer cement is similar to that of Portland cement.

1. Introduction

The evolution of low-carbon binders is realized as one of the possible solutions to reduce the environmental impact associated with Portland cement binders [1, 2, 3]. Aluminosilicates materials react very slowly with water but react under hydrolysis and condensation reactions in an alkaline medium to form inorganic polymers capable of resisting loads. The binding property resulting from the amorphous aluminosilicate gels and the reactivity of the source materials is strongly affected by the structure of the aluminosilicates, and various research has been committed to this problem [4]. Depending on the calcium composition of the source materials, the synthesis of aluminosilicates with alkalis can be categorized into two forms: low and high calcium aluminosilicates. Aluminosilicates materials containing low metal content result in the formation of a metallic element aluminosilicate hydrate (N-A-S-H) product, whereas high metal content causes the formation of calcium aluminosilicate hydrate (C-A-S-H) almost like Portland cement C-S-H. However, the former contains essential replacement in connecting

tetrahedral structure, offering extended chains that can be cross-linked based on the activator used [2, 5]. Geopolymer binders are environmentally-friendly material used as a substitute for ordinary Portland cement (OPC) binder. Geopolymer's polymerization mechanism is an intensely rapid chemical reaction on silica-alumina minerals in an alkaline environment that yields a 3-dimensional polymeric sequence and ring structure consisting of Si–O–Al–O bonds. Geopolymer concrete production does not require the use of any OPC, but the binder is produced by the reaction of an aluminosilicate ingredient with strong alkaline liquids. It is evident that, in addition to depleting existing resources in Portland Cement production, an enormous amount of carbon dioxide has been emitted into the surrounding atmosphere. A substitute green material is required to mitigate these effects. It is therefore essential to use substitute ingredients for the development of environmentally favorable concrete [6]. Collectively, cement gel geopolymer binds aggregates and unreacted material to yield geopolymer concrete [7, 8]. It is worth mentioning that the production of geopolymer cement releases 80 to 90 percent less CO₂ (greenhouse effect gas) compared to

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the production of Portland cement [9].

Notwithstanding the reality that the geopolymer system has been developed since the 1970s [7,10,11] and has numerous beneficial attributes, yet it is not widely used in concrete compared to ordinary Portland cement. Due to its restrictions in terms of slow setting, high porosity, elevated temperature curing and sluggish strength growth, comprehensive use of low calcium fly-ash geopolymers is hampered. Curing geopolymer concrete at ambient temperature led to slow growth of strength at the initial stage while curing at relatively high temperatures resulted in a better development of strength [12]. Haruna et al. [13], reported that outdoor cured geopolymer concrete has developed reasonable strength, but high molarity degrades the strength properties under the same condition. Improving the stated properties is vital for maximizing the use of geopolymers in construction as a substitute for the energy-demanding OPC binder. Efforts have been made to produce one-part mix geopolymer [5, 14, 15], their findings focused on producing commercially and environmentally suitable one-part binders.

One-part geopolymer binders are known as alkaline cement, they are a new geopolymer production approach that has been recently developed with the aim of reducing complications in dealing with silicate solution activated geopolymers, produced by mixing aluminosilicate source materials with solid activators [14, 16]. Unlike normal geopolymer binders where solutions are used to trigger the activation process, the activator remains in dry powdered form in the one-part binder and the reaction starts as water is poured into the binder resembling ordinary Portland cement [14, 17, 18]. This method helps to avoid using corrosive and viscous solutions for large-scale geopolymer concrete production and encourages the commercial suitability of geopolymer binder, this concept is called just adding water. There are many attempts to synthesize aluminosilicate materials together with alkaline solutions at higher temperatures to produce one-part binders [14, 15, 19, 20, 21, 22]. As reported by Provis [18] the activating agent in a one-part geopolymer binder can be any material that supplies alkali cations, amplifies the pH level of the combination, and accelerates the dispersal of silica and alumina into the system. Different types of activators were used in the formation of one-part geopolymers consisting of sodium aluminate, sodium silicate, sodium sulfate, sodium carbonate, sodium silicate pentahydrate and many more. Koloušek et al. [19] developed one-part geopolymer systems by burning kaolinite with powdered hydroxides. After 7 days of curing, which was inferior to conventional geopolymer systems, the strength gain was only 1 MPa. Other interesting findings [16] also used red mud and silica fume to produce one-part geopolymer and realized a compressive strength of about 32 MPa at 28 days of ambient curing which proves to be a vital improvement to the findings reported by [19]. In addition, one part of the geopolymer binder for ambient healing was produced by [23, 24] using fly ash and slag as the primary ingredients and acting with sodium silicate and sodium hydroxide powder. The binder produced with 100 percent slag gained a compressive strength of about 50 MPa, while that of 100 percent fly ash yields 9.45 MPa strength after 28 days of healing at room temperature.

One-part geopolymers are reported to have low green gas emissions compared to conventional geopolymers as the polymerization process constitutes a small portion of the matrix [25, 26, 27]. They also revealed that the one-part geopolymers binding phases are similar to the two-part geopolymers. Alkali activation process includes concentrated fluid arrangements that are harmful, sticky, making it difficult to deal with and not easy to use. Therefore, the improvement of alleged one-part geopolymers or "simply include water" geopolymers may have more prominent potential than traditional two-part geopolymers, particularly in cast-in-situ applications [26, 27]. It is interesting to note that one-part geopolymers have proved cost-effective than the two-part counterpart geopolymers [28], but it is still costly compared to the one-part geopolymers [28].

Recent studies have used anhydrous sodium metasilicate powder to synthesize one-part geopolymers from fly ash blended slag [14, 20, 29]. High calcium fly ash has rarely been used in the production of one-part

geopolymer binders. Therefore, this research may be of significant interest in the production of one-part geopolymers based on fly ash. In addition, there are limited studies of the fresh, mechanical and microstructural properties of high calcium fly ash one-part geopolymer binders synthesized using anhydrous sodium metasilicate powder. In this study, one-part geopolymer pastes are prepared and cured at ambient temperature. Flowability, setting time, strength and effect of one-part geopolymer paste molar ratios at different activator dosage were examined to determine the fundamental properties of one-part geopolymer paste cured in the ambient. The optimum mixture of one-part geopolymer cement achieved could be a substitute for Portland cement and further use by adding water similar to OPC to produce geopolymer concrete and mortar. The idea of using C-class fly ash in geopolymer manufacturing would be helpful for this by-product's long-term applications.

2. Materials and methods

2.1. Materials

High calcium fly ash was used as the primary binder in this investigation. The chemical components of the high calcium fly ash used were detected by X-Ray fluorescence (XRF) and presented in Table 1. The fly ash was acquired from the Manjung Power Plant in Perak, Malaysia as a source of aluminosilicate materials for the production of one-part geopolymer binder. Field emission scanning electron microscope (FESEM) examined the morphology of fly ash and shown by the micrograph in Fig. 1. Based on the FESEM micrograph, all fly ash particles appeared to be spherical in different sizes, allowing it to mix freely in mixtures. It is well recognized that the percentage of CaO used for HCFA was greater than 10 percent. Additionally, the total sum of SiO₂, Al₂O₃ and Fe₂O₃ is less than 70 percent, hence the fly ash in this study meets the requirement of C-class fly ash as per ASTM 618–10 [30].

Granular sodium metasilicate anhydrous (50 percent Na₂O, 46 percent SiO₂ and 4 percent H₂O) and modulus ratio of 0.92 was used as a solid activator to activate fly ash. The use of granular alkaline activators in geopolymer systems is easier and less time consuming than the frequently used alkaline solutions as there is no need to prepare NaOH solution before mixing. The granular activator was used by weight of the primary ingredients as 8 percent–16 percent. Portray Sdn Bhd located in Selangor, Malaysia, provided the granular sodium metasilicate anhydrous used in this work.

2.2. Mixing procedure and test methods

One-part geopolymer paste was made in accordance with the standard practice of ASTM C305-14 [31]. Five different levels of geopolymer pastes were produced by mixing fly ash with anhydrous sodium metasilicate at five different levels as shown in Table 2. HCFA and granular anhydrous sodium metasilicate were thoroughly mixed in a small Hobart

Table 1
Fly ash chemical composition.

Chemical oxide	Percentages (%)
Al ₂ O ₃	14.9
SiO ₂	37.3
Fe ₂ O ₃	16.5
CaO	17.1
MgO	3.72
P ₂ O ₅	2.59
SO ₃	2.56
Na ₂ O	1.74
K ₂ O	1.66
TiO ₂	1.07
Others	0.69
LOI	0.17
Blaine fineness (m ² /Kg)	385
Specific gravity	2.35

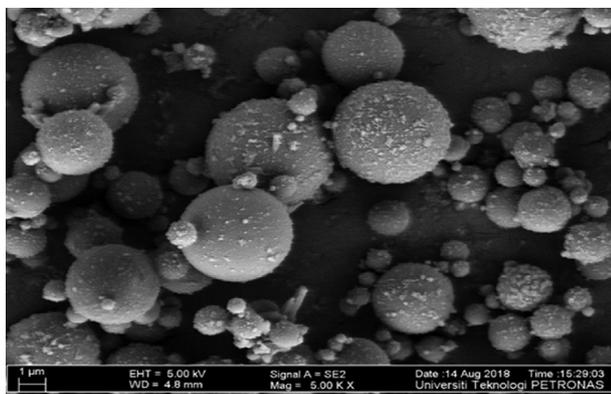


Fig. 1. FESEM micrograph of high calcium fly ash.

mixer for about 2 min. Potable tap water was gently poured into the dry mix and mixed for 3 more minutes until it becomes homogeneous and consistent. The details of the proportions of the mixture were presented in Table 2. Mixing was performed at approximately 25 °C in a laboratory. Immediately after mixing, the fresh geopolymer paste was poured into 50/50/50 mm cubic moulds. After 24 h, the specimens were dislodged from the moulds and cured at ambient temperature (25–27 °C). The samples and moulds were left in the laboratory until the 3, 7, and 28 day test period. The percentage of granular sodium metasilicate anhydrous used is the parameter considered in this investigation.

The flowability of the fresh one-part geopolymer paste was measured using a flow table. The flow table test was used to evaluate the fresh properties of the one-part geopolymer paste as it is easy to perform and uses simple handling equipment. The flow table consists of a complete cast immovable iron frame and a circular static table top. Other flow table information is mounting and lubrication as mentioned in ASTM C230/C230M-14 [32]. Mini slump test was used to determine paste flowability. Each mixture was twice tried. The mini slump test procedure was performed in accordance with [14, 33, 34]. The conical mold was used to cast the one-part geopolymer paste while the caliper was used to measure the mortar diameter. The fresh one-part geopolymer pastes were designed to weigh the binder at 8 percent, 10 percent, 12 percent, 14 percent and 16 percent of granular anhydrous sodium metasilicate. Setting time for one-part geopolymer pastes was performed as per ASTM C807 [35] to determine the influence of sodium metasilicate activator content on one-part geopolymer pastes. Vicat apparatus was used to conduct the one-part geopolymer cement setting time behaviour. One-part geopolymer pastes were placed in an exceedingly conic Vicat equipment mould. Vicat needle infiltration of approximately 1 mm in diameter on the geopolymer paste was recorded at orderly time intervals for initial setting. The initial stiffening time of the one-part geopolymer is recorded as the period during which the needle does not penetrate 3 mm–5 mm from the bottom of the conical mould. The final setting time of the paste was also observed once the needle failed to make any impression on the surface of the paste. The test was performed at 25–27 °C laboratory temperature.

The cubes specimens are weighed at the test date to obtain their respective densities. The compressive strength of the specimens was

Table 2
Mix proportion of one-part geopolymer paste.

Mix. No.	Fly ash	Na ₂ SiO ₃ Anhydrous (%)	water [g]	w/s ratio
N1	1	0.08	225	0.25
N2	1	0.10	225	0.25
N3	1	0.12	225	0.25
N4	1	0.14	225	0.25
N5	1	0.16	225	0.25

evaluated using ASTM C109/109M [36] for 3, 7 and 28 days. At each testing age, three sets of the specimens were used to conduct the compressive strength test. Similarly, the tensile strength of the geopolymer paste was split in accordance with BSEN12390-6 [37].

3. Results and discussion

3.1. Flow characteristics

The flowability of the mixtures decreases with an increase in sodium oxide content in the paste. This is attributed to the Na₂O's higher molecular mass, which immediately binds the fly ash particles as water was added to the mix. Basically, the flow resistance of the mix worsened at a higher percentage of Na₂O and therefore reduces the flowability as shown in Fig. 2(a) and subsequently result in highly viscous mixtures [24, 38]. The behavior of the one-part geopolymer paste is found to be thixotropic in nature as shown in Fig. 2(b) When stirred, the gels become more workable and less viscous. It is predicted that the powder-activated geopolymer has a moderate rate of alkali release into the binder resulting in a steady initial reaction, making the paste flowable for a relatively longer time [17]. It is worth noting that for one-part geopolymer pastes, increasing the quantity of the solid activator leads to a decrease in flowability as shown in Fig. 3, this finding is in line with the work of [14]. Nevertheless, improving water content enhanced workability but leads to a decline in compressive strength. The results shown in Fig. 3 show that sodium metasilicate content in the mixes relatively influenced the flowability of one-part geopolymer paste. The flowability of the developed pastes was measured in terms of relative slump values, a relative slump value of 6.13 was recorded for N1 samples. The relative slump values were obtained in a decreasing pattern for N1 by 2.5 percent, 4.9 percent, 6.7 percent, and 7.3 percent for N2, N3, N4, and N5 respectively. It is interesting to note that the flowability of N4 and N5 remains almost constant at high activator content. It should be noted that the relative slump values depend on the mini-slump test without any compaction or external pressure. Subsequently, there was no problem with the casting and compaction of the samples and these results are in good agreement with those published by [14, 36, 39].

3.2. Setting time of one-part geopolymer paste

With the help of the Vicat apparatus, the setting time behavior of one-part geopolymer paste was performed. For each test, as suggested by [40], two sets of readings were recorded for initial and final setting time, and the average was reported. The initial setting time was recorded as the moment from which water is added to the dry mixture at which time the needle resists penetration from the bottom plate to a depth of 3 mm–5 mm, while the final setting time is recorded as the time from which water is added to the dry mixture at which time the needle does not affect the surface of the one-part geopolymer paste. The results shown in Fig. 4 show that sodium metasilicate content in the mix considerably influenced the setting time of one-part geopolymer paste. One-part geopolymer pastes quickly hardened, which is associated with the heat produced from the dissolution of anhydrous sodium metasilicate resulting in rapid geopolymerization on the surfaces of fly ash and sodium silicate grains. It is noteworthy that the mixing time significantly affects the setting time of the one-part geopolymer paste, the higher the mixing time and the faster the setting time. The initial setting time examines the mixing time of 3 and 5 min is found to be almost the same while the final setting time changes slightly. In the process of releasing alkali and silica ions from the granular sodium metasilicate activator, one-part geopolymers made with higher activator content (N3, N4 & N5) tend to set rapidly than N1 and N2, which is attributed to an increase in temperature. The optimum mixing time for one-part geopolymer paste is found to be 5 min at normal mixing speed based on the data obtained from this study. The initial setting time varied from 45 to 25 min as shown in Fig. 4, and the final setting time was recorded from 125 to 55 min. It was also



Fig. 2. (a). Flowability property of the one-part geopolymer paste before stirred, (b) one-part geopolymer paste after stirred.

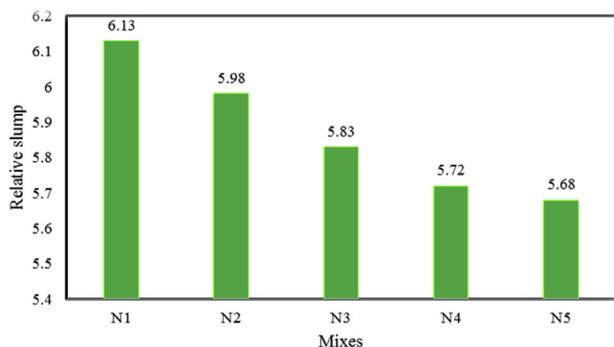


Fig. 3. Relative slump the one-part geopolymer pastes.

observed that the setting time of the designed one-part geopolymer paste decreases for both final and initial setting time at higher concentration of sodium metasilicate activator. This is associated with the number of reactive elements in the content of the source materials that ultimately led to the development of N-A-S-H and C-A-S-H gels. It is noteworthy that in the NaOH solution medium, silica, alumina and other minerals are leached from fly ash [41], while in the system with high calcium content in the precursor material, silica and alumina with high CaO content lead to C-A-S-H formation which coexisted with N-A-S-H at the initial stage which later contributed to the rapid reaction process [42, 43].

3.3. Hardened density

The hardened density of one-part geopolymer paste was evaluated by weighing the mass and dimensions of the cube specimens per mix, initially two cubes per mix were measured, and if the difference in densities between the two is more than 5 percent, then a third cube was used. The investigation revealed that hardened densities of the developed one-part geopolymer pastes decrease slightly with age attributable to a relatively low geopolymerization reaction at a later age. During the

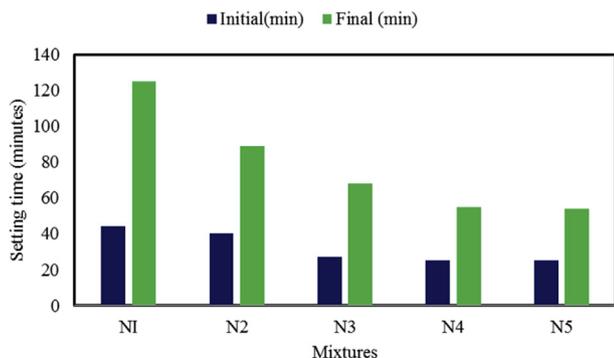


Fig. 4. Setting time of one-part geopolymer pastes.

reaction process, which later evaporated from the paste surface, the decrease may also be associated with the water loss. Accordingly, it can be deduced from these findings that at the initial stages the reaction process is faster. These findings are quite the opposite of [44]. Fig. 5 shows the densities of the different mixes and it was observed that the density of the one-part geopolymer pastes is almost the same. This shows that the amount of solid activator present in the mixtures has a negligible or small influence in the mixtures.

3.4. Compressive strength of one-part geopolymer pastes

The compressive strength growth of various combinations of geopolymer paste was evaluated at 3–28 days of ambient healing. Fig. 6 represents the compressive strength profile of user-friendly geopolymer pastes at different activator content. The results shown in Fig. 6 are the mean result of three specimens tested during the testing period. The compressive strength of nearly 50 MPa was obtained for N3 geopolymer paste which was the highest strength and occurred at 12 percent of the solid activator (6.1 percent Na₂O). It is pointed out that the strength gained at lower activator content is minimal leading to lower strength development, this is due to insufficient alkali that will trigger the system's geopolymerization process. However, increasing the activator content from 8 to 10 percent increases the strength by 24.6 percent. The high early strength in the geopolymeric system may be associated with the Si/Al ratio. The presence of comparatively lower Si constituents in N1 to N5 geopolymer pastes with a Si/Al ratio of 2.75–3.0 as shown in Table 3 makes it viable that additional Al species were present at the initial stages for contraction in the ingredients. It is noteworthy that the higher Si/Al ratio reduced the quantity of silicon in the geopolymer paste and the rate of geopolymerization reaction in an excessively fly ash-based one-part geopolymer. This could be due to the development of fewer reactive silicon dioxide oligomers instead of small reactive species at high SiO₂/Al₂O₃ ratios [5]. Subsequently, the aluminum element is likely to dissolve faster compared to the silicon element and thus allows a faster condensation rate between silicate and aluminum elements than the condensation between silicate elements. It is worth mentioning that N3's

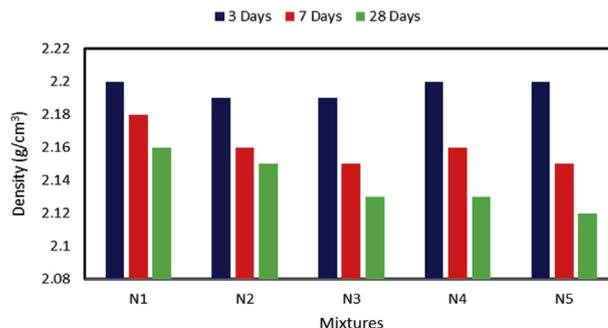


Fig. 5. Densities of one-part geopolymer pastes.

compressive strength increased significantly by nearly three times that of N1, which is attributed to sufficient Na₂O in the one-part geopolymer paste. Further increase in activator content (Na₂SiO₃) to 14 and 16 percent, which is 7 percent and 8 percent Na₂O content tends to decrease compressive strength by 12 percent and 14.5 percent respectively. This can be attributed to heat energy released at the higher activator during the geopolymerization process, resulting in the formation of minor cracks on the surface of the specimens. These cracks are associated with heat evolving during the process of reaction. In addition, the presence of excess sodium oxide in the mix is suspected to slow down the development of strength slightly.

In addition, the compressive strength of one-part geopolymer paste gradually decreases with higher activator content. It is therefore deduced that the improvement of the water content of the one-part geopolymer with the remaining uninterrupted variables leads to a significant increase in flowability and subsequent decreases in the compressive strength of the one-part geopolymer mix [14, 29].

3.5. Split tensile strength of the pastes

Fig. 7 shows the split tensile result of the one-part geopolymer paste for all design mixes considered. The test is conducted in accordance with BS EN 12390-6 [37]. To follow the same trend with compressive strength, the split tensile strength of one-part geopolymer paste cured at ambient temperature was obtained. At 4 percent Na₂O that is N1, the tensile strength of nearly 1.5 MPa is realized, but adding more Na₂O content in fly ash by 5 percent (N2) and 6 percent (N3) tends to increase strength by 26 percent and 66 percent respectively. It is noteworthy to explain that the split tensile strength of the developed one-part geopolymer pastes increases with an increase in anhydrous sodium metasilicate and subsequently decreases at 14 percent and 16 percent equivalent to (7 percent and 8 percent Na₂O) respectively. As shown in Fig. 7, it is noteworthy that the split tensile strength of the one-part geopolymer paste at 14 percent and 16 percent activator decreases slightly by 2 percent and significantly decreases by 22 percent compared to the N3 mix. This is attributed to the geopolymer's fast setting behavior whereby the Na₂O content did not participate fully in the reaction process. As such, some of the unreacted sodium oxides react with surrounding CO₂ to form sodium bicarbonate on the surface of the specimens and subsequently contribute to lower strength growth. The heat produced when water was added to the geopolymer cement is due to the dissolution of the solid sodium metasilicate which resulted in significant water loss resulting in some minor crack formation on the surface of the specimens. The cracks are associated with the stability of the geopolymer paste.

3.6. Effect molar ratios on strength

One of the notable parameters in making one-part alkali activated paste is the alkali activator content of the geopolymer paste as the

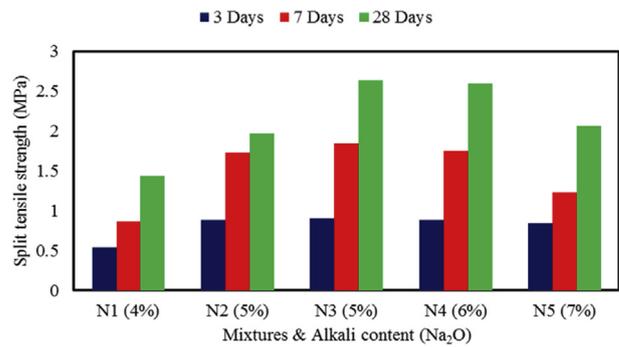


Fig. 7. Splitting tensile strength of one-part geopolymer pastes.

geopolymer's rheology and strength development was governed by the alkali content. The reaction occurs immediately as soon as water reacts with the solid precursors, the solid activator begins to dissolve in the mix, thereby generating an exothermic reaction process which results in elevating the temperature of the one-part geopolymer paste. The dissolution of the granular activator dissociates the alkali and silica ions thereby generating a heat of geopolymerization that accelerates the geopolymerization process on the source materials and the granular anhydrous sodium metasilicate activator. The molar ratios of all mixes of one-part geopolymer pastes were calculated and presented in Table 3. The Silica-alumina ratio of the developed geopolymer paste ranges from 2.75-3.0 with a ratio of 2.87 appeared to be the best in this work. Alumina emanates from the source materials of fly ash, while silica evolves from fly ash and anhydrous sodium metasilicate. The higher Si/Al values result from a high percentage of anhydrous sodium metasilicate in the blended paste. The formation of multiple polymeric silicate families at higher sodium metasilicate is suspected to have slowed down the reaction process and consequently affected the strength of the geopolymer pastes. Accordingly, the Silica-alumina ratio can not be increased infinitely as thoroughly deliberated for conventional geopolymer mixtures [5, 45, 46] as such patterns were adopted in this study.

It is observed that the strength of the one-part geopolymer paste increases rapidly with a decrease in H₂O/Na₂O ratios with the highest ratio having the lowest strength. The compressive strength of the one-part geopolymer significantly improved as the percentage of H₂O/Na₂O in the mixes decreases by 14.8 percent, 25.6 percent, 34.1 percent, and 40.8 percent for N2, N3, N4, and N5 respectively. The Na/Al ratio is found to be increasingly high at relatively higher activator content, resulting in excess concentration of Na⁺ in paste systems. The excess sodium oxide in the mix combines with water to produce more hydroxyl ions in the geopolymer paste which later reacts with calcium to form calcium hydroxide and thus decreases its strength [47]. Similarly, the surplus amount of Na⁺ ions in the mix reacts with CO₂ in the surrounding area to form a white precipitate on the surface of the specimens, which significantly contributed to the lower strength evolution at higher H₂O/Na₂O proportions, particularly at ambient temperature. This is due to the fact that one-part geopolymer paste contains higher soluble concentrations of alkaline ions than Portland cement as such when it comes into contact with water or due to higher humidity efflorescence, and this study is in agreement with that of [48].

Table 3
Molar ratios of the one-part geopolymer pastes.

Mix ID	SiO ₂	Al ₂ O ₃	Na ₂ O	H ₂ O	Si/Al	Na/Al	H ₂ O/Na ₂ O
N1	368.82	134.1	52.38	227.16	2.75	0.39	4.34
N2	377.1	134.1	61.56	227.7	2.81	0.46	3.70
N3	385.38	134.1	70.74	228.24	2.87	0.53	3.23
N4	393.66	134.1	79.92	228.78	2.94	0.60	2.86
N5	401.94	134.1	89.1	229.32	3.00	0.66	2.57

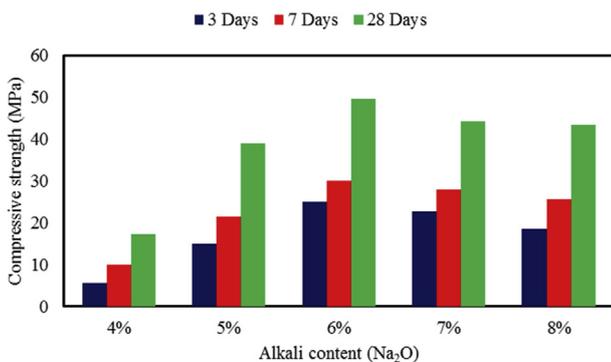


Fig. 6. Compressive strength of one-part geopolymer paste.

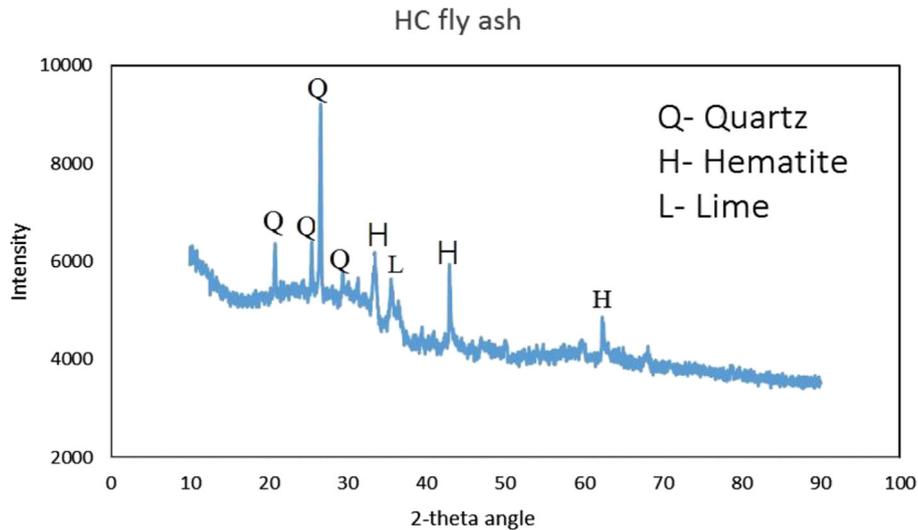


Fig. 8. X-ray diffraction (XRD) pattern of the fly ash.

3.7. Microstructural properties

3.7.1. X-ray diffractometer (XRD) investigation

The XRD patterns of high calcium fly ash (HCFA) and one-part geopolymer paste were obtained by Bruker's X-ray diffraction instrument and presented in Figs. 8 and 9. HCFA's primary mineral ingredients were quartz, magnetite, and lime. HCFA's XRD pattern in Fig. 8 shows a broad peak indicating that most of the HCFA structure is amorphous. The intensity of disturbances can be taken by the approach that diffracts X-rays to make a pattern. The intensity patterns were recorded using a constant divergence slit of 0.38 mm with Cu as the anode material using two distinct reflections coefficient $K\alpha_1$ of 1.5406 and $K\alpha_2$ of 1.5444. All samples are scanned from 10° 2 θ to 90° 2 θ at 0.0262 step size and scan range angle. Small portions of the geopolymer pastes were used for XRD analysis after 28 days of ambient healing and the existence of distinctive phases within the developed one-part geopolymer pastes was observed. The phases identified were Na aluminosilicate and Ca aluminosilicate

hydrate. The sharp characteristic quartz peak is associated with the development of alkali-silicate glasses with the partial amorphization of the phases [49]. The crystalline phases (quartz and hematite) observed in the fly ash remained almost unchanged after synthesis. X-ray diffraction characteristic for one-part geopolymer mixtures revealed the formation of different reaction products that differed depending on the percentage of activator material used. It is interesting to note that the presence of calcite ($CaCO_3$) was also observed in the N3 geopolymer sample as depicted in Fig. 9 in all geopolymer pastes, low-intensity pyrrhotite ($Fe_{11}S_{12}$ is observed between 30° and 43° 2 angles). There is the appearance of a new phase of dorite compound for N4 and N5 geopolymers at higher activator content.

3.7.2. FESEM analysis

Field emission scanning electron microscope (FESEM) was used to investigate the microstructural pattern of one-part geopolymer based on fly ash. Before viewing the specimen in the FESEM, the specimens were

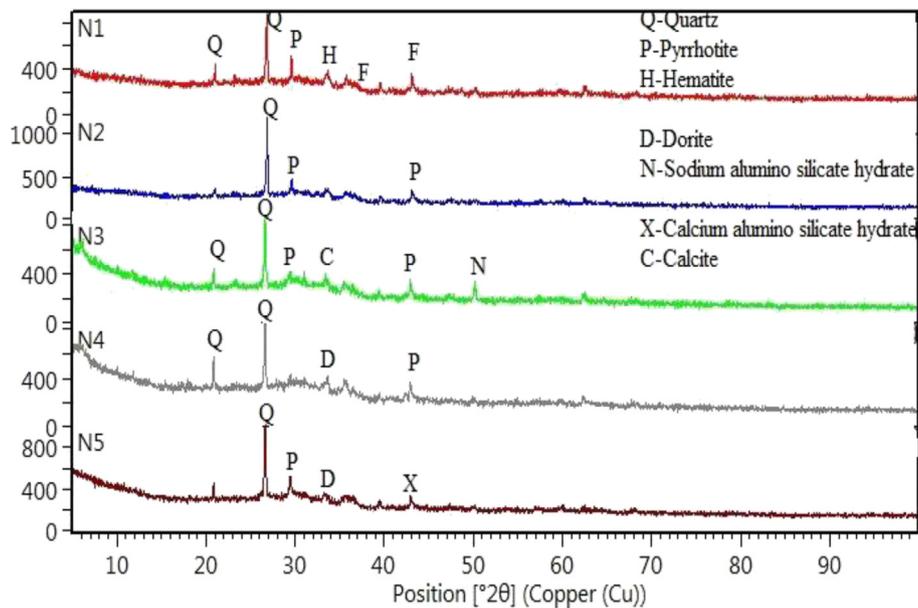


Fig. 9. X-ray diffraction (XRD) of one-part geopolymer mixtures cured at ambient temperature. Key to phases: Q- SiO_2 ICDD#01-075-8320; P- $Fe_{11}S_{12}$ ICDD#06-89-7439; C- $CaCO_3$ ICDD#01-05-0586; H- Fe_2O_3 ICDD#01-33-0664; X- $CaAl_2Si_2O_8 \cdot 4H_2O$ ICDD#00-039-1373; N- $NaAlSiO_3 \cdot 5H_2O$ ICDD#09-00-1344; D- $CaMg(CO_3)_2$ ICDD#01-36-0426.

coated with 200 Å Gold–Palladium and the micrograph image was obtained for each surface area examined under the microscope. After 28 days of ambient curing, a small portion of the hardened geopolymer pastes was taken for observations and comparison of the microstructure as shown in Fig. 10. As shown in Fig. 10(a-d), the microstructures of the paste show reacted amorphous microspheres, and some partially reacted spheres of fly ash appear in the entire matrix. It is interesting to see that the formation of the geopolymer paste structure was based on the reactions at the outer edge of the particles, forming reaction products adjoining cement particles [50, 51]. Different micrograph image is observed by increasing activator dosage from 8 percent to 12 percent with some reaction products covering fly ash particles. N3 micrographs were observed to be very compact and dense with fewer cracks for N3 as shown in Fig. 10b. It is interesting to note that some isolated voids or pores and microcracks were observed in microstructures with higher activator dosage (14 percent -16 percent) i.e. N4 and N5 as shown in Fig. 10(c-d). The decrease in strength of one-part geopolymer pastes is attributable to the appearance of these pores and cracks. Partially reacted particles develop some pores in the structure spread in pores of small shapes consequently small openings are noticed from the spots after the fly ash particles have been dissolved [52]. High CaO content in fly ash yields some Ca–Al–Si-hydrate gels in the microstructure.

3.7.3. Fourier transform infrared spectroscopy (FT-IR) analysis

FT-IR spectra are frequently used in characterizing products and are useful in characterizing the established HCFA one-part geopolymer paste in this job. FT-IR spectra were registered using the Perkin Elmer FT-IR spectrometer between 4000 cm^{-1} and 500 cm^{-1} . Measurements were done using 2 cm^{-1} and 40 scans resolution. The peaks of the one-part geopolymer paste happen for all mixtures at distinct wavelengths (i.e. N1, N2, N3, N4 and N5), as shown in Fig. 11. The absorption band at 3430 cm^{-1} , 3438 cm^{-1} and 1646 cm^{-1} corresponds to OH stretching and bending vibration of adsorbed water molecules on the surface of binder components, the band at 1416 cm^{-1} is connected with the asymmetric increase of Si–O and Al–O bonds [53]. The band at 991 cm^{-1} through 956 cm^{-1} corresponds to the loss of undistorted CO_3^{2-} gel in the scheme, absorption peaks at 742 cm^{-1} , 695 cm^{-1} , 618 cm^{-1} correspond to unequal stretching of Si–O–Si and Si–O–Al. These peaks show the magnitude of the source ingredients' reactivity and are common in silicate compounds [53, 54]. A combination of aluminum–oxygen, silicon–oxygen, silicon–oxygen–silicon, or silicon–oxygen–aluminum bonds presented by unequal crest places is helpful in interpreting the geopolymerization mechanism. It is worth mentioning that higher activator content did not result in major structural modifications of the geopolymer pastes, but a slight movement in the peak position was noticed

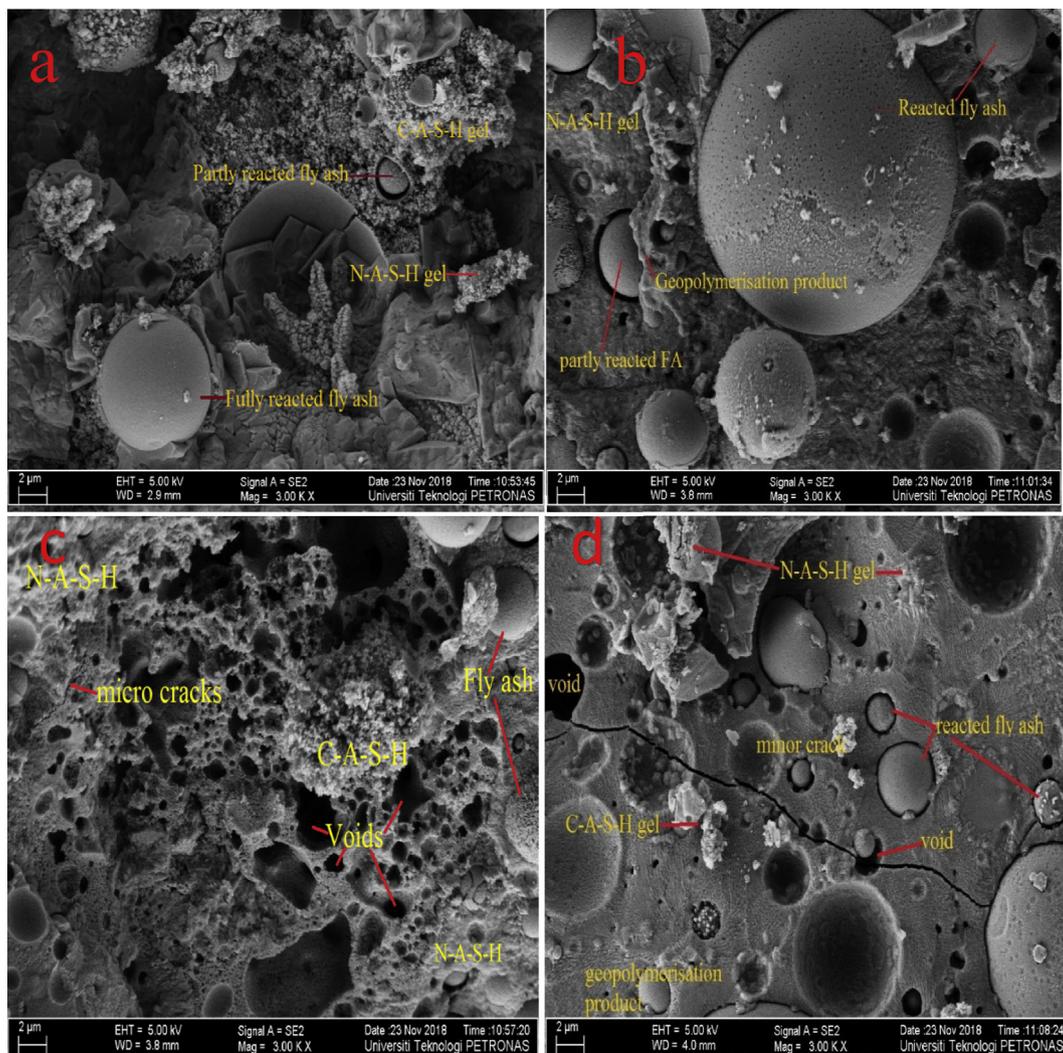


Fig. 10. FESEM micrographs of one-part geopolymers: (a) N1 with 4% Na_2O (b) N3 with 6% Na_2O (c) N4 with 7% Na_2O (d) N5 with 8% Na_2O .

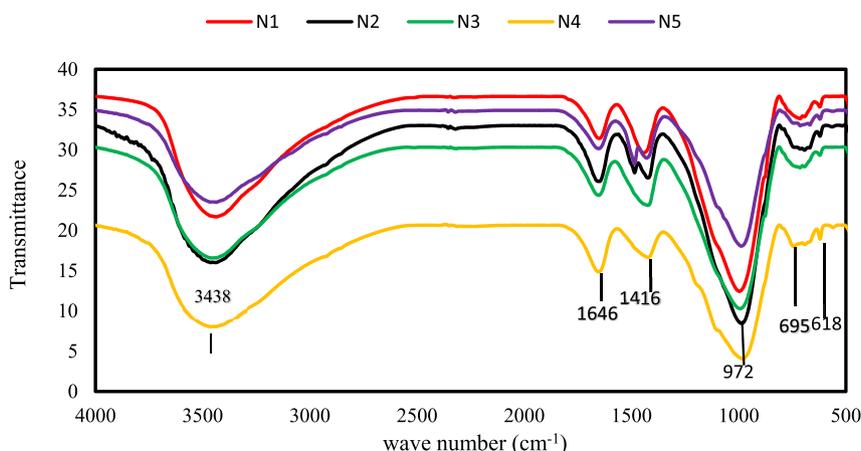


Fig. 11. Combined FT-IR spectra of fly ash based one-part geopolymer paste at different activator content.

for all the mixtures. Although N4's transmittance is lower than the remaining mixtures, it follows the same patterns as N1, N2, N3, and N5 respectively.

4. Conclusion

This study shows a crucial step towards using high-calcium fly ash as a substitute building material for in-situ and large-scale concrete manufacturing. Granular anhydrous sodium metasilicate has been effectively used to create one-part activated alkali pastes. Understanding the perfect alkali activation circumstances is essential for producing environmentally friendly products that will be used almost as worthy replacements for ancient cement material. Five mixtures of one-part geopolymer pastes were intended for distinct activator structure. Workability, setting time, density and strength behaviour, impact of molar proportions and microstructural characteristics were envisaged. The following findings are drawn from the present experimental consequence:

- i. Results showed that raising the granular activator by weight of the fly ash beyond 12 percent slightly decreases the strength and workability of the one-part geopolymer cement pastes. The optimum mix by weight of the fly ash was discovered to be 12 percent (i.e. 6 percent Na_2O) at 28 days curing.
- ii. The developed one-part geopolymer paste exhibited a compressive strength of nearly 50 MPa. The flowability of one-part geopolymer paste was comparatively affected by the content of sodium metasilicate in the mixtures. Flowability reduces with regard to N1 by 2.5 percent, 4.9 percent, 6.7 percent, and 7.3 percent for N2, N3, N4, and N5 mixtures respectively.
- iii. The setting time of the designed one-part geopolymer paste decreases for both final and initial setting time at higher concentration of sodium metasilicate activator. The setting ranges from 25 min to 125 min. The hardened density of the one-part geopolymer paste is nearly the same regardless of the mixes.
- iv. With a rise in $\text{H}_2\text{O}/\text{Na}_2\text{O}$ ratios with the largest proportion having the smallest strength, the strength of the one-part geopolymer paste reduces quickly. The strength improved by almost 2 to 3 times when the $\text{H}_2\text{O}/\text{Na}_2\text{O}$ was lowered from 14.8 % to 40.8 %.
- v. FESEM, FTIR and XRD microstructural analyzes have shown that the advanced geopolymer paste includes quartz, pyrrhotite, sodium aluminosilicate hydrate and calcium aluminosilicate hydrate gels.
- vi. The advanced geopolymer cement can be used for the building, maintenance and patching of significant structural and non-structural components as it can be healed in the ambient setting without any heat healing owing to fast hardening process.

Declarations

Author contribution statement

Sani Haruna: Performed the experiments; Analyzed and interpreted the data; Wrote the paper.

Bashar Mohammed: Conceived and designed the experiments.

Mohamed Mubarak & Abdulrahman Haruna: Analyzed and interpreted the data.

Mohd Shahir Liew: Contributed reagents, materials, analysis tools or data.

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Competing interest statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

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