

Fly ash-GGBS blended geopolymer mortar for early engineering characteristic at ambient temperature

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ABSTRACT

Geopolymer concept is by using industrial waste materials which is rich in silica and alumina, then activating them with alkaline solution. An appropriate binder combination that achieves high early age strength at ambient temperature using FA is still subject required. Thus, this study has been designed in two stages which involving the paste and mortar studies. The alkaline activator as a blend of sodium hydroxide and sodium silicate solution with SS/SH ratio of 2.5, NaOH molarity of 12 M and L/B ratio of 0.43 are used for geopolymer composite. Compressive strength, water absorption and shrinkage are tested on geopolymer mortar with various level of blended combination. High volume content (60–80 %) gave a promising early age compressive strength at ambient temperature. Metakaolin gave a further strength improvement on geopolymer mortar. Overall, the finding suggested that the geopolymer with high FA content is suitable for cement-based material repair product.

1. Introduction

The geopolymer concept is no longer new in the concrete industry, as the construction sector has come under scrutiny for the significant contribution of the cement industry, which is the second-largest industry that contributes to the increase of global carbon emissions [1,2]. Aside from environmental concerns, the cement industry also faces complications due to the depletion of raw materials in cement production [3]. According to Zahid et al. [4], about 1.6 tonnes of natural raw resources are needed to produce a tonne of ordinary Portland cement (OPC). With this regard, geopolymer technology is considered as an approach to overcome each problem that arises [3,5].

Geopolymer is also known as one of the methods for producing an alternative binder to the traditional binder of OPC, in which the basic principle of geopolymer is to create a zero-cement binder [3,5]. The geopolymer binder concept consists of two main constituents: the binder material, which possesses aluminosilicate material, and the alkaline solution. The aluminosilicate material is commonly known as pozzolan, which is mostly a by-product of materials such as fly ash (FA), ground granulated blast furnace slag (GGBS), metakaolin (MK), and silica fume. By reducing the need for cement and utilizing by-products, geopolymers are considered green construction materials for sustainable construction

[6].

Literature indicates that the geopolymer was discovered by Davidovits in the 1970 s, and research on the development of geopolymer composites for construction applications has been conducted over the last two decades [7,8,9]. Among the aluminosilicate materials for geopolymer production, FA stands out as the most favorable binder due to its widespread availability and low cost [5,8]. Geopolymer composite materials using FA have proven to be cost-effective compared to Portland cement products and beneficial to the environment, and they help eliminate greenhouse gases, making them a viable prevention method [10]. Extensive research on geopolymer composites using FA has been carried out by numerous researchers over the years [11]. In addition to the advantage of being rich in silica and alumina, the round shape of FA is favorable for the fluidity characteristics of geopolymer composites. Furthermore, the use of FA provides high compressive strength, reduces permeability, and enhances durability. Nevertheless, the use of FA hinders early strength development, particularly ambient curing, which limits the amount of FA that can be used [12,13]. In contrast, geopolymer composites, particularly those incorporating FA, exhibit high strength at an early age when the composites are subjected to high-temperature curing [5]. In addition, FA-based geopolymer composites require heat curing, which is not a practical approach, particularly for

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industries [9]. Under high-temperature conditions, the geopolymerization reaction produces sodium aluminosilicate hydrate (NASH) and calcium aluminosilicate hydrate (CASH) gels, which form a dense microstructure of composites and result in strength enhancement. The hindrances of curing conditions in geopolymer composites, which affect the strength development, also limit the application of geopolymers in the construction industry, for example, their use is limited to the precast concrete industry due to their requirement of heat curing [14].

Atis et al. [15] observed that FA geopolymer mortar achieved a very high compressive strength of 120 MPa with 14 M of sodium hydroxide (NaOH) and a curing temperature of 115 °C for 24 h. The authors also discovered that the strength was not properly developed at the curing temperature of 45 °C, even though the curing duration was 72 h. Assi et al. [12] found that geopolymer concrete with 100 % FA achieved high early strength (approximately 105 MPa) by using class F FA and silica fume as the activator solution. The findings also reported that no significant effect was observed on the strength due to the variation of the curing period under combined curing conditions (ambient temperature and 75 °C).

Numerous modifications have been made to the geopolymer system using various sources of binder materials [9,16]. According to Guo et al. [17], the type of binder materials affects the properties of geopolymer composites. Literature reports a significant focus on the evaluation of geopolymer composites produced from FA and GGBS [9]. The geopolymer composites produced by the activation of both FA and GGBS result in better compressive strength and promising engineering properties compared to that of OPC composites [16,18]. On another note, GGBS exhibits high early strength development, but rapid setting and low workability have been significant issues. Given these discrepancy behaviors, it is essential to determine the optimum mix combinations between the two source materials, particularly to achieve an early high strength characteristic.

Zhang et al. [16] reported that the compressive strength of blended FA/GGBS geopolymer composites improved with increasing GGBS content. Furthermore, the apparent porosity of geopolymer mortar was found to decrease with an increase in GGBS content [19]. Nevertheless, GGBS decreased the workability due to its angular shape [19]. Puertas et al. [20] revealed that FA/GGBS geopolymer pastes achieved strength of more than 50 MPa at 28 days with an FA/GGBS ratio of 1.0 and 10 M NaOH at a temperature of 25 °C. Pan et al. [21] discovered that the compressive strength of blended geopolymer composites using GGBS and FA showed good performance at a curing temperature of 60 °C, but a high temperature was required for the mixture based solely on ash.

Another important parameter in geopolymerisation reaction is a strong alkaline solution, which is known as alkaline activator [15]. A strong alkaline solution, which is a high concentration of alkaline activator, is required to dissolve the amorphous content of aluminosilicate material for geopolymerisation reaction which in turn produce the hardened geopolymer composite. Although there are arguments on the parameter of alkaline activator, most findings indicate that sodium hydroxide with 12 M concentration has a potential in achieving a high early strength of geopolymer composites [16,22].

Regarding other aspects of studies, Bouzoubaa et al. [23] discovered that a high volume of coarse FA provided better mechanical and durability properties compared to those with unground FA for cement-based concrete. Furthermore, the replacement of cement with FA exceeding 40 % has been implemented in cement-concrete applications, leading to the attainment of promising properties [24]. Most studies on geopolymer composites related to ash/FA focus on the effects of different curing conditions. Although blended FA/GGBS geopolymer composites have favorable outcomes, the main parameters such as the FA/GGBS ratio, type, and concentration of alkalis are crucial in achieving the targeted properties. On the other hand, investigating the combination of FA and GGBS is not uncommon in concrete research, with studies dating back to the 1980s. In addition, the effect of high volume of FA on the

properties of concrete has been well documented. Nevertheless, the coverage of the combination utilizing FA as a primary component in the geopolymer field is still lacking, where the binder is a key factor in geopolymer properties.

The main objective of this research is to determine the appropriate mix combinations using primary binder material of FA with GGBS and MK as combined binders to achieve high early strength at ambient temperature. The effect of varying FA contents on compressive strength was also investigated to explore the beneficial use of FA. The alkaline activator concentration and SS/SH ratio were kept constant to understand the effect of aluminosilicate materials used in the geopolymer composite system. A preliminary investigation into identifying the optimum mix combinations was performed on FA/GGBS paste, with compressive strength tested at an early age of up to 3 days. The optimization analysis using response surface methodology (RSM) was also carried out. Moreover, the compressive strength development, water absorption, and drying shrinkage of geopolymer mortar were also evaluated. Microstructure analysis using field emission scanning electron microscopy (FESEM) and energy dispersive X-ray (EDX) were also conducted to examine the difference in depolymerization reactivity and morphology of geopolymer composites. The research outcomes were analyzed to find an optimum mix that exhibits favorable engineering properties for geopolymer mortar, which has the potential to be used as an alternative green material in the construction industry.

2. Materials and methodology

2.1. Materials

In this study, three aluminosilicate binder materials were used: FA, GGBS, and MK. Metakaolin was considered as a third component, while the other two materials served as primary binders. Fly ash was obtained from the Kapar Energy Ventures power plant in Malaysia, while both GGBS and MK were obtained from local suppliers in Malaysia. The chemical compositions and physical characteristics of the aluminosilicate binder materials are presented in Table 1. The chemical compositions were determined using X-ray fluorescence analysis. It is well noted that the current FA is classified as Class F Type FA according to ASTM C618 [23] because its calcium oxide (CaO) content is less than 10 % and the total amount of silicon dioxide (SiO₂), aluminum oxide (Al₂O₃), and iron oxide (Fe₂O₃) is more than 70 %. Meanwhile, the GGBS used consists of more than 40 % CaO, which classifies it as a latent hydraulic

Table 1
Properties of the binder constituents.

Chemical Composition (% by Mass)	Binder			
	OPC	Fly ash	GGBS	Metakaolin
SiO ₂	17.28	38.32	16.03	60.44
Al ₂ O ₃	3.65	16.48	11.28	28.11
Fe ₂ O ₃	3	22.48	0.35	3.55
CaO	60.90	12.39	41.39	1.10
MgO	2.80	0.28	6.38	–
K ₂ O	1.08	2.79	0.37	1.05
TiO ₂	–	3.78	0.45	3.48
P ₂ O ₅	–	–	–	–
SO ₃	3.82	1.06	1.08	1.19
SiO ₂ + Al ₂ O ₃ + Fe ₂ O ₃	–	77.28	–	–
ASTM C618	–	Class F Minimum of 70 %	–	–
Physical Properties				
Type	–	Class F	–	–
Color	Grey	Grey	White	White
Specific Gravity (g/cm ³)	–	2.24	2.67	2.6
Specific surface area, (m ² /g)	–	4.54	5	19–20

* The chemical results obtained from X-ray fluorescence analysis.

binder. The microstructure of the aluminosilicate binder materials was examined using a field emission scanning electron microscope and is shown in Fig. 1. It can be noticed that the FA particles have a round shape and smooth surface texture, whereas GGBS particles have an angular and more to flaky-elongated shape with a rough surface layer (see Fig. 1(a) and 1(b)). For MK, the particles have a similar angular shape as GGBS particles, but the surface is platy, resulting in a more porous surface (Fig. 1(c)).

The alkaline solution used is single alkaline prepared using sodium silicate (Na_2SiO_3) and NaOH with an SS/SH ratio of 2.5. According to Zhang et al. [16], a ratio of 2.5 is optimal for enhancing the geopolymerization reaction irrespective of the binder combinations. The chemical composition of Na_2SiO_3 used is SiO_2 (32.1 %), Na_2O (16.5 %), and H_2O (51.4 %). Meanwhile, NaOH was supplied in pellet form with 12 M concentration. A commercial sulfonated naphthalene-based superplasticizer was used to achieve the desired level of workability for geopolymer mortar. Natural river sand was used as fine aggregate, which was obtained from a local supplier in Malaysia. The fine aggregate size ranged between 300 μm and 1.18 mm. Table 2 presents the physical properties of the fine aggregate.

2.2. Experimental program and mixture proportions

This study involved two phases. The first phase focused on geopolymer paste with four main mix combinations of FA/GGBS containing different levels of FA between 20 % and 80 %. A further study was then conducted with the inclusion of 10 % MK as a third component in the main mix combinations. This investigation aims to determine the most feasible mix combinations of FA/GGBS that can achieve high early strength at the age of 3 days. The identification of the optimum mix combinations was performed using both analytical and experimental methods. In the first phase, 12 M NaOH, liquid-to-binder ratio (L/B) of 0.43, and SS/SH of 2.5 were kept constant for all mix combinations. In phase two, the most favorable FA/GGBS mix combination obtained from the paste was then applied to the geopolymer mortar. For the mortar mixture, a similar geopolymer parameter was employed with a binder-to-sand ratio of 1:3 and a superplasticizer to achieve flowability. The mortar ratio used complies with the ASTM standard. The details of the mix proportion for paste and mortar are described in Table 3.

2.3. Mixing and curing

A similar mixing procedure was applied for geopolymer paste and mortar, where the binders were dry-mixed for 3–4 min, followed by the addition of the blended alkaline solution. The mixture was further mixed for another 6 min until it became homogeneous. Then, the geopolymer mixture was cast into molds and vibrated on a vibration table for 25–30 s. After 24 h, the specimens were demolded and stored at ambient temperature until the testing age.

Table 2

Physical properties of fine aggregate.

Properties of Fine Aggregate	
Specific gravity	2.64
Colour	Brown
Water Absorption (%)	1.20
Fineness Modulus	2.64

2.4. Experiments

The compressive strength of the geopolymer specimen was measured in accordance with ASTM C109 [25] on a cube measuring 50 mm \times 50 mm \times 50 mm. The tests were conducted at the age of 1, 3, 7, and 28 days, with three specimens for each test.

The total water absorption test was conducted on a 50 mm \times 50 mm \times 50 mm cube in accordance with ASTM C642 [26]. The geopolymer mortar specimens were dried in an oven for 24 h at 100–105 $^\circ\text{C}$. The mass of the dried geopolymer mortar specimen was weighed as the initial reading (W_d). Then, the dried specimens were immersed in water for testing periods of 1, 3, and 7 days. The mass of wet geopolymer mortar specimens was weighed (W_w) and the percentage of water absorption was determined using Eq. (1). Meanwhile, the percentage of porosity of the specimens was measured using Eq. (2).

$$\text{Water absorption (\%)} = \frac{(W_w - W_d)}{W_d} \times 100 \quad (1)$$

$$\text{Porosity (\%)} = (W_w - W_d) \times 100 \quad (2)$$

The drying shrinkage test was carried out on a specimen prism of 25 mm \times 25 mm \times 275 mm in accordance with ASTM C157 [27] at the age of 1, 3, 7, 28, and 56 days, with three specimens tested for each day. The test was conducted upon demolding of the specimens for the initial reading measurement. All specimens were cured at ambient temperature throughout the testing. For microstructure imaging assessment, the samples were analyzed using FESEM/scanning electron microscopy (SEM). The morphological characteristics of the samples may provide further explanation of the engineering behavior of the geopolymer blended composites. Furthermore, the chemical analysis of the geopolymer paste composites was conducted using EDX. The measurement was performed on the blended geopolymer paste with 12 M, and the sample was obtained from the mechanical test.

3. Results and discussion

3.1. Compressive strength of geopolymer paste

Fig. 2 shows the early compressive strength development of FA/GGBS geopolymer paste with various contents of FA ranging from 20 % to 80 %. In general, the FA/GGBS compound paste exhibited higher

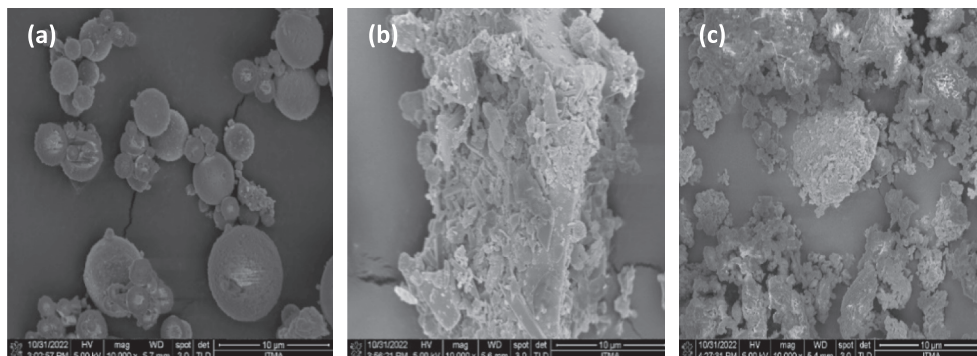


Fig. 1. FESEM images of (a) fly ash, (b) ground granulated blast slag (GGBS) and (c) metakaolin.

Table 3
Detail geopolymer composite mix proportions.

Geopolymer Paste		Binary Compound				Ternary Compound			
Mix Combinations	100 % OPC	FA > 50 %		FA < 50 %		FA > 50 %		FA < 50 %	
		80FA20GGBS	60FA40GGBS	40FA60GGBS	20FA80GGBS	80FA10GGBS10MK	60FA30GGBS10MK	40FA50GGBS10MK	20FA70GGBS10MK
OPC (kg/m ³)	1440	–	–	–	–	–	–	–	–
FA (kg/m ³)	–	1152	864	576	288	1152	864	576	288
GGBS (kg/m ³)	–	288	576	864	1152	144	432	720	1008
MK (kg/m ³)	–	–	–	–	–	144	144	144	144
Water (litres)	620	–	–	–	–	–	–	–	–
Activator (kg/m ³)	–	620	620	620	620	620	620	620	620

Geopolymer Mortar		Binary Compound		Ternary Compound	
Mix Combinations	100 % OPC	FA > 50 %		FA > 50 %	
		80FA20GGBS	60FA40GGBS	80FA10GGBS10MK	60FA30GGBS10MK
OPC (kg/m ³)	468	–	–	–	–
FA (kg/m ³)	–	374	281	374	281
GGBS (kg/m ³)	–	94	187	47	140
MK (kg/m ³)	–	–	–	47	47
Water (litres)	201	201	201	201	201
Activator (kg/m ³)	–	–	–	–	–
Fine Aggregate (kg/m ³)	1424	1424	1424	1424	1424

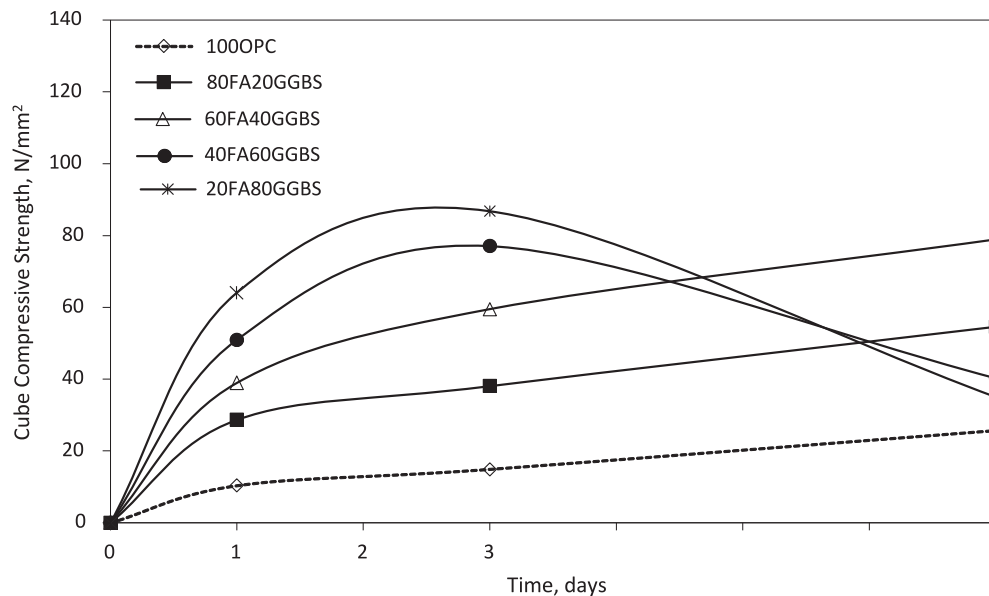


Fig. 2. Early age compressive strength for FA/GGBS geopolymer paste at 12 M of NaOH.

strength compared to the 100OPC paste, regardless of the FA content and age. The results also indicate that the early strength development is governed by the changes in geopolymerization reactivity with FA content. Fig. 2 also demonstrates good early strength development with a decrease in FA content at the early age of 1–3 days. The compressive strengths for the geopolymer paste of 60FA40GGBS at the age of 1, 3, and 7 days were 39.0, 59.50, and 78.95 N/mm², respectively. Meanwhile, the compressive strengths of 100OPC paste at the age of 1, 3, and 7 days were 10.36, 14.92, and 25.71 N/mm². Good early strength development up to 7 days for geopolymer paste of FA/GGBS with high FA content (>50 %) was also observed. The presence of GGBS consistently enhanced the early-age strength of geopolymer paste, except for mixes with GGBS content exceeding 50 %.

The FA geopolymerization results in NASH bonding gel for strength

development of the geopolymer matrix due to the limited calcium (Ca) in the geopolymer system. The crystalline phase of FA, however, requires a longer time to dissolve and heat cure to accelerate geopolymerization, which is responsible for slow early strength development [28,29]. The presence of GGBS accelerates the geopolymerization reactivity due to its glassy material characteristic, which is easily activated when it reacts with the alkaline activator. GGBS, also known as a latent hydraulic binder, contains CaO in addition to SiO₂ and Al₂O₃. The reaction between the alkali activator and CaO releases heat, which enhances the geopolymerization reactivity, leading to early strength development [30]. Furthermore, the progressive increase in strength with increasing GGBS content is due to the increase of Ca compound in the dissolution of the binder, which enhances geopolymerization reactivity. Subsequently, Ca reacts with the alkaline

activator to form the binding gel of calcium silicate hydrate (CSH), along with geopolymer gels (CASH and NASH). The formation of binding gels results in a more compact geopolymer microstructure, contributing to the high early strength in the FA/GGBS geopolymer system [28,30,31]. Previous researchers reported a similar trend where high early strength and strength development were achieved by increasing the GGBS content, but the compressive strength decreased with excessive GGBS content ($\geq 50\%$), particularly after 7 days [31]. This is because the gel formation is affected by the slow dissolution rate of FA, which leads to low aluminosilicate availability for Ca reactivity, as well as low NASH gel production [30,31,32]. Excessive Ca results in the formation of calcite despite the reasonable compressive strength achieved [30]. On the other aspect, a high amount of GGBS may lead to high autogenous shrinkage, which can result in cracking and surface dry conditions, which are also responsible for strength reduction [32].

The findings from the EDX spectrum analysis shown in Fig. 3 confirm the increase of Ca content with increasing GGBS content, which is associated with rapid geopolymerization reactivity for high early strength development. Furthermore, the EDX spectrum analysis also observed that a high Ca content led to an increase in the Ca/Si ratio. A higher Ca/Si ratio enhanced the formation of CSH gel, which improved the microstructure of the geopolymer system [33]. A similar trend of an increase in Si/Al ratio and a decrease in Al content was observed with an

increasing GGBS content as FA replacement in all FA/GGBS geopolymer paste mixes. The Si/Al ratio indicates the rate of dissolution of chemical elements within the geopolymer system, which affects the formation of geopolymer gels for a denser geopolymer microstructure [5]. A high Si/Al ratio leads to the formation of geopolymer gels with Si–O–Si bonds, which are stronger than the geopolymer gels with Si–O–Al bonds, resulting in the high strength of the geopolymer matrix [5]. Thus, the finding suggests that incorporating GGBS into the FA geopolymer system effectively accelerates the geopolymerization reactivity and consequently, enhances the early strength development of the FA geopolymer system at ambient temperature. This finding is in agreement with studies by Shang et al. [31] and Nath & Sarker [34].

However, the results showed that the progressive strength beyond 3 days decreased for geopolymer paste containing low FA content ($< 50\%$), in contrast to high GGBS content as FA replacement (60%–80%). The strength reduction can be anticipated due to the excessive amount of Ca resulting from the high GGBS content. As seen in Fig. 3, the Ca content is 9.51 and 12.59 for geopolymer paste with high GGBS content of 60% and 80%, respectively. In comparison to the Si content, the EDX spectrum analysis indicates that the Ca content is low, and this can affect the strength as NASH gel may govern the geopolymer matrix. For the same mixes (40FA50GGBS and 20FA80GGBS), the study also observed a decrease in the amount of Ca between the age of 3 days and 7 days. This

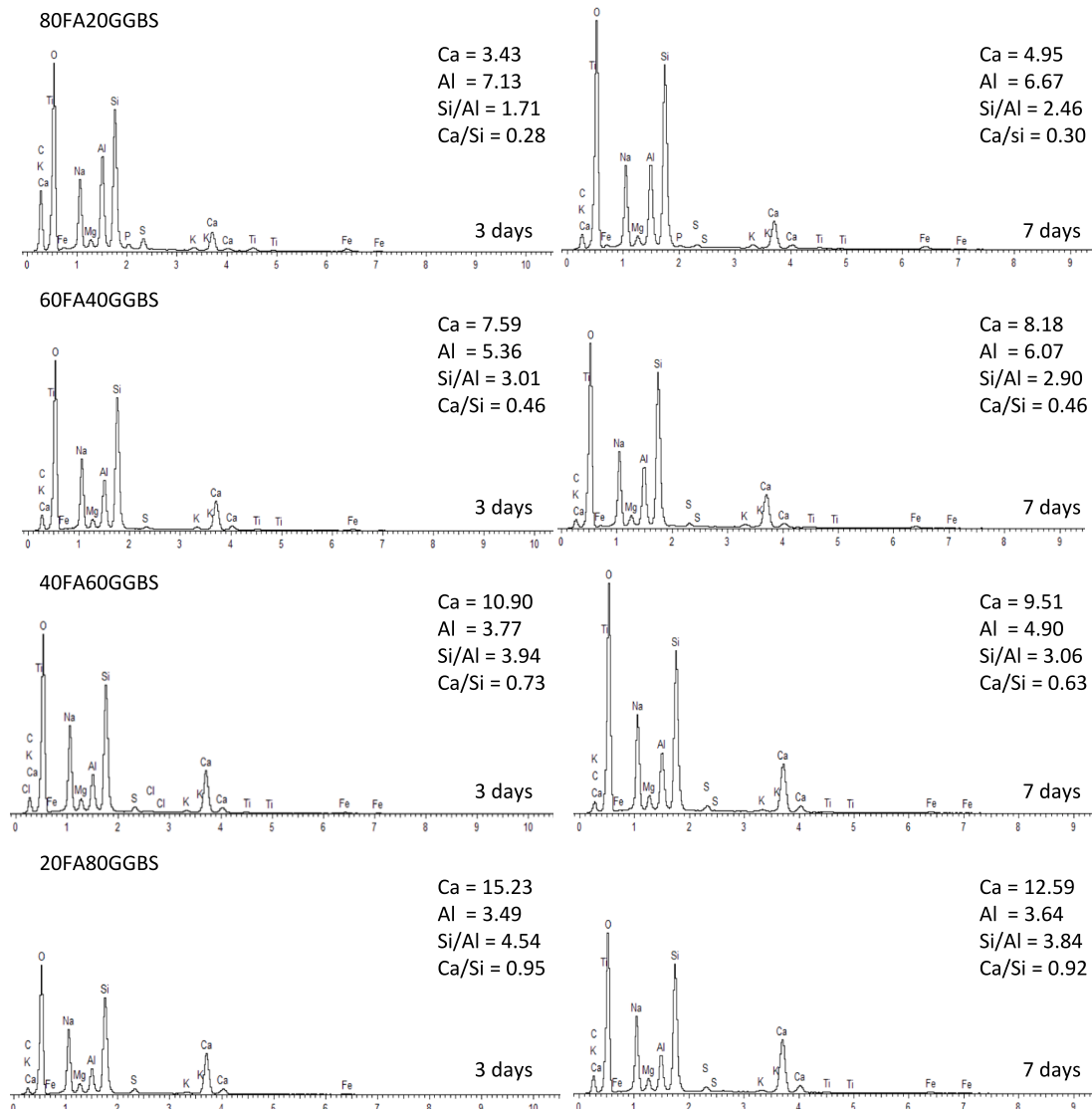


Fig. 3. EDX spectrum analysis of FA/GGBS geopolymer paste.

may suggest that the Ca did not react within the FA/GGBS geopolymer matrix, which inhibits the formation of geopolymer gels [30]. In this condition, the excessive Ca may also cause microcracks due to the expansion of calcium hydroxide [33]. Visible microcracks can be

observed in the microstructure images of geopolymer paste containing low FA content (<50 %), as shown in Fig. 4. A dense geopolymer matrix paste can be seen in the geopolymer mixes of 40FA50GGBS and 20FA80GGBS. Despite the dense geopolymer matrix, the images also

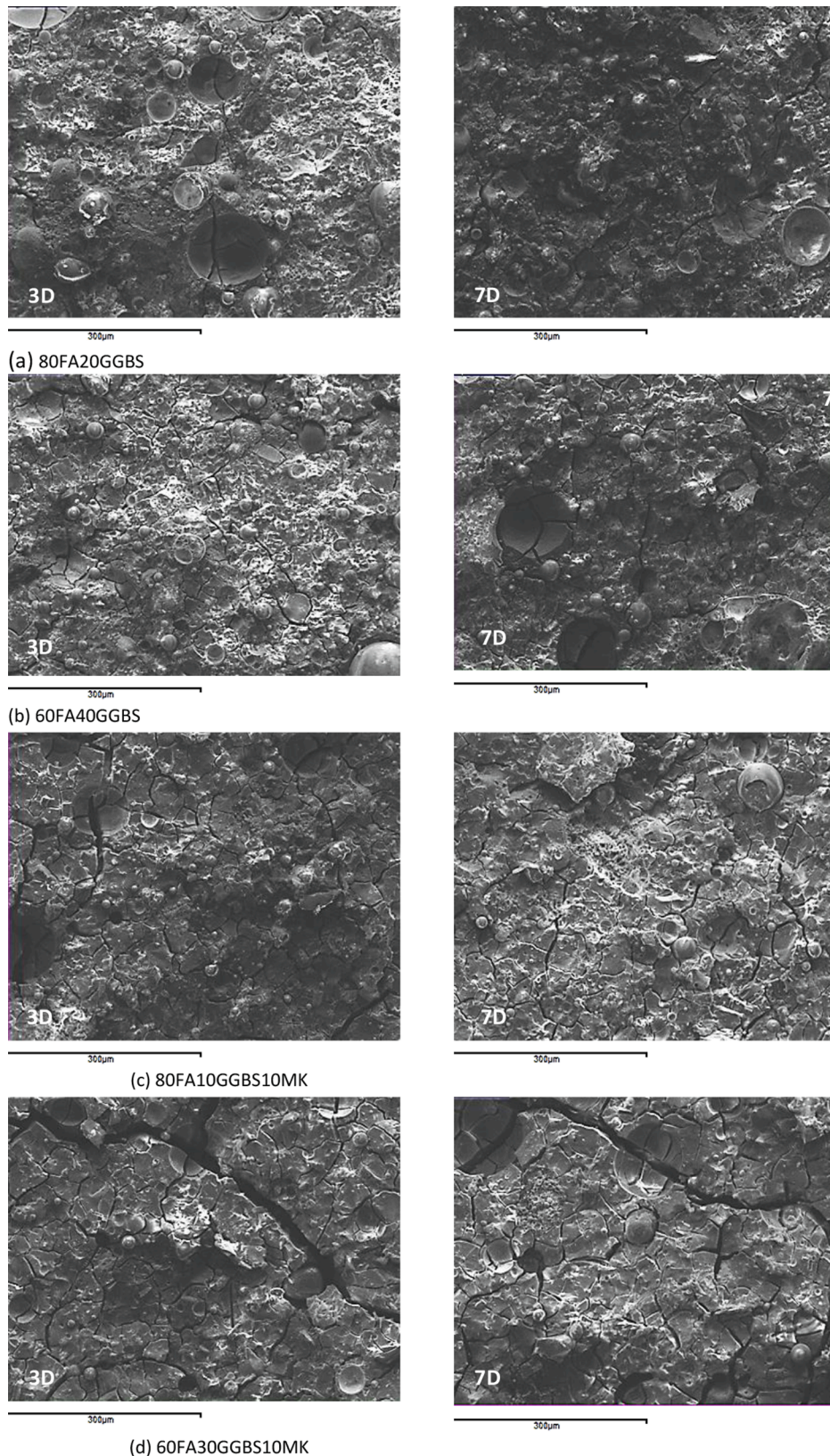


Fig. 4. SEM of FA/GGBS geopolymer paste.

show non-reacted or partially reacted FA particles within the geopolymer matrix. Furthermore, the microcracks induced in the geopolymer paste of 20FA80GGBS appeared to be wider than that of 40FA50GGBS regardless of age. It was also observed that the geopolymer matrix of 20FA80GGBS exhibited a surface dry condition. Collin & Sanjayan [35] reported that the strength reduction could also be caused by the surface dry condition of the geopolymer matrix. Shang et al. [31] determined that the higher the GGBS content (80 % GGBS) incorporated into the FA geopolymer mix, the slower the progressive strength after 7 days.

The effect of MK on enhancing the early strength development of the FA/GGBS geopolymer matrix is illustrated in Fig. 5. The peak strength was achieved at 3 days, where the strength increased with increasing GGBS content. When 10 % MK was used to partially replace GGBS, the strength increased by about 100 %, 55 %, 45 %, and 46 % for the mixes of 80FA/20GGBS, 60FA/40GGBS, 40FA/60GGBS, and 20FA/80GGBS, respectively. Metakaolin has a finer particle grain compared to FA and GGBS particles, where the finer the particles, the faster the geopolymerization reactivity occurs [36]. Furthermore, MK consists of Al₂O₃ and SiO₂, and the inclusion of MK into the FA/GGBS geopolymer compound increases the Si/Al ratio, in addition to the presence of Ca. Thus, the geopolymerization products result in NASH, CASH, and CSH, and these bonding gels enhance the strength of the geopolymer matrix. A similar trend of strength reduction beyond 3 days due to high GGBS content was observed in the FA-ternary compound geopolymer paste with low FA content. The strength decrement is expected because geopolymerization reactivity is governed by the main compound of the geopolymer system, which is similar to FA/GGBS binary blending.

3.2. Analytical analysis on Early-Age strength of FA/GGBS geopolymer paste

As the geopolymerization reactivity is governed by the major compound of the binder in the geopolymer system, analytical analysis using RSM was employed to evaluate the ideal mix combination of FA/GGBS geopolymer for high early strength. The analysis was conducted by considering the maximum compressive strength at 3 days. For the optimization analysis, three independent variables (or factors) and one dependent variable (or response) were defined for response surface modeling. The FA content, the GGBS content as a replacement for FA, and the curing period (for the early age of 1–3 days) were considered as

the independent variables, whereas compressive strength was chosen as the dependent variable. The details of the independent factors are shown in Table 4. Other factors such as NaOH molarity, SS/SH ratio, and curing temperature were held constant at 12 M, 2.5, and 21 °C, respectively. The experimental design involved full factorial points, six axial points, and six center points. Twenty test runs (mixtures) and the experimental responses based on phase 1 of the study were imported and analyzed using Design-Expert software. The data were analyzed using non-linear fitting models, and the results identified that a second-order model had the best interaction between variables. The specific regression model for predicting early-age compressive strength is shown in Eq. (3). The regression coefficient (R²) of the equation model for early-age compressive strength is 0.9981, and the regression diagram is shown in Fig. 6. The result of the data regression indicates that the equation fitting effect is good. The equation of the early-age compressive strength developed is only valid for the boundary conditions that have been designed.

$$\begin{aligned} \text{Compressive Strength} = (\text{N/mm}^2) & 14.12 + 1.168 \text{FA} - 0.395 \text{GGBS} \\ & - 21.78 \text{Time} - 0.010346 \text{FA} * \text{FA} \\ & + 0.010587 \text{GGBS} * \text{GGBS} + 1.114 \text{Time} * \text{Time} \\ & - 0.00924 \text{FA} * \text{GGBS} + 0.2550 \text{FA} * \text{Time} \\ & + 0.3237 \text{GGBS} * \text{Time} \end{aligned} \tag{3}$$

The analysis of variance (ANOVA) was employed to examine the efficiency of the model, and the results are presented in Table 5. The results of the analysis show that the *p*-value of each model is less than 0.01, indicating that the regression effect is significant. The *p*-value of the lack of fit of the compressive strength model is greater than 0.05, suggesting that the proportion of abnormal errors in model fitting is small. Thus, this analysis indicates that the three factors have significant effects on the development of early-age compressive strength. Furthermore, Fig. 7

Table 4
Factors in the optimization analysis.

Factors	Low Level Limit	High Level Limit
Factor 1: FA %	20	80
Factor 2: GGBS %	20	80
Factor 3: Time	1	3

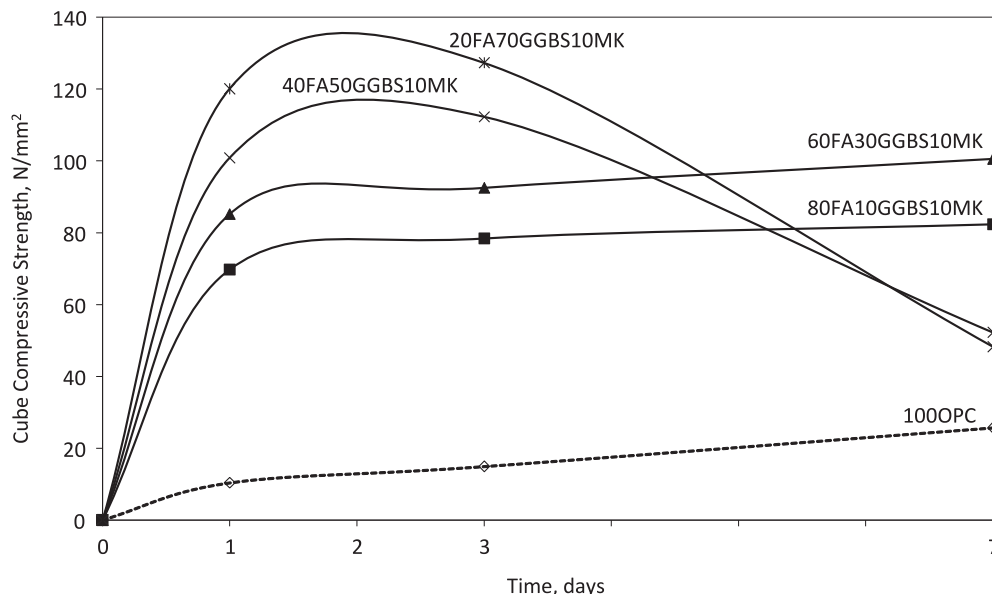


Fig. 5. Early age compressive strength for FA-based ternary system of geopolymer paste.

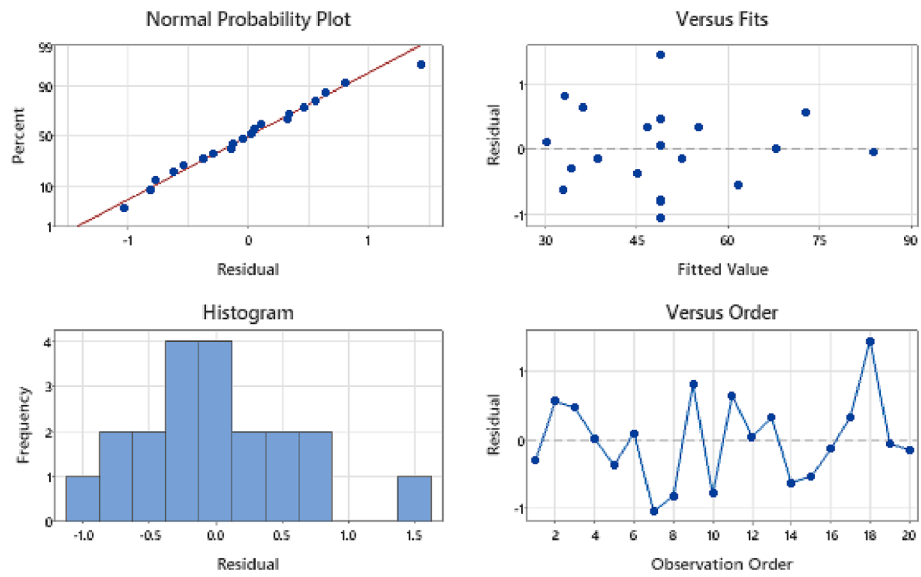


Fig. 6. Residual plots for early strength.

Table 5
Regression coefficient for the RSM model.

Source	Df	Adj SS	Adj MS	F-Value	P-Value	Remarks
Model	9	3719.95	413.33	573.73	0.000	Signification
Linear	3	3251.99	1084.00	1504.67	0.000	Signification
FA	1	117.94	117.94	163.71	0.000	Signification
GGBS	1	2594.88	2594.88	3601.90	0.000	Signification
Time	1	539.17	539.17	748.41	0.000	Signification
Square	3	356.94	118.98	165.15	0.000	Signification
FA*FA	1	136.25	136.25	189.12	0.000	Signification
GGBS*GGBS	1	142.68	142.68	198.05	0.000	Signification
2-Way Interaction	3	111.01	37.00	51.37	0.000	Signification
FA*GGBS	1	34.61	34.61	48.04	0.000	Signification
FA*Time	1	29.26	29.26	40.62	0.000	Signification
GGBS*Time	1	47.14	47.14	65.44	0.000	Signification
Error	10	7.20	0.72	—	—	—
Lack of Fit	5	2.64	0.53	0.58	0.720	Insignification
Pure Error	5	4.57	0.91	—	—	—
R ²	0.9981					
Adjusted R ²	0.9963					

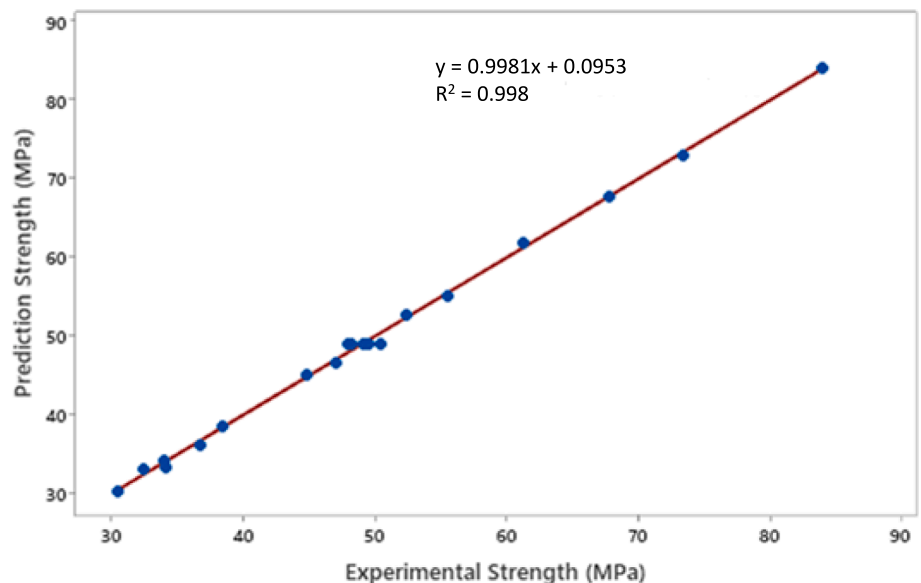


Fig. 7. Parity plots for Experimental and Fit Strengths.

illustrates a good fit between the experimental and predicted values, where the R^2 is close to 1. This indicates a significant correlation between the experimental and predicted results. Therefore, this model suggests that GGBS plays a significant role in the development of early strength, which can be associated with the high activity of GGBS due to its CaO content that promotes polymerization. Although CaO enhances strength development, excessive CaO adversely affects the properties of the geopolymer composite. According to Diaz et al. [37], CaO content should not be more than 20 % as it affects the setting of geopolymer composites and is not recommended for practical application.

3.3. Engineering properties of geopolymer mortar

Based on the previous findings from experimental and statistical analysis, a further investigation was carried out on the behavior of geopolymer mortar with a high content of FA. The assessment was conducted on the compressive strength, drying shrinkage, and water absorption.

3.3.1. Compressive strength

Fig. 8 shows that the compressive strength of mortar increases with age, confirming the trend observed in the geopolymer. The strength was also found to increase with an increase in GGBS content. A good early-age strength development (up to 7 days) with about 20–50 % strength increment compared to OPC control was observed in FA/GGBS geopolymer mortar mixes. The improvement in the strength of the geopolymer mortar could be attributed to the rapid formation of CSH and NASH gels during its geopolymerization. From the FESEM images of FA/GGBS geopolymer mortar shown in Fig. 9(a) and (b), it appears that the microstructure of the geopolymer mortar is relatively dense with an increase in the age of the mortar. A denser microstructure was also formed for mortar with a higher FA content replaced by GGBS, resulting in high compressive strength. The results also indicated that the compressive strength at 1 day, 3 days, and 7 days increased by about 19 %, 10–13 %, and 9 %, respectively, using 10 % MK as the GGBS replacement in the mixes in comparison to FA/GGBS geopolymer mortar only. The presence of MK in the ternary system has increased the content of Al_2O_3 and SiO_2 , which improved the geopolymerization process and produced additional NASH and CASH gels in addition to CSH gel. Thus, the formation of gels contributed to the early strength properties of geopolymer mortar. Despite the binder characteristics, fine aggregate

particles also play a key role. Generally, the finer the particle, the greater the surface area of the particles; thus, more binder gel is required to coat the particles and bind the composite components. Furthermore, the fine aggregate also possesses its own mechanical strength, which depends on the fine aggregate category. Medium sand and coarse sand have higher mechanical strength than fine sand [38]. In this study, the fine aggregate used consisted of smaller particles with a fine modulus of 2.64. This classifies the fine aggregate as medium sand. From the microstructure images shown in Fig. 9(c) and (d), it can be concluded that the presence of MK forms a denser structure, which leads to strength gain in the ternary geopolymer system. Furthermore, there is a strong binding within the microstructure of geopolymer mortar, indicating that there is sufficient geopolymer binder to coat and bind the aggregate particles in the geopolymer mixes.

It is also worth discussing the three-dimensional response surface plot and contour diagram for the model of early-age compressive strength, as shown in Fig. 10. The response surface illustrates the relationship among FA content, time, and compressive strength. The surface and contour plots show that the FA content (20–80 %) has a positive correlation with compressive strength as curing time increases. The RSM analysis suggests that the factor influencing the early-age compressive strength is the curing time, which is required for geopolymer mixes with a high FA content.

In addition, all geopolymer mortar mixes met the early-age compressive strength requirement specified for repair products by the ASTM standard [39] (1–7 days), as observed in Fig. 8. For 28-day compressive strength, Fig. 8 shows that the strength increased with higher GGBS content that replaced FA. A further increase was observed with the inclusion of 10 % MK as a partial replacement for GGBS. Generally, the strength performance of all mixes at 28 days also surpasses the maximum strength of 35 N/mm^2 specified by the ASTM standard [39] for repair product Class R3, which is also applicable for structural repair. The results also indicate that only mixes with FA content exceeding 50 % exhibited compressive strength above 45 N/mm^2 at 7 and 28 days. The finding suggests that the geopolymer mortar with 60 % FA content is suitable for structural repair product Class R4 [40]. The high strength of geopolymer mortar is obtained at ambient temperature regardless of the FA content, making it practical for concrete repair materials. Nevertheless, other properties such as flowability and setting are still deemed important factors to be considered.

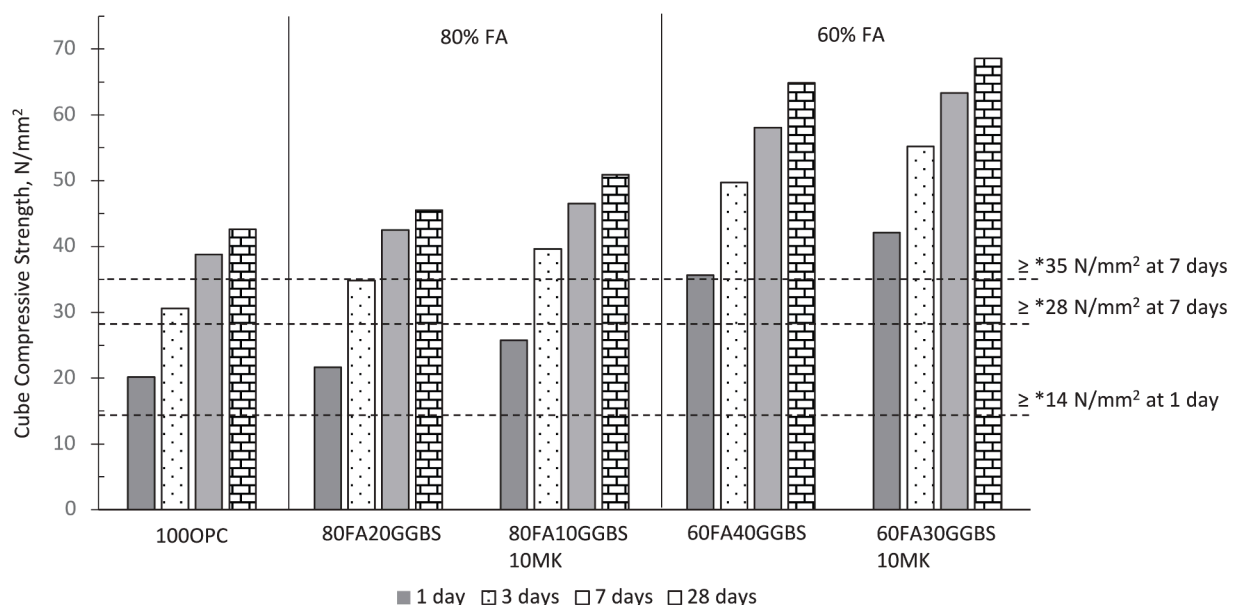


Fig. 8. Compressive strength of FA-based geopolymer mortar.

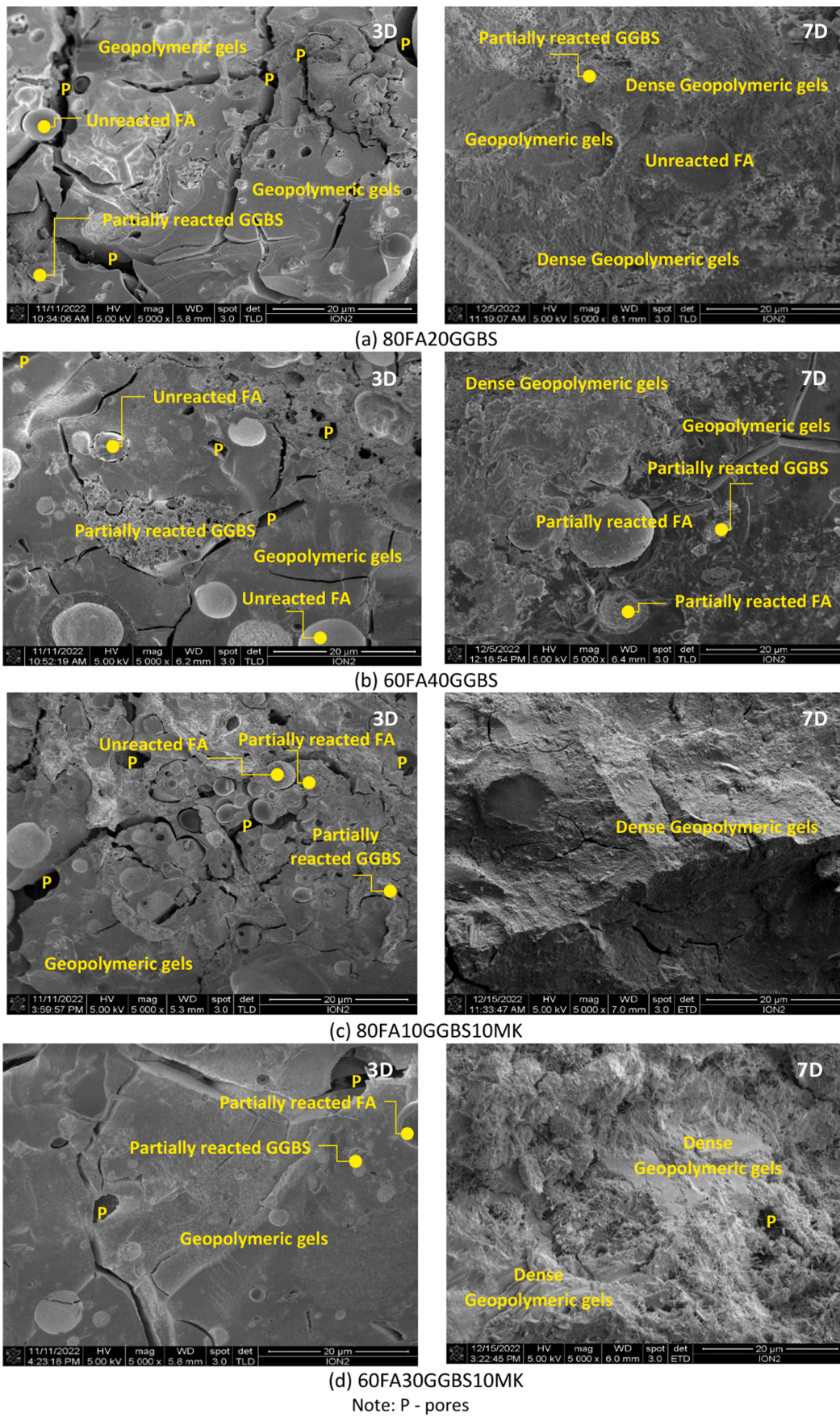


Fig. 9. FESEM images of FA-based geopolymer mortar with high fly ash content.

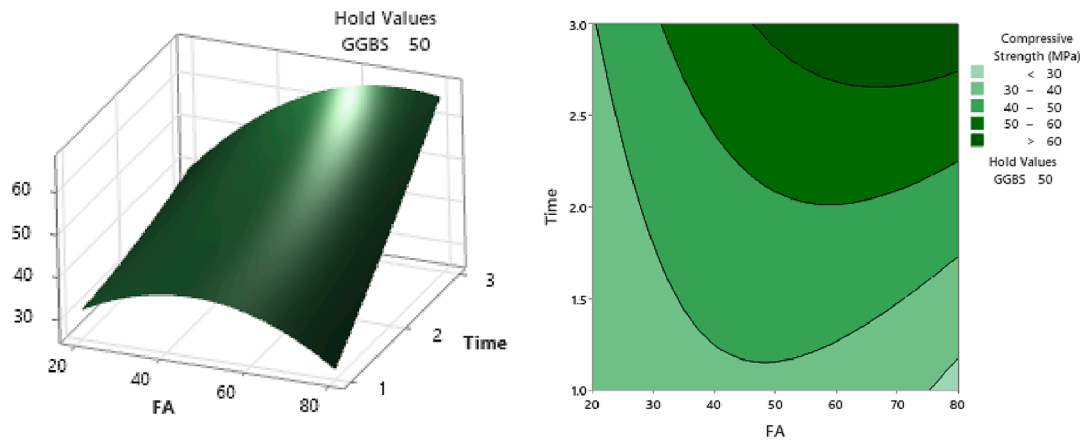


Fig. 10. Surface and contour plots showing the relation between FA content, time and compressive strength.

Table 6

Water Absorption and porosity of geopolymer mortar.

Specimen	Weight of oven dried specimen, W1 (kg)	Weight of specimen after 1 day of immersion, W2 (kg)	Weight of specimen after 3 day of immersion, W3 (kg)	Weight of specimen after 7 day of immersion, W4 (kg)	Initial Water absorption (%)	Final water absorption (%)	Porosity (%)
100OPC	0.2736	0.2743	0.2765	0.2773	0.2254	1.3351	0.3650
80FA20GGBS	0.2132	0.2134	0.2135	0.2135	0.0829	0.1610	0.0343
60FA40GGBS	0.2310	0.2312	0.2313	0.2314	0.0880	0.1443	0.0333
80FA10GGBS 10MK	0.2102	0.2104	0.2104	0.2105	0.0682	0.0650	0.0220
60FA30GGBS 10MK	0.2112	0.2113	0.2113	0.2113	0.0110	0.0521	0.0110

3.3.2. Water absorption

Water absorption of geopolymer mortar at 7 days is presented in Table 6. The results show that the water absorption of FA/GGBS geopolymer mortar is lower than that of 100OPC, and increasing FA content replaced by GGBS decreases water absorption. A significant decrease in

water absorption was observed for the geopolymer mortar with ternary compound systems compared to that of FA/GGBS only. About 64 % lower water absorption was observed for the 60FA30GGBS10MK mortar compared to that of the binary compound only. It is well noted that the ternary system consisting of GGBS and MK exhibited lower water

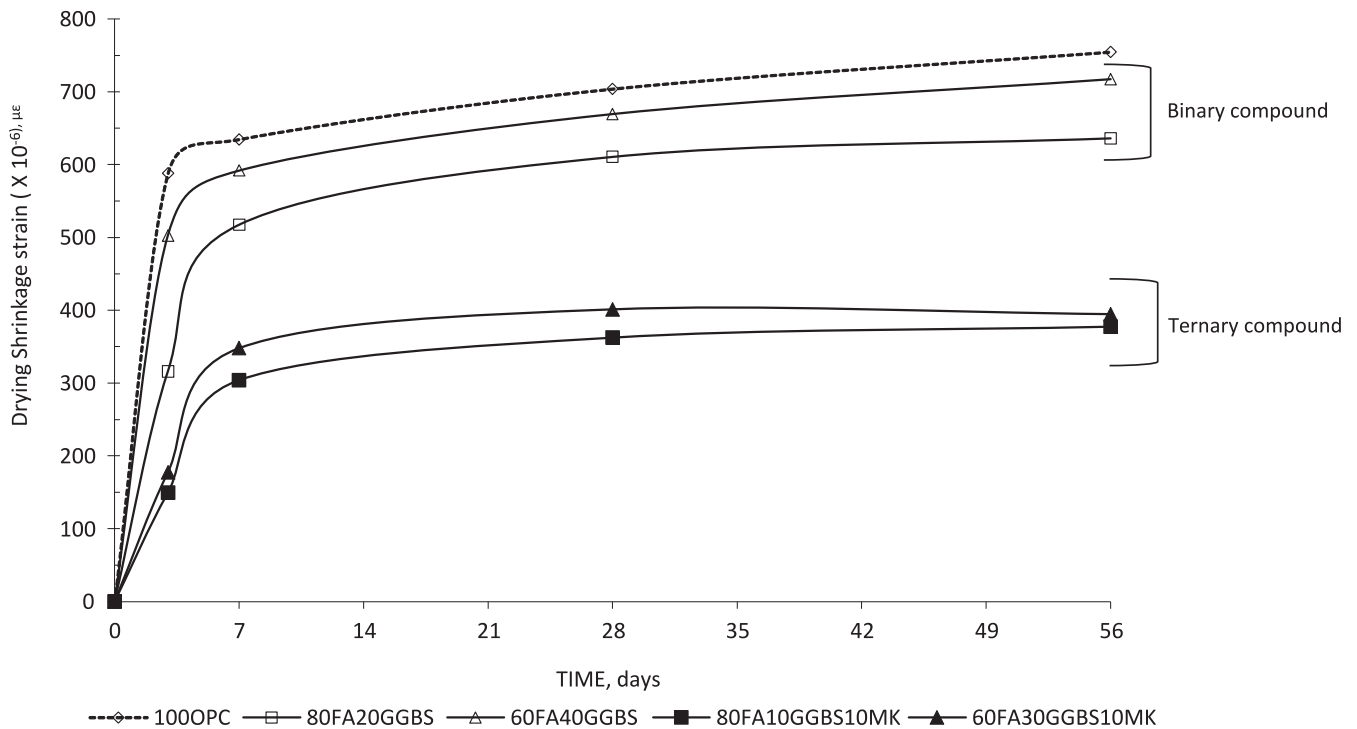


Fig. 11. Drying shrinkage of geopolymer mortar.

absorption due to the improvement in the dense geopolymer matrix. As discussed earlier in Section 3.1, the presence of GGBS enhances the geopolymerization reactivity, resulting in additional sources of Ca and alumina-silica that influence the formation of the binding gel phase [28]. Thus, Ca reacts to form additional CSH and NASH gels, resulting in a denser microstructure within the geopolymer mortar incorporating GGBS. Furthermore, the presence of MK enhanced the geopolymeric matrix both chemically and physically.

3.3.3. Drying shrinkage

Fig. 11 shows the drying shrinkage of geopolymer mortar, which was monitored for 2 months. The results clearly indicate that the binder combination plays a significant role in the formation of the cementitious matrix within the microstructure, which subsequently influences the drying shrinkage behavior of the geopolymer mortar. Shrinkage occurs because the internal water evaporates within the pore network of the microstructure. From the results, the shrinkage rate was high in the early age (up to 3 days), and a minimal rate occurred after this age regardless of binder combinations. Additionally, drying shrinkage increased rapidly at an early age. Zhao et al. [41] and Castel et al. [42] also observed a rapid increase in shrinkage in blended geopolymer FA/GGBS-based composites at an early age, particularly between 3 and 7 days. According to Yang et al. [43], the rapid internal loss of relative humidity at the surface of the sample resulted in early-age drying shrinkage. In this study, a high drying shrinkage was observed for FA/GGBS geopolymer mortar, but the mortar with a high FA content exhibited the lowest shrinkage at the age of 56 days. Yang et al. [43], reported a similar result, where geopolymer composites incorporating a high content of FA exhibited a lower drying shrinkage, which suggested that the refinement of the pore structure restricted the evaporation of internal water. The FESEM results in Fig. 9(a) and 9(b) also support the finding that there is a less porous microstructure on 80FA20GGBS geopolymer mortar in comparison with 60FA40GGBS. Furthermore, a similar trend was observed in the ternary compound system: the higher the FA content, the lower the drying shrinkage of mortar at a later age. However, a significant decrease in drying shrinkage was observed when 10 % MK replaced GGBS in the ternary system. The drying shrinkage decreased by about half compared to that of FA/GGBS geopolymer mortars. The lower drying shrinkage could be attributed to dense geopolymeric microstructure and less interconnected pores within the geopolymer matrix. This finding is in agreement with the study by Kouamo and Tonnayopas et al., where the incorporation of MK densified the geopolymer mortar microstructure and resulted in a significant reduction in drying shrinkage [16].

4. Conclusions

The following conclusions can be drawn from this study:

- Geopolymer composites with high FA content (more than 50 %) exhibit promising strength characteristics at ambient curing. The RSM results show that there is a highly significant relationship among the early strength, FA content, and curing period, as demonstrated by the developed model equation.
- The decrease in strength over time in FA/GGBS geopolymer composites is attributed to the excessive Ca content, which leads to the formation of microcracks within the geopolymer matrix. Additionally, it causes surface dryness within the geopolymer microstructure.
- A combination of FA/GGBS with a high FA content, where the GGBS content ranged from 20 % to 40 %, resulted in high early strength at 3 days. Also, the strength increased steadily at 28 days. Furthermore, the R^2 between the test and predicted values indicated a high level of accuracy in predicting the early strength prediction for the mix design combination.
- There was a significant increase in compressive strength with the incorporation of 10 % MK, which replaced GGBS. The FA-based

geopolymer with a ternary compound achieved the highest compressive strength among all the other mixes.

- The FESEM analysis also shows that the geopolymer mortar microstructure of the ternary compound is denser than that of the binary compound. This is due to the presence of additional CSH, NASH, and CASH gels developed from the geopolymer reactivity. This results in high strength and low shrinkage effects. Also, the water absorption capacity of the geopolymer mortar decreased.
- The SEM-EDX analysis indicates that high Ca/Si and Si/Al ratios lead to more gel formation, which enhanced the strength of geopolymer mortar when combined with a high FA content (maximum GGBS of 40 %). However, the SEM-EDX analysis found that the amount of Ca decreased with age for mixes with a high GGBS content (more than 60 %). This indicates the presence of unreacted particles, which affect gel formation and consequently reduce the strength at later ages.
- 60FA40GGBS and 60FA30GGBS10MK are the most favorable mix combinations that have the potential to be used as cement-based repair products for structural applications.

CRedit authorship contribution statement

Archanaah Nadarajah: Data curation, Formal analysis, Investigation, Writing – original draft. **Noor Azline Mohd Nasir:** Conception and design of study, Funding acquisition, Writing – review & editing, Supervision, Methodology, Formal analysis. **Nabilah Abu Bakar:** Software, Writing – review & editing. **Nor Azizi Safiee:** Methodology, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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