Research Article

Influence of Molarity and Chemical Composition on the Development of Compressive Strength in POFA Based Geopolymer Mortar

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The investigation concerns the use of the optimum mix proportion of two locally available pozzolanic waste materials, namely, ground granulated blast furnace slag (GGBS) and palm oil fuel ash (POFA), together with metakaolin (MK) as binders. In addition, another local waste material, manufactured sand (M-sand), was used as a replacement for conventional sand in the development of green geopolymer mortar. Twenty-four mortar mixtures were designed with varying binder contents and alkaline activators. The oven dry curing was also kept consistent for all the mix proportions at a temperature of 65°C for 24 hours. The highest 28-day compressive strength of about 48 MPa was obtained for the mortar containing 20% of MK, 35% of GGBS, and 45% of POFA. The increment of MK beyond 20% leads to reduction of the compressive strength. The GGBS replacement beyond 35% also reduced the compressive strength. The entire specimen achieved average 80% of the 28-day strength at the age of 3 days. The density decreased with the increase of POFA percentage. The finding of this research by using the combination of MK, GGBS, and POFA as binders to wholly replace conventional ordinary Portland cement would lead to alternate eco-friendly geopolymer matrix.

1. Introduction

The utilization of waste materials or industrial by-products, such as fly ash (FA), metakaolin (MK), silica fume (SF), ground granulated blast furnace slag (GGBS), rich husk ash (RHA), and palm oil fuel ash (POFA), as binders in the development of geopolymer concrete is gaining momentum [1–3] due to the overuse of virgin materials in the production of ordinary Portland cement (OPC). The use of OPC has resulted in a high volume of carbon dioxide (CO₂) emissions, as the construction industries cannot replace the use of OPC entirely with the other cementitious materials due to the huge demand for concrete. The extraction of natural deposits in the production of cement and concrete has led to an ecological imbalance due to the continuous depletion of natural resources. The present annual consumption of 2.9 billion tons of cement globally could increase to 4 billion

tons by 2020 [4]. The estimated value of CO₂ emission in the production of 1 ton OPC is about 0.6 tons [5]. The cement industry is responsible for approximately 5% of global CO₂ emissions and it is likely to reach 6% annually by 2015 [6].

The ratio of clinker to cement is the main fact upon which the CO₂ emissions per ton of cement depend. Normally this ratio varies from 0.5 to 0.95 [7]. The CaO share in clinker amounts to 64–67%. The rest consists of silicon oxides, iron oxides, and aluminium oxides. On the other hand, the production of industrial by-products and other waste materials, such as FA, GGBS, RHA, SF, POFA, and MK, continues to rise and the waste disposal of these materials in an environmentally friendly manner is becoming a challenge. The use of these waste materials in the production of concrete could reduce the greenhouse gas emissions [8]. Geopolymers are usually reported to be much more sustainable than Portland cement, in terms of reduced production energy

FIGURE 1: Geopolymer terminology [11].

necessity, lower CO_2 emissions comparing with the OPC concrete; Turner and Collins (2013) [9] have pointed out only 9% less CO_2 emission from geopolymer concrete production considering curing as well as FA, sodium silicate, and sodium hydroxide production. The emission of CO_2 from geopolymer concrete may control considering emissions during curing as well as production of FA, sodium silicate, and sodium hydroxide. Geopolymers are a class of synthetic alkali-activated alumina silicate inorganic polymers (AIPs) featuring a predominantly X-ray amorphous three-dimensional network [10]. Glukhovskiy carried out the maiden work on alkali-activated alumina silicate materials in the 1950s, which consisted of mixed alkalis with a burnt mixture of kaolinite, limestone, and dolomite. Later, binders other than cement could be produced by the reaction between alkaline solution and source materials that are rich in silica (SiO_2) and alumina (Al_2O_3). Davidovits was the pioneer in introducing the term known as geopolymer and [11] further categorized the geopolymer structure based on the ratio of Si/Al (Figure 1).

Duxson et al. [12] proposed another model for the mechanism of geopolymerisation (Figure 2), which consisted of (a) dissolution, (b) speciation equilibrium, (c) gelation, (d) reorganization, and (e) polymerization and hardening. The type of alkaline activator plays an important role in the geopolymerisation process and has a significant effect on the mechanical strength of the geopolymer.

Provis and van Deventer (2009) pointed out the reality of the geopolymer gel structure and the Davidovits sialate nomenclature implies certain aspects of the geopolymer gel structure which do not correspond to reality [14]. Provis (2014) has recently presented latest definition of the geopolymer structure. Provis [15] reported that geopolymers are produced through the reaction of an aluminosilicate, normally supplied in powder form as an industrial by-product or other inexpensive material, with an alkaline activator, which is

FIGURE 2: Geopolymerisation-conceptual model [12].

usually a concentrated aqueous solution of alkali hydroxide, silicate, carbonate, or sulfate.

The commonly used alkaline activators in current times are combinations of sodium hydroxide solution (NaOH)

and sodium silicate solution (Na_2SiO_3) with different $\text{Na}_2\text{SiO}_3/\text{NaOH}$ mass ratios [16]. Geopolymers can be progressed through a series of several distinct reaction processes from initial pozzolanic activation to final microstructure development, which is very similar to zeolites. The rate of strength development and the chemical reaction of geopolymer concrete are influenced by several factors based on the chemical composition of the source materials, alkaline activators, and curing conditions [17].

Due to the pure alumina silicate content of metakaolin (MK), it was used in the early development of geopolymers and has continued to be used as a raw material. MK is obtained by the calcination of kaolinitic clay at a temperature ranging between 500°C and 800°C , and, of course, it is a pozzolanic material. The comparison between OPC and MK shows that the latter is environmentally friendly as its production requires a much lower calcining temperature and emits 80–90% less CO_2 than the former [18]. MK is a good source of Al_2O_3 and SiO_2 and is highly reactive with alkaline activators and produces a higher degree of geopolymerization. MK is a better material for geopolymers because it has a higher amorphous phase content and smaller sized particles, the compressive strength of MK-based geopolymers increases with an increase of alumina content by up to 20% [19]. Islam et al. [13] reported that with the increase of GGBS content in the matrix containing FA and POFA the compressive strength of geopolymer mortar could be increased. The addition of calcium from other sources in geopolymer based on metakaolin improved its mechanical strength [20]. Depolymerised siliceous structures with raw materials, such as GGBS, exhibit higher dissolution rates under high alkalinity conditions compared with raw materials composed of laminar structures such as MK. Therefore, blending these components can contribute to an improvement in the stability of the system, and, potentially, also to the durability, when compared to the binders prepared from a single raw material.

Malaysia is one of the largest producers of POFA. There are nearly 200 palm oil mills operating in Malaysia, which produce about 100 tons of POFA annually. In addition, although due to the increased plantation of palm oil trees this production rate is likely to increase [21], this material has no market value and is simply dumped into ponds/lagoons [22], which causes environmental pollution. However, recent research [23, 24] has paved the way for the development of sustainable material using POFA and related waste materials from the palm oil industry. Such development has become more significant as a number of studies have identified it as being rich in silica. Alengaram et al. [23] reported that the compressive strength of concrete can improve with the increase in sand content coupled with a reduction in OPS content. POFA is a cementitious material that could be used with recycled aggregate concrete that could result in a higher compressive strength than that of recycled aggregate concrete without POFA [25].

For centuries, river or mining sand was used as fine aggregate even though river sand is a nonrenewable natural resource. The exploitation of river sand endangers the stability of riverbanks and creates environmental problems, such

as flooding and the reduction of ground water level. Thus, the urgency to find an alternate to natural sand is vital. Cai [26] suggested the use of alternative materials for river sand, such as manufactured sand (M-sand), industrial by-products (some forms of slag, bottom ash), and recycled aggregates. Among these, the use of manufactured sand is gaining momentum [26]. M-sand, which is a by-product of quarry dust (QD), is obtained by centrifuging it using a technology known as the vertical shaft impact (VSI). QD, which is the by-product of rock crushing, contains angular and flaky edged particles. Hence, the process of VSI technology produces particle sizes with greater integrity and a more isometric shape. M-sand fines contribute to an increase in paste volume [27] and the shape and texture of M-sand lead to improved strength due to better interlocking between the particles.

During the last two decades, there have been a number of researchers who focused their efforts to utilize geopolymer concrete due to the wide range of potential applications [8, 9, 15]. The effect of single and binary combination of binders among MK, GGBS, and POFA has been reported by previous researchers [28–30]. Islam et al. [13] reported the highest compressive strength of about 66 MPa by using 70% GGBS with 30% POFA. However, the use of large amount of GGBS leads to workability and setting time problems. Sahana [31] reported that the inclusion of GGBS at different replacement levels below 40% increases the setting time of mortar, but beyond this level, the setting time decreases and this could lead to loss of workability and casting of concrete; Deb et al. [32] described that the workability decreased with the high content of GGBS due to accelerated reaction of the calcium and the angular shape of GGBS. Islam et al. [13] also used 12-molarity sodium hydroxide solution to obtain the maximum compressive strength; however the effect of higher molarity was not investigated.

Thus these research questions, such as the loss of workability and early setting due to high GGBS content, have to be addressed. In order to address this issue, an attempt has been made through this research work to minimize the use of GGBS content within 20–45%; and at the same time to ensure the required compressive strength, POFA and MK were used along with GGBS. Therefore, the research focus of this work is to utilize ternary binders and at the same time to optimize the local waste material, POFA as binder. Utilization of 45% of POFA along with 35% and 20% of GGBS and MK, respectively, was experimented and the main flaw of low workability was investigated. The use of hydroxide solution with 12 and 14 molarities on the compressive strength was also carried out.

The variables investigated in the research are different percentages of binder, sand, and two molarities of alkali activators to obtain maximum compressive strength. In this study M-sand was used as fine aggregate for the replacement of conventional mining sand. The other parameters, such as M-sand and water contents, were kept constant. The flow ability, oven dry density, and compressive strength at different ages of cube specimens were tested and reported in this investigation. A total of 24 mixes were prepared to obtain the optimum mix that could be used for further works.

FIGURE 3: MK, GGBS, and POFA's particle size distribution (log scale).

2. Experimental Programme

2.1. Materials

2.1.1. Metakaolin. Metakaolin (MK) is an industrial by-product material, it has significant potential in the development of concrete composites, and it is a highly reactive pozzolanic admixture. MK is manufactured from a natural geological mineral that often contains impurities and inconsistent Si:Al atomic ratios. The chemical composition of MK and the physical properties are given in Tables 1 and 2, respectively. It can be seen from the chemical composition that MK contains 90% silica and alumina. The physical properties show that MK has a specific gravity and specific surface area of 2.5 and 2.16 m²/gm, respectively. The particle size distribution of MK is shown in Figure 3, which shows that MK is much finer than GGBS or POFA.

2.1.2. Ground Granulated Blast Furnace Slag. Ground granulated blast furnace slag (GGBS) was used as one of the source materials to produce a cementless binder. GGBS was collected from YTL Cement Marketing Sdn Bhd, Malaysia. GGBS has both cementitious and pozzolanic properties and is different from other supplementary cementitious materials. GGBS develops its own hydraulic reaction when mixed with water. GGBS is off-white in colour. The combination of calcium, silicates, and alumina comprising about 90% of GGBS shows that it satisfies the requirements of a pozzolanic material. Table 1 shows the chemical composition of GGBS, and its physical properties are given in Table 2. The specific gravity is 2.89 g/cm³ and specific surface area is 2.36 m²/gm. The particle size distribution of GGBS is shown in Figure 3.

2.1.3. Palm Oil Fuel Ash. Palm oil fuel ash (POFA) is a waste material; it is obtained from palm oil mill boilers by

TABLE 1: The raw materials (MK, GGBS, and POFA) chemical composition (wt %).

Chemical composition	MK	GGBS	POFA
CaO	0.04	45.83	4.34
SiO ₂	52.68	32.52	63.41
Al ₂ O ₃	42.42	13.7	5.55
MgO	0.12	3.27	3.74
Na ₂ O	0.07	0.25	0.16
SO ₃	0.05	1.8	0.91
P ₂ O ₅	0.4	0.04	3.78
K ₂ O	0.34	0.48	6.33
TiO ₂	1.46	0.73	0.33
MnO	0.08	0.35	0.17
Fe ₂ O ₃	2.01	0.76	4.19
SrO	0.03	0.08	0.02
Cl	—	0.02	0.45
CuO	—	—	6.54
LOI	—	0.6	6.2

TABLE 2: Materials physical properties.

Properties	Materials			
	MK	GGBS	POFA	M-sand
Specific gravity	2.5	2.89	2.2	2.78
Specific surface area (m ² /gm)	2.16	2.36	1.65	—
Colour	Off-white	Off-white	Dark	—

the burning of palm oil husk and shell as fuel. However, this ash has pozzolanic properties that play an active role in making strong and durable concrete by the replacement of cement. Raw POFA was collected from the local palm oil industry, Malaysia. The incompletely combusted fibres and kernel shells were separated by using a 300 μm sieve. Before that, they were dried in an oven for at least 24 h at 100°C to remove the moisture. The POFA was then ground in a ball mill to obtain particle sizes of about 10 μm. Forty mild steel rods of 10 mm diameter and 400 mm length were placed in the rotating drum to grind approximately 10 kg of POFA one time before being sieved through 300 μm. To obtain the desired level of fineness (>66%), the grinding of POFA was carried out for 30,000 cycles over 16 h. The mass of natural pozzolan passing a 45 μm wet sieve should be at least 86% based on ASTM: C618-12a. The POFA was greyish in colour. The physical properties of POFA are shown in Table 2. Its particle size distribution is shown in Figure 3. The fineness of POFA was found to be 88.7%. Table 1 shows the chemical analysis of POFA. The chemical analysis suggests that, in general, this POFA satisfies the requirement to be considered pozzolanic and may be grouped between Class C and Class F pozzolana, as specified in ASTM C618-92a.1. Figure 4 shows the palm fruits and the raw POFA in factory.

FIGURE 4: Palm fruits (left) and raw palm oil fuel ash (right).

2.1.4. Manufactured Sand (M-Sand). M-sand was collected from the local industry in Malaysia. The shape and texture of M-sand particles could lead to improvements in the strength of concrete due to better interlocking between particles. The physical properties of M-sand are shown in Table 2. The fine aggregate is separated into three grades based on the percentage passing through standard sieves according to BS 882-1992. M-sand falls into the category of grade C sand. The packing ability of M-sand's finer particles enables the mortar to achieve higher packing density, which could enhance the durability of the concrete. Figure 6 shows that the M-sand has a wide range of particles (metakaolin (MK), palm oil fuel ash (POFA), ground granulated blast furnace slag (GGBS), and manufactured sand (M-sand) are shown in Figure 5).

2.1.5. Alkaline Activators. Sodium hydroxide, silicate, carbonate, and sulphate are the commonly used activators and one of the most common and effective activators are sodium silicate. For the development of geopolymerisation in geopolymer materials, the main contribution of silicate ions is through alkali activation. An activator's efficiency depends on certain factors, such as the type of activator, dosage, ambient temperature, and water to FA (W/FA) ratio. The NaOH was of commercial grade in pellet form (5 mm), with a specific gravity of 2.13 and 97% purity. Na_2SiO_3 in liquid form with a density of about 1.5 gm/mL at 20°C , a modulus ratio of 2.5 ($\text{SiO}_2/\text{Na}_2\text{O}$, $\text{SiO}_2 = 30\%$ and $\text{Na}_2\text{O} = 12\%$, water = 57% by mass), and specific gravity of 1.5 was used along with NaOH as an alkali activator according to [33]. Two different molarities (namely, 12 M and 14 M) were used by using sodium hydroxide solution to obtain the optimum strength. For instance, NaOH solution with a concentration of 12 M consists of $12 \times 40 = 480$ g of NaOH solids per litre of the solution, where 40 is the molecular weight of NaOH. The mass of NaOH solids was measured as 361 g per kg of NaOH solution of 12 M concentration [34]. The reaction was exothermic, so the solution was made to cool down before use. The ratio of $\text{Na}_2\text{SiO}_3\text{aq}/\text{NaOH}$ was kept constant at 2.5 for all the mixtures.

2.1.6. Water. Potable water with a pH value of 6 and free from impurities and chemical contaminants was used in all

the mixes. The water to binder ratio of 0.25 was used for all the mixes.

2.2. Specimen Preparation and Curing

2.2.1. Mixing Procedure and Casting. A total of 24 mixes were prepared. The mix proportions of all the concrete mixes are shown in Tables 3 and 7. By varying the MK, POFA, GGBS, and activator contents, the mixtures were prepared. To investigate the effect of the binders, the M-sand and water contents were kept constant. The ratio of binder to sand content was kept at 1:2 in all the mixes. In order to mix the ternary binders of MK, POFA, and GGBS along with sand, these materials were mixed in dry condition in a bowl mixer at a low rate of speed for about 5 min. The alkali activator solution was then slowly added into the mix for another 5 min and the mode of speed was then changed from slow to high. Then, additional water was added to the mix to increase the workability and homogeneity of the mortar. The mortar was then cast in 50 mm cube moulds and poured in three phases with appropriate compaction. A total of 12 specimens were cast for each mix proportion. To remove the entrained air and bubbles the samples were vibrated with standard compaction using a rod and vibrating table. A comparison between Islam et al. [13] and the present research work on mixing and casting time shows that the former required finishing the casting within 7 min, while the duration could be extended to 10 min in the present research. Another salient point in the comparison is the total binder content; the present research has total binder content of about 760 kg/m^3 compared to 460 kg/m^3 used by Islam et al. [13]. Thus, the use of such high volume of binder with large quantity of GGBS would have reduced the setting time drastically [31]. Hence, the proposed mixing time with different speed of mixer is crucial in balancing the mix and workability.

2.2.2. Curing Regime. Immediately after casting, the specimens along with the moulds were covered using plastic film to reduce water loss during curing in an oven for 24 h at 65°C . Subsequently, the specimens were taken out of the oven and kept at ambient condition with an average temperature and humidity of 28°C and 70%, respectively,

FIGURE 5: (a) Metakaolin (MK), (b) palm oil fuel ash (POFA), (c) ground granulated blast furnace slag (GGBS), and (d) manufactured sand (M-sand).

FIGURE 6: The particle size distribution of M-sand [13].

until testing day. This procedure was adopted from the method suggested by Hardjito and Rangan [34]. To gain high early strength geopolymers with acceptable physical and mechanical properties, the optimum geopolymer oven curing temperature was between 60°C and 65°C [35]. Therefore,

the selection of 65°C was justified as the 50 mm cubes have high surface to volume ratio and are more susceptible to curing heat and to moisture loss.

2.2.3. Compressive Strength. The compressive strength test was done using an ELE Auto Compressive Testing Machine in accordance with ASTM: C109/C109M-13. The compressive strength of the specimens was tested after 3, 7, 14, and 28 days. Each day's compressive strength value was determined as the average of three specimens. The compressive strength of the specimen was determined by dividing the maximum load carried by the specimen during the test by the average cross-sectional area. The failure mode of the specimens is shown in Figure 7(a) and that was found satisfactory as specified in BS EN 12390-3:2002 (Figure 7(b)). Compressive strength = Failure load/Cross-sectional area.

2.2.4. Flow Test. The flow test was done according to the ASTM C230 standard [36]. The used apparatus comprised a flow table, flow mould, tamper, trowel, and measuring tape. The flow test can give an indication as to the consistency, filling ability, and workability of the mortar.

TABLE 3: Mix design for development of compressive strength of geopolymer mortar.

Mortar designation	Mix proportion (%) MK/GGBS/POFA	MK content (kg/m ³)	GGBS content (kg/m ³)	POFA content (kg/m ³)	Compressive strength (MPa)	
					3-day	28-day
M1	5/35/60	38.3	268.3	460	18.88	23.5
M2	10/30/1960	76.7	230	460	33.89	39.2
M3	10/35/55	76.7	268.3	421.7	21.91	30.34
M4	10/45/45	76.7	345	345	36.59	40.85
M5	15/20/65	115	153.3	498.3	31.01	38.57
M6	15/35/50	115	268.3	383.3	22.14	32.47
M7	15/40/45	115	306.7	345	35.51	40.24
M8	20/20/60	153.3	153.3	460	25.18	33.65
M9	20/25/55	153.3	191.7	421.7	37.15	41.05
M10	20/28/52	153.3	214.7	398.7	32.92	37.78
M11	20/30/50	153.3	230	383.3	36.46	42.19
M12	20/35/45	153.3	268.3	345	43.2	48
M13	20/40/40	153.3	306.7	306.7	34.93	41.62
M14	20/45/35	153.3	345	268.3	33.85	40.92
M15	25/30/45	191.7	230	345	35.86	41.43
M16	25/35/40	191.7	268.3	306.7	33.45	37.23
M17	30/40/30	230	306.7	230	33.5	39.6
M18	30/35/35	230	268.3	268.3	32.95	40.73

Note: MK: metakaolin; GGBS: ground granulated blast furnace slag; POFA: palm oil fuel ash.

FIGURE 7: (a) Failure mode of cubes. (b) Satisfactory failures of BSEN 12390-3:2002.

3. Results and Discussion

3.1. Flow. The flow of mortar is shown in Figure 8, while the flow values are shown in Table 4. A lower flow ability of the mortar was observed for mix M5, which could be due to the higher percentage of POFA. POFA decreases the flow tendency because of the porous and spongy nature of the microstructure (Figure 9) [2] of these materials and an increased fineness or surface area. Figure 11 and Table 4 show that the flow percentage increased in mixes M16, M17, and M18, which could be attributed to a decrease in the percentage of POFA (Table 3) in the mortars. It is also observed in Figure 11 that the flow percentage increased with the increase in the MK percentage in the mortars because MK has a much finer particle size than POFA and GGBS and requires less water to obtain a flow [37]. Workability in geopolymer depends on the viscous properties of the binder matrix. Si plays major role in producing viscosity [13]. The percentage of SiO₂ is not so much different between mixes M1 and M18; that

is why the flow has no significant differences between each of the mixtures.

3.2. Oven Dry Density (ODD). The oven dry density (ODD) was taken for all specimens after oven-drying at a temperature of 65°C for a period of 24-h. Figure 10 and Table 4 show the 3-day oven dry density (ODD) of the specimens with different replacement levels of MK, GGBS, and POFA. The ODD of the mortar varied between 1639 kg/m³ and 2181 kg/m³. The spherical particles of all the materials, compared to the crushed particles in a wet state, could produce a higher packing density, resulting in lower water retention in the spherical case, and, consequently, lower water demand [38]. Thus, the use of a high percentage of GGBS with spherical particles produced the highest ODD of 2181 kg/m³ among all the mixes. The POFA, due to its agglomerated and crushed shape and increasing interparticle friction particles, cannot easily roll over one another. Therefore, this highlights

TABLE 4: Flow of mortar and 3-day average oven dry density (ODD) (kg/m^3) of mortar for all mixes.

	Mix number																	
	M1	M2	M3	M4	M5	M6	M7	M8	M9	M10	M11	M12	M13	M14	M15	M16	M17	M18
Flow (%)	61	62	64	67	60	67	67	68	69	72	71	72	70	73	75	76	79	78
Density (kg/m^3)	1874	1789	1870	2115	1639	1896	2007	1691	1739	1756	1821	1914	2067	2181	1833	1943	2089	1961

FIGURE 8: Flow of mortar.

FIGURE 9: Field emission scanning electron microscopy (FESEM) images of POFA [2].

the need for more evaporable water in mixes with high POFA content in order to obtain a workable mix. Hence, the use of a higher dosage of POFA decreases the specimen density [2]. The use of 65% of POFA in mix 5 shows the lowest ODD of 1639 kg/m^3 . Another factor that influences the density is the ability of finer particles to fill the voids within the mortar. Figure 3 shows that POFA has relatively coarser particles within a narrow range compared to that of GGBS and MK. Thus, a high percentage of GGBS (mix 14) with finer particles enhances its density by about 25% compared to mortar with a high percentage of POFA (mix 5). Khatib and Wild [39] reported that the threshold value for pastes decreased with the increase in MK content. The ODD of the mortar decreased slightly by about 2.2% in the first few weeks but remained almost constant thereafter. Figure 11 shows the reduction in density for all mixes (mix 14).

3.3. Compressive Strength Development. The development of the compressive strength of hardened geopolymer mortar

FIGURE 10: 3-day average density (kg/m^3) for all mixes. FIGURE 11: Density (kg/m^3) reduction for all mixes from the peak of density and the percentage of flow of mortar.

is the basic indicator of the performance of alternative source materials, since it provides a fundamental description concerning the quality of geopolymerization products. The development of compressive strength at the ages of 3, 7, 14, and 28 days is shown in Figure 12. As indicated in the methodology, the specimens were cured at 65°C for 24-h. Curing at an elevated temperature allows a reduction in the time of heat treatment to achieve high strength [40]. It can be observed from Figure 12 that mix M1, which contained 5% MK, 35% GGBS, and 60% POFA and was cured at 65°C for 24-h, produced the lowest compressive strength. However, an increase in the MK from 5% to 10% and

FIGURE 12: Compressive strength development of mortar at different ages with varying binder content ratios.

subsequent decrease in POFA in mix M3 showed an increase in the compressive strength. It can be seen that this trend continued until the MK was increased to 20%. It was observed that with up to 20% replacement of MK by POFA the highest compressive strength of about 48 MPa could be obtained. With any further increase in the MK content beyond 20%, the strength decreased, as seen from the mixes M16 and M18, which had 25% and 30% of MK, respectively. A similar trend was observed for mixes with an increase in both the GGBS and POFA contents. Thus, it can be concluded that mix M12 with 20% MK, 35% GGBS, and 45% POFA with high SiO_2 and Al_2O_3 contents produced the highest compressive strength. It is evident that POFA contained fewer amounts of Al_2O_3 and Ca, when mixed with GGBS, which enhanced the compressive strength. From the results, it is also observed that up to 45% of the binders could be replaced by POFA to achieve a compressive strength of 48 MPa. However, an increase of POFA beyond 45% had a negative effect on the compressive strength. Thus, the amount of Ca and Al_2O_3 had a significant influence on the compressive strength of the mortar [41]. From Figure 12, it can also be seen that the compressive strength curve is steep at the age of 3 days with a slight drop at 7 days, which may be because the Si/Al ratio varies during the geopolymerization process [42]. Table 5

TABLE 5: The percentage (%) increase in the compressive strength at 3, 7, and 14 days compared with the 28-day strength.

Mortar designation	Increase of compressive strength (%)		
	3-day (%)	7-day (%)	14-day (%)
M1	80	85	90
M2	87	90	96
M3	72	78	91
M4	90	93	97
M5	80	84	89
M6	68	73	85
M7	88	93	97
M8	75	81	88
M9	91	94	100
M10	87	90	98
M11	86	88	93
M12	90	94	97
M13	84	87	94
M14	83	91	96
M15	87	87	95
M16	90	96	100
M17	85	89	96
M18	81	81	86

shows the increase in the compressive strength at 3, 7, and 14 days compared to 28 days expressed as a percentage. It can be seen from a comparison of the 3-day test results with the 28-day strength that most of the specimens achieved about 80% of the 28-day strength. Correspondingly, the 7-day and 14-day strengths were 87% and 94%, respectively, of the 28-day strength (Figure 15).

3.4. Analysis of Chemical Composition on the Development of Strength. The parameters, such as curing temperature, water content, alkali concentration, initial solids content, silicate and aluminate ratio, pH, and the type of activators used, influence the rate of polymerization [13]. The comparison of the major oxide composition of the three materials, that is, MK, GGBS, and POFA, is shown in Table 6. As explained in Section 3.3, the mixes M1 and M12 produced the lowest and the highest compressive strengths, respectively. It can be seen from Table that the SiO_2 content was found to be higher in mix M12 compared to the other mixes, for which the ratio of Si/Al for mix M12 was 3.26. In contrast, the Si/Al ratio for M1 was found to be 5.12. The initial Si/Al ratio will not be constant all the way through the geopolymerization process. During different stages of geopolymerization the Si/Al ratio increases [42]. The trend of compressive strength development is affected by the change in the Si/Al ratio in the original particles, the reactive ones, and the reacted product during the reaction process [43]. Geopolymerization was almost complete after 7 days and that the strength gain beyond this period was found to be insignificant. Lime (CaO) plays a very important role as it controls the strength and soundness, whereas excess CaO

TABLE 6: The major chemical components of mortar.

Chemical components	Mortar designation																	
	M1	M2	M3	M4	M5	M6	M7	M8	M9	M10	M11	M12	M13	M14	M15	M16	M17	M18
CaO	18.68	16.42	18.5	22.65	12.09	18.32	20.39	11.91	13.99	15.23	16.06	18.14	20.21	22.28	15.88	17.95	19.85	17.77
SiO ₂	52.01	52.97	51.43	48.34	55.48	50.84	49.3	54.89	53.35	52.42	51.8	50.26	48.71	47.17	51.22	49.67	47.54	49.09
Al ₂ O ₃	10.16	11.5	11.91	12.73	12.44	13.66	14.07	14.19	14.6	14.84	15.01	15.42	15.82	16.23	16.76	17.17	19.33	18.92
MgO	3.44	3.32	3.3	3.25	3.23	3.16	3.14	3.09	3.07	3.05	3.04	3.02	3	2.97	2.9	2.88	2.72	2.74
Fe ₂ O ₃	2.81	2.81	2.63	2.29	2.97	2.46	2.29	2.79	2.62	2.52	2.45	2.28	2.11	1.94	2.27	2.1	1.75	1.92
Na ₂ O	0.2	0.2	0.21	0.22	0.2	0.21	0.22	0.21	0.21	0.22	0.22	0.22	0.23	0.23	0.22	0.23	0.24	0.24
K ₂ O	4.07	4.14	3.85	3.26	4.51	3.63	3.34	4.29	4	3.83	3.71	3.42	3.12	2.83	3.49	3.2	2.69	2.98
SiO ₂ /Al ₂ O ₃	5.12	4.61	4.32	3.8	4.46	3.72	3.5	3.87	3.65	3.53	3.45	3.26	3.08	2.91	3.06	2.89	2.46	2.59

content makes the material unsound and causes expansion and disintegration. An excessive quantity of lime (CaO) is considered for the hardening mechanism of mortar [44]. Mix M4 contained the highest percentage of CaO among all the mixes but produced the lowest compressive strength of about 37 MPa. The formation of Ca compounds in geopolymers is greatly dependent on the pH and Si/Al ratio. The SiO₂ content provides greater strength, but, at the same time, it extends the setting time [13]. Although mix M5 contained the highest percentage of SiO₂ among all the mixes, the Si/Al ratio was quite high (4.45). Further, mixes M5 and M1 contained high percentages of K₂O and MgO, respectively, which are harmful ingredients in cement. Mixes M17 and M1 contained the highest percentage of Na₂O and MgO. If the amount of Na₂O and K₂O exceeds 1%, it leads to the failure of concrete and if the content of MgO exceeds 5%, it causes cracks in the hardened concrete. Further, the Al component tends to dissolve easier than the Si components, which enables a higher rate of condensation between the silicate and aluminate species than the condensation between just the silicate species [45].

3.5. Effect of Metakaolin on the Compressive Strength. In general, MK is a poorly crystallized white powder with a specific surface of 12,000 m²/kg (Table 2) and an average particle size between 1.5 and 2.5 μ m [46]. MK is effective at reducing the rate of diffusion of the Cl⁻ and Na⁺ ions in mortar and also the rate of water absorption [47]. The effect of MK content on the compressive strength is shown in Figure 13. The results of Wild et al. [48] derived the maximum compressive strength possible at the different percentage replacements of MK. The use of 20% MK with 45% of POFA and 35% of GGBS in mix M12 produced the highest strength of about 48 MPa. The mixes M1, M3, and M6 that contained 5%, 10%, and 15% MK with 35% GGBS and different amounts of POFA produced about 51%, 37%, and 32% lower compressive strength than mix M12. This is because M12 contained more Al₂O₃ compared to the other mixes. Al plays an important role, as, to a large degree, it controls the properties of geopolymers, and the

FIGURE 13: Effect of MK on compressive strength mixed with 45% POFA and 35% GGBS.

absolute amount of available Al throughout the reaction affects the final compressive strength [49]. Similarly, for mixes M7 and M14, which contained 15% MK, 40% GGBS, and 45% POFA and 20% MK, 45% GGBS, and 35% POFA, respectively, the compressive strength decreased by 16% and 15% compared to mix M12. MK grains are not fully dissolved during the geopolymerization process as the reaction takes place on the surface layer of the solid particles [50]. The reaction rate is then mainly controlled by diffusion of the hydroxide and silicate ions through the primary geopolymer gel, which is mainly affected by the curing temperature at the early stage of the reaction. In the case of increasing the percentage of MK, the compressive strength also reduced compared to mix M12. The inclusion of a small amount of MK in GGBS-rich alkali silicate-activated binders produces stable Al-substituted calcium silicate hydrate (C-S-H) gels and promotes enhanced workability. A low activator content does not provide sufficient alkalinity to promote the dissolution of MK to produce a highly stable coexisting geopolymer gel. Therefore, a large proportion of MK needs activator content with very high concentration that will affect the

rate of polymerization. Thus, the inclusion of MK after 20% reduces the compressive strength. Mixes M16 and M18, which contained 25% MK, 35% GGBS, and 40% POFA and 30% MK, 35% GGBS, and 35% POFA, showed a reduction in the compressive strength by about 22% and 15% compared to M12. According to Austell [51] the rate of water absorption of mortar is halved by replacing 20% of MK. So, mix M12 with 20% MK, 35% GGBS, and 45% POFA produced the highest compressive strength and is considered the optimum mix to develop geopolymer mortar with a compressive cube strength of 40 MPa. Wild et al. [52] reported that increasing the specific surface of MK from 12,000 to 15,000 m²/kg reduces the age at which the maximum strength enhancement occurs in MK mortar. The effect of MK on the compressive strength at 28 days with 35% GGBS and 45% POFA is shown in Figure 13. One of the reasons for mixing MK with GGBS in this study is to provide better control of the setting time. GGBS binders have a tendency to set extremely rapidly and the addition of a small amount of MK can retard setting to provide a more convenient period of workability.

3.6. Effect of GGBS on the Compressive Strength. GGBS plays an important role for compressive strength development. A higher concentration of GGBS results in a higher compressive strength of geopolymer concrete [53]. GGBS contains higher CaO, and, as a result, it is a good potential source of soluble Ca in the mixture. The compressive strength of mortar is shown in Figure 12. Mix number M12, with 35% GGBS, produced the highest strength, while a further decrease in the GGBS content reduced the compressive strength. M11 (30% GGBS with 20% MK, 50% POFA) and M9 (25% GGBS with 20% MK, 55% POFA) showed a decrease in strength of about 12% and 14% with respect to M12. Yunsheng et al. [54] also reported that the addition of GGBS leads to a considerable increase in the compressive strength but only when the GGBS replacement percentage exceeds 30%. GGBS is a latent hydraulic product, which can be activated by suitable activators, and, without activation, the development of the strength of the GGBS is extremely slow and the development of the slag necessitates a pH12 [44]. Mix M13 contained 40% GGBS with 20% MK, 40% POFA, while mix M12 contained 35% GGBS with 20% MK, 45% POFA. A comparison between mixes M12 and M13 showed that the former with 40% GGBS produced 13% lower compressive strength compared to the mix with 35% GGBS and that mix M14 with 45% GGBS showed that the reduction is 15%. Therefore, if the percentage of GGBS exceeds 35%, then the compressive strength also reduces. This might be because the GGBS undergoes a slower hydration process; therefore at higher GGBS replacement levels, the hydration process becomes increasingly slower, which leads to a reduction in the compressive strength [55]. Another reason is that the quantity of soluble Ca depends on the volume of GGBS present in the mixture, which has a direct effect on the compressive strength. Figure 14 shows the effect of GGBS on the compressive strength with POFA and 20% MK.

3.7. Effect of POFA on the Compressive Strength. POFA, which is also a by-product from thermal power plants, contains

FIGURE 14: Effect of GGBS on compressive strength mixed with POFA and 20% fixed MK.

high amounts of Si and Al oxides in the amorphous state and is considered to be a pozzolanic material [56]. The high fineness of POFA produces a greater pozzolanic reaction and acts as a filler in the voids and thus increases the compressive strength of the concrete [21]. Figure 16 shows the effect of varying POFA contents on the compressive strength with MK varying from 5% to 30% with nonvarying GGBS at 35%. The compressive strengths of mixes M1 and M3, which contained 60% POFA and 55% POFA, respectively, produced about 51% and 37% lower compressive strength than mix M12. Mix M12 with 45% of POFA, 35% of GGBS, and 20% MK produced the highest strength of 48 MPa. Ariffin et al. [57] opined that due to its low content of Al₂O₃ it produces low compressive strength; the use of high POFA content could be attributed to the incomplete geopolymerization, because Al₂O₃ tends to dissolve at a higher rate during the early stage of geopolymerization. The comparison between mixes M12 and M13 showed that the former with 40% POFA produced 13% lower compressive strength compared to the mix with 45% POFA. Then, from the comparison between mixes M12 and M14 (35% POFA), the strength reduced by about 15% for the latter.

The particles of the POFA with cohesive characteristics could not be mixed properly, and, hence, the strength development was poor. Tonnayopas et al. [58] explained that the low early compressive strength is due to the slow pozzolanic activity of POFA. Figure 14 shows the 28 days' compressive strength of all mixes.

3.8. Effect of Curing. All the specimens were covered with plastic wrapping and cured in an oven at 65°C for 24 h, as stated in the methodology; the results for the compressive strengths are shown in Figure 12. The average difference between the 14- and 28-day compressive strength for the mixes was about 10%, and, between the 7- and 14-day strengths, it was 8%. This shows that the rate of the geopolymerization is accelerated by elevated temperature at the early stage [59]. Kusbiantoro et al. [60] reported that compared to the other curing methods—ambient and external exposure—the oven-cured geopolymer concrete at 65°C exhibited superior mechanical properties. They reasoned that this could

FIGURE 15: 28-day compressive strength of different mixes.

FIGURE 16: Effect of POFA on compressive strength mixed with MK and 35% fixed GGBS.

be attributed to the oven curing as the suitable condition to accelerate the dissolution and polycondensation of alumina silicate gel in the geopolymer framework. Furthermore, geopolymer concrete could achieve high early compressive strength when oven-cured, instead of undergoing ambient curing [61]. Hence, all the mixes, M1 to M18, gathered maximum strength within 3 days, as shown in Table 5.

3.9. Alkaline Solution to Binder Ratio by Mass on Compressive Strength Analysis. The solution to binder ratio was kept constant at 0.5 for all the mixtures, as stated in Table 4. Abdullah et al. [62] reported that the ratio of S/B of 0.5 by mass produces higher compressive strength compared to the ratio of 0.67. Sathonsaowaphak et al. [63] found a very low compressive strength of 8 MPa for the mix with the AK/B ratio of 0.325 by mass; however, they reported that the high compressive strengths were in the range of 42 to 52 MPa, as the ratios of S/B by mass varied between 0.429 and 0.709. Thus, the use of a solution to binder ratio of 0.5 for all the mixtures produced acceptable compressive strength.

3.10. Effect of Molarity of Alkaline Activated Solution (NaOH Solution) on the Compressive Strength. Table 7 shows the effect of molarity of alkaline activated solution on the compressive strength.

The compressive strength of mix M4 with 14 molarities produced 17% higher than the mix M4A with 12 molarities. Other parameters except the molarity, such as binder, sand, and water, were kept constant for both the mixes. The use of high molarity based alkaline solution increases the compressive strength because of the release of silica and alumina [64]. From Figure 17 it was also found that the mixes M9, M12, and M16 with 14 molarities show higher compressive strength than the mixes M9A, M12A, and M16A with 12 molarities. All the parameters are same, respectively, except the molarity (NaOH solution). NaOH solution with high concentration increases the geopolymerization reaction [43].

4. Conclusions

This research into optimizing the use of MK, GGBS, and POFA as binders in the production of geopolymer mortars produced significant conclusions. Twenty-four geopolymer mortar mixtures incorporating sustainable binders—MK, GGBS, and POFA—were prepared. The cube compressive strength of the mortars was investigated up to a period of 28 days to study the feasibility of the replacement with GGBS and POFA in MK-based geopolymer mortar. Based on the tests conducted, the following conclusions were drawn:

- (i) The compressive strength of the geopolymer mortar increased with the MK content up to 20%, while further replacement of MK showed a significant reduction in the strength.
- (ii) Mix M12, with the addition of GGBS up to 35%, POFA up to 45% with 20% MK, produced the highest strength among the mixes.
- (iii) It is possible to improve the compressive strength of the geopolymer mortars at the early ages by introducing initial heating at 65°C for 24 h after demoulding. Improvements are more significant at 3 days.
- (iv) In most of the specimens, 90% of the compressive strength of the geopolymer mortar was achieved at the age of 7 days.
- (v) POFA produces a cohesive mix, in which the density of mortar decreases with the increase in the percentage of POFA, resulting in a decrease in density of approximately 12%.
- (vi) The finer particles of GGBS produce a dense mix; hence, the partial substitution of GGBS in the mortar should be maintained at 35%.
- (vii) The compressive strength increases with the increases of molarity of sodium hydroxide solution.
- (viii) It can also be concluded that the combined maximum volumes of MK, GGBS, and POFA can be used for the development of a sustainable construction material to replace OPC for the production of eco-environmentally friendly geopolymer mortars.

TABLE 7: Comparison of the compressive strength between different molarities.

Mortar designation	Binder mix proportion MK/GGBS/POFA (%)	M-sand (kg/m ³)	Alkali activator molarity	Compressive strength (MPa)	
				3-day	28-day
M4	10/45/45	1532	14	36.6	40.85
M6	15/35/50	1532	14	22.15	32.5
M9	20/25/55	1532	14	37.15	41
M12	20/35/45	1532	14	43.15	48
M16	25/35/40	1532	14	33.4	37.5
M18	30/35/35	1532	14	34	40.8
M4A	10/45/45	1532	12	30.8	34.1
M6A	15/35/50	1532	12	17.2	25.8
M9A	20/25/55	1532	12	32.5	37
M12A	20/35/45	1532	12	36	41.5
M16A	25/35/40	1532	12	27.6	33
M18A	30/35/35	1532	12	31.1	35.6

Note: MK: metakaolin; GGBS: ground granulated blast furnace slag; POFA: palm oil fuel ash.

FIGURE 17: Effect of molarity in compressive strength.

This research work was focused on the development of the compressive strength of geopolymer mortar produced by different proportion of three pozzolanic materials and activated by two concentrated alkaline solutions. The test results were analysed based on the chemical compositions. Geopolymerization process could be affected by the particle size and reactivity rate of the binding materials, curing condition, and casting process. This could be recommended for further research work on the microstructural analysis for the proposed binding materials with different particle size and various curing conditions. Since water/binder ratio is an important factor and affects significantly the development of compressive strength for different proportion of binder/aggregate in concrete and mortar, mix design with different type and volume of aggregate could be carried on for optimization.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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