1. Introduction

The Portland cement is produced from finely ground clinker. The finely ground clinker is a substance composed mainly by hydraulically active calcium silicate minerals. This substance is formed via high temperature burning of limestone and other materials in kiln [1]. During the process of producing cement, it demanded about 1.7 tons of raw materials per ton of clinker, and the process discharges approximately 1 ton of carbon dioxide to the atmosphere. Therefore, this horrifying carbon dioxide discharge causes the cement industry to become main sources of greenhouse gases in the aspect of industrial manufacturer [2, 3]. Rapid economic growth has increased cement production in the construction industry sector, thus causing problems to the environment [4]. The cement industry is facing a lot of struggles including environmental issue and sustainable issue [4]. This is because the cement manufacture will not only discharge carbon dioxide but also nitrogen oxides (NOx), sulfur dioxide (SO2), and grey dust to the atmosphere [5]. The activities that lead to air pollution in cement manufacture including excavation activities, crushing mills, and kiln emissions. A survey regarding the air pollutants emission in Kuala Lumpur was carried out and the result clearly observes that the respirable...
dust (PM10) emission from the cement industry is the highest among the other pollutants [6]. Thus, cement industry can cause severe air pollution if no proper solution is taken.

Besides, the study carried out by Sharma et al. [7] proved that the cement industry pollutant can cause 5% to 17% of reduction in Forced Expiratory Volume (FEV). Thus, it could lead to severe respiratory diseases if the person is exposed to the pollutant for a long time. Therefore, a method was proposed to reduce the negative effect toward the environment and increase the quality of concrete, which is applied certain type of material for partial replacement for the manufacture of Portland cement. This material can be industrial or agriculture wastes. The blending between Portland cement and this waste will produce a mixture known as “composite cement” or “blended cement” [8]. The performance of composite cement will still be similar with the general Portland cement as they will react when mixed with water and become harder, in which finally the same hydration products are formed. The most common material used as the replacement material is pozzolanic material such as fly ash, condensed silica fume, and rice husk ash [3, 8]. When the Portland cement is used to carry out the hydration reaction, a chemical substance Ca(OH)₂ will produce as one of the by-products and lead to the deterioration of concrete. However, by using pozzolanic material which is composed of amorphous silica to carry out the hydration process of cement, additional calcium silicate hydrate (CSH) will produce, which is also the dominant cementing component. Thus, it is obvious that the application of pozzolanic material can significantly increase the quality of concrete, however, a study from Zeyad et al. [9] stated that the replacement of pozzolanic material should not contain more than 60% of cement as the excess replacement will cause increasing demand for superplasticiser, water, and reduce the workability [9]. Therefore, in this project, treated palm oil fuel ash and metakaolin were chosen as the pozzolanic material. Palm oil fuel ash (POFA) was chosen to use as the pozzolanic material in this project because palm oil industry is one of the most developed agriculture industry in Malaysia. According to the global palm oil production research in 2017, Malaysia was known as the second most in the production of palm oil [10].

According to the research from the United States Department of Agriculture, the combination of palm oil production globally in year 2016 and 2017 will be about 64.5 million metric tons [10]. Large quantity of waste in the form of remnants of fibers and kernels will be produced after the extraction of palm oil. According to the research, about 3.14 million tons of palm oil fuel ash was produced in Malaysia annually, while Indonesia produced about 3.7 million tons of POFA annually [11]. Thus, we can conclude that our country can supply large amount of palm oil fuel ash. Furthermore, accumulation of large amount of palm oil fuel ash may lead to several environmental issues and health problem if failed to maintain or dispose it properly. Besides, the palm oil fuel ash is also being applied to enhance the durability characteristic of concrete. Another reason for choosing palm oil fuel ash in this project is that research stated that 40% of microfine palm oil fuel ash replacement can enhance the workability and viscosity of concrete [12]. Moreover, the POFA also proved that it can decrease heat emission throughout the hydration process as it can be applied to treat the thermal cracking due to drastic increase in temperature in mass mortar [13]. POFA also can improve the resistance of concrete toward acidic and sulfate environment. The POFA also have the advantage of the capability to increase shrinkage of concrete and decrease the heat development [13].

The metakaolin was chosen as the pozzolanic material in this project because it can bring several advantages to the concrete including increase the compressive strength, increase the flexural strength, and increase the resistance toward chemicals. The metakaolin can also decrease shrinkage that is caused by particle packing and indirectly raising the strength and density of concrete [14]. The properties of metakaolin stated above were related to this project which is effect toward workability and compressive strength. According to the research, concrete composed of metakaolin was proved to decrease the expansion caused from alkali-silica reaction and chloride ions penetration process [15]. It also proved that the metakaolin can refine the pore structure of cement paste of concrete by forming a secondary CSH [15]. The application of metakaolin as pozzolanic material can also bring advantages such as improving the resistance of concrete toward acidic and sulfuric environment. The other reason for selecting metakaolin as pozzolanic material applied in this project is that it can decrease porosity as an addition of 2% of metakaolin and result in the decrease of porosity by 10% [16].

The research regarding application of palm oil fuel ash has been studied and investigated deeply since the late nineteenth century. The optimum percentages of palm oil fuel ash replacement for cement would be 10% to 20% as the higher percentage of POFA will decrease the compressive strength of concrete [17]. There was also much research showing that the ground POFA can be used as supplementary cementing material in high strength concrete due to its excellent pozzolanic property. The POFA will be sieved before being applied in the experiment because fine POFA produced will form a more precise result and indirectly increase the strength of concrete. The purpose of the application of metakaolin is to enhance the compressive strength of the concrete. However, the accurate percentage of the application of POFA and metakaolin was also an issue. The outcome of this research will summarize the application of palm oil fuel ash, superplasticiser, and metakaolin together with their effectiveness in the compressive strength of concrete. From this finding, it could help the other researchers in this field in having more understanding and information about the application of POFA and MK and how it affects the compressive strength of sustainable high strength concrete. The information can be used for further study in this scope.

2. Materials and Method

The primary materials of this study were ordinary Portland cement (OPC), POFA, MK, sand, gravel, water, and superplasticiser. POFA and MK wastes used as semi-cement
substitutes were ensured that their fineness was below 10 micrometres and was heat-treated at 500°C and 700°C for one hour, respectively. The POFA and MK are Class F and Class N, respectively, based on ASTM 618 [18]. The sizes of the fine and coarse aggregates used averaged under 2.36 mm and 14 mm, respectively. The superplasticiser used is compliant with the ASTM C494 type F [19] admixture which improves the workability of concrete.

Table 1 shows the properties of the materials used through the particle size analyser and air permeability methods. Specific surface area and particle size were obtained from the particle size analyser while Blaine surface area was obtained using BS EN 196 [20]. Table 2 shows the chemical composition of the binders used in this study. OPC contains a total amount of silicon dioxide, aluminum oxide, and iron oxide of 28.86%. In comparison, POFA and MK were 73.43% and 93.23%, respectively. Figures 1–3 shows element analysis using Scanning Electron Microscope Energy Dispersive X-ray Analysis (SEM-EDX), particle size analysis for POFA, MK, fine aggregate, and coarse aggregate.

2.1. Specimen. The mixing of concrete containing semi-substitute cement is carried out based on the substitution of the cement mass with the mass of cement substitute material. Eight concrete mixtures containing semi-substitute cement and a control concrete mixture were conducted for the success of this study. These nine mixtures were conducted to prepare the samples for workability testing, microstructural analysis. Slump test was carried out on fresh concrete mix before the concrete mix was put into prism for compressive strength tests. Concrete water absorption tests were also conducted on mature samples using 55 mm diameter and 40 mm thickness concrete cores extracted from 500 mm × 100 mm × 100 mm prism concrete and the tests were performed based on RILEM CPC11.3 [23]. The compound phrases in the dust samples were characterised using XRD equipment that complied with BS EN 13925-1 [24] and then analysed using X’Pert HighScore Plus software. Based on the analysis, portlandite or calcium hydroxide (CH) and tobermorite or calcium silicate hydrate compounds were detected in most of the dust samples in zones 1 and 2 as shown in Figure 4. Counts of CH in zones 1 were analysed to understand the concrete mechanisms in this study that contribute to concrete strength, and for counts of CSH analysed in zone 2 on dust samples aged 7, 28, and 90 days as in Figure 5. Zone 1 is between 34° and 35° and zone 2 is between 51° and 52° of 2Θ. Nima et al. [25] studied that there is a CH intensity value in the scan range 34°. Haider et al. [26] also stated that there is portlandite material at scan range 34°. The study of Bamaga et al. [27] also showed portlandite material at scan range 34°. Data for scan range 51° has been found to have minerals such as quartz which with the addition of chemical components such as SiO₂ can give more compressive strength to concrete. Yusof et al. [28] stated in their research of POFA samples also found that quartz is a chemical composition found in the scan range 51°. Elahi et al. [29] found similar results on the MK sample that said the chemical composition of quartz on the scan range 51°. Research from Haider et al. [26] also said that the chemical composition for OPC control in the scan range 51° is quartz. The increase in value for each sample indicates CSH formation at scan range 51°.

3. Results and Discussion

3.1. Slump Test. Results from a slump test conducted on nine fresh concrete mixes found that the partial replacement of cement with MK and POFA reduced the workability of the concrete. The partial replacement of cement with 10% MK reduced the workability of concrete by 61.7%, while the partial replacement of cement with POFA by 40% only experienced a workability reduction of 29.4%. The workability of concrete containing POFA is better than concrete containing MK by 32%. The Blaine surface area of MK (0.881 m²/g) which is wider than POFA (0.79 m²/g) causes the concrete containing MK to absorb higher water than the concrete containing POFA, hence the concrete containing MK experiences a higher reduction in workability. Apart from this, Zoe et al. [30] stated that fine MK acts as a filler filling the spaces between binder, sand, and coarse aggregate causing fresh concrete to be more stiff and lower workability. The workability of concrete binaries containing MK and POFA is very low, and the slump difference is very high compared to concrete binaries containing only MK or POFA. Therefore, using a higher superplasticiser is required to improve the workability of concrete ternary (Figure 6).
3.2. Slump Flow Table Test. The results of the slump flow test of concrete mix containing MK and POFA are better than the slump test. The results of the slump flow test for nine fresh concretes are shown in Figure 7. The test results found that the partial replacement of cement with POFA and MK reduced the workability of fresh concrete by reducing the diameter of fresh concrete. Partial replacement of cement with 10% MK reduced the diameter of fresh concrete by 25.4%, while partial replacement of cement with POFA by 20% and 40% showed a reduction in the diameter of fresh concrete by 26% and 25.4%, respectively. The ternary diameter of concrete also experienced a reduction in workability between 15.9% and 31% compared to OPC concrete.

3.3. Density. Figure 8 shows the density of concrete in this study from the age of 7 days to 180 days. Partial replacement of cement with MK and POFA lowers the overall concrete density. Thus, the density of concrete containing MK and POFA is lower than that of ordinary concrete. Concrete containing a total of 40% cement substitute material showed the lowest concrete density compared to other concretes in this study. All concretes in this study experienced an increase in density from the age of 7 days to 180 days. The increase in concrete density from the age of 7 days to 180 days indicates the increase of mass in the concrete due to the reaction between cement, MK, POFA, and water that causes the addition of cement hydration and secondary hydration products. Sofri et al. [31] also experience the same situation where total concrete densities decreases when cement is replaced by POFA.

3.4. Water Absorption of Concrete. From Figure 9 concrete containing MK and POFA cement substitutes has lower water absorption than ordinary concrete. Concrete containing more cement substitutes shows lower water absorption. Concrete in the group of total cement substitutes of 40% showed the lowest water absorption followed by the group of total cement substitutes of 20%, 10%, and 5%. The pores of the concrete become smaller or smaller causing the water absorption in the concrete to decrease. The cause of reduced water absorption is from the addition of hydration cement and secondary hydration product in concrete in parallel with the increase in concrete density from the age of 7 days to 180 days. In addition, there is a strong trend in the absorption of water that reduces as POFA material increases [32].

3.5. Compressive Strength. Table 4 and Figure 10 are the results of compressive strength tests for nine different types of concrete from the age of 7 days to 180 days. M5 concrete containing 5% MK and 95% cement was the strongest concrete at 7 days concrete age with 11% stronger than OPC concrete, while P40 concrete containing 40% POFA and 60% cement was the weakest concrete in this study at 7 days age with 11% weaker than OPC Concrete. Next is P15M5 concrete containing 15% POFA and 5% MK, i.e., the strongest ternary concrete at the age of 7 days with 0.8% stronger than OPC concrete. This study of Ahmad et al. [33] obtained the same result, whereby the replacement of POFA with 15% has obtained the highest optimum compressive strength. The finer MK than cement acts as a filler to reduce the pores in the concrete and the aluminum Oxide content in the MK influences the hydration reaction by increasing the initial strength of the M5 and P15M5 concrete. To add on, Arslan et al. [34] mentioned that MK in cement functions as a filler, accelerating cement hydration at the early age of 3 days. By speeding up the hydration process, more CH is formed, and this CH combines with the active silica and aluminate phases of pozzolans, resulting in secondary calcium aluminosilicate hydrates at an early stage. The study by Narmatha & Felixkala [35] also obtained similar results, where the replacement of cement with MK increased the early strength of the concrete. MK concrete strength increases are effective only at the early age of concrete and the strength increase is only marginal in the long term [35].

Excessive cement replacement inhibits the hydration reaction resulting in a reduction of the early strength of P40 concrete. Studies also state that the compressive strength of

### Table 1: Properties of material.

| Material   | Type/properties                                                                 | Specific surface area (m²/g) | Particle size mean (μm) | Blaine surface area (m²/g) |
|------------|--------------------------------------------------------------------------------|-----------------------------|-------------------------|---------------------------|
| Cement     | Normal Portland cement                                                        | 1.16                        | 16.4                    | 0.32                      |
| POFA       | POFA passing sieve 300μm, refined for 8 hours in ball mill and Heat treatment 500°C for 1 hours | 1.72                        | 9.64                    | 0.79                      |
| MK         | Kaolin via heat treatment 700°C for 1 hours                                    | 1.48                        | 8.36                    | 0.881                     |
| Coarse aggregate | Passing sieve 14 mm                                                              |                             |                         |                           |
| Fine aggregate | Passing sieve 5 mm                                                              |                             |                         |                           |
| Water      | Tap water                                                                     |                             |                         |                           |

### Table 2: Chemical composition of materials.

| Chemical composition (%) | OPC     | POFA    | MK      |
|--------------------------|---------|---------|---------|
| Silicon dioxide (SiO₂)   | 20.63   | 60.78   | 55.04   |
| Aluminum oxide (Al₂O₃)   | 4.71    | 6.96    | 37.44   |
| Iron oxide (Fe₂O₃)       | 3.52    | 5.69    | 0.75    |
| Calcium oxide (CaO)      | 60.98   | 5.20    | 0.02    |
| Magnesium oxide (MgO)    | 0.98    | 4.17    | 0.81    |
| Sodium oxide (Na₂O)      | 0.06    | 0.10    | —       |
| Potassium oxide (K₂O)    | 1.09    | 5.40    | 2.31    |
| Sulfur trioxide (SO₃)    | 4.97    | 0.22    | —       |
| Loss of ignition (LOI)   | 2.38    | 3.00    | 2.50    |
| SiO₂+Al₂O₃+Fe₂O₃         | 28.86   | 73.43   | 93.23   |
POFA at an early age is lower than OPC [32]. Conditions changed when the concrete age reached 28 days, P40 concrete underwent a high strength change with a strength increase of 36% from 56 MPa to 76.5 MPa. P40 concrete at 28 days of age was 6% stronger than OPC concrete.

M5 concrete which is the strongest concrete at the age of 7 days experienced a strength increase at a low rate of 9% from 69.9 MPa to 76.5 MPa at the age of 28 days. M5 concrete was 6.3% stronger than OPC concrete at 28 days of age. For ternary concrete, P35M5 concrete showed a better increase in strength than other ternary concretes and P35M5 concrete was 3.6% stronger than OPC concrete at 28 days. Ismail et al. [36] obtained a similar result as well, whereby the replacement of POFA partially in cement increases the
compressive strength. It also highlights that higher compressive strength is achieved due to its fines [36].

Concrete containing heat-treated POFA contains high silica content that begins to be reactive after the concrete age of 7 days. The high silica content in concrete and the smaller size of POFA than cement led to an increase in hydration and pozzolanic reactions which resulted in an increase in strength in concrete containing POFA at 28 days.

After 28 days of concrete age, concrete containing MK cement substitute showed a decreasing pattern of increasing concrete strength. In comparison, binary and ternary concretes containing POFA and MK cement substitutes continue to show a pattern of continuous increase in strength. Sam et al. [37] documented that the progressive strength of concrete containing POFA and MK increases due to the pozzolanic response, which usually improves over age. The synergetic effect of POFA and MK in concrete can be seen in ternary concrete where the increase in strength of ternary concrete continues to increase with the age of concrete.

3.6. X-Ray Diffraction Analysis. Figure 11 shows the counts of CH in zone 1 (Figure 5), while Figure 10 shows the counts of CSH in zone 2 (Figure 5) analysed from XRD scan results up to a concrete age of 90 days. Substitution of cement with POFA and MK causes the CaO content to decrease in concrete causing the hydration product of CH to decrease [38]. The results of zone 1 analysis found that concrete containing POFA and MK as cement substitutes contained less CH content than OPC concrete in line with the study [38]. However, CH content continued to increase in parallel with hydration reaction until the concrete age of 90 days. No increase in CH content was recorded for P40, CH content for P40 continued to decrease with concrete age. The decrease in CH content in concrete containing more than 5% cement substitute material at 28 days of concrete age is the effect of the reaction of CH with silicon dioxide and aluminum oxide to form more CSH and calcium aluminosilicate hydrates (CASH) in concrete. The synergetic effect of POFA and MK can be seen in ternary concrete with higher CH content for ternary concrete containing 20% cement substitute material compared to ternary concrete containing 40% cement substitute material. The unhydrated CaO in POFA and unhydrated cement reacts with the water present promoting the growth of CH content in concrete. This is because, when the degree of hydration of the mineral admixture is low and the amount of CH that has not reacted with the mineral admixture is significant, the consumption rate of CH increases due to a large number of mineral admixtures [39].

The results of zone 2 analysis in Figure 12 found that the replacement of cement with silica-rich cement substitute increased the CSH content in concrete. Concrete containing more than 5% cement substitutes experienced a decrease in CSH content at an early stage compared to OPC. Replacement of cement with POFA or MK resulted in a decrease in CaO content in concrete resulting in a reduction in CSH production compared to OPC concrete [40]. Nevertheless, the CSH content in concrete continues to increase with the pozzolanic reaction in which silicon dioxide reacts with CH to form more CSH between the concrete ages of 7 to 90 days. However, the CSH content in concrete containing POFA and MK could not match the CaO-rich OPC concrete. The CSH content of M5 concrete was higher than OPC concrete at 7 days, but the production of CSH content of M5

Figure 2: Particle size analysis of OPC, POFA, and MK (Mastersizer 2000 Ver 5.4, Malvern Instruments Ltd).

Figure 3: Coarse and fine aggregate sieve analysis.
concrete decreased after 7 days. M5 concrete containing MK is more likely to react in the early stages of the hydration process. The high CSH production for P40 and P30M10 concretes after 28 days of concrete age is due to the result of a pozzolanic reaction in concrete, in which CH reacts with silicon dioxide to form more CSH. According to Ash et al. [38], the pozzolanic reactions involve a secondary reaction whereby the CH and silica content integrate to form the strength by contributing CSH. This will develop the strength of concrete beyond 28 days.

3.7. Influence of Water Absorption, Calcium Hydroxide, and Calcium Silicate Hydrate on Compressive Strength. The strength of concrete is influenced by the rate of absorption of water, CH, and CSH in the concrete. OPC concrete experiences high water absorption compared to other concretes. OPC concrete also contains high CH and CSH compared to other concretes. Although the CSH content of OPC concrete is high, it does not contribute to a higher strength than other binary and ternary concrete because OPC concrete contains high porosity, which contributes to high water absorption. High porosity of OPC concrete results in low strength of OPC concrete. Cement replacement with POFA and MK over 5% substitution causes the concrete to contain less CaO, which is required in the hydration process to produce CH and CSH. The lack of CaO in concrete causes the production of CH, and CSH is to be reduced. Nevertheless, the presence of more silicon dioxide than POFA and MK contributes to the pozzolanic reaction which increases the CSH content in concrete. This unhydrated silicon dioxide reacts with CH to produce CSH in concrete which causes a decrease in CH content in concrete. Like OPC concrete, the strength of these binary and ternary concretes is influenced by the porosity of the concrete, CH, and CSH content. Binary and ternary concretes with a low water absorption rate, lower CH

| Mix    | OPC (kg/m³) | POFA (kg/m³) | MK (kg/m³) | Total (kg/m³) | Sand (kg/m³) | Gravel (kg/m³) | Water (kg/m³) | SP (kg/m³) |
|--------|-------------|--------------|------------|---------------|--------------|----------------|---------------|------------|
| OPC    | 500         | —            | —          | 500           | 750          | 1250           | 150           | 15         |
| M5     | 475         | —            | 25         | 500           | 750          | 1250           | 150           | 15         |
| M10    | 450         | —            | 50         | 500           | 750          | 1250           | 150           | 15         |
| P20    | 400         | 100          | —          | 500           | 750          | 1250           | 150           | 15         |
| P40    | 300         | 200          | —          | 500           | 750          | 1250           | 150           | 15         |
| P15M5  | 400         | 175          | 25         | 500           | 750          | 1250           | 150           | 15         |
| P35M5  | 300         | 175          | 25         | 500           | 750          | 1250           | 150           | 15         |
| P10M10 | 400         | 50           | 50         | 500           | 750          | 1250           | 150           | 15         |
| P30M10 | 300         | 150          | 50         | 500           | 750          | 1250           | 150           | 15         |

Figure 4: Compound scanning using X'pert highscore software.
Figure 5: Samples counts analysis by zone for 7 to 90 days age.

Figure 6: Concrete slump test results.
content than OPC concrete, and have high CSH production increase contributes to higher concrete strength. P40 concrete showed low water absorption results, in low CH content but increased in high CSH production resulting in P40 concrete being stronger than other concretes. Adebayo et al. [41] also explained in their research that the compressive strength of concrete is influenced by the presence of CH and CSH in the pozzolanic reaction.
Concrete containing POFA and MK has lower workability compared to OPC concrete. By adding superplasticiser content, the workability of concrete containing POFA and MK can be improved. In addition, concrete containing POFA and MK has a lower density of concrete than OPC. Concrete containing POFA and MK also has a lower water absorption rate than OPC concrete, where the high-water absorption in concrete is the cause of the low strength of concrete. Meanwhile, concrete containing POFA and MK has lower CH and CSH content than OPC concrete. Nevertheless, the increase in high CSH production contributed to the increase in the strength of concrete containing POFA and MK.

Data Availability

The data used to support the findings of this study are included within the article.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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