

Research Article

Development of a Zero-Cement Binder Using Slag, Fly Ash, and Rice Husk Ash with Chemical Activator

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The increasing demand and consumption of cement have necessitated the use of slag, fly ash, rice husk ash (RHA), and so forth as a supplement of cement in concrete construction. The aim of the study is to develop a zero-cement binder (Z-Cem) using slag, fly ash, and RHA combined with chemical activator. NaOH, Ca(OH)₂, and KOH were used in varying weights and molar concentrations. Z-Cem was tested for its consistency, setting time, flow, compressive strength, XRD, SEM, and FTIR. The consistency and setting time of the Z-Cem paste increase with increasing RHA content. The Z-Cem mortar requires more superplasticizer to maintain a constant flow of 110 ± 5% compared with OPC. The compressive strength of the Z-Cem mortar is significantly influenced by the amounts, types, and molar concentration of the activators. The Z-Cem mortar achieves a compressive strength of 42–44 MPa at 28 days with 5% NaOH or at 2.5 molar concentrations. The FTIR results reveal that molecules in the Z-Cem mortar have a silica-hydrate (Si-H) bond with sodium or other inorganic metals (i.e., sodium/calcium-silica-hydrate-alumina gel). Therefore, Z-Cem could be developed using the aforementioned materials with the chemical activator.

1. Introduction

Cement and concrete are important materials for rapid urbanization. Concrete is considered as one of the key elements in the social, economic, and infrastructural development of human society. Ordinary Portland cement- (OPC-) based concrete is one of the significant construction materials in the globe. Up to now, we are still dependent on cement and concrete for the construction of many infrastructures such as buildings, towers, roads, bridges, flyovers, tunnels, industries, and river training works. Approximately 25 billion tons of concrete are produced and consumed annually all over the world. Cement consumption increased to 2.9 billion tons globally in a year and is predicted to increase to 4 billion tons by 2020 [1]. An alternative technology for the production of concrete without the use of cement does not currently exist. OPC is an energy-intensive material after steel and aluminum. OPC emits huge quantities of CO₂ gas to the atmosphere, about 7% of global emission [2]. These phenomena have necessitated the use of supplementary and/or

pozzolanic materials such as slag, fly ash (FA), and rice husk ash (RHA) in cement and concrete manufacturing. However, the use of pozzolanic materials, either natural, agricultural, or industrial by-products, in the production of composite cements has gained increasing interest because of various ecological, economical, technical, and diversified product quality reasons [3]. Sustainable concrete manufactured from pozzolanic materials is one of the solutions recommended by several researchers in the past few decades [2, 4, 5].

Pozzolans are fine materials that contain silica and/or alumina. They do not exhibit any cementing properties of their own, only in the presence of calcium oxide or calcium hydroxide. Silica and alumina in pozzolans also react and form cementitious materials [6]. Huge quantities of pozzolanic materials are regularly produced as wastes in every part of the world. All of these pozzolanic wastes are usually discharged into ponds, rivers, and lagoons without any commercial return. For instance, slag is a pozzolana generated as industrial by-product from steel industries. Approximately 100 million tons of slag is produced annually

worldwide. However, only 35 million tons of slag is used and the rest is disposed [2]. FA is another by-product generated from coal-operated power stations. About 900 million tons of FA is produced annually worldwide [5]. RHA is generated as a waste from rice-processing mills. Approximately 110 million tons [7] of rice husk (about 20% of 550 million tons of rice) and 16 million tons to 22 million tons of RHA are produced globally. No potential and/or alternative uses have been proposed for RHA. These wastes are not only accountable for spending huge expenses for transportation and/or removal but are also the cause of enormous environmental pollutions. These pozzolanic materials contain a high amount of silica and alumina in amorphous or crystal forms. Many researchers have recommended the use of these pozzolans as supplement of cement and as constituent of concrete because of their engineering, technical, financial, and environmental significance. The utilization of slag [8], FA [9], and RHA [10] significantly improves the strength and durability of mortar and concrete. A quaternary-blended cement was developed by Lai [11] by using slag, palm oil fuel ash, RHA, and timber ash with 66% OPC replacement, which was also used in high-strength (100 MPa to 120 MPa), sustainable, and high-performance concrete. Ehrenberg and Geiseler [12] reported that in manufacturing blended cement, the inclusion of slag and FA considerably reduces energy demand and CO₂ emission. Therefore, the use of pozzolans in cement and concrete construction is a valuable and appropriate step for solving or minimizing the important issues on CO₂ emission, sustainable concrete, environmental pollution, and demand/consumption of cement.

Pozzolans were used as partial replacement of cement in most of the past research. Some studies were conducted on the production of geopolymers [13–16]. Geopolymer concrete is produced from aggregates with FA/slag that are activated using highly concentrated sodium hydroxide with sodium silicate (or potassium hydroxide with potassium silicate). The curing process of geopolymer concrete is usually performed at approximately 60°C to 90°C. Recently, Karim et al. [17, 18] reported a preliminary investigation for the development of an alternative cementitious binder by using slag + FA [17] and slag + palm oil fuel ash + RHA with only 5% sodium hydroxide [18] and calcium hydroxide by weight of binder, separately. However, they did not investigate the effects of types and contents (by weight and molar concentration) of the activator and its different content on the strength of the alternative cementitious binder. Considering all of these parameters, the current study investigates the possibility of developing a zero-cement binder (Z-Cem) using slag, FA, and RHA through the mechanical (i.e., grinding of materials) and chemical (using chemical activators) activation technique. To activate these materials, sodium hydroxide (NaOH), calcium hydroxide (Ca(OH)₂), and potassium hydroxide (KOH) were used by weight of Z-Cem and by their molar concentrations.

2. Materials and Methodology

2.1. Materials. Ground granulated blast furnace slag, FA, and RHA were used as raw materials. OPC type I was used to

compare the different properties of Z-Cem (i.e., physical, chemical, binding, flow, compressive strength, morphological, and microstructure). The slag was provided by a local cement company. FA was obtained from Klang power station, Selangor, Malaysia. RHA was produced using a special type of furnace available at the concrete and structure laboratory in University Kebangsaan Malaysia. The details of the furnace were reported by Zain et al. [19]. Standard sand manufactured by Societe Nouvelle Du Littoral, France (certified by the international organization of standardization) was used as fine aggregate for the preparation of the mortar. NaOH, KOH, and Ca(OH)₂ flakes of analytical grade, which were used as chemical activators, were obtained from Merck. Viscocrete 315 brand obtained from Sika Manufacturer was used as a superplasticizer (SP) to increase and maintain a sufficient flow for casting. The Viscocrete 315 is a high-range water reducer based on polycarboxylate-ether (PCE). It is a low-viscosity liquid that has been formulated by the manufacturer for use as received. Drinking water that is available in the concrete laboratory was used for the preparation of the mortar and in curing purposes.

2.2. Tests on Materials. The physical and chemical properties of the materials and those of Z-Cem were determined in different laboratories. The following tests and instruments were used to examine several properties of materials: the fineness of the materials was determined using an automatic Blaine machine. The grain size of the materials was analyzed using Malvern Mastersizer 2000. The amorphous or crystal phase of the materials was examined using X-ray diffraction (XRD) test. The chemical composition of materials was determined using X-ray fluorescence (XRF) test with a Bruker brand XRF machine. The morphological view and shape of the particles of the materials were examined by scanning electron microscopy (SEM) analysis by using Supra 55 VP (ZEISS).

2.3. Preparation of Paste and Mortar Specimens. Table 1 presents five different mixing proportions of the raw materials (i.e., slag, FA, RHA, SP, activators, and sand) for the development of Z-Cem. The fineness of Z-Cem was significantly improved by grinding RHA in a grinding machine. However, slag and FA were used as received from the industry. NaOH, KOH, and Ca(OH)₂ were used as chemical activators. The water-to-binder ratio (W/B) was kept at 0.55 to obtain a reasonable flow for easily casting. The solution-to-binder (S/B) ratio of the activator was kept as required to maintain a constant flow of 110 ± 5% for the mortars. The paste and mortar were mixed and prepared using a Hobart mixing apparatus according to ASTM C305 specification [20] via the two methods described below. The activators were used by weight (%) of Z-Cem and by their molar concentrations. Given that the same amount (%) of Ca(OH)₂, KOH, and NaOH produces different molar concentrations in the same amount of water, we used them in the same molar concentration.

2.3.1. Preparation of Mortar Using Activator by Weight. The activators were used according to the weight of the Z-Cem

TABLE 1: Materials used for the preparation of Z-Cem and OPC mortars (wt.%).

Binder	Raw materials (%)				SP	Activators	BSEN 196-1		ASTM C109	
	Slag	FA	RHA	OPC			W/B	Sa/B	S/B	Sa/B
Z-Cem 1	60	40	0	0	According to necessity	NaOH or KOH or Ca(OH) ₂	0.55	3.0	0.55 to 0.62	2.75
Z-Cem 2	55	35	10	0						
Z-Cem 3	50	30	20	0						
Z-Cem 4	40	30	30	0						
Z-Cem 5	70	20	10	0						
Cement	0	0	0	100	—	—	0.50	3.0	0.50	2.75

binder (i.e., 2.5%, 5.0%, and 7.5%). The W/B, S/B, and sand-to-binder (Sa/B) ratios are listed in Table 1. The required activator was dissolved in water. Generally, Ca(OH)₂ dilutes instantly, whereas KOH and NaOH need a little more time to be diluted. The NaOH flakes were diluted within 60 ± 10 seconds. The Z-Cem mortars can be cast easily for a minimum flow of 50%. Thus, to maintain a minimum mortar flow of 50% for easy casting, SP was used in various doses when necessary. After dilution of the activator, all raw materials (powder sample of Z-Cem, sand, chemical activator, water, and SP) were mixed using a Hobart mixture machine based on ASTM C305 [20] testing specification. Mortar prisms of 40 mm × 40 mm × 160 mm in size were prepared according to BS EN196-1 [21] testing procedure. Mortar specimens of 50 mm × 50 mm × 50 mm in size were also cast according to ASTM C109 specification [22]. The mortars were poured into the moulds and were compacted using a mechanical shaking table.

2.3.2. Preparation of Mortar Using Activator with Different Molar Concentrations. The mortar was prepared using different molar concentrations of the activators. The activators were produced at 1.0, 2.5, 5.0, and 7.5 M before preparing the Z-Cem mortar. Mortar cubes were cast according to ASTM C109 [22] specification by using the required materials in Table 1. The mortar was poured into the cube molds and was uniformly compacted using a vibrator table. Finally, molds of the Z-Cem mortars were opened after two days because the specimens could collapse if immersed into water for less than two days. The mortar specimens were then immersed in a water tank at room temperature of $25 \pm 2^\circ\text{C}$ for curing until desired testing ages of 3, 7, 14, and 28 days.

2.4. Testing of Paste and Mortar. The consistency and setting time of the pastes were determined according to ASTM C187 [23] and ASTM C191 [24] by using a Vicat apparatus. Mortar flow spread test was performed using a flow table based on the ASTM C1437 testing procedure [25]. The compressive strength of the mortar was determined according to BS EN196-1 [21] and ASTM C109 [22] testing procedures by using a universal testing machine (unit test Sdn Bhd brand with 1000 kN capacity). The mortar was also evaluated using SEM and XRD analysis. To determine the types of chemical bonding characteristics in the mortar, Fourier transform infrared (FTIR) test was performed using a PerkinElmer machine.

3. Results and Discussion

3.1. Properties of Materials. Various material properties such as chemical composition, fineness, specific gravity, XRD analysis, SEM analysis, and strength are presented and discussed below.

3.1.1. Chemical Composition. Table 2 presents the chemical properties of the materials. Among all materials, RHA contains the highest amount of silica (87.55%). The loss on ignition (LOI) is higher for FA but is less than 10%, as mentioned in ASTM C618 [6]. The SO₃ content of these materials is less than 4% based on the same standard, whereas the SO₃ content is nearly 1% for Z-Cems. LOI falls within the ASTM limit for all Z-Cem samples. Z-Cem has more than double amounts of silica, three to four times of alumina, and one-half to one-third part of the CaO content compared with OPC. The sum of the major oxides (SiO₂ + Al₂O₃ + Fe₂O₃) of Z-Cems ranges between 50% and 70%, which is greater than 50% (minimum) according to ASTM C618 for Class-C pozzolan [6]. Thus, Z-Cem could be considered as Class-C pozzolana.

3.1.2. Fineness. The particle sizes of the pozzolanic materials and Z-Cem samples are also shown in Table 2. The average particle sizes of slag, FA, RHA (ungrounded, UG), RHA (grounded, G), and OPC are 14.87, 14.45, 8.80, 6.72, and 16.79 μm, respectively. These results indicate that the slag, FA, and RHA (G) have smaller particle sizes compared with OPC. The materials have high specific surface areas (lower grain size), except for FA. The slag has an extensively high specific surface area of 1.155 m²/cm³, which is more than 1.5 times that of OPC. All of the Z-Cem samples have an average particle size of 10 ± 2 μm. All of the Z-Cems also have fineness that is 3.5 times greater than that of OPC. Z-Cems have specific surface areas that are 2.5 times greater than that of the OPC sample.

3.1.3. Microstructure of Materials. Figure 1 presents the SEM images of the raw materials. The SEM image of the slag shows square and diamond shapes, whereas the OPC sample has box-type and stone-type shapes. FA, as received from the industry, contains mostly spherical particles with rough surfaces. The SEM image of RHA (UG) shows very porous and spongy structures, which explain their high specific surfaces. The particles of RHA (UG) are angular, cellular, and

TABLE 2: Chemical and physical properties of the materials (wt.%).

(a) Chemical properties and oxide compositions (%)												
Materials	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	SO ₃	Na ₂ O	K ₂ O	P ₂ O ₅	TiO ₂	MnO	LOI
Slag	33.75	16.76	0.55	45.40	6.71	1.29	0.12	0.44	—	—	—	3.10
FA	58.95	20.24	5.18	3.04	0.52	0.48	0.14	1.32	0.65	1.08	0.12	3.16
RHA	87.55	0.39	0.20	1.03	0.67	0.57	0.05	2.85	1.31	0.02	0.07	3.14
OPC	20.94	4.63	4.46	67.17	2.53	2.98	0.04	0.15	—	—	—	1.29
*Z-Cem 1	43.41	17.91	2.39	28.22	4.05	0.92	0.13	0.78	—	—	—	3.13
*Z-Cem 2	47.59	16.12	2.12	25.92	3.78	0.89	0.13	0.98	—	—	—	3.12
*Z-Cem 3	51.76	14.33	1.86	23.62	3.50	0.86	0.12	1.17	—	—	—	3.11
*Z-Cem 4	57.23	12.73	1.82	19.22	2.93	0.80	0.11	1.41	—	—	—	3.11
*Z-Cem 5	43.70	15.54	1.43	32.21	4.66	1.00	0.12	0.84	—	—	—	3.09

*Calculated values.

(b) Physical properties									
	Specific gravity	Grain size (μm)			Fineness (m ² /gm)		Specific surface area (m ² /cm ³)	Pore volume (cm ³ /gm)	
		d ₁₀	d ₅₀	d ₉₀	Blaine	BET			
Slag	2.84	2.65	14.87	37.57	0.3935	1.98	1.155	0.00617	
FA	2.42	3.27	14.45	64.33	0.3567	2.34	0.785	0.00444	
RHA (UG)	1.96	1.99	8.80	38.75	0.5780	—	1.225	—	
RHA (G)	2.09	1.66	6.72	29.20	0.6975	12.70	1.502	0.00674	
OPC	3.15	3.34	16.79	47.37	0.2302	1.54	0.715	0.00312	
Z-Cem 1	2.65	1.24	11.23	65.19	0.8529	—	1.83	—	
Z-Cem 2	2.59	1.24	11.03	70.61	0.8507	—	1.87	—	
Z-Cem 3	2.55	1.10	9.28	46.83	0.9309	—	2.03	—	
Z-Cem 4	2.49	1.03	8.90	47.20	0.9503	—	2.09	—	
Z-Cem 5	2.68	1.32	11.65	66.32	0.8326	—	1.80	—	

spongy. The SEM image of grounded RHA (G) is presented in Figure 1(d), which indicates that most plerospheres and large, irregular-shaped particles of RHA samples were crushed into smaller sizes after grinding. Therefore, Z-Cems produced from these pozzolanic materials contain very irregularly shaped particles with a porous cellular surface, which consequently increases the fineness and surface area of Z-Cem.

3.1.4. Morphology of Materials. Figure 2 shows the XRD patterns of the pozzolanic materials and OPC. RHA mainly contains amorphous materials with a small quantity of crystalline phase as cristobalite (high-temperature phase of SiO₂) and sylvite. The XRD pattern of the slag shows a crystalline phase of amorphous silica. The XRD pattern of FA shows that it is mainly composed of crystalline phases such as quartz and mullite. Only a few small peaks could be identified as crystalline silica, which is different from the XRD patterns of the slag and RHA. The amorphous silica present in these materials is responsible for the pozzolanic activity. Similar observations have been reported by other researchers [26, 27].

3.1.5. Strength Activity Index of Pozzolans. According to ASTM C311, the strength activity index (SAI) is the ratio between the compressive strength of a mortar containing substitute materials 20% by mass of binder and the average compressive strength of a reference cement mortar at

a designated age [28]. The SAI of the raw materials (FA and RHA) was determined according to this standard. Slag activity index, which is defined as similar to the SAI, only differs by 50% replacement of slag [29]. The compressive strength of the mortar was determined according to ASTM C109 [22]. The SAI of the materials was determined and the results are presented in Table 3. The results show that the SAI of slag is more than 100% for both 7 and 28 days. Thus, slag is considered as grade 100 based on ASTM C989 [29] classification. The SAI of FA mortars is just over 75% at 7 days; however, the value is over 88% at 28 days. Therefore, FA is considered as class F according to ASTM C618 [6] specification. The SAI of RHA (UG) is below 50% at 7 days and more than 63% at 28 days. The SAI of RHA was low because original and/or RHA (UG) were used in preparing the testing mortar. Usually, the SAI of any pozzolans can be improved by grinding or increasing their fineness, which can easily be seen from Table 3. Thus, the SAI of RHA (G) is much higher than that of RHA (UG) samples. Pozzolans can be activated greatly by improving their fineness [10, 30, 31].

3.2. Consistency of Paste. According to ASTM C187, consistency is defined as the amount of water required for a paste to penetrate the Vicat testing needle (10 mm in diameter) by 10 mm [23]. Table 4 provides the obtained consistencies of OPC and Z-Cem pastes. Table 4 indicates

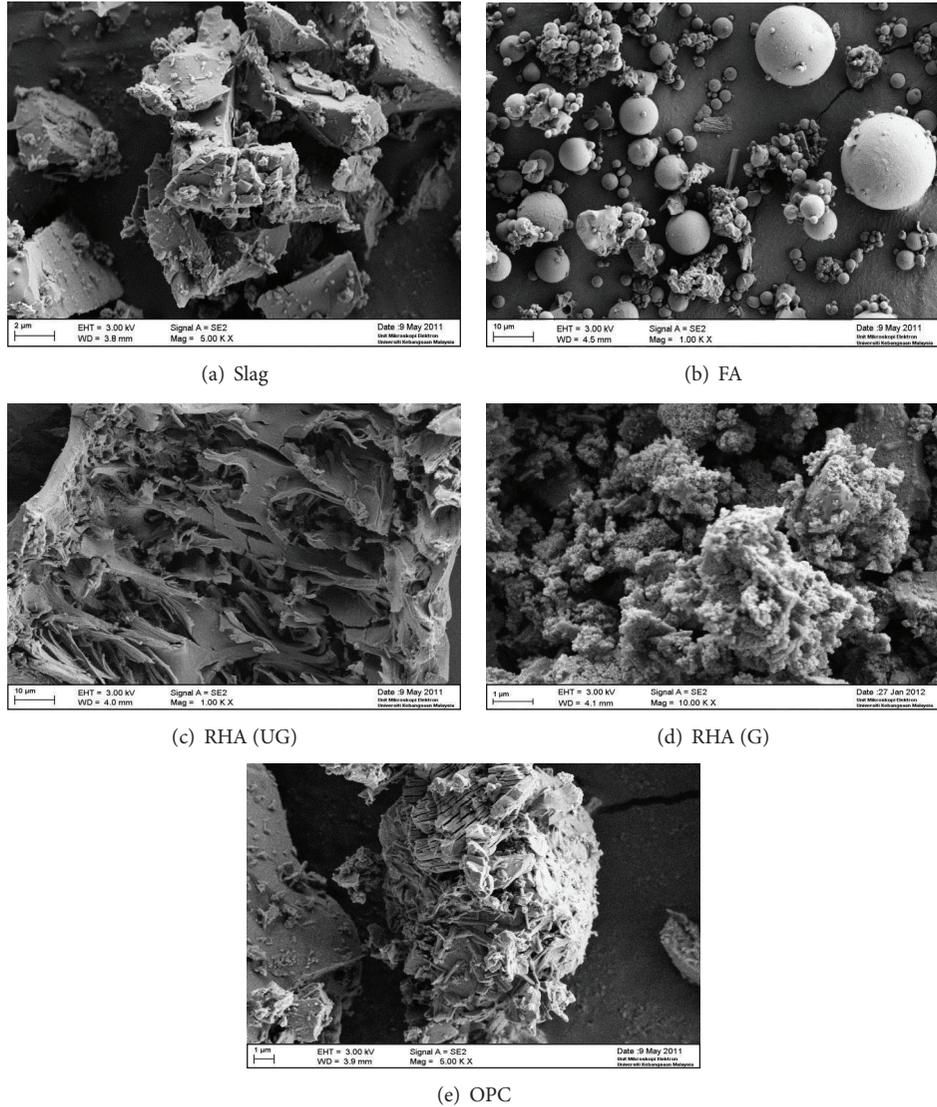


FIGURE 1: SEM images of the used materials.

TABLE 3: Strength activity index of pozzolans.

Materials	Strength activity at 7 days		Strength activity at 28 days	
	Activity index	ASTM requirement	Activity index	ASTM requirement
Slag	100.44	95 for 120 grade	103.88	95 for 100 grade
FA	75.36	75 for Class F	88.07	75 for Class F
RHA	48.58	—	63.04	—
RHA (G)	86.76	—	101.61	—

that the consistency of OPC is 30.5%, whereas that of Z-Cem pastes show increased values. Z-Cem pastes, as obtained from the slag, FA, and RHA, show a higher consistency (or more water demand) compared with OPC. The consistency for Z-Cem pastes varies between 32.5% and 42.5%. The Z-Cem paste possesses more consistency because of the porous and spongy particles of RHA (Figures 1(c) and 1(d)), more voids in the particles, and greater fineness of Z-Cem. Usually,

consistency increases for any blended paste of pozzolan with OPC. Ganesan et al. [32] reported that the consistency of RHA-blended pastes gradually increases with the addition of RHA. A similar finding was also reported by Cheerarot and Jaturapitakkul [33]. They claimed that the consistency of FA-blended paste is higher than that of OPC. Therefore, the greater consistency of the paste of Z-Cem (as produced from the blended materials of slag, FA, and RHA) is rational.

TABLE 4: Consistency and setting time of Z-Cem and OPC.

Binder	Normal consistency (%)	Setting time (h:min) with 5% activators					
		Ca(OH) ₂	Initial KOH	NaOH	Ca(OH) ₂	Final KOH	NaOH
Z-Cem 1	32.5	5:45	1:50	0:21	8:10	3:40	1:30
Z-Cem 2	35.8	7:10	3:40	0:42	—	—	2:20
Z-Cem 3	39.0	—	—	0:50	—	—	3:15
Z-Cem 4	42.5	—	—	1:10	—	—	3:40
Z-Cem 5	33.5	6:05	2:05	0:30	8:40	4:20	2:05
OPC	30.5		2:35			5:25	

—: not determined exactly because setting time is more than 12 hours.

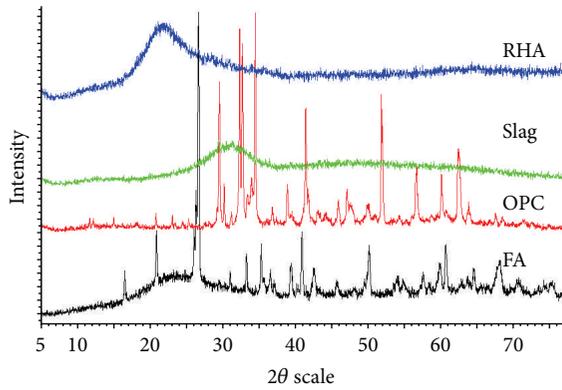


FIGURE 2: XRD patterns of the used materials.

3.3. Setting Time of Paste. The initial and final setting times of OPC and Z-Cem pastes are presented in Table 4. The setting time of Z-Cem is lesser when slag and FA are used but increases when RHA is added to Z-Cems (for Z-Cem 2, Z-Cem 3, Z-Cem 4, and Z-Cem 5). The initial setting time of OPC pastes is 155 min. The initial setting time of 30% RHA (Z-Cem 4) is the highest among all of the Z-Cem binders. The final setting time also increases for Z-Cem 4. The final setting times of Z-Cem pastes are within the ASTM C150 limit for the final setting time of OPC (≤ 375 min) [34]. An excess amount of silica increases the strength of the cement but prolongs its setting time. Larger quantities of alumina quicken the setting time but weaken the cement [35]. For the same reasons, among all of the Z-Cem pastes, Z-Cem 1 sets early because of the larger amount of alumina and lesser amount of silica. By contrast, Z-Cem 4 takes a longer time to set because it has more silica content and less alumina (as shown in Table 2). Although Z-Cems contain higher silica contents than that of OPC, Z-Cems set earlier than OPC. The setting time of Z-Cem is not only affected or controlled by its silica and alumina contents but also by the amount/concentration of the activator. Even though the same amount of activator was used in all of the Z-Cem pastes, the concentration of the activator was reduced for Z-Cem 2, Z-Cem 3, Z-Cem 4, and Z-Cem 5 because more amount of water was used (i.e., greater values of normal consistency of Z-Cem 2, Z-Cem 3, Z-Cem 4, and Z-Cem 5). Therefore, the setting times of Z-Cem 2, Z-Cem 3,

Z-Cem 4, and Z-Cem 5 gradually increase. The coarser particle size of FA and the spongy particle size of RHA could be the reasons for the longer setting time of the paste because they absorb more amount of water (can be seen from the normal consistency data), thereby resulting in the delay of the hydration process. Usually, smaller-sized particles react quickly than coarser particles. Thus, particles with a diameter between 1 and 10 μm will react within 1 day and 1 month, respectively, whereas particles over 50 μm will not completely react with the presence of sufficient amount of water [36]. The setting time of FA-blended paste increases compared with that of OPC [37]. Among the activators, NaOH contributes to the earlier setting of the Z-Cem because of its smaller cation (Na^+) compared with the other activators.

3.4. Flow of Mortar. Tables 5 and 6 show the measured flow of the mortar in different Z-Cem mixes and OPC in which the mortars were prepared according to BS EN196-1 [21] and ASTM C109 [22] testing standards, respectively. The flow value of the mortars depends on various factors including mix proportion, fineness of Z-Cem, amount of used SP, and types of activators. The flow value of the OPC mortar is over 80% without SP. For the utilization of NaOH, the flow decreases for Z-Cems 3 and 4 mortars, even though SP was added in the mortars. Although 3.5% SP was used in Z-Cem 4, its flow is less compared with those of other Z-Cems and OPC mortars. The lesser flow rate of the Z-Cem mortar is due to more water demand from slag, FA, and RHA, which were used in preparing Z-Cem. To maintain a constant flow of $110 \pm 5\%$, the highest amount of the activator's solution and SP are required with the use of NaOH, whereas $\text{Ca}(\text{OH})_2$ and KOH require less amount of solution. Similar observation has been reported in the literatures for mortar [38] and paste [39] prepared with potassium-based activators, which exhibits greater flow compared with those prepared with sodium-based activator. The lesser flow of mortar containing NaOH may be caused by the following reasons:

- (i) among the activators, NaOH has the smallest cation (Na^+), which may attract the molecules/constituents of the binder and the mortar and, consequently, reduce the flow of mortar;
- (ii) the lower flow tendency and the higher water demand are due to the porous and spongy nature of these

TABLE 5: Flow and SP requirements for 0.55 W/B ratio with Sa/B ratio of 3.0.

Binder	SP (%)	Flow (%) for using 5% activators by wt. of binder		
		Ca(OH) ₂	KOH	NaOH
Z-Cem 1	1.0	79	78	73
Z-Cem 2	1.0	68	59	51
Z-Cem 3	2.5	62	58	48
Z-Cem 4	3.5	74	70	58
Z-Cem 5	1.0	80	76	71
OPC	0.0	81		

TABLE 6: Activator's solution and SP requirement to maintain constant flow (110 ± 5%).

Binder	2.5 M Ca(OH) ₂ solution			2.5 M KOH solution			2.5 M NaOH solution		
	B/S	SP (%)	Flow	B/S	SP (%)	Flow	B/S	SP (%)	Flow
Z-Cem 1	0.55	4.0	113	0.58	4.0	109	0.62	4.2	112
Z-Cem 2	0.55	4.2	111	0.58	4.2	108	0.62	4.3	106
Z-Cem 3	0.55	4.5	106	0.58	4.4	113	0.62	4.5	109
Z-Cem 4	0.55	4.7	110	0.58	4.6	108	0.62	4.8	105
Z-Cem 5	0.55	4.2	108	0.58	4.0	107	0.62	4.2	106
OPC	Flow – 107, SP 0.0%								

materials (particularly for RHA) and the higher fineness or larger surface area; thus, more amount of water is required. Several researchers have reported that more amount of water is required to obtain the desired consistency; a lower flow ability is common among pozzolans [40, 41].

3.5. SP Requirement of Mortar. The amounts of SP are presented in Tables 5 and 6. Table 5 indicates that the Z-Cem mortar requires more SP. Moreover, SP requirement is varied for different Z-Cem mortars. This result could be due to the high fineness and porous surface of FA and RHA. In the Z-Cem mortar, SP requirement is greatly influenced by RHA content and the activators. For example, with NaOH, Z-Cem 1 (60% slag, 40% FA, and 0% RHA) shows 73% flow with 1% SP, whereas Z-Cem 4 (40% slag, 30% FA, and 30% RHA) provides 58% flow with 3.5% SP. Table 6 presents the SP content required for the mortar prepared with 2.5 M solution to maintain a constant flow of $110 \pm 5\%$, as specified in ASTM C109 [22]. The results show that compared with KOH and NaOH, Ca(OH)₂ produces a constant flow ($110 \pm 5\%$) with less S/B and SP content for the same Z-Cem mortar (i.e., Ca(OH)₂ requires less amount of solution and SP). Thus, the flow of the Z-Cem mortar is also greatly influenced by different activators. Among the three activators, Ca(OH)₂ possesses the highest flow ability. Given the application of molar-concentrated solution, SP requirement also increases within creasing RHA content (i.e., Z-Cem 2 to Z-Cem 5). Pozzolan materials require greater amount of SP [41, 42].

3.6. Compressive Strength of Mortar. The compressive strengths of Z-Cem mortars are significantly influenced by the amount, type, and concentrations of activators, as discussed below.

3.6.1. Strength of Mortar as Influenced by Amount of Activators. To investigate the effect of the amount of activators (by weight of binder) on the strength of the mortar, Z-Cem 4 with three different activators was used because this mix gives the best compressive strength (Table 7). Mortar cubes were cast according to ASTM C109 [22]. Figure 3 presents the compressive strengths of the mortar with different activator contents. The figure demonstrates that when the same amount of activators was used, the strength of the Z-Cem mortar is less with Ca(OH)₂, whereas KOH and NaOH give better results. The rate of strength development of the Z-Cem mortar with KOH is slow compared with those of Ca(OH)₂ and NaOH at early ages, up to 7 days. It could be expected that there would be a retardation of reaction kinetics due to the utilization of potassium-based activator [38].

Due to the wide range of factors affecting the mechanical strength of the alkali activated binders and/or geopolymer, and the physical and chemical differences of the materials (slag, fly ash, metakaolin, and activators) and curing temperature used in the different researches, it is difficult to make direct comparison between strengths and other properties reported by the various authors. However, considering the relationship between the strength with molarity of activators and the types of activators, obtained results of this study have been compared with the others research findings in the relevant sections.

Figure 3 shows that Z-Cem achieves lesser strength at lower doses of activators (2.5%). A lower amount (2.5%) may not be sufficient to activate all of the molecules of Z-Cem. Besides, no significant effects on the strength development are observed with the addition of higher amount (7.5%) of Ca(OH)₂ and NaOH. The strengths of the mortars were almost the same for doses of 5.0% and 7.5% of the activators. However, at a dose of 7.5%, KOH imparted the highest strength to the Z-Cem mortar at 28 days but shows reduced

TABLE 7: Strength of mortar as influenced by types of activators.

Mortar	Activators (5% by weight)	Compressive strength (MPa) at different ages			
		3 days	7 days	14 days	28 days
Z-Cem 1	Ca(OH) ₂	—	2.61	6.59	18.10
	KOH	—	4.66	5.45	20.26
	NaOH	9.21	13.25	18.12	22.12
Z-Cem 2	Ca(OH) ₂	—	2.38	5.60	6.10
	KOH	—	3.32	6.20	26.35
	NaOH	10.24	14.65	20.12	28.32
Z-Cem 3	Ca(OH) ₂	—	5.22	10.42	13.35
	KOH	—	15.20	22.55	30.60
	NaOH	12.12	16.22	21.23	33.12
Z-Cem 4	Ca(OH) ₂	—	5.50	13.76	20.65
	KOH	—	9.53	19.10	27.46
	NaOH	13.87	20.12	34.15	42.24
Z-Cem 5	Ca(OH) ₂	—	6.30	12.70	14.90
	KOH	—	12.40	28.21	30.52
	NaOH	10.12	14.12	26.13	30.22
OPC	—	24.94	30.32	35.08	44.08

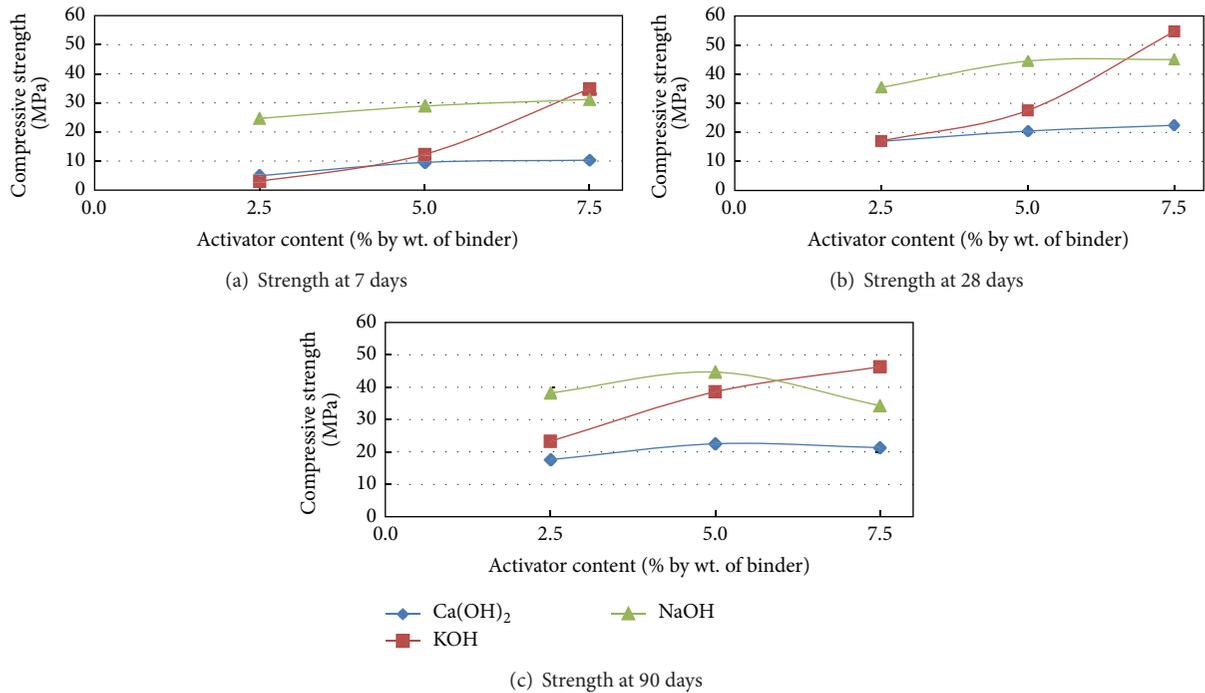


FIGURE 3: Compressive strength of mortar (Z-Cem 4) as influenced by amount of activators.

strength at 90 days. The strength of the mortar remains the same or decreases, which is most probably due to the excess amounts of activators that did not participate in the chemical reaction or weak intrabonds that have formed between them. These weak bonds may collapse at prolonged periods of time.

3.6.2. *Strength of Mortar as Influenced by Types of Activators.* To investigate the effect of different types of activators on the strength of the Z-Cem mortar, mortar prisms were cast

according to BS EN196-1 [21] standard. Table 7 presents the compressive strengths of Z-Cem mortars with different activators at various ages of mortar. Although 7.5% KOH imparts the best strength to the Z-Cem mortar, it develops the strength of the mortar very slowly at an earlier age. Therefore, activators were used by 5% weight of binder because this percentage of activators (NaOH and Ca(OH)₂) imparts better strength to the Z-Cem mortar (Figure 3). Table 7 shows that the strength development of the Z-Cem mortar is slow at

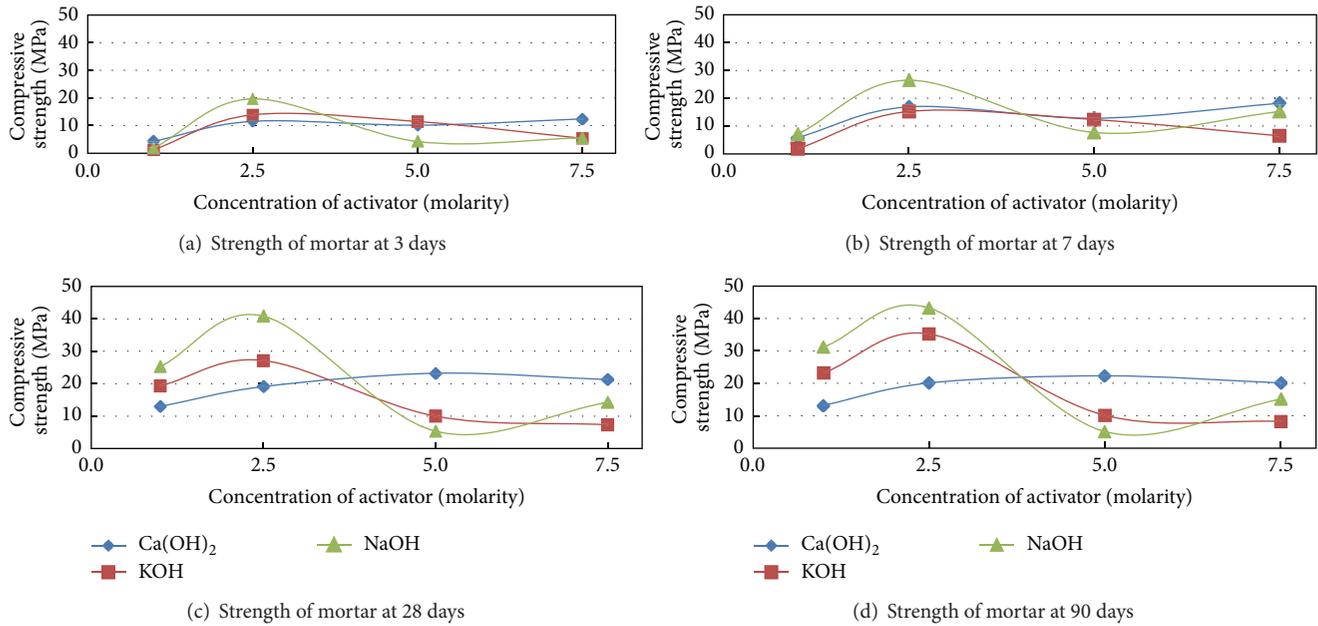


FIGURE 4: Strength of mortar (Z-Cem 4) for different concentrations of activators.

an early age (3 days), particularly for $\text{Ca}(\text{OH})_2$ and KOH , but it is considerable for the same amount of NaOH (5% weight of binder). Z-Cem 4 achieves the highest compressive strength (42.24 MPa at 28 days) with NaOH . The strength development of Z-Cem mortars with NaOH is similar to that of OPC, in which a reasonable strength is achieved at early and later ages. The Z-Cem 1 mortar has lesser strength compared with the OPC mortar. The compressive strengths of the Z-Cem 4 mortar at 3 and 7 days are greater than those specified in ASTM C150 (the minimum strength of OPC type I cement is 12.0 and 19.0 MPa at days 3 and 7, resp.) [34]. The Z-Cem mortars achieve considerable strengths because of greater fineness, which is an important property in increasing the strength of the mortar and the chemical action of activators. The grinding technique of slag and RHA could be demonstrated as a kind of mechanical activation for Z-Cem. Thus, grinding and the particle size of pozzolans are vital factors in increasing the strength of the mortar. Kiattikomol et al. [43] reported that a mortar with 20% OPC replacement had greater strength with higher fineness of FA. A comparable strength was found in other studies where greater fineness was observed in grounded RHA. OPC mortar and concrete with finer RHA were reported to have better strengths [10, 31, 44]. However, in the present study, the strength of the Z-Cem 1 (without RHA) mortar is less but increases with the presence of RHA (for Z-Cem 2, Z-Cem 3, Z-Cem 4, and Z-Cem 5). RHA might function as another activator for Z-Cem. Therefore, the developed strength of Z-Cem may be due to the following combined reasons:

- (i) better strength which was observed because of the presence of more silica in Z-Cem. Thus, Z-Cem 4 achieves the highest compressive strength because of its greater silica and lesser alumina contents [35] that

can be seen from the oxide composition of Z-Cem (Table 2);

- (ii) the chemical cross-linking action of RHA;
- (iii) the grinding effects of materials that lead to the increase in surface area or greater fineness of Z-Cem;
- (iv) effect of the binary or ternary blends of pozzolans in Z-Cem, because Joshi and Kadu [45] reported that FA-based geopolymer concrete (activated by 12 M–14 M NaOH solution) achieves a compressive strength of 14 MPa due to heat curing at 75°C for 24 hours. Ravikumar et al. [46] also found that FA-based geopolymer concrete (activated by 8 M NaOH solution) achieves a compressive strength of 29 MPa due to heat curing at 75°C for 48 hours;
- (v) chemical action of NaOH , $\text{Ca}(\text{OH})_2$, and KOH .

The strength development rate of the Z-Cem mortar is faster with NaOH , followed by $\text{Ca}(\text{OH})_2$ and KOH at an early age of up to 14 days. Thus, NaOH contributes to gain strength quickly which is similar argument as reported by Oh et al. [16] and Fernandez-Jimenez et al. [47]. This result may be due to the following reasons: (i) among the activators, NaOH has the smallest cation (Na^+), which may attract the molecules/constituents of the binder and the mortar, and consequently increase the intrabond of molecules in the mortar. To obtain better strength, a similar argument was reported by Pimraksa et al. [48] where they utilized NaOH . Rodríguez et al. [38] have reported that mortars produced with sodium-based activator present higher strength than those with potassium-based activator. Table 7 also indicates that with the same amount of the activator (5% by weight), NaOH contributes to the development of the highest strength

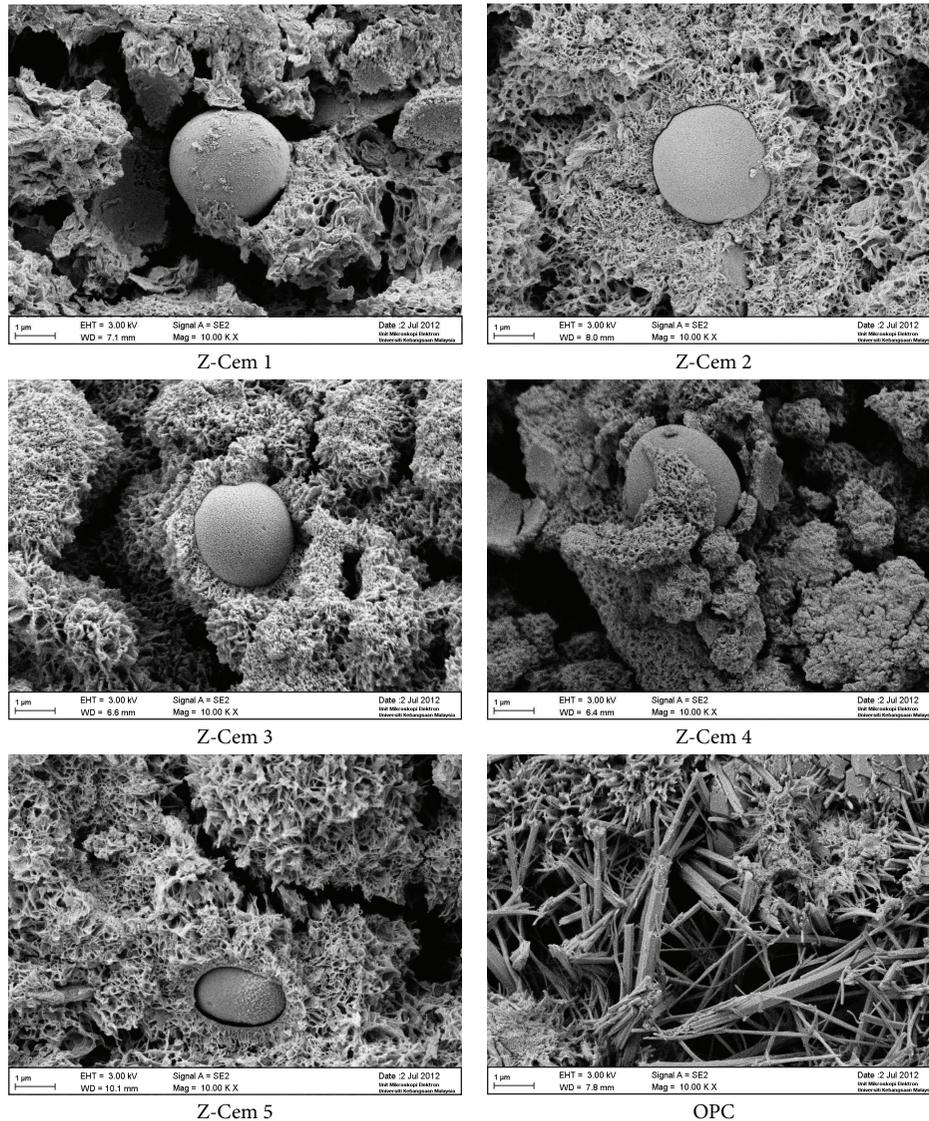


FIGURE 5: SEM images of OPC and Z-Cem mortars at 28 days of curing age.

of mortar among the activators. Therefore, to obtain a reasonable strength of the Z-Cem mortar, the use of grounded pozzolans with NaOH activator is the best technique.

3.6.3. Effects of Concentration of Activator on Mortar Strength.

To investigate the effects of the concentration of activators on the strength of the mortar, Z-Cem 4 was considered because it has the highest compressive strength. Mortar cubes were prepared according to ASTM C109 [22]. The compressive strengths of those specimens are presented in Figure 4. The figure shows that strength of mortar is influenced significantly by the concentration of activators which is similar conclusion reported by other researchers [46, 49]. However, the strength of the mortar is less when $\text{Ca}(\text{OH})_2$ is used with different concentrations. By contrast, the strength of the Z-Cem mortar is better with different molar concentrations of KOH and NaOH. The rate of strength

development of the Z-Cem mortar is slow for KOH compared with those of $\text{Ca}(\text{OH})_2$ and NaOH at early ages, up to 7 days and concentrations of 1.0 and 2.5 M. Rodríguez et al. [38] claimed that a retardation of reaction kinetics occurs due to the utilization of potassium-based activator. The higher concentrated solutions (over 5.0 M concentrations) of activators are insignificant in increasing the strength of the Z-Cem mortar, even though the strength of the mortar was reduced for 5.0 and 7.5 M activators. This result may be due to following causes:

- (i) a lower concentration (1.0 M) may not be sufficient to activate all of the molecules of Z-Cem;
- (ii) for a higher molarity (5.0 and 7.5 M), excess amounts of activators remain without forming weak intra-bonds inside the Z-Cem mortar. Residual activators or weak bonds of activators may collapse when the mortar is immersed in water for curing. As a result,

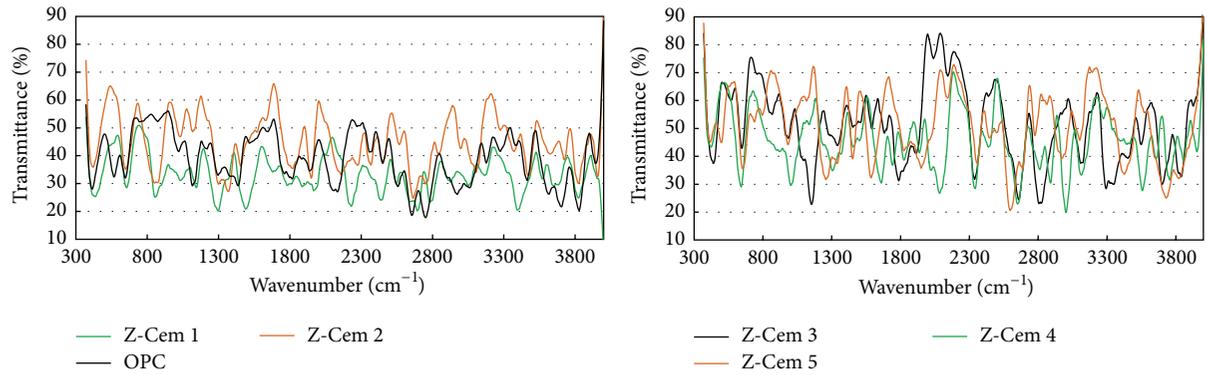


FIGURE 7: FTIR data of OPC and Z-Cem mortars.

molecules in the Z-Cem mortar are observed to have a silica-hydrate (Si-H) bond with sodium or other inorganic metals (i.e., sodium/calcium-silica-hydrate-alumina gel).

4. Conclusions

Based on the overall test results, the following conclusions are drawn from this study.

- (i) Slag and FA contain moderate amounts of SiO_2 , whereas RHA has a high percentage of SiO_2 . Z-Cem (combination of slag, FA, and RHA) contains over 50% major oxides ($\text{SiO}_2 + \text{Al}_2\text{O}_3 + \text{Fe}_2\text{O}_3$). As a result, Z-Cem functions as a binder with the presence of chemical activators such as NaOH, KOH, or $\text{Ca}(\text{OH})_2$ at an ambient temperature.
- (ii) Slag, FA, and RHA could be used as Z-Cem binders with the presence of a chemical activator provided that these materials should be processed properly while maintaining a high fineness value (combined Blaine fineness should be more than $8000 \text{ cm}^2/\text{gm}$).
- (iii) Z-Cem shows a reasonable consistency and setting time. The initial and final setting times of Z-Cem 4 (slag 40%, FA 30%, and RHA 30%) with 5% NaOH are 70 and 220 min., respectively. The flow value of the mortar of Z-Cem is satisfactory with the presence of SP.
- (iv) The strength of the Z-Cem mortars is significantly influenced by the amount, molar concentration, and type of activators. When the same amount or molar concentration of activators is used, NaOH contributes in achieving the highest strength of the Z-Cem mortar, except 7.5% KOH.
- (v) The mortar of Z-Cem 4 with NaOH has the highest strength. A minimum of 2.5% and/or 1.0 M solution of NaOH is required to obtain reasonable strength, which is more than 25 MPa at 28 days. NaOH could be used by 5% weight of binder or 2.5 M solution to obtain better and acceptable strength of more than 42 MPa at 28 days. The utilization of an activator of more than 5% by weight of binder or over 5.0 M is

insignificant to increase the strength of the Z-Cem mortar.

- (vi) The FTIR results reveal that molecules in the Z-Cem mortar are connected and linked with a silica-hydrate (Si-H) bond with sodium or other inorganic metals (i.e., sodium/calcium-silica-hydrate-alumina gel).
- (vii) The test results ensure that Z-Cem could be developed using locally available slag, FA, and RHA combined with the chemical activator. If slag, FA, and RHA are used as supplement for cement, the demand of cement should be reduced. Therefore, these materials can be consumed properly as Z-Cem binders that could also be used as sustainable binders in the near future.

Conflict of Interests

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

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