PROPERTIES OF CONCRETE UTILIZING SELF-CEMENTING HIGH-CALCIUM FLY ASH AS SOLE BINDER MATERIAL

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PROPERTIES OF CONCRETE UTILIZING SELF-CEMENTING HIGH-CALCIUM FLY ASH AS SOLE BINDER MATERIAL

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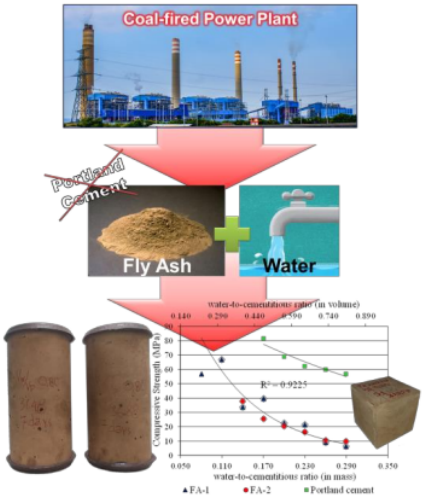

Abstract

Fly ash, a waste material derived from the coal burning in power plants, could be utilized in concrete mixtures as a filler or as a cement replacement material, either partially or fully. Some papers also report that high-calcium fly ash can be utilized as a cementitious material through the hydration process. However, there are just a few papers that studied the behavior of concrete that utilizes high-calcium fly ash as the sole binder material without any chemical activators. Furthermore, there are no reports about whether the effect of the different water-to-cementitious ratios of this concrete is the same as the ordinary Portland cement concrete. This paper presents an initial development on the use of fly ash as a sole binder material, i.e., 100% fly ash concrete without any chemical solution as an activator. This research utilizes the high calcium content in the fly ash to produce concrete with a commonly used method (just add the water without any alkali activator) and investigates how the water-to-cementitious ratio can influence the compressive strength of the paste and the concrete as well. The calcium content in the fly ash used in this study was in the range of 19–22 percent of the total weight of fly ash. Paste compressive strength of 66.78 MPa was obtained at 28 days with a very low water-to-cementitious ratio of 0.110; on the other hand, the concrete compressive strength of 37.05 MPa was obtained at 28 days with a water-to-cementitious ratio of 0.143 and continued to increase to 49.78 MPa at 365 days. This research proved that the calcium content in high-calcium fly ash can act as self-cementing material and has the potential to be a sole binder material in concrete.

Keywords: High-calcium fly ash, self-cementing, very low water-to-cementitious ratio, workability, slump loss, compressive strength

1.0 INTRODUCTION

Concrete has become one of the most widely used building materials in construction since ancient times. Generally, it is composed of Portland cement, water, and aggregate as filler. In the production of Portland cement, carbon dioxide is released into the atmosphere, which is a greenhouse effect. Because global warming has become a major global concern, there is a need to reduce or replace Portland cement with more sustainable materials.

Fly ash is already well known as a substitute material for Portland cement. It has been widely studied related to the concept of reducing greenhouse gas emissions in the construction industry. However, the consumption of fly ash is still much less than the amount of fly ash (waste) produced [1].

Fly ash is waste derived from coal combustion in steam power plants along with other products such as bottom ash, boiler slag, and flue gas desulfuration (FGD) [2]. Hence, the properties of fly ash will strongly depend on the type of coal used as boiler fuel. The boiler type, its operating conditions, and other combustion parameters also affect the properties of the fly ash [3]. ASTM C618 divides fly ash into class F and class C based on its chemical properties. The difference between F and C classes is in the amount of Calcium Oxide (CaO), where class C fly ash should have more than 18% of CaO. ASTM C618 also states that class C fly ash has some cementitious properties in addition to its pozzolanic properties [4]. Similar to ASTM, the Canadian standard (CAN/CSA-A3001-13) classifies fly ash based on its calcium oxide content and divides it into three types. Type F fly ash has a calcium oxide content below 8 percent. For calcium oxide content between 8 and 20 percent, the fly ash is classified as type CI, and above 20 percent, the fly ash is classified as type CH [5].

Fly ash has performed successfully in replacing some amount of Portland cement in the concrete. Replacement amounts of 10–30 percent are commonly used for ready-mix concrete these days. In some cases, up to 50 percent replacement has also been achieved [6]–[8]. Some research also shows the utilization of fly ash at a higher percentage of replacement. This kind of concrete is generally called "high-volume fly ash concrete" [9] or "very high-volume fly ash concrete" (for up to 80 percent replacement) [10]–[14]. Regardless of how high the percentage of fly ash used to replace, some amount of Portland cement is still needed to start the pozzolanic reaction. That is because most studies use class F fly ash, which naturally has pozzolanic properties only due to the high content of silica and alumina but low in calcium.

On the other hand, besides as a pozzolanic material, Class C fly ash has been known as an alternative cementitious material and can act as self-cementing material due to the presence of alkaline substances in it [15]. Its high calcium content can form either calcium silicate hydrate or calcium aluminate hydrate when in contact with water [16]. Therefore, the level of utilization of fly ash still has a chance to be increased by totally replacing Portland cement to produce more sustainable construction material.

Utilizing fly ash as a total replacement for Portland cement can be achieved through chemical activation [17], [18]. In this case, fly ash acts as an aluminosilicate material that should be activated using a high-alkaline solution to start the polymerization. The final product of this process is called "geopolymer concrete" or, in several studies, "alkali-activated material" [19], [20]. In most cases, low-calcium fly ash, i.e., class/type F, is more favorable to synthesize than high-calcium fly ash regarding the flash setting time [21]. However, this geopolymer concrete is still not widely applied because its curing regime needs heat in most cases [22]. The involvement of some chemical liquid in the production process may also become an obstacle because it takes a particular person and more caution.

Fly ash that contains high amounts of calcium oxide can react with water to produce hydrates even without the presence of calcium hydroxide [23], [24]. Some research also showed a promising application of 100% fly ash concrete as a structural element [25]. In all existing research, the mix composition was presented as a "ready-to-use" composition. However, there is no further discussion about the parameters of mix design, especially the effect of the water-to-cementitious ratio. Other research also shows that some verification must be done before utilizing the fly ash as some types might not meet the standards for structural concrete [26]. It is said that the flash set would occur when the fly ash is used as a sole binder material and a retarder should be used to regulate the setting time. However, there is also no further information about how fast the setting time will occur without the addition of a retarder. On the other hand, which properties of the fly ash would affect the setting time is also essential to be studied.

The Paiton steam power plant in East Java, Indonesia, consists of nine coal-fueled units that are owned and operated by different companies. Some of these units are among the largest steam power plants in Indonesia, with a total electricity production capacity of around 4715 MW, which supplies 20–30 percent of the total electricity demand in Java and the Bali Islands. The amount of fly ash derived from coal combustion in all steam power plant units is estimated at more than five million tons annually. Utilizing the local fly ash obtained from the Paiton power plant, this research focuses on developing self-cementing properties in high-calcium fly ash. How the water-to-fly ash ratio affects the compressive strength and whether it is different from the Portland cement will be the main discussion of this paper.

2.0 METHODS

This research has been conducted in two stages. The first one was to analyze the self-cementing properties of the fly ash by mixing fly ash itself with water to produce paste mixtures. Both fine and coarse aggregates were used in the second stage along with the fly ash paste to produce concrete. Paste mixture was poured into a three-gang mold (5 cm x 5 cm x 5 cm) while the concrete mix was cast into a 10 cm diameter and 20 cm high cylinder mold. A flow table, as described by ASTM C230, was used to determine the workability of the fly ash paste mixture and its setting time was determined by calculating the heat changes in the paste. The data for paste temperature evolution were collected using thermocouples and a data logger. A slump test, as described by ASTM C143, was used to determine concrete workability. Compressive strength tests were applied for each sample at every specified age to determine the strength of the paste and the concrete samples.

Water bath curing was avoided after several observations in which some calcium compounds were leached by the water. The increased pH level of the water bath indicates that calcium in the paste dissolves faster than the hydration reaction of the matrix itself. Therefore, the curing was carried out either in calcium-saturated water or in sealed plastic bags. For concrete specimens, the curing regime was performed by wrapping the concrete cylinders in plastic and store at room temperature until the compressive strength test day.

2.1 Materials

Fly ash used in this research was obtained from the PLTU (Steam Power Plant) Paiton Unit 5 & 6, located in Probolinggo, East Java, Indonesia. Table 1 presents the results of the XRF analysis of two fly ash samples collected at two different times from the same source. According to ASTM, both fly ash samples were classified as class C. However, based on Canadian standards, FA-1 is in the upper limit of CI-type fly ash while FA-2 is CH-type. A pH value of 12.6 has been observed from a mixture of 20 grams of FA-2 with 80 grams of distilled water. While the physical properties of the FA-2 are a brownish color, have a specific gravity of 2.80, and the total retained fly ash on sieve #325 (45-micron opening) is 23%.

Table 1 Chemical composition of fly ash by XRF analysis

Analyzed Element	FA-1 (wt%)	FA-2 (wt%)
SiO_2	33.04	29.50
Al_2O_3	15.79	15.02
Fe_2O_3	16.17	17.71
CaO	19.59	22.34
MgO	8.24	8.37
SO_3	1.89	1.90
LOI	0.25	0.36

As the self-cementing capability comes from the quicklime (CaO) inside the fly ash, special attention is required in terms of storage to prevent the calcium compound from reacting with carbon dioxide in the air. In conditions where quicklime (CaO) reacts with atmospheric carbon dioxide (CO_2), lime will turn into limestone ($CaCO_3$), which cannot react with water; in other words, the fly ash loses its self-cementing capability. In this experiment, the fly ash samples were divided and stored in several airtight plastic bags to prevent carbonation.

Table 2 Physical properties of aggregates

Properties	Fine Aggregate	Coarse Aggregate
Bulk Density	1.604 g/cm^3	1.459 g/cm^3
Specific Gravity	2.661	2.609
Absorption	0.444%	3.321%
Fineness Modulus	2.651	7.298

Silica sand and natural crushed stone were used as fine and coarse aggregates, respectively, to make concrete specimens. The maximum aggregate size was 25 mm. Table 2 shows the physical properties of both fine and coarse aggregates in saturated surface dry (SSD) conditions.

2.2 Mix Design

2.2.1 Paste Specimens

To produce paste specimens, fly ash was mixed with tap water only, without the addition of any chemical activators. Eight mix compositions were designed by varying the ratio of water-to-cementitious material (w/cm). The lowest and highest ratios of w/cm used were 0.080 and 0.290, respectively. Some amount of superplasticizers were added to maintain the workability of the mixtures that have a low (≤ 0.140) w/cm. Table 3 shows the weight of each material for each w/cm to produce the same volume of fly ash paste mixture.

Table 3 Mix design of fly ash paste

w/cm		Fly Ash	Water
(In mass)	(In volume)	(grams)	(grams)
0.080	0.224	3088	247
0.110	0.308	2890	318
0.140	0.392	2716	380
0.170	0.476	2561	435
0.200	0.560	2423	485
0.230	0.644	2299	529
0.260	0.728	2188	569
0.290	0.812	2086	605

2.2.2 Concrete Specimens

Table 4 shows the material composition of five concrete mix designs with different w/cm. The water-to-cementitious ratio in the concrete's mix design was calculated based on volume ratio instead of mass ratio, considering the wide variation in the specific gravity of fly ash. This volume ratio is also in line with the volumetric-based mix design that will keep the paste volume for all mix designs is the same. However, the mass ratio (volume ratio divided by the specific gravity of fly ash) of w/cm is also shown in the table.

The paste volume in the concrete mixture was constant at 40 percent for all mix designs. This value is expected to fully cover each aggregate's particles to create a good quality ITZ (Interfacial Transition Zone). Instead of increasing the paste volume, adding an amount of superplasticizer in a concrete mixture is chosen to improve the workability of the concrete. Thus, the compressive strength of concrete will not be affected by the paste volume but by w/cm only.

Table 4 Mix design of concrete specimens

w/cm		Fly Ash	Water	Fine	Coarse	SP
(In mass)	(In volume)	(kg)	(kg)	(kg)	(kg)	(%)
0.143	0.400	600.0	85.7	745.1	1155.0	0.60
0.179	0.500	560.0	100.0	745.1	1155.0	0.50
0.214	0.600	525.0	112.5	745.1	1155.0	0.20
0.250	0.700	494.1	123.5	745.1	1155.0	0.10
0.286	0.800	466.7	133.3	745.1	1155.0	0.00

Table 5 Workability of fly ash paste compared with Portland cement paste

w/cm		Fly Ash (FA-2)		Portland Cement	
		Super-plasticizer	Flow Diameter	Super-plasticizer	Flow Diameter
(in mass)	(in volume)	(%)	(cm)	(%)	(cm)
0.140	0.392	0.15	13.50	Cannot be done	
0.170	0.476	-	13.50	1.50	> 25.00
0.200	0.560	-	17.00	0.75	> 25.00
0.230	0.644	-	> 25.00	0.50	14.50
0.260	0.728	-	> 25.00	-	13.50
0.290	0.812	-	> 25.00	-	17.50

3.0 RESULTS AND DISCUSSION

3.1 Fly Ash Paste

Both FA-1 and FA-2 were used to make two sets of fly ash paste. The first set, made from FA-1, was tested for its compressive strength only. While the second set, which used FA-2, was tested for its compressive strength as well as its workability using the flow table. The second set also used w/cm 0.140 as the lowest ratio.

Table 5 shows the flow diameter of fly ash paste for each w/cm as compared with the Portland cement paste. A bigger diameter means higher flowability of the paste. It also shows the amount of superplasticizer needed for each mixture. At the same w/cm, fly ash paste offers better workability than Portland cement paste. No superplasticizer was required to provide favorable workability for w/cm higher than 0.170. However, for w/cm 0.230 or higher, the fly ash paste mixture starts to become watery as indicated by the flow that exceeds the plate (>25.00 cm).

On the other hand, some superplasticizers are needed to maintain the workability for the mixtures with w/cm 0.140 or lower. The addition of superplasticizer to the fly ash paste is relatively small compared with Portland cement paste. Fly ash paste with w/cm 0.110 and 0.080, which used FA-1, needs the addition of superplasticizer as much as 0.46 and 2.64 percent, respectively, to become workable. The lowest w/cm for the fly ash paste that is still possible to make is 0.080. However, at this ratio, the amount of superplasticizer used is considered as economically disadvantageous. Moreover, the high amount of superplasticizer (2.64%) might increase the actual w/cm due to its water content.

Better workability of the fly ash paste at very low w/cm can occur due to the spherical shape [3] of the fly ash particles. These physical properties cause a ball-bearing effect, which makes the paste flow easily. The solid-spherical shape also has a lower permeability and smaller surface area compared with angular-shaped particles of the same size. Therefore, the free water content needed is less for the fly ash having solid spherical-shaped particles.

An adequate working time, i.e., the initial setting time, is crucial in concrete work to provide sufficient time for compaction to reduce cavities that can reduce the concrete's compressive strength. Figure 1 shows that the setting time of fly ash paste decreases as the w/cm becomes lower. This trend is almost the same for both fly ash samples despite having a different values. For FA-1, the initial setting time (IS) is in the range of 37–103 minutes and the final setting time (FS) is between 139 and 270 minutes for all mixtures. A shorter setting time occurs when using FA-2, where the initial setting time is in the range of 37–62 minutes and the final setting time is between 66 and 129 minutes for all mixtures having w/cm between 0.140 and 0.290.

Compared with Portland cement paste (not less than 45 minutes for initial setting time, according to ASTM C150), the setting time of fly ash paste is relatively fast. This fast-setting time is probably due to the rapid chemical reaction between water and calcium oxide in the fly ash. It also occurs with Portland cement; thus, gypsum is always added to the cement clinker in order to prevent flash sets. However, all fly ash paste in this experiment did not show any flash set problem [26] even without gypsum addition. The initial setting time of about 30–40 minutes is enough to do proper compaction and finishing.

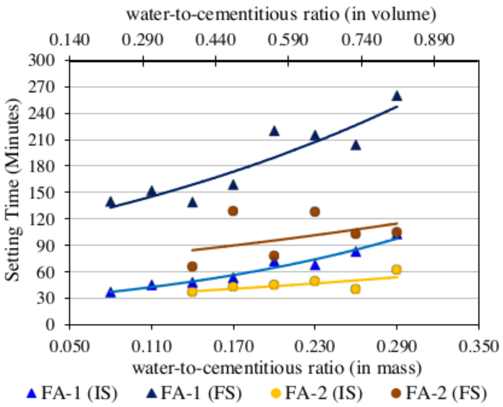

Figure 1 Setting time of fly ash pastes with different w/cm

Regardless of the same trend, Figure 1 also shows that FA-2 has a faster setting time for both the initial and the final times. It also shows the time difference between the final and the initial setting time, which is shorter for FA-2 (2.23 times on average) than FA-1 (3.03 times on average), which could be due to the higher calcium content in FA-2 than in FA-1.

The use of a poly-carboxylate-ether type of superplasticizer also shows compatibility with the fly ash pastes as indicated by the absence of surge in the fly ash pastes setting time. Whether the amount of superplasticizer used is a lot (2.48%) or a little (less than 0.5%) or even not used, Figure 1 shows that the setting time of fly ash pastes is always close to the trend line.

3.2 Compressive Strength of Fly Ash Paste

The compressive strength of fly ash paste will reflect the degree of self-cementing properties in the fly ash. These cementing capabilities will be the first phase material, which determines the strength of the concrete in the next step of the experiment. Higher compressive strength indicates higher cementing ability in the fly ash, which means better binding ability between aggregates. Table 6 shows the compressive strength test results for all fly ash paste at ages 7 and 28 days. Compressive tests at 56 days were carried out on paste specimens using FA-2 to evaluate its strength development.

Figure 2 shows the 28-day compressive strength of the pastes with various w/cm. Although the compressive strength produced is lower, fly ash paste still exhibits the same trend of behavior as the Portland cement paste, where a lower water-to-cementitious ratio results in higher compressive strength. The highest compressive strength of the fly ash pastes, 66.78 MPa, was achieved with w/cm 0.110. That ratio is considered the lowest ratio possible to produce the best result as the lower w/cm, e.g., 0.080, results in lower compressive strength. It might happen due to the lack of water that may cause either insufficient compaction or incomplete hydration reaction.

In the case of different fly ash used, both FA1 and FA2 show similar results, which means the little variation in the amount of calcium compound contained in fly ash may not affect the 28-day compressive strength. Furthermore, it also proved that both fly ash samples consistently demonstrate self-cementing ability.

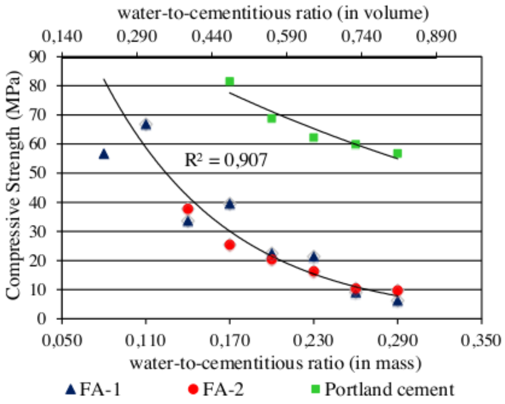

Figure 2 Compressive strength of fly ash pastes at 28 days

Compressive strength development also occurs in the fly ash paste as shown in Figure 3. Assuming compressive strength at 28 days as 100 percent, the strength development of 7 days of the fly ash paste varies from around 70 to 95 percent, with 87.66 percent on average. Meanwhile, at 56 days, the strength development of the fly ash paste is 111.87 percent on average. The development of the paste compressive strength also confirms that a hydration reaction occurs [24].

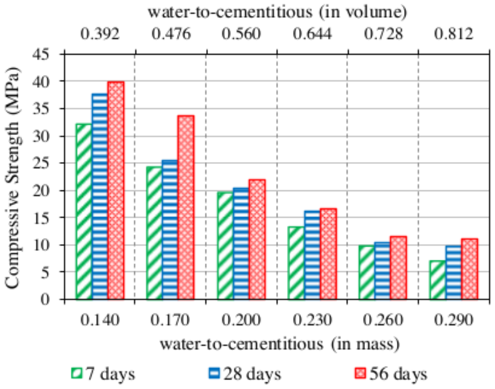

Figure 3 Strength development of the fly ash pastes utilizing FA-2

Table 6 Compressive strength of fly ash paste (in MPa)

w/cm		FA-1		FA-2		
(in mass)	**(in volume)**	**7 days**	**28 days**	**7 days**	**28 days**	**56 days**
0.080	0.224	48.86	56.74	-	-	-
0.110	0.308	52.67	66.78	-	-	-
0.140	0.392	36.67	33.60	32.13	37.73	39.87
0.170	0.476	26.40	39.56	24.27	25.47	33.60
0.200	0.560	19.46	22.51	19.60	20.40	21.87
0.230	0.644	16.46	21.41	13.33	16.27	16.67
0.260	0.728	6.87	9.07	9.87	10.40	11.47
0.290	0.812	4.90	6.27	7.07	9.73	11.07

Table 7 Compressive strength of fly ash concrete (in MPa)

w/cm		Slump	Age (days)							
(In mass)	**(In volume)**	**(cm)**	**3**	**7**	**14**	**28**	**56**	**90**	**180**	**365**
0.143	0.400	24.0	29.92	33.10	34.50	37.05	41.89	46.22	48.51	49.78
0.179	0.500	22.5	19.74	21.39	25.15	28.84	33.74	35.97	37.43	38.64
0.214	0.600	20.5	13.43	16.17	19.93	21.96	22.98	23.43	24.26	25.15
0.250	0.700	21.5	7.96	10.12	11.78	14.07	15.41	16.30	17.06	17.38
0.286	0.800	9.0	3.69	5.41	5.92	7.77	8.59	9.23	9.93	10.12

3.3 Fly Ash Concrete

In general, the workability and compressive strength of concrete will differ among paste specimens due to the addition of aggregates. Lower workability could be caused by the angular shape or the rough surface of the aggregate particles. These problems can be easily overcome by adding superplasticizer to improve workability. Besides the paste quality, the quality of aggregate together with the interfacial transition zone (ITZ) between the aggregate and the paste will determine concrete compressive strength. Table 7 shows the compressive strength of all concrete mix designs utilized with FA-2 at the specified test ages.

The highest 28-days compressive strength of 37.05 MPa was obtained from the mixture with the lowest water-to-cementitious ratio of 0/143 (0.600 in volume). The compressive strength continues to develop after 28 days.

The workability of fly ash concrete starts to need aid—in this case, the addition of superplasticizer—when w/cm is 0.250 (0.700 in volume) or lower. Despite the target slump of 10±2cm can be achieved in a mixture without additive (w/cm 0.286 or 0.800 in volume), a slight addition of superplasticizer (in mixture with w/cm 0.250 or lower) will drastically improve the workability of fly ash concrete, which showed by a high slump of above 20 centimeters.

Regardless of the benefits provided, to ensure that the use of superplasticizer does not have a negative impact on setting time, a slump loss test was carried out. A mortar mixture with w/cm 0.214 (0.600 in volume) and paste volume of 40 percent was prepared. Some amount of superplasticizer was gradually added until the standard flow diameter (15±1cm), shown by the horizontal dash-space line in Figure 4, was reached. A slump flow test was carried out every 10 minutes, and the changes in diameter were measured.

Figure 4 shows the results of the slump loss test. It shows that the fly ash mortar can maintain its workability for at least 180 minutes. It also confirms the compatibility of fly ash with superplasticizer to improve the workability.

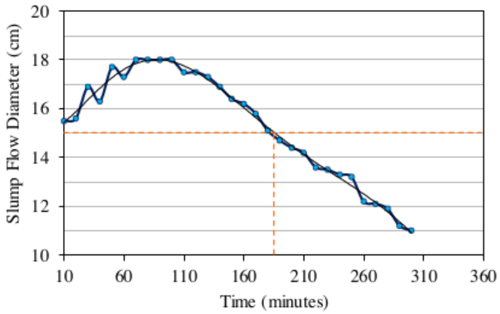

Figure 4 Slump loss of fly ash concrete

Figure 5 shows the strength development of fly ash concrete with five different w/cm. As with the fly ash paste, fly ash concrete also shows strength development even at 365 days. Assuming the concrete compressive strength at 28 days as 100 percent, the strength development of five mix designs increases as the w/cm decrease, especially at an earlier age (3,7, and 14 days). For example, the strength development of concrete with w/cm 0.286, 0.250, 0.214, 0.179, and 0.143 at 3 days is 47.5%, 56.5%, 61.2%, 68.4%, and 80.8%, respectively. However, at a later age (after 28 days), strength development is less significant for concrete with w/cm 0.214 and lower. For concrete with w/cm 0.179 and 0.143, its compressive strength increased more than 10 percent at each age of testing after 28 days up to 90 days. It continues to increase up to 365 days with a lower development percentage (below 10 percent).

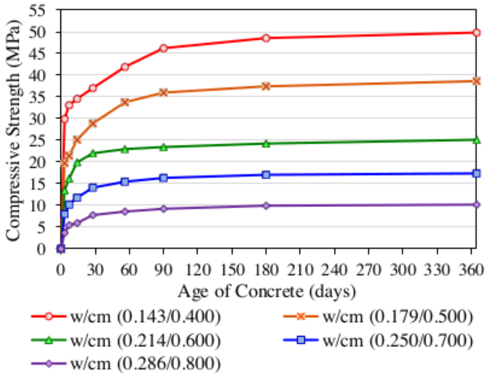

Figure 5 Development of concrete compressive strength for various w/cm (in mass/in volume)

The fact that high-calcium fly ash can react with water and the compressive strength of concretes was developed over time suggests that hydration reaction was involved. However, it needs further investigation to analyze how much the pozzolanic reaction affects later strength development.

Figure 6 shows the 28 days of concrete compressive strength plotted on the paste compressive strength trend line. It clearly shows that the compressive strength of concrete is close to the paste trend line, which confirms the good quality of both the aggregates and the ITZ in the concrete.

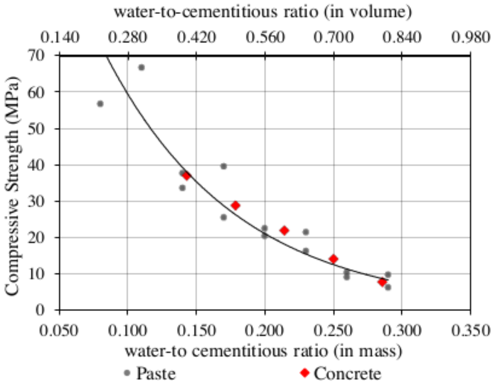

Figure 6 Concrete compressive strength compared with paste at 28 days

4.0 CONCLUSION

By utilizing the self-cementing properties in high-calcium fly ash, it only needs water to start the hydration reaction and produce a "cement-like" material. This research also proved that high-calcium fly ash could be used as a sole binder material in concrete without using any alkali activator.

Furthermore, from the above discussion, fly ash with enough calcium oxide content (in this experiment, it is above 19 percent) will have self-cementing properties and the potential to be a sole binder material in concrete through a hydration reaction that needs water only (no activator needed). However, these self-cementing properties will also depend on several factors, such as the amount of calcium oxide and the form of calcium itself. When calcium oxide in fly ash reacts with carbon dioxide in the air, it will produce calcium carbonate, which can no longer react with water and eliminates the fly ash's self-cementing properties. Therefore, proper fly ash storage or utilizing fly ash as soon as possible after discharging is highly recommended to prevent the reaction of calcium oxide with carbon dioxide.

The water-to-cementitious ratio affects the compressive strength of the concrete. Using lower w/cm increases the compressive strength of the concrete. This trend is the same as the Portland cement concrete but with a much lower value. To achieve concrete compressive strength above 20 MPa at 28 days, the w/cm used is as low as 0.600 in volume or 0.214 in mass. The physical properties of fly ash (especially the spherical shape), which is derived from pulverized coal combustion, can compensate for the use of very low w/cm. Some amount of superplasticizer (polycarboxylate ether type) can also be used to improve workability.

The setting time of fly ash paste is also affected by w/cm. Lower w/cm gives a shorter setting time. Furthermore, different properties of fly ash also show different setting times. Fly ash with a higher amount of calcium oxide tends to give a shorter setting time. Nevertheless, there is enough time to do proper concrete work for all mix designs. Therefore, no retarder is needed in the concrete mixture.

Both paste and concrete that utilize fly ash as a sole-binder material show strength development as the concrete ages. As in the Portland cement concrete, it needs time to achieve a specified strength due to the hydration reaction. The development of concrete compressive strength occurs at a high rate from an early age because the reaction starts immediately and does not wait for hydration by-product (calcium hydroxide) as in the pozzolanic reaction.

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