Effects of High Loss on Ignition Fly Ash as a Partial Replacement for Sand on the Properties of Mortars

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ABSTRACT

This study investigates the effects of fly ash with a high unburned carbon content as a partial sand replacement material on the properties of mortars. Fly ash was added to the mortar in four different ratios: 0, 10, 30, and 50% by volume. This study examined two series of mortar mix proportions, one having a controlled water-to-cement ratio and the other having a controlled flow. The fresh properties and compressive strength of the mortars were investigated. The experimental results showed that the compressive strength increased with increasing fly ash-to-aggregate ratio (FA/A). However, the increase in FA/A resulted in higher water requirements, lower flow, and longer setting times. The calcium hydroxide content and total porosity of the mortars were also examined to support the results of the compressive strength tests. Based on the results, the FA/A ratio had a significant impact on the fresh properties and compressive strength of the mortars. To ensure fresh properties and compressive strength of mortars, a fly ash-to-sand ratio of up to 30% is recommended.

Keywords-flow; water requirement; setting times; compressive strength; TGA

I. INTRODUCTION

Nowadays, the demand for concrete in construction is constantly increasing. This leads to an increasing demand for natural river sand as a fine aggregate. After fresh water, natural sand is the most widely consumed natural resource in the world. The UN estimates that almost 40 billion tons of sand are extracted and consumed per year worldwide, particularly for the construction industry. To ensure geological safety and social stability, each country has very strict regulations on the amount of sand mining allowed in each locality. However, in terms of sand demand and economic benefits, much illegal sand mining still occurs. This results in landslides and riverbank instability, affecting people's lives. Therefore, the solution of using other alternative materials is attractive to many researchers and concrete manufacturers. In addition to sand, several other alternative materials can be used as fine aggregates in concrete, such as crushed stone, demolition waste, bottom ash, slag, etc., which have been previously studied for use as fine aggregates in concrete [1-4]. The use of alternative fine aggregates has many benefits, including a reduced environmental impact, improved concrete properties, and cost savings.

Fly ash has a wide range of industrial uses. In the cement industry, fly ash is used as a mineral admixture to improve some concrete properties. As a Supplementary Cementitious Material (SCM) in concrete, fly ash can help increase its workability and performance [5, 6]. It can also help reduce the heat of hydration, which can help prevent cracking [7]. Fly ash is known as a raw material in a variety of products, such as lightweight aggregate, bricks, and tiles. The use of fly ash in industrial applications has many benefits. It can help reduce the environmental impact of cement production, as it can replace some portions of cement. However, the utilization rate of fly ash in cement and concrete is unsatisfactorily low. This is due to several factors, including the cost of transporting fly ash, the lack of awareness of its benefits, and the regulatory requirements for fly ash disposal. Effective utilization of disposed fly ash can improve sustainability by reducing environmental pollution. The use of fly ash as a sand replacement has the potential to improve the environmental and economic sustainability of the construction industry. Some studies examined the influence of using fly ash as a replacement material for sand in mortar and concrete.

In [8], concrete blocks containing fly ash as a partial replacement for fine aggregates were studied, showing an improvement in compressive strength. The results showed that fly ash concrete blocks could be applied for building construction, and concrete blocks with a 20% replacement ratio had the highest compressive strength. In [9], the replacement of fine sand with fly ash in concrete was studied in concrete, finding that the compressive strength, especially in later ages, and the elastic modulus of fly ash concrete were higher than that of the control concrete. In [10], it was shown that sand can be replaced up to 60% with fly ash without greatly affecting the workability and required strength of the concrete. The use of fly ash to replace sand in concrete is both environmentally friendly and very economical. In [11], class C fly ash was studied as lightweight pelletized aggregates, finding that the properties of concrete using fly ash were comparable to those of concrete using river sand and lightweight aggregates and had a better environmental performance. In [12], fly ash was used instead of fine aggregates in concrete. The experimental results showed that a 15% substitution ratio was optimal to reduce both drying shrinkage and autogenous shrinkage of concrete. In [13], the properties of mortar with fly ash as a fine aggregate were studied, showing that the workability and unit weight of mortars were significantly reduced in mixtures containing fly ash. The reduced unit weight of fly ash concrete contributed to the reduction of dead load, which is very important to ensure that the fly ash concrete structure is safe and resistant to earthquakes.

However, all fly ashes studied in the previously mentioned literature had low Loss On Ignition (LOI) content. According to TCVN 10302:2014 [14], the maximum LOI of class F fly ash for heavy concrete with and without reinforcement is 12 and 15%, respectively. Meanwhile, there is a maximum limit for LOI content according to many standards. For example, the LOI in TIS 2135 is limited to 6% [15]. ASTM C618 specifies the LOI limit for normal concrete at 6% [16]. JIS A 6201 classifies fly ash into four (4) types, and their LOI limits range from 3% to 8% [17]. In Vietnam, many fly ashes contain high unburned carbon content due to coal sources and combustion systems. The LOI limitation of TCVN 10302:2014 is higher than other standards. Therefore, to use high-LOI fly ash in concrete production, a flotation process is usually applied to reduce the LOI content before it is used in concrete [18]. Fly ash, after filtering to remove the amount of unburned carbon, is called selected ash. This flotation process is very expensive and increases the cost of fly ash. It is noted that fly ashes from some sources meet the standard for the LOI content, but supply is still limited.

Fly ash with high LOI content has been studied for use as a cement replacement material in concrete, but there are several disadvantages, such as increased water demand, higher air content, greater requirement for superplasticizer, etc. [19, 20]. Therefore, high-LOI fly ash is usually used as fill or embankment material. The application of high-LOI fly ash as a fine aggregate in the production of mortar or concrete has not been investigated until recently. If high LOI fly ash can be used as a fine aggregate to replace a part of natural sand, it can help solve the problems of natural sand shortage and environmental pollution caused by fly ash while eliminating the cost of treating the unburned carbon in fly ash. This study focuses on investigating the influence of high LOI fly ash when partially replacing sand on the properties of cement mortars. The sand was replaced at 0, 10, 30, and 50% by volume with fly ash. The influence of high-LOI fly ash as a fine aggregate in cement mortars was examined on some basic properties such as flow, water requirements, setting times, and compressive strength of mortars. The results of this study provide valuable data that can be used to evaluate the feasibility of using high-LOI fly ash in concrete.

II. EXPERIMENTAL PROGRAM

A. Materials

The tested raw materials include OPC type I cement, class F fly ash, natural sand, and water. Fly ash is used as a fine aggregate to partially replace sand in cement mortars. Table I shows the physical properties of the materials, while Table II shows the chemical compositions of the cement and fly ash. The fly ash was a class F fly ash with low calcium oxide (CaO) content. Its LOI was 9.7%, which is higher than the maximum allowable value according to ASTM C618. Figure 1 shows the SEM image of the fly ash sample. It includes both fly ash particles and unburned carbon particles. The porous structure of unburned carbon particles can be seen clearly in Figure 2. Moreover, many fly ash particles can be seen in the pores of the unburned carbon particles.

TABLE I. PHYSICAL PROPERTIES OF MATERIALS

Physical Properties	Cement	Fly ash	Sand
Specific gravity (g/cm^3)	3.15	2.28	2.66
Fineness (% passing seize 90μm)	9.65	6.25	-
Fineness Modulus	-	-	2.37

TABLE II. CHEMICAL COMPOSITIONS OF CEMENT AND FLY ASH

Chemical Compositions	Content (% by mass)	
	Cement	Fly ash
SiO_2	18.15	49.50
Al_2O_3	3.15	23.41
Fe_2O_3	3.23	7.65
CaO	70.12	2.58
MgO	1.32	1.30
Na_2O	0.04	0.22
K_2O	0.52	3.44
SO_3	2.84	0.35
LOI	-	9.70

The percentages of fly ash to partially replace sand by volume were 0, 10, 20, 30, and 50%. This study used two series of mortar samples. In series 1, the water-to-cement ratio (w/c) was controlled at 0.6 while in series 2, the mortar flow was controlled at 110±5 mm. Table III presents the mixed proportions of the mortar samples in both series. The sand-to-cement ratio (s/c) of the mixtures without fly ash (F100 and F200) was 3:1. The fly ash mixtures were designed to maintain the same total volume as the F100 and F200 mixtures for series 1 and 2, respectively, so their s/c ratios were not constant.

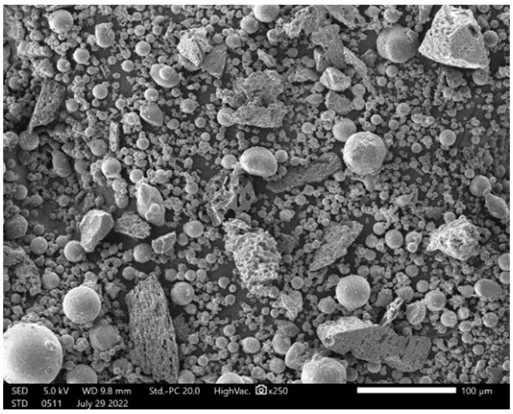

Fig. 1. SEM image of the fly ash sample.

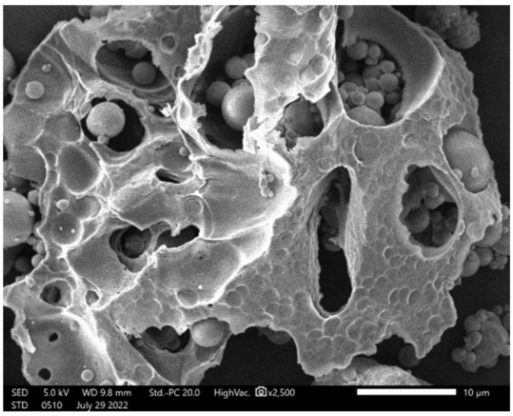

Fig. 2. SEM image of an unburned carbon particle.

TABLE III. MIX PROPORTIONS OF THE MORTARS

Series	Mixtures	w/c	Flow (mm)	s/c (by weight)	FA/A (% by volume)
Series 1	F100	0.6	-	3:1	0
	F110				10
	F130				30
	F150				50
Series 2	F200	-	110 ± 5	3:1	0
	F210				10
	F230				30
	F250				50

B. Experimental Methods

For the controlled w/c series, the flow of the mortars was determined by the diameter of the fresh cement mortars on the flow table, according to ASTM C1437 [21]. The water requirement of the mortars was determined when the flow of the fresh mortars achieved 110±5 mm. Setting times were measured according to ASTM C403 [22]. The initial and final setting times were taken by the time of penetration resistance of 3.5 MPa and 27.6 MPa, respectively.

The mortar cube samples to test compressive strength had dimensions of 50×50×50 mm. The compressive strength of the mortars was determined according to ASTM C109 [23]. To clarify the influence of fly ash on the compressive strength of mortars, the total porosity, the $Ca(OH)_2$ content, and the $Ca(OH)_2$ consumption of the mortars were measured. The total porosity of the mortar cube specimens was tested according to ASTM C642 [24]. The Portlandite content and its consumption in mortars were measured using a thermal gravimetric apparatus. Mortar specimens at 91 days of age were selected for the TGA analysis. The samples were prepared for TGA analysis using the following steps: crush and pass the crushed mortar through sieve No.100 (150 μm), submerge the sieved mortar in an acetone solution for 24 hours to stop hydration, and dry the samples at 100°C for 24 hours. The test temperature range was 20°C to 900°C with a heating rate of 10°C/min carried out under an N_2 atmosphere.

III. RESULTS AND DISCUSSION

A. Flow and Water Requirement of Mortars

Figure 3 presents the results of the flow and water requirement of the mortars tested. The flows of mortars incorporating fly ash are lower than those of the control mortar (non-fly ash mortar). When the fly ash-to-aggregate ratio is increased, the flow decreases. The results of the water requirement and mortar flow correlate well. Mortars require more water when the percentage of fly ash to replace sand increases. The effects of fly ash content on mortar flow and water requirement are due to two reasons. First, since fly ash is much finer than sand, the use of fly ash as a sand replacement material leads to an increased total surface area of the fine aggregate in the mortars, increasing the amount of water absorbed on the fine aggregate surface. Second, the fly ash used in this study contains a very high LOI content. Fly ash with a high LOI content means that it contains a large amount of unburned carbon particles. The unburned carbon particles in the fly ash have a highly porous structure, as seen in Figure 2.

Thus, they increase the water retention and water requirement of the mixtures.

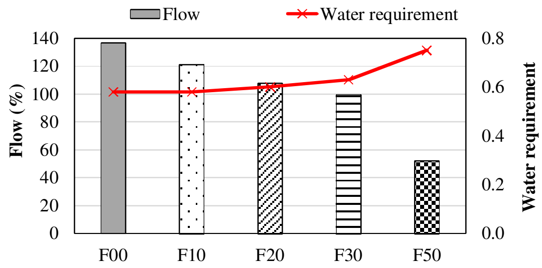

Fig. 3. Flow and water requirement of mortars.

B. Setting Times

Figures 4 and 5 show the effects of fly ash on the setting times of the two series of mortars, including a control w/c ratio and a control flow, respectively. The results of the setting times in both series indicate that both the initial and final setting times of the mortars are delayed when using fly ash as a fine aggregate.

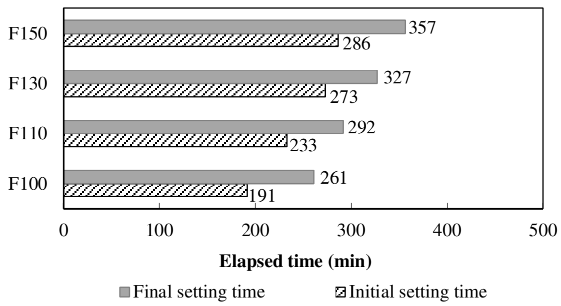

Fig. 4. Setting times of mortars (series 1: controlled w/c ratio).

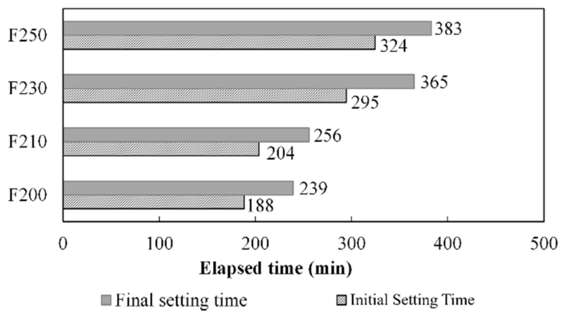

Fig. 5. Setting times of mortars (series 2: controlled flow).

C. Compressive Strength

Figures 6 and 7 show the effects of fly ash on the compressive strength of mortars in the controlled w/c ratio and controlled flow series, respectively. For the series of mortars with a controlled w/c ratio (Figure 6), in the first week, the compressive strength of the mortars using fly ash as a partial replacement for sand was not significantly affected by the replacement ratio. However, at later ages, the compressive strength of the mortars with the fly ash partially replacing the sand was significantly improved. Although the effects of the replacement ratios on compressive strength at 28 days and 91 days of age were not significantly different (almost similar strength improvement for F110, F130, and F150), the compressive strength of mortars containing fly ash as a fine aggregate at later ages was significantly improved compared to the control mortar F100 (without using fly ash). For the series of mortars with controlled flow (Figure 7), at 3 days of age, the compressive strength of the mortars was not improved when using fly ash. The compressive strength of the mortar sample F250 was the lowest. This is because mortars containing fly ash require more water to obtain the same flow. However, at later ages, the compressive strength of mortars containing fly ash was still higher than that of non-fly ash mortar. From the results in Figure 7, it can be concluded that the optimal replacement of sand with fly ash should not exceed 30%.

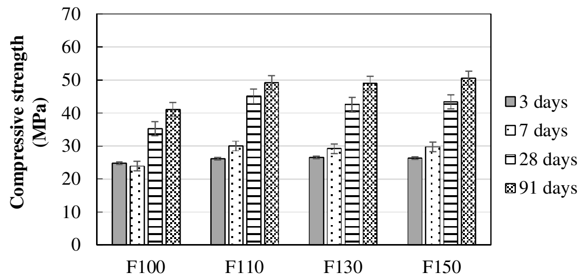

Fig. 6. Compressive strength of mortars (series 1: controlled w/c).

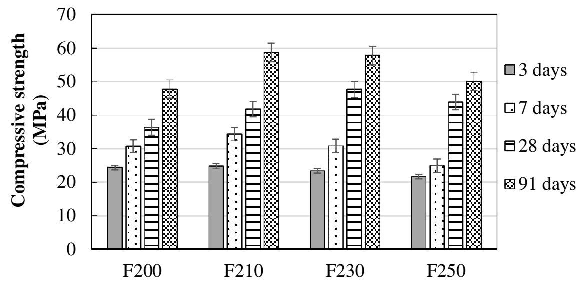

Fig. 7. Compressive strength of mortars (series 2: controlled flow).

The compressive strength of mortars depends greatly on the amount of mixing water. Figures 8 and 9 present the relationship between the water-to-powder ratio (w/p) and the compressive strength at 91 days of the mortars in series 1 and 2, respectively. The mortars in series 1 have a controlled w/c ratio of 0.6, but the w/p ratios (p includes cement and fly ash) are different. The w/p ratios of the fly ash mortars are lower than that of the control mortar. This is the main reason for improving the compressive strength of mortars when sand is partially replaced by fly ash. It is interesting to note that the relationship between compressive strength and w/p of mixtures with fly ash does not follow the typically known Abram's law, as in the case of using fly ash to partially replace cement, especially for the controlled flow series in Figure 9, where the increase in strength was detected when increasing w/p in mortars with fly ash as a partial replacement for sand.

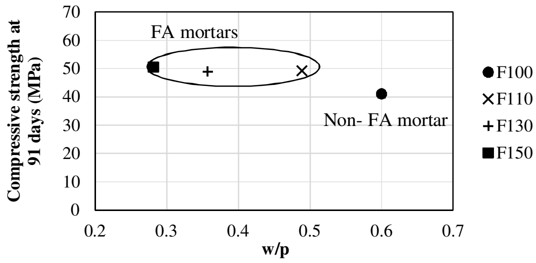

Fig. 8. Compressive strength at 91 days and w/p ratio of mortars (series 1: controlled w/c = 0.6).

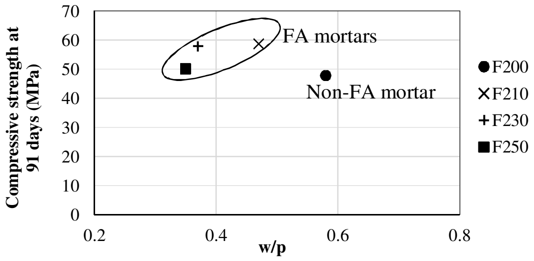

Fig. 9. Compressive strength at 91 days and w/p ratio of mortars (series 2: controlled flow).

D. Total Porosity

Figure 10 shows the total porosity measurements of series 1 mortars with a controlled w/c of 0.6. Total porosity was measured at 3, 28, and 91 days in samples with the same mix proportions and curing conditions as those of the compressive strength test. The total porosity of the mortars incorporating fly ash was lower than that of the non-fly ash mortar. At 3 days of age, the total porosity of mortars containing fly ash was slightly lower than that of the control mortar. However, at later ages (28 and 91 days), the mortars with fly ash became much denser. Fly ash behaves as a filler material, filling between the smallest aggregate size and the largest cement size, improving the overall particle gradation and providing a pozzolanic reaction, making the structure of the mortars denser. Therefore, the compressive strength of mortar was improved using fly ash as a partial replacement for sand compared to the non-fly ash mortar.

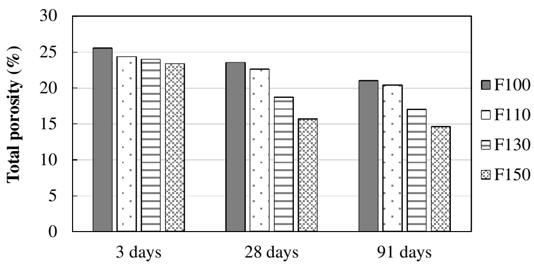

Fig. 10. Total porosity of mortar controlled w/c = 0.6.

Figure 11 shows the trend of higher strength for lower total porosity when considering the same mixture condition. Mixtures with different replacement ratios of sand with fly ash have different relationships. The differences in compressive strength are more obvious at lower total porosity. This confirms that total porosity is not the only factor in controlling the strength.

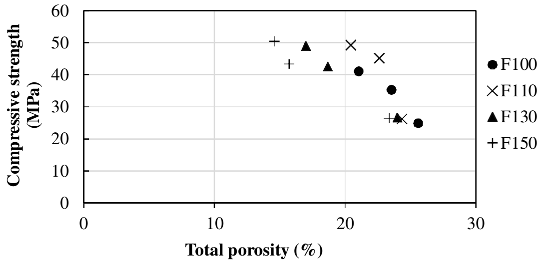

Fig. 11. Total porosity and compressive strength of mortars (series 1: controlled w/c = 0.6).

E. TGA Analysis

The mechanism of influence of fly ash in cement mortar is very complicated because fly ash works as both a fine aggregate and a reactive mineral additive. The filling effect of fly ash is provided in mortar when fly ash is used as a part of fine aggregate. However, fly ash is also reactive. The pozzolanic reaction of fly ash occurs, especially at late ages. This study performed TGA analysis to assess the reaction of fly ash in mortar, measuring the portlandite contents $Ca(OH)_2$ and the consumption of $Ca(OH)_2$ by the pozzolanic reaction of fly ash. Figure 12 shows the $Ca(OH)_2$ content measured in the mortars. The pozzolanic reaction of fly ash consumes a part of $Ca(OH)_2$, reducing the $Ca(OH)_2$ content in the mortars containing fly ash. In [25], it was reported that the pozzolanic reaction of fly ash started and consumed $Ca(OH)_2$ after 28 days of age. The mix proportions of this study had the same cement content. The consumption of $Ca(OH)_2$ in mortars containing fly ash as a partial replacement for sand was calculated by:

$$CH_{cons} = CH_{OPC}^{91d} - CH_{FA}^{91d} \quad (1)$$

where CH_{cons} is the consumption of $Ca(OH)_2$ in the mortar, CH_{OPC}^{91d} is the $Ca(OH)_2$ content in the mortar without fly ash at 91 days, and CH_{FA}^{91d} is the $Ca(OH)_2$ content in the mortar with fly ash at 91 days. This equation may not precisely determine the consumption of $Ca(OH)_2$ in mortars but is considered sufficient to demonstrate its trend. Figure 13 shows that the consumption of $Ca(OH)_2$ in both controlled flow and controlled w/c series of mortars at 91 days of age increased with increasing fly ash content. Fly ash consumes $Ca(OH)_2$ and helps improve compressive strength in addition to the total gradation improvement by the filling effect.

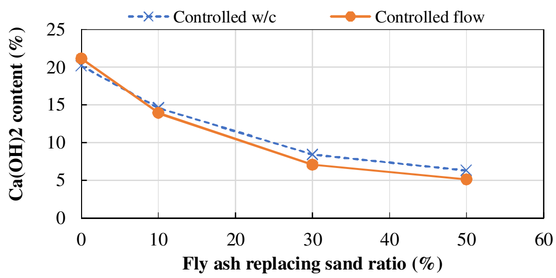

Fig. 12. $Ca(OH)_2$ content of mixtures.

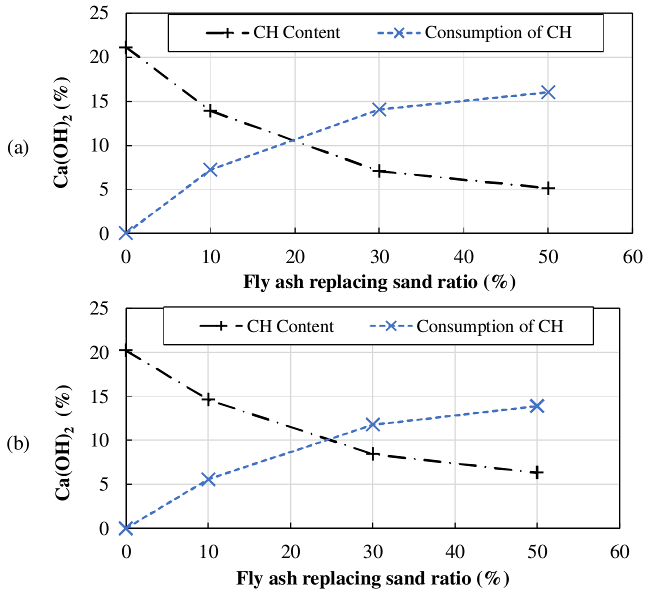

Fig. 13. $Ca(OH)_2$ content and consumption in mortars: (a) Controlled flow series, (b) controlled w/c=0.6 series.

IV. CONCLUSIONS

High-LOI fly ash is usually used as cement replacing material in concrete, as fill or embankment material. The application of high-LOI fly ash as a fine aggregate to produce mortar or concrete has not been investigated until recently. This study used high-LOI fly ash in mortar as a partial replacement for sand. This approach can solve the problem of natural sand shortage. Moreover, the pozzolanic reaction of fly ash can improve the performance of concrete. The replacement sand ratios were 0, 10, 30, and 50%. The effect of fly ash content was investigated in two series of mortar samples. In series 1, the water-to-cement ratio (w/c) was controlled at 0.6, while in series 2, the mortar flow was controlled at 110±5 mm. Experimental tests were carried out to determine flow, water requirement, setting times, and compressive strength. To clarify the influence of fly ash on the compressive strength of mortar, total porosity, $Ca(OH)_2$ content, and $Ca(OH)_2$ consumption were measured using TGA analysis. Based on the test results, the following conclusions can be drawn:

- A higher fly ash-to-aggregate ratio leads to lower flow, higher water requirement, and longer setting times of mortar. The flow of mortar is in reverse relation to the water requirement.
- At the first week of age, the compressive strength of mortars containing fly ash as a partial replacement for sand is not much different from that of the control mortar without fly ash. However, at later ages, the compressive strength increases significantly when using fly ash as a sand-replacing material.
- The improvement in compressive strength is supported by the results of total porosity. Especially at a later age, the total porosity of mortar decreases when using fly ash as a partial replacement for sand. However, there is no unique relationship between porosity and strength.
- The consumption of $Ca(OH)_2$ content in mortar increases with increasing fly ash content in place of sand. Higher consumption of $Ca(OH)_2$ content shows a higher degree of fly ash reaction, which also supports the improved compressive strength of mortars containing fly ash.
- The test results indicate a high potential for the use of high-LOI fly ash as a partial replacement for sand in concrete.

The results obtained were compared with previous studies, showing similarities. The compressive strength of cement mortar is only improved when the sand replacement ratio is low and does not exceed 30%. It can be said that there is a high potential to apply high-LOI fly ash as a partial sand replacement material in concrete, and more studies are needed in the future.

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