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Development of strong lightweight cementitious matrix for lightweight concrete simply by increasing a water-to-binder ratio in $Ca(OH)_2$-Na_2CO_3-activated fly ash system

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HIGHLIGHTS

- Class F fly ash was activated with varying weights of water, $Ca(OH)_2$, and Na_2CO_3.
- Its specific gravity (SG) was largely reduced simply by increasing water content.
- However, no excessive bleeding and strength reduction were not generated.
- Even the strongest sample (56.5 MPa) had a low oven-dried SG of 1.47.

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ABSTRACT

Reducing the specific gravity (SG) of structural binder pastes is one possible way to prevent material segregation between binder paste and lightweight aggregates, which often occurs in producing lightweight concrete due to their SG differences. To this end, this study suggests the use of a new lightweight fly ash binder paste, given that its SG can largely be reduced simply by increasing the water-to-binder (w/b) ratio without both excessive bleeding and significant strength reduction. Class F fly ash was activated with varying weights of water (i.e., w/b), $Ca(OH)_2$, and Na_2CO_3 to prepare the samples. Compressive strength, SG, and water absorption were measured with microstructural study using XRD, TG, and MIP. The results showed satisfactory performances with strengths of 27.8–56.5 MPa, an oven-dried SG of 1.24–1.47, and water absorption of 13.3–24 wt% for selected representative mixtures at 28 days. In particular, the strongest sample, at 56.5 MPa, also obtained a low value of oven-dried SG of 1.47, which lay within the range of oven dried SG (~ 0.6–1.5) of commercial lightweight aggregates. In this study, strength, SG, water absorption, reaction products, and pore characteristics were mainly determined by w/b ratio rather than $Ca(OH)_2$ and Na_2CO_3 contents; thus, this binder would be more advantageous for quality control than other types of fly ash-based cementless binders in structural construction.

1. Introduction

Recently, installation of long-span precast concrete (PC) members has increased in the modular construction of such things as storage buildings and parking facilities. While applying a long-span PC member, its self-weight should be less than the maximal lifting capacity of the crane. Weight reduction of the PC member is also important to significantly reduce the risk of earthquake damage, because natural period and base shear of building, which affect seismic load, are mainly influenced by the self-weight of the structural member. The use of structural lightweight concrete would be a possible solution to obtain longer spans, thinner sections, and less earthquake load responses [1–3]. However, in the process of lightweight concrete production, material separation (=segregation) and bleeding often occur due to substantial differences in specific gravity (SG) between ordinary portland cement (OPC) (SG = ~3.15) and commercial lightweight aggregates (LWA) (oven dried SG = ~0.6–1.5 [4,5]) such as pumice, VersaLite™, bottom ash, expanded shale, etc. Therefore, reducing the specific

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gravities of structural binder pastes (e.g., OPC, cementless binders), comparable to that of LWA, is desirable to prevent any segregation between binder paste and aggregates.

Adding foaming agents is a common way of reducing the self-weight of binder pastes to produce lightweight concrete. Foaming agents generate a large volume of artificial pores in hardened pastes, leading to reduced specific gravities of the pastes [6]. However, the foamed binder pastes are not suitable for structural purposes, due to their significantly low strength; thus, they are limited in their use to improvements in nonstructural properties such as thermal insulation or soundproofing capacity [7,8].

Increasing the weight ratio of water-to-binder (w/b) might be a simple way to reduce the specific gravity of binder paste, because increasing the volume of dried pores, originally filled with residual mixing water before drying due to the ambient humidity, would result in a decreased volume fraction of heavy solids in the paste. However, an excessive increase in w/b may cause severe bleeding in the binder paste [9]. It is worth noting that because a large difference in specific gravities between binder and water (SG = 1.0) is often responsible for bleeding, the extent of bleeding might be reduced after a large fraction of OPC is replaced with lighter cementitious materials (e.g., fly ash (SG = ~2.1), ground granulated blast furnace slag (SG = ~2.9)) [10,11]. In particular, because the specific gravity of fly ash is ~1.9–2.3, which is significantly lower than that of OPC, as the fraction of fly ash increases, the overall specific gravity of the binder paste also largely decreased. Thus, cementless fly ash binders with a high w/b may be a decent solution not only for reducing bleeding but also for decreasing the specific gravity of binder paste.

Commonly, fly ash is used to produce strong cementless binders (e.g., alkali-activated fly ash, geopolymer) through alkaline activation using alkaline activators (e.g., NaOH, sodium silicates) under relatively high temperature conditions (~60–90 °C) [12–14]. However, this type of fly ash binder is limited in practical use, because alkaline activators are normally very expensive and caustic (pH > ~14) [15,16].

The use of calcium hydroxide ($Ca(OH)_2$) for fly ash activation, hereafter referred to as "$Ca(OH)_2$ activation", may overcome the drawbacks of alkaline activators, as it is not only less expensive, but it also has a relatively low pH (~12.5 in saturated solution) compared to alkaline activators. In earlier studies [17–21], the $Ca(OH)_2$ activations for pozzolans (e.g., fly ash, volcanic ash) generally produced calcium silicate hydrate (C—S—H) as a main reaction product, similar to that of portland cement; however, they mostly generated notably lower compressive strengths, compared to those of alkali activation, when only $Ca(OH)_2$ was used without any other additive chemicals. Thus, the use of various additional chemicals was attempted to promote strength development. Shi et al. [17,21] found that the additions of Na_2SO_4 and $CaCl_2$, respectively, significantly improved the early hydration degree and later strength development in $Ca(OH)_2$ activation of volcanic ash; however, because these additives might lead to durability problems such as sulfate attack or steel corrosion, their use should be limited [18,19]. Other earlier studies also used $Na_2O{\cdot}SiO_2$ [20], NaOH, gypsum, or Na_2SO_4 [18] in $Ca(OH)_2$ activation for fly ash, but they did not provide any clear evidence of strength improvement.

Recently, Jeon et al. [22] reported that the use of Na_2CO_3 as an additive had a significant effect on the strength improvement of $Ca(OH)_2$-activated fly ash, mainly due to the substantial promotion of C—S—H formation from more dissolution of fly ash and the subsequent pore-size refinement.

The main purpose of this study is to reduce the self-weight of hardened binder paste simply by increasing the w/b of cementless fly ash binder paste activated with $Ca(OH)_2$ and Na_2CO_3 to produce a strong lightweight binder matrix. To this end, various types of mixtures were designed for the purpose of exploring their mechanical properties and microstructures. Mechanical and physical properties such as compressive strength, specific gravity in a saturated surface dried condition (SSD), specific gravity in an oven-dried condition (OD), and the extent of water absorption for hardened pastes were measured. To identify important experimental factors for these measured properties, powder X-ray diffraction (XRD), a thermogravimetric (TG) analysis, and mercury intrusion porosimetry (MIP) were also conducted.

2. Materials and test methods

In this study, Class F fly ash (FA) was obtained from the Boryeong power plant in the Republic of Korea. The chemical composition of FA was examined using an X-ray fluorescence (XRF) spectrometer (LAB CENTER XRF-1800, Shimadzu, Japan). The result is shown in Table 1. The particle size distribution of FA was measured with a laser diffraction particle size analyzer (HELOS with a RODOS dispersing unit, Sympatec, Germany), as presented in Fig. 1. The fly ash showed typical characteristics of fly ash in Korea in terms of oxide composition and particle size distribution.

The mixture proportions are shown in Table 2. The FA was activated with analytical grades of calcium hydroxide ($Ca(OH)_2$) (Junsei Chemical, Japan) and sodium carbonate (Na_2CO_3) (Sigma Aldrich, US). In this study, calcium hydroxide and sodium carbonate are denoted CH and SC, respectively, and the dry powder mixture of FA, CH, and SC are denoted the CNF powder. Thirty-nine paste mixtures were designed to have various weight ratios of w/b, CH/FA, and SC/FA. As shown in Table 2, the estimated material costs were relatively low compared to the market price of portland cement in Korea (~US$65/ton). In the mixture proportions, representative mixtures were selected to measure specific gravities, water absorption, XRD, TG, and MIP; the 0.6W/30CH/7SC was firstly selected as a reference sample because (1) it used w/b = 0.6, which was a maximal value of w/b without excessive bleeding, and (2) CH = 30% and SC = 7% were found to be suitable as reference values in trial tests, which were conducted prior to this study. After setting 0.6W/30CH/7SC as a reference sample, we varied w/b, CH, and SC. These samples were marked with 'R' in Table 2.

In this study, at 60 min after mixing, volumes of bleeding water of CNF binder pastes were measured at a range of w/b ratios to find proper w/b values that produced only acceptable degrees of bleeding water below 3% vol., almost invisible to the naked eye. To this end, a powder of CNF binder with a weight ratio of FA:CH:SC = 100:30:7 was prepared. Using this powder, 40 ml of CNF binder paste was made at a range of w/b from 0.4 to 1.0 in a 50 ml plastic tube; then, after 60 min, the volume of bleeding water

Table 1
Oxide composition of raw fly ash.

Oxide	Content (wt%)
SiO_2	62.6
Al_2O_3	19.5
CaO	5.4
Fe_2O_3	5.3
K_2O	1.7
Na_2O	1.4
MgO	1.2
TiO_2	1.1
SO_3	0.6
P_2O_5	0.5
SrO	0.2
BaO	0.2
ZrO_2	0.1
MnO	0.1