

Durability Controlled Hydration Mechanisms of High-Volume FABA-Based Binders with Nano Silica and Silica Fume Modification

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Abstract

High-volume supplementary cementitious materials (SCMs) offer significant potential for reducing clinker consumption and CO₂ emissions, but maintaining hydration efficiency and long-term durability remains a major challenge. This study investigated the hydration behavior, microstructural evolution, and transport-related durability of high-volume fly ash–bottom ash (FABA) binders incorporating mechanically activated bottom ash, nano-silica, and silica fume. The binder systems were evaluated using mechanical, transport, and integrated microstructural characterization. The 60% FABA (M1) binder exhibited the optimum performance, achieving a compressive strength of 44.5 MPa at 56 days together with the lowest sorptivity and water absorption. Integrated SEM, XRD, TGA/DTG, and BET/BJH analyses confirmed enhanced secondary hydration, progressive matrix densification, and mesopore refinement, with the highest specific surface area (11.9 m²/g) and the smallest average pore diameter (12.6 nm). These findings demonstrate that the durability of high-volume FABA binders is governed primarily by hydration-induced pore refinement and reduced pore connectivity, providing a scientific basis for the design of durable low-carbon cementitious binders with high clinker replacement.

Keywords

Fly Ash-Bottom Ash (FABA), Mechanical Activation, Supplementary Cementitious Materials, Hydration, Low-Carbon Cementitious Binders

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1. INTRODUCTION

The cement industry contributes significantly to global CO₂ emissions, with clinker production emitting approximately 0.8–0.9 t CO₂ per ton of clinker (Scrivener et al., 2018; ASTM International, 2022). High-volume supplementary cementitious materials (SCMs) can substantially reduce clinker consumption and associated emissions, particularly at replacement levels of 60–80% (ASTM International, 2022; Global Cement and Concrete Association, 2021). Among these, high-volume fly ash–bottom ash (FABA) binders offer a promising route for clinker reduction and valorization of coal-combustion residues, provided that their mechanical performance and durability can be maintained (Bhandari et al., 2023; Oyejobi et al., 2024).

Fly ash is widely used as an SCM because its pozzolanic constituents promote secondary C–S–H formation and contribute to long-term performance (Deng et al., 2024; Schackow et al., 2025; Ho and Huynh, 2023; Tosti et al., 2020). However,

high fly ash replacement can slow hydration, reduce early-age strength, and increase fluid transport (Choi, 2025; Kim et al., 2022; Wang et al., 2020). Bottom ash is generally less reactive and is more commonly used as aggregate than as an SCM (Kumar et al., 2021; Li et al., 2025; Pramudiyanto et al., 2025). Mechanical activation can increase its reactivity by reducing particle size and increasing reactive surface area (Mousa, 2023). Meanwhile, silica fume and nano-silica can promote secondary pozzolanic reactions, C–S–H nucleation, and pore refinement (Kalinkin et al., 2025; Pormmoon et al., 2024; Bhandari et al., 2023). Their combined use can enhance hydration and mechanical performance, although excessive SCM replacement may reduce the calcium availability required for sustained secondary hydration (Ho and Huynh, 2023; Kim et al., 2022; Tao and Massoudi, 2024).

Despite these advances, the coupled relationship between mechanically activated bottom ash reactivity, calcium avail-

ability, secondary hydration, pore refinement, and transport-related durability in high-volume FABA binders remains insufficiently established (Alnezami et al., 2025; Gómez-Pérez et al., 2025). In particular, it remains unclear how increasing FABA replacement affects the balance between clinker-derived hydration and supplementary pozzolanic reactions, and how this balance governs pore connectivity and fluid transport. Sorptivity and water absorption therefore provide important complementary indicators of durability beyond compressive strength because they reflect the accessibility and connectivity of the pore network (Scrivener et al., 2018; Tibbetts et al., 2020; Ho and Huynh, 2023).

Accordingly, this study investigates the hydration behavior, microstructural evolution, mechanical performance, and transport-related durability of FABA binders containing 60-80% FABA, with mechanically activated bottom ash and constant additions of nano-silica and silica fume. Compressive strength, sorptivity, and water absorption were evaluated together with SEM, XRD, TGA/DTG, and BET/BJH analyses to establish the relationship between hydration, microstructure, and transport resistance (Kim et al., 2022; Wang et al., 2020; Kang et al., 2024; Ho and Huynh, 2023).

The novelty of this study is the integrated hydration-microstructure-durability assessment of high-volume FABA binders in which mechanically activated bottom ash is utilized as a reactive SCM. By combining quantitative SEM, XRD, TGA/DTG, and BET/BJH analyses with transport measurements, the study elucidates how secondary hydration and pore refinement are associated with improved transport resistance. The results further indicate that the 60% FABA mixture provides an effective balance between clinker-derived calcium availability and FABA reactivity, consistent with greater pore refinement and improved transport resistance. This provides a mechanistic basis for developing durable low-carbon binders with high clinker replacement.

2. EXPERIMENTAL SECTION

2.1 Materials

Ordinary Portland cement (OPC) and Portland composite cement (PCC), supplied by PT Semen Indonesia (Persero) Tbk, were used as reference binders. OPC complied with ASTM C150, whereas PCC conformed to SNI 7064:2014. Fly ash (FA) and bottom ash (BA) were obtained from the Suralaya coal-fired power plant, Indonesia, and used as supplementary cementitious materials. Silica fume (SF), supplied by PT Sika Indonesia, consisted of ultrafine amorphous silica (0.1-1 μm). Nano-silica (NS) powder, supplied by Wacker Chemicals, had an average particle size of approximately 20 nm and a SiO_2 purity of 99.8%.

A polycarboxylate ether (PCE)-based superplasticizer supplied by CV John Hi-Tech Contrindo was used to improve workability and ensure uniform particle dispersion. Mixing water was obtained from the Engineering Laboratory of CV John Hi-Tech Contrindo, Depok, Indonesia.

2.2 Binder Composition

FABA binders were prepared using a fixed fly ash-to-bottom ash ratio of 80:20, with total FABA replacement levels of 60%, 70%, and 80% by mass of the binder. The contents of nano-silica (0.1%), silica fume (0.2%), and polycarboxylate ether (PCE) superplasticizer (0.2%) were kept constant to isolate the effect of FABA replacement. These dosages were selected based on previous studies demonstrating their effectiveness in promoting hydration and pore refinement without excessive agglomeration or increased water demand (Kim et al., 2022; Wang et al., 2020).

Reference mixtures containing 100% OPC and 100% PCC were included for comparison. All mixtures were prepared with a constant water-to-binder ratio (w/b) of 0.45, following durability-oriented mix design principles for high-volume SCM systems (Ho and Huynh, 2023; Ribeiro et al., 2025). The detailed mixture proportions are presented in Table 1.

Control mixtures consisting of 100% OPC and 100% PCC were prepared for comparison. The water-to-binder ratio was maintained at 0.45 for all mixtures to ensure comparable hydration conditions and enable meaningful comparison of performance indicators. The detailed mix proportions are summarized in Table 1.

2.3 Mixing Procedure and Specimen Preparation

All dry materials were thoroughly pre-mixed before nano-silica, pre-dispersed in the mixing water, and a polycarboxylate ether (PCE) superplasticizer were added to minimize particle agglomeration and ensure homogeneous mixing (Wang et al., 2020; Choi, 2025). Cement paste was cast into $50 \times 50 \times 50$ mm cube molds following ASTM C109, demolded after 24 h, and water-cured at room temperature until testing at 7, 28, and 56 days. All tests were conducted on three replicate specimens, and the reported results represent the mean values.

2.4 Methods

2.4.1 Chemical Characterization

The chemical compositions of OPC, PCC, FA, BA, SF, and NS were determined using X-ray fluorescence (XRF) analysis performed with a PAN alytical Epsilon 1 spectrometer (Malvern Panalytical, UK). XRF characterization of cementitious materials was conducted following established practices for oxide composition determination (Scrivener et al., 2018).

2.4.2 Scanning Electron Microscopy (SEM)

Scanning electron microscopy (SEM) was performed using a JEOL JSM-6510LV microscope (JEOL Ltd., Japan) to examine the morphology of representative paste samples (OPC, PCC, and selected FABA mixtures). Quantitative image analysis was conducted on SEM micrographs using image-processing software to determine particle size distribution based on at least 100 particles per sample. The analysis included particle segmentation, identification, and equivalent particle diameter measurement to evaluate the effect of mechanical activation

Table 1. Mix Proportions of Cement Paste Mixtures

Code	OPC (%)	PCC (%)	FABA (%)	NS (%)	SF (%)	PCE (%)	W/B ratio
C0	100	-	-	-	-	-	0.45
C1	-	100	-	-	-	-	0.45
M1	39.5	-	60	0.1	0.2	0.2	0.45
M2	29.5	-	70	0.1	0.2	0.2	0.45
M3	19.5	-	80	0.1	0.2	0.2	0.45

Table 2. Result of X-Ray Fluorescence (XRF) Materials

Chemical Properties	Pozzolan Type		High-Reactivity Pozzolan		OPC	PCC	M1
	FA	BA	NS	SF			
SiO ₂	55.60	50.40	99.8	94.20	18.02	17.40	30.40
Al ₂ O ₃	26.80	21.20	-	0.22	5.02	4.75	12.72
Fe ₂ O ₃	6.90	9.70	-	0.29	3.35	2.10	18.34
CaO	3.70	4.16	-	0.78	64.95	57.06	31.61
MgO	1.80	2.90	-	0.46	1.89	1.77	0.89
SO ₃	1.1	0.71	-	0.75	0.92	2.12	1.04
Na ₂ O + K ₂ O	0.60	1.3	-	0.56	0.46	0.87	1.71

on bottom ash particle refinement and its relationship with hydration reactivity.

2.4.3 X-Ray Diffraction (XRD)

Phase identification was performed using X-ray diffraction (XRD) with a PANalytical X’Pert Pro MPD diffractometer (Malvern Panalytical, UK). X-ray diffraction (XRD) analysis was conducted at 28 days to identify the representative hydration and pozzolanic reaction phases.

2.4.4 Thermogravimetric and Derivative Thermogravimetric (TGA/DTG)

Thermogravimetric and derivative thermogravimetric (TGA/DTG) analyses were performed on selected paste samples at 28 and 56 days to evaluate hydration products and pozzolanic reactions (Li and Scrivener, 2025; Snehal et al., 2023). The calcium hydroxide (CH) content was calculated from the mass loss between 400 and 500 °C using the stoichiometric relationship $CH = \text{mass loss} \times 74/18$. Hydration behavior was interpreted through integrated XRD–TGA/DTG analysis.

2.4.5 Surface Area and Pore Structure Characterization

Pore structure characterization was conducted using nitrogen adsorption–desorption (BET/BJH) analysis to evaluate specific surface area and pore size distribution. These parameters provide indirect assessment of pore connectivity and microstructural refinement in the binder system.

2.4.6 Compressive Strength

Compressive strength tests were conducted on 50 mm cubic specimens in accordance with ASTM C109 at curing ages of 7, 28, and 56 days. The reported values represent the average of at least three specimens per mixture. Statistical significance of compressive strength results was evaluated using one-way

ANOVA followed by a post hoc t-test at a 95% confidence level ($p < 0.05$) to assess differences between mixtures.

2.4.7 Durability-Related Transport Properties

Water absorption and sorptivity tests were performed at 28 and 56 days to evaluate the transport-related durability of the binders. Water absorption was measured in accordance with (ASTM International, 2022), while sorptivity was determined following (ASTM International, 2020) and expressed in mm/√s. Both initial sorptivity (Si) and secondary sorptivity (Ss) were evaluated to characterize capillary water transport at early and later absorption stages (ASTM International, 2022, 2020; Tibbetts et al., 2020).

2.4.8 Surface Area and Pore Structure Characterization

Specific surface area and pore structure were analyzed by N₂ adsorption-desorption using a Micromeritics ASAP 2020 analyzer. After degassing at 105–150 °C for 12 h, the specific surface area was determined by the BET method (ISO 9277:2010), while pore size distribution and cumulative pore volume were calculated using the BJH method. The results were used to evaluate pore refinement in the developed FABA binders.

3. RESULTS AND DISCUSSIONS

3.1 Chemical Composition and Reactivity Implications

The chemical compositions of OPC, PCC, fly ash, bottom ash, silica fume, and nano-silica determined by X-ray fluorescence analysis are summarized in Table 2. Fly ash exhibits a high combined content of SiO₂, Al₂O₃, and Fe₂O₃, confirming its pozzolanic nature and suitability as a supplementary cementitious material (Scrivener et al., 2018; Deng et al., 2024). In contrast, bottom ash shows lower silica content and a comparatively higher CaO fraction, explaining its lower intrinsic

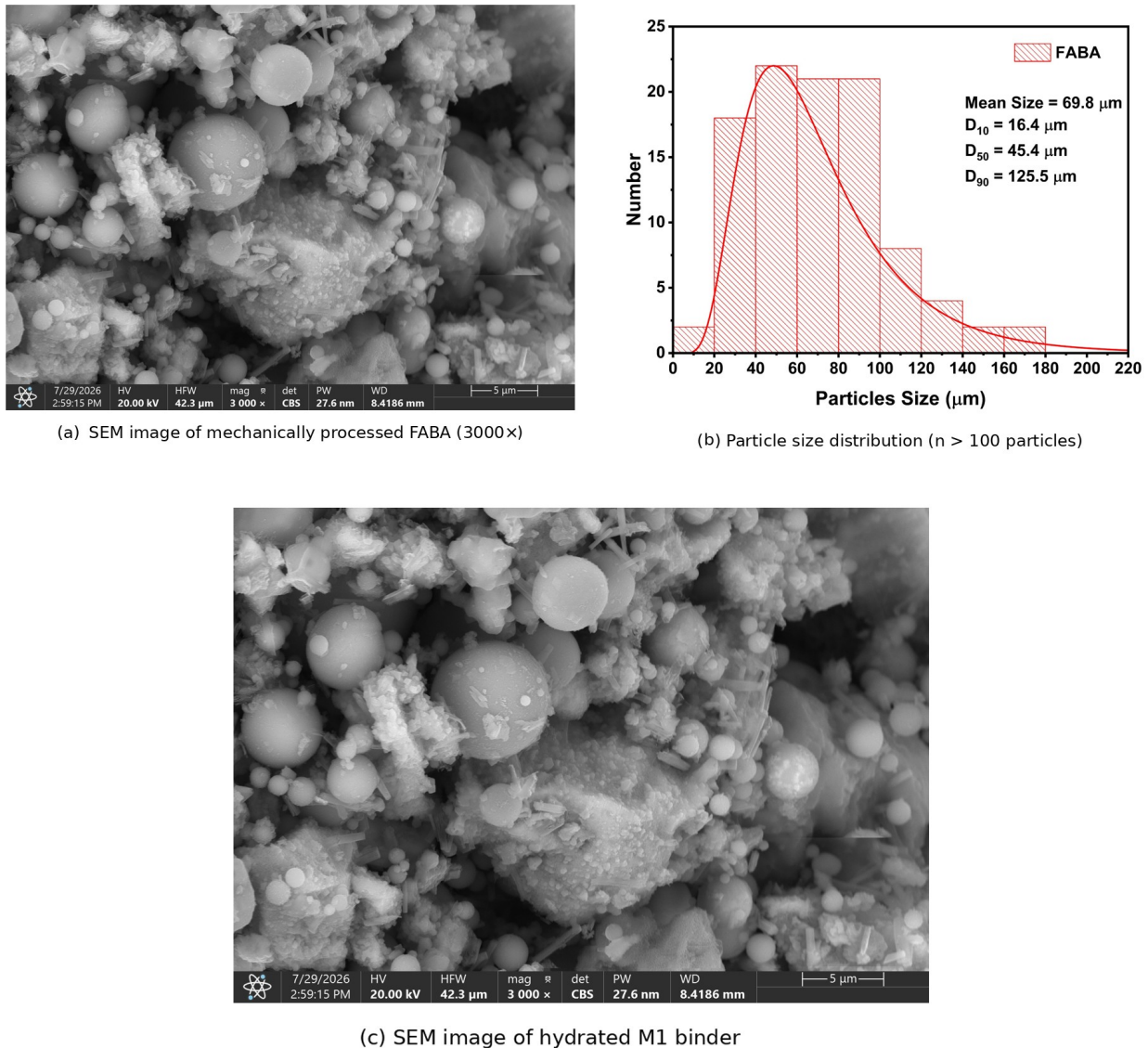


Figure 1. SEM Images of Mechanically Processed FABA and Hydrated M1 Binder with Quantitative Particle Size Distribution

pozzolanic reactivity when used in its as-received form (Mousa, 2023; Schackow et al., 2025).

The high SiO_2 contents of nano-silica and silica fume further support their function as reactive micro and nano-scale modifiers, providing additional sources of amorphous silica for secondary hydration reactions rather than acting as inert fillers.

3.2 Microstructural Evolution and Hydration Products

3.2.1 Quantitative SEM Image Analysis of Mechanically Processed FABA and Hydrated M1 Binder

SEM was employed to evaluate the morphology of mechanically processed fly ash-bottom ash (FABA) and the hydrated microstructure of the optimized M1 binder. As shown in Figure 1(a-b), mechanical activation produced irregular and angular particles with fractured surfaces, indicating effective particle

refinement and increased reactive surface area. Quantitative image analysis revealed a unimodal particle size distribution with a mean particle size of 69.8 μm , and characteristic diameters of $D_{10} = 16.4 \mu\text{m}$, $D_{50} = 45.4 \mu\text{m}$, and $D_{90} = 125.5 \mu\text{m}$. Approximately 50% of the particles were smaller than 45.4 μm , demonstrating the effectiveness of mechanical processing in increasing the proportion of reactive particles.

The hydrated M1 binder (Figure 1(c)) exhibited a dense and continuous matrix dominated by interconnected C-S-H and C-(A)-S-H gels with limited visible capillary pores, indicating progressive matrix densification through secondary pozzolanic reactions. This microstructure suggests enhanced hydration and the formation of additional secondary hydration products. The reduction in particle size increased the reactive surface available for the dissolution of amorphous silica and alu-

Table 3. Mineralogical Phase Composition of FA and BA (XRD Analysis)

Phase	Chemical Formula	Fly Ash (FA)	Ash Bottom Ash (BA)	Reactivity Implication
Amorphous phase	-	High	Moderate	Primary source of pozzolanic reactivity
Quartz	SiO ₂	Minor	High	Inert, reduces overall reactivity
Mullite	Al ₆ Si ₂ O ₁₃	Moderate	Minor-Moderate	Low reactivity, stable phase
Calcite	CaCO ₃	Minor	Moderate	Potential Ca source, limited reactivity
Hematite	Fe ₂ O ₃	Minor	Minor	Largely inert
Anhydrite/Gypsum	CaSO ₄ / CaSO ₄ ·2H ₂ O	Trace	Trace-Minor	Sulfate source, affects hydration kinetics

mina, thereby accelerating secondary pozzolanic reactions and promoting the nucleation of additional C–S–H/C–(A)–S–H gels. As a result, the refined particle morphology facilitated matrix densification and reduced capillary pore connectivity, which contributed to the improved transport resistance of the M1 binder.

These observations are consistent with the integrated XRD, TGA/DTG, and BET/BJH results, which confirmed progressive portlandite consumption, increased specific surface area, and pore refinement. The reduced pore connectivity is further supported by the lower sorptivity and water absorption of the M1 mixture. Collectively, the SEM analysis demonstrates that mechanical activation enhances the reactivity of FABA and contributes to improved durability through hydration-induced microstructural refinement rather than compressive strength.

These SEM observations are quantitatively supported by BET/BJH characterization (Section 3.2.4), where the M1 binder exhibited the highest specific surface area (11.9 m²/g) together with the smallest average pore diameter (12.6 nm), confirming hydration-induced pore refinement and development of a dense mesoporous network.

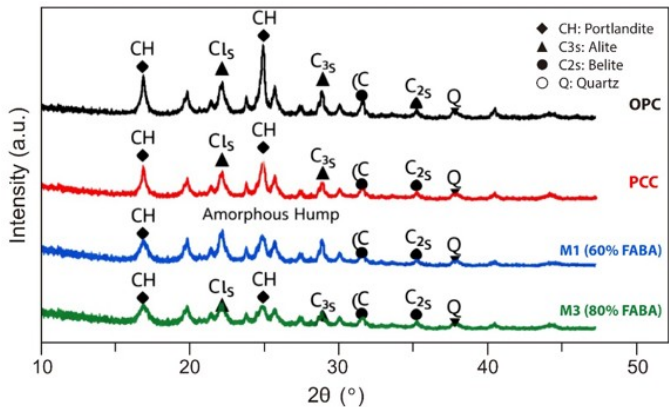


Figure 2. XRD Patterns of OPC, PCC, M1, and M3 at 28 Days with CH Peaks and an Amorphous Hump (20-35° 2θ)

3.2.2 XRD Analysis of Raw Materials and Hydrated Binders

The XRD-based phase identification indicates (Table 3) that fly ash is dominated by amorphous aluminosilicate phases, which are responsible for its high pozzolanic reactivity. In contrast, bottom ash exhibits a higher fraction of crystalline phases such as quartz and calcite, which contributes to its lower intrinsic reactivity. This difference in phase composition explains the need for mechanical activation to enhance the reactivity of bottom ash in high-volume SCM systems.

3.2.3 Integrated Hydration and Pozzolanic Reaction Mechanisms (XRD–TGA/DTG)

The hydration and pozzolanic reaction mechanisms were investigated using integrated X-ray diffraction (XRD) and thermogravimetric/derivative thermogravimetric (TGA/DTG) analyses. XRD identifies the evolution of crystalline hydration products, whereas TGA/DTG quantifies calcium hydroxide (CH) consumption, providing complementary evidence of secondary hydration in high-volume FABA binders (Li and Scrivener, 2025).

The XRD patterns (Figure 2) show pronounced portlandite peaks in OPC, reflecting clinker-dominated hydration, while PCC exhibits lower peak intensity because of partial clinker replacement. In contrast, the FABA mixtures, particularly M1 (60% FABA), display significantly reduced portlandite peaks together with a broad amorphous hump (20-35° 2θ), indicating effective pozzolanic reactions and the formation of secondary C–S–H and C–(A)–S–H gels. At higher replacement levels (M3), the further reduction of clinker-related phases suggests that excessive clinker dilution limited calcium hydroxide availability for continued secondary hydration.

The TGA/DTG results (Figure 3) support these observations. Portlandite contents decreased from 18.5 wt.% (OPC) and 15.6 wt.% (PCC) to 10.3 wt.% (M1) and 7.4 wt.% (M3), confirming progressive CH consumption through pozzolanic reactions. Between 28 and 56 days,

the FABA binders exhibited continued CH consumption accompanied by increased bound water, demonstrating sustained secondary hydration. The M1 mixture retained sufficient cal-

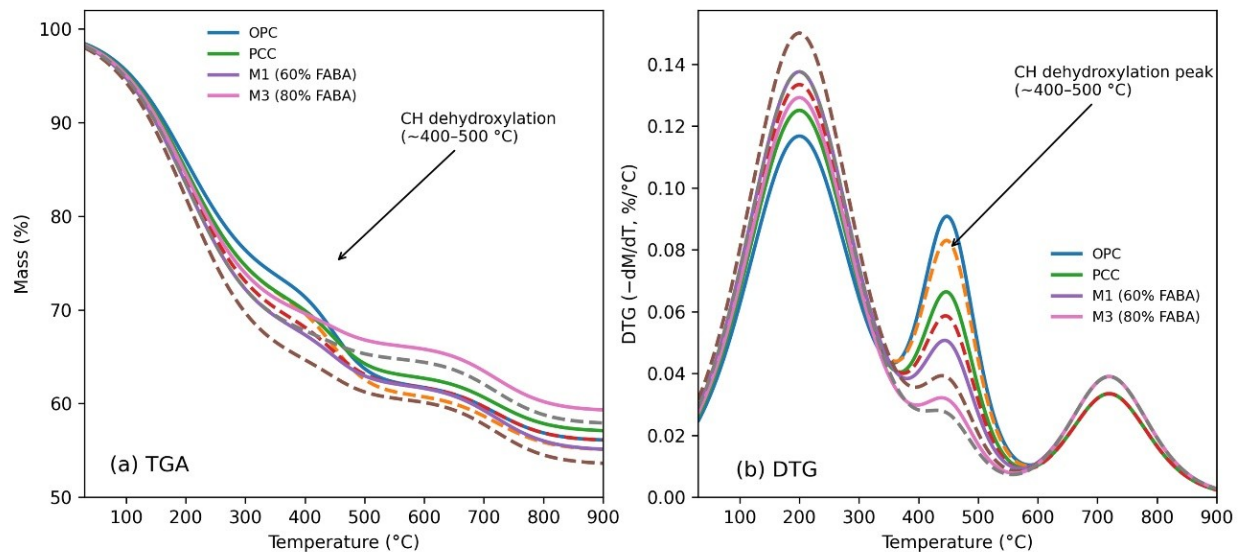


Figure 3. TGA and DTG Curves of OPC, PCC, M1, and M3 Paste Samples at 28 and 56 Days

cium hydroxide to support ongoing hydrate formation, whereas hydration in M3 became calcium-limited because of excessive clinker replacement (Snehal et al., 2023).

The integrated XRD-TGA/DTG results demonstrate that mechanically activated bottom ash, together with nano-silica and silica fume, enhanced secondary hydration by promoting CH consumption and the formation of additional C-S-H and C-(A)-S-H gels. These findings agree with the SEM observations of matrix densification and the BET/BJH results showing pore refinement, providing a mechanistic explanation for the improved transport resistance and durability of the optimized 60% FAB binder (Kang et al., 2024).

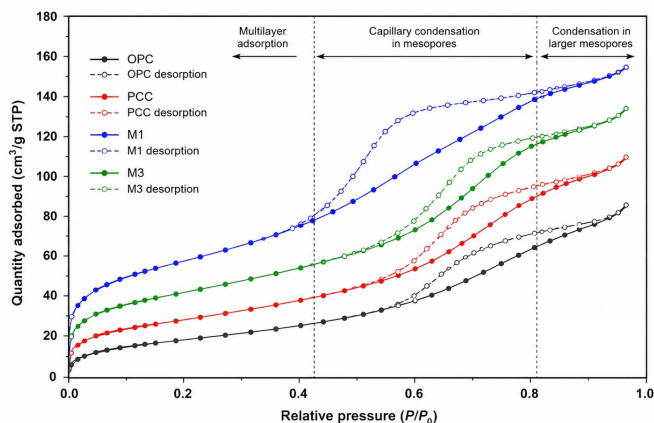


Figure 4. Nitrogen Adsorption-Desorption Isotherm (Type IV-H3) of OPC, PCC, M1 and M3

3.2.4 BET/BJH Surface Area and Pore Structure Characterization

Nitrogen adsorption-desorption (BET/BJH) analysis was conducted to evaluate the evolution of pore structure in the investigated binders. The specific surface area, average pore diameter, and total pore volume are summarized in Table 4, while the corresponding adsorption-desorption isotherms, pore size distributions, and cumulative pore volume curves are presented in Figures 4-6.

Table 4 shows that the optimized M1 (60% FAB) mixture exhibited the highest BET surface area ($11.9 \text{ m}^2/\text{g}$) and the smallest average pore diameter (12.6 nm), whereas the M3 (80% FAB) mixture showed a lower surface area ($9.3 \text{ m}^2/\text{g}$) and a larger average pore diameter (17.8 nm). Although M3 presented a slightly higher total pore volume, its larger pore diameter indicates a less refined pore structure.

The nitrogen adsorption-desorption isotherms (Figure 4) exhibit typical Type IV isotherms with H3 hysteresis loops, confirming that all binders are predominantly mesoporous. The higher adsorption capacity of the M1 mixture indicates the development of a larger accessible internal surface associated with continuous formation of secondary C-S-H and C-(A)-S-H gels. In contrast, the lower adsorption capacity of M3 suggests that excessive FAB replacement restricted hydration because of limited calcium hydroxide availability.

The BJH pore size distributions (Figure 5) further demonstrate that the M1 mixture developed a narrower pore size distribution concentrated within the fine mesopore range, indicating a more homogeneous pore network. Conversely, the broader distribution observed in M3 reflects the persistence of larger interconnected mesopores caused by incomplete secondary hydration.

The cumulative pore volume curves (Figure 6) support these observations. Although M3 exhibited the highest cumulative pore volume, its larger average pore diameter indicates a greater proportion of coarse mesopores. In contrast, the refined pore structure of M1 is characterized by smaller pore diameters and lower pore connectivity, which are more effective in restricting fluid transport.

The refined mesoporous structure of the M1 mixture explains its lower sorptivity and water absorption compared with the higher FABAs mixtures. Therefore, the improved durability of the optimized binder is primarily attributed to hydration-induced pore refinement and reduced pore connectivity rather than total pore volume or compressive strength alone.

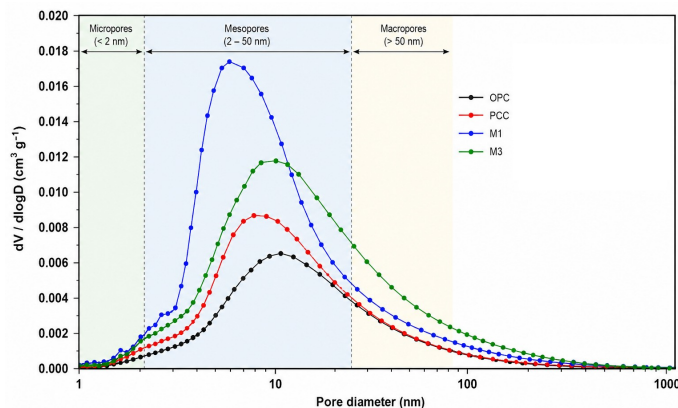


Figure 5. BJH Pore Size Distribution of OPC, PCC, M1 and M3

3.3 Compressive Strength Development

Compressive strength results (mean $\pm$ standard deviation, $n = 3$) are presented in Figure 7. The OPC mixture exhibited the highest compressive strength at all curing ages because of its higher clinker content and faster hydration, while PCC showed slightly lower strength due to partial clinker replacement.

The FABAs binders showed reduced early-age strength with increasing replacement level. The M1 (60% FABAs) mixture achieved 15.9 MPa at 7 days, whereas the 70% and 80% FABAs mixtures exhibited progressively lower strengths owing to clinker dilution and slower hydration. However, significant strength development occurred between 28 and 56 days, with M1 reaching 44.5 MPa at 56 days. This improvement is attributed to continued secondary pozzolanic reactions that generated additional C–S–H and C–(A)–S–H gels, consistent with the SEM observations of matrix densification and the XRD–TGA/DTG evidence of progressive portlandite consumption (Kim et al., 2022; Wang et al., 2020).

In contrast, mixtures containing $\geq 70\%$ FABAs exhibited lower ultimate strengths because excessive clinker replacement limited calcium hydroxide availability for sustained secondary hydration. These findings indicate that 60% FABAs provides an optimum balance between clinker hydration and pozzolanic

reactivity, enabling satisfactory later-age strength while supporting the pore refinement and transport resistance discussed in the following sections (Kang et al., 2024; Ho and Huynh, 2023).

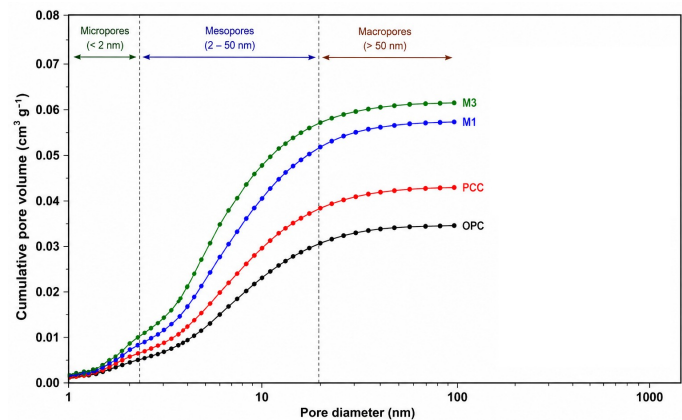


Figure 6. Cumulative Pore Volume Versus Pore Diameter for OPC, PCC, M1 and M3

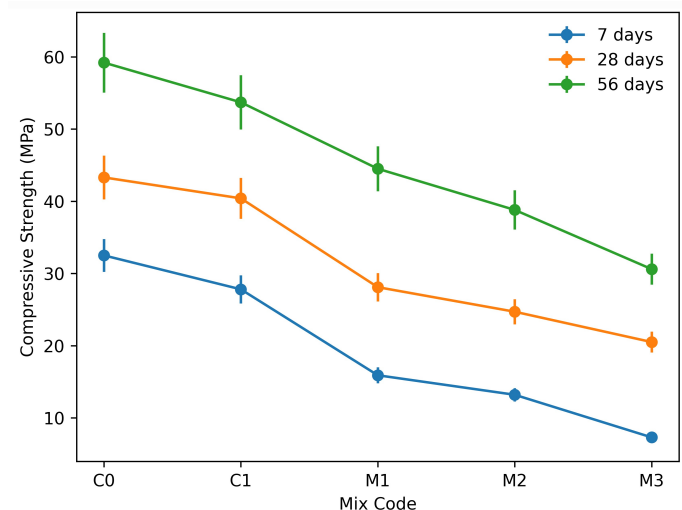


Figure 7. Compressive Strength of Cement Paste Mixtures at 7, 28, and 56 Days

3.4 Sorptivity

Sorptivity results (mean $\pm$ standard deviation, $n = 3$) are presented in Figure 8. At 28 days, sorptivity increased with increasing FABAs replacement, indicating higher capillary pore connectivity in high-volume SCM binders. Accordingly, the OPC and PCC mixtures exhibited the lowest initial sorptivity owing to their denser hydration.

At 56 days, all FABAs mixtures showed reduced sorptivity, with the M1 (60% FABAs) mixture exhibiting the greatest reduction (approximately 25–35%) compared with its 28-day value. This improvement is attributed to continued secondary

Table 4. BET Surface Area and Pore Structure Characteristics

Sample	BET Surface Area (m ² /g)	Average Pore Diameter (nm)	Total Pore Volume (cm ³ /g)
OPC	4.9	22.8	0.034
PCC	6.2	19.7	0.043
M1 (60% FABA)	11.9	12.6	0.058
M3 (80% FABA)	9.3	17.8	0.062

pozzolanic reactions, which promoted the formation of additional C–S–H and C–(A)–S–H gels, refined the pore structure, and reduced capillary pore connectivity. In contrast, the M3 (80% FABA) mixture maintained comparatively higher sorptivity because excessive clinker replacement limited calcium hydroxide availability for sustained secondary hydration (Ho and Huynh, 2023; Pormmoon et al., 2024; Tibbetts et al., 2020; Kang et al., 2024).

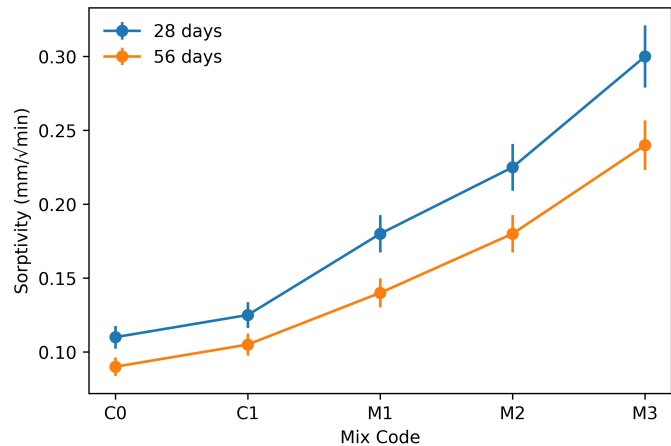


Figure 8. Sorptivity of Cement Paste Mixtures at 28 and 56 Days

3.5 Water Absorption

Water absorption results values represent mean ± standard deviation (*n* = 3) presented in Figure 9. At 28 days, Water absorption increased with increasing FABA replacement, indicating a larger volume of accessible pores due to clinker dilution and slower hydration in high-volume SCM systems. Accordingly, the OPC and PCC mixtures exhibited the lowest water absorption because of their denser hydration matrices (Ho and Huynh, 2023; Kumar et al., 2021). With continued curing, all mixtures showed reduced water absorption, reflecting progressive pore refinement through secondary hydration. The reduction was most pronounced for the M1 (60% FABA) mixture, indicating that an appropriate balance between clinker hydration and pozzolanic reactions promoted the formation of additional C–S–H and C–(A)–S–H gels, thereby reducing pore connectivity and limiting water ingress. In contrast, the M3 (80% FABA) mixture exhibited comparatively higher water absorption because excessive clinker re-

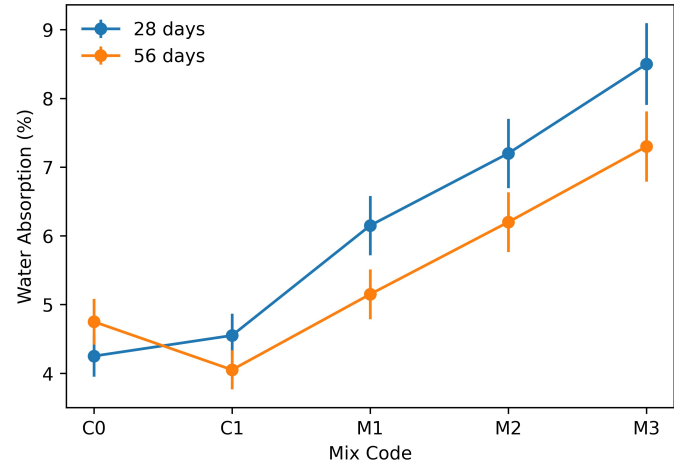


Figure 9. Water Absorption of Cement Paste Mixtures at 28 and 56 Days

placement limited calcium hydroxide availability for sustained secondary.

These observations indicate that continued secondary hydration refined the pore structure and reduced pore connectivity, thereby lowering water absorption in the M1 mixture (Ho and Huynh, 2023; Pramudiyanto et al., 2025; Cui et al., 2025).

3.6 Microstructure Durability Relationship of High Volume FABA Based Binders

The durability of high-volume FABA binders is governed primarily by hydration-induced microstructural evolution, particularly pore refinement and pore connectivity, rather than compressive strength alone. The integrated interpretation of SEM, XRD, TGA/DTG, BET/BJH, sorptivity, and water absorption consistently demonstrates the relationship between hydration reactions, microstructure development, and transport-related durability (Tibbetts et al., 2020; Pramudiyanto et al., 2025). As shown in Figure 10, the microstructure of both M1 and M3 evolved significantly between 28 and 56 days. At 28 days, both mixtures exhibited heterogeneous matrices containing partially connected capillary pores, resulting in relatively high sorptivity and water absorption. With continued curing, the M1 (60% FABA) mixture developed a denser and more continuous matrix dominated by secondary C–S–H and C–(A)–S–H gels, effectively reducing pore connectivity. In contrast, the M3 (80% FABA) mixture exhibited limited microstructural

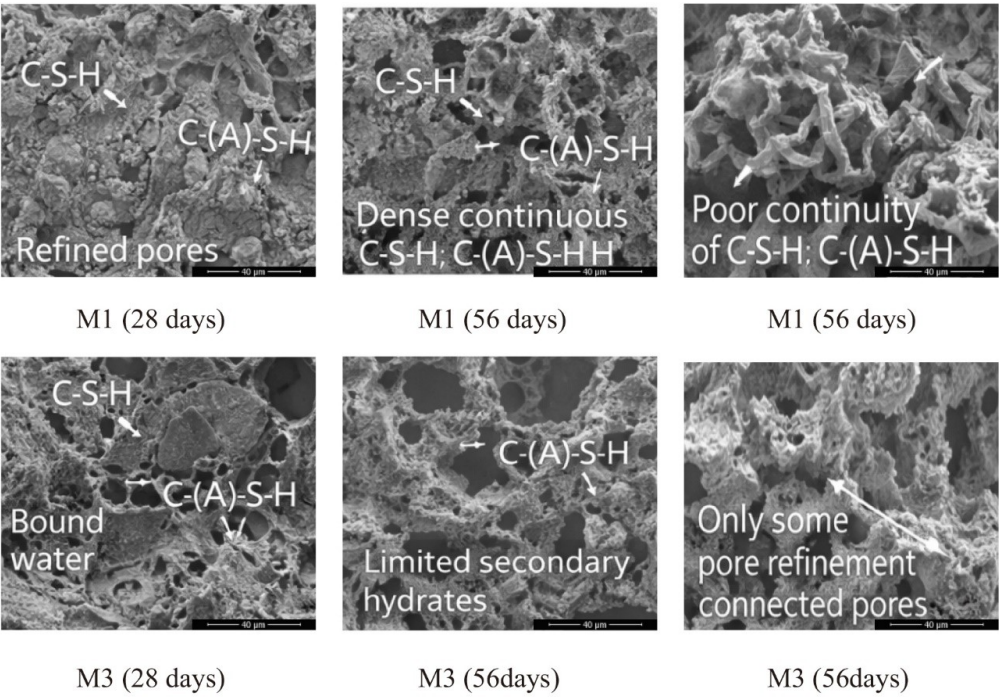


Figure 10. The Microstructural Evolution of M1 and M3 at 28 and 56 Days

Table 5. Comparison of Durability Performance with Recent Literature

Study	Binder System	SCM Replacement (%)	Sorptivity (mm/√s) ×10 ⁻³	Water Absorption (%)
(Ho and Huynh, 2023)	High-volume FA cement	50-60	6.5-7.5	7.2-8.4
(Kang et al., 2024)	Mechanically activated BA cement	40-50	5.6-6.6	6.3-7.4
(Cui et al., 2025)	FA + nano-silica cement	55-65	4.5-5.3	5.4-6.3
This study (M1–M3)	High-volume FABA + SF + NS	60-80	3.2-4.5	4.3-6.0

refinement because excessive clinker replacement reduced calcium hydroxide availability, restricting secondary hydration and leaving a more interconnected pore network.

The refined mesoporous structure explains the lower sorptivity and water absorption measured for M1, indicating that improved durability is primarily associated with reduced pore connectivity rather than total porosity or compressive strength (Tibbetts et al., 2020).

The results demonstrate that the optimum durability of high-volume FABA binders is achieved through a balanced interaction between clinker-derived calcium hydroxide and supplementary cementitious material reactivity. The synergistic effects of mechanically activated bottom ash, nano-silica, and silica fume promoted continuous secondary hydration, matrix densification, and pore refinement, providing an effective strategy for developing durable low-carbon cementitious binders with high clinker replacement.

3.7 Benchmarking of Durability Performance with Previous Studies

To evaluate the competitiveness of the proposed binder, the durability performance of the optimized high-volume FABA system was compared with representative low-carbon cementitious binders reported in recent studies (Table 5). The comparison was based on sorptivity and water absorption, which are widely accepted indicators of transport-related durability (Ho and Huynh, 2023; Pormmoon et al., 2024; Pramudiyanto et al., 2025).

As shown in Table 4, the high-volume FABA-based binders Table 5 shows that the developed FABA binder achieved lower sorptivity ($3.2-4.5 \times 10^{-3} \text{ mm}/\sqrt{s}$) and water absorption (4.3-6.0%) than previously reported fly ash- and bottom ash-based systems, despite incorporating a higher SCM replacement level (60-80%). These results indicate that the combined use of mechanically activated bottom ash, nano-silica, and silica fume

effectively enhances pore refinement and transport resistance (Kim et al., 2022; Wang et al., 2020; Tibbetts et al., 2020).

The superior durability performance is attributed to the synergistic effects of particle refinement, sustained secondary pozzolanic reactions, and matrix densification, as demonstrated by the integrated SEM, XRD, TGA/DTG, and BET/BJH analyses. The comparison confirms that the proposed binder provides improved durability at substantially higher clinker replacement, supporting its potential as a durable low-carbon cementitious material (Ho and Huynh, 2023; Pormmoon et al., 2024; Pramudiyanto et al., 2025).

4. CONCLUSIONS

This study demonstrated that mechanical activation effectively enhanced the reactivity of bottom ash, promoting secondary pozzolanic reactions and improving the hydration performance of high-volume fly ash–bottom ash (FABA) binders. Among the investigated mixtures, the 60% FABA binder (M1) exhibited the optimum performance, achieving the highest later-age compressive strength together with the lowest sorptivity and water absorption. Integrated SEM, XRD, TGA/DTG, and BET/BJH analyses confirmed that these improvements were associated with progressive matrix densification, mesopore refinement, and reduced pore connectivity. The findings demonstrate that the durability of high-volume FABA binders is governed primarily by hydration-induced microstructural refinement rather than compressive strength alone, providing a scientific basis for the development of durable low-carbon cementitious binders with high clinker replacement.

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