

Prediction of microstructural evolution in fly ash-modified cementitious system: A computational study

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The intricate interaction between supplementary cementitious materials (SCMs) and cementitious systems profoundly influences the performance and sustainability of cementitious composites. This study explores the microstructural evolution of fly ash (FA)-modified cement paste by employing a three-dimensional cement hydration and microstructure development (CEMHYD3D) modeling package. Through comprehensive simulations, the influence of varying FA content on hydration phase evolution and pore structure within the cementitious system is revealed. As the proportion of FA within the cementitious mixtures increases, there is a substantial enhancement in the rate of hydration. Notably, the incorporation of FA introduces a significant augmentation in the hydration rate, a phenomenon with potential implications for the long-term performance of FA-modified cementitious materials. The prediction results also highlight that increasing FA substitution in cement leads to finer and more interconnected pore networks due to the pozzolanic reaction. These perceptions hold significant implications for optimizing cementitious mixes and advancing sustainable construction practices. The model-predicted results have been validated with experiments, and they are successful in predicting the microstructural evolution in FA-modified cement paste. In summary, the prediction model bridges the theoretical and practical implementation gaps by providing a thorough understanding of the microstructural evolution of FA-modified cement paste. Furthermore, it provides invaluable guidance for tailoring FA-blended cement compositions, thus promoting their enhanced performance and sustainability in the realm of cementitious materials.

Keywords: *cementitious system; FA, prediction model; microstructural evolution; pore structure*

1. Introduction

Microstructural properties of cementitious materials determine the macro performance of structures. The microstructural evolution during cement hydration is a complicated process due to the multi-scale nature of the porous material. Microstructures of cementitious composites are studied using a wide variety of experimental methods, such as scanning electron microscopy (SEM) [1], X-ray diffraction (XRD) [2], Vicat needle and penetration resistance [3], electrical resistivity [4], and ultrasonic pulse velocity [5]. The microstructural and mechanical properties of various supplementary cementitious materials (SCMs) incorporating cementitious systems were previously studied [6]. The formation of microstructure in blended cements has previously been experimentally studied [7]. However, in all

cases, an experimental approach takes more time and resources to execute.

The recent growth in computer programs such as three-dimensional cement hydration and microstructure development (CEMHYD3D) has made microstructure modeling of cementitious materials achievable. Based on the hydration of cementitious systems that use only Portland cement, a number of computer-based models such as CEMHYD3D [8], HYMOSTRUC3D [9, 10], μ ic [11], and DuCOM [12] were developed to predict various properties of microstructure. Several multi-scale models for cement hydration [13] and the microstructure simulation of Portland cement-only systems [14] were established and have been in use throughout the last few decades. The existing models of hydration were developed to predict the hydration of composites made of only pure Portland cement. Previously, a prediction model to determine the microstructure of the Portland cement system was developed by [15].

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Computer models were employed to simulate the microstructure of Portland cement-based materials, as detailed in [16].

Even though many models have been established to predict different properties of the microstructure of Portland cement-based composites, they cannot fully replace experimental investigations, but they can shed light on the process, especially in light of the growing use of numerous SCMs such as fly ash (FA). FA is a byproduct of the combustion of pulverized coal in power plants and is one of the most commonly utilized SCMs in concrete. Researchers have explored the incorporation of FA into cementitious systems to understand its effect on the microstructure [17], mechanical properties [18], and long-term performance of cementitious materials [19]. The primary reason to incorporate SCMs in cement is to minimize the use of Portland cement in cementitious composites and thereby reduce the carbon footprint [20]. More specifically, the substitution of FA in cement improves the durability properties of cementitious materials [21].

The hydration of cements modified with FA is more complex than that of cementitious systems made only with Portland cement due to the reaction of minerals available in FA. The microstructure of FA-modified cements has been extensively studied with experiments [18]. However, there is currently no model available for simulating the microstructural evolution of blended cement pastes with FA. Therefore, there is a need to propose an appropriate model to predict the evolution of microstructures in FA-modified cement pastes.

While the utilization of FA in cementitious materials has gained considerable attention in the construction industry due to its sustainability benefits, a comprehensive understanding of the microstructural evolution within such systems remains incomplete. Existing literature primarily focuses on experimental investigations and rudimentary theoretical models, lacking a detailed predictive model that can elucidate the complicated microstructural changes over time. This study seeks to bridge this gap by employing advanced computational modeling techniques to predict and analyze the evolution of microstructure

in FA-modified cementitious systems. By bridging this gap and offering a comprehensive understanding of the evolution of microstructure in cement systems containing FA, this study contributes to the advancement of science, aiding engineers and researchers in making informed decisions for the development of more sustainable cementitious materials.

2. Materials and methods

2.1. Materials

The materials used in this study include Type I cement or ordinary Portland cement (OPC) and FA. The OPC utilized in this study was a commercially available cement, confirming ASTM C 150 specifications. FA cement confirming ASTM C 618 (AASHTO M 295) was also commercially available and was used to make an FA-blended mixture. All materials were stored in a controlled environment to prevent moisture absorption and contamination.

2.2. Method

First of all, the investigation was initiated by determining the particle size distribution (PSD) and chemical composition of each cement. This step was undertaken to establish the PSD and oxide compositions, which are essential for subsequent computational modeling purposes. The PSD of the cements was obtained through the application of the laser diffraction (LD) technique, as detailed in Figure 1 below.

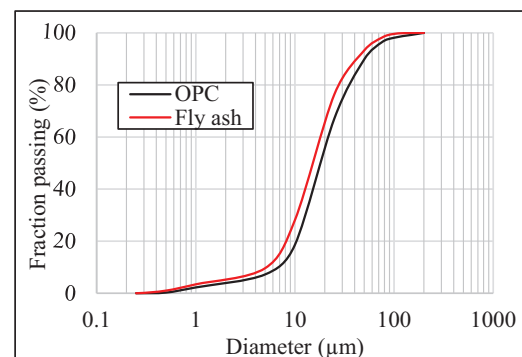


Fig. 1. Particle size distribution of OPC and FA used in the model

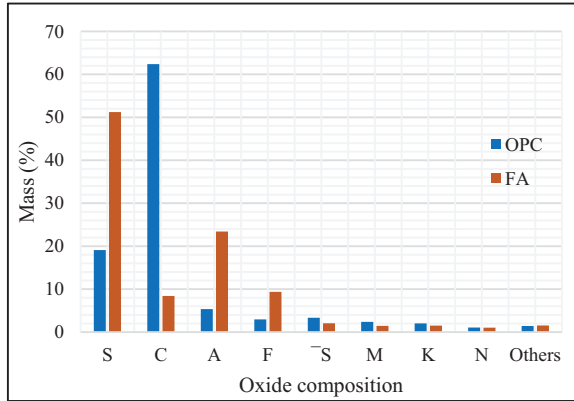


Fig. 2. Oxide composition of OPC and FA used in the model (mass%)

In cement, various oxides are found in the composition of cementitious materials. Some of the common oxides include SiO_2 , CaO , Al_2O_3 , Fe_2O_3 , SO_3 , MgO , K_2O , and Na_2O . These oxides are typically represented using chemical symbols and short forms as $\text{S}=\text{SiO}_2$, $\text{C}=\text{CaO}$, $\text{A}=\text{Al}_2\text{O}_3$, $\text{F}=\text{Fe}_2\text{O}_3$, $\bar{\text{S}}=\text{SO}_3$, $\text{M}=\text{MgO}$, $\text{K}=\text{K}_2\text{O}$, $\text{N}=\text{Na}_2\text{O}$, $\text{T}=\text{TiO}_2$, $\bar{\text{C}}=\text{CO}_2$, and $\text{H}=\text{H}_2\text{O}$. In this study, the oxide composition of the cements was obtained using the X-ray diffraction (XRD) technique, as presented in Figure 2. Using the characteristics of the cements, various cement pastes were created to replicate the microstructural evolution of cement pastes modified with FA.

The experimental investigation aimed to analyze the microstructural evolution within FA-modified cement paste. Cement paste specimens were created, each featuring distinct FA replacement levels of 10%, 20%, 30%, and 40%. To ensure a standardized approach, we rigorously maintained a constant w/c of 0.45 for all samples. The curing of the samples took place under controlled conditions at a standard temperature of 20°C , ensuring a saturated environment for optimal hydration development. The curing duration was determined based on the specific testing age required for analysis. A thermogravimetric analysis (TGA) technique was utilized to measure and assess the levels of hydrate water and calcium hydroxide (CH) present in both unmodified and FA-modified cement pastes. This technique involves subjecting samples to controlled temperature changes while

measuring the weight variations and providing insights into the hydration process and resulting products. The porosity of FA-modified cement pastes was measured using the mercury intrusion porosimetry (MIP) technique. In order to determine the pore size distribution and porosity, this approach includes introducing mercury into the pore network of the sample while applying increasing pressure.

The extended version of a CEMHYD3D model was employed in this study to predict the microstructural evolution of FA-modified cement paste. This computational approach considers the various factors influencing microstructure formation, such as FA content, cement chemistry, and curing conditions. The modeling process involved inserting the experimental parameters, including FA replacement levels, w/c ratio, and curing conditions, into the CEMHYD3D model. The model then performed simulations based on these inputs to predict the progression of microstructural features over time, including hydration phases, porosity, and pore distribution. The predicted microstructural properties were subsequently compared to the experimental measurements obtained through TGA and MIP analyses. This validation process aimed to assess the accuracy of the prediction of the CEMHYD3D model and its application to simulate the microstructural evolution within FA-blended cement paste. By combining experimental analysis with the predictive capabilities of the CEMHYD3D model, this research helps to advance a comprehensive insight into the microstructural changes occurring in FA-blended cement paste over time.

3. Microstructure modeling

While various characteristics of cement-based materials can be readily ascertained, the reaction processes of hydration and microstructure formation remain complex. Nonetheless, advancements in computational science and a diverse array of models have facilitated the simulation of microstructures in recent times. These projections can be effectively validated using experimental outcomes, given the utilization of precise

data that aligns with the experimental conditions in the model. The microstructure was represented through discrete entities referred to as volume pixels, which portray hydrated or non-hydrated pores within the model.

The modeling process involves the creation of a virtual microstructure by discretizing the cementitious system into a grid or lattice. The various constituents of the system, including cement particles, FA particles, water, and other additives, are represented within this grid. CEMHYD3D then simulates the chemical reactions and physical interactions among these components as the system undergoes hydration and other aging processes.

Therefore, using the extended CEMHYD3D model, the microstructure of cement pastes containing FA has been simulated in this study. Based on w/c, PSD, chemical compositions, and the distribution of mineral phases, a preliminary microstructure was developed in this model. Then, a cellular-automata algorithm was applied to run through multiple reaction cycles to complete the hydration process. The output of the CEMHYD3D simulation provides valuable understanding of the microstructural evolution of FA-modified cementitious systems over time. Finally, at any hydrated stage, one can obtain microstructure information such as capillary pores and the rate of hydration.

3.1. Hydration of Portland cement

The hydration process of Portland cement can be computed using the basic hydration equation expressed in Equation (1) for each chemical composition in the cement [22]. The chemical reactions as well as the diffusion processes during hydration of a single cement are all considered in this model:

$$\frac{d\alpha}{dt} = \left[\frac{3 \left(\frac{S_w}{S_0} \right) \rho_w C_{w-free}}{(v + w_g) r_0 \rho_c} \right] \times \left[\frac{1}{\left(\frac{1}{k_d} - \frac{r_0}{D_e} \right) + \frac{r_0}{D_e} (1 - \alpha)^{-\frac{1}{3}} + \frac{1}{k_r} (1 - \alpha)^{-\frac{2}{3}}} \right] \quad (1)$$

where α is the rate of hydration of cement, v is the mass ratio of water to cement, w_g is the physically-bound water in the C-S-H gel, S_w is the effective surface area of the cement particle that are in contact with water, S_0 is the total surface area, C_{w-free} is the physically free water from the C-S-H gel, r_0 is the radii of an unhydrated cement particle, ρ_c is the density of cement, and ρ_w is the density of water.

The reaction coefficient, k_d , is supposed to be proportional to the rate of hydration of the cement, whereas the values of the coefficients B and C govern the initial rates of shell formation and decay, respectively. This can be expressed with Equation (2) below:

$$k_d = \frac{B}{\alpha^{1.5}} + C\alpha^3 \quad (2)$$

In the diffusion process, the effective diffusion coefficient value of the water, D_e , changes based on the radii of the gel pore in the cementitious system. Equation (3) presents an expression for this phenomenon in terms of the rate of hydration.

$$D_{e0} = \ln \left(\frac{1}{\alpha} \right) \quad (3)$$

While the mineral components in each cement undergo hydration, the quantity of free water, C_{w-free} within the capillary pores decreases. Therefore, the amount of water within the capillary pores changes in accordance with the hydration rate, which can be mathematically expressed by Equation (4) below:

$$C_{w-free} = \frac{W_0 - 0.4\alpha C_0}{W_0} \quad (4)$$

where C_0 and W_0 represent the initial percentages by mass of cement and water, respectively, in the mix.

3.2. Hydration of FA-blended cement

Hydration models essentially provide microstructural properties at different stages of hydration. Microstructural properties such as elastic modulus, rate of hydration, autogenous shrinkage, and transport properties can be

predicted using these images. Cement particles are typically made up of tri-calcium silicate (C_3S), di-calcium silicate (C_2S), tri-calcium aluminate (C_3A), and tetra-calcium aluminate ferrite (C_4AF) mineral phases. The hydration model for a blended cement system is formulated by summing up the hydration processes involved in cement clinkers and the SCMs in the mix as a single component when considering the interaction between mineral components and the reaction of SCMs.

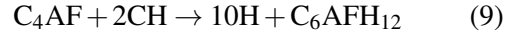
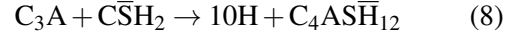
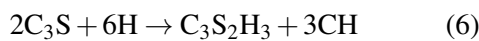
The rate of hydration in FA-blended cement can be computed based on the method described in [9, 12], and [23]. Therefore, the cumulative rate of hydration of both Portland cement and FA at a specific time, $t_j(\alpha_j^{cem}, \alpha_j^{FA})$ can be expressed with Equation (5) as follows:

$$\alpha_j^i = \frac{1}{G_i(x_{max}^i - 1)} \sum_{z=x_{min}^{cem}}^{x_{max}^i-1} \alpha_{z,j}^i \cdot W_z^i \quad (5)$$

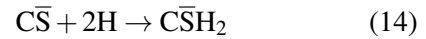
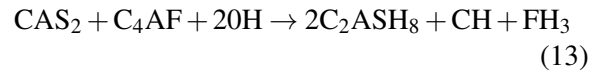
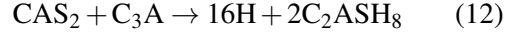
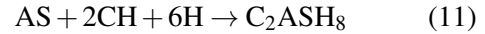
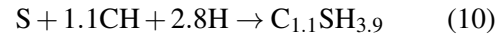
where $G_i(x)$ is the Rosin-Rammler function, which was used to determine the PSD of i particles. x_{min}^i and x_{max}^i represent the minimum and maximum particle size of i particle, respectively. W_z^i is the mass of i particles in a given fraction. With information on the PSD of i particles, it is possible to calculate the values of $G_i(x)$ and W_z^i . $\alpha_{z,j}^i$ denotes the reaction degree of i particles with size j at time t_j ; it can be calculated using the penetration depth of the i particles with size z , which can be determined using the basic hydration equation.

3.3. Formation of hydration products

Hydration modeling basically covers the chemical effects of the cement minerals on hydration reactions. The amount of hydration products like calcium-silicate-hydrate (C-S-H) and calcium hydroxide (CH) in a cement paste can be calculated by the balance equation of pozzolanic reaction and cement hydration. According to the theories put forth by [24], the reactions that the minerals in Portland cement undergo during the hydration stage can be described with Equations (6)–(9) as follows:



Similar to other pozzolanic materials, the main effect of FA substitution into cement is to lower the ratio of Ca/Si in C-S-H by reducing the amount of portlandite generated. The main constituents of FA are calcium, silicon, and aluminum oxide, which combine to form a glassy material [25]. Higher concentrations of SiO_2 can be found in the glassy phases, while magnetite, mullite, quartz, and hematite are the most common crystalline phases. Some FA have a higher concentration of CaO in their glassy phases, and their crystalline phases are more likely to be quartz, lime, and periclase [26]. Based on the study of [27], the equation for the chemical reaction of FA-modified cementitious systems can be described with Equations (10)–(14) as follows:



The amount of CH in the paste decreases due to the pozzolanic reaction involving FA in cementitious systems. Therefore, the CH content, m_{CH} , in the system can be computed with Equation (15). This computation accounts for the chemical composition and reactivity of the specific FA being used:

$$m_{CH} = m_{CH_0} - m_{CH_{FA}} \quad (15)$$

where m_{CH} is the total CH content in the system after the pozzolanic reactions, m_{CH_0} is the initial CH content in the system before the pozzolanic reactions, and $m_{CH_{FA}}$ is the amount of CH consumed due to the pozzolanic reactions with FA.

Over past few decades, numerous distinct hydration models have been meticulously formulated and widely adopted, each specifically applied for the analysis of pure Portland cement.

The best approach to developing a hydration model for a cement system containing FA blends would be to extend the verified model for Portland cement. Therefore, this study employed the extended CEMHYD3D modeling program from the open source.

4. Results and discussions

The utilization of computational modeling tools, such as CEMHYD3D, to predict the microstructural evolution of FA-modified cement paste considering the reaction between cementitious phases and FA particles. This section undertakes a comprehensive analysis and discussion of the outcomes derived from CEMHYD3D simulations, casting light on the influence of FA content on hydration phase evolution, pore structure, and connectivity within the cementitious system. The simulation started with the initialization of the CEMHYD3D model, where the user defines the parameters and properties of the cementitious system being studied. This includes specifying the composition of the cementitious mix, which consists of Portland cement and FA. The model requires input data such as the chemical composition of the Portland cement and FA, PSD, w/c ratio, and curing conditions. The key aspect of the simulation involves modeling the hydration of cementitious materials. CEMHYD3D considers the chemical reactions that occur as water interacts with cement and FA particles. This includes the dissolution of cement and FA phases, the formation of hydration products such as C-S-H gel and CH, and the evolution of pore solution chemistry. The model predicts the kinetics of these reactions in cement paste containing FA.

4.1. Phase evolution and hydration process

The microstructural evolution of FA-modified cement pastes with various substitution levels of FA was simulated using the extended CEMHYD3D model. In the simulation, it is possible to calculate a wide range of microstructural parameters such as

heat of hydration, phase volume fractions, chemical shrinkage, and porosity. In the computer model, microstructural properties like heat of hydration, CH formation, and pore structure were simulated.

The prediction yielded valuable insights into the progression of phases and the distribution of hydration products within the cementitious system containing FA. An intriguing observation was made as the FA content increased, subtle shifts occurred in the composition of reaction products. The introduction of FA particles brought about additional nucleation sites, fostering the generation of hydrates and consequently promoting a more extensive formation of C-S-H. This phenomenon bears significance for the material's mechanical properties, as the secondary C-S-H gel enhances both the acceleration of strength development and the improvement of microstructural compaction.

Additionally, the simulations revealed the formation of C-A-S-H gels in the presence of FA, further contributing to the overall densification of the microstructure of cement paste. The simulated figures (a, b, c, and d) in Figure 3, corresponding to various levels of FA replacement, vividly depict this phenomenon. Over a span of 180 days, the extended CEMHYD3D model aptly predicted the rate of hydration and the evolution of microstructure in FA-modified cement pastes, underscoring its reliability.

From the model predictions, the influence of FA on cement hydration appears to be minimal when up to 10% FA is incorporated into the cement mixture. As the FA content increases to 20% in Portland cement, the rate of hydration for the blend surpasses that of a cement system containing only Portland cement, particularly during the later stages beyond 90 days. However, the rate of hydration in early ages, up to 28 days, is a little slower than that of the later ages when compared with the hydration of pure Portland cement pastes. When about 30% FA is replaced with cement, the rate of hydration in the system is still higher than that of pure Portland cement, and this is true when up to 40% FA is substituted. The simulation shows that the rate of hydration for FA-blended cement demonstrates an increased trend after 28 days,

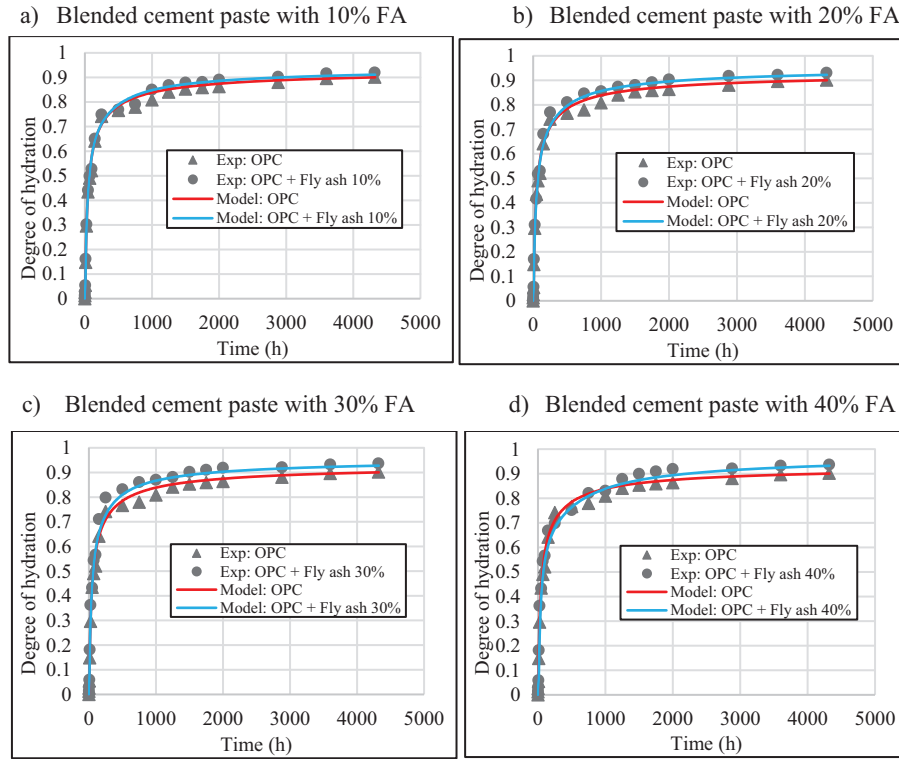


Fig. 3. Simulated degree of hydration in blended cement with 10%, 20%, 30%, and 40% FA substitution for $w/c = 0.45$

which is consistent with an increase in the FA substitution level.

When FA is added to cementitious systems, it can have a significant effect on the formation of CH and the microstructural evolution of the resulting cement pastes. FA reacts chemically with CH to form additional cementitious compounds, such as C-S-H. As a result, when FA is added to cement paste, it reduces the amount of CH as illustrated in Figure 4. This reduction in CH content can be simulated in terms of volume fractions with the CEMHYD3D model.

4.2. Effect on pore structure

The initial parameters for pore structure analysis of the cement paste were set before the simulation started. The chemical composition of the mixture, FA content, w/c ratio, temperature, and curing condition were normally considered when determining the pore structure. Based on these parameters, the CEMHYD3D model simulated

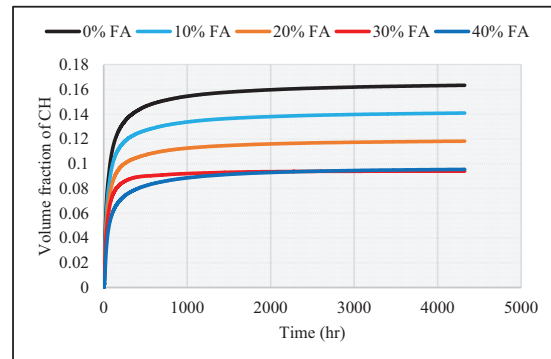


Fig. 4. Comparison of CH formation in FA-modified cement paste

the microstructure images of hydrated blended cements with various substitution levels of FA at the age of 180 days.

The microstructure images in Figure 5 shows the pore structure of various cement pastes denoted as A, B, C, and D, each featuring different levels of FA substitution. It is evident that as the FA content in the cement increases, the rate of hydration

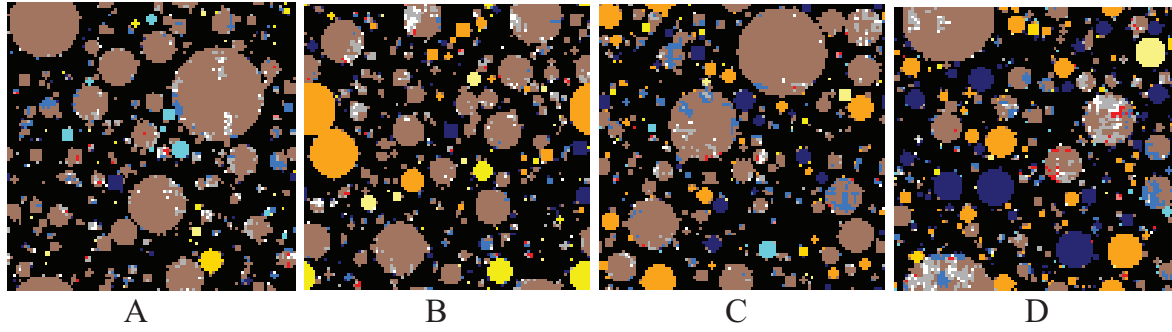


Fig. 5. Simulated microstructure of FA-blended cement pastes from the CEMHYD3D model with various FA substitutions. The image is $256\mu\text{m} \times 200\mu\text{m}$

notably increases in the later stages, beyond 90 days, and this continues until the particular curing period of 180 days. When the hydration time increases, the hydrated products take up a larger volume and the mineral phases shrink, and this determines the pore structure of the system. The colors in the microstructure images indicate the various mineral phases present in cement.

The CEMHYD3D modeling program tracks changes in the pore structure of the cement paste, including the size and distribution of pores. FA can affect the pore structure by filling some of the voids or influencing the formation of smaller pores. Therefore, understanding the evolution of porosity is crucial for predicting the mechanical and durability properties of cementitious materials.

The microstructure simulation encompassed various levels of FA substitution, denoted as A (10% FA), B (20% FA), C (30% FA), and D (40% FA), all simulated at the age of 180 days with a constant w/c of 0.45. Distinct cement phases are represented by different colors within the images. Specifically, C_3S is depicted in brown, C_2S in blue, C_3A in gray, C_4AF in white, K_2SO_4 in red, gypsum in yellow, free lime in green, free silica in aqua, periclase in pink, and the pore structure in black.

The simulations revealed that elevating the FA replacement level yielded finer and better-connected pore networks. This phenomenon can be attributed to the pozzolanic reaction of FA, which fosters the creation of supplementary reaction products and the refinement of pore sizes. The intensified pore interconnectivity signifies improved durability, reduced permeability, and

Table 1. Pore size distribution of FA-blended cement paste at the age of 180 days

FA Replacement (%)	Pore Size < 10 nm (%)	Pore Size 10–100 nm (%)	Pore Size > 100 nm (%)
0	12.50	56.50	31.00
10	11.10	55.30	33.60
20	10.20	54.90	34.90
30	9.30	54.20	36.50
40	8.40	53.40	38.20

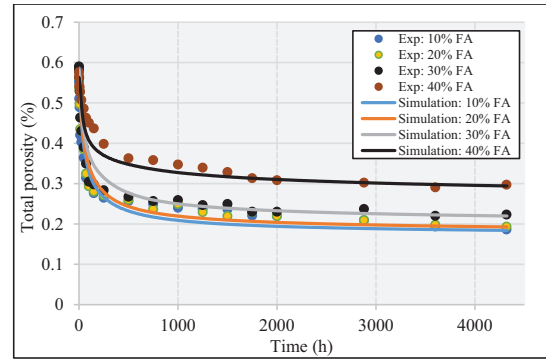


Fig. 6. Model-predicted effect of FA on pore structure of cement paste at the age of 180 days

heightened resistance against detrimental agents like chlorides and sulfates.

From the simulation results, the porosity of cement blended with FA demonstrates an upward trend with increasing percentages of FA addition, extending up to the age of 180 days, as illustrated in Figure 6. These simulations also probed the interactions between FA particles and the cementitious system. It was observed that FA particles

played a role as nucleation spaces for the formation of reaction products, consequently fostering an intensified interfacial transition zone between the FA and the encompassing cement paste. This phenomenon impacted the connectivity of the pore network, adding to the overall reduction in pore sizes and the improvement of mechanical performance. The presence of FA particles also exerted an influence on the distribution of CH crystals, which formed around the FA particles, influencing the overall pore filling and connectivity. These findings underscore the role of FA in modifying the microstructure and mechanical behavior of the cementitious system. Microstructures from a scale of nanometers to micrometers govern the permeability of cement-based materials [14]. That is why microstructural properties are important to exhibit excellent durability properties in concrete. Using sufficiently small voxels, a detailed microstructural property can be obtained with simulation [28]. From the simulation results, as the degree of hydration increases, the outer hydrate layer expands while the volume of small capillary pores in the system decreases.

The small capillary pores result from insufficient packaging of hydrates in the layer of outer products, while the large capillary pores are left out of the layer of outer products. The large capillary pores can percolate throughout the entire material; high permeability can be anticipated, or they can be connected through small capillary pores, and low permeability can be anticipated. Further developments in computational models will incorporate some algorithms to simulate the pore structure of blended cement pastes, which will result in more refined microstructures and may improve the accuracy of the pore structure of hydrated phases in blended cements.

5. Conclusions

The exploration of the study of predicting the microstructural evolution in FA-blended cement paste through CEMHYD3D modeling has provided a profound understanding of the intricate interactions between FA particles and the cementitious system. The impact of FA content on the

pore structure of cement paste, as elucidated by CEMHYD3D simulations, has proven the crucial role of pozzolanic reactions in shaping the microstructure. The fine and interconnected pore networks observed with increasing FA replacement levels offer promising prospects for enhanced durability, reduced permeability, and resistance to harmful agents. This aligns harmoniously with experimental findings and emphasizes the potential of FA as an advantageous additive in cementitious mixtures.

The implications of the CEMHYD3D modeling results are profound. The tailored cement pastes, which leverage insights into phase evolution and pore structure refinement, hold the potential to contribute to durable and high-performance cementitious systems. However, the significance of this study is grounded in its acknowledgment of the need for experimental validation to ensure the accuracy of modeling predictions. Integrating experimental data and computational simulations paves the way for a holistic understanding of the microstructural evolution of FA-modified cement paste.

The predictive capabilities of CEMHYD3D modeling have proven to be helpful in unraveling the details of microstructural evolution in FA-modified cement paste. The comprehensive analysis and discussions presented in this study contribute to a broad understanding of the influence of FA on the microstructure of cementitious systems.

However, while this computational study provides a crucial understanding of the microstructure development of FA-modified cementitious systems, the combined effects of material composition and various environmental conditions on the durability properties remain areas needing further investigation. Future research should therefore focus on comprehensive microstructure-based durability assessments, including exposure to aggressive environments and service life simulations through advanced predictive models. These studies will ensure the reliable and widespread application of FA as a sustainable cementitious material in cementitious materials, ultimately contributing to an eco-friendly built environment.

Acknowledgments

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