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Review Article

High-Volume Fly Ash-Based Cementitious Composites as Sustainable Materials: An Overview of Recent Advances

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High-volume fly ash (HVFA) cementitious composites (paste, grout, mortar, and concrete) have been widely investigated as a class of sustainable materials due to their lower carbon footprint and often better life cycle performance than conventional Portland cement mixtures. Recent years have seen increased research in HVFA-based materials, and the potential of this type of mixtures in engineering applications has significantly improved. In this context, this work reviews the renewed knowledge of HVFA mixtures, focusing on the relevant papers published over the last decade. The effects of replacing cement with a HVFA binder on the fresh properties, mechanical properties, durability performance, and environmental impact of HVFA cementitious composites are explored. Measures that can compensate for the main drawbacks that limit the wider application of HVFA mixtures are discussed in detail. At last, we summarize the research needs and remaining challenges of HVFA cementitious composites.

1. Introduction

Fly ash, the main by-product of coal combustion in thermal power plants, is one of the most commonly used supplementary cementitious materials (SCMs) in concrete. The global annual generation of coal combustion products is approximately 1.1 billion metric tonnes, more than 85% of which is fly ash, resulting in about 935 metric tonnes of fly ash produced every year [1]. The estimated worldwide production of fly ash will rise over 50% by 2030 [2]; however, only about 35% of fly ash can be recycled or reutilized [3]. The considerable production and low utilization rate of fly ash could result in the material ending up in a landfill polluting soil and water due to the heavy metal elements it leaches [4]. Using fly ash in cementitious composites to partially replace cement is an effective way to solidify the potential hazardous elements in cementitious composites. It is also an economical approach to improving the engineering properties of the cementitious composite.

The history of the use of fly ash in concrete spans more than half a century. Fly ash was used in the late 1940s as a mineral admixture in mass concrete to reduce hydration heat and early-age cracking [5]. Since then, acting as a SCM, fly ash has been used widely in cementitious composites, including paste, grout, mortar, and concrete. Traditionally, the cement replacement ratio by fly ash in cementitious composites is typically limited to about 20% to 25% by the mass of binder. Reasons for this limitation include the variability of fly ash sources, negative effects on air entrainment, and reduced early-age strength of the mixtures [6]. At such a replacement level, fly ash could not only improve the workability and cost economy of cementitious composites but could also improve the resistance to sulfate attack, alkali-aggregate reactions, and thermal cracking [7].

Although fly ash is a valuable mineral admixture for concrete, its low utilization rate raises the interest to replace cement with a high volume of fly ash in cement-based materials [8]. In the 1980s, Malhotra proposed a novel use of fly ash in cement-based materials, i.e., further increasing the cement replacement level to more than 50%, through which high-volume fly ash (HVFA) mixtures can be generated [9]. This type of mixture is fabricated to contain more fly ash than cement by mass, usually with a water-to-cementitious material ratio (w/cm) less than 0.35 and an adjustable amount of high-range water reducing admixture (HRWRA) to obtain the desired workability. Since HVFA mixtures contain substantial quantities of industrial by-product, they feature both low production cost and reduced environmental carbon footprint [10, 11].

In the span of more than three decades, a growing number of studies have been devoted to investigate the fresh properties, mechanical properties, durability performance, and environmental impact of HVFA-based materials, including paste, grout, mortar, and concrete [12]. In addition, HVFA-based materials have been successfully applied in some conventional construction domains such as beams, pavements, and dams. Recently, increased research on the potential of these types of mixtures in emerging engineering applications has significantly improved. For example, HVFA mortar can be modified by admixing nanomaterials or other pozzolanic materials to improve the rheological behavior and structural build-up for successful implementation in 3-D printing [13, 14]. Some techniques in the field of novel concrete materials, such as self-healing concrete and ultra-high performance concrete, have also seen increasing application of HVFA binder in concrete. In this context, there is a need to synthesize the renewed knowledge with the latest information of HVFA mixtures as sustainable construction materials, especially regarding the fresh and hardened properties and environmental impact.

To develop a comprehensive overview of recent advances in HVFA mixtures, a thorough literature search was conducted with a focus on relevant publications over the last decade. The topics for keywords searched in the Google Scholar, Springer, ASCE, ICE, Taylor & Francis, and SAGE databases included fresh properties, mechanical properties, durability performance, life cycle assessment, and measures or treatments used to improve the application potential of HVFA-based materials. Through this approach, a total of about 147 publications were identified and included in this review by using the following criteria: relevance to the review topics, quality and credibility, observation of issues related to the application, and indication of future trends of HVFA mixtures.

This work firstly summarizes the fresh properties of HVFA cementitious composites. Then, we proceed to provide an overview of the mechanical properties, durability performance, and environmental impact of HVFA cementitious composites. Given that low early-age strengths are a main concern of HVFA mixtures, this review also discusses measures that can address this concern. Finally, the knowledge gaps related to the HVFA mixtures (based on the examined references) are identified to promote the proper use of this sustainable material in industry.

2. Fresh Properties

While HVFA cementitious composites exhibit very good potential in engineering applications, some practical issues related to construction processes remain. The main challenges are related to the workability and setting time of HVFA mixtures, as detailed in this section.

2.1. Workability. Generally, the slump value of concrete increases with the replacement level of cement by fly ash at low volume [15], as the spherical-shaped fly ash particles serve to reduce interparticle friction. The vast majority of fly ash particles feature a smooth surface and spherical shape, working as “ball-bearings” in mixtures, which can overcome the internal friction between grains and mitigate the agglomeration of flocs and fragmentation to release the trapped water [16]. The shear force between the fine aggregates can also be reduced by the ball-bearing effect of fly ash [17], as shown in Figure 1. As shown, cementitious mixtures with a low-volume fly ash replacement of cement feature reduced water demand and improved workability.

However, with a further increase of fly ash replacement of cement, a decrease of slump value may occur in HVFA mixtures [11, 15, 18–23]. Figure 2 summarizes the effect of the fly ash replacement ratio on the slump value of HVFA mixtures from relevant references. In general, replacing cement with fly ash in the range of 40% to 60% by mass provides the best workability of FA-based cementitious composites. In this range, the slump value of mixtures is usually increased by about 20–40 mm compared with ordinary Portland cement (OPC) mixtures that contain no fly ash. Beyond the optimum range, the slump will fall due to the higher surface area of fly ash [15]. Normally, cement particles feature a specific surface area of about 303–636 m^2/kg [19, 20, 22, 24], while most fly ash particles feature a higher specific surface area of 320–1521 m^2/kg [19, 22, 24]. An even lower workability of HVFA mixtures can be expected when ultrafine fly ash (with specific surface areas as high as 2510 m^2/kg) is incorporated [25], making less water available for mixing due to the higher water demand of fly ash particles [15].

2.2. Setting Time. It is well known that the setting time of concrete can be retarded by replacing cement with fly ash. HVFA mixtures require a longer initial setting time and final setting time relative to the equivalent mixtures containing no fly ash [26–34]. Moreover, higher retardation times are obtained for the concrete with higher fly ash replacement levels [20]. Figure 3 shows the box plot of the initial and final setting times of OPC and HVFA mixtures based on information from relevant studies. Generally, HVFA mixtures exhibit initial and final setting times that are 3 h to over 10 h longer than those of OPC mixtures without fly ash.

Normally, the concrete setting mainly depends on a variety of parameters including cement fineness, cement-to-water ratio (w/c), and dispersion/flocculation of the cement particles [35]. Figure 4 illustrates the concept of cement dilution and deflocculation in a 60% fly ash system. It is

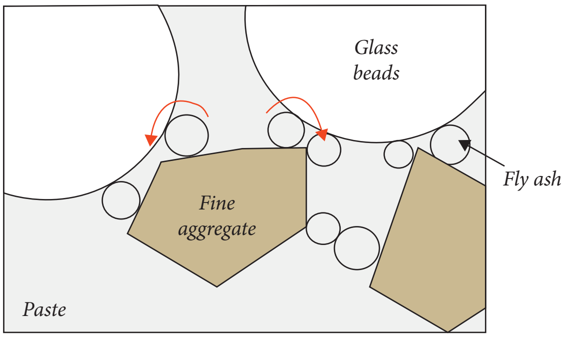


Figure 1: Ball-bearing effect of fly ash on the interaction between fine aggregates, reproduced from [17], with permission from Elsevier, 2021.

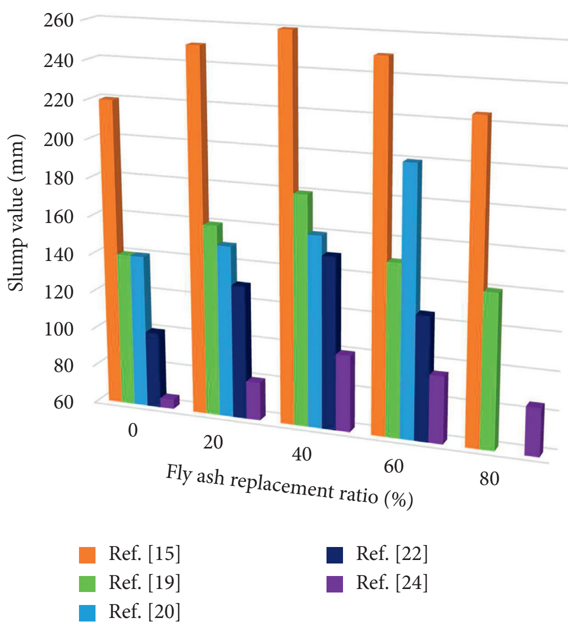


Figure 2: Slump value of FA-based cementitious composites as a function of the fly ash replacement ratio, based on data collected from relevant studies.

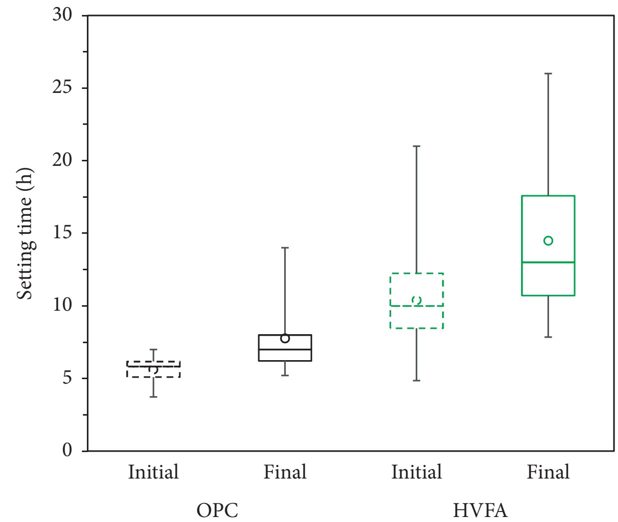


Figure 3: Comparison of setting times between OPC and HVFA mixtures, based on data collected from relevant studies.

obvious that the presence of fly ash particles in HVFA mixture reduces the number of contacts between cement particles. A decrease in the flocculation level of cement particles can be achieved as the fly ash particles break up some of the flocculated cement 3-D network structure [31]. If the fly ash particles are removed (Figure 4(c)), the cement particles' spacing is significantly increased compared with the plain OPC system, resulting in the dilution of cement in a HVFA mixture. As a result, the reduced concentration of cement and the slow pozzolanic reaction in HVFA mixtures account for the observed retardation in the setting times [31].

HVFA mixtures containing Class F fly ash exhibit longer setting times than their counterparts containing Class C fly ash [33]. According to the newly released ASTM C618, Class F fly ash features low CaO content (18% maximum), while Class C fly ash features a higher CaO content (>18%). Class F fly ash generally exhibits pozzolanic properties, whereas Class C fly ash exhibits pozzolanic and cementitious properties simultaneously [31]. The pozzolanic reaction of Class F fly ash only occurs in the presence of calcium hydroxide [33]. As a result, Class F fly ash remains relatively inert during the first 24 h. In comparison, Class C fly ash can be quite reactive at early ages, resulting in a shorter retardation time than Class F fly ash [26].

3. Mechanical Properties

3.1. Compressive Strength. Compressive strength is a fundamental property of concrete, making it the most common mechanical property investigated in almost every study of HVFA mixtures. Given the same w/cm and curing regime, a reduction in the compressive strength at various curing ages (mainly ranging from 1 d to 365 d) is generally reported when replacing cement with HVFA [36–44]. The level of strength reduction increases with increasing fly ash content and decreases with increasing curing age.

Figure 5 depicts a typical example that contrasts the compressive strength development between HVFA and OPC concretes. For HVFA concrete, both the early-age and later-age compressive strength development trends can be explained by the delay in calcium-silicate-hydrate (C-S-H) formation because the pozzolanic reaction becomes the major reaction as the curing time increases [45]. At early ages (up to 14 d), the strength of HVFA concrete is mainly contributed by the hydration of cement [38]. As fewer cement particles are available for hydration due to the dilution effect, HVFA concrete has fewer hydration products and thus a lower compressive strength than its OPC counterpart. At later ages (beyond 28 d), the compressive strength gain of HVFA concrete mainly results from the pozzolanic reaction of fly ash [33, 42], because such reaction becomes fast enough to contribute to the strength development of HVFA mixtures [46, 47]. As a result, as the curing age increases, the difference in compressive strength between HVFA and OPC mixtures diminishes [20, 48]. At the curing time of 90 days, the compressive strength of HVFA mixtures may reach

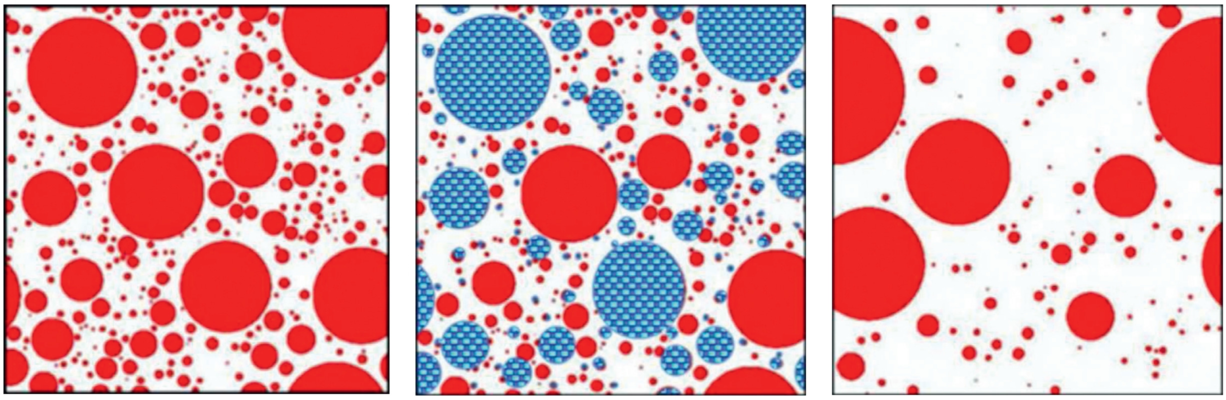


FIGURE 4: (a) Cement particles in OPC; (b) deflocculation; and (c) dilution of cement particles in HVFA system (red part, cement particles; blue patterned part, fly ash particles), reproduced from [31], with permission from Elsevier, 2021.

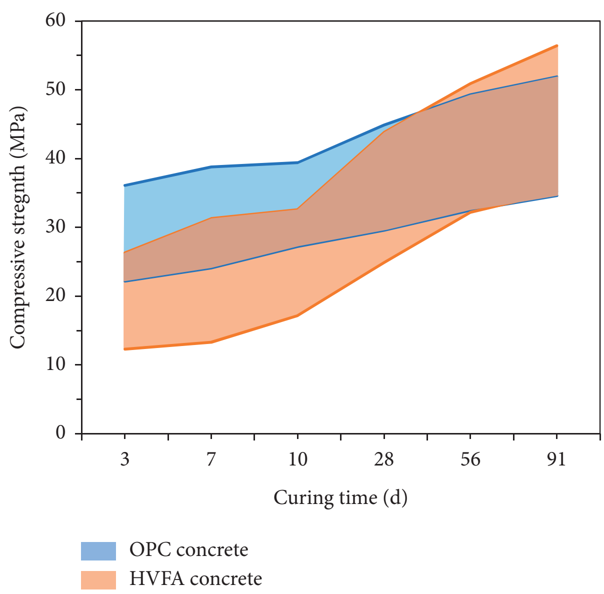


FIGURE 5: Temporal evolution of compressive strength of OPC and HVFA concretes, based on the data collected from reference [19].

comparable or even higher values than that of their OPC counterparts, as illustrated in Figure 5.

The enhanced compressive strength of HVFA mixtures observed at later ages is influenced by several factors, including the amorphous SiO_2 and Al_2O_3 contents, alkalinity of the amorphous phase, and the specific surface area of fly ash [33]. The physical, chemical, and mineralogical properties of fly ash tend to fluctuate from one source to another and sometimes within the same source. These changes in fly ash properties could have a profound impact on the mechanical properties of HVFA mixtures [49].

To date, there have been some limited studies aimed at correlating the properties or performance of HVFA mixtures with the characteristics of fly ash. For instance, isothermal calorimetry and rheological measurements have been used as screening tools to assess the early-age behavior of HVFA mixtures and as a means of quality control to evaluate and qualify fly ashes [49]. With the aid of soft computing techniques, the artificial neural network model [50] or support vector machine model [51] can be developed to predict the compressive strength of HVFA mixtures using relevant parameters such as the content and type of fly ash as model inputs. Mohammed et al. [52] demonstrated the use of neuro-swarm and neuro-imperialism models for predicting the compressive strength of fly ash concretes, using 9 mix design parameters as model inputs and 379 data points from published literature. They also demonstrated the use of soft computing techniques [53] to accurately predict the compressive strength of HVFA concrete based on curing time and several key mix design parameters, using experimental data of 450 tested HVFA concrete mixes.

A recent study proposed that the efficiency factor (k value) of fly ash is a function of its chemical composition and density based on the database of 440 HVFA mixtures obtained from the last 31 years. This factor was capable of reasonable prediction of the compressive strength of HVFA concretes made with 40–75% Class F fly ash and normal-weight aggregate [12]. The proposed k mainly considers the ratio of CaO to the sum of SiO_2 and Al_2O_3, abbreviated as $C/(S+A)$ ratio, and the fly ash fineness indirectly characterized by the fly ash density, as expressed in the following equation:

$$k = a \cdot \gamma_{FA} \cdot \frac{C}{S + A}, \qquad (1)$$

where a is the coefficient of correction and γ_{FA} is the density of fly ash. The incorporation of density as a measure of fineness in the calculation of k value of fly ash led to a decrease in the results scattering of compressive strength of HVFA concretes [12, 54]. The predicted improvement was based on the fact that the density of fly ash can be determined more reliably than the Blaine specific surface area [12].

3.2. Flexural Strength. Flexural strength, a.k.a. modulus of rupture, is another important mechanical property to consider in studies of HVFA mixtures, especially for the fabrication of beams and concrete slabs. Similar to the compressive strength, the flexural strength of HVFA mixtures is lower than that of OPC mixtures with the same w/cm at ages up to 1 year [20, 32, 48, 55–57]. Moreover, the flexural strength of HVFA mixtures increases with age and decreases with fly ash content. With sufficiently low w/cm, HVFA mixtures can achieve a 28 d flexural strength greater than the lower limit for pavement design, which is considered to be in the range of 4.0 to 4.5 MPa [57]. Furthermore, in later ages, HVFA mixtures may develop superior flexural strength to OPC mixtures, attributing to the pozzolanic reaction between fly ash and calcium hydroxide (CH) and resulting in the formation of additional hydration products. This process can decrease the porosity of the composite matrix and the transition zone [32].

Similar to OPC mixtures, there is a close relationship between flexural strength and compressive strength of HVFA mixtures, and various studies reported either a power fit [32] or a linear fit [20, 48]. This discrepancy may be ascribed to the fact that the w/cm and fly ash contents used differ from study to study. Figure 6 presents the overall trend between the compressive strength and flexural strength of HVFA mixtures from relevant references, which follows a power relationship. In other words, the flexural strength of HVFA mixtures increases as the compressive strength increases, but the slope of the increment gradually declines with the strength increase. The likely explanation is as follows. The compressive strength of HVFA mixtures is defined by both the pozzolanic reaction and the pore-filling effect of fly ash; however, the flexural strength of HVFA mixtures is primarily defined by the pozzolanic reaction [58].

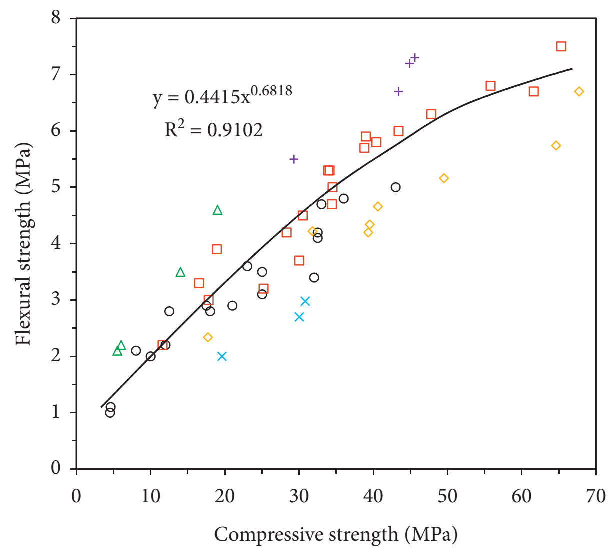


Figure 6: Relationship between compressive strength and flexural strength of HVFA mixtures, based on the data collected from relevant studies.

3.3. Splitting Tensile Strength. In practice, splitting tensile strength is generally used to characterize the tensile strength of concrete since it is easily conducted as a conventional and conservative test [59]. For HVFA mixtures, many studies have reported a reduction in the splitting tensile strength with the inclusion of 50–70% fly ash as cement replacement [20, 48, 57, 59, 60]. Usually, the splitting tensile strength of HVFA concrete is slightly lower than the flexural strength as they correspond to 7–10% and 10–11% of the compressive strength, respectively [48].

The lower splitting tensile strength of HVFA mixtures likely results from the weaker bond between the cementitious matrix and the aggregates. The addition of fibers can increase the splitting tensile strength of HVFA concrete that contains 50% fly ash [57]. Particularly, longer fibers that have hooked ends are more effective in increasing the tensile strength than short and straight fibers [57].

The vibration method utilized during the production of HVFA mixtures is also reported to affect their splitting tensile strength. For instance, self-compacting HVFA concretes usually feature a higher splitting tensile strength than normally vibrated concretes or roller compacted concretes, even if with a lower compressive strength [36]. This might be attributed to the higher deformability and better homogeneity of self-compacting concrete, which result in an improved bond in the interfacial transition zone (ITZ) and thus an increased splitting tensile strength.

3.4. Modulus of Elasticity. Modulus of elasticity is used to characterize the stiffness of hardened concrete. Each component of concrete and its intrinsic modulus have a determinative effect on the elastic modulus of the concrete as a composite material. As such, HVFA mixtures that partially replace Portland cement with fly ash exhibit a different elastic modulus than their OPC counterparts [23]. In general, the modulus of elasticity of HVFA mixtures is lower than that of OPC mixtures with equivalent w/cm [20, 23, 32, 59, 61]. A further reduced modulus of elasticity can be expected with the increase of fly ash content in HVFA mixtures; however, at mature ages, HVFA mixtures can develop a comparable modulus of elasticity to their OPC counterparts [59]. It should be noted that the tensile modulus of HVFA mixtures usually exceeds the compressive modulus, and it is more rational to evaluate the tensile stress by using tensile modulus [59].

4. Durability Performance

In cement-based materials, the incorporation of HVFA binder reduces the cost of production and makes the composites more sustainable in terms of lower footprint and, in many cases, more durable. This section focuses on the durability performance of HVFA mixtures, including abrasion resistance, carbonation resistance, freezing-thawing resistance, deicing salt scaling resistance, shrinkage, transport properties, resistance to alkali-aggregate reactions, and resistance to chemical attack.

4.1. Abrasion Resistance. Abrasion resistance is an important property of concrete to consider, especially for applications such as pavement. Generally, the abrasion resistance of HVFA mixtures is lower than their OPC counterparts [18, 46, 62, 63]. During the abrasion test, the depth of wear or mass loss increases by increasing the fly ash content and decreases by increasing the curing age of HVFA mixtures. The change in the abrasion resistance is mainly attributed to the change in compressive strength of HVFA mixtures [46, 62, 63]. Generally, the abrasion resistance of HVFA mixtures increases when increasing the compressive strength [18], and this is similar to OPC mixtures.

In addition to compressive strength, surface finishing and curing conditions have been reported to strongly influence the surface abrasion resistance of HVFA mixtures [62]. For instance, improper surface finishing could result in an unusually high content of fine-grained material (including pastes of cement and SCMs) in the surface layer of HVFA concretes, causing lower abrasion resistance than that prepared with proper finishing [64]. Surface finishing also leads to distinct pore structures and hardness in the surface zone, contributing to the change in the abrasion resistance of HVFA mixtures [65]. If not cured properly, the surface quality of HVFA concretes is negatively affected, thus reducing their abrasion resistance [46]. Our recent study found that the incorporation of trace amounts of a novel nanomaterial, graphene oxide, improved the microhardness, scratch surface roughness, scratch hardness, and friction coefficient of 60% fly ash-contained HVFA concretes resulting in their increased abrasion resistance [66].

4.2. Carbonation Resistance. As mentioned earlier, HVFA concrete with 50% fly ash in the binder generally exhibits a lower level of strength, which translates to higher permeability, thus facilitating the carbonation of concrete by allowing a higher rate of CO_2 ingress into the concrete. One study reported that the negative correlation between compressive strength and carbonation depth of HVFA mixtures was strong and linear [67]. Many studies have concluded that HVFA mixtures are more susceptible to carbonation than their OPC counterparts of equivalent w/cm, with the depth of carbonation increasing with the fly ash content [19, 67–70].

In addition to the lower strength, the reduction of CH content (due to the pozzolanic reaction of fly ash) also explains the reduced carbonation resistance of HVFA concrete [67]. Moreover, the existence of excessive ettringite, which results from the noncompletion of pozzolanic reactions, further deteriorates the carbonation resistance of HVFA concrete [69]. Figure 7 shows the split sections after the phenolphthalein spray of a HVFA concrete vs. OPC concrete after 7 days in the accelerated carbonation test, respectively [71]. Phenolphthalein is naturally colorless but turns pink in the presence of high alkalinity. In the case of concrete, the carbonated area features lower alkalinity, thus failing to turn pink. Figure 7 illustrates that the 50% fly ash HVFA concrete experienced severe carbonation, while its OPC counterpart exhibited a clear carbonation front and much less carbonation.

4.3. Freezing-Thawing Resistance. The freezing-thawing (F-T) resistance of HVFA mixtures is generally lower than their OPC counterparts of equivalent w/cm [72–75]. Moreover, the F-T damage in HVFA mixtures tends to increase with the fly ash content. Interestingly, the air-void characteristic (e.g., spacing factor) is a more significant factor affecting the F-T performance of HVFA mixtures than the fly ash content or w/cm [75]. Therefore, the most commonly used strategy for improving the F-T durability of HVFA mixtures is air entrainment. The F-T resistance of the air-entrained HVFA mixtures can be excellent, with the durability factor being higher than 95% at the end of the rapid F-T cycling test [73, 75, 76]. However, the free carbon in fly ash particles could absorb the air-entraining admixture, compromising the F-T durability of HVFA mixtures [75]. Besides air entrainment, improving the quality of the ITZ is another approach to enhance the F-T resistance of HVFA mixtures [72]. Improved ITZ can slow down the liquid transport in the HVFA mixture, resulting in less water available in the cementitious composite and thus less F-T damage caused by ice crystallization pressure [72], hydraulic pressure, or osmotic pressure.

Adequate air-void parameters can provide HVFA mixtures with sufficient resistance against repeated F-T cycles but may fail to prevent salt scaling. In general, HVFA mixtures perform poorly in the F-T cycling test, even though they feature improved F-T resistance when air entrained [75, 77]. The performance of nonair-entrained HVFA concretes in a salt scaling test would be even more worse [75]. One reasonable explanation for the failure of HVFA mixtures in a salt scaling test may be the reduction in their surface strength due to the carbonation [75]. In addition, the ITZ is easily deteriorated and the integrity of ITZ's microstructure is compromised when HVFA concretes are subjected to salt scaling damage [76], as shown in Figure 8. At the beginning of the test, the gaps and air voids in the ITZ around the aggregate are full of salt solution, leading to the separation of ITZ by ice expansion force, shrinkage of cementitious paste [78], etc. Therefore, improving the ITZ could result in an increased salt scaling resistance of HVFA mixtures, by slowing down the ingress of water and reducing the concentration of ions in the pore solution [78].

4.4. Shrinkage. In HVFA mixtures, cracking occurs when the tensile stress induced by shrinkage strain exceeds the local tensile strength of the composite material. Generally, the amount of cement paste accounts for the shrinkage in concrete. HVFA mixtures usually exhibit lower shrinkage than their OPC counterparts when a large amount of cement in the mixture is replaced with fly ash [32, 79–82]. Furthermore, the unhydrated fly ash particles in HVFA mixtures may act as microaggregates and thus restrain shrinkage [81]. This microaggregate characteristic of fly ash is attributed to the good dispersibility of its particles in the HVFA mixture as well as the high strength (over 700 MPa) of the glass microbeads in the fly ash [83]. Therefore, it is reasonable to state that the use of Class F (instead of Class C) fly ash in HVFA mixtures could further reduce the shrinkage strain [80], considering the higher amount of Class F fly ash that remains unhydrated.

4.5. Alkali-Aggregate Reactions. The resistance of HVFA mixtures to alkali-aggregate reactions (AARs) benefits from the high-level replacement of cement by fly ash. This is attributed to the fact that the replacement of cement by fly

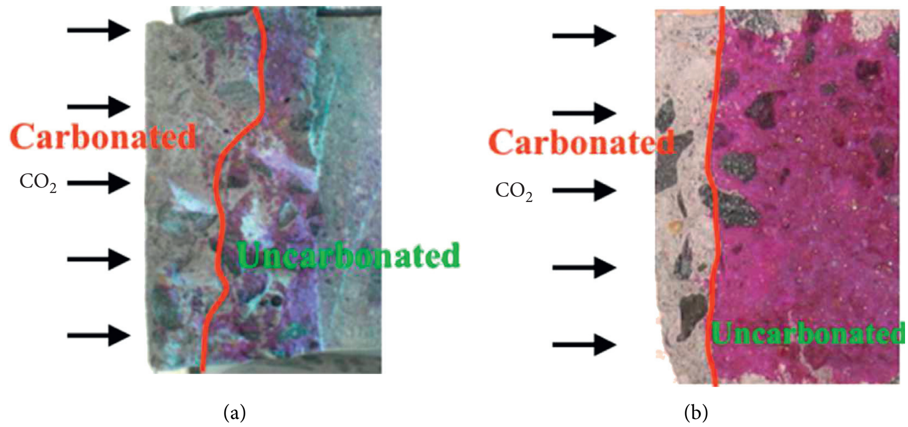


Figure 7: Split section after the phenolphthalein spray of (a) 50% fly ash HVFA concrete and (b) OPC concrete, reproduced from [71], with permission from Elsevier, 2021.

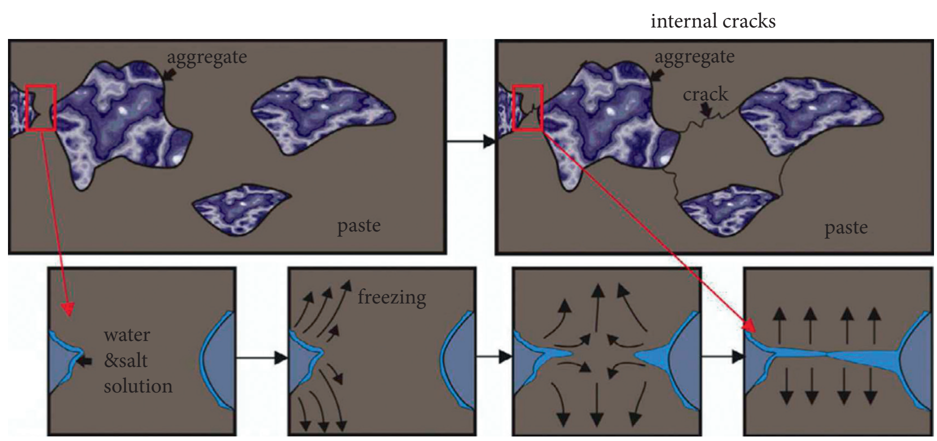


Figure 8: Propagation of internal cracks of HVFA concretes during F-T cycles in the presence of salt, reproduced from [78], with permission from Elsevier, 2021.

ash could reduce the amount of dissolvable alkali released from the binder [84]. For instance, one study reported that concretes containing HVFA binder exhibited no adverse expansion when highly reactive aggregates were used [85]. Another study reported that even with added alkali activator, the use of HVFA binder in concrete still effectively inhibited the expansion due to AARs [84]. The inhibited expansion could also be ascribed to the densification of the concrete microstructure, thereby lowering the release rate of alkalis from the cementitious binder.

Ca- and S-rich phases in fly ash particles are released more rapidly than the alkalis [84, 86], as illustrated in Figure 9. Specifically, Figure 9 shows that calcium and sulfur exhibit higher elemental concentration in the hydration products than in the fly ash particles; in contrast, potassium and sodium exhibit higher values in the fly ash particles. This indicates a slower depletion of alkali (Na and K) from the fly ash particles [86], contributing to the increased AAR resistance of HVFA mixtures. Furthermore, some larger fly ash particles in the 28 d HVFA mortar did not fully participate in the cementitious phase because their pozzolanic reaction only occurred on the surface layer of a few microns. In other words, much of the “core” of these fly ash particles remained intact and actually served as a fine aggregate phase. This highlights the need to activate the fly ash and some activation treatments are discussed in a later section.

In addition, the alkali’s binding effect of calcium-alu minate-silicate-hydrates (C-A-S-H) that formed in HVFA mixtures can reduce the alkalinity of the pore solution [87]. Another possible mechanism is that the aluminate sourced from fly ash could slow down the dissolution kinetics and reduce the extent of dissolved silica from the aggregate, contributing to the low AAR expansion [88]. A recent study confirmed that the presence of unbound aluminum discharged from fly ash is effective in limiting the dissolution of amorphous reactive aggregates, which is beneficial for the AAR mitigation of concrete [89].

4.6. Transport Properties. The use of HVFA binder in cement-based materials can cause pore and grain refinement, which leads to the improved transport properties of HVFA mixtures, including reductions in water permeability, water sorptivity, gas permeability, and chloride ion penetration [61, 74, 77, 80, 90–96]. The presence of HVFA binder can also increase the capillary network tortuosity, decrease pore interconnectivity, and generate additional C-S-H by the pozzolanic reaction. HVFA binder also reduces the susceptibility to the

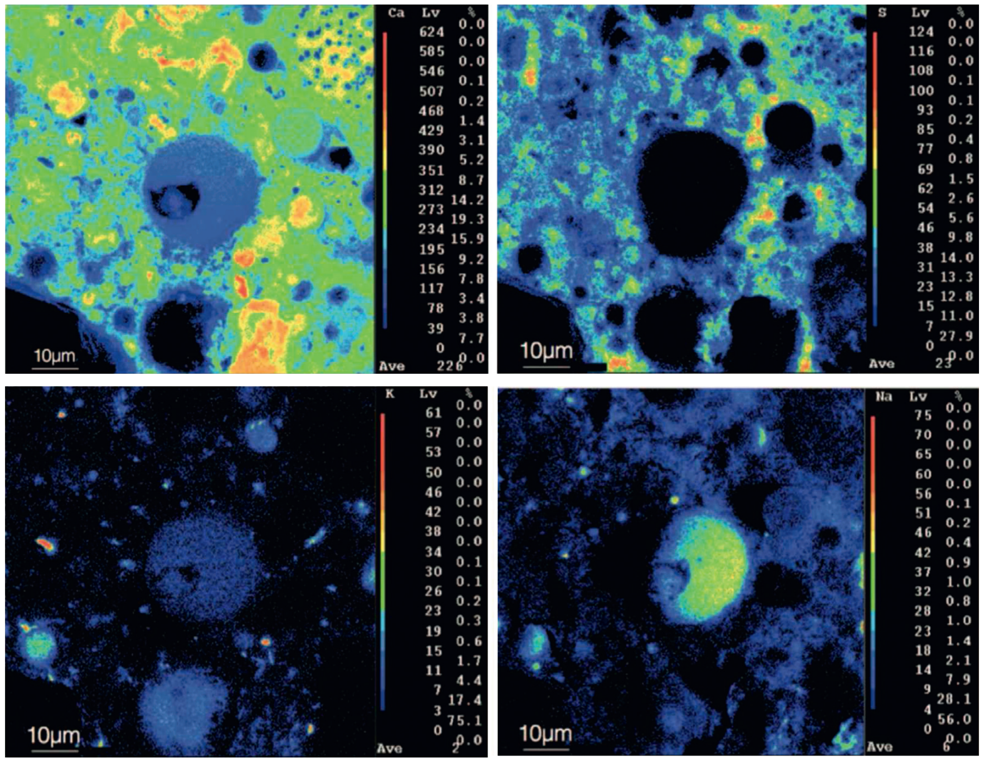


Figure 9: Element mapping of Ca, S, K, and Na in the same area in HVFA mortar at the curing age of 37 d, reproduced from [86], with permission from American Society of Civil Engineers, 2021.

penetration of gas and migration of free water or chloride ions [92, 94]. Moreover, HVFA mixtures usually feature a reduced number of capillary pores, thus lower capillary absorption and lower water sorptivity [97]. Some studies suggested the reduction of microcracking in the ITZ in HVFA mixtures as another reason for the improved transport properties [80, 92].

For chloride ion penetration, the alumina in fly ash can react with chlorides to form stable chloro complexes [72]. Therefore, HVFA mixtures usually feature a significantly reduced chloride ion penetration and their chloride binding capacity tends to increase with the fly ash content [70, 72, 77, 95]. Figure 10 shows an example of the colorimetric test of a HVFA concrete, where the area with color change to white corresponds to the chloride penetration depth [92]. The chloride penetration depth in HVFA mixtures could be reduced to one-third of the original value in OPC mixtures with the addition of HVFA binder [72, 92]. Meanwhile, the electrical resistivity of HVFA mixtures is increased due to the incorporation of HVFA binder [86, 98]. The electrical resistivity of cement-based materials is an indirect indicator that provides a rapid evaluation of their chloride permeability [99]. The higher electrical resistivity could translate to lower chloride permeability of HVFA mixtures, even though there are ions other than Cl^- in the pore solution that also affect the electrical resistivity of the concrete.

4.7. Chemical Attack. The deterioration of cementitious composites by chemicals is mainly due to the paste erosion and expansion caused by the reactions of chemical ions with Portlandite and other hydration products. Relative to their OPC counterparts, the resistances of HVFA mixtures to

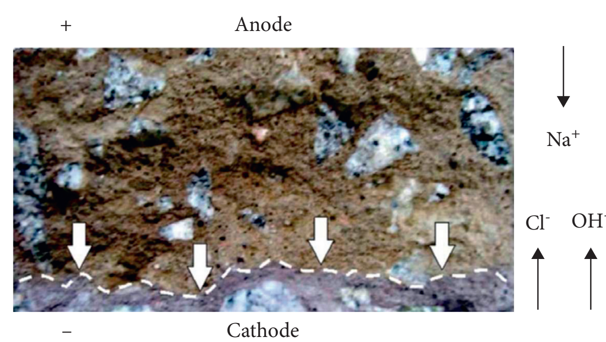


Figure 10: Colorimetric test of concrete with 50% fly ash (wide arrows indicate the precipitated chloride reaction front), reproduced from [92], with permission from American Society of Civil Engineers, 2021.

sulfate, lactic acid, acetic acid, and hydrochloric acid are improved [90, 100]. This can be explained by the reduction in CH content in the hydrated HVFA binder and the dense and discontinuous pore structure of HVFA mixtures (due to the pozzolanic reaction of fly ash). In a 75-day test where concretes were exposed to 2% hydrochloric acid, the HVFA concretes only exhibited minor surface erosion, whereas the OPC concretes already experienced edge loss [90].

HVFA concretes also feature less susceptibility to external or internal forms of sulfate attack, relative to their OPC counterparts. This is due to the pozzolanic behavior of fly ash that leads to increased consumption of CH [100]. Figure 11 depicts a comparison (by visual observation) between OPC and HVFA concretes after exposure to 10 wt% sodium sulfate

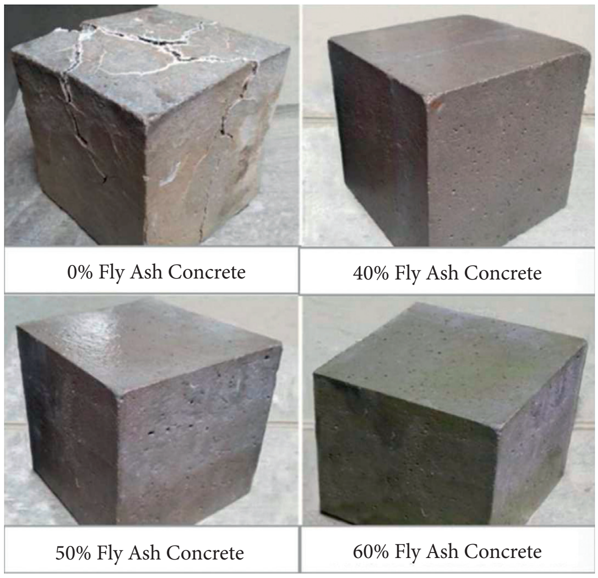


Figure 11: Concrete cubes after exposure to 10 wt.% sodium sulfate solution, reproduced from [90], a paper published in an open access journal.

solution for 550 days in an alternating drying and wetting scheme [90]. It is obvious that the OPC concrete is seriously deformed, whereas the HVFA concrete stays intact (no significant change to the physical appearance).

5. Environmental Assessment

As mentioned above, HVFA mixtures generally feature superior long-term durability performance compared with conventional OPC mixtures, except for the resistances to carbonation and deicing salt scaling. Therefore, from a life cycle perspective, it is likely that HVFA mixtures could offer greater cost savings, as well as reduced CO_2 emissions generated in the production of a unit volume of the mixture [101]. Normally, a life cycle assessment (LCA) methodology is recommended to evaluate the environmental impact of cement-based materials. In this methodology, a full description of the product from the extraction of raw materials to the waste treatment, i.e., cradle-to-grave approach, is required to quantify its impact on the environment [102]. However, for HVFA-based materials, a cradle-to-gate approach is usually used to exclude the influence of the end-of-life scenarios, thus emphasizing on the production process. Generally, the cradle-to-gate approach includes extraction of cement raw materials, manufacturing of cement, extraction and processing of aggregates, manufacturing of superplasticizers, preparation and collection of fly ash, transportation of raw materials, and concrete batching; the analysis ends at the concrete plant with the final product ready to be used in the construction field [103, 104], as illustrated in Figure 12. In evaluating the environmental impact of HVFA mixtures, all emissions during the cradle-to-gate approach are converted into CO_2 equivalent (CO_2-eq) to calculate the global warming potential (GWP) [105, 106]. These emissions may include greenhouse gas (mainly CO_2) and air pollutants associated with the use of electricity, fuel, transportation, and production, such as CO, NO_x, PM_{10}, and SO_2 [103].

A function unit of 1 m^3 HVFA concrete with a service life of 50 years was adopted to conduct the LCA analysis, and the results revealed that the CO_2-eq footprint of HVFA concrete was about 25%–40% less than the reference OPC concrete exposed to different deterioration processes [102, 105]. Using the concept of the CO_2-eq intensity, which represents the CO_2-eq per unit volume of concrete per compressive strength, one study concluded that a lower CO_2-eq intensity was achieved by Portland cement being replaced with HVFA binder in concrete [103]. Although the HVFA mixtures possessed high compressive strength, in that case, the reduced GWP associated with the HVFA mixtures was more notable [39, 107]. This is mainly because that the environmental impact of manufacturing fly ash is considered as zero since it is treated as waste [108], while about 70% of the total greenhouse gas emission in concrete production is related to the production of clinker in cement [104, 109]. Figure 13 depicts the information related to the total consumption of fuel and electricity for fly ash. It is obvious that the majority greenhouse gas emission related to fly ash occurs in the transportation stage [108].

When using different allocations (to none, by mass or by economic value) associated with fly ash, economic allocation is preferable since the mass allocation results in an environmental impact that is an order of magnitude higher [110]. A similar conclusion was drawn from another study [111], in which the environmental impact of concrete was strongly dependent on the allocation scenario applied. Yet, the environmental impact of HVFA mixtures decreased with increasing fly ash content in all allocation scenarios [111]. The incorporation of a HVFA binder reduces the environmental impact of concrete by about 50% [112, 113], regardless of the locations of the coal power plant (fly ash supplier) and the concrete plant [109, 114–116]. However, if the impacts from allocation or transportation are large enough to reduce the benefits associated with the use of the HVFA binder in mitigating GWP, increasing fly ash replacement may become less favorable in concrete production [117].

6. Measures to Improve the Strengths of HVFA Mixtures

As discussed earlier, HVFA cementitious composites have relatively low early-age strengths, which is a main hurdle that limits the implementation of HVFA mixtures [30]. Some of the relatively poor durability performance, which correlate closely with mechanical properties (especially compressive strength), are also concerns for HVFA mixtures in various service environments. Fabricating HVFA mixtures with lower w/cm can compensate for the delayed strength development. Yet, the pozzolanic reaction of fly ash only occurs in the presence of sufficient moisture [118]. In addition, the content of CH produced from the hydration of cement is usually less than that required for the pozzolanic reaction of the whole mass of fly ash present in the concrete [119]. Therefore, a critical combination of the fly ash replacement level and w/cm would exist in the HVFA mixtures. In this case, the amount of

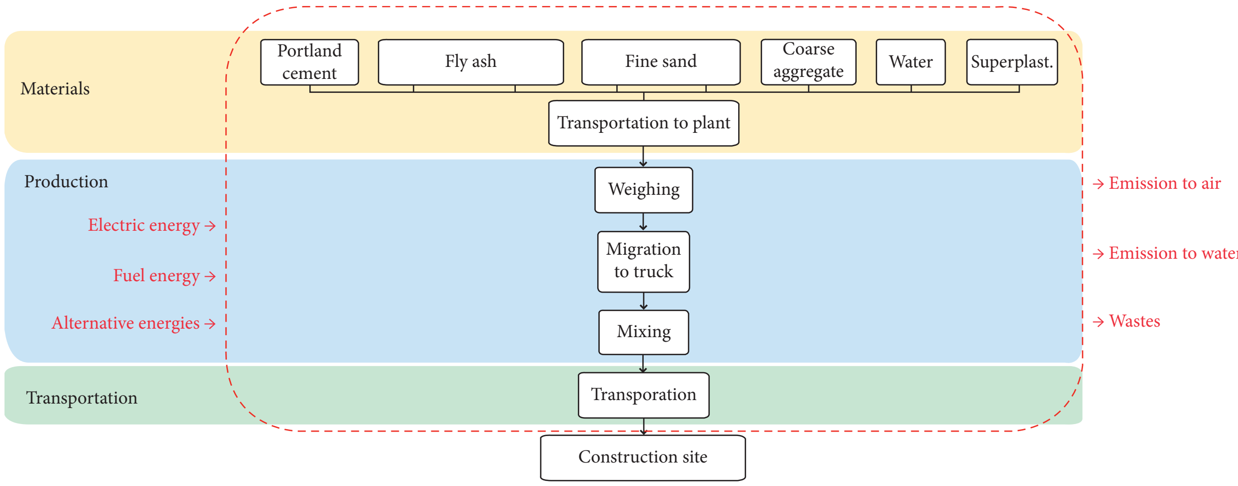


Figure 12: Schematic diagram of the cradle-to-gate approach LCA of HVFA concrete, reproduced from [39], with permission from Elsevier, 2021.

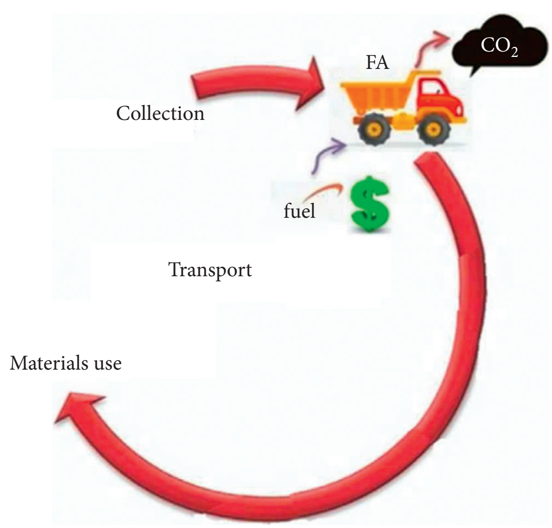


Figure 13: Various stages of the life cycle of fly ash, reproduced from [108], with permission from Elsevier, 2021.

CH or moisture is exactly enough for participating in the pozzolanic reaction of fly ash [23]. As the physicochemical characteristics of fly ash used vary from study to study, the optimum substitution rates of fly ash and w/cm lie in the range of 50%–60% and 0.3–0.4, respectively [23, 120]. Besides, there exists the risk of significant shrinkage if low w/cm are employed [30].

Other than lowering the w/cm, the limitations of HVFA mixtures in practice can be addressed using different curing conditions, activation treatments, finer SCMs, nanomaterials addition, and fiber inclusion, as detailed in this section.

6.1. Special Curing Conditions. Special curing conditions, including elevated-temperature curing and internal curing, have been employed to induce an increase in the strength development rate of HVFA mixtures [30, 80, 98, 121]. Warm-air curing can increase the compressive strength at the early age of HVFA concretes by expediting the hydration process of cement and the pozzolanic reaction of fly ash [80]. In contrast, HVFA concretes cured in warm water feature both increased early-age and long-term compressive strengths, compared with those cured at room temperature [98]. Internal curing is a method employed to provide additional moisture from the presaturated porous aggregate to support the pozzolanic reaction of fly ash [30], as illustrated in Figure 14. The water used for curing can be delivered internally to the concrete as "packets or inclusions" rather than just being placed on the surface of the mixtures. The internal curing reportedly enhanced the compressive strength by 61%, 32%, and 49% in a 60% fly ash HVFA mortar at 1 d, 28 d, and 365 d, respectively [30]. These strength improvements mainly resulted from higher degrees of hydration of cement and the pozzolanic reaction of fly ash.

6.2. Activation Treatments. In addition to specific curing conditions, some activation treatments have been employed in HVFA mixtures to improve their mechanical properties. Generally, the activation treatments include mechanical activation and chemical activation. Mechanical activation, such as the grinding of fly ash, endows HVFA concretes with higher compressive strength than those without activation applied [122], by improving the reactivity of the fly ash. As mentioned earlier, the specific surface area (particle size) of fly ash is a significant factor that affects the compressive strength of HVFA mixtures [33]. It should be noted that there exists an optimum grinding duration for the fly ash (about 2 hours), beyond which the size of the fly ash particles cannot be further reduced and the strength of HVFA mixtures cannot be further improved. This is because of the agglomeration of the particles or the overgrinding of the softer material with the harder material, resulting in noneffectiveness of further grinding [122]. A similar conclusion was drawn in other studies [123, 124], in which the median particle size of fly ash could be reduced from 19.70 μm to 2.67 μm by 2 hours of wet grinding. Through this process, both the pozzolanic activity and dissolution of fly ash are significantly promoted, contributing to the formation of hydration products and thus the strength development [123].

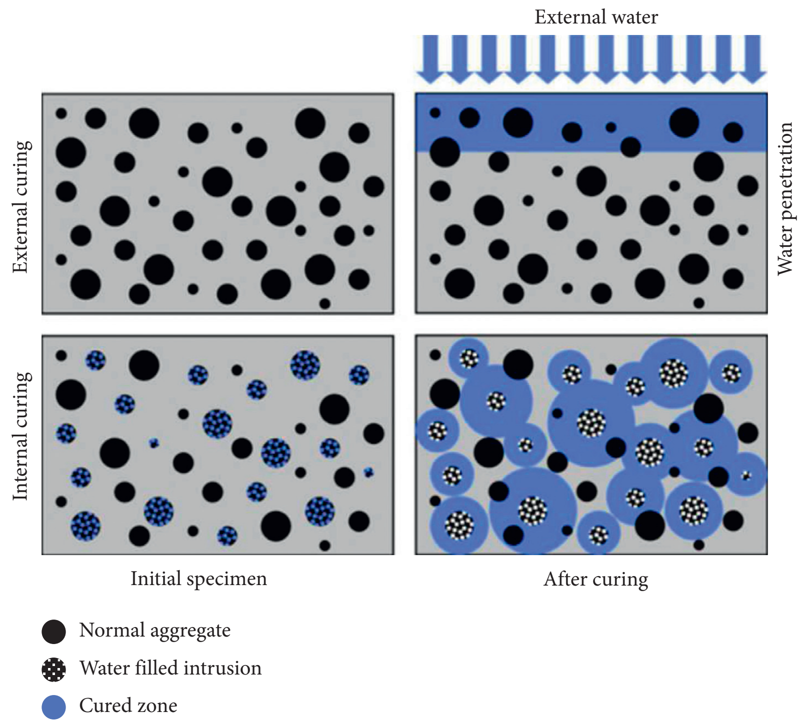


FIGURE 14: Conceptual illustration of the differences between external and internal curing, reproduced from [30], with permission from Elsevier, 2021.

Sodium sulfate, a neutral pH chemical activator, is found effective in improving the compressive strength of HVFA mortars at both the early and later ages [82, 100]. The improved compressive strength correlates with the process of increased ettringite formation and CH consumption, suggesting an accelerated pozzolanic reaction and improved hydration degree of fly ash. Moreover, with the addition of 3%–5% Na_2SO_4 by mass, the HVFA mixtures showcased the stability of free sulfate ions present within the samples during the expansion test, which demonstrates the applicability of this activator in practice [100]. With both 3 wt% Na_2SO_4 (SS) and 2 wt% nanosilica (NS) added, the HVFA mortars exhibited a sharper increase in their compressive strength, relative to the increases seen in the single system. Figure 15 depicts the presence of a synergistic effect between SS and NS in promoting the strength development of HVFA mortars [125], with the strength enhancement notably stronger than that of the sum of these single systems at the same dosage.

With high alkali and sulfate contents, cement kiln dust (CKD) has been shown to enhance the early-age strength of HVFA mixtures through the activation of fly ash [122, 126]. Figure 16 presents the contour plot illustrating the effect of the three binder constituents on the 2 d compressive strength of a HVFA paste. With the contents of fly ash, cement, and CKD set as 60%, 30%, and 10% by mass, respectively, the 2 d compressive strength can be maximized to 184% higher than that of the control paste without CKD [126].

Different from the chemical activation of fly ash used during the fresh mixture stage, electro-mutagenesis is an interesting technique that activates HVFA cementitious

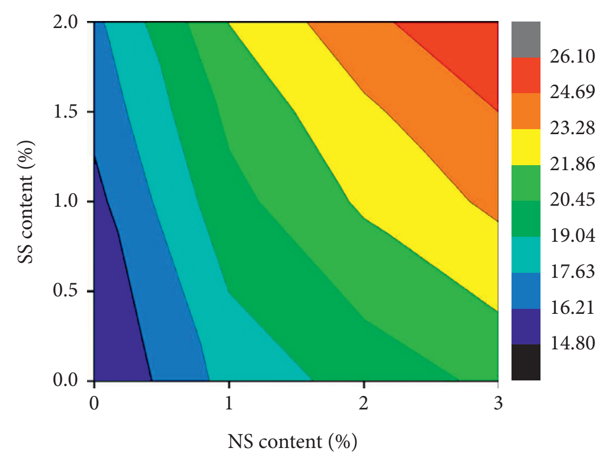


FIGURE 15: Effect of NS and SS on the compressive strength of HVFA mortars, reproduced from [125], with permission from Elsevier, 2021.

composites during the hardened stage. Conceptually, this technique uses sodium hydroxide and sodium silicate as the source of alkaline ions and applies an electric field across the composite matrix [127]. As illustrated in Figure 17, this treatment forces external alkaline ions into the capillary pores in HVFA concretes, where they react with unhydrated fly ash particles and contribute to the formation of hydration products and thus a denser microstructure. Depending on the detailed process parameters used, this electrochemical treatment may induce risks such as AARs, undesirable alternation of the microstructure, and hydrogen embrittlement of the embedded metal. These risks are similar to some cases of electrochemical chloride extraction and

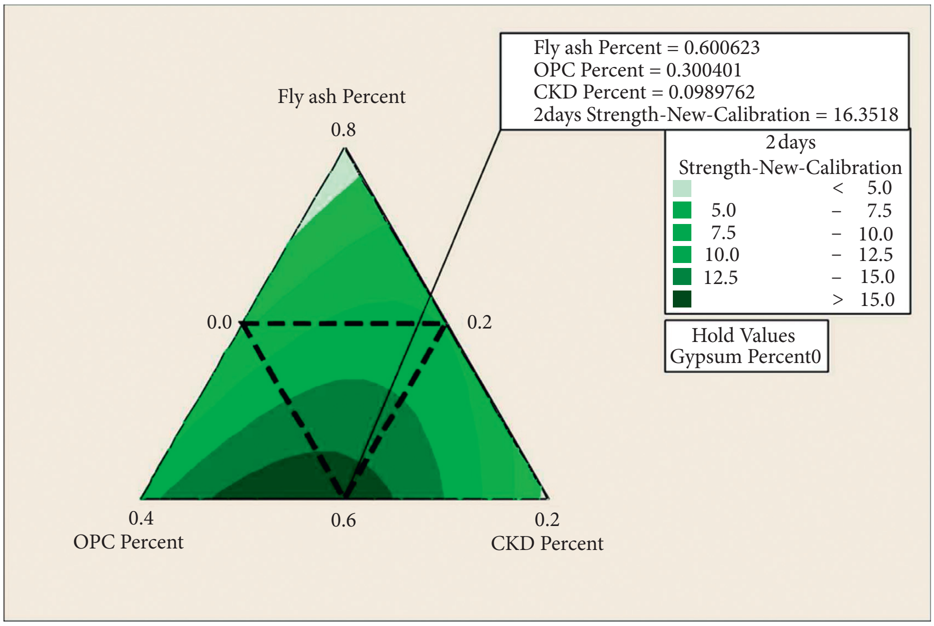


Figure 16: Proportion optimization of a HVFA paste based on 2 d compressive strength, reproduced from [126], with permission from Elsevier, 2021.

electrochemical injection of corrosion inhibitors [115]. Accordingly, it is crucial to design the electrochemical treatment process with appropriate levels of current density, external electrolyte, electrode configuration, and treatment duration in light of the condition of the existing HVFA cementitious composite.

6.3. Replacement by Finer SCMs. Supplementary cementitious materials with finer sized fly ash, such as ultrafine fly ash, fine limestone powder, glass powder, and silica fume, show an ability to improve the mechanical properties of HVFA mixtures at both early and later stages when replacing normal fly ash at a low dosage (i.e., 10%–15% by mass). An ultrafine fly ash, which features the mean size of 3.4 μm and the amorphous content of about 81%, consumes CH faster than normal fly ash (mean size of 10 μm and amorphous content of 63%) and hence generates more amorphous C-S-H in HVFA mixtures [25].

The incorporation of limestone powder with a median size of 0.7 μm in diameter could recover a portion of the loss in compressive strength of HVFA mixtures [128]. The fine limestone powder is able to compact the structure of HVFA mixtures through the pore-filling effect, resulting in the improved compressive strength at early ages (first 3 days) [129]. At more than 56 days, limestone powder in HVFA mixtures facilitates the pozzolanic reaction of fly ash by providing additional surfaces for the nucleation and growth of reaction products and participates in the production of carboaluminate hydrates via reaction with aluminate phases [128, 129]. Admixed limestone can also stabilize ettringite, lower the CH content, and increase the amount of C-S-H and C-A-S-H gels in concrete incorporating other SCMs [118].

With a mean size of 20 μm and a significant amorphous content of about 90%, a glass powder has been reported to positively influence the mechanical properties of a HVFA mixture, through the nucleation effect at an early age and pozzolanic reaction at later age [130].

When silica fume is added to concrete to replace fly ash at levels from 2% to 20% by mass, it significantly increases the compressive strength of HVFA concrete [24, 80, 131–133]. Replacing fly ash with silica fume at levels of 10% and 20% could reportedly result in an enhancement of 140% and 191% in the compressive strength of HVFA concrete, respectively [131]. As the finest SCM (other than nanomaterials) usually admixed in cement-based materials, silica fume features a specific surface area as high as 20200 m^2/kg [132], as shown in Figure 18. Therefore, silica fume significantly benefits the compressive strength of HVFA mixture through multiple effects, by filling the pores (i.e., "packing"), serving as nucleation sites, improving the pore size distribution, and being a high-reactive pozzolan [131].

6.4. Addition of Nanomaterials. Recently, nanomaterials have become increasingly popular in cement-based materials, showing great potential in improving their resistance to physical and chemical deterioration [134]. There are cumulative studies on HVFA cementitious composites dedicated to enhancing the mechanical properties of HVFA mixtures by admixing nanomaterials. At the optimum dosage of 1-2 wt%, nano-$CaCO_3$ contributes to the improvement of compressive strength of HVFA concretes because its reaction with the silicate and aluminate contents in fly ash, resulting in the generation of additional hydration products (e.g., carboaluminates) and the densification of the microstructure [43, 58, 93, 135, 136]. The increased consumption of CH and the presence of ettringite (AFt) in HVFA mixtures both indicate that $CaCO_3$ nanoparticles facilitate the pozzolanic reaction of fly ash and the hydration of cement, which also contribute to the strength

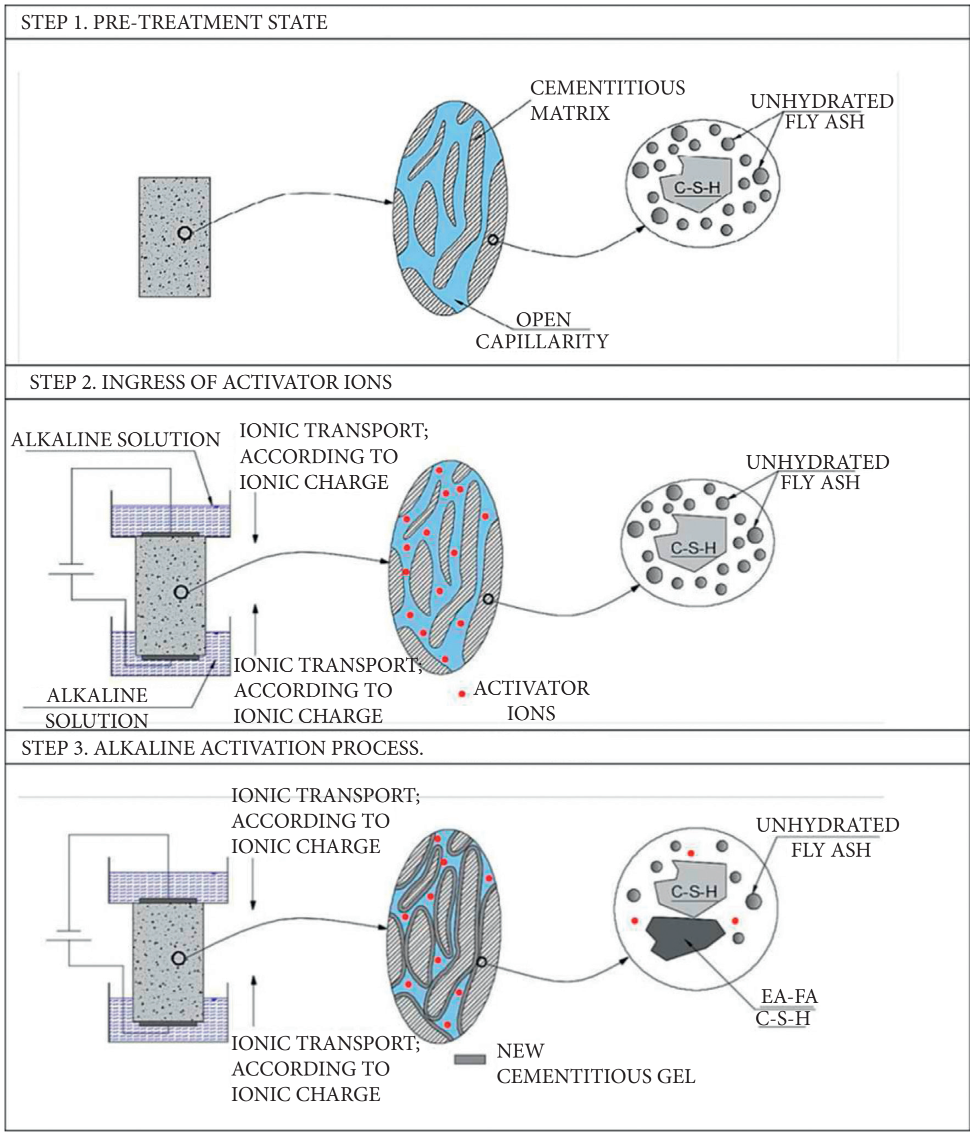


Figure 17: Preliminary conceptual model of the electrochemical activation process, reproduced from [127], with permission from Elsevier, 2021.

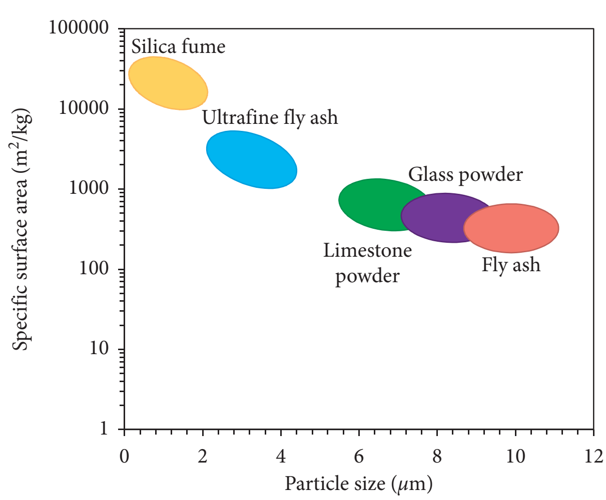


Figure 18: Particle size and specific surface area related to SCMs, based on data collected from relevant studies.

development of HVFA mixtures [93]. As shown in Figure 19 [97], the $CaCO_3$ nanoparticles-modified HVFA paste (FA59NC1) showed reductions in the intensity of CH peaks and calcium silicate peaks, as well as the presence of AFt peaks, relative to the pure HVFA paste (FA60). In the case of HVFA mortar containing 60% fly ash, the addition of 1% nano-$CaCO_3$ increased the 7 d and 28 d compressive strengths by about 100% and 111%, respectively [136]. Another study confirmed that the strength loss of HVFA concretes due to the partial replacement of cement with fly ash can be recovered by the incorporation of 1% nano-$CaCO_3$ [58], especially at 28 d [135].

Nanosilica is a highly reactive pozzolanic nanomaterial that can react with CH at an early age and increase the pozzolanic activity of fly ash at a later age, both of which contribute to better mechanical properties of HVFA mixtures [133]. Nanosilica enables better particle packing (due to its extremely small particle size), which also benefits the compressive strength development [137]. The addition of 1–5 wt% powdery or colloidal nanosilica in HVFA mixtures could compensate for the decrease in the early-age strength of HVFA mixtures [25, 36, 38, 40, 44, 133, 138]. Specifically, the addition of 2 wt% nanosilica increased the compressive strength of a 60% fly ash HVFA concrete by 95%, 7%, and 48% at 3, 7, and 28 d, respectively [21]. The ITZ of HVFA concrete can also be compacted by the admixed nanosilica [36, 38]. Figure 20 shows the microstructure of ITZ in a HVFA concrete, in which the reduction of CH and generation of C-S-H in the ITZ due to nanosilica can be observed, resulting in a densified ITZ and consequently an increased strength.

It should be noted that an adverse effect was observed in the later-age strength of HVFA mixtures when admixing colloidal nanosilica. This adverse effect was due to the lower hydration degree of fly ash that resulted from the lack of CH as well as the hindered hydration of cement by colloidal nanosilica [139]. Moreover, although HVFA mixtures could feature smaller pores with a high amount of nanosilica (such as 7.5%) admixed, the growth of AFt crystals in them exerts expansive pressure on the pore walls, inducing the development of microcracks [138].

6.5. Inclusion of Fibers. The admixed fibers either have no significant effect on the compressive strength of HVFA mixtures or the common increase is limited to about 10%. In contrast, they can increase the flexural strength and splitting tensile strength by about 20%, given the same dosage of fiber [44, 57]. The inclusion of 0.03–1 vol% fiber, especially straight steel fiber and polyester fiber, can slightly improve the compressive strength of HVFA mixtures [37, 46, 57, 137, 140, 141]. This improvement is attributed to the ability of the small fibers to delay the microcracks formation and arrest their propagation [142]. As a result, the admixed fibers can delay the cracking in concrete upon loading and thus compensate for the strength loss due to the fly ash substitution for cement [57]. The fact that the fibers act as crack arresters contributes more to enhancing the tensile and ductility properties of HVFA mixtures relative to compressive strength [46].

The fiber strands are usually discretely distributed throughout the HVFA mixtures [37]. If the fiber is admixed at high dosage (above 3 vol%), a decrease in the compressive strength of HVFA mixtures can be observed; in other words, there is an optimum dosage of fiber, beyond which the agglomeration of fibers would occur [140].

The fibers and nanomaterials can be used synergistically to benefit the mechanical properties of HVFA mixtures [44, 143, 144]. Figure 21 depicts the positive effect of nanosilica (NS) on the microstructure of HVFA mortars incorporating polyvinyl acetate (PVA) microfibers. It is clearly shown that due to the 1 wt% admixed NS, the surface of the PVA microfibers is covered by hydration products (mainly additional C-S-H). This mechanism improved the bonding between fibers and HVFA matrix, resulting in an efficient load transfer by fiber bridging and thus increased mechanical properties [44]. Another study similarly revealed the synergistic effects of montmorillonite nanoclay and polyethylene microfiber in a 70% fly ash HVFA foamed cementitious paste [145].

7. Research Needs and Remaining Challenges of HVFA Mixtures

Cumulative studies have investigated multiple aspects of HVFA cementitious composites and most of them demonstrated that HVFA mixtures can be successfully applied in construction if implemented appropriately. Figure 22 presents the overview of this review, including the superior aspects (workability, later-age strength, shrinkage, alkali-aggregate reaction, transport properties, chemical attack, and environmental impact) and inferior aspects (setting time, early-age strength, abrasion, carbonation, freezing-thawing, and salt scaling) of HVFA mixtures relative to OPC counterparts, as well as the measures that can be taken to improve their main hurdle (early-age strength) for wider acceptance. However, the following gaps (in the current knowledge base) and/or challenges need to be addressed, likely through multidisciplinary innovations and collaboration, before the construction industry can efficiently move forward with the optimal use of this promising type of material and introduce it into mainstream use.

There is a very limited understanding to enable quantitative and reliable prediction of the mechanical properties and durability performance of HVFA mixtures, based on a selection of physical and chemical characteristics of fly ash, such as chemical composition, fineness, crystallinity, and loss-on-ignition. Depending on the supply source and upstream process, fly ash is an industrial by-product with great variability in its physicochemical characteristics, making its quality assurance difficult yet critical. Although there are studies aiming to correlate the performance of HVFA mixtures with the characteristics of fly ash, the obtained correlations are unique for the fly ash used in each study and vary from one study to another.

Furthermore, in some areas, the supply of conventional fly ash is dwindling because of the rapid construction using up the supply or the changes of fuel sources at the electric power plants (e.g., from coal to natural gas). In this context,

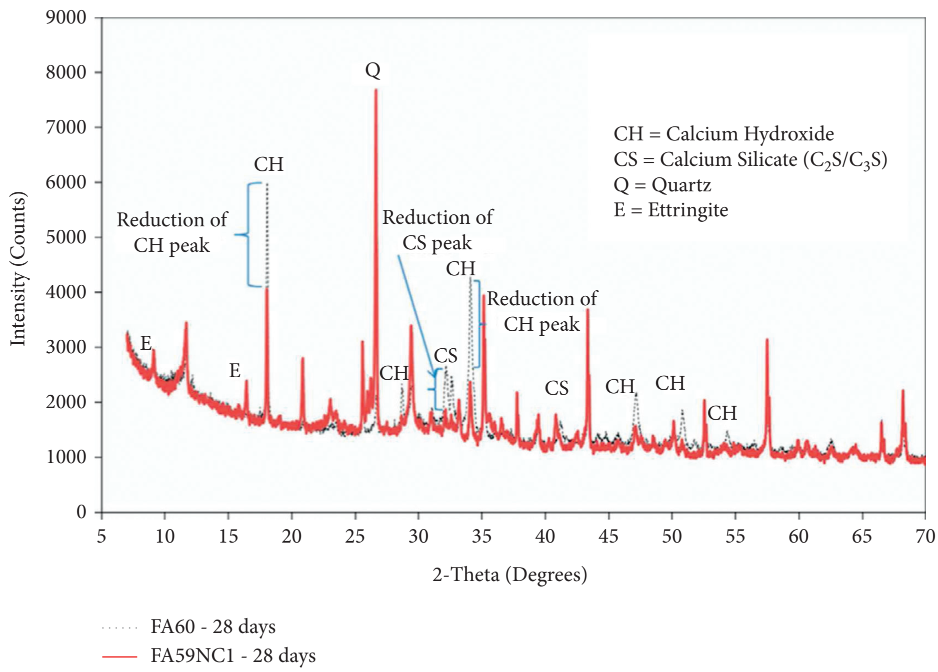
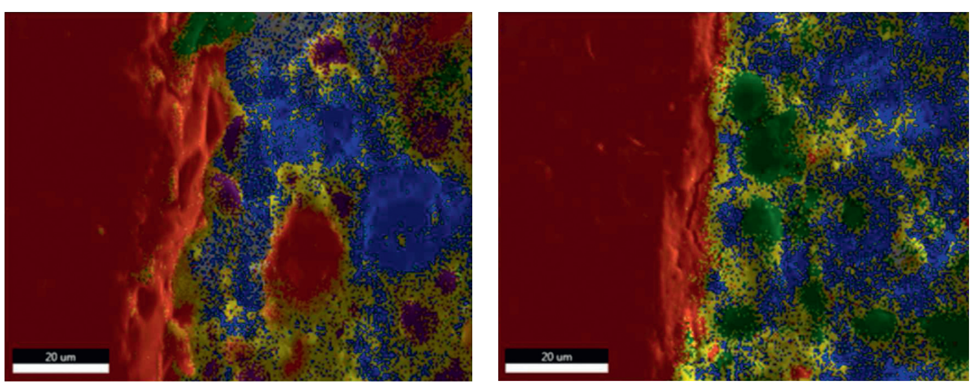
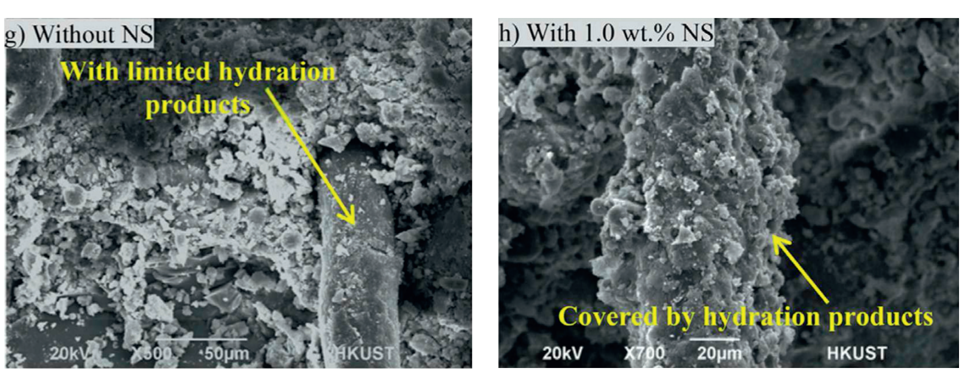


Figure 19: XRD analysis of HVFA pastes with and without nano-$CaCO_3$ at 28 d, reproduced from [93], with permission from Elsevier, 2021.



(a) (b)

Figure 20: SEM-EDS analysis of the ITZ in HVFA concrete: (a) no addition of nanosilica; (b) admixing 1 wt% nanosilica (Red part, Si-rich phase such as aggregate; blue and yellow parts, Ca-rich phase such as CH; green part, C-S-H), reproduced from [38], with permission from Elsevier, 2021. SEM, scanning electron microscopy; EDS, energy-dispersive X-ray spectroscopy.



(a) (b)

Figure 21: SEM images of a microfiber-modified HVFA mortar with and without NS, reproduced from [44], with permission from Elsevier, 2021.

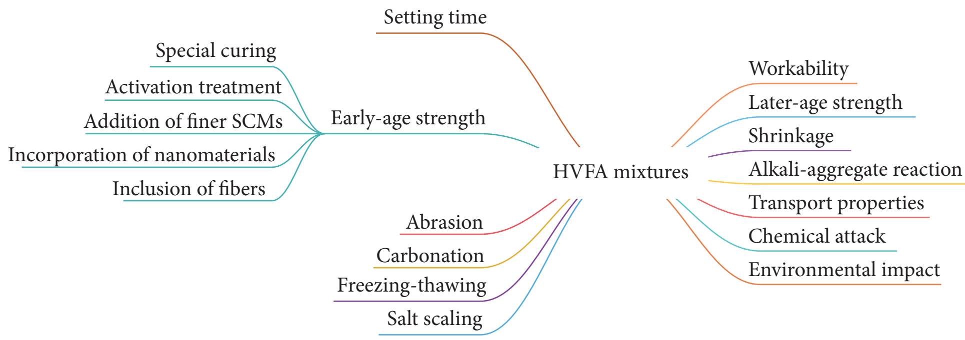


Figure 22: Renewed perspective about the superior and inferior aspects of HVFA mixtures relative to OPC counterparts.

some unconventional sources of fly ash have been considered for use in HVFA mixtures, such as ash disposed of in landfills or impoundments and biomass fly ash. This makes the prediction of the performance of HVFA mixtures based on the physicochemical characteristics of fly ash more complicated yet increasingly necessary.

Nearly every study about the HVFA mixture claims its superiority to OPC mixture in terms of carbon footprint, environmental friendliness, and sustainability. However, to reduce the setting time and increase the early-age strength of HVFA mixtures, some measures are taken to facilitate its application in the field, including the use of special curing regimes, activators, finer SCMs, nanomaterials, or fibers. These measures may induce extra costs and environmental impact for the production of HVFA mixtures. In these cases, it is in an urgent need to conduct the LCA analysis of the modified HVFA mixtures beyond the plain HVFA mixtures [39]. In addition, field implementation of in-place measurement methods for estimating the setting time and compressive strength is crucial for successful application of HVFA mixtures [146, 147].

There is a lack of fundamental understanding of HVFA mixtures as types of multiscale, heterogeneous composite materials, particularly in terms of deterioration under coupled mechanical and environmental loadings and potential chemical and physical synergies between various constituents (cement, fly ash, admixtures, other SCMs, fibers, and nanomaterials). Many of the existing studies are limited to treating concrete as a homogenous material and empirically evaluating its properties and performance in a macroscopic manner. The fundamental understanding of composition-property-performance relationships is crucial to guide best practices in selecting materials, preserving, maintaining, and possibly rehabilitating and recycling HVFA cementitious composites.

8. Concluding Remarks

This work provides an overview regarding recent advances in fresh properties, engineering performance, and environmental impact of HVFA cementitious composites. Some key findings from the review are provided as follows:

(i) The replacement of cement with HVFA binder in cementitious materials could increase the slump with the fly ash content lower than 60% due to the ball-bearing effect of fly ash, beyond which the slump of HVFA mixtures begins to decline because of the high surface area of fly ash. The setting time of HVFA mixtures is retarded, resulting from the dilution of cement and the slow pozzolanic reaction of fly ash.

(ii) There exists a reduction in the mechanical properties of HVFA mixtures compared with OPC mixtures, especially at early ages. This reduction increases with increasing fly ash content and decreases with increasing curing age. The combination of 50%–60% fly ash and w/cm of 0.3–04 is optimal for the pozzolanic reaction of fly ash.

(iii) Relative to their OPC counterparts, HVFA cementitious composites exhibit lower resistances to abrasion and carbonation. Freeze-thaw and salt scaling resistances are also lowered by the replacement of cement by HVFA binder, but proper air entrainment can endow HVFA mixtures with excellent freeze-thaw durability. HVFA mixtures exhibit lower environmental impact and better durability performance in terms of shrinkage, AARs, transport properties, and chemical attack.

(iv) To compensate the loss in mechanical properties due to the introduction of HVFA in cement-based materials, many measures can be taken, including elevated-temperature curing, internal curing, physical or chemical activation treatments, partially replacing normal fly ash by SCM with a finer size, admixing nanomaterials at appropriate dosage, and incorporating fibers.

(v) Both nanoscience and nanoengineering hold great promise in advancing the technology of HVFA cementitious composites.

(vi) Some aspects of HVFA-based materials, such as the application of HVFA binder in emerging concrete technologies (3-D printing concrete, ultra-high performance concrete, self-healing concrete, etc.), beneficial use of marginal fly ashes (biomass fly ash, municipal solid waste incineration fly ash, disposed fly ash, and ponded fly ash), and life cycle assessment considering all possible constituents and footprint, need to be researched and reviewed in future work.

Data Availability

All the relevant data are presented in this review article.

Additional Points

Highlights. (i) HVFA cementitious composites as a class of sustainable materials were reviewed. (ii) The combination of 50%–60% fly ash and w/cm of 0.3–04 is optimal for HVFA mixtures. (iii) Lower environmental impact and better durability in terms of shrinkage, AARs, transport properties, and chemical attack. (iv) Low early-age strength and low resistance to abrasion, carbonation, and salt scaling. (v) Alternative curing, activation, and use of finer SCM, fibers, and nanomaterials can help. (vi) Nanotechnology holds great promise in advancing HVFA concrete technology. (vii) Beneficial uses of marginal fly ashes, LCA of HVFA mixtures containing all possible constituents, etc., need to be further explored.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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