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Article

Nano-Silica–Enhanced Binder Synergy and Multi-Index Performance of Resource-Efficient Alkali-Activated Composites for Sustainable Infrastructure Applications

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Abstract

This study presents a multi-index performance method to measure the synergy of nano-silica-enhanced binders in resource efficient alkali-activated composites based on the Strength Activity Index (SAI) as a reference index according to ASTM C618. High-calcium fly ash (HCFA) and low-calcium fly ash (LCFA) were used with fine aggregate replacement level is kept constant at 20% by mass and nano-silica was incorporated at 0, 1, 2, and 3 wt% of the binder to prepare alkali-activated slag fly ash composites. The fresh state performance was assessed using the Initial Flow Index (IFI) and Flow Retention Index (FRI), while the mechanical performance was evaluated using the compressive, tensile, and flexural indices (SAI, TSI, and FSI). These results indicate that with an increase in nano-silica content, flowability and workability retention reduces systematically, with LCFA-based mixtures always exhibiting higher fresh-state retention than HCFA systems. Optimal mechanical performance was achieved with an intermediate nano-silica concentration of about 2 wt%, with consequent maximum SAI performance of about 120% at 28 days with HCFA-based mixtures and 118% at 28 days with LCFA-based mixtures, as well as a uniform improvement in TSI and FSI. Correlation analyses between SAI and tensile and flexural indices revealed clear linearity (R^2 of about 0.91-0.95) which indicated that compressive strength is not a sufficient measure of total mechanical performance. The mineralogical and microstructural analyses assisted by X-ray diffraction (XRD) and scanning electron microscopy (SEM) showed that the performance trends observed depend on the interactions of the calcium supply, amorphous aluminosilicate and the nucleation effects of nano-silica. The proposed solution provides a performance-based approach towards the optimal utilization of nano-silica-modified alkali-activated composites to be used in sustainable infrastructure applications.

Keywords: nano-silica; alkali-activated composites; resource efficiency; binder synergy; multi-index performance; sustainable infrastructure applications

1. Introduction

The construction industry worldwide is experiencing a paradigm shift toward resource-efficient and low-impact material systems, with growing environmental limitations, resource depletion, and performance requirements of the contemporary infrastructure [1-3]. Traditional substances that use Portland cement are still considered to be among the most energy-consuming construction products, which also leads to a substantial contribution to global carbon emissions and the exhaustion of natural resources [2,4,5]. The challenges have been heating research on alternative binder systems that would provide high mechanical performance with minimum environmental burdens [6,7].

Alkali-activated materials have been considered as an exciting category of cementitious composites since they can use industrial by-products like slag and fly ash as the main precursors [8-10]. Alkali-activated composites have significantly lower carbon footprints, greater chemical performance, and superior durability in hostile conditions compared to normal Portland cement systems [2,11]. The fact that they are versatile in the sense of adapting to various sources of aluminosilicate further increases their applicability to the practice of constructing resources efficiently [7]. The behavior of alkali-activated systems is, however, quite sensitive to the binder chemistry, precursor activity, and microstructural growth, which requires special attention to the optimization of the mixture [12].

The role of fly ash in alkali-activated composites is important because of its aluminosilicate composition and the morphology of the particles in the form of a sphere [13,14]. Chemical and mineralogical properties of fly ash, most notably the content of calcium, affect the kinetics of the reaction, as well as mechanical performance, to a significant extent [15-17]. The early-age reactivity of a high-calcium fly ash is generally higher, and these fly ashes assist in the creation of products rich in calcium, contributing to higher rates of strength development, whereas fly ashes with low calcium levels can enhance the base of polymerization-related processes that help to enhance stability over time [18-20]. Although fly ash tends to be used as a precursor to a binding agent, its application in the form of a partial fine aggregate replacement has been of interest recently because it could enhance the properties of particle packing and minimize the use of natural sand resources [15,21].

Although these are the benefits, the balancing of the binder synergy in alkali-activated slag fly ash systems is still not easy to achieve [22,7]. Over incorporation can cause dilution effects, low load-bearing capability, and deteriorated fresh-state properties, and under incarceration can restrict sustainability advantages [23,24]. Additionally, it has been established that alkali-activated systems are sensitive to the parameters of the mixture, like water demand, activator concentration, and size distribution, which have a direct influence on the workability and development of strength [25,7]. To overcome these issues, advanced modifiers that can be used to refine the microstructure and increase reaction efficiency to the detriment of fresh-state performance are necessary.

Nano silica has also drawn much interest as a nano-scale cementitious and alkali-activated systems nano-additive because of its tremendously high specific surface area and chemical reactivity [26-28]. As a physical filler and a chemically active nucleation agent, nano-silica can hasten reaction kinetics and enhance the development of extra binding phases, as well as refit pore structure [9,28]. With alkali-activated composites, nano-silica has also been cited to help in increasing the interfacial transition zone, better packing density of particles, and raising the degree of polymerization, which results in better mechanical performance [29,30]. Nevertheless, nano-silica is highly dosage-sensitive, and its over-incorporation can lead to particle agglomeration, an increase in water requirement, and low workability [26,31].

Although many research works have analyzed nano-silica-modified cementitious and alkali-activated systems, most tests have been largely based on compressive strength as the key outcome of performance [10]. This method will give little information on tensile and flexural behavior, which will be critical in determining cracking resistance, redistribution of stress, and the overall structural reliability [30,12]. Furthermore, the interplay of nano-silica and various types of fly ash, under regulated alkali-activated circumstances, has not been assessed in systematic appraisals of combined performance-based systems [9].

Recently, it has been highlighted that there is a necessity to have a multi-indexed performance measurement system that goes beyond compressive strength to reflect the intricate mechanical action of advanced cementitious composites [31,32]. Normalized performance indices consisting of tensile and flexural performances give a better measure of material efficiency, particularly under systems where microstructural refinement and interface bonding are of primary importance [12]. The use of fresh-state performance measures combined with mechanical indexes also increases the trustworthiness of mixture optimization, as it ensures constructability as well as mechanical efficiency [33].

From an infrastructure engineering perspective, these performance-based evaluation systems are becoming increasingly demanded to ensure that advanced cementitious composites not only meet the strength requirements but also meet the constructability, crack resistance, and long-term service performance requirements under a realistic loading environment [34-37].

In recent infrastructure usage, complex stress conditions, time-dependent loading and aggressive service conditions to which materials are commonly subjected make durability and mechanical reliability equally important. As a result, assessment systems that combine fresh-state behavior and multi-directional mechanical performance give a more realistic assessment of in-service behavior when compared to strength-based measures alone. With these approaches, careful selection of materials, and optimization of mixtures can be made in a way that it addresses the needs of infrastructure systems where consistency in performance, resilience, and sustainability are critical design goals.

In line with this, the current study aims to examine the synergistic effect of nano-silica to control the interaction between the binder and the development of the resource-efficient alkali-activated composite using a holistic multi-index performance platform, which is based on the Strength Activity Index (SAI) as a reference index according to ASTM C618 [38].

The composites that were prepared with slag and fly ash as alkali-activated slag and with low-calcium and high-calcium fly ash, with a fixed degree of fine aggregate replacement, and nano-silica added at different dosages. Flow-based indices were used to measure fresh-state behavior, and normalized compressive-, tensile-, and flexural-based indices were used to measure mechanical performance. An integration of these indicators provides the study with the complete perspective of nano-silica-enhanced synergy and establishes a rational base for the creation of high-performance, resource-effective alkali-activated composite systems with clear perspectives in terms of sustainable infrastructure applications.

It is essential to note that the proposed framework is intended to be a performance-integration methodology, rather than a mechanistic approach or a predictive modeling approach. Unlike the classical study of compressive strength as the selection of a performance metric, the current study involves an integrative multi-index design that integrates fresh-state indices (IFI, FRI) and normalized hardened-state indices (SAI, TSI, and FSI). To make such a performance-based measure, mineralogical (XRD) and microstructural (SEM) characterizations are given to decode the phase evolution, amorphous phase, and microstructural refinements of the evident fresh-state and mechanical performance of the identified materials.

The experimental and characterization methodology facilitates the study of the alkali-activated slag fly ash composites more comprehensively and performance-based, particularly systems where constructability and mechanical response are highly conditional on the nano-scale adjustment and interaction of binders.

2. Experimental Program and Methodology

As depicted in Figure 1, the sequence of actions followed in the experimental outline applied in this study includes the selection of raw materials and mix proportions prior to the testing procedure which was conducted with the aim of studying nano-silica adjusted synergy in alkali-activated, resource-saving composites.

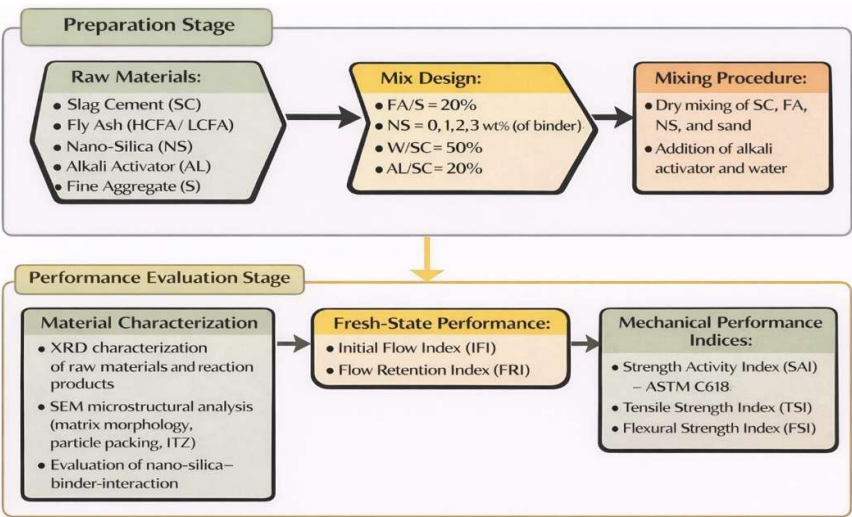


Figure 1. Experimental framework for nano-silica enhanced alkali-activated composites.

The framework also involves the mineralogical and microstructural characterization using the X-ray diffraction (XRD) and scanning electron microscopy (SEM) to give a fundamental insight into the mechanics of the interaction between the binder system and nano-silica incorporation. This combined description allows relating phase composition and microstructural refinement with reaction kinetics, matrix densification as well as fresh-state and mechanical performance of alkali-activated composites.

The workability and flowability retention properties were quantitatively measured by the use of the Initial Flow Index (IFI) and Flow Retention Index (FRI) in assessing fresh-state quantitative performance.

Mechanical performance was measured using a multi-index of the Strength Activity Index (SAI), which is defined in ASTM C618 [38] and complemented by tensile-based and flexural-based indices, the Tensile Strength Index (TSI) and Flexural Strength Index (FSI). The synergistic method enables the comprehensive evaluation of the influence of nano-silica inclusion on fresh and hardened properties that act as a performance-driven basis to the knowledge of the synergy of the role of binder and mixture optimization in the alkali-activated composite systems.

2.1. Materials and Constituent Properties

Slag cement (SC), high-calcium fly ash (HCFA), low-calcium fly ash (LCFA), nano-silica (NS), an alkaline activator (AL), and fine aggregate (S) were used as the main constituents of this study. Their combination in the simultaneous application permitted the performance of the systematic analysis of the synergetic effect of slag binders, alkali-activated fly ash, and nano-silica activeness on the formation of resource-efficient alkali-activated composite systems.

The slag cement (SC) is a by-product of municipal waste, and it contains high proportions of amorphous materials. It was selected as a green option since it can minimize the environmental impact that is usually associated with the production of Portland cement.

The sources of the high-calcium fly ash (HCFA), low-calcium fly ash (LCFA) were coal power plants, which utilize a wide variety of fuels. The HCFA is rich in CaO, and LCFA is rich in amorphous silica and alumina.

The nano-silica (NS) is recycled waste glass as a nano-scale modifier to bring synergy in binders and micro-structure fineness in the alkali-activated system. The nano-silica is largely amorphous, as shown by its chemical composition and physical characteristics (Tables 1 and 2) and a very small particle size and very high specific surface area.

These properties make nano-silica a useful physical filler and chemically active moiety, inducing nucleation of reaction products and increasing the density of particle packing and the interfacial bonding of the alkali-activated matrix.

This paper used nano-silica in the percentage of total binder to provide a systematic assessment of the dosage-related impact of the nano-silica and fly ash types on the fresh-state behavior and hardened mechanical performance.

Mixed care was taken to eliminate agglomeration of nano-silica and to achieve even distribution of the nano-silica so that its full involvement in the alkali-activation process could be achieved.

The alkaline activator (AL) is sodium silicate (Na_2SiO_3) with a molar ratio of $\text{SiO}_2/\text{Na}_2\text{O}=1.0$ (50% SiO_2 and 50% Na_2O), which was chosen as an activator of slag-fly ash binders. Activation dosage was kept unchanged in all mixtures ($\text{AL}/\text{SC}=20\%$ by mass) in order to isolate the effect of the addition of nano-silica.

The fine aggregate (S) was used in the aggregate phase and consisted of crushed sandstone with specific gravity of 2.58 and fineness modulus of 2.83; fly ash ($\text{FA}/\text{S}=20\%$ by weight) was incorporated in the aggregate phase to partially substitute the fine aggregate, enhancing the ability of the particle to pack efficiently and decreasing the use of natural sand.

X-ray fluorescence (XRF) analysis was used to establish the chemical composition of slag cement, types of fly ash, and nano-silica, and loss on ignition (LOI) was determined according to the standard procedures. The specific surface area and particle size distribution of slag cement and fly ash were calculated using the Blaine permeability technique and laser diffraction method, respectively. Conversely, the Brunauer-Emmett-Teller (BET) method was used to determine the specific surface area of nano-silica because it is nano-scale in nature.

Table 1 shows the chemical compositions of slag cement (SC), high-calcium fly ash (HCFA), low-calcium fly ash (LCFA), and nano-silica (NS), whereas Table 2 presents the physical properties of these materials in a summary.

These characterizations form the requisite background on understanding the effect of nano-silica on the interaction of binders, the fresh-state behavior and the mechanical performance of the alkali-activated composites.

Table 1. Chemical compositions of slag cement (SC), high-calcium fly ash (HCFA), low-calcium fly ash (LCFA), and nano-silica (NS).

Chemical Compositions (%)	CaO	SiO ₂	Al ₂ O ₃	MgO	Fe ₂ O ₃	Na ₂ O	TiO ₂	P ₂ O ₅	LOI
Slag Cement - (SC)	43.1	32.5	13.5	2.9	2.7	1.8	1.3	0.8	1.4
High-Calcium Fly Ash - (HCFA)	18.8	48.8	19.8	1.5	3.8	1.2	3.9	0.5	1.7
Low-Calcium Fly Ash - (LCFA)	6.3	57.6	26.5	1.2	4.2	0.5	1.9	0.3	1.5
Nano-Silica - (NS)	0.6	98.2	0.4	0.1	0.2	0.3	<0.05	<0.05	0.2

Table 2. Physical properties of slag cement (SC), high-calcium fly ash (HCFA), low-calcium fly ash (LCFA), and nano-silica (NS).

Physical Properties	Specific gravity (g/cm ³)	Specific surface area, Blaine (cm ² /g)	Specific surface area, BET (m ² /g)	Average particle size D ₅₀ (μm)
Slag Cement - (SC)	2.80	3750	---	6.48
High-Calcium Fly Ash - (HCFA)	2.80	3780	---	16.25
Low-Calcium Fly Ash - (LCFA)	2.14	3630	---	18.35
Nano-Silica - (NS)	2.20	---	85	0.04

2.2. Mixture Design and Proportioning

Table 3 summarizes the mix proportions used in this work. Each of the alkali-activated composite mixtures was prepared to examine the effects of the incorporation of nano-silica and fly ash types on binder synergy and performance at constant baseline mixture parameters. The primary binder in all mixtures was slag cement (SC) and fly ash was added as a partial replacement of fine aggregates in all mixtures at 20% fly ash-sand (FA/S) proportion by weight. High-calcium fly ash (HCFA) and low-calcium fly ash (LCFA) were used to investigate the relationship between fly ash types and nano-silica dosage.

To determine the impact of nano-silica and fly ash types, the water-to-slag ratio (W/SC), alkali activator-to-slag ratio (AL/SC) were kept constant throughout the mixtures at 50% and 20%, respectively. Nano-silica was added in 0, 1, 2, and 3 wt% to the total binder content. The reference mixtures containing no nano-silica (NS = 0%) were used as control mixes to normalize the performance and make a comparative evaluation.

Fine aggregate was a mixture of crushed sandstone, which was controlled in terms of grading and was partially substituted by fly ash to increase the efficiency of particle packing and minimize the use of natural sand. To compare reliably and consistently different dosages of nano-silica and fly ash types, the mixtures were prepared under the same conditions in terms of mixing, casting, and curing conditions.

Table 3. Mix proportions of nano-silica–modified alkali-activated composites.

Mix ID	Fly Ash Type	NS (%)	FA/S (%)	W/SC (%)	AL/SC (%)	Mix Proportioning (kg/m ³)					
						Slag Cement (SC)	Alkali				Nano-Silica (NS)
							Water (W)	- Activator (AL)	Fly Ash (FA)	Sand (S)	
HCFA-NS0 – Ref.	HCFA	0	20	50	20	600	300	120	200	1000	0
HCFA-NS1		1				594					6
HCFA-NS2		2				588					12
HCFA-NS3		3				582					18
LCFA-NS0 – Ref.	LCFA	0	20	50	20	600	300	120	200	1000	0
LCFA-NS1		1				594					6
LCFA-NS2		2				588					12
LCFA-NS3		3				582					18

2.3. Experimental Procedures and Testing

The mineralogical properties, fresh-state behavior and mechanical performance of nano-silica-modified alkali-activated composites were compared through an organized experimental program that was carried out under controlled and consistent testing conditions. The experimental processes were planned to allow the comparison of the various nano-silica dosages and fly-ash types to be made in a reliable way so that they are relevant to the infrastructure-based performance assessment.

2.3.1. Mineralogical Characteristics of Constituent Materials

Mineralogical characterization of the raw materials was made to provide a basic platform on which the reactivity and interaction mechanisms of the raw materials in the nano-silica modified alkali-activated composite system could be understood. The analysis by X-ray diffraction (XRD) was used to determine crystalline and amorphous phases in slag cement (SC), high-calcium fly ash (HCFA), low-calcium fly ash (LCFA), and nano-silica (NS). The resulting patterns of diffraction were qualitatively evaluated to determine the presence of crystalline phases containing calcium and amorphous aluminosilicate components, which are important in regulating reactivity and reaction between alkali activation and other components.

2.3.2. Fresh-State Flow Assessment

Flow-based indices were used to quantify the fresh-state performance of nano-silica modified alkali-activated composites to measure flowability and the retention of workability. Immediately after mixing, the Initial Flow Index (IFI) was obtained in a standard flow-table test following the guidelines of JIS A1150-JSA 2014 [39] wherein the spread diameter of each mixture was normalized against the respective reference mixture devoid of nano-silica (NS = 0%). Flow Retention Index (FRI) was established after casting 15 minutes to evaluate the potential of the mixtures to retain their flowability. The measures of both indices were based on standardized procedures that made it possible to compare mixtures with varying nano-silica dosages and types of fly ash.

2.3.3. Mechanical Performance Evaluation

The nano-silica modified alkali-activated composites were tested in mechanical performance with respect to compressive, splitting tensile, and flexural strength. Multi-index methodology was used to assess mechanical performance. The compressive strength was measured in terms of Strength Activity Index (SAI) which according to ASTM C618 [38] is determined as the ratio of compressive strength of the nano-silica modified mixtures to the compressive strength of the corresponding reference mixtures without nano-silica (NS = 0%). The tensile and flexural performance was determined by testing in terms of Tensile Strength Index (TSI) and Flexural Strength Index (FSI) respectively. Both the indices were determined by dividing a measured splitting tensile and flexural strength of each mixture by the relevant reference mixtures, using the same normalization strategy as SAI. Compressive, splitting tensile and flexural strength tests were performed based on JIS A1108-JSA 2006a [40], JIS A1113-JSA 2006c [41], and JIS A1106-JSA 2006b [42], respectively. All of the tests were conducted at curing ages of 1, 3, 7 and 28 days of specimens exposed to a controlled steam-curing regime. Cylindrical specimens (50 × 100 mm) were used to measure compressive as well as splitting tensile strengths, and prismatic specimens (40 × 40 × 160 mm) were used to measure flexural strength. Each mixture was tested on three specimens at each curing age.

2.3.4. Microstructural Characterization (SEM)

To support the interpretation of the trends of fresh-state and mechanical performance, Scanning Electron Microscopy (SEM) was used to scan the microstructural characteristics of the chosen alkali-activated composites. At 28 days of curing, representative specimens were made of reference mixtures and mixtures containing nano-silica. The morphology of the matrices, the distribution of the particles, the structure of the pore, and the relationship between the product of the reactions and the particles that had not reacted were observed using SEM.

3. Results and Discussion

3.1. Flow-Based Performance Indicators (IFI–FRI)

Quantitative evaluation of fresh-state behavior of nano-silica-modified alkali-activated composite was done by Initial Flow Index (IFI) and the Flow Retention Index (FRI) as shown in Figures 2 and 3.

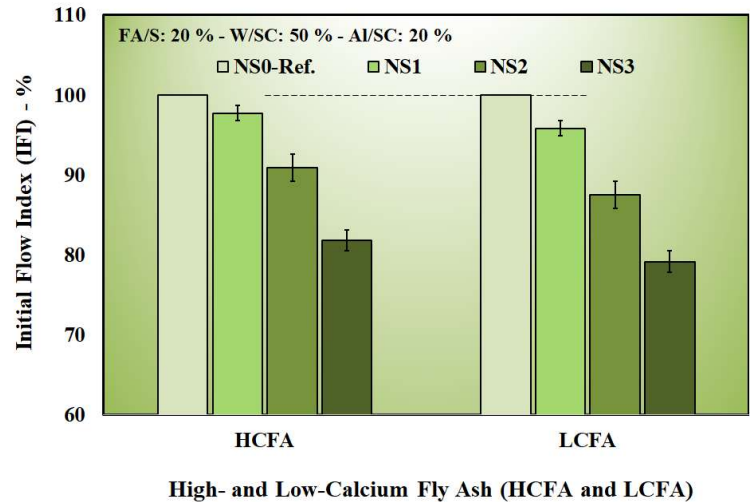


Figure 2. Initial Flow Index (IFI) of HCFA- and LCFA-based mixtures at different nano-silica dosages.

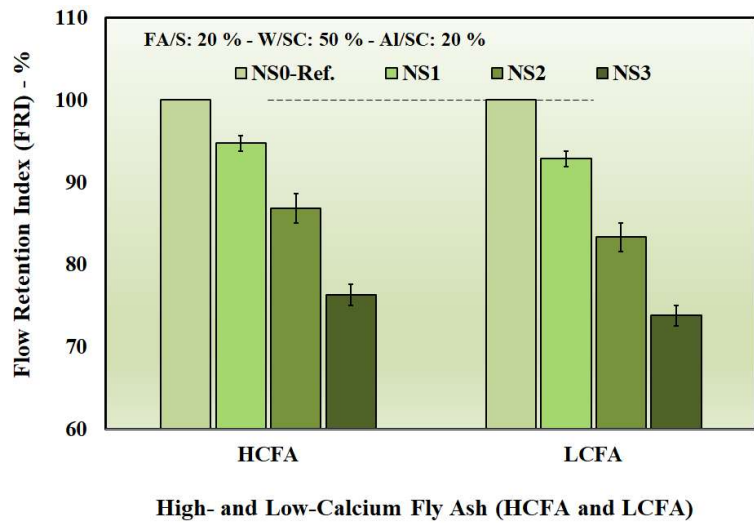


Figure 3. Flow Retention Index (FRI) of HCFA- and LCFA-based mixtures at different nano-silica dosages.

These indices have been used to measure the initial flowability immediately after mixing and the capacity of the mixtures to keep the workability with time as a function of the dosage of nano-silica and the types of fly ash.

In both HCFA-based and LCFA-based systems, there was a steady decrease in fresh-state performance as nano-silica content was increased. The reference mixtures without nano-silica (NS0) had defined values of IFI and FRI of 100% and were used as normalized baselines to be used in comparative evaluation instead of reflecting the absolute performance of the flow. Initial flow and 15 min post-cast reference mixtures were normalized based on corresponding average flow diameters (220 mm and 190 mm) for HCFA, while (240 mm and 210 mm) for LCFA, respectively, as the basis of flowability normalization across all mixtures.

With a dosage of nano-silica of 1 wt% (NS1), the values of IFI showed a moderate reduction of about 98% with HCFA mixtures and 96% with LCFA mixtures (Figure 2). This reduction at the beginning is indicative of the effect of the specific surface area and fine particle incorporation that is related to the addition of nano-silica. With further addition of nano-silica, there was observed a further reduction in initial flowability. At a 2 wt% nano-silica (NS2) concentration, IFI reduced to almost 91% in the case of HCFA and about 88% in the case of LCFA, which indicates that there is

increased internal friction and low particle movement within the fresh matrix. In the highest doses of nano-silica 3 wt% (NS3), IFI was found to have minimum values of about 82% and 79% of HCFA and LCFA respectively, which indicate a severe loss of original flowability.

The same but even more noticeable trend was reflected in flow retention behavior (Figure 3). With nano-silica at 1 wt% concentration, FRI values were relatively high with HCFA and LCFA mixtures maintaining about 95% and 93% of the initial flowability, respectively. However, when nano-silica was added at 2 wt% FRI decreased further to about 87% in HCFA and 83% in LCFA, indicating the increasing susceptibility of the blends to the effects of time in stiffening. At the maximum level of nano-silica concentration (3 wt%), HCFA mixtures lost the ability to retain workability noticeably, with FRI values decreasing to almost 76%, whereas LCFA mixtures retained a slightly better stability with FRI values being about 74%.

The IFI-FRI indices show that an increase in dosage of nano-silica has a negative result on the fresh-state flowability and workability retention of the mixtures containing the two types of fly ash.

However, mixtures using LCFA were always slightly more stable at high contents of nano-silica than their HCFA counterparts. This observation indicates that the reduced calcium level and delayed early-age reactivity of LCFA system schemes partially counterbalances the rapid hardening caused by nano-silica add-in, but mixtures of HCFA, with larger calcium content, are more inclined to increased rate-build and lose its operating ability more rapidly at higher levels of nano-silica incorporation.

3.2. Strength-Based Performance Indicators

A multi-index based on strength was used to assess the mechanical performance of the nano-silica modified alkali-activated composites based on compressive, tensile, and flexural outputs.

The methodology allows the study of mechanical behavior in a comprehensive way to characterize the load-bearing capacity and mechanisms of resistance. Compressive strength response was mainly measured by the Strength Activity Index (SAI), which used tensile- and flexural-based indices to give more details on the effect of nano-silica on the overall mechanical performance. These findings are addressed in the subsequent subsections against nano-silica dosage and the types of fly ash.

3.2.1. Compressive Strength Activity Index (SAI)

The Strength Activity Index (SAI) was used to assess the development of compressive strength of the nano-silica modified alkali-activated composites with nano-silica as demonstrated in Figures 4 and 5, respectively, of the systems based on the HCFA and LCFA.

The corresponding reference mixtures without nano-silica (NS0), which by definition had a value of 100%, were used to normalize the SAI values by their respective values to facilitate comparative assessment across the various dosage levels of nano-silica and age of curing.

In HCFA-based mixtures (Figure 4), the addition of nano-silica led to a progressive increase in compressive strength activity at all curing ages.

SAI values at 1 day increased above the level at the reference level to about 104% in NS1 and about 110% in NS2, which is a sign of faster early-age development of strength. There was a slight decrease at the highest nano-silica dosage (NS3), which indicates that diminishing returns were beginning at high levels of nano-silica content.

The same tendencies were observed at 3 and 7 days, with NS2 having the largest SAI values at both points, 112% and 116%, respectively.

At 28 days, HCFA mixtures with NS2 gave peak SAI values near 120% and NS3 mixtures continued to perform well in comparison to the reference at a lower level.

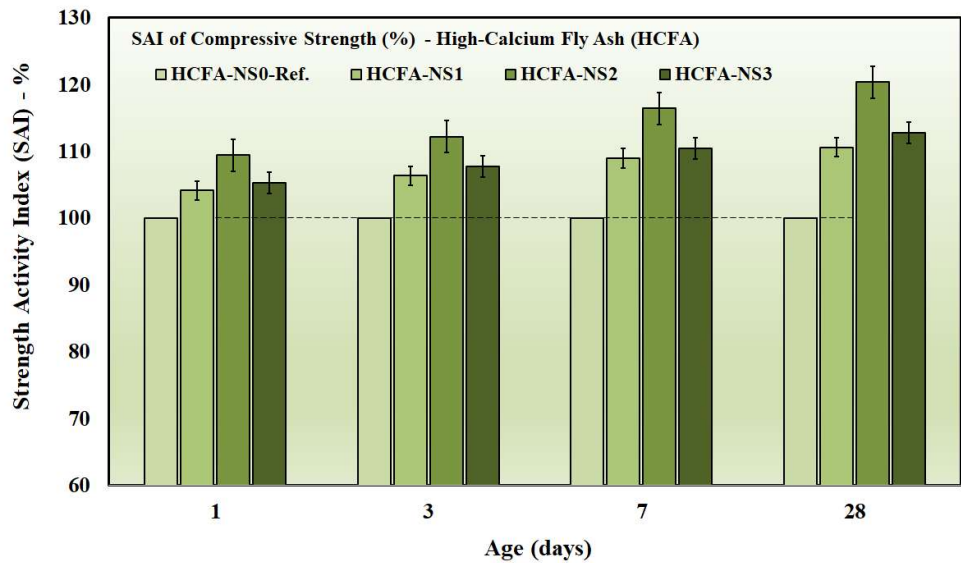


Figure 4. Compressive Strength Activity Index (SAI) of HCFA-Based Alkali-Activated Composites Modified with Nano-Silica.

A similar improvement trend was noted on LCFA-based mixtures (Figure 5), but the magnitudes of SAI generally were lower than those of HCFA systems at the same dose of nano-silica. This is because the calcium content in LCFA is low, and it moderates the reaction kinetics of early age and restricts the degree of strength gains compared to the calcium-enriched mixtures of HCFA.

At early curing ages, NS2 showed the greatest improvement and SAI value of about 108 and 110 percent at 1 and 3 days respectively.

LCFA-based mixtures with NS2 achieved SAI of almost 115% and 118 at 7 and 28 days, respectively, which confirms the positive effect of nano-silica in enhancing strength increases over time.

As in the case of HCFA systems, addition of more nano-silica to 3 wt% led to slightly lower SAI values in comparison to NS2, still higher than the reference mixtures.

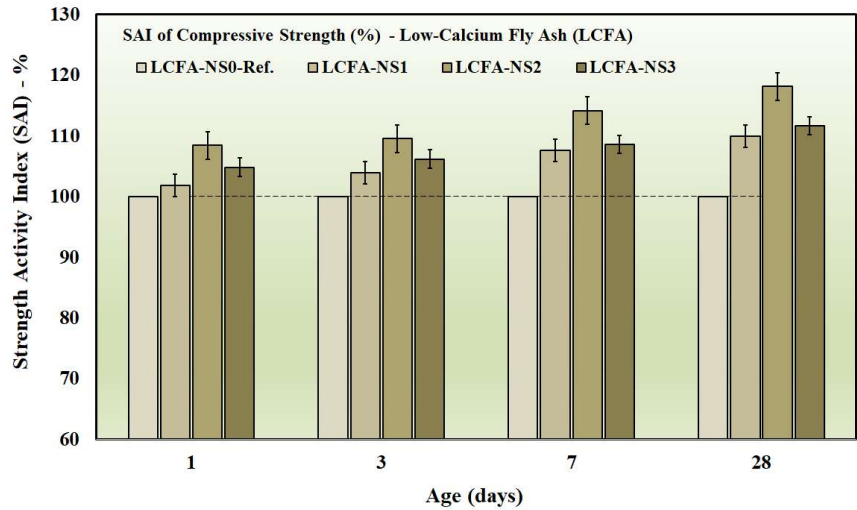


Figure 5. Compressive Strength Activity Index (SAI) of LCFA-Based Alkali-Activated Composites Modified with Nano-Silica.

Comprehensively, the results of the SAI show that the incorporation of nano-silica improves compressive strength activity in both HCFA-based and LCFA-based alkali-activated composites with an optimum dosage of nano-silica of about 2 wt% (NS2).

The steady performance peak of NS2 during curing ages and fly ash types indicates a compromising nucleation performance and dispersion efficiency of particles.

Excessive nano-silica concentration appears to limit additional strength enhancement, and this might be due to the surface area demand and reduced workability that may prevent effective transfer of stress in the hardened matrix.

3.2.2. Tensile Strength Performance Index (TSI)

The tensile performance of alkali-activated composites that were modified with nano-silica was measured based on the Tensile Strength Index (TSI), as shown in Figures 6 and 7, respectively, of the HCFA- and LCFA-based systems.

The TSI values were adjusted using the respective reference mixtures that had been prepared without nano-silica (NS0), which was the standard set to 100% to maintain the same comparing result between curing ages and nano-silica dosages.

With mixtures based on HCFA (Figure 6), tensile strength activity was evidently increased with the addition of nano-silica at all ages of curing.

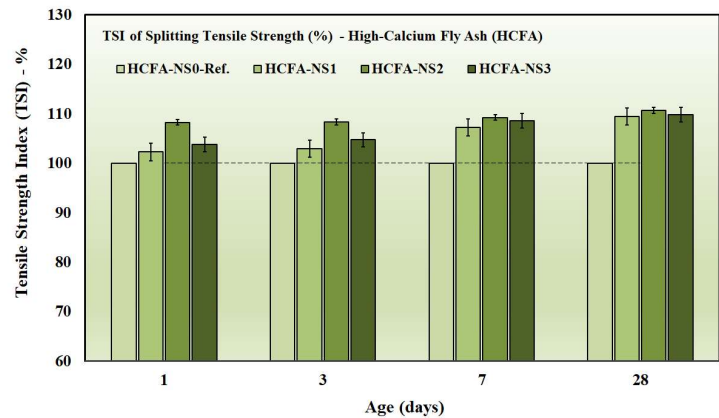


Figure 6. Tensile Strength Index (TSI) of HCFA-Based Alkali-Activated Composites Modified with Nano-Silica.

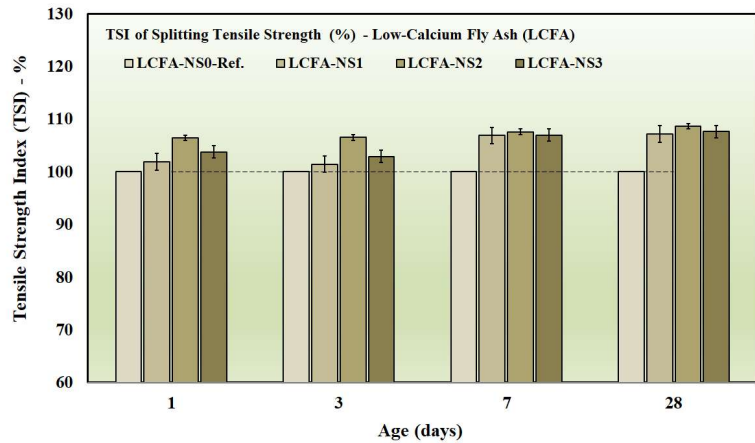


Figure 7. Tensile Strength Index (TSI) of LCFA-Based Alkali-Activated Composites Modified with Nano-Silica.

It was found that the values of TSI at 1 day were higher than the reference value (around 102 percent in NS1 and around 108 percent in NS2) indicating faster tensile strength at an early age.

At NS3, there was a small but significant decrease of approximately 104%, which indicates the development of diminishing returns to increasing nano-silica contents.

The same pattern was observed at 3 days, with NS2 giving the highest TSI values of about 108%, with 103% and 105% by NS1 and NS3, respectively.

The improvement was more pronounced at later ages at 7 days; TSI values at 7 days in NS2 were approximately 109% and at 28 days, TSI values in NS2 were approximately 110%.

Although NS3 mixtures retained better tensile properties compared to the reference, they had lower values of TSI compared to those of NS2 at all ages. Such a pattern indicates that excessive nano-silica levels might be restrictive to the tensile strength improvement in comparison to the optimum intermediate level.

The same trend of gradual improvement was found in LCFA-based mixtures (Figure 7). The value of TSI at 1 day was slightly more, approximately 102% in NS1 and 106% in NS2, which is an indication of slower reaction kinetics using low-calcium systems.

At 3 days, NS2 once again showed the most notable improvement with a TSI value of around 106% with slightly lower values in NS3. LCFA-based mixtures of NS2 obtained LCFA 7 and 28 days gave TSI values of about 107% and 108%, which corroborates the long-term time-dependent contribution of nano-silica to tensile strength development, whereas NS3 maintained lower TSI values than NS2.

The TSI findings provide evidence that incorporation of nano-silica produces noticeable tensile performance improvement to both the HCFA- and LCFA-based systems, with an optimum nano-silica dosage of about 2 wt% (NS2). The high tensile performance peak over the constant NS2 indicates the positive effect of nano-silica in fine-tuning the matrix microstructure, interfacial bonding, and the efficiency of crack-bridging. Higher dosage seems to be accompanied by higher surface area and particle-aggregation requirements, which limit additional tensile performance improvement, as with the compressive development trends.

3.2.3. Flexural Strength Performance Index (FSI)

Flexural performance of the nano-silica-modified alkali-activated composites was evaluated by Flexural Strength Index (FSI), which is shown in Figures 8 and 9, respectively, in HCFA-based and LCFA-based systems.

As with compressive and tensile indices, all FSI values were calculated concerning the reference mixtures without nano-silica (NS0) (FSI = 100%).

In the case of HCFA-based mixtures (Figure 8), the flexural strength of the mixtures was systematically increased with the addition of nano-silica at all curing ages.

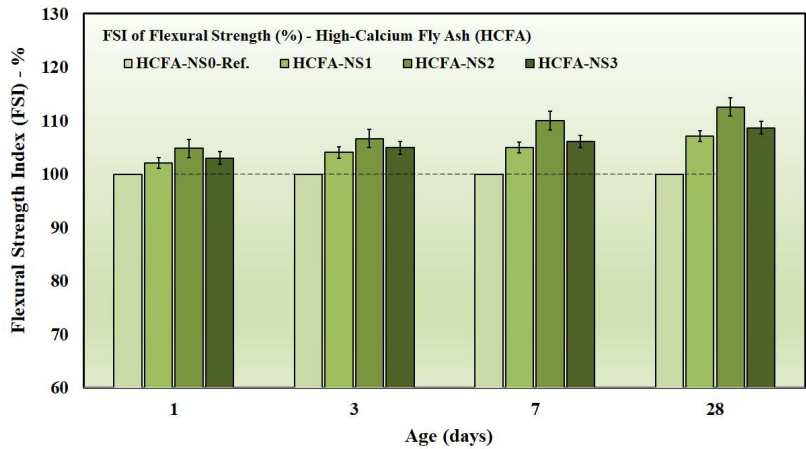


Figure 8. Flexural Strength Index (FSI) of HCFA-Based Alkali-Activated Composites Modified with Nano-Silica.

At 1 day, the FSI values were greater than the reference value and were about 102% under NS1 and about 105% under NS2, and at the same time, the values of NS3 were slightly lower at about 103%.

The improvement was more noticeable at 3 days, as NS2 had an FSI value of about 107% versus 104% in NS1 and 105% in NS3.

A further improvement in NS2 was also demonstrated at 7 days with an FSI value of approximately 110%, and at 7 days, NS3 was also lower with an FSI of about 106 percent. At 28 days, HCFA mixtures with NS2 recorded the highest FSI values of around 112%, which confirms the ongoing role of nano-silica in the development of flexural resistance.

With LCFA-based mixtures (Figure 9), the strength enhancement trend in flexural strength was moderate but still followed the same trend.

On day 1, the FSI values increased by a few percent (102% NS1, 104% NS2) because the low-calcium systems accreted their structure more slowly.

At 3 days, the highest FSI values were seen in NS2 at a range of about 106%, although the improvement in NS3 was slightly less.

The FSI was noticeably increased in 7 and 28 days by LCFA mixtures with NS2 and yielded a more stable but less pronounced improvement as compared to HCFA systems, 108% and 110%, respectively.

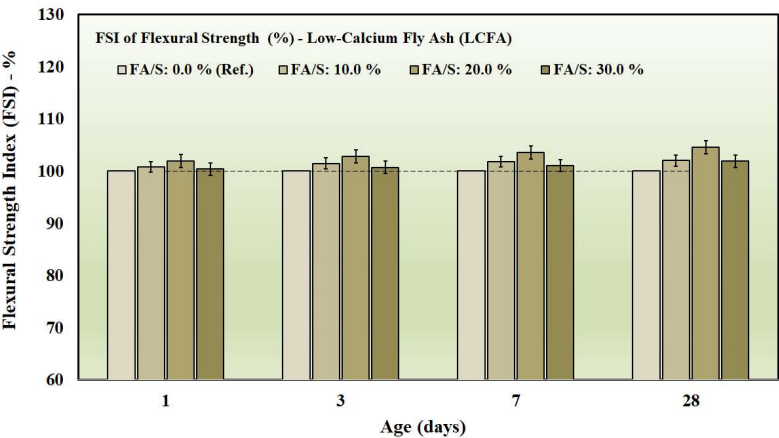


Figure 9. Flexural Strength Index (FSI) of HCFA-Based Alkali-Activated Composites Modified with Nano-Silica.

The FSI measurements indicate that the flexural performance of both an HCFA- and LCFA-based alkali-activated composite is improved by incorporating nano-silica, and the optimal performance is always recorded at around 2 wt% nano-silica.

This enhanced flexural response at a range of intermediate nano-silica dosages can be explained by the increased continuity of the matrix, finer pore structure, and increased stress transfer across microcracks. High nano-silica concentrations, though, seem to have a restrictive effect on additional flexural gains because of dispersion inefficiencies and surface area demand, which were observed to follow similar trends in compressive and tensile performance development.

3.2.4. Integrated Multi-Index Performance Assessment

The multi-index evaluation is an integrated assessment of the mechanical performance of nano-silica modified alkali-activated composites through correlation of compressive, tensile, and flexural strength responses through a single performance framework. Instead of focusing only on compressive strength, the joint analysis of the Strength Activity Index (SAI), Tensile Strength Index (TSI), and Flexural Strength

Index (FSI) was applied to make a more reliable evaluation of the binder synergy, stress transfer efficiency, and redistribution of loads induced by the introduction of nano-silica.

The compressive, tensile, and flexural performance trends are generally in line with the prior research carried out on the alkali-activated slag-fly ash systems, where moderate levels of supplementary aluminosilicate materials are reportedly found to increase the homogeneity and mechanical efficiency of the matrix, with higher rates found to substitute the reactive phases negatively on the strength development process [7,15,17,19]. Consistent with the previous results on fly ash reactivity and calcium effect [4,10,22], the current experiment findings reveal that the two HCFA-based and the LCFA-based systems exhibit an optimum mechanical capability at the middle nano-silica concentrations.

From a mechanical performance perspective, the current findings reveal that compressive strength is not sufficient to reflect the important changes in tensile and flexural behavior, especially in nano-silica modified alkali-activated materials. This finding supports previous results that, to estimate cracking resistance, redistribution of stress, and efficiency of load transfer in advanced alkali-activated composites, overall mechanical assessment is necessary [2,30,32].

In this regard, the suggested multi-index performance framework adds further analytical value in this aspect by allowing a more consistent and performance-based explanation of binder synergy and nano-scale effects of modification.

Therefore, to support this performance-based interpretation, the relationships between compressive strength development and the associated tensile and flexural performance were quantitatively determined using correlation analysis. Figures 10 and 11 show the SAI-TSI of the HCFA-based and LCFA-based systems, respectively. In the case of HCFA mixtures, a good linear relationship was achieved, with a coefficient of determination of $R^2 = 0.91$ (Fig. 10).

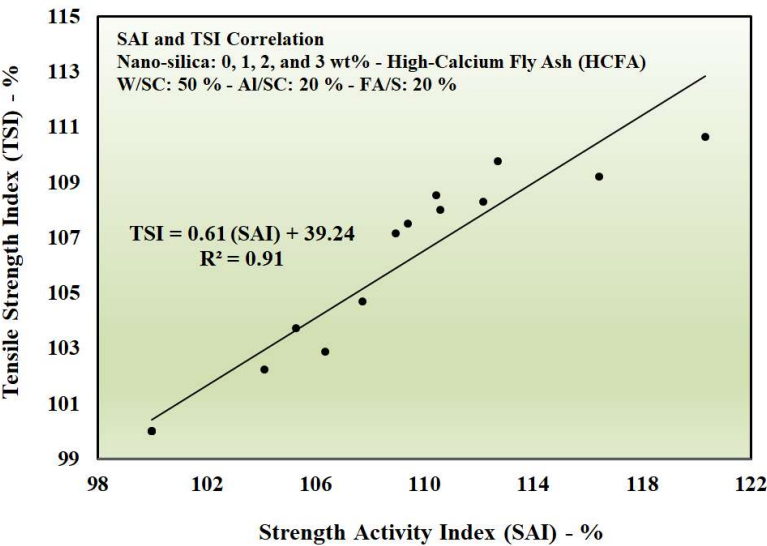


Figure 10. SAI-TSI correlation for HCFA-based nano-silica-modified composites.

It means that the increases in compressive strength are successfully converted into tensile resistance but with certain dispersion explained by the calcium-rich reaction environment and localized cracking behavior. Conversely, LCFA mixtures had a somewhat more consistent correlation, with a larger value of $R^2 = 0.94$ (Fig. 11), indicating a more homogeneous stress transfer under the control of aluminosilicate-dominated reaction products. The same pattern was revealed in flexural performance.

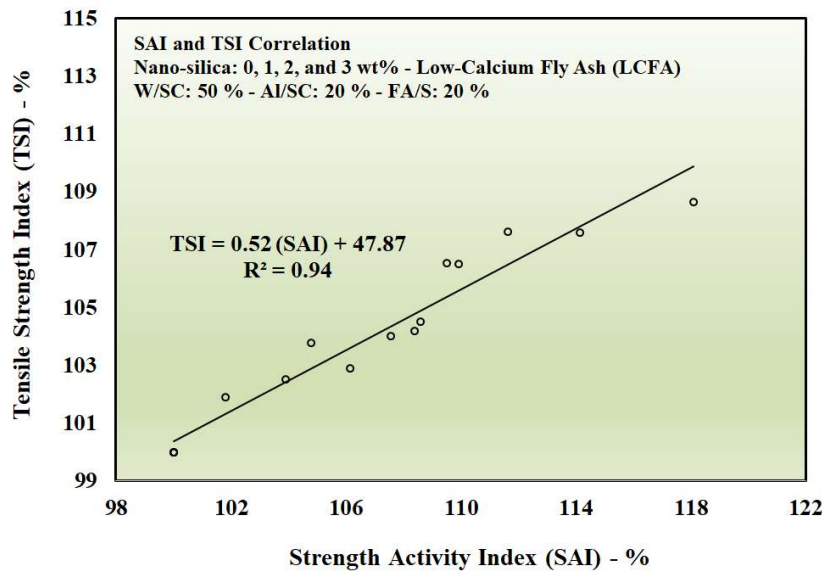


Figure 11. SAI-TSI correlation for LCFA-based nano-silica-modified composites.

Figures 12 and 13 demonstrate that mixtures of HCFA provided a good relationship between SAI and FSI with $R^2 = 0.93$ (Fig. 12), which verified a high level of coupling between compressive strength improvement and flexural resistance in calcium-rich mixtures.

Even greater correlation was observed with LCFA mixtures, with a notably high $R^2 = 0.95$ (Fig. 13), indicating that compressive strength is translated into flexural performance coherently because of geopolymeric gel formation, which was gradual and homogeneous. In both fly ash systems, the multi-index analysis has always determined mixtures with intermediate nano-silica contents (NS2 level) to exhibit the most balanced mechanical behavior, with corresponding improvements in SAI, TSI and FSI. This action proves that nano-silica is not just effective in increasing compressive strength but also crack-bridging capacity and flexural load resistance, which contribute to the structural efficiency of the composite.

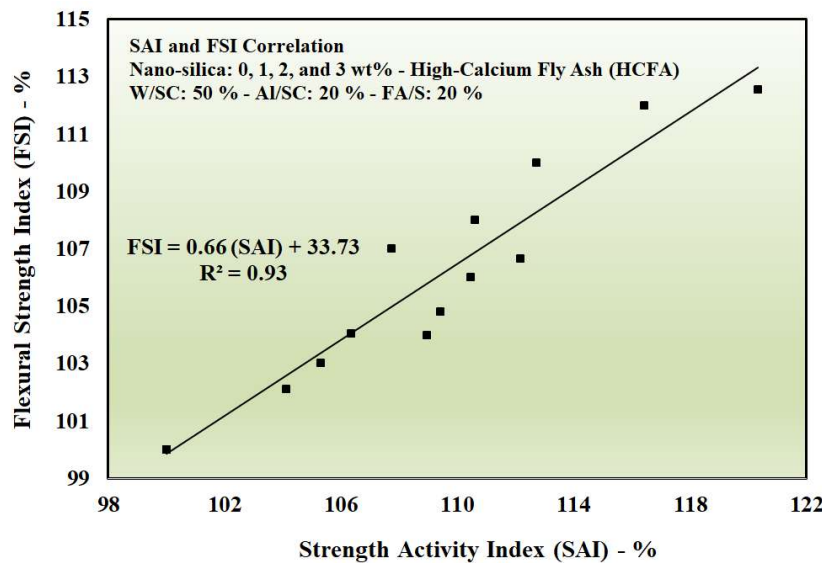


Figure 12. SAI-FSI correlation for HCFA-based nano-silica-modified composites.

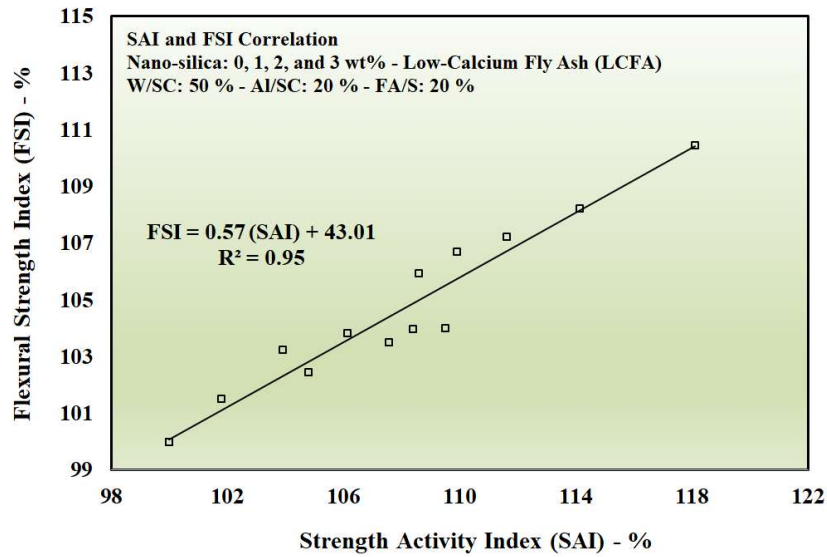


Figure 13. SAI-FSI correlation for LCFA-based nano-silica-modified composites.

The SAI-TSI-FSI correlation analysis represents an empirical observation of trends in performance, but not a predictive relationship, which means that compressive strength is not a reliable predictor of tensile and flexural performance, especially in alkali-activated systems whose reactivity is governed by differing calcium and alumino-silicate. The specific correlation tendencies of HCFA and LCFA mixtures indicate the determining role of the binder chemistry and nano-silica induced refinement of the microstructure in regulating mechanical synergy. The results confirm the multi-index framework proposed as an effective, performance-based methodology to evaluate nano-silica-modified alkali-activated composites to be used in the context of infrastructure applications.

Although the integrated multi-index evaluation illustrates the internal mechanical synergy and performance interdependence of nano-silica-modified alkali-activated composites, one would need to consider the implications of such laboratory-scale data on the practical engineering application in terms of infrastructure-based applications.

3.2.5. Performance Implications for Infrastructure Applications

As observed, the combined multi-index evaluation focuses on the functionality of nano-silica-modified alkali-activated composites to infrastructure-based composites, where fresh-state constructability and multi-directional mechanical strength are required. It has been established through a combination of the flow-based indices (IFI, and FRI) and strength-based indices (SAI, TSI, and FSI) that enhancement of compressive strength alone cannot make plants in an ideal material performance, but that controlled synergy between binder chemistry, nano-scale modification, and reaction kinetics is required, as has been shown in recent performance-based infrastructure material studies [34,35]. This interaction between fresh-state behavior, mechanical indices, and infrastructure performance requirements is schematically shown in Figure 14.

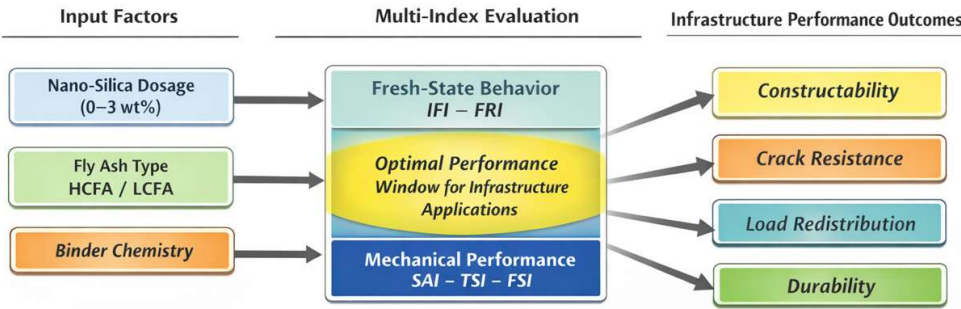


Figure 14. Schematic of multi-index performance implications for infrastructure applications of alkali-activated composites.

In the case of an intermediate nano-silica dosage of approximately 2 wt% (NS2), both HCFA-based and LCFA-based systems had a favorable balance of maintaining workability and forming compressive strength, tensile resistance, and flexural capacity. Balanced performance applies particularly to those infrastructure elements under more complicated loading circumstances, such as cracking, bending, and stress redistribution, such as pavements, precast structural members, and repair overlays. The tensile and flexural performance achieved at this dosage is high, suggesting that there is high efficacy of crack-bridging and stress transmission, most critical to infrastructure design focused on durability [35,36]. In addition to this, the difference between the HCFA and LCFA systems provides feasible tips on how mixtures should be customized according to the usage requirements. HCFA-based composites showed higher early age strength development as compared to LCFA-based systems, albeit with poorer fresh-state stability, and a slower strength development, which is more constructable and long-term stable. These contrary steps may be associated with the recent findings, which claim that the infrastructure materials should be selected based on the performance criteria and not on the optimization of a single parameter [34,37]. Thus, the findings indicate that the proposed multi-index system offers a rational and performance-driven basis of the process of translating the laboratory-scale maximization of the materials to the actual infrastructure implementation. The framework enables the establishment of resource-efficient alkali-activated composites that can fulfil the mechanical, constructability, and sustainability demands of modern infrastructure systems, based on fresh-state behavior, mechanical synergy, and nano-scale modification mechanisms [35-37].

3.3. Mineralogical and Microstructural Interpretation of Performance Trends

3.3.1. XRD-Based Mineralogical Interpretation

The XRD patterns shown in Figure 15 provide a fundamental indication of the mineralogical properties of the constituent materials, which are utilized to interpret the trends of fresh-state and mechanical performance of the nano-silica-modified alkali-activated composites.

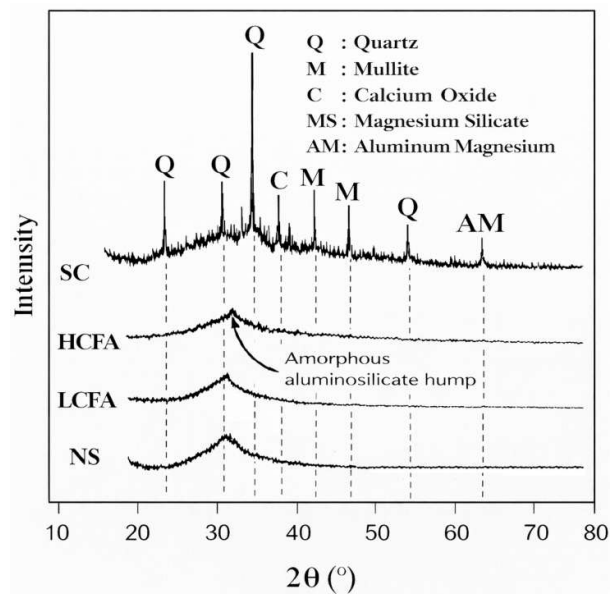


Figure 15. XRD Patterns of slag cement (SC), high-calcium fly ash (HCFA), low-calcium fly ash (LCFA), and nano-silica (NS).

The assemblage of crystalline phases and amorphous content of slag cement (SC), high-calcium fly ash (HCFA), low-calcium fly ash (LCFA) and nano-silica (NS) directly determine the reaction kinetics, binder synergy and load-transfer efficiency of the composite matrix.

The XRD pattern of slag cement represents pronounced crystalline peaks with the main participants as quartz and calcium-based phase, which are superimposed over a partially amorphous background.

This mineralogical reagent is the reason the rapid early-age response in HCFA-based systems is achieved, whereby the presence of calcium enhances faster dissolution-precipitation reactions during alkaline activation. This action is in line with the stiffer losses in IFI and FRI with progress in dosage of HCFA mixtures of nano-silica, as early reaction products form faster, leading to increased particle interlocking and lowered retention in workability.

On the contrary, both HCFA and LCFA show a wide amorphous aluminosilicate hump centered in the 2θ range of about $25\text{--}35^\circ$, which is more intense and broader in LCFA. This increased amorphous aluminosilicate is responsible for slower, more controlled reaction kinetics and the better flow retention behavior (FRI) that is always witnessed in LCFA based mixtures. Lower LCFA early-age reactivity reverses the premature stiffening and therefore increases the fresh-state stability relative to the systems based on HCFA.

Nano-silica has a predominant hump of amorphous type and little crystalline peaks indicating that it is highly reactive and has a glassy structure. This mineralogical property is the basis of the mentioned above improvement of mechanical indices at an intermediate dose of nano-silica of about 2 wt% (NS2). The amorphous silica offered by NS helps to enhance the way the particles are packed, the speed at which the reaction products are nucleated, as well as the optimization of the interfacial transition zones. The same effects manifest in the parallel increases in SAI, TSI, and FSI in each of the two systems based on the incorporation of nano-silica (HCFA- and LCFA-based).

The mineralogical description is also supported by correlation analysis of SAI-TSI and SAI-FSI (Figures 10-13). Close linear correlations with large coefficients of determination (R^2 of approximately 0.91-0.95) are a sign that the development of compressive strength is closely associated with tensile and flexural performance when the balance of mineralogical reactivity is achieved. In mixtures made with LCFA, these correlations are a little more coherent, which can be explained by the fact that it is the reaction products that are based on aluminosilicate, so it facilitates more coherent transfer of stress and crack-bridging.

On the other hand, the HCFA systems have a larger scatter, indicating the effects of calcium-enriched crystalline phases, which are characterized by fast strengthening but add heterogeneity at a microscopic level of development.

At increased dosages of nano-silica (NS3), the mineralogical balance is less desirable. Amorphous silica in excess causes demand on surface area and insufficient binder packing resulting in marginal mechanical gains and noticeable loss in fresh-state performance. This action proves that it is mineralogical synergy, or complete amorphous content, and not absolute amorphous content, that governs performance of composites.

Altogether, the mineralogical support of the trends of the performance of IFI-FRI and SAI-TSI-FSI tests is presented by Figure 15. The optimal performance of the middle doses of nano-silica is an indication of a balanced reaction among the availability of calcium, amorphous aluminosilicate content and the nano-scale reactivity.

The findings verify that the multi-index framework provided is a mineralogically based, performance-based approach to create resource-efficient alkali-activated composites to be utilized in infrastructure applications.

3.3.2. SEM-Based Microstructural Interpretation

Scanning electron microscopy (SEM) was used to investigate the microstructural features of the alkali-activated composites to justify the mineralogical interpretation as well as elucidate the fresh-state and mechanical performance tendencies as discussed previously. SEM micrographs of an HCFA- and LCFA-based reference system (nano-silica free) and the systems with 2 wt% nano-silica at 28 days is shown in Figure 15 in order to assess the effect of calcium content and nano-scale modification on the morphology of the matrix, particle interaction, and microstructural homogeneity.

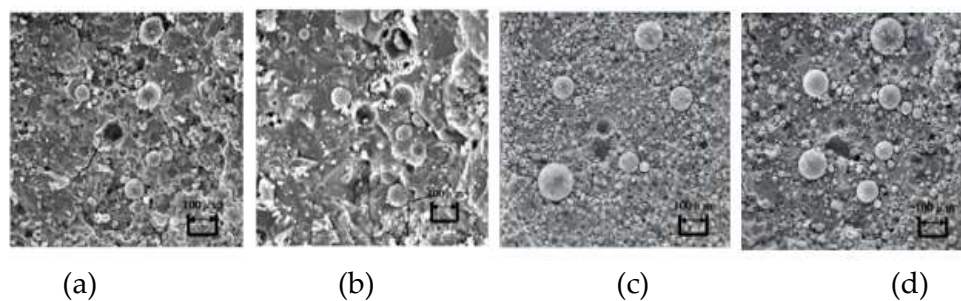


Figure 15. SEM micrographs of alkali-activated composites: (a) HCFA-NS0, (b) LCFA-NS0, (c) HCFA-NS2, and (d) LCFA-NS2.

In reference mixtures, that is, mixtures that do not contain nano-silica (NS0), the microstructure differs with the fly ash chemistry. In the HCFA-based reference system (Figure 15a), the microstructure is characterized by partially reacting fly ash spheres and embedded in a calcium-based reaction matrix. Even though a relatively thick binder phase is formed, distinctive heterogeneities, micro voids, and localized microcracks are observed, particularly with regard to unreacted or partially reacted particles. These properties indicate a rapid dissolution precipitation reaction aided by heavy calcium contents, which leads to premature strength increase and formation of disproportionate distribution of stress and localized regions of reduced mechanical integrity in the matrix. These microstructural anomalies are also in line with an increased dispersion of tensile and flexural performance of HCFA mixtures.

On the other hand, the reference system of LCFA (Figure 15b) shows a rather homogenous and continuous structure of silicate aluminosilicate gels. SEM images reveal less product reaction microcracks, smoother product, and particle connectivity. The LCFA systems are low in calcium, which leads to slower and more controlled geopolymerization, less premature precipitation, and slower formation of binding steps. It is this microstructural homogeneity that contributes to the

enhanced flow retention and more homogeneous mechanical behavior of LCFA-based mixtures even without nano-silica.

Addition of nano-silica at 2 wt% (NS2) causes both systems to be refined to a remarkable degree. In the case of the HCFA-based system, which contains nano-silica (Figure 15c), all SEM images depict a general densification of the matrix, where micro voids are filled with nano-scale reaction products. Nano-silica is an active filler and nucleation agent that is capable of speeding up gel formation and interparticle bonding. Nevertheless, localized heterogeneities and microcracks can be seen, especially in the calcium-rich regions, which suggest that the combined presence of large quantities of calcium and nano-silica can enhance local reactions and stress concentration. This finding compares with enhanced compressive behavior but comparatively dispersed tensile and flexural response in the systems of HCFA.

The LCFA-based system with the additive of nano-silica (NS2) (Figure 15d) has a comparatively refined and uniform microstructure. SEM micrographs indicate that reaction products are well-distributed with lower pore connectivity and are better packed in. The aluminosilicate-based gel creates a continuous matrix that plays a good role in bonding fly ash particles and reducing microstructural defects. The reduced reaction kinetics of LCFA enable the nano-silica to be more favorably involved in the nucleation and microstructural refining without precipitating early stiffening. This improved morphology is aligned with the increase in flow stability and the enhanced correlation among compressive, tensile, and flexural performance indices of LCFA-composites.

Thus, when comparing reference and nano-silica-modified systems, it can be concluded that the nano-silica can substantially enhance the densification of the matrices, interfacial transition zones, and microstructural coherence when introduced at an optimal dosage.

The results of the SEM observations confirm that performance growth is burdened by harmonious mineralogical interaction rather than the excessive nano-scale supplementation. Such microstructural observations offer direct physical support for the multi-index performance framework and the reasons behind the optimum mechanical and fresh-state performance of intermediate doses of nano-silica when applied to alkali-activated composites to be used in sustainable infrastructure applications.

3.3.3. Integrated Mineralogical–Microstructural Interpretation

The combined explanation of mineralogical and microstructural features provides an explanation of the observed fresh-state and mechanical performance tendencies of the nano-silica-modified alkali-activated composites in a unitary manner.

Where XRD analysis elucidates the phase constituent, amorphous content, and dominating reaction mechanisms that govern the systems founded on calcium- and silicate alumina, SEM observations represent the actual findings of the microstructural organization, particle packing, and interface properties. The enhanced amorphous aluminosilicate hump localization is seen in the LCFA-based systems in the XRD sense, which is translated into SEM in the form of a more homogenized, compact, and well-linked structure to enhance the capacity of holding flow and transferring stress in an efficient way.

On the other hand, the localized densification, high microstructural heterogeneity, and vulnerability to premature stiffening, which are seen in the SEM micrographs, are related to the presence of localized densification, crystalline phases rich in calcium, as revealed by XRD.

At the intermediate dosage of nano-silica (2 wt%), both techniques consistently demonstrate that balanced mineralogical synergy rather than the highest proportion of amorphous content regulates the process of microstructural refinement and overall performance enhancement.

This integrative mineralogical-microstructural analysis is conducive to the validity of the proposed multi-index framework as a viable, performance-based approach methodology to the creation of resource-efficient alkali-activated composites with specific prospects to utilize them in the context of sustainable infrastructural applications.

4. Conclusions

This study revealed that the proposed multi-index performance is a comprehensive and systematic framework thorough foundation measurements the synergies of the nano-silica incorporation into the resource-efficient alkali-activated composites. The proposed framework facilitates the use of performance-based assessment, integrating the fresh-state flow indicators and strength-based mechanical indicators, thereby providing material assessment other than the conventional compressive strength-based techniques.

The performance of fresh-state systems based on the Initial Flow Index (IFI) and the Flow Retention Index (FRI) gradually declined as the dosage of the nano-silica increased in both HCFA-based systems and LCFA-based systems. This behavior is attributed to the increase in the surface area and the rate of reaction in the initial stage. Nevertheless, LCFA systems never showed limited flow retention and fresh-state stability as compared to HCFA systems because of the influence of low calcium concentrations and low reaction rates in inhibiting quick stiffening.

An intermediate nano-silica dosage of about 2 wt% was optimum, yielding the enhanced compressive, tensile, and flexural response with both fly ash systems. At this dosage concentration, the Strength Activity Index (SAI), Tensile Strength Index (TSI) and Flexural Strength Index (FSI) showed improved development of strength. An increase in the nano-silica content led to a decrease in the performance gains, and the results illustrated the need to optimize dosage to prevent the negative effects of excessive fineness and particle agglomeration.

Correlation analyses of Strength Activity Index (SAI) and Tensile and Flexural Strength Index (TSI and FSI) revealed that it is not possible to reliably estimate tensile and flexural performance based only on compressive performance, particularly in nano-silica, fly ash, and alkali-activated composites, which react in different ways. Such findings suggest that mechanical synergy as well as load-transfer properties of alkali-activated composites require a multi-index evaluation.

Mineralogical and microstructural analyses (XRD and SEM) revealed that the effectiveness of the reaction, as well as the densification of the matrix and stress transfer, is determined by the interaction of amorphous aluminosilicate components, the availability of calcium, and the reactivity of nano-silica. An intermediate dosage of nano-silica (2 wt) was favorable to balance the nucleation and refinement of the microstructure, but a high dosage favored agglomeration, non-homogeneous nucleation, and low performance. These values agree with the experimentally determined patterns of fresh-state behavior and mechanical performance.

Collectively, the results endorse that the proposed multi-index framework is an effective methodological approach in the optimization of nano-silica dosage and directing the design of resource-efficient alkali-activated composites, which have proven to be relevant in sustainable infrastructure applications.

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