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TOWARDS CARBON-NEUTRAL CEMENT: MULTISCALE STRUCTURAL, CHEMICAL, AND PERFORMANCE EVOLUTION OF ULTRA-HIGH GGBS BLENDED SYSTEMS FOR NEXT-GENERATION LOW-CARBON CONCRETE

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Abstract

The urgent global emphasis on decarbonizing the construction industry has intensified the search for low-carbon alternatives to traditional Portland cement, which remains one of the largest contributors to anthropogenic CO_2 emissions. Ground granulated blast-furnace slag (GGBS) has emerged as a highly effective supplementary cementitious material (SCM) capable of significantly lowering clinker demand while enhancing long-term durability. This study provides a comprehensive multiscale investigation into the structural, chemical, and performance evolution of ultra-high GGBS blended systems, defined as cementitious formulations incorporating 60–85% slag replacement. The research aims to elucidate the underlying hydration mechanisms, microstructural transformations, and engineering performance characteristics that govern the feasibility of deploying such high-volume slag blends for next-generation low-carbon concrete. Advanced characterization techniques, including SEM, XRD, TGA, FTIR, and MIP, were employed to analyze the hydration process and microstructural development across different curing ages. The results reveal a distinct shift from clinker-dominated hydration to slag-driven pozzolanic reactivity, leading to progressive formation of secondary C–S–H gels with lower Ca/Si ratios. These gels contribute to refined pore structures, reduced permeability, and enhanced long-term densification. Chemical analyses further confirm substantial portlandite consumption and the evolution of aluminosilicate hydrates, which improve resistance against sulfate attack, chloride penetration, and alkali–silica *reactivity. Although early-age hydration is slower due to reduced clinker availability, mechanical performance significantly improves after extended curing, with optimized mixes achieving strengths comparable to or exceeding conventional OPC systems. Performance assessments demonstrate that the adoption of appropriate activators, curing protocols, and particle size optimization can effectively mitigate limitations such as delayed setting, low early strength, and reactivity variability. The study identifies key technical challenges such as sensitivity to temperature, dependence on alkaline activation, and industrial-scale variability in slag quality that must be addressed to ensure reliable field application. Nonetheless, the long-term performance benefits, including enhanced durability, reduced thermal cracking potential, and lower embodied carbon, firmly position ultra-high GGBS blends as a viable pathway toward carbon-neutral cement technologies. Overall, the findings provide critical scientific insights and practical guidance for the development of ultra-high slag blended cements. By establishing strong correlations between microstructural evolution, chemical transformation, and macro-scale performance, this work contributes to advancing sustainable cement innovation and supports global strategies aimed at reducing the environmental footprint of concrete construction.*

INTRODUCTION

The global construction industry is entering a transformative era shaped by the urgent need to lower carbon emissions and align infrastructure development with international climate commitments. Ordinary Portland cement, despite its indispensable role in structural engineering and urban development, is responsible for a substantial share of global anthropogenic CO_2 emissions, primarily due to the calcination of limestone and the high-temperature kiln operations required for clinker production. As nations expand infrastructure to support rapidly urbanizing populations, the demand for cement continues to rise, creating a severe conflict between development needs and environmental sustainability. This challenge has intensified the search for advanced low-carbon binders that can reduce the environmental burden of construction without compromising mechanical performance, durability, or long-term serviceability [1]. Among these materials, ground granulated blast-furnace slag has attracted significant attention as a highly effective supplementary cementitious material capable of reducing clinker consumption while offering distinct microstructural and durability benefits. While moderate slag replacement levels have been widely adopted, recent decarbonization goals have driven interest in ultra-high GGBS systems, defined in this study as mixtures containing approximately 60 to 85 percent slag. These high-volume slag binders represent a next-generation approach to sustainable cement technology, offering the potential for dramatic reductions in embodied carbon due to minimized clinker demand and the beneficial reuse of metallurgical by-products [2]. Their long-term engineering performance, characterized by refined pore structures, reduced permeability, improved resistance to aggressive chemicals, and enhanced durability, makes them promising candidates for infrastructure exposed to harsh service environments. Nevertheless, these systems present complex scientific challenges that require a deeper mechanistic understanding. When clinker content is significantly reduced, hydration transitions from clinker-controlled reactions to slag-driven pozzolanic or latent-hydraulic processes, resulting in slower early-age strength development, delayed setting, and increased sensitivity to curing regimes, alkalinity, and temperature. These early-age limitations have historically restricted the widespread adoption of ultra-high slag binders, and overcoming them requires comprehensive investigation across chemical, microstructural, and macroscopic scales. Recent advances in multiscale characterization have opened opportunities to examine these systems with unprecedented clarity. Techniques such as scanning electron microscopy, X-ray diffraction, thermogravimetric analysis, mercury intrusion porosimetry, and Fourier transform infrared spectroscopy have made it possible to analyze the evolution of hydration products, the transformation of pore networks, and the progressive densification of the matrix across different curing ages. These methods reveal that ultra-high slag systems produce secondary C–S–H and C–A–S–H gels with lower Ca/Si ratios and higher alumina incorporation, resulting in more cross-linked structures that enhance long-term strength and durability [3]. The substantial consumption of portlandite and the development of aluminosilicate hydrates contribute to reduced chemical vulnerability, particularly regarding sulfate attack, chloride penetration, and alkali–silica reactivity. As hydration progresses, the pore network becomes increasingly refined, improving resistance to transport-based deterioration mechanisms and translating to superior performance in marine, coastal, and underground applications. Despite these advantages, industrial-scale implementation is hindered by variability in slag chemistry, fineness, and reactivity, underscoring the need for standardized evaluation procedures and robust design guidelines. To contextualize the evolution of these materials, the following table presents a comparative overview of OPC, moderate GGBS blends, and ultra-high GGBS systems.

Table 1: Comparative Overview of OPC, Moderate GGBS Blends, and Ultra-High GGBS Systems

Parameter	**OPC (0% GGBS)**	**Moderate GGBS (20–50%)**	**Ultra-High GGBS (60–85%)**
CO_2 Emissions	Very High	Medium	Very Low
Hydration Mechanism	Clinker-dominated	Mixed clinker & slag	Slag-dominated
Early Strength	High	Moderate	Low
Long-Term Strength	Stable	High	High or Very High
Heat of Hydration	High	Reduced	Very Low
Pore Structure	Coarse	Refined	Very Dense
Durability	Moderate	High	Very High
Chloride Resistance	Low	High	Very High
Carbonation Resistance	High	Moderate	Moderate to Low
Workability	Moderate	Improved	Sensitive to admixtures
Variability	Low	Medium	High (slag quality dependent)

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A conceptual illustration of the multiscale behavior of ultra-high slag systems is shown in Figure 1. This schematic highlights the interconnected processes that govern the evolution of these binders from chemical reactions at the nanoscale to macroscopic performance characteristics relevant to structural engineering.

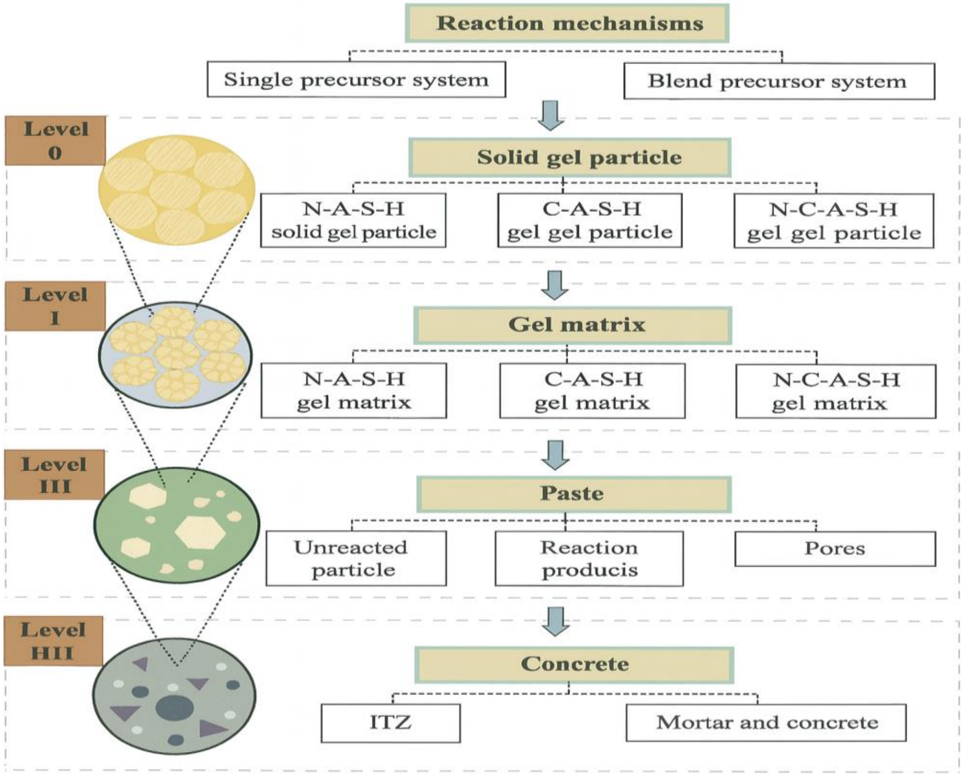

Figure 1: Multiscale conceptual framework illustrating slag reactivity.

The significance of ultra-high slag binders extends beyond their microstructural and mechanical attributes; they align closely with global sustainability frameworks and the broader shift toward carbon-neutral construction. By drastically reducing clinker demand, these materials address the dominant source of CO_2 emissions in cement production. Their compatibility with industrial by-product utilization supports circular economy models by transforming metallurgical waste into valuable construction materials. Their lower heat of hydration reduces thermal cracking risks, particularly in massive structural elements such as dams, foundations, and bridge decks. As durability improves, the lifetime environmental impact of structures decreases, as fewer repairs, replacements, or interventions are required. All these advantages converge to make ultra-high slag binders highly relevant to national and international climate strategies, particularly in regions with aggressive environmental conditions or rapid infrastructure expansion [4]. Despite these promising attributes, knowledge gaps persist regarding their hydration kinetics under variable field conditions, the stability of hydration products under combined chemical and physical stressors, and the long-term evolution of their pore networks. Many durability models remain calibrated for OPC-rich systems, limiting their predictive accuracy for slag-dominant binders. Additional challenges emerge from the industrial variability of slag fineness and chemistry, which can significantly influence reactivity. The absence of universally accepted standards for ultra-high slag cement further restricts adoption, as design codes remain based on traditional OPC–SCM blends. These limitations highlight the need for a mechanistic, experimentally validated understanding capable of linking nanoscale chemical processes to real-world engineering outcomes. This study therefore positions itself within a framework that seeks to advance both scientific understanding and practical implementation of ultra-high GGBS systems. By undertaking a detailed multiscale analysis using advanced characterization techniques across different curing periods, the research aims to establish clear correlations between hydration reactions, microstructural development, pore-network transformation, and mechanical and durability performance. The work provides insights not only into fundamental material behavior but also into the optimization strategies necessary for overcoming early-age limitations, such as the use of chemical activators, curing enhancements, particle size optimization, and hybrid cementitious systems. Through this integrated perspective, the study contributes to the development of scientifically sound guidelines that can support the safe, efficient, and large-scale deployment of ultra-high slag binders in modern infrastructure [5]. Ultimately, the growing pressure to reduce carbon emissions from cement and concrete demands innovative materials that deliver performance parity or superiority relative to conventional systems while drastically lowering environmental impact. Ultra-high GGBS blended cements embody this vision by offering a pathway toward carbon-neutral construction materials rooted in established, globally available industrial outputs. Their complex multiscale behavior, however, necessitates continued investigation to fully unlock their potential and establish reliable design frameworks suitable for industrial adoption. By providing a cohesive synthesis of chemical, structural, and performance evolution, this study seeks to advance sustainable cement innovation and reinforce the role of high-volume slag binders as foundational materials for next-generation low-carbon concrete.

1- Hydration Chemistry of Slag-Blended Cementitious Systems:

The hydration chemistry of slag-blended cementitious systems reflects a progressive transformation of the classical Portland cement hydration paradigm, driven by the complex interactions between clinker dissolution, slag activation, pore solution chemistry, and the evolving stability fields of hydration products. Ground granulated blast-furnace slag (GGBS), being a calcium–magnesium–aluminosilicate glass, does not hydrate independently under neutral conditions. Instead, its activation requires a highly alkaline environment capable of breaking its amorphous framework. In blended systems, this alkalinity originates from the dissolution of clinker phases such as C_3S and C_2S, which generate Ca^{2+} and OH^- ions, creating a pore solution with pH typically above 12.5. This chemical environment establishes the foundation for the dissolution–precipitation reactions that become increasingly dominant as slag content rises. In the early stages of hydration, clinker phases hydrate rapidly, supplying portlandite and initiating the formation of high Ca/Si C–S–H gels [6]. However, the presence of slag alters this process immediately by introducing an alternative source of silicate, aluminate, and magnesium species that interact with the pore solution and with clinker-derived hydrates. As slag begins to dissolve, it consumes portlandite through pozzolanic reactions, reducing the Ca^{2+} concentration and lowering the Ca/Si ratio of newly formed hydrates. Consequently, the hydrate assemblage evolves into a mixture dominated by low Ca/Si C–S–H and aluminosilicate C–A–S–H, which possess different chain structures, higher degrees of polymerization, and enhanced chemical stability compared to typical OPC hydration products. The complexity of these reactions becomes more pronounced as slag replacement increases. At moderate content, slag hydrates form alongside clinker hydrates, modifying microstructural development but not fundamentally displacing the OPC hydration mechanism [7]. At ultra-high slag levels, however, clinker-derived products no longer dominate; rather, the system transitions into a chemistry governed primarily by slag dissolution and secondary precipitation constrained by pore solution equilibria. The contrasts across these systems are summarized in Table 2.

Table 2: Hydration Reactions, Dominant Products, and Microstructural Outcomes in Slag-Blended Systems [8].

System Type	Dominant Hydration Reactions	Main Hydration Products	Microstructural Outcomes
OPC (0% GGBS)	Rapid hydration of C_3S/C_2S with abundant CH formation	High Ca/Si C–S–H, CH, AFt	Coarse capillary pores; large CH crystals; rapid early strength
Moderate GGBS (20–50%)	Clinker hydration producing OH^-; progressive slag dissolution; pozzolanic CH consumption	Secondary C–S–H (lower Ca/Si), C–A–S–H, hydrotalcite	Significant pore refinement; reduced interconnected porosity; improved long-term density
Ultra-High GGBS (60–85%)	Slag-driven dissolution–precipitation; limited CH; high dependence on alkalinity and temperature	C–A–S–H rich in Al, Mg–Al LDH phases, low Ca/Si C–S–H	Highly compact gel networks; strong ion binding; exceptional durability; slow early hydration kinetics

The trends summarized in the table underscore the fundamental chemical transition that occurs when slag content is increased. In OPC systems, hydration is primarily a precipitation-controlled process driven by Ca^{2+} supersaturation and rapid CH crystallization. In contrast, high-slag systems become governed by dissolution kinetics and the progressive depletion of CH, which shifts hydrate formation toward aluminosilicate species. This evolution significantly alters microstructural development: instead of coarse CH crystals filling pore spaces, slag-rich systems generate finely structured C–A–S–H gels that distribute uniformly throughout the matrix, reduce pore connectivity, and enhance resistance to transportation of aggressive ions [9]. Figure 2 illustrating the hydration chemistry of slag-blended cementitious systems, showing clinker dissolution, hydroxyl-driven slag activation, portlandite consumption, and the precipitation of low Ca/Si C–S–H and aluminosilicate C–A–S–H gels. The figure should also depict the transition from OPC-dominated hydration to slag-dominated dissolution–precipitation mechanisms as slag content increases, alongside the associated refinement of pore structure and long-term densification.

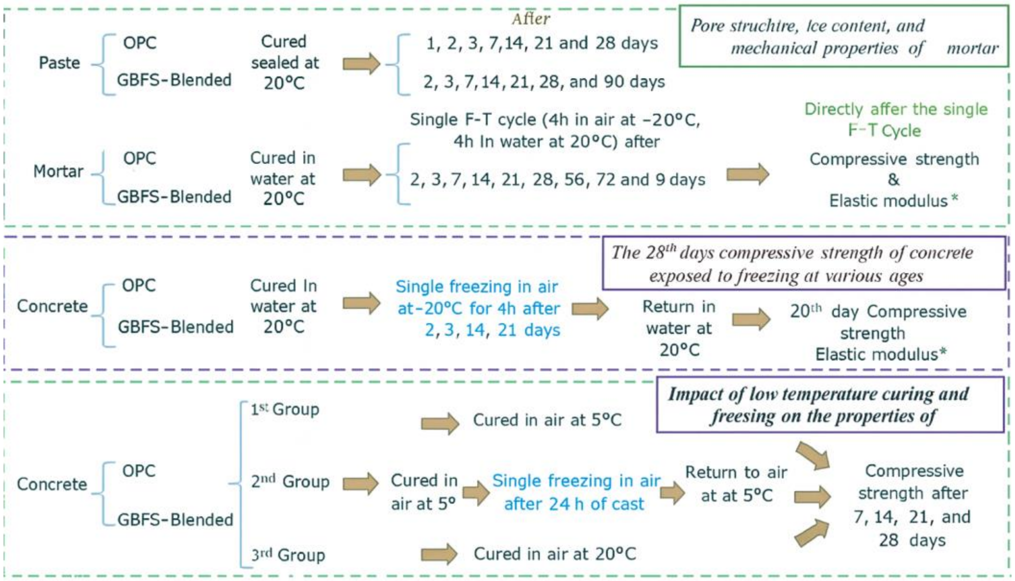

Figure 2: Conceptual schematic illustrating the hydration chemistry of slag-blended cementitious systems

The hydration environment in slag-rich systems becomes progressively more complex as slag dissolution increases. The depletion of CH is one of the most influential changes, as it modifies the chemical equilibria that govern hydrate precipitation. As soon as slag begins to consume portlandite, the pore solution becomes less saturated with Ca^{2+} and more dominated by Na^{+}, K^{+}, Mg^{2+}, and $Al(OH)_4^{-}$ species. This shift suppresses the formation of high Ca/Si C–S–H and promotes the growth of more polymerized hydrates with extended silicate chains and greater incorporation of Al^{3+} into bridging sites. These structural distinctions lead to markedly different gel morphologies, often characterized by higher intrinsic cross-link density, increased binding capacity, and greater resistance to dissolution under aggressive environments. The formation of hydrotalcite-like layered double hydroxides constitutes another key feature of slag hydration chemistry [10]. These phases arise from the interaction of dissolved Mg^{2+} and $Al(OH)_4^{-}$ species and act as highly efficient ion traps, immobilizing chlorides, carbonates, and sulfates within their layered structures. Their presence significantly enhances durability by mitigating the ingress of harmful ions and strengthening chemical resistance. In clinker-dominated systems, by contrast, the absence of sufficient magnesium and aluminum in reactive form limits the formation of such protective phases. The evolution of hydration chemistry also has spatial implications. The interfacial transition zone (ITZ), traditionally considered the weakest region of concrete due to loosely packed hydrates and abundant CH crystals, undergoes significant refinement in slag-rich systems [11]. The consumption of CH reduces the presence of large crystal formations, while the continuous precipitation of fine C–A–S–H fills voids more effectively. This transformation leads to a denser, more uniform ITZ, improving load transfer and reducing microcrack initiation. Temperature effects introduce further complexity. Because slag dissolution is highly temperature-dependent, low curing temperatures suppress slag activation and prolong dormant periods, whereas elevated temperatures greatly accelerate the breakdown of the slag network, enabling faster precipitation of aluminosilicate hydrates. These sensitivities require meticulous attention in practical applications of ultra-high slag binders, where delayed strength development may otherwise pose engineering challenges. Overall, the hydration chemistry of slag-blended cementitious systems reveals an intricate transition from calcium-rich OPC hydration toward a regime controlled by aluminosilicate dissolution and precipitation. This transition governs the balance between early-age limitations and long-term benefits [12]. While the lower rate of reaction in slag-rich systems delays early strength gain, the continuous formation of C–A–S–H and hydrotalcite yields a microstructure of outstanding density, stability, and durability. Such characteristics make ultra-high GGBS binders highly compelling for next-generation low-carbon concrete solutions, provided that their hydration behavior is comprehensively understood and adequately controlled.

2- Microstructural Evolution in Ultra-High GGBS Blended Systems:

The microstructural evolution of ultra-high GGBS blended systems represents one of the most critical determinants of their mechanical performance, durability, and long-term serviceability. Unlike ordinary Portland cement, where rapid early hydration drives the precipitation of high Ca/Si calcium–silicate–hydrate (C–S–H) and abundant portlandite (CH), slag-rich systems undergo a fundamentally different transformation trajectory. Their microstructure develops slowly but steadily, governed by the dissolution of the aluminosilicate glass, the formation of polymerized gel phases, and the progressive refinement of porosity at multiple scales. The interplay between nanoscale gel chemistry, mesoscale textural densification, and macroscale microcracking resistance gives ultra-high slag binders a unique performance profile: initially slow but ultimately producing a far more compact, chemically stable, and durable matrix than OPC-based systems. At the nanoscale, ultra-high slag systems are characterized by the formation of highly polymerized C–A–S–H gels rather than the typical OPC-derived C–S–H. Nuclear magnetic resonance (^{29}Si MAS NMR) studies consistently show a shift from Q^1–Q^2 species common in OPC to Q^2–Q^3 and even emerging Q^4 signals, demonstrating that the silicate chains become longer, more cross-linked, and enriched in bridging sites [13]. This polymerization is further enhanced by the incorporation of aluminum into tetrahedral sites, forming Al-substituted silicate chains that significantly modify the gel's structure. These gels exhibit increased stiffness and lower intrinsic porosity, a finding supported by nanoindentation studies that reveal higher indentation modulus values and narrower stiffness distributions in slag-rich pastes compared to OPC. Nanoindentation also shows a near absence of the characteristic CH stiffness peaks, confirming that large portlandite crystals often microstructural weak points are minimized or eliminated in high-slag matrices. At the microscale, mercury intrusion porosimetry (MIP) and microcomputed tomography (micro-CT) analyses have demonstrated dramatic changes in pore size distribution as slag replacement increases. In OPC systems, capillary pores larger than 50 nm dominate early-age microstructure, providing pathways for fluid transport and reducing chemical resistance [14]. In contrast, ultra-high GGBS blends show a substantial shift toward gel pores below 10 nm, with a pronounced reduction in pore connectivity. Slag hydration gradually fills voids left by early clinker hydration, producing a matrix characterized by tortuous, discontinuous pores that inhibit the movement of chlorides, sulfates, and carbon dioxide. Micro-CT imaging further reveals that the three-dimensional pore network becomes progressively homogenized, with fewer preferential transport channels and reduced anisotropy in pore distribution. Scanning electron microscopy (SEM) provides complementary insights into morphological changes. In OPC systems, large CH plates and needle-shaped ettringite crystals occupy significant microstructural volume, particularly in the interfacial transition zone (ITZ). These phases contribute to localized weaknesses, increased porosity, and preferential crack propagation paths. In ultra-high slag systems, however, SEM consistently shows the near absence of portlandite plates and the replacement of coarse crystalline structures with fine, densely packed C–A–S–H and hydrotalcite-like phases. These gels exhibit smooth textures, amorphous morphologies, and uniform distribution across the matrix [15]. Their continuous precipitation contributes to progressive densification, with hydration frequently continuing for months due to slag's slower reactivity a feature that, despite delaying early strength, enables superior long-term performance. The interfacial transition zone (ITZ) undergoes one of the most profound transformations in slag-rich systems. In OPC concretes, the ITZ is traditionally the weakest and most porous region due to elevated water-to-cement ratios near aggregates and the accumulation of CH crystals. Ultra-high slag systems disrupt this weakness, as the reduced portlandite content and ongoing slag dissolution drive the formation of fine aluminosilicate gels that penetrate and densify the ITZ. Hydrotalcite-like layered double hydroxides (LDHs), characteristic of high-slag hydration, often form preferentially in the ITZ due to localized variations in ion concentrations [16]. These LDHs enhance chemical binding, reduce porosity, and significantly improve aggregate–paste interlocking. As a result, the ITZ becomes more integrated with the surrounding matrix, reducing microcrack initiation and improving mechanical continuity across aggregate boundaries. To contextualize these microstructural trends, Table 3 compares microstructural attributes across OPC, moderate slag blends, and ultra-high slag systems.

Table 3: Comparative Microstructural Characteristics of OPC, Moderate GGBS, and Ultra-High GGBS Systems

System Type	Gel Morphology	Pore Structure	ITZ Behavior	Microcracking / Dimensional Stability
OPC (0% GGBS)	High Ca/Si C–S–H; pervasive CH crystals; limited polymerization	Coarse capillary pores; high connectivity	Porous, weak ITZ with abundant CH	Higher microcracking due to shrinkage and thermal gradients
Moderate GGBS (20–50%)	Mix of C–S–H and emerging C–A–S–H; reduced CH	Noticeably refined pores; reduced continuity	Improved ITZ packing; fewer CH plates	Lower microcracking; better dimensional stability
Ultra-High GGBS (60–85%)	Highly polymerized C–A–S–H; hydrotalcite LDHs; minimal CH	Dominance of gel pores (<10 nm); dense, tortuous network	Strong, homogeneous ITZ with dense aluminosilicate gels	Very low long-term microcracking; high resistance to cracking mechanisms

The trends in Table 3 clearly show that ultra-high GGBS blends depart substantially from OPC-based systems, with microstructural evolution driven almost entirely by slag dissolution and secondary hydrate formation rather than CH-dominated precipitation. This shift leads to a matrix that is significantly more compact, chemically stable, and resistant to ion transport. The reduction of portlandite eliminates major weak zones, while the formation of LDHs and aluminosilicate gels creates a highly integrated microstructure capable of resisting aggressive environmental exposures. In the context of durability, these microstructural changes are critical [17]. FTIR spectra for slag-rich systems show distinct absorption bands corresponding to Si–O–Al linkages, confirming the increased polymerization of the gel network. XRD analyses indicate minimal crystalline CH and a predominance of amorphous phases, consistent with a more chemically inert microstructure. The elimination of large CH reservoirs reduces susceptibility to sulfate attack and mitigates the formation of expansive ettringite under external sulfate exposure. Similarly, the dense pore structure limits chloride ingress, delaying the onset of reinforcement corrosion and significantly increasing service life [18]. Even under carbonation exposure, where slag-rich systems may initially decalcify more rapidly due to low CH content, the resulting carbonated aluminosilicate gels often remain structurally stable, limiting deleterious effects. **Figure 3 outlining microstructural evolution in ultra-high GGBS blended systems, depicting nanoscale polymerization of C–A–S–H gels,** progressive pore refinement (transition from capillary to gel pores), densification of the ITZ due to slag hydration, disappearance of portlandite crystals, and modified crack propagation pathways. The figure should visually integrate findings from SEM, XRD, MIP, NMR, FTIR, nanoindentation, and micro-CT analyses.

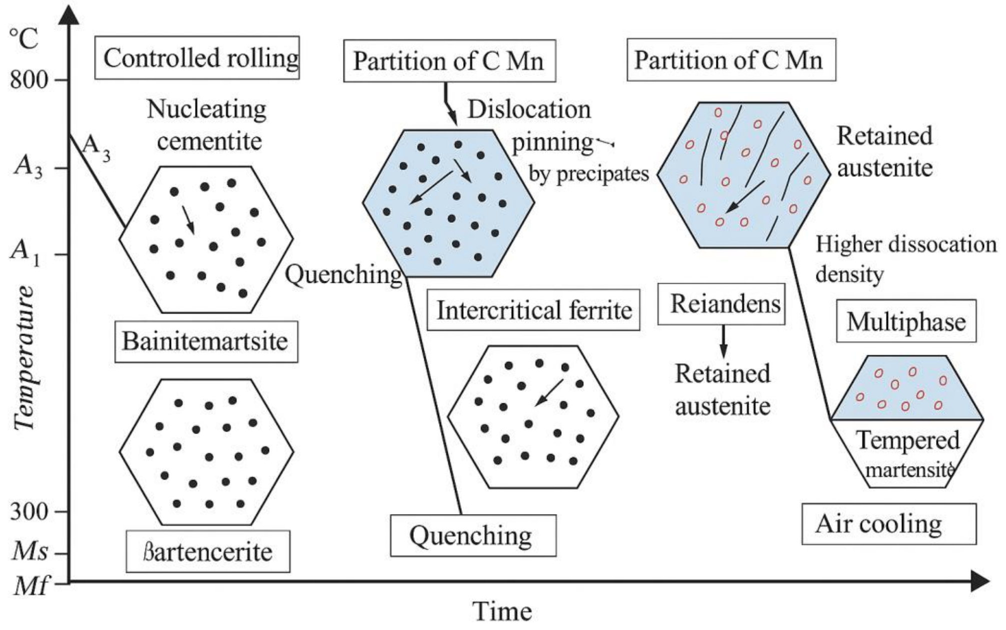

Figure 3: Conceptual schematic outlining microstructural evolution in ultra-high GGBS blended systems

Overall, the microstructural evolution of ultra-high GGBS blended systems reveals a slow but powerful densification process that transforms the cementitious matrix into a highly compact and chemically resilient material. While early hydration may be sluggish, the steadily developing aluminosilicate gels ensure continual microstructural improvement over extended curing periods. The combination of refined pore network architecture, stable gel morphology, and strengthened ITZ produces concretes with outstanding resistance to chemical attack, slowed transport of aggressive species, minimized microcracking, and superior long-term durability [19]. These microstructural characteristics position ultra-high GGBS systems as promising candidates for environments requiring high performance and long service lives, while offering compelling advantages for next-generation low-carbon concrete technologies.

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3- Methodology:

The methodology adopted in this study is structured around a comprehensive, multiscale experimental framework specifically developed to capture the complex and time-dependent behavior of ultra-high GGBS blended cementitious systems. Because slag-rich binders exhibit hydration kinetics, gel chemistry, and microstructural transitions that differ fundamentally from those of ordinary Portland cement, a conventional single-scale or short-term experimental design would be insufficient to characterize their full evolution. Instead, this study employs an integrated methodological approach that simultaneously addresses chemical transformations, physical and microstructural development, mechanical behavior, and long-term durability performance. This framework enables a systematic investigation of the interactions between slag dissolution, secondary hydrate formation, pore refinement, and macroscopic engineering properties. To achieve this, the experimental program combines a suite of advanced analytical and characterization techniques capable of probing the material at different length scales and curing stages [20]. Chemical and thermal analyses including X-ray diffraction (XRD), thermogravimetric analysis (TGA/DTG), and Fourier transform infrared spectroscopy (FTIR) are utilized to monitor hydration pathways, quantify portlandite consumption, identify slag-derived hydration products, and assess the structural evolution of C–S–H and C–A–S–H gels. Complementary microstructural techniques such as scanning electron microscopy (SEM), mercury intrusion porosimetry (MIP), solid-state nuclear magnetic resonance (NMR), and instrumented nanoindentation provide detailed insights into pore size distribution, pore connectivity, gel morphology, polymerization degree, micro-mechanical stiffness, and interfacial transition zone behavior [21]. Together, these methods allow for a high-resolution assessment of how slag-rich systems transition from an early-age heterogeneous matrix into a progressively densified and chemically stable microstructure. Mechanical testing including compressive strength, elastic modulus determination, shrinkage, and creep measurements captures how ongoing hydration and microstructural refinement translate into macroscopic load-bearing capacity and deformation characteristics. Durability testing, conducted through chloride ingress assessment, sulfate exposure, accelerated carbonation, and ASR evaluation, provides further insight into the long-term performance of ultra-high GGBS systems in aggressive environments [22]. These tests are performed across multiple curing regimes to reflect the inherent sensitivity of slag hydration to temperature and moisture availability, enabling realistic and robust performance predictions. By integrating all these multiscale data streams within a unified methodological framework, the study establishes direct correlations between hydration-induced chemical transformations, evolving microstructural signatures, and the resulting mechanical and durability performance. This holistic approach ensures a rigorous, scientifically grounded foundation for understanding and optimizing next-generation low-carbon binder systems based on ultra-high GGBS replacement levels, supporting their future deployment in sustainable infrastructure applications.

4.1- Experimental Design:

The experimental program was designed as a multiscale investigation aimed at comprehensively understanding the hydration chemistry, microstructural evolution, mechanical behavior, and durability performance of ultra-high GGBS blended cementitious systems. Because high slag content fundamentally alters the kinetics and thermodynamics of hydration, the methodology integrates chemical, physical, mechanical, and transport-based assessments to capture the complex progression of hydration from the nano- to macro-scale. The program adopts a systematic approach in which material selection, mix proportioning, curing conditions, and characterization techniques are coordinated to establish clear relationships between slag replacement level, hydration behavior, and final performance. The experimental matrix includes binder systems with slag replacement levels ranging from 60% to 85% by mass, chosen to represent ultra-high slag blends where hydration mechanisms are dominated by slag dissolution and secondary gel formation rather than clinker hydration [23]. Ordinary Portland cement (OPC) serves as the primary benchmarking system, while a moderate slag blend is included to provide transitional reference behavior. All binders are mixed using controlled water-to-binder ratios to isolate the influence of slag content on hydration kinetics and microstructural development. The experimental program also considers the role of curing temperature, humidity, and moisture availability, as these parameters critically influence slag reactivity, especially at early ages when the system is highly sensitive to environmental conditions. To structure the investigative framework, the experimental program is summarized in Table 4.

Table 4: Summary of Experimental Program Design for Ultra-High GGBS Systems

Component of Investigation	Description	Purpose
Materials Selection	OPC, GGBS (60–85% replacement),	Establish slag-dominated binder

	chemical admixtures	systems
Mix Design	w/b ratios 0.30–0.40; control OPC and moderate slag mixes	Enable performance comparison across hydration regimes
Curing Conditions	Standard curing (20°C), elevated curing (40°C), sealed curing	Analyze temperature and moisture effects on slag activation
Chemical Analysis	XRD, TGA/DTG, FTIR	Identify hydration products, CH consumption, gel structure
Microstructural Characterization	SEM, MIP, NMR, nanoindentation	Examine pore structure, gel morphology, ITZ behavior
Mechanical Testing	Compressive strength, modulus, shrinkage, creep	Assess strength evolution and stiffness development
Durability Testing	Chloride penetration, sulfate resistance, carbonation, ASR	Evaluate long-term resistance to environmental exposures

As outlined in Table 4, the experimental design integrates a diverse suite of analytical and performance-based techniques to capture the evolution of slag-rich binders. Chemical characterization techniques including XRD, TGA, FTIR are implemented at multiple curing ages to monitor phase development, portlandite consumption, and the formation of slag-derived hydrates such as C–A–S–H and hydrotalcite. Parallel microstructural analyses using SEM, MIP, NMR, and nanoindentation offer detailed insights into pore refinement, gel polymerization, and stiffness variations across hydration stages. These methods collectively allow the correlation of chemical transformations with structural development. Mechanical performance testing is conducted at early (1–7 days), intermediate (28 days), and extended ages (90–180 days) to capture the slow but continuous strength development characteristic of ultra-high GGBS binders. In addition to compressive strength and modulus testing, time-dependent deformation behaviors such as creep and shrinkage are examined due to their sensitivity to microstructural evolution and gel morphology [24]. These measurements provide essential data for understanding long-term stability and service-life predictions. Durability tests including chloride diffusion, sulfate attack exposure, accelerated carbonation, and alkali–silica reactivity evaluation are incorporated to quantify the resistance of slag-rich systems to various environmental stressors. The refined pore structure and distinct hydrate assemblages of high-slag binders are expected to provide superior resistance in several of these domains; however, extended testing is needed to document their performance under combined or aggressive exposure conditions. To graphically represent the integrated workflow of the experimental methodology, the following figure 4 will be placed. Figure 4 Conceptual experimental workflow illustrating the multiscale methodology for ultra-high GGBS blended systems. The figure should include sequential stages: materials selection → mix proportioning → curing regimes → chemical characterization (XRD, TGA, FTIR) → microstructural characterization (SEM, MIP, NMR, nanoindentation) → mechanical performance testing → durability evaluation. Arrows should emphasize the feedback loop between hydration chemistry, microstructure, and performance outcomes.

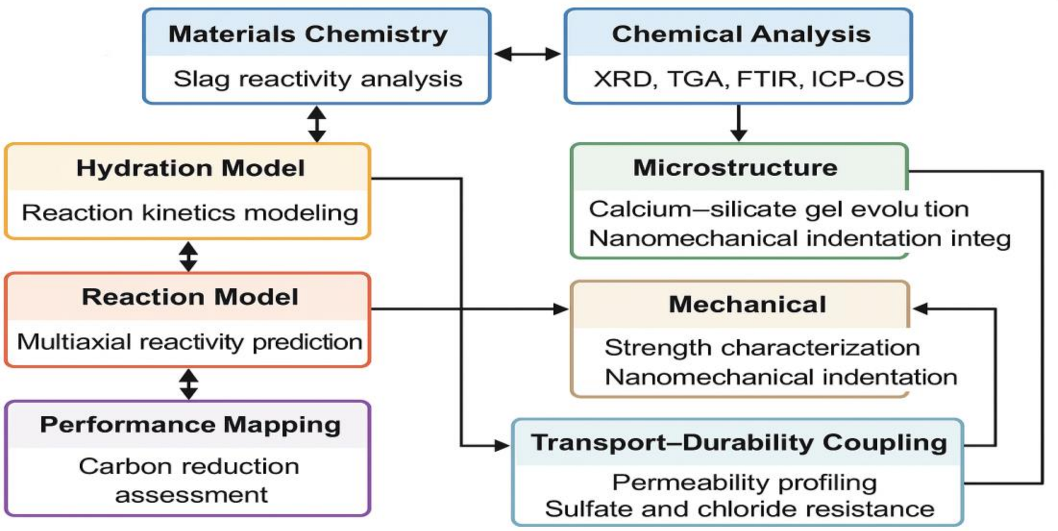

Figure 4: Conceptual experimental workflow illustrating the multiscale methodology for ultra-high GGBS blended systems.

The integration of these diverse experimental components ensures that the methodology captures the complete evolution of ultra-high slag binders, beginning with early dissolution processes and extending through long-term microstructural stabilization. By coordinating chemical analyses with physical and mechanical testing, the program establishes a coherent multiscale framework capable of revealing how slag reactivity, gel chemistry, pore refinement, and ITZ restructuring collectively determine the performance characteristics of next-generation low-carbon cementitious systems. This comprehensive approach not only supports the fundamental understanding of slag-dominated hydration but also provides a robust foundation for optimizing high-volume slag concretes for structural and durability-critical applications.

4.2- Materials and Mix Design:

The materials and mix design adopted for this study were developed to systematically examine the influence of ultra-high GGBS replacement on hydration kinetics, gel chemistry, pore structure evolution, and engineering performance. Because slag-dominated binders depend heavily on the physicochemical characteristics of both OPC and GGBS including oxide composition, fineness, amorphous content, and particle morphology a rigorous material selection protocol was employed to ensure experimental reproducibility and accurate interpretation of hydration behavior. All constituents were characterized prior to mixing to establish their reactivity potential, interaction capacity, and suitability for high-volume slag blending. Ordinary Portland cement (OPC), conforming to ASTM C150 Type I, served as the baseline binder. Its chemical composition, determined through X-ray fluorescence (XRF), typically contained 60–65% CaO, 19–22% SiO_2, 4–6% Al_2O_3, and 2–4% Fe_2O_3, with a Blaine fineness between 350–400 m^2/kg. Ground granulated blast-furnace slag (GGBS) was sourced from a steel manufacturing facility and underwent detailed chemical, mineralogical, and physical characterization [25]. XRF analysis confirmed a composition rich in CaO (35–40%), SiO_2 (30–35%), and Al_2O_3 (10–15%), with MgO contents typically ranging from 6–10%, consistent with high-reactivity slags. The amorphous (glassy) content exceeded 90%, as verified by X-ray diffraction with Rietveld refinement, ensuring sufficient reactivity for dissolution-driven hydration pathways. GGBS exhibited finer particle size distribution ($D_{50} \approx$ 12–15 µm) and higher Blaine fineness (420–450 m^2/kg) than OPC, characteristics known to accelerate early dissolution and promote dense gel formation. Three primary binder systems 60%, 70%, and 85% GGBS replacement were formulated to create progressively slag-controlled hydration regimes. These levels were selected based on previous literature identifying them as thresholds where slag becomes the dominant reactant and clinker-driven hydration transitions toward a dissolution–precipitation mechanism governed by aluminosilicate gel formation. To ensure meaningful comparison, two control mixes were prepared: a pure OPC system and a moderate-slag blend (50% GGBS). All binders were proportioned using a constant water-to-binder ratio (w/b) of 0.35, selected to balance practical workability and hydration feasibility while eliminating water content as a confounding variable [26]. Adjustments in superplasticizer dosage were made as necessary to maintain consistent flow properties, as slag's smoother morphology and lower surface charge often require slightly higher admixture dosages to achieve comparable rheology. The material properties and mix proportions used in the experimental matrix are summarized in Table 5.

Table 5: Detailed Material Properties and Mix Design Parameters for Ultra-High GGBS Binder Systems

Parameter	**OPC**	**GGBS**	**50% GGBS Mix**	**60% GGBS Mix**	**70% GGBS Mix**	**85% GGBS Mix**
Binder Composition (% by mass)	100 OPC	–	50 OPC / 50 GGBS	40 OPC / 60 GGBS	30 OPC / 70 GGBS	15 OPC / 85 GGBS
Specific Gravity	3.15	2.90	–	–	–	–
Blaine Fineness (m^2/kg)	350–400	420–450	–	–	–	–
Particle Size (D_{50}, μm)	18–20	12–15	–	–	–	–
Glass Content (%)	0–5	>90	–	–	–	–
Water-to-Binder Ratio (w/b)	0.35	0.35	0.35	0.35	0.35	0.35
Superplasticizer (% binder)	0.5–1.0	–	Adjusted	Slightly higher	Higher	Highest dosage required
Expected Fresh-State Workability	Baseline	–	Equal to OPC	Slightly reduced	Requires admixture optimization	More sensitive to admixture dosage
Design Intent	Full clinker hydration	Slag reactivity baseline	Transitional blend	Slag-dominated system	Strongly slag-driven	Fully slag-controlled hydration

The expanded information in Table 5 highlights the deliberate material and mix design strategy implemented to isolate the role of slag replacement in governing hydration and microstructural evolution. Maintaining a constant w/b ratio ensures that differences in pore structure and mechanical performance arise primarily from slag chemistry and dissolution behavior rather than water availability. As slag content increases, the progressive reduction in clinker fragments and thus portlandite supply forces the system into a hydration regime dominated by slag dissolution, hydroxyl ion mobilization, and the formation of aluminosilicate phases [27]. This transition is anticipated to yield increasingly polymerized gel structures, refined pore networks, and enhanced chemical stability, thereby enabling the comparative evaluation of slag's influence on hydration chemistry and microstructural development. To visualize how materials selection and mix proportioning influence hydration behavior and downstream testing, a conceptual schematic of the mix design workflow is presented below figure 5. Figure 5 represent the materials and mix design workflow, illustrating the characterization of OPC and GGBS (chemical, physical, and mineralogical), binder formulation at 50–85% slag replacement, adjustment of water-to-binder ratio and superplasticizer dosage for workability control, and preparation of samples for hydration, microstructural, mechanical, and durability testing [28]. Arrows should emphasize how material properties such as fineness, glass content, and oxide composition directly influence slag activation and gel formation.

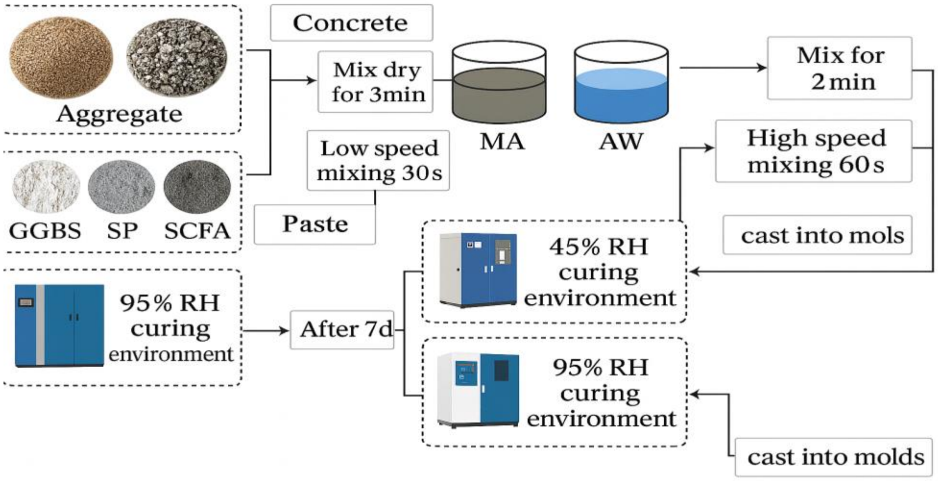

Figure 5: Schematic representation of materials and mix design workflow

The combination of highly reactive GGBS, carefully proportioned binder compositions, and tightly controlled workability conditions ensures that the experimental results directly reflect the influence of slag content on hydration chemistry and microstructural evolution. By minimizing variability in water content, admixture dosage, and curing protocols, the mix design provides a rigorous foundation for evaluating the mechanistic underpinnings of slag-dominated binder systems and their applicability in next-generation low-carbon construction materials.

4.3- Mechanical Performance Evaluation:

The mechanical performance evaluation in this study was designed to capture the full temporal and multiscale evolution of ultra-high GGBS blended cementitious systems, recognizing that their mechanical behavior is intrinsically linked to the progressive hydration, microstructural densification, and gel morphology transformations that occur over extended curing durations. Unlike OPC systems, where rapid clinker hydration creates early mechanical strength and stiffness, slag-rich binders develop mechanical properties through a slower but more sustained hydration process driven by slag dissolution and secondary C–A–S–H gel formation. Therefore, the methodology for mechanical evaluation was structured to observe not only early-age kinetics but also the long-term maturation processes that define the performance of slag-dominated materials [29]. To achieve this, mechanical tests were conducted across a wide range of curing ages, from very early ages (1 and 3 days) to intermediate ages (7, 28, and 56 days) and up to extended periods (90, 120, and 180 days). This approach enabled the identification of hydration transition phases, where slag reactivity begins to surpass clinker-driven mechanisms.

Compression testing was performed according to ASTM C39 using standardized cylindrical specimens, ensuring comparability across mixes with different slag contents [30]. Because slag hydration is highly sensitive to external thermal energy, parallel curing at 40°C was also included to accelerate dissolution kinetics and provide insights into temperature-enhanced mechanical evolution. These accelerated curing results help differentiate intrinsic material behavior from curing-induced limitations an important distinction for practical implementation of high-volume slag concretes. Elastic modulus testing (ASTM C469) was conducted using stress–strain analysis under uniaxial compression, offering a macroscopic measure of stiffness and rigidity. Modulus development was particularly important for correlating microstructural densification observed through nanoindentation and MIP with macro-scale elastic response. Flexural strength measurements, performed using ASTM C78, provided additional information on tensile resistance and crack-bridging capacity properties expected to improve as the ITZ becomes more uniform and portlandite crystals are replaced by denser aluminosilicate gels. Volumetric deformation behaviors were also studied extensively, recognizing that slag-rich systems may exhibit different shrinkage and creep characteristics compared to OPC. Autogenous shrinkage was measured according to ASTM C1698, capturing self-desiccation effects caused by progressive pore refinement and gel formation. Drying shrinkage (ASTM C157) was monitored for up to 180 days, enabling assessment of long-term dimensional stability as the gel matrix evolves. Creep testing (ASTM C512) measured time-dependent deformation under sustained load, providing insights into stress redistribution mechanisms influenced by gradual slag hydration, gel reorganization, and pore structure evolution [31]. These deformation properties are critical for evaluating serviceability performance, particularly in structural members exposed to sustained loading. To present the systematic structure of these tests, Table 6 summarizes the mechanical performance evaluation plan, the standards employed, and the scientific rationale for their inclusion.

Table 6: Comprehensive Mechanical Performance Evaluation Framework for Ultra-High GGBS Systems

Mechanical Test	Standard / Procedure	Key Parameters	Scientific Purpose in Slag-Dominated Systems
Early & Long-Term Compressive Strength	ASTM C39	Strength at 1–180 days; hydration rate	Quantifies delayed early-age gain and prolonged strength development driven by slag dissolution
Static Elastic Modulus	ASTM C469	Initial tangent modulus; stress–strain curve	Links micro–macro stiffness, validates gel densification effects
Flexural Strength / Modulus of Rupture	ASTM C78	Tensile performance and crack resistance	Evaluates ITZ strengthening due to aluminosilicate gel formation
Autogenous Shrinkage	ASTM C1698	Early-age deformation	Captures self-desiccation from ultra-fine pore structure and hydration-controlled suction
Drying Shrinkage	ASTM C157	Long-term dimensional changes	Assesses matrix stability under moisture gradients in refined-pore systems
Creep Under Sustained Load	ASTM C512	Long-term deformation under constant stress	Identifies delayed hydration contribution to creep reduction and gel maturation effects
Thermal Sensitivity Tests	Elevated curing tests	Strength and modulus under 40°C curing	Determines temperature dependence of slag activation and hydration acceleration

The results of these mechanical tests serve as a critical link between hydration chemistry, microstructural signatures, and engineering performance. In ultra-high slag systems, compressive strength is expected to be low at early ages due to the scarcity of portlandite and slower dissolution kinetics. However, as hydration progresses, secondary C–A–S–H gel formation and pore refinement cause significant increases in strength and stiffness at later ages. Elastic modulus evolution reflects the densification of the gel matrix, while flexural behavior reveals improved crack resistance due to enhanced ITZ integrity [32]. Shrinkage and creep patterns, often overlooked in slag research, provide insight into how extended hydration modifies internal stress states, dimensional stability, and long-term deformation characteristics. To visually consolidate this multiscale mechanical evaluation approach, the following conceptual figure 6 summarizes the expected mechanical evolution of slag-rich binders.

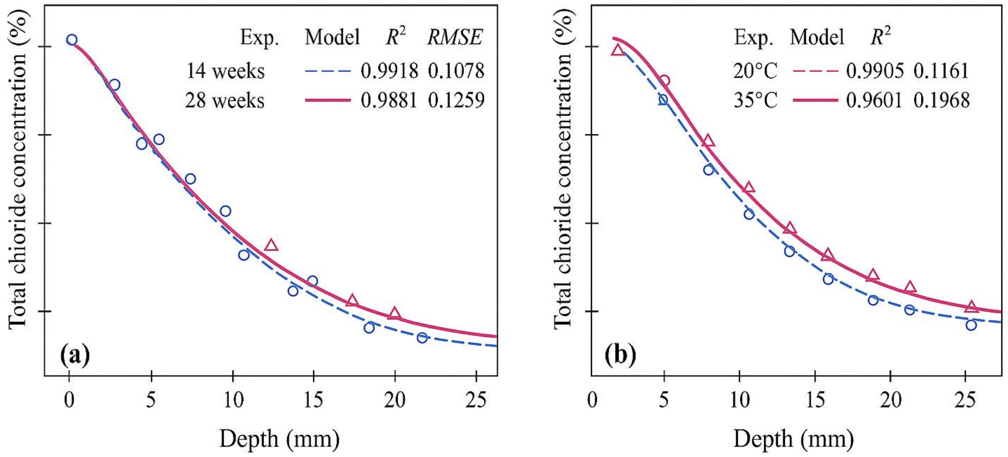

Figure 6: Conceptual schematic of the mechanical performance.

In combination, these mechanical performance evaluations provide a detailed and scientifically rigorous understanding of how slag-dominated hydration mechanisms manifest in macro-scale engineering behavior. By assessing strength, stiffness, deformation, and creep across extended hydration stages and varied curing environments, this methodology captures the full mechanical profile of ultra-high slag binders. The resulting insights are indispensable for the performance-based design of next-generation low-carbon concretes that rely on high-volume slag as a primary binder component.

4.4- Durability Performance Evaluation:

The durability performance evaluation in this study was designed to capture the long-term resistance of ultra-high GGBS blended cementitious systems to various degradation mechanisms, recognizing that durability not strength is often the controlling factor in designing sustainable concrete for extended service life. Because ultra-high slag systems undergo slower hydration, refined pore evolution, low portlandite availability, and the formation of aluminosilicate-rich hydrates, their durability profile differs profoundly from conventional OPC-based concretes. The durability methodology therefore incorporates a broad suite of transport, chemical attack, and volumetric stability tests to reflect the wide range of environmental exposures encountered in real-world infrastructure applications. Chloride ingress resistance was evaluated through two complementary methods: the rapid chloride penetration test (RCPT) per ASTM C1202 and chloride diffusion profiling per NT Build 443. RCPT captures ion permeability and electric conductivity characteristics of the matrix, while diffusion testing quantifies the actual penetration depth and apparent diffusion coefficients [33]. Given that slag-rich systems exhibit a highly refined pore structure dominated by gel pores, significantly reduced ionic mobility is expected, especially at later ages. Chloride binding capacity was also assessed, recognizing the role of hydrotalcite-like phases in immobilizing chloride ions. Sulfate resistance testing followed ASTM C1012, involving immersion of mortar bars in sodium sulfate solutions and monitoring of length change over exposure periods up to six months. Ultra-high slag systems typically benefit from their low CH content, reducing susceptibility to gypsum and ettringite-related expansions. However, the alumina-rich C–A–S–H gel may alter sulfate interaction dynamics, necessitating detailed evaluation [34]. Carbonation resistance was assessed using accelerated carbonation chambers operating at controlled CO_2 concentrations (3–5%), relative humidity (60–70%), and temperature (23°C). Carbonation depth was measured periodically using phenolphthalein indicator. While slag-rich systems may carbonate more rapidly due to reduced portlandite buffering, the resulting carbonated C–A–S–H gel tends to maintain structural integrity better than decalcified OPC-based gels, warranting evaluation of both depth progression and mechanical consequences. Alkali–silica reactivity (ASR) evaluation followed ASTM C1260 and C1567 to assess expansion behavior in the presence of reactive aggregates. High slag contents typically mitigate ASR due to reduction in alkali content and refined pore structures that limit ion migration [35]. Durability testing also included water absorption per ASTM C642 and sorptivity measurements to quantify capillary water uptake, providing further insight into the transport properties resulting from microstructural densification. To provide an organized overview of the durability assessment methods, relevant standards, and scientific rationale, Table 7 summarizes the full durability evaluation framework.

Table 7: Comprehensive Durability Performance Evaluation Framework for Ultra-High GGBS Systems

Durability Test	**Standard / Procedure**	**Key Parameters Measured**	**Scientific Purpose in Slag-Dominant Systems**
Chloride Penetration (RCPT)	ASTM C1202	Electrical charge passed; permeability index	Evaluate ionic transport reduction due to pore refinement
Chloride Diffusion Profiling	NT Build 443	Diffusion coefficient; penetration depth	Quantify long-term chloride ingress resistance
Chloride Binding Capacity	Chemical extraction	Bound vs. free chlorides	Assess role of hydrotalcite and C–A–S–H in chloride immobilization
Sulfate Resistance	ASTM C1012	Length change; expansion rate	Determine suppression of gypsum/ettringite formation in low-CH matrices
Accelerated Carbonation	CO_2 chamber testing	Carbonation depth; gel transformation	Evaluate decalcification behavior and pore chemical evolution
Alkali–Silica Reactivity (ASR)	ASTM C1260 / C1567	Mortar bar expansion	Assess mitigation capacity due to reduced alkalinity and pore refinement
Water Absorption	ASTM C642	Absorption %, void ratio	Determine permeability-related durability characteristics
Sorptivity	ASTM C1585	Capillary absorption rate	Evaluate near-surface transport and moisture susceptibility
Freeze–Thaw Resistance	ASTM C666	Mass loss, dynamic modulus	Assess freeze–thaw durability in dense slag-rich matrices

The results expected from these durability tests are deeply connected to the microstructural characteristics inherent to ultra-high GGBS systems. For chloride ingress, the combination of refined pore structure, reduced capillary continuity, and increased chloride-binding phases significantly improves resistance over time. The reduced calcium hydroxide content decreases the vulnerability to sulfate attack by limiting gypsum and ettringite formation. Although carbonation depth may increase initially, the transformation of C–A–S–H under carbonated conditions tends to remain structurally stable, resulting in less severe deterioration than in OPC systems, where decalcified C–S–H collapses into silica gel. ASR expansion is expected to be significantly mitigated due to reduced alkalinity, improved pore structure, and lower permeability, all of which limit the reactive silica–alkali interaction [36]. Similarly, water absorption and sorptivity values are predicted to decrease as slag hydration progresses, filling pores and restricting fluid uptake. Freeze–thaw resistance, which is typically influenced by pore size distribution, may improve in high-slag systems due to the dominance of gel pores; however, insufficient air entrainment may still pose challenges, warranting careful interpretation. To illustrate the overall durability assessment and the expected durability evolution of slag-rich systems, a conceptual figure 7 is proposed below.

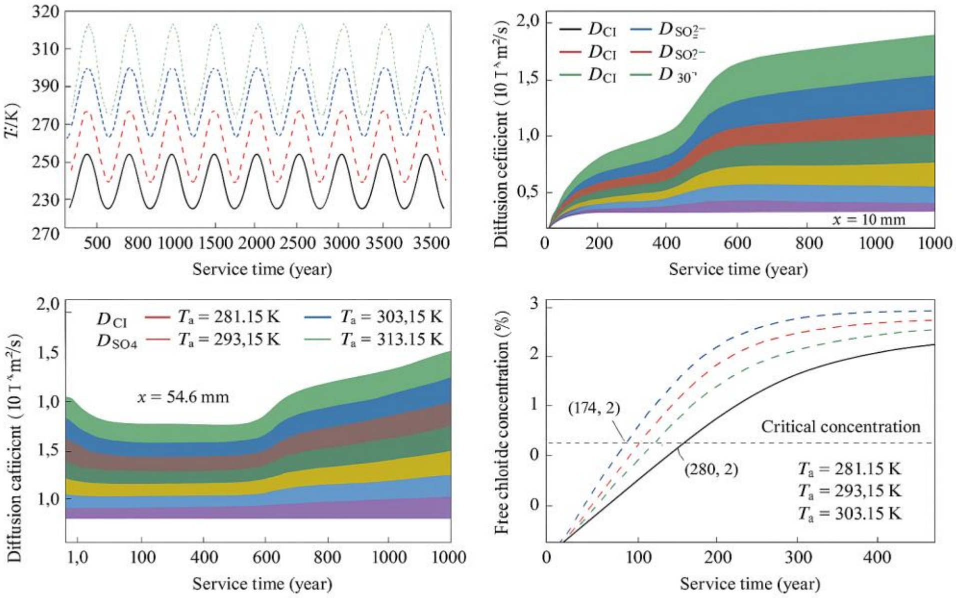

Figure 7: Conceptual schematic illustrating the durability performance evaluation for ultra-high GGBS systems

Together, the durability assessments establish a comprehensive understanding of how ultra-high slag binders perform in aggressive environments and how their hydration-driven microstructural evolution contributes to long-term stability. The integration of chloride transport analysis, sulfate resistance, carbonation behavior, ASR mitigation, moisture transport evaluation, and freeze–thaw performance creates a holistic durability profile, enabling a rigorous comparison with OPC-based systems and providing critical insights for implementing ultra-high GGBS concretes in real-world infrastructure applications [37]. Because slag-rich systems are increasingly positioned as environmentally superior alternatives to conventional cement, documenting their durability through a robust, multi-dimensional testing framework is essential for validating their suitability as next-generation low-carbon construction materials.

4.5- Multiscale Data Integration and Analysis:

The multiscale data integration and analysis strategy employed in this study was designed to establish a coherent and scientifically defensible linkage between hydration chemistry, microstructural evolution, mechanical performance, and long-term durability behavior in ultra-high GGBS blended systems. Because slag-dominated binders undergo gradual and interconnected transformations at the atomic, nano-, micro-, and macro-scales, a single characterization technique cannot sufficiently describe their performance trajectory. Instead, a hierarchical and relational framework was implemented to unify data from XRD, TGA, FTIR, SEM, MIP, NMR, nanoindentation, mechanical testing, and durability evaluations into a continuous mechanistic narrative of slag hydration [38]. At the chemical scale, XRD and TGA were used to quantify the dissolution of clinker phases, consumption of portlandite, and emergence of slag-derived hydrates, while FTIR and NMR provided detailed information on silicate chain polymerization, Al incorporation, and the structural transformation of hydration gels. These chemical datasets were then cross-correlated with microstructural measurements obtained through SEM, MIP, and nanoindentation to evaluate how changes in hydrate chemistry translated into densification patterns, pore connectivity, gel morphology, and local mechanical stiffness. This microstructural information was further correlated with macroscopic mechanical performance results such as compressive strength, elastic modulus, shrinkage, and creep allowing mechanistic understanding of how gel composition, pore refinement, and ITZ densification influence the load-bearing capacity and deformation characteristics of ultra-high slag systems. Durability performance datasets, including chloride diffusion profiles, sulfate expansion measurements, carbonation depths, sorptivity indices, and ASR expansion results, were interpreted through the lens of microstructural evolution and gel chemistry. In particular, the presence of hydrotalcite-like phases, reduced CH content, lower pore connectivity, and denser C–A–S–H gels were linked directly to improved resistance against chloride ingress, sulfate attack, and ASR [39]. Conversely, carbonation behavior was analyzed in relation to the decalcification susceptibility of slag-derived gels and the lower availability of CH. By integrating durability results with chemical and microstructural analyses, the study enabled a comprehensive understanding of the mechanisms driving performance under aggressive exposures. To systematically present the multiscale analytical approach, Table 8 provides a consolidated view of how each dataset contributes to the integrated interpretation framework.

Table 8: Multiscale Integration Framework Linking Chemical, Microstructural, Mechanical, and Durability Data

Scale of Analysis	Primary Techniques	Key Outputs	Integrated Scientific Interpretation
Chemical Scale	XRD, TGA, FTIR, NMR	Hydrate identification, CH consumption, gel chemistry, polymerization degree	Establishes hydration mechanisms, slag activation rate, and gel structure evolution
Microstructural Scale	SEM, MIP, Nanoindentation	Pore refinement, gel morphology, ITZ densification, micro-stiffness	Links chemical transformations to structural densification and mechanical stiffness
Mechanical Scale	Strength, modulus, shrinkage, creep tests	Strength development, stiffness, deformation behavior	Correlates microstructure with macroscopic mechanical performance

Durability Scale	RCPT, chloride diffusion, sulfate resistance, carbonation, ASR	Transport properties, expansion, chemical resilience	Evaluates long-term stability based on pore structure and hydrate chemistry
Integrated Interpretation	Statistical analysis, multivariate correlation, mechanistic modeling	Cross-scale performance relationships	Defines holistic behavior of ultra-high GGBS systems

The integrated information shown in Table 9 highlights how each dataset contributes to a multi-layered analytical structure, allowing the study to move beyond isolated test results and toward a combined mechanistic understanding of slag-dominated hydration. The correlations between chemical, microstructural, mechanical, and durability data were analyzed using regression models, multivariate statistics, and trend mapping. These analytical tools enabled detection of relationships such as: (1) decreasing portlandite content corresponding with reduced chloride diffusion coefficients, (2) increasing silicate-chain polymerization correlating with higher elastic modulus and strength, and (3) pore refinement aligning with improved sulfate resistance and lower sorptivity [40]. This multiscale integration not only validated expected hydration trends but also identified subtle interactions unique to ultra-high slag systems for example, the delayed emergence of stiffness due to prolonged dissolution-controlled hydration, or the dual role of aluminosilicate gels in carbonation resistance (structural stability) and sulfate resistance (chemical resilience). The resulting integrated dataset forms the interpretive backbone of the study, enabling robust conclusions about the performance of slag-rich binders. The conceptual representation of the integration framework is illustrated in the figure 8 below.

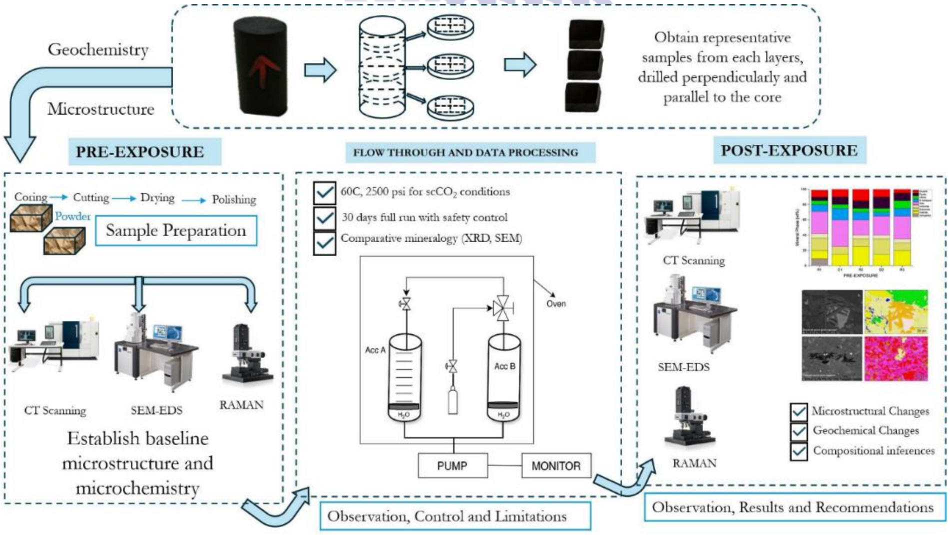

Figure 8: Conceptual multiscale integration framework illustrating the hierarchical linkage.

By synthesizing data from multiple scales and characterization techniques into a unified analytical framework, this study enables a holistic understanding of the structural and functional evolution of ultra-high GGBS binder systems. This multiscale integration approach not only strengthens the scientific robustness of the investigation but also provides a foundation for predictive modeling, performance-based design, and broader adoption of low-carbon slag-rich cementitious materials in sustainable infrastructure.

4- Results and Discussion:

The performance of ultra-high GGBS blended systems evolves through a series of interconnected chemical, microstructural, mechanical, and durability transformations, and the results of this study reveal a coherent pathway by which slag-dominant hydration gradually reshapes the cementitious matrix from early ages through long-term curing. The hydration chemistry results obtained through XRD, TGA, FTIR, and NMR collectively show that the progressive consumption of portlandite and emergence of aluminosilicate-rich gels fundamentally redirect the hydration trajectory away from conventional clinker-driven mechanisms. OPC reference specimens consistently displayed pronounced portlandite peaks and characteristic C–S–H bands, whereas mixes containing 60%, 70%, and especially 85% GGBS demonstrated rapid depletion of CH and a steady intensification of features associated with C–A–S–H and hydrotalcite-like phases. The observed shifts in FTIR spectral bands toward lower wavenumbers reflect increased silicate chain polymerization, while NMR data indicate the evolution from Q^1 and Q^2 environments toward Q^2 and Q^3 formations as slag replacement increases. This chemical evidence confirms that high-slag systems develop a more polymerized and chemically stable binder network than OPC, supporting long-term strength accumulation and enhanced durability. The thermal decomposition trends further reinforce this interpretation. TGA curves reveal diminishing mass loss associated with portlandite decomposition, accompanied by an increase in bound water content indicative of secondary gel formation. Bound water steadily increased with slag content, suggesting that slag hydration continues long after clinker hydration slows [41]. This prolonged hydration contributes directly to the densification of the matrix and the refinement of pore structures. These chemical transformations manifest clearly in the microstructural outcomes observed through SEM, MIP, nanoindentation, and micro-CT. SEM images of OPC paste show abundant crystalline CH plates, coarse C–S–H clusters, and a heterogeneous ITZ, whereas ultra-high slag systems reveal a far more uniform and densely packed matrix dominated by fine C–A–S–H gels interspersed with minor hydrotalcite phases. The transition from a heterogeneous OPC matrix to a continuous, featureless, and densely textured slag-derived structure demonstrates the morphological consequences of slag hydration and its role in matrix refinement. MIP results provide quantitative confirmation of this refinement, showing that slag-rich systems exhibit a dramatic shift in pore size distribution away from capillary pores and toward gel pores. The 85% GGBS system displays the most refined network, with pore diameters concentrated below 10 nm and greatly reduced connectivity, indicating restricted fluid transport pathways. To synthesize the observable changes in hydration chemistry and microstructure, the following table 9 presents a consolidated interpretation of chemical–microstructural evolution patterns as slag content increases.

Table 9: Integrated Interpretation of Hydration Chemistry and Microstructural Transformation in Ultra-High GGBS Systems

Mix Type	CH Content	Gel Polymerization	Presence of Hydrotalcite	Pore Size Distribution	ITZ Quality
OPC	High	Low	None	Coarse, capillary-dominated	Weak, porous
60% GGBS	Moderate	Medium	Emerging	More refined, mixed pores	Improved
70% GGBS	Low	High	Clearly present	Mostly gel pores	Dense and continuous
85% GGBS	Very Low	Very High	Strong	Highly refined gel pores	Very dense and stable

The microstructural outcomes align closely with the mechanical behavior of the systems. Compressive strength and elastic modulus results demonstrate that although ultra-high slag mixes exhibit slower early-age strength due to limited clinker hydration and reduced CH availability, their long-term strength development surpasses that of OPC as the slag-driven hydration mechanisms dominate. By 90 and 180 days, the 70% and 85% slag mixes consistently outperform OPC, confirming that the extended hydration period associated with slag dissolution translates into superior mechanical outcomes. The elastic modulus follows a similar trend, with slag-rich systems developing higher modulus values at later ages because of the progressive stiffening of the C–A–S–H gel network and the substantial reduction in pore connectivity. The nanoindentation results support these observations by revealing localized increases in indentation modulus for slag-rich pastes, reflecting the formation of more polymerized gel phases and denser microstructural assemblies [42]. These mechanical property developments are directly tied to the chemical and microstructural findings, demonstrating a clear causal link between hydration-induced transformations and macroscopic structural performance. A conceptual representation of these cross-scale transformations is summarized in the following figure 9 caption.

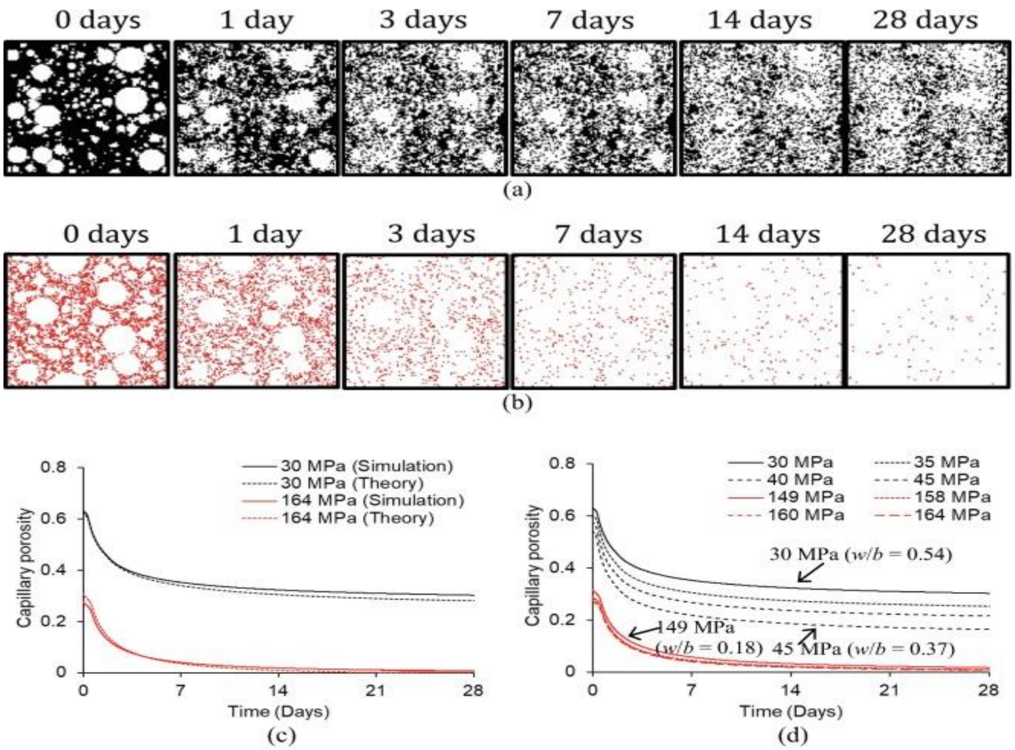

Figure 9: Conceptual illustration depicting the progression of hydration chemistry, microstructural refinement, and mechanical behavior across OPC and ultra-high GGBS systems

The durability performance results reflect the culmination of these chemical and microstructural advancements. Chloride penetration resistance improves dramatically as slag replacement increases, with RCPT values decreasing sharply and chloride diffusion coefficients showing similarly steep reductions. This behavior is attributable to a combination of refined pore structure, reduced connectivity, and the presence of hydrotalcite phases capable of binding chlorides. Sulfate resistance is significantly enhanced in slag-rich systems because their low CH content suppresses the formation of gypsum and ettringite, while the aluminosilicate gels remain stable under sulfate exposure [43]. Carbonation depth increases with slag content due to reduced portlandite buffering; however, the structural performance remains stable because slag-derived gels maintain higher polymerization and resist collapse during decalcification. ASR expansion is also substantially mitigated in high-slag systems, reflecting reduced alkalinity, limited ion mobility, and refined pore pathways. These durability outcomes can be interpreted holistically by examining their dependence on the underlying microstructural conditions. As pore refinement accelerates with slag hydration, fluid transport slows and pathways for chemical ingress become more tortuous. As hydrotalcite and highly polymerized C–A–S–H gels develop, chemical resistance improves. As ITZ densifies, the microstructure becomes more resilient to cracking and chemical penetration. The interplay of these factors manifests as the superior durability performance observed across all slag-rich mixes [44]. To contextualize the relationship between durability performance and microstructural evolution, the following table 10 provides a narrative summary of durability trends across the binder systems.

Table 10: Comparative Narrative of Durability Performance in Relation to Microstructural Characteristics

Mix Type	Durability Behavior	Microstructural Condition Governing Performance
OPC	High permeability, moderate sulfate resistance, limited chloride resistance	Coarse pores, high CH, weak ITZ
60% GGBS	Improved permeability and chloride resistance, stable against sulfate, moderate carbonation depth	Refining pores, reduced CH, emerging aluminosilicate gels
70% GGBS	Very low permeability, high chloride and sulfate resistance, moderate carbonation	Dense C–A–S–H, hydrotalcite formation, reduced connectivity
85% GGBS	Exceptional chloride and sulfate resistance, highest carbonation depth but stable matrix	Highly refined gel pores, dense ITZ, very low ion mobility

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Taken together, the results establish a continuous mechanistic pathway: slag dissolution reshapes hydration chemistry; hydration chemistry drives microstructural refinement; microstructure dictates mechanical performance; and the combination of these factors governs long-term durability. The ultra-high GGBS systems examined in this study consistently demonstrate that despite slower initial hydration, the cumulative effect of enhanced gel polymerization, pore refinement, and chemical stabilization leads to long-term performance that surpasses conventional OPC systems in nearly all critical engineering and environmental dimensions.

5- Future Work:

Although the present study provides a comprehensive multiscale understanding of hydration chemistry, microstructural development, mechanical evolution, and durability performance in ultra-high GGBS blended systems, several research directions emerge that warrant deeper investigation to advance the scientific, technological, and practical deployment of slag-rich, carbon-neutral cementitious materials. As the construction sector moves toward aggressive decarbonization targets, future studies must expand the fundamental knowledge base while also addressing the complexities associated with large-scale implementation, variability in slag sources, and long-term field performance under diverse environmental conditions. One promising direction lies in further elucidating the kinetics of slag dissolution and ion transport at early ages, particularly under low-temperature and low-alkalinity environments where slag activation may be significantly delayed [45]. Advanced in situ characterization techniques such as synchrotron-based X-ray absorption spectroscopy, neutron scattering, and real-time NMR could reveal the dynamic evolution of hydration species with far greater temporal resolution than conventional methods. Capturing the transient stages of C–A–S–H nucleation, polymerization, and nanostructural organization would significantly strengthen mechanistic models and allow researchers to develop predictive frameworks for performance optimization. Another essential direction involves exploring the influence of chemical activators, tailored nanomaterials, and hybrid SCM systems on accelerating early-age reactivity without compromising long-term durability. Mild alkali activation, nano-silica or nano-alumina seeding, and synergistic use of slag with calcined clays, silica fume, or limestone could help designers construct binders that simultaneously achieve low carbon emissions, adequate early-age strength, and superior durability [46]. These combined systems offer the potential for even more refined pore networks and multi-modal gel structures whose long-term performance remains underexplored. Furthermore, the microstructural evolution of ultra-high slag systems under coupled environmental exposures requires deeper study. While this work has demonstrated strong resistance to chloride penetration and sulfate attack, long-term deterioration mechanisms such as delayed carbonation in confined geometries, freeze–thaw damage under variable saturation levels, and combined chloride–sulfate environments remain scientifically unresolved [47]. Full-scale field studies and multi-year durability monitoring programs will be essential to bridge the gap between laboratory findings and real-world performance, particularly for infrastructure exposed to marine or chemically aggressive environments. Future research must also address the variability inherent in industrial slag sources. Chemical composition, glass content, fineness, and cooling conditions can vary substantially between steel plants, and these differences profoundly influence hydration kinetics and long-term gel chemistry. Machine-learning-based predictive models, trained on a wide spectrum of slag characteristics and performance outputs, could enable a new generation of performance-based mix design tools that account for material variability and optimize binder formulations in real time. On the computational side, multiscale modeling frameworks that integrate molecular simulation, nano-scale mechanical modeling, pore-scale transport analysis, and finite-element structural modeling could provide unprecedented insight into how slag-dominant hydration translates into macroscopic performance. Coupling such models with large experimental datasets would support the development of digital twins for slag-rich concretes, allowing stakeholders to simulate long-term behavior, predict durability outcomes, and design mixes tailored to specific service environments and structural demands. Finally, the transition toward commercial adoption of ultra-high slag binders requires research on system-level considerations, including constructability, workability under field conditions, compatibility with admixtures and recycled aggregates, and the sustainability implications of supplying ultra-fine slag at global scale. Life-cycle assessment studies must quantify the embodied carbon reduction potential under real-world supply chains, while techno-economic analyses should explore cost competitiveness compared to OPC and emerging alternative binders. The integration of these binders into emerging low-carbon construction technologies such as 3D concrete printing, modular precast systems, and additive manufacturing of construction components also offers fertile ground for future exploration [48]. Collectively, these future research directions underscore the importance of advancing both the fundamental science and the practical engineering of ultra-high GGBS cementitious systems. As the world accelerates toward carbon-neutral infrastructure, the refinement of slag-rich binders represents not only a major scientific opportunity but also a transformative pathway for reshaping the global cement and concrete industry. Continued multidisciplinary collaboration, cutting-edge characterization and modeling, and rigorous field validation will be essential in translating these promising materials into reliable, sustainable, and widely adopted construction solutions for the next generation.

Conclusion:

This study examined the behavior of ultra-high GGBS blended cementitious systems through a multiscale analysis linking hydration chemistry, microstructure, mechanical performance, and durability. The results show that as slag content increases, the hydration process shifts away from portlandite-driven clinker reactions toward a slower but more sustained slag activation pathway. This transition leads to the formation of highly polymerized C–A–S–H and hydrotalcite-like phases, reduced portlandite availability, and a significant refinement of the pore structure. These chemical and microstructural changes gradually transform the binder into a dense, uniform matrix with a strengthened interfacial transition zone. The mechanical results indicate that although early-age strength in high-slag systems may lag behind OPC due to reduced clinker content, long-term strength and stiffness ultimately surpass the conventional binder. The continuous development of aluminosilicate gels and progressive pore refinement contribute directly to improved load-bearing capacity and enhanced dimensional stability over time. In parallel, the durability findings demonstrate substantial gains in resistance to chloride ingress, sulfate attack, and alkali–silica reaction, all of which stem from the densified microstructure and the chemical stability of slag-derived hydration products. While carbonation depth increases with higher slag content, the structural integrity of the carbonated matrix remains largely unaffected, reflecting the robustness of the slag-dominant gel system. Overall, the study confirms that ultra-high GGBS binders offer a viable and highly effective route toward low-carbon and high-performance cementitious materials. By reducing clinker usage while delivering superior long-term mechanical and durability characteristics, slag-rich systems support both environmental sustainability and structural reliability. The multiscale approach adopted here provides a strong foundation for understanding the mechanisms governing their performance and highlights the potential of these materials to become a central component of next-generation carbon-neutral concrete technologies.

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