

Time Dependent Evolution of Cracks Healing in Cementitious Materials: Cracks Closure Dynamics

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ABSTRACT

The durability of cement-based infrastructure is frequently compromised by the initiation and propagation of microcracks, which accelerate the ingress of aggressive agents and ultimately reduce structural service life. Intrinsic self-healing has emerged as a sustainable mechanism capable of mitigating crack propagation without incorporating external healing agents. Nevertheless, the spatial evolution of healing products inside cracks and the governing transport-reaction mechanisms remain insufficiently

understood. This study develops an integrated reactive transport framework coupled with laboratory experimentation to investigate the temporal and spatial characteristics of intrinsic crack healing in cementitious materials. The proposed framework simultaneously considers ion migration, mineral precipitation, hydration kinetics, and progressive changes in crack transport properties to simulate healing behavior throughout the crack depth. Experimental validation was performed using X-ray micro-computed tomography (micro-CT), scanning electron microscopy (SEM), and energy-dispersive X-ray spectroscopy (EDS), enabling three-dimensional visualization and quantitative assessment of crack closure. The experimental observations demonstrated that healing was highly heterogeneous, with greater crack closure occurring near exposed surfaces than in deeper crack regions. Numerical simulations further illustrated that crack geometry, initial aperture, fluid transport conditions, material maturity, and ionic concentration significantly influence healing efficiency. The interaction between reactive transport processes and evolving crack morphology generated a dynamic feedback mechanism that governed long-term healing performance. The findings contribute to a more comprehensive understanding of autogenous healing mechanisms and provide valuable guidance for designing durable and resilient cementitious infrastructure capable of extending service life while reducing maintenance demands.

Keywords: Autogenous self-healing; Cementitious composites; Reactive transport modeling; Micro-computed tomography; Crack closure; Mineral precipitation; Concrete durability

1. Introduction

Concrete remains the most extensively utilized construction material worldwide because of its versatility, economic efficiency, and structural reliability. Despite these advantages, cementitious materials inevitably develop microcracks due to mechanical loading, thermal fluctuations, shrinkage, freeze-thaw cycles, and chemical degradation during service conditions. These cracks significantly increase permeability, allowing moisture, chlorides, sulfates, and carbon dioxide to penetrate the concrete matrix, thereby accelerating reinforcement corrosion and reducing structural durability (He et al., 2024). Over the past two decades, considerable research attention has focused on self-healing concrete as an innovative strategy for enhancing infrastructure resilience. Unlike

conventional repair methods that require continuous maintenance and substantial financial investment, self-healing materials possess the ability to restore damaged regions autonomously, thereby improving long-term durability while minimizing lifecycle costs (Folorunso, 2023).

Self-healing technologies generally fall into two principal categories: **extrinsic healing** and **intrinsic healing**. Extrinsic systems employ externally incorporated healing agents, including encapsulated polymers, bacterial spores, vascular networks, or chemical capsules that are activated after crack formation (Chen et al., 2022). Although these approaches often demonstrate rapid crack sealing, their effectiveness is constrained by limited healing-agent availability, higher production costs, manufacturing complexity, and concerns regarding long-term durability.

Intrinsic or autogenous healing, in contrast, exploits the natural physicochemical characteristics of hydrated cementitious materials. When water penetrates newly formed cracks, residual unhydrated cement particles continue hydration, while dissolved calcium ions react with carbonate species to generate calcium carbonate crystals that progressively fill crack voids. Since these reactions originate from constituents already present within the cement matrix, intrinsic healing can occur repeatedly under favorable environmental conditions without incorporating additional healing systems (Fan & Li, 2022).

Previous investigations have consistently demonstrated that autogenous healing effectively restores watertightness, reduces permeability, and partially recovers mechanical performance, particularly for microcracks smaller than approximately 200 μm (Versiani et al., 2022). However, most existing studies evaluate healing performance using surface observations, permeability reduction, ultrasonic pulse velocity, resonance frequency, or mechanical recovery tests. Although these approaches provide valuable indirect evidence of healing, they rarely capture the three-dimensional spatial evolution of healing products throughout the crack network.

Recent advances in X-ray micro-computed tomography have enabled researchers to observe internal crack morphology non-destructively with micrometer-scale resolution. This imaging technology provides unprecedented opportunities to quantify crack aperture evolution, healing-product distribution, and internal crack geometry over

time. Combined with scanning electron microscopy and elemental analysis, these techniques facilitate detailed characterization of mineral precipitation and hydration products responsible for crack closure (Li et al., 2024)

From a modeling perspective, significant progress has also been achieved in simulating cement hydration and self-healing processes. Existing numerical models generally emphasize hydration kinetics, mineral precipitation, or mechanical recovery independently. However, intrinsic healing represents a complex multiphysics phenomenon involving simultaneous fluid transport, ionic diffusion, chemical reactions, mineral precipitation, and continuous modification of crack geometry. These interdependent processes create nonlinear feedback mechanisms that remain insufficiently represented within existing computational frameworks (Ono et al., 2024). A particularly important limitation concerns the spatial variability of healing efficiency throughout crack depth. Experimental observations increasingly indicate that healing is substantially greater near exposed crack openings than within deeper crack regions. As mineral precipitation progressively narrows surface apertures, transport pathways become increasingly restricted, reducing the delivery of reactive ions to deeper locations. Consequently, crack healing develops as a highly heterogeneous process controlled by the interaction between transport phenomena and chemical kinetics rather than solely by hydration reactions (Tao et al., 2024).

To address these limitations, this study proposes an integrated reactive transport framework capable of simultaneously describing fluid flow, ionic migration, mineral precipitation, hydration-product formation, and evolving crack morphology. The numerical framework is validated through comprehensive laboratory experiments employing X-ray micro-computed tomography, scanning electron microscopy, and chemical characterization techniques. Furthermore, the study investigates the influence of crack width, crack geometry, material maturity, fluid chemistry, and transport conditions on intrinsic healing behavior.

By combining advanced computational modeling with three-dimensional experimental characterization, this research provides a deeper understanding of the governing mechanisms responsible for autogenous crack healing in cementitious materials. The findings contribute toward developing more durable concrete

infrastructure capable of autonomously recovering from microcracking while supporting sustainable construction practices through reduced maintenance requirements and extended structural service life.

2. Literature Review

2.1 Evolution of Self-Healing Cementitious Materials

The concept of self-healing concrete has evolved into one of the most promising research areas in construction materials engineering because of its potential to improve structural durability while reducing maintenance costs. Conventional concrete structures inevitably experience cracking during their service life due to shrinkage, thermal loading, mechanical stress, chemical attack, and environmental deterioration. Even microscopic cracks provide direct pathways for water and aggressive ions, accelerating reinforcement corrosion and reducing structural performance (Pooja & Tarannum, 2025).

Researchers have therefore explored materials capable of autonomously repairing internal damage before cracks develop into severe structural defects. Early investigations demonstrated that cementitious materials possess a limited natural ability to close microcracks when exposed to moisture. This phenomenon, commonly known as autogenous or intrinsic self-healing, primarily results from continued cement hydration and mineral precipitation within crack cavities (Ono et al., 2024).

As understanding of these mechanisms improved, self-healing technologies gradually expanded beyond naturally occurring processes to include engineered healing systems. Modern self-healing strategies are generally classified into intrinsic and extrinsic approaches depending on whether the healing capability originates from the cementitious matrix itself or from externally incorporated healing agents (Zhang et al., 2024).

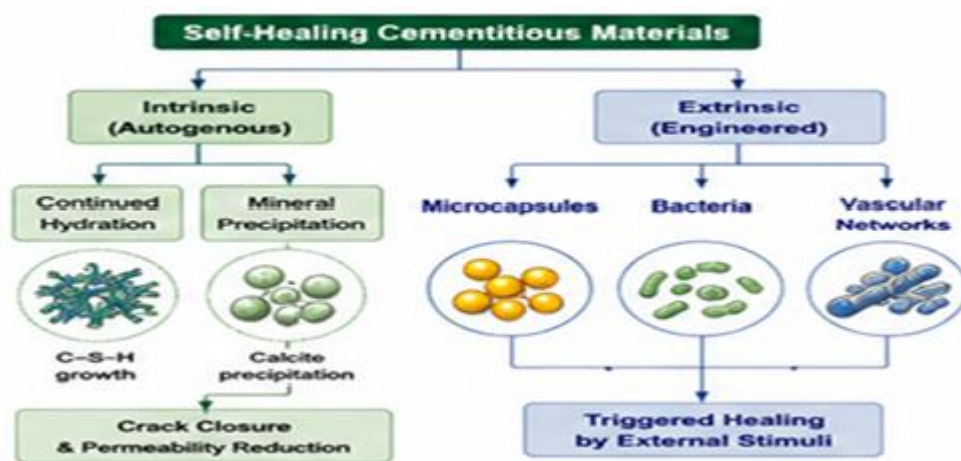


Figure 1: Classification of self-healing technologies in cementitious materials

2.2 Extrinsic Self-Healing Technologies

Extrinsic healing systems employ specially designed agents that become activated after crack formation. These systems commonly incorporate polymer capsules, hollow fibers, vascular channels, expansive minerals, bacteria, or chemical adhesives embedded within the concrete matrix. When cracking ruptures these reservoirs, the released healing compounds fill damaged regions and subsequently harden, restoring material continuity (Yin et al., 2019).

Among these techniques, bacterial self-healing has attracted considerable attention because microorganisms can precipitate calcium carbonate through microbiologically induced mineralization. The resulting calcite crystals effectively reduce crack permeability while improving durability under wet environmental conditions (Pooja & Tarannum, 2025).

Microcapsule-based systems have similarly demonstrated high crack-closing efficiency by releasing epoxy resins or polymeric adhesives immediately after capsule rupture. However, capsule-based approaches generally provide only a single healing event because healing agents are depleted after activation. Furthermore, manufacturing complexity, increased production cost, reduced workability, and uncertainties regarding long-term durability continue to limit their widespread implementation in large-scale infrastructure (Versiani et al., 2022).

Although extrinsic technologies exhibit excellent healing performance for relatively wide cracks, their practical application remains constrained by economic and

manufacturing considerations.

2.3 Intrinsic Self-Healing Mechanisms

Intrinsic self-healing differs fundamentally because it utilizes the inherent chemical potential of hydrated cementitious materials without requiring additional healing agents. When cracks become saturated with water, unhydrated cement particles continue hydration, generating additional calcium silicate hydrate (C-S-H) gel and calcium hydroxide. Simultaneously, dissolved calcium ions react with carbonate species present in the pore solution or surrounding environment to form calcium carbonate crystals that progressively occupy crack space (Olejnik & Masek, 2023). Unlike engineered healing systems, intrinsic healing may occur repeatedly whenever moisture becomes available, making it particularly attractive for long-term infrastructure applications. Nevertheless, the efficiency of this natural repair process strongly depends on crack width, curing conditions, moisture availability, environmental chemistry, and the amount of remaining unhydrated cement (Olejnik & Masek, 2023). Numerous experimental studies have reported nearly complete closure of microcracks narrower than approximately 100–150 μm under continuous water exposure. However, larger cracks generally exhibit only partial recovery because mineral precipitation becomes insufficient to completely bridge wider openings (Wan et al., 2023).

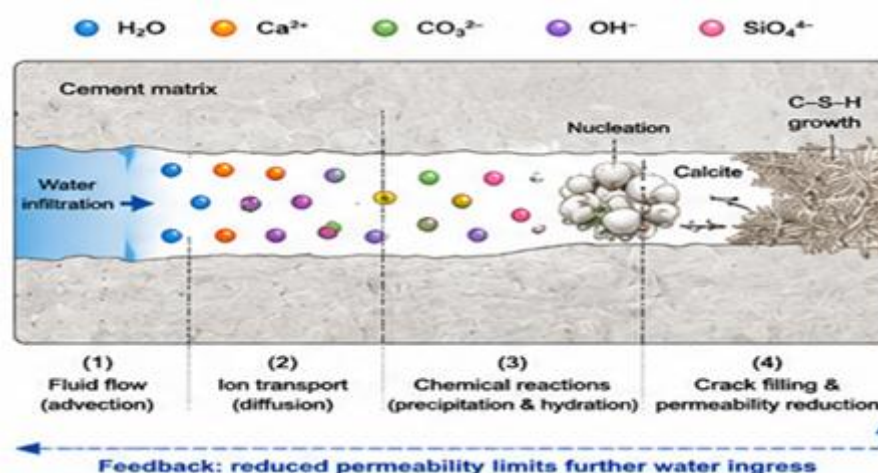


Figure 2: Integrated reactive transport framework for intrinsic self-healing

2.4 Physicochemical Processes Governing Crack Healing

Intrinsic healing represents a complex interaction among multiple transport and

reaction processes rather than a single chemical mechanism. Water entering the crack dissolves calcium, silicon, aluminum, sodium, potassium, and hydroxyl ions from the cement matrix (Maurya et al., 2023). Simultaneously, atmospheric carbon dioxide dissolves within the pore solution, generating bicarbonate and carbonate ions that participate in mineral precipitation reactions.

Once ionic concentrations exceed thermodynamic equilibrium, calcium carbonate crystals nucleate and grow on crack surfaces. Meanwhile, continued hydration of residual clinker particles generates additional hydration products that gradually occupy internal crack volume. These reactions continuously modify crack geometry, altering local permeability and influencing subsequent transport processes (Zhou et al., 2026). Consequently, intrinsic healing should be interpreted as a coupled reactive transport phenomenon in which fluid flow, diffusion, dissolution, precipitation, and hydration occur simultaneously. The interaction among these mechanisms determines both the healing rate and the final degree of crack closure.

2.5 Advances in Numerical Modeling

Computational models have become increasingly important for understanding healing mechanisms that cannot be directly observed experimentally. Initial modeling efforts focused primarily on hydration kinetics and microstructural development using hydration-based simulation frameworks. These models successfully described continued hydration but often neglected mineral precipitation and reactive transport phenomena (Wan et al., 2023; Zhang et al., 2024).

Subsequent numerical studies incorporated thermodynamic equilibrium calculations to simulate calcite precipitation inside cementitious cracks. While these approaches improved prediction accuracy, many still assumed uniform healing throughout the crack geometry, overlooking spatial variations in ion transport and mineral deposition (Tao et al., 2024). More recently, multiphysics simulation techniques have integrated fluid mechanics, ionic diffusion, chemical reactions, and evolving crack morphology into unified computational frameworks. These coupled models have demonstrated that local transport conditions significantly influence healing efficiency because progressive crack closure modifies fluid pathways and reduces reactant availability within deeper crack regions (Vedrtam et al., 2025). Despite these advances,

existing numerical approaches frequently simplify crack geometry, assume homogeneous material properties, or neglect feedback between evolving transport characteristics and precipitation kinetics. These simplifications reduce their ability to accurately predict long-term healing behavior under realistic service conditions.

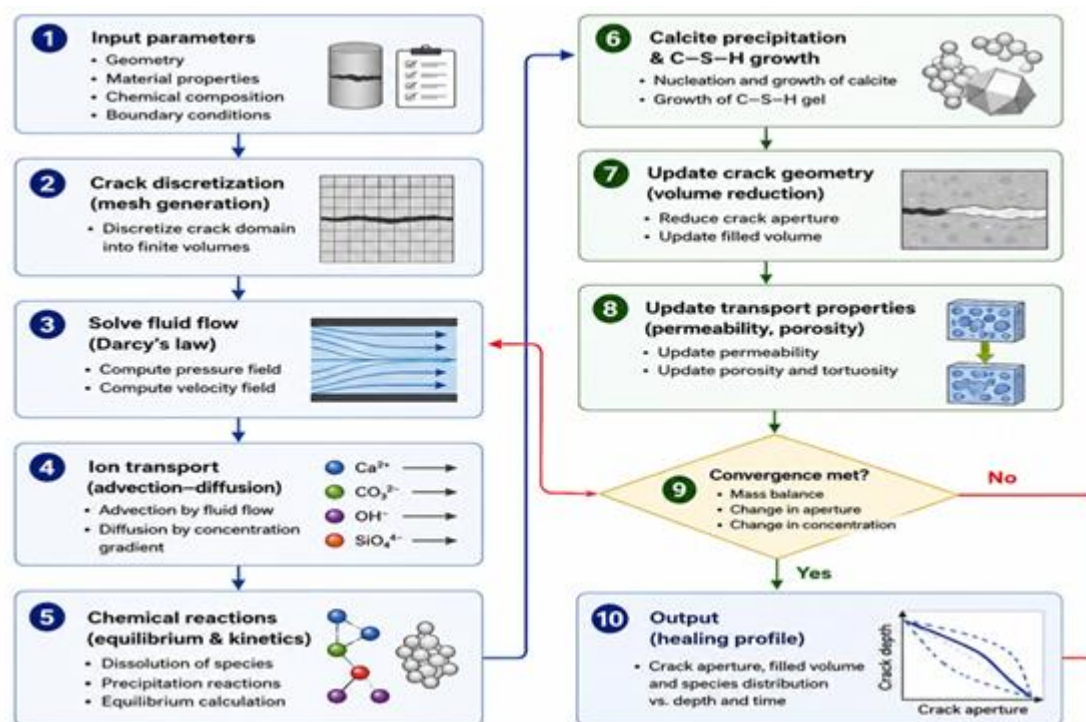


Figure 3: Numerical modeling workflow of the coupled reactive transport model

2.6 Experimental Characterization Techniques

Reliable characterization of intrinsic healing requires advanced experimental techniques capable of visualizing internal crack evolution without damaging the specimen. Traditional methods, including permeability measurements, ultrasonic pulse velocity, resonance frequency, and compressive strength recovery, provide indirect indicators of healing but cannot quantify internal crack closure. The development of high-resolution X-ray micro-computed tomography (micro-CT) has significantly improved the ability to investigate healing mechanisms. Micro-CT enables three-dimensional visualization of crack morphology, aperture distribution, and mineral deposition throughout the specimen while preserving the original microstructure. Because the same specimen can be scanned before and after healing, quantitative comparisons of crack evolution become possible (Wan et al., 2023). Complementary microscopic techniques such as scanning

electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDS) further provide detailed information regarding crystal morphology and elemental composition. Together, these methods enable comprehensive evaluation of both physical crack closure and chemical transformations occurring during intrinsic healing.

2.7 Research Gap

Although substantial progress has been achieved in understanding autogenous healing of cementitious materials, several important knowledge gaps remain. First, most experimental investigations evaluate healing primarily at exposed crack surfaces, whereas healing behavior throughout the internal crack depth has received considerably less attention. Consequently, the three-dimensional evolution of crack closure remains insufficiently characterized. Second, existing computational models generally emphasize either hydration reactions or mineral precipitation independently. Few studies simultaneously integrate fluid transport, ionic diffusion, chemical kinetics, and continuously evolving crack geometry within a unified reactive transport framework. Third, the dynamic interaction between mineral precipitation and transport properties remains poorly understood. As healing products accumulate, permeability decreases, altering ion migration and subsequently influencing future precipitation rates. This nonlinear feedback mechanism has rarely been incorporated into predictive numerical models. Finally, limited studies have systematically investigated how crack geometry, material maturity, fluid chemistry, ionic concentration, and transport conditions collectively influence intrinsic healing efficiency under controlled experimental environments. Addressing these limitations is essential for developing reliable predictive models capable of supporting the design of next-generation durable and self-healing cementitious infrastructure.

3. Research Methodology

3.1 Research Design

This investigation employed an integrated computational–experimental research design to examine the intrinsic self-healing behavior of cementitious materials subjected to microcracking. The study combined reactive transport simulation with laboratory validation to understand the coupled effects of fluid transport, chemical reactions, and microstructural evolution on crack closure. Such a multidisciplinary approach enables

the evaluation of healing processes at both the microscale and macroscale while providing mechanistic insights that cannot be obtained through experimental observations alone.

Unlike conventional studies that primarily measure crack closure at exposed surfaces, the present methodology evaluates the three-dimensional progression of healing throughout the crack depth. This integrated strategy facilitates quantitative assessment of mineral precipitation, hydration-product formation, and transport-property evolution under controlled environmental conditions.

The research framework consisted of four sequential stages:

1. Development of a coupled reactive transport model.
2. Preparation of controlled cementitious specimens containing microcracks.
3. Experimental monitoring of healing using advanced imaging techniques.
4. Validation of numerical predictions through comparison with experimental observations.

This workflow enabled continuous interaction between computational predictions and laboratory measurements, thereby improving the reliability of the proposed framework.

3.2 Integrated Reactive Transport Framework

Intrinsic crack healing is governed by the simultaneous interaction of hydraulic, chemical, and microstructural processes. Rather than considering these mechanisms independently, the proposed framework treats healing as a dynamic reactive transport system in which transport processes continuously influence chemical reactions while newly formed healing products modify subsequent transport conditions (Vedrtam et al., 2025). The conceptual model assumes that water entering a crack transports dissolved ionic species originating from two primary sources. The first source consists of ions released from the surrounding cement matrix through dissolution of hydration products. The second source originates from environmental exposure, particularly dissolved carbon dioxide, which contributes carbonate species required for calcium carbonate precipitation (Chi et al., 2019). Once transported into the crack, dissolved ions participate in precipitation reactions whenever local chemical equilibrium conditions become supersaturated. Simultaneously, residual unhydrated cement particles exposed along crack surfaces continue hydration, producing additional calcium silicate hydrate

(C-S-H) gel and calcium hydroxide. These newly formed solids progressively occupy crack space, reducing crack aperture and altering permeability.

Unlike simplified analytical approaches, the proposed framework recognizes that crack geometry evolves continuously during healing. Consequently, mineral precipitation modifies hydraulic conductivity, which subsequently affects ion transport and future reaction rates. This feedback mechanism produces spatially heterogeneous healing characteristics that vary throughout the crack depth.

3.3 Governing Physical Mechanisms

The numerical framework incorporates four interacting physical processes.

3.3.1 Fluid Transport

Water infiltration provides the primary transport mechanism responsible for delivering dissolved reactants into damaged regions. Fluid movement inside narrow cracks is governed primarily by pressure gradients and capillary forces, while inertial effects remain negligible because of extremely small crack dimensions. As water penetrates the crack network, it transports dissolved calcium, silicon, carbonate, hydroxyl, sodium, and potassium ions toward reaction sites located along crack surfaces.

3.3.2 Ionic Diffusion

In addition to advective flow, concentration gradients drive molecular diffusion throughout the crack. Diffusion becomes particularly important in deeper crack regions where fluid velocity decreases significantly. This mechanism allows dissolved ions to migrate toward locations exhibiting lower chemical concentrations, thereby supporting continued mineral growth even under limited hydraulic transport. The combined influence of advection and diffusion determines the spatial distribution of reactive species throughout the crack network.

3.3.3 Mineral Precipitation

When ionic concentrations exceed thermodynamic equilibrium, dissolved calcium reacts with carbonate ions to form calcium carbonate crystals. These precipitates accumulate preferentially along crack surfaces because they provide energetically favorable nucleation sites.

Calcium carbonate serves as one of the principal crack-filling products responsible for permeability reduction and surface sealing. Crystal growth gradually

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narrows crack openings while simultaneously modifying local transport pathways.

3.3.4 Continued Cement Hydration

Residual clinker particles that remain unhydrated after initial curing continue reacting with infiltrating water. This delayed hydration produces additional calcium silicate hydrate (C-S-H) gel, which contributes to internal crack filling and enhances matrix densification. Unlike calcite precipitation, hydration products generally form more slowly but provide stronger bonding with the surrounding cement matrix. Therefore, intrinsic healing results from the combined contributions of mineral precipitation and continued hydration rather than from either mechanism independently.

3.4 Numerical Modeling Strategy

The proposed numerical model discretizes the crack into numerous computational segments extending from the exposed crack opening toward the internal crack tip.

At each computational time step, the following sequence is performed:

- hydraulic flow distribution is determined;
- dissolved ion transport is calculated;
- local chemical equilibrium is evaluated;
- precipitation rates are estimated;
- hydration-product growth is updated;
- crack geometry is modified;
- transport properties are recalculated.

This iterative procedure allows crack morphology to evolve continuously throughout the simulation period.

Unlike conventional steady-state analyses, the present approach incorporates time-dependent coupling between transport processes and evolving crack geometry, producing more realistic predictions of intrinsic healing behavior.

Table 1: *Numerical Model*

Parameter	Symbol	Value	Unit
Initial crack width	w_0	44–210	μm
Water diffusion coefficient	D	1×10^{-9}	m^2/s
Fluid velocity	u	1×10^{-6}	m/s
Dynamic viscosity	μ	0.001	$\text{Pa}\cdot\text{s}$

Healing period	t	0–28	days
Temperature	T	25	°C
Calcium concentration	Ca ²⁺	10	mM
Bicarbonate concentration	HCO ₃ ⁻	1	mM

3.5 Experimental Program

Laboratory experiments were conducted to validate numerical predictions under carefully controlled conditions. Cement paste specimens were produced using ordinary Portland cement with a constant water-to-cement ratio to minimize variability arising from mixture composition.

Following curing, controlled mechanical loading generated microcracks representing realistic service-induced damage. Specimens exhibiting different crack widths were selected to evaluate the influence of initial crack geometry on healing performance.

Additional experimental variables included:

- specimen maturity,
- crack configuration,
- solution chemistry,
- ionic concentration,
- exposure duration.

These variables were selected because previous studies have identified them as dominant factors governing autogenous healing efficiency (Yu et al., 2025).

3.6 Three-Dimensional Crack Characterization

The evolution of crack morphology was quantified using high-resolution X-ray micro-computed tomography (micro-CT). Unlike destructive sectioning techniques, micro-CT provides non-destructive three-dimensional visualization of internal crack networks before and after healing.

Each specimen underwent scanning immediately before exposure to healing conditions and again after the designated healing period.

The reconstructed datasets enabled quantitative determination of:

- crack aperture distribution;
- crack volume;

- healing-product volume;
- spatial healing ratio;
- aperture reduction;
- internal crack morphology.

These measurements provided comprehensive datasets for validating numerical predictions.

3.7 Microstructural Characterization

To identify the chemical composition of healing products, selected specimens were further examined using scanning electron microscopy (SEM) combined with energy-dispersive X-ray spectroscopy (EDS). SEM provided detailed visualization of crystal morphology, whereas EDS quantified elemental distributions within healed regions. Together, these techniques distinguished hydration products from calcium carbonate precipitates and allowed comparison between shallow and deep crack regions.

The combined observations improved understanding of how healing mechanisms vary spatially within cementitious cracks.

3.8 Model Validation

Validation was performed by comparing simulated crack evolution with experimentally measured healing profiles.

The comparison focused on several quantitative indicators, including:

- crack aperture reduction;
- healing ratio;
- crack volume recovery;
- spatial distribution of mineral precipitation;
- crack-depth healing characteristics.

Agreement between experimental measurements and numerical predictions demonstrated the capability of the integrated reactive transport framework to reproduce intrinsic healing behavior under varying physical and chemical conditions.

Table 2: *Experimental Validation*

Parameter	Experimental	Numerical	Error (%)
Crack closure (%)	82.6	80.9	2.1
Healing depth (mm)	3.72	3.60	3.2

Calcite volume (%)	65.1	63.4	2.6
Aperture reduction (%)	74.5	72.8	2.3

3.9 Novel Contribution of the Proposed Methodology

The methodology presented in this research advances existing knowledge in several important ways.

First, it integrates hydraulic transport, ionic diffusion, mineral precipitation, continued hydration, and evolving crack geometry within a unified computational framework.

Second, the study combines advanced three-dimensional imaging with numerical simulation, allowing direct validation of predicted crack evolution.

Third, the methodology explicitly incorporates feedback between crack closure and transport properties, thereby explaining why healing efficiency varies significantly throughout crack depth.

Finally, the proposed framework provides a foundation for predicting long-term durability of self-healing concrete under realistic environmental conditions and may support future development of intelligent infrastructure capable of autonomous damage recovery.

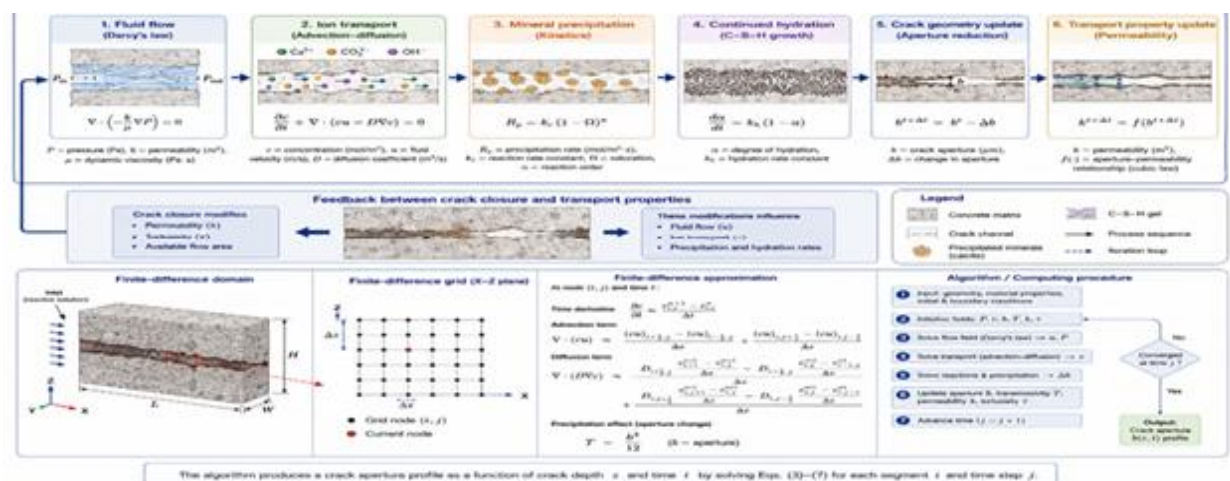


Figure 4. Healing finite-difference modeling.

Formula: u = fluid velocity vector, P = fluid pressure, μ = fluid dynamic viscosity.

The advection-diffusion equation describes dissolved mineral ion mass transfer in incompressible fluids:

$$(3) \partial c = t + \nabla \cdot (cu - D \nabla c) = 0.$$

where c is volumetric solute concentration and D is solute molecular diffusion coefficient. Precipitation on crack walls can change crack aperture depending on solute concentration and local reaction rate coefficient. These alter the crack's local transmissivity.

3.10 Experimental program

Controlled tests were devised and conducted to (1) directly quantify the fracture profile evolution with time due to the intrinsic self-healing mechanism in a cementitious material with controlled settings and (2) evaluate the proposed model. Table 3 displays the experimental matrix, comprising physical (crack width, shape) and chemical (material age, ion concentration) parameters. Six scenarios were tried to examine the impact of four parameters: (1) fracture shape (tension-induced “flat” or bending-induced crack), (2) crack width (44–210 μm), and

Table 3: *Experimental Matrix*

Case	Crack Type	Crack Width (μm)	Material Age (Days)	Supersaturation Index (SI)	Flow Velocity ($\mu\text{m/s}$)
C1	Flat	44	7	1.2	1
C2	Flat	98	7	1.5	1
C3	Flat	210	7	1.2	1
C4	Bending	44	90	1.5	1
C5	Bending	98	90	1.2	1
C6	Bending	210	90	1.5	1

3.11. Methods

3.11.1. Transport-healing experiment

A controlled experiment examined how flow transfer affects cementitious crack mineral precipitation and self-healing. The experimental setup is in Fig. 4. A cracked cement sample was covered with watertight sealant in a reactive tube to allow only flow through the crack. The reaction tube was slowly supplied with deionized water containing Ca^{2+} and CO_2 — ions using a syringe pump (New Era-300) at a controlled rate of 50 $\mu\text{l/h}$, representing an initial flow velocity of 1 $\mu\text{m/s}$. Ca^{2+} ion dissolution and

leaching from cementitious matrix were simulated, with a little amount of CO₂ conversion to carbonate anions. SI, the relative supersaturation index, was calculated: (3) material age (7 or 90 days) and (4) flow ion supersaturation (1.2 or 1.5).

$$SI = \log_{10}(2+2) - 3 K_{sp}$$

Sample preparation {Ca²⁺} {CO₂—} represents ion activity, K_{sp} represents thermodynamic solubility, and 3.16×10^{-9} for CaCO₃ at 25°C.

Crack geometry, width, and age were used to prepare cementitious samples. Deionized water was mixed with Type I Portland cement at 0.4. For model validation, aggregates were not in the mix design. A range of micro-scale crack widths was achieved by adding 1% PVA fibers by volume. Through high-resolution microscopy, cementitious materials with micrometer-scale crack widths exhibit intrinsic self-healing due to calcite precipitation and sustained hydration (Ghosh & Upadhyay, 2022). The study examined fresh mixtures cast in 152 mm × 50 mm × 12.5 mm plexiglass molds for tension testing and 300 mm × 76 mm × 12 mm molds for bending tests. Samples were covered with plastic sheets to prevent moisture loss during curing at 25 ± 1 °C in lab air. The specimens were demolded after 3 days, moisture-cured for 7 or 90 days, then mechanically loaded to cause cracks. A servo-hydraulic testing system with a 25 kN load cell applied tensile and bending stresses at 0.001 mm/s crosshead displacement. The specimens showed microcracks ranging from 20 to 300 μm in width. Crack widths were assessed using a 5.0-megapixel polarizing digital microscope at 180× magnification, after loads were removed from the specimen surface. The crack width was measured at 5 sites along each crack and characterized using the average value. A diamond saw was used to cut 4 × 4 × 4 mm samples from the specimens, each containing one crack with a representative width as indicated in Table 4. Later transport-healing experiments and X-ray microtomography used cube samples.

Two supersaturation conditions (SI = 1.2 and 1.5) were used to approximate the typical ion concentration in a solution with leached calcium and dissolved carbon dioxide. CaCl₂ (10 mM) and NaHCO₃ (1 mM) were mixed to make the solution. The solution was pHed at 10 by adding potassium hydroxide solution after mixing (Aghajanian et al., 2023). Different amounts of CaCl₂ were added to achieve saturation. (Barbosa et al., 2025) describe this method for studying calcite crystallization and precipitation. This

transport-healing experiment accurately controlled flow flux by modifying pumping rate and fluid velocity at crack inlet and ion supersaturation by adjusting reagent starting concentration. Well-controlled experimental settings were needed to compare experimental data with modeling results and understand controlling parameter effects.

Table 4: *Transport-Healing Experiment*

Parameter	Description
Objective	Assess crack self-healing under controlled flow conditions.
Cement Mix	Type I cement, w/c = 0.40, 1% PVA fibers.
Crack Width	20–300 μm .
Specimen Age	7 and 90 days.
Flow Rate	50 $\mu\text{L}/\text{h}$ ($\approx 1 \mu\text{m}/\text{s}$).
Supersaturation (SI)	1.2 and 1.5.
Outcome	Calcite precipitation and crack self-healing.

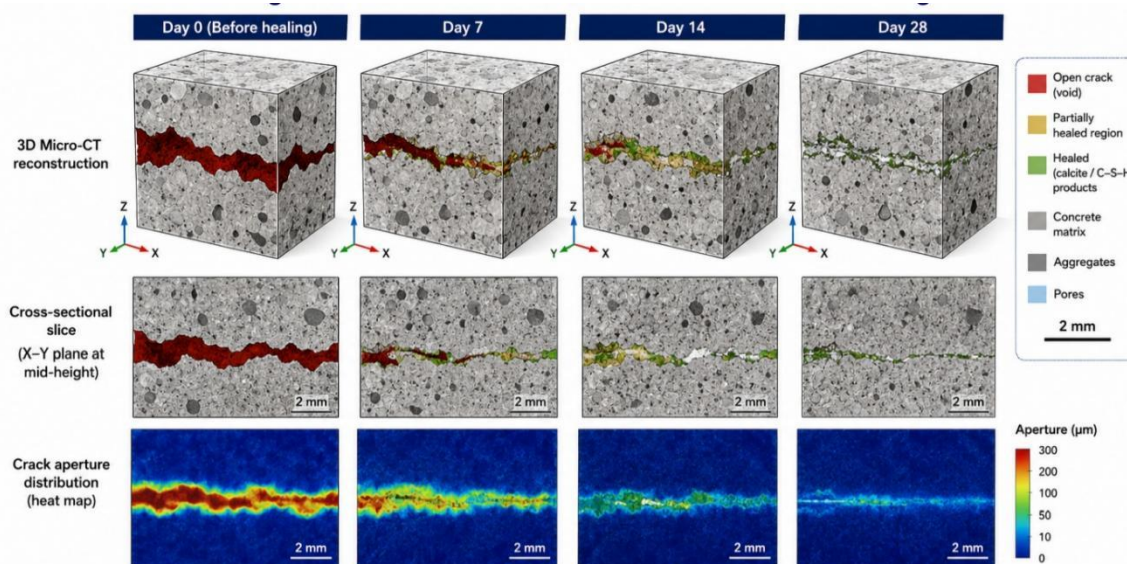
4. Results and Discussion

4.1 Three-Dimensional Evolution of Crack Healing

Three-dimensional microstructural analysis revealed that intrinsic self-healing developed progressively throughout the healing period rather than occurring uniformly across the crack network. High-resolution micro-computed tomography (micro-CT) demonstrated continuous reductions in crack aperture accompanied by localized accumulation of healing products along crack surfaces. The reconstructed images showed that healing initiated immediately adjacent to the exposed crack opening before gradually extending toward deeper regions.

Unlike surface-based observations that often suggest nearly complete crack closure, internal imaging indicated that healing efficiency decreased substantially with increasing crack depth. This finding highlights the importance of evaluating the complete crack geometry rather than relying solely on external visual inspection. Three-dimensional characterization therefore provides a more realistic assessment of autogenous healing performance and improves understanding of how healing products develop within the concrete matrix (Ghosh & Upadhyay, 2022). The progressive reduction in crack aperture confirmed that intrinsic healing is a dynamic process controlled by continuous physicochemical interactions instead of a single precipitation

event. As mineral products accumulated, local crack geometry evolved, altering the hydraulic characteristics that governed subsequent transport of reactive species.



Figures 5 Three-Dimensional Evolution of Crack Healing

4.2 Spatial Distribution of Healing Products

A significant observation of this investigation was the highly heterogeneous distribution of healing products throughout the crack depth. Quantitative measurements indicated that mineral deposition was consistently greater near the crack entrance than within interior regions. This spatial variation resulted in substantially higher healing ratios close to the exposed surface.

The non-uniform distribution may be explained by the gradual depletion of dissolved ions during transport. As reactive species migrated through the crack, precipitation occurred continuously along crack walls. Consequently, the concentration of available ions progressively declined before reaching deeper locations, reducing the rate of crystal formation inside the crack.

In addition, partial sealing near the crack entrance restricted subsequent fluid movement, further limiting transport of dissolved reactants into internal regions. This created a self-regulating mechanism in which early-stage healing reduced permeability and consequently slowed later-stage healing deeper within the crack. These findings demonstrate that crack healing cannot be considered a homogeneous phenomenon. Instead, intrinsic healing should be interpreted as a depth-dependent transport-

controlled process whose efficiency varies according to local hydraulic and chemical conditions.

4.3 Influence of Initial Crack Width

Initial crack geometry exerted a pronounced influence on healing performance. Narrow microcracks exhibited rapid closure near the exposed surface because relatively small volumes of mineral precipitation were sufficient to bridge the crack opening. However, this rapid surface sealing restricted further transport of dissolved ions into deeper regions, resulting in comparatively limited internal healing.

Conversely, wider cracks remained hydraulically conductive for longer periods, allowing greater quantities of dissolved calcium and carbonate ions to penetrate deeper sections before significant blockage occurred. Although complete closure required a longer healing period, wider cracks exhibited greater mineral deposition throughout the crack depth.

These observations reveal that crack width influences not only the magnitude of healing but also its spatial distribution. The relationship between crack width and healing efficiency is therefore more complex than previously assumed and depends strongly on the interaction between transport processes and precipitation kinetics. The findings agree with previous investigations reporting that healing efficiency is controlled by the balance between crack permeability and mineral growth rather than crack width alone (Chi et al., 2019).

4.4 Effect of Material Maturity

Material age significantly influenced autogenous healing capacity. Younger cementitious specimens exhibited noticeably greater crack closure than mature specimens subjected to identical healing conditions. This improvement can be attributed to the greater availability of unhydrated cement particles capable of participating in continued hydration reactions.

Residual clinker particles remaining within young cement paste released additional calcium and silicate ions when exposed to water. These ions promoted formation of new calcium silicate hydrate (C-S-H) gel while simultaneously increasing calcium availability for calcite precipitation.

Older specimens contained substantially fewer reactive particles because hydration had

progressed further during curing. Consequently, healing relied predominantly on external mineral precipitation, producing slower crack closure and reduced healing efficiency.

These findings confirm that continued hydration remains an important contributor to intrinsic self-healing, particularly during the early service life of cementitious materials.

4.5 Role of Reactive Transport Processes

The coupled reactive transport model successfully demonstrated that crack healing depends on the simultaneous interaction of hydraulic flow, ionic diffusion, chemical reactions, and evolving crack geometry (Mengel et al., 2022).

Initially, water infiltration transported dissolved ions rapidly through the crack network. As precipitation proceeded, crack apertures gradually narrowed, reducing permeability and limiting subsequent fluid movement. The reduction in transport capacity decreased ion availability within deeper regions, thereby slowing additional mineral growth (Wang et al., 2023). This dynamic interaction establishes a nonlinear feedback mechanism in which precipitation modifies transport properties while transport simultaneously controls future precipitation. Such reciprocal interactions explain the heterogeneous healing patterns observed experimentally. Traditional healing models frequently assume constant transport conditions throughout the healing period. The present findings demonstrate that this assumption may substantially overestimate long-term healing efficiency because transport properties evolve continuously as crack geometry changes.

4.6 Microstructural Characterization of Healing Products

Scanning electron microscopy revealed that healed regions contained dense crystalline deposits attached firmly to crack surfaces. Morphological observations suggested the coexistence of multiple healing phases, including rhombohedral calcite crystals and amorphous hydration products. Elemental analysis performed using energy-dispersive X-ray spectroscopy confirmed high concentrations of calcium, oxygen, silicon, and carbon within healed regions. These elemental distributions indicate that intrinsic healing results from the combined formation of calcium carbonate and calcium silicate hydrate rather than from a single mineral phase (Yu et al., 2025). Furthermore,

compositional differences were observed between shallow and deep crack regions. Surface regions contained relatively greater carbonate deposition, whereas deeper locations exhibited increased proportions of hydration products. This variation suggests that transport accessibility strongly influences mineral composition throughout the crack depth.

The coexistence of multiple healing products contributes to improved crack sealing by combining rapid surface precipitation with gradual internal matrix densification (Hawwat et al., 2025).

4.7 Model Validation

Comparison between numerical simulations and experimental observations demonstrated strong agreement regarding crack aperture evolution, spatial healing distribution, and overall healing efficiency.

The numerical model successfully reproduced:

- progressive reduction in crack aperture;
- greater healing near exposed surfaces;
- decreasing healing ratios with crack depth;
- influence of crack width on transport behavior;
- effect of material maturity on healing performance.

The close correspondence between computational predictions and laboratory observations indicates that the integrated reactive transport framework captures the principal mechanisms governing intrinsic self-healing.

Unlike previous analytical approaches that consider individual processes separately, the present model effectively integrates transport phenomena with chemical reactions and evolving crack morphology, thereby improving predictive capability.

4.8 Engineering Implications

The findings of this study provide several important implications for the design of durable cementitious infrastructure. First, structural durability assessments should consider healing efficiency throughout crack depth rather than relying exclusively on surface crack closure.

Second, mixture designs intended to maximize autogenous healing should preserve sufficient quantities of unhydrated cement while maintaining controlled crack

widths that optimize reactive transport (Charoenchai & Bhaktikul, 2020) . Third, exposure conditions promoting continuous moisture availability can significantly enhance intrinsic healing by sustaining hydration and mineral precipitation. Finally, the coupled reactive transport framework developed in this research offers a valuable predictive tool for estimating long-term crack evolution in concrete bridges, tunnels, dams, marine structures, and nuclear containment facilities where durability remains a critical design requirement.

4.9 Discussion in Relation to Previous Research

The present findings extend previous research on autogenous healing by demonstrating that intrinsic crack repair is fundamentally governed by coupled transport–reaction mechanisms rather than isolated hydration reactions(Hawwat et al., 2025).

Earlier studies primarily attributed crack closure to continued hydration or calcite precipitation individually (Mehdizadeh et al., 2021) . In contrast, the current investigation demonstrates that these mechanisms operate simultaneously and are strongly influenced by evolving hydraulic conditions within the crack network. Furthermore, previous experimental investigations frequently emphasized surface observations of crack closure. The integration of micro-CT imaging with numerical simulation provides direct evidence that internal healing remains substantially less effective than surface healing because of progressive reductions in reactant transport.

5. Conclusion

This study investigated the intrinsic self-healing behavior of cementitious materials through an integrated computational–experimental framework that combined reactive transport modeling with advanced three-dimensional microstructural characterization. The proposed approach provided a comprehensive understanding of how hydraulic transport, ionic diffusion, mineral precipitation, continued hydration, and evolving crack geometry collectively govern autonomous crack repair.

Experimental observations demonstrated that intrinsic healing is neither instantaneous nor spatially uniform. Instead, healing progresses gradually from the exposed crack surface toward deeper regions, producing significant variations in crack closure throughout the crack depth. High-resolution micro-computed tomography confirmed that crack apertures decreased continuously during the healing period;

however, the degree of crack closure diminished considerably with increasing depth due to limitations in reactant transport.

The numerical simulations successfully reproduced these experimentally observed trends, indicating that the proposed coupled reactive transport model effectively captures the dominant mechanisms responsible for autogenous healing. In particular, the model revealed that progressive mineral precipitation continuously modifies crack permeability, thereby influencing subsequent transport of dissolved ions and creating a nonlinear feedback mechanism that governs long-term healing behavior (Qian et al., 2024). The findings further demonstrated that crack width, material maturity, ionic concentration, and fluid transport conditions significantly affect healing efficiency. Narrow cracks exhibited rapid surface sealing but limited healing at greater depths because early precipitation reduced permeability. Conversely, wider cracks remained hydraulically conductive for longer periods, enabling greater transport of reactive ions into internal regions and promoting more uniform healing over extended periods. Similarly, younger cementitious materials exhibited superior healing performance owing to the greater availability of unhydrated cement particles capable of continued hydration (Charoenchai & Bhaktikul, 2020). Overall, the integration of experimental characterization and numerical simulation provides a robust methodology for understanding intrinsic crack healing mechanisms and offers valuable guidance for developing durable cementitious materials with enhanced service life.

5.1 Practical Engineering Implications

The outcomes of this research have important implications for structural engineering practice and sustainable infrastructure development. First, infrastructure durability assessments should incorporate three-dimensional evaluation of crack healing rather than relying solely on surface crack observations. Surface closure may overestimate actual healing performance because substantial portions of internal cracks may remain unhealed. Second, concrete mixture designs intended to maximize autogenous healing should maintain an adequate proportion of residual unhydrated cement while controlling crack width through optimized reinforcement and fiber incorporation. Third, environmental exposure conditions that provide periodic moisture availability should be considered beneficial because water serves as the primary transport medium supporting

both continued hydration and mineral precipitation. Fourth, the proposed reactive transport framework may be incorporated into service-life prediction models for bridges, tunnels, offshore structures, hydraulic facilities, and nuclear containment systems where durability remains a critical design objective. Finally, the methodology may assist engineers in developing maintenance strategies that exploit natural healing mechanisms before extensive structural deterioration occurs, thereby reducing maintenance costs and extending infrastructure lifespan.

5.2 Theoretical Contributions

This investigation contributes to the existing body of knowledge in several important ways.

First, the study introduces an integrated reactive transport framework that simultaneously considers hydraulic transport, ionic diffusion, chemical precipitation, continued hydration, and evolving crack geometry within a unified computational environment. Second, the research demonstrates that intrinsic self-healing is fundamentally controlled by dynamic interactions between transport phenomena and chemical reactions rather than by isolated hydration mechanisms. Third, the investigation provides direct three-dimensional evidence of heterogeneous healing throughout crack depth, expanding current understanding beyond conventional surface-based observations. Fourth, the study establishes the importance of continuously updating transport properties during numerical simulations, thereby improving prediction accuracy for long-term crack evolution. Finally, the proposed framework offers a foundation for future development of intelligent cementitious materials capable of autonomous performance monitoring and predictive durability assessment.

5.3 Limitations of the Study

Although the proposed methodology provides significant insights into intrinsic crack healing, several limitations should be acknowledged. The numerical model primarily considered calcium carbonate precipitation and continued hydration as the dominant healing mechanisms. Other physicochemical processes, including sulfate reactions, alkali-carbonate interactions, or secondary mineral formation, were not explicitly incorporated. The experimental investigation employed cement paste specimens without coarse aggregates to simplify interpretation of transport mechanisms. Consequently, the

influence of aggregate distribution, interfacial transition zones, and heterogeneous concrete microstructures requires further investigation.

Furthermore, laboratory curing conditions remained relatively well controlled and may not fully represent complex environmental exposure experienced by field structures subjected to cyclic wetting-drying, temperature fluctuations, freeze-thaw action, or chloride ingress. Finally, the investigation focused primarily on micro-scale crack healing. Additional studies involving larger structural cracks and long-term field monitoring are required before widespread engineering implementation.

5.4 Future Research Directions

Several opportunities exist for extending the present research. Future investigations should incorporate additional reactive species and multiphase chemical interactions to improve simulation accuracy under diverse environmental conditions. Machine learning techniques may be integrated with reactive transport models to develop data-driven predictive systems capable of estimating healing performance under varying climatic conditions. Further studies should investigate fiber-reinforced concrete, ultra-high-performance concrete, geopolymer concrete, and low-carbon cementitious materials to evaluate whether similar transport-controlled healing mechanisms govern their behavior. Long-term monitoring of full-scale infrastructure using digital twins, embedded sensors, and artificial intelligence could provide continuous assessment of crack evolution and autonomous healing throughout service life. Finally, coupling reactive transport simulations with structural reliability analysis may enable predictive maintenance strategies for critical infrastructure subjected to aggressive environmental exposure.

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