Discover Concrete and Cement

Review

Insights into cementitious materials: exploring atomic structures and interactions

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Abstract
Understanding cement at the atomic level is essential for optimizing its performance, durability, and sustainability. This review explores the atomic structure and defects of key cement phases—C-S-H, C_3S, C_2S, C_3A, and C_4AF—and their impact on hydration reactions and phase formation. Calcium silicate hydrate (C-S-H), the primary hydration product, is examined for its amorphous structure, which governs strength and durability. Structural defects, including vacancies and dislocations in cement phases, influence hydration kinetics and mechanical properties. Interactions at the atomic scale, such as hydration reactions driven by water molecules and the formation of C-S-H gel, are discussed alongside the role of temperature and pressure in accelerating or decelerating these processes. Insights into atomic arrangements, defects, and environmental conditions provide pathways to improve cement formulations, enhance durability, and reduce environmental impact. This comprehensive review deepens the understanding of cement's fundamental behaviour, enabling advancements in sustainable and high-performance cementitious materials.

Keywords Cementitious materials · Atomic structure · Calcium silicate hydrate (C-S-H) · Hydration reactions

1 Introduction

Understanding cement at the atomic level is critical for enhancing its performance, durability, and long-term sustainability in construction applications. Cement is composed of several key components, including calcium silicates, aluminates, and ferrites, whose atomic arrangements and interactions during hydration dictate the material's strength, stability, and overall behaviour [114, 117]. These hydration reactions, initiated by water molecules, lead to the formation of critical phases such as calcium silicate hydrate (C-S–H), which serves as the primary binding phase in concrete. The ability to analyze the atomic structure and associated defects within these phases provides deeper insight into the mechanisms governing cementitious material properties, allowing for significant advancements in cement formulation and optimization.

The atomic-level understanding of cement phases, such as tricalcium silicate (C_3S), dicalcium silicate (C_2S), tricalcium aluminate (C_3A), and tetracalcium aluminoferrite (C_4AF), highlights their individual roles in influencing cement hydration and performance. For instance, C_3S and C_2S are primarily responsible for strength development through their reactions with water to produce C-S–H and calcium hydroxide [98, 153]. C_3S contributes to early strength due to its high reactivity, while C_2S reacts more slowly, enhancing long-term strength [70]. Tricalcium aluminate (C_3A), with its cubic crystal structure, influences the setting time of cement and is particularly reactive with water, while C_4AF, which adopts an

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orthorhombic structure, provides moderate sulphate resistance and impacts the colour of cement [47, 88, 99]. These phases' atomic arrangements are critical for their reactivity and the resulting hydration products.

Defects and structural disorder at the atomic scale are equally significant in determining cement performance. Imperfections such as vacancies, dislocations, and variable cation distributions arise during the production and hydration of cement phases. For example, the amorphous or poorly crystalline nature of C-S-H makes it prone to structural variability, including irregularities in calcium-silicate layers and interlayer water distributions [104, 113, 152]. These defects can influence mechanical properties, load-bearing capacity, and durability. Similarly, the monoclinic structure of C_3S may include chain disruptions and vacancies that affect its reactivity, while the β-polymorph of C_2S exhibits inconsistencies that impact hydration kinetics [132, 153]. Understanding and addressing these defects through optimized manufacturing techniques, additives, and admixtures are essential for improving material strength, stability, and resistance to environmental challenges.

Recent advances in understanding cement at the atomic level have significantly enhanced our knowledge of its hydration processes, structural characteristics, and mechanical properties. These insights are pivotal for developing more sustainable and durable cementitious materials. One notable advancement involves the use of molecular dynamics simulations with reactive force fields to investigate the initial hydration processes of tricalcium silicate (C_3S) and dicalcium silicate (C_2S) [8, 108, 136]. These simulations have elucidated the rapid initial hydration of C_3S compared to C_2S, providing a molecular-level understanding of calcium ion dissolution pathways. Such knowledge is essential for designing environmentally friendly cements with reduced carbon emissions. Another significant development is the application of solid-state nuclear magnetic resonance (NMR) spectroscopy in studying cementitious materials [15, 126]. This technique has enabled researchers to characterize the atomic structure of calcium silicate hydrate (C-S-H), the primary hydration product of cement. For instance, studies have revealed that the addition of zinc enhances the mechanical strength of tricalcium silicates upon hydration [29, 93]. The incorporation of zinc leads to the formation of new silicon species, where zinc substitutes specific silicon sites in the C-S-H structure, thereby modifying its properties. Furthermore, the development of realistic molecular models of cement hydrates has provided atomistic-level structural insights into C-S-H. These models, developed from a bottom-up perspective and validated against experimental data, have improved our understanding of the structural organization and mechanical behaviour of C-S-H at the nanoscale. Collectively, these advancements at the atomic level are instrumental in guiding the design of next-generation cementitious materials with improved performance and sustainability.

Interactions at the atomic scale, particularly during hydration reactions, are fundamental to cement's functionality. Water molecules, driven by their polar nature, interact with cement components to dissolve ions and facilitate the formation of C-S-H [35, 155, 164]. This phase, composed of silicate tetrahedral chains interspersed with calcium ions and water, forms a highly flexible and amorphous gel-like network. The resulting C-S-H microstructure controls the strength, porosity, and overall performance of cement paste. The influence of external factors such as temperature and pressure also play a critical role at the atomic level, as they can accelerate or decelerate hydration, modify microstructure density, and impact mechanical properties [62, 166, 172]. This review provides a comprehensive analysis of cementitious materials at the atomic scale, exploring how their chemical composition, crystal structures, and inherent defects influence hydration reactions, phase formation, and key mechanical properties. By understanding these processes in detail, researchers can develop improved cement formulations that are more durable, sustainable, and tailored to specific construction requirements. Such advancements are essential for addressing the growing demand for high-performance and environmentally friendly cementitious materials in modern construction.

2 Atomic structure of cement phases

2.1 Crystal structures of C-S–H, C_3S, C_2S, C_3A, and C_4AF

The crystal structures of essential cement phases—Calcium-Silicate-Hydrate (C-S–H), Tricalcium Silicate (C_3S), Dicalcium Silicate (C_2S), Tricalcium Aluminate (C_3A), and Tetracalcium Aluminoferrite (C_4AF)—play a fundamental role in determining cement's mechanical and chemical properties. These phases influence critical attributes, including strength development, durability, and the overall performance of cementitious materials.

Calcium-Silicate-Hydrate (C-S–H), the most significant hydration product, serves as the primary binding phase in cement. Unlike other phases, C-S–H lacks a well-defined crystalline structure and is instead described as amorphous or poorly crystalline [23, 91, 112]. Its atomic-scale arrangement consists of layered calcium silicate sheets, with interlayer spaces filled by calcium ions and water molecules [51, 80, 140]. This structure provides C-S–H with flexibility, enabling it to accommodate varying compositions and water contents. These attributes are essential for the material's binding properties, as they allow C-S-H to act as the "glue" that binds aggregate particles together in concrete. Its amorphous nature, however, results in variability in mechanical properties, making it sensitive to defects and hydration conditions.

Tricalcium Silicate (C_3S), a key cementitious phase, crystallizes in a monoclinic structure and is known for its high reactivity. This phase is the most significant contributor to the early strength development of cement, reacting rapidly with water to produce C-S-H and calcium hydroxide (CH). The atomic structure of C_3S features chains of silicate tetrahedra that are linked together through calcium ions [71, 111]. This chain-like arrangement promotes reactivity by facilitating the release of calcium and silicate ions during hydration. The monoclinic crystal symmetry of C_3S, although stable, can include defects such as vacancies or dislocations, which influence its reactivity and hydration kinetics. Dicalcium Silicate (C_2S) is another calcium silicate phase that is typically found in its β-polymorph, which also adopts a monoclinic crystal structure. In comparison to C_3S, C_2S exhibits slower hydration kinetics, contributing primarily to the long-term strength of cement [41, 134, 144]. The atomic arrangement of β-C_2S closely resembles that of C_3S, with silicate tetrahedra interconnected through calcium ions. However, its lower reactivity is attributed to differences in the stability and energy required to dissolve its crystal lattice during hydration. Over time, the slower release of calcium and silicate ions from C_2S ensures a gradual development of strength, improving cement's durability. Tricalcium Aluminate (C_3A) is a critical phase that features a cubic crystal structure. It reacts rapidly with water, significantly influencing the early setting time of cement. The cubic symmetry of C_3A results in a relatively open structure, which facilitates the diffusion of water molecules and accelerates hydration [47, 97]. However, this rapid reaction can lead to flash setting, a phenomenon where cement hardens prematurely [24, 83]. The addition of gypsum mitigates this effect by forming ettringite, which moderates the reactivity of C_3A and extends the setting time.

Figure 1 illustrates the atomic structure and surface configuration of a crystal supercell, particularly showing the cleavage of the (0 4 0) surface. The top-left diagram highlights the atomic arrangement within a 4 × 4x2 supercell, where red spheres represent oxygen atoms and green spheres likely depict calcium or silicon atoms. The top-right image displays the cleaving process to expose the (0 4 0) surface. The bottom-left section provides the dimensions of the cleaved surface, measuring 48.73 Å × 50.54 Å. Finally, the bottom-right zoomed-in view presents the orderly atomic arrangement of the cleaved (0 4 0) plane, revealing its structural periodicity and symmetry.

Figure 2 depicts the atomic structure of a tricalcium aluminate (C_3A) unit cell, a key phase in cement. The cubic crystal structure is clearly represented, with different atoms color-coded: red spheres for oxygen (O), pink tetrahedra for aluminum (Al), and green spheres for calcium (Ca). The oxygen atoms form interconnected frameworks around the aluminum tetrahedra, which serve as the primary structural units. Calcium ions (green) are distributed within the structure, balancing the negative charge of the aluminum-oxygen network. This arrangement creates an open, symmetrical cubic lattice that facilitates water diffusion, enabling rapid hydration reactions during cement setting. The uniform distribution of calcium ions is critical for stabilizing the crystal lattice. This structure plays a significant role in influencing the early setting time of cement, as C_3A reacts quickly with water. Understanding its atomic arrangement helps optimize cement performance and mitigate flash setting issues through the addition of gypsum.

2.2 Defects and disorder in cement phases

Defects and structural disorder in cement phases play a significant role in influencing the material's performance, mechanical behaviour, and long-term durability. These imperfections often originate during production processes, including clinker formation, grinding, and hydration reactions, and impact key cement phases such as Calcium-Silicate-Hydrate (C-S-H), Tricalcium Silicate (C_3S), Dicalcium Silicate (C_2S), Tricalcium Aluminate (C_3A), and Tetracalcium Aluminoferrite (C_4AF). Understanding these defects is essential, as they can alter hydration kinetics, mechanical strength, and chemical stability.

Calcium-Silicate-Hydrate (C-S-H), the primary hydration product responsible for cement's binding properties, is particularly prone to structural disorder. Unlike other crystalline phases, C-S-H is poorly crystalline or amorphous, consisting of calcium silicate sheets interspersed with water molecules and calcium ions. Variations in the arrangement of these calcium silicate layers and irregularities in interlayer water distribution introduce significant disorder [109, 151, 152]. These defects lead to inconsistencies in the mechanical properties, such as reduced load-bearing capacity and durability, as the structure becomes less efficient in distributing stress. Additionally, disorder can impact the material's porosity and permeability, making it susceptible to chemical degradation over time.

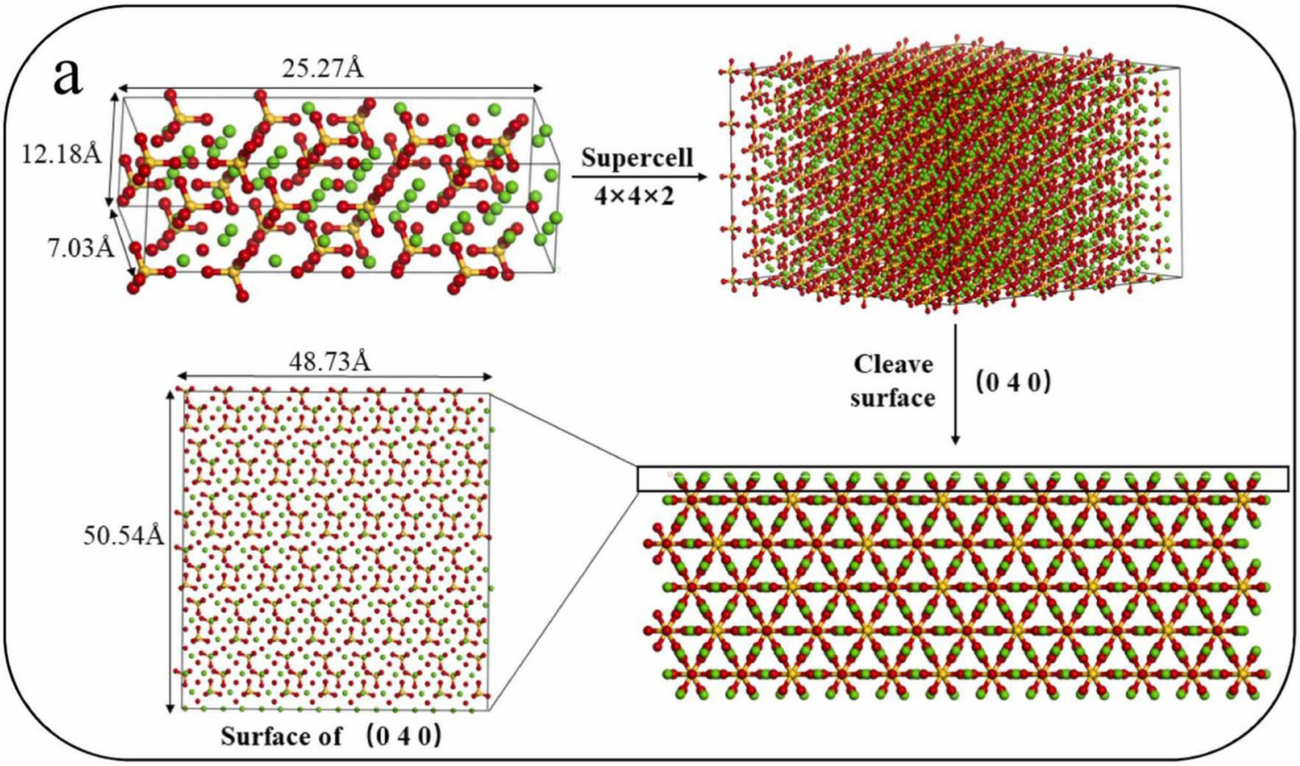
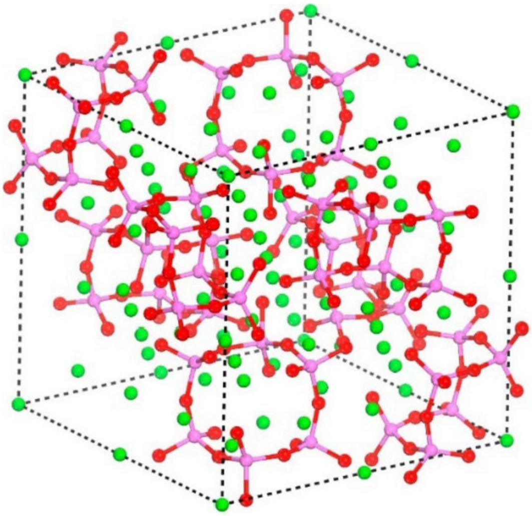


Fig. 1 Atomic structure representation and surface cleaving procedure of the material [37]. Top left panel: A single unit cell of the material, with dimensions 25.27 Å × 12.18 Å × 7.03 Å. Red and green spheres represent different atomic species. Top right panel: Construction of a 4 × 4 × 2 supercell from the unit cell, which is then cleaved along the (040) crystallographic plane to expose the desired surface. Bottom panels: Detailed views of the cleaved (040) surface. The bottom left shows the top view with surface dimensions 48.73 Å × 50.54 Å, and the bottom right provides a zoomed-in perspective highlighting the atomic arrangement on the surface

Fig. 2 Unit cell C_3A (Ca:green; Al:Pink; O:red) [115]



Tricalcium Silicate (C_3S), a major phase responsible for early strength development in cement, may exhibit defects within its monoclinic crystal structure. Vacancies, dislocations, and chain interruptions in the arrangement of silicate tetrahedra can occur during production and cooling [17, 92, 103]. These imperfections disrupt the ideal lattice structure, potentially reducing the phase's reactivity with water. As hydration reactions are dependent on the availability of reactive sites, such defects can slow down the hydration process, delaying the production of C-S-H and calcium hydroxide (CH) and, consequently, reducing early-age strength development. Dicalcium Silicate (C_2S), particularly in its β-polymorph, also experiences structural inconsistencies that influence its hydration behaviour [13, 85, 132]. Compared to C_3S, β-C_2S is less reactive and contributes to long-term strength development. However, defects such as lattice distortions, vacancies, or impurities can further slow hydration kinetics, diminishing the phase's long-term strength contribution. These structural irregularities make the hydration of C_2S highly dependent on environmental conditions like temperature and pressure, leading to variability in cement performance [19], Link et al. [77]; [1]. Tricalcium Aluminate (C_3A) and Tetracalcium Aluminoferrite (C_4AF) are prone to disorder in the form of cation substitution and distribution irregularities [14, 81, 90]. Variations in calcium, aluminium, and iron ion positions disrupt the stability of their respective cubic and orthorhombic structures. Such defects impact their reactivity with water and susceptibility to chemical attacks. For instance, cation disorder in C_3A accelerates its reaction with water, which can cause flash setting if not controlled. In C_4AF, impurities or cation substitutions reduce sulphate resistance, compromising cement's stability in aggressive environments.

Figure 3 provides a detailed representation of the atomic structure of calcium silicate phases, emphasizing the interplay between silicate chains and calcium oxide (CaO) layers, which are critical for cement properties. In part (a), silicate chains, shown in blue, are arranged in a linear configuration, alternating with CaO layers, depicted in yellow. This layered arrangement forms the structural backbone of calcium silicate phases, essential for the strength and durability of cement. Part (b) highlights the silicate tetrahedral units with their bridging sites and pairing sites. The bridging sites connect neighbouring silicate units, ensuring chain continuity, while pairing sites stabilize the tetrahedral framework. Parts (c), (d), and (e) illustrate the three-dimensional crystal structure, with calcium and silicate units arranged periodically. Part (c) shows a compact atomic view, while parts (d) and (e) emphasize the alignment of silicate chains and CaO layers along different crystallographic axes (a, b, and c). These atomic arrangements dictate the reactivity and mechanical performance of calcium silicate phases like C_3S and C_2S during hydration, directly influencing cement's strength development.

Improving cement performance and durability requires addressing defects that compromise its structure and functionality. Advanced production techniques play a key role in minimizing these defects. Controlled cooling rates, precise chemical formulations, and cutting-edge grinding processes are effective in reducing structural irregularities. Additionally, incorporating mineral admixtures or additives into the cement composition can stabilize its phases and diminish

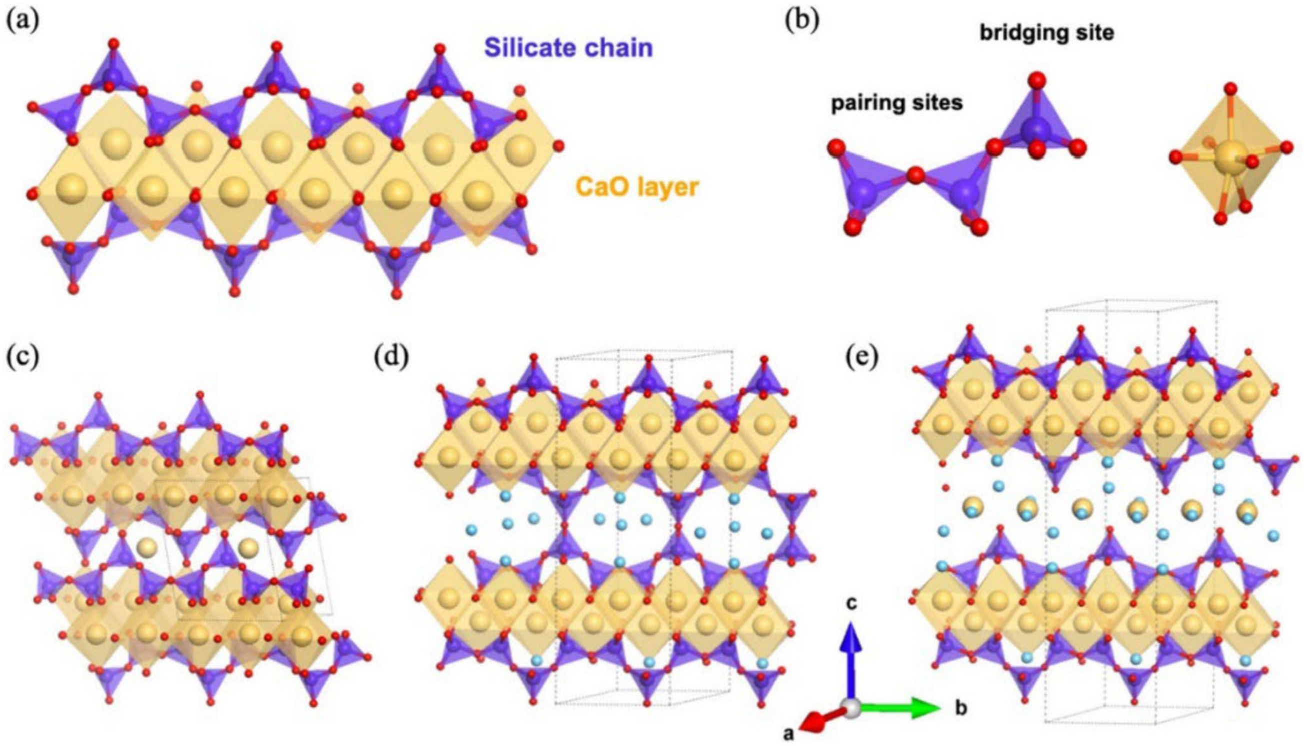


Fig. 3 Crystal structure and interlayer site characterization in the material [35]. **a** Side view showing alternating silicate chains (purple) and CaO layers (yellow). **b** Schematic highlighting oxygen coordination: bridging and pairing sites within silicate chains, with a single Ca-centred octahedron shown. **c**–**e** Different crystallographic projections of the structure, illustrating the stacking of silicate and CaO layers and interlayer cavity distributions along various axes (**a**, **b**, and **c**).

the effects of structural disorder. By focusing on defects at the atomic and microscopic levels, manufacturers can significantly enhance the material's strength, resilience, and service life. This meticulous management of imperfections not only improves the functional attributes of cement but also contributes to its environmental sustainability. Optimized processes and carefully selected additives reduce waste, energy consumption, and the overall carbon footprint of cement production. In essence, minimizing defects through innovative approaches enables the development of more durable and eco-friendly cement-based materials, meeting the demands of modern construction while promoting sustainability.

3 Interactions at the atomic scale

3.1 Hydration reactions

Hydration reactions at the atomic scale involve the interaction of water molecules with other substances, resulting in significant chemical and physical transformations. These reactions are primarily driven by the strong polar nature of water molecules. Each water molecule possesses partial positive charges on its hydrogen atoms and a partial negative charge on its oxygen atom, creating a dipole. This polarity enables water to interact effectively with both ionic compounds and polar molecules, facilitating processes like dissolution and phase changes. For ionic compounds, water molecules induce solvation, a process in which individual ions are surrounded and stabilized by water molecules [34, 105, 120, 154]. The oxygen atoms in water, carrying partial negative charges, align toward positively charged cations, while the hydrogen atoms, with partial positive charges, orient toward negatively charged anions. This alignment results in electrostatic interactions that lower the overall energy of the system. As a result, the ionic bonds within the compound weaken, leading to the dissociation of the ions and their uniform dispersion in water. For polar molecules, hydration reactions involve the formation of hydrogen bonds [64, 67, 106]. Water molecules interact with functional groups of polar substances by forming bonds between the partially positive hydrogen atoms of water and electron-rich regions of the solute. These hydrogen bonds disrupt existing intermolecular forces in the solute, breaking apart molecular structures and leading to dissolution. Such interactions allow polar molecules, like sugars or organic compounds, to form stable aqueous solutions where water uniformly surrounds and interacts with the solute molecules.

At the atomic scale, hydration reactions are essential in various chemical and biological processes. For example, in biochemical systems, hydration stabilizes macromolecules like proteins, ensuring proper folding and functionality. Water's role in solvation and hydrogen bonding also underpins key biological processes, such as enzyme–substrate interactions and ion transport in cells. In construction materials like cement, hydration reactions govern the transformation of cement phases into key products that contribute to the mechanical strength and durability of concrete. The primary cement phases, including tricalcium silicate (C_3S), dicalcium silicate (C_2S), tricalcium aluminate (C_3A), and tetracalcium aluminoferrite (C_4AF), undergo hydration to form various compounds that define the properties of hardened cement paste. Tricalcium silicate (C_3S) and dicalcium silicate (C_2S) react with water to form calcium silicate hydrate (C-S-H) and calcium hydroxide (CH), both of which contribute to concrete strength. The rapid reaction of C_3S dominates early strength development, while the slower hydration of C_2S provides long-term durability.

Tricalcium aluminate (C_3A) hydrates rapidly and interacts with gypsum ($CaSO_4 \cdot 2H_2O$) in the cement mix to form ettringite ($C_6A\acute{S}_3H_{32}$) in the early stages. As hydration progresses, ettringite may transform into monosulfate ($C_4A\acute{S}H_{12}$), depending on the availability of sulfate ions. Tetracalcium aluminoferrite (C_4AF) undergoes similar reactions but contributes less to strength development. The hydration of these aluminate phases significantly influences setting time, sulfate resistance, and overall durability of concrete [16, 58]. Ultimately, hydration reactions are fundamental for dissolving ionic and polar substances, enabling the formation of homogeneous solutions, and driving chemical and physical transformations. The unique polar nature of water, combined with its ability to form hydrogen bonds, highlights its central role in maintaining aqueous systems in chemistry, biology, and industrial applications, particularly in the hydration of cement, where it governs the formation of strength-giving phases in concrete.

Table 1 comprehensively explains the atomic-scale mechanisms of hydration reactions and their effects across various systems. It highlights the role of water's polarity, enabling interactions with ionic, polar, and nonpolar substances through electrostatic forces, hydrogen bonding, or weak van der Waals forces. Key mechanisms like solvation for ionic compounds and hydrogen bonding for polar molecules are shown to promote dissolution or stabilization. The hydration of cement phases (C_3S, C_2S) and energy transformations illustrate water's role in driving chemical and physical processes, such as cement hardening. Additionally, the table emphasizes temperature and pressure influences, which accelerate hydration

Table 1 Mechanisms and effects of hydration reactions across different systems

Aspect	Description	Mechanism	Effect on solute	Examples	Relevance	References
Nature of water molecule	Strongly polar; partial positive charge on hydrogen atoms and partial negative charge on oxygen	Electrostatic attraction and hydrogen bonding	Enables strong interactions with solutes	Dipole moment of water	Drives solvation and chemical reactions	[56, 157]
Ionic compounds	Water molecules surround and stabilize ions through solvation	Oxygen aligns with cations; hydrogen with anions	Disrupts ionic bonds, dissolves compounds	NaCl dissolving into Na^+ and Cl^- ions	Enables dissolution in aqueous systems	[49, 75]
Polar molecules	Hydration involves the formation of hydrogen bonds between water molecules and the solute	Hydrogen bonds with polar functional groups	Weakens solute's intermolecular forces	Glucose dissolving in water	Ensures solubility of organic compounds	[18, 20]
Nonpolar substances	Limited hydration as water cannot form strong interactions with nonpolar molecules	Weak van der Waals forces	Low solubility; molecules cluster together	Oil–water separation	Drives immiscibility in mixtures	Lomax & Lomax, [82]; [137]
Mechanism for Ions	Electrostatic forces lower system energy as water stabilizes individual ions	Solvation shell forms around ions	Stabilizes solute, promotes dissociation	Salt ions dispersing in water	Critical for electrolyte formation	[38, 135]
Mechanism for polars	Water molecules form hydrogen bonds with solute molecules	Hydrogen bonds disrupt intermolecular forces	Distributes molecules uniformly in solution	Dissolution of sugar	Supports energy transfer in solutions	[76, 130]
Hydration of cement phases	Interaction of water with cement compounds (C_3S, C_2S) to form hydration products like C-S–H	Water molecules react chemically	Formation of C-S–H gel, hardening of cement	$C_3S + H_2O \rightarrow$ C-S–H + CH	Drives concrete strength and durability	Cui et al. [28]; [70]
Energy transformation	Reduction in overall energy of the system through hydration	Thermodynamic stabilization	Promotes spontaneous dissolution or reactions	Exothermic reactions in cement	Provides stability to aqueous systems	[114, 162]
Physical processes	Dissolution and solvation: Uniform dispersion of solute in water	Molecular dispersion	Formation of homogeneous solutions	Salt or sugar dissolving in water	Crucial for material behaviour in solutions	[12, 86]
Chemical transformations	Hydration may trigger phase or structural changes in solutes	Reactivity between water and solute	New compounds form (hydration products)	Cement hardening, gypsum hydration	Essential for construction applications	Lesovik, 2021; [21]
Role in biological systems	Maintains aqueous environments required for biochemical reactions	Hydrogen bonding with biomolecules	Stabilizes structures like proteins/enzymes	DNA stabilization in water	Fundamental to life processes	[4, 107]

Table 1 (continued)

Aspect	Description	Mechanism	Effect on solute	Examples	Relevance	References
Temperature influence	Higher temperatures accelerate hydration reactions	Increases molecular motion	Faster solute dissolution, faster hydration	Rapid hydration of cement phases	Controls kinetics in industrial processes	Wen et al. [148]; [61]
Pressure influence	Higher pressure enhances water's interaction with solutes	Improves solute solubility	Denser hydration products, greater stability	High-pressure hydration of minerals	Relevant in geochemical environments	[65, 69]

reactions, affecting solubility, stability, and structural transformations. It links these processes to industrial, biochemical, and environmental relevance, demonstrating water's central role in systems like cement hydration, electrolyte formation, and biological stabilization.

The relationship between calcium-silicate-hydrate (C-S-H) gel and calcium content is multifaceted and controlled by numerous interdependent factors. A primary limitation lies in the variability of raw materials, notably the type of cement and supplementary cementitious materials (SCMs) employed. These materials significantly influence the calcium-to-silicon (C/S) ratio of the C-S-H gel, which in turn governs key properties such as microstructure, porosity, and mechanical behaviour [112, 118]. The hydration process itself—affected by water availability, curing conditions, and temperature—further complicates C-S-H formation [74, 142]. An excess of calcium during hydration can lead to the precipitation of portlandite ($Ca(OH)_2$), potentially increasing material permeability and compromising durability. Conversely, a low C/S ratio may yield a denser but more brittle microstructure, negatively impacting long-term performance [133].

Environmental exposures, including carbonation, sulphate attack, and chemical ingress, can substantially modify C-S-H composition, resulting in gradual degradation [43, 55, 110, 125]). However, equating carbonation alone with deterioration overlooks nuances such as the role of pore structure refinement during carbonation, which can sometimes reduce permeability despite chemical vulnerabilities. Furthermore, impurities and chemical admixtures such as superplasticizers and pozzolanic additions alter the nano- and microstructure of the C-S-H, affecting its resilience to environmental stresses [89, 95, 171]. Thus, while controlling calcium content and achieving a balanced C/S ratio are essential to optimizing the performance and durability of cementitious materials, the precise regulation of these parameters remains challenging due to the numerous influencing variables. Simplified correlations between carbonation, degradation, and durability risk obscuring the intricate mechanisms that actually govern material behaviour.

3.2 Calcium silicate hydrate (C-S-H) formation

Calcium Silicate Hydrate (C-S–H) is the primary hydration product in cementitious materials and is fundamental to the strength, durability, and overall performance of concrete. At the atomic scale, the formation of C-S–H involves complex interactions among calcium ions (Ca^{2+}), silicate tetrahedra (SiO_4), and water molecules [79, 109, 165]. These interactions begin during the hydration process of Portland cement, particularly through the reaction of tricalcium silicate (C_3S) with water. This reaction produces two main products: calcium silicate hydrate (C-S–H) and calcium hydroxide (CH). While CH contributes to the alkalinity of concrete, it is the C-S–H gel that provides the essential binding properties needed for strength development [59, 63, 133].

The formation of C-S–H involves calcium ions combining with silicate anions to create a gel-like network. This network consists of chains of interconnected tetrahedral silicate units, where each silicon atom is coordinated to four oxygen atoms in a tetrahedral arrangement [9, 30, 50], 84]. Calcium ions are dispersed throughout the structure, balancing the negative charge of the silicate chains. The presence of calcium ions within the C-S-H framework is critical for maintaining structural stability and mechanical integrity. Unlike most crystalline phases in cement, C-S-H is highly amorphous, meaning it lacks a well-defined long-range order. This amorphous nature gives C-S-H its unique flexibility, allowing it to accommodate varying ratios of calcium to silicate. This variability in composition directly influences the physical and chemical properties of C-S-H, such as its density, strength, and porosity. For example, higher calcium content tends to create a denser C-S-H structure, enhancing its mechanical properties, whereas lower calcium content may result in a more open and less compact network [23, 52, 124]. Water molecules play a crucial role in the formation and structure of C-S-H. They fill the interlayer spaces within the gel, creating voids that contribute to the material's microstructure. These water-filled voids influence both the density and mechanical properties of C-S-H. For instance, higher water content within the gel increases porosity, which can weaken the material, whereas reduced water content results in a denser and stronger C-S-H network [54, 76]. The dynamic balance between water, calcium, and silicate ensures that the C-S-H gel forms a cohesive and robust matrix capable of binding aggregate particles in concrete.

Figure 4 illustrates the hydration process and structural changes in a material, likely focusing on cement phases or calcium silicate hydrate (C-S-H), across varying conditions or hydration degrees. Each subfigure—(a), (b), (c), and (d)—represents different stages or levels of hydration, where the red spheres indicate oxygen atoms, green spheres likely denote calcium ions, and brown or pink structures represent silicate tetrahedra chains. In (a) and (c), the system appears highly hydrated, as evidenced by the dense distribution of water molecules (small red and white clusters), indicating extensive interaction with the material. This hydration causes increased disorder and separation of silicate structures. In contrast, (b) and (d) display a more ordered, drier structure with fewer water molecules, reflecting reduced hydration or a

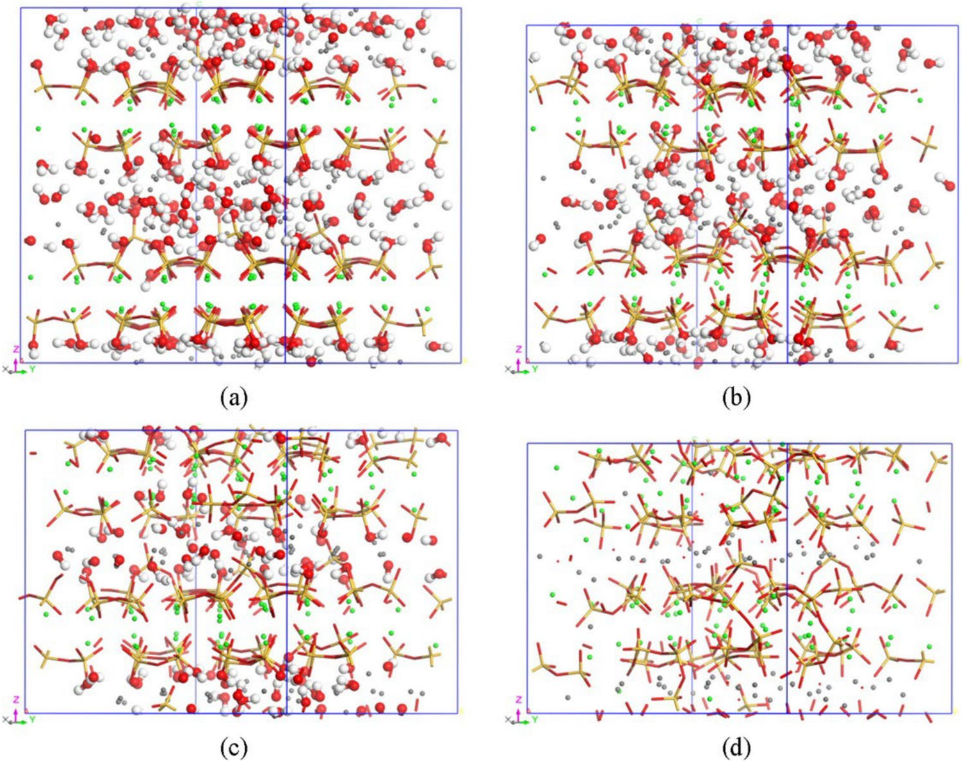


Fig. 4 Snapshots of molecular dynamics simulations showing adsorption behaviour on the material surface [152]. **a**–**c** Increasing concentrations of water molecules interacting with the surface, highlighting the structural changes and hydrogen bonding over time. **d** Simulation with CO_2 molecules showing their distinct adsorption configuration compared to water. Atoms are color-coded: red (O), white (H), green (Ca), yellow (Si), and gray (C), with simulation box boundaries marked in blue

later stabilization stage. The evolution of the arrangement highlights the role of water in structural changes, influencing mechanical properties such as strength and durability in cementitious materials.

The unique atomic-scale characteristics of C-S-H underscore its significance in cement science. Its amorphous structure provides a degree of adaptability that crystalline phases lack, enabling concrete to perform well under variable environmental conditions. However, this same amorphous nature introduces variability in mechanical properties, making it essential to understand and control the hydration process to optimize the formation of C-S-H. Advances in understanding these atomic-scale interactions allow for the development of enhanced concrete formulations, incorporating additives or supplementary materials to improve performance, durability, and sustainability. In summary, C-S-H is the backbone of cementitious materials, driving their strength and stability. Its formation through hydration reactions and its amorphous, flexible structure make it uniquely suited to serve as the binding phase in concrete. By examining the atomic interactions among calcium, silicate, and water, researchers can refine concrete formulations to achieve better mechanical properties, enhanced durability, and greater environmental efficiency.

The calcium-to-silicon (Ca/Si) ratio in the calcium-silicate-hydrate (C-S-H) matrix plays a crucial role in defining its water content and consequently its mechanical properties. As the Ca/Si ratio increases, the water content in the C-S-H structure also increases due to the formation of a more disordered and porous network. This is because higher calcium concentrations lead to the formation of additional interlayer water and increased gel porosity. Conversely, lower Ca/Si ratios result in a denser structure with reduced water retention. Mechanically, these changes directly impact strength, stiffness, and durability. Higher Ca/Si ratios (above ~ 1.5) introduce more free water, which weakens inter-particle cohesion and reduces compressive strength and stiffness. This leads to lower elastic modulus and higher creep deformation under load. In contrast, lower Ca/Si ratios (around 1.0 or lower) form a denser, more cross-linked C-S-H network, improving mechanical performance by increasing strength and rigidity. Additionally, water content influences shrinkage and cracking. Excess water leads to higher drying shrinkage, reducing durability, while a low water content results in a more brittle matrix. Thus, optimizing the Ca/Si ratio is key to balancing strength, durability, and long-term stability in cementitious materials.

While SCMs such as fly ash, silica fume, and slag enhance C-S-H formation and densify the cementitious matrix, recent studies have shown that their impact on durability is highly condition-dependent. Although SCMs reduce CH content and refine porosity, certain SCMs—especially those with lower early pozzolanic reactivity—can increase carbonation rates and permeability at early ages, potentially compromising durability [45, 46, 128]. Highly reactive SCMs like silica fume and metakaolin can mitigate early-age weaknesses, but the optimization of blending strategies is crucial, as inappropriate combinations may reduce resistance to chloride ingress or sulphate attack [72]. Additionally, while particle packing improvements and the incorporation of nano-materials such as nano-silica show promise for enhancing microstructure, these benefits are highly sensitive to dosage and dispersion techniques, with improper use leading to agglomeration and microcracking [5, 66]. Controlled heat curing can accelerate hydration kinetics,however, it may also increase thermal cracking risk if not properly managed [33, 123, 150]. Thus, while SCM-modified concrete presents opportunities for superior strength, durability, and sustainability, achieving these benefits requires a nuanced understanding of material behaviour, optimized mix design, and alignment with the intended exposure conditions, as emphasized in recent research.

3.3 Role of water molecules

Water molecules play a critical role in cement paste, serving as the driving force behind hydration reactions that lead to the hardening and strength development of concrete. At the molecular level, water initiates the reaction of cement compounds, particularly tricalcium silicate (C_3S) and dicalcium silicate (C_2S), to produce calcium silicate hydrate (C-S-H) gel and calcium hydroxide (CH) [68, 127, 149]. The formation of C-S-H gel is particularly significant because it serves as the primary binding phase in concrete, providing mechanical strength and durability. Simultaneously, calcium hydroxide contributes to the alkalinity of the system, which helps protect embedded steel reinforcements from corrosion.

The water-to-cement (w/c) ratio is a crucial factor in determining the overall performance of cement paste. A well-balanced w/c ratio ensures sufficient water is available to complete the hydration process while minimizing the formation of excess pores. In an ideal scenario, the hydration of tricalcium silicate and other cement phases progresses efficiently, creating a dense and interconnected C-S-H network. This dense microstructure reduces the overall porosity of the cement paste, enhancing strength, durability, and resistance to chemical attacks. Conversely, an improper w/c ratio can have detrimental effects. Excessive water increases the volume of capillary pores, leading to a weaker, less dense paste. Such a microstructure compromises mechanical strength and permeability, making the concrete more susceptible to external chemical attacks, freeze–thaw cycles, and other forms of degradation. On the other hand, insufficient water content can result in incomplete hydration, leaving unreacted cement particles in the system [10, 60]. This not only reduces strength but also increases long-term susceptibility to cracking.

Water also influences the cement paste's microstructure by controlling pore size, distribution, and connectivity. During hydration, water-filled voids are transformed into capillary pores, which can vary in size depending on the water content [39, 57, 94]. Large, connected pores facilitate the movement of fluids and gases through the cement matrix, negatively affecting durability. A denser microstructure, achieved through proper hydration, minimizes pore connectivity, significantly improving the material's resistance to permeability and chemical intrusion. In addition to hydration, water plays a role in the long-term durability of cement paste, particularly in its interactions with external chemical agents. For instance, in sulphate-rich environments, water facilitates the transport of sulphate ions into the paste, where they react with hydration products like calcium hydroxide and tricalcium aluminate [3, 25, 26]. This reaction forms expansive products such as ettringite, leading to cracking and degradation of the concrete, a phenomenon known as sulphate attack. Similarly, water is involved in carbonation, where atmospheric carbon dioxide dissolves in pore water to form carbonic acid, which reacts with calcium hydroxide [6, 96, 138]. This process reduces the alkalinity of concrete, compromising its ability to protect steel reinforcements. Managing water content is therefore critical for ensuring the performance and longevity of cement paste. Proper water management through an optimized w/c ratio not only promotes complete hydration but also minimizes the risk of chemical deterioration. By controlling pore structure, strength, and durability, water significantly influences the quality and service life of cementitious materials.

Figure 5 illustrates the wetting behaviour and local structure of water molecules interacting with cement hydration products, specifically highlighting calcium silicate hydrate (C-S-H) or related phases. In the top panel, a 3D representation shows a water layer (blue spheres) interacting with the cementitious surface composed of green, red, and yellow atoms. This represents the formation of a water film that plays a critical role in the hydration process by wetting the surface and facilitating molecular interactions. The bottom panels provide a detailed atomic-level view of the interaction. Water molecules (cyan and red spheres) form an intricate hydrogen-bond (H-bond) network with surface atoms, such as calcium and silicate oxygens. The middle panel highlights water's coordination, where oxygen atoms (Ow) from water interact with surface oxygen atoms (Os) and other functional groups, stabilizing the water layer. The surface line demarcates the boundary of water alignment with the cement hydration products. This figure demonstrates how water wetting influences hydration kinetics, facilitating the formation of C-S-H gel and promoting microstructural development essential for cement paste strength and durability.

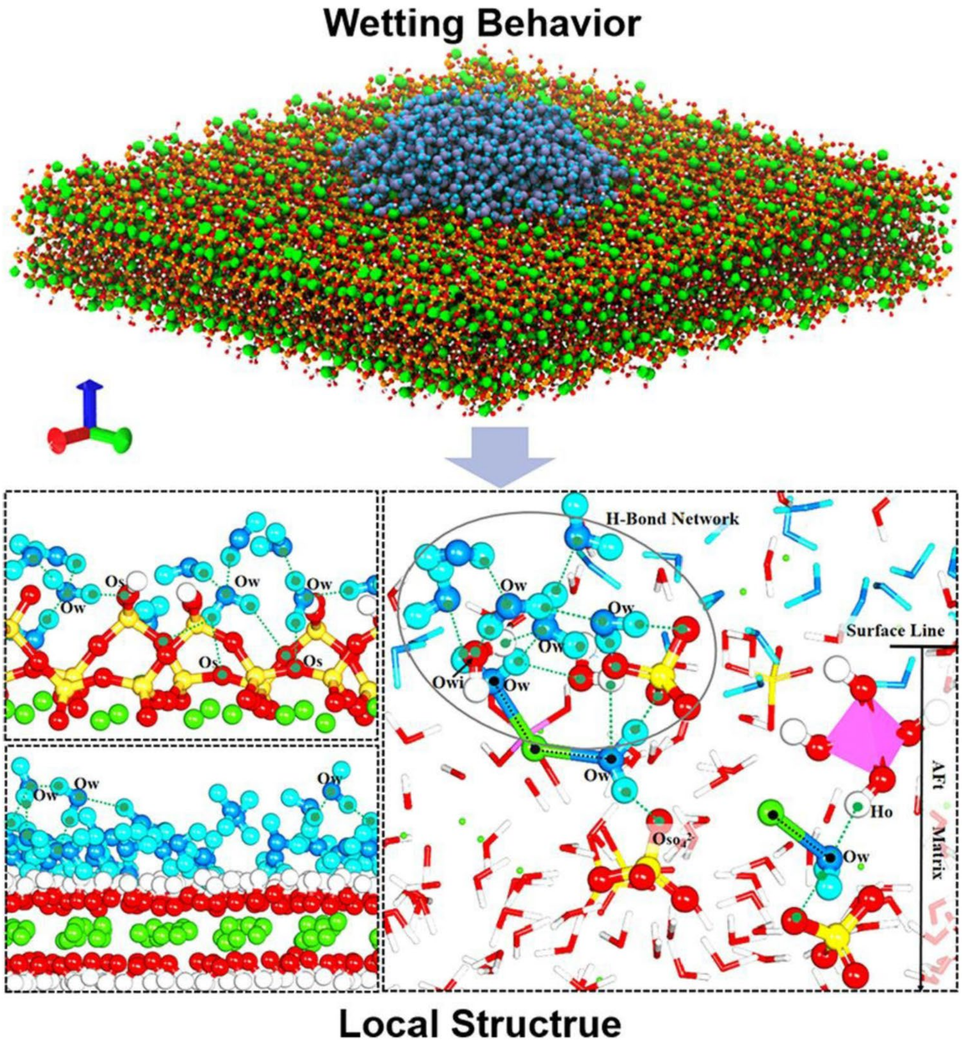


Fig. 5 Wetting Behaviour and Hydrogen Bond Network on Cement Hydration Product Surfaces [143]

Table 2 highlights the effects of water content on hydration, microstructure, and durability of cement paste. Proper water content ensures complete hydration of cement phases, forming a dense C-S-H network that provides strength and durability while minimizing pore size and permeability. Excess water increases porosity, weakening the structure and making it prone to chemical attacks like sulphate ingress and freeze–thaw damage. Insufficient water leads to incomplete hydration, reducing strength and increasing vulnerability to cracking and carbonation. By controlling the water-to-cement ratio, the microstructure can be optimized to enhance strength, durability, and resistance to environmental degradation, ensuring long-term performance.

3.4 Influence of temperature and pressure

Temperature and pressure play significant roles in influencing cement paste behaviour at the atomic scale, primarily by altering the rate and mechanisms of hydration reactions. These factors directly impact the formation of calcium silicate hydrate (C-S-H) gel, calcium hydroxide (CH), and the overall microstructure, which determines the paste's strength, setting time, and long-term durability. At higher temperatures, the kinetic energy of water molecules increases, which accelerates the reaction between water and cementitious compounds, particularly tricalcium silicate (C_3S) and dicalcium silicate (C_2S) [16, 159, 160]. This increased molecular activity enhances the rate of hydration, promoting the rapid formation of C-S–H gel and calcium hydroxide. Consequently, the cement paste sets faster and gains strength more quickly during the early stages. While this can be beneficial in some applications requiring fast setting, it may introduce complications. Rapid hydration generates significant heat, causing

Table 2 Effects of Water Content on Hydration, Microstructure, and Durability of Cement Paste

Aspect	Description	Effect of proper water content	Effect of excess water	Effect of insufficient water	References
Hydration Reactions	Water reacts with cement phases (C_3S, C_2S) to form C-S-H gel and CH	Complete hydration, strong C-S-H network	Weak gel formation, excess porosity	Incomplete hydration, unreacted cement	[40, 122]
Strength Development	Formation of C-S-H provides mechanical strength to cement paste	Optimal strength and durability	Reduced strength due to large pores	Lower strength, brittle microstructure	[73, 139]
Microstructure	Water controls pore size, distribution, and connectivity during hydration	Dense and low-porosity structure	High porosity, weak matrix	Poorly connected structure, cracks	[44, 78]
Permeability	Water content influences the paste's resistance to fluid penetration	Low permeability, improved durability	High permeability, fluid ingress	Higher permeability, weak protection	[7, 158]
Chemical Durability	Water enables reactions with external chemicals (e.g., sulphates, CO_2)	Resilient to chemical attacks	Increased risk of sulphate attack	Lower durability, carbonation risks	[100, 170]
Freeze–Thaw Resistance	Excess water in pores can freeze and expand, causing internal cracking	Good freeze–thaw resistance	Vulnerable to freeze–thaw damage	Limited resistance due to weak bonds	[146, 169]
Sulphate Attack	Water facilitates sulphate ion transport, causing ettringite formation	Minimal risk due to dense structure	High risk of cracking and damage	Insufficient hydration reduces defense	[2, 53]
Carbonation	Water enables CO_2 to dissolve and react with calcium hydroxide (CH)	Alkaline environment protects steel	Faster carbonation, corrosion risk	Lower alkalinity, steel protection loss	[48, 131]

thermal gradients within the paste [22, 116, 119]. These gradients can lead to internal stresses, uneven expansion, and eventual shrinkage cracking as the material cools and solidifies. Such thermal cracking reduces durability, particularly in large concrete structures where temperature control is challenging.

In contrast, lower temperatures reduce molecular activity, slowing down hydration reactions [89, 102]. The decrease in kinetic energy limits the dissolution of cement particles and the diffusion of ions, delaying the formation of C-S-H gel and CH. This slower reaction results in delayed strength development, which can be problematic in cold weather conditions. Furthermore, incomplete hydration of cement particles may occur, leaving unreacted material within the paste. This not only lowers the overall strength but also weakens the microstructure, increasing the material's permeability and reducing long-term durability. However, slower hydration at lower temperatures can sometimes lead to improved ultimate strength, as the gradual formation of hydration products allows for a more uniform and refined microstructure [36, 101].

Pressure also influences hydration reactions by modifying the solubility and diffusion rates of cementitious compounds. Increased pressure enhances the solubility of cement particles, accelerating their dissolution into ions [11, 32, 117]. This facilitates the formation of C-S-H gel and other hydration products, leading to a denser and more compact microstructure. The improved microstructural density reduces porosity, increasing the material's strength, impermeability, and resistance to chemical degradation. However, at extremely high pressures, the behaviour of hydration products may change [161, 163]. The altered pore structures and density of C-S-H gel can lead to a brittle microstructure, reducing the material's capacity to accommodate stress and potentially compromising long-term durability.

Overall, the combined effects of temperature and pressure on hydration reactions highlight their importance in controlling the setting behaviour, strength development, and durability of cement paste. Higher temperatures accelerate hydration but risk thermal cracking and uneven expansion, while lower temperatures delay hydration and strength gain. Similarly, moderate pressures enhance dissolution and densify the microstructure, improving performance, whereas excessive pressures may alter the material's properties. Proper management of temperature and pressure conditions during cement hydration is essential to optimize its microstructure and ensure the desired mechanical and durability characteristics for a wide range of applications.

4 Overview of techniques used to study cement at the atomic level

X-ray diffraction (XRD) remains a widely used method for studying crystalline phases in cement due to its rapid data acquisition and non-destructive nature. Researchers [27, 129, 156], have confirmed that XRD effectively identifies major phases like alite and belite and monitors phase changes during hydration. However, XRD struggles with detecting amorphous or nano-crystalline phases such as calcium-silicate-hydrate (C-S-H), which dominate hydrated products. Peak overlap further complicates quantification unless Rietveld refinement is applied, introducing modelling assumptions and limiting complete characterization, particularly at the nanoscale.

Electron microscopy techniques like SEM and TEM are vital for visualizing cement micro- and nanostructures. SEM provides insights into morphology and porosity, while TEM reveals nano-scale features with atomic resolution [42, 121, 168]. However, both are sensitive to preparation artifacts such as micro-cracking and beam damage and only capture localized regions, limiting their statistical representativeness. Solid-state nuclear magnetic resonance (NMR) spectroscopy offers unique insights into atomic-scale structures, particularly disordered phases like C-S-H. Wang et al. [141] illustrated NMR's power in revealing silicate polymerization. Yet, challenges include low sensitivity for certain nuclei, long acquisition times, and the need for complex deconvolution, introducing potential interpretative biases.

Molecular dynamics (MD) and density functional theory (DFT) modelling increasingly predict atomic interactions and hydration mechanisms. Researchers [147, 167, 170] have shown their value, but simulations often simplify system size, composition, and surface conditions, limiting realism and transferability. X-ray absorption spectroscopy (XAS), including XANES and EXAFS, provides element-specific local structural data. Researchers [31, 87, 145] have highlitged its effectiveness when crystalline information is lacking. However, reliance on synchrotron facilities and model-dependent data interpretation remain significant barriers. Given these limitations, a multi-technique, cross-validated approach is essential for fully understanding cement chemistry and performance.

5 Concluding remarks

Understanding cement at the atomic scale provides crucial insights into its performance, durability, and sustainability. This study emphasizes the importance of atomic-level structures, crystal arrangements, and defects in key cement phases, including tricalcium silicate (C_3S), dicalcium silicate (C_2S), tricalcium aluminate (C_3A), and tetracalcium aluminoferrite (C_4AF). Calcium silicate hydrate (C-S-H), the primary binding phase, plays a vital role due to its amorphous, flexible structure, which influences concrete's mechanical properties and microstructure. Defects such as vacancies, dislocations, and structural disorder significantly impact hydration kinetics and long-term stability. Optimizing production methods and additives can mitigate these defects, enhancing mechanical strength, reducing permeability, and improving durability. Hydration interactions with water molecules underscore the influence of water content, temperature, and pressure on microstructural development. However, limitations in atomic-scale characterization techniques and computational modelling present challenges in fully understanding cement's complex behaviour. Addressing these gaps is essential for refining predictive models and optimizing cement formulations. By incorporating these advancements, researchers can drive innovations in concrete technology, balancing strength, sustainability, and environmental resilience to create more durable and eco-friendly infrastructure.

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