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Molecular dynamics simulation in concrete research: A systematic review of techniques, models and future directions

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ABSTRACT

This paper presents a comprehensive review of the application of molecular dynamics simulation in concrete research. The study addresses the background and significance of the topic, providing an overview of the principles, applications, and types of molecular dynamics simulation, with a particular focus on its role in enhancing the understanding of concrete properties. Moreover, it critically examines existing research studies that employ molecular dynamics simulation in concrete research, highlighting the associated benefits and limitations. The paper further investigates various simulation techniques and models employed in concrete research, offering a comparative analysis of their effectiveness. Additionally, the study explores future directions and identifies research needs in the field of molecular dynamics simulation in concrete, while also discussing the potential impact of this approach on the sustainability of the construction industry. By providing a comprehensive overview and critical analysis, this review serves as a valuable resource for researchers and practitioners interested in leveraging molecular dynamics simulation for advancing concrete science and engineering.

1. Introduction

Molecular dynamics (MD) simulation is a computational technique that models the behaviour and interactions of atoms and molecules over time. It has become a powerful tool in the field of materials science, including the study of concrete. Concrete, a widely used construction material, is composed of cement, aggregates, water and various additives. Understanding the complex structure and properties of concrete at the atomic level is crucial for improving its performance, durability, and sustainability. MD simulation offers a unique approach to investigate the atomic-scale behaviour of concrete materials. It allows researchers to simulate the movement, energy and interactions of individual atoms and molecules within the concrete matrix. By considering the forces and energies at play, MD simulations can provide insights into the mechanical, thermal, and chemical properties of concrete, as well as the effects of external factors such as temperature, pressure and moisture [1–3].

Through MD simulations, researchers can explore phenomena that are difficult to observe experimentally, gaining a deeper understanding of the fundamental processes and mechanisms governing concrete behaviour. This knowledge can then be used to optimize concrete formulations, enhance its mechanical strength, improve resistance to environmental degradation, and design more sustainable construction materials. However, it is important to note that MD simulations have certain limitations. They require accurate force field parameters, adequate computational resources and appropriate modelling assumptions. The complexity and size of concrete systems pose challenges in terms of simulation time and computational cost. Despite these limitations, MD simulations continue

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to contribute significantly to the understanding and advancement of concrete materials, paving the way for innovative developments in the construction industry.

The application of MD simulation in the study of concrete and cementitious materials has gained significant attention in recent years. Kalinichev et al. (2007) [4] focused on MD modelling of mineral-water interfaces, with specific application to cement materials. They explore the structure, dynamics and energetics of these interfaces, providing insights into the interactions between water and minerals in cement. Cho et al. (2020) [5] presented a comprehensive review of MD simulation of calcium-silicate-hydrate (C-S-H), a crucial component of cement. They discussed the use of MD to investigate the nano-engineering of cement composites, highlighting the importance of understanding C-S-H at the atomic level for the development of advanced cement materials. Lau et al. (2018) [6] discussed the prospects and challenges of using MD simulations in nano-engineering construction materials, including concrete. They emphasize the potential of MD to provide insights into the behaviour and properties of construction materials at the nanoscale, enabling the development of innovative and high-performance materials.

Wang et al. (2019) [7] provided a review on modelling techniques for cementitious materials, including MD simulation, across different length scales. They discuss the development and future prospects of these techniques, highlighting the importance of multiscale modelling in capturing the complex behaviour of cementitious materials. Valavi et al. (2022) [8] focused on the molecular dynamic simulations of cementitious systems using a newly developed force field suite, ERICA FF. They demonstrate the significance of accurate force fields in capturing the behaviour and properties of cement materials at the atomic level, leading to more reliable simulations. Hou (2020) [9] presented a comprehensive book on molecular simulation specifically focused on cement-based materials. It covers various aspects of MD simulation, including the characterization of cement microstructures, hydration processes and mechanical properties, providing an in-depth understanding of the application of MD in this field. Li et al. (2022) [10] investigated the chloride ion erosion mechanism of cement mortar in coastal areas using a combination of experiments and MD simulations. They demonstrate the capability of MD simulation to provide insights into the degradation processes of cement materials under aggressive environments. These studies highlight the significant contributions of MD simulation in advancing our understanding of concrete and cementitious materials. They showcase the potential of MD to unravel the atomic-scale behaviour of these materials, aiding in the development of more durable, sustainable, and high-performance concrete for various applications in the construction industry.

The advancement in computing power has opened up new possibilities for exploring materials at the nanoscale, leading to the emergence of nanoscale modelling in materials science. Atomistic simulation methods, depicted in Fig. 1 [11], have proven to be highly valuable in deepening our fundamental understanding of cementitious materials. These simulation techniques offer a great potential to investigate the behaviour and properties of cementitious materials at the atomic level, enabling researchers to gain insights into their structural, mechanical, and chemical characteristics. By simulating the interactions and dynamics of individual atoms and molecules, atomistic simulations contribute to the advancement of cement science and pave the way for the development of innovative materials and improved performance in various applications.

MD simulation offers a significant advantage by providing fundamental insights into the initiation and evolution of material damage at the nanoscale, which are often challenging to obtain through traditional laboratory experiments. By simulating the atomic interactions and dynamics within a material, MD simulations allow researchers to observe and analyse processes such as crack propagation, dislocation movement, and fracture behaviour at a level of detail not achievable experimentally. This nanoscale perspective enables a deeper understanding of the underlying mechanisms governing material failure, leading to improved material design and the potential for enhanced performance in various applications.

This review paper is motivated by the increasing interest and potential of molecular dynamics simulation in the study of concrete materials. With concrete being a widely used construction material, understanding its behaviour and properties at the atomic level is crucial. Molecular dynamics simulation offers a unique opportunity to explore the intricate atomic interactions and processes in concrete, providing valuable insights into crack propagation, material damage, and other phenomena. By consolidating existing knowledge and highlighting the benefits of molecular dynamics simulation, this review paper aims to inspire researchers and practitioners to further explore this approach and contribute to advancements in concrete science and engineering. This paper also aims to address several gaps in the existing literature. These gaps include:

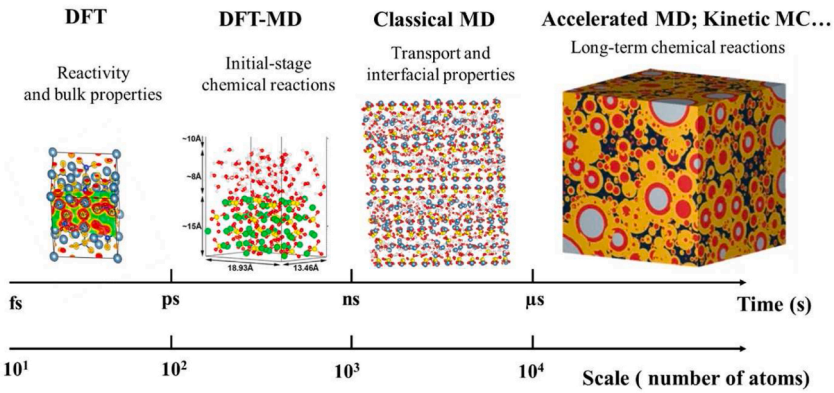

Fig. 1. Schematic of atomistic simulation methods in various time and space scales [11].

1. Limited coverage: The current literature lacks a comprehensive overview of the principles, applications, and types of molecular dynamics simulation specifically applied to concrete. This review paper aims to provide an extensive examination of the utilization of molecular dynamics simulation in the context of concrete research.
2. Inadequate emphasis on understanding concrete properties: Although some studies mention the use of molecular dynamics simulation in concrete research, there is a lack of emphasis on its significance in enhancing the understanding of concrete properties at the atomic level. This review paper seeks to address this gap by highlighting the contributions of molecular dynamics simulation in uncovering the intricate behaviour and properties of concrete.
3. Insufficient evaluation of simulation techniques and models: The existing literature does not adequately evaluate and compare different simulation techniques and models employed in concrete research. This review paper intends to bridge this gap by critically examining and comparing the effectiveness of various simulation approaches, providing insights into their strengths, limitations, and potential advancements in the context of concrete research.
4. Limited discussion on sustainability implications: The literature does not sufficiently explore the future directions and potential impact of molecular dynamics simulation on the sustainability aspects of the construction industry. This review paper aims to fill this gap by discussing the potential implications of molecular dynamics simulation in promoting sustainable practices within the construction sector and identifying research needs in this area.

By addressing these literature gaps, this review paper aims to provide researchers and practitioners with a comprehensive and up-to-date resource that consolidates existing knowledge and identifies avenues for further exploration in the field of molecular dynamics simulation in concrete research.

This review paper has some limitations that should be acknowledged. Firstly, the selection of references may introduce some bias, as the inclusion of specific studies could influence the overall findings and conclusions. Additionally, the review's scope may not cover every aspect or recent advancements in the field of molecular dynamic simulation and concrete. The limitations of the individual studies included in the review, such as the assumptions and simplifications made in their simulations, should also be considered. Furthermore, language and access limitations could affect the comprehensiveness of the literature search. Despite these limitations, the review paper provides a valuable synthesis of the existing literature on molecular dynamics simulation and concrete.

For researchers, this study provides a novel and comprehensive overview of the use of molecular dynamics simulation in concrete research. By addressing the existing literature gaps and consolidating knowledge, it offers a valuable resource for researchers to deepen their understanding of the principles, applications, and techniques of molecular dynamics simulation in the context of concrete. The critical evaluation of simulation techniques and models will guide researchers in selecting appropriate methods for their studies. Additionally, the exploration of the potential impact of molecular dynamics simulation on the sustainability of the construction industry opens new avenues for future research and promotes the adoption of sustainable practices. For end users, such as engineers, architects, and construction professionals, the findings of this study have practical implications. The insights gained from molecular dynamics simulation can inform the design and engineering of concrete structures, leading to improved performance, durability, and safety. By understanding concrete properties at the atomic level, end users can make more informed decisions regarding material selection, construction methods, and maintenance strategies. This ultimately contributes to the development of more sustainable and resilient infrastructure.

2. Molecular dynamics simulation

2.1. Definition of molecular dynamics simulation and its applications

Molecular dynamic (MD) simulation is a computational modelling technique used to study the behaviour and interactions of atoms and molecules over time. It employs numerical algorithms to solve Newton's equations of motion for a system of particles, allowing for the simulation of their movements and interactions at the atomic level. MD simulations rely on force fields, which describe the interatomic forces and potential energy of the system.

Molecular dynamics (MD) simulation has emerged as a powerful tool with diverse applications in various scientific disciplines. In the field of polymer science, MD simulations have been extensively utilized to investigate the conformational behaviour, phase transitions, and mechanical properties of polymers [12] (Binder, 1995). In biomolecular research, MD simulations have played a crucial role in elucidating the dynamics, folding pathways, and interactions of biomolecules such as proteins and nucleic acids [13]. Notably, MD simulations have contributed to the understanding of four-stranded DNA structures [14]. In computational chemistry, MD simulations have been applied to predict molecular properties such as diffusion coefficients and density [15]. They have also been employed in studies related to asphalt materials [1] and computational toxicology [16], providing valuable insights. Furthermore, MD simulations have been instrumental in structure prediction of peptides and proteins [17]. Overall, MD simulations have revolutionized research across multiple domains by enabling detailed investigations and predictions of molecular behaviour and properties.

The applications of MD simulation in the context of concrete research are extensive. It enables the investigation of various phenomena, such as the hydration process, binding mechanisms, and the behaviour of different cementitious phases. MD simulations provide insights into the structural, mechanical, and thermal properties of concrete materials. They can be used to study the influence of additives, admixtures and environmental conditions on concrete performance. Additionally, MD simulations facilitate the exploration of interfaces, such as the mineral-water interface, and their impact on the properties and durability of concrete. Overall, MD simulation plays a vital role in advancing the understanding and design of concrete materials.

2.2. Principles of molecular dynamics simulation

Molecular dynamics (MD) simulation is a computational technique that models the behaviour and interactions of atoms and molecules over time. It is based on Newton's laws of motion and statistical mechanics principles. The principles of molecular dynamics simulation can be summarised as follows [18,19]:

1. Newton's Equations of Motion: MD simulations solve the equations of motion for each atom in a system. These equations describe the forces acting on the atoms and their resulting motions.
2. Force Field: A force field is used to calculate the forces between atoms. It consists of mathematical functions that describe the potential energy of the system and the forces derived from it. Force fields include parameters for bonded and non-bonded interactions such as bond stretching, angle bending, torsional rotation, and van der Waals and electrostatic interactions.
3. Time Integration: MD simulations divide time into discrete steps, called time steps. During each time step, the positions and velocities of atoms are updated based on the forces acting on them. The time step should be small enough to accurately capture the dynamics of the system.
4. Ensemble: MD simulations can be performed in different ensembles, such as the microcanonical ensemble (NVE), canonical ensemble (NVT) and isothermal-isobaric ensemble (NPT). These ensembles control the temperature, volume, and number of particles in the simulated system.
5. Thermostats and Barostats: To maintain a desired temperature and pressure, MD simulations often employ thermostats and barostats. These algorithms adjust the velocities and positions of atoms to control temperature and pressure fluctuations.
6. Boundary Conditions: MD simulations use periodic boundary conditions to mimic an infinite system. This allows for the simulation of a small unit cell repeated in space, eliminating artificial surface effects.
7. Sampling and Averaging: MD simulations generate a trajectory that represents the time evolution of the system. Statistical analysis of the trajectory provides thermodynamic and dynamic properties of interest, such as energy, temperature, pressure, and diffusion coefficients.
8. Initial Configuration: MD simulations require an initial configuration of atoms. This can be obtained from experimental data, crystal structures, or other simulation techniques like molecular mechanics or quantum mechanics.

2.3. Types of molecular dynamics simulation and their characteristics

Molecular dynamics (MD) simulations can be classified into different types based on the specific aspects they focus on and the techniques employed. Here are some common types of MD simulations and their characteristics:

1. Classical MD: Classical MD simulations treat atoms as classical particles, neglecting quantum mechanical effects. They use empirical force fields to describe interatomic interactions. Classical MD is computationally efficient and widely used for studying large systems over longer timescales [20,21].
2. Ab initio MD: Ab initio MD simulations incorporate quantum mechanical principles, considering electronic structure and interactions. They employ density functional theory (DFT) or wavefunction-based methods to calculate forces and energies. Ab initio MD is particularly suitable for studying small molecules, reactions, and systems with strong electron-electron interactions [22–24].
3. Coarse-Grained MD: Coarse-grained MD simulations represent groups of atoms as single particles, reducing the computational complexity. They capture the essential features of the system while sacrificing atomic-level details. Coarse-grained MD is useful for studying large biomolecular systems and self-assembly processes [25–27].
4. Reactive MD: Reactive MD simulations focus on chemical reactions and the dynamics of bond formation and breaking. They employ potential energy surfaces that explicitly include reaction pathways and transition states. Reactive MD is employed in studying reaction kinetics, catalysis, and exploring complex reaction mechanisms [28,29].
5. Quantum MD: Quantum MD simulations combine molecular mechanics and quantum mechanics. They treat a subset of atoms quantum mechanically while treating the remaining atoms classically. Quantum MD is useful for systems where quantum effects play a significant role, such as proton transfer, hydrogen bonding and electronic structure properties [30,31].
6. Enhanced Sampling MD: Enhanced sampling methods aim to overcome limitations in traditional MD simulations, such as the sampling of rare events and slow processes. Techniques like replica exchange MD, meta dynamics and umbrella sampling bias the simulation to explore different regions of the potential energy surface more efficiently [32,33].
7. Non-Equilibrium MD: Non-equilibrium MD simulations investigate systems far from equilibrium, where energy and/or momentum are actively exchanged with the environment. These simulations are used to study processes such as heat transfer, shearing, and mechanical deformation [34,35].

Each type of MD simulation has its advantages and limitations. The choice of simulation type depends on the specific research question, system size, timescale of interest, and computational resources available. By utilizing different types of MD simulations, researchers can gain insights into a wide range of molecular and materials phenomena.

2.4. Role of molecular dynamic simulation in understanding concrete properties

Molecular dynamics (MD) simulation plays a significant role in understanding concrete properties by providing insights into the atomic-scale behaviour and interactions within the material. Here are some key aspects where MD simulation contributes to our understanding of concrete:

2.4.1. Atomic-level interactions

Concrete, a complex material composed of various components, can be better understood through molecular dynamics (MD) simulations. By virtually simulating the interactions of individual atoms, MD simulations provide insights into the bonding, hydration, and overall structure of concrete at the atomic level. These simulations enable researchers to study phenomena such as the growth of hydration products, pore structure development, and mechanical properties. Additionally, MD simulations allow for investigating the influence of factors like admixtures, environmental conditions, and loading on concrete behaviour. By enhancing our understanding of concrete at the fundamental level, MD simulations contribute to optimizing concrete materials and designs for improved performance and sustainability [36,37].

2.4.2. Hydration process

MD simulations play a significant role in enhancing our understanding of the hydration process in cement, a critical aspect of concrete formation. By simulating the interactions between water molecules and cement particles at the atomic level, MD simulations provide valuable insights into the nucleation and growth of hydration products, particularly the formation of calcium-silicate-hydrate (C-S-H) gel and other phases in cementitious materials [4,38]. MD simulations enable researchers to observe the dynamics and molecular-level mechanisms involved in cement hydration. They capture the diffusion of water molecules into the cement matrix, the adsorption and incorporation of water molecules by cement particles, and the subsequent development and growth of hydration products. By simulating these processes, MD simulations reveal the kinetics, morphology, and atomic structure of hydration products. Understanding the hydration process at the atomic level is crucial for predicting and optimizing concrete properties. The formation and growth of hydration products significantly impact the development of concrete strength, porosity, and other mechanical characteristics. By providing detailed insights into cement hydration, MD simulations contribute to the prediction and design of concrete with desired properties, such as strength, durability, and performance.

2.4.3. Mechanical behaviour

Molecular dynamics (MD) simulations have the capability to provide valuable insights into the mechanical properties of concrete, including elasticity, strength, and fracture behaviour. By subjecting virtual concrete models to external forces, MD simulations allow researchers to observe the atomic-level deformation and stress distribution within the material [39–41]. Through MD simulations, researchers can investigate the mechanisms of load transfer, crack initiation, and crack propagation in concrete. By analysing the atomic-scale behaviour of the material under different loading conditions, MD simulations provide a detailed understanding of the mechanical response of concrete. By examining the atomic-level deformation and stress distribution, MD simulations offer insights into the factors that influence the overall mechanical behavior of concrete. This information aids in understanding the fundamental processes governing concrete's response to external forces, including the interactions between different components and the formation and propagation of cracks. The insights gained from MD simulations of concrete's mechanical properties have important implications for optimizing concrete materials and designs. By understanding the mechanisms of deformation and fracture at the atomic level, researchers can develop strategies to enhance concrete's strength, durability, and resistance to cracking.

2.4.4. Transport phenomena

A comprehensive understanding of the transport properties of concrete, including the diffusion of ions, water, and gases, is essential for evaluating its durability and sustainability. MD simulations offer a powerful approach to gaining detailed information about the diffusion mechanisms, transport rates, and preferential pathways within the concrete matrix [42,43]. By simulating the interactions between individual atoms and considering the intricate pore structure of concrete, MD simulations provide insights into the factors that influence transport behaviour. Researchers can examine how atoms diffuse through the matrix, the effects of pore connectivity and size on transport rates, and the presence of preferential pathways for different species. MD simulations enable the observation of transport phenomena at the atomic level, allowing researchers to investigate the diffusion mechanisms and understand the factors that control transport properties. By providing detailed information on the interactions between atoms and the pore structure, MD simulations contribute to the design and optimization of concrete with improved permeability and durability. Understanding the transport properties of concrete through MD simulations has significant implications for enhancing its performance and sustainability. By guiding the development of concrete with optimized permeability, researchers can improve resistance to harmful ions, mitigate degradation processes, and enhance overall durability.

2.4.5. Defects and degradation

Molecular dynamics (MD) simulations provide a powerful platform to study defect formation and degradation mechanisms within concrete. MD simulations enable researchers to investigate various types of defects, including vacancies, dislocations, and grain boundaries, which have a significant impact on the mechanical and chemical properties of the material. By simulating the interactions between atoms and considering environmental factors such as temperature, moisture, and chemical exposure, MD simulations can shed light on the degradation mechanisms of concrete. For instance, MD simulations can explore phenomena like alkali-silica reaction, sulphate attack, and corrosion of reinforcement, which are critical degradation processes affecting concrete structures. Understanding the formation and behaviour of defects and degradation mechanisms at the atomic level is crucial for developing strategies to mitigate degradation and improve the long-term performance of concrete structures. By gaining insights into the atomistic processes involved, researchers can propose methods to prevent or mitigate the detrimental effects of defects and degradation phenomena.

The knowledge obtained from MD simulations aids in the optimization of concrete formulations and the development of strategies to enhance durability, mitigate degradation, and ensure the long-term performance of concrete structures in diverse environmental conditions.

2.4.6. Material design and optimization

MD simulations provide a valuable tool for designing and optimizing concrete mixes by investigating the effects of additives, admixtures, and material configurations at the atomic scale [9,44]. By virtually modelling the interactions between individual atoms, MD simulations can predict the impact of different components on concrete properties. Through MD simulations, researchers can explore the effects of additives and admixtures on important concrete characteristics such as workability, strength, and durability. By considering the atomic-level interactions between these components and the concrete matrix, MD simulations can provide insights into how they influence the overall performance of the material. The ability of MD simulations to predict the impact of various components on concrete properties enables researchers to guide the development of new concrete formulations and optimize existing ones. By virtually testing different combinations and concentrations of additives and admixtures, researchers can identify optimal mix designs that meet specific performance requirements. The knowledge gained from MD simulations contributes to the advancement of concrete technology by facilitating the development of innovative materials and improved mix designs. This ultimately leads to the production of concrete with enhanced properties, such as increased strength, improved workability, and enhanced durability. By leveraging the capabilities of MD simulations, researchers and engineers can make informed decisions in the design and optimization of concrete mixes, resulting in more efficient and sustainable construction practices.

3. Use of molecular dynamic simulation in concrete research

3.1. Research on the use of molecular dynamic simulation in concrete

Research on the use of molecular dynamic simulation in concrete has significantly contributed to our understanding of the material's properties and behaviour at the atomic and molecular levels. By simulating the interactions between cementitious components, water and additives, molecular dynamic simulations provide valuable insights into the hydration process, mechanical properties and durability of concrete. These simulations help in predicting the structure and dynamics of cementitious materials, studying the effects of different factors on performance, and designing more sustainable and efficient concrete formulations. Furthermore, molecular dynamic simulations enable the exploration of novel materials and the development of advanced modelling techniques, facilitating the optimization and innovation of concrete technology.

The use of MD simulation in concrete research has proven to be a valuable tool for studying the properties and behaviour of cementitious materials at the atomic and molecular levels. By simulating the interactions and movements of individual atoms and molecules, MD simulations provide insights into the structural, mechanical and chemical aspects of concrete. One area of research where MD simulations have made significant contributions is the investigation of the structure and properties of calcium-silicate-hydrate (C-S-H), the primary binding phase in cement paste. Faucon et al. (1997) [45] utilized MD simulations to study the structural properties of C-S-H, revealing important details about its composition, atomic arrangement and hydration mechanism. These findings have advanced our understanding of the fundamental characteristics of C-S-H and its role in the overall performance of concrete.

In addition to studying the bulk properties of cementitious materials, MD simulations have been employed to explore interfacial phenomena and the behaviour of interfaces between different materials. Wang et al. (2020a) [40] investigated the interlayer bonding between 3D printed concrete and polymer modified mortar using both experimental methods and MD simulations. By analysing the molecular interactions and mechanical behaviour at the interface, the study provided insights into the improvement of interlayer bonding and the overall performance of the composite material. The interfacial shear strength between C-S-H and polymer fibres was investigated by Wang et al. (2020b) [46] through MD simulations. This study shed light on the mechanisms governing the adhesion and load transfer between the fibre and matrix, contributing to the design and optimization of fibre-reinforced concrete composites.

Fracture toughness is a critical property for understanding the mechanical behaviour of concrete. Bauchy et al. (2015) [47] conducted MD simulations to explore the fracture toughness of C-S-H, providing insights into the crack propagation mechanisms and the influence of microstructure on the material's resistance to fracture. This knowledge is essential for the development of more durable and crack-resistant concrete materials. MD simulations have also been employed to investigate chemical interactions, such as chloride binding to the surfaces of calcium hydroxide (CH) and calcium silicate phases. Kalinichev and Kirkpatrick (2002) [48] used MD simulations to elucidate the binding mechanisms and energetics of chloride ions, which are known to contribute to the degradation of concrete structures in chloride-rich environments. Understanding these interactions at the atomic scale aids in the development of strategies to mitigate chloride-induced deterioration.

Furthermore, MD simulations have provided insights into the early stages of cement hydration. Qi et al. (2021) [38] conducted comprehensive MD simulations to compare the initial hydration processes of different calcium silicates in Portland cement. The initial hydration process can be categorized into three distinct stages, as illustrated in Fig. 2, with the time scale represented logarithmically. The first stage ($t \leq 0.001$ ns) is characterized by rapid hydration of the exposed surface. During this stage, approximately half of the total hydroxylation degree and the number of Os-H bonds are achieved. Water molecules are quickly adsorbed onto the exposed surface, while the solid surface atoms exhibit minimal vibrations near their initial positions. The second stage occurs between 0.001 ns and 3 ns. During this stage, continuous hydration of the exposed surface is observed. Surface Ca atoms move outward, and some H atoms diffuse into the solid, penetrating beneath the surface. The third stage encompasses a longer time scale ($t > 3$ ns). In this stage, the increase in both the hydroxylation degree and the number of Os-H bonds is slower compared to the previous stages. The key characteristics of this stage include the diffusion of water molecules into the solid and the ongoing translocation of surface atoms. These

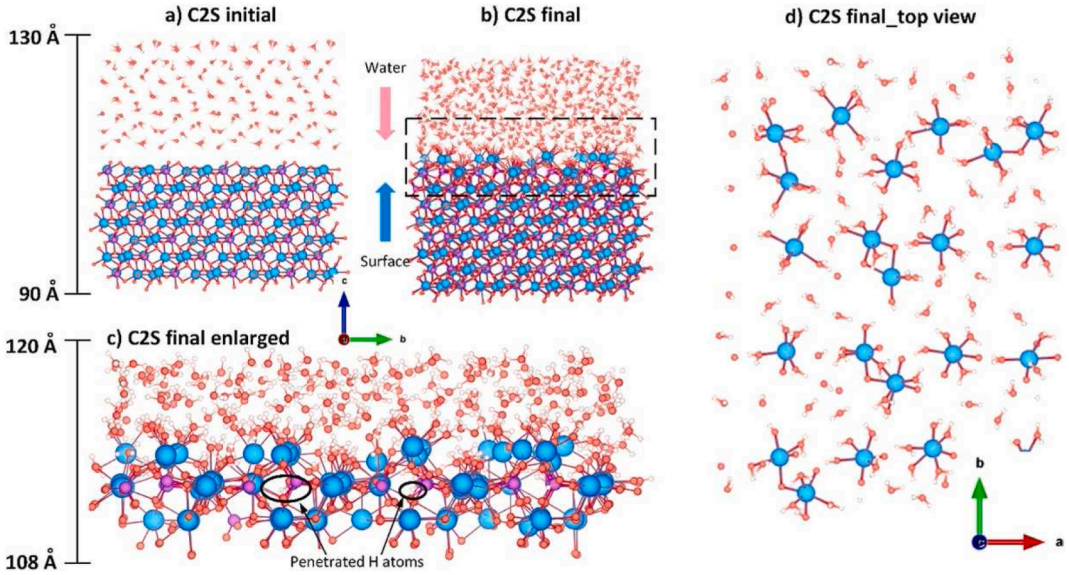

Fig. 2. The comparison of structures around the low surface of β-C_2S: a) initial structure around the low surface, b) side view of the final structure around the low surface at c) side view of the enlarged final structure, d) top view of the final structure [38].

findings provide valuable insights into the temporal evolution of the initial hydration process, highlighting the dynamic behaviour of water molecules, surface atoms, and their interactions within the cementitious system.

Portlandite, a key product of cement hydration, exhibits a relatively straightforward structure (Fig. 3a) [8]. On the other hand, ettringite, a fascinating cementitious material, possesses a more intricate structure (Fig. 3b). Its chemical formula is ($Ca_6Al_2(SO_4)3(OH)_{12}\cdot 26H_2O$), and the unit cell structure contains water molecules with varying occupancy. Within the unit cell, there are two equivalent positions (top and bottom), each accommodating six water molecules with half occupancy.

While MD simulations have proven valuable in advancing our understanding of concrete properties, it is important to acknowledge their limitations. MD simulations are computationally intensive and typically limited to small system sizes and short timescales. The complexity of concrete, which involves a hierarchical structure spanning multiple length and time scales, poses challenges for MD simulations to capture real-world behaviour. Therefore, it is crucial to complement MD simulations with experimental techniques and to integrate multiscale modelling approaches that bridge different length and time scales. MD simulations have also been employed to investigate the interface between graphitic structures and calcium-silicate-hydrate [49]. These simulations provided insights into interaction energies, structure and dynamics, aiding in the understanding of interfacial phenomena.

Two distinct oxygen atom types have been characterised in the alite crystal structure through charge density analysis (Fig. 4a) [50]. The reactive site was further elucidated using the Local Density of States (LDOS) of the valence band maximum (VBM) and conduction band minimum (CBM). The study reveals that calcium atoms can exhibit reactivity upon nucleophilic attacks, specifically from OH (Fig. 4b). Conversely, oxygen atoms bonded to silicon atoms in SiO^4 tetrahedra are expected to remain inert but play a significant role in reactivity during electrophilic attacks, such as H^+ (Fig. 2c) [50]. The distinction between two types of oxygen atoms was

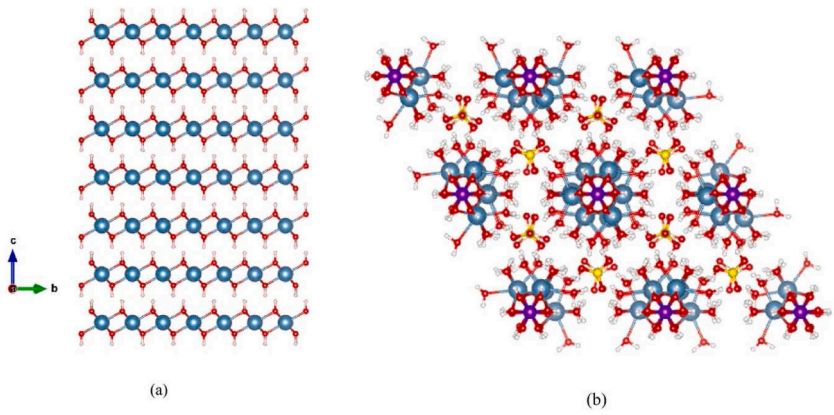

Fig. 3. Bulk structure of (a) portlandite (6x6x6 supercell) and (b) ettringite (2x2x1 supercell). Colour code Light Blue: Ca, Purple: Al, Red: O, White: H, yellow: S [8]. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

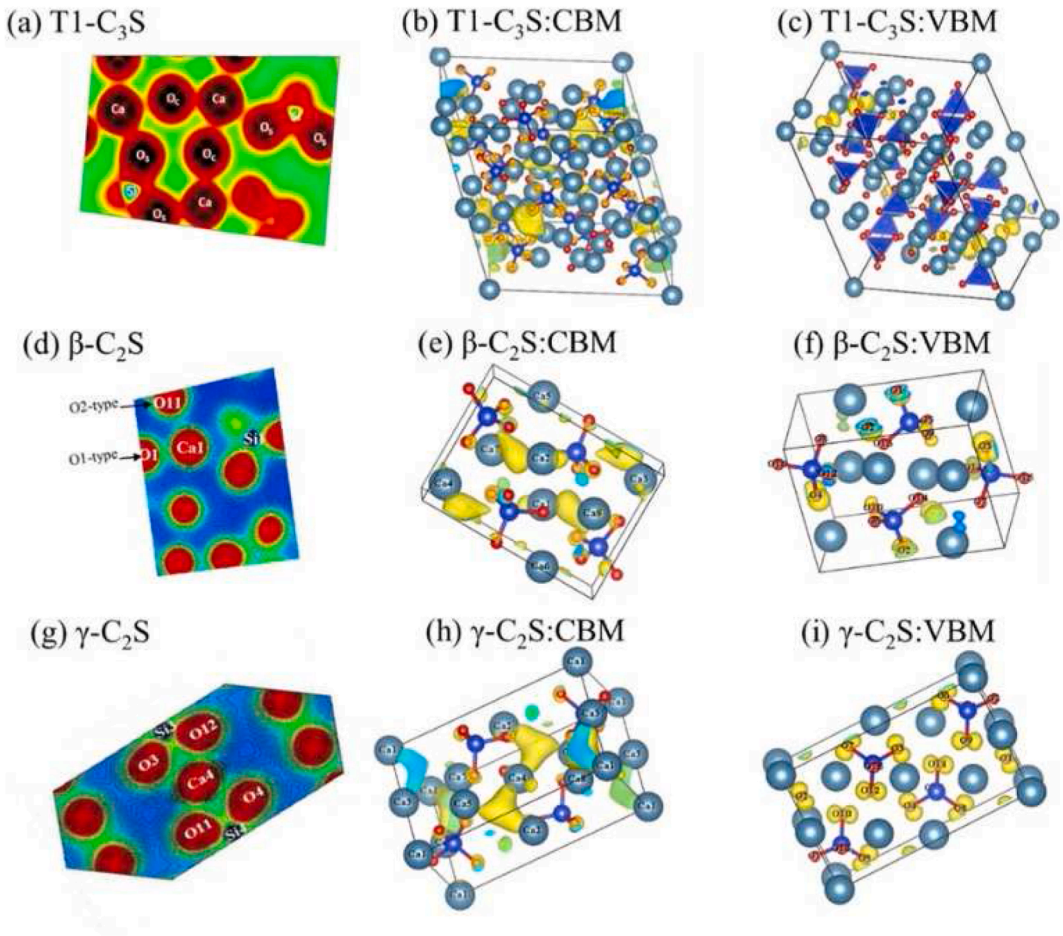

Fig. 4. Charge density distribution and LDOS of VBM and CBM respectively for: (a) (b) and (c) T1-C3S; (d), (e) and (f) β-C_2S; (g), (h) and (i) γ-C_2S. Atomic colour codes are oxygen: red, calcium: turquoise, silicon: blue [50,51]. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

also observed in β-C_2S (Fig. 4d), whereas γ-C_2S displayed a high level of symmetry in the charge distribution across all oxygen atoms (Fig. 2g). The localised density of states (LDOS) of the valence band maximum (VBM) around one type of oxygen atoms in β-C_2S indicated a high reactivity towards electrophilic attacks (Fig. 2e). Conversely, the uniformly distributed LDOS of VBM in γ-C_2S explained its non-reactivity towards water (Fig. 2i) [51].

In a study, Kunhi Mohamed et al. (2020) [52] utilized the brick model and experimental NMR measurements to investigate the coordination geometries of aluminum in C-A-S-H (calcium-aluminum-silicate-hydrate) (Fig. 5a). The researchers employed a GIPAW (Gauge Including Projector Augmented Wave) approach based on DFT (Density Functional Theory) calculations to calculate NMR chemical shifts of aluminum and silicon atoms. Their findings provided evidence against the existence of a third aluminate hydrate (TAH) proposed to explain the six-fold coordinated aluminum in C-S-H. These atomistic simulation methods proved to be highly valuable in unravelling the nanostructure of C-S-H. Moreover, the structural insights obtained from the brick model of C-S-H contributed to the development of a new C-A-S-H+ sublattice solid solution model within the GEMS thermodynamic modelling software [53]. The free energy of complexation for few early-age hydration species in solution is presented in Fig. 5b.

Overall, the application of MD simulation in concrete research has advanced our understanding of concrete materials at the atomic and molecular levels. It has provided valuable information about their structural properties, mechanical behaviour, hydration process, and interfacial interactions. However, it is important to note that MD simulations have limitations, such as the size and timescale constraints. The complexity of concrete at larger scales and longer timeframes still poses challenges for MD simulations. Therefore, a combination of experimental and computational approaches is necessary to gain a comprehensive understanding of concrete behaviour. Future research should focus on addressing these challenges and further refining MD models to enhance their accuracy and predictive capabilities in concrete studies.

3.2. Benefits and limitations of using molecular dynamic simulation in concrete research

Benefits of using molecular dynamics simulation in concrete research:

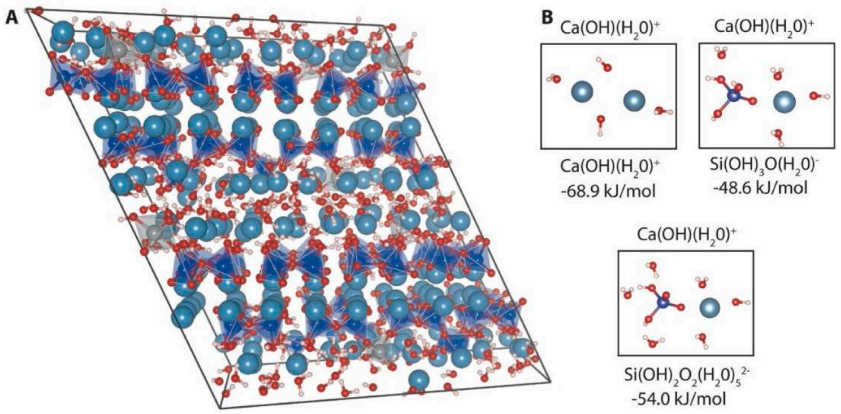

Fig. 5. (A) A bulk atomic-level structure of aluminum incorporated C-S-H) [52]. (B) Free energy of complexation of a few early age hydration species in solution [53]. Atomic color codes are oxygen: red, hydrogen: white, calcium: turquoise, silicon: blue, aluminum: grey. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

1. Atomistic Understanding: Molecular dynamics simulation provides an atomistic-level understanding of the behaviour of concrete constituents, such as cement particles, water and aggregates. It allows researchers to investigate the interactions, structural properties, and dynamic processes at the molecular scale, which are not easily accessible through experimental techniques alone.
2. Time and Length Scale: MD simulations can cover longer time scales compared to experiments, enabling the study of processes that occur over microseconds to milliseconds. Additionally, simulations can span larger length scales, providing insights into the behaviour of concrete at the nanoscale and mesoscale.
3. Cost-Effectiveness: MD simulations can be a cost-effective alternative to experimental testing, as they reduce the need for extensive laboratory experiments and material synthesis. Simulations can provide preliminary insights and guide experimental efforts by narrowing down the parameter space.
4. Exploration of Multiple Parameters: Simulations offer the flexibility to explore a wide range of parameters, such as temperature, pressure, composition and hydration conditions. This enables researchers to investigate the effects of different factors on concrete properties and behaviour systematically.
5. Predictive Capabilities: Molecular dynamics simulations have the potential to predict material properties and behaviour, allowing researchers to gain insights into the mechanical, thermal and chemical properties of concrete. This predictive capability can guide material design and optimization processes.

Limitations of using molecular dynamics simulation in concrete research:

1. Computational Resources: MD simulations require significant computational resources, including high-performance computing clusters or supercomputers, to handle the large number of atoms and complex interactions involved in concrete systems. This can limit the size and time scale of simulations that can be performed.
2. Simplified Models: Simulations involve approximations and simplifications to describe complex phenomena accurately. Force fields and potential models used in MD simulations have inherent limitations, and the accuracy of results depends on the chosen models and parameterisation.
3. Time Scale Limitations: While MD simulations can cover longer time scales than experiments, simulating the entire hydration process of concrete, which spans weeks to months, is still challenging. Coarse-grained or accelerated simulation techniques are often used to bridge the gap between atomistic simulations and experimental time scales.
4. Uncertainty in Parameters: Accurate parameterisation of force fields and potential models is crucial for obtaining reliable results. However, parameterisation can be challenging, and uncertainties in parameters can affect the accuracy and reliability of simulations.
5. Complexity of Interactions: Simulating the complex interactions between different components of concrete, such as aggregates, interfaces and additives, requires advanced modelling techniques and accurate representation of intermolecular forces. Developing accurate models for complex systems is an ongoing challenge in MD simulations.

Overall, molecular dynamics simulation offers valuable insights into the behaviour of concrete at the molecular level. It provides a complementary approach to experimental techniques, allowing researchers to gain a deeper understanding of concrete properties, explore new materials, and optimize design parameters. However, careful consideration of limitations and validation against experimental data is necessary to ensure the accuracy and reliability of MD simulations in concrete research.

3.3. Effect of molecular dynamic simulation on understanding the mechanical and durability properties of concrete

Molecular dynamics (MD) simulation has emerged as a powerful tool for understanding the mechanical and durability properties of concrete at the atomic and molecular level. By simulating the behaviour of concrete constituents and their interactions, MD simula-

tions provide valuable insights into the fundamental mechanisms that govern the mechanical strength and durability performance of concrete.

3.3.1. Mechanical properties

MD simulation has revolutionized our understanding of the mechanical properties of concrete by providing detailed insights into critical aspects such as stress transfer and deformation. By modelling the interactions between individual atoms, MD simulations allow researchers to observe how forces are transmitted and distributed within the material. This atomic-level information is invaluable for comprehending the load-bearing capacity and overall mechanical behaviour of concrete, enabling the design of more resilient and structurally sound structures.

Crack initiation and propagation are fundamental phenomena in concrete mechanics, and MD simulations play a crucial role in unravelling the intricacies of these processes. By simulating the behaviour of atoms, researchers can observe the nucleation and growth of cracks, study their propagation pathways, and examine the underlying factors that influence crack formation and development. This knowledge is essential for predicting and mitigating structural failures, improving the safety and durability of concrete infrastructure.

The interfacial bonding between different components of concrete, such as aggregates and the surrounding matrix, significantly affects the material's mechanical properties. MD simulations provide a powerful tool for investigating interfacial bonding strength and understanding its influence on the overall performance of concrete. By analysing the interactions between atoms at the interface, researchers can gain insights into the mechanisms governing adhesion, cohesion, and interfacial fracture. This understanding aids in optimizing the composition and design of concrete, enhancing its strength, durability, and long-term performance.

In addition to stress transfer, crack initiation and propagation, and interfacial bonding, MD simulations have been extensively utilized to explore other mechanical properties of concrete. These include elasticity, plasticity, and fracture behaviour. By considering the atomic-level interactions and deformation mechanisms, researchers can delve deeper into the fundamental behaviours of concrete, shedding light on its response to external loading, deformation characteristics, and failure mechanisms. This knowledge facilitates the development of advanced computational models and predictive tools for designing concrete structures with enhanced performance and resilience.

3.3.2. Durability properties

MD simulation has significantly advanced our understanding of the durability properties of concrete by providing detailed insights into various aspects such as transport behaviour, degradation mechanisms, and mechanical response.

Transport behaviour in concrete, particularly the ingress and diffusion of aggressive substances, is a critical factor influencing its durability. MD simulations allow researchers to investigate the transport mechanisms, rates, and preferential pathways of ions, water, and gases within the concrete matrix. By simulating the interactions between atoms and molecules, MD simulations can provide a microscopic view of how these substances move through the pore structure of concrete. This knowledge helps in understanding phenomena such as moisture migration, ion diffusion, and gas permeability, which play vital roles in the deterioration and performance of concrete structures. The insights gained from MD simulations assist in optimizing the composition and design of concrete mixtures to achieve reduced permeability and enhanced durability.

The study of degradation mechanisms is another key area where MD simulations have made significant contributions. By simulating the interactions between concrete constituents and environmental factors, such as temperature, moisture, and chemical exposure, MD simulations can elucidate the underlying processes that lead to deterioration. For instance, MD simulations can be employed to study the alkali-silica reaction (ASR), a common durability issue in concrete. By modelling the interactions between alkalis, silica, and water molecules at the atomic level, MD simulations provide insights into the mechanisms of ASR-induced expansion and associated damage. Similarly, MD simulations can investigate sulphate attack by simulating the interactions between sulphate ions and cementitious phases, helping to understand the degradation mechanisms and develop mitigation strategies. The knowledge obtained from MD simulations guides the development of more durable concrete materials and the formulation of effective preventive measures.

In addition to transport and degradation, MD simulations contribute to understanding the mechanical response of concrete, which is closely linked to its durability. By modelling the behaviour of individual atoms, MD simulations provide valuable insights into crack initiation and propagation, interfacial bonding strength, and the effects of loading on concrete structures. These simulations can help identify the mechanisms underlying crack growth and the factors influencing the overall durability of concrete structures. Furthermore, MD simulations enable the study of interfacial bonding between different phases of concrete, such as the interface between aggregates and the surrounding matrix. By analysing the interactions between atoms at these interfaces, researchers can gain insights into the adhesion, cohesion, and interfacial fracture behaviour, all of which significantly affect the durability of concrete under mechanical stresses and environmental exposures.

In summary, MD simulations have revolutionized our understanding of the durability properties of concrete by providing insights into transport behaviour, degradation mechanisms, and mechanical response at the atomic level. This knowledge assists in the development of more durable and sustainable concrete materials, leading to improved durability performance and reduced life-cycle costs. By combining experimental observations with MD simulations, researchers can gain a comprehensive understanding of concrete's durability, enabling them to design more resilient structures and optimize maintenance strategies.

3.3.3. Material design and optimization

Additive and Admixture Effects: MD simulations allow for the virtual screening and optimization of concrete additives and admixtures, helping to identify promising candidates for enhancing specific mechanical or durability properties of concrete.

Particle Size and Shape Effects: By studying the influence of particle size, shape, and distribution on concrete properties, MD simulations contribute to the design of tailored concrete materials with improved performance characteristics.

Nanostructuring and Self-Healing: MD simulations facilitate the exploration of novel nanostructured materials and self-healing mechanisms, enabling the development of advanced concrete materials with enhanced mechanical strength and improved durability.

3.3.4. Multiscale modelling

Bridging Length and Time Scales: MD simulations serve as a valuable tool for bridging the gap between the atomic-scale details and macroscopic behaviour of concrete, contributing to the development of multiscale models that capture the hierarchical nature of concrete materials.

Validation and Integration: MD simulations are integrated with experimental techniques to validate and refine computational models, enabling a more comprehensive understanding of concrete behaviour across different length and time scales.

While MD simulations have significantly advanced our understanding of the mechanical and durability properties of concrete, it is important to note that experimental validation and collaboration between computational and experimental approaches are crucial. The combination of MD simulations with experimental testing allows for a more robust characterization of concrete behaviour, leading to improved material design and optimized performance.

4. Simulation techniques and models

4.1. Techniques for simulating concrete at different scales

Molecular dynamics (MD) simulations of concrete can be performed at different scales to study its behaviour and properties. Here are some techniques commonly used for MD simulations of concrete at various scales:

1. Atomistic Scale: At the atomistic scale, two main approaches are commonly used for simulating concrete materials: All-Atom Molecular Dynamics (MD) and Reactive Force Fields. All-Atom MD represents each atom explicitly and uses interatomic potentials to describe their interactions [54–59]. This approach provides detailed information about atomic positions, bond breaking and formation, and chemical reactions during cement hydration. However, it is computationally intensive and limited to small system sizes and short timescales. Reactive Force Fields, such as ReaxFF, incorporate bond-breaking and bond-forming processes within the force field formalism [60–63]. These models can capture chemical reactions occurring during cement hydration and are suitable for studying complex reaction mechanisms in concrete.
2. Coarse-Grained Scale: At the coarse-grained scale, Mesoscale MD and Dissipative Particle Dynamics (DPD) are commonly employed. Coarse-grained MD simulations represent several atoms as a single interaction site or a coarse-grained particle, reducing computational cost and enabling longer simulation times and larger system sizes. Mesoscale models are useful for studying phenomena occurring over larger length and time scales, such as self-assembly of cement particles or diffusion in cementitious materials. DPD is a mesoscale simulation method that combines atomistic and hydrodynamic behaviours [64–66]. It can capture the mesoscopic behaviour of water and cement particles, including their interactions and transport properties, in a computationally efficient manner.
3. Continuum Scale: At the continuum scale, the Finite Element Method (FEM) is commonly used. FEM combines MD simulations with continuum mechanics to model the behaviour of concrete at a larger scale [67,68]. It bridges the atomistic and macroscopic scales by incorporating properties obtained from MD simulations into a continuum framework. FEM is suitable for studying the mechanical behaviour of concrete structures and predicting their response under various loading conditions.
4. Multi-Scale Approaches: Multi-Scale MD simulations aim to integrate different length and time scales to accurately capture the behaviour of concrete materials. They combine atomistic, coarse-grained, and continuum models to simulate phenomena occurring at different scales [69,70]. These approaches provide a hierarchical description of concrete properties and can account for the interactions between different scales in a computationally efficient manner. They offer a comprehensive understanding of concrete behaviour, considering both atomistic details and macroscopic responses.

The choice of simulation technique depends on the research objectives, the level of detail required, and the available computational resources. While atomistic MD provides the most detailed information, it is computationally expensive and limited in terms of system size and timescale. Coarse-grained and continuum models offer a compromise between computational efficiency and accuracy, enabling the study of larger systems and longer timescales. Multi-scale approaches provide a comprehensive understanding of concrete behaviour by combining multiple simulation techniques at different scales.

4.2. Models for simulating the behaviour of different components in concrete

In molecular dynamics (MD) simulations of concrete, various models are employed to simulate the behaviour of different components. These models capture the interactions and properties of specific constituents of concrete. Here are some commonly used models for simulating the behaviour of different components in concrete:

4.2.1. Cement particles

Classical Force Fields: Classical force fields, such as the ClayFF and CHARMM force fields, are widely employed to describe the interactions between atoms in cement particles. These force fields include parameters for calcium, silicon, aluminium, oxygen, and hydrogen atoms, enabling simulations of cement hydration and mechanical properties of cement particles. They provide valuable insights into the atomic-level behaviour of cement particles [71,72].

4.2.2. Water

Flexible and Rigid Water Models: Molecular dynamics simulations of water in cementitious materials can utilize flexible or rigid water models depending on the desired level of detail. Flexible water models, like the TIP3P and SPC/E models, account for explicit atomic interactions and accurately capture the flexibility of water molecules. Rigid water models, such as the TIP4P and TIP5P models, simplify water molecules as rigid entities, significantly reducing computational costs while still capturing essential properties [73,74].

4.2.3. Aggregate particles

Silica Models: Simulating silica aggregates requires appropriate models such as the BKS (Beest-Kramer-van Santen) potential or the ClayFF force field. These models accurately represent the atomic interactions within silica and can capture its structural properties and mechanical behaviour, providing insights into the behaviour of silica-based aggregates in cementitious materials [75,76].

Calcium Silicate Models: Calcium silicate hydrate (C-S-H) is a crucial component of cementitious materials and can be modelled using various empirical potentials. Potentials like the clay-like potential describe the interactions between calcium, silicon, oxygen, and hydrogen atoms in C-S-H, enabling simulations to investigate the structural and mechanical properties of C-S-H [38,46,47].

4.2.4. Interfaces and interactions

Interface Models: Modelling interfaces between different components in concrete, such as the cement-aggregate interface or the polymer-cement interface, requires specialized models. These models consider the interactions between different materials, taking into account surface energies, interfacial bonding, and adhesion properties. They provide valuable insights into the behaviour of interfaces within concrete [77,78].

ReaxFF: The ReaxFF force field is commonly utilized in concrete research to capture complex chemical reactions occurring during cement hydration. It enables simulations of bond-breaking and bond-forming processes, making it suitable for studying reactive interfaces and chemical reactions within concrete systems. ReaxFF offers a powerful tool to investigate the reactivity and behaviour of concrete at the atomic level [8,60].

These models enable researchers to simulate the behaviour of individual components in concrete, such as cement particles, water, aggregates, and interfaces. By combining these models and simulating their interactions, researchers can gain insights into the overall behaviour and properties of concrete at the molecular scale. It is important to select appropriate models that accurately represent the desired components and phenomena of interest in order to obtain reliable and meaningful results in MD simulations of concrete.

5. Future directions and research needs

5.1. Recommendations for future research on molecular dynamic simulation in concrete

While molecular dynamics (MD) simulations have already shown great promise in studying concrete at the atomic and molecular level, there are several areas where future research can further enhance our understanding and application of MD simulations in this field. Here are some recommendations for future research:

1. Multi-scale modelling: Concrete is a complex material with hierarchical structures spanning from the nano-to macroscale. Future research can focus on developing multi-scale MD models that can bridge the gap between atomistic simulations and macroscopic behaviour. This will enable a more comprehensive understanding of the mechanical, thermal, and transport properties of concrete.
2. Incorporating realistic environmental conditions: Concrete is exposed to various environmental conditions, including temperature variations, moisture, chemical exposure, and external loading. Future research can explore the incorporation of these realistic environmental conditions into MD simulations to study the degradation mechanisms and long-term performance of concrete structures. This will help in designing more durable and sustainable concrete materials.
3. Modelling of cement hydration: Cement hydration is a complex chemical process that governs the development of concrete strength and durability. Further research can focus on developing accurate MD models to simulate cement hydration reactions, including the nucleation and growth of hydration products. This will provide insights into the early-age properties of concrete and enable the optimization of mix designs.
4. Incorporating admixtures and supplementary cementitious materials: Concrete often includes admixtures and supplementary cementitious materials (SCMs) to improve its properties and sustainability. Future research can investigate the interactions between cementitious materials, admixtures, and SCMs at the molecular level using MD simulations. This will help in understanding their influence on concrete performance and guide the development of greener and more effective concrete mixtures.
5. Simulation of concrete interfaces: Interfaces play a crucial role in the behaviour of concrete, such as the bond between aggregates and matrix, and the interaction between concrete and reinforcing materials. Future research can focus on simulating and characterizing these interfaces using MD simulations to better understand their mechanical properties, adhesion and potential failure mechanisms. This knowledge can lead to improved design and performance of concrete structures.
6. Validation and experimental correlation: MD simulations need to be validated against experimental data to ensure their accuracy and reliability. Future research can emphasize the validation of MD models and the correlation of simulation results with experimental observations. This will enhance the confidence in using MD simulations as a predictive tool for concrete behaviour.

7. High-performance computing and algorithm development: MD simulations can be computationally demanding, especially for large-scale systems and long-time simulations. Future research can explore the development of efficient algorithms and parallel computing techniques to accelerate MD simulations of concrete. This will enable the study of larger and more realistic systems, leading to more accurate predictions and practical applications.

By addressing these research areas, we can advance the capabilities of MD simulations in studying concrete and contribute to the development of sustainable and high-performance construction materials and structures.

5.2. The challenges and opportunities for scaling up molecular dynamic simulation for concrete research

Scaling up molecular dynamics (MD) simulations for concrete research presents both challenges and opportunities. Here are some key considerations:

Challenges:

1. System size: Concrete is a complex material with a large number of atoms, making it challenging to simulate at the atomic scale. The computational cost increases exponentially with the system size, limiting the ability to simulate large-scale concrete structures using MD. Developing efficient algorithms and utilizing high-performance computing resources are essential to overcome this challenge.
2. Time scale: Concrete behaviour involves processes that occur over a wide range of time scales, from nanoseconds to years. Capturing long-term phenomena, such as creep and aging, within the limitations of MD simulations remains a challenge. Advanced sampling techniques, accelerated molecular dynamics methods, and coupling MD with other simulation techniques can be explored to overcome time scale limitations.
3. Accuracy of force fields: MD simulations rely on empirical force fields to describe the interactions between atoms. Developing accurate force fields that can capture the complex behaviour of concrete, including its hydration, aging and failure mechanisms, is a significant challenge. Improving force field parameters and developing more sophisticated models that capture specific properties of concrete, such as calcium-silicate-hydrate (C-S-H) gel, will enhance the accuracy of MD simulations.
4. Experimental validation: Validating MD simulations with experimental data is crucial to ensure their reliability and accuracy. However, obtaining detailed experimental data at the atomic level for concrete is challenging. Collaboration between experimentalists and computational researchers is essential to provide reliable data for model validation and to iteratively improve the simulation accuracy.

Opportunities:

1. Insight into atomic-scale processes: MD simulations offer the unique opportunity to investigate concrete behaviour at the atomic scale. By capturing the interactions between individual atoms, MD simulations can provide insights into fundamental mechanisms underlying concrete properties, such as hydration, mechanical behaviour and transport phenomena. This knowledge can guide the development of new concrete formulations and optimize existing ones.
2. Prediction of material properties: MD simulations can predict material properties of concrete, such as elastic modulus, strength and fracture toughness, by simulating the behaviour of concrete at the atomic level. These predictions can complement experimental testing and aid in the design of tailored concrete materials with desired properties.
3. Understanding degradation mechanisms: MD simulations can shed light on the degradation mechanisms of concrete, including chemical attack, alkali-silica reaction and corrosion of reinforcement. By simulating these processes at the atomic scale, researchers can gain a deeper understanding of degradation mechanisms and develop mitigation strategies to improve the durability of concrete structures.
4. Optimization of concrete mixes: MD simulations can assist in the optimization of concrete mixtures by exploring the effects of different materials, additives and mix proportions on the resulting properties. This can contribute to the development of sustainable concrete mixes with improved performance and reduced environmental impact.
5. Virtual testing and design: MD simulations can serve as virtual testing platforms, allowing researchers to evaluate the behaviour of concrete under various loading conditions and environmental exposures. This virtual testing can help reduce the need for costly and time-consuming experimental tests, enabling more efficient design and optimization of concrete structures.

Addressing these challenges and capitalising on the opportunities can pave the way for scaling up MD simulations for concrete research. Continued advancements in computational methods, collaboration between experimentalists and computational researchers, and access to high-performance computing resources will be crucial in realising the full potential of MD simulations for concrete research and development.

6. Conclusions

6.1. Summary of the key findings and implications for the use of molecular dynamic simulation in concrete

Molecular dynamics (MD) simulations have been used to study various aspects of concrete at the atomic and molecular level. Here are some key findings and implications for the use of MD simulations in the context of concrete:

1. Understanding cement hydration: MD simulations have provided insights into the hydration process of cement, including the nucleation and growth of hydration products such as calcium-silicate-hydrate (C-S-H) gel. These simulations have helped in understanding the structural and mechanical properties of C-S-H gel, which is a major component of hardened concrete.
2. Predicting mechanical properties: MD simulations have been used to predict the mechanical properties of concrete, such as its elastic modulus, strength, and fracture behaviour. By modelling the interactions between cementitious materials, aggregates and water, MD simulations can provide information on the microstructural origins of mechanical behaviour and guide the design of high-performance concrete.
3. Investigating transport phenomena: MD simulations have shed light on transport phenomena in concrete, including water permeability, diffusion of ions and gas adsorption. These simulations have helped in understanding the factors influencing durability and the transport of harmful substances, such as chloride ions or carbon dioxide, within concrete structures.
4. Exploring interfaces and interfacial properties: MD simulations have been used to study the interfaces between cementitious materials and aggregates, as well as the interactions between cement and reinforcing materials. These simulations have provided insights into interfacial bonding, interfacial strength and the mechanisms of crack initiation and propagation in concrete.
5. Assessing durability and aging effects: MD simulations have been utilized to investigate the aging and deterioration processes in concrete, including the effects of moisture, temperature, and chemical attack. By simulating the interactions between concrete constituents and external environments, MD simulations can help assess the long-term durability and performance of concrete structures.

Overall, MD simulations offer a valuable tool for studying the behaviour of concrete at the atomic and molecular level, providing insights into its mechanical, transport, and durability properties. The findings from MD simulations can inform the development of improved concrete materials, optimize mix designs, and enhance the understanding of concrete performance in different conditions, contributing to more sustainable and durable infrastructure.

6.2. Final thoughts and potential impact of molecular dynamic simulation on the sustainability of the construction industry

Molecular dynamics (MD) simulations have the potential to significantly impact the sustainability of the construction industry. By providing detailed insights into the behaviour of construction materials at the atomic and molecular level, MD simulations can contribute to the development of more sustainable and efficient practices in several ways:

1. Material design and optimization: MD simulations can aid in the design and optimization of construction materials, such as concrete, by providing a deeper understanding of their structural, mechanical, and transport properties. This knowledge can be utilized to develop materials with improved performance, durability and environmental impact, leading to more sustainable construction practices.
2. Reduced experimental costs and time: MD simulations offer a cost-effective and time-efficient alternative to extensive experimental testing. By simulating various scenarios and properties, MD simulations can guide the selection of materials, mix designs, and construction methods, reducing the need for resource-intensive trial-and-error approaches. This can lead to significant cost savings and faster development of sustainable construction solutions.
3. Improved energy efficiency: MD simulations can assist in optimizing energy efficiency in construction processes. By modelling heat transfer, energy storage and energy conversion mechanisms, simulations can guide the design of energy-efficient building materials, insulation systems, and renewable energy integration strategies. This can contribute to reduced energy consumption and greenhouse gas emissions in the construction industry.
4. Enhanced durability and resilience: MD simulations can help in assessing the long-term durability and resilience of construction materials and structures. By studying aging processes, deterioration mechanisms and the effects of external factors, simulations can guide the selection of materials and design strategies that improve the lifespan and performance of structures. This can reduce the need for frequent maintenance and replacement, resulting in reduced resource consumption and waste generation.
5. Environmental impact assessment: MD simulations can support the assessment of the environmental impact of construction materials and processes. By modelling emissions, pollution, and resource utilization, simulations can provide valuable data for life cycle assessments and sustainability evaluations. This information can guide decision-making towards more environmentally friendly practices and contribute to the overall sustainability of the construction industry.

In conclusion, molecular dynamics simulations have the potential to revolutionise the construction industry by providing valuable insights into material behaviour, optimizing designs, reducing costs and time, improving energy efficiency, improving mechanical properties, enhancing durability, and facilitating environmental impact assessments. Integrating MD simulations into construction practices can drive the development of sustainable and resilient infrastructure, paving the way for a greener and more sustainable future for the construction industry.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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