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Opening letter of RILEM TC ASM: Atomistic simulation for cement-based materials: Recommendations and link to experiments

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Abstract

Over the past decades, molecular simulations have become a central tool in understanding the fundamental physico-chemical processes in cement-based materials that in turn enable rational design and development of cementitious systems and chemical admixtures for practical applications. Despite the breadth of work, approaches remain heterogeneous. Different research groups employ varied atomic models, force fields, and simulation protocols, often leading to discrepancies, inconsistencies preventing direct comparisons of different results, difficulties in reproducibility, and lack of systematic multiscale validation frameworks which can connect atomistic predictions with meso- and macroscale experiments. The *RILEM TC ASM Atomistic Simulations for Cement-Based Materials: Recommendations and Link to Experiments* has been established to address these issues by reviewing existing modeling practices, establishing recommended protocols, and creating open-access resources to support both academic and industrial communities.

Keywords: Molecular simulations, Cement physical-chemistry, Force fields (FFs), Multiscale modeling, Model validation, Nanoscale experiments.

1 Introduction

Molecular scale modeling over the last decades has become an important tool to gain insights into fundamental physico-chemical processes occurring at the micro- and nano-structural scales in cementitious materials which otherwise remain difficult to capture experimentally. With the rapid development of materials and the current drive towards low-carbon and sustainable concrete, it becomes increasingly necessary to fundamentally understand these materials without relying on empiricism to ensure accurate and reliable service life predictions for structures. Nevertheless, the modeling approaches from different research groups remain largely varied, to the extent that algorithms with unreasonable atomic structures, force fields (FF), boundary conditions or sampling methods might be reported. There is a need to review and recommend a baseline for molecular modeling work in cementitious systems to bring homogeneity to the field and to guide researchers. Additionally, with the rise in the digitalization technology and science, alongside the

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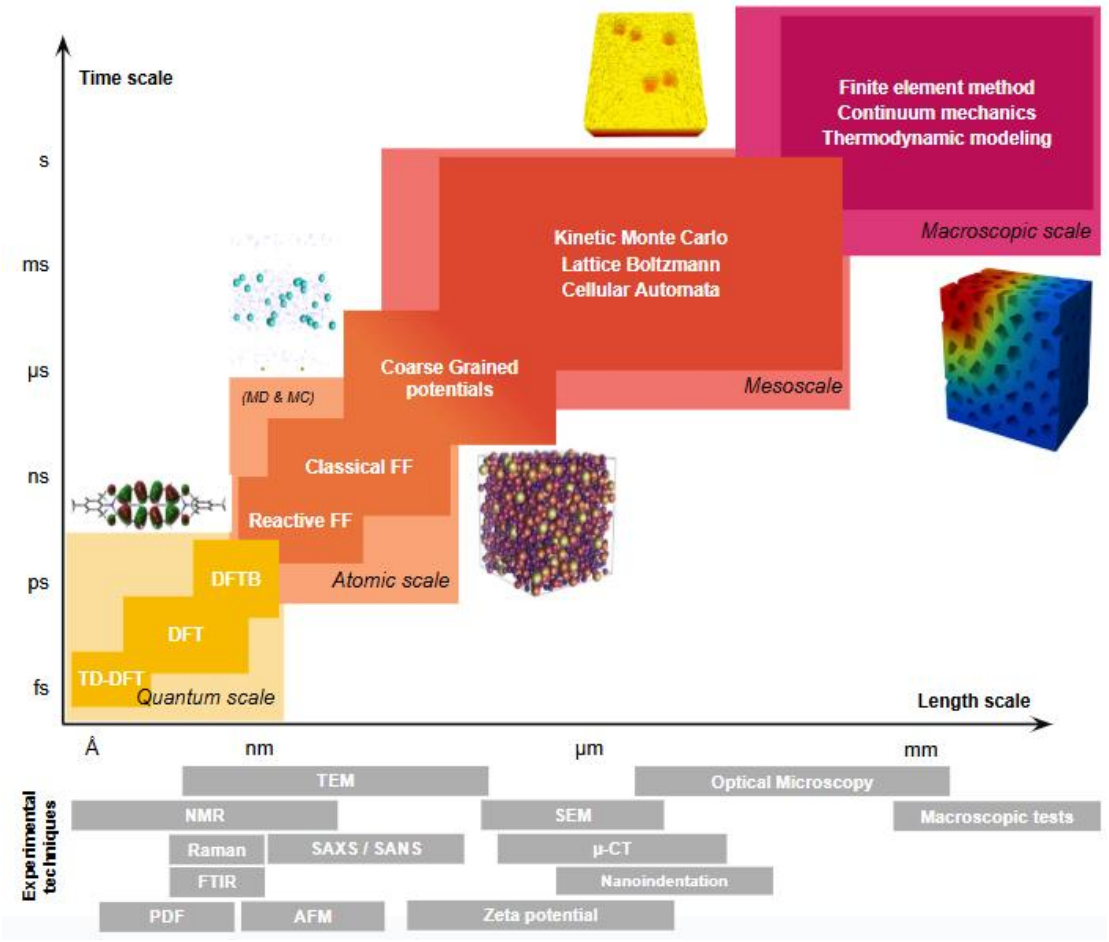

Figure 1. Multiscale modeling and scope of TC ASM including quantum-scale methods, classical molecular scale and mesoscale approaches, which can find experimental support in various techniques (TEM, NMR, FTIR, Raman, AFM, SAXS/SANS, etc.). The time and length scales shown are indicative, with a focus on small scales; however, some techniques can straightforwardly span much larger ranges. For example, Lattice Boltzmann can simulate days at the pore scale, while continuum models can reach years.

growing reliance on data-driven approaches that demand standardized databases, it is essential to address the current fragmentation in molecular modeling work within cementitious systems and establish a reliable baseline. The TC is inscribed within the scope of multiscale modeling, and multi-technique experimental evaluations across scales are depicted in Figure 1.

The *TC ASM Atomistic Simulations for Cement-Based Materials: Recommendations and Link to Experiments* aims to comprehensively review the atomic structures and force fields used in the modeling of cementitious materials, review and recommend the tools and algorithms for different modeling purposes, review and recommend upscaling strategies (including mesoscale modeling) and to benchmark the modeling results with experimental data. The primary focus will be minerals, solutions, surfaces and chemicals that are most common in modern cementitious systems, such as clinker minerals, cement-water suspensions, and hardened cement pastes. However, we may extend our activities to include other materials of interest for RILEM.

2 Current status of atomistic and molecular simulations research

2.1 Current status in other research fields

Molecular simulations have become a fundamental tool across a wide range of disciplines, including, e.g., protein research, drug discovery, polymer design, materials for energy storage, optoelectronics. Ongoing advances in computational power, algorithmic efficiency, and the integration of machine learning (ML) techniques are rapidly expanding their scope, accuracy, and accessibility. These developments are transforming molecular simulations from exploratory tools into predictive frameworks capable of guiding experimental design and materials optimization. For instance, in *biomolecular research* and *pharmaceutical sciences*, molecular simulations routinely provide atomistic insights into protein dynamics, ligand–receptor interactions, and disease-related conformational changes, thereby informing rational drug design and optimization strategies [1, 2]. In *molecular sciences*, they are extensively employed to

predict phase equilibria and to investigate self-assembly, crystallization, and chemical reactions in solutions, suspensions, polymers, and other macromolecular systems [3]. In *materials science*, molecular simulations play a crucial role in elucidating the structural, thermodynamic, and mechanical behavior of complex materials, enabling the design of novel compounds with tailored properties and improved performance [4] .

Machine learning techniques are increasingly being integrated into molecular simulations, particularly for the construction of interatomic potentials and for accelerating sampling strategies [2, 5–7]. Rather than replacing physics-based modeling approaches, these methods enable simulations that retain atomistic resolution while approaching quantum-mechanical accuracy for larger and more chemically complex systems. In this context, ML-based force fields should be understood as extensions of atomistic modeling frameworks trained on physically meaningful datasets, rather than purely empirical fitting tools. Their main contribution lies in improving transferability across chemical environments, enabling uncertainty quantification, and facilitating integration with multiscale and experimental datasets. At the same time, challenges remain regarding robustness, reproducibility, and transferability across the chemically heterogeneous systems typical of cementitious materials. Addressing these challenges will be essential for consolidating ML-enhanced simulations as reliable predictive tools for cement science. Databases dedicated for engineering materials such as Atomgptlab, NOMAD, Alexandria and Materials Project enable acceleration of simulation-based strategies using ML. The emergence of cloud computing and open-source platforms, such as OpenMM and Google Colab, has significantly broadened access to high-performance molecular simulations, making them more attainable for educational use and resource-limited research environments. Parallel advances in interactive, web-based visualization tools are further enhancing the interpretability and reusability of molecular dynamics data, thereby fostering transparency, reproducibility, and collaborative research [8]. Also, educational platforms such as nanoHUB are further broadening access to simulation tools across the materials science community

Despite these achievements, several challenges remain. Ensuring the scalability and transferability of ML-enhanced models across chemically diverse systems continues to be a major limitation [9]. Moreover, achieving reproducibility and interoperability requires the standardization of simulation workflows, metadata, and data-sharing practices. Addressing these challenges will be critical for consolidating molecular simulations as reliable, predictive, and interpretable tools for the next generation of scientific and engineering applications.

2.2 Current status in modeling of cementitious systems

The application of molecular simulation to cementitious materials began in the late 1990s, emerging from methodologies pioneered in clay and geochemical research. The very first ab-initio study on aluminum in tobermorite, which mentioned the C-S-H gel, was reported by Kashihara et al. [10], laying the groundwork for subsequent atomistic investigations. The first molecular dynamics simulations of C-S-H were later conducted at the *Commissariat à l'énergie atomique et aux énergies alternatives* (CEA) in France at the end of the 1990s [11, 12], marking the beginning of atomistic studies on cementitious materials. These early works were followed by pioneering investigations of water-tobermorite and ettringite interfaces by Kalinichev [13, 14], stimulated in the sequel by the development of the ClayFF force field [15].

By the early 2000s, molecular simulation of cement became a subfield of cement science. Several research groups began specializing in atomistic modeling of cement hydrates and related phases, rather than treating cement as merely an offshoot of clay or mineral studies. For example, White and Provis applied molecular simulation methods to supplementary cementitious materials [16]. Around the same time, the doctoral work by Manzano and colleagues exemplified this new focus by using MD to explore the nanostructure and mechanics of C–S–H and other cement [17, 18]. The field's momentum further accelerated in the late 2000s, highlighted by the introduction of increasingly realistic models for C–S–H chemistry and structure. A milestone achievement was the "realistic molecular model" of C–S–H proposed by Pellenq et al. [19]. This model provided, for the first time, an atomistic representation of C–S–H that captured its disordered, semi-crystalline nature and matched many experimental observations. Its publication opened the door to a new wave of simulations targeting the structure and properties of cement hydrates. In subsequent years, many improved structural models for C–S–H (and C–A–S–H, its aluminum-containing variant) were put forward, refining our understanding of compositional variability and atomic arrangement in this phase (Casar et al., 2024; Kovačević et al., 2015; Kunhi Mohamed et al., 2020, 2020, 2018; Pellenq et al., 2009; Qomi et al., 2014; Richardson, 2014). Each new model integrated progressively more realistic features (such as variable Ca/Si ratios, defect sites, and interlayer water structuring), bringing simulations closer to the observed experimental features. Concurrently, force field development kept pace with these structural advances. With ClayFF as a starting point Shahsavari et al. formulated improved empirical potentials for C–S–H, and Mishra et al. developed the Interface force field (IFF) tuned for cementitious mineral interactions and Galmarini et al. worked on the CementFF suite of FFs [20–27]. Furthermore, ReaxFF reactive force fields were parameterized for cement constituents [28, 29], enabling the simulation of chemical reactions such as hydration and dissolution at the atomic scale [30]. The end of the decade witnessed a proliferation of dedicated research teams. Multiple groups in France, in Switzerland (ETH Zürich, EPFL Lausanne), and the Basque Country were among the most active in Europe, applying MD to cementitious systems. In the USA, we should mention Princeton and especially California, which became a hub with groups at UCLA, UC Irvine, and Berkeley.

In the last decade, atomistic simulation of cementitious materials has entered a phase of expansion and generalization. These methods are now employed by a wide global community in both academia and industry. One of the most remarkable developments has been the rapid growth of activity in Asia. Pioneering work by researchers like Dongshuai Hou in the early 2010s introduced molecular modeling of cement to China [31], and this effort was soon followed by a proliferation of groups across the country. Today, numerous universities and institutes in China host teams working on cement MD, including Southeast University (Nanjing), Wuhan University, Qingdao University of Technology, Tsinghua University (Beijing), Tongji University (Shanghai), Nanjing Tech, and Harbin Institute of Technology, among others, collectively producing a high volume of cement simulation research. Similar efforts have taken root in other parts of the Asia-Pacific, with active researchers in places such as Japan, Singapore, Hong Kong, and Macau, and India applying atomistic simulations to cement and concrete problems. This worldwide adoption has effectively created a new generation of cement chemists fluent in computational methods, accelerating the pace of discovery and broadening the range of questions addressed.

Atomistic simulations have extended to an expanding roster of phases relevant to cement chemistry: from clinker minerals like alite (C_3S) and belite (C_2S) [24, 32], to various hydrated phases including portlandite (CH), gypsum, AF-phases [33–36], hydrogarnets, and even the magnesium-silicate-hydrate (M–S–H) [37] and N-A-S-H [38, 39] gels found in alternative cements. Simulations also tackled degradation products and deleterious phases such as the alkali–silica reaction (ASR) products [40–42] and other phases arising from durability problems. Properties such as elastic moduli, creep behavior [43, 44], water and ion diffusion coefficients [45–48], permeability [49], thermal conductivity [33, 50], and dielectric response [51–53] have been derived from atomistic models, yielding insights into how atomic structure and dynamics underpin macroscale behavior.

Furthermore, simulations have progressively extended toward the mesoscale domains, elucidating the mesostructural organization of C–S–H and other colloidal assemblages in hydrated cements [54–60]. This line of mesoscale simulation, using atomistically-informed effective potentials has simulated the development of a percolating, stress-bearing network of C-S-H gel [61]. Goyal et al. [62] further extended this approach to understand the role of heterogeneous precipitation gradients near cement grains. More recently, significant advances have been achieved in the simulation of chemical transformations, including dissolution, precipitation, and initial steps of hydration reactions, contributing to a deeper understanding of cement chemistry from a molecular perspective [32, 63–65].

Reliable and accurate model developments of cementitious systems and organic admixtures constitute the backbone of molecular modeling in cement science. Over the last three decades, atomistic simulations have been primarily driven by the need to establish physically and chemically consistent atomic-scale descriptions of both unhydrated clinker phases and hydrated cement products, as well as of organic admixtures interacting with these phases. This sustained effort has led to major advances in the development and validation of atomistic models for both anhydrous and hydrated cement phases. In parallel, substantial progress has been achieved in modeling organic admixtures, where atomistic simulations have successfully elucidated adsorption mechanisms, structure–property relationships, and dispersing action for small-molecular-weight molecules [66], providing actionable insights that directly support admixture and product development. For high-molecular-weight polymeric systems, while conceptual frameworks and initial models exist [67, 68], fully predictive and transferable atomistic descriptions remain challenging and create an important open frontier for the field. Simulations have increasingly contributed to interpreting spectroscopic observations, parameterizing multiscale transport and thermodynamic models, and supporting the design of low-carbon binders and admixture interactions. Overall, these developments illustrate how advances in atomistic modeling have progressively evolved from feasibility demonstrations toward tools capable of informing formulation strategies, interpreting experimental observations, and supporting rational design particularly on the admixtures side while highlighting the remaining gaps that motivate continued methodological and multiscale developments.

While these advances have substantially improved our mechanistic understanding and our ability to interpret experimental observables and parameterize multiscale models, their translation into practical engineering applications remains uneven. A key objective of the present TC is therefore to tighten this gap by identifying the most promising applications and pathways through which atomistic simulations can contribute to cement technology, whether by enabling the design of new binders and admixtures, optimizing mix designs for durability, or supporting the management of aging infrastructure through predictive, physics-based tools.

Despite these advances, several open questions and disconnect with the experiments remain:

- **Force Field Accuracy and Validation**. The reliability of molecular simulations critically depends on the accuracy of the underlying force fields describing atomic interactions. Many of these force fields have not been extensively benchmarked for the complex chemistries of cement systems particularly at interfaces and in the presence of admixtures leading to uncertainties in predicted structures and properties. Furthermore, the transferability of these force fields across different interfaces remains an open challenge. Earlier studies have combined multiple force fields within a single simulation without validating interfacial properties, which can lead to non-physical results and misinterpretation of adsorption or hydration mechanisms. Standardized validation protocols are required to address these gaps.

- **Multiscale and Multiphase Complexity**. Cementitious materials are inherently multiscale and multiphase, characterized by defects, impurities, and dynamically evolving interfaces. Bridging atomistic insights to macroscopic behavior, while capturing the influence of structural heterogeneity and chemical diversity, remains a major challenge. Moreover, the relevant processes span timescales from femtoseconds (molecular vibrations) to several years (hydration, aging, creep, degradation), making direct atomistic simulation of long-term behaviour computationally prohibitive.
- **Modeling Interfaces and Additives**. A molecular-level understanding of the interactions between cement hydrates and chemical admixtures, nanomaterials, or polymers is still limited [69]. Accurately simulating adsorption, bonding, and transport processes at these complex interfaces is computationally demanding and methodologically challenging. Recent atomistic simulations involving low-molecular-weight polymers in the presence of pore solutions have provided only limited insights. Two major factors contribute to this limitation: (i) the use of simplified or prototype polymer structures that do not represent the chemical complexity of commercial admixtures, and (ii) the absence of explicit cement surfaces in most simulations, which neglects critical surface-specific interactions governing adsorption and dispersion.
- **pH-Specific Surface Models:** Most existing surface models neglect the dynamic protonation and deprotonation behavior of cement phases under highly alkaline conditions. Incorporating pH-dependent surface speciation is essential for accurately simulating early hydration processes and adsorption phenomena. This aspect is particularly critical for modeling interactions with dissolved ions in pore solutions and capturing phase-specific reactivity at partially hydrated clinker surfaces. Furthermore, surface charge variations strongly influence electrostatic interactions with charged functional groups in chemical admixtures, affecting both adsorption strength and molecular orientation at different hydration stages [70].
- **Data Scarcity and Computational Cost**. High-fidelity molecular simulations require substantial computational resources, and the scarcity of high-quality experimental data for model calibration and validation—particularly for emerging low-carbon and nanomodified systems—further constrains progress.
- **Link to experiments**. One of the biggest challenges is that the cementitious systems are incredibly complex and not very easy to simulate exact or very close representative systems with such techniques. For instance, dissolution of minerals or adsorption of ions on surfaces can be simulated; however the dissolution of cement minerals or adsorption of ions on phases are driven by surface defects which are often not easily simulated. This boils down to narrowing down the question from the experimental community to the atomistic level to essentially solve the right problem that is otherwise hard to address. This requires constant dialogue and exchanges between the experimental and computational community which is the essence of this TC.

2.3 Current status in RILEM

Molecular simulations have not yet been a primary focus of RILEM TCs, but several current TCs could benefit from, or engage in dialogue with, research on nanoscale processes that can be modeled using the techniques considered here. The TC is part of Cluster B: Transport and Deterioration Mechanisms (Convener: Liberato FERRARA), and its outcome will benefit multiple TCs in Clusters A B, C, and D. For example, it will support those focused on materials where phases with atomics structures yet to be fully understood are critical, such as Mg-rich phases (311-MBC: Magnesia-based binders in concrete and 284-CEC: Controlled expansion of concrete by adding MgO-based expansive agents taking the combined influence of composition and size of concrete elements into consideration), ASR products (300-ARM: Alkali-aggregate reaction mitigation), and alkali-activated products (294-MPA : Mechanical properties of alkali-activated concrete). It will also aid TCs investigating the transport of moisture and ions (313-MMS: Modeling and experimental validation of moisture state in bulk cementitious materials and at the steel-concrete interface, 285-TMS: Test method for concrete durability under combined role of sulphate and chloride ions), where nanoscale processes play a key role, as well as those exploring the functionalities of cement systems requiring a fundamental understanding of physical processes (302- CNC: Carbon-based nanomaterials for multifunctional cementitious matrices and 299-TES: Thermal energy storage in cementitious composites). Additionally, it can also be relevant to the TC 315-DCS : Data-driven concrete science, as data collection and curation will be increasingly relevant for the training of ML-based approaches. These links are particularly important for durability-related mechanisms such as ion transport, carbonation, sulfate attack, alkali–silica reaction products, and Mg-rich hydrate stability, where nanoscale processes control macroscopic performance and where atomistic simulations can help interpret experimental observations and define physically grounded modeling parameters.

2.4 Current status in other materials considered in RILEM

Besides cement-based materials, RILEM involves activities in other materials for which molecular simulation can also play a crucial role as a tool:

- **Asphalt**. Molecular modeling of asphalt has also progressed rapidly over the past decades, providing valuable insights into its complex chemical composition and multi-phase structure [71, 72]. Despite these advances, several challenges remain concerning model realism, chemical diversity, interfacial mechanisms, and data integration [71, 73, 74]. In particular, a molecular

understanding of adhesion, moisture-induced damage, and debonding mechanisms at asphalt–aggregate interfaces, and chemical aging is still limited [74, 75]. Addressing these challenges will require the development of more comprehensive molecular databases, systematic model validation against experimental data, and the integration of molecular simulations with experimental characterization and machine learning approaches [71].

- **Geomaterials, clays, and earth-based materials**. Geomaterials in general are also relevant for concrete, as the various phases present in aggregates have been studied using molecular modeling, particularly in applications involving radiation exposure [76–79]. Clays are especially important for cementitious systems, since calcined clays are today among the main supplementary cementitious materials, playing a pivotal role in reducing the emissions associated with ordinary Portland cement production. Molecular models of calcined clays (e.g., metakaolin [80, 81]) have been proposed. Phases such as M–S–H have been understood as being structurally related to clay minerals such as talc, lizardite, or saponite [37, 82]. As layered silicates with complex mesoscale structures that can exist as both dilute and dense colloidal suspensions, clay minerals share several similarities with C–S–H. Therefore, developments in clay science can be directly transferred to the study of C–S–H, and vice versa.
- **Wood and bio-based building materials**. Molecular modeling is transforming the understanding and design of wood and bio-based building materials by enabling detailed analyses from the atomic to the macroscopic scale. Recent studies employing atomistic and molecular dynamics models have simulated the arrangement and interactions of wood polymers—cellulose, hemicellulose, and lignin—revealing how water interacts with these components and how mechanical properties emerge at the nanoscale [8, 83, 84]. Such models are essential for understanding deformation mechanisms and interfacial behavior in wood-based composites.
- **Nanomaterials**. Atomistic simulations in nano-reinforced cementitious materials have revealed fundamental interactions between nanoscale additives and hydrates such as C–S–H [85]. Most studies focus on a few nanomaterials, particularly graphene oxide, carbon nanotubes, and nano-silica, whose chemical, structural, and long-term effects remain partially understood [86–88]. There is also a growing need for molecular-level studies of emerging nanomaterials with new functionalities [89]. A deeper atomistic understanding of how nanomaterials influence hydration, crack resistance, and transport is essential to their practical use. Realistic, experimentally validated molecular models linked to environmental conditions can further accelerate evaluating these effects and integrating them into multiscale models.

2.5 Current status in RILEM

The initiative to propose the Technical Committee (TC) originated from discussions held during the 1st International Conference on Atomistic Simulations of Cementitious Materials (ICASCM). The TC proposal was subsequently prepared over the following months by the chair and co-chair, with the active participation of H. Manzano, G. Geng, and E. Masoero. The draft proposal was broadly circulated to researchers worldwide who are engaged in molecular modeling and simulations of cementitious and related materials at the end of 2024 and beginning of 2025.

At the time of submission to RILEM, 37 researchers had formally expressed their willingness to participate in the TC upon its approval. As of today, the TC comprises more than 60 listed participants. This participation reflects a diverse international community, including participants from Europe, Americas, and Asia, representing both academic institutions, research organizations and the industry.

Since its establishment, the TC has held several online coordination meetings and one hybrid meeting during the RILEM Spring Convention 2025, where members discussed the TC's scope, initial objectives, and working structure. Key points of discussion have included the definition of benchmark systems for simulation, strategies for data sharing and model validation, and the integration of multiscale and machine learning approaches to link molecular insights with engineering applications.

3 Atomistic and molecular simulation and the link to experiments

A central goal of the TC ASM is to establish strong, quantitative links between molecular simulations and experimental observations. While molecular modeling provides unique insights into the structure, dynamics, and chemistry of cementitious systems at the atomic scale, its predictive power relies on systematic validation and calibration against experimental data. Bridging these scales is therefore essential to ensure that simulation outcomes are physically meaningful and relevant for experts on the experimental side to make better materials characterization, performance prediction, and design.

Atomistic simulations—whether based on classical force fields, reactive molecular dynamics, first-principles methods, or more recently, machine learning force fields— allow the direct calculation of structural and thermodynamic quantities of cementitious materials such as bond lengths, coordination numbers, interlayer spacings, diffusion coefficients, and elastic moduli. These quantities can be quantitatively compared with data obtained from various experimental techniques including nuclear magnetic resonance (NMR), X-ray and neutron diffraction, pair distribution function (PDF) analysis, vibrational spectroscopy (IR and Raman), nanoindentation, Brillouin spectroscopy, and calorimetry. The simulation of spectroscopy data (NMR, Raman, FTIR, XAS, and XPS) using DFT combined with ab-initio MD have been recently shown to capture molecular-scale features related to local environments and dynamics (e.g., T_1, T_2 in NMR).

Reverse Monte Carlo, possibly combined with classical or ML force fields, provides an additional route to refine structures against experimental data. These approaches not only reproduce experimental spectra but also yield parameters—such as atomic charges, dipole moments, and coordination environments—essential for validation and predictive modeling across scales. Such cross-validation supports the refinement of force fields, the verification of atomistic models, and eventually the identification of nanoscale mechanisms controlling macroscopic behavior establishing relationships between ionic coordination environments and diffusion coefficients, interlayer spacings and cohesion, free energies of formation with phase development, or mechanical properties and durability.

Additionally, atomistic simulations provide a powerful means to explore mineral-aqueous solution interfaces and investigate the complexation and adsorption of hydrated ions, and water behaviour. These methods enable the determination of key properties including, e.g., adsorption sites, adsorption free energies, acidity constants, ion exchange constants, electric double layer (zeta-potential), and interfacial hydrogen bonds behaviour. Combining modeling with experimental techniques like EXAFS (extended X-ray absorption fine structure), TRLFS (time-resolved laser-induced fluorescence spectroscopy), INS (inelastic neutron scattering), and laser Doppler electrophoresis technique allows us to refine models of mineral surfaces and edges where ion adsorption and complexation occur [90–94].

Beyond direct comparison, atomistic simulations complement experimental studies by exploring conditions that are challenging to reproduce and probe experimentally, such as extreme temperatures, pressures, radiation environments, or confined hydration states. They also provide atomistic interpretations for phenomena observed at larger scales—such as creep, shrinkage, and chemical degradation—by linking them to local structural rearrangements and reaction pathways. Machine learning and data-driven modeling further strengthen this connection by enabling efficient parameter optimization, uncertainty quantification, and multi-scale data fusion between simulations and experiments.

4 Organization of the Technical Committee

4.1 Current status in RILEM

The TC is organized in five working groups (WG) and related tasks (Figure 2):

WG1: Atomic structures and force fields: The focus is collecting and reviewing atomic structures of cementitious and related phases, including compositionally variable systems (e.g., C-S-H, AF-phases) and relevant surfaces, as well as organic materials such as admixtures and polymers. It will address data management challenges by structuring databases for structures, force fields, and simulation results. WG1 also evaluates experimental data for structural validation, identifies understudied phases (e.g., Mg- and Fe-rich phases, alkali-containing C-(A)-S-H, ASR products), and reviews existing and emerging force fields, including machine-learning-based potentials. The link to experiments can be established through simulations of spectroscopic data (NMR, Raman, FTIR, etc), PDF (including structure refinement with Reverse MC).

WG2: Chemical transformations: This WG focuses on the molecular mechanisms underlying chemical processes in cementitious systems, including dissolution, precipitation, growth, and hydration. It reviews enhanced sampling strategies (e.g., metadynamics) and free energy methods. WG2 also examines high-temperature/pressure processes, carbonation, durability issues, and strategies for upscaling molecular insights to mesoscale and continuum models.

WG3: Interfacial processes, confined fluids, sorption-induced deformations focusing on modeling (physio-)adsorption and ion-exchange processes in hydration products and cementitious phases, including effects of electrical double layers and zeta potential. It reviews hydration states of phases containing molecular water (e.g., C-S-H, AFm, AFt, ASR), their stability under changes in relative humidity and temperature, and phenomena such as hysteresis, cavitation in nanopores, surface tension, disjoining pressures, and sorption-induced swelling or shrinkage. WG3 also emphasizes comparison with experimental data, recommends suitable simulation methodologies, and explores upscaling strategies.

WG4: Mechanical, transport, thermal, and electromagnetic properties focusing on modeling the physical properties of cementitious systems. It reviews calculations of mechanical properties, thermal properties, ionic conductivity, dielectric permittivity, diffusion, and permeability. WG4 also emphasizes the comparison of modeling results with experimental data, the compilation of essential property tables (including gaps relevant for thermodynamic and multiscale modeling), the recommendation of simulation methodologies, and strategies for upscaling to macroscopic models.

WG5: Mesoscale modeling focusing on bridging molecular-level insights to mesoscale behavior in cementitious systems. It reviews effective interactions derived from molecular simulations, coarse-grained modeling approaches, and the assessment of structural, chemical, and physico-mechanical properties at the mesoscale. WG5 also emphasizes the comparison of simulation results with experimental data (including for instance SAXS) and explores strategies for upscaling molecular and mesoscale findings to macroscopic models. In this way, WG5 is intended not only to integrate results from the other working groups but also to help formulate transferable challenges and modeling targets that can guide future developments in atomistic and multiscale simulations within the RILEM community.

4.2 List of the planned TC activities

The deliverables of this TC mainly include Topical Collection in *Materials and Structures* related to WG activities, open-access database of structures and tools, tutorial documents for recommended modeling methodologies.

- The TC has been launched in 2025, with a projected five-year timeline.

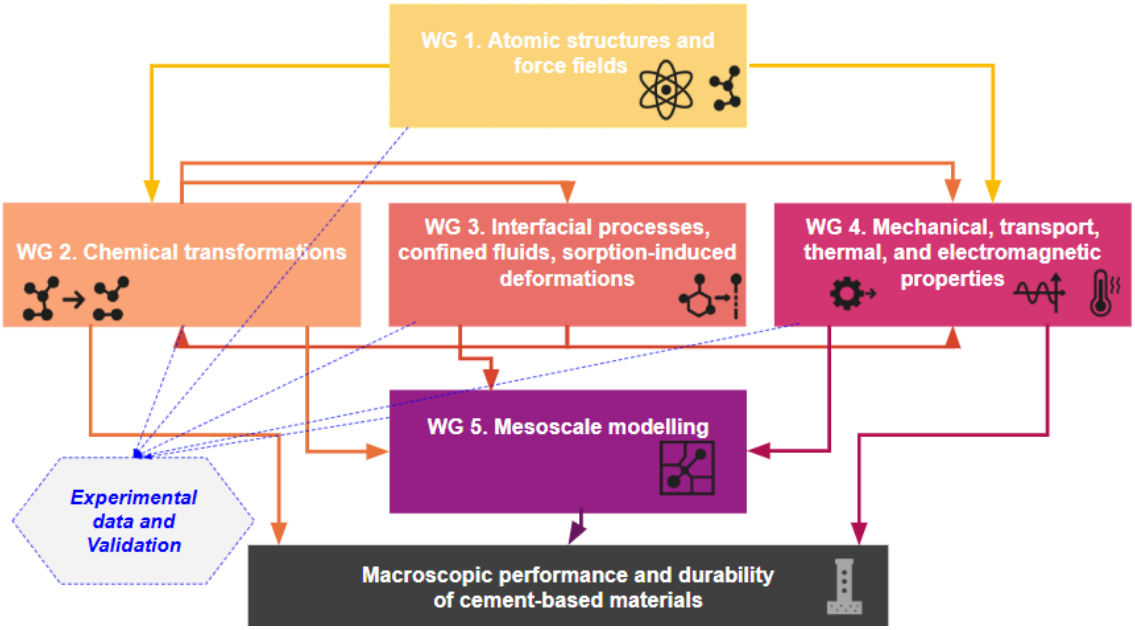

Figure 2. Schematic representation of the WGs in the TC ASM.

- Key objectives will be achieved by engaging a diverse membership of academic and industrial research entities across Europe, America, Asia, and Oceania.
- The committee will focus on bibliographical research, database construction, and benchmarking within specific WGs, producing review articles and an open-access database to foster a cohesive modeling community and improve simulation consistency.
- Educational activities and conferences are also planned. Industry participation will be encouraged through links with cement and additive producers, ensuring that TC activities support both fundamental research and applied insights relevant to industrial stakeholders.

The TC will operate through online and hybrid meetings twice per year with the entire TC, synchronized preferentially with RILEM events and the (biennial) International Conference on Atomistic Simulation of Cementitious Materials (ICASCM).

4.3 Expected achievements

The deliverables of the proposed TC will be the following:

- Open-access database of phase structures relevant to cementitious systems, along with their properties, which can support thermodynamic and multiscale modeling.
- Open-access database of relevant force fields with benchmarked outcomes.
- State-of-the-art review (STAR) and WG-related work published as a topical collection in Materials and Structures.
- Recommendations for established simulation protocols aimed at harmonizing adopted methodologies.
- Educational materials (tutorials, courses) covering established simulation protocols and emerging techniques.
- Two letters to RILEM Technical Letters: one at the beginning of the TC (outlining the state-of-the-art, research needs, and rationale for the TC's creation) and one at the end of the TC (providing an overview of the work accomplished, remaining open questions, and future directions).
- Organization of the biennial International Conference of Atomistic Simulation of Cementitious Materials (ICASCM) combined with advanced schools.

4.4 Setting the stage for modeling research on nanoscale processes in RILEM

Understanding and predicting the behavior of cementitious materials requires a comprehensive view that connects atomic-scale mechanisms to macroscopic properties. Nanoscale processes—such as transport properties, interfacial adsorption, dissolution–precipitation reactions, and nanoscale deformation—govern key aspects of hydration, durability, and long-term performance in concrete. However, these phenomena are often difficult to isolate experimentally and are influenced by complex chemical and structural heterogeneity. Atomistic and molecular modeling offers a unique opportunity to elucidate these mechanisms, quantify their effects, and provide input parameters for larger-scale models that underpin engineering predictions.

The TC ASM provides a coordinated framework to advance nanoscale modeling as an integral part of materials research within RILEM. By standardizing methodologies, developing benchmark datasets, and fostering strong links between modeling and experiments, the TC lays the foundation for a

more coherent and reproducible approach to studying nanoscale processes. These efforts will support existing RILEM TCs working on transport phenomena, durability mechanisms, alternative binders, and data-driven materials design, while also creating a platform for future cross-disciplinary collaborations. By setting this foundation, RILEM can strengthen the integration of modeling within its scientific and technical activities. The TC will contribute to the digital transformation of cement and concrete research through open databases, standardized workflows, and education initiatives that promote accessibility and reproducibility. This effort will also facilitate the creation of shared reference models and benchmark problems, ensuring that nanoscale simulations can reliably inform experimental design and multiscale analysis across the RILEM community. These benchmarks are intended not only to harmonize modeling practices but also to define validation targets and predictive challenges that can stimulate advances in atomistic and multiscale simulations across the cement and concrete community. Ultimately, this initiative positions RILEM at the forefront of the global movement toward data-driven, multiscale, and mechanism-based materials science, enhancing the collective capacity to develop durable, sustainable, and low-carbon cementitious materials through a deep understanding of their nanoscale foundations.

5 Conclusions

The TC will establish a reference database of structures and force fields, along with standardized protocols for molecular modeling, providing a robust framework that improves the accuracy and consistency of simulations across research groups. By advancing the scientific understanding of nanoscale processes in cement systems, it will contribute to the development of more durable and sustainable materials. The TC's outputs, including a comprehensive database and validated methodologies, will benefit both academia and industry by enabling reliable virtual testing, reducing experimental costs, better understanding of cementitious materials, and accelerating material innovation. In the long term, the TC's findings are expected to deliver economic benefits by enabling research on low-carbon cement formulations—spanning new chemistries, novel phases, and innovative admixture solutions—while also amplifying scientific impact through stronger collaboration and greater research efficiency.

Authorship statement (CRediT)

Tulio Honorio: Conceptualization; Writing – Original Draft; Writing – Review & Editing; Visualization. **Hegoi Manzano:** Conceptualization; Writing – Original Draft; Writing – Review & Editing. **Guoqing Geng:** Conceptualization. **Enrico Masoero:** Conceptualization; Writing – Original Draft; Writing – Review & Editing. **Elvys Dias Reis:** Writing – Original Draft; Writing – Review & Editing. **N. M. Anoop Krishnan:** Writing – Original Draft; Writing – Review & Editing. **Aleena Alex:** Writing – Original Draft; Writing – Review & Editing. **Priyanka Beta:** Writing – Original Draft; Writing – Review & Editing. **Eduardo Duque-Redondo:** Writing – Original Draft; Writing – Review & Editing; Visualization. **Evgeny Tararushkin:** Writing – Original Draft; Writing – Review & Editing. **Thibault Charpentier:** Writing – Original Draft; Writing – Review & Editing. **Ratan K. Mishra:** Writing – Original Draft; Writing – Review & Editing. **Aslam Kunhi Mohamed:** Conceptualization; Writing – Original Draft; Writing – Review & Editing.

References

[1] S. A. Hollingsworth, R. O. Dror, Molecular dynamics simulation for all, Neuron (2018) 99: 1129-1143. https://doi.org/10.1016/j.neuron.2018.08.011

[2] F. Noé, A. Tkatchenko, K.-R. Müller, C. Clementi, Machine learning for molecular simulation, Annu Rev Phys Chem (2020) 71: 361-390. https://doi.org/10.1146/annurev-physchem-042018-052331

[3] J. Potoff, I. Economou, Molecular simulation, Fluid Phase Equilib (2019) 498: 160. https://doi.org/10.1016/j.fluid.2019.05.027

[4] J. Choi, A review of the mechanical design of materials based on molecular dynamics simulations, Multiscale Sci Eng (2023) 5: 86-103. https://doi.org/10.1007/s42493-024-00105-x

[5] A. Glielmo, B. E. Husic, A. Rodriguez, et al., Unsupervised learning methods for molecular simulation data, Chem Rev (2021) 121: 9722-9758. https://doi.org/10.1021/acs.chemrev.0c01195

[6] S. Mehdi, Z. Smith, L. Herron, et al., Enhanced sampling with machine learning, Annu Rev Phys Chem (2024) 75: 347-370. https://doi.org/10.1146/annurev-physchem-083122-125941

[7] R. Jacobs, D. Morgan, S. Attarian, et al., A practical guide to machine learning interatomic potentials - Status and future, Curr Opin Solid State Mater Sci (2025) 35: 101214. https://doi.org/10.1016/j.cossms.2025.101214

[8] M. Abraham, R. Apostolov, J. Barnoud, et al., Sharing data from molecular simulations, J Chem Inf Model (2019) 59: 4093-4099. https://doi.org/10.1021/acs.jcim.9b00665

[9] J. Zhang, D. Chen, Y. Xia, et al., Artificial intelligence enhanced molecular simulations, J Chem Theory Comput (2023) 19: 4338-4350. https://doi.org/10.1021/acs.jctc.3c00214

[10] S. Kashihara, S. Yamanaka, T. Inoue, et al., Quantum chemical determination of the Al-substituted site in tobermorite, J Am Ceram Soc (1994) 77: 3023-3026. https://doi.org/10.1111/j.1151-2916.1994.tb04540.x

[11] P. Faucon, J. M. Delaye, J. Virlet, et al., Study of the structural properties of the CSH(I) by molecular dynamics simulation, Cem Concr Res (1997) 27: 1581-1590. https://doi.org/10.1016/S0008-8846(97)00161-0

[12] P. Faucon, J. M. Delaye, J. Virlet, Molecular dynamics simulation of the structure of calcium silicate hydrates: I. Ca4+xSi6O14+2x(OH)4−2x(H2O)2(0 ⩽ x ⩽ 1), J Solid State Chem (1996) 127: 92-97. https://doi.org/10.1006/jssc.1996.0361

[13] A. G. Kalinichev, J. Wang, R. J. Kirkpatrick, Molecular dynamics modeling of the structure, dynamics and energetics of mineral-water interfaces: Application to cement materials, Cem Concr Res (2007) 37: 337-347. https://doi.org/10.1016/j.cemconres.2006.07.004

[14] R. J. Kirkpatrick, Experimental and molecular dynamics modeling studies of interlayer swelling: Water incorporation in kanemite and ASR gel, Mater Struct (2005) 38: 449-458. https://doi.org/10.1617/14344

[15] R. T. Cygan, J.-J. Liang, A. G. Kalinichev, Molecular models of hydroxide, oxyhydroxide, and clay phases and the development of a general force field, J Phys Chem B (2004) 108: 1255-1266. https://doi.org/10.1021/jp0363287

[16] C. E. White, J. L. Provis, T. Proffen, et al., Combining density functional theory (DFT) and pair distribution function (PDF) analysis to solve the structure of metastable materials: The case of metakaolin, Phys Chem Chem Phys (2010) 12: 3239. https://doi.org/10.1039/b922993k

[17] H. Manzano, J. S. Dolado, A. Ayuela, Elastic properties of the main species present in Portland cement pastes, Acta Mater (2009) 57: 1666-1674. https://doi.org/10.1016/j.actamat.2008.12.007

[18] H. Manzano, A. Ayuela, J. S. Dolado, On the formation of cementitious C-S-H nanoparticles, J Comput Aided Mater Des (2007) 14: 45-51. https://doi.org/10.1007/s10820-006-9030-0

[19] R. J.-M. Pellenq, A. Kushima, R. Shahsavari, et al., A realistic molecular model of cement hydrates, Proc Natl Acad Sci USA (2009) 106: 16102-16107. https://doi.org/10.1073/pnas.0902180106

[20] Z. Casar, T. B. Montandon, M. Cordova, et al., CementFF4: Formal atomic charge polarizable force field for cementitious systems - Bulk and surface, Cem Concr Res (2025) 187: 107708. https://doi.org/10.1016/j.cemconres.2024.107708

[21] S. Galmarini, A. Kunhi Mohamed, P. Bowen, Atomistic simulations of silicate species interaction with portlandite surfaces, J Phys Chem C (2016). https://doi.org/10.1021/acs.jpcc.6b07044

[22] O. Heinz, H. Heinz, Cement interfaces: Current understanding, challenges, and opportunities, Langmuir (2021) 37: 6347-6356. https://doi.org/10.1021/acs.langmuir.1c00617

[23] K. Kanhaiya, M. Nathanson, P. J. in 't Veld, et al., Accurate force fields for atomistic simulations of oxides, hydroxides, and organic hybrid materials up to the micrometer scale, J Chem Theory Comput (2023) 19: 8293-8322. https://doi.org/10.1021/acs.jctc.3c00750

[24] R. K. Mishra, A. K. Mohamed, D. Geissbühler, et al., cemff: A force field database for cementitious materials including validations, applications and opportunities, Cem Concr Res (2017) 102: 68-89. https://doi.org/10.1016/j.cemconres.2017.09.003

[25] R. K. Mishra, R. J. Flatt, H. Heinz, Force field for tricalcium silicate and insight into nanoscale properties: Cleavage, initial hydration, and adsorption of organic molecules, J Phys Chem C (2013) 117: 10417-10432. https://doi.org/10.1021/jp312815g

[26] R. Shahsavari, R. J.-M. Pellenq, F.-J. Ulm, Empirical force fields for complex hydrated calcio-silicate layered materials, Phys Chem Chem Phys (2010) 13: 1002-1011. https://doi.org/10.1039/C0CP00516A

[27] M. Valavi, Z. Casar, A. Kunhi Mohamed, et al., Molecular dynamic simulations of cementitious systems using a newly developed force field suite ERICA FF, Cem Concr Res (2022) 154: 106712. https://doi.org/10.1016/j.cemconres.2022.106712

[28] H. Manzano, S. Moeini, F. Marinelli, et al., Confined water dissociation in microporous defective silicates: Mechanism, dipole distribution, and impact on substrate properties, J Am Chem Soc (2012) 134: 2208-2215. https://doi.org/10.1021/ja209152n

[29] H. Manzano, R. J.-M. Pellenq, F.-J. Ulm, et al., Hydration of calcium oxide surface predicted by reactive force field molecular dynamics, Langmuir (2012) 28: 4187-4197. https://doi.org/10.1021/la204338m

[30] D. Axthammer, S. Darouich, M. Collin, et al., Mechanistic insights into the anion-driven suppression of the initial cement reaction of tricalcium aluminate, J Am Ceram Soc (2026) 109: e70480. https://doi.org/10.1111/jace.70480

[31] D. Hou, H. Ma, Y. Zhu, Z. Li, Calcium silicate hydrate from dry to saturated state: Structure, dynamics and mechanical properties, Acta Mater (2014) 67: 81-94. https://doi.org/10.1016/j.actamat.2013.12.016

[32] X. Xu, C. Qi, X. M. Aretxabaleta, et al., The initial stages of cement hydration at the molecular level, Nat Commun (2024) 15: 2731. https://doi.org/10.1038/s41467-024-46962-w

[33] T. Honorio, Thermal conductivity, heat capacity and thermal expansion of ettringite and metaettringite: Effects of the relative humidity and temperature, Cem Concr Res (2022) 159: 106865. https://doi.org/10.1016/j.cemconres.2022.106865

[34] T. Honorio, P. Guerra, A. Bourdot, Molecular simulation of the structure and elastic properties of ettringite and monosulfoaluminate, Cem Concr Res (2020) 135: 106126. https://doi.org/10.1016/j.cemconres.2020.106126

[35] E. V. Tararushkin, G. S. Smirnov, N. D. Kondratyuk, A. G. Kalinichev, Structure and properties of thaumasite and its aqueous interfaces revealed by molecular dynamics simulations, Cem Concr Res (2025) 197: 107944. https://doi.org/10.1016/j.cemconres.2025.107944

[36] E. V. Tararushkin, V. V. Pisarev, A. G. Kalinichev, Atomistic simulations of ettringite and its aqueous interfaces: Structure and properties revisited with the modified ClayFF force field, Cem Concr Res (2022) 156: 106759. https://doi.org/10.1016/j.cemconres.2022.106759

[37] A. Pedone, F. Palazzetti, V. Barone, Models of aged magnesium-silicate-hydrate cements based on the lizardite and talc crystals: A periodic DFT-GIPAW investigation, J Phys Chem C (2017) 121: 7319-7330. https://doi.org/10.1021/acs.jpcc.7b00708

[38] F. Lolli, H. Manzano, J. L. Provis, et al., Atomistic simulations of geopolymer models: The impact of disorder on structure and mechanics, ACS Appl Mater Interfaces (2018) 10: 22809-22820. https://doi.org/10.1021/acsami.8b03873

[39] E. Shamo, T. Charpentier, A. Rousselet, et al., Atomistic simulations of water-driven structural and mechanical changes in N-A-S-H gels, J Am Ceram Soc (2025) 108: e70124. https://doi.org/10.1111/jace.70124

[40] R. Dupuis, R. J.-M. Pellenq, Alkali silica reaction: A view from the nanoscale, Cem Concr Res (2022) 152: 106652. https://doi.org/10.1016/j.cemconres.2021.106652

[41] T. Honorio, O. M. C. Tamouya, Z. Shi, Specific ion effects control the thermoelastic behavior of nanolayered materials: The case of crystalline alkali-silica reaction products, Phys Chem Chem Phys (2020) 22: 27800-27810. https://doi.org/10.1039/D0CP04955G

[42] T. Honorio, O. M. Chemgne Tamouya, Z. Shi, A. Bourdot, Intermolecular interactions of nanocrystalline alkali-silica reaction products under sorption, Cem Concr Res (2020) 136: 106155. https://doi.org/10.1016/j.cemconres.2020.106155

[43] E. Duque-Redondo, E. Masoero, H. Manzano, Nanoscale shear cohesion between cement hydrates: The role of water diffusivity under structural and electrostatic confinement, Cem Concr Res (2022) 154: 106716. https://doi.org/10.1016/j.cemconres.2022.106716

[44] M. F. Kai, L. W. Zhang, K. M. Liew, New insights into creep characteristics of calcium silicate hydrates at molecular level, Cem Concr Res (2021) 142: 106366. https://doi.org/10.1016/j.cemconres.2021.106366

[45] W. Chen, K. Gong, Insights into ionic diffusion in C-S-H gel pore from molecular dynamics simulations: Spatial distributions, energy barriers, and structural descriptor, J Phys Chem B (2025) 129: 10550-10567. https://doi.org/10.1021/acs.jpcb.5c04534

[46] E. Duque-Redondo, K. Yamada, H. Manzano, Cs retention and diffusion in C-S-H at different Ca/Si ratio, Cem Concr Res (2021) 140: 106294. https://doi.org/10.1016/j.cemconres.2020.106294

[47] M. J. A. Qomi, M. Bauchy, F.-J. Ulm, R. J.-M. Pellenq, Anomalous composition-dependent dynamics of nanoconfined water in the interlayer of disordered calcium-silicates, J Chem Phys (2014) 140: 054515. https://doi.org/10.1063/1.4864118

[48] M. Youssef, R. J.-M. Pellenq, B. Yildiz, Glassy nature of water in an ultraconfining disordered material: The case of calcium–silicate–hydrate, J Am Chem Soc (2011) 133: 2499-2510. https://doi.org/10.1021/ja107003a

[49] T. Honorio, Permeability of C-S-H, Cem Concr Res (2024) 176: 107408. https://doi.org/10.1016/j.cemconres.2023.107408

[50] M. J. A. Qomi, F.-J. Ulm, R. J.-M. Pellenq, Physical origins of thermal properties of cement paste, Phys Rev Appl (2015) 3: 064010. https://doi.org/10.1103/PhysRevApplied.3.064010

[51] S. Ait Hamadouche, T. Honorio, T. Bore, et al., Dielectric permittivity of C-S-H, Cem Concr Res (2023) 169: 107178. https://doi.org/10.1016/j.cemconres.2023.107178

[52] T. Honorio, W. B. Bonfim, O. Cascudo, Impedance and electrical conductivity of C-S-H, Cem Concr Res (2026) 201: 108123. https://doi.org/10.1016/j.cemconres.2025.108123

[53] T. Honorio, T. Bore, F. Benboudjema, et al., Dielectric properties of the pore solution in cement-based materials, J Mol Liq (2020) 302: 112548. https://doi.org/10.1016/j.molliq.2020.112548

[54] S. Ait Hamadouche, T. Honorio, Nanomechanics of ASR gels from coarse-grained simulations, J Eng Mech (2023). https://doi.org/10.1061/JENMDT.EMENG-6926

[55] K. Ioannidou, C. Labbez, E. Masoero, A review of coarse grained and mesoscale simulations of C-S-H, Cem Concr Res (2022) 159: 106857. https://doi.org/10.1016/j.cemconres.2022.106857

[56] K. Ioannidou, K. J. Krakowiak, M. Bauchy, et al., Mesoscale texture of cement hydrates, Proc Natl Acad Sci USA (2016) 113: 2020487. https://doi.org/10.1073/pnas.1520487113

[57] K. Ioannidou, R. J.-M. Pellenq, E. Del Gado, Controlling local packing and growth in calcium-silicate-hydrate gels, Soft Matter (2014) 10: 1121-1133. https://doi.org/10.1039/C3SM52232F

[58] E. Masoero, E. Del Gado, R. J.-M. Pellenq, et al., Nano-scale mechanics of colloidal C-S-H gels, Soft Matter (2013) 10: 491-499. https://doi.org/10.1039/C3SM51815A

[59] E. Masoero, E. Del Gado, R. J.-M. Pellenq, et al., Nanostructure and nanomechanics of cement: Polydisperse colloidal packing, Phys Rev Lett (2012) 109. https://doi.org/10.1103/PhysRevLett.109.155503

[60] S. Masoumi, D. Ebrahimi, H. Valipour, M. J. A. Qomi, Nanolayered attributes of calcium-silicate-hydrate gels, J Am Ceram Soc (2020) 103: 541-557. https://doi.org/10.1111/jace.16750

[61] K. Ioannidou, M. Kanduč, L. Li, et al., The crucial effect of early-stage gelation on the mechanical properties of cement hydrates, Nat Commun (2016) 7: 12106. https://doi.org/10.1038/ncomms12106

[62] Goyal, A., Palaia, I., Ioannidou, K., Ulm, F.-J., van Damme, H., Pellenq, R.J.-M., Trizac, E., Del Gado, E., The physics of cement cohesion, Sci Adv 7 (2021) https://doi.org/10.1126/sciadv.abg5882

[63] B. Chen, M. Wang, H. Manzano, et al., Molecular elucidation of cement hydration inhibition by silane coupling agents, Nat Commun (2025) 16: 1597. https://doi.org/10.1038/s41467-025-56877-9

[64] Y. Li, H. Pan, Q. Liu, et al., Ab initio mechanism revealing for tricalcium silicate dissolution, Nat Commun (2022) 13: 1253. https://doi.org/10.1038/s41467-022-28932-2

[65] Y. Tao, P. Martin, H. Manzano, M. J. A. Qomi, Mesoscopic mechanisms of dicalcium silicate dissolution, Cem Concr Res (2025) 189: 107660. https://doi.org/10.1016/j.cemconres.2024.107660

[66] R. K. Mishra, M. Weibel, T. Müller, et al., Energy-effective grinding of inorganic solids using organic additives, CHIMIA (2017) 71: 451-451. https://doi.org/10.2533/chimia.2017.451

[67] A. Kunhi Mohamed, S. A. Weckwerth, R. K. Mishra, et al., Molecular modeling of chemical admixtures; opportunities and challenges, Cem Concr Res (2022) 156: 106783. https://doi.org/10.1016/j.cemconres.2022.106783

[68] R. K. Mishra, S. Darouich, P. J. in 't Veld, et al., Understanding hydration reactions, mechanical properties, thermal expansion, and organic interfacial interactions of calcium sulfate hydrates from the atomic scale, Cem Concr Res (2025) 189: 107740. https://doi.org/10.1016/j.cemconres.2024.107740

[69] A. Kunhi Mohamed, S. A. Weckwerth, R. K. Mishra, et al., Molecular modeling of chemical admixtures; opportunities and challenges, Cem Concr Res (2022) 156: 106783. https://doi.org/10.1016/j.cemconres.2022.106783

[70] E. Pustovgar, R. K. Mishra, M. Palacios, et al., Influence of aluminates on the hydration kinetics of tricalcium silicate, Cem Concr Res (2017) 100: 245-262. https://doi.org/10.1016/j.cemconres.2017.06.006

[71] Z. Wang, Y. Zhou, J. Zhang, et al., Molecular components and molecular modeling for asphalt: A review, Adv Phys Res (2025) 4: 2400128. https://doi.org/10.1002/apxr.202400128

[72] H. Yao, J. Liu, M. Xu, et al., Discussion on molecular dynamics (MD) simulations of the asphalt materials, Adv Colloid Interface Sci (2022) 299: 102565. https://doi.org/10.1016/j.cis.2021.102565

[73] D. D. Li, M. L. Greenfield, Chemical compositions of improved model asphalt systems for molecular simulations, Fuel (2014) 115: 347-356. https://doi.org/10.1016/j.fuel.2013.07.012

[74] J. Xu, B. Ma, W. Mao, et al., Review of interfacial adhesion between asphalt and aggregate based on molecular dynamics, Constr Build Mater (2023) 362: 129642. https://doi.org/10.1016/j.conbuildmat.2022.129642

[75] M. Xu, J. Yi, D. Feng, Y. Huang, Diffusion characteristics of asphalt rejuvenators based on molecular dynamics simulation, Int J Pavement Eng (2019) 20: 615-627. https://doi.org/10.1080/10298436.2017.1321412

[76] T. Fujimura, Y. Hakozaki, S. Sakuragi, et al., The mechanical properties of irradiated concrete aggregates: Insights from molecular dynamics simulations, J Am Ceram Soc (2025) 108: e20318. https://doi.org/10.1111/jace.20318

[77] N. M. A. Krishnan, Y. Le Pape, G. Sant, M. Bauchy, Disorder-induced expansion of silicate minerals arises from the breakage of weak topological constraints, J Non Cryst Solids (2021) 564: 120846. https://doi.org/10.1016/j.jnoncrysol.2021.120846

[78] N. M. A. Krishnan, Y. Le Pape, G. Sant, M. Bauchy, Effect of irradiation on silicate aggregates' density and stiffness, J Nucl Mater (2018) 512: 126-136. https://doi.org/10.1016/j.jnucmat.2018.10.009

[79] Y. Le Pape, J. Sanahuja, M. H. F. Alsaid, Irradiation-induced damage in concrete-forming aggregates: Revisiting literature data through micromechanics, Mater Struct (2020) 53: 62. https://doi.org/10.1617/s11527-020-01489-6

[80] S. Sperinck, P. Raiteri, N. Marks, K. Wright, Dehydroxylation of kaolinite to metakaolin-a molecular dynamics study, J Mater Chem (2011) 21: 2118-2125. https://doi.org/10.1039/C0JM01748E

[81] C. E. White, J. L. Provis, T. Proffen, et al., Combining density functional theory (DFT) and pair distribution function (PDF) analysis to solve the structure of metastable materials: The case of metakaolin, Phys Chem Chem Phys (2010) 12: 3239. https://doi.org/10.1039/b922993k

[82] H. Sreenivasan, E. Bernard, H. S. Santos, et al., A critical review of magnesium silicate hydrate (M-S-H) phases for binder applications, Cem Concr Res (2024) 178: 107462. https://doi.org/10.1016/j.cemconres.2024.107462

[83] D. Adler, M. Buehler, Mesoscale mechanics of wood cell walls under axial strain, Soft Matter (2013) 9: 7138-7144. https://doi.org/10.1039/c3sm50183c

[84] K. Jin, Z. Qin, M. J. Buehler, Molecular deformation mechanisms of the wood cell wall material, J Mech Behav Biomed Mater (2015) 42: 198-206. https://doi.org/10.1016/j.jmbbm.2014.11.010

[85] R. G. Merodio-Perea, M.-J. Terrón-López, I. Lado-Touriño, Molecular dynamics simulation of CNT reinforced cement: A step toward sustainable construction, Sustainability (2025) 17: 3185. https://doi.org/10.3390/su17073185

[86] W. A. Al-Awsh, M. A. Al-Osta, A. A. Bahraq, et al., Development of a universal atomistic cement model incorporating nanomaterials: From laboratory investigation to molecular simulation, J Build Eng (2024) 95: 109975. https://doi.org/10.1016/j.jobe.2024.109975

[87] J. Song, H. Zheng, S. Chai, et al., Influence of functionalized carbon nanotubes on the mechanical properties of cementitious materials: Insights from molecular to coarse-grained modeling, Case Stud Constr Mater (2025) 22: e04551. https://doi.org/10.1016/j.cscm.2025.e04551

[88] Y. Yang, J. Cao, Interfacial heat transfer behavior of graphene-based filler and calcium-silicate-hydrate in cement composites, Int J Heat Mass Transf (2021) 176: 121165. https://doi.org/10.1016/j.ijheatmasstransfer.2021.121165

[89] Y. Chi, B. Huang, M. Saafi, et al., Carrot-based covalently bonded saccharides as a new 2D material for healing defective calcium-silicate-hydrate in cement: Integrating atomistic computational simulation with experimental studies, Compos Part B Eng (2020) 199: 108235. https://doi.org/10.1016/j.compositesb.2020.108235

[90] F. Colmenero, A. M. Fernández, O. Almendros-Ginestà, T. Missana, Density functional theory study of the crystal structure and infrared spectrum of a synthetized ettringite mineral, Minerals (2024) 14: 824. https://doi.org/10.3390/min14080824

[91] A. Javadi, T. Jamil, E. Abouzari-Lotf, et al., Working mechanisms and design principles of comb-like polycarboxylate ether superplasticizers in cement hydration: Quantitative insights for a series of well-defined copolymers, ACS Sustain Chem Eng (2021) 9: 8354-8371. https://doi.org/10.1021/acssuschemeng.0c08566

[92] C. A. Lee, A. van Veelen, K. Morris, et al., Uranium (VI) adsorbate structures on portlandite [Ca(OH)2] type surfaces determined by computational modeling and X-ray absorption spectroscopy, Minerals (2021) 11: 1241. https://doi.org/10.3390/min11111241

[93] Z. Zhakiyeva, V. Magnin, A. Poulain, et al., Water dynamics in calcium silicate hydrates probed by inelastic neutron scattering and molecular dynamics simulations, Cem Concr Res (2024) 184: 107616. https://doi.org/10.1016/j.cemconres.2024.107616

[94] C. Zhang, X. Liu, R. M. Tinnacher, C. Tournassat, Mechanistic understanding of uranyl ion complexation on montmorillonite edges: A combined first-principles molecular dynamics-surface complexation modeling approach, Environ Sci Technol (2018) 52: 8501-8509. https://doi.org/10.1021/acs.est.8b02504