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Editorial: Molecular Simulation on Cementitious Materials: From Computational Chemistry Method to Application

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Editorial on the Research Topic

Molecular Simulation on Cementitious Materials: From Computational Chemistry Method to Application

The use of molecular simulation has made great progress over recent years in the field of cement chemistry. The progress includes, but is not limited to, deriving a series of accurate force fields, decoding the realistic structure of calcium-silicate-hydrate (C-S-H), unraveling the interfacial reactivity and chemistry of cement minerals, and studying interactions between cement hydrates and organic/inorganic additives (e.g., cellulose) or corrosive substances (e.g., chloride ions) (Mishra et al., 2017; Pellenq et al., 2009; Zhang, et al., 2021). Molecular simulation can be used as a powerful tool to yield a profound scientific understanding of cementitious materials and their behaviors across several length scales (as shown in **Figure 1**). Based on the understanding of intrinsic mechanisms, molecular simulation can complement experimental studies to offer more explicit and efficient guides for the design of cementitious materials.

It is our great pleasure to present this research topic on *Molecular Simulation on Cementitious Materials: From Computational Chemistry Method to Application*. The collection of 11 articles in this research topic further discovers the nanoscale nature of cementitious material and guides applications of nanotechnologies in cement-based materials. We expect that these studies will advance cement chemistry and concrete technology through the application of modern computational methodologies alone, or in conjunction with experimental techniques. These studies will also push forward development of eco-efficient cementitious materials along the three directions pointed out by Scrivener et al. (2018): enhancing usage of supplementary cementitious materials (SCMs); developing alternative cements; and enhancing the efficiency of cement (production and application).

So far, utilization of SCMs is still the most effective way to improve the eco-efficiency of cement and concrete. The most popular SCMs are normally from industrial waste streams (e.g., coal fly ash and ground granulated blast-furnace slag), which contain amorphous alumina apart from amorphous silica. Thus, when such SCMs are used to replace part of cement in the binder phase of concrete, the formed C-S-H gel often involves Al-uptake, forming calcium-aluminosilicate-hydrate (C-A-S-H) gel. To better understand the effect of Al-uptake, Zhang et al. used reactive force field molecular dynamics to reproduce the Al-induced cross-linking effect on the aluminosilicate chains in the C-A-S-H gel, which is a major hydrate phase of cement with SCMs. The chemical and fracture processes coupled in this study demonstrated the significant

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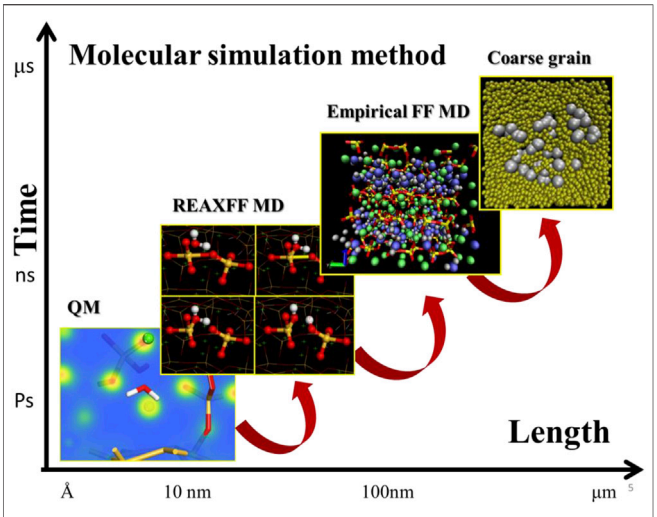

FIGURE 1 | Molecular simulation of cement-based materials at several length scales.

improvement of mechanical properties due to the incorporation of cross-linking aluminate structures.

When it comes to alternative binders, this research topic involves computational studies about alkali-activated materials, carbonation-hardening binders, and magnesium phosphate cement (MPC). Li et al. simulated a series of geopolymers using reactive force field molecular dynamics and Monte Carlo method. They found that the structure and mechanical properties of sodium aluminosilicate hydrate (N-A-S-H) gel is greatly dependent on sodium content: with Na/Al ratio rising, the aluminosilicate skeleton is transformed from an integral network to partially destroyed branch structures, reducing the stiffness and cohesive force of the gel. Wan et al. investigated the intrinsic mechanism for sodium dissolution (i.e., leaching) from the N-A-S-H gels of geopolymers. This study showed that sodium dissociation promotes a hydrolytic reaction, exacerbating the non-conservation of charge of the N-A-S-H system, and in the long term reduces the stability of the aluminosilicate skeleton. This finding is consistent with existing knowledge (Hou et al., 2014).

Carbonation-hardening binders have been attracting increasing attention because of their direct involvement of CO_2 mineralization. It has been known that nanotechnology can effectively improve the carbonation reactivity and, thus, the mechanical properties, of carbonation-hardening binders. Tao et al. presented first-principles calculations for screening potential dopants for increasing the carbonation reactivity of γ-Ca_2SiO_4 crystals. It is found that the carbonation reactivity is related to the reactive site distribution and the binding strength of γ-Ca_2SiO_4; and Ba, P, and F elements can potentially decrease the overall binding strength, which benefits the dissolution and carbonation reactions. MPC is a widely studied alternative binder for niche functional applications (e.g., fast repair, metal protection, fire retardant, etc.) (Ma et al., 2014). Similar to carbonation-hardening binders, the hardening process of MPC relies on reactions between a base oxide and an acidic component, which is completely different from traditional cements and alkali-activated materials. Li et al. proposed an experimental-computational approach to study the mechanical properties of magnesium potassium phosphate hexahydrate–the basic binding phase in MPC.

To improve the efficiency of cement utilization, a commonly considered approach is to use nano-reinforcement/modifier for improving the mechanical and/or durability properties of cement-based materials. Zhou et al. used reactive molecular simulation to investigate the effect of an emerging two-dimensional (2D) material—2D-silica–that is intentionally intercalated into the interlayer defective sites of C-S-H. This work may shed new light on the interaction mechanisms between 2D-materials and inorganic hosts, and provide solutions to modifying concrete against its high brittleness. Cement efficiency can also be improved by increasing strength and durability of concrete, since improved strength and durability imply needs of smaller volume of material in the construction stage and the whole life-cycle, respectively. Wang et al. advanced a well-known type of high-strength concrete (i.e., reactive powder concrete), and proved theoretically and experimentally that a well-selected delay (of start of steam curing after molding) can benefit the development of microstructure and strength. Realizing that high-strength concrete requires improved cracking resistance, Yue et al. proposed a methodology to optimize high-strength concrete mix proportion towards better cracking resistance using artificial neural network and genetic algorithm. To better understand the durability of concrete so as to improve it, Xu et al. simulated the coupled penetration of sulfate ions and chloride ions into concrete, say, in marine service environment; and Liu et al. developed a meso-scale five-phase model to decode the complexity of the microstructure, as well as to describe the transport and distribution of water in recycled aggregate incorporated concrete. In addition, to a large extent, the true quality of concrete (no matter how well it is designed) is determined by its workability (how well it can be manipulated in the fresh state). To better understand the nature of workability, Li et al. simulated the slump and J-ring tests of highly flowable concrete using discrete element method.

The Guest Editorial team is confident that the successful application of computational methods will facilitate the fundamental research and technology development of cementitious materials.

AUTHOR CONTRIBUTIONS

All authors listed have made a substantial, direct, and intellectual contribution to the work and approved it for publication.

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