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Unravelling CSH atomic structure via computational and experimental physical chemistry

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ABSTRACT

Calcium Silicate Hydrate (CSH) is the main binding phase for the cement paste, which is responsible for its strength and creep behavior. This is a nonstoichiometric hydration phase with calcium to silicon ratio (C/S) ranging from 1 to 2.2. At low C/S ratios, the molecular structure of CSH resembles to that of Tobermorite minerals, whereas in high C/S ratios it mostly looks like disordered glasses. By taking advantage of tools of statistical physics, it is shown that CSH at a given C/S can be associated with degenerate molecular structures called CSH polymorphs. Polymorphs are energetically competitive, i.e., they have the same free energy content, which means they can coexist under equilibrium conditions. To start, SiO₂ groups are randomly removed from the layered atomic structure Tobermorite 11A. One hundred and fifty structures are created. Grand Canonical Monte Carlo simulation of water adsorption is performed to adsorb water in the interlayer spacing and nanoscale porosities in defected CSH structures. The amount of adsorbed water scales linearly with the number of defects in the calcium–silicate layer. Samples are relaxed using a reactive potential in canonical and isothermal–isobaric ensembles. We observe that the confined water reacts with the free interlayer calcium atoms and nonbridging oxygen to form hydroxyl groups. The number of hydroxyl groups scales linearly with the amount of defects. The amount of water in CSH and Ca-OH content match well with drying and Neutron Scattering experiment. Although the reactive modeling of CSH impacts the water molecules in CSH's nanoconfinement environment, it does not significantly affect the silica chain length. This means that the reactive atomistic modeling does not affect the calico-silicate backbone of CSH structures. The silica mean chain length from atomistic simulation aligns perfectly with experimental NMR data. The elastic properties and hardness of all CSH polymorphs are measured at a given C/S and are directly compared with nano-chemo-mechanical testing via coupled nanoindentation and X-ray WDS. Atomistic simulation matches with the experimental data in both elastic and plastic regimes. The correlation of mechanical properties to structural observables of the molecular structures such as dimer content, mean silicate chain length, density, basal distance, water content, number of hydroxyl groups, and topological constraints parameter are calculated. No direct correlations were found at short ranges. The search was extended to the medium range order analysis and it is found that the polymorphism is closely related to the medium range order of Si-O bonds.