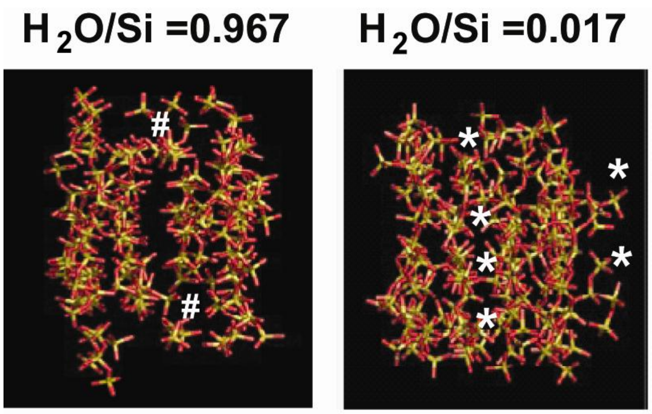
Why Wet C-S-H is Weak

Problem

It is known empirically that key properties of cement paste and concrete depend significantly on water content. Why do the stiffness and strength of cement paste vary with water content and relative humidity, and how is this related to changes in the nanoscale calcium-silicate-hydrate (C-S-H) binding phase itself? The strength of "dry" cement pastes derives largely from the C-S-H phase, and it is hypothesized that the mechanical characteristics of synthetic C-S-H depend substantially on the presence of water between silicate sheets. However, experiments alone cannot validate such hypotheses, due to material purity and instrument resolution at the relevant length- and time-scales.



Connectivity among silica tetrahedra in C-S-H at different water content levels, where yellow chains are silicates. At molar ratio H_2O/Si = 0.967 or ~80%RH, only two points of connection exist between the two silicate-rich layers of a C-S-H unit cell (at #), However, at H_2O/Si = 0.017 or ¡1%RH, three times more connections exist (at *).

Approach

The C-S-H structure was equilibrated via molecular dynamics, using the CSHub's recently developed classical force field, CSH-FF (Shahsavari et al., 2010). Water content was varied via the Grand Canonical Monte Carlo (GCMC) method: for a given temperature and system volume, the number of water molecules adsorbed increase with increasing partial pressure, relating to local environmental factors including relative humidity (%RH) or temperature of C-S-H. Structural and dynamic properties of each structure were calculated via molecular dynamics.

Findings

We find that the stiffness and strength of C-S-H increase with decreasing water content, accessible via decreased humidity or increased temperature (below C-S-H decomposition at >400 K). Extreme variation from saturated water vapor in cured cement pastes to 0%RH within C-S-H increases stiffness by 40% and hardness by 90%. Changes in stiffness for modest excursions from ~11%RH agree quantitatively with experimental characterization in cement pastes (Alizedeh et al., 2011). Importantly, our simulations allow us to attribute these trends to three correlated mechanisms. With decreasing water content within the nanoscale C-S-H, we find decreased distance between silicate-rich layers within the C-S-H, increased connectivity among silica tetrahedra, and decreased contributions of the mechanically weak water to mechanical deformation.

Impact

These results provide new understanding of how C-S-H structure and properties vary with water content. The proposed mechanisms governing increased stiffness and strength of C-S-H with low water content, particularly the humidity-dependent silica connectivity and resistance to shear deformation, can now be validated experimentally.

More

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This research was carried out by the CSHub@MIT with sponsorship provided by the Portland Cement Association (PCA) and the National Ready Mixed Concrete Association Research & Education Foundation. For more information, write to CSHub@mit.edu.