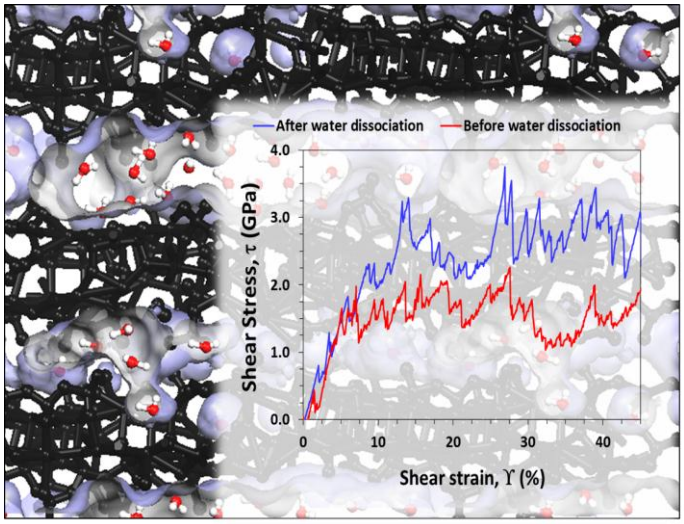
Gaining strength by splitting water

Problem

While, it is well known that structural water coexists in the C-S-H gel as molecular water (H_2O) and hydroxyl groups (OH^-), in fact, the "H" in the C-S-H acronym accounts for both with no distinction. However, even the most refined experimental methods cannot directly access the H_2O/OH^- ratio, not to mention any effects of that ratio on C-S-H properties. Molecular simulations, on the other hand, are well suited to study this in detail.

It has been long recognized that water has an influence on the mechanical response of the C-S-H gel particles, i.e. the same importance as, for example, the Ca/Si ratio. We are interested in how the H_2O/OH^- ratio might induce changes in the solid structure of the C-S-H gel, and the impact of such modifications on properties. We report here new information about the molecular state of water in order to quantify the relationship between chemistry and mechanical properties. This can lead to new approaches in the design of cement matrices with improved properties.



Molecular scale representation of the water trapped in C-S-H gel nanopores. The Ca-O-Si layers are depicted in black, molecular water molecules in white and red, and the pore space in blue. The insert shows the increase in the maximum shear stress after molecular water dissociation to OH^-.

Approach

The study of chemical reactions in large complex systems over large time scales is a challenging topic in the field of atomistic simulation. To overcome the size and time limitations, we have enlarged the capabilities of the reactive force field (ReaxFF). This gives us the opportunity to study chemical reactions within C-S-H at the atomic scale, which can be used to explore aging and water speciation (H_2O/OH^- ratio) as a function of environmental (pressure and temperature) conditions.

Findings

At room temperature about 40% of the water in the C-S-H model (Ca/Si=1.65) dissociates, forming silanol groups from the non-bridging oxygen atoms (NBOs) of the silicate chains. Small interlaminar spaces also trap molecular water, which is unable to diffuse throughout the texture, and do not contribute to relaxation mechanisms of C-S-H under strain. Increased dissociation correlates with fewer mechanically weak regions within the nanoparticles of C-S-H and greater strength. Interestingly elastic properties remained unchanged upon water dissociation.

Impact

Besides giving a more refined description of the C-S-H gel atomic structure, we have demonstrated that water speciation has a great effect on the strength properties of C-S-H nanoparticles. It is important to note that controlling the speciation of water within the C-S-H model through the Ca/Si ratio and temperature, provides a new degree of freedom from which one can master/improve the strength of the solid C-S-H phase.

More Research presented by Dr. Hegoi Manzano, Post-doctoral Fellow of the CSHub (now at UPV/EHU, Spain), in collaboration with Mr. S. Moeini and Profs. A.C.T. van Duin, F.-J. Ulm and R.J.-M. Pellenq. More information can be found in: H. Manzano et al., *J. Am. Chem. Soc.* **2012,** *134* (4).

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