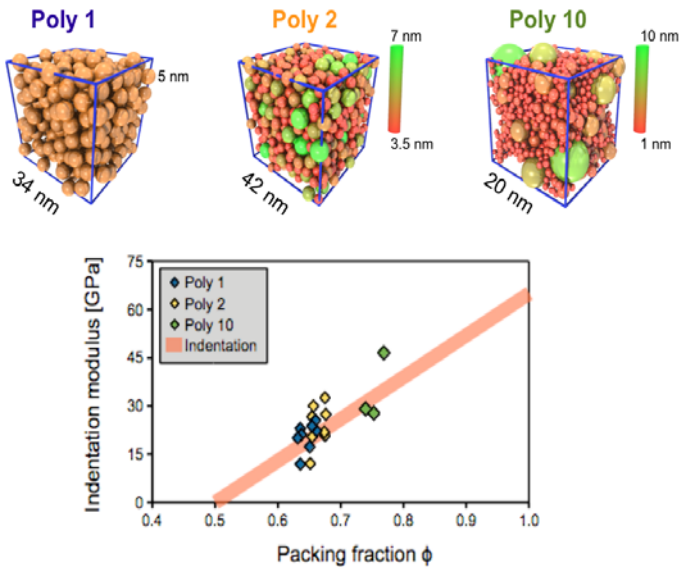
Setting from Statistical Principles

Problem

Cement reacts with water to set and develop strength and other rheological and mechanical properties as the result of the dissolution of clinker phases and the nearly simultaneous precipitation and growth of hydration products. Previous attempts to model cement setting either addressed precipitation at the atomistic scale on very short time scale, or employed percolation theory of expanding micron scale cement grains. The formation of hydration products at the intermediate scale of 1–100 nm is still largely unexplored and may provide a high potential for innovation. Indeed, controlling the formation of microstructure at the atomistic scale could yield cements with optimal rheo-mechanical performance that will enable the development of "setting-on-demand" materials. But to reach this goal, a new approach is needed to quantitatively use information from atomistic scale modeling to determine factors that control growth of hydration products and concurrent development of mechanical properties.



Nanoindentation modulus of C-S-H: Linear regression of experimental data [from Ulm et al., J.A.Ceram.S. 90(9), 2677-2692, 2007] and simulations of microstructures of different polydispersity ratios (difference in grain size). Grain size polydispersity increases the solid packing fraction and the mechanical properties.

Approach

Based on a statistical mechanics framework, cement hydration products are modeled as grains with variable size within a prescribed range. Interaction energies and forces between the grains are quantified in a bottom-up fashion through atomistic simulations. Grand Canonical Monte Carlo (GCMC) method is used to fill an initially empty simulation box with nano-sized grains. This yields predictions of the dynamics of microstructure evolution. Throughout this process, mechanical properties are monitored to track the liquid to solid transition.

Findings

Using interaction potentials for solid C-S-H and controlling grain size and grain size distribution, we generate microstructures with solid packing fractions between 0.63 and 0.77, that are representative of "low density" and "high density" C-S-H. The final microstructures are amorphous with local crystalline subregions. It is found that a higher polydispersity in grain size favors the formation of high-density C-S-H, resulting in enhanced mechanical properties. These model predictions are found to be consistent with nanoindentation, small angle neutron scattering and other measurements.

Impact

A new approach to quantitatively model the formation of cement hydration products, including their mechanical properties is proposed based on basic statistical mechanics. With the bottom-up philosophy, the input comes directly from the atomistic composition of solution and precipitates. Captured by cement-specific inter-particle potentials, investigation and control of microstructure and property evolution during setting become possible.

More

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