Water Content Dependence of the Structure and Properties of Nanoscale Calcium-Silicate-Hydrate

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Abstract

Calcium-silicate-hydrate, commonly abbreviated as C-S-H, is the chief nanoscale hydration product comprising Portland cement. In the synthesis of such complex multiscale materials, water content affects hydration kinetics, structure and properties of the hardened cementitious composite, and the subsequent changes in those properties during variations of temperature or humidity. Moreover, it has been observed experimentally that the stiffness and strength both of cement and of the C-S-H phase itself depend significantly on water content. The mechanisms driving these phenomena at the nanoscale, however, remain unclear. To this end, we conducted molecular simulations of C-S-H comprising varying water content. Our results show that the stiffness and strength increase as water content decreases, which can be accessed by decreasing relative humidity or increasing temperature. We 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 region to mechanical deformation. These results provide new predictive understanding of the dependences of C-S-H structure and properties on water content.