

**All-Atom Models of Tobermorite 11 Å and 14 Å - Benchmarks for Realistic Modelling of C-S-H**

T. Jamil<sup>1</sup>, R. K. Mishra<sup>2</sup>, R. J. Flatt<sup>2</sup>, H. Heinz<sup>1</sup>

<sup>1</sup>Chemical and Biological Engineering Department, University of Colorado Boulder, CO, USA, <sup>2</sup>  
Institute for Building Materials, ETH Zurich, Zurich, Switzerland

Calcium silicate hydrate is the main binding phase in cementitious materials. Its accurate atomic-scale models and force fields are crucial in the development of a greener construction industry. Tobermorite 11 Å and 14 Å minerals are known to be the closest crystalline structural analogs for calcium silicate hydrate (C-S-H). Therefore, dependable tobermorite models can serve as a benchmark for C-S-H, which exhibits complex poorly crystalline structures and variable surface chemistry at solid-liquid interfaces and for which nanoscale details are difficult to obtain from experiments. The Interface force field describes models that reproduce covalent versus ionic bonding and surface properties in agreement with experiment in contrast to other models. A new version of tobermorite parameters with extensive validation is presented in this study. Atomic charges of calcium ions of 1.5e in the intra-layer region and 1.7e in the partially hydrated inter-layer region were found to best describe chemical bonding. The lattice parameters of both minerals deviate less than 0.5% on average from X-ray data. The surface energy of tobermorite 14 Å ( $405 \pm 10$  mJ/m<sup>2</sup> least for 001 facet) agrees with available experimental measurements for C-S-H (Ca<sub>3</sub>Si<sub>2</sub>O<sub>7</sub>·2H<sub>2</sub>O) of  $386 \pm 20$  mJ/m<sup>2</sup>, whereas other models lead to deviations over 50%. The computed bulk moduli of tobermorite 11 Å and 14 Å are  $71 \pm 2$  GPa and  $45 \pm 1$  GPa, respectively, matching experiment within the uncertainty. Parameters developed using the INTERFACE approach (IFF) are consistent with other important cement phases such as CH, C<sub>3</sub>S, C<sub>3</sub>A, CaO, water, and polymers. This platform is therefore helpful in defining full scale atomistic models of C-S-H multiphase mixtures with local and global morphologies. Brief applications to understand conformations and working mechanisms of polycarboxylate ether superplasticizers in comparison to experiment are demonstrated using these models.