Understanding the Nanoscale Structure, Mechanics, Hydration, and Organic Interfaces of Calcium Sulfate Phases Using an Accurate Force Field

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Calcium sulfates are major components in materials for construction, casts, implants, and tissue healing. Despite their broad usage, the relationship between nanoscale properties and functions delivered by these minerals has remained largely unknown. This study reports insight into the structure and dynamics of calcium sulfate minerals and organic interfaces in atomic-detail using a novel force field and molecular dynamics simulations. Data from X-ray diffraction, interfacial, and mechanical measurements are reproduced in high accuracy, one to two orders of magnitude better than generic atomistic force fields. Nanoscale origins of hydration, solubility, and modification of crystal growth by polymers are described. Thereby, structures of the minerals and their hydrated phases, (h k l) cleavage planes, hydration reactions and energies, anisotropic mechanical and thermal properties, as well as facet-specific adsorption of organic modifiers are explained. Using our validated atomistic model of calcium sulfate minerals, we have also verified the transition temperature for the gypsum to anhydrite conversion and the thermal stability of these phases at a given temperature range. The force field matches available experimental measurements, exceeds the accuracy of common DFT methods, and can be applied to systems as large as 100 million atoms. The models can be used with multiple force fields (INTERFACE FF, CHARMM, AMBER, GROMACS, PCFF, OPLS-AA) and for similar sulfate/sulfonate nanomaterials.