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Density Functional Theory on the Structure of Montmorillonite

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Total-energy calculations with ultrasoft pseudopotentials and plane-wave basis functions were performed on montmorillonites of varying layer charge using density functional theory. Full atomic coordinates with the unit cell geometry of montmorillonite were determined. Important structural features, including interatomic distances, interatomic angles, surface corrugation, and tetrahedral rotation were compared with theoretical and/or experimental values for montmorillonite and for pyrophyllite, which is structurally isomorphic to montmorillonite but without layer charge. The effect of charge substitution and interlayer cation on the structure, especially its hydroxyl groups, was studied systematically. Simulated X-ray diffraction patterns and IR/Raman spectra also were obtained for montmorillonite and pyrophyllite. These quantum mechanical calculations of clay mineral structure, which require no adjustable parameters, gave excellent results for both equilibrium structures and total energies. All ab-initio calculations were performed with Cray T-3E and IBM SP RS/6000 supercomputers at the National Energy Research Scientific Computing Center (NERSC).