Intercalation of CO_2 and H_2O in Na-hectorite under geological carbon sequestration conditions using GCMD simulations

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We report computer simulations using grand canonical molecular dynamics (GCMD) to understand the intercalation of CO_2 and H_2O at different combinations of temperature and pressure relevant to petroleum reservoirs and geological carbon sequestration. The simulations are based on the smectite mineral hectorite with Na^+ as the charge compensating cation. In the GCMD simulations the interlayer spacings were fixed at values from 9.5Å (dry state) to 18.0Å (3-layer hydrate) at intervals of 0.2Å. The interlayer mole fractions imposed by the chemical potential of the CO_2/H_2O bulk phase were quantitatively analyzed using equilibrium adsorption isotherms. The results agree well with experiments and confirm the notion that the presence of H_2O aids CO_2 intercalation, which is inhibited under dry conditions. Maximum CO_2 adsorption occurs at 12.2Å which is in the range of monolayer structures. The adsorbed CO_2 molecules exhibit parallel orientations with respect to basal hectorite surface and are coordinated to the basal surface. The interlayer species exhibit significant redistribution between different coordinations as the temperature and pressure changes. The orientation of the structural hydroxyl groups plays an important role in determining the interlayer CO_2/H_2O content. The variation in adsorption structure, nearest neighbour coordinations, residence time correlation functions, and diffusional mobility of H_2O and CO_2 as functions of interlayer spacing are consistent with available experimental and simulation data and provide a detailed structural and dynamical understanding of CO_2 and H_2O intercalation on a molecular scale.