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Carbon dioxide intercalation in Na- and Ca-exchanged montmorillonite

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Carbon capture and storage (CCS) is expected to mitigate anthropogenic CO_2 emissions from discrete CO_2 sources, such as electric power stations. A widely considered CCS option is underground geologic storage, and this requires, among other things, a reservoir confined by an impermeable caprock. Because this caprock is commonly clay-dominated, understanding the interactions between clay minerals, brines, and CO_2 is essential for site selection for long-term CO_2 storage. Two types of high-pressure environmental chambers (HPECs) developed at UIC were used for X-ray diffraction studies of Na- and Ca-exchanged montmorillonite (CMS source clay SWy-2): a transmission mode HPEC (THPEC) and a reflection mode HPEC (RHPEC).

The THPEC experiments used random powder aggregates of Na-exchanged SWy-2. The clay was grain size sorted by sedimentation to select the fine fraction ($< 2\ \mu$m), cation exchanged, dried at 200^{o}C, then stored in a desiccator. This was followed by exposure to moist air for varying lengths of time to produce samples with different amounts of interlayer H_2O. Samples were then exposed to gaseous or supercritical CO_2 and held at a temperature of 40-45^{o}C. The THPEC results show that the position and shape of the (001) peak changed for most samples under low CO_2 pressures (40-50 bars). This response to CO_2 is strongly related to the initial H_2O content. For Na-exchanged SWy-2 with an initial d(001) = $\sim$10.5 Å, the peak position changes to $\sim$12.25 Å. However, nearly dry [d(001) = 10.10 Å] samples do not exhibit significant changes to the (001) peak. Increasing pressure to P = 650 bars (supercritical) does not produce any further significant changes to the (001) peak compared to the P = 50 bars (gaseous) results.

The RHPEC experiments used fine fraction, Na- and Ca-exchanged SWy-2 in oriented aggregates on glass slides, which were either stored in a desiccator or in a vessel above a saturated salt solution. The RHPEC cannot be used above $\sim$50 bars CO_2 due to attenuation of the X-rays by the pressurizing gas; these experiments were conducted at ambient T ($\sim$22^{o}C). The RHPEC results for NaSWy-2 and CaSWy-2 indicate that both expand under CO_2 with a non-monotonic dependence of CO_2-induced expansion on the initial H_2O content of the interlayer. Expansion is minimal for near-dry samples and for samples with an initial d(001) = $\sim$12.5 Å, $i.e.$, NaSWy-2 or CaSWy-2 having one "plane" of interlayer H_2O. For samples with a significantly different initial d(001), expansion of up to $\sim$10% is observed.

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