



Strain development in smectite clays upon exposure to CO₂

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Smectites (or swelling clays) are common constituents of claystones, mudstones and shales and are often present in the caprocks and faults sealing potential CO₂ storage reservoirs. Their crystal structure is comprised of alternating silicate layers separated by an interlayer region, containing cations and water molecules. As the water molecules are easily exchanged between this region and the intergranular pore space, the structure can expand or shrink depending on factors such as temperature, water activity and clay composition. Whereas the water uptake and swelling properties of smectite clays have been studied extensively, fewer studies have been directed at possible interactions with CO₂. However, several scenarios including shrinkage (dehydration) and swelling (surface adsorption or uptake of CO₂ into the interlayer region) of the crystals are conceivable, which could have significant implications for caprock and fault integrity.

To investigate possible effects of CO₂ on the swelling properties of smectite clays, we performed unconfined volumetric strain measurements on compacted pellets of montmorillonite (SWy-1), which is a common type of smectite, and on smectite-bearing shale. This was done using an optical cell. We probed the macroscopic response of the pressed samples to assess the overall strain response to exposure to CO₂ at typical P-T conditions expected in carbon dioxide storage sites, i.e. at a temperature of 45°C and CO₂ pressures up to 15MPa. Samples were heat-treated prior to exposure to CO₂ to obtain a defined hydration state (d001-spacing). This was determined independently using X-ray diffraction methods.

Our results show that montmorillonite SWy-1 swells almost instantaneously (in a few seconds) to an equilibrium state, when placed in contact with (supercritical) CO₂ for the conditions $PCO_2 \leq 8$ MPa, $T = 45^\circ\text{C}$. Maximum swelling is observed for an initial d001 spacing of 11Å, reaching $2.4 \pm 0.45\%$ at a CO₂ pressure of 15MPa. Only minor swelling ($0.55 \pm 0.46\%$ at 10MPa CO₂ pressure) is found for clay with an initial d001 spacing equivalent to 9.8Å. For an initial spacing of $9.8\text{Å} \leq d001 \leq 11\text{Å}$, swelling decreases systematically with decreasing d-spacing at all pressures investigated. The strain effect is already observed at low CO₂ pressure (1MPa) and expansion appears to be completed at $PCO_2 \sim 8$ MPa. Shale/claystone samples containing 53% smectite displays a volume increase of $\sim 1.0\%$, at $T = 45^\circ\text{C}$ and $PCO_2 = 10$ MPa. No swelling effects were observed in identical experiments performed using Argon gas.

Our results demonstrate that montmorillonite swells by up to a few percent due to uptake of CO₂. Whether the CO₂ actually penetrates the interlayer region or is adsorbed at clay crystallite surfaces remains unclear. No evidence is found for CO₂ induced dehydration (shrinkage) of the crystal structure. For smectite-bearing caprocks above carbon sequestration reservoirs, our results suggest that a minor swelling can be expected if CO₂ migrates into caprocks and faults, probably closing any open fractures or joints and thus reducing bulk permeability.

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