In-situ Powder X-ray Diffraction of a Smectite-CO_2-Brine Sytem Using a High-Pressure Environmental Chamber

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The process of carbon dioxide (CO_2) capture and storage (CCS), where supercritical CO_2 is injected in deep saline aquifers [1] and sealed by clay-rich impermeable cap rocks, has been considered an effective approach to curb the increase of anthropogenic CO_2 in the environment. Smectite, a primary clay mineral constituent in both the aquifer and the cap rock, has the capability to control porosity and permeability with changes in local conditions. Here, we experimentally investigated the activity of H_2O [$a(H_2O)$] by studying the effects of varying temperature (T), partial pressure of CO_2 [$P(CO_2)$], and brine composition and concentration using a high-pressure X-ray environmental chamber [2] to assess the volume change of smectite from the $d(001)$ peak. Increase in brine concentrations from 0.17 M to saturation, and T from 0 to 150 ˚C, for both Na- and Ca-smectite, produces a large decrease of $d(001)$ values of up to 25%. Furthermore, the effect of increase in $P(CO_2)$ to 500 bars at constant T and brine concentration shows a small effect on the $d(001)$. Changing the interlayer cation from Na^+ to Ca^{2+} can decrease the $d(001)$ by 20%. Experiments performed at He pressure show identical results with CO_2, within error, suggesting that CO_2 does not enter the interlayer. The results show that brine composition and concentration, T, $P(CO_2)$, and interlayer cation composition are important parameters affecting the $d(001)$ spacing and the $a(H_2O)$. The changes in $d(001)$, due to the migration of H_2O in to smectite, may compromise the safety and integrity of reservoirs targeted for CO_2 sequestration because of volume changes and the formation of fractures of the cap rock.

[1]IPCC (2014) Climate Change 2014: Synthesis Report. Contribution of Working Groups I, II and III to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, IPCC, Geneva, Switzerland, 151 pp.

[2]Guggenheim, S. and Koster van Groos, A. F. (2014) An Integrated Experimental System for Solid-Gas-Liquid Environmental Cells. Clays and Clay Minerals, 62, 479-485.