



Interactions between scCO₂ and scCO₂-SO₂, formation fluids and rock forming minerals: an experimental approach under in-situ conditions of potential geological storages

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The storage of CO₂ in the deep underground is regarded to be one possibility to reduce the release of anthropogenic CO₂ in the atmosphere. Therefore, experiments have been performed to investigate the interactions between CO₂ and concomitant gases with potential reservoir rocks considering both the injection and storage phases of the sequestration process. With respect to the composition of CO₂ captured industrially from coal-fired power plants using the Oxyfuel technology contaminants like SO₂ (plus possibly NO_x and O₂) were chosen to investigate chemical corrosion of rock-forming minerals with impure supercritical CO₂ (scCO₂) at possible storage conditions of >73.7 bar and >31°C. Co-injection of SO₂ (and NO_x and O₂) has been expected to cause a stronger brine acidification of the deep saline aquifers than CO₂ alone. Experiments up to 1000h have been performed with 3n NaCl solution, 12 natural mineral concentrates and pure scCO₂ or scCO₂+SO₂ (99.5+0.5vol-%; "impure scCO₂") gas mixture. The brine composition reflects the average salinity of deep formation waters of the North German Basin. To increase the reaction rates all minerals were ground and the reagents agitated either by stirring or shaking in 1L-reaction autoclaves. The autoclaves consist of HastelloyTM or ferromagnetic stainless steel fully coated with PTFE to avoid contact between metal and reaction agents. In average, we used 15 g of solids and 700 ml liquid. The autoclaves were pressurised up to 100 bar with CO₂ or CO₂+SO₂ mixture. Before, during and after the experiments reaction fluids were sampled and analysed by ICP-MS and IC. Solid phases were characterised by XRF, ICP-MS, and EMPA before and after the experiment and pH, pressure and temperature are monitored. Interactions with pure scCO₂ show slight changes in the mineral composition of albite as it gained in a not yet identified carbonate phase that might be dawsonite. The compositions of e.g. clay minerals, microcline, anhydrite and micas remain nearly unchanged at a min. pH of 4. Carbonates, like calcite, dolomite and siderite were corroded at min. pH of 7-8. Experiments using scCO₂+SO₂ have a lower pH than using scCO₂ alone, at which those with silicates or anhydrite have a lower pH (2-3) than experiments with carbonates (>5). Fluid-mineral interactions are generally stronger using impure scCO₂+SO₂ and reaction fluids contain much higher amounts of cations, especially Fe, Si and Al when silicates were studied. However, the intensity and rate of reaction is controlled by the availability of SO₂.