

An experimental study of the geochemical impact of CO₂ leakage in siliclastic aquifers

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Leakage of CO₂ from deep storage formations into an overlying potable aquifer may mobilize trace metals leading to undesirable water quality degradation. Understanding CO₂-water-rock interactions under leakage conditions is therefore an important step toward the safe implementation of geologic carbon sequestration. In this study we investigated the geochemical response of three sandstone samples from the Mesaverde Group in northwestern Colorado. Two batch dissolution experiments were conducted in which samples were reacted with water and CO₂ at partial pressures of 0.01 and 1 bar, representing natural background levels and levels expected in an aquifer impacted by a small leak of CO₂, respectively. The pH dropped sharply after CO₂ was introduced into the system, and then rebounded slightly as the minerals in the rocks dissolved. Concentrations of major (e.g., Ca, Mg, Fe) and trace (e.g., As, Ba, Cd, Pb, Sr, U) elements in the fluids increased over the 4 week duration of the experiments. These concentrations increased more in the 1 bar CO₂ experiments compared to the 0.01 bar CO₂ experiments. Throughout the experiments the concentrations of potentially toxic constituents remained below regulatory limits. Sequential extraction results suggest that carbonate minerals, although volumetrically insignificant in sandstones, are the dominant source of metals. A simple geochemical model was developed that simulates observed changes in fluid composition and supports the interpretation that carbonate minerals are an important source of metals in siliclastic aquifers impacted by CO₂ leakage.

Reaction path geochemical modelling of CO₂-SO₂-water-rock experiments

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The CO₂CRC has conducted an in depth study of the geochemical impacts of CO₂ storage with co-contaminant SO₂. In this study, laboratory experiments were conducted in static reaction vessels to investigate the chemical evolution of the CO₂-SO₂-water-rock system of core samples from 3 different geologic formations in the Surat Basin of southern Queensland, Australia. Numerical modelling of the experiments was undertaken to evaluate modelling capabilities as well as to determine which factors contribute significantly to the chemical behaviour of the system. This work reports on the geochemical modelling component of the study.

The experiments were conducted at 60°C and 120 bar with scCO₂ and scCO₂ + SO₂ in a low salinity water and 3 different rock types (see Pearce *et al.*, Goldschmidt 2013). Changes in the gas and water chemistry as well as mineralogy were determined. Static reaction path geochemical modelling using Geochemist's Workbench was conducted to simulate the experimental conditions and output of the experiments. The simulations generated provided a reasonable history match to the experiments. It was noted that the experiments and even more so the numerical models were very sensitive to redox state. In the case of the experiments it appeared that the most important contribution to redox related to aqueous phase species, particular O₂ and Fe. In the geochemical modelling, the presence of ferric iron (usually initially in the form of hematite) played a significant role in determining model outcomes. The presence of oxidised iron drove SO₂ oxidation to H₂SO₄ to take place as well as the disproportionation reaction to H₂SO₄ and H₂S leading to increased mineral dissolution and pyrite precipitation in the simulations. However, without oxidised iron the disproportionation reaction dominated leading to elemental sulphur and pyrite precipitation. The experiments and modelling clearly indicate that understanding the redox state in terms of minerals and aqueous phase of any potential sites will be critical to the geochemical behaviour.