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Interaction Between CO₂-Rich Sulfate Solutions and Carbonate Reservoir Rocks from Atmospheric to Supercritical CO₂ Conditions: Experiments and Modeling

Details

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Abstract

A test site for CO₂ geological storage is situated in Hontomín (Spain) with a reservoir rock that is mainly composed of limestone. During and after CO₂ injection, the resulting CO₂-rich acid brine gives rise to the dissolution of carbonate minerals (calcite and dolomite) and gypsum (or anhydrite at depth) may precipitate since the reservoir brine contains sulfate. Experiments using columns filled with crushed limestone or dolostone were conducted under different P-pCO₂ conditions (atmospheric: 1-10-3.5 bar; subcritical: 10-10 bar; and supercritical: 150-34 bar), T (25, 40 and 60 °C) and input solution compositions (gypsum-undersaturated and gypsum-equilibrated solutions). We evaluated the effect of these parameters on the coupled reactions of calcite/dolomite dissolution and gypsum/anhydrite precipitation. The CrunchFlow and PhreeqC (v.3) numerical codes were used to perform reactive transport simulations of the experiments. Under the P-pCO₂-T conditions, the volume of precipitated gypsum was smaller than the volume of dissolved carbonate minerals, yielding an increase in porosity (Δ porosity up to $\approx$ 4%). A decrease in T favored limestone dissolution regardless of pCO₂ owing to increasing undersaturation with decreasing temperature. However, gypsum precipitation was favored at high T and under atmospheric pCO₂ conditions but not at high T and under 10 bar of pCO₂ conditions. The increase in limestone dissolution with pCO₂ was directly attributed to pH, which was more acidic at higher pCO₂. Increasing pCO₂, carbonate dissolution occurred along the column whereas it was localized in the very inlet under atmospheric conditions. This was due to the buffer capacity of the carbonic acid, which maintains pH at around 5 and keeps the solution undersaturated with respect to calcite and dolomite along the column. 1D reactive transport simulations reproduced the experimental data (carbonate dissolution and gypsum precipitation for different P-pCO₂-T conditions). Drawing on reaction rate laws in the literature, we used the reactive surface area to fit the

models to the experimental data. The values of the reactive surface area were much smaller than those calculated of the geometric areas.

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