Gypsum precipitation during interaction between CO_2-rich sulfate solution and carbonate rocks (from atmospheric to supercritical CO_2 conditions)

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Introduction

A test site for a prospective CO_2 geological storage is situated in Hontomín (northern Spain) with a reservoir rock that is mainly composed of limestone. During and after CO_2 injection, the resulting CO_2-rich acid brine will likely promote the dissolution of carbonate minerals (calcite and dolomite) and, since the reservoir brine contains sulfate, gypsum (or anhydrite at depth) may precipitate.

Methodology

Column experiments filled with crushed limestone or dolostone rock fragments were conducted at different P-pCO_2 conditions (atmospheric: $1\text{-}10^{-3.5}$ bar; subcritical: 10-10 bar; and supercritical: 150-34 bar), T (25, 40 and 60 ºC) and input solution compositions (gypsum undersaturated and gypsum equilibrated solutions). We evaluated the effect that these parameters exert 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.

Results and discussion

In the range of P-pCO_2 and T of this study only gypsum precipitation took place (no anhydrite was detected) and it only occurred if the injected solution was equilibrated with respect to gypsum. Comparing the two reservoir rock reactivities, limestone dissolution induced late gypsum precipitation (long induction time), in contrast to dolostone dissolution that promoted fast gypsum precipitation. A decrease in T favored limestone dissolution regardless of pCO_2. However, gypsum precipitation was favored at high T and atmospheric pCO_2 conditions and unfavored at high T and 10 bar of pCO_2. The 1D reactive transport simulations reproduced the experimental data (carbonate dissolution and gypsum precipitation for different P-pCO_2-T). Reactive surface area was used to fit the models to the experimental data. It was inferred that reactive surface area values were much smaller than the calculated geometric areas.