Effect of S impurities on the interaction between CO_2-rich solutions and limestone and marl rocks at P = pCO_2 = 10 bar

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Introduction

The effects of impurities (e.g. H_2S and SO_2) on geological storage of CO_2 are uncertain. Oxidation of these impurities leads to the formation of sulfuric acid and a decrease in pH of the formation water. We investigate the effect of added H_2SO_4 on the reactivity of the reservoir (limestone) and cap (marl) rocks of the Hontomin site (Spain). The increased acidity may induce a decrease in the solubility of CO_2, an increase in carbonate mineral dissolution and additional precipitation of gypsum.

Methodology

Flow-through column experiments filled with crushed limestone or marl were conducted under subcritical CO_2 conditions (P = pCO_2 = 10 bar and 60 ºC). As the Hontomin groundwater is in equilibrium with respect to gypsum, we evaluate the effect that an addition of 3 % S (as H_2SO_4) with respect to the dissolved CO_2 exerts on the reactivity of the rocks. The CrunchFlow numerical code is used to perform 1D reactive transport simulations of the experiments.

Results and discussion

In the P-pCO_2 and T of this study, calcite dissolution fostered gypsum precipitation. The addition of H_2SO_4 lowered the pH in the injected solution by 1.5 pH units with respect to the pH of 3.5 of the H_2SO_4-free brine. A comparison between the reactivity of the rocks reacted in H_2SO_4-free and H_2SO_4-rich brine shows that calcite dissolution and gypsum precipitation in both rocks were larger in the H_2SO_4-rich brine. As a consequence, a higher increase in porosity was observed, even with the larger gypsum precipitation.