Geophysical Research Abstracts
Vol. 13, EGU2011-737, 2011
EGU General Assembly 2011

Experimental study of calcite dissolution and gypsum precipitation in acid brines

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Injection of CO2 into suitable host rock formations represents one of the most promising options to limit the atmospheric concentration of CO2. A test site for a prospective CO2 reservoir is the Hontomin site (Spain) in which the repository rocks for CO2 injection are dominantly limestones and dolostones. The injection of CO2 will cause the formation of CO2-rich acid brines, which will induce the dissolution of the carbonate minerals. Since the brine contains sulfate, gypsum (or anhydrite at depth) will precipitate, which may cover the surface of the dissolving carbonate causing its passivation. These reactions imply changes in porosity and in the structure of pores in the repository rocks. Therefore, changes in permeability and fluid flow are expected.

To understand the effect that dissolved CO2 exerts on the reactivity of calcite in the acidic brine, experimental work is carried out to study the kinetics of the dissolution of calcite coupled to the precipitation of gypsum at atmospheric PCO2.

The dissolution of calcite and precipitation of gypsum is studied by means of flow-through experiments. An acidic solution (H2SO4, pH 2.5) at equilibrium with gypsum is continually injected into a cell containing known mass of calcite with variable grain size. Variation of calcium, total sulfur and pH is monitored with time. The evolution of solution composition indicates that the ratio between the volumes of gypsum precipitated and calcite dissolved equals 1.4, which would translate into a decrease in porosity of the rock matrix.

Microscopic examination of the surface of reacted grains suggests that gypsum precipitates heterogeneously upon the calcite surface. AFM experiments performed on the calcite cleavage surface (10-14) reacting with the acidic brine allow studying the mechanism by which gypsum precipitates on the dissolving calcite cleavage surface.