Dissolution experiments and simulations: implications for CO2 underground sequestration into aquifers of the Boso Peninsula

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1. Background

Underground sequestration of CO2 is expected to play an important role in mitigating further increases in atmospheric CO2. Among the three principle methods that constitute CO2 underground sequestration - structural trapping, solubility trapping (eq. (1)-(3)), and mineral trapping (eq. (4)) - the latter two have received little attention especially in terms of experimental research. Here, we present temporal changes in solubility and mineral trapping deduced from changes in abundance of aqueous and solid phase species.

CO2 + H2O = H2CO3 = H+ + HCO3- (1) CaCO3 + CO2 + H2O = Ca2+ + 2HCO3- (2) CaSiO3 + 2CO2 + H2O = Ca2+ + 2HCO3- + SiO2 (3)Ca2+ + 2HCO3- = CaCO3 + CO2 + H2O (4)

2. Materials & Methods

We conducted dissolution experiments and computer simulations using 3 rock samples from the Boso Peninsula, Chiba. The main constituents of the samples were as follows: A (quartz and feldspars), B (quartz, feldspars, and dolomite), C (quartz, feldspars, and calcite). Each sample was disintegrated into powder with a pot mill and was left to react with solutions of 5 different pH in the range pH = 3-11. HCl and Na2CO3 were used for acidic and basic adjustments, respectively. On seven occasions between 1-30 days after starting the reaction, the solid and aqueous phases were separated and the compositions of each phase were analyzed.

3. Results & Discussion

Progress in reaction (2) led to rapid increases in bicarbonate concentrations within 1 day after starting the experiment using samples B and C. At the same time, pH rose from 3 to 7.7 using samples B and C, and from 3 to 3.8 using sample A. This implies that reaction (3) did not make much progress within 30days, and that solubility trapping occurred through the dissolution of carbonates within this timeframe.

Simulation results show that most of the feldspars' dissolution start 1 year from the start of the reaction. Progress in reaction (3) causes the aqueous phase to become saturated with CaCO3, and calcite begins to precipitate subsequently. Therefore, we expect mineral trapping to take effect 1 year from the start of the reaction.