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Chemical Interactions between CO₂-saturated Brines and Carbonate Minerals at Reservoir Conditions

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SUMMARY

Injection of carbon dioxide into deep saline aquifers results in an acidic solution that can react with the reservoir rocks, especially carbonate minerals, with consequent changes in the porosity and permeability of the formation. Numerous factors control the extent and rate of these processes and in this paper we present a fundamental experimental and modelling study of the kinetics and equilibria of the underlying chemical reactions.

Abstract

Injection of carbon dioxide into deep saline aquifers results in an acidic solution that can react with the reservoir rocks, especially carbonate minerals, with consequent changes in the porosity and permeability of the formation. Numerous factors control the extent and rate of these processes and in this paper we present a fundamental experimental and modelling study of the kinetics and equilibria of the underlying chemical reactions.

Experimentally, we have studied the kinetics of carbonate-mineral dissolution in the CO_2 -water system at temperatures up to 373 K and pressures up to 14 MPa. As illustrated in Figure 1, the kinetics are influenced by both mass transfer and chemical reaction rates, but our work has focused on an regime in which the latter are the controlling factor. Pure calcite and dolomite have been studied, as well as samples of chalk and limestone. The results show that two parallel reaction pathways, one involving protons and the other $\text{CO}_2(\text{aq})$, are important under far-from-equilibrium conditions. We have also quantified the impact of certain adsorbed organic films on the rate of calcite dissolution in CO_2 -saturated water.

Figure 1 Chemical and physical processes associated with carbonate mineral dissolution in CO_2 -saturated brine.

Additionally, we have measured the pH of CO_2 -saturated brines as a function of temperature (308 to 423 K), pressure (1 to 15 MPa) and salt molality (0 to 5 mol/kg) using a pair of carefully-calibrated high-pressure pH and reference electrodes. Interestingly, while increasing the salt concentration has the effect of reducing the amount of CO_2 dissolved at equilibrium, the pH of the CO_2 -saturated brines is similar to that of the CO_2 -water system at the same temperature and pressure.

The results of these experiments have been used to optimise parameters in appropriate kinetic and thermodynamic models that can be applied in field simulations. Robust and computationally-efficient numerical methods have been developed to allow the kinetic and equilibrium relations to be solved without the need for *a priori* assumptions concerning the number of phases present or the selection of dominant reaction pathways. A computer code implementing these methods is available.

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