



1 Experimental study of dissolution of siderite in high pressure high temperature H₂O-CO₂ fluids with implications for geological storage of CO₂

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Reduced or partially reduced iron phases are among the most reactive phases that CO₂ will encounter in deep subsurface environments and the major formed solid carbonate is the iron carbonate FeCO₃ siderite. For storage purposes, it is thus important to precisely model the stability of siderite: in these environments, the solubility of siderite is indeed the major limiting factor to mineral sequestration of CO₂. Existing data sets at storage conditions are not extensive and kinetic data are even scarcer.

An original experiment was designed for monitoring in-situ the dissolution of siderite in H₂O-CO₂ fluids, by X-ray absorption spectroscopy. A high pressure cell, well adapted to dissolution studies at deep storage conditions [1, 2] was installed at beam-line BM30B (FAME) of the ESRF synchrotron ring. The effect of pressure (0-800 bar), temperature (25-300°C), pH and ionic strength on siderite solubility were measured, from which a thermodynamic model was constructed. Data on dissolution kinetics were also measured. Speciation of Fe²⁺ ions in the H₂O-CO₂ fluids was studied by interpreting the XANES part of the X-ray absorption spectra.

Solubilities of the order of 0.01 M were measured at typical injection conditions (i.e.

300 bars and 100°C). Measured solubilities are lower, yet still quite elevated, in presence of NaCl 1M. Consistency of this effect with the thermodynamic models used will be discussed. It is shown that the presence of CO₂ and Cl⁻ has no major effect on iron speciation. Finally, the effect of a reduction of CO₂ by Fe²⁺ above 200°C was examined.

REFERENCES:

1. D. Testemale et al., Review of Scientific Instruments, submitted.
2. G. S. Pokrovski et al., Chemical Geology, accepted