

## The solubility of siderite $(FeCO_3)$ as a function of temperature

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Iron is the second most abundant metal on Earth occurring in a variety of rock and soil minerals in oxidation states II and III. Under anoxic conditions, the solubility of ferrous iron ( $Fe^{2+}$ ) is frequently controlled by the ferrous carbonate, siderite ( $FeCO_3$ ), by the equilibrium:

 $FeCO_{3(s)} = Fe^{2+} + CO_3^{2-}$ 

Siderite, a widespread mineral in near-surface sediments and ore deposits, occurs in hydrothermal veins, lead-silver ore deposits, sedimentary concretions formed in limestones and sandstones, and Precambrian banded iron formations that precipitated under acidic conditions. Siderite formation is known to be facilitated by both mesophilic and thermophilic iron reducing bacteria (*e.g.*, Zhang *et al.* 2001), and has been interpreted to be microbially mediated in many natural environments (see Mortimer and Coleman 1997). Siderite has also been mentioned lately as potential CO<sub>2</sub> mineral trapping in numerous computer simulation of CO<sub>2</sub> geological sequestration (Johnson *et al.*, 2002; Zerai *et al.*, 2006, Xu *et al.*, 2003) and was confirmed experimentally at 200°C and 20MPa by Kaszuba *et al.* (2003, 2005).

A number of previous studies have focus on the determination of the solubility product of  $\text{FeCO}_{3(s)}$  at low temperature (<90°C), various ionic strengths (from 0.1 to 1 molal NaClO<sub>4</sub> or 0.1 to 5.5 molal NaCl medium), and CO<sub>2</sub> pressure (from 0.05 to 0.01 atm pCO<sub>2</sub>). At 25°C, the values are widespread and ranged from  $10^{-11.03}$  to  $10^{-10.24}$ , and the values of the standard enthalpy of formation differ by more than 10 kJ/mol.

In this study, the solubility of a natural siderite (from Peyrebrune, Quarry, France) was investigated from 25 to 200°C at 0.1 molal NaCl and saturated vapor pressure

using a hydrogen-electrode concentration cell (HECC), which provided continuous, *in situ* measurement of hydrogen ion molality. Ferrous iron was analyzed by a revised Ferrozine-spectrophotometric method. The solubility products  $(Q_s)$  obtained were extrapolated to infinite dilution  $(K_s)$  for comparison with previous work.

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