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Siderite dissolution kinetics in acidic aqueous solutions from 60 to 100˚C and 0 to 50 atm pCO_2.

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The rate of dissolution of siderite has been measured at 60 and 100˚C in 0.1 M NaCl and pH from 2.1 to 4.6 at far from equilibrium conditions as a function of partial pressure of CO_2. Dissolution experiments on siderite crystal planes (from Peyrebrune, Quarry, France) were performed in batch titanium high pressure reactor under controlled hydrodynamic conditions using the rotating disk technique with stirring speed of 425 rpm. The pH was measured in-situ using a solid-contact electrode in a cell without liquid junction. Carbon dioxide was delivered in the reactor through a Ti porous filter and pCO_2 was controlled by calibrated pressure manometer. Solution samples were taken each 0.5-1.5 hrs through an in situ Ti 2 μm porous filter. Duration of experiments varied from 5 to 10 hrs depending on solution pH. The total amount of solution sampled (5-10 samples of $\sim$ 4 mL each) did not exceed 10 % of the initial mass of solution, but necessary corrections were made.

Rates were generated from measured solution composition as a function of time using

$R =$(d[Fe]/dt)$/s$

where t(sec) designates the elapsed time, [Fe] stands for the number of iron moles released from the solid, and s is crystal plane geometric area. In all experiments presented in this study, the same disk was used only once. Fe total concentration was measured by flame atomic absorbtion. Fe(III) was not detected in sampled solutions by spectrophotometric analyses using the ferrozine method, which allows determination of Fe(II) and Fe_{tot}.

Experimental results show a linear dependence of the logarithm of dissolution rates on pH consistent with R (mol/cm^2/s) = k·a_{H+}^n, where k = 6.21·10^{-7} mol/cm^2/s, n

= 0.96 at 60˚C and k = $3.37 \cdot 10^{-6}$ mol/cm^2/s, n = 0.90 at 100˚C. Activation energy for siderite dissolution varies from 54 kJ/mol at pH = 3.0 to 57 kJ/mol at pH=4.0 and is in good agreement with values recently determined by Dufauld (2006). Very weak (catalizing) effect of pCO_2 on siderite dissolution kinetics has been observed to pCO_2= 50 atm. Results obtained in this study can be used for modeling CO_2 reactive transport in sandstones and basic and ultrabasic rocks.

Reference.

Dufaud F. (2006) Etude expérimentale des réactions de carbonatation minérale du CO_2 dans les roches basiques et ultrabasiques. Unpublished PhD thesis, IPG, Paris.