

Experimental study of dissolution rates of some silicate minerals at high temperatures up to 400°C

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Steady-state dissolution rate measurements of pyroxene, wollastonite, actinolite and albite were carried out in the temperature (T) range from 25 to 400°C at 23 MPa. Minerals used in experiments are collected through microscope, cleaned and analyzed chemically. Most experiments were performed in mixed-flow reactors with Zr liner. Usually, all reactive solutions were undersaturated with respect to the mineral and the secondary product phases on the reacted surface.

The experiments suggest that the measured dissolution rates of the minerals at $T \leq 300^\circ\text{C}$ coincide with previous experiments published in literatures. The dissolutions are stoichiometric for those experiments, in which cases the release ratio of molar concentrations of metal M_i versus molar concentration of Si in outlet solutions $\Delta M_i/\Delta \text{Si}$ is identical to the stoichiometric number N_i in solid. The stoichiometric dissolution of albite in water is present at 300°C. Stoichiometric dissolution of pyroxene in water is at T near 200°C, and that of actinolite is at 300°C. Dissolution rates (r_{Si}) for albite, actinolite, pyroxene, wollastonite in water were found to increase with increasing T from 25 to 300°C, and then decrease with increasing T from 300 to 400°C. The maximum release rates of Si for those minerals are reached at 300°C. But $\Delta M_i/\Delta \text{Si}$ varies with T. The different metals of the minerals often behave the different release rates at a fixed temperature. Usually, the release rates of Na, Ca, Mg, Fe, Al of minerals are often higher than Si at $T < 300^\circ\text{C}$. In contrast, release rates of Si are higher than others at $T \geq 300^\circ\text{C}$.

Experiments prove that the hydrolysis of Si-O-Si bond and metal $M_i\text{-H}^+$ exchange reactions at $T < 300^\circ\text{C}$ are different with reactions at $T \geq 300^\circ\text{C}$, 23 MPa. At T range from 300 to 400°C and 23MPa, strong lowering water density and dielectric constant lead to break water hydrogen bond network, and decrease hydration of Si-O-Si bond, thus, dissolution rates of Si decrease.

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Mobility of iodine (^{129}I and ^{127}I) species in sediment columns from the Savannah River Site

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^{129}I is a major by-product of nuclear fission and is among the top three risk drivers for waste disposal at the Yucca Mt., Hanford, and Savannah River Sites (SRS). ^{129}I is of major concern because of its perceived mobility in the environment, excessive inventory, toxicity, and long half-life (~16 million yrs). Little research has been carried out at these facilities beyond simple monitoring of groundwater plumes. Different iodine species exhibit dramatically different mobility in aquatic and sediment environments as inorganic and organic species may be hydrophilic, atmophilic and biophilic. In this study, we conducted column experiments to investigate the sorption and transportation of iodine species using a sandy aquifer sediment from the SRS and deionized water with CaCl_2 to simulate the SRS ground water ionic strength. This study is unique in that we mimic ambient concentrations of iodide (10^{-8} to 10^{-7} M) in the experiments instead of using artificially high concentrations of iodide (≥ 0.1 mM/L) used in most laboratory analyses. Results demonstrate that iodine mobility varies greatly with iodine concentration, mostly due to covalent binding of iodine to organic carbon moieties in soils and aquifer sediments. At the ambient concentration, significant retardation of iodide in the sediment was found, while there was no retardation at the artificially high concentration of 0.1 mM/L. The data from this study supports findings from sediment and ground water slurry experiments on SRS samples [1] and Swiss Glattfelden samples (unpublished).

[1] Schwehr *et al.* (2009) *ES&T* **43**, 7258–7264.