Diffusion characteristics of dissolved Si in pore water at pH 5-7 and 11

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Diffusion of dissolved Si in pore water is one of the major factors controlling the rate of chemical weathering. The charge and polymerization degree of dissolved Si depend on pH and the concentration of Si (Fig. 1). To evaluate how the effective diffusion coefficient of Si ($D_{e,Si}$) is affected by the charge and polymerization degree and how $D_{e,Si}$ differs from D_e of K and Cl ($D_{e,K}$, $D_{e,Cl}$), through-diffusion experiments were conducted at 25 °C using two sandstone and one rhyolite. In this experiment, a rock sample is sandwiched between a diffusion source reservoir and a measurement reservoir, and D_e is determined by monitoring solute concentration in the measurement reservoir. Comparison of $D_{e,Si}$, $D_{e,K}$, and $D_{e,Cl}$ at pH 5-7 revealed that $D_{e,Si}$ were 2.3-3.0 times smaller than $D_{e,K}$ and $D_{e,Cl}$ [1]. This is largely because the diffusion coefficient of Si in free water is smaller than those of K and Cl by a factor of ~1.7 but might be also partly attributed to the precipitation of Si in the pores. While the dominant Si species at pH 5-7 is $Si(OH)_4$ (Fig. 1), at pH 11 the dominant species is $SiO(OH)_3^-$ when Si concentration is low and the fractions of multimeric species increase with increasing Si concentration. For Si source concentrations of ~0–20 mM, $D_{e,Si}$ values at pH 11 and pH 6 were similar [1]. $D_{e,Si}$ value at pH 11 decreased with increasing Si source concentration, suggesting that the diffusion coefficients of Si multimers were lower than those of monomer.

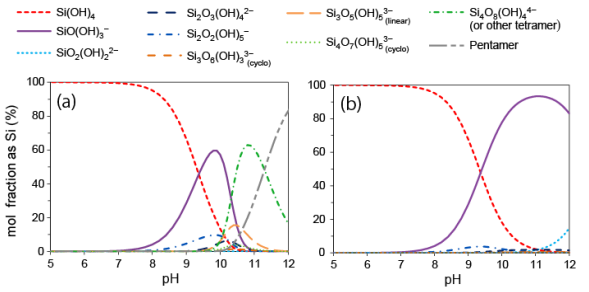

Figure 1: Speciation of dissolved Si vs. pH in equilibrium with amorphous silica (a) and at $[Si]_{total}$=2 mM (b), calculated using equilibrium constants reported in [2] (0.6 M NaCl).

[1] Yokoyama (2013) *Water Resour. Res.* **49**, 1-11. [2] Sjöberg *et al* (1985) *Acta Chem. Scand.* **A 39**, 93-107.