M-S-H precipitation via simultaneous dissolution of chalcedony

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In the cement-clay interface, precipitation of M-S-H (magnesium silicate hydrate) occurs as a result of the interaction between the high-pH pore water and the phases composing the interface. The M-S-H precipitation kinetics depends on the dissolution rate of the primary minerals (e.g. chalcedony, amorphous SiO_2) and the flow conditions.

To better understand the M-S-H precipitation, a two-step flow-through experiment was carried out. In the reactor, a surface of 1.6 cm^2 of a chalcedony fragment was allowed to react. The temporal variation of the aqueous chemistry was reproduced using The Geochemist's Workbench code.

In the first step, the chalcedony dissolution kinetics was studied injecting a 10^{-3} M NaOH solution (eventually adding Si up to 2.2×10^{-6} M) at 0.08-0.2 mL/min flow rate. The released Si was used to calculate the chalcedony dissolution rate.

In the second step (dissolution and precipitation), 5.8×10^{-6} M of Mg and 2.3×10^{-6} M of Si were added to the injected solution. The flow rate was initally diminished from 0.08 mL/min to 0.02 mL/min and thereafter increased to 0.12 mL/min. The Mg/Si ratios of the M-S-H phases were estimated from the calculated dissolution rate of chalcedony (first step) and the measured concentrations of Mg and Si of the output solutions.

At 0.08 mL/min, a deficit in aqueous Mg was attributed to precipitation of a M-S-H phase with high Mg/Si ratio (M-S-H(h)). However, at the lower and faster flow rates (0.02 and 0.12 mL/min, respectively), the formation of newly precipitates with a lower Mg/Si ratio seemed to occur. The Geochemist's Workbench calculations point out the possibility of newly precipitates with low Mg/Si ratios to grow on the reacting surface of the formed M-S-H(h). During this process the precipitation rate of M-S-H(h) decreases, and the precipitates with low Mg/Si ratio continue growing.