

Formation of hydrous Mg-silicates at low temperatures: New insights from sepiolite precipitation experiments

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The spatiotemporal changes in the distribution and abundance of hydrous Mg-silicates have been frequently used to reconstruct sedimentary facies in modern and past epicontinental seas and lakes, lacustrine settings and in marine environments; albeit the physicochemical conditions and the mineral-forming processes of hydrous Mg-silicates remain questionable. In this experimental study, sepiolite [$\text{Mg}_4\text{Si}_6\text{O}_{15}(\text{OH})_2 \cdot 6\text{H}_2\text{O}$] was precipitated from silica-doped seawater and silica-doped synthetic MgCl_2 -brines over a three months period at aqueous Si/Mg molar ratios ranging from 1:27.5 to 1:110, initial pH of 8.3 ± 0.03 at $25 \pm 1^\circ\text{C}$. The evolution of the solution chemistry and solid-phase composition was monitored using UV-vis spectroscopy, ICP-OES, XRD, ATR-FTIR and TEM analysis.

The reactive fluids were, at any time, undersaturated in respect to amorphous silica [$\text{SiO}_2 \cdot n\text{H}_2\text{O}$] and brucite [$\text{Mg}(\text{OH})_2$]; thus, a Mg-rich phyllosilicate with a modulated, sepiolite-like structure was the only precipitates in our experiments. The crystallites were poorly crystalline, fibrous (20 to 100 nm in length) and had a $(\text{MgO} + \text{Al}_2\text{O}_3)/\text{SiO}_2$ ratio of 0.44 ± 0.02 , which is almost equal to that of ideal and naturally-grown sepiolite. An increase in the intensity of the striking infrared lattice vibration at $\sim 1205 \text{ cm}^{-1}$ is in accord with an elevated Si/Mg molar ratio of the reactive solutions. This feature results from the periodic inversion of the Si tetrahedra in the evolving 2:1 layer and subsequently denotes the formation of “polysome units” in sepiolite-palygorskite group minerals. For the first time, we determined the apparent growth rate of sepiolite to be $172 \pm 16 \times 10^{-6}$ up to $279 \pm 29 \times 10^{-6} \text{ mole L}^{-1} \cdot \text{day}^{-1}$, which mainly depended on the evolution of pH of the reactive fluids. The presence of MgSO_4 aquo-complexes seems to have insignificant influence on the precipitation rate of sepiolite. Our results demonstrate that hydrous Mg-silicates can form in most (peri)marine and diagenetic environments, if sufficient time and an additional source of silicic acid is provided through, i.e. dissolution of marine silicifiers, volcanic ash and/or silicate detritus. We suppose that the low crystallinity degree of the incipient precipitates, the presence of reactive intermediates and the formation of polyphase products at low temperatures could mask the widespread precipitation of hydrous Mg-silicates in modern (marine) sediments.