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Mg-silicate hydrate formation via simultaneous MgO dissolution at pH 10 and 50 °C in an open system

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Magnesium silicate hydrate (M-S-H) is a low-crystalline phase formed at low temperature (< 100 ºC) in Mg-SiO_2-H_2O systems. In previous studies related to cement chemistry, M-S-H formation was investigated in a closed system (e.g. batch experiments) under conditions of low water/solid ratio and high solution saturation states with respect to M-S-H[1]. However, there are no experimental studies for the formation of this compound in open systems (e.g. flow-through experiments) that mimic the geochemical reactions under Earth surface conditions. This work aims to study the formation of M-S-H in an aqueous open system. To this end, MgO powder reacted at pH 10 and 50 ºC in two flow-through experiments, in which two solutions with different concentration of dissolved silica (0.15 mM and 1.5 mM) were injected. The aqueous chemistry of the outflow was monitored over time and the reacted solids were analyzed by XRD, SEM, TEM and ^{29}Si MAS NMR.

The steady-state output solutions were undersaturated with respect to Mg-hydroxide and supersaturated with respect to M-S-H. XRD patterns of the reacted solids did not reveal the presence of M-S-H. However, SEM images showed secondary silicate phases on the surface of the MgO particles. TEM and ^{29}Si MAS NMR analyses of the newly formed phases exhibited crystallographic characteristics similar to those of M-S-H synthesized in the previous study[1]. Moreover, Mg/Si ratios of 1.00 ± 0.09 and 1.59 ± 0.15 were obtained for the M-S-H formed in the high and low Si solutions, respectively.

Results demonstrated that: (i) M-S-H forms in an open system; (ii) the chemical composition of the M-S-H depends on the solution concentration, and (iii) in comparison with the low Si experiment, the high Si experiment shows lower saturation index with respect to Mg-hydroxide and higher saturation index with respect to M-S-H, suggesting that an increase in aqueous silica enhances Mg-(hydr)oxide dissolution and M-S-H precipitation. This work helps to improve our understanding of the geochemical reactions under field conditions in which dissolved silica coexists with Mg-bearing minerals at alkaline pH.

[1] Nied et al. (2016) Cem. Concr. Res., 79, 323–332.