Parameterization of a new C-S-H solid solution model for alkali uptake

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A new structurally consistent sublattice solid solution model of calcium-silicate-hydrate (C-S-H) [1] is proposed for describing the ion uptake, solubility, water content, and mean silicate chain length (MCL). Taking advantage of recent structural and atomistic views of C-S-H, the model considers two sublattice sites with substitutions of ionic moieties and vacancies.

Due to the complexity of multi-site solid solution models with many parameters to be adjusted at the same time, we used GEM-Selektor and GEMS3K codes [2] with TSolMod library of solution models [3] for calculation of equilibria in complex aqueous-solid solution systems, and the GEMSFITS code [4] for multiple parameter optimization.

In the first step, selected experimental datasets for solubility were used for parameter optimization covering the C-S-H sub-system. To improve the MCL fit to data derived from ^{29}Si NMR spectroscopy, we added MCL data to the objective function. For the C-S-H system, standard Gibbs energies of 6 end members, together with 4 regular site interaction parameters were simultaneously optimized against a large selection of Ca and Si partitioning data, as well as reported and interpolated MCL values.

In the second step, selected experimental datasets on the Na and K uptake in C-S-H were used to refine standard Gibbs energies of related end members and interaction parameters; while keeping constant those previously optimized for the C-S-H sub-system. Initially we assumed that K and Na can enter both sublattices, but the results show that alkalis most probably only enter IC (interlayer cation) sites. This reduces number of added end members from 6 to 3 for Na or K, and number of site interaction parameters from 5 to 2 per cation. Good fits also show that the model is indeed incremental and can be extended stepwise for aluminum and minor cations of interest for waste management.

The resulting internally consistent set of parameters can be used to model the uptake of Na and K in fully hydrated C-S-H at various Ca/Si from low to high Na/Si or K/Si ratios.

[1] Kulik et al. (2018), current abstract volume.
[2] Kulik et al. (2013) Computat. Geosci. 17, 1-24.
[3] Wagner et al. (2012) Canad. Min. 50, 1173-1195.
[4] Miron et al. (2015) Appl. Geochem. 55, 28-45.