A realistic three-site solid solution model of C-S-H

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Structurally consistent solid solution models of calcium-silicate-hydrate (C-S-H) are relevant in cement chemistry [1]. We present a new CNKASH3 sublattice solid solution (sss) model, capable of describing the solubility, non-gel water content, and mean silicate chain length (MCL) of C-S-H.

The model is built upon some new ideas on atomistic defects in 14Å tobermorite structure [2] and its non-gel water content [3]. For a fully-hydrated case, two sublattices with substituting moieties and vacancies are used; third sublattice is foreseen to model partially dehydrated systems. Main assumption is that charges acting on interlayer cationic (IC) sublattice sites originate in bridging tetrahedral (BT) sites due to deprotonated –OH groups, either on bridging silica or aluminate or on dimeric silica (if bridging ones are missing); on average, one negative charge per BT site. Thus, any end member is constructed using a template $DU:BT:IC:IW_2$: where DU is (a moiety on) a dimeric unit of tobermorite structure; IW is H_2O or vacancy on interlayer water site. Ca and Al can enter both BT and IC sites, whereas Na and K can enter IC sites and optionally BT sites. Moieties and vacancies on BT and IC sites are chosen such that the model provides a realistic non-gel water content at various Ca/Si ratios.

For the C-S-H sub-system, the model has 6 end members (EM) and 4 Berman regular interaction parameters (IP); it can be easily computed in GEM codes and parameterized using GEMSFITS [4]. This results in a very good fit to both solubility and MCL data for co-precipitated C-S-H, including refinement of logK of formation of $CaSiO_3$,aq complex. Thus obtained 11 parameters were fixed in the subsequent fitting. Extension of the model with K and Na required these cations in IC sites only to obtain good fits, hence, only 3 EMs and 2 IPs are added per alkali cation. Further extensions for Al (+8 EMs and +7 IPs) and minor cations can also be done incrementally. The CNKASH3 sss model performs in general better than any previous one (e.g. [5]) and can be efficiently used for hydrated ordinary, CSA, and blended cements.

[1] Kulik et al. (2011) Cem.Con.Res. 41, 477-495.
[2] Mohamed et al. (2018) Cem.Con.Res. 107, 221-235.
[3] Lothenbach et al. (2018), to be submitted.
[4] Miron et al. (2018), current abstract volume.
[5] Myers et al. (2014) Cem.Con.Res. 66, 27-47.