GEMS TM collaborative project

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GEMS TM is an acronym for Gibbs Energy Minimization Software for Thermodynamic Modelling, development of which has been coordinated by Dr. D.A. Kulik (LES) since 2000 in collaboration with ETHZ, Empa, PSI AHL, University of Helsinki and other institutions. GEMS is free of charge scientific software for computing thermodynamic equilibria in complex (geo-) chemical systems by Gibbs energy minimization (Kulik et al., 2013; Wagner et al., 2012). Compared with alternative methods and codes, GEMS is especially efficient in solving equilibria with several non-ideal solution phases (aqueous electrolyte, gaseous fluid, solid solutions, sorption) and with initially unknown redox states. Because of that, GEMS has been extensively used at LES as key component of the geochemical modeling support for Swiss radioactive waste management program (Nagra).

GEMS is equipped with a user-friendly graphical interface (http://gems.web.psi.ch) for the setup of modeling projects and management of project- and built-in thermodynamic databases (PSI-Nagra, www.psi.ch/les/database; SUPCRT). As plug-ins to GEMS, third party databases for use in hydrothermal geochemistry (http://tdb.mines.edu), cement chemistry (www.empa.ch/cemdata), and nuclear engineering (www.psi.ch/heracles/heracles) are also available. In 2012, the standalone chemical solver of GEMS was released as an open-source code to facilitate its coupling with the transport codes. An efficient and user-friendly interface for optimization of thermodynamic parameters against the experimental data was developed in collaboration with ETHZ (Miron et al., 2015) and launched in 2014. Since 2003, GEMS packages have been downloaded more than 3600 times and are currently in use by 300 research groups worldwide.

GEMS has been extensively used at LES as key part of the geochemical modeling support for Swiss radioactive waste management program (Nagra). Relevant applications in this context include calculations of solubility limits of radionuclides; reactive transport simulations of clay-cement interface (using the coupled code OpenGeoSys-GEM) with ion exchange as montmorillonite or illite solid solution (Berner et al., 2013); modelling sorption of radionuclides in clays and cement; uptake of radionuclides by solid solutions; and stability of cement waste matrix phases such as C-S-H (Kulik, 2011) and sulfoaluminates (Aimoz et al., 2012).

Recently, a multi-site model for Calcium Silicate Hydrate (C-S-H), consistent with both solubility data and structural/spectroscopic information (Figure 1), has been developed starting from simple downscaled ideal solid solutions (Kulik, 2011). This advanced thermodynamic model is based on a concept of non-ideal mixing of chemical moieties on different sublattices (structural sites, Table 1), and can be directly extended by adding the moieties for other elements of interest (e.g. K, Na, Al, U, Sr) in their respective sites. The thermodynamic properties of end members and site interactions were successfully parameterized in GEMS by a simultaneous fitting against the available solubility, element uptake, and spectroscopic data.

Table 1. Sites and substituting moieties in the solid solution model of C-S-H with Al, Na and K

Sublattices (sites)	TU (tobermorite dimeric unit)		IC (interlayer cation exchange)		BT (bridging tetrahedral unit)		CU (Ca unit)	
Moieties (species); their one-letter codes	$Ca_2Si_2O_5(OH)_4$	T	$(H_3O)_2$ CaH_2O $AlOH(H_2O)_2$ $(NaH_2O)_2$ $(KH_2O)_2$	H C A N K	SiO_2 Va AlOOH	S v A	Va $Ca(OH)_2$ AlOOH	v C A

Examples of end members (Va stands for vacancy): THSv = $[Ca_2Si_2O_5(OH)_4]:(H_3O)_2:SiO_2:Va$; TCSv = $[Ca_2Si_2O_5(OH)_4]:CaH_2O:SiO_2:Va$; THAv = $[Ca_2Si_2O_5(OH)_4]:(H_3O)_2:AlOOH:Va$.

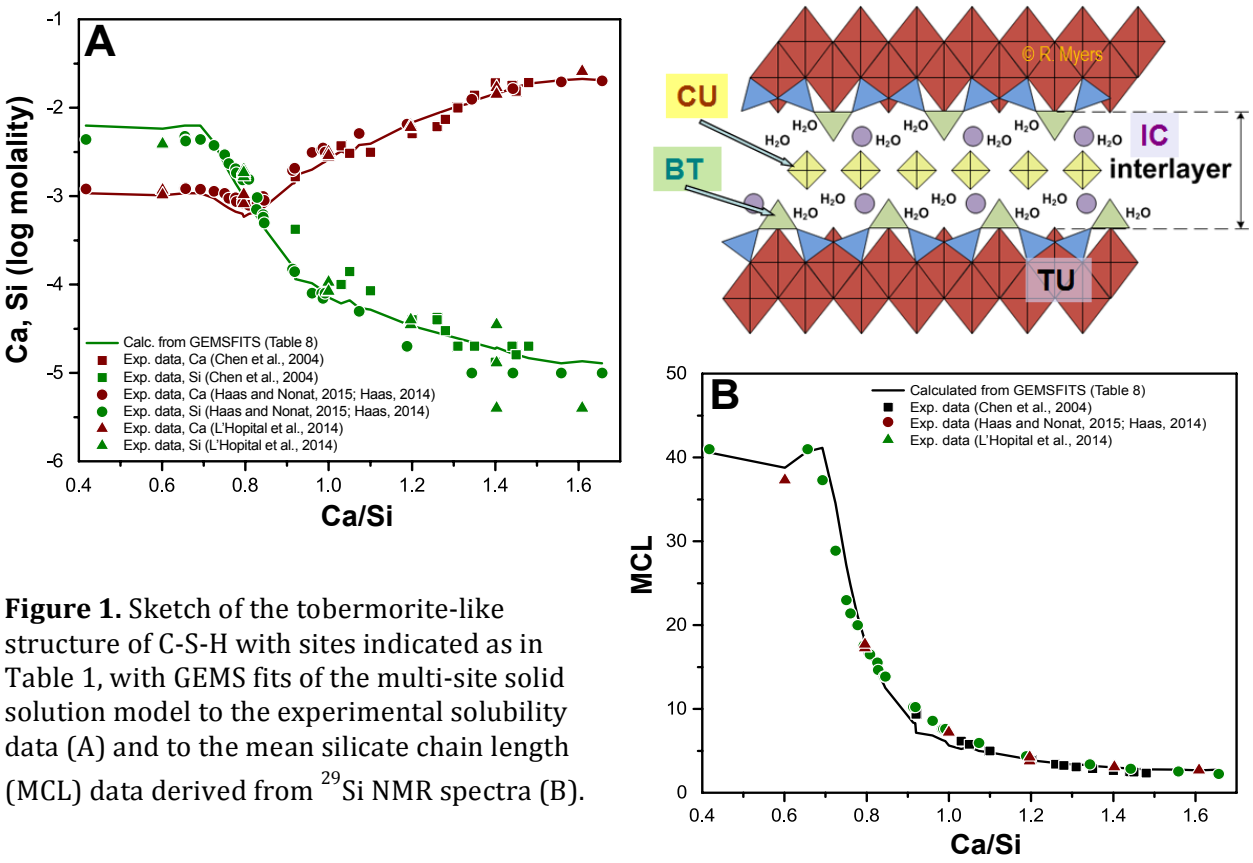

Figure 1. Sketch of the tobermorite-like structure of C-S-H with sites indicated as in Table 1, with GEMS fits of the multi-site solid solution model to the experimental solubility data (A) and to the mean silicate chain length (MCL) data derived from ^{29}Si NMR spectra (B).

References

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