A COMSOL Multiphysics® Software Interface With GEMS3K for Modeling (Geo)Chemical Reactive Transport Processes

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Introduction

- Application of reactive-transport (RT) models

- Two main type of RT models: (1) Fully Coupled and (2) Operator Splitting methods[1]: (THOUGHREACT, OpenGeySys-GEMS, CrunchFlow.)

- Previously, COMSOL was couple with PhreeqC[2].

[1] Xu et al. 2006, Steefel and Lasaga 1994, Koldiz et al. 2012
[2] Nardi et. al. 2014

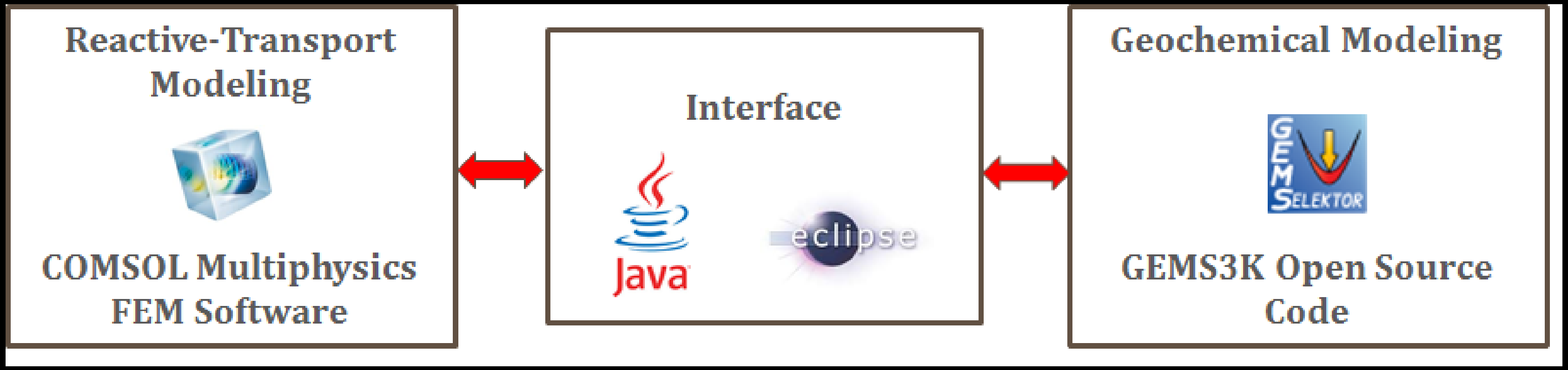
Introduction: GEMS PSI

- GEMS PSI: a Gibbs Energy Minimization (GEM) thermodynamic software for multi-phase multicomponent (geo)chemical systems[1]
- GEMS can work with built-in and formatted input thermodynamic databases (e.g. CEMDATA[2])
- The core methods are publically available in C++ (GEMS3K[3])

[1] Kulik et al. 2013, Wagner et al. 2012
[2] http://www.empa.ch/
[3] gems.web.psi.ch

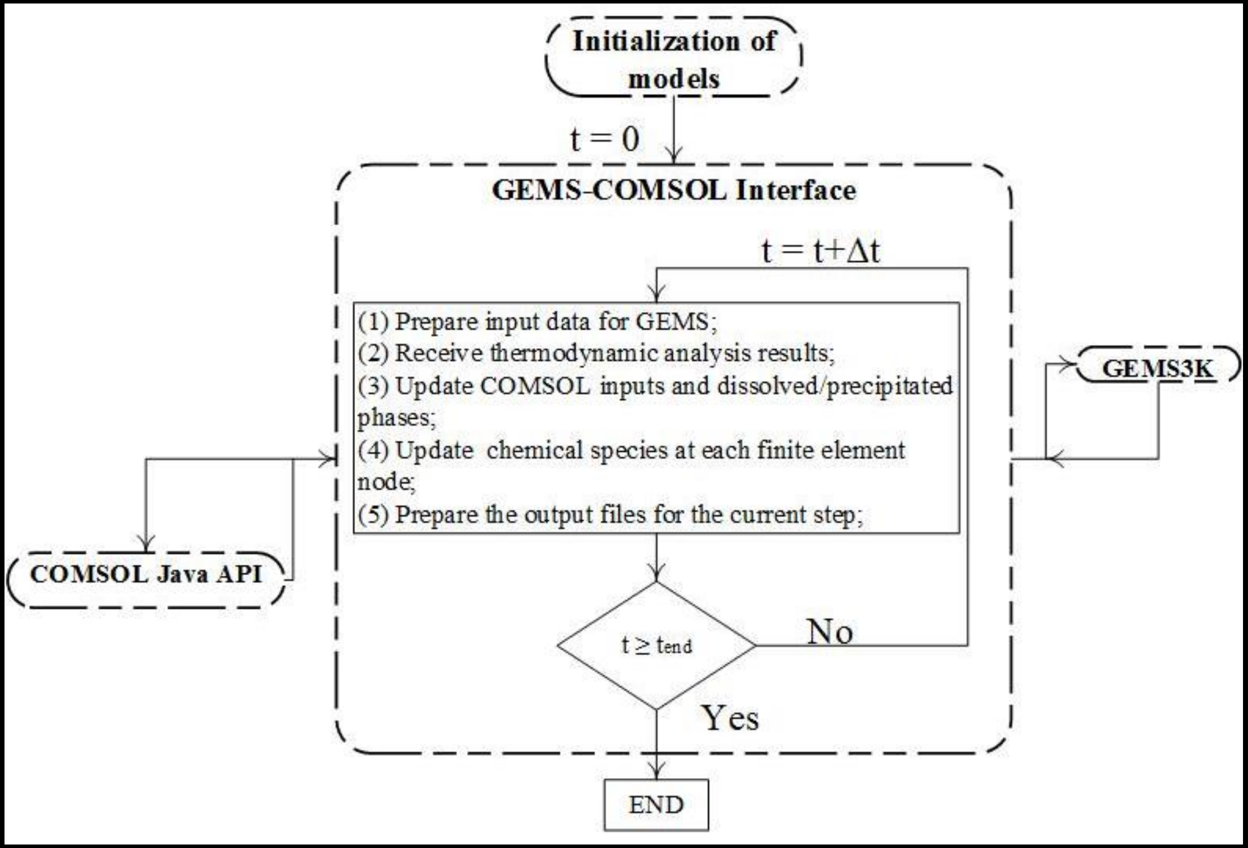
Developed Interface for RT analysis

- An interface was developed to connect COMSOL Multiphysics and GEMS3K.



[1] www.comsol.com

Interface Architecture[1]: initialization



[1] Jafari Azad et al. (2016), "Computers and Geosciences"

Cementitious system (geo)chemical models: theory

Mass balance → For atoms: Ca, Si, Na, K, Mg, S, Al, Fe, H, O, C and Cl

$$\frac{\partial c_i}{\partial t} = \frac{\partial\left[c_{s,i} + \varphi\, S_L\, c_{aq,i} + \varphi(1 - S_L)\, c_{G,i}\right]}{\partial t} = -\sum_{n_{DC}} \nabla \cdot J_{flow}$$

J_{flow} from Nernst-Planck equation.
S_L from the phase flow equations.
FCT solution techniques needed for advection.

Porosity change → based on solid volume changes

$$\varphi - \varphi_0 = -\sum_j \overline{V_j}\,(n_j - n_j^0)$$

Cementitious system (geo)chemical models: Nernst-Planck equation for flow

$$J_{flow} = \underbrace{-D\,\nabla c_{aq}}_{\text{diffusion}} \underbrace{-\,Dc_{aq}\frac{Fz}{RT}\nabla\emptyset}_{\substack{\text{electrical}\\ \text{migration}}} \underbrace{-\,Dc_{aq}\,\nabla\ln\gamma}_{\substack{\text{chemical}\\ \text{activity}}} + \underbrace{c_{aq}\,v_L + c_G\,v_G}_{\substack{\text{aqueous and gaseous}\\ \text{advection}}}$$

Diffusion[1] Base effective values in water are modified by temperature, tortuosity and porosity.

$$D_i = 2.9\times 10^{-4}\exp(9.95\varphi)\,r_\tau\,D_i^0$$

$$D_i^0 = \frac{k_b\,T}{6\pi\mu_L\,r}$$

$$r_\tau = \frac{1}{1+625(1-S_L)^4}$$

Cementitious system (geo)chemical models: Nernst-Planck equation for flow: cont.

Potential: $\nabla i = \sum_i z_i \nabla J_{flow,i} = 0$

No external potential

Activities: from GEMS3K

Debye–Hückel activity model was used in this work

Cementitious system (geo)chemical models: Phase flow and heat transfer

Phase flow → time dependent saturation degree profiles

$$(\varphi - \varphi_r)\,\frac{\partial S_L}{\partial t} = \nabla v_L \quad \rightarrow \quad v_L = \frac{k_{init}^{L}\, k_{rL}}{\mu_L}(\nabla P_L + \rho_L g\, \nabla h)$$

$$(\varphi - \varphi_r)\,\frac{\partial S_G}{\partial t} = \nabla v_G$$

Cementitious system (geo)chemical models: Phase flow and heat transfer: cont.

Velocity field → From porosity, viscosity and saturation

$$k_{init}^{L} = k_{init}^{L,0}\left(\frac{\varphi}{\varphi_0}\right)\left(\frac{1-\varphi_0}{1-\varphi}\right)^2$$

$$k_{rL} = \sqrt{S_L}\left(1-\left(1-S_L^{\frac{1}{\beta}}\right)^{\beta}\right)^2$$

$$P_C = \frac{-\rho_w RT}{\alpha M}\left(S_L^{\frac{1}{\beta}}-1\right)^{1-\beta}$$

$$v_L = \frac{k_{init}^{L}\, k_{rL}}{\mu_L}\left(\nabla P_L + \rho_L g\, \nabla h\right)$$

Cementitious system (geo)chemical models: Kinetics of dissolutions and precipitations

Cement phases in hydration → Based on nucleation, growth and diffusion, corrected with w/c, RH, surface area and temperature[1]

$$R_{phase} = \min\{R_{1,phase}, R_{2,phase,}, R_{3,phase}\}\, f_{w/c}\, \beta_{RH}\, \frac{A}{A_0} \exp\left(\frac{E_i}{R}\left(\frac{1}{T_0} - \frac{1}{T}\right)\right)$$

[1] Parrot&Killoh 1984, Lothenbach et al. 2006

Cementitious system (geo)chemical models: Kinetics of dissolutions and precipitations: cont.

Other minerals for dissolution[1] → $\frac{dm}{dt} = A \sum k_i \left(1 - \Omega^{p_i}\right)^{q_i}$

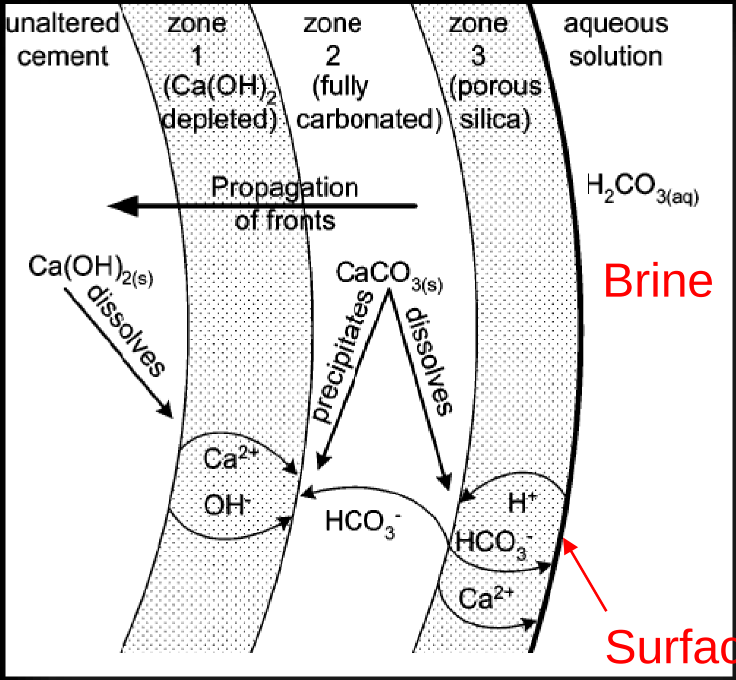
and precipitations

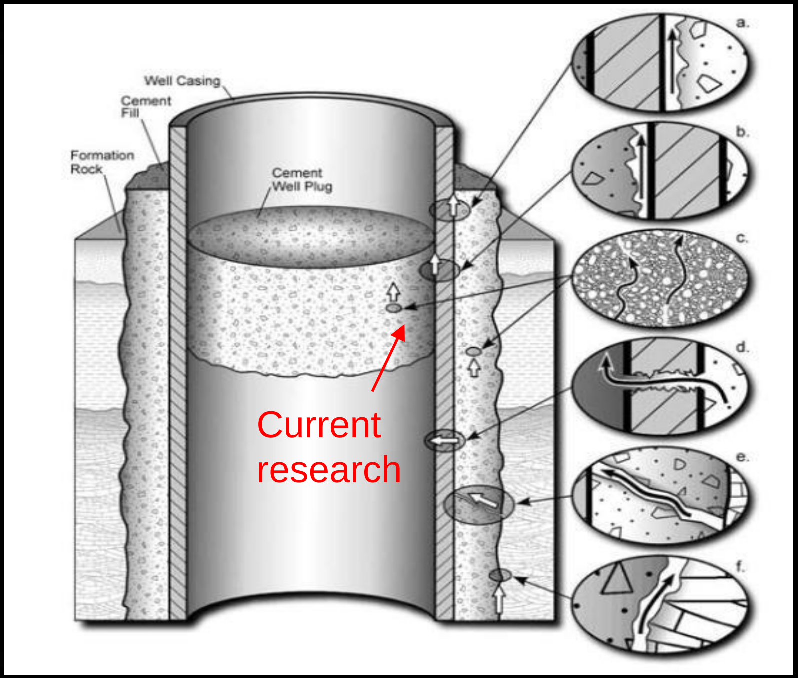
$$\frac{dm}{dt} = -A\left[k_{acid}\gamma_{H^+}{}^{n_1}\left(1 - \Omega^{p_1}\right)^{q_1} + k_{neutral}\left(1 - \Omega^{p_2}\right)^{q_2}\right.$$

[1] Lasaga 1984

Numerical Example:

Carbonation of wellbore cement samples in extreme downhole conditions



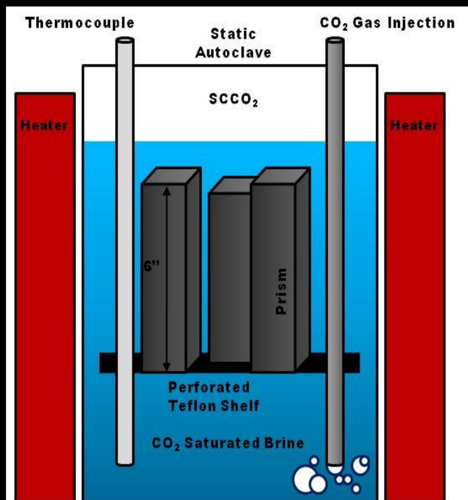


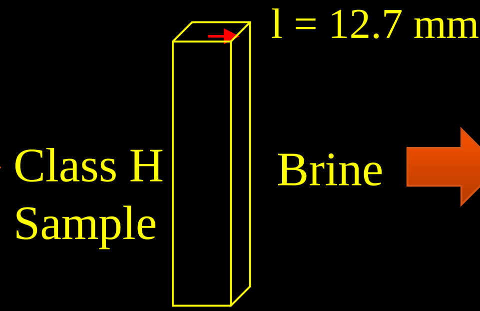
Schematic formation of distinct zones in the cement and Leakage pathways through an abandoned well[1,2]

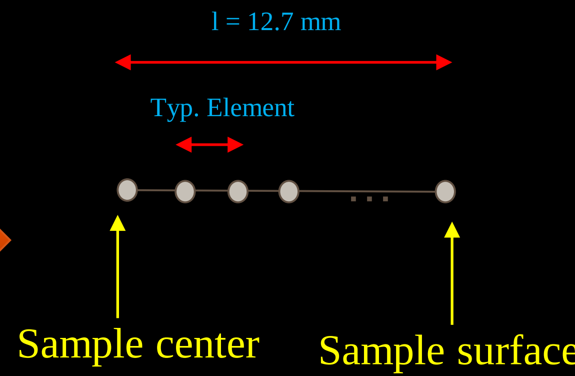
[1] Kutchko et al. 2007

2 Gasda et al., 2004

Case study: A one dimensional model was developed



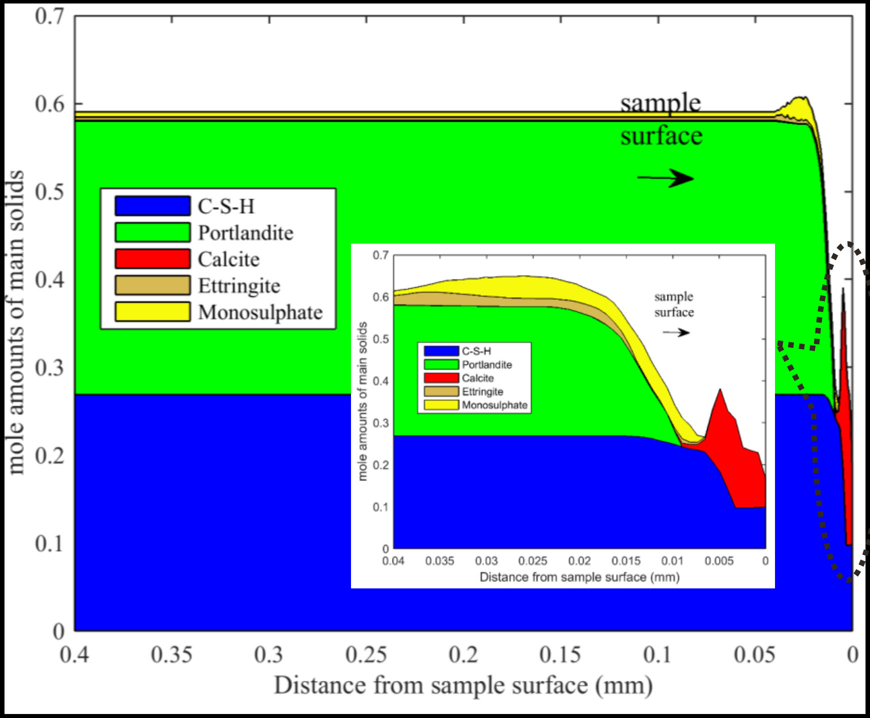




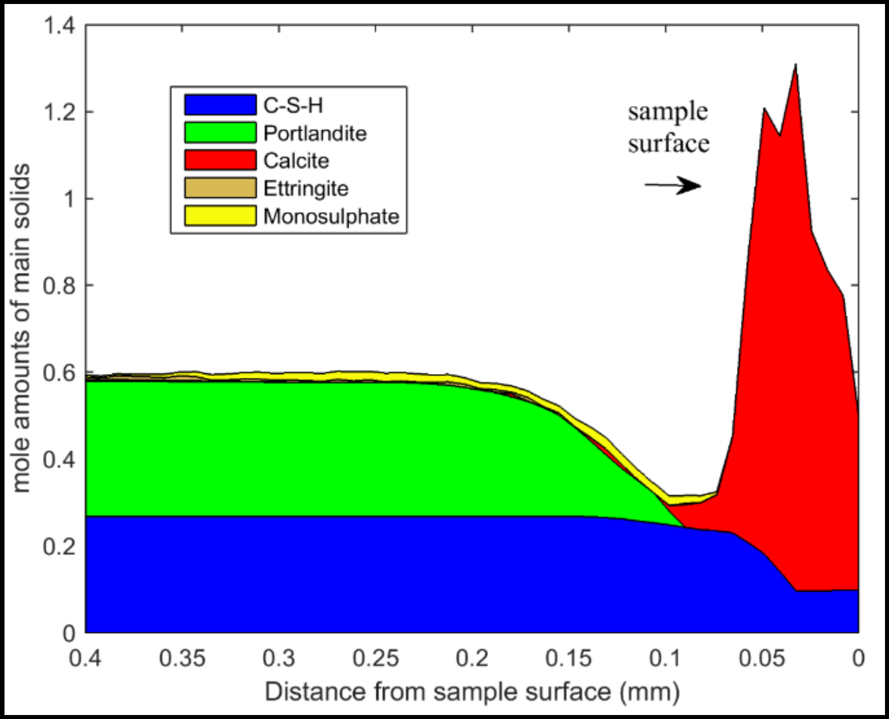
Synthetic Mt. Simon Brine | Class H cement | Typical 1-D model domain

Analysis case: High Temperature (85°C), CO_2 exposure;

Case 1: High Temperature, CO_2 exposure, Mt Simon Brine, P = 14.7 psi, T = 85 °C.



Day 0: alteration depth: 0.01 mm



42 days exposure: alteration depth 0.20 mm.

Concluding remarks:

- The developed GEMS-COMSOL interface can be applied for RT modeling with available database in GEMS format.

- More detailed information can be found in:

Vahid Jafari Azad, Chang Li, Circe Verba, Jason H. Ideker, O. Burkan Isgor, “A COMSOL–GEMS interface for modeling coupled reactive-transport geochemical processes,” Computers & Geosciences, Volume 92, July 2016, 79-89.

Acknowledgments

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Thank you