Coupling Multiphysics with Geochemistry

The Comsol-PhreeqC Interface: a Powerful Tool for Reactive Transport

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Introduction

PhreeqC [1] is one of the most widely spread, freely available geochemical computer programs for simulating chemical reactions.

In this work, an interface has been developed that combines the key capabilities of PhreeqC and Comsol in a single reactive transport (RT) simulator.

Implementation

The RT system of equations [8] is solved using the widely spread Sequential Non Iterative Approach (SNIA), which is based on the Operator Splitting concept. For pros and cons of SNIA see [6-7].

reactive transport equations

$$\boldsymbol{U}_a \frac{\partial(\phi\rho_l \boldsymbol{c}_a)}{\partial t} + \boldsymbol{U}_d \frac{\partial(\phi\rho_l \boldsymbol{c}_d)}{\partial t} + \boldsymbol{U}_m \frac{\partial((1-\phi)\rho_m \boldsymbol{c}_m)}{\partial t} = \boldsymbol{U}_a \boldsymbol{L}_l(\boldsymbol{c}_a) + \boldsymbol{U}\boldsymbol{S}_{\boldsymbol{k}}{}^{\boldsymbol{t}} \boldsymbol{r}_{\boldsymbol{m}}(\boldsymbol{c})$$

SNIA scheme

- *Conservative transport step*

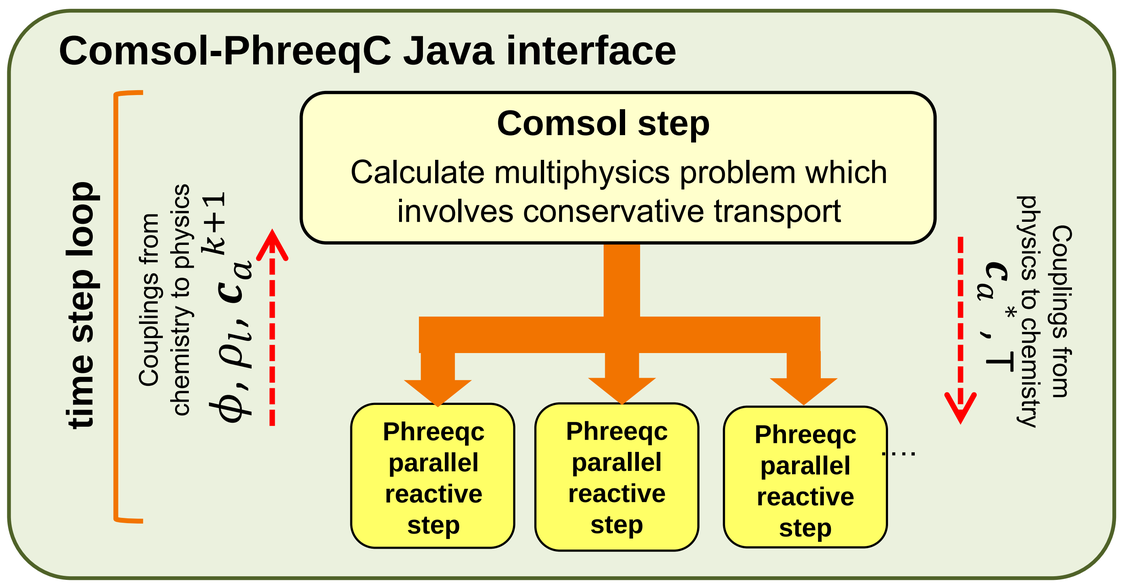
$$\boldsymbol{U}_a \frac{\partial(\phi\rho_l \boldsymbol{c}_a)}{\partial t} = \boldsymbol{U}_a \boldsymbol{L}_l(\boldsymbol{c}_a) \longrightarrow \boldsymbol{c}_a{}^*$$

- *Reactive step*

$$\boldsymbol{U}_a \frac{d(\phi\rho_l \boldsymbol{c}_a{}^*)}{dt} + \boldsymbol{U}_d \frac{d(\phi\rho_l \boldsymbol{c}_d)}{dt} + \boldsymbol{U}_m \frac{d((1-\phi)\rho_m \boldsymbol{c}_m)}{dt} = \boldsymbol{U}\boldsymbol{S}_{\boldsymbol{k}}{}^{\boldsymbol{t}} \boldsymbol{r}_{\boldsymbol{m}}(\boldsymbol{c})$$

Coupling details

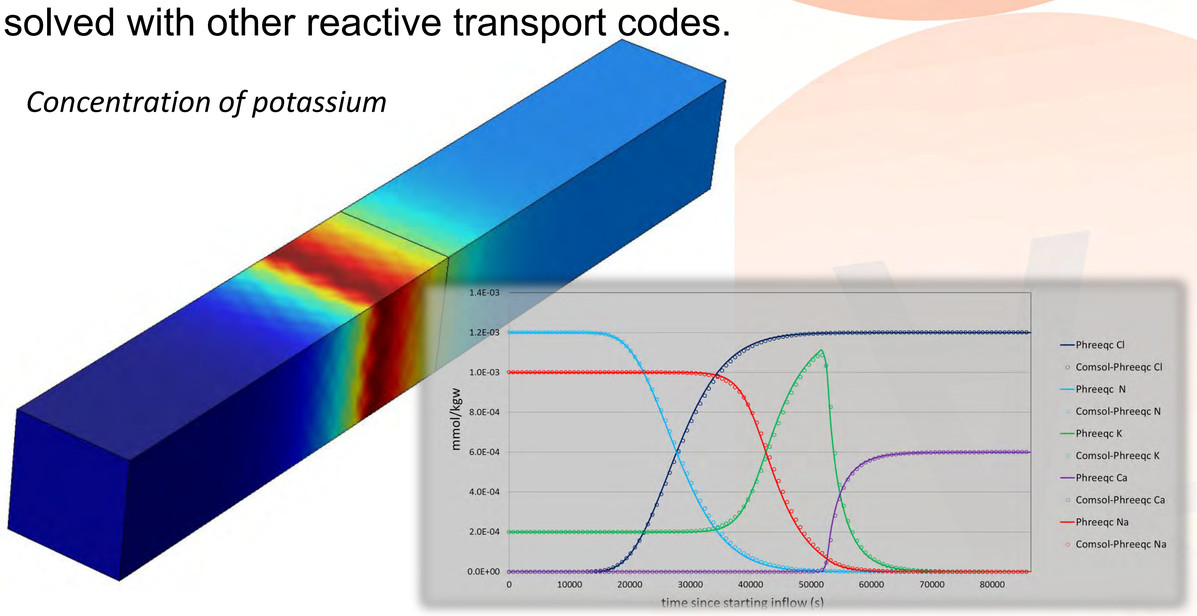
- Through a Java interface, it uses the Comsol API and the IPhreeqc dynamic library [5]
- The chemical step is parallelized over multicore processors



Code verification

The interface has been extensively verified by comparison of simulation results of 1D, 2D and 3D benchmark examples solved with other reactive transport codes.

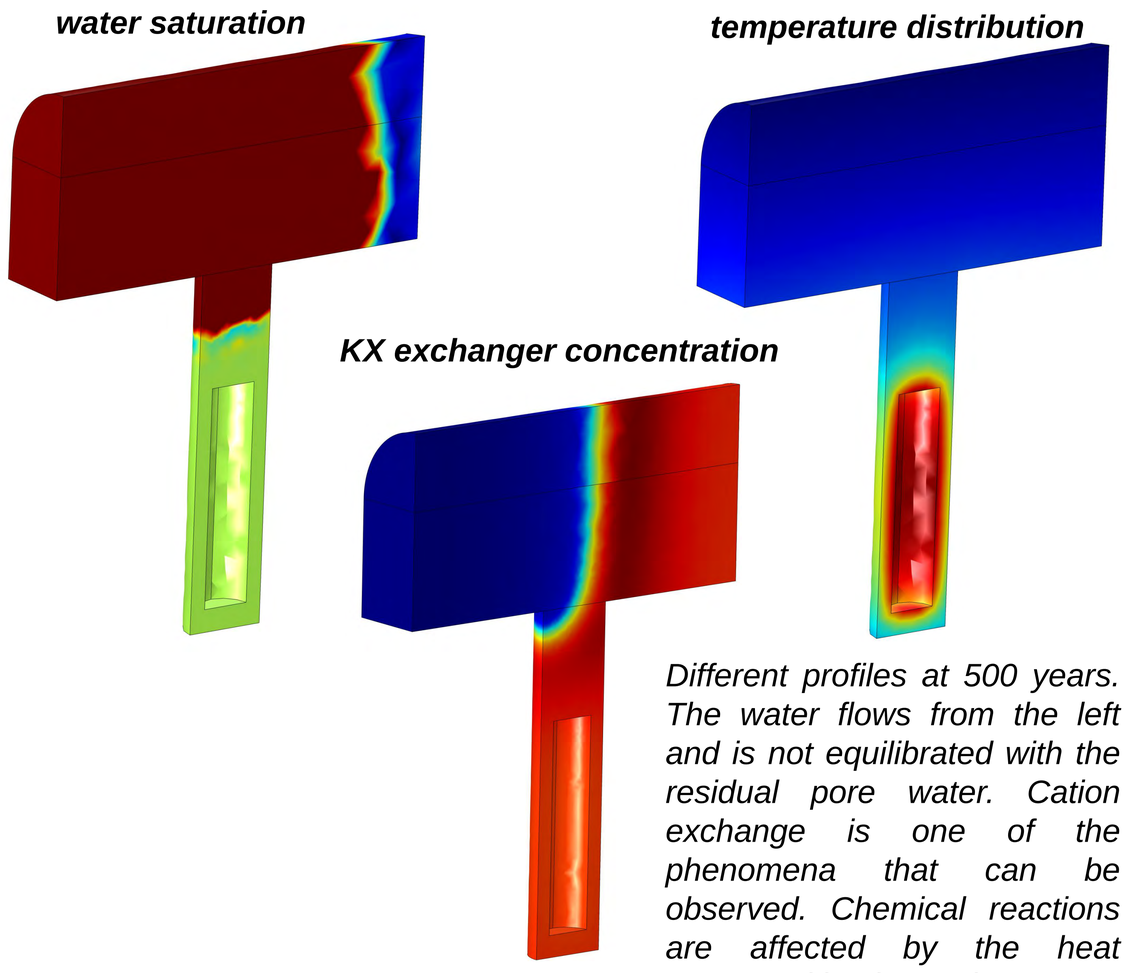
Concentration of potassium



Quantitative comparison of results between PhreeqC 1D RT and Comsol-PhreeqC Interface (using a 3D geometry): example number 11 of PhreeqC manual.

Application case

A 3D RTM in clay material has been simulated to illustrate the capabilities and the potential of the framework. It consists of the Thermo-Hydro-Chemical (THC) evolution of a deposition hole of a spent nuclear fuel repository. Simulations considering variably saturated flow coupled to chemical reactions and heat transfer have been performed to study the expected geochemical evolution of the clay barriers.



Different profiles at 500 years. The water flows from the left and is not equilibrated with the residual pore water. Cation exchange is one of the phenomena that can be observed. Chemical reactions are affected by the heat generated by the canister.

Conclusions

- The developed interface combines the key capabilities of PhreeqC (wide range of chemical reactions) and Comsol (flexibility to solve all kind of PDE systems)
- Substantial efforts have been devoted to enhance numerical performance to face highly coupled non-linear THMC simulations with a large number of elements (around 100k).
- The correctness of the interface has been verified using well-known benchmarks.

References

[1] Parkhurst D.L., Appelo C.A.J. (1999) User's guide to PHREEQC (v2) – A computer program for speciation, batch-reaction, one-dimensional transport, and inverse geochemical calculations: USGS Water-Resources Investigations Report 99-4259, 312 p. (freely available from USGS website)

[2] Parkhurst D.L. et al. (2004) Phast, a program for simulating ground-water flow, solute transport, and multicomponent geochemical reactions. USGS Techniques and Methods 6-A8, 154 p.

[3] Šimunek, J. et al., Multicomponent geochemical transport modeling using the HYDRUS computer software packages, *J. Am. Water Resour. Assoc.*, 42(6), 1537-1547, 2006.

[4] Wissmeier L., Barry D.A. (2011) Simulation tool for variably saturated flow with comprehensive geochemical reactions in two- and three-dimensional domains. *Env. Modelling & Software*, 26:210-218.

[5] Charlton S.R., Parkhurst D.L. (2010) Modules based on the geochemical model PhreeqC for use in scripting languages and reactive-transport calculations. *Computers & Geosciences*, 37:1653-1663.

[6] Barry D.A. et al. (1996). Temporal discretisation errors in non-iterative split-operator approaches to solving chemical reaction/groundwater transport models. *Journal of Contaminant Hydrology*, 22:1-17.

[7] Saaltink M.W. et al. (2001) On the behavior of approaches to simulate reactive transport. *Journal of Contaminant Hydrology*, 48:213-235.

[8] Saaltink M.W. et al. (2004) Retraso, a code for modeling reactive transport in saturated and unsaturated porous media. *Geologica Acta*, 2:235-251.

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