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OpenGeoSys-Gem: A numerical tool for calculating geochemical and porosity changes in saturated and partially saturated media

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ABSTRACT

Reactive transport codes that use a Gibbs Energy Minimization (GEM) to solve chemical equilibria are uncommon. We present a new coupling of the Richards flow module of the Finite Element (FE) based OpenGeoSys code with the GEM based chemical solver GEMS3K. The coupled code is highly parallelized using an overlapping domain decomposition approach in combination with execution of multiple threads that solve chemical equilibria in parallel. FE reactive transport schemes are often affected by spurious concentration oscillations. We effectively suppress these oscillations with a linearized algebraic flux corrected transport (FCT) algorithm. An application example is presented which investigates the evolution of material interfaces in a deep geological repository for nuclear waste. The example uses all features of the new coupled code: flow and multi-component transport in variably saturated media, and a very complex chemical setup which makes extensive use of (non-linear) solid solution formulations for mineral phases.

1. Introduction

(Bio-)Geochemical reactions in saturated and in partially fluid saturated porous media are important in various geoscientific and geoengineering fields. They influence transfer processes between atmosphere and groundwater, they are used for the remediation of contaminant spills, or they might influence high enthalpy geothermal systems. In this paper we present a numerical code which is able to simulate coupled reactive transport processes in partially saturated media without considering gas flow. An illustrative application example is shown that investigates the geochemical changes in a partially saturated deep geological repository for radioactive waste.

Relatively well-developed concepts for transport by diffusion or by advection or both in fully saturated media are currently available (see e.g. Steefel and Maher, 2009). If water is partly replaced by a gas phase, however, we are faced with a more complex system because we have to deal with two phases (fluid and gas) which occupy variable portions of the pore space. The actual gas/fluid ratio is itself a complex function of, for example, pore size distribution, pressure, temperature, external and internal sinks/sources for gas and water. Saturation does significantly affect the transport of solutes. It is well known that the diffusive and advective transport of solutes is significantly slowed down (Martys, 1999; Revil and Jougnot, 2008) in partially saturated media.

In the last decades several numerical codes have been developed that can solve multi-component transport in partially saturated media, for example FLOTRAN (Hammond et al., 2007), TOUGHREACT (Xu et al., 2011), CRUNCH (Steefel et al., 2005), HYDROGEOCHEM (Yeh et al., 2009), MIN3P (Mayer et al., 2002), CORE2D (Yang et al., 2008) or COMSOL-PHREEQC (Wissmeier and Barry, 2010). The majority of the codes can solve the coupled equations for gas and liquid flow, whereas some are restricted to one fluid phase and solve only the Richards flow equation. All the above reactive transport codes use chemical solvers that are based on the law of mass action (LMA) approach. The LMA approach is fast, accurate and relatively easy to implement. For the geochemical simulation of realistic, multi-component, multiphase systems, which include for example multiple solid solutions, chemical solvers based on the Gibbs Energy Minimization (GEM) method are advantageous (Kulik et al., 2013). In order to use the advantages of the GEM method, the open source, object-oriented Finite Element (FE) simulator OpenGeoSys (Kolditz et al., 2012a) was coupled to the open source numerical kernel of the GEM-Selektor code (GEMS3K) (Shao et al., 2009a,b), the commercially available solver ChemApp (Xie et al., 2011), the Biogeochemical Reaction Network Simulator BRNS (Centler et al., 2010). OpenGeoSys provides several modules for modeling kinetic reactions (Beyer et al., 2006).

In this work we describe an extension of the coupling between OpenGeoSys and GEMS3K in order to calculate reactive transport in partially saturated media in combination with the possibility to use a complex geochemical model.

The numerical solution of advection–diffusion–reaction equations with Finite Element (FE) methods is often affected by

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spurious oscillations in the transport equations. Due to the strong coupling between solute concentrations and mineral reactions, such oscillations might cause the accumulation of precipitates (or enhanced dissolution of minerals). Generally, such problems are related to advection dominated systems (John and Schmeyer, 2008), but also the artificial precipitation of minerals at a dissolution front was observed for equilibrium mineral reactions in a diffusion dominated transport regime (Perko et al., 2013). Such numerical artifacts should be avoided especially in applications that have a complex spatial and temporal transport regime, which is strongly coupled to a complicated geochemical setup via multiple mineral dissolution/precipitation reactions that change porosity and transport parameters.

We therefore added a flux correction scheme that suppresses effectively the accumulation (or reduction) of minerals introduced by the concentration oscillations from the FE method.

The applicability of the coupled code is demonstrated with the help of a simplified application example related to the geochemical and porosity evolution in the Eingineered Barrier System (EBS) of a deep geological repository for low- and intermediate level waste. Recent literature reviews and numerical simulations tailored specifically to the Swiss disposal concepts for radioactive waste (Savage, 2010, 2011; Savage et al., 2010; Berner et al., 2013; Kosakowski and Berner, 2013; Bradbury, 2013; Kosakowski et al., 2013) indicate that long-term interactions between cement and clay containing materials under fully water saturated conditions may lead to localized precipitation of minerals which clog the pore space. A widespread occurrence of porosity clogging in the EBS of a deep geological repository could influence the functionality of the system by decreasing porosity and permeability. The application gives indication, that the spatial and temporal evolution of porosity near such interfaces is strongly influenced by several competing processes, the transport regime, the water saturation and the kinetic control of precipitation/dissolution reactions.

2. Approach and code description

The OpenGeoSys code, which is used to solve the flow and transport equations, is a general purpose FE code. Verification and application examples for single and coupled thermal–hydraulic–mechanical–chemical (THMC) processes can be found in Kolditz et al. (2012b). For the chemical calculations the numerical kernel GEMS3K of the chemical solver GEM-Selektor v3 (Kulik et al., 2013) was utilized. The general coupling between both codes for fully fluid saturated media is described in Shao et al. (2009a,b). Here we only describe the newly implemented coupling to the Richards flow module of OpenGeoSys.

The Richards flow equation is implemented in OpenGeoSys to describe fluid flow in partially saturated porous media (Wang et al., 2010):

$$\phi\rho_f\frac{\partial S}{\partial p_c}\frac{\partial p_c}{\partial t}+\nabla\cdot\left(\rho_f\frac{k_{rel}\mathbf{k}}{\mu_f}(\nabla p_f-\rho_f\mathbf{g})\right)=Q_f. \qquad (1)$$

With porosity ϕ (–), fluid density ρ_f (kg m^{-3}), saturation S (–), capillary pressure $p_c = -p_f$, fluid pressure p_f (Pa), time t (s), gravitational acceleration vector $\mathbf{g}$ (m s^{-2}), relative permeability k_{rel} (–), intrinsic permeability tensor $\mathbf{k}$ (m^2), fluid viscosity μ_f (Pa s), and source term Q_f (kg s^{-1}).

Different relations between capillary pressure and saturation are implemented in OpenGeoSys. We use the following formulation after Mualem (1976) and Van Genuchten (1980):

$$p_c=\frac{\rho_f g}{\alpha}\left(S_e^{-1/m}-1\right)^{(1-m)}, \qquad (2)$$

with $m = 1 - (1/n)$ where n is a dimensionless pore size distribution index and α $(1/m)$ the van Genuchten parameter, which is related to the gas entry pressure. The effective saturation S_e is:

$$S_e=\frac{S-S_r}{S_{max}-S_r}, \qquad (3)$$

where S_{max} and S_r are the maximum and residual saturation, respectively.

The relative permeability k_{rel} for the fluid phase is related to the effective saturation S_e by

$$k_{rel}=S_e^{0.5}\cdot\left[1-\left(1-S_e^{\frac{1}{m}}\right)^m\right]^2. \qquad (4)$$

The capillary pressure–saturation and the permeability–saturation relations are empirical relations that depend on the pore size distribution and the shape of the pores in a specific medium. These relations are normally determined experimentally, or have to be obtained by upscaling procedures (see e.g. Toussaint et al., 2012; Gray et al., 2013).

In the coupled code the transport of dissolved chemical species is described by the commonly used advection–dispersion (–diffusion) equation (ADE) (Bear and Bachmat, 1990) which is supplemented by source/sink terms originating from chemical reactions.

$$\begin{aligned}\frac{\partial SC_i}{\partial t}&=-\nabla(S\mathbf{v}C_i)+\nabla(S\mathbf{D}_i\nabla C_i)+Q_i\\ \frac{\partial SC_i}{\partial t}&=\Gamma_i(C_1\ldots C_m)\end{aligned}. \qquad (5)$$

C_i (mol m^{-3}) stands for the molar concentration of species i, $\mathbf{v}$ (m s^{-1}) for the pore velocity in the fluid phase, $\mathbf{D}_i$ (m^2 s^{-1}) for the dispersion tensor of species i, Q_i (mol m^{-3} s^{-1}) for a source/sink term and $\Gamma_i(C_1\cdots C_m)$ (mol m^{-3} s^{-1}) is a source/sink term for species i due to chemical reactions with m other species. The Scheidegger dispersion tensor is implemented in two dimensions as

$$\mathbf{D}_{kl}=\alpha_T|\mathbf{v}|\delta_{kl}+(\alpha_L-\alpha_T)\frac{\mathbf{v}_k\mathbf{v}_l}{|\mathbf{v}|}+D_e. \qquad (6)$$

where α_L (m) and α_T (m) are the longitudinal and transversal dispersion length, respectively. δ_{kl} (–) is the Kronecker symbol, $v_{k,l}$ (m s^{-1}) is the fluid pore velocity in direction k,l and D_e (m^2 s^{-1}) is the effective diffusion coefficient. In our implementation the effective diffusion coefficient is calculated based on an Archie relation with exponent b and a pore diffusion coefficient D_p (m^2 s^{-1})

$$D_e=(\phi\cdot S_e)^b\cdot D_p. \qquad (7)$$

The chemical system is solved with the numerical kernel GEMS3K of the GEM-Selektor package (Kulik et al., 2013). The overall coupling employs the sequential non-iterative coupling scheme as described in Shao et al. (2009a). After solving the flow Eq. (1) and several transport Eq. (5), GEM-Selektor calculates the thermodynamic equilibrium of the system at each FE node taking into account the changed solute concentrations. In contrast to law of mass action (LMA) based chemical solvers, GEMS3K does not work with (solid and solute) concentrations. GEMS3K uses a Gibbs Energy Minimization (GEM) algorithm and requires as input the absolute amount of all independent components (base species). From this it calculates the complete liquid/gas/solid phase composition. Fluid saturation and solute concentration changes are therefore first transformed into appropriate changes of the independent components. After the new solution is provided by GEMS3K, the new fluid phase composition in terms of solute concentrations is passed to the OpenGeoSys transport solver. Eventual changes of porosity due to precipitation/dissolution of mineral phases are calculated and the porosity in the OpenGeoSys part is updated. In addition, changes in the volume of the fluid phase are back-coupled to the flow equation for the next time step via source-sink terms.