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Advances in Modelling Salt Stock Deposition of Nuclear Wastes

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GRS, Braunschweig, Germany

Content

- Introduction
- Reactive Transport Modelling
- Reactions and Database
- Application – UO2(s) dissolution and transport
- Outlook

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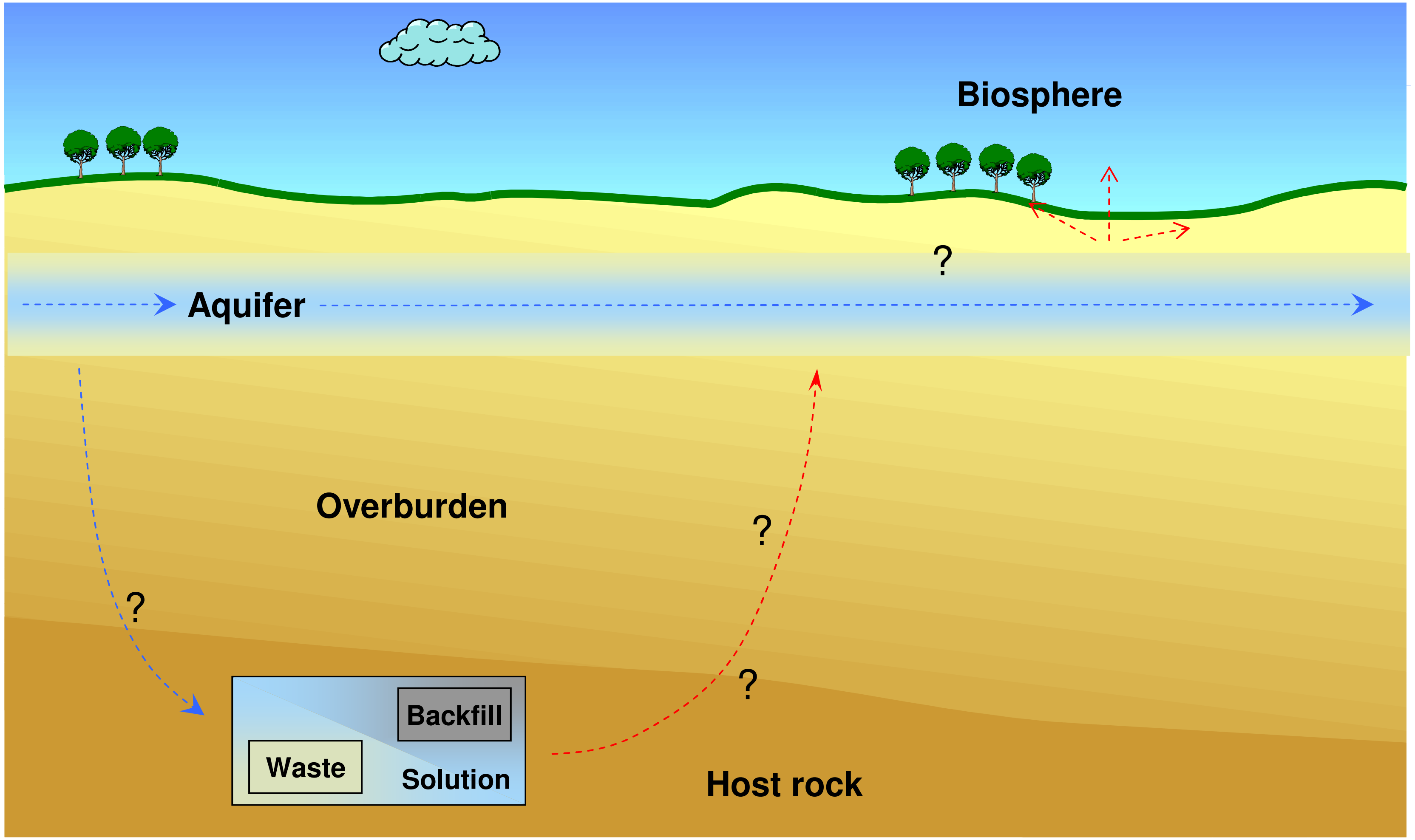
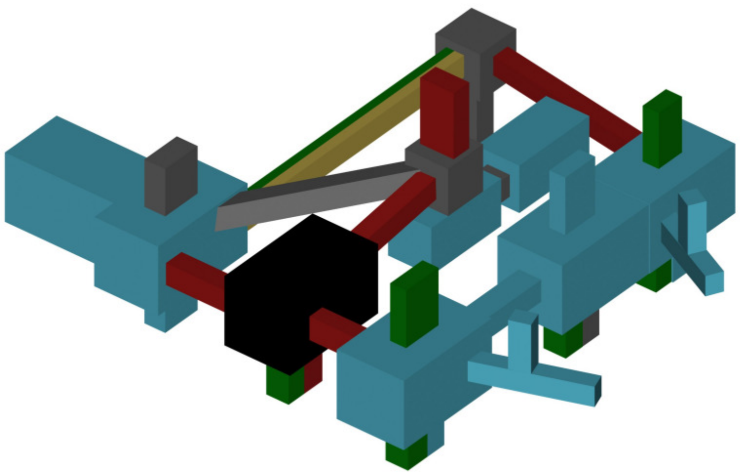

Biosphere
?
Aquifer
Overburden
?
?
?
Backfill
Waste
Solution
Host rock

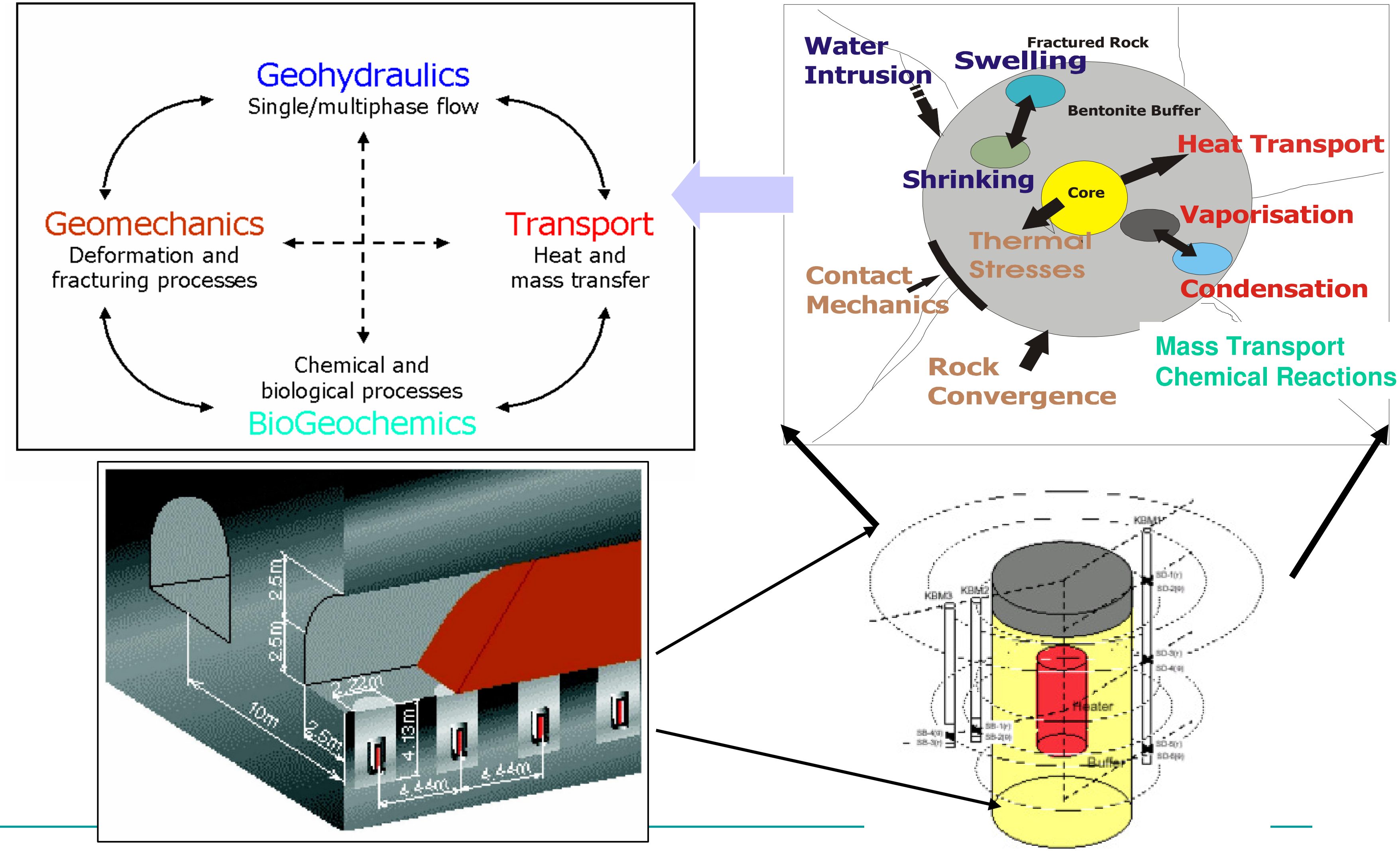
Of course, a real underground structure of a disposal site is much more complex…
This is just ONE base of an underground disposal site

- **Red** = Barriers
  Green = solid salt
  Blue = crushed salt
  Black = Disposal cavern
  Grey = Excavation-disturbed zone

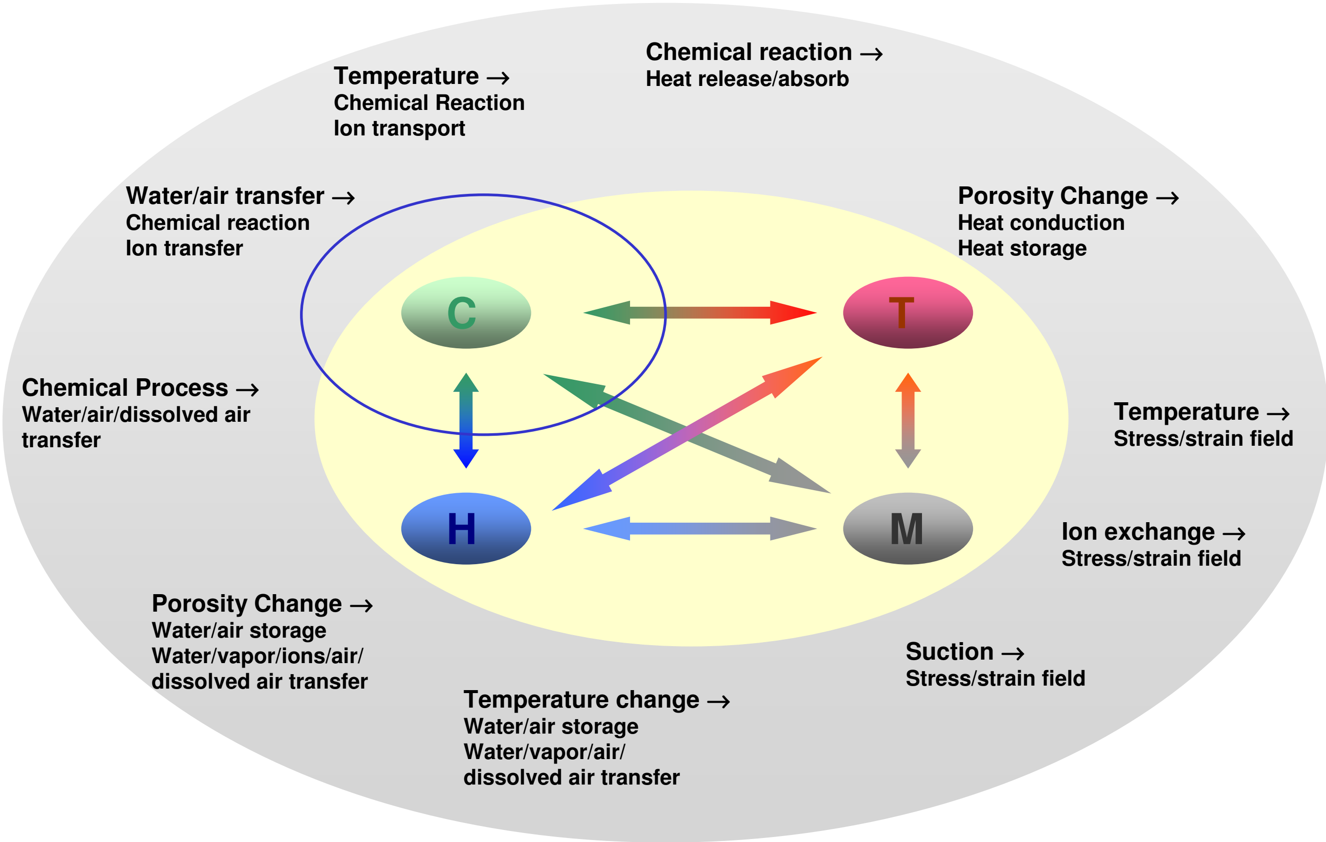
- Up to 15 bases possible



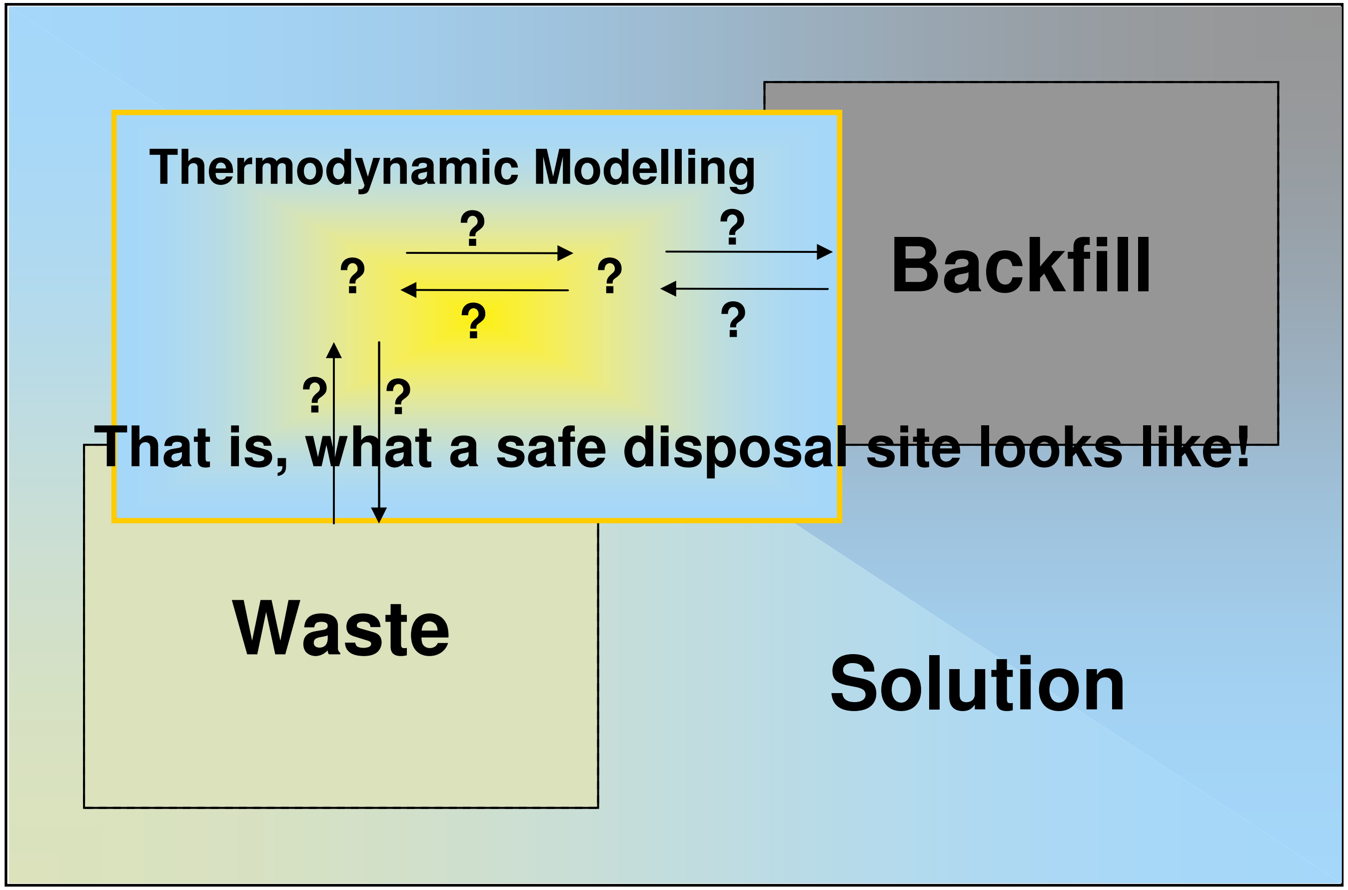
Coupled Processes in Geotech Systems



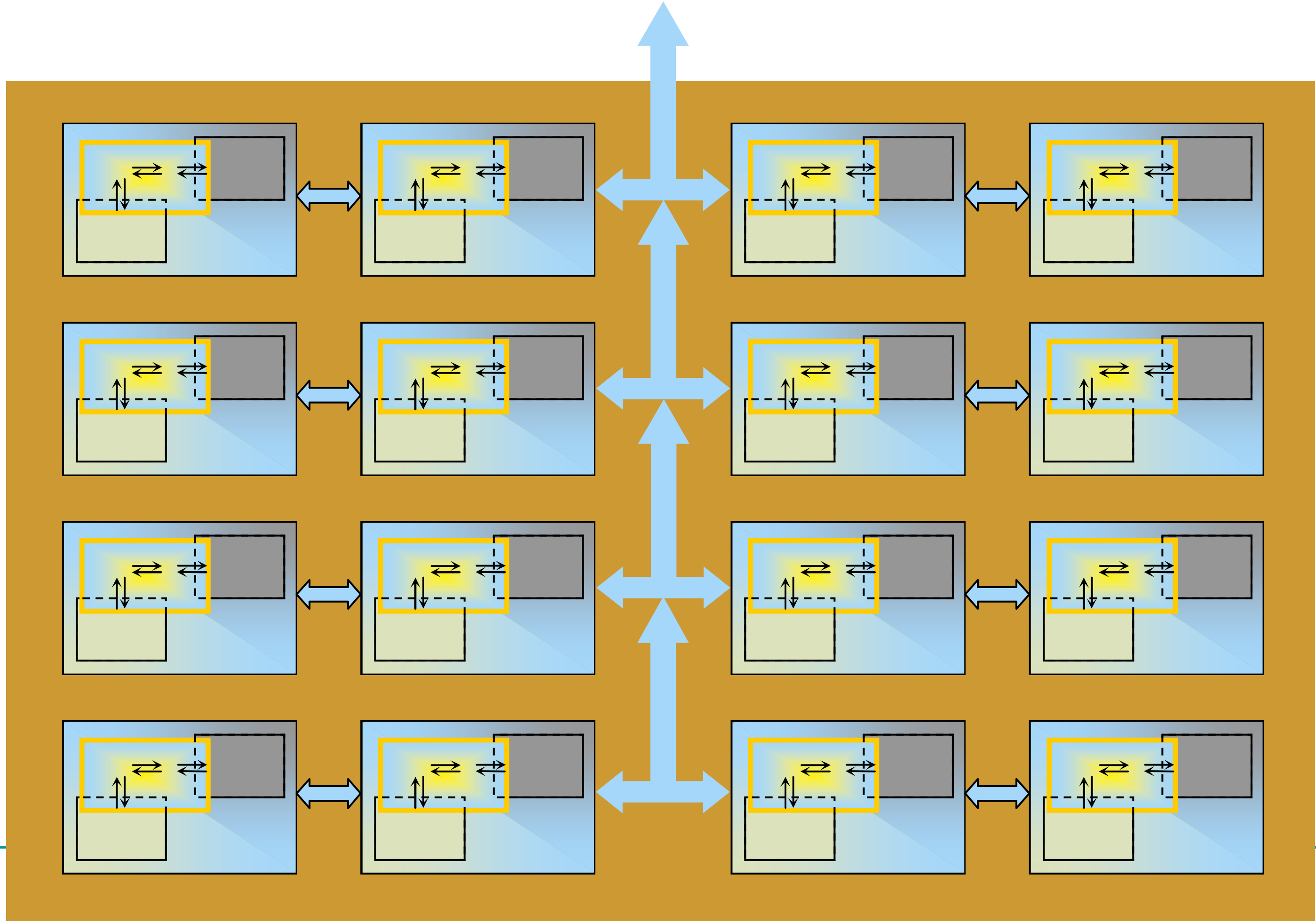
Concept of Coupled Processes



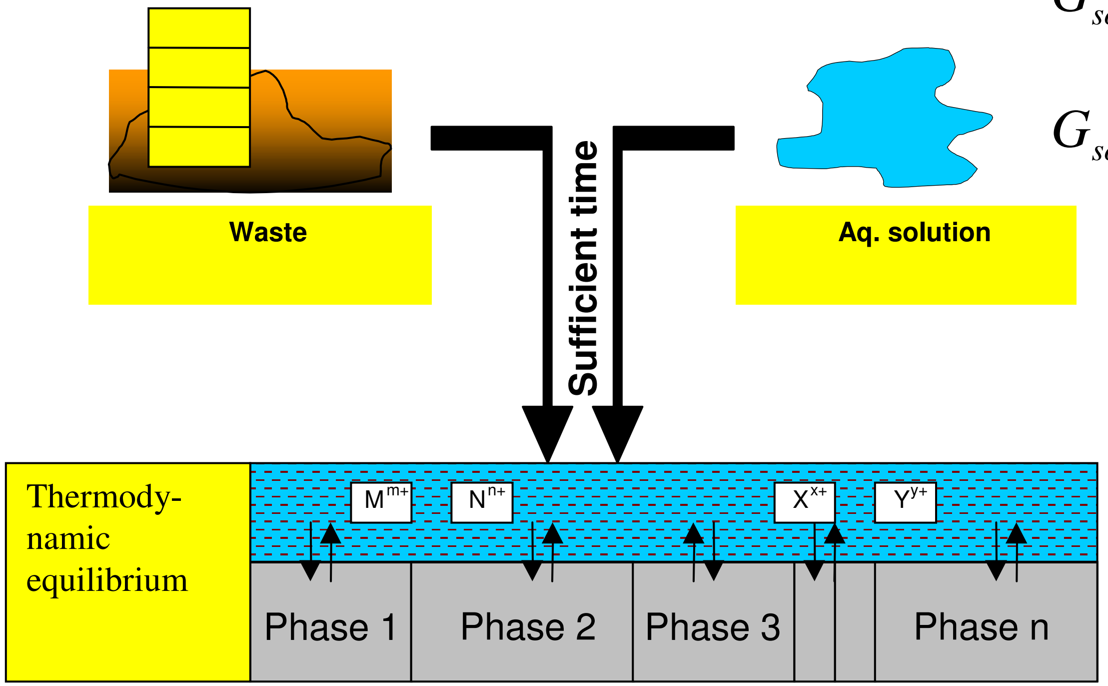
Disposal chamber for waste



Many caverns form one system…



Interaction Waste - Solution



Phasecomposition:

$$G_{solids} = \sum_{Phases\ i} n_i \cdot G^{\otimes}_{f,i}$$

$$G_{solution} = \sum_{i} n_i \left(G^{\otimes}_{f,i} + RT\left(\ln m_i + \ln \gamma_i\right)\right)$$

Equilibrium

$$K = \prod_j a_j^{\nu_j} = \prod_j m_j^{\nu_j} \cdot \gamma_j^{\nu_j}$$

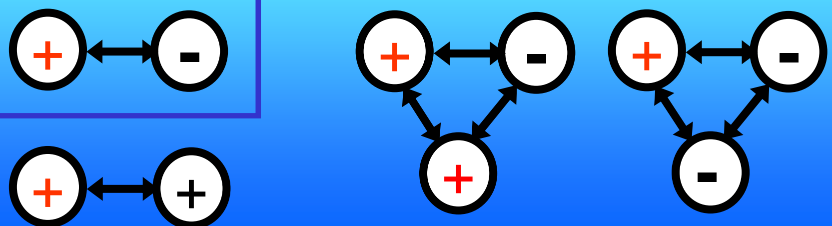
Modells for the calculation of activity coefficients

- Debye-Hückel (1923): purely electrostatic interactions
- Extensions to the DH-equation…
- Approaches considering specific ionic iteractions …

$$\ln \gamma_i = DH \quad + \quad \sum_j \lambda_{i,j}(I) m_j \quad + \quad \sum_j \sum_k \mu_{i,j,k} m_j m_k$$

SIT -
Single Ion
Interaction Theory

Pitzer-Modell



- Material microstructure $\longrightarrow n_0$
- Dissolution/precipitation (C)

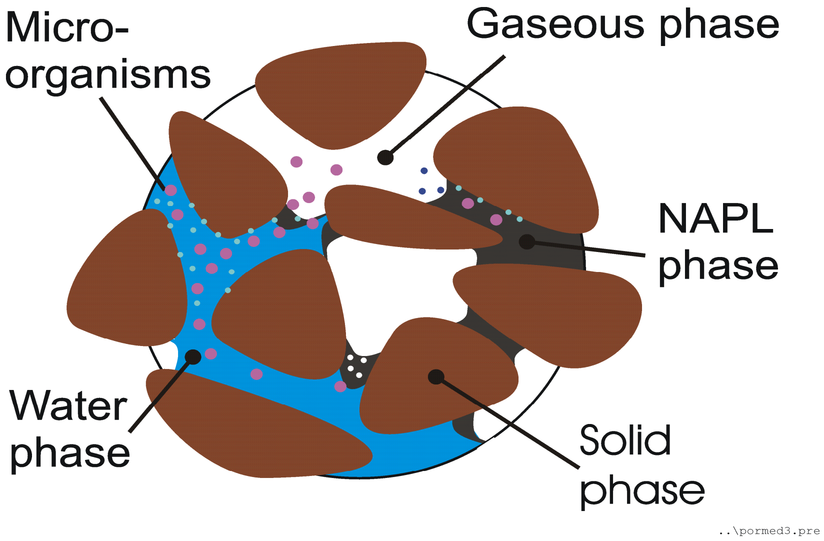
- $$n = n_0 - \sum_{j=1}^{nm} V_j M_j$$

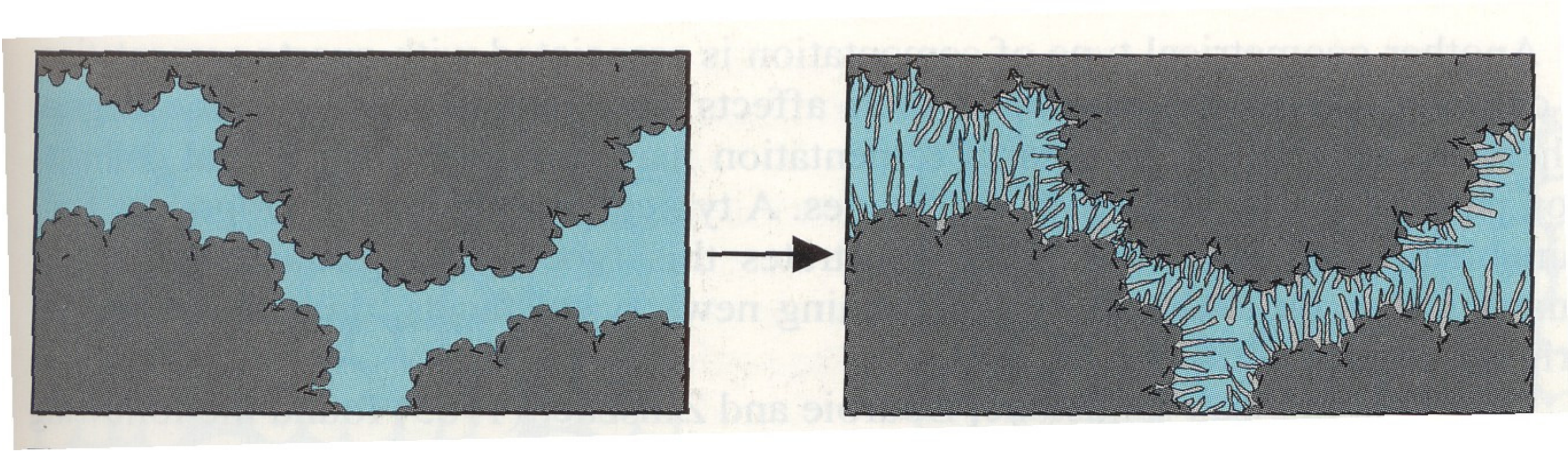
- Swelling (bentonite)

$$n = n(\epsilon, \text{other factors})$$

- Permeability k=k(n)

$$n_{\mathrm{IL}} = (S^l)^\eta \cdot \beta \cdot n_{\mathrm{IL}max}$$

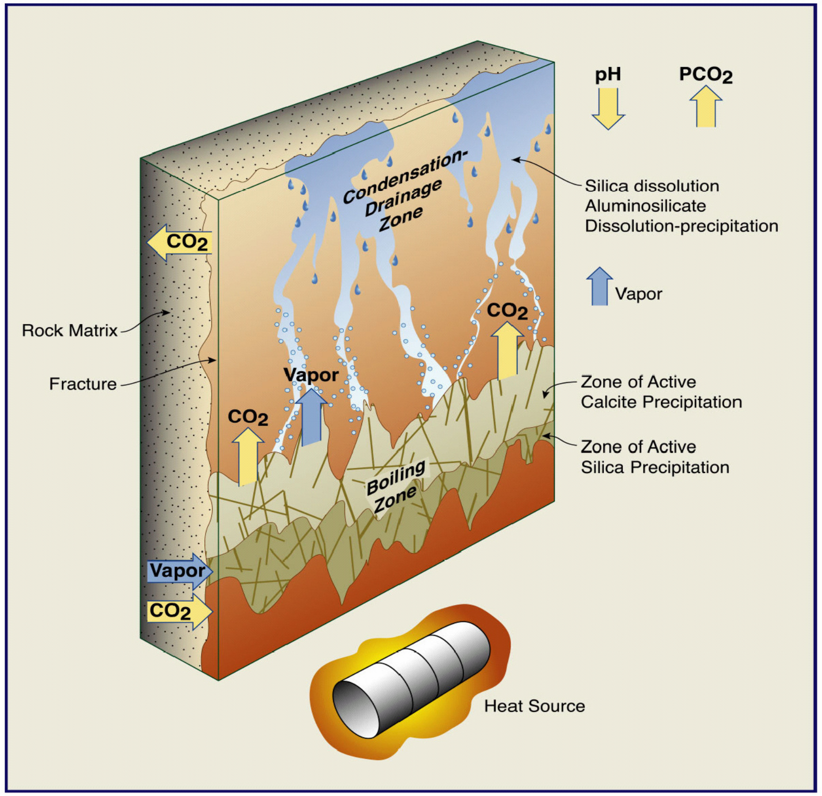




(Clauser 2003)

Geochemical processes

- Chemical evolution of waters, gases and minerals intimately coupled to TH processes
  - Drying concentrates aqueous species in remaining liquid phase
  - Boiling forces mineral precipitation
  - Dilute water in condensation zones promotes dissolution of minerals
- Mineral-water reactions limited by kinetics and water saturation
- pH affected by
  - CO_2 degassing and transport
  - Mineral dissolution/precipitation
- Mineral dissolution and precipitation
  - Porosity and permeability



THC Processes

(Sonnenthal et al 2005)

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Saturated

$$\frac{\partial C_i}{\partial t} = \underbrace{-v_a \nabla C_i + \nabla \cdot (D \nabla C_i) + Q_{C_i}} + \underbrace{Reakt(C_1, ..., C_n)}$$

i=1, 2,...,n

n x conservative Transport

Chemical reactions with PHREEQC, ChemApp or GEMS-PSI

$$\frac{\partial C_i}{\partial t} = -v_a \nabla C_i + \nabla \cdot (D \nabla C_i) + Q_{C_i}$$

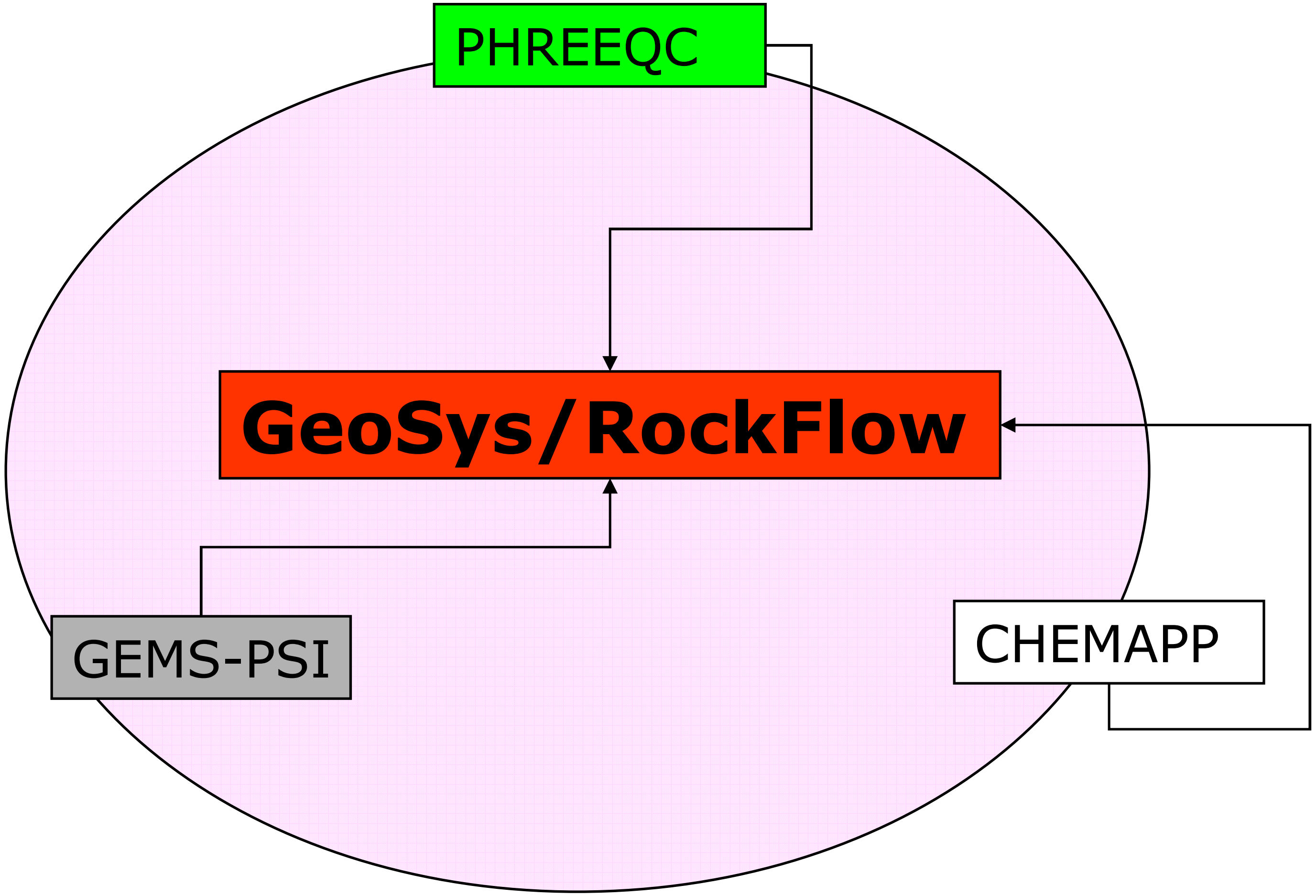
$$\frac{\partial C_i}{\partial t} = Reakt(C_1, ..., C_n)$$

Unsaturated

$$\frac{\partial(nS_\gamma C_{i,\gamma})}{\partial t} = -v_\gamma \nabla C_{i,\gamma} + \nabla(nS_\gamma D_{i,\gamma} \nabla C_{i,\gamma}) + nS_\gamma Q_{i,\gamma} + nS_\gamma \Gamma_{i,\gamma}(C_{1,\gamma}...C_{n,\gamma})$$

where $C_{i,\gamma}$ is the concentration of the i-th component of a n component system in phase γ, v_γ is the Darcy velocity of phase γ, S_γ is saturation of phase γ, D is the diffusion - dispersion coefficient of component i in phase γ, $Q_{i,\gamma}$ is the source/sink term and $\Gamma_{i,\gamma}(C_{1,\gamma}...C_{n,\gamma})$ is the source/sink term of component i in phase γ due to equilibrium chemical reactions with all other species in the same phase.

(Suarez & Simunek, 1996)

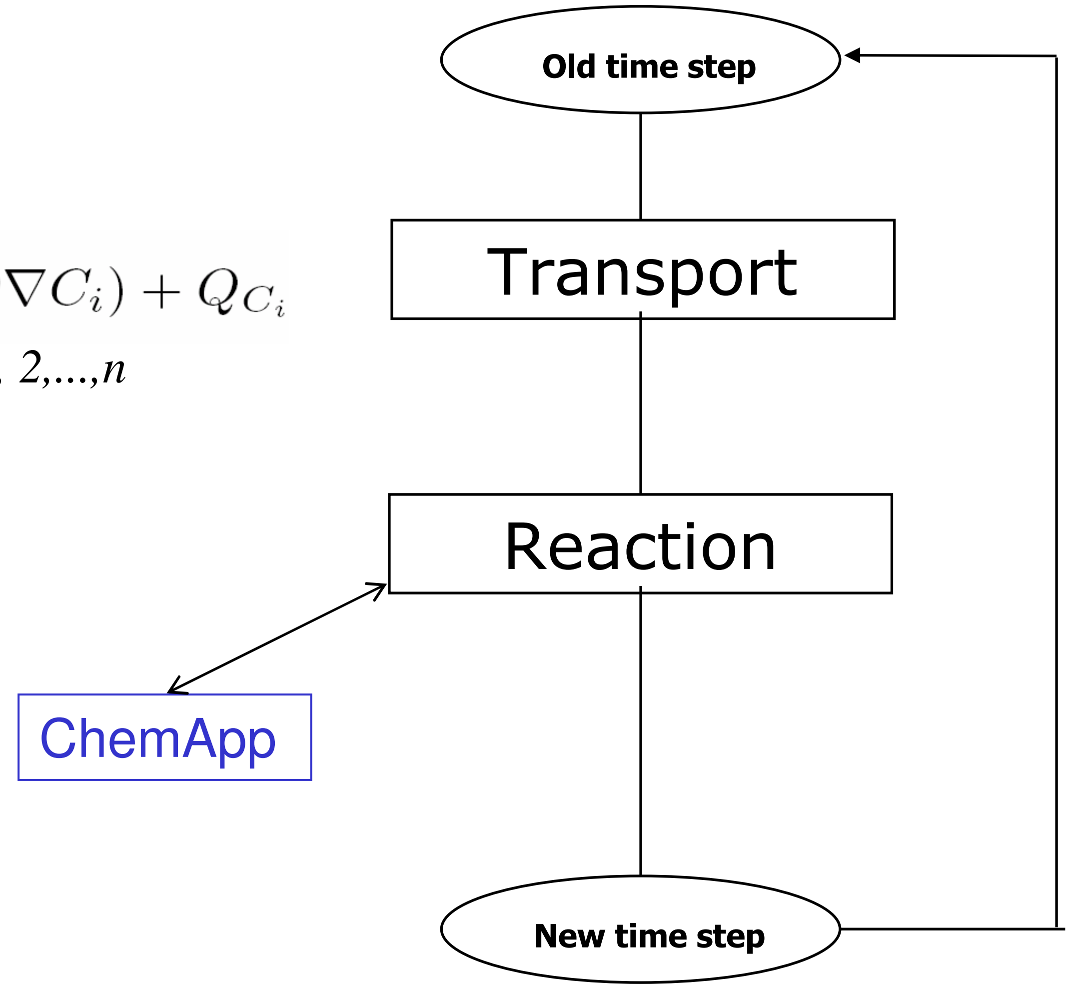

PHREEQC
GeoSys/RockFlow
GEMS-PSI
CHEMAPP

RockFLow/GeoSys+ChemApp

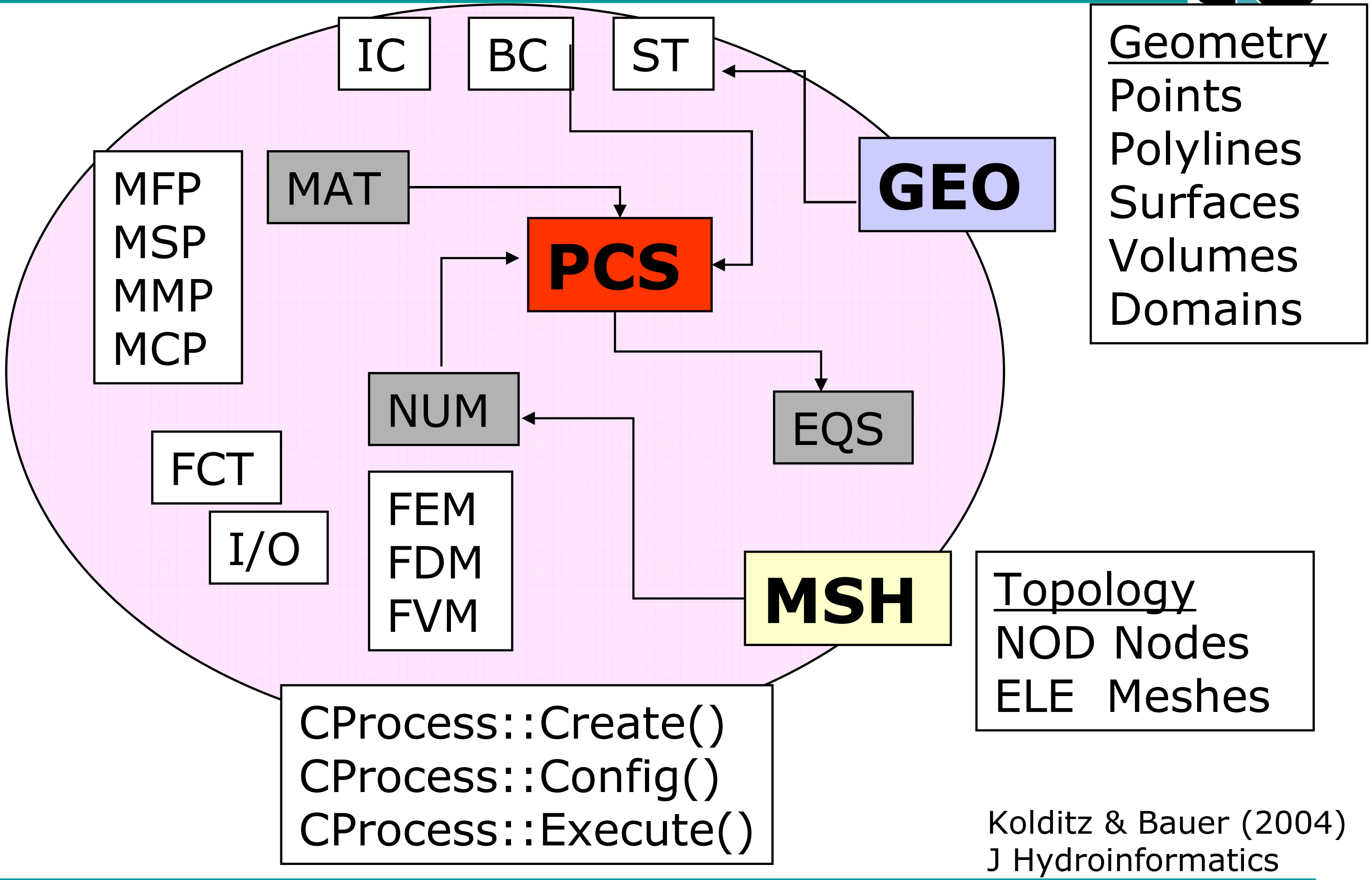
$$\frac{\partial C_i}{\partial t} = -v_a \nabla C_i + \nabla \cdot (D \nabla C_i) + Q_{C_i}$$

$i=1, 2,\ldots,n$

$$\frac{\partial C_i}{\partial t} = Reakt(C_1, \ldots, C_n)$$

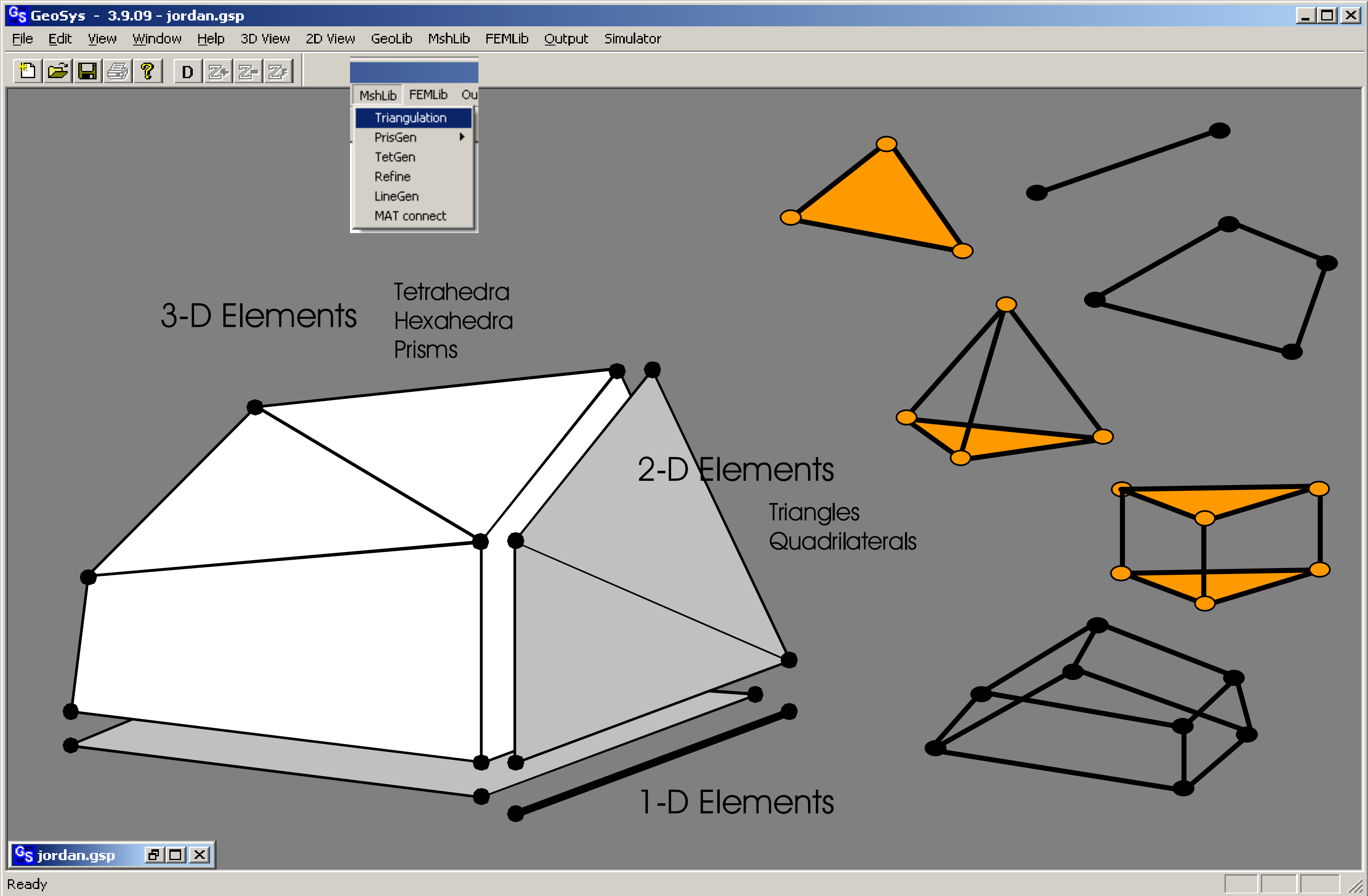


Object-Orientation:Multifield Problems



Kolditz & Bauer (2004)
J Hydroinformatics

Geometric Element Types



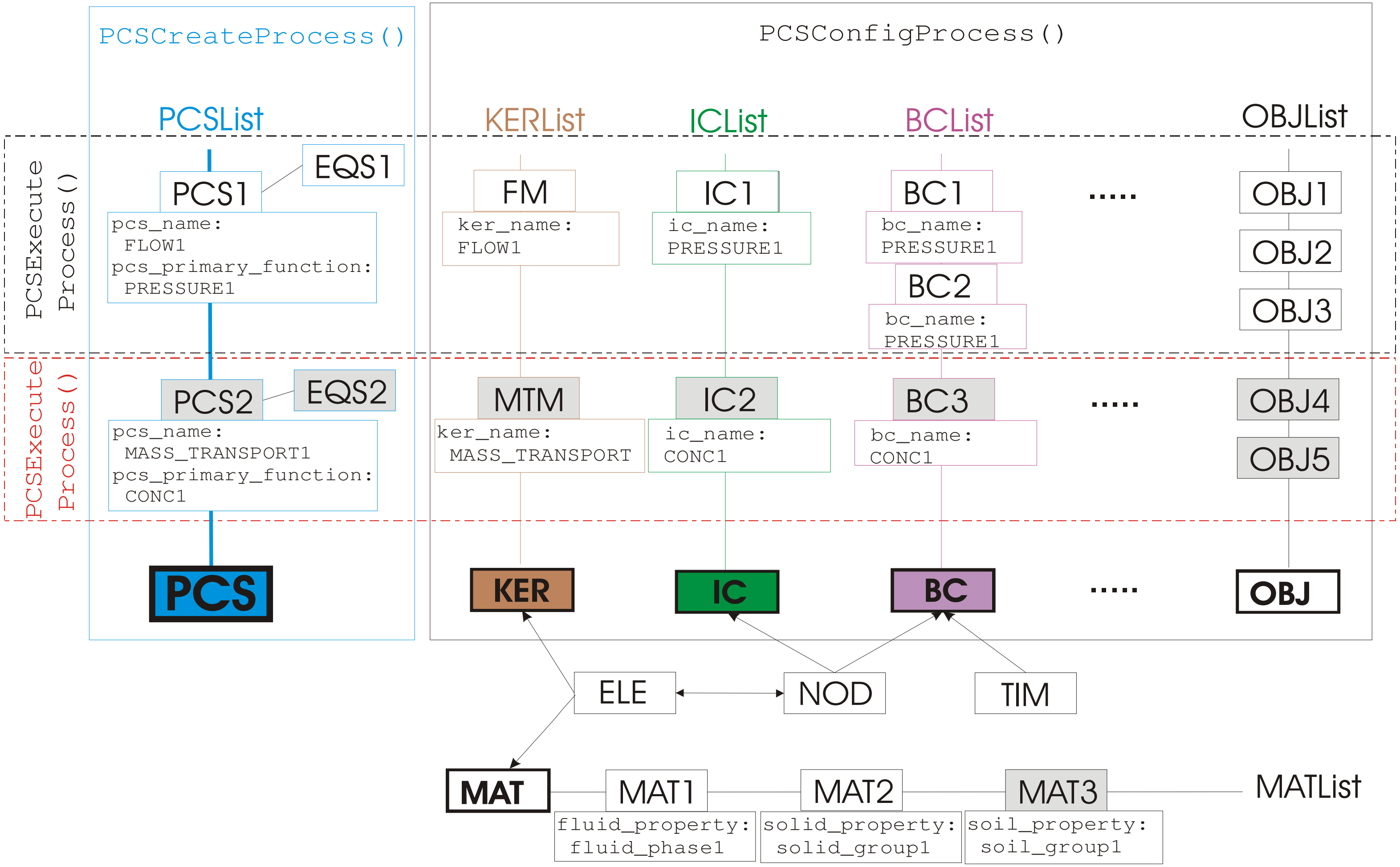
$$\begin{pmatrix} A_1 & B_{12} & B_{13} & & \dots & B_{1n} \\ B_{12} & A_2 & . & B_{24} & \dots & \\ B_{13} & . & A_3 & & \dots & \\ & B_{24} & & A_4 & \dots & B_{4n} \\ \dots & \dots & \dots & \dots & \dots & \dots \\ B_{1n} & & & B_{4n} & \dots & A_n \end{pmatrix} * \begin{pmatrix} C_1 \\ C_2 \\ C_3 \\ C_4 \\ C_5 \\ C_6 \end{pmatrix} = \begin{pmatrix} R_1 \\ R_2 \\ R_3 \\ R_4 \\ \dots \\ R_n \end{pmatrix}$$

Dimension: (number of grid nodes * number of components)^2

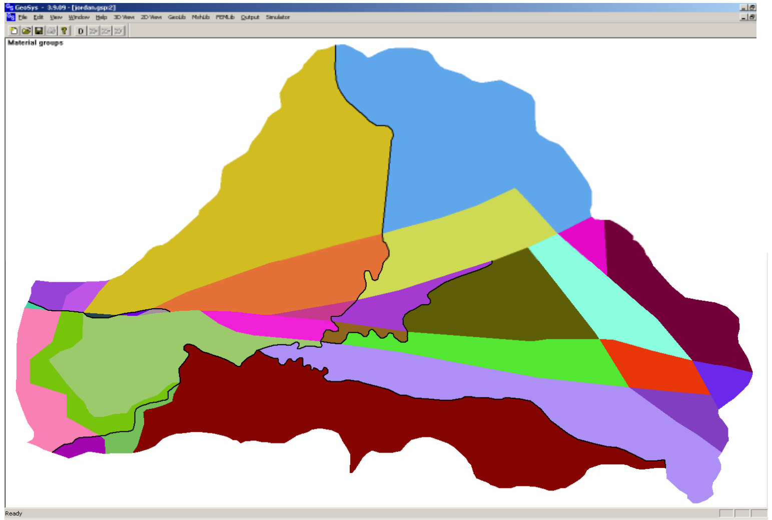
For real world applications: $(10^5 * 20)^2 = 4*10^{12}$

Memory requirement: $4*10^{12}$

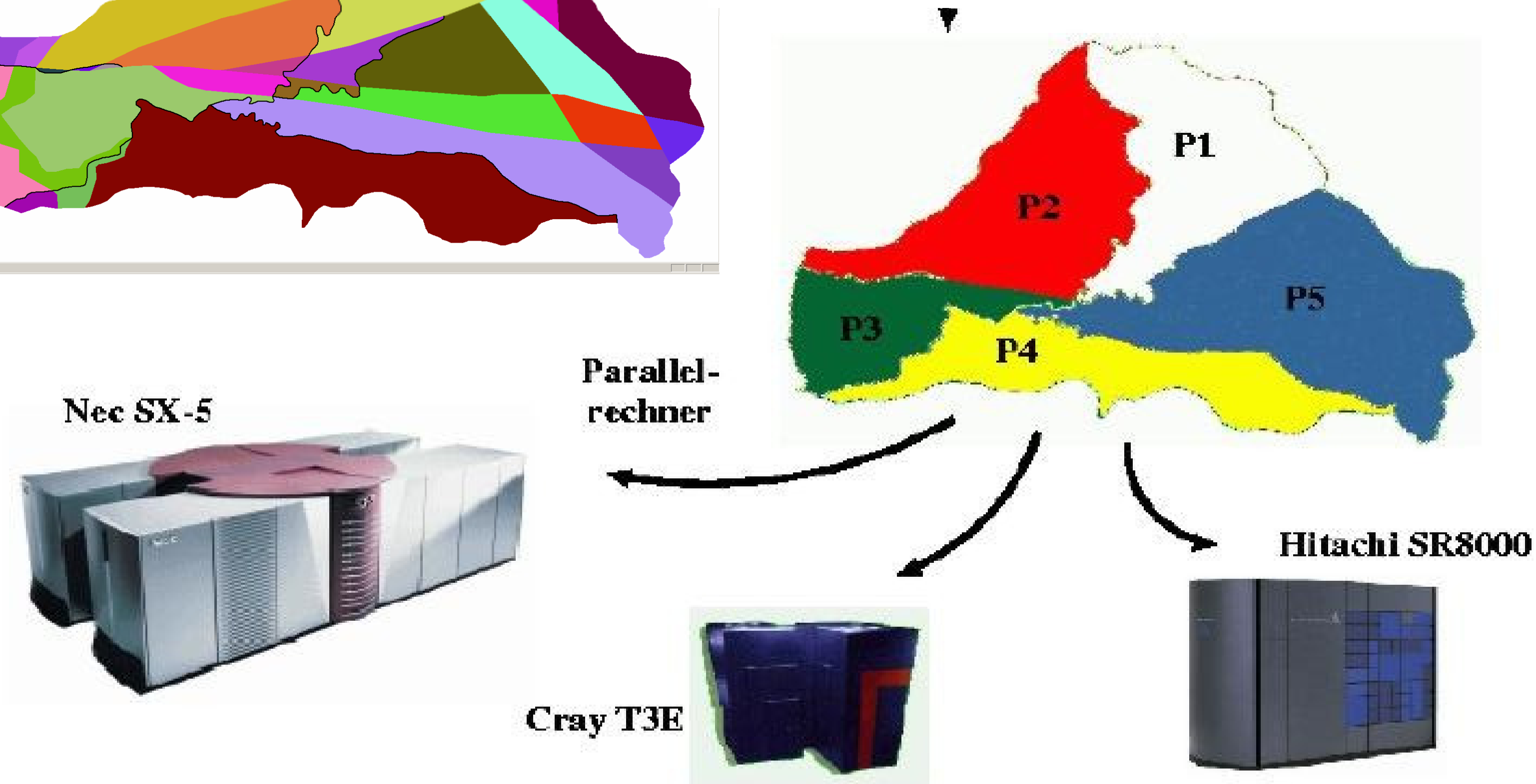
PCS Implementation



Parallelization



High Performance Computing
Parallel Computing
Visualization

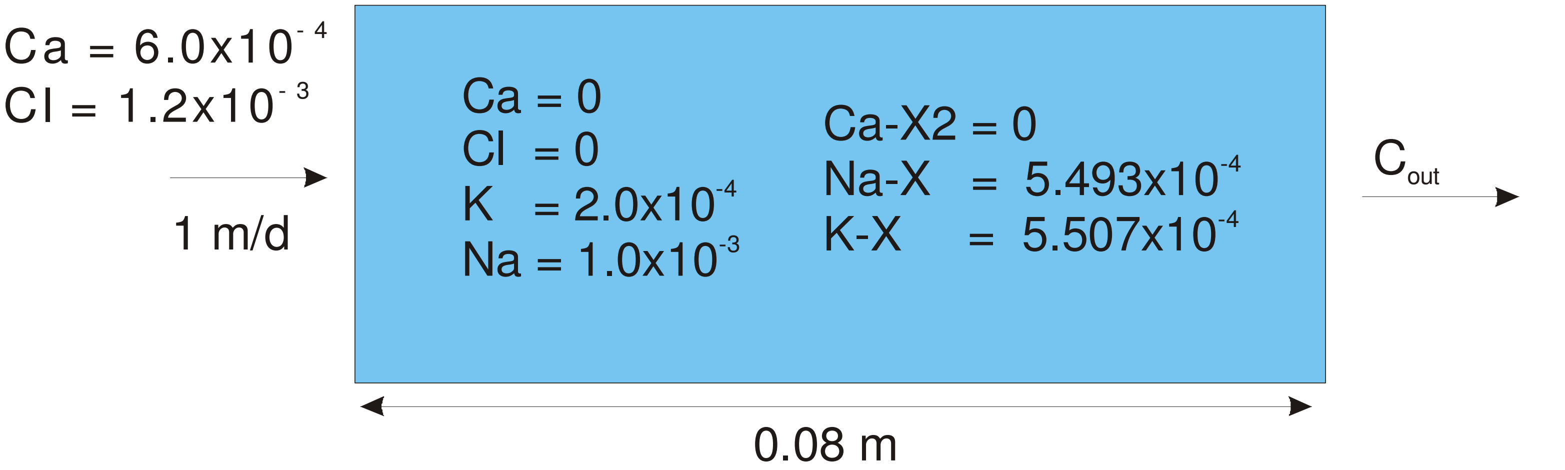


In cooperation with HLRS Stuttgart

Verification example: Comparison with 1D PHREEQC

1D Transport and Cation exchange (Example 11, PHREEQC – User Instructions)

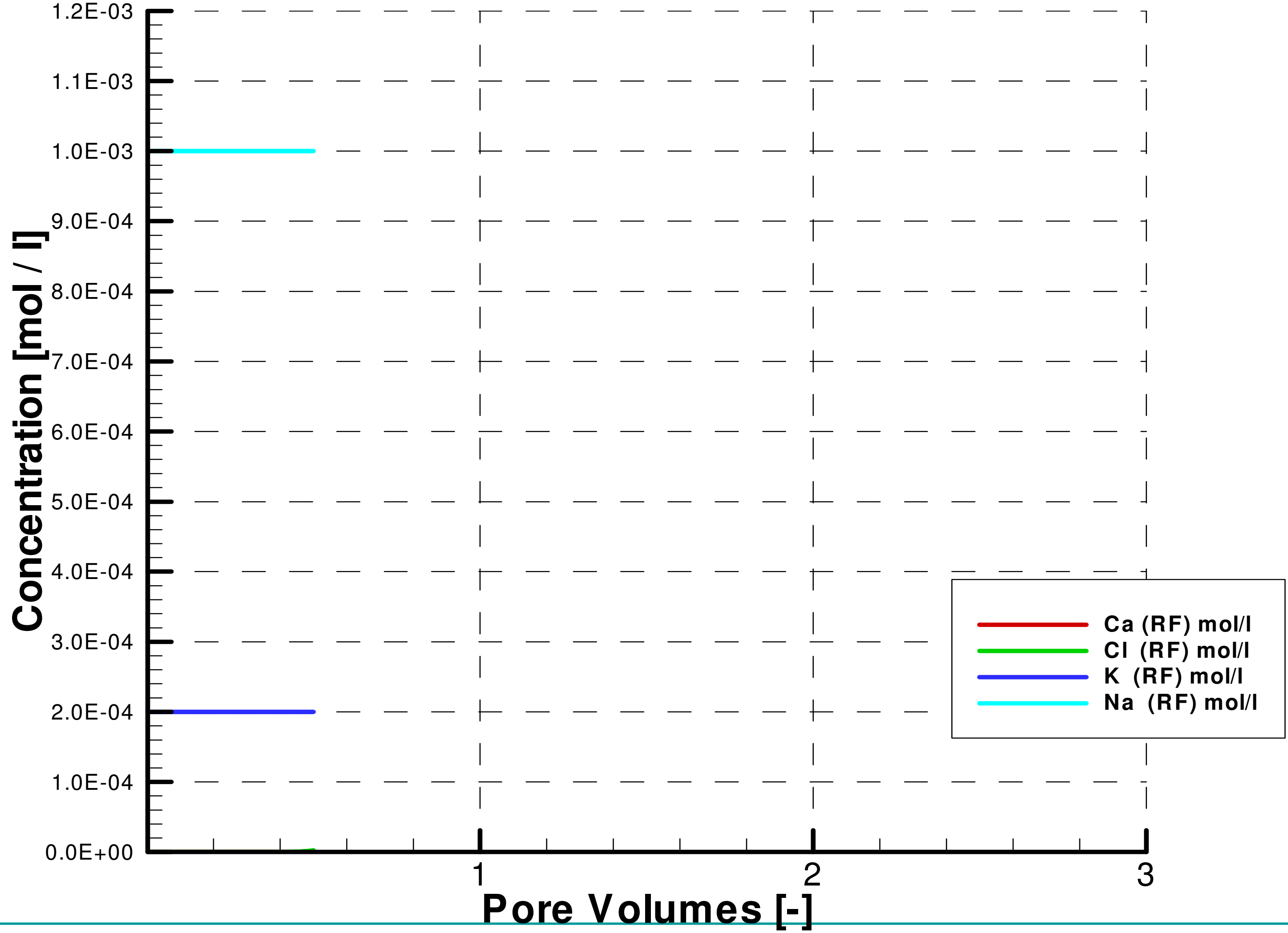
Flushing a 1D column with $CaCl_2$.
Initial pore water: Na, K
Exchange: Ca-X2, Na-X, K-X



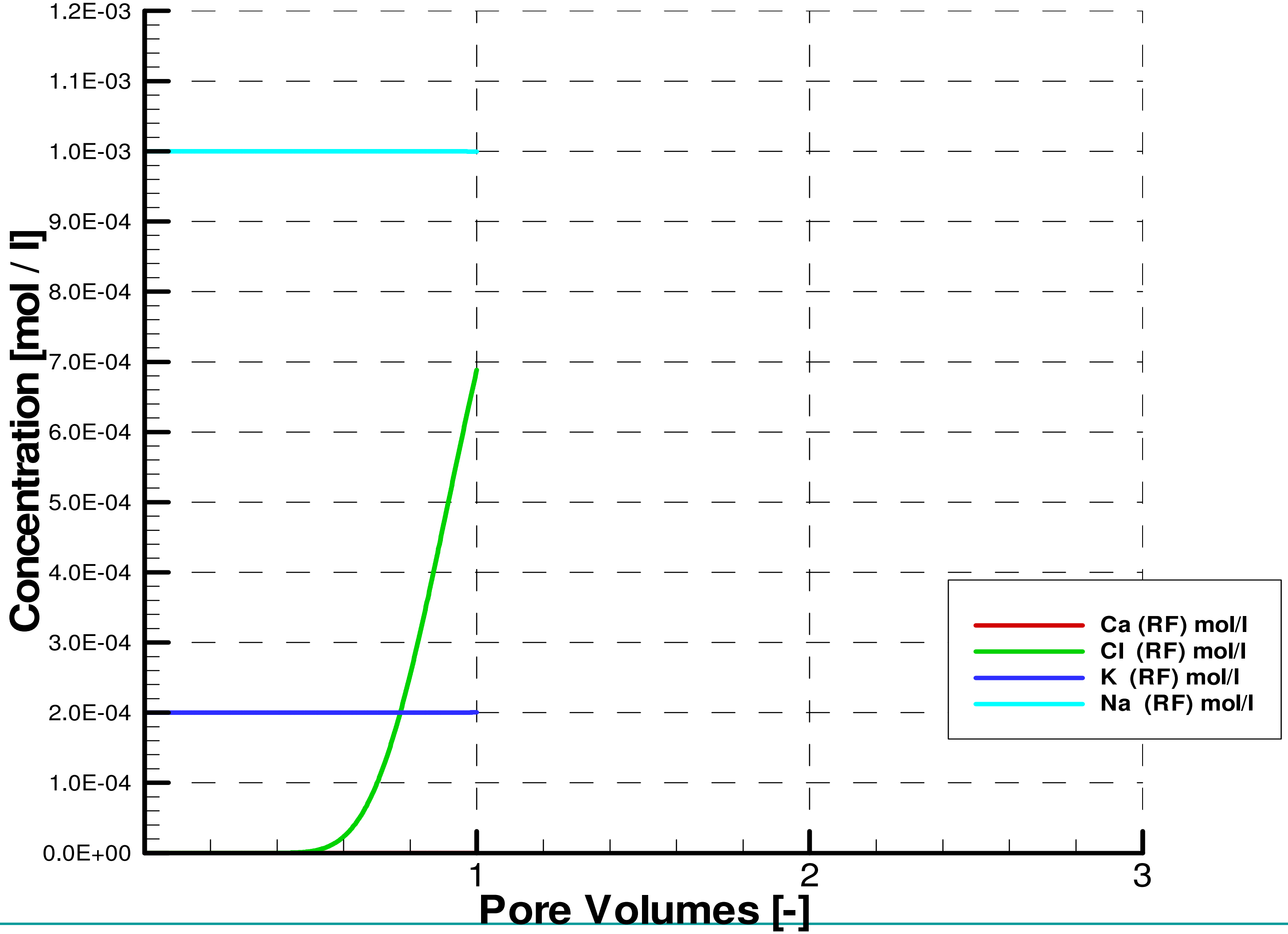
D = 0.002 m, n = 1.0, t=0.24 d

C in mol/l

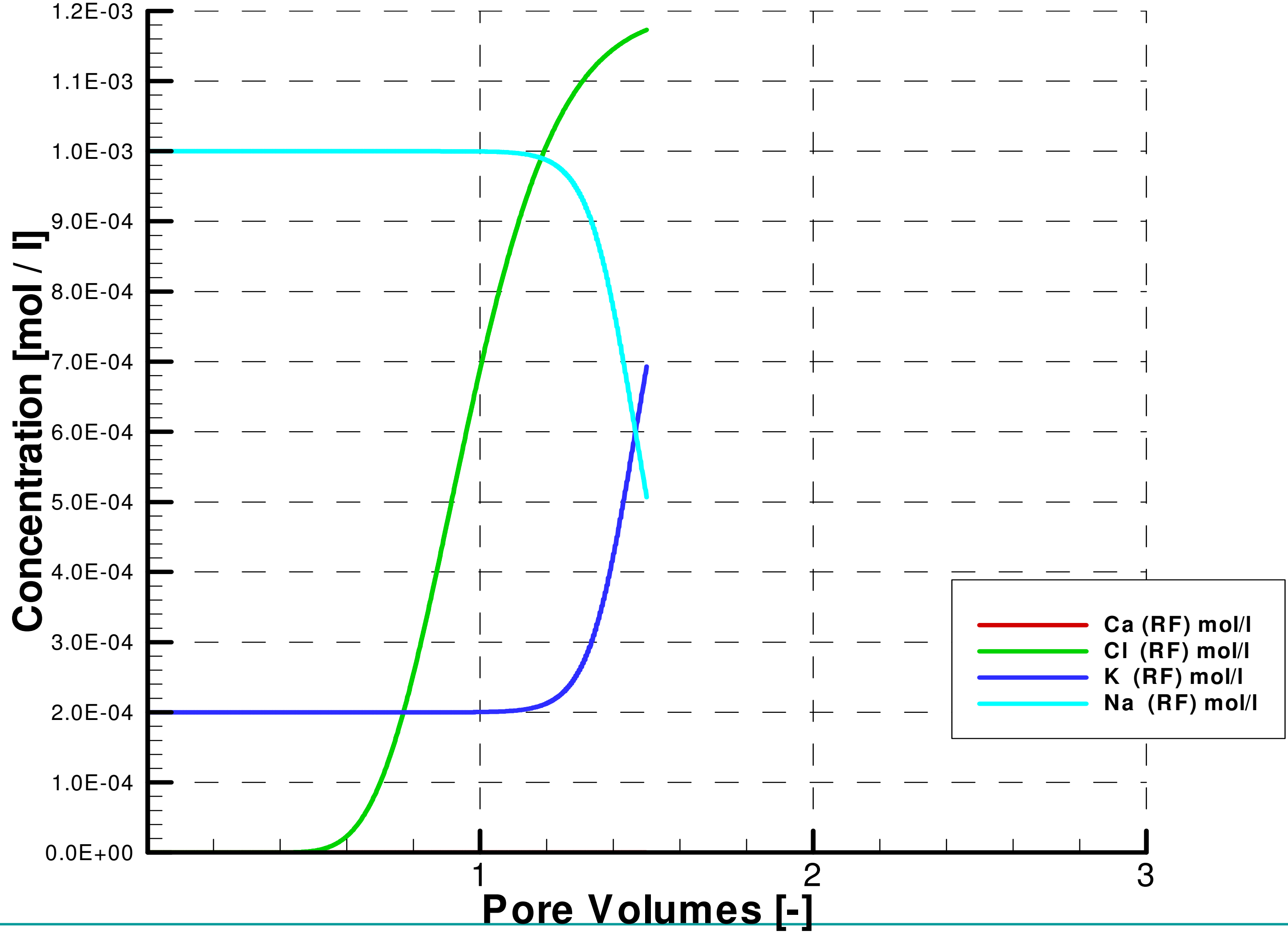
Verification Benchmark



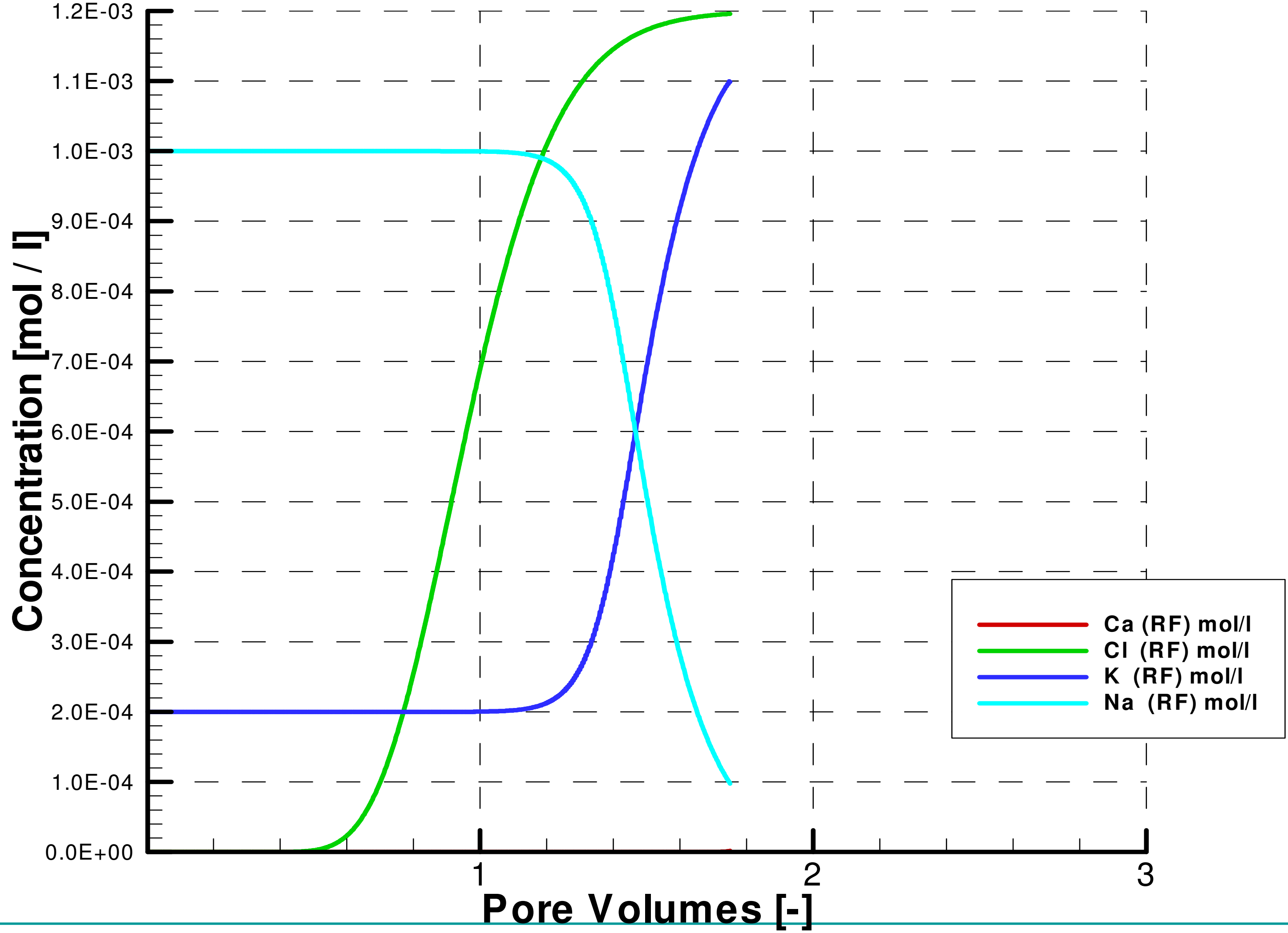
Verification Benchmark



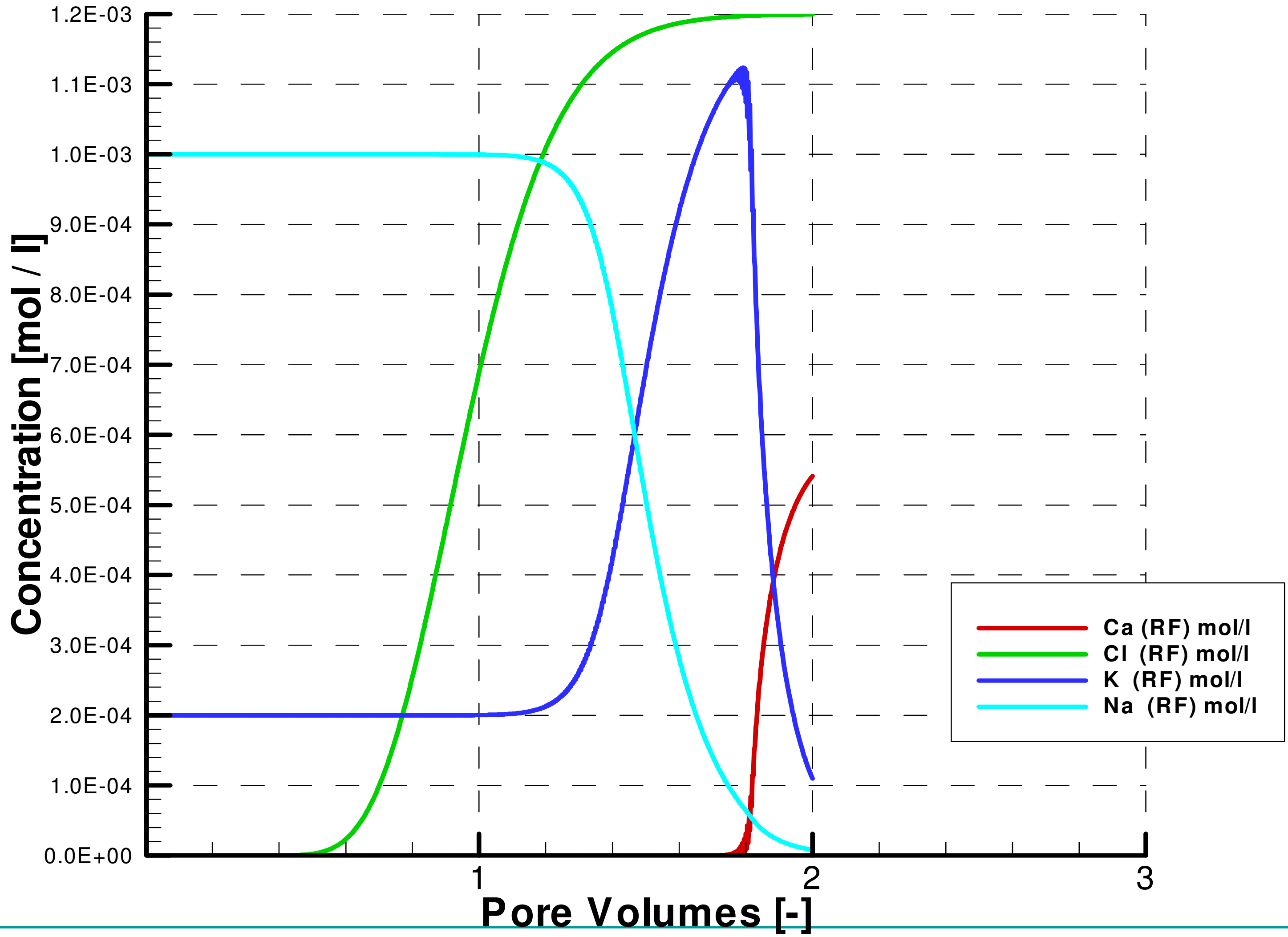
Verification Benchmark



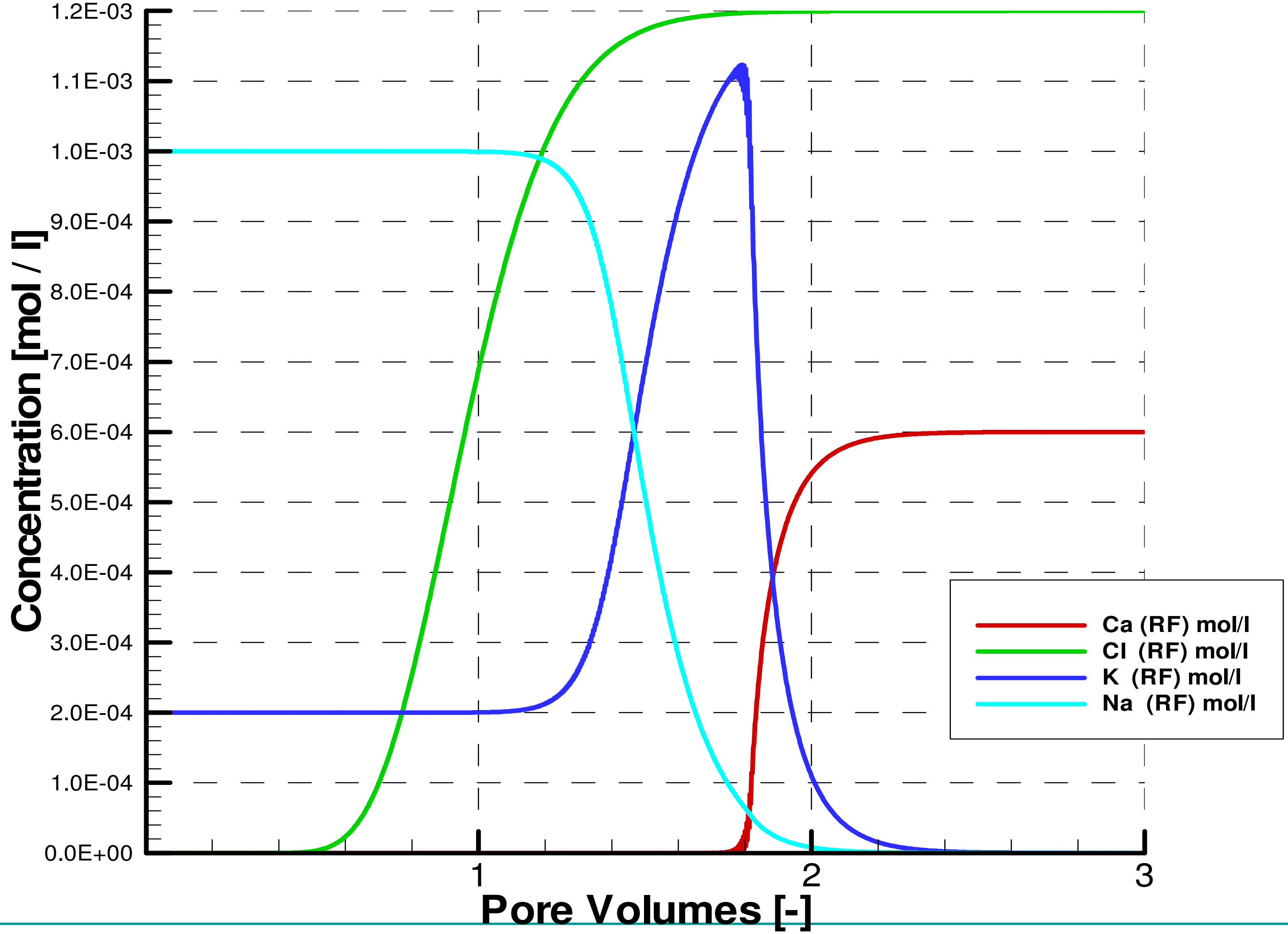
Verification Benchmark



Verification Benchmark



Verification Benchmark



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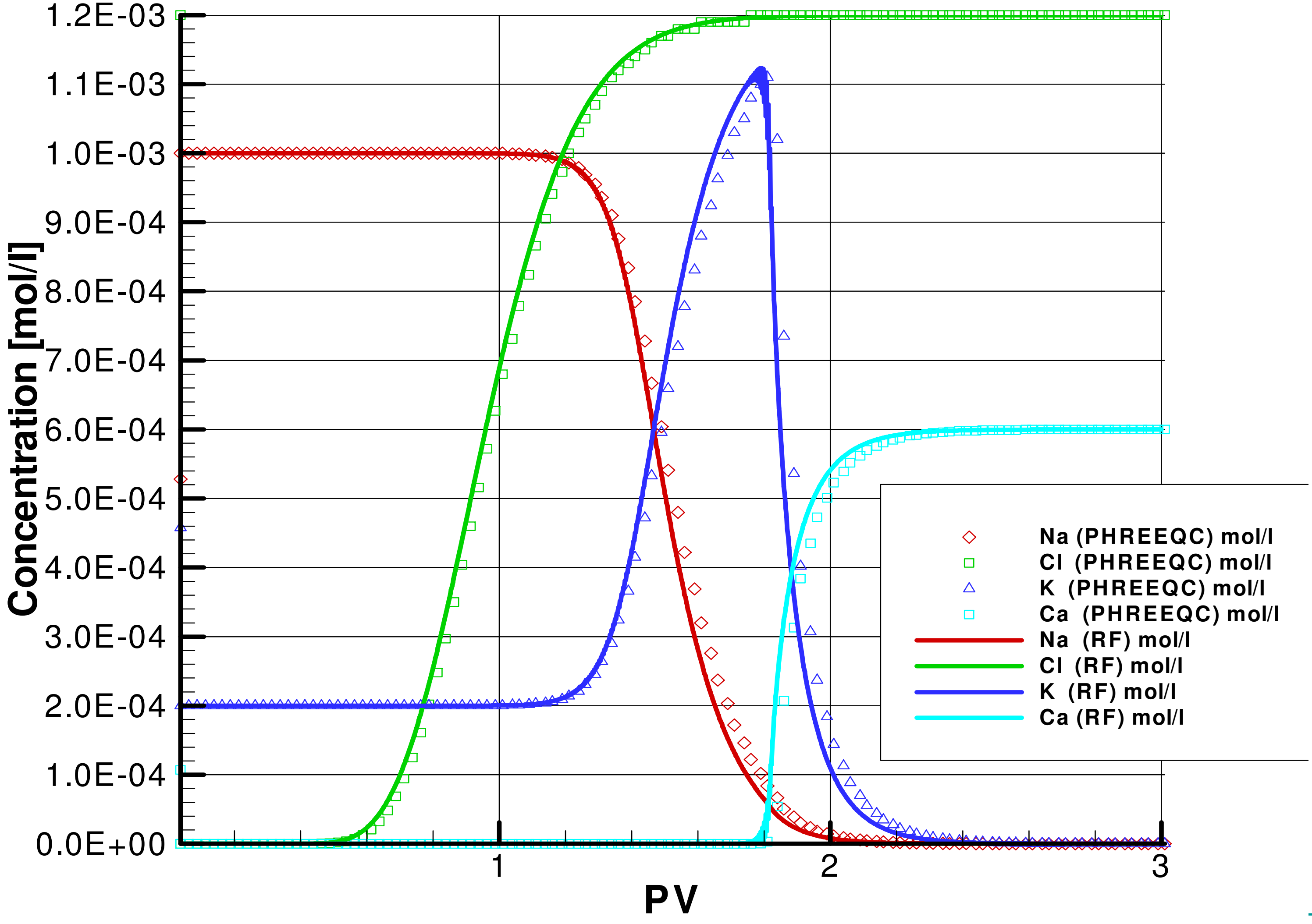
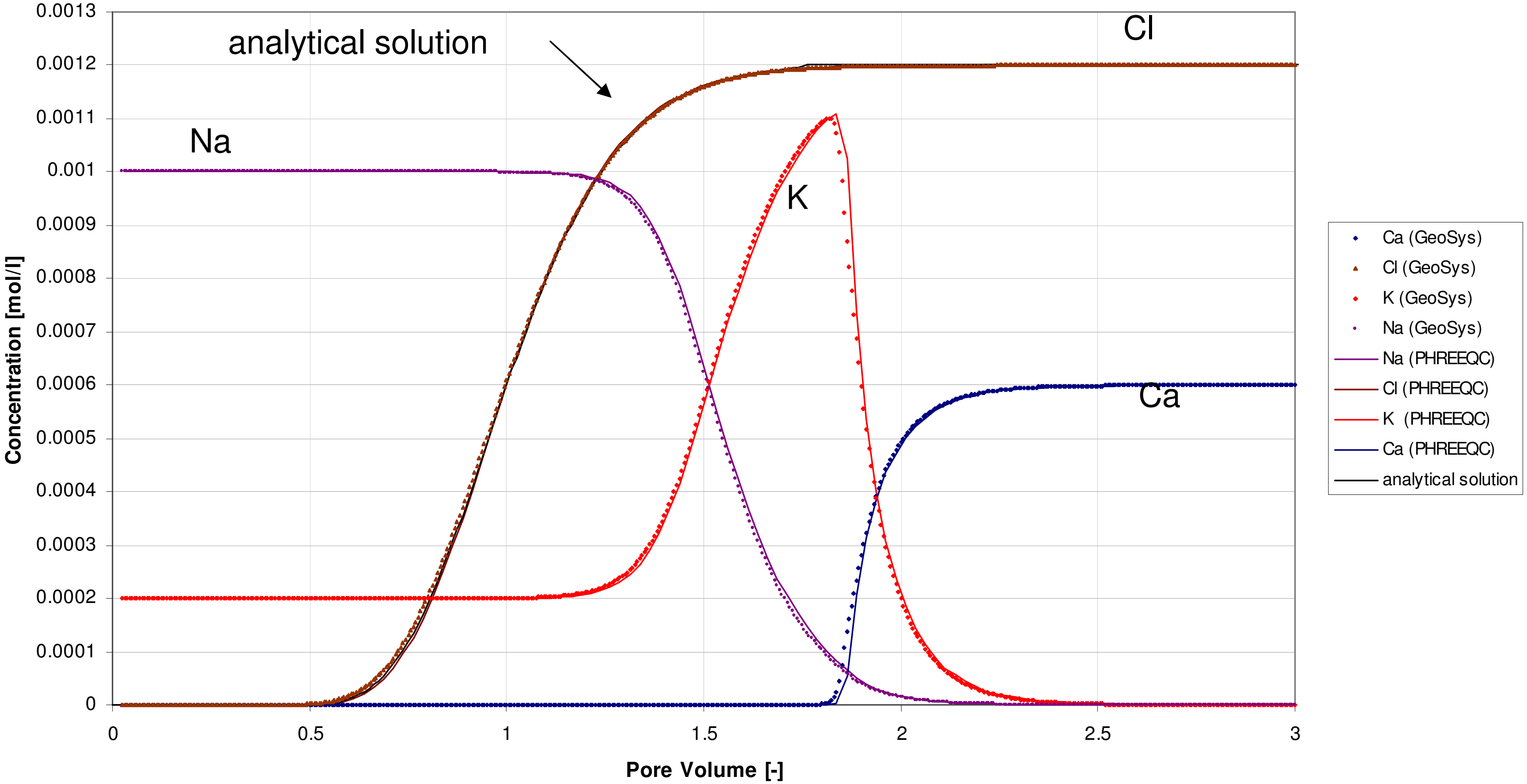


Concentrations at the column outlet



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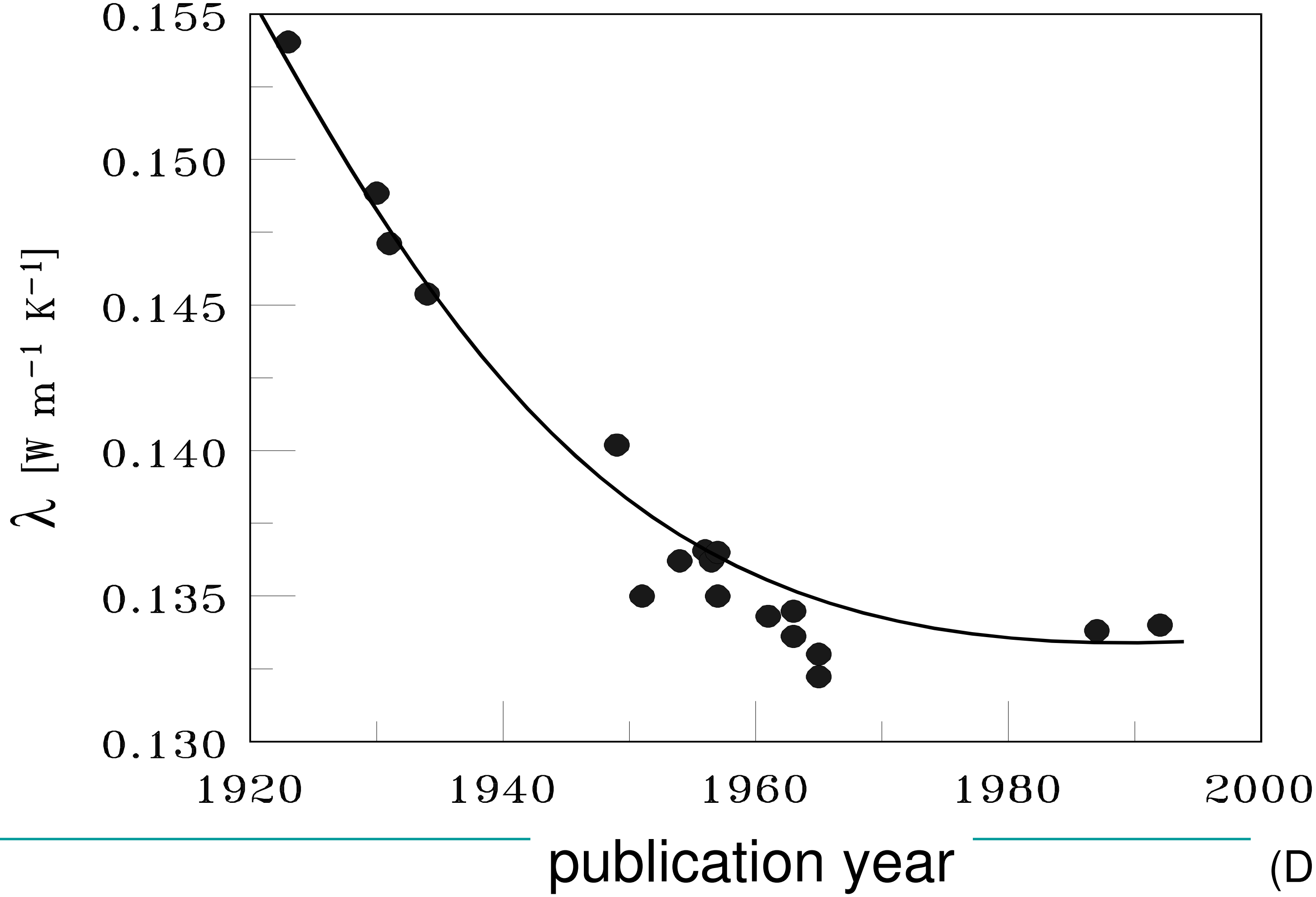
Cause of Database Difference

Example: Thermal Conductivity λ of Liquid Toluene at 25°C



(DDBST, 2006)

Verification Example

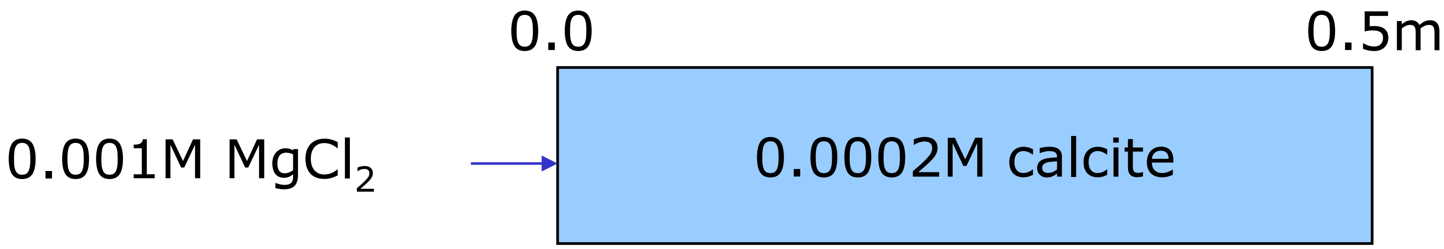
1D Transport and calcite dissolution

Simulated Program

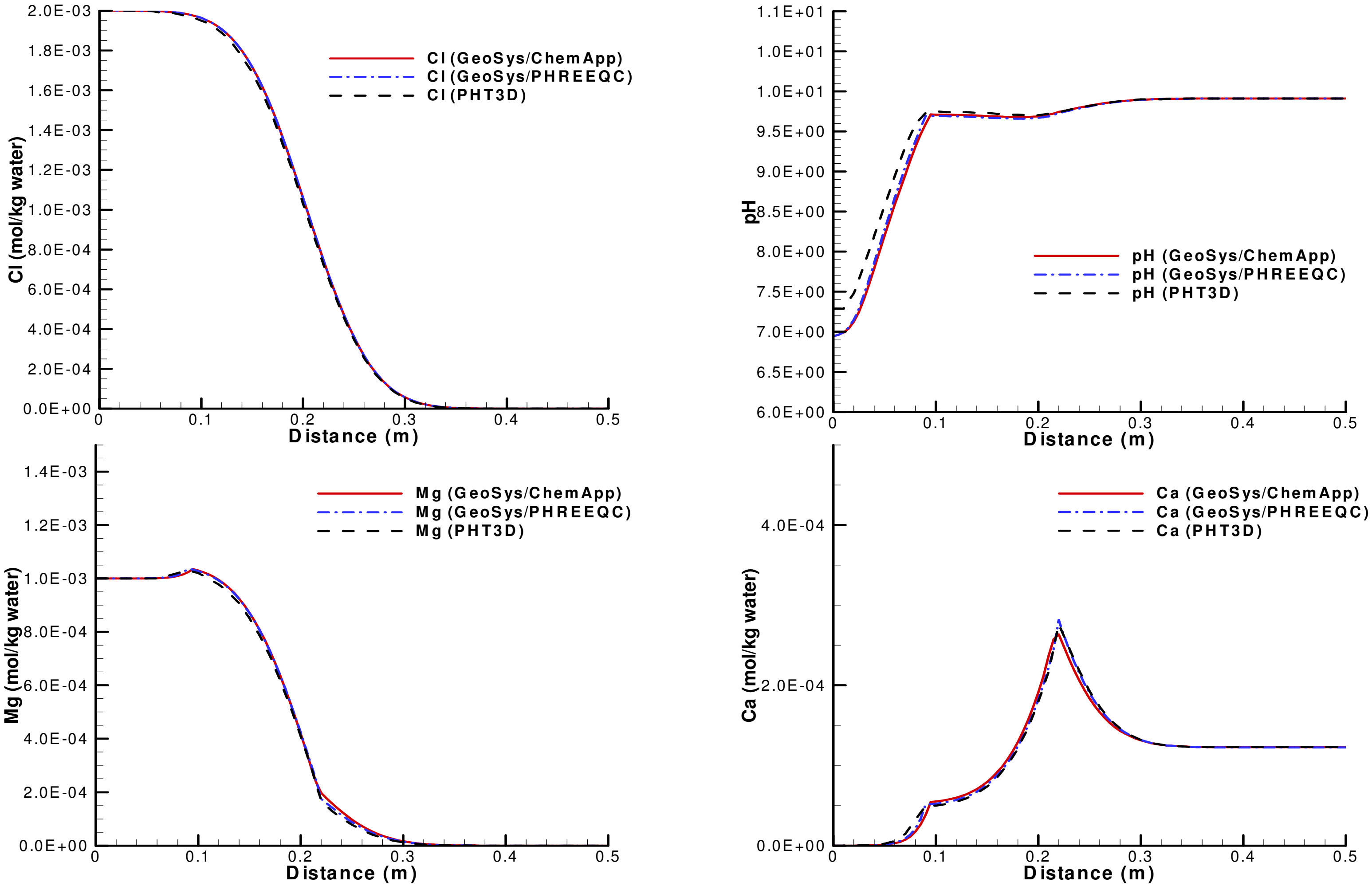
-- MST1D (by Engesgaard and Kipp 1992)

-- PHT3D (by Prommer 2002)

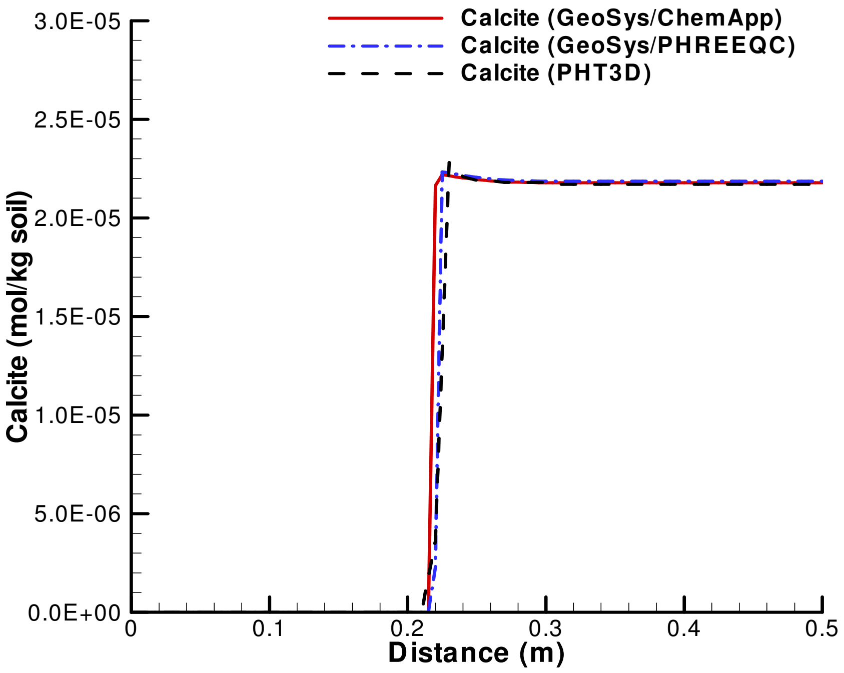
-- GeoSys/PHREEQC (by Xie et al 2005)

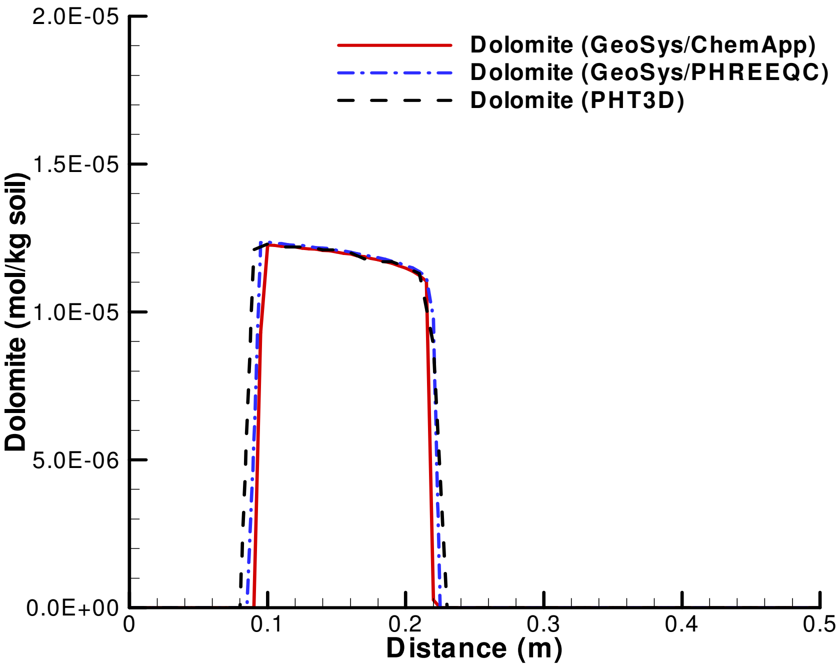


Simulation results comparison



Simulation results comparison





Database sensitivity analysis

Species	$\triangle_f G_m^0$ $(J \cdot mol^{-1})$ YMF	NAGRA/PSI	Error	Percentage error (%)
H_2O	-237182.3302	-237140	-42.33015071	0.018
OH^-	-157297.7015	-157230	-67.70151593	0.043
Mg^{2+}	-455375	-455375	0	0.000
Ca^{2+}	-552806	-552806	0	0.000
Cl^-	-131217	-131217	0	0.000
$(HCO_3)^-$	-586845	-586875	30	-0.005
$(CO_3)^{2-}$	-527887.7684	-527917	29.23161984	-0.006
$(CO_2)^0$	-385878.4885	-385992	113.5115214	-0.029
$CaCO_3^0$	-1099684.997	-1099127	-557.9973053	0.051

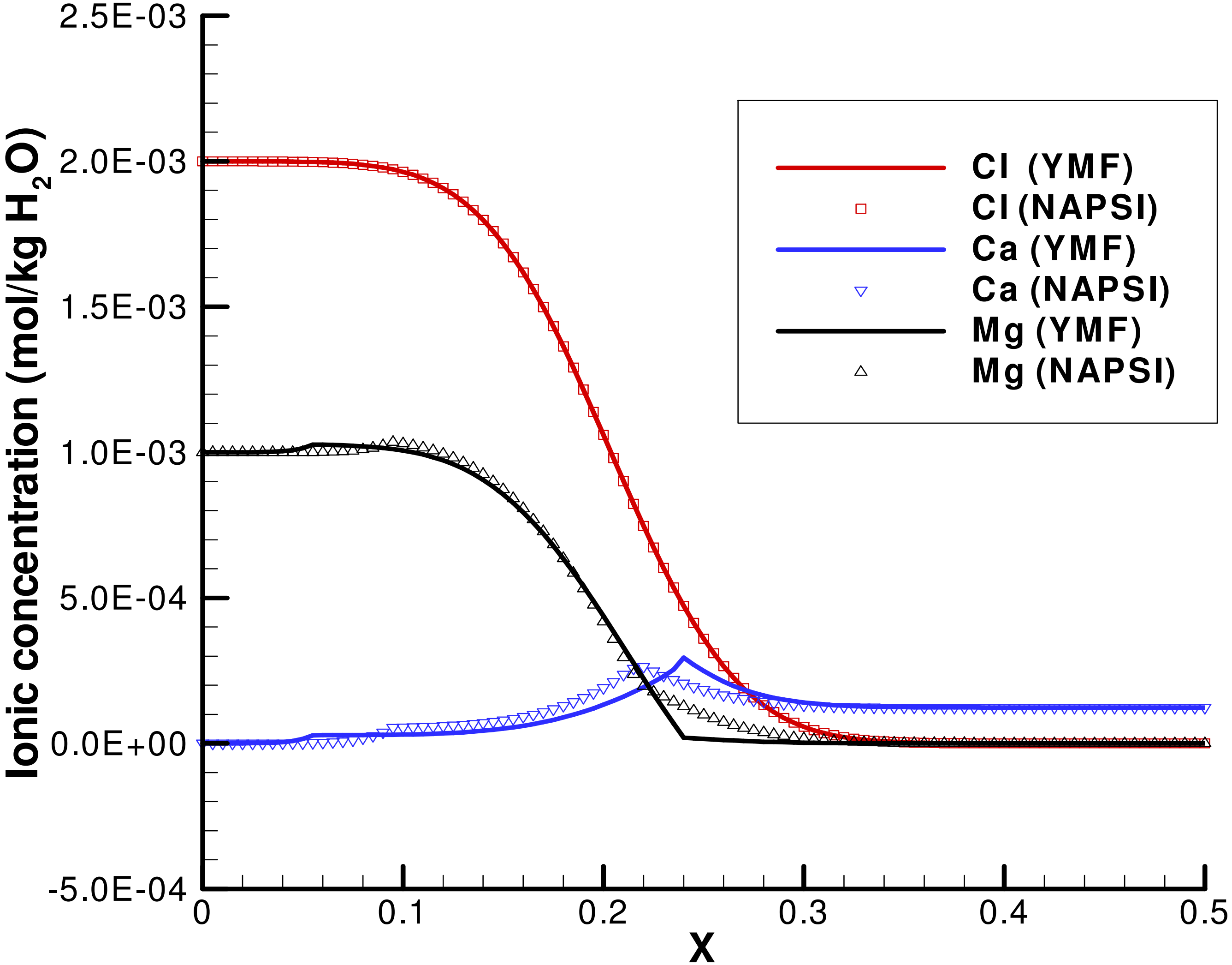
Database sensitivity analysis

Species	$\triangle_f G_m^0$ $(J \cdot mol^{-1})$ YMF	NAGRA/PSI	Error	Percentage error (%)
$MgCO_3^0$	-1000266.457	-1000300	33.54316887	-0.003
$Mg(OH)^+$	-625873.1211	-627215	1341.878896	-0.214
$Mg(HCO_3)^+$	-1048131.82	-1048347	215.1801576	-0.021
$Ca(HCO_3)^+$	-1145625.608	-1145992	366.3916877	-0.032
$Ca(OH)^+$	-716735.3055	-716997	261.6944549	-0.036
$CaCO_3\_Aragonite$	-1128274.3	-1128306	31.69994012	-0.003
$CaCO_3\_Calcite$	-1129098.541	-1129127	28.45857172	-0.003
$CaMg(CO_3)_2\_Dolomite$	-2167523.835	-2161565	-5958.83463	0.276

[a] Nagra/PSI database [*Hummel et al.*, 2002].

[b] Database based on EQ3/6, data0.ymf.

Database sensitivity analysis



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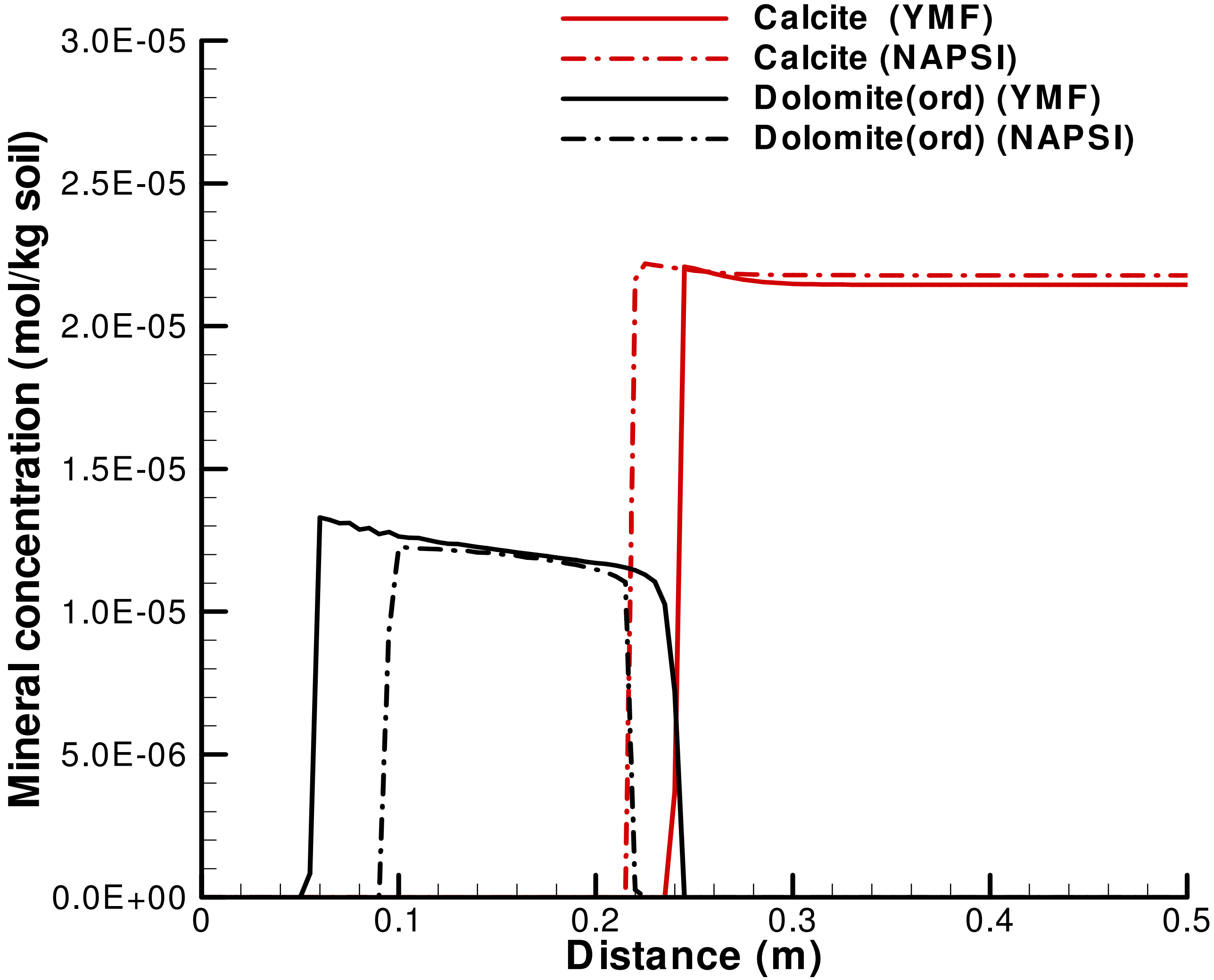

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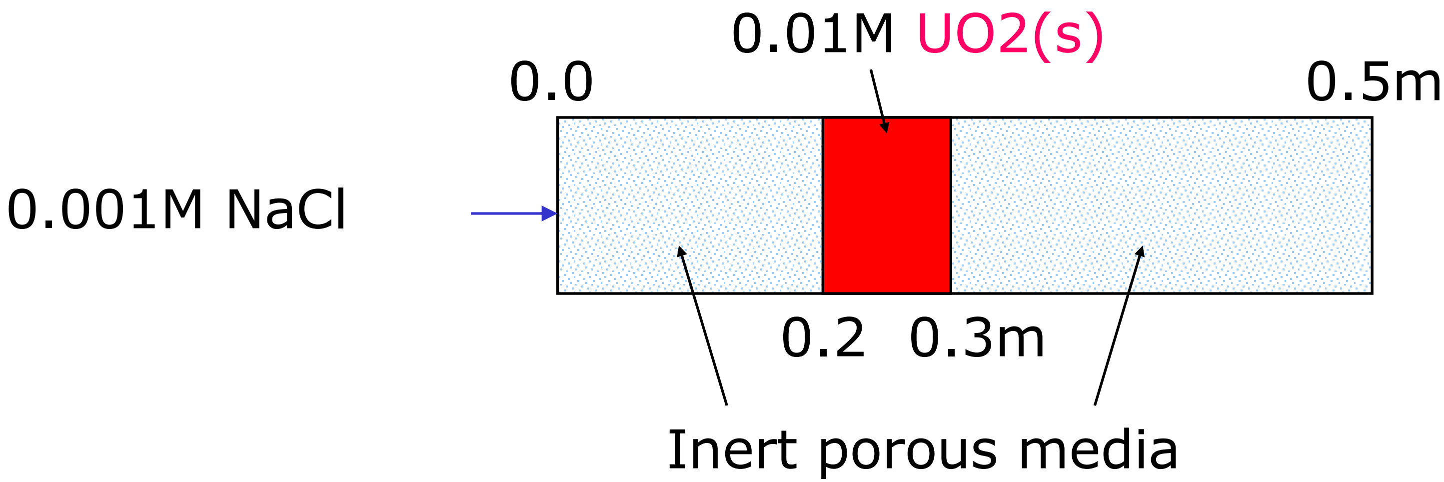
Application Example

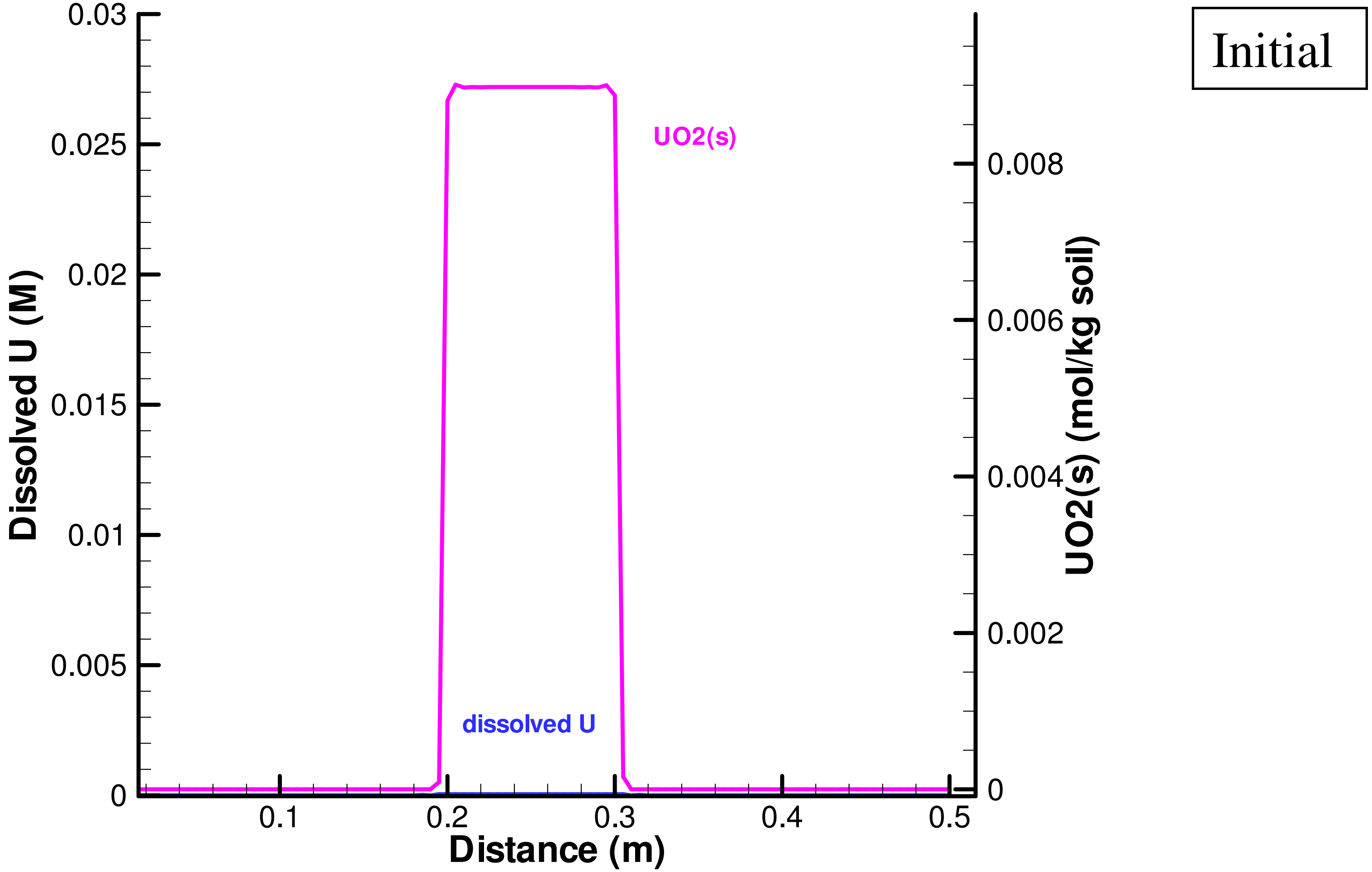
1D Transport and UO2(s) dissolution

Simulated Program

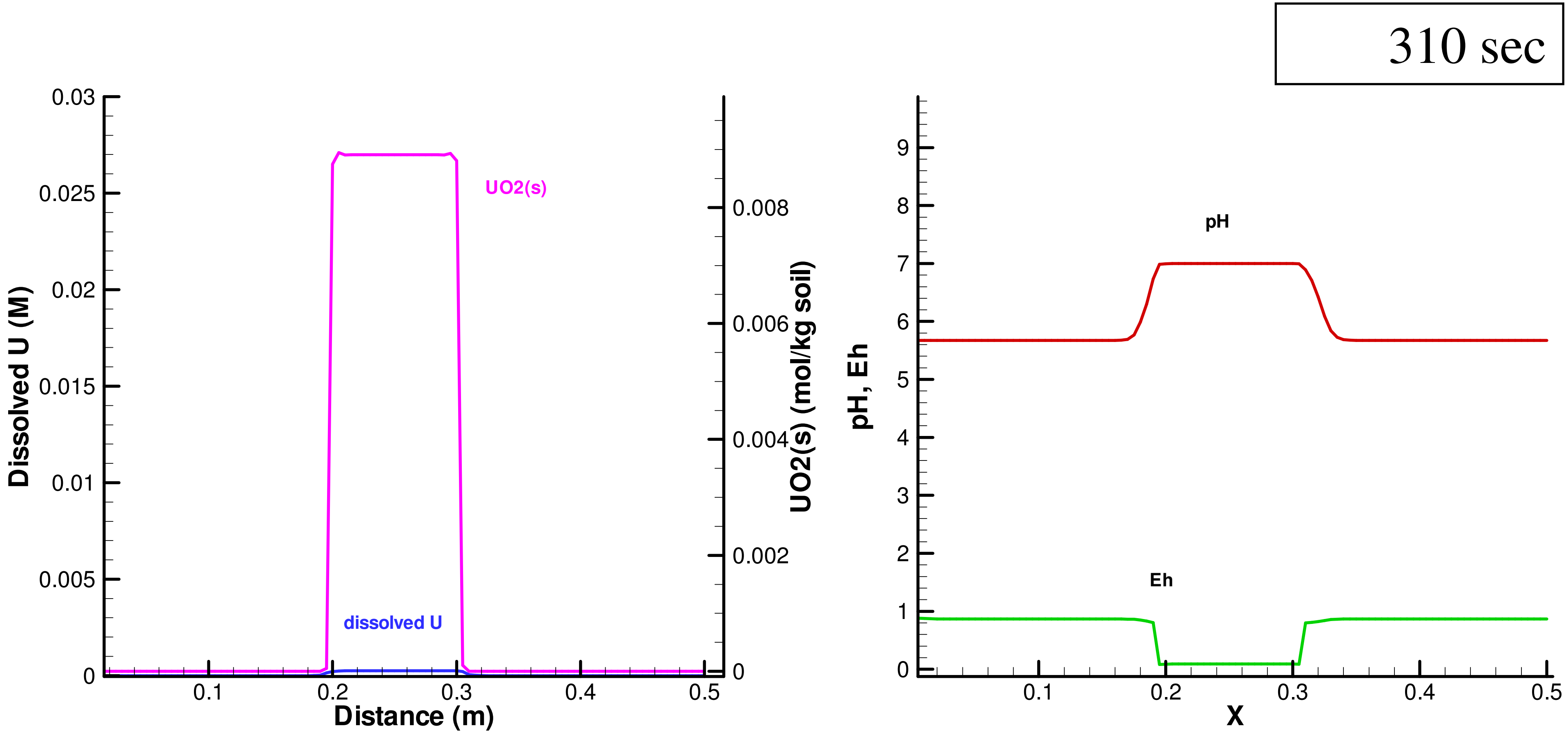
-- GeoSys/Rockflow+ChemApp (by Xie et al 2005)



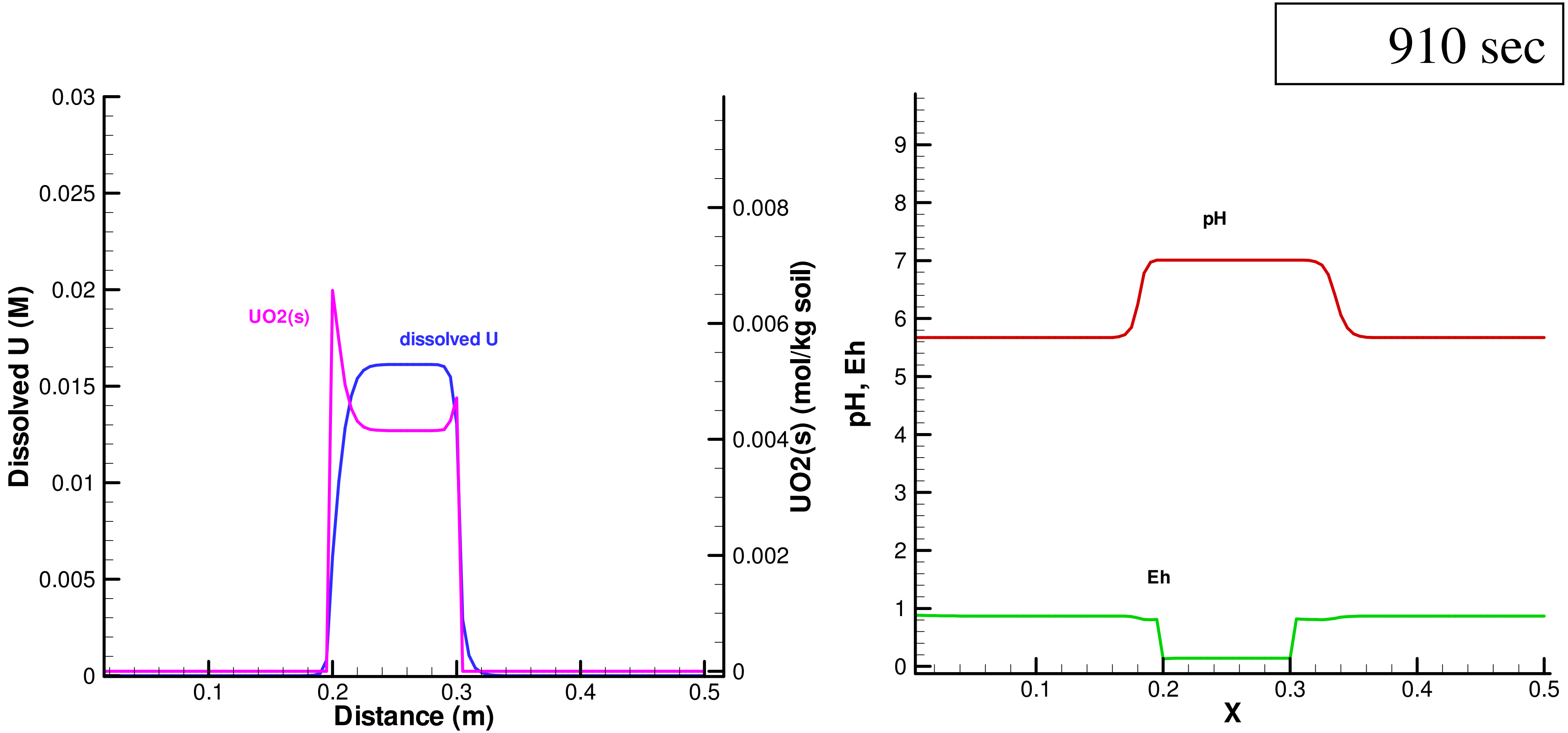
UO2(s) dissolution and transport



UO2(s) dissolution and transport

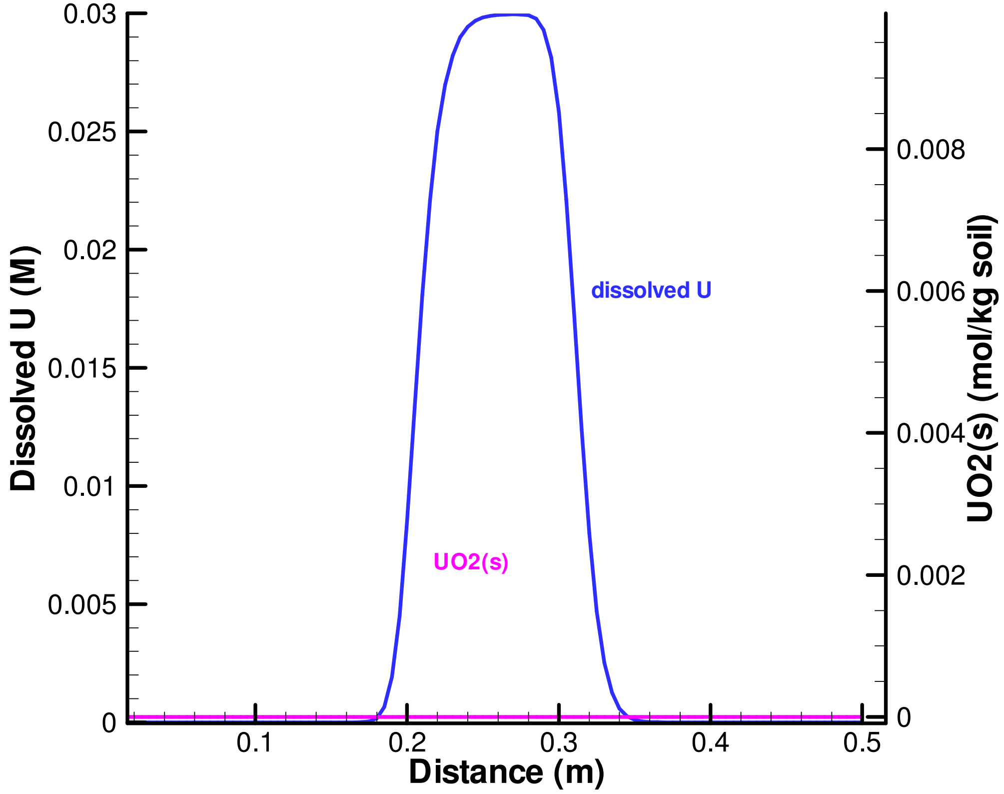


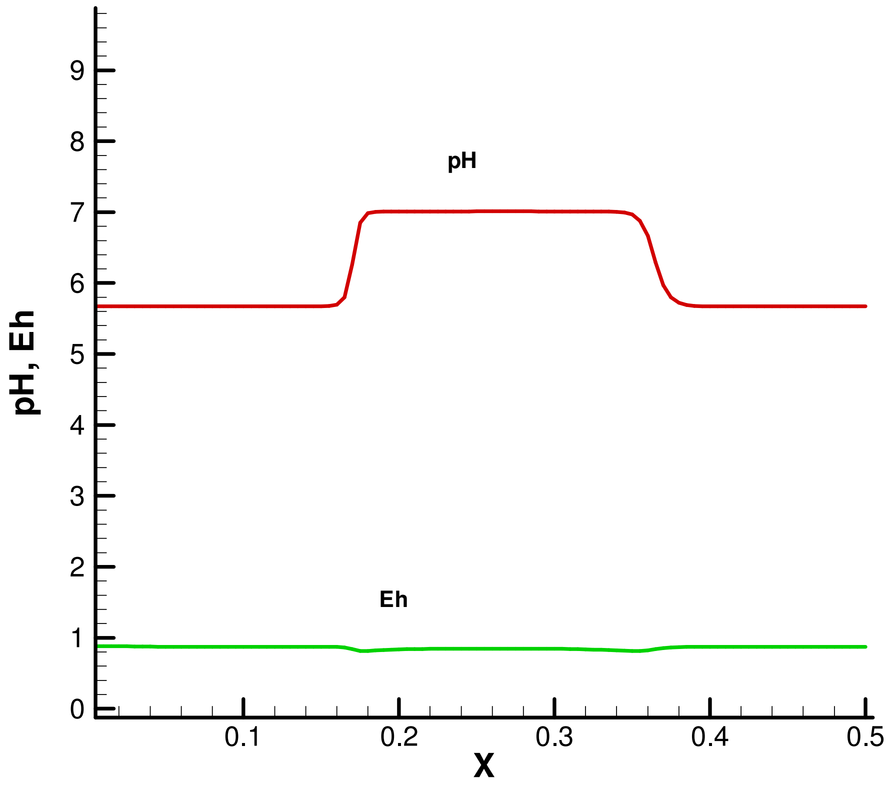
UO2(s) dissolution and transport



UO2(s) dissolution and transport

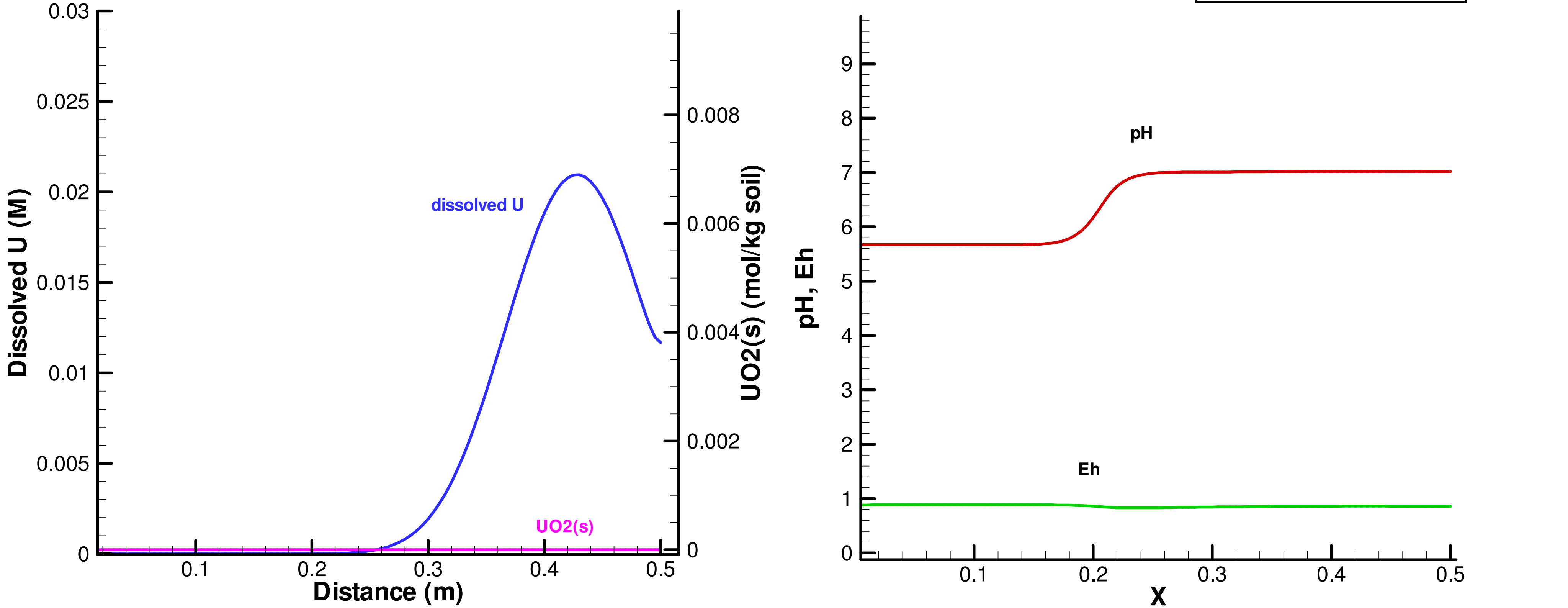
1110 sec





UO2(s) dissolution and transport

2e4 sec



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Outlook

- Implementation of coupled effect of volumetric changes due to chemical reaction on HM-behaviour
- Corrosion of HLW container and gas production
- Performance assessment: Application to real systems
- Paralel computing