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Flow, transport and chemistry in porous media : numerical methods for coupled problems

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1. Motivations

2. Basic models and methods
   - Flow model
   - Transport model
   - Chemistry

3. Reactive transport
   - Single species with sorption (joint work with A. Taakili)
   - Multi-species equilibrium chemistry

Outline

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Nuclear waste storage (1)

- Assess safety of deep geological nuclear waste storage (clay layer)
- Long term simulation of radionuclide transport (one million years)
- Wide variation of scales : from package (meter) to regional (kilometers)
- Geochemistry: large number of species
- Strong government regulation
- Main actors : ANDRA, cea, EDF
- Research in mathematical and numerical modeling is conducted in the CNRS MOMAS group (Director A. Ern).

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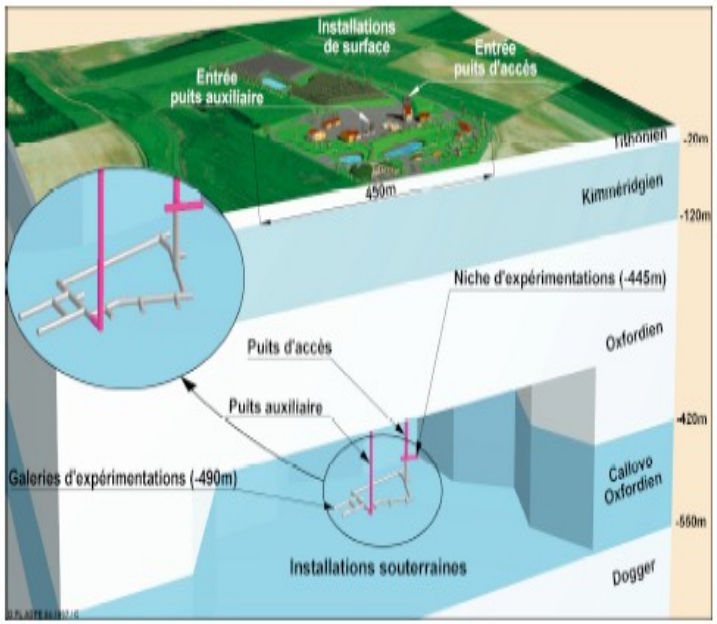
Nuclear waste storage (2)

Present choice in France: a sedimentary geological formation (in the Meuse region)



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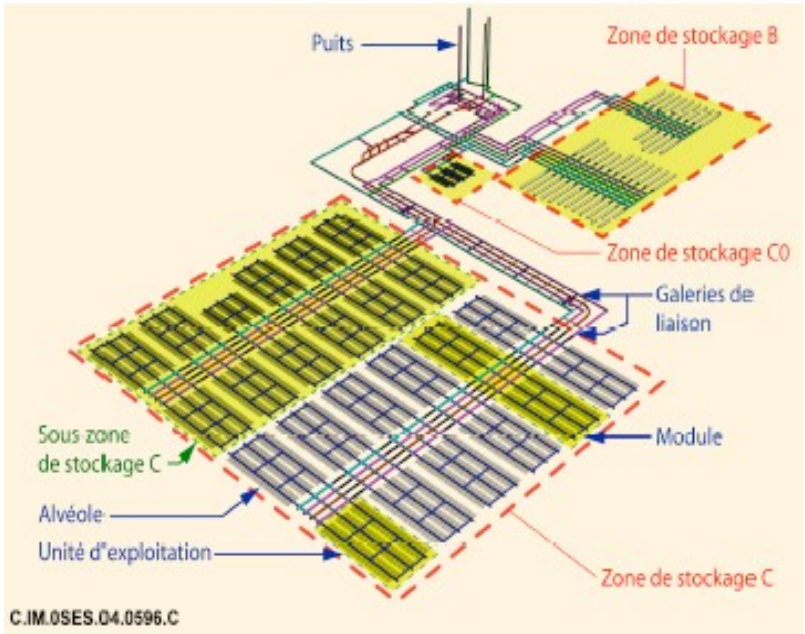

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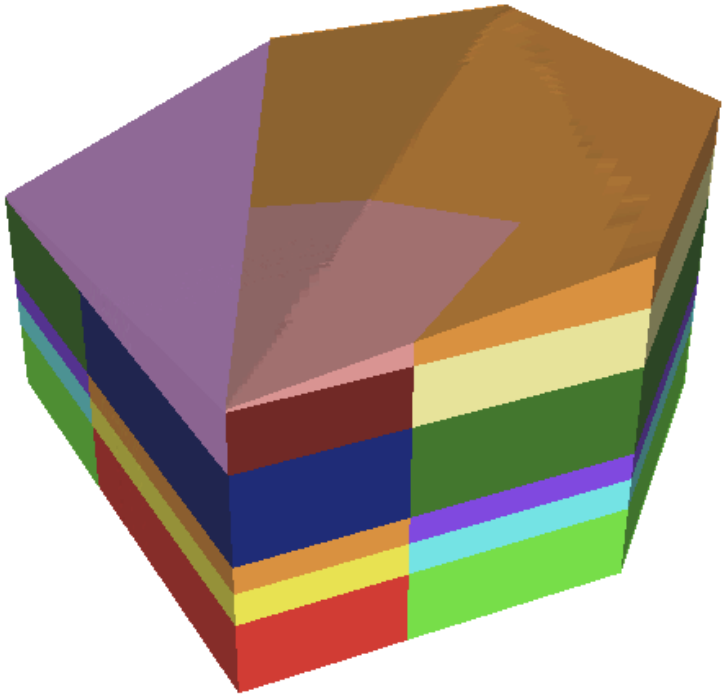
A 3D far field model

Used as a benchmark, similar to Andra safety model



Blown-up 30 times vertically

Difficulties

- Distorted geometry (horizontal $\approx$ 40 km, vertical 700 m)
- Strong heterogenities (permeability varies by 8 orders of magnitude)
- General hexahedral mesh
- Simulation over 500 000 years

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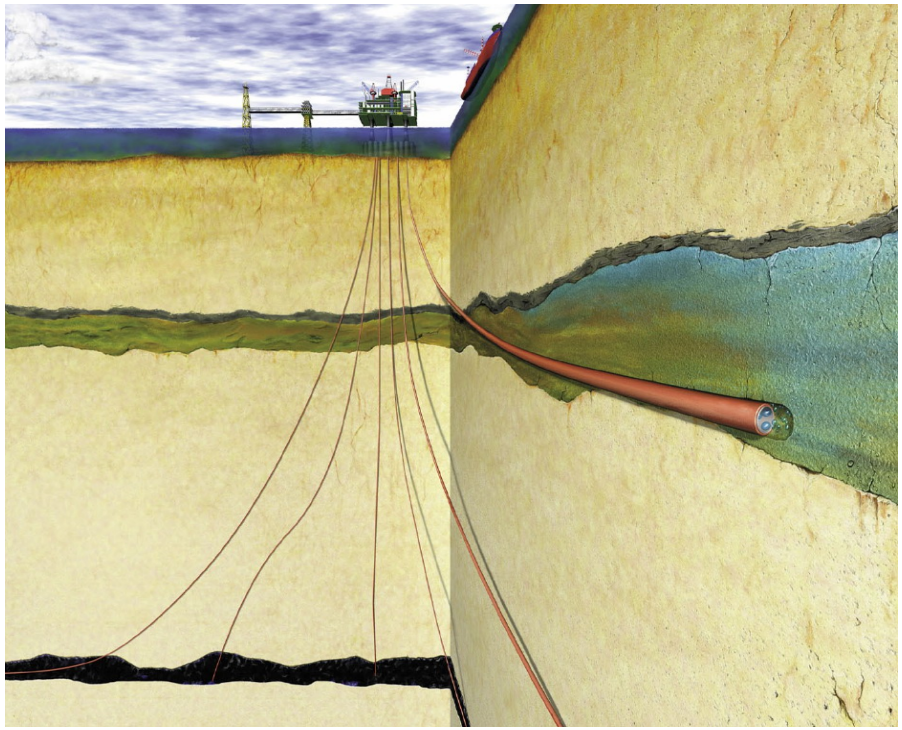
CO_2 sequestration



Sleipner project, Norway

- Long term capture of CO_2 in saline aquifer
- Simulation to understand CO_2 migration through salt
- Coupling of liquid and gas phase, reactive transport

SHPCO project (funded by ANR) High Performance Simulation of CO2 sequestration

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Darcy's law and mixed finite element

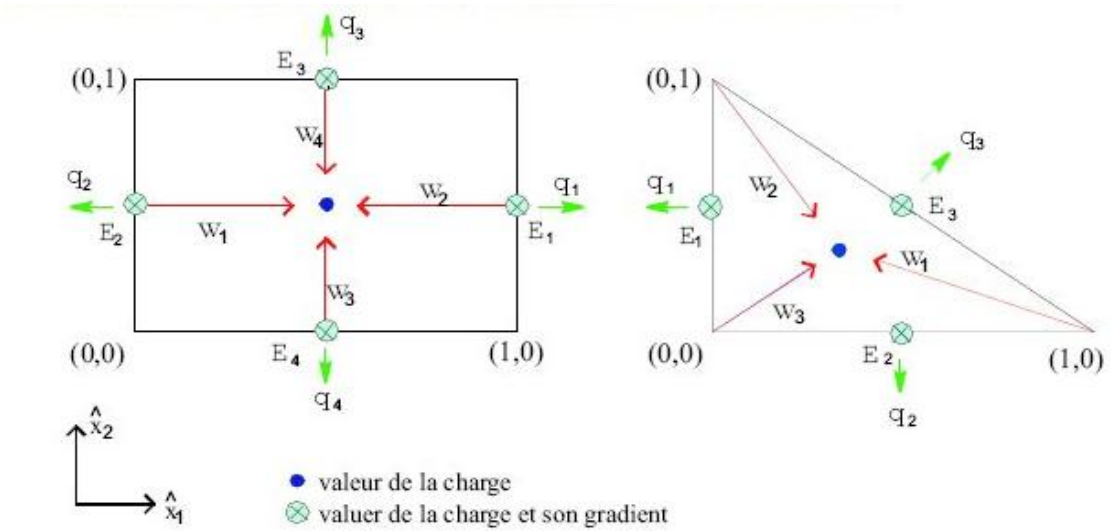
Flow equations

$q = -K\nabla h$ Darcy's law h piezometric head

$\nabla \cdot q = 0$ incompressibility q Darcy velocity

Mixed finite elements

- Approximate both head and velocity
- Continuous flux across element faces
- Locally mass conservative
- Allows full diffusion tensor

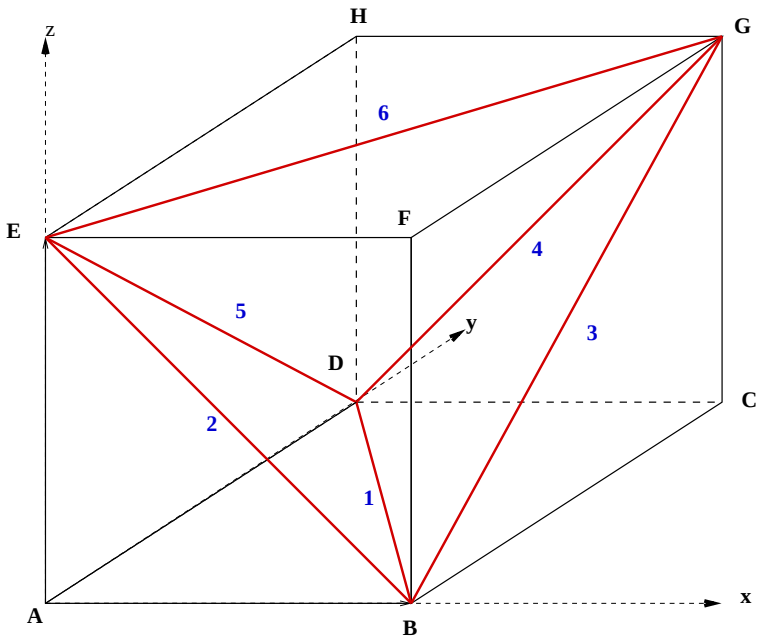


A composite mixed finite element for hexahedra (1)

The problem

Standard convergence theory not valid for RTN space over general (deformed) hexahedra, pressure space does not containt constant functions (T. Russell)

Kuznetsov, Repin (2003): construct macroelement on a hexahedron by subdividing it into 5 tetrahedra



A composite mixed finite element for hexahedra (2)

Features

- Same DOFs as before (average pressure in each element, flux across each face)
- Standadrd error estimates shown to apply: optimal order error (under regularity assumption).

Construction

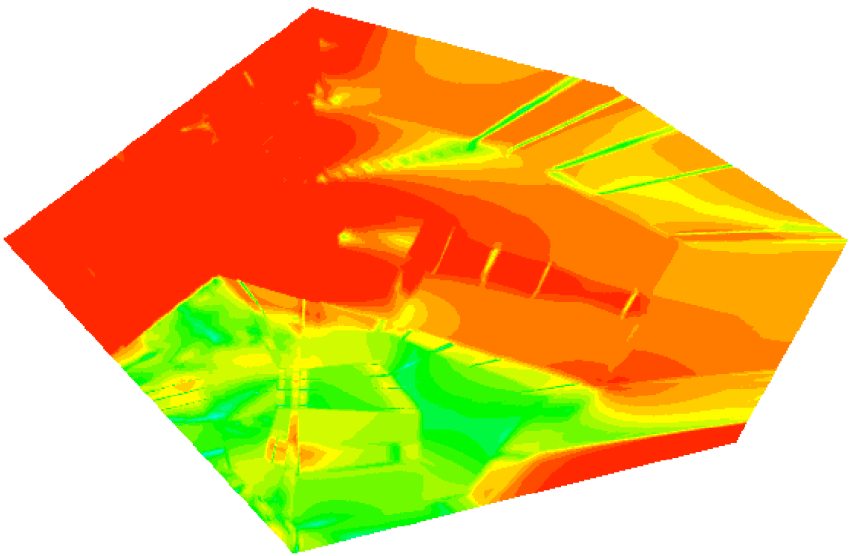
$$\mathcal{W}_h = \left\{ \mathbf{v}_h \in H(\mathrm{div}, \Omega);\ \mathbf{v}_{h|T_i} \in RTN_0(T_i), i = 1, \cdots, 5, \right.$$
$$\left. \mathrm{div}\, \mathbf{v}_h \text{ const. on } H,\ \mathbf{v}_h \cdot \mathbf{n} \text{ constant on faces of } H \right\}.$$

$\mathbf{v}_{hH}$ is uniquely defined by its normal components across the 6 faces, so $\mathcal{W}_h$ contains constants.

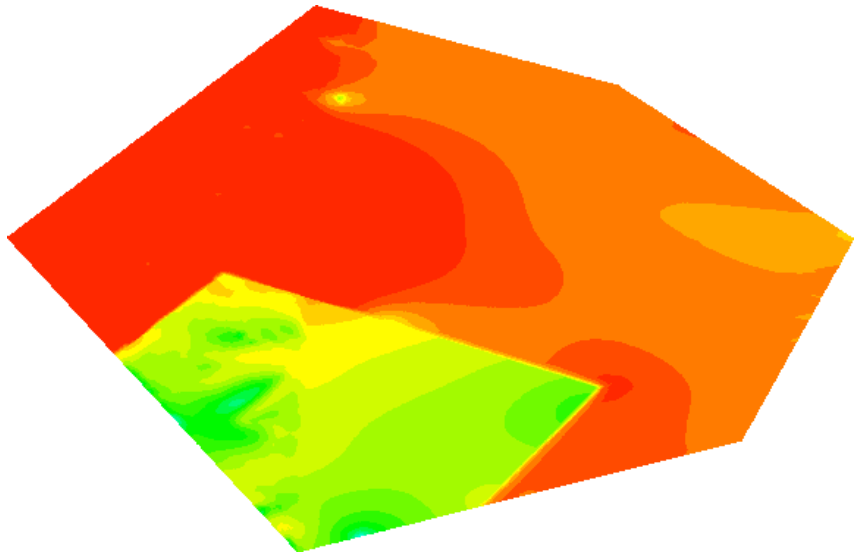
A. Sboui's PhD thesis, Sboui, Jaffr , Roberts to appear in SIAM J. SCi. Comp.

A composite mixed finite element for hexahedra (3)

Simulation for 3D far field benchmark model, horizontal cros section of modulus of velocity



RTN FE



New FE

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Transport model

Convection–diffusion equation

$$\omega \frac{\partial c}{\partial t} - \underbrace{\operatorname{div}(\mathbf{D} \operatorname{grad} c)}_{\text{dispersion}} + \underbrace{\operatorname{div}(\mathbf{u} c)}_{\text{advection}} + \omega \lambda c = f$$

- c: concentration $[\mathrm{mol/l}]$
- ω: porosity [–]
- λ radioactive decay $[\mathrm{s}^{-1}]$
- $\mathbf{u}$ Darcy velocity $[\mathrm{m/s}]$

Dispersion tensor

$$\mathbf{D} = d_e \mathbf{I} + |\mathbf{u}|[\alpha_l \mathbf{E}(\mathbf{u}) + \alpha_t (I - \mathbf{E}(\mathbf{u}))], \quad E_{ij}(\mathbf{u}) = \frac{u_i u_j}{|\mathbf{u}|}$$

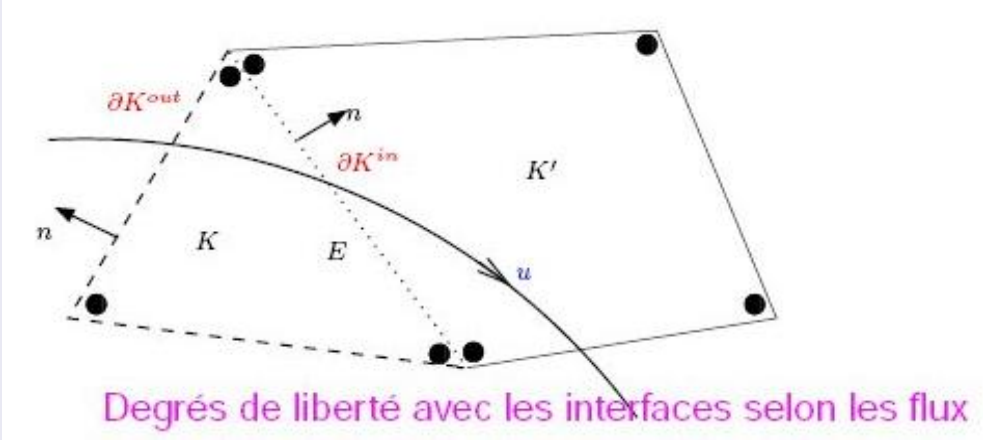
α_l, α_t dispersicity coeff. $[\mathrm{m}]$, d_e molecular diffusion $[\mathrm{m/s^2}]$

Solution by operator splitting

Advection step

Explicit, finite volumes / discontinuous Galerkin

- Locally mass conservative
- Keeps sharp fronts
- Small numerical diffusion
- Allows unstructured meshes
- CFL condition: use sub–time–steps



Degrés de liberté avec les interfaces selon les flux

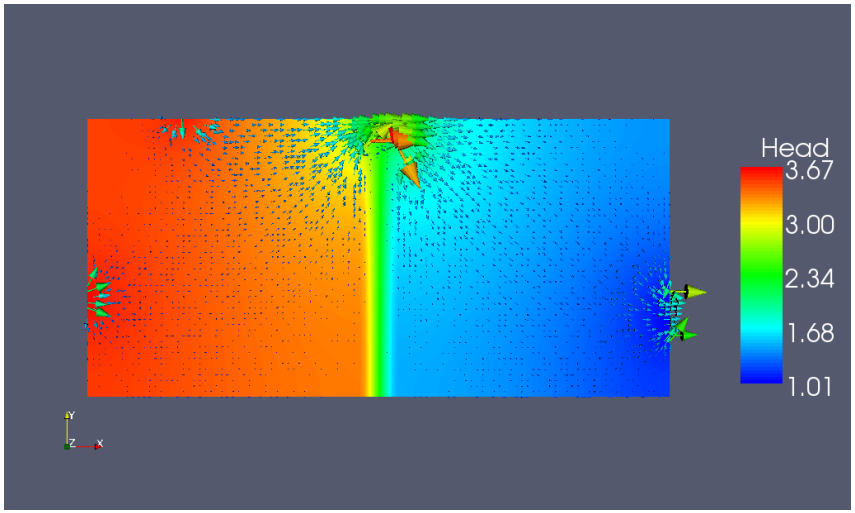
Dispersion step

Like flow equation (time dependant): mixed finite elements (implicit)

First order method

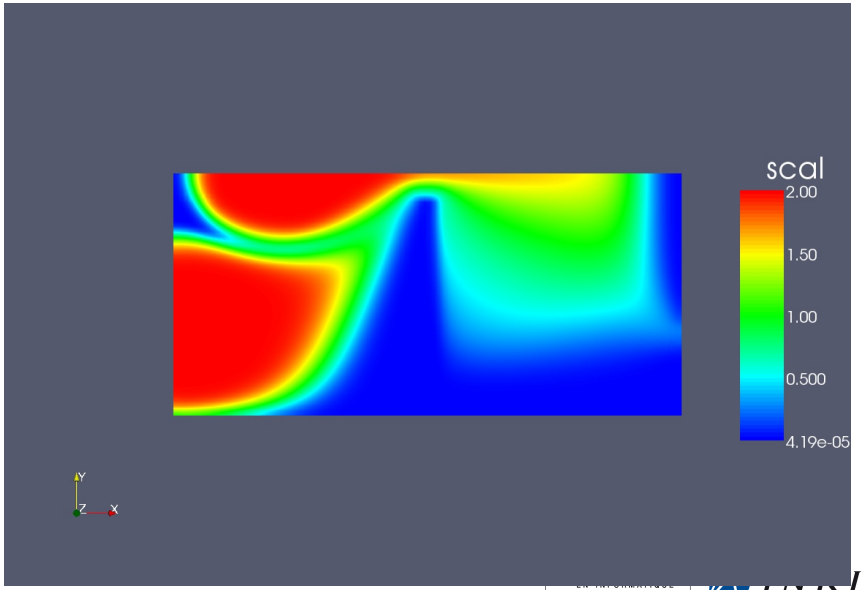
Example: transport around an obstacle

MoMaS benchmark for reactive transport. Here transport only



Head and velocity

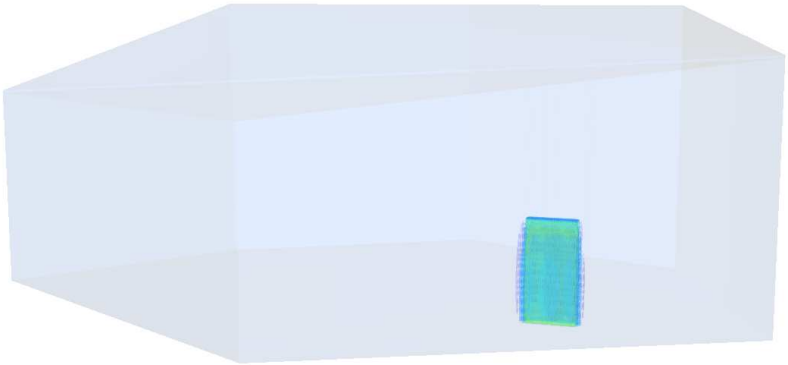
Concentration at $t = 25$

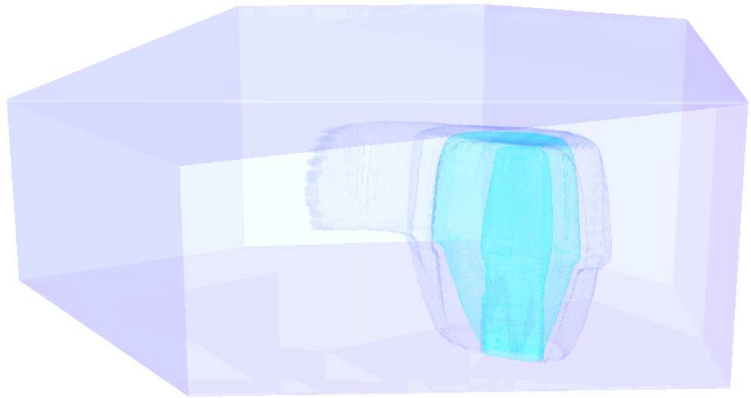


J. B. Apoung, P. Hav, J. Houot, MK, A. Semin, O. Saouli

Transport around a nuclear waste storage site

GdR MoMaS benchmark, Andra model





Concentration at 130 000 years

Concentration at 460 000 years

A. Sboui, E. Marchand (INRIA, Estime)

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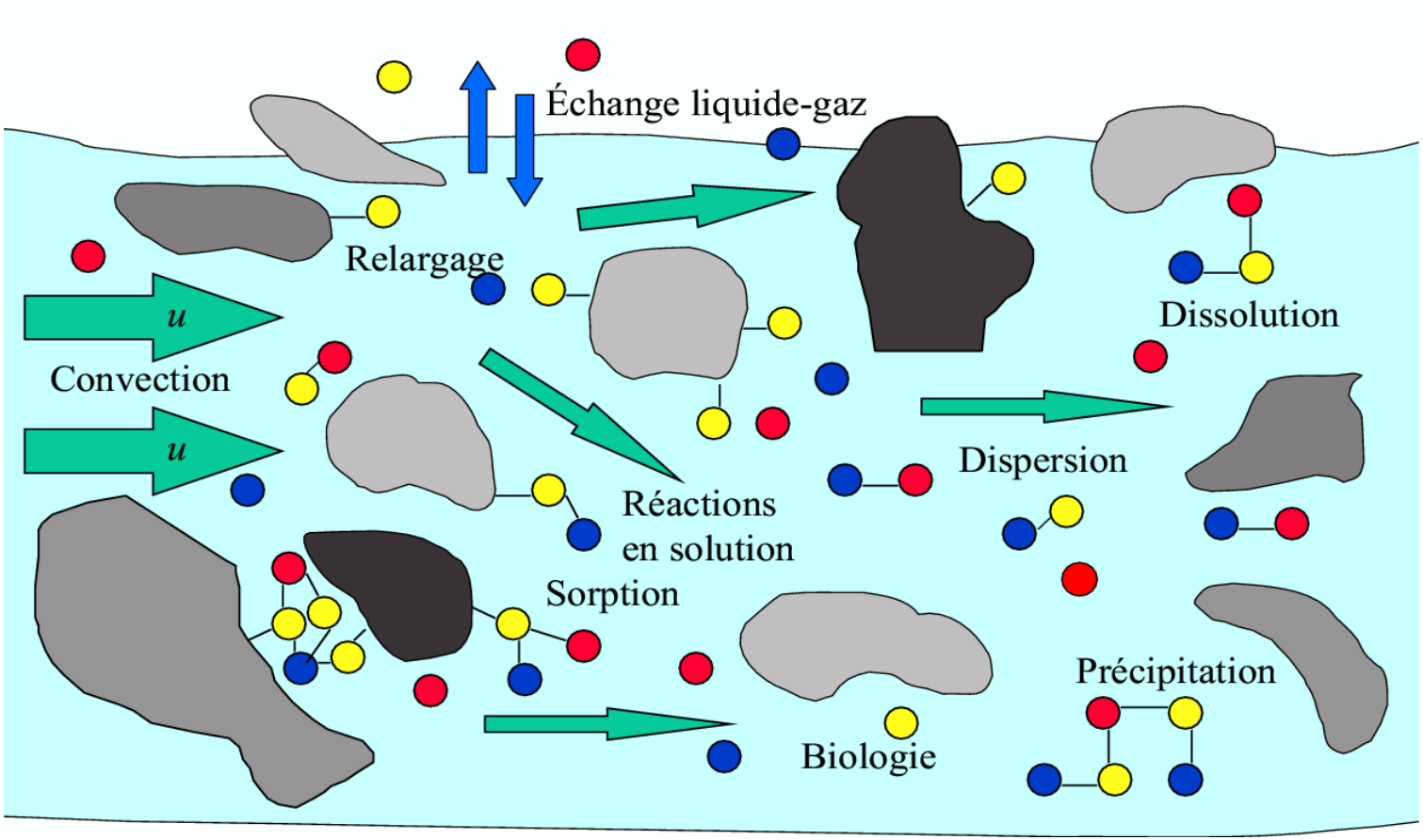
Chemical phenomena



Classification of chemical reactions

According to nature of reaction

Homogeneous	In the same phase (aqueous, gaseous, ...) Examples: Acid base, oxydo–reduction
Heterogeneous	Involve different phases Examples: Sorption, precipitation / dissolution, ...

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Slow reactions	Irreversible, modeled using kinetic law
Fast reactions	Reversible, modeled using equilibrium

Depends on relative speed of reactions and transport.

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In this talk: Equilibrium reactions, with sorption.

Sorption processes

Definition

Sorption is the accumulation of a fluid on a solid at the fluid–solid interface.

Main mechanism for exchanges between dissolved species and solid surfaces.

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Several possible mechanisms

Surface complexation Formation of bond between surface and aqueous species, due to electrostatic interactions. Depends on surface potential.

Ion exchange Ions are exchanged between sorption sites on the surface. Depends on Cationic Exchange Capacity.

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Can be modeled as mass action law

Modeling general equilibrium models

General chemical reactions : N_s species, N_r reactions

$$\sum_{j=1}^{N_s} \nu_{ij} Y_j \leftrightarrows 0, \quad i = 1, \ldots, N_r$$

ν_{ij} stoichiometric coefficients. Matrix equation $\nu Y = 0$

Modeling general equilibrium models

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Basis for null-space of ν has dimensions $N_c = N_s - N_r$.

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Partition $\nu = \begin{pmatrix} G & N \end{pmatrix}$, $B \in \mathbf{R}^{N_r \times N_r}$ invertible, $N \in \mathbf{R}^{N_c \times N_r}$. Let $H = -G^{-1}N$

General solution of $\nu Y = 0$: $Y = \begin{pmatrix} x \\ c \end{pmatrix}$, $x = Hc$. $c \in \mathbf{R}^{N_c}$, $x \in \mathbf{R}^{Nr}$.

The chemical problem

c (resp $\bar{c}$) mobile (resp. fixed) primary species
x (resp $\bar{x}$) mobile (resp. fixed) secondary species

System of non-linear equations

$$\left.\begin{aligned} c + S^T x + A^T \bar{x} &= T, \\ \bar{c} + B^T \bar{x} &= W, \end{aligned}\right\} \quad \text{Mass conservation}$$

$$\left.\begin{aligned} \log x &= S \log c + \log K, \\ \log \bar{x} &= A \log c + B \log \bar{c} + \log \bar{K}. \end{aligned}\right\} \quad \text{Mass action law}$$

Role of chemical model

Given T (and W, known), split into mobile C and fixed F concentrations.

$$C = c + S^T x = \Phi(T), \qquad F = A^T \bar{x} = \Psi(T)$$

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Sorption models

One species reacts with rock matrix, description by a sorption isotherm : $v = \Psi(u)$.

u aqueous concentration, v fixed "concentration"

Common isotherms

Linear $v = K_d u$

Langmuir $v = \dfrac{k_f \sigma_0 u}{k_f u + k_b}$

Freundlich $v = \gamma u^{1/p}$ $(p > 1$ possible$)$

Coupled model

$$\omega \frac{\partial u}{\partial t} + \omega \frac{\partial v}{\partial t} + Lu = 0, \quad L \text{ adv. diff operator}$$
$$v = \Psi(u).$$

Mathematical, numerical analysis: van Duijn, Knabner, Barrett, Kacur, Frolkovic

Formulations of coupled system

After space and time discretization,

Coupled formulation $F(u,v)=0$ with

$$F\begin{pmatrix}u\\v\end{pmatrix}=\begin{pmatrix}(M+\Delta L)u+Mv+b\\v-\Psi(u)\end{pmatrix}$$

Eliminate v $F_1(u)=(M+\Delta tL)u+M\Psi(u)-b^n$

Eliminate u $F_2(u)=v-\Psi\left((M+\Delta L)^{-1}(b-Mv)\right)$

Jacobian for coupled formulation,with $D=\mathrm{diag}(\Psi'(u_1),\ldots,\Psi'(u_N))$

$$J=\begin{pmatrix}M+\Delta L & M\\ -D & I\end{pmatrix}$$

$J_2=I+D(M+\Delta tL)^{-1}M$ is Schur complement of J

Solution by Newton–Krylov

- Solve the linear system by an iterative method (GMRES)
- Requires only jacobian matrix by vector products.

Used for CFD, shallow water, radiative transfer(Keyes, Knoll, JCP 04), and for reactive transport (Hammond et al., Adv. Wat. Res. 05)

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Inexact Newton

- Approximation of the Newton's direction $\|f'(x_k)d + f(x_k)\| \leq \eta \|f(x_k)\|$
- Choice of the forcing term η?
  - Keep quadratic convergence (locally)
  - Avoid oversolving the linear system
- $\eta = \gamma \|f(x_k)\|^2 / \|f(x_{k-1})\|^2$ (Kelley, Eisenstat and Walker)

Preconditioning

- Essential for good linear performance
- Difficult for matrix free formulation

Possible choices

Block diagonal $P = \mathrm{diag}\,(M + \Delta t L, I)$,

Block Gauss Seidel "Physics based", equivalent to (lienar) sequential method

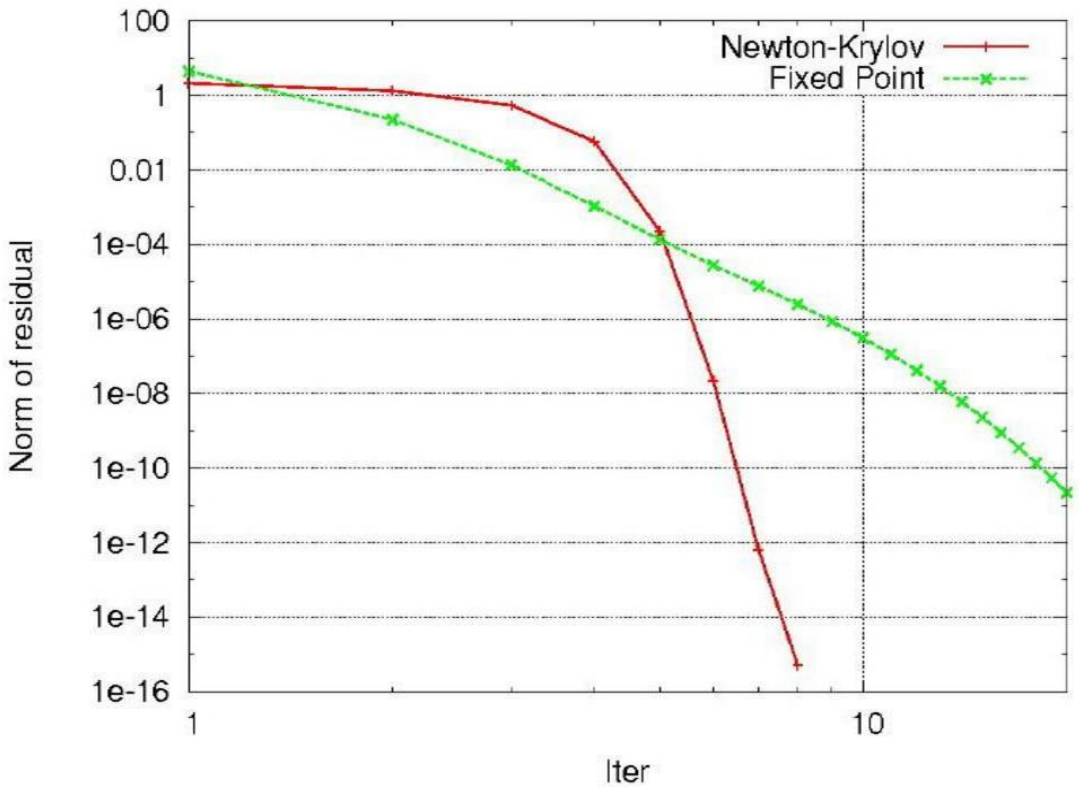
$$P = \begin{pmatrix} M + \Delta t L & 0 \\ -D & I \end{pmatrix}$$

Aproximate block factorization ?

Numerical results

Geometry of MoMaS reactive transport benchmark (2D), LifeV + Kinsol (Sundials)

σ_0	0.125	0.25	0.4	0.45	0.5
nb nonlin iter	5	6	6	6	7
nb lin. iter	12	19	27	43	86



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The coupled system

Transport for each species (same dispersion tensor for all species)
Eliminate (unknown) reaction rates by using conservation laws ($T = C + F$)

$$\omega \frac{\partial T^{ic}}{\partial t} + L(C^{ic}) = 0, \quad ic = 1, \ldots, N_c$$

$$T^{ic}_{ix} = C^{ic}_{ix} + F^{ic}_{ix} \qquad ic = 1, .., N_c \text{ and } ix = 1, .., N_x$$

$$F_{ix} = \Psi(T_{ix}) \qquad ix = 1, \ldots, N_x.$$

Coupling methods

Iterative, based on fixed point (Yeh Tripathi '89, Carrayrou et al. '04)

Substitution, global (Saaltink '98, Hammond et al. '05)

Reduction method (Knabner, Kratle, '06)

Coupling formulations and algorithms(1)

CC formulation, explicit chemistry

$$\begin{cases} \omega \dfrac{dC}{dt} + \dfrac{dF}{dt} + LC = 0 \\ H(z) - \begin{pmatrix} C+F \\ W \end{pmatrix} = 0 \\ F - F(z) = 0. \end{cases}$$

- $+$ Explicit Jacobian
- $+$ Chemistry function, no chemical solve
- $-$ Intrusive approach (chemistry not a black box)
- $-$ Precipitation not easy to include

Coupled system is index 1 DAE

$$K\frac{dy}{dt} + f(y) = 0$$

Use standard DAE software

C. de Dieuleveult (Andra thesis), J. Erhel, MK (JCP '09)

Coupling formulations and algorithms(2)

TC formulation, implicit chemistry

$$\begin{cases} \omega \dfrac{dT}{dt} + LC = 0 \\ T - C - F = 0 \\ F - \Psi(T) = 0 \end{cases}$$

- $+$ Non-intrusive approach (chemistry as black box)
- $+$ Precipitation can (probably) be included
- $-$ One chemical solve for each function evaluation

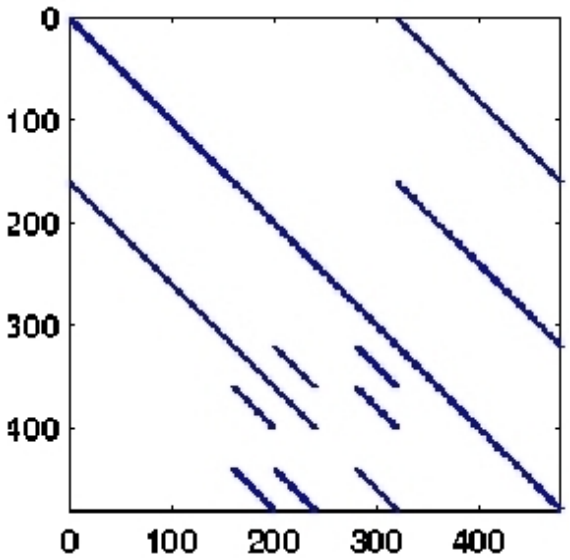
$$\begin{cases} C^{n+1} = (M + \Delta t L)^{-1} \left(C^n + F^n - F^{n+1} \right) \\ T^{n+1} = C^{n+1} + F^{n+1} \\ F^{n+1} = \Psi(T^{n+1}) \end{cases}$$

Fixed point problem, can be solved by block Gauss Seidel or by Newton's method

Solution by Newton–Krylov

Structure of Jacobian matrix

$$f'(C,T,F) = \begin{pmatrix} I & 0 & (I+\Delta t L)^{-1} \\ -I & I & -I \\ 0 & -\Psi'(T) & I \end{pmatrix}$$



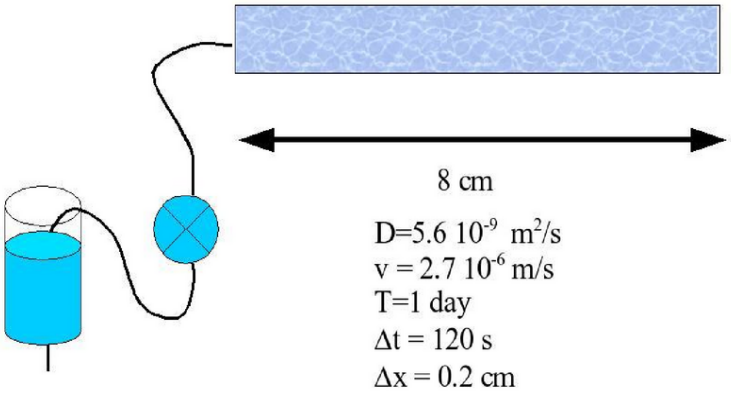
- Transport independant for each species
- Chemistry independant for each grid cell

Find a good preconditioner ?

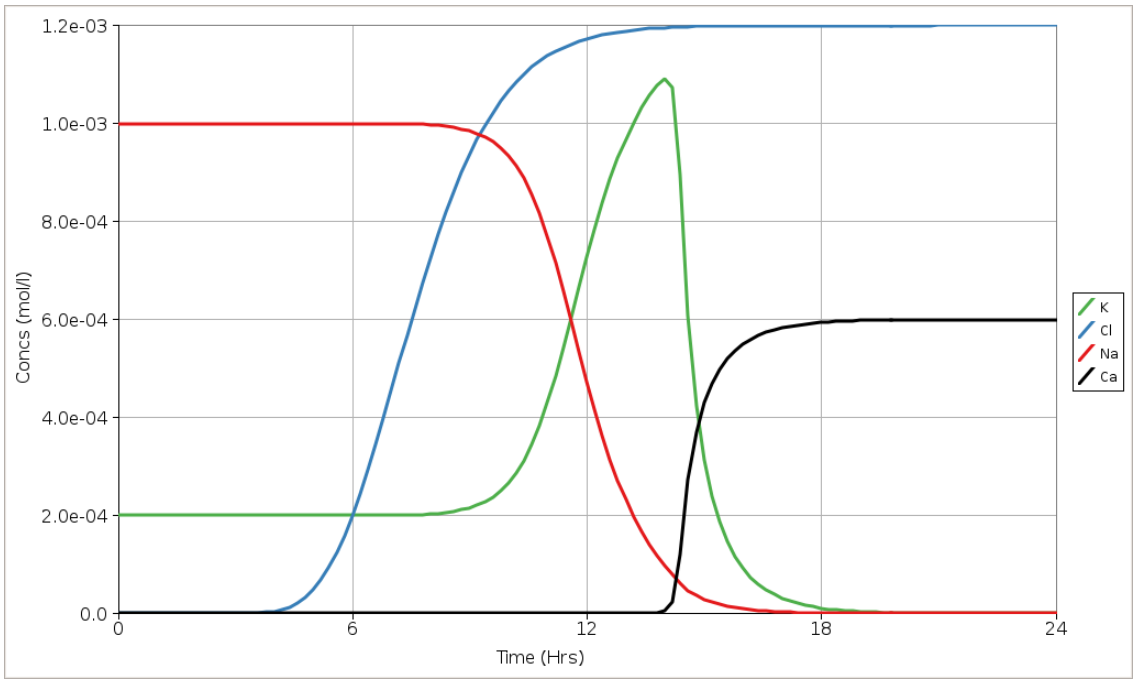
- Is block diagonal good enough ?
- Physics based (cf Hammond et al.) ?

Example: ion exchange

Column experiment (Phreeqc ex. 11, Alliances ex. 3)



Column contains a solution with $1\,\mathrm{mmol\,Na}$, $0.2\,\mathrm{mmol\,K}$ and $1.2\,\mathrm{mmol\,NO_3}$. Inject solution with $1.2\,\mathrm{mmol\,CaCl_2}$. $CEC = 1.1\,10^{-3}$.



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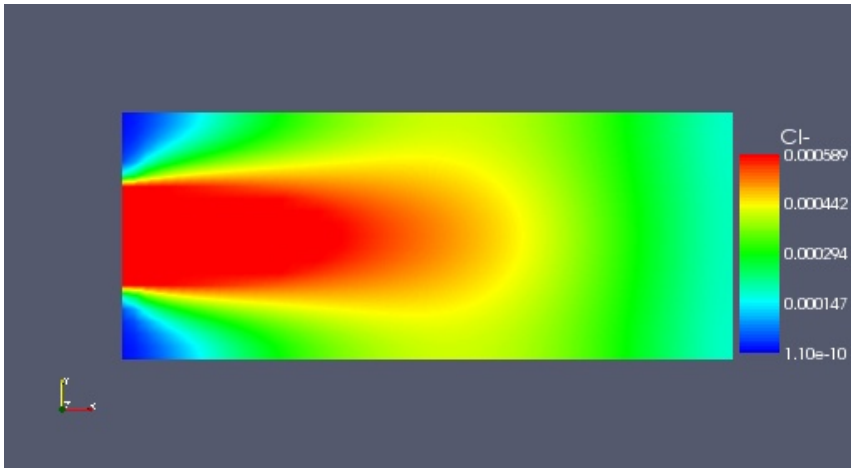
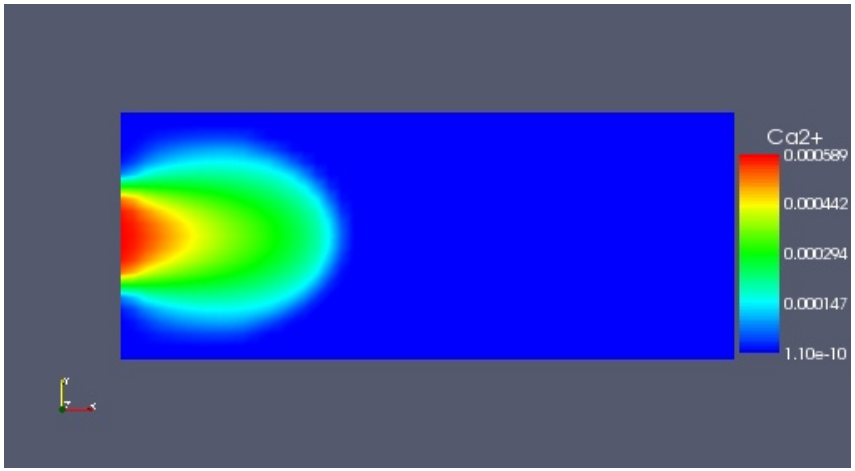
Ion exchange example (ctd)

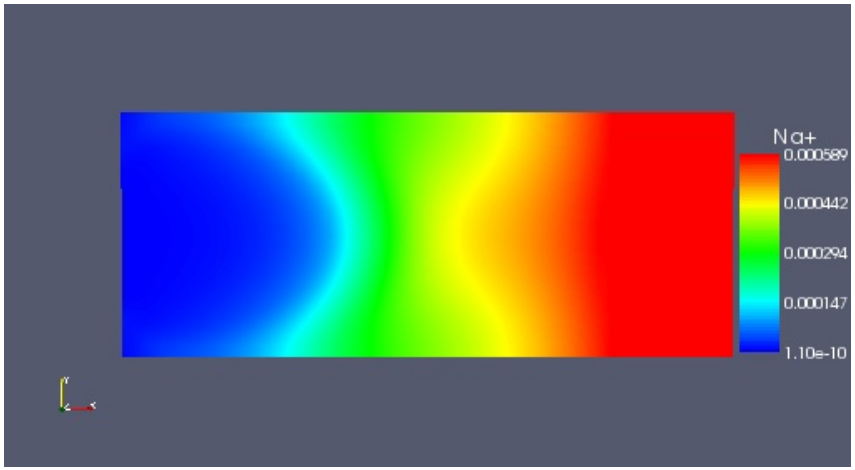
Snapshots at $t = 35$



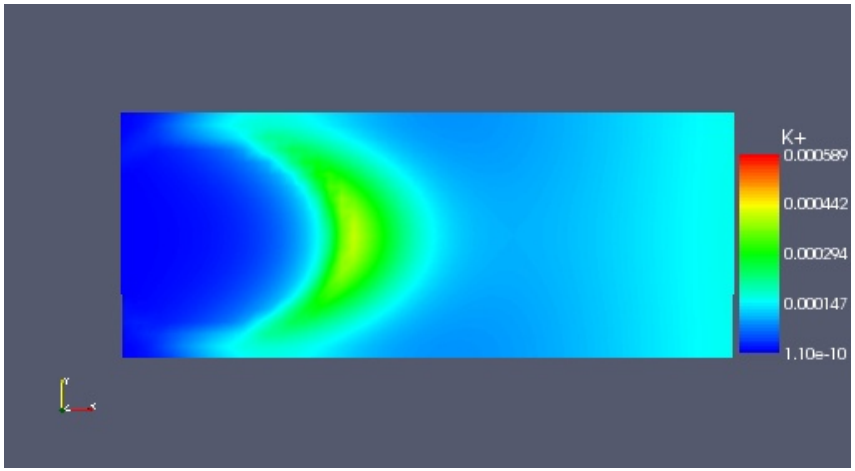
Cl



Ca



Na



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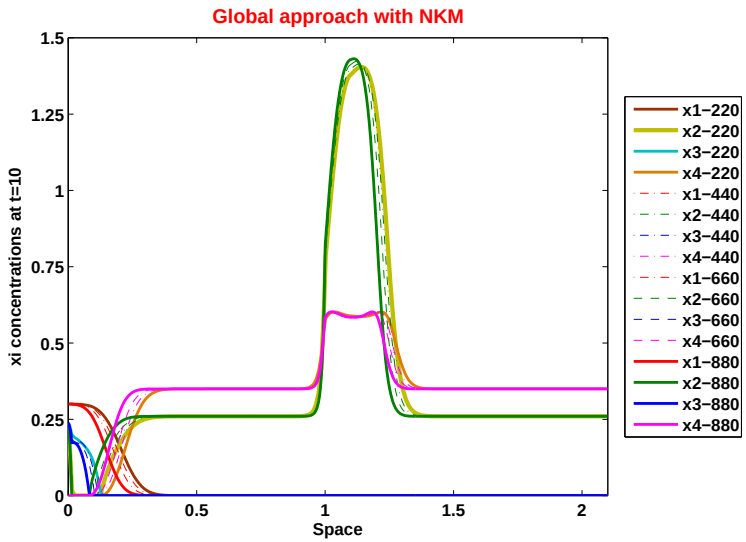
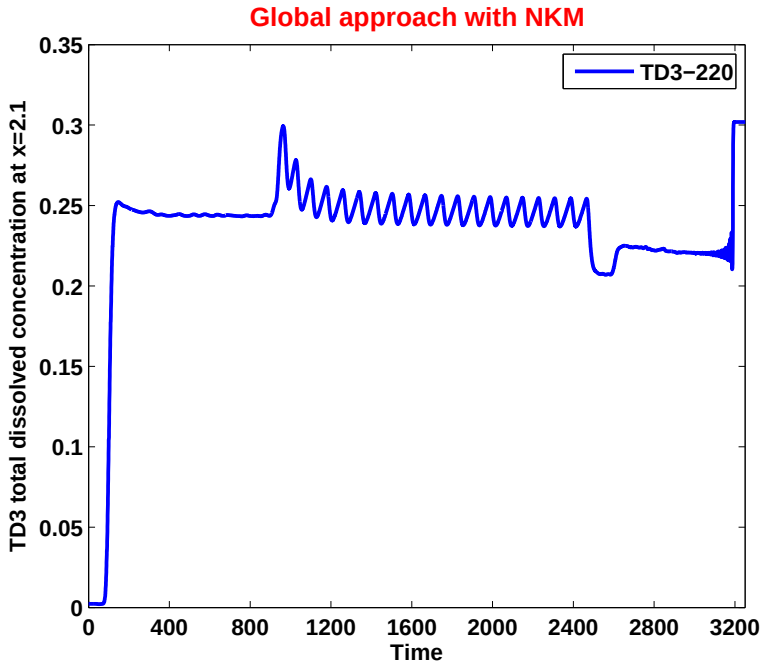
MoMaS benchmark

Difficult test case, heterogenous medium, with complex chemistry (even for “easy” level).
12 species, eq. constants vary by 45 orders of magnitude (1D, Matalb code, L.Amir’s thesis).





Conclusions – perspectives

- Robust methods for solving flow and transport in porous media
- Preliminary results for reactive transport
- Newton–Krylov promising framework, implementation in progress

- Move to two-phase (multiphase) flows (water and gas)
- Transport in fractured media
- For chemistry, take into account “real” phenomena (minerals, kinetics,...)