Flow and transport of pollutants in the subsurface: coupled models and numerical methods

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Outline

1 Motivations

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

3 Coupled models
   - Density driven flow
   - Reactive transport
Plan

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Contaminant transport

Underground water

- 22% of all natural water resources
- 51% of all drinking water
- 37% of agricultural water

Possible contamination of groundwater by industrial waste

Microbial remediation

Variant: saltwater intrusion: coupling to flow
Nuclear waste storage

- Assess safety of deep geological nuclear waste storage (clay layer)
- Long term simulation of radionuclide transport
- Wide variation of scales: from package (meter) to regional (kilometers)
- Geochemistry: large number of species
CO₂ sequestration

- Long term capture of CO₂ in saline aquifer
- Simulation to understand CO₂ migration through salt
- Coupling of liquid and gas phase, reactive transport

Sleipner project, Norway
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Flow: Darcy’s law

Henry Philibert Gaspard Darcy, (1803-1858) French engineer

Darcy’s law

\[ Q = AK \frac{\Delta h}{L} \]

- \( Q \) flow (m\(^3\)/s)
- \( K \) Hydraulic conductivity (m/s)
- \( h \) Piezometric head (m)
  
  \( h = \frac{p}{\rho g} + z \)
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Modern, differential version

\[ q = -K \nabla h, \quad q \text{ Darcy velocity} \]

Flow equations

\[ \nabla \cdot q = 0 \quad \text{incompressibility} \]
Mixed finite elements

- Approximate both head and Darcy velocity
- Locally mass conservative
- Flux is continuous across element faces
- Allows full diffusion tensor

Raviart-Thomas space
Flow around nuclear waste storage area

Computed by domain decomposition (Robin–Robin)

Subdomain code in C++ (LifeV), interface solver in Ocaml

Parallelism in OcamlP3l (skeleton based)

F. Clément, V. Martin (thesis), P. Weis (INRIA, Estime)
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Physics of advection–dispersion

**Convection**  Transport by velocity field

**Diffusion**  motion due to concentration gradient

**Dispersion**  due microscopic velocity heterogeneity

**Reaction**  between species, interaction with host matrix
Transport model

Convection–diffusion equation

\[ \omega \frac{\partial c}{\partial t} - \text{div}(D \text{grad } c) + \text{div}(uc) + \omega \lambda c = f \]

- **c**: concentration [mol/l]
- **\(\omega\)**: porosity (–)
- **\(\lambda\)**: radioactive decay [s\(^{-1}\)]
- **\(u\)**: Darcy velocity [m/s]

Dispersion tensor

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

- \(\alpha_l, \alpha_t\): dispersivity coeff. [m], \(d_e\): molecular diffusion [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

**Dispersion step**

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

Order 1 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. Havé, J. Houot, MK, A. Semin
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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Classification of chemical reactions

According to nature of reaction

**Homogeneous**  In the same phase (aqueous, gaseous, ...)
Examples: Acid base, oxydo–reduction

According to speed of reaction

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

Depends on relative speed of reactions and transport.

In this talk: Equilibrium reactions, with sorption.
## Classification of chemical reactions

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In this talk: Equilibrium reactions, with sorption.
**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.
Sorption processes

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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.
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Can be modeled as mass action law
The chemical problem

System of non-linear equations

\[
\begin{align*}
  &c + S^T x + A^T y = T, \\
  &\bar{c} + B^T \bar{x} = W,
\end{align*}
\]

\[
\begin{align*}
  \log x &= S \log c + \log K_x, \\
  \log \bar{x} &= A \log c + B \log \bar{c} + \log K_y.
\end{align*}
\]

Mass conservation

Mass action law

Dissolved total: \( C = c + S^T x \), Fixed total: \( F = A^T \bar{x} \).

Role of chemical model

Given totals \( T \) (and \( W \), known), split into mobile and immobile total concentrations.

\[
C = \Phi(T), \quad F = \Psi(T)
\]
Numerical solution of chemical problem

Take concentration logarithms as main unknowns
Use globalized Newton’s method (line search, trust region).

Ion exchange: 6 species, 4 components (vary initial guess)
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Motivations

Seawater intrusion can threaten drinking water reservoir

Synthetic model (Elder): fingering instability

\[ \rho = 1200 \text{ kg m}^{-3} \]

\[ k = 4.8 \times 10^{-13} \text{ m}^2 \]

Simulated concentration at \( t = 20 \text{ years} \) for the Elder benchmark
Physical model

Flow

Mass conservation for fluid
\[ \frac{\partial (\rho \omega)}{\partial t} + \nabla \cdot (\varepsilon \rho \vec{V}) = \rho Q_S, \]

Generalized Darcy’s law
\[ \varepsilon \vec{V} = -\frac{1}{\mu} K (\nabla P + \rho g \hat{n}_z), \]

Equation of state
\[ \rho = \rho_0 + \frac{\partial \rho}{\partial C_m} C_m, \quad \rho_0 = 1000, \quad \frac{\partial \rho}{\partial C_m} = 200. \]

Transport

Salt mass conservation
\[ \varepsilon \rho \frac{\partial C_m}{\partial t} + \varepsilon \rho \vec{V} \cdot \nabla C_m = \nabla \cdot (\varepsilon \rho D(\vec{V}) \nabla C_m), \]

Dispersion tensor
\[ D(\vec{V}) = D_m I + (\alpha_L - \alpha_T) \frac{\vec{V} \otimes \vec{V}}{||\vec{V}||} + \alpha_T ||\vec{V}|| I \]
Distributed implementation

Coupling between flow and transport:

Flow code

Controller

Transport code

Use Corba for coupling components
Joint work with J. Erhel, Ph. Ackerer, Ch. Perez, M. Mancip
Elder model
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The coupled system

Transport for each species (same dispersion tensor for all species)

\[
\begin{align*}
\frac{\partial x_i}{\partial t} + L(x_i) &= r^x_i, \\
\frac{\partial c_j}{\partial t} + L(c_j) &= r^c_j, \\
\frac{\partial y_i}{\partial t} &= r^\gamma_i, \\
\frac{\partial s_j}{\partial t} &= r^s_j,
\end{align*}
\]
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\frac{\partial y_i}{\partial t} = r^y_i, \quad \frac{\partial s_j}{\partial t} = r^s_j,
\]

Eliminate (unknown) reaction rates by using conservation laws \((T = C + F)\)

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

\[
T^{ic}_{ix} = C^{ic}_{ix} + F^{ic}_{ix} \quad ic = 1, \ldots, N_c \text{ and } ix = 1, \ldots, N_x
\]

\[
F_{ix} = \Psi(T_{ix}) \quad ix = 1, \ldots, N_x.
\]

Number of transport equations reduced from \(N_x + N_y\) to \(N_c + N_s\)
Coupling formulations and algorithms

CC formulation, explicit chemistry

\[ \frac{dC}{dt} + \frac{dF}{dt} + LC = 0 \]
\[ H(z) - \left( \begin{array}{c} C + F \\ W \end{array} \right) = 0 \]
\[ F - F(z) = 0. \]

Coupled system is index 1 DAE

\[ M \frac{dy}{dt} + f(y) = 0 \]

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

Use standard DAE software

J. Erhel, C. de Dieuleveult (Andra thesis)
**TC formulation, implicit chemistry**

\[
\begin{align*}
\frac{dT}{dt} + L C &= 0 \\
T - C - F &= 0 \\
F - \Psi(T) &= 0
\end{align*}
\]

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

\[
\begin{align*}
C^{n+1} - C^n &= \frac{F^{n+1} - F^n}{\Delta t} \quad + \quad L(C^{n+1}) = 0 \\
T^{n+1} &= C^{n+1} + F^{n+1} \\
F^{n+1} &= \Psi(T^{n+1})
\end{align*}
\]

Solve by **Newton’s method**
Solution by Newton–Krylov

Structure of Jacobian matrix

\[
f'(C, T, F) = \begin{pmatrix} (I + \Delta tL) & 0 & I \\ -I & I & -I \\ 0 & -\psi'(T) & I \end{pmatrix}
\]

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

Choice of the forcing term \( \eta \)?

- Keep quadratic convergence (locally)
- Avoid oversolving the linear system

\[ \eta = \gamma \frac{\|f(x_k)\|_2}{\|f(x_k^1)\|_2} \] (Kelley, Eisenstat and Walker)
Solution by Newton–Krylov

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

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 \( \eta \)?
  - Keep quadratic convergence (locally)
  - Avoid oversolving the linear system
- \( \eta = \gamma \| f(x_k) \|^2 / \| f(x_{k-1}) \|^2 \) (Kelley, Eisenstat and Walker)
MoMaS reactive transport benchmark

Numerically difficult test case, 12 chemical species (J. Carrayrou)
Concentration of species 2 at $t = 50$, $t = 1000$, $t = 2000$, $t = 5010$.

C. de Dieuleveult (Andra Thesis, INRIA, Sage)