A global reactive transport model

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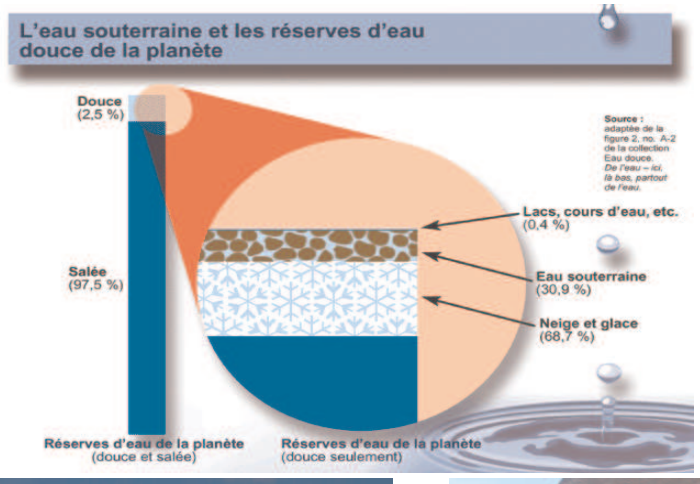
1 Introduction

Water resources



Pictures: Yves Chaux, Rennes, France

Diagram: `http://www.ec.gc.ca/water/f_main.html`

Reactive transport modeling

- Coupling transport by advection-dispersion with geochemistry
- System of Partial Differential Algebraic equations
- Model with thermodynamic equilibrium
- Method of lines: first discretize in space then in time $\Rightarrow$ DAE system
- Explicit scheme (SNIA): decoupling but stability restrictions
- Implicit scheme (Global): stability but nonlinear coupled system

Our method: global approach GDAE

 S. Kräutle, P. Knabner, (2005); A new numerical reduction scheme for fully coupled multicomponent transport-reaction problems in porous media; Water Resources Research, Vol. 41, W09414, 17 pp.

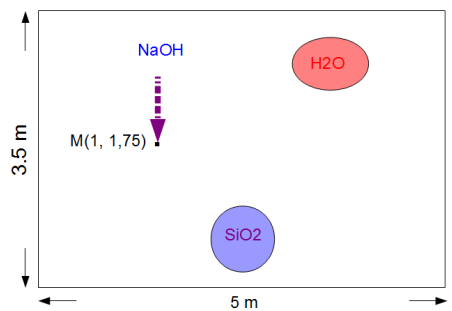
 S. Molins, J. Carrera, C. Ayora, Carlos and M.W. Saaltink, (2004); A formulation for decoupling components in reactive transport problems; Water Resources Research, Vol.40, W10301, 13 pp.

 C. de Dieuleveult, J. Erhel , M. Kern; A global strategy for solving reactive transport equations; Journal of Computational Physics,2009.

Numerical example

Andra qualification test

Injection of alcaline water $NaOH$ in a porous medium with quartz SiO_2



$$\begin{cases} \epsilon = 1. \\ v = \begin{pmatrix} 5.7\,10^{-7} \\ 0. \end{pmatrix} \text{m.s}^{-1} \\ d_m = 0 \\ \alpha_L = 0.2\,\text{m} \\ \alpha_T = 0.05\,\text{m} \\ T = 30\,\text{days} \\ \text{no flux boundary conditions} \end{cases}$$

Mugler, G. and Bernard-Michel, G. and Faucher, G. and Miguez, R. and Gaombalet, J. and Loth, L. and Chavant, C.), Projet ALLIANCES: plan de qualification ; CEA, ANDRA, EDF.

Chemistry conditions

Chemistry equations

$$\begin{array}{lcll} H_2O & \leftrightarrow & H^+ + \mathrm{OH}^- & K_1 = 10^{-14} \\ H_4\mathrm{Si}O_4 & \leftrightarrow & H_3\mathrm{Si}O_4^- + H^+ & K_2 = 10^{-9.8} \\ H_4\mathrm{Si}O_4 & \leftrightarrow & \mathrm{Si}O_2(s) + 2H_2O & K_3 = 10^{3.6} \end{array} \quad (1)$$

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Stoichiometric coefficients

	Na^+	OH^-	H_4SiO_4
H^+	0	-1	0
$H_3SiO_4^-$	0	1	1
SiO_2	0	0	1

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Initial values

	Na^+	OH^-	H_4SiO_4
Outside M	0	0	10.
At M	10^{-2}	10^{-2}	10.

Mass action laws

Aqueous reactions

$$x_i(c) = K_{ci} \prod_{j=1}^{N_c} c_j^{S_{ij}}, \quad i = 1, \dots N_x \tag{2}$$

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Sorption reactions

$$y_i(c, s) = K_{si} \prod_{j=1}^{N_c} c_j^{A_{ij}} \prod_{j=1}^{N_s} s_j^{B_{ij}}, \quad i = 1, \dots N_y, \tag{3}$$

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Precipitation reactions

$$\Pi_i(c) = K_{pi} \prod_{j=1}^{N_c} c_j^{E_{ij}}, \quad i = 1, \dots N_p \tag{4}$$

Mass conservation laws

Chemical variables and functions

$$X = \begin{pmatrix} c \\ s \\ p \end{pmatrix}, \quad \Phi(X) = \begin{pmatrix} c + S^T x(c) + A^T y(c,s) + E^T p \\ s + B^T y(c,s) \\ \Pi(c) \end{pmatrix} \tag{5}$$

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Chemical model

$$\begin{cases} \Phi(X) = \begin{pmatrix} T \\ W \\ 1 \end{pmatrix}, \\ c \geq 0, \\ s \geq 0, \\ p > 0. \end{cases} \tag{6}$$

Transport model

Advection-Dispersion operator

$$\mathcal{L}(u) = \nabla \cdot (vu - D\nabla u)$$

$$D = d_m I + \alpha_T \|v\| I + (\alpha_L - \alpha_T) \frac{vv^T}{\|v\|}$$

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Transport of mobile species

$$C(X) = c + S^T x(c) \tag{7}$$

$$\omega \frac{\partial T_i}{\partial t} + \mathcal{L}(C_i) = Q_i, \quad i = 1, \ldots, N_c \tag{8}$$

with boundary and initial conditions

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Space discretization

with (for example) a finite difference method

$$T = (T_1, \ldots, T_{N_m}) \tag{9}$$

Coupling transport with chemistry

Semi-discrete reactive transport model

$$\begin{cases} \omega \frac{dT_i}{dt} + LC_i(X) = Q_i + G_i, \quad i = 1, \ldots, N_c, \\ \Phi(X_j) - \begin{pmatrix} T_j \\ W_j \\ 1 \end{pmatrix} = 0 \quad j = 1, \ldots, N_m, \\ \text{initial condition for } T, \text{positivity constraints for } \Phi \end{cases} \tag{10}$$

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DAE formulation

$$\begin{cases} \omega \frac{d\mathbf{vec}\, T}{dt} + (L \otimes I)\mathbf{vec}\, C(X) - \mathbf{vec}\, Q - \mathbf{vec}\, G = 0, \quad i = 1, \ldots, N_c, \\ \mathbf{vec}\, \Phi(X) - (I \otimes N)\mathbf{vec}\, T - \mathbf{vec}\, F = 0 \end{cases} \tag{11}$$

DAE Global approach with substitution

Time discretization: BDF scheme

$$\frac{d\mathbf{vec}\, T}{dt} \simeq \frac{a}{\Delta t}\mathbf{vec}\, T + \frac{1}{\Delta t}\mathbf{vec}\, Z,$$

$$\begin{cases} \frac{a\omega}{\Delta t}\mathbf{vec}\, T + (L \otimes I)\mathbf{vec}\, C(X) - \ldots = 0, \\ -(I \otimes N)\mathbf{vec}\, T + \mathbf{vec}\, \Phi(X) - \ldots = 0, \end{cases}$$

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Substitution

$$R(X) = \frac{\Delta t}{a\omega}(L \otimes N)\,\mathbf{vec}\, C(X) + \mathbf{vec}\, \Phi(X) - \ldots$$

The Jacobian of R is

$$J_R(X) = \frac{\Delta t}{a\omega}(L \otimes N)\,\mathbf{diag}\,(\frac{dC}{dX}(X_j)) + \mathbf{diag}(J_\Phi(X_j)).$$

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Nonlinear system

$$R(X) = 0$$

solved with Newton method

Newton method

Jacobian matrix

$$J_\Phi(X) = \begin{pmatrix} I + S^T \frac{dx}{dc} + A^T \frac{\partial y}{\partial c} & A^T \frac{\partial y}{\partial s} & E^T \\ B^T \frac{\partial y}{\partial c} & I + B^T \frac{\partial y}{\partial s} & 0 \\ \frac{d\Pi}{dc} & 0 & 0 \end{pmatrix}$$

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Difficulties

- The derivatives $\frac{dx}{dc}$, etc, are not easy to compute
- The positivity constraints must be satisfied at each time step

Chemistry with logarithmic variables

Change of variables

- assuming $c > 0, s > 0$, use $\hat{X} = (\log(c), \log(s), p)$
- The positivity constraints are satisfied
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Jacobian matrix with logarithmic variables

$$J_{\hat{\Phi}}(\hat{X}) = \begin{pmatrix} \mathbf{diag}(\exp(\hat{c})) + S^T \mathbf{diag}(x)\, S + A^T \mathbf{diag}(y)\, A & A^T \mathbf{diag}(y)\, B & E^T \\ B^T \mathbf{diag}(y)\, A & \mathbf{diag}(\exp(\hat{s})) + B^T \mathbf{diag}(y)\, B & 0 \\ E & 0 & 0 \end{pmatrix}$$

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Difficulties

- Concentrations can become very small
- The matrix becomes almost singular for a component with almost null concentration

Precipitation Dissolution

Complementary problem

$\Pi(c) = 1$ and $p > 0$ replaced by

$$\begin{cases} (1 - \Pi(c)) \times p = 0, \\ 1 - \Pi(c) \geq 0 \text{ and } p \geq 0 \end{cases}$$

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Semismooth Newton method

- Generalized derivative of min function
- The positivity constraints must be satisfied

GRT3D software

Transport modules

- The velocity v is computed with MODFLOW
- The transport operator L is computed with MT3D

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Chemistry modules

- The functions $\Phi(X)$ and $C(X)$
- The derivatives $J_\Phi(X)$ and $dC(X)/dX$

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Coupling modules

- The function $R(X)$
- The derivative $J_R(X)$

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Coupling modules

- The function $R(X)$
- The derivative $J_R(X)$

Solving modules

- The DAE solver IDA in SUNDIALS using Newton-LU method
- The sparse linear solver UMFPACK

Versions of GRT3D

First version with logarithms

No elimination of T and C

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Reduced version with logarithms

Elimination of T and C in the linearized equations

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Optimized version without logarithms

Elimination of T and C in the linearized equations

Andra test case: performance results

CPU time of GRT3D

Computations done on two six-core Intel Xeon processors X5690, with a frequence of 3.46 GHz and 4GB of RAM per core.

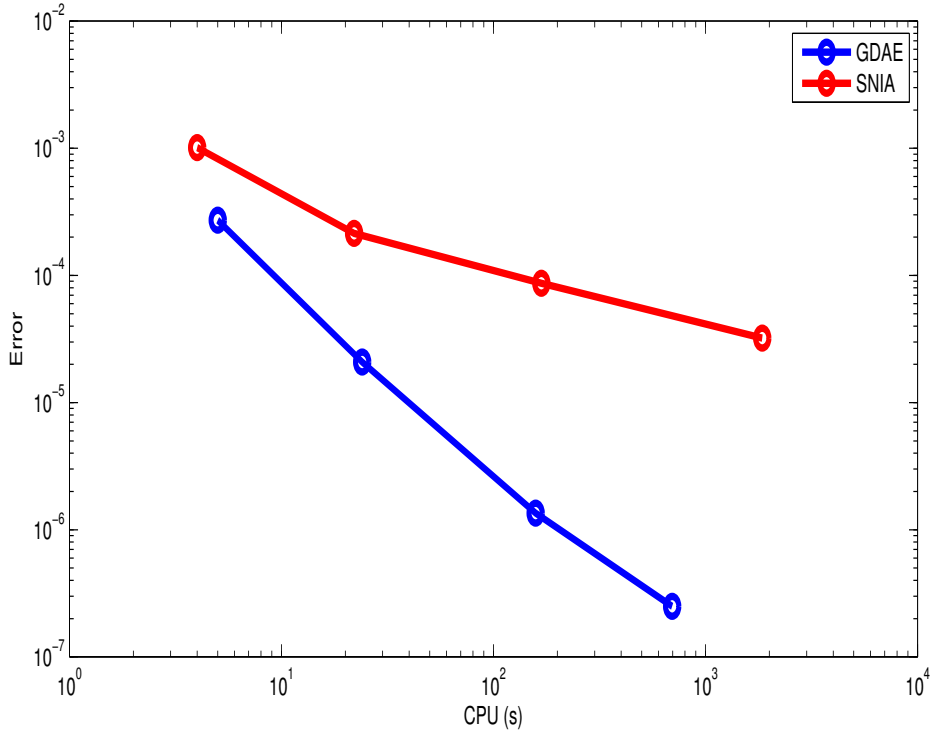
Mesh	first GRT3D	reduced GRT3D	optimized GRT3D
21x14	18 s	4 s	2 s
48x28	1 min 36 s	21 s	8s
81x56	6 min 33 s	1 min 53 s	50 s
71x101	11 min 55 s	3 min 28 s	1 min 21 s
161x112	32 min 43 s	16 min 30 s	4 min 32 s
322x224	-	1 h 52 min	37 min 38 s

Andra test case: comparison results

Performance/Accuracy results

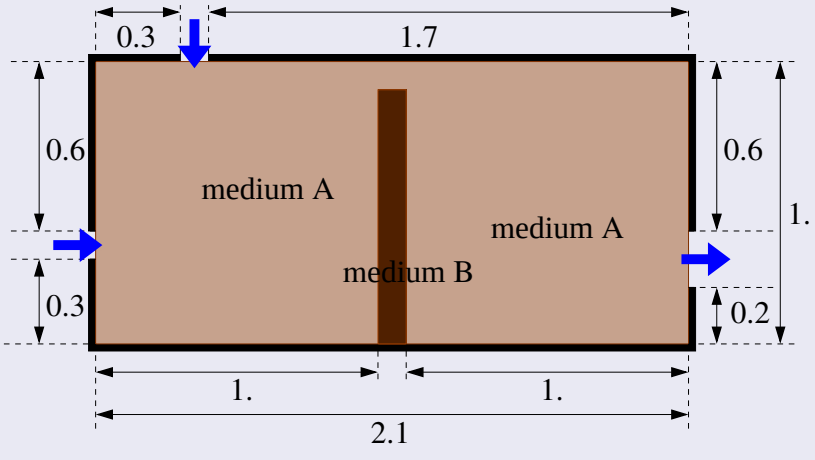
Comparison with an analytical solution

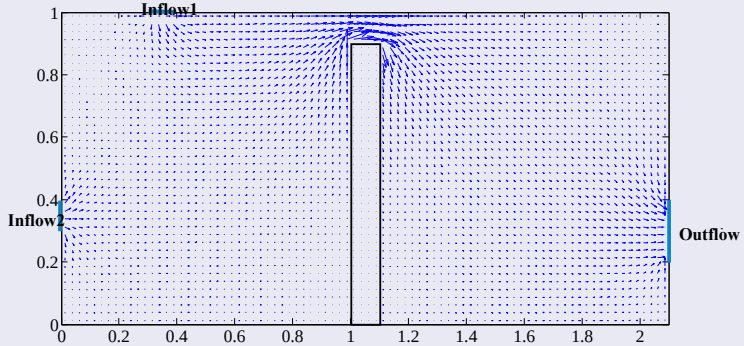
$$E_{Na^+} = [\frac{1}{N_m N_t} \sum_{n,j} (\tilde{x}_{Na^+}(m_j, t_n) - x_{Na^+}(m_j, t_n))]^{1/2}$$



Numerical experiment: MoMaS 2D easy test case

Geometry and computed velocity field





J. Carrayrou, M. Kern and P. Knabner, Reactive transport benchmark of MoMaS, Comput. Geo., 2010

MoMaS benchmark: transport conditions

Flow and transport data

Data	Medium A	Medium B
Porosity ω	0.25	0.5
Permeability $K(L.T-1)$	10^{-2}	10^{-5}
Molecular diffusion d_m	0	0
Dispersivity $\alpha_L(L)$	10^{-2}	6.10^{-2}
Dispersivity $\alpha_T(L)$	10^{-3}	6.10^{-3}

MoMaS benchmark: Flow and transport boundary conditions

Inflow	Outflow	Other
velocity $= 2.2510^{-2}(LT^{-1})$	head $= 1(L)$	no flow
Injection period $0 \leq t \leq 5000$	no diffusive flux	no total flux
Leaching period $t \geq 5000$	any time	any time

Chemistry conditions

Stoichiometric coefficients

	c_1	c_2	c_3	c_4	s	K
x_1	0	-1	0	0	0	10^{-12}
x_2	0	1	1	0	0	1
x_3	0	-1	0	1	0	1
x_4	0	-4	1	3	0	0.1
x_5	0	4	3	1	0	10^6
y_1	0	3	1	0	1	10^6
y_2	0	-3	0	1	2	10^{-1}

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x_4	0	-4	1	3	0	0.1
x_5	0	4	3	1	0	10^6
y_1	0	3	1	0	1	10^6
y_2	0	-3	0	1	2	10^{-1}

The component c_1 is a spectator ion.

Modified benchmark: the constant K_5 is changed from 10^{35} to 10^6.

Chemistry conditions

Total W of fixed component

Medium A	Medium B
1	10

Chemistry conditions

Total W of fixed component

Medium A	Medium B
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Initial and boundary conditions of total aqueous components

	T_1	T_2	T_3	T_4
Initial value	0	-2	0	2
Injection value	0.3	0.3	0.3	0
Leaching value	0	-2	0	2

$T_i = 0 \rightarrow c_i = 0$ for $i = 1, 3, 4$

Performance results

Results with GRT3D ; Mesh size 40×84

GRT3D version	with c_1		without c_1	
	CPU time	system size	CPU time	system size
First version	9h 52 mn	43680	7h 20 mn	33600
Reduced	4h 44 mn	16800	4h 4 mn	13440
Optimized	3h 9 mn	16800	2h 12 mn	13440

Results with GRT3D ; Mesh size 80×168

GRT3D version	with c_1		without c_1	
	CPU time	system size	CPU time	system size
First version	135 h 12 mn	174720	66 h 22 mn	134400
Reduced	83 h 24 mn	67200	54 h 56 mn	53760
Optimized	50 h 12 mn	67200	32 h 20 mn	53760

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with logarithms, $c > 0$ but when $c \simeq 0$, the Jacobian is ill-conditioned

MoMaS test: movies

components c2 and s

Conclusion

Summary: accuracy and efficiency

- DAE global approach (implicit scheme and Newton method)
- Control of time step and Newton iterations
- Substitution
- No logarithms but positivity constraints

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Future work

- Precipitation-dissolution with vanishing p
- Parallel computations
- Adaptive mesh refinement