

Chemistry-Transport Coupling by a DAE approach : Application on the Benchmark

Caroline de Dieuleveult ^a Jocelyne Erhel ^b



^aNational Radioactive Waste Management Agency



^bNational Institute for Research in Computer Science and Control

Sequential and global approaches

Sequential and split-operator approaches :

Separate solving

- non iterative
- iterative

Global approach : Simultaneous solving

- with substitution
- with DAE

Sequential and global approaches

Sequential and split-operator approaches :

- ▶ Decoupled transport and chemistry
- ▲ Additional error

Global Approach :

- ▲ High memory requirements
- ▶ Fast convergence

Chemistry model

Assumptions :

- Local equilibrium
- Precipitated species are known

Chemistry reactions :

$$\sum_{j=1}^{N_e} \alpha_{ij} X_j \rightleftharpoons 0 \quad i = 1, N_r$$

Chemistry model

Mass action equations (log. form) :

$$\begin{aligned}u &= \exp(\ln(K_u) + A \ln(c)) \\ 0 &= \ln(K_p) + E \ln(c)\end{aligned}$$

Variables :

- c : vector of component species
- u : vector of secondary species
- p : vector of precipitated species

Chemistry model

Mass action equations (log. form) :

$$\begin{aligned}u &= \exp(\ln(K_u) + A \ln(c)) \\ 0 &= \ln(K_p) + E \ln(c)\end{aligned}$$

Conservation laws :

$$\begin{aligned}T &= C + F \\ &= \mathcal{C}(\ln(c), p) + \mathcal{F}(u) \\ &= \exp(\ln(c) + A^T u(\log(c))) + E^T p\end{aligned}$$

With

T : vector of total analytical concentration

C : mobile part of T

F : fixed part of T

Chemistry model

Chemical system :

$$\Phi(y(x, t), T(x, t)) = 0$$

with

$$\Phi \begin{pmatrix} \ln(c) \\ p \\ T \end{pmatrix} = \begin{pmatrix} T - \exp(\ln(c)) - A^T u(\ln(c)) - E^T p \\ \ln(K_p) + E \ln(c) \end{pmatrix}$$

and $y = (\ln(c) \ p)^T$

\implies **Non linear equations**

Transport model

$$\varepsilon(\mathbf{x}) \frac{\partial T_i(\mathbf{x}, t)}{\partial t} + \mathcal{L}(C_i(\mathbf{x}, t)) = 0 \quad i = 1, N_c$$

with

$$\mathcal{L}(C_i(\mathbf{x}, t)) = \underbrace{\nabla \cdot \mathbf{v} C_i(\mathbf{x}, t)}_{\text{Advection}} - \underbrace{\nabla \cdot \mathbf{D} \nabla (C_i(\mathbf{x}, t))}_{\text{Dispersion}}$$

ε : porosity of media

\mathbf{v} : pore velocity

\mathbf{D} : dispersion tensor

\implies **Partial differential equations**

Chemical and transport system

$$\left\{ \begin{array}{l} \varepsilon(\mathbf{x}) \frac{\partial T_i(\mathbf{x}, t)}{\partial t} + \mathcal{L}(C_i(\mathbf{x}, t)) = 0 \quad i = 1, N_c \\ \Phi(y(\mathbf{x}, t), T(\mathbf{x}, t)) = 0 \\ C(\mathbf{x}, t) - C(y(\mathbf{x}, t)) = 0 \\ + \quad \text{boundary conditions} \\ + \quad \text{initial conditions} \end{array} \right.$$

⇒ **Non linear partial differential equations**

Method of lines

Spatial discretisation :

- Advection : combination of centering and upwind schemes
- Dispersion : finite difference scheme

$$\left\{ \underbrace{\begin{pmatrix} \varepsilon I & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}}_M \frac{d}{dt} \underbrace{\begin{pmatrix} T(t) \\ y(t) \\ C(t) \end{pmatrix}}_Y + \underbrace{\begin{pmatrix} \mathcal{L}_d C(t) + q_d \\ \Phi_d(y(t), T(t)) \\ C(t) - C_d(y(t)) \end{pmatrix}}_{f(Y(t))} = 0 \right.$$

+ initial conditions

$$\Rightarrow \begin{cases} M \frac{dY(t)}{dt} + f(Y(t)) = 0 \\ Y(t_0) = Y_0 \end{cases}$$

Differential algebraic equations (DAE)

$$\begin{cases} g(t, Y(t), \dot{Y}(t)) = 0 \\ Y(t_0) = Y_0 \end{cases}$$

With $g(t, Y(t), \dot{Y}(t)) = M\dot{Y}(t) + h(Y(t))$

\implies **DAE of index 1**

\implies DAE Initial-value system :

$$\begin{cases} g(t, Y(t), \dot{Y}(t)) = 0 \\ Y(t_0) = Y_0 \\ \dot{Y}(t_0) = \dot{Y}_0 \end{cases}$$

Method of lines

Time discretisation variable order and time step with "Backward Differential Formula" (BDF).

$$G(Y_n) = 0$$

with

$$G(Y_n) = g(t_n, Y_n, \frac{1}{t_n - t_{n-1}} \sum_{i=0}^q \alpha_{n,i} Y_{n-i})$$

\implies **Non linear algebraic system**

Jacobian of G is equal to :

$$J(Y) = \frac{\partial G(Y)}{\partial Y} = \frac{\partial g}{\partial Y} + \frac{\alpha_{n,0}}{t_n - t_{n-1}} \frac{\partial g}{\partial \dot{Y}}$$

Method of lines

Modified Newton method

at iteration m :

$$\bar{J}_n(Y_n^{m+1} - Y_n^m) = -G(Y_n^m)$$

with $\bar{J}_n \simeq J(Y_n)$

- Update of \bar{J}_n : depends on the calculation history.
- If convergence failure : reduction of the step size

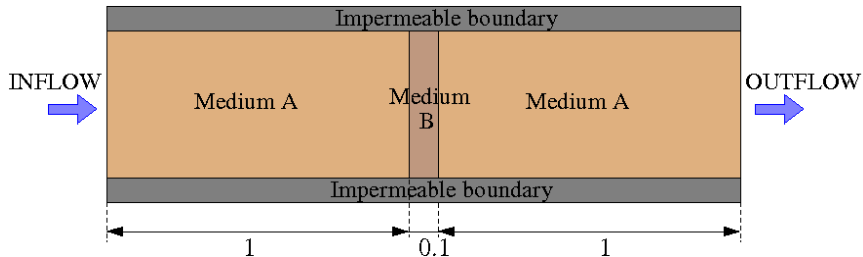
\implies **Sparse linear system**

Implementation

Numerical tools

- IDA, a solver of the SUNDIALS (Suite of Nonlinear and Differential/Algebraic Equation Solvers) family developed by the Lawrence Livermore National Laboratory
- Introduction of a module for the solution of a sparse linear system : UMFPACK

Benchmark 'Easy Test Case' 1D



Two subcases :

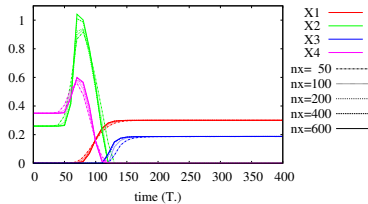
- Advective case
- Dispersive case

Benchmark 'Easy Test Case' 1D

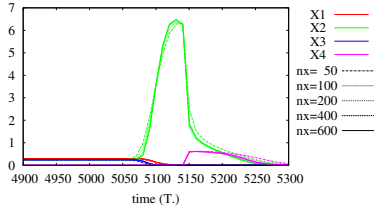
	X_1	X_2	X_3	X_4	S	K
C_1	0	-1	0	0	0	10^{-12}
C_2	0	1	1	1	0	1.
C_3	0	-1	0	1	0	1.
C_4	0	-4	1	3	0	0.1
C_5	0	4	3	1	0	10^{+35}
CS_1	0	3	1	0	1	10^{+6}
CS_2	0	-3	0	1	2	0.1
Initial total concentration (mol.L ⁻³)						
Medium A	0.	-2.	0.	2.	1.	
Medium B	0.	-2.	0.	2.	10.	
Imposed inflow condition (mol.L ⁻³)						
$t \in [0; 5000]$	0.3	0.3	0.3	0.		
$t \in [5000; 6000]$	0.	-2.	0.	2.		

Advective test case

Concentration at point $x=0.08$ m (mol/l)

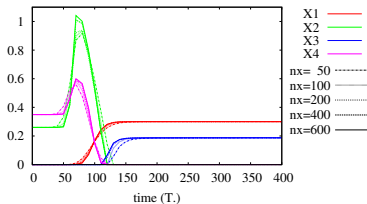


Concentration at point $x=0.08$ m (mol/l)

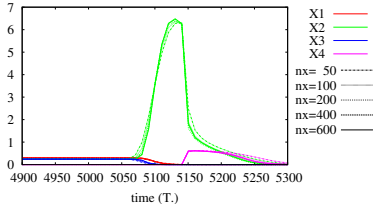


Advective test case

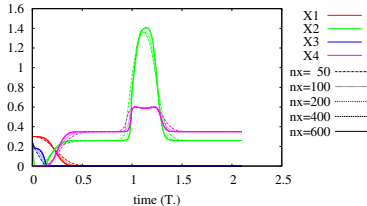
Concentration at point $x=0.08$ m (mol/l)



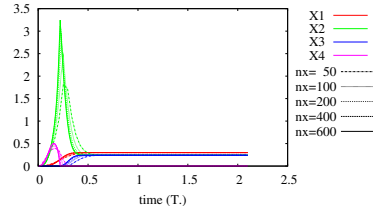
Concentration at point $x=0.08$ m (mol/l)



Concentration at $t=10$ T. (mol/l)

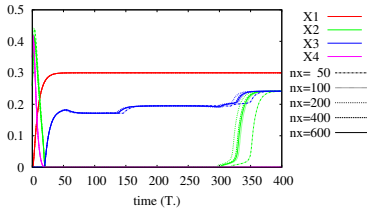


Concentration at $t=10$ T. (mol/l)

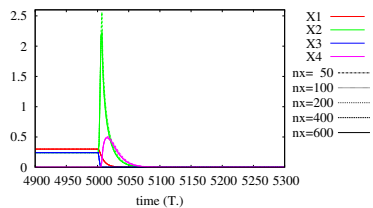


Dispersive test case

Concentration at point $x=0.08$ m (mol/l)

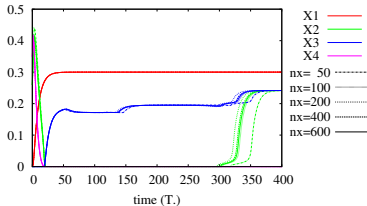


Concentration at point $x=0.08$ m (mol/l)

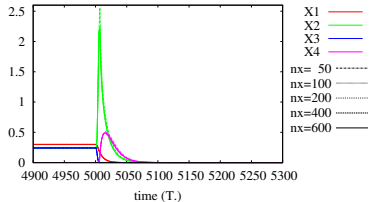


Dispersive test case

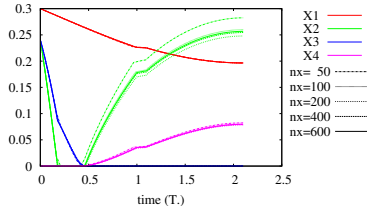
Concentration at point $x=0.08$ m (mol/l)



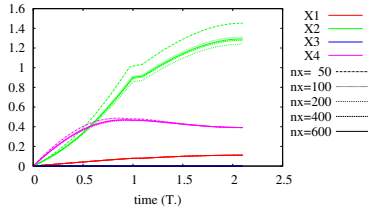
Concentration at point $x=0.08$ m (mol/l)



Concentration at $t=10$ T. (mol/l)



Concentration at $t=10$ T. (mol/l)



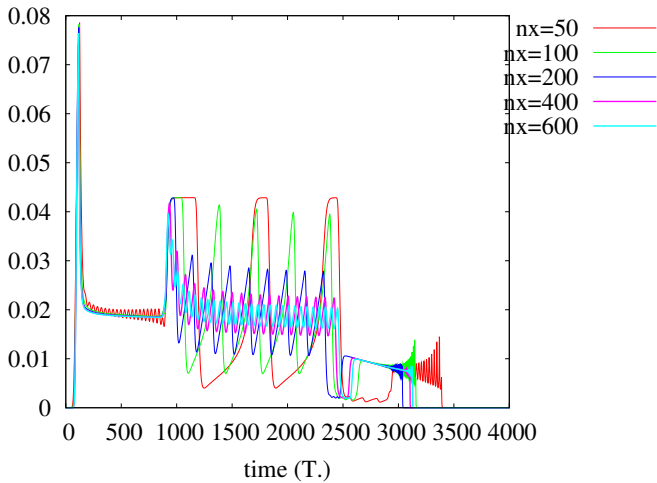
Current work and prospect

Current work

- A more realistic precipitation/ dissolution model
- Different space discretisation (MT3D)

Prospect

- A more complex chemistry model (PhreeqC)
- Introduction in the ANDRA platform



Concentration at $t=10 T.$ (mol/l)

