Numerical simulation of multicomponent two-phase reactive flow

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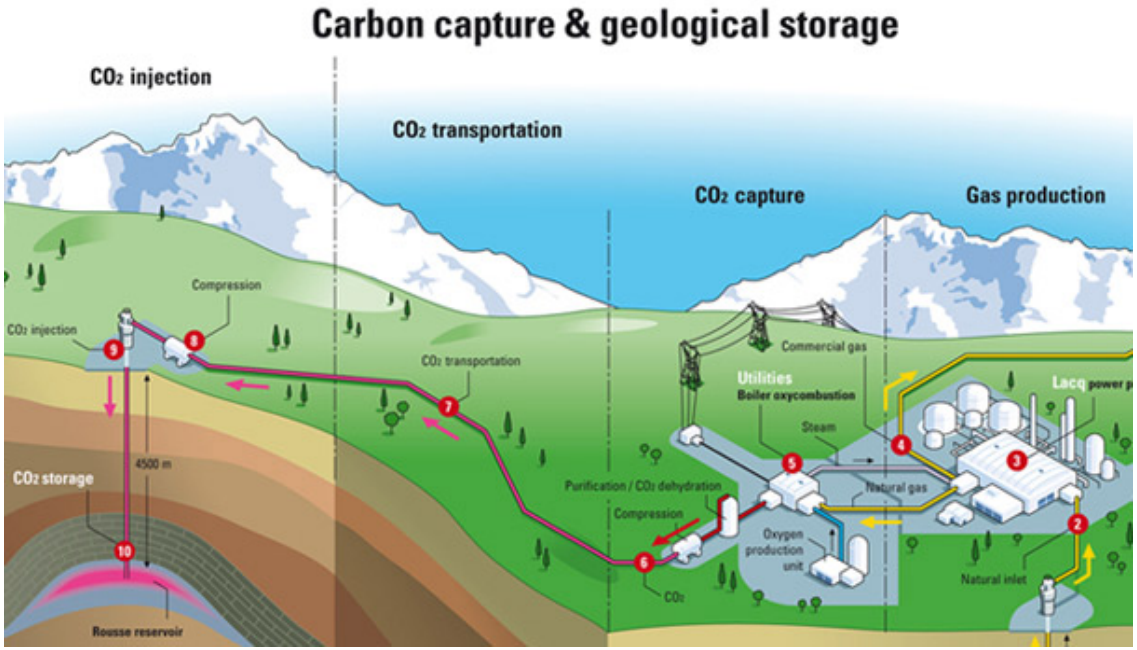
Outline

Motivation: CO2 sequestration



Capture and storage of CO_2 (Lacq pilot site)

Goals

- Numerical simulation of multicomponent two-phase flow with chemical reactions
- Implementation in DuMu^X of a reactive transport module

Simplified chemical system

Chemical components

Liquid phase (l)	Gas phase (g)	Solid phase (s)
H_2O, CO_2, H^+, OH^-, HCO_3^-, Ca^{2+}	$CO_{2(g)}$	$CaCO_3$

Chemical reactions (all equilibrium)

Homogeneous reactions

$$\begin{aligned} OH^- &\rightleftharpoons H_2O - H^+, \\ HCO_3^- &\rightleftharpoons H_2O - H^+ + CO_{2(l)}, \end{aligned}$$

Heterogeneous reactions

$$\begin{aligned} CO_{2(g)} &\rightleftharpoons CO_{2(l)}, \\ CaCO_3 &\rightleftharpoons H_2O - 2H^+ + CO_{2(l)} + Ca^{2+}, \end{aligned}$$

Modeling chemical equilibrium

N_s chemical species, N_r reactions, S stoichiometric matrix .

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

(Usually) $N_r \geq N_s$, S full rank: define Kernel matrix U st $US^T = 0$
(Saaltink et al. (98), Knabner et al. (07), Amir, K. (10)).

Nonlinear system

- Mass action laws $S \log c = \log k$
- Mass conservation $Uc = C$, C known from transport

Newton's method with line search, use $\log c$ as unknown.

Local elimination of chemical concentrations $c = \Psi_C(T)$.

Mathematical model for two-phase multicomponent flow

Notation

Phase index $\alpha = l, g, s$ i - species index. $\alpha_i =$ index of the phase that contains species i.
Phase flow operator
$L_\alpha(c) = -\nabla \cdot (\phi S_\alpha D_\alpha \nabla c) + \nabla \cdot (c \overrightarrow{q_\alpha}),\ \alpha = l, g,\ L_s = 0.$

- Mass conservation law for each species:

$$\partial_t(\phi S_{\alpha_i} c^i) + L_{\alpha_i}(c^i) = \sum_j \mathbb{S}_{ji} r_j, \quad i = 1 \dots N_s,$$

  together with Darcy's law and state equations (for each phase), capillary pressure, closure relations.

- r_j rate for jth reaction (unknown for equilibrium)

Primary variables: phase pressures p_α, phase saturations S_α, chemical concentrations c_i.

Coupled problem

Elimination of reaction rates

Multiply conservation laws by U :

$$\sum_{\alpha} \left(\partial_t(\phi S_\alpha C_\alpha^k) + L_\alpha C_\alpha^k \right) = 0, \quad k = 1, \ldots, N_s - N_r$$

with $C_\alpha^k = \sum_{i \text{ st } \alpha_i = \alpha} U_{ki} c_i$ total component concentrations.

System closed by mass action laws

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System closed by mass action laws

Solving the coupled system

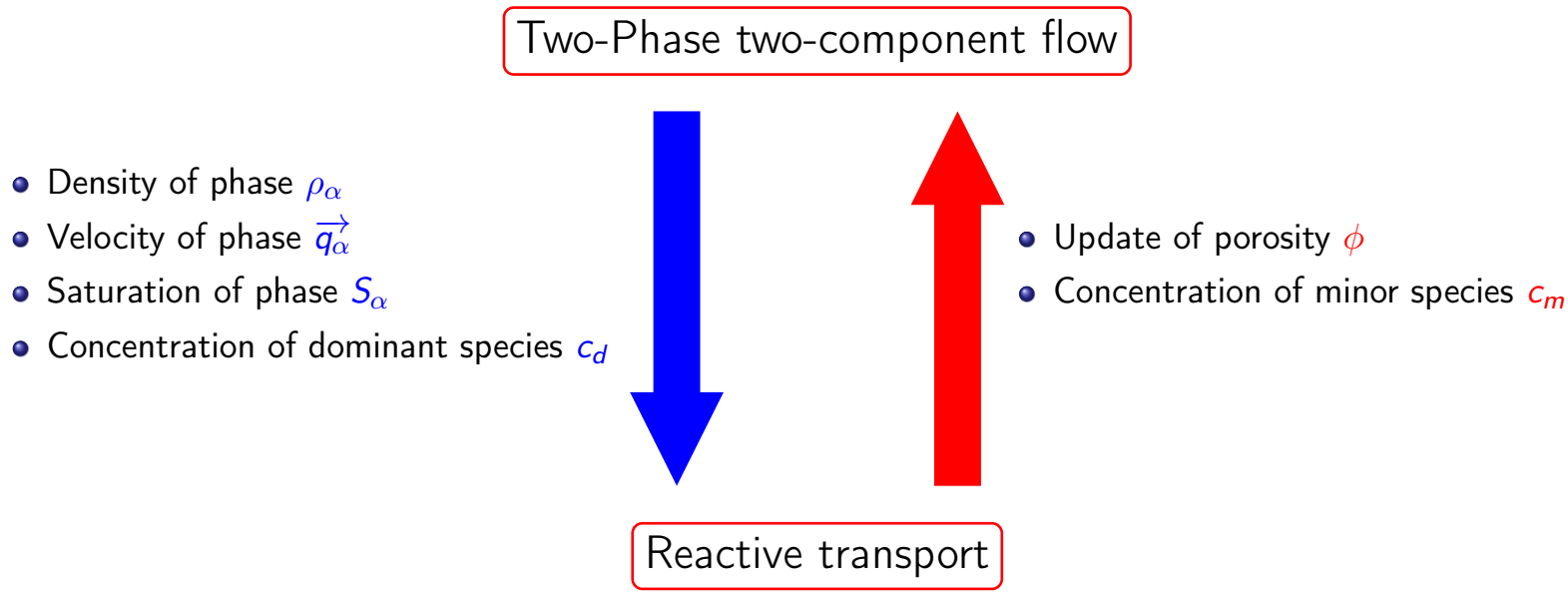
- Fully coupled approach Solve full system for phase pressures, phase saturations and all species concentrations (Fan et al., 2012, Saaltink et al, 2013). Large nonlinear system
- Sequential approach Solve separately for flow (pressures, saturations, mass fractions of H_2O and CO_2) and reactive transport (other species concentrations). Used in ToughReact (Xu et al., 2012).

A consistent decoupled approach

Decoupling strategy

Identify a dominant component in each phase: H_2O in liquid, CO_2 in gas.

- Dominant components form a compositional two-phase flow system, include CO_2 dissolution (Henry's law), variables are p_α, S_α, component concentrations.
- Other components follow reactive transport system with given velocity q_α, variables c_i



Decoupled system

Two phase, two components flow

$$
\begin{aligned}
H_2O \quad & \partial_t(\phi S_l c_{H_2O}) + L_l c_{H_2O} = R_1(c_{OH^-}, c_{HCO_3^-}, c_{CaCO_3}), \\
CO_{2(l)} \quad & \partial_t(\phi S_l c_{CO_{2(l)}} + \phi S_l c_{CO_{2(g)}}) + L_l c_{CO_{2(l)}} + L_g c_{CO_{2(g)}} = R_2(c_{HCO_3^-}, c_{CaCO_3}), \\
CO_{2(g)} \quad & \gamma(c_{CO_{2(g)}}) p_g = c_{CO_{2(l)}}.
\end{aligned}
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\end{aligned}
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Reactive transport

$$
\begin{aligned}
H^+ \quad & \partial_t\left(\phi S_l C_l^{H^+} + C_s^{H^+}\right) + L_l C_l^{H^+} = 0,\\
Ca^{2+} \quad & \partial_t\left(\phi S_l C_l^{Ca^{2+}} + C_s^{Ca^{2+}}\right) + L_l C_l^{Ca^{2+}} = 0,\\
OH^- \quad & c_{OH^-} = K_{OH^-}\, c_{H^+},\\
HCO_3^- \quad & c_{HCO_3^-} = K_{HCO_3^-}\, c_{H^+}^{-1} c_{CO_{2(l)}},\\
CaCO_3 \quad & 1 = K_{CaCO_3}\, c_{H^+}^{-2} c_{CO_{2(l)}} c_{Ca^{2+}}.
\end{aligned}
$$

$$
\begin{aligned}
C_l^{H^+} &= c_{H^+} - c_{OH^-} - c_{HCO_3^-},\\
C_s^{H^+} &= -2 c_{CaCO_3},\\
C_l^{Ca^{2+}} &= c_{Ca^{2+}},\\
C_s^{Ca^{2+}} &= c_{CaCo_3}
\end{aligned}
$$

Numerical methods

- Implementation in DuMu$^{\mathrm{X}}$: DUNE for Multi-Phase, Component, Scale, Physics, ... flow and transport in porous media
- Flow is 2p2c, fully coupled approach, vertex centered finite volumes.
- Reactive transport is new module, extends 1pNc, coupling through Standard Iterative Approach (Yeh and Tripathi, 89).

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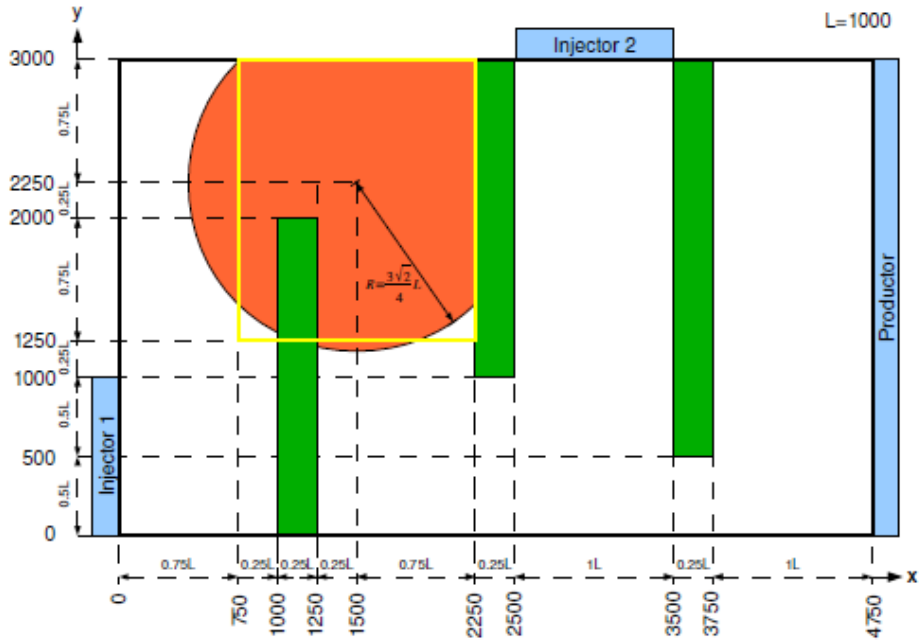
Estimation of splitting error

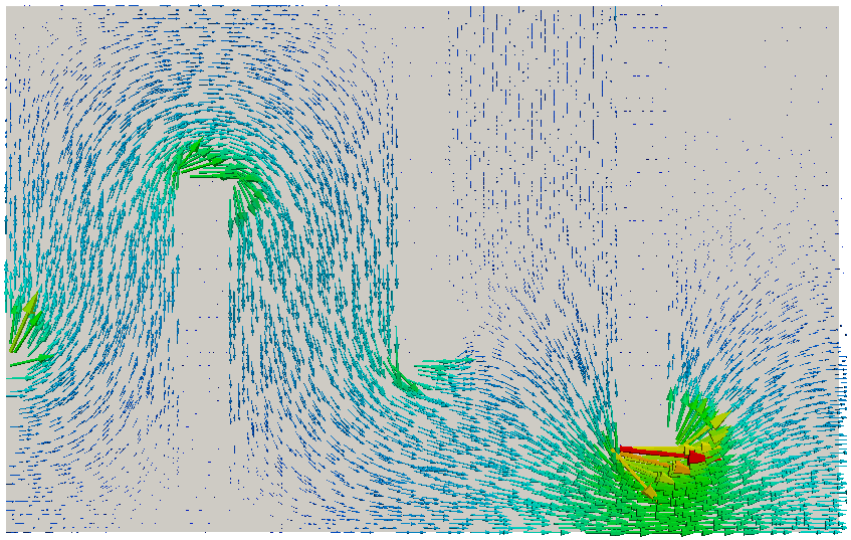
Variation of liquid CO_2 total concentration.

Time (years)	Without R	With R
50 years	334590649	335411173
100 years	334127585	335409334
200 years	333338905	335408767
Absolute change	−1928479	−7775
Relative change	−0.58%	−0.002%

Numerical example: the SHPCO2 benchmark

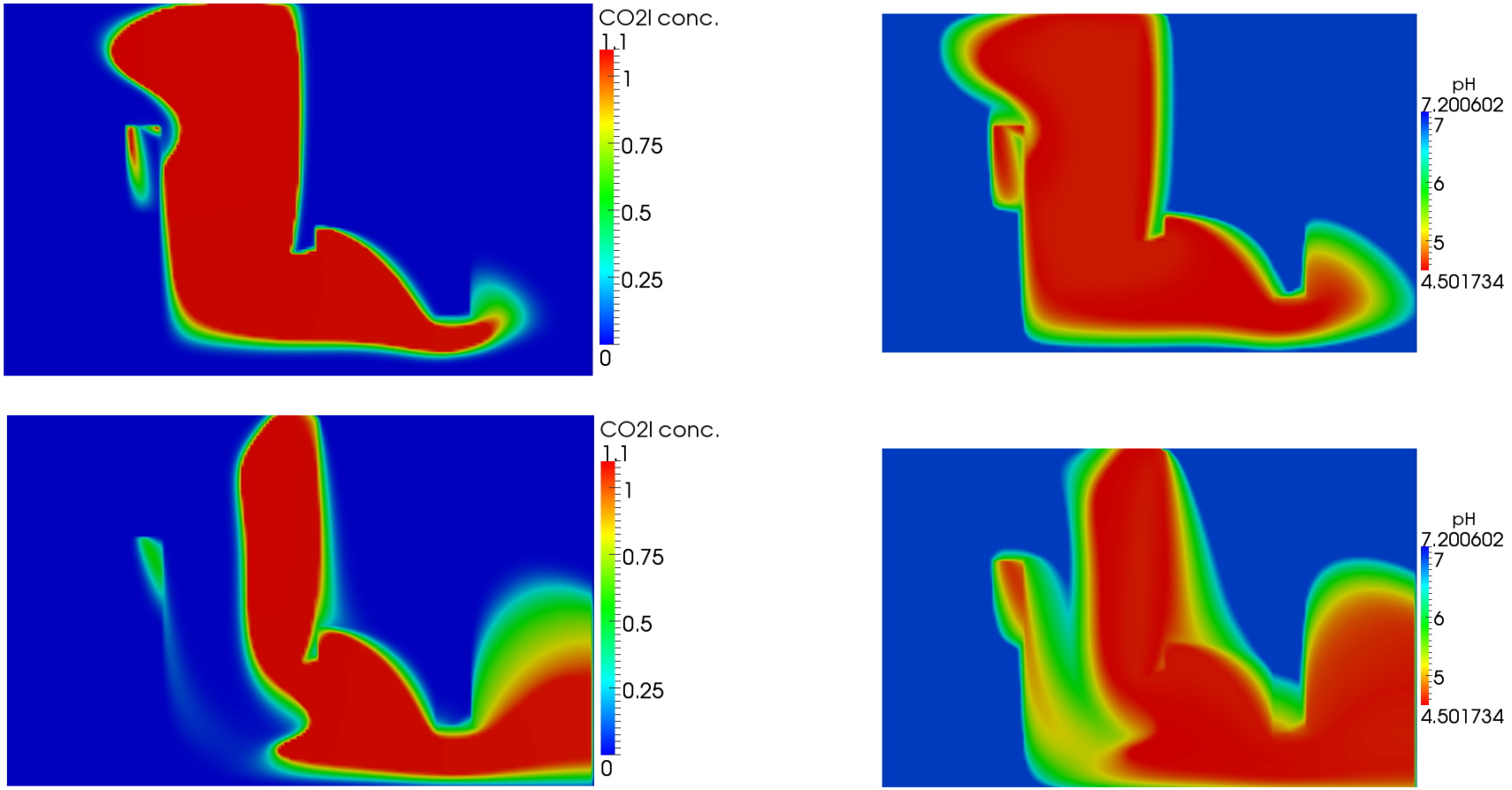
Study dissolution of gas bubble after injection of water.
Compare one phase (immobile gas) and two-phase simulations





Geometry and (one phase) velocity field
A. Michel, F. Haeberlein, L. Trenty (2009)

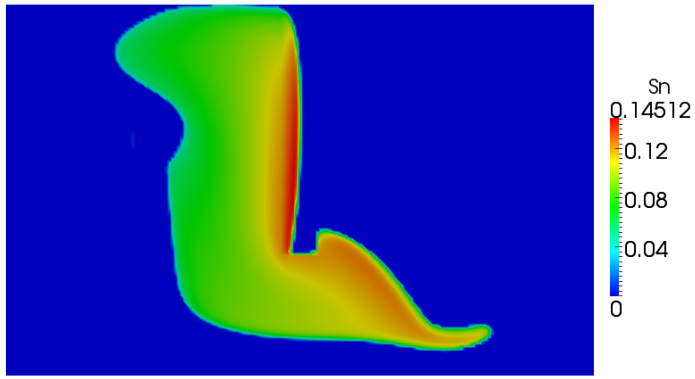
SHPCO2 benchmark: two-phase results

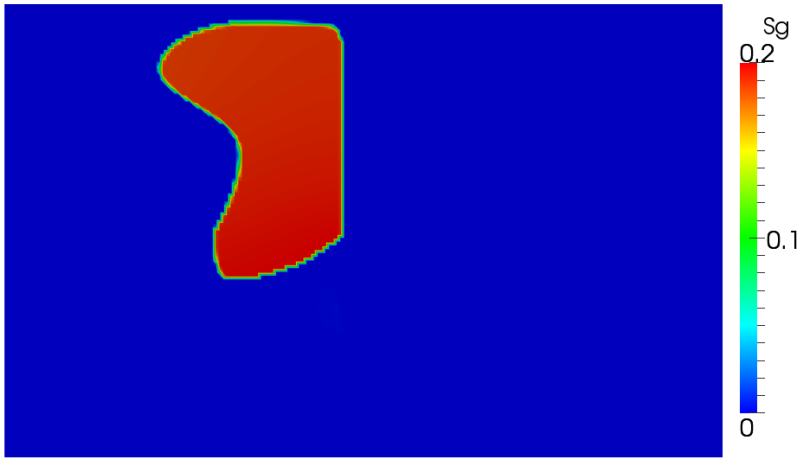


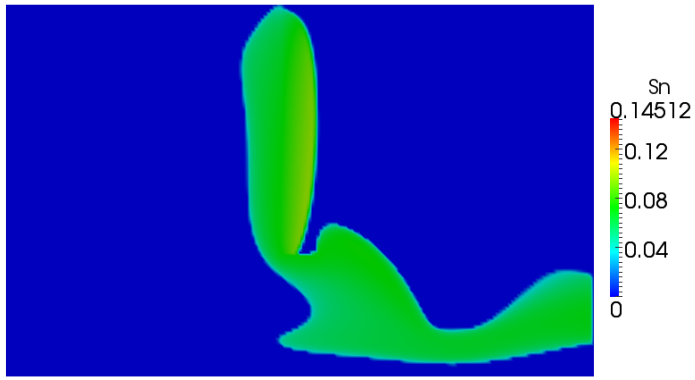
Liquid CO_2 (left) and pH (right).
Top: 400 years, bottom: 1200 years.

SHPCO2: one phase and two-phase comparison

Gas concentration.



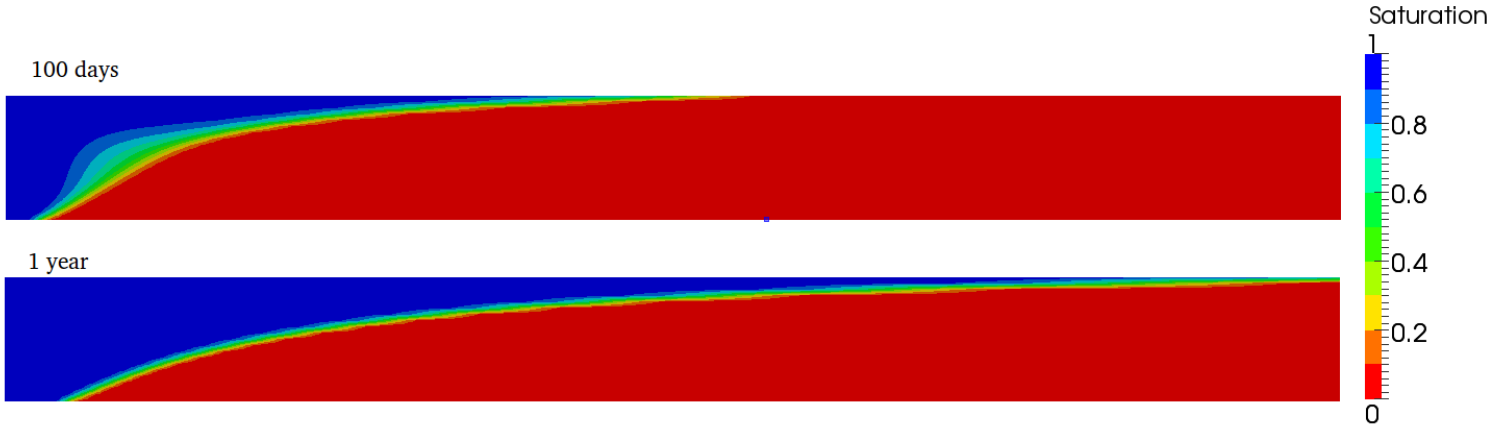




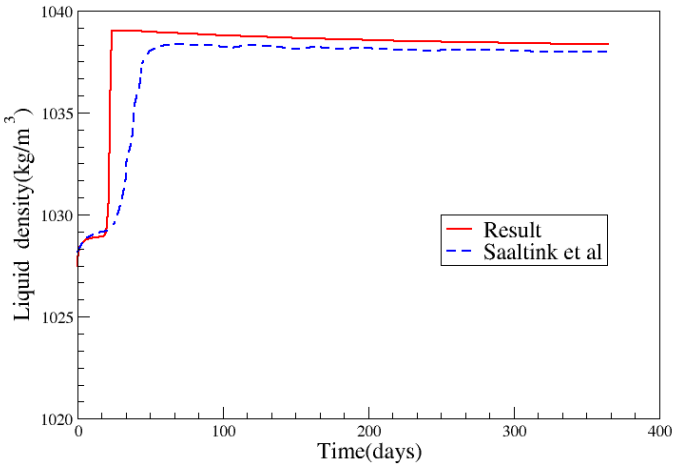
Left: immobile gas (one phase), right: mobile gas (two phases)

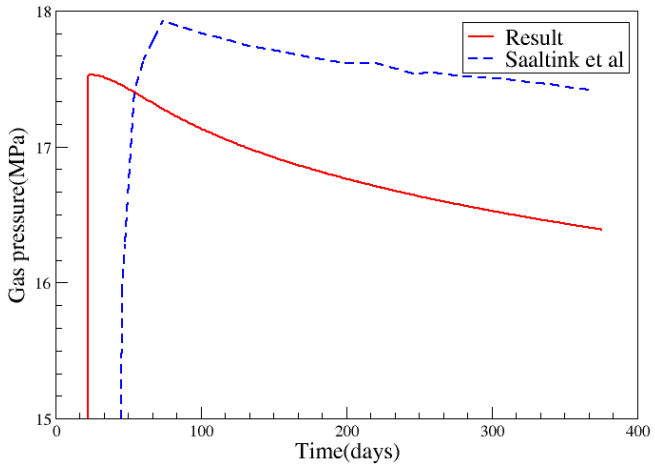
Top: 400 years, bottom: 1200 years.

CO_2 injection (after Saaltink et al., 2013)



Gas saturation after100 days, and 1 year of CO2 injection

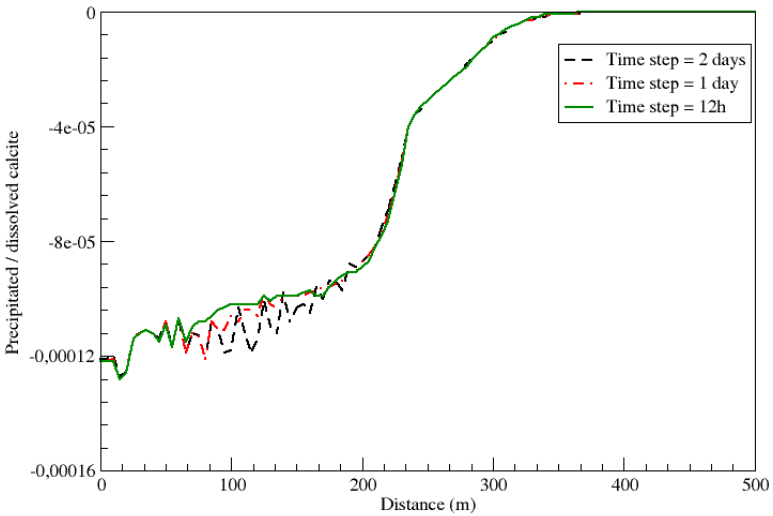




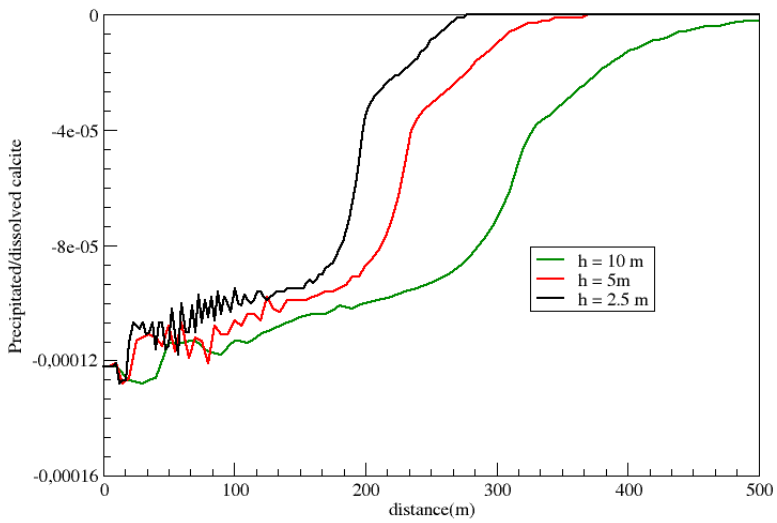
Density and pressure variation at 20 m from injection well. Comparison with Saaltink et al.

Precipitation of calcite: sensitivity to mesh and time step

Precipitated / dissolved calcite volume fraction after 100 days



Different time steps



Different meshes

Conclusions – perspectives

- Uncoupled algorithms for a two-phase multicomponent flow with reactive transport
- Validation example: behavior different than one phase case
- Quantitative estimate for splitting error

E. Ahusborde, M. K., V. Vostrikov, *Numerical simulation of two-phase multicomponent flow with reactive transport in porous media: application to geological sequestration of CO2*, ESAIM: Proc., 2015.

In progress (Ahusborde, Amaziane, El Ossmani , Poncet)

- Taking into account porosity changes from mineral reactions
- Extension to kinetic reactions