Simulation of reactive transport in heterogenous porous media with a Newton-Krylov method

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Motivations and scope of the talk

- Reactive transport = coupling of flow + transport with geochemistry,

- Applications: nuclear waste storage, CO2 or H2 underground storage.

- Variety of chemical reactions: aqueous (acid - base, redox), surface reactions (ion exchange), mineral precipitation dissolution

Carbon capture & geological storage

This talk

- One phase flow and transport, equilibrium chemistry

- Globally coupled approach that keeps chemistry and transport codes separate

- Solve mineral precipitation-dissolution with an interior point method.

- Large non-linear system solved with a Newton–Krylov method. Nonlinear preconditioning ⇒ convergence independant of mesh size

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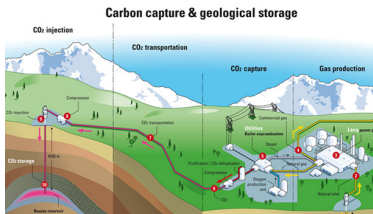
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Chemical laws for equilibrium

Species : $(X_j^a)_{j=1,..,N_a}$ mobile , $(X_j^s)_{j=1,..,N_s}$ immobile, $(X_j^p)_{j=1,..,N_p}$ mineral

Chemical reactions

$$S \begin{pmatrix} X^a \\ X^s \\ X^p \end{pmatrix} \leftrightarrows \begin{pmatrix} 0 \\ 0 \\ 0 \end{pmatrix}, \; S = \begin{pmatrix} S_{aa} & 0 & 0 \\ S_{sa} & S_{ss} & 0 \\ S_{pa} & 0 & -I \end{pmatrix}$$

- N_r homogeneous reactions,
- $\bar{N}_r$ heterogeneous reactions,
- N_p precipitation reactions.

c_j conc. species X_j^a, s_j conc. species X_j^s, p_j conc. species X_j^p

Aqueous and sorption reactions

Reactions involving minerals

Mass action law (log. form)

$$\begin{pmatrix} S_{aa} & 0 \\ S_{sa} & S_{ss} \end{pmatrix} \begin{pmatrix} \log c \\ \log s \end{pmatrix} = \begin{pmatrix} \log K_a \\ \log K_s \end{pmatrix}$$

Mass action law for minerals

($\Pi = 0$ and $p \geq 0$) or ($\Pi \geq 0$ and $p = 0$)

Solubility product

$$\Pi = \log K_p - S_{pa} \log c$$

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Mass conservation with total concentrations

Mass conservation

$$\phi \partial_t \mathsf{c} + \mathcal{L}\mathsf{c} = S_{aa}^T r_a + S_{sa}^T r_s + S_{pa}^T r_p,$$

$$\rho_s(1-\phi)\partial_t \mathsf{s} \quad = \quad S_{ss}^T r_s,$$

$$\rho_s(1-\phi)\partial_t \mathsf{p} \quad = \quad - r_p.$$

r_a, r_s, r_p: unknown reaction rates

$\mathcal{L}(\mathsf{c}) = \nabla \cdot (q\mathsf{c} - D\nabla \mathsf{c})$ (advection-diffusion)

Elimination of the reaction rates ([Saaltink et al. WRR, 1998])

Introduce a kernel matrix U such that $US^T = 0$. Let $U = \begin{pmatrix} U_{aa} & U_{as} & U_{ap} \\ 0 & U_{ss} & 0 \end{pmatrix}$

We define : $\mathsf{C} = U_{aa}\,\mathsf{c}, \quad \overline{\mathsf{C}} = U_{as}\,\mathsf{s} + U_{ap}\mathsf{p}, \quad \mathsf{T} = \mathsf{C} + \overline{\mathsf{C}},$

Transport for total concs.

$$\phi \partial_t \mathsf{C} + \rho_s(1-\phi)\partial_t \overline{\mathsf{C}} + \mathcal{L}\mathsf{C} = 0,$$

$$\rho_s(1-\phi)\partial_t \overline{\mathsf{T}} = 0.$$

Solution of the chemical sub-problem

Mass action laws

$S_{aa} \log \mathsf{c} + \log K_a = 0$

$+$

Definition of the totals

$\mathsf{T} = U_{aa}\mathsf{c} + U_{pa}\mathsf{p}$

Complementarity condition

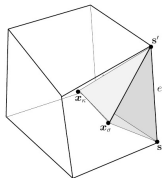$(\log K_p - S_{pa}\xi^{\mathsf{p}} = 0, \mathsf{p} \geq 0)$ or $(\log K_p - S_{pa}\xi^{\mathsf{p}} \geq 0, \mathsf{p} = 0)$.

- Nonlinear system with complementarity conditions
- Solve by Newton interior points method (F. Saaf (96)) or semi-smooth Newton (S. Kraütle (11))

Chemical solution operator

$\Psi_{\mathsf{C}} : \mathsf{T} \mapsto \Psi_{\mathsf{C}}(\mathsf{T}) = \overline{\mathsf{C}} = U_{as}\,\mathsf{s} + U_{ap}\mathsf{p}.$

pE-pH diagram for iron
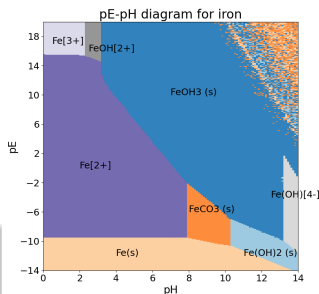
Discrete advection–diffusion model

Discrete advection diffusion

$Ac^{n+1} = Bc^n + M\Delta tq^{n+1}$

Vertex **A**pproximate **G**radient scheme (Eymard et al., 2012)

ComPASS code (Lopez et al., 2018)

- Multiphase compositional thermal Darcy flow model (Coats formuation)
- 2D discrete fracture or fault network coupled with the surrounding 3D matrix
- 3D conforming polyhedral meshes
- Newton Raphson algorithm with phase appearance and disappearance
- CPR-AMG preconditioner [Lacroix et al, 2001], [Scheichl e al., 2003]
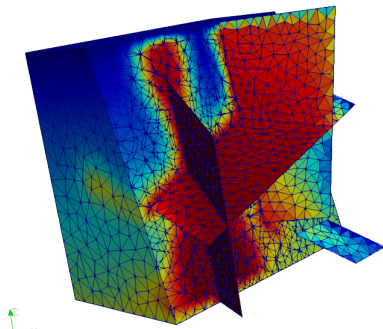
Temperature in medium with fractures

The coupled formulation

SIA : sequentially solve transport and chemistry, easy to code, slow convergence, loss of mass.

GIA : solve full coupled system, accurate, robust, difficult to code, needs efficient solver.

Chemistry is local $\Rightarrow$ eliminate individual concentrations at each point by using the operator Ψ_C. Only unknowns : C, $\bar{C}$.

Coupled formulation

$$\phi\partial_t C + \phi\partial_t \bar{C} + \mathcal{L}(C) = 0,$$

$$\bar{C} - \Psi_C\left(C + \bar{C}\right) = 0.$$

Discrete non-linear system ($b^n = B C^n + M^n$)

$$f\begin{pmatrix} C \\ \bar{C} \end{pmatrix} = \begin{pmatrix} (A \otimes I)C + (M \otimes I)\bar{C} - b^n \\ \bar{C} - \Psi_C(C + \bar{C}) \end{pmatrix} = 0$$

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Newton-Krylov with (non-linear) preconditioning

Inexact Newton method [Kelley, 1995])

- Solve the linear system by an *iterative* method (GMRES),
- GMRES requires only **Jacobian matrix by vector products**, can be computed analytically or **approximated by finite differences**.

Elimination of C

Reduced system $h(\bar{C}) = \bar{C} - \Psi_C \left((A^{-1} \otimes I) \left(b^n - (M \otimes I)\bar{C} \right) + \bar{C} \right) = 0$.
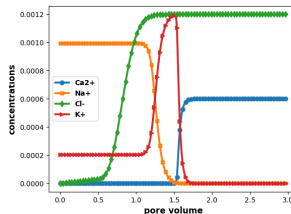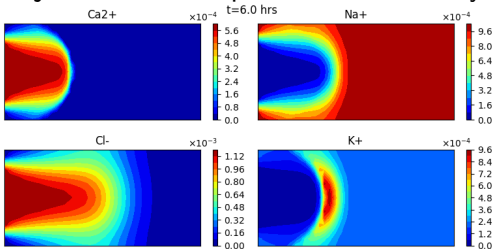
Jacobian * vector $J_h v = v - J_C J_T v = v - J_C v + J_C \left((A^{-1}M) \otimes I \right) v$.

Details: [L. Amir, MK, IJNAM, 2019], [L. Amir, MK, Water, 2021]

- Links between elimination and block preconditioning,
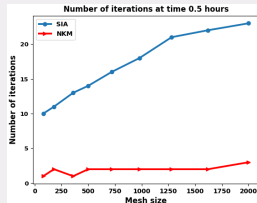- **The number of iterations becomes independent of the mesh**.

Inject calcium on part of left boundary into a K-Na solution

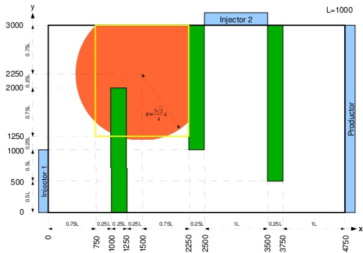## Fixed-point – precond. Newton-Krylov comparison

- Nb. of iterations ↗ with the mesh resolution for SIA while it remains stable, small for NKM.
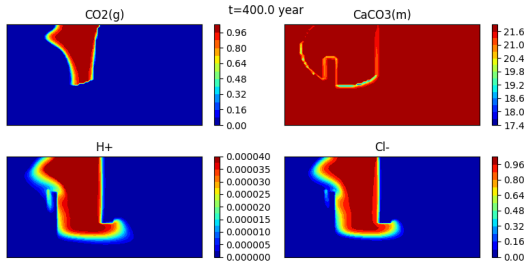- NKM gives **convergence independent of the mesh size**.

Parameters (Heterogenous)

Barriers with low permeability
Initial state : two zones with
different initial concentrations,
bubble contains $CO_2(g)$ equal to 1, 0
elsewhere.

Conclusion and perspectives

- Extension of the capabilities of the equilibrium chemical solver to handle mineral precipitation and dissolution;
- Heterogenous, 2D configurations : Ex11, SHPCO2
- Number of iterations is independent of the mesh size

- Improve the robustness of the NKM method in the presence of minerals.
- Improve the computation of the Jacobian matrix by vector product, by exploiting the known block structure of the matrix
- Extension to two–phase flow (cf [E. Ahusborde, M. Kern, V. Vostrikov, 2015], [E Ahusborde, B Amaziane, M El Ossmani, 2018])

Some references

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Geothermal Modeling in Complex Geological Systems with the ComPASS Code.

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Call for participation: two-phase reactive benchmark

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`https://hal.archives-ouvertes.fr/hal-03635080v1`
Contact one of us !