



A nonlinear preconditioner for reactive transport in heterogenous porous media

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Outline

- 1 Chemical laws and transport equation
- 2 Coupled problems and coupling algorithms
- 3 Linear and non-linear preconditionning
- 4 Numerical results

Goals of this work

- Study **preconditioning** methods for the linearized coupled problem, and their **relationship** to **elimination methods** where the **mobile concentrations are eliminated**
- Obtain methods (Newton and GMRES) with **convergence independent of the mesh size**.
- Application to 1D and 2D configurations

Previous work

- Solve reactive-transport models by Newton-Krylov,
- Keep transport and chemistry modules separate,

Equilibrium chemical reactions

$(\mathbf{x}_j)_{j=1,\dots,N_S}$ mobile species, $(\bar{\mathbf{x}}_j)_{j=1,\dots,\bar{N}_S}$ immobile species,

Chemical reactions

$$S \begin{pmatrix} \mathbf{x} \\ \bar{\mathbf{x}} \end{pmatrix} = \begin{pmatrix} S_{cc} & 0 \\ S_{\bar{c}c} & S_{\bar{c}\bar{c}} \end{pmatrix} \begin{pmatrix} \mathbf{x} \\ \bar{\mathbf{x}} \end{pmatrix} \rightleftharpoons \begin{pmatrix} 0 \\ 0 \end{pmatrix}$$

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

\mathbf{c}_j (resp. $\bar{\mathbf{c}}_j$) concentration of species \mathbf{x}_j (resp. $\bar{\mathbf{x}}_j$),

Mass action law (log. form)

$$\begin{pmatrix} S_{cc} & 0 \\ S_{\bar{c}c} & S_{\bar{c}\bar{c}} \end{pmatrix} \begin{pmatrix} \log \mathbf{c} \\ \log \bar{\mathbf{c}} \end{pmatrix} = \begin{pmatrix} \log K \\ \log \bar{K} \end{pmatrix} +$$

Mass conservation

$$\begin{aligned} \phi \partial_t \mathbf{c} + \mathcal{L}(\mathbf{c}) &= S_{cc}^T \mathbf{r} + S_{\bar{c}c}^T \bar{\mathbf{r}}, \\ \phi \partial_t \bar{\mathbf{c}} &= S_{\bar{c}\bar{c}}^T \bar{\mathbf{r}}, \end{aligned}$$

$\mathbf{r}, \bar{\mathbf{r}}$:
unknown
reaction
rates

\mathcal{L} denotes advection–diffusion operator : $\mathcal{L}(\mathbf{c}) = -\operatorname{div}(\mathbf{D} \operatorname{grad} \mathbf{c}) + \operatorname{div}(\mathbf{u} \mathbf{c})$

Mass conservation with total concentrations

Elimination of the reaction rates

We follow the approach of Saaltink and al. [*Water Resour. Res.*, 1998] by introducing a kernel matrix U such that $US^T = 0$. We can choose U of the form $\begin{pmatrix} U_{cc} & U_{c\bar{c}} \\ 0 & U_{\bar{c}\bar{c}} \end{pmatrix}$,

We define the total analytic concentration, the total mobile and immobile concentrations for the species as :

$$\mathbf{T} = \mathbf{C} + \bar{\mathbf{C}}, \quad \mathbf{C} = U_{cc} \mathbf{c}, \quad \bar{\mathbf{C}} = U_{c\bar{c}} \bar{\mathbf{c}},$$

The system of the mass conservation can be rewritten as

Mass conservation with total concentrations

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

$$\phi \partial_t \bar{\mathbf{T}} = 0.$$

The chemical equilibrium problem

The subsystem formed by :

Mass action laws

$$\begin{pmatrix} S_{cc} & 0 \\ S_{\bar{c}c} & S_{\bar{c}\bar{c}} \end{pmatrix} \begin{pmatrix} \log \mathbf{c} \\ \log \bar{\mathbf{c}} \end{pmatrix} = \begin{pmatrix} \log K \\ \log \bar{K} \end{pmatrix},$$

+

Definition of the totals

$$\begin{pmatrix} \mathbf{T} \\ \bar{\mathbf{T}} \end{pmatrix} = \begin{pmatrix} U_{cc} & U_{c\bar{c}} \\ 0 & U_{\bar{c}\bar{c}} \end{pmatrix} \begin{pmatrix} \mathbf{c} \\ \bar{\mathbf{c}} \end{pmatrix}.$$

is a closed system that enables computation of the individual concentrations \mathbf{c} and $\bar{\mathbf{c}}$ given the totals \mathbf{T} and $\bar{\mathbf{T}}$.

The chemical sub-problem as a function

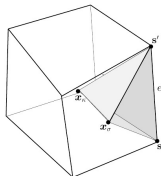
$$\Psi_{\mathbf{C}} : \mathbf{R}^{N_c} \rightarrow \mathbf{R}^{N_c}$$

$$\mathbf{T} \mapsto \Psi_{\mathbf{C}}(\mathbf{T}) = \bar{\mathbf{c}} = U_{c\bar{c}} \bar{\mathbf{c}}$$

its jacobian

$$J_{\mathbf{C}} = \Psi_{\mathbf{C}}'(\mathbf{T}_{:j})_{j=1,\dots,N_h}$$

Discrete advection–diffusion model



Discrete advection diffusion

$$A\mathbf{c}^{n+1} = B\mathbf{c}^n + M\Delta t\mathbf{q}^{n+1}$$

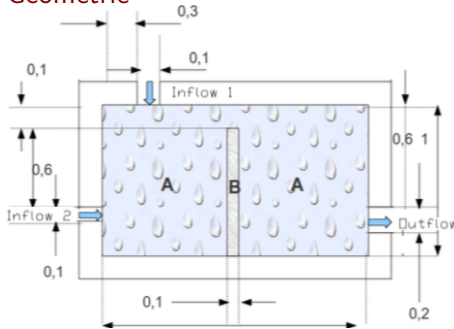
Vertex **A**pproximate **G**radient scheme (Eymard et al., 2012)

ComPASS code(Lopez et al., 2018)

- Multiphase **compositional** thermal Darcy flow model (Coats formation)
- 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, Vassilevski, Wheeler, 2001], [Scheichl, Masson, Wendeburg, 2003]

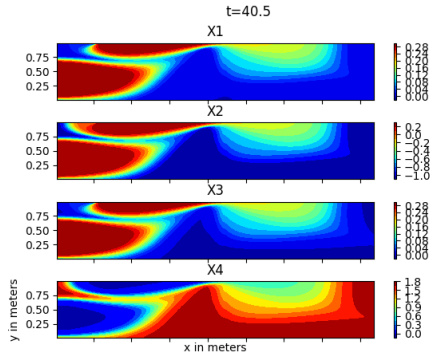
Transport simulation (2D MoMaS Benchmark)

Geometrie



Total	T_1	T_2	T_3	T_4
Init. conc.	0	-2	0	2
Injection	0.3	0.3	0.3	0

Movie
Run



Parameters (Heterogenous)

	k	ω	α_L	α_T
A	0.01	0.25	1e-2	1e-3
B	10 ⁻⁵	0.5	6e-2	6e-3

The coupled formulation

Fixed point (operator splitting):
sequentially solve transport and
chemistry, **easy to code**, **slow**
convergence, **loss of mass**

Chemistry is local \Rightarrow eliminate
individual concentrations at each
point by using the operator Ψ_C .
Only unknowns **C**, **C̄**, **T**

Direct substitution: solve full coupled
system, **accurate**, **robust**, **difficult to**
code, **needs efficient solver**

Coupled formulation

$$\begin{aligned}\phi \partial_t \mathbf{C} + \phi \partial_t \bar{\mathbf{C}} + \mathcal{L}(\mathbf{C}) &= 0, \\ \mathbf{T} &= \mathbf{C} + \bar{\mathbf{C}}, \\ \bar{\mathbf{C}} &= \Psi_C(\mathbf{T}).\end{aligned}$$

Discrete non-linear system ($\mathbf{b}^n = B\mathbf{C}^n + M\bar{\mathbf{C}}^n$)

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

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Using Newton Krylov method

Full Jacobian cannot be stored, size of matrix is $3N_x N_c \times 3N_x N_c$

Inexact Newton method ([Kelley, 1995])

- Solve the linear system by an **iterative** method (GMRES),
- **Approximation** of Newton direction: $\|f'(x_k)d + f(x_k)\| \leq \eta_k \|f(x_k)\|$,
 η_k forcing term
- GMRES requires only **Jacobian matrix by vector products**,

Numerical approximation

[Keyes and Knoll, 2004]

$$Jv = f'(x) v \approx \frac{f(x + \epsilon v) - f(x)}{\epsilon}$$

Expensive, requires solving chemistry

Analytical calculation

$$J_f \begin{pmatrix} v_C \\ v_T \\ v_{\bar{C}} \end{pmatrix} = \begin{pmatrix} (A \otimes I)v_C + (M \otimes I)v_{\bar{C}} \\ -v_C + v_T - v_{\bar{C}} \\ v_{\bar{C}} - J_C v_T \end{pmatrix}$$

Linear preconditioning

Jacobian matrix **not explicitly computed**, but \mathbf{J}_f has a known **block structure**, use only **block preconditioners**

Block Jacobi

$$\mathbf{P}_{\text{BJ}}^{-1} \mathbf{J}_f \mathbf{v} = \begin{pmatrix} v_C & + ((\mathbf{A}^{-1} \mathbf{M}) \otimes \mathbf{I}) v_{\bar{C}} \\ v_C + & v_T - & v_{\bar{C}} \\ - J_C v_T & & \end{pmatrix}$$

Block Gauss Seidel

$$\mathbf{P}_{\text{BGS}}^{-1} \mathbf{J}_f \mathbf{v} = \begin{pmatrix} v_C + & ((\mathbf{A}^{-1} \mathbf{M}) \otimes \mathbf{I}) v_{\bar{C}} \\ v_T - & v_{\bar{C}} + & ((\mathbf{A}^{-1} \mathbf{M}) \otimes \mathbf{I}) v_{\bar{C}} \\ v_{\bar{C}} - J_C v_{\bar{C}} + J_C & ((\mathbf{A}^{-1} \mathbf{M}) \otimes \mathbf{I}) v_{\bar{C}} \end{pmatrix}$$

Remark

Neither \mathbf{A}^{-1} nor the Kronecker product are computed.

$\mathbf{w} = ((\mathbf{A}^{-1} \mathbf{M}) \otimes \mathbf{I}) v_{\bar{C}}$ is computed by solving $\mathbf{A} \mathbf{W} = \mathbf{M} \mathbf{V}_{\bar{C}}^T$ with

$v_{\bar{C}} = \text{vec}(\mathbf{V}_{\bar{C}})$, and $\mathbf{w} = \text{vec}(\mathbf{W})$.

Could use *approximate* solves

Non-linear preconditioning and elimination

Elimination of the unknowns T and C

Reduced system $h(\bar{\mathbf{C}}) = \bar{\mathbf{C}} - \Psi_C \left((A^{-1} \otimes I) (\mathbf{b}^n - (M \otimes I) \bar{\mathbf{C}}) + \bar{\mathbf{C}} \right) = 0.$

Jacobian * vector $J_h \mathbf{v} = \mathbf{v} - J_C J_T \mathbf{v} = \mathbf{v} - J_C \mathbf{v} + J_C \left((A^{-1} M) \otimes I \right) \mathbf{v}.$

Elimination is a **linear change of variables** leads to a **block factorization** of original Jacobian

$$J_f = J_{\tilde{f}} B = \begin{pmatrix} A \otimes I & 0 & 0 \\ -I & I & 0 \\ 0 & -J_C & \mathbf{J}_h \end{pmatrix} \begin{pmatrix} I & 0 & (A^{-1} M) \otimes I \\ 0 & I & -(I - A^{-1} M) \otimes I \\ 0 & 0 & I \end{pmatrix}.$$

Links between elimination and block preconditioning

\mathbf{J}_h is Schur complement of J_f , both BJ and BGS
replace \mathbf{J}_h by I

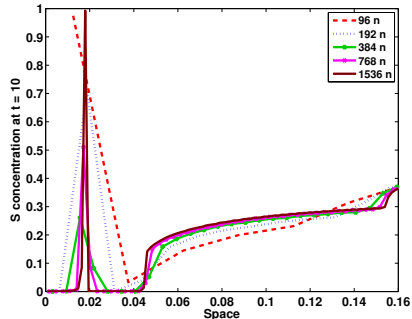
Details: [Amir & Kern, IJNAM, 2019]

MoMaS Benchmark : 1D easy advective case

1D MoMaS Benchmark, [Results : Comp. Geosc. (2010)]

Synthetic benchmark for reactive transport with **significant numerical difficulties** :

- Large variations in equilibrium constants,
- Large stoichiometric coefficients,

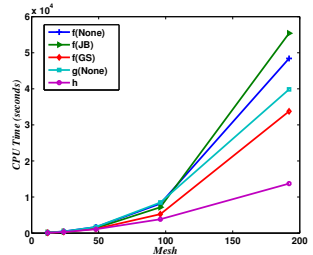
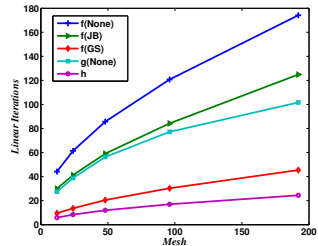
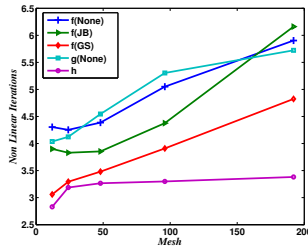


- Our results are in **good agreement** with those of other teams.
- **Intensity** of the peak and its **localization** are close to the reference solution ($x=0.0175$, $S=0.985$)

Influence of preconditioning strategies

* the alternative formulation h has the smallest number both for NL. and L. iterations.

* independence of the mesh size.



* h requires less CPU time than other methods.

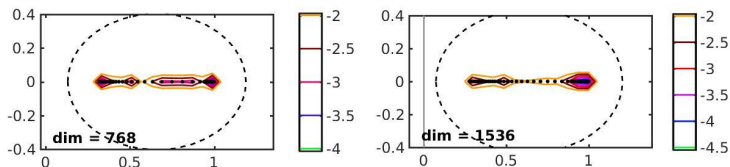
* Good efficiency of h , with BGS preconditioning as a distant second.

* the BJ preconditioning and g (elimination 1 unknown) does not bring an improvement;

Mesh independence convergence of GMRES: field of value explanation

Convergence of GMRES depends of Field of Values

$$W(J_h) = \left\{ \frac{x^* J_h x}{x^* x}, x \in \mathbf{C}, x \neq 0 \right\}. [Eigtool(Embree, Trefethen)]$$



Field of values and pseudo-spectra of J_h for $N_x = 768$ and $N_x = 1536$.

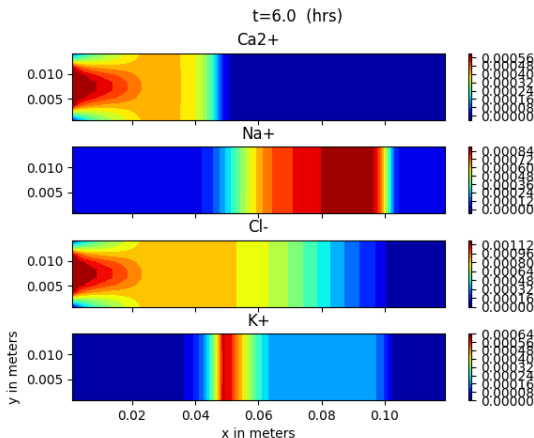
The field of values is inside of the dashed curves, it is bounded away from zero, indepently of mesh size.

Ion exchange with h formulation

EX11 Phreeqc (2D): injection of CaCl_2 only on part of left boundary,

Results

- Chlorine (Cl^-) perfect tracer, **good agreement** with analytical solution [Feike and Dane 1990].
- Ca^{2+} reacts with X^-
 - \Rightarrow a disappearance of KX and NaX
 - \Rightarrow the increase of K^+ and Na^+ .



Conclusion and perspectives

- An **alternative formulation** and **block preconditioners** were used to accelerate the convergence of the Krylov method.
- **Alternative formulation** requires less CPU time, and the number of linear and non linear iterations becomes independent of the mesh.
- More studies and tests are under way for 2D configuration
- Extensions to handle kinetic reactions & minerals.
- Two-phase flow (cf [Ahusborde, M.K., Vostrikov, 2015], [Ahusborde et al., 2017])

Some references



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