



# A non linear preconditioner for coupling transport with chemistry in porous media

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# Outline

- 1 Chemical laws and transport equation
  - Chemical laws in the equilibrium
  - Chemical and transport equations
- 2 Coupled models
  - Coupling by global approach
  - Using Newton-Krylov methods
- 3 Non linear and linear preconditionning
  - Block Jacobi and Block Gauss Seidel
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- 4 Numerical results
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  - EX11 Phreeqc (2D configuration)

## 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,

## Chemical reactions in the equilibrium

We can write  $\Rightarrow$  Chemical system

$$\sum_{j=1}^{N_s} (S_{cc})_{ij} \mathbf{x}_j \Leftrightarrow 0 \quad i = 1, \dots, N_r$$

$$\sum_{j=1}^{N_s} (S_{\bar{c}c})_{ij} \mathbf{x}_j + \sum_{j=1}^{\bar{N}_s} (S_{\bar{c}\bar{c}})_{ij} \bar{\mathbf{x}}_j \Leftrightarrow 0 \quad i = 1, \dots, \bar{N}_r,$$

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

Condensed form

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

# Chemical and advection-diffusion equations

Ideal solution : activity = concentration.

We denote by  $\mathbf{c}_j$  (resp.  $\bar{\mathbf{c}}_j$ ) the concentration of species  $\mathbf{X}_j$  (resp.  $\bar{\mathbf{X}}_j$ ).

Mass action law (logarithmic 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

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

where  $\mathcal{L}$  denotes the advection–diffusion operator :

Advection–diffusion operator (2D)

$$\mathcal{L}(\mathbf{c}) = -\text{div}(\mathbf{D} \text{grad} \mathbf{c}) + \text{div}(\mathbf{u} \mathbf{c})$$

## Mass conservation with total concentrations

### Elimination of the reaction rates

We follow the approach of Saaltink et 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

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

# The coupled formulation

## Transport equation

$$A\mathbf{c}^{n+1} = B\mathbf{c}^n + M\Delta tq^{n+1},$$

Chemistry is local  $\Rightarrow$  we can eliminate the individual concentrations at each point by using the operator  $\psi_C$ . This only leaves  $\mathbf{C}$ ,  $\bar{\mathbf{C}}$ ,  $\mathbf{T}$  as unknowns :

## Coupled formulation

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

## Non linear system

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

with  $\mathbf{b}^n = B\mathbf{C}^n + M\bar{\mathbf{C}}^n$ .



## Using Newton Krylov method

Storage of Jacobian matrix is expensive, size of matrix is  $3N_x N_c \times 3N_x N_c$

Use inexact Newton method :

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

Numerical approximation

$$J_v \approx \frac{f(x + \epsilon v) - f(x)}{\epsilon}$$

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 preconditionning

Since the **Jacobian** matrix **isn't explicitly computed**, and in our model  $\mathbf{J}_f$  has a **block structure**, we choose a **preconditioner defined by block**.

### Block Jacobi

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

### Block Gauss Seidel

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

- Neither  $A^{-1}$  nor the Kronecker product are computed.
- $\mathbf{w} = ((\mathbf{A}^{-1} \mathbf{M}) \otimes \mathbf{I}) \mathbf{v}_{\bar{C}}$  is computed by solving  $\mathbf{A} \mathbf{w} = \mathbf{M} \mathbf{V}_{\bar{C}}^T$  with  $\mathbf{v}_{\bar{C}} = \text{vec}(\mathbf{V}_{\bar{C}})$ , and  $\mathbf{w} = \text{vec}(\mathbf{W})$ .

## Non linear preconditionning

### Elimination of the unknowns T

Elimination of the unknown T from the original system leads to a system with only **C** and  $\bar{\mathbf{C}}$  as unknowns.

### Elimination of the unknowns T and C

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

the Jacobian by vector product

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

## Elimination as a block factorization of the Jacobian

**Elimination** seen as a **linear change of variables**, with matrix

$$B = \begin{pmatrix} I & 0 & (A^{-1}M) \otimes I \\ 0 & I & -(I - A^{-1}M) \otimes I \\ 0 & 0 & I \end{pmatrix}$$

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

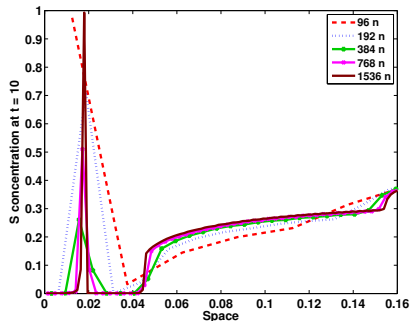
$\mathbf{J}_h$  is Schur complement of  $J_f$ , both BJ and BGS replace  $\mathbf{J}_h$  by  $I$

## MoMaS Benchmark : 1D easy advective case

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

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

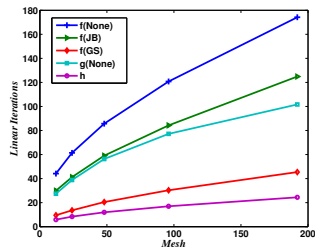
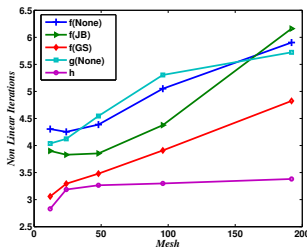
- Large variations in equilibrium constants,
- Large stoichiometric coefficients,



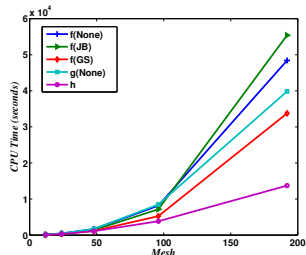
- Our results are in **good agreement** with those of other teams.
- **Intensity** of the peak and its **localization** are close to the reference solution ( $x=0.0175$ ,  $S=0.985$ )

# Influence of preconditioning strategies

- \* the alternative formulation  $h$  has the smallest number both for NL. and L. iterations.
- \* independence of the mesh size.



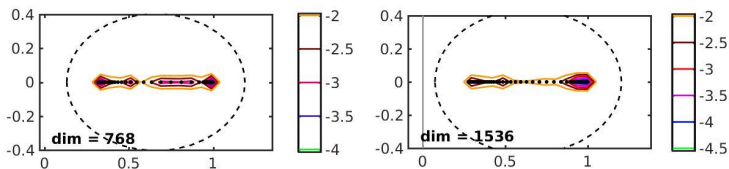
- \*  $h$  requires less CPU time than other methods.
- \* Good efficiency of  $h$ , with BGS preconditioning as a distant second.
- \* the BJ preconditioning and  $g$  (elimination 1 unknown) does not bring an improvement;



# Mesh independence convergence of GMRES: field of value explanation

Convergence of GMRES depends of Field of Values

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



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, independtly of mesh size.

## Application to 2D with $h$ formulation

### Test case : EX11 Phreeqc (2D)

The injection of  $CaCl_2$  occurs only on part of the left boundary, giving rise to 2D solution.

$$\bar{C}_{BC} = [6 * 10^{-4}, 0, 1.2 * 10^{-3}, 0]$$

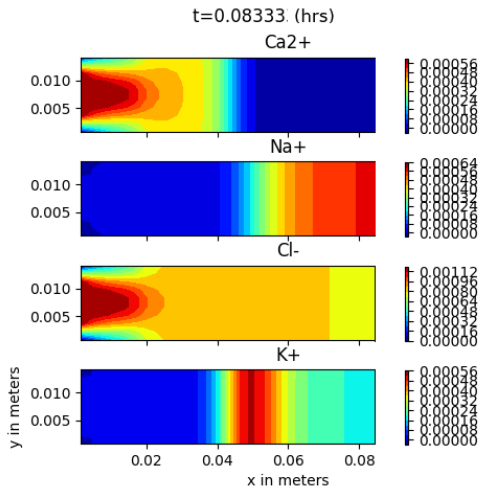
### Results

\* Chlorine ( $Cl^-$ ) is a perfect tracer. Our results show a **good agreement** with the analytical solution [Feike and Dane 1990].

\*  $Ca^{2+}$  reacts with  $X^-$

⇒ a disappearance of  $KX$  and  $NaX$

⇒ 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.
- The **alternative formulation** **requires less CPU time**, and the **number of linear and non linear iterations becomes independent of the mesh**.
- The alternative formulation is applied to **2D test case for ion exchange**, and it gives a good results.
- More studies and tests are under way for 2D configuration
- Extensions to handle kinetic reactions.

## Some references



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- **Code ComPASS** (BRGM and LJAD-INRIA)   
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- **VAG scheme** (Vertex Approximate Gradient scheme)  
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