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LINEAR AND NONLINEAR PRECONDITIONING FOR REACTIVE TRANSPORT

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Summary. Reactive transport in porous media leads to a large nonlinear system of equations, coupling one advection-dispersion type partial differential for each species, to algebraic (if local equilibrium is assumed) or ordinary differential (if a kinetic model is chosen) equations for each grid point. The coupled problem is formulated as a fixed point problem, by using local inverses for each of the subproblems. By applying the Newton-Krylov framework on the resulting non-linear system, a globally coupled method is obtained with the complexity of a fixed point method, making it possible to keep the transport and chemistry codes separated. At each time step, the linear system for Newton's method is solved by an iterative method of Krylov type, requiring only the computation of the Jacobian of the system with a given vector, but not the computation of the Jacobian matrix itself. A critical aspect of the method is an efficient matrix-free preconditioner. Preconditioning techniques that respect the block structure of the system are introduced,a and several block preconditioners are compared. As the transport problem is solved as part of the nonlinear preconditioning, a con- vergence rate independent of the mesh size is expected. Indeed, an analysis of a simplified chemical system for a single species with sorption shows that the eigenvalues of the preconditioned Jacobian matrices are bounded independently of the mesh size, so that the number of outer Newton iterations, as well as the number of inner GMRES iterations, are independent of the mesh. The resulting preconditioner can be extended to the case with multi- species chemistry.

1 INTRODUCTION – PROBLEM STATEMENT

Reactive transport models lead to a set of coupled partial differential equations, coupled with algebraic (in the case of equilibrium reactions) or ordinary differential (in the case of kinetic reactions) equations. The system may be very large, as the number of unknowns

is the number of grid points times the number of chemical species. In Amir and Kern[1] a method was introduced where the chemical equations are eliminated, and a set of transport equations are solved, with a source term implicitly representing the effect of chemistry. The resulting problem is solved by the Newton–Krylov method, where the linear system is solved by an iterative method. It was seen that an efficient preconditioning was a crucial component of the method. However, finding a preconditioner is difficult, as no matrix is constructed in the Newton–Krylov method, and one would like to preserve the decoupling between transport and chemistry that is the main advantage of the formulation in Amir and Kern.[1]

In this work, we consider a simplified model with one species undergoing a sorption reaction, given by a known equilibrium isotherm. This choice is motivated by the facts that the resulting mathematical problem has the same structure as that considered in the more general multicomponent model, that it is amenable to a more complete analysis, and that it can still be seen as representative of a physically relevant model (see for example the book by Logan[2]).

We denote by c the aqueous concentration (in mol/L) of the species, and by $\bar{c}$ that of the solid part. The mathematical model given by writing the mass balance equation, and the adsorption relation is:

$$\begin{aligned} &\omega\partial_t c + \omega\partial_t \bar{c} - \nabla.(D\,\nabla c - qc) = 0 \\ &\bar{c} = \psi(c) \end{aligned} \qquad \text{in } \Omega \times [0,T] \tag{1}$$

with appropriate initial and boundary conditions. Here Ω is a bounded domain in $\mathbf{R}^d$, $1 \leq d \leq 3$, $[0,T]$ a fixed time interval, q is the Darcy velocity, ω is the porosity and D is the diffusion–dispersion tensor.

The *sorption isotherm* ψ in equation (1) will be taken as the Langmuir isotherm (see the above references)

$$\psi(c) = \sigma \frac{Kc}{1+Kc}, \tag{2}$$

where σ and K are two constants.

For completeness, we briefly describe how equation (1) is discretized. We assume the Darcy velocity has been computed (typically by mixed finite elements, see for instance Mosé et al.[3]). We follow the method described in Siegel et al.[4] and use a splitting method, where advection and diffusion-dispersion are solved separately at each time step. This has the advantage that each physical phenomenon is solved with an appropriate method. The drawbacks are that the method is (formally) first order and that is may be difficult to implement boundary conditions. The advective part is approximated with an upwind cell centered finite volume method, and the dispersive part is approximated by a mixed finite element method. For the time derivative, we restrict to a simple Euler method. The advective pat is treated explicitly, and the dispersive part is treated implicitly, so that a

discrete version of (1) can be written as

$$\begin{aligned} &M\frac{C^{n+1}-C^n}{\Delta t}+M\frac{\bar{C}^{n+1}-\bar{C}^n}{\Delta t}+L_a\,C^n+L_d\,C^{n+1}=b^n,\\ &\bar{C}^{n+1}=\psi(C^{n+1}). \end{aligned} \tag{3}$$

where C^n (resp. $\bar{C}^n$) is a vector representing an approximation of c (resp. $\bar{c}$) on the given mesh at time t^n, M is a mass matrix (in this work we assume M is diagonal) , L_a and L_d are matrices for the advective and dispersive parts respectively and b^n takes into account non-homogeneous boundary conditions.

2 COUPLED FORMULATIONS

At each time step, one has to solve the nonlinear system given by equation (3). We continue to pretend that equation (3) is representative of more realistic reactive transport problem, and strive to keep the "transport" part and the "chemistry" part (evaluating ψ) separate. Thus we assume that we can solve the problem (pure transport)

$$M\frac{C^{n+1}-C^n}{\Delta t}+L_a\,C^n+L_d\,C^{n+1}=f^n \tag{4}$$

where f^n now stands for a known source term, and we write $A=M+\Delta t L_d$ for the matrix to be inverted at each time step (in our case, A is symmetric and positive definite). We note that for realistic 3D problems, this is a computationally demanding task, but that is a well understood problem, for which robust and efficient methods exist, see the above references.

The nonlinear system takes the form

$$\begin{aligned} &AC^{n+1}=g^n-M\bar{C}^{n+1}\\ &\bar{C}^{n+1}=\psi(C^{n+1}), \end{aligned} \tag{5}$$

where g^n is a know term that lumps together the boundary conditions and the initial conditions. For simplicity, in the following we drop the time superscripts from the notation.

We can obtain two mathematically equivalent formulations by eliminating one the unknowns:

Eliminiation of $\bar{C}$ We solve the nonlinear system

$$AC+M\psi(C)=g \tag{6}$$

This is actually very close to the well known Global Implicit Approach commonly used in reactive transport studies (see the survey by Yeh and Tripathi[5]). This formulation makes it difficult to keep transport and chemistry separate.

Elimination of C We solve the nonlinear system

$$\bar{C} = \psi(A^{-1}(g - M\bar{C})). \qquad (\bar{CP})$$

This formulation looks complicated because of the presence of A^{-1}, but is actually fairly easy to implement. As usual, the inverse of A is not actually computed. Rather, when one needs to evaluate the residual, one simply solves a linear system with the matrix A, and this turns out to the building block singled out before, namely the solution of one transport step, with a source term given by $M\dfrac{\bar{C}^{n+1} - \bar{C}^n}{\Delta t}$, which is assumed known in this context.

Note that all three formulations (5), (6) and ($\bar{CP}$) have the form of a fixed point problem. They could be solved by means of a fixed point algorithm. As an example, or the coupled formulation, the iterations take the form of the familiar operator splitting method (also called Standard Iterative Algorithm, or SIA, see for instance Yeh and Tripathi[5]):

$$\begin{aligned} AC^{k+1} &= g - M\bar{C}^k \\ \bar{C}^{k+1} &= \psi(C^{k+1}), \end{aligned} \qquad (7)$$

where the superscript k now denotes an iteration index.

However, we wish to solve the nonlinear systems by means of Newton's method.In addition to computing the residual of the nonlinear system function, the main computational task is the solution of a linear system at each iteration. In the Newton–Krylov method, this linear system at each Newton iteration is itself solved by means of an iterative method, typically GMRES (a survey on Newton–Krylov methods can be found in[6]). The Newton–Krylov method has been succesfuly applied to reactive transport problems by Hammond et al.,[7] who used a substittuion approach for the coupled problem (substitue the chemical equation in the transport operator, and solve the resulting nonlinear problem). This work follows the formulation proposed by Amir and Kern,[1] where the formulation rests on using the solution operators for both transport and chemistry.

From an implementation point of view, the main feature of the method is that one does not need to know the full Jacobian matrix, but only its application to a given vector. For the problem at hand, the Jacobian matrices are, respectively

$$J_{CP} = \begin{pmatrix} A & M \\ -D^k & I \end{pmatrix} \qquad (8)$$

for the coupled formulation,

$$J_C = A + MD^k \qquad (9)$$

for the formulation where $\bar{C}$ is eliminated, and

$$J_{\bar{C}} = I + D^k A^{-1} M \qquad (10)$$

for the formulation where C is eliminated.

We have denoted by D^k the diagonal matrix $D^k = \mathrm{diag}(\psi'(C_1), \ldots, \psi'(C_N))$, where N is the number of discrete unknowns.

When iterative methods are applied to solve linear system, finding an efficient preconditioner is crucial to the success of the method. For the coupled problem, we can only use preconditioners that respect the block structure of the method. Two simple choices are

Jacobi preconditioner, where one uses the diagonal of the matrix. This completely uncouples transport and chemistry.

Gauss–Seidel preconditioning, where one uses the block-lower part of the matrix. It can be shown that one step of Gauss–Seidel preconditioning is equivalent to one iteration of the SIA method, thus providing a justification for the use of SIA as a preconditioner for a fully coupled approach, albeit in a linear

It is also possible to carry out the eliminations at the linear level (that is starting with the linear system (8)). Then one sees that the Jacobian matrix obtained when C is eliminated is the Schur complement of the matrix of the coupled problem. Because the transport equation is linear, and for the "full" Newton's method, the iterates obtained are identical. We still prefer to perform the elimination at the nonlinear level, as the method carries over to more general situations. We thus view the methods where one of the unknowns is eliminated as an instance of nonlinear preconditioning in the sense of.[8]

In all cases, the important feature is that we invert the transport operator, and this will lead to a bounded operator. At the discrete level, this will translate to a mesh-independent convergence of the linear iterations.

3 SPECTRAL ANALYSIS OF THE LINEAR SYSTEMS

We start by computing the eigenvalues of the various matrices. We denote by μ_j, $j = 1, N$ the eigenvalues of the (generalized) eigenproblem $Aw = \mu M D^k w$ (the μ_j are real positive numbers because M was assumed diagonal and D^k is diagonal and positive, if ψ is an increasing function).

Jacobi preconditioning The eigenvalues of the preconditioned matrix are the numbers $\lambda_j = 1 \pm i\dfrac{1}{\sqrt{\mu_j}}$, $j = 1, \ldots, N$.

Gauss–Seidel preconditioning The eigenvalues of the preconditioned matrix are the numbers 1, with multiplicity N and $\lambda_j = 1 + \dfrac{1}{\mu_j}$, $j = 1, \ldots, N$.

Elimination of C The eigenvalues are the numbers $\lambda_j = 1 + \dfrac{1}{\mu_j}$, $j = 1, \ldots, N$. They are the same as for Gauss–Seidel preconditioning.

We now assume that, for large N, μ_j behaves as $1/h^2$, where h is a measure of the mesh-size. This is a natural assumption since the matrix L approximates a diffusion operator. One then sees that in all three cases, the eigenvalues are tightly clustered around 1.

However, it is well known that the convergence of the GMRES method is not determined by the spectrum of the matrix alone (see Greenbaum et al.[9]). One needs further information, which could be the condition number of the eigenvector matrix, or information on the field of values of the matrix (see Embree[10]).

The field of values of a matrix is defined as the set of Rayleigh quotients of the matrix

$$W(A) = \left\{ \frac{x^* A x}{x^* x},\ x \in \mathbf{C}^N,\ x \neq 0 \right\}.$$

It is a convex set that contains the eigevalues of A (and is strictly larger than the eigenvalues, unless A is a normal matrix).

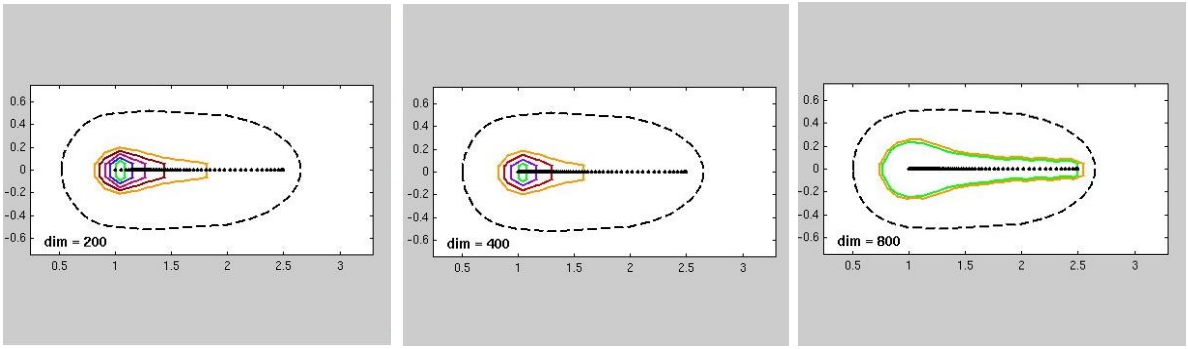

Figure 1: Field of values of the Jacobian matrix, for different mesh sizes

We have numerically computed the field of values for several mesh sizes, using the Eigtool software[11] . The field of values is the inside of the dashed curves, and one sees it is bounded away from zero, independently of h.

4 NUMERICAL RESULTS

Figure 2 compares the convergence of the fixed-point and Newton–Krylov methods, showing clearly the advantage of Newton's method, at least for the number of iterations.

We have compared the performance of the Newton–Krlyov method, wkith different preconditioners. In the tables below, "None" refers to no preconditioning (using the matrix J_{CP} in (8) directly, "BJ" and "GS" refer to block Jacobi (resp. block Gauss–Seidel preconditioning) and "Elim. of C" means using matrix $J_{\bar{C}}$ in (10).

We have compared two implementations of the Newton–Krylov method. In the first one ("constant forcing term", table 1 below), the tolerance used when solving the linear system is kept constant (with a value of 10^{-12} in this experiment). In the second one, an

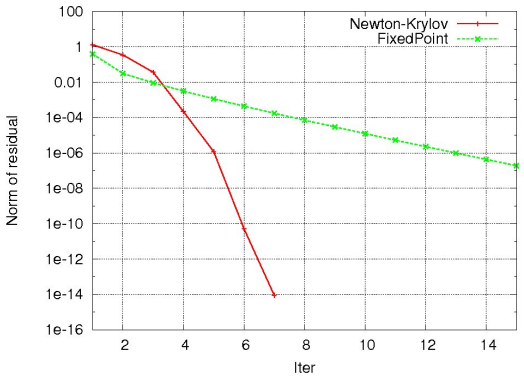

Figure 2: Comparison of fixed–point and Newton performance

adaptive tolerance (strategy 2 in Eisentat and Walker[12]) is used, where the tolerance is tightened as the nonlinear iterations progress.

Mesh/PC	h		$h/2$		$h/4$		$h/8$		$h/16$	
	NNI	NLI	NNI	NLI	NNI	NLI	NNI	NLI	NNI	NLI
None	3	104	3	167	3	275	3	453	—	—
BJ	3	68	3	67	3	63	3	60	3	62
BGS	3	48	3	48	3	47	3	45	3	44
Elim. of C	3	41	3	41	3	41	3	40	3	40

Table 1: Performance of preconditioners, constant forcing term: NNI is Number of Nonlinear Iterations, NLI is Number of Linear Iterations

As expected, the number of nonlinear iterations is either independent of the mesh (for a fixed tolerance) or grows slowly as the mesh is refined. The number of nonlinear iterations increases when an adaptive tolerance is used. However, the number of linear iterations is smaller, by a factor of almost 3.

As far as the preconditioners are concerned, it can be seen that when using non preconditioner, the number of linear iterations grows quickly when the mesh is refined. When a preconditioner where transport is eliminated is used, the number of linear iterations becomes almost independent of the mesh, as predicted by the spectral analysis from the previous section.

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Mesh/PC	h		$h/2$		$h/4$		$h/8$		$h/16$	
	NNI	NLI	NNI	NLI	NNI	NLI	NNI	NLI	NNI	NLI
None	8	42	8	76	10	105	10	177	—	—
BJ	7	27	7	27	7	26	7	26	7	26
BGS	8	23	7	24	7	22	8	25	8	24
Elim. of C	5	15	5	15	5	15	5	15	5	15

Table 2: Performance of preconditioners, adaptive forcing term: NNI is Number of Nonlinear Iterations, NLI is Number of Linear Iterations

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