INTERNATIONAL JOURNAL OF
NUMERICAL ANALYSIS AND MODELING
Volume 16, Number 1, Pages 18–48

PRECONDITIONING A COUPLED MODEL FOR REACTIVE TRANSPORT IN POROUS MEDIA

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Abstract. Reactive transport problems involve the coupling between the chemical interactions of different species and their transport by advection and diffusion. It leads to the solution of a non-linear systems of partial differential equations coupled to local algebraic or differential equations. Developping software for these two components involves fairly different techniques, so that methods based on loosely coupled modules are desirable. On the other hand, numerical issues such as robustness and convergence require closer couplings, such as simultaneous solution of the overall system. The method described in this paper allows a separation of transport and chemistry at the software level, while keeping a tight numerical coupling between both subsystems. We give a formulation that eliminates the local chemical concentrations and keeps the total concentrations as unknowns, then recall how each individual subsystem can be solved. The coupled system is solved by a Newton–Krylov method. The block structure of the model is exploited both at the nonlinear level, by eliminating some unknowns, and at the linear level by using block Gauss-Seidel or block Jacobi preconditioning. The methods are applied to a 1D case of the MoMaS benchmark.

Key words. Reactive transport, porous media, preconditioning, non-linear systems.

1. Introduction

Reactive transport in porous media studies the coupling of reacting chemical species with mass transport in the subsurface, see [5, sec. 7.9] for an introduction, or [3, 7] for more comprehensive references. It plays an important role in several applications when modeling subsurface flow [64, 63, 74] :

- chemical trapping is one of the mechanism by which the safety of geological sequestration of carbon dioxide in deep saline aquifers can be ensured [4, 25, 57, 66];
- nuclear waste storage is based on a multiple barrier concept so as to delay the arrival of radionuclides in the bio-sphere. The concrete barrier that seals the repository may be attacked by oxidized compounds, and again its safety needs to be assessed [18, 45, 58];
- bio-geochemistry involves interactions with organic chemicals, and is important for studies of soil pollution, and also for planning bio-remediation [15, 47].

The numerical simulation of reactive transport has been the topic of numerous works. The survey by Yeh and Tripathi [72] has been very influential in establishing a mathematical formalism for setting up models, and also for establishing the "operator splitting" approach (see below) as a standard. More recent surveys, detailing several widely used computer codes and their applications, can be found in the book [74] and the survey article [64].

One traditionally distinguishes two main approaches for solving reactive transport problems:

Received by the editors November 22, 2017.
2000 *Mathematics Subject Classification.* 65F10, 65H10, 65M22, 76S05.

Sequential Iterative Approach (SIA): in this family of methods (also known as operator splitting), transport and chemistry are solved alternatively [14, 41, 48, 59, 73]. This has the advantage that a code for reactive transport can be built from pre-existing transport and chemistry codes [33], and that no global system of equations needs to be solved. On the other hand, the splitting between chemistry and transport may restrict the time step in order to ensure convergence of the method, and the splitting may also introduce mass errors that need to be controlled [67]. As the above references show, the method can be quite successful if it is implemented carefully.

Globally implicit Approach (GIA): Here the fully coupled system involving transport and chemistry is solved in one shot (see for instance [24, 56] and the references below). The balance is the opposite of what it was for SIA: the method does not introduce spurious mass errors, and can converge with large time steps, but it leads to a large and difficult to solve system of non-linear equations coupling all the chemical species at all grid points. The system can be solved by substituting the mass action laws into the conservation equations, a variant known as Direct Substitution approach (DSA) as in [25, 28]. Recently, methods that solve the coupled problem without direct substitution have been introduced. In [29, 39, 40], local chemical systems are solved, and the solution becomes an additional term in a transformed form of the transport. In [16, 21, 22], the overall coupled system is solved as a Differential Algebraic System.

In addition to the surveys mentioned above, the methods have been compared in several studies or benchmarks [11, 46, 55]. It is fair to say that no method or code emerges as a clear winner for all situations.

We finish this short (and far from exhaustive) review of the literature by noting that most of the work cited deal with one-phase flow and transport. The methods have recently been extended to the case of two-phase flow [1, 25, 57, 61] .

In a previous paper [2], the authors introduced a method that belongs to the GIA family without sacrificing the ease of implementation of the SIA methods, due to the separation of software modules for chemistry and transport. The method solves the nonlinear coupled system by a Jacobian-free Newton–Krylov method.

This paper presents several improvements to [2]:

- a systematic study of preconditioning methods for the *linearized coupled problem*, and their relationship to elimination methods. This results in a method where the mobile concentrations are eliminated, and one solves for the total fixed concentrations. We give both heuristic arguments and experimental evidence that the convergence rate for both the Newton and GMRES iterations is bounded independently of the mesh size;
- a formulation of the *chemical equilibrium problem* that does not need the a priori separation of the chemical species between primary and secondary components, but that keeps the distinction between mobile and immobile species.

The method originally introduced in [2] approximated the Jacobian matrix by vector product by finite difference, so that only solvers for transport and chemistry were needed, and they could be applied as black boxes. In this work, we slightly "open the boxes", as we propose to compute exactly the Jacobian matrix by vector products, so that access to the Jacobians of the transport and chemical solvers are needed. We feel the added accuracy and reduced cost (for the chemical part,