

APPLICATION OF FLUX CORRECTED TRANSPORT FOR MULTICOMPONENT REACTIVE TRANSPORT MODELING

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Summary. Reactive transport describes the coupling among flow, transport and chemical reactions occurring in porous media; and therefore it is important in areas such as aquifer remediation, groundwater contamination, geochemical reaction modeling and CO₂ sequestration. Due to the large time scales involved and/or the lack of accessibility to the subsurface, in most of these studies an adequate representation of the important physical and chemical processes require numerical simulation. Conventional numerical methods to solve the reactive transport equations can produce non-physical values (negative concentrations), particularly for cases with highly heterogeneous flow fields, irregular grids and full tensor dispersion. Negative concentrations can cause numerical convergence problems and therefore cannot be tolerated, specially when transport is coupled to nonlinear reactions. These problems can be solved using a methodology called flux corrected transport, in which a positive solution can be obtained combining a high-order solution (high accuracy but non-positive) and a low-order solution (positive but dispersive). The numerical dispersion of the low-order solution is corrected using an antidiffusion component obtained from high-order solution in order to improve its accuracy. In regions of non-physical values, the low-order solution predominates while the high-order solution is prevalent in the areas of sharp changes. In this study, flux corrected transport is tested in reactive transport problems with spatial variability and diagonal flow, and the results indicate that this is a simple and flexible method to obtain a positive solution in a reactive transport framework. This methodology can be easily adapted to existing discretizations, therefore there is no need to extensively modify existing codes.

1 Introduction

Reactive transport describes the coupling among flow, transport and chemical reactions occurring in porous media; and therefore it is important in areas such as aquifer reme-

diation, groundwater contamination, geochemical reaction modeling and CO₂ storage in geological formations. Modeling of mixing in reactive transport requires the specification of full dispersion tensor, nonlinear reaction rates, incorporation of spatial heterogeneity and irregular grids. In general reactive transport models require the solution of a nonlinear system of partial differential equations, and Newton’s method is often used. Certain authors argue that convergence is facilitated for solving the logarithm of the concentration change because in many geochemical problems the concentration can range over many orders of magnitude. This is called the log formulation approach to reactive transport modeling^{1,2,11} Nonnegative solutions are a requirement for the convergence of the nonlinear system of reactive transport equations but in some cases negative concentrations appear³. These spurious results can cause numerical convergence problems and therefore cannot be tolerated, especially when the log formulation is used. Oscillations and non-physical negative concentrations can arise due to high-order discretization of the advection term, use of full-tensor dispersion, and irregular grids³

Possible solutions to address these non-physical negative concentrations include the Total Variation Diminishing (TVD) approach⁴, Flux limiters⁵ and Flux Corrected Transport (FCT)⁶. Approaches for avoiding difficulties arising from full-tensor dispersion include multi-point flux approximation (MPFA)^{9,10} and nonlinear finite volume discretizations¹² In recent years, an extension of FCT to Finite Element Modeling (FEM) and fully implicit timestepping has been developed⁷. The purpose of this work is to apply FCT with a fully implicit time stepping to obtain a positive solution of the log-formulation for solution of multicomponent reactive transport equations.

2 Governing equations

The Advection-Dispersion-Reaction equation for an aqueous component Ψ_k is given by¹:

$$\frac{\partial \phi \Psi_k}{\partial t} = \underbrace{\frac{\partial}{\partial x_i} \left(\phi D_{ij} \frac{\partial \Psi_k}{\partial x_j} \right)}_{\text{Dispersion}} - \underbrace{\frac{\partial (\phi v_i \Psi_k)}{\partial x_i}}_{\text{Advection}} \pm R_k \quad (1)$$

where ϕ is the porosity, v_i is the velocity of species i , D_{ij} are the components of the dispersion tensor and R_k is the reaction rate. Speciation can be represented as $\mathbf{c} = f(\mathbf{\Psi})$ where \mathbf{c} is the vector that contains the free-ion concentration. The reaction rates R_k are in general nonlinear for most of the problems of interest. This reaction rate term (source/sink) for a precipitation/dissolution reaction can be for example:

$$R_k = k_r(T)[c_{H^+}]^\alpha \left(1 - \frac{Q_p(c_j)}{K_{sp}} \right) \quad (2)$$

The discretization of equation 1 via finite volume or finite element methods leads to

$$\mathbf{L}[\Psi^{n+1}] = \mathbf{R}[\Psi^n] \quad (3)$$

where the superscript n refers to the time step, \mathbf{L} and \mathbf{R} are matrices and the criteria for a positive solution of this linear system is given by two conditions⁷:

1. \mathbf{L} must be a M-Matrix, that is, $L_{ii} > 0$ and $L_{ij} < 0$, for $i \neq j$
2. All entries in \mathbf{R} must be positive, $R_{ij} > 0$

Using standard finite volume or finite element discretization of full dispersion tensor on a structured grid results in condition 2 is being satisfied but not condition 1 leading to the occurrence of negative concentrations³ (Note that we use a fully implicit time discretization; use of Crank-Nicolson time discretization may also lead to negative concentrations under some conditions). There are additional numerical oscillations if the advection component is discretized using central difference. The next section describes how we use the FCT approach to correct these deficiencies.

3 Methodology

The general idea of the FCT is to use a combination of a monotone low-order scheme where the solution does not comply with the physical limits (in this case negative concentrations), and a high order scheme when the solution is smooth. In other words, using the high-order solution (low numerical dispersion but with oscillations), some amount of antidiffusion is added in such a way that the mass balance and positivity is preserved. The amount of antidiffusion is defined using the low order solution (high numerical dispersion but monotone). The methodology is the following^{6,7}:

- Discretization of the transport equation. In this case the, Advection-Dispersion-Reaction Equation can be discretized in space as:

$$\mathbf{M} \frac{d\Psi}{dt} = \mathbf{K}\Psi + R(\Psi) \quad (4)$$

where \mathbf{K} is the discrete operator that includes advection-dispersion, and \mathbf{M} is the consistent mass matrix (Note that for standard finite volume methods, \mathbf{M} equals the Identity matrix), and $R()$ is the reaction rate (For simplicity it is assumed that the reaction rate depends on the concentration of the components Ψ_k). concentrations for the.

- Replace \mathbf{M} by the diagonal matrix $\mathbf{M}_L = \text{diag}(\mathbf{M})$
- Add artificial diffusion to eliminate negative off-diagonal terms of \mathbf{K} and define the low order operator \mathbf{L} :

$$\mathbf{L} = \mathbf{K} + \mathbf{D} \quad (5)$$

where \mathbf{D} is just a linear combination of coefficients in the original matrix \mathbf{K} (see references^{6,7}).

- Then the original equation is transformed to:

$$\begin{aligned} M_L \frac{d\Psi}{dt} &= \mathbf{L}\Psi + R(\Psi) \\ &= (\mathbf{K} + \mathbf{D})\Psi + R(\Psi) \end{aligned} \quad (6)$$

The solution of this equation is always positive but with high numerical dispersion.

- To deal with the numerical dispersion introduced before, add an antidiffusion component \mathbf{F} to the low order operator \mathbf{L} to preserve mass balance and positivity:

$$M_L \frac{d\Psi}{dt} = \mathbf{L}\Psi + \mathbf{F}\Psi + R(\Psi) \quad (7)$$

where the antidiffusion is defined as

$$\mathbf{F} = f_{ij} = -\alpha \mathbf{D} \quad (8)$$

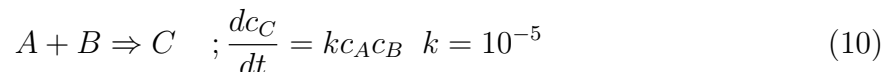
The specific amount of antidiffusion added to preserve these properties α is called Zalesak flux limiter⁸. We do not include details for computing α which is a function of the concentration; (usually the concentration at the prior time step or iteration). The Computation of the flux limiter is an important part of the method and can be computationally demanding. For more details consult the original references^{7,8}

- The corrected solution (without negative concentrations and mass preserving) in this case is given by:

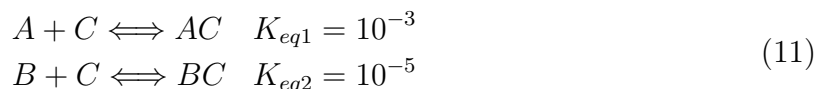
$$M_L \frac{d\Psi}{dt} = \mathbf{L}\Psi + \alpha \underbrace{(\mathbf{K} - \mathbf{L})}_{\mathbf{D} \text{ from equation 5}} \Psi + R(\Psi) \quad (9)$$

4 Test Problem

The FCT methodology has been tested for a simple problem where a pulse is injected in the middle of a heterogeneous domain. The chemical system is composed of only 1 reaction:



The speciation reactions are:



And with them, the following components are defined:

$$\begin{aligned} \Psi_A &= c_A + c_{AC} \\ \Psi_B &= c_B + c_{BC} \\ \Psi_C &= c_C + c_{AC} + c_{BC} \end{aligned} \quad (12)$$

In the pulse region, a high concentration of species A and low concentrations of B and C are specified, while the background region is the opposite situation (see figure 1). Anisotropic dispersion is assumed as the only transport mechanism occurring in this problem. Although this is unrealistic in practice, we select this case in order to focus on the non-physical oscillations caused solely by the full-tensor dispersion. We follow the standard model of Bear^{13,14} and others, where the dispersion tensor is a function of the longitudinal and transverse dispersivity components and the local velocity vector. The boundary conditions for the flow problem vary linearly to induce a diagonal flow. The hydraulic conductivity is assumed heterogeneous and the streamlines indicate the presence of a complex flow pattern with diagonal trend (see figure 1). Constant flow boundary conditions are assumed for the transport problem in the left and lower sides of the domain.

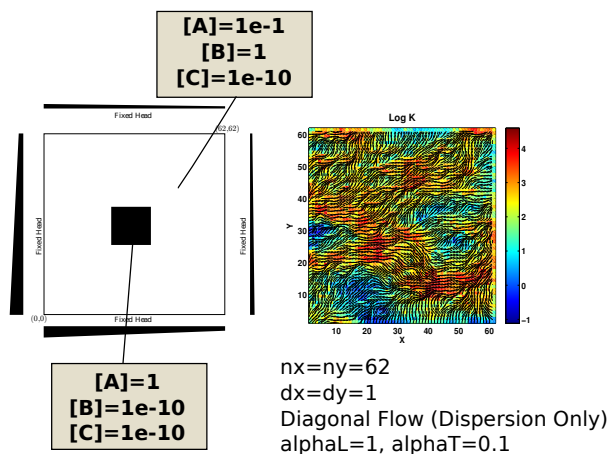


Figure 1: Configuration of the test problem

5 Results

Figure 2 shows the concentration maps for species A , B and C obtained for the high-order (see figure 2(a)) and FCT solutions (figure 2(b)). The negative concentrations of species A , C and reaction rate are evident in the high-order solution (white areas in figure 2(a)). The solution obtained using the FCT approach preserves positivity in the concentrations of the chemical species and reaction rate (see figure 2(b)).

Figure 3 includes some profiles of the solutions taken along the main diagonal of the domain. From these, the low-order solution is positive but shows a clear diffusive behaviour as expected (red line). The FCT solution is closer to the high-order solution (conventional solution) than the low-order (lines black and blue, and zoomed box on the upper left of figure 3). The low-order and FCT solutions preserve the positivity and mass conservation (not shown) when compared with the high-order solution (zoomed box in the lower right of figure 3)

6 Preliminary conclusions

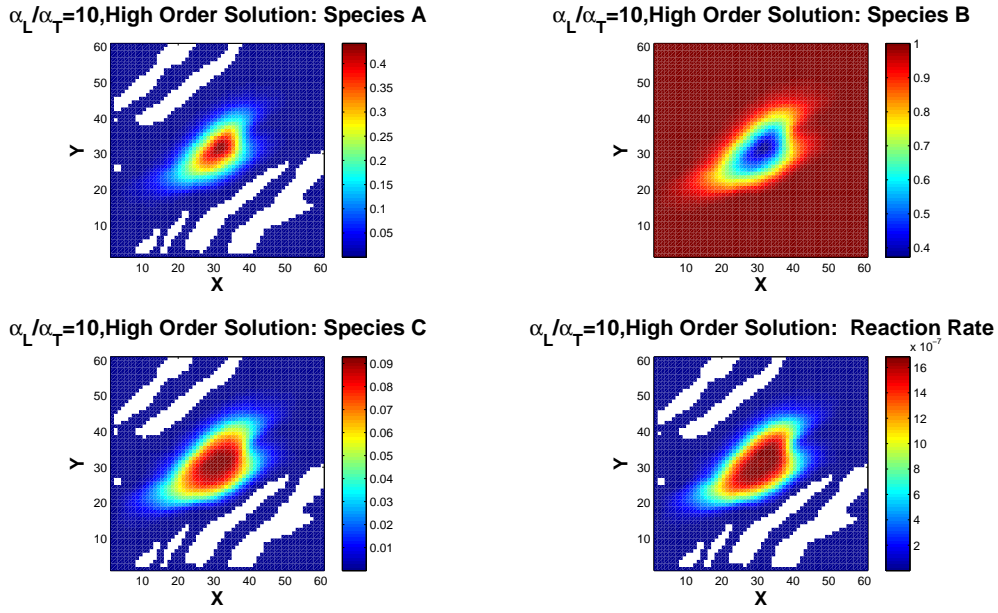
Flux Corrected Transport is a useful methodology to correct the negative concentrations that appear during reactive transport simulations while preserving the mass balance. This is especially important when the solution of the nonlinear systems of equations uses the logarithm of the concentration change³ because this only converges with the low-order and FCT methods when the negative concentrations are present. The FCT approach can be readily incorporated into existing FDM, FV, FEM codes in a non-intrusive way.

Acknowledgements

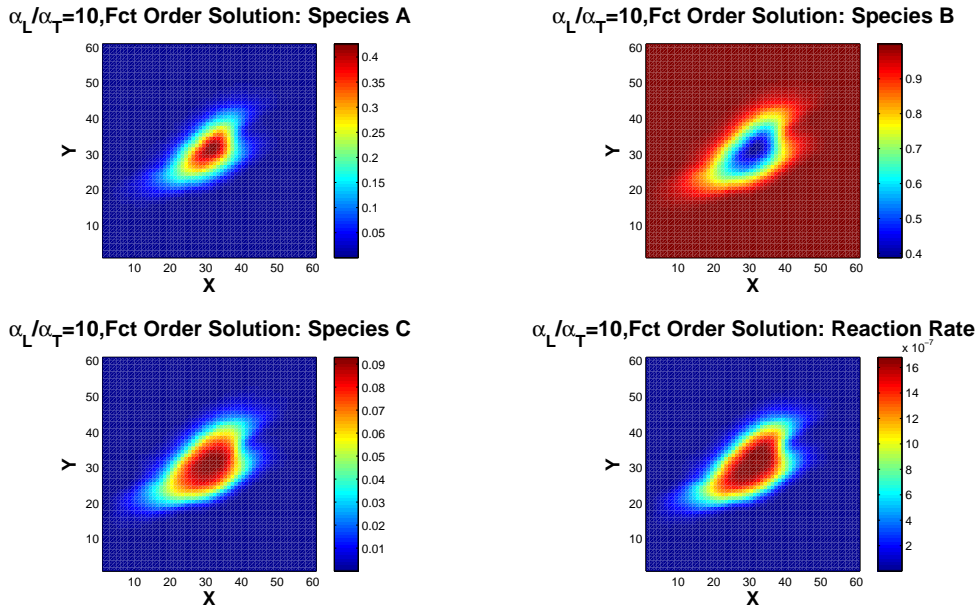
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(a) High Order Solution for species A, B and C . White areas (negative concentrations)



(b) FCT solution

Figure 2: Spatial distribution of the concentration of species A, B and C and reaction rate

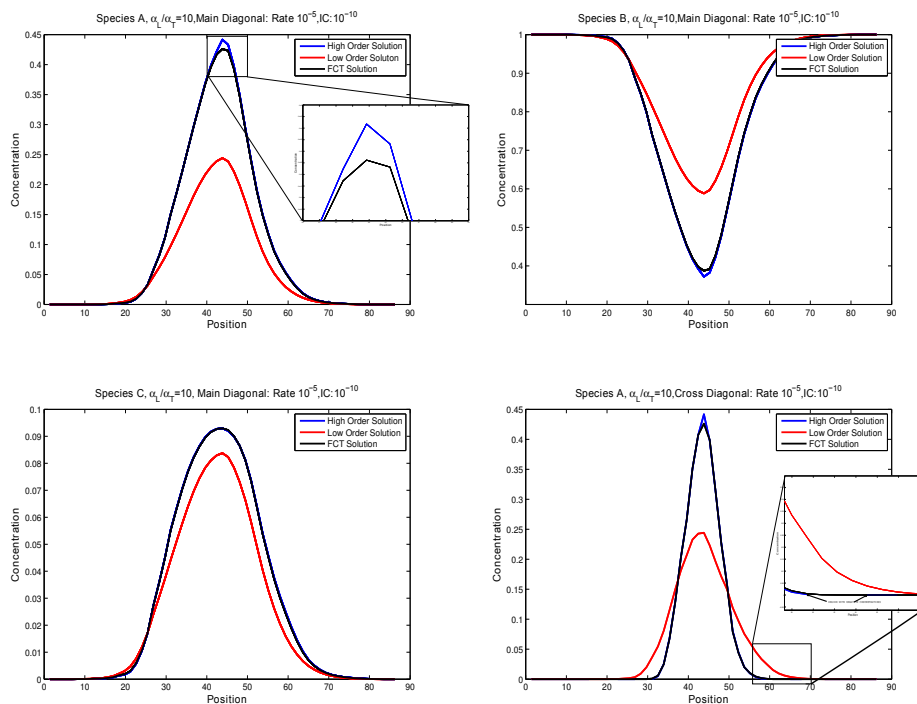


Figure 3: Concentration profiles for species A, B and C .

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