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Newton method for reactive solute transport with equilibrium sorption in porous media

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ABSTRACT

We present a mass conservative numerical scheme for reactive solute transport in porous media. The transport is modeled by a convection–diffusion–reaction equation, including equilibrium sorption. The scheme is based on the mixed finite element method (MFEM), more precisely the lowest-order Raviart–Thomas elements and one-step Euler implicit. The underlying fluid flow is described by the Richards equation, a possibly degenerate parabolic equation, which is also discretized by MFEM. This work is a continuation of Radu et al. (2008) and Radu et al. (2009) [1,2] where the algorithmic aspects of the scheme and the analysis of the discretization method are presented, respectively. Here we consider the Newton method for solving the fully discrete nonlinear systems arising on each time step after discretization. The convergence of the scheme is analyzed. In the case when the solute undergoes equilibrium sorption (of Freundlich type), the problem becomes degenerate and a regularization step is necessary. We derive sufficient conditions for the quadratic convergence of the Newton scheme.

1. Introduction

The extensive use of chemical substances in industry in the last 100 years enhanced also considerably the number of possible contaminated sites. There are hundreds of thousands of such places only in the developed countries. Due to this an active remediation is practically impossible. To decide how dangerous such a site is or can become is a very difficult task. A reliable and efficient simulation tool for contaminant transport in saturated/unsaturated soil is needed. This includes a comprehensive mathematical model, mass conservative discretization tools, robust and fast convergent methods for solving the nonlinear discrete problems and finally efficient linear solvers.

The diffusive–advective–reactive transport with a delay caused by the sorption on the soil skeleton of a one-component solute can be mathematically modeled by the equation

$$\partial_t(\Theta(\psi)c) + \rho_b\partial_t\phi(c) - \nabla\cdot(D\nabla c - \mathbf{Q}c) = \Theta(\psi)r(c) \quad \text{in } J\times\Omega, \tag{1}$$

with $c(t, \mathbf{x})$ denoting the concentration of the solute, D the diffusion–dispersion coefficient, ρ_b the soil density, $\phi(\cdot)$ a sorption isotherm, $\psi(t, \mathbf{x})$ the pressure, $\Theta(\cdot)$ the water content, $\mathbf{Q}(t, \mathbf{x})$ the water flux and $r(\cdot)$ a reaction term. In this paper we assume D to be a constant. For the ease of presentation we take $D = 1$. Further, $J = (0, T]$ $(0 < T < \infty)$ is the time interval, whereas $\Omega \subset \mathbb{R}^d (d \geq 1)$ is the computational domain having a Lipschitz continuous boundary Γ. Initial $c(t = 0) = c_I$ and homogeneous Dirichlet boundary conditions complete the model. We considered for the ease of

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presentation the case of one-component transport, but the present results can be extended to the case of a multi-component reactive transport, as long as the reactive term $r(\cdot)$ remains Lipschitz continuous. The same holds for more realistic boundary conditions (but regular enough) and a strictly positive non-constant diffusion coefficient.

For adsorption we consider two situations: Lipschitz continuous isotherms, as well as the commonly used Freundlich type isotherm

$$\phi(c) = c^{\alpha}, \quad \text{with } \alpha \in (0, 1]. \tag{2}$$

In the last case the derivative is singular at $c = 0$, so ϕ is not Lipschitz. To apply the Newton method for Freundlich type isotherms we employ a regularization step.

The water flux $\mathbf{Q}$ appearing in (1), as well as the water content $\Theta(\psi)$, are obtained by solving the mass balance equation for water, which is assumed incompressible

$$\partial_t \Theta(\psi) + \nabla \cdot \mathbf{Q} = 0 \tag{3}$$

and the Darcy's law

$$\mathbf{Q} = -K(\Theta(\psi))\nabla(\psi + z), \tag{4}$$

together with initial $\Psi(t = 0) = \Psi_I$ and homogeneous Dirichlet boundary conditions. Combining the Eqs. (3) and (4) one obtains the Richards equation, which is a typical mathematical model for water flow through saturated/unsaturated soil. For the coefficient functions $\Theta(\cdot)$ and $K(\cdot)$ functional dependencies of the pressure are assumed, so that the unknowns in (3)–(4) are reduced to two.

In [1,2] we have proposed and analyzed a mass conservative scheme for the Eq. (1) based on the MFEM for the spatial discretization and Euler implicit (EI) for the discretization in time. We have shown (see [2]) that the difference between the solution of the nonlinear fully discrete problems and the exact solution (which is called in the following the discretization error) vanishes as the time step and the mesh diameter are approaching zero. The order of convergence naturally depends on the accuracy of the scheme for the water flow. However, at each time step one has to solve the fully discrete nonlinear problems resulting after the mixed finite element (MFE) discretization, this being a challenging problem in itself. The objective of this work is to analyze the applicability of the Newton method for solving these nonlinear systems. Sufficient explicit conditions for the quadratic convergence of the method are derived.

Most of the papers dealing with numerical schemes for transport equations cope to estimate the discretization error and assume that the fully discrete nonlinear problems are solved exactly. We mention [3] for a conformal FEM discretization and [4] for finite volume schemes. Furthermore, a characteristic mixed method is studied in [5], upwind MFEM is considered in [6], whereas combined finite volume mixed hybrid finite elements are employed in [7,8]; see also [9] for a review. In [1,2] we presented and analyzed an EI-MFE scheme that is based on the lowest-order Raviart–Thomas (RT_0) elements for the Eq. (1). The resulting fully discrete nonlinear problems are commonly solved by different methods: the Newton scheme (which is locally quadratic convergent), some robust first-order linearization schemes (see [10–12]), or the Jäger–Kačur scheme [13]. The convergence of the Newton method applied to the system provided by a MFE discretization of an elliptic problem is studied in [14]. Concerning the systems provided by the MFE discretization of degenerate parabolic equations we mention [10] for a robust linear scheme and [15] for the Newton method. There, the fast diffusion case is considered, whereas here we have slow diffusion. Furthermore, the nonlinear term in the cited papers depends only on the solution and not on time, whereas the convection vector is constant and there is no reaction term.

The paper is structured as follows. The next section provides the continuous problem, the assumptions and presents the discretization scheme. In the first part of Section 3 we define the Newton scheme and prove its convergence. This is done for the diffusive–convective–reactive solute transport without sorption. We also explain how to include equilibrium, Lipschitz continuous sorption isotherms. Next we consider equilibrium non-Lipschitz sorption and perform a regularization step. We show the consistence of the regularization scheme and present the Newton scheme for this case, proving its convergence. In Section 5 we give some concluding remarks.

2. Euler implicit mixed finite element discretization

Throughout this paper we use common notations in the functional analysis. By $\langle \cdot, \cdot \rangle$ we mean the inner product on $L^2(\Omega)$. Further, $\| \cdot \|$ and $\| \cdot \|_{L^4(\Omega)}$ stand for the norms in $L^2(\Omega)$ and $L^4(\Omega)$, respectively. The functions in $H(\text{div}; \Omega)$ are vector valued, having a L^2 divergence. By C we mean a positive constant, not depending on the unknowns or the discretization parameters and by L_f the Lipschitz constant of a function $f(\cdot)$. Furthermore, $\mathcal{T}_h$ is a regular decomposition of $\Omega \subset \mathbb{R}^d$ into closed d-simplices; h stands for the mesh diameter. Here we assume $\overline{\Omega} = \cup_{T \in \mathcal{T}_h} T$, hence Ω is polygonal. Correspondingly, we define the discrete subspaces $W_h \subset L^2(\Omega)$ and $V_h \subset H(\text{div}; \Omega)$:

$$\begin{aligned} W_h &:= \{p \in L^2(\Omega) |\ p \text{ is constant on each element } T \in \mathcal{T}_h\}, \\ V_h &:= \{\mathbf{q} \in H(\text{div}; \Omega) | \mathbf{q}_{|T} = \mathbf{a} + b\mathbf{x} \text{ for all } T \in \mathcal{T}_h\}. \end{aligned} \tag{5}$$

In other words, W_h denotes the space of piecewise constant functions, while V_h is the RT_0 space (see [16]). For the discretization in time we let $N \in \mathbb{N}$ be strictly positive, and define the time step $\tau = T/N$, as well as $t_n = n\tau (n \in \{1, 2, \ldots, N\})$.

The continuous mixed variational formulation of (1) is:

Problem 2.1 (*The Continuous Problem*)**.** Find $c \in L^2(J; L^2(\Omega))$ and $\mathbf{q} \in L^2(J; H(\mathrm{div}; \Omega))$ such that for almost all $t \in J$ there holds for all $w \in L^2(\Omega)$

$$\langle \Theta(\psi(t))c(t) - \Theta(\psi_I)c_I, w\rangle + \rho_b\langle \phi(c(t)) - \phi(c_I), w\rangle + \left\langle \nabla \cdot \int_0^t \mathbf{q}\,\mathrm{d}s, w \right\rangle = \left\langle \int_0^t \Theta(\psi) r(c)\,\mathrm{d}s, w \right\rangle \tag{6}$$

and for all $\mathbf{v} \in H(\mathrm{div}; \Omega)$

$$\langle \mathbf{q}, \mathbf{v}\rangle - \langle c, \nabla \cdot \mathbf{v}\rangle - \langle c\mathbf{Q}, \mathbf{v}\rangle = 0. \tag{7}$$

The scheme for solving the water flow (3)–(4) is based on MFEM and Euler implicit and reads

Problem 2.2. Let ψ_h^{n-1} be given. Find $(\psi_h^n, \mathbf{Q}_h^n) \in W_h \times V_h$ such that

$$\langle \Theta(\psi_h^n) - \Theta(\psi_h^{n-1}), w_h\rangle + \tau\langle \nabla \cdot \mathbf{Q}_h^n, w_h\rangle = 0, \quad \forall w_h \in W_h, \tag{8}$$

$$\langle K^{-1}(\Theta(\psi_h^n))\mathbf{Q}_h^n, \mathbf{v}_h\rangle - \langle \psi_h^n, \nabla \cdot \mathbf{v}_h\rangle + \langle \mathbf{e_z}, \mathbf{v}_h\rangle = 0, \quad \forall \mathbf{v}_h \in V_h, \tag{9}$$

where $\mathbf{e_z}$ denotes the constant gravitational vector.

Remark 2.1. We solve the fully discrete scheme for the flow Problem 2.2 by a robust linearization scheme, which is first-order convergent (see [10]) or by the Newton method (see [15] for the analysis). So, on each time step, we know numerically $(\psi_h^n, \mathbf{Q}_h^n) \in W_h \times V_h$.

We define now the fully discrete scheme for Problem 2.1 at level $n \in \{1, \dots, N\}$:

Problem 2.3 (*The Fully Discrete Problem*)**.** Let $\Theta(\psi_h^n), \Theta(\psi_h^{n-1}), c_h^{n-1} \in W_h$ and $\mathbf{Q}_h^n \in V_h$ be given. Find $(c_h^n, \mathbf{q}_h^n) \in W_h \times V_h$ such that for all $w_h \in W_h$ there holds

$$\langle \Theta(\psi_h^n)c_h^n - \Theta(\psi_h^{n-1})c_h^{n-1}, w_h\rangle + \rho_b\langle \phi(c_h^n) - \phi(c_h^{n-1}), w_h\rangle + \tau\langle \nabla \cdot \mathbf{q}_h^n, w_h\rangle = \tau\langle \Theta(\psi_h^n) r(c_h^n), w_h\rangle, \tag{10}$$

and for all $\mathbf{v}_h \in V_h$ we have

$$\langle \mathbf{q}_h^n, \mathbf{v}_h\rangle - \langle c_h^n, \nabla \cdot \mathbf{v}_h\rangle - \langle c_h^n\mathbf{Q}_h^n, \mathbf{v}_h\rangle = 0. \tag{11}$$

Throughout this paper we make use of the following assumptions:

(A1) The rate function $r : \mathbb{R} \to \mathbb{R}$ is differentiable with r' bounded and Lipschitz continuous. Furthermore, $r(c) = 0$ for all $c \le 0$.
(A2) $1 \ge \Theta_S \ge \Theta(x) \ge \Theta_R > 0,\ \forall x \in \mathbb{R}$.
(A3) The initial c_I is essentially bounded and positive; furthermore, $\Psi_I \in L^2(\Omega)$.
(A4) $\mathbf{Q} \in L^\infty(J \times \Omega) \cap L^2(J; H^1(\Omega))$ and $\mathbf{Q}_h^n \in L^\infty(\Omega)$ for all $n \in \{1, \dots, N\}$.
(A5) The sorption isotherm $\phi(\cdot)$ is nondecreasing, nonnegative and Hölder continuous with an exponent $\alpha \in (0, 1]$, i. e. $|\phi(a) - \phi(b)| \le C|a - b|^\alpha \quad \forall a, b \in \mathbb{R}$. Moreover, $\phi(c) = 0$ if $c \le 0$.

We refer to [2] for the validity of the assumptions above. Under these assumptions one can show existence and uniqueness for the solutions of Problems 2.1 and 2.3 (see again [2]). Moreover, the assumptions on the nonlinearities $r(\cdot)$ and $\phi(\cdot)$ in the negative subdomain guarantee that the solution c remains nonnegative. Since c models some concentration, extending the rates by 0 for negative (thus unrealistic) concentrations is physically reasonable. By using a suitable quadrature formula (see [17]) for computing the term $\langle \mathbf{q}_h^n, \mathbf{v}_h\rangle$ in (11) one can guarantee also a discrete maximum principle, so we can further assume that the discrete concentrations are also nonnegative. Furthermore, in [2] the following a priori error estimates are proven:

Theorem 2.1. *Let* $(c, \mathbf{q})$ *solve* Problem 2.1 *and* $(c_h^n, \mathbf{q}_h^n)$ *solve* Problem 2.3 *for* $n \in \{1, \dots, N\}$. *Assuming* (A1)–(A5), *there holds*

$$\sum_{n=1}^N \int_{t_{n-1}}^{t_n} \|c(t) - c_h^n\|^2\,\mathrm{d}t + \left\| \sum_{n=1}^N \int_{t_{n-1}}^{t_n} \mathbf{q} - \mathbf{q}_h^n\,\mathrm{d}t \right\|^2 \le C \left\{ \tau^{\frac{4\alpha}{1+\alpha}} + h^{1+\alpha} + \sum_{n=1}^N \frac{1}{\tau} \left\| \int_{t_{n-1}}^{t_n} \mathbf{Q} - \mathbf{Q}_h^n\,\mathrm{d}t \right\|^2 \right.$$
$$\left. + \ \tau\|\Theta(\psi(t_n)) - \Theta(\psi_h^n)\|^2 + \int_{t_{n-1}}^{t_n} \|\Theta(\psi) - \Theta(\psi_h^n)\|^2\,\mathrm{d}t \right\}. \tag{12}$$

The result in Theorem 2.1 can be further simplified if one knows the accuracy of the scheme for the water flow. For example in the case of strictly unsaturated flow and a Lipschitz continuous sorption (or without sorption) we get a $\tau^2 + h^2$ order of convergence [2].

In the next section we will use the following elementary lemma (see [18], p. 350):

Lemma 2.1. *Let $f : \mathbb{R} \to \mathbb{R}$ differentiable with $f'(\cdot)$ Lipschitz continuous. Then there holds*

$$|f(x) - f(y) - f'(y)(x-y)|^2 \le \frac{L_{f'}}{2}|x-y|^2, \quad \forall x, y \in \mathbb{R}. \tag{13}$$

3. Newton schemes

In this section we present a Newton scheme to solve the nonlinear problem (10)–(11). For an easier understanding of the proof ideas we first consider the case of reactive transport without sorption. The same ideas apply then for Lipschitz continuous sorption rates. Finally, for dealing with non-Lipschitz sorption rates we employ a regularization step. In this case we show the convergence of the solution of the regularized problem to the solution of (10)–(11) and continue by analyzing the Newton scheme.

Throughout this section we let $n \in \{1, \dots, N\}$ index the time step, while i is used to index the iteration. Accordingly, $\{c_h^n, \mathbf{q_h^n}\}$ denotes the solution pair at the nth time step, $\{c_h^{n,\mathrm{reg}}, \mathbf{q}_h^{n,\mathrm{reg}}\}$ the solution of the regularized problem. In the same spirit, for a given $n \ge 1$ and for a known (previously determined) c_h^{n-1}, as well as $\psi_h^n, \psi_h^{n-1}, \mathbf{Q}_h^n$, we let $\{c_h^{n,i}, \mathbf{q}_h^{n,i}\}$ stand for the solution pair at iteration $i \ge 1$. The iteration process starts with $c_h^{n,0} = c_h^{n-1}$ for a Lipschitz sorption or in the absence of such effects, respectively with $c_h^{n,0} = c_h^{n-1,\mathrm{reg}}$ for non-Lipschitz rates. In proving the convergence of the iteration in the case without sorption (or with an equilibrium, Lipschitz one) it is sufficient to show that

$$\|c_h^n - c_h^{n,i}\| + \|\mathbf{q}_h^n - \mathbf{q}_h^{n,i}\| \to 0$$

as $i \to \infty$. This will be achieved for a sufficiently small time step τ. In the case of a non-Lipschitz sorption, for the convergence of the scheme we have to show that simultaneously it holds

$$\begin{cases} \|c_h^n - c_h^{n,\mathrm{reg}}\| + \|\mathbf{q}_h^n - \mathbf{q}_h^{n,\mathrm{reg}}\| \overset{\epsilon\to 0}{\to} 0 \\ \|c_h^{n,\mathrm{reg}} - c_h^{n,i}\| + \|\mathbf{q}_h^{n,\mathrm{reg}} - \mathbf{q}_h^{n,i}\| \overset{i\to\infty}{\to} 0. \end{cases}$$

Sufficient conditions for this are derived in Section 3.2.

3.1. Reactive solute transport

In the absence of sorption the Newton scheme for Problem 2.3 reads:

Problem 3.1. Let $c_h^{n,i-1} \in W_h$ be given, $i \ge 1$. Find $(c_h^{n,i}, \mathbf{q}_h^{n,i}) \in W_h \times V_h$ such that for all $w_h \in W_h$ there holds

$$\langle \Theta(\psi_h^n)c_h^{n,i}, w_h\rangle + \tau\langle \nabla\cdot \mathbf{q}_h^{n,i}, w_h\rangle = \langle \Theta(\psi_h^{n-1})c_h^{n-1}, w_h\rangle + \tau\langle \Theta(\psi_h^n)r(c_h^{n,i-1}) + \Theta(\psi_h^n)r'(c_h^{n,i-1})(c_h^{n,i} - c_h^{n,i-1}), w_h\rangle \tag{14}$$

and for all $\mathbf{v}_h \in V_h$ we have

$$\langle \mathbf{q}_h^{n,i}, \mathbf{v}_h\rangle - \langle c_h^{n,i}, \nabla\cdot\mathbf{v}_h\rangle - \langle c_h^{n,i}\mathbf{Q}_h^n, \mathbf{v}_h\rangle = 0. \tag{15}$$

Theorem 3.1. *Assuming* (A1)–(A4) *there holds for all $i \ge 1$*

$$\|c_h^{n,i} - c_h^n\|^2 + \tau\|\mathbf{q}_h^{n,i} - \mathbf{q}_h^n\|^2 \le C\tau h^{-d}\|c_h^{n,i-1} - c_h^n\|^4. \tag{16}$$

Proof. Let $\mathrm{e}_c^i := c_h^n - c_h^{n,i}$ and $\mathbf{e}_\mathbf{q}^i := \mathbf{q}_h^n - \mathbf{q}_h^{n,i}$. Subtracting (14) and (15) from (10) (where $\phi \equiv 0$) and (11), respectively, taking $w_h = \mathrm{e}_c^i$ and $\mathbf{v}_h = \tau\mathbf{e}_\mathbf{q}^i$ and adding the results we obtain

$$\begin{aligned}\langle \Theta(\psi_h^n)\mathrm{e}_c^i, \mathrm{e}_c^i\rangle + \tau\|\mathbf{e}_\mathbf{q}^i\|^2 &= \tau\langle \mathrm{e}_c^i\mathbf{Q}_h^n, \mathbf{e}_\mathbf{q}^i\rangle + \tau\langle \Theta(\psi_h^n)r'(c_h^{n,i-1})\mathrm{e}_c^i, \mathrm{e}_c^i\rangle \\ &\quad + \tau\langle \Theta(\psi_h^n)(r(c_h^n) - r(c_h^{n,i-1}) - r'(c_h^{n,i-1})(c_h^n - c_h^{n,i-1})), \mathrm{e}_c^i\rangle.\end{aligned} \tag{17}$$

Using (A1), (A2), (A4), the Cauchy–Schwarz and the mean inequalities, Lemma 2.1 gives

$$\Theta_R\|\mathrm{e}_c^i\|^2 + \frac{\tau}{2}\|\mathbf{e}_\mathbf{q}^i\|^2 \le \tau\left(\frac{1}{2} + \Theta_S L_r + C\right)\|\mathrm{e}_c^i\|^2 + \tau\frac{L_{r'}^2}{8}\|\mathrm{e}_c^{i-1}\|_{L^4(\Omega)}^4. \tag{18}$$

With τ small enough, the inverse estimate $\|\mathrm{e}_c^{i-1}\|_{L^4(\Omega)} \le Ch^{-d/4}\|\mathrm{e}_c^{i-1}\|$ (see e.g [19]) gives (16). ■

A straightforward consequence of Theorem 3.1 is the following

Theorem 3.2. *Assuming* (A1)–(A4)*, the Newton scheme in* Problem 3.1 *converges quadratically if*

$$\tau\|c_h^n - c_h^{n-1}\|^2 < Ch^d. \tag{19}$$

Remark 3.1. One can easily show that $\|c_h^n - c_h^{n-1}\| \to 0$, when $\tau \to 0$, which ensures the quadratic convergence of the Newton method at least when $\tau = O(h^d)$. Proceeding as in [20,2], one can show that $\|c_h^n - c_h^{n-1}\|^2 \leq C\tau$. In this case the condition (19) is satisfied whenever $\tau = O(h^{d/2})$, which is not too restrictive.

Remark 3.2. The condition (19) also holds for the case of transport with equilibrium sorption, when the derivative of the sorption isotherm is Lipschitz continuous (e.g. linear or Langmuir). Due to the second term in (10), we add

$$\langle \phi(c_h^{n,i-1}) + \phi'(c_h^{n,i-1})(c_h^{n,i} - c_h^{n,i-1}), w_h\rangle - \langle \phi(c_h^{n-1}), w_h\rangle$$

in the left side of (14). For proving then (16) we repeat the steps above and subtract

$$\langle \phi(c_h^n) - \phi(c_h^{n,i-1}) - \phi'(c_h^{n,i-1})(c_h^{n,i} - c_h^{n,i-1}), e_c^i\rangle$$

from the right-hand side of (17). The convergence condition (19) becomes $(\tau + L_{\phi'}^2)\|c_h^n - c_h^{n-1}\|^2 < Ch^d$, which is satisfied if $\tau = O(h^d)$.

3.2. *Reactive solute transport with Freundlich type equilibrium sorption*

In this section we discuss the case of non-Lipschitz sorption. We restrict the discussion to the case of a Freundlich isotherm, which is commonly used in modeling: $\phi(x) = x^\alpha$, $\alpha \in (0, 1]$. The case $\alpha = 1$ can be included in the previous section, therefore we consider here $\alpha \in (0, 1)$. Clearly, ϕ is not Lipschitz, but Hölder continuous. We notice here that the above isotherm is defined only for nonnegative arguments. We extend it by 0 on for (non-physical) negative values of x.

3.2.1. *Regularization*

To solve the problem (10)–(11) by the Newton method we first regularize the sorption isotherm. Specifically, for a given $\epsilon > 0$ we define

$$\phi_\epsilon(x) = \begin{cases} \phi(x) & \text{if } x \notin [0, \epsilon], \\ \alpha\epsilon^{\alpha-1}x + (1-\alpha)\epsilon^\alpha & \text{if } x \in [0, \epsilon]. \end{cases} \tag{20}$$

Lemma 3.1. *The regularized sorption isotherm is nondecreasing. Further, $\phi_\epsilon(\cdot)$ and $\phi'_\epsilon(\cdot)$ are Lipschitz continuous on $[0, \infty)$ with the Lipschitz constants $L_{\phi_\epsilon} = \alpha\epsilon^{\alpha-1}$, respectively $L_{\phi'_\epsilon} = \alpha(1-\alpha)\epsilon^{\alpha-2}$. Finally, we have*

$$0 \leq \phi_\epsilon(x) - \phi(x) \leq (1-\alpha)\epsilon^\alpha \tag{21}$$

if $x \in (0, \epsilon)$, whereas $\phi(x) = \phi_\epsilon(x)$ whenever $x \notin (0, \epsilon)$.

Remark 3.3. The regularization above is just the simplest example of a regularization function which will have the properties required for the analysis below. However, a quadratic isotherm as

$$\phi_\epsilon(x) = \begin{cases} \phi(x) & \text{if } x \notin [0, \epsilon], \\ (\alpha-1)\epsilon^{\alpha-2}x^2 + (2-\alpha)\epsilon^{\alpha-1}x & \text{if } x \in [0, \epsilon]. \end{cases}$$

is more realistic from a physical point of view. Approximation properties similar to the above are also satisfied, and the proofs below can be adapted along the same lines.

We can now define the regularized problem:

Problem 3.2. Given $\Theta(\psi_h^n)$, $\Theta(\psi_h^{n-1})$, $c_h^{n-1,\text{reg}} \in W_h$ and $\mathbf{Q}_h^n \in V_h$, find $(c_h^{n,\text{reg}}, \mathbf{q}_h^{n,\text{reg}}) \in W_h \times V_h$ such that for all $w_h \in W_h$ there holds

$$\begin{aligned} &\langle \Theta(\psi_h^n)c_h^{n,\text{reg}} - \Theta(\psi_h^{n-1})c_h^{n-1,\text{reg}}, w_h\rangle + \rho_b\langle \phi_\epsilon(c_h^{n,\text{reg}}) - \phi_\epsilon(c_h^{n-1,\text{reg}}), w_h\rangle + \tau\langle \nabla\cdot\mathbf{q}_h^{n,\text{reg}}, w_h\rangle \\ &\quad = \tau\langle \Theta(\psi_h^n)r(c_h^{n,\text{reg}}), w_h\rangle, \end{aligned} \tag{22}$$

and for all $\mathbf{v}_h \in V_h$ we have

$$\langle \mathbf{q}_h^{n,\text{reg}}, \mathbf{v}_h\rangle - \langle c_h^{n,\text{reg}}, \nabla\cdot\mathbf{v}_h\rangle - \langle c_h^{n,\text{reg}}\mathbf{Q}_h^n, \mathbf{v}_h\rangle = 0. \tag{23}$$

We choose $c_h^{0,\text{reg}}$ such that there holds $\Theta(\psi_h^0)c_h^0 + \phi(c_h^0) = \Theta(\psi_h^0)c_h^{0,\text{reg}} + \phi_\epsilon(c_h^{0,\text{reg}})$.

Defining

$$\Omega_\epsilon^{n,h} := \{T \in \mathcal{T}_h \mid 0 < c_{h|T}^n < \epsilon\},$$

and denoting by $\sigma(\omega)$ the area of ω, we have

$$\sum_{n=1}^{N} \sigma(\Omega_\epsilon^{n,h}) \leq C\tau^p\epsilon^q h^l. \tag{24}$$

The inequality above holds obviously for $p = -1, q = 0, l = 0$. Such an inequality is assumed in [3] for the continuous case, where $p = 0, q = 1, l = 0$; furthermore, a similar situation is considered in [21] for phase transition problems. Here we consider a general context and derive the convergence condition in terms of p, q and l, where $p \le 0, q \ge 0$, and $l \ge 0$.

Theorem 3.3. *Assuming* (A1)–(A5), *if* $(c_h^n, \mathbf{q}_h^n)$ *and* $(c_h^{n,\text{reg}}, \mathbf{q}_h^{n,\text{reg}})$ *are the solutions of Problems* 2.3 *and* 3.2, *respectively, then there holds*

$$\sum_{n=1}^{N} \|c_h^n - c_h^{n,\text{reg}}\|^2 + \sum_{n=1}^{N} \langle \phi_\epsilon(c_h^n) - \phi_\epsilon(c_h^{n,\text{reg}}), c_h^n - c_h^{n,\text{reg}} \rangle + \tau \sum_{n=1}^{N} \|\mathbf{q}_h^n - \mathbf{q}_h^{n,\text{reg}}\|^2 \le C\epsilon^{1+\alpha}\tau^p \epsilon^q h^l. \tag{25}$$

Proof. With $n = 1, \dots, N$ we define the errors at $t = t_n$:

$$e_c^n := c_h^n - c_h^{n,\text{reg}} \quad \text{and} \quad \mathbf{e}_q^n := \mathbf{q}_h^n - \mathbf{q}_h^{n,\text{reg}}.$$

Subtracting (22) from (10) and summing up the result for $n = 1, \dots, K$ we obtain for all $w_h \in W_h$

$$\sum_{n=1}^{K} \langle \Theta(\psi_h^n) e_c^n, w_h \rangle + \rho_b \sum_{n=1}^{K} \langle \phi(c_h^n) - \phi_\epsilon(c_h^{n,\text{reg}}), w_h \rangle + \tau \left\langle \sum_{n=1}^{K} \nabla \cdot \mathbf{e}_q^n, w_h \right\rangle = \tau \sum_{n=1}^{K} \langle \Theta(\psi_h^n)(r(c_h^n) - r(c_h^{n,\text{reg}})), w_h \rangle. \tag{26}$$

Further, subtracting (23) from (11) we have for all $\mathbf{v}_h \in V_h$

$$\langle \mathbf{e}_q^K, \mathbf{v}_h \rangle - \langle e_c^K, \nabla \cdot \mathbf{v}_h \rangle - \langle e_c^K \mathbf{Q}_h^K, \mathbf{v}_h \rangle = 0. \tag{27}$$

Taking now $w_h = e_c^K$ and $\mathbf{v}_h = \tau \sum_{n=1}^{K} \mathbf{e}_q^n$ in (26) and (27) respectively, adding the results and summing up from $K = 1$ to N gives

$$\begin{aligned}
&\sum_{n=1}^{N} \langle \Theta(\psi_h^n) e_c^n, e_c^n \rangle + \rho_b \sum_{n=1}^{N} \langle \phi(c_h^n) - \phi_\epsilon(c_h^{n,\text{reg}}), e_c^n \rangle + \tau \sum_{n=1}^{N} \left\langle \mathbf{e}_q^n, \sum_{k=1}^{n} \mathbf{e}_q^k \right\rangle \\
&\quad = \sum_{n=1}^{N} \tau \sum_{k=1}^{n} \langle \Theta(\psi_h^k)(r(c_h^k) - r(c_h^{k,\text{reg}})), e_c^n \rangle + \sum_{n=1}^{N} \tau \left\langle e_c^n \mathbf{Q}_h^n, \sum_{k=1}^{n} \mathbf{e}_q^k \right\rangle.
\end{aligned} \tag{28}$$

Using (A1), (A2), (A4), the Cauchy–Schwarz inequality, as well as the identity

$$2 \sum_{n=1}^{N} \left\langle \mathbf{a}_n, \sum_{k=1}^{n} \mathbf{a}_k \right\rangle = \left\| \sum_{n=1}^{N} \mathbf{a}_n \right\|^2 + \sum_{n=1}^{N} \|\mathbf{a}_n\|^2 ,$$

valid for any set of d-dimensional real vectors $\mathbf{a}_k \in \mathbb{R}^d$ $(k \in \{1, \dots, N\}, d \ge 1)$ we obtain

$$\begin{aligned}
&\frac{\Theta_R}{4} \sum_{n=1}^{N} \|e_c^n\|^2 + 2\rho_b \sum_{n=1}^{N} \langle \phi_\epsilon(c_h^n) - \phi_\epsilon(c_h^{n,\text{reg}}), e_c^n \rangle + \tau \left\| \sum_{n=1}^{N} \mathbf{e}_q^n \right\|^2 + \tau \sum_{n=1}^{N} \|\mathbf{e}_q^n\|^2 \\
&\quad \le 2\rho_b \sum_{n=1}^{N} \langle \phi_\epsilon(c_h^n) - \phi(c_h^n), e_c^n \rangle + \sum_{n=1}^{N} \tau \sum_{k=1}^{n} \|e_c^k\|^2 + C \sum_{n=1}^{N} \tau^2 \left\| \sum_{k=1}^{n} \mathbf{e}_q^k \right\|^2 .
\end{aligned} \tag{29}$$

Recalling the positivity and boundedness of the concentrations and using (24), (21) we get

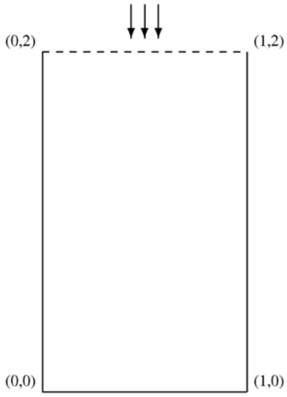
$$\begin{aligned}
\rho_b \sum_{n=1}^{N} \langle \phi_\epsilon(c_h^n) - \phi(c_h^n), c_h^n - c_h^{n,reg} \rangle &= \rho_b \sum_{n=1}^{N} \langle \phi_\epsilon(c_h^n) - \phi(c_h^n), c_h^n \rangle \\
&\quad - \rho_b \sum_{n=1}^{N} \langle \phi_\epsilon(c_h^n) - \phi(c_h^n), c_h^{n,\text{reg}} \rangle \le C\tau^p \epsilon^q h^l \epsilon^{1+\alpha}.
\end{aligned} \tag{30}$$

Using (29) and (30) and applying the Gronwall inequality immediately gives (25). ■

Remark 3.4. Alternatively, one may give up (24) and follow the steps in [2] to show that the error in (25) is at least of order $(\epsilon^{2\alpha} + \tau^{2\alpha} + h^{2\alpha})/\tau$. This result also does not assume the positivity of the concentrations, but is weaker than the one in Theorem 3.3.

3.2.2. Convergence of the Newton scheme

We can formulate now the Newton scheme for Problem 2.3



Initial conditions

$$\psi_I(x,y) = 1 - y\,, c_I(x,y) = 0.$$

Boundary conditions

$$\text{At } y = 2: \quad \psi = \begin{cases} 4t-1, & 0 < t < 0.5 \\ 1, & t \geq 0.5 \end{cases},$$

$$c = \begin{cases} 2t, & 0 < t < 0.5 \\ 1, & t \geq 0.5 \end{cases}.$$

$$\text{Elsewhere:} \quad \mathbf{Q}\cdot\mathbf{n} = 0,\ \mathbf{q}\cdot\mathbf{n} = 0.$$

Fig. 1. Domain and initial and boundary conditions for the test problem.

Problem 3.3. Let $c_h^{n,i-1}$ be given, $i \geq 1$. Find $(c_h^{n,i}, \mathbf{q}_h^{n,i}) \in W_h \times V_h$ such that for all $w_h \in W_h$ there holds

$$\langle \Theta(\psi_h^n)c_h^{n,i}, w_h\rangle + \rho_b\langle \phi_\epsilon(c_h^{n,i-1}) + \phi'_\epsilon(c_h^{n,i-1})(c_h^{n,i} - c_h^{n,i-1}) - \phi_\epsilon(c_h^{n-1,\text{reg}}), w_h\rangle + \tau\langle \nabla\cdot\mathbf{q}_h^{n,i}, w_h\rangle$$
$$= \langle \Theta(\psi_h^{n-1})c_h^{n-1,\text{reg}}, w_h\rangle + \tau\langle \Theta(\psi_h^n)r(c_h^{n,i-1}) + \Theta(\psi_h^n)r'(c_h^{n,i-1})(c_h^{n,i} - c_h^{n,i-1}), w_h\rangle, \tag{31}$$

and for all $\mathbf{v}_h \in V_h$ we have

$$\langle \mathbf{q}_h^{n,i}, \mathbf{v}_h\rangle - \langle c_h^{n,i}, \nabla\cdot\mathbf{v}_h\rangle - \langle c_h^{n,i}\mathbf{Q}_h^n, \mathbf{v}\rangle = 0. \tag{32}$$

Proceeding as in Theorem 3.1 one obtains

Theorem 3.4. *Assuming* (A1)–(A5), *if* $(c_h^{n,i}, \mathbf{q}_h^{n,i})$ *and* $(c_h^{n,\text{reg}}, \mathbf{q}_h^{n,\text{reg}})$ *are the solutions of Problems* 3.3 *and* 3.2, *respectively, then there holds*

$$\|c_h^{n,i} - c_h^{n,\text{reg}}\|^2 + \langle \phi'_\epsilon(c_h^{n,i-1})(c_h^{n,i} - c_h^{n,\text{reg}}), c_h^{n,i} - c_h^{n,\text{reg}}\rangle + \tau\|\mathbf{q}_h^{n,i} - \mathbf{q}_h^{n,\text{reg}}\|^2 \leq C(\tau + L^2_{\phi'_\epsilon})h^{-d}\|c_h^{n,i-1} - c_h^{n,\text{reg}}\|^4. \tag{33}$$

As a consequence of Theorem 3.4, we can derive now a sufficient condition for the quadratic convergence of the Newton scheme (31)–(32):

Theorem 3.5. *Assuming* (A1)–(A5), *the Newton scheme in Problem* 3.3 *converges quadratically if*

$$(\tau + L^2_{\phi'_\epsilon})\|c_h^{n-1,\text{reg}} - c_h^{n,\text{reg}}\|^2 < Ch^d. \tag{34}$$

Remark 3.5. If $\|c_h^{n-1,\text{reg}} - c_h^{n,\text{reg}}\|^2 \leq C\tau$ (see also Theorem 3.1), using Theorem 3.3 and (34), the condition for quadratic convergence of the Newton and of the regularization step becomes

$$C\tau^{1+p}\epsilon^{3\alpha+q-3}h^{l-d} < 1. \tag{35}$$

In other words, if either $1 + p > 0$, or $3\alpha + q > 3$, or $l > d$, one can correlate the discretization parameters to ensure the quadratic convergence of the Newton scheme, as well as the convergence of the regularization scheme.

Remark 3.6. An alternative to the Newton method is the linearization method in [10]. The method is robust, but converges only linearly. Further, it also requires a regularization step.

4. Numerical results

In this section we test the convergence of the Newton scheme on a realistic model. For test problems with explicit solutions we refer to [20,22,2], where the convergence of the MFE-scheme was investigated. We solve the flow equations (3)–(4) and the transport equation (1) in $J \times \Omega$ given by $J = (0, 1]$ and $\Omega = [0, 1] \times [0, 2]$, together with initial and boundary conditions as given in Fig. 1.

For the coefficient functions $\Theta(\cdot)$ and $K(\cdot)$ in (3)–(4) we assume a van Genuchten–Mulaem parametrization:

$$\Theta(\psi) = \Theta_R + (\Theta_S - \Theta_R)\Phi(\psi),$$
$$\Phi(\psi) = \frac{1}{(1 + (\alpha_{vG}\psi)^n)^m}, \quad m = 1 - \frac{1}{n},$$
$$K(\psi) = K_S\sqrt{\Phi(\psi)}(1 - (1 - \Phi(\psi)^{\frac{1}{m}})^m)^2,$$

Table 1
Convergence of the Newton method for the flow and transport equations with $\alpha = 0.25$, $\epsilon = 0.01$, $h = 0.05$.

	Flow $\tau = 0.02$	Transport $\tau = 0.02$	Flow $\tau = 0.0002$	Transport $\tau = 0.0002$
$T = 0.1$	1.3279066e−05 1.2966466e−07 9.1991567e −11	1.0510516e−01 9.4736256e−03 5.2516599e−04 2.0940888e−06 5.7297630e−08	4.8917332e−07 3.7437728e−09	6.6471896e−03 1.3066840e−04 5.7562211e−06 5.2655217e−08
$T = 0.5$	1.3090357e−05 1.2978282e−07 9.3385660e−11	9.4464958e−02 6.2066456e−03 2.8405743e−04 7.3637625e−07 7.5065276e−10	1.3091112e−07 9.7107833e −10	9.4651347e−03 1.4749051e−04 3.4909208e−06 2.4276207e−07 2.6345758e−08
$T = 1.0$	Steady state	1.7647909e−02 1.8168214e−03 3.7108137e−05 7.6127179e−08	Steady state	2.7065780e−03 1.1538238e−05 3.1800262e−08

Table 2
Convergence of the Newton method for the transport equation with $\alpha = 0.25$, $\epsilon = 0.005$, $h = 0.05$.

	$\tau = 0.0002$	$\tau = 0.0001$
$T = 0.1$	8.5241823e−03 3.8427892e−04 2.6027274e−05 1.5023433e−06 1.3971493e−07 1.4578884e−08	7.2895060e−03 5.1684957e−04 2.4287001e−05 1.5508735e−06 1.8033486e−08
$T = 0.5$	1.0623940e−02 3.2977608e−04 4.5016427e−06 8.8306969e−08	8.3193900e−03 3.7367723e−04 1.4082861e−05 3.0702861e−07 2.6335528e−08
$T = 1.0$	3.0027793e−03 1.7393033e−05 2.7778862e−07 2.8181040e−08	2.2740664e−03 1.0249958e−04 4.3239342e−07 2.5401588e−08

for negative values of ψ and $\Theta(\psi) = \Theta_S$, $K(\psi) = K_S$ otherwise. For the computations we use $\Theta_R = 0.1$, $\Theta_S = 0.5$, $n = 2.6$, $\alpha_{vG} = -0.0189$, $K_S = 2 \times 10^{-5}$. Further, for the transport problem we use a Freundlich type isotherm with two values for α: 0.25 and 0.5, $\rho_b = 1$, the diffusion coefficient is taken $D = 0.5$ and the rate function $r(c) = 0.2c$. All the data should be understood in some compatible units. We point out here that the chosen problem shows the main difficulties considered in this work: saturated/unsaturated flow and a Freundlich type isotherm in the transport equation. Consequently, the flow equation as well as the transport equation are degenerate, so solving this problem is a challenging task. Physically, the problem models the infiltration of contaminated water in soil. The numerical scheme was implemented in the software package *ug* [23].

We perform simulations with a constant mesh diameter $h = 0.05$. We vary the regularization number ϵ, the Hölder exponent α and, accordingly, the time step τ. We always give the residuum of the nonlinear problem, computed in the L^2 norm, at different times $T = 0.1$, $T = 0.5$ and $T = 1$. For times $T > 0.5$, the flow equation is stationary (steady state). In Table 1 we present the numerical results for $\alpha = 0.25$, $\epsilon = 0.01$, $\tau = 0.02$ and $\tau = 0.0002$. The convergence of the Newton scheme is as expected. Although is not the subject of this paper, Table 1 also presents the convergence of the Newton scheme for the flow equation (see [15] for details). We mention that a relative large time step is allowed. The results presented in Table 2 are obtained with a smaller regularization parameter $\epsilon = 0.005$ and time steps $\tau = 0.0002$ and $\tau = 0.0001$. Now a much smaller time step was required, due mainly to the very small Hölder exponent $\alpha = 0.25$. Nevertheless, the qualitative behaviour of the Newton scheme is similar. To have a comparison, we gave also the results for $\tau = 0.0002$ and $\epsilon = 0.01$ in Table 1. To enhance the role of the Hölder exponent α for such problems, we finally performed simulations with a moderate $\alpha = 0.5$. In this case, much higher time steps are allowed. In Table 3 are presented the results with $\epsilon = 0.01$ and $\tau = 0.05$, $\epsilon = 0.005$ and $\tau = 0.02$, and $\epsilon = 0.001$ and $\tau = 0.002$. Finally, we mention that for the all performed simulations we saw a mean of 3 Newton steps for the flow problem and 4 Newton steps for the transport one.

As follows from the numerical calculations, the numerical results enhance the theoretical findings. A time step that is small enough ensures the convergence of the scheme. If the Hölder exponents in the Freundlich isotherm, as well as the regularization parameters are very small, convergence is achieved only for similarly small time steps. Moderate Hölder

Table 3
Convergence of the Newton method for the transport equation with $\alpha = 0.5$, $h = 0.05$.

	$\epsilon = 0.01$ $\tau = 0.05$	$\epsilon = 0.005$ $\tau = 0.02$	$\epsilon = 0.001$ $\tau = 0.002$
$T = 0.1$	2.2703281e−01	9.9128224e−02	6.0362811e−03
	1.0751856e−02	4.1539348e−03	5.9018657e−05
	6.7193777e−04	1.3678799e−04	7.7609150e−07
	5.9708054e−06	2.6514627e−07	4.4115517e−08
	1.2973834e−09	7.8647372e−10	
$T = 0.5$	2.1440355e−01	9.4211876e−02	9.1613280e−03
	3.2307266e−03	1.4017927e−03	4.8510185e−05
	7.4759504e−05	1.1564611e−05	1.1043093e−07
	9.2559006e−08	5.1118141e−09	5.3040865e−09
$T = 1.0$	1.2966579e−02	1.2219626e−02	2.6568105e−03
	7.7630178e−04	3.6332678e−04	1.9664538e−05
	9.3841209e−06	1.5746036e−06	5.8744814e−07
	6.1387277e−09	1.2604563e−09	2.9589405e−08

exponents allow for relatively large time steps. Clearly, the time step ensuring the convergence of the Newton scheme depends nonlinearly on the Hölder exponent and on the regularization parameter.

5. Conclusions

We have considered a mass conservative scheme for diffusive–advective–reactive transport with equilibrium sorption of a solute in a porous medium. The scheme is based on EI and MFEM for the time and spatial discretization, respectively. The algorithmic aspects of the scheme were already presented in [1] and the discretization error was estimated in [2]. Here we focus on solving the nonlinear, fully discrete systems provided by the time and space discretization, and investigate the Newton method. If a Freundlich type sorption is considered, a regularization step is required. We derive sufficient conditions for the rigorous quadratic convergence of the Newton method. Here the flow problem and the transport problem are both solved on the same mesh and with the same time step. In general, one should also consider the conditions on the discretization parameters ensuring the convergence of the iterative method for the flow (see [10,15]), and not only for the transport problem. The paper is concluded by numerical examples sustaining the theoretical findings.

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