Numerical simulation of a phase-field model for reactive transport in porous media

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UHasselt Computational Mathematics Preprint
Nr. UP-20-02

Jan. 22, 2020

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Abstract We consider a Darcy-scale model for mineral precipitation and dissolution in a porous medium. This model is obtained by homogenization techniques starting at the scale of pores. The model is based on a phase-field approach to account for the evolution of the pore geometry and the outcome is a multi-scale strongly coupled non-linear system of equations. In this work we discuss a robust numerical scheme dealing with the scale separation in the model as well as the non-linear character of the equations. We combine mesh refinement with stable linearization techniques to illustrate the behaviour of the multi-scale iterative scheme.

1 Introduction

Soil salinization and harvesting of geothermal energy are examples from real life in which the pore-scale geometry can be affected by mineral precipitation and dissolution. While these processes are active at the pore scale (micro scale) and affect the pore-scale structures, their effects are reflected in the Darcy-scale (macro-scale) parameters such as the porosity and permeability.

Several approaches are available to account for the evolution of the micro-scale geometry. To locate the micro-scale interfaces a layer thickness function is proposed

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in [8, 15], whereas a level set approach is considered in [2, 13, 14]. In both approaches, upscaled models can be derived by solving micro-scale problems involving moving interfaces. This makes the development of numerical schemes a challenging task as it requires a very fine mesh reproducing the micro-scale details such as tracking the movement of the interfaces.

Here we model the evolution of the micro-scale boundary through a phase-field equation. We consider the phase-field model proposed in [3] in which the freely moving interfaces (separating the fluid and the mineral) are approximated by a thin diffuse interface layer described by a phase-field variable ϕ. This function is an approximation of the characteristic function that approaches 1 in the fluid phase and 0 in the mineral. Using the phase-field approach we avoid the difficulties related to discontinuities in the domain. This model is hence defined over the entire domain where the evolution of the phase field accounts the moving interface. Such models are derived from a minimization of the free energy in [5]. In [16] a phase-field model is extended to a precipitation and dissolution problems.

Since the main interest is the behaviour of the system at the macro scale, homogenization techniques are employed to derive upscaled models. The outcome is a coupled and non-linear system of equations addressing flow, chemistry and the phase-field evolution. Focusing on the two-scale model in [3], the main goal of this paper is to develop a robust numerical scheme accounting for both scale separation and the non-linearities in the model. This multi-scale iterative scheme borrows ideas from [4], where a stabilized iterative coupling scheme is introduced for a phase-field approach for fracture propagation.

This paper is organized as follows. In Section 2 the two-scale model is presented briefly, where the governing equations in two different scales are displayed and the strong coupling between the scales is discussed. In Section 3 we introduce the multi-scale iterative scheme in order to solve the upscaled model. There we give some details about handling the non-linearities and the convergence of the multi-scale iterative scheme. Finally, Section 4 provides a numerical example and the discussion of the results.

2 The two-scale phase-field model

We consider the two-scale phase-field model formulation of single-phase fully saturated flow with constant density and viscosity introduced in [3]. There, the details about the formal homogenization procedure can be found. Here we restrict to presenting the upscaled model only.

We consider a periodic porous medium $\Omega \subseteq \mathbb{R}^{\mathrm{d}}$. At each $\mathbf{x} \in \Omega$ we identify the variations at the micro-scale defining a fast variable. In other words, for each macro-scale point $\mathbf{x} \in \Omega$ we use one micro-scale cell $Y := [0,1]^{\mathrm{d}}$ to capture the fast changes encountered locally.

The unknowns $\mathbf{q}(\mathbf{x}, t)$, $p(\mathbf{x}, t)$ denote the macro-scale velocity and pressure in the fluid and $u(\mathbf{x}, t)$ is the upscaled solute concentration. We hence have the macro-scale flow and solute transport problems

$$
\begin{aligned}
(\mathcal{P}_1^M) \quad & \begin{cases} \nabla \cdot \overline{\mathbf{q}\phi} = 0, & \text{in } \Omega_{\mathrm{T}} := \Omega \times (0, \mathrm{T}], \\ \overline{\mathbf{q}\phi} = -\mathcal{K}\nabla p, & \text{in } \Omega_{\mathrm{T}}, \end{cases} \\
(\mathcal{P}_2^M) \quad & \begin{cases} \partial_t(\overline{\phi}(u - u^\star)) + \nabla \cdot (\overline{\mathbf{q}\phi} u) = D\nabla \cdot (\mathcal{A}\nabla u), & \text{in } \Omega_{\mathrm{T}}, \end{cases}
\end{aligned} \tag{1}
$$

completed by initial and boundary conditions. Here D is the solute diffusivity and $u^\star > u$ is the constant concentration of the mineral in the immobile domain. Moreover, the variable $\overline{\phi}$ defines the porosity and it is nothing but the average of the phase field ϕ over the micro-scale Y. The matrices $\mathcal{A}$ and $\mathcal{K}$ are the effective diffusion and permeability, respectively.

First, the phase field $\phi(\mathbf{x}, \mathbf{y}, t)$ is updated for all $\mathbf{x} \in \Omega$ and $t > 0$, by solving the following micro-scale problem

$$
(\mathcal{P}_\phi^\mu) \quad \begin{cases} \lambda^2 \partial_t \phi + \gamma P'(\phi) = \gamma\lambda^2 \nabla^2 \phi - 4\lambda\phi(1-\phi)\dfrac{1}{u^\star} f(u), \text{ in } Y, \\ \phi \text{ is } Y\text{-periodic.} \end{cases} \tag{2}
$$

The phase field ϕ has a smooth transition layer of width $\lambda > 0$ separating the phases. This equation is coupled with the macro scale through the reaction rate, which is chosen as $f(u) := \frac{u^2}{u_{\mathrm{eq}}^2} - 1$ with u_{eq} being a given equilibrium concentration. The term $P(\phi) = 8\phi^2(1-\phi)^2$ is the double-well potential, which ensures that the phase field approaches 0 and 1. The parameter γ is the diffusivity of the interface that separates the fluid and the mineral.

The macro-scale porosity in (1) is defined by the phase field $\overline{\phi}(\mathbf{x}, t) := \int_Y \phi(\mathbf{x}, \mathbf{y}, t) d\mathbf{y}$ and the elements of the effective matrices $\mathcal{A}(\mathbf{x}, t)$ and $\mathcal{K}(\mathbf{x}, t)$ are given by

$$
\mathcal{A}_{\mathtt{rs}}(\cdot, t) = \int_Y \phi \left(\delta_{\mathtt{rs}} + \partial_{\mathtt{r}} \omega^{\mathtt{s}}\right) d\mathbf{y} \quad \text{and} \quad \mathcal{K}_{\mathtt{rs}}(\cdot, t) = \int_Y \phi\, \mathbf{w}_{\mathtt{r}}^{\mathtt{s}} d\mathbf{y} \tag{3}
$$

for $\mathtt{r}, \mathtt{s} = 1, \ldots, \mathtt{d}$. The functions $\omega^{\mathtt{s}}$ and $\mathbf{w}^{\mathtt{s}} = [\mathbf{w}_1^{\mathtt{s}}, \ldots, \mathbf{w}_{\mathtt{d}}^{\mathtt{s}}]^t$ solve the following cell problems, defined for each $\mathbf{x} \in \Omega$

$$
\begin{aligned}
(\mathcal{P}_A^\mu) \quad & \begin{cases} \nabla \cdot (\phi(\nabla \omega^{\mathtt{s}} + \mathbf{e}_{\mathtt{s}})) = 0, & \text{in } Y, \\ \omega^{\mathtt{s}} \text{ is } Y\text{-periodic} \quad \text{and} \quad \displaystyle\int_Y \omega^{\mathtt{s}} d\mathbf{y} = 0, \end{cases} \\
(\mathcal{P}_K^\mu) \quad & \begin{cases} (\nabla \Pi^{\mathtt{s}} + \mathbf{e}_{\mathtt{s}}) + \mu_f \nabla^2(\phi \mathbf{w}^{\mathtt{s}}) = \dfrac{g(\phi, \lambda)}{\phi + \delta} \mathbf{w}^{\mathtt{s}}, & \text{in } Y, \\ \nabla \cdot (\phi \mathbf{w}^{\mathtt{s}}) = 0, & \text{in } Y, \\ \Pi^{\mathtt{s}} \text{ is } Y\text{-periodic} \quad \text{and} \quad \displaystyle\int_Y \Pi^{\mathtt{s}} d\mathbf{y} = 0. \end{cases}
\end{aligned} \tag{4}
$$

Here $\delta > 0$ is a small regularization parameter. The function $g(\phi, \lambda)$ ensures that there is zero flow in the mineral phase. This function is such that $g(1, \lambda) = 0$ and $g(0, \lambda) > 0$ (see [6]).

3 The multi-scale iterative scheme

We propose an iteratively coupled scheme to simulate the multi-scale behaviour of the phase-field model presented in Section 2. In [4, 10] similar approaches can be found but we remark that in the present case the coupling of different scales is encountered.

We let $N \in \mathbb{N}$ be the number of time steps and $\Delta t = \mathrm{T}/N$ be the time step size. For $n \in 1, \ldots, N$ define $t^n = n\Delta t$ and denote the time discrete solutions by $\nu^n := \nu(\cdot, t^n)$ for $\nu \in \{\phi, \mathcal{A}, \mathcal{K}, p, \mathbf{q}, u\}$.

Applying the Euler implicit discretization, at each time step a fully coupled non-linear system of equations has to be solved. For each $n > 0$, the iterative algorithm defines a multi-scale sequence $\left\{\phi_j^n, \mathcal{A}_j^n, \mathcal{K}_j^n, p_j^n, \mathbf{q}_j^n, u_j^n\right\}$ with $j \geq 0$ being the iteration index. Naturally, the initial guess for ϕ_0^n and u_0^n are ϕ^{n-1} and u^{n-1}.

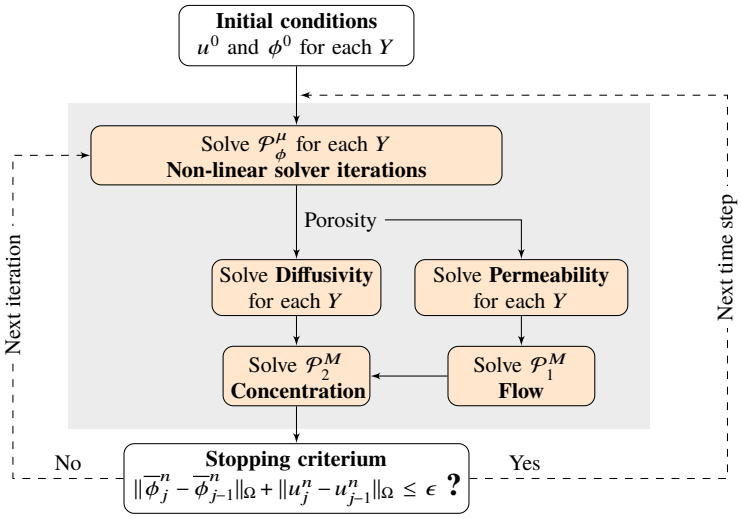

Fig. 1: Sketch of the iterative scheme.

The iterative scheme follows the idea in [4]. We let $L_\phi > 0$ be a stabilization parameter and for $j > 0$ with given u_{j-1}^n and ϕ_{j-1}^n, one performs the following steps:

Step 1. *For each* $\mathbf{x} \in \Omega$, *find* ϕ_j^n *such that*

$$\begin{aligned} \phi_j^n + \Delta t \gamma \nabla \cdot \phi_j^n - \frac{\Delta t}{\lambda^2} F(\phi_j^n, u_{j-1}^n) + L_\phi \left(\phi_j^n - \phi_{j-1}^n \right) = \phi^{n-1}, \quad \text{in } Y \\ \phi_j^n \text{ is } Y - \text{periodic.} \end{aligned} \tag{5}$$

where $F(\phi_j^n, u_{j-1}^n) := -\gamma P'(\phi_j^n) - 4\lambda \phi_j^n (1 - \phi_j^n) \frac{1}{u^\star} f(u_{j-1}^n)$.

Step 2. *Given* ϕ_j^n *find the effective matrices* $\mathcal{A}_j^n$ *and* $\mathcal{K}_j^n$ *in* (3) *by solving the cell problems* (4).

Step 3. *Given* $\mathcal{K}_j^n$ *and* $\mathcal{A}_j^n$ *find* p_j^n, $\mathbf{q}_j^n$ *and* u_j^n *by solving the system* (1).

In Figure 1 we sketch the multi-scale iterative scheme. Here it is important to remark that the behaviour of an efficient and robust non-linear solver for (5) affects directly the convergence of the complete multi-scale iterative scheme. To deal with the non-linearities we use a fixed-point iteration scheme, called L-scheme (see [9, 11]).

The convergence of the iterative scheme in Steps 1-3 is a non-trivial task as it involves multiple scales and couples non-linear and possible degenerate systems of equations. Preliminary results are obtained in a simplified setting. Specifically, we assume that the pore space is never clogged and that the mineral never disappears completely. In other words, there exists two constants $\overline{\phi}_m, \overline{\phi}_M \in (0,1)$ such that $0 < \overline{\phi}_m \leq \overline{\phi}(\mathbf{x}) \leq \overline{\phi}_M < 1$ for a.e $\mathbf{x} \in \Omega$. Moreover, the flow component is disregarded and the diffusion tensor is assumed not depending on the phase field. With $M_1 = \max_{\substack{\phi \in [0,1] \\ u>0}} \{|\partial_1 F(\phi, u)|\}$, $M_2 = \max_{\substack{\phi \in [0,1] \\ u>0}} \{|\partial_2 F(\phi, u)|\}$ and $\bar{u} = \max_{\substack{\mathbf{x} \in \Omega \\ n \in \mathbb{N}}} \{|u^\star - u^n(\mathbf{x})|\}$ one can prove the following.

Proposition 1 *Let* M_1, M_2, $\bar{u}$ *and* $\overline{\phi}_m$ *be as above. If the time step is small enough, namely*

$$\Delta t \leq 2\lambda^2 \min \left\{ \frac{1}{M_1 + M_2 + 0.5\bar{u}}, \frac{\overline{\phi}_m^2}{M_2} \right\}$$

the scheme in Steps 1-3 is convergent.

The proof uses contraction arguments, we omit the details here.

4 A numerical example

We consider a simplified 2D situation where the processes are expected to be uniform in the vertical direction. The macro-scale domain is $\Omega = [0,1]^2$, where a dissolution process is triggered by imposing a Dirichlet condition for the concentration on the right boundary of Ω. This configuration is displayed in Figure 2, while Table 1 shows the parameters used for the simulation. In the following, all the solutions are computed using the lowest order Raviart-Thomas elements.

Solute Diff.	$D = 1$
Mineral concent.	$u^\star = 1$
Equilibrium	$u_{\text{eq}} = 0.5$
Diffusivity	$\gamma = 0.01$
Transition zone	$\lambda = 0.08$
Initial porosity	$\overline{\phi}_0 = 0.5$
Max. porosity	$\overline{\phi}_M = 0.87$
Initial condition	$u_0 = 0.5$
Stabilization	$L_\phi = 1\text{E}{-4}$

Table 1: The parameters

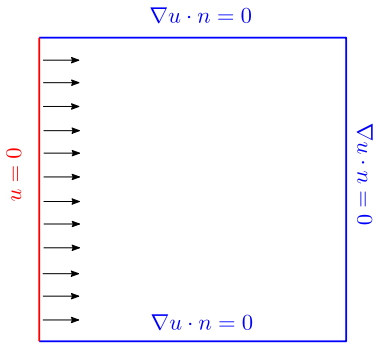

Fig. 2: The configuration of the macro-scale problem.

Figure 3 shows the evolution of the phase field corresponding to the macro scale location $(0.5, 0.5)$. At the micro scale we use a mesh refinement strategy to capture the movement of the phase-field transition zone.

At each time step we construct a micro-scale mesh with 800 elements. This mesh is refined in the first iteration of the scheme by following a prediction-correction strategy. We refer to [1, 7] for more details about handling similar meshes.

As mentioned before, we use an L-scheme dealing with the non-linearities at the micro scale. The non-linear term $F(\phi, u)$ needs to be split in a convex and concave part. Only the concave part is treated implicitly and the linearization parameter corresponds to the Lipschitz constant of F with respect to ϕ (which depends on the concentration u) at every multi-scale iteration.

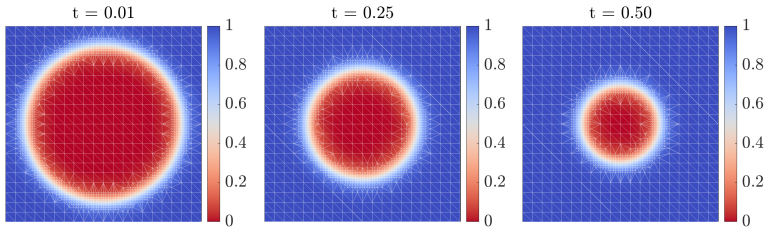

Fig. 3: The phase-field evolution at the macro-scale location $(0.5, 0.5)$. From left to right, the phase field at $t = 0.2$, 0.25 and 0.5.

The Darcy-scale solute concentration is displayed in Figure 4. Due to the chosen boundary and initial conditions, this solution does not depend on the vertical component and therefore the 1D projection in the horizontal direction is sufficient. The results for the porosity and the effective parameters are shown in Figures 4 and 5.

We highlight that even if we are not computing the flow in this case, the effective permeability can still be calculated. Where the concentration decreases, it induces a dissolution of the mineral, which then increases the diffusivity and the permeability until the system reaches the maximum porosity $\overline{\phi}_M$.

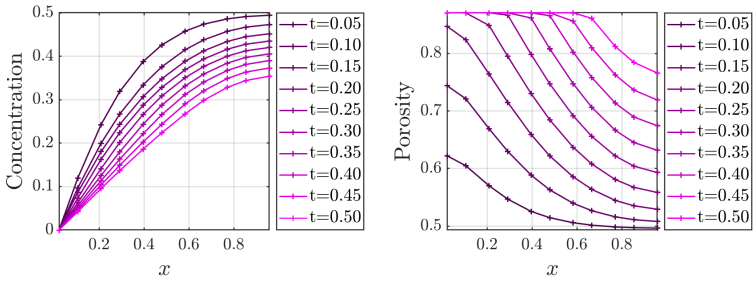

Fig. 4: The 1D projection of the concentration and the porosity at different times.

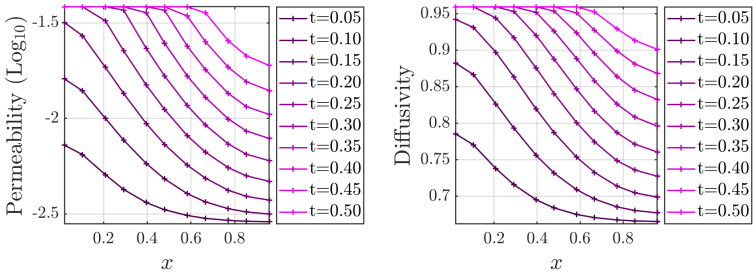

Fig. 5: The 1D projection of the effective parameters at different times.

Finally, in Figure 6 we show the convergence of the norm of the residual $\|\overline{\phi}_j^n - \overline{\phi}_{j-1}^n\|_\Omega + \|u_j^n - u_{j-1}^n\|_\Omega$ at different time steps. The non-linear solver at each micro-scale domain Y is stopped once the residual is below 1E−10.

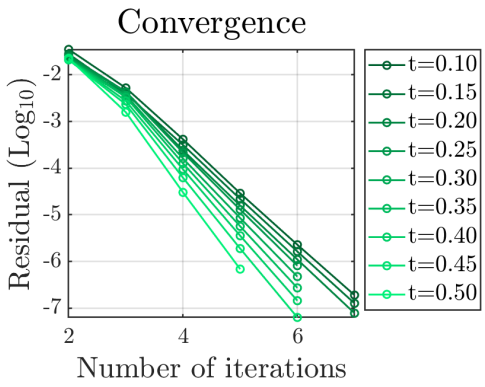

Fig. 6: The convergence of the residual in the multi-scale iterative scheme.

In this numerical example the averaged number of degrees of freedom is 7.623.300 per time step. At the macro scale we have 512 elements and for each element the porosity and the effective parameters must be updated at each iteration. Due to the

local mesh refinement the micro-scale degrees of freedom vary between 1.200 and 1.400. However, the micro-scale problems are solved in parallel and this can be improved by employing an adaptive strategy at the macro scale (see [12]).

We conclude that the multi-scale iterative scheme presented here is a valid approach to solve the two-scale phase-field model of precipitation and dissolution processes. This scheme can easily be parallelized and the resulting simulations show the influence of the micro-scale structural changes on the macro-scale parameters.

The next research steps are in the direction of proving the convergence of the full numerical scheme, including the error analysis of the micro-cell problems. Moreover, the study of the optimal choice of the stabilization parameter L_ϕ and the macro-scale adaptivity are important to enhance the efficiency of the scheme.

Acknowledgements The authors are supported by the Research Foundation - Flanders (FWO) through the Odysseus programme (Project G0G1316N). We thank Thomas Wick and Florin Adrian Radu who made valuable suggestions and contributed to the ideas behind this manuscript.

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