

# Density-driven convection with dissolution in porous media: A competition among density ratio, reaction rate and porosity evolution

Xuhui Meng, Huazhong University of Science and Technology, Wuhan 430074, China

Xiaofan Yang\*, Beijing Normal University, Beijing 100875, China

Zhaoli Guo\*, Huazhong University of Science and Technology, Wuhan 430074, China

**Key words:** dissolution, variable density flows, interfacial instability

## Introduction

Geological storage of the CO<sub>2</sub> in subsurface saline aquifers is a promising way to reduce the CO<sub>2</sub> emissions. To predict the transport of CO<sub>2</sub> in subsurface environment becomes critical yet difficult since the transport process (density-driven convection with dissolution) is highly heterogenous and complex. Cardoso and Andres [1] conducted both experiments and the linear stability analysis to investigate the effect of the heterogeneous chemical reaction on the transport of CO<sub>2</sub> in brine. Their results showed that the chemical reaction can suppress the density-driven fingering instability, suggesting that the chemical reaction in-between phases needs to be considered in predicting the transport of CO<sub>2</sub>. However, the variation of the porosity/permeability caused by the heterogeneous reaction was not taken into consideration. In the present study, pore-scale simulations using the lattice Boltzmann method [2], Darcy-scale linear stability [3] analysis and laboratory experiment are integrated to investigate the competition among density ratio, reaction rate and porosity/permeability evolution and their impacts on the transport processes.

## Methodology

**Problem setup** A 2D micromodel is used in the current study with regular pore arrangement (experiment ongoing). At the initial time, the upper part of the domain is filled with fluid A, which can react with the solid phase; while the bottom part of the domain is filled with fluid B. Fluid A is denser than fluid B that will be driven down by the gravity. Furthermore, the reaction between the fluids and the solids will induce the dissolution on the solid phase that changes the pore structure.

**Pore-scale simulation** The dimensionless governing equations for the mentioned process coupling the reactive flows and the porosity/permeability evolution can be expressed as follows

$$\nabla \cdot \mathbf{u} = 0, \quad (1)$$

$$\partial_t \mathbf{u} + \mathbf{u} \cdot \nabla \mathbf{u} = -\nabla p + \sqrt{\frac{Sc}{Ra}} \nabla^2 \mathbf{u} + \Delta C \mathbf{j}, \quad (2)$$

$$\partial_t C + \mathbf{u} \cdot \nabla C = \frac{1}{\sqrt{ScRa}} \nabla^2 C, \quad (3)$$

$$\frac{\partial C}{\partial \mathbf{n}} = Da(C - 1) \quad (4)$$

where  $\mathbf{u}$  is the velocity vector,  $p$  is the pressure,  $C$  is the solute concentration,  $\mathbf{n}$  is the normal, with the dimensionless numbers (Schmidt number  $Sc$ , Rayleigh number  $Ra$  and Damköhler number  $Da$ ). The above governing equations are solved by a recently developed multiple-relaxation-time lattice Boltzmann equation (MRT-LBE) model with a localized boundary scheme for the reactions, which is proved to have good numerical stability [2].

**Linear stability analysis** For linear stability analysis, Darcy-scale governing equations for flow and reactive transport are summarized as follows:

$$\partial_t \phi + \nabla \cdot \mathbf{u} = 0, \quad (5)$$

$$\mathbf{u} = -\frac{K(\phi)}{\mu} (\nabla P - \mathbf{g} \beta \Delta C), \quad (6)$$

$$\partial_t C + \mathbf{u} \cdot \nabla C = \nabla \cdot (D \nabla C) - k_r s C, \quad (7)$$

$$c_{sol} \partial_t \phi = \nu_{sol} k_r s C, \quad (8)$$

where  $\phi$  is the porosity of the porous media,  $\mathbf{u} = (u, w)$  is the velocity,  $K$  denotes the permeability,  $\mu$  is the fluid dynamic viscosity,  $P$  is the pressure at the pore space,  $\mathbf{g}$  is the gravitational acceleration,  $C$  is the solute

concentration,  $D$  represents the effective diffusion coefficient,  $k_r$  is the effective reaction rate,  $s$  is the surface area,  $c_{sol}$  is the concentration of the solid phase that can be dissolved, and  $\nu_{sol}$  is the stoichiometric number. Once the above governing equations are written in a dimensionless form, they can be solved numerically using a spectral method.

## Results

**Effect of chemical reaction on the interfacial instability** Figure 1 shows the instantaneous concentration distribution from pore-scale simulations in three different scenarios: (a) no reaction; (b) with reaction but without consideration of dissolution-induced structure change; (c) with reaction and the pore structure change. It is obvious to see that the interfacial instability is triggered by the density ratio between two miscible fluids, suppressed by the heterogeneous surface reaction between the fluid and solid phases, and influenced by the porosity (permeability) evolution due to the dissolution. Specifically the dissolution-induced pore morphology change has great impact on the interfacial instability thus plays an important role in the transport processes.

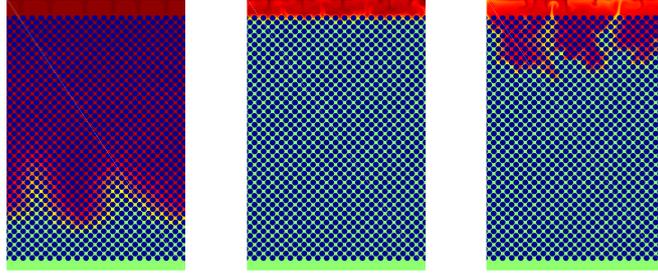


Figure 1: Instantaneous concentration plots for three different scenarios: (a) no reaction  $Da = \gamma = 0$ , (b) with reaction but without consideration of dissolution-induced structure change  $Da = 1, \gamma = 0$  and (c) with reaction and the pore structure change  $Da = 1, \gamma = 0.1$ .  $\gamma$  is the acid capacity.  $Ra = 10^4$  for all the cases.

**Theoretical analysis** To analyze the interfacial instabilities, linear stability analysis is conducted. We first use density-driven flows with the first-order reaction (without chemical dissolution) to validate our code by comparing with literature data reported in [3] (Figure 2a). Then we study the effect of the chemical dissolution on the interfacial instability (Figure 2b).

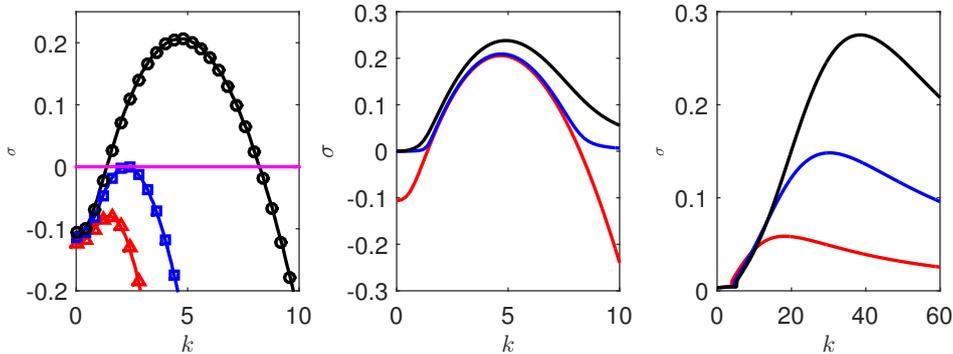


Figure 2: Linear stability analysis: (a) comparison between current results (line) and literature data [3] (dots)  $Ra = 25$  (red), 41.6 (blue) and 100 (black); (b)  $\gamma = 0$  (red), 0.01 (blue) and 0.1 (black); (c)  $Ra = 50$  (red), 150 (blue) and 250 (black).

## References

- [1] S.S.S. Cardoso and J.T.H. Andres. Geochemistry of silicate-rich rocks can curtail spreading of carbon dioxide in subsurface aquifers. *Nature Comm.*, **5**, 5743–5748, (2014).
- [2] X.H. Meng, Z.L. Guo. Multiple-relaxation-time lattice Boltzmann model for incompressible miscible flow with large viscosity ratio and high Péclet number. *Phys. Rev. E*, **92**, 043305, (2015).
- [3] J.C. Ward, K.A. Cliffe, O.E. Hensen. Dissolution-driven porous-medium convection in the presence of chemical reaction. *J. Fluid Mech.*, **747**, 316–349, (2014).