



# Pore-scale study of dissolution-induced changes in permeability and porosity of porous media



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## SUMMARY

We apply a reactive transport lattice Boltzmann model developed in previous studies to study the dissolution-induced changes in permeability and porosity of two porous media at the pore scale. The permeability–porosity relationship is explored for a wide range of Peclet and Damkohler numbers. It is found that this relationship depends not only on different dissolution regimes characterized by  $Pe$  and  $Da$ , but also on the specific porous medium structure. The permeability–porosity relationship for the more geometrically complex porous medium shows much more complexity than that for the simple fractured medium. While a very small  $Da$  sets an upper bound for the permeability–porosity relationship for the simple medium, a combination of a high  $Da$  and  $Pe$  results in wormholing, and the fastest permeability increase for the complex medium. At a moderate  $Pe$  but large  $Da$ , a transition from transport-limited dissolution regime to wormholing is also observed for the complex medium.

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## 1. Introduction

Hydrologic properties that govern fluid flow and reactive transport in the subsurface include porosity, permeability, pore/grain size distribution, tortuosity, relative permeability, fluid–fluid and fluid–solid interfacial areas. In many geologic processes, these properties, as well as constitutive relationships among them, may change due to various processes including dissolution/precipitation of minerals, colloid release and capture, ion exchange, and clay swelling (Saripalli et al., 2001). Because these hydrologic properties and constitutive relationships are required in numerical models based on continuum approaches, a better understanding and accurate description of these changes is an issue critical for reducing uncertainties in large-scale numerical modeling. For problems such as geological carbon sequestration, anticipating the modification of reservoir properties is also critical for designing the injection operations and assessing storage performance and safety.

Processes that cause the changes of hydrologic properties often occur at the pore scale. However, hydrologic properties, such as

porosity and permeability, are defined at the continuum scale. This mismatch in length scales makes it difficult to correctly understand and describe the changes of hydrologic properties at the continuum scale. Pore-scale studies are necessary to better characterize these changes, because detailed information from pore-scale modeling can be used to derive hydrologic properties and constitutive relationships among them; and time-dependent information can be used to characterize their changes with time.

In the past two decades, reactive transport processes at the pore scale have been studied extensively using both numerical and experimental approaches under a wide range of simplified conditions (Bekri et al., 1995, 1997; Daccord, 1987; Dijk and Berkowitz, 1998; Fredd and Fogler, 1998; Hoefner and Fogler, 1988; Kang et al., 2003, 2002b, 2004; Kelemen et al., 1995; Reeves and Rothman, 2012; Sallès et al., 1993; Verberg and Ladd, 2002; Verhaeghe et al., 2005, 2006; Walsh and Saar, 2010; Wells et al., 1991; Willingham et al., 2008; Yu and Ladd, 2010). More recently, much greater complexity involving strongly coupled flow, transport, and precipitation/dissolution reactions in realistic geochemical systems has been explicitly included in numerical models (Algive et al., 2010; Chen et al., 2013; Huber et al., 2014; Kang et al., 2010, 2006, 2007; Li et al., 2006; Molins et al., 2012; Parmigiani et al., 2011; Tartakovsky et al., 2007, 2008; Yoon et al., 2012).

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However, pore-scale studies dedicated to the determination of hydrologic property changes are few (Algive et al., 2010; Egermann et al., 2010; Gouze and Luquot, 2011; Luquot and Gouze, 2009; Sadhukhan et al., 2012). Most of the modeling studies were based on the pore network model, and the study by Sadhukhan et al. (2012) was based on solving the Navier Stokes's equation using a finite difference method.

In this paper, we present a pore-scale study of dissolution-induced changes in permeability and porosity – two basic hydrologic properties of porous media, using the lattice Boltzmann (LB) method. We focus on the permeability–porosity relationship for different dissolution regimes, as characterized by the Peclet ( $Pe$ ) and Damkohler ( $Da$ ) numbers. Although some initial work on this topic has been done for some simple porous medium structures, here we aim to provide a more comprehensive study, emphasizing the effect of a more complex pore structure and investigating the permeability–porosity relationship on a more quantitative basis. For this purpose, here we only consider the dissolution of a single mineral into a solution according to linear reaction kinetics, although the LB model and computer code used can handle multi-component reactive transport in porous media (Kang et al., 2010, 2006, 2007).

## 2. Mathematical model

### 2.1. Governing equations

For isothermal incompressible fluid flow through porous media, the pore-scale continuity and momentum equations can be written as (Landau and Lifshitz, 1959)

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

$$\rho \frac{\partial \mathbf{u}}{\partial t} + \rho \mathbf{u} \cdot \nabla \mathbf{u} = -\nabla p + \rho \nu \nabla^2 \mathbf{u}, \quad (2)$$

where  $\rho$  represents the density of the fluid,  $t$  the time,  $\mathbf{u}$  the velocity vector,  $p$  the pressure, and  $\nu$  the kinematic viscosity.

If the solute concentration is sufficiently low so that its effect on the density, viscosity, and velocity of the solution is negligible, then the transport of the solute species can be described using the following pore-scale advection–diffusion equation (Dawson et al., 1993):

$$\frac{\partial C}{\partial t} + (\mathbf{u} \cdot \nabla)C = \nabla \cdot (D \nabla C). \quad (3)$$

In the above equation,  $C$  is the solute concentration and  $D$  is the molecular diffusion coefficient which is considered isotropic in this study.

### 2.2. Dissolution reaction at the pore-mineral interface

At the pore-solid interface, the no-slip boundary condition is enforced for fluid flow, and dissolution of a single mineral into the solution according to linear reaction kinetics is considered:

$$D \frac{\partial C}{\partial n} = \hat{T} = k_r(C - C_s), \quad (4)$$

where  $n$  denotes the unit normal perpendicular to the fluid–mineral interface pointing toward the fluid phase;  $\hat{T}$  denotes the reaction flux of the mineral at its surface, taken as positive for precipitation and negative for dissolution;  $k_r$  is the reaction rate coefficient; and  $C_s$  is saturated concentration. The system is in chemical equilibrium when solute concentration  $C$  is equal to  $C_s$ . When  $C$  is lower than  $C_s$ , dissolution will take place.

In this study, the solid movement with fluid flow is not considered. Therefore, the mineral volume fraction ( $\varphi$ ) of the stationary solid satisfies the following equation:

$$\frac{\partial \varphi}{\partial t} = \bar{V} a \hat{\Gamma}, \quad (5)$$

where  $\bar{V}$  and  $a$  are molar volume and specific surface area of the solid phase.

## 3. Numerical method

### 3.1. Lattice Boltzmann method for coupled flow, solute transport, and dissolution

Rather than solve the pore-scale governing Eqs. 1–3 directly using a conventional numerical method (e.g., finite difference, finite volume, finite element), the LB method is used to solve these equations. Due to its kinetic nature, the LBM has the advantage of treating geometrically complex boundaries of porous media and intra-pore transport and interfacial processes. In the LBM framework, flow of a single phase fluid is described by the following evolution equation:

$$f_i(\mathbf{x} + \mathbf{c}_i \Delta t, t + \Delta t) - f_i(\mathbf{x}, t) = -\frac{1}{\tau_v} (f_i(\mathbf{x}, t) - f_i^{eq}(\mathbf{x}, t)), \quad (6)$$

where  $f_i$  is the density distribution function along the  $i$  direction,  $f_i^{eq}$  is the corresponding equilibrium distribution function,  $\Delta t$  is the time increment,  $\mathbf{c}_i$  is the discrete lattice velocity, and  $\tau_v$  is the collision time related to the kinematic viscosity. With an appropriate choice of  $\mathbf{c}_i$  and  $f_i^{eq}$ , Eq. (6) can be proved to recover Eqs. (1) and (2), with fluid density, velocity, and viscosity given by  $\rho = \sum_i f_i$ ,  $\rho \mathbf{u} = \sum_i f_i \mathbf{c}_i$ , and  $\nu = c_s^2 (\tau_v - 0.5) \Delta t$ , respectively.

Similarly, the transport of a solute in the fluid can be described by the following equation:

$$g_i(\mathbf{x} + \mathbf{c}_i \Delta t, t + \Delta t) - g_i(\mathbf{x}, t) = -\frac{1}{\tau_s} (g_i(\mathbf{x}, t) - g_i^{eq}(\mathbf{x}, t)). \quad (7)$$

With an appropriate choice of  $\mathbf{c}_i$  and  $g_i^{eq}$ , Eq. (7) can be proved to recover Eq. (3), with solute concentration and diffusion coefficient given by  $C = \sum_i g_i$  and  $D = \frac{1}{2} (1 - J_0) (\tau_s - 0.5)$ , respectively, where  $J_0$  is a constant between 0 and 1 (Chen et al., 2012).

A boundary condition for unknown distribution functions at the reactive fluid–solid interface was developed in Kang et al. (2007) to implement the dissolution reaction described by Eq. (4). This boundary condition has been shown to guarantee mass conservation during dissolution, and is used in this study. The detail will not be repeated here for brevity.

### 3.2. Update of solid phase

In the present study, mineral volume fraction  $\varphi$  is updated explicitly at each time step according to

$$\varphi(t + \delta t) = \varphi(t) + \bar{V} a \hat{\Gamma} \delta t, \quad (8)$$

which is simply the forward difference of Eq. (5). Initially  $\varphi$  is set to 1 for all solid nodes (including solid–fluid interface nodes) and is updated according to Eq. (8) for interface nodes. When its value reaches zero for an interface node, that node is then converted to a fluid node, and its neighboring interior solid nodes may become interface nodes, depending on the detailed local geometric feature. Accordingly, the flow field will also be updated to account for the change of porous medium structure.

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