

COUPLING LATTICE BOLTZMANN AND FINITE VOLUME METHODS FOR SIMULATION OF REACTIVE TRANSPORT

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ABSTRACT

A numerical model is developed to study reactive transport at pore scale. It couples lattice Boltzmann and finite volume methods to compute fluid flow and reaction processes. The model is validated by comparing numerically simulated reactive flow against the published results of dynamic imaging of carbonate dissolution. The porosity profiles and permeability-porosity relationship are compared with the experimental results. The variation of the porous structure is quantitatively analysed and compared with those obtained from dynamic imaging experiments. The findings show that the numerical framework provides robust and accurate prediction of porous media alteration due to flow of reactive fluids and can be used as a predictive tool for resolving reaction mechanisms in porous media with applications in reservoir engineering, carbon dioxide sequestration, hydrological and environmental studies.

INTRODUCTION

Reactive transport in porous media is of great importance in a wide range of science and engineering applications including reservoir engineering, carbon dioxide sequestration, hydrogeological and environmental studies. Chemical reactions at the pore scale can dramatically alter the pore structure and change the petrophysical properties of reservoir rocks [1]. Pore scale modelling methods for reactive transport studies include network modelling and direct modelling [2]. Pore network modelling has been used to study the permeability and porosity variation due to reaction [3,4], but it cannot accurately predict the alteration of local microstructure in very heterogeneous porous media [5]. Direct modelling addresses this issue by simulating fluid flow directly in voxels of micro-computed tomography (micro-CT) images. As the most popular method of direct simulation, the lattice Boltzmann (LB) method has been widely used to investigate the dissolution and precipitation in two-dimensional porous media [6,7]. Apart from the LB approach in direct simulation, finite difference, volume and element methods [8-11] and

particle-tracking methods [12,13] have been applied to simulate flow and predict reaction rates in porous media. However, numerical model with the lattice Boltzmann method have mainly focused on 2D geometries. Although 3D investigations of reactive flow have been performed by other direct modelling methods [13], the maximum size of the micro-CT image is limited due to computational limitations.

In this paper, an efficient pore scale model is developed for simulating the reactive transport directly on 3D micro-CT images. We simulate reactive transport directly on the micro-CT image and compare our results with published dynamic micro-CT imaging performed at Imperial College London [14].

MATHEMATICAL FORMULATIONS

The Lattice Boltzmann method and Finite Volume method are coupled in Fortran codes to calculate fluid flow and chemical reaction directly on micro-CT images.

Lattice Boltzmann Simulation of Flow

We use the lattice Boltzmann method based on D3Q19 to solve the incompressible fluid flow [9] by applying the discrete Boltzmann equation,

$$f_i(\mathbf{x} + \mathbf{e}_i \Delta t, t + \Delta t) - f_i(\mathbf{x}, t) = -\frac{1}{\tau} \left(f_i(\mathbf{x}, t) - f_i^{eq}(\mathbf{x}, t) \right), \quad (1)$$

where f_i is the distribution function along the i -direction, \mathbf{e}_i is the discrete lattice velocity, Δt is the time step, τ is the relaxation time parameter and f_i^{eq} is the equilibrium distribution function. The fluid flow density and velocity can be calculated by $\rho = \sum_i f_i$ and $\underline{\mathbf{v}} = \frac{1}{\rho} \sum_i \mathbf{e}_i f_i$, respectively. The no-slip boundary condition is enforced at the solid surface by applying the bounce-back scheme [15]. The solution obtained based on the lattice Boltzmann simulations reproduces the incompressible Newtonian Navier-Stokes equations with constant kinematic viscosity of $\frac{1}{3}(\tau - \frac{1}{2})$ [16].

Finite Volume Simulation of Reaction

The solute concentration can be updated by solving the advection-diffusion equation,

$$\frac{\partial C}{\partial t} + (\underline{\mathbf{v}} \cdot \nabla) C = \nabla \cdot (D \nabla C), \quad (3)$$

in which C [$mol\ m^{-3}$] is concentration and D [$m^2\ s^{-1}$] is the diffusion coefficient. A finite volume scheme is applied to discretize the concentration equation on micro-CT images. The first order kinetic is enforced at solid surface via

$$D \frac{C(t + \Delta t) - C(t)}{\Delta x} = -\kappa(C(t) - C_s), \quad (4)$$

where κ [ms^{-1}] is the reaction rate constant, Δx [m] is the image resolution, Δt [s] is time step, and C_s [$mol\ m^{-3}$] is the saturated concentration.

The solid volume fraction update is related with the reaction flux by

$$\frac{p(t + \Delta t) - p(t)}{\Delta t} = -V_m S \kappa (C(t) - C_s), \quad (5)$$

where p is the solid volume fraction, $V_m [m^3 mol^{-1}]$ is the molar volume, and $S [m^{-1}]$ is the specific surface area. The concentration is initially set to $C = 0.79 mol m^{-3}$ and a Dirichlet boundary condition is applied at the inlet. Table 1 provides the constant values used in the simulations to reproduce the experimental reactive and transport regimes. For more details regarding the mathematical formulation and the numerical method, refer to [9].

Table 1. Reaction and transport parameters used for simulations

Pe	Da	Reaction rate constant [ms^{-1}]	Diffusion coefficient [$m^2 s^{-1}$]	Characteristic length [m]
2100	0.05	1.08×10^{-7}	7.5×10^{-10}	4.07×10^{-4}

RESULTS

The numerical results for reactive transport are compared with dynamic imaging experiment of a Ketton carbonate dissolution performed at Imperial College London [14]. The Peclet and Damkohler numbers are set to be identical to the experimental conditions; $Pe=2100$ and $Da=0.05$. The developed numerical method with no tuning parameter and only setting the Peclet and Damkohler numbers can reproduce the experimental observations. Note that the Peclet and Damkohler numbers are the dimensionless number characterising the regimes of transport and reaction, respectively. The dissolved core sample and orthogonal slices are presented in Figure 1, which demonstrate a uniform dissolution. Our numerical results show very similar uniform dissolution pattern as observed in the experiment observations using identical reaction parameters.

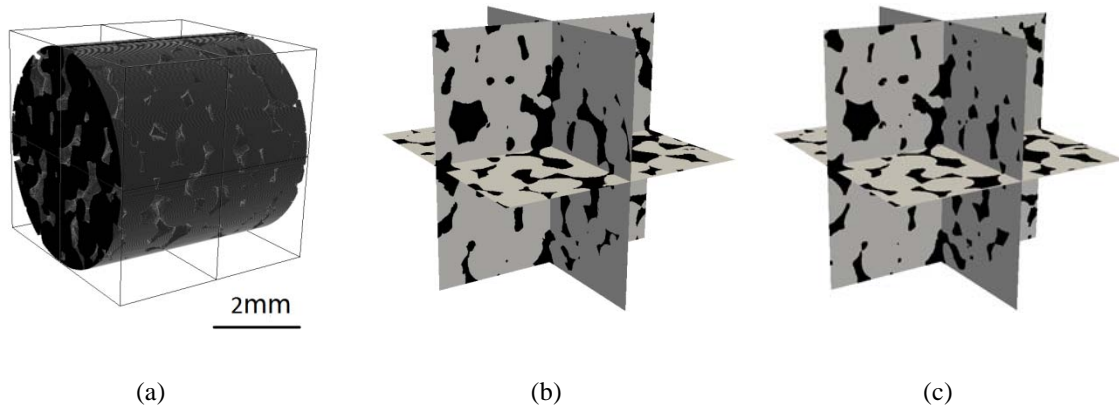


Figure 1 The 3D carbonate geometry after dissolution; (a) the full sample after 50 min, and orthogonal slices of (b) experiments, (c) simulation after 50 min of reaction.

The comparison of porosity variations between the numerical results and experimental measurements is demonstrated in Figure 2(a). There is minor difference between the predicted porosity and the experimental observations and on average our results show 3.8% difference to the experimental measurements.

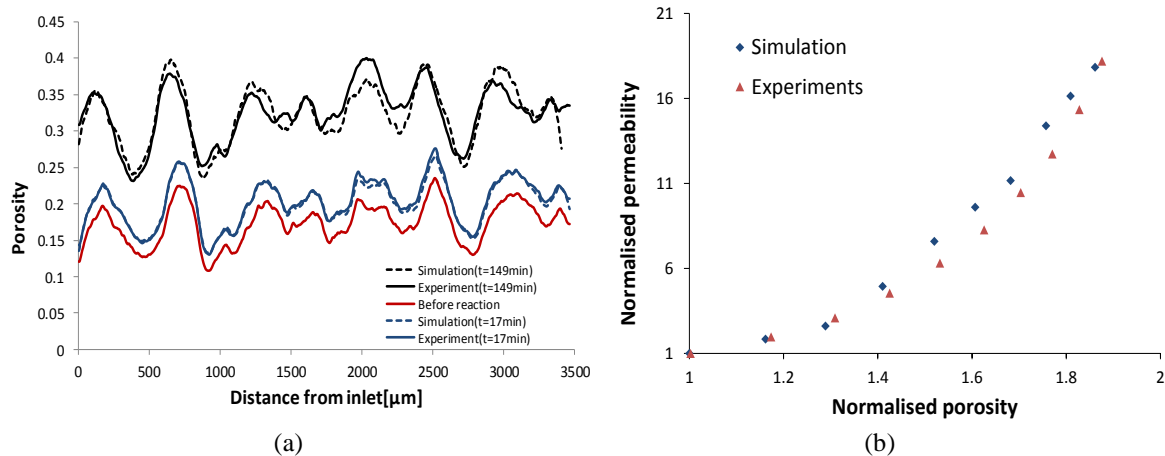


Figure 2 (a) Porosity profiles across layers normal to the main direction of flow before reactions (red); after 17 min (blue); and 149 min (black) of reaction. Solid lines are experimental measurements and dash lines are simulation results. (b) Permeability-porosity relationships of experiment and numerical results (Normalised porosity and permeability are calculated as the ratio of the porosity and permeability of the sample divided by the initial values).

The permeability-porosity relationship of both experimental and simulation results are shown in Figure 2(b). The value of n in the power-law correlation ($K=\phi^n$) is calculated as 4.58 for the experiment and for the simulation, the exponent is 4.83 (5.4% difference). The simulation results match very well with those calculated from experimental data.

In our recent published work [9], we have applied the numerical method to analyse the sensitivity of rock permeability, porosity and their relationships to the Peclet and Damkohler regimes. It was shown that the values and scaling of rock permeability and porosity is strongly dependent upon the reaction and transport regimes as well as the rock heterogeneity.

During the process of reaction, the microstructure of the pore space changes. The chord length distribution can be used to discriminate the pore structure of porous media by presenting the probability of forming a random chord length in the pore space. Chord length is described as the length of any line-segment lying in one specific phase with its endpoints at the phase interface [17]. The chord length distribution is measured directly on the images of experiments and simulation results to reflect the pore structure alteration during the dissolution. The chord length distributions of simulation and experiments in solid phase are presented in Figure 3(a). The distribution of the predicted chord lengths is identical to that of experimental measurements after 149 min of reaction.

Critical radius is defined as the radius of the largest sphere percolating through a porous medium from one side to the opposite one. A larger critical radius indicates a better connectivity of the porous media. We calculate the critical radius from images of experiments and results from simulations after different time of dissolution as shown in Figure 3(b). All the values are normalised by the critical radius of the original image prior to reaction. As can be seen in the figure, the critical radius of simulation shows a very

similar trend to the one in experiments. This justifies the good match between numerical simulations of permeability and experimentally measured permeability since permeability is strongly correlated with the critical radius. The average difference of the numerical prediction and experimental results is less than 6% during the reaction process.

In brief, critical radius and chord length are applied as two quantities expressing the morphological changes of the porous structure due to dissolution. The observed variations justify the permeability increase due to reaction; the critical radius increases with time which means larger throats are available for fluid flow and the probability of short chord lengths in the solid phase increases indicating occurrence of dissolution.

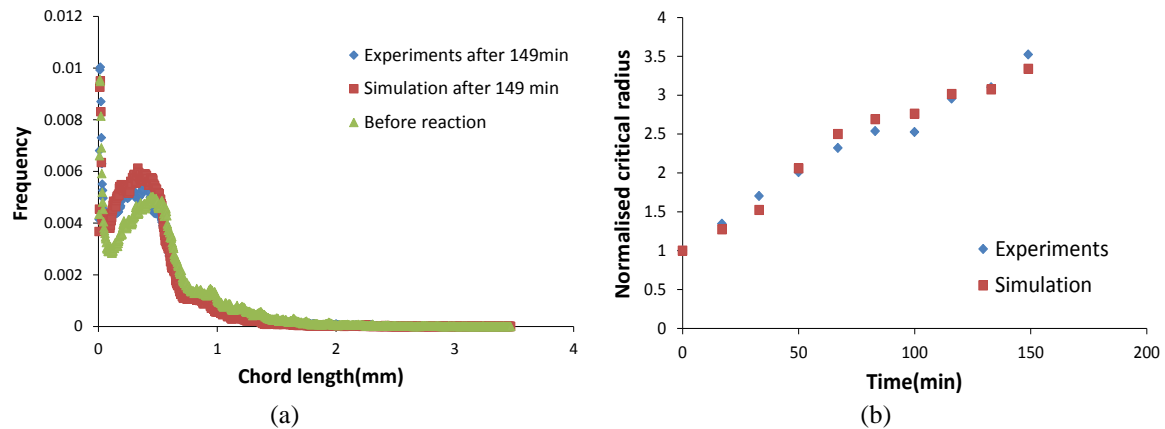


Figure 3 (a) Distribution of chord length in the solid phase for experiments and simulations; (b) Variation of critical radius of experiments and simulation with time.

CONCLUSION

We have developed a numerical model to simulate the reactive flow on micro-CT images of carbonate. We compare the numerical results with dynamic imaging observations to validate the model. The permeability-porosity relationship of simulations and experiments are measured. The predicted permeability in the simulations presents very good agreement with the experimental measurements. The local porosities and chord length distribution as well as critical radius in simulation match very well with experimental measurements. The numerical framework coupling lattice Boltzmann method and finite volume formulations can accurately predict the reactive flow and provide important understanding in pore scale reactive transport.

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