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High-resolution pore-scale simulation of dissolution in porous media

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HIGHLIGHTS

- A scalable numerical framework for modelling reactive transport is developed.
- The migration of solid particles released due to dissolution is included.
- It provides accurate prediction of pore structure and hydrological properties.
- Results are compared against published dynamic micro-CT imaging of a carbonate.

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ABSTRACT

Reactive flow is imperative in a wide range of chemical sciences, hydrogeological and environmental applications. A parallel numerical framework is presented for modelling the dissolution of a carbonate rock at the pore scale. Mass transport, chemical reactions, solid updates and migration are included in the model which are solved by the combination of lattice Boltzmann and finite volume methods. For calculation of the flow field, the incompressible Stokes equation is solved by applying an efficient lattice Boltzmann method with the D3Q19 scheme. The solid-fluid interaction is computed with the finite volume method. The numerical method includes the migration of solid particles released due to dissolution within the porous medium. The solid migration is realised by the cluster analysis and local movement. We validate this model by comparing against published dynamic micro-CT imaging experiments for dissolution of a Ketton carbonate. To measure the local dissolution, the porosity profiles are compared with the published experimental observations. The increases in permeability and porosity are investigated and a power law is derived to describe their relationship. Then, the significance of capturing the migration of solid particles released due to dissolution on hydrological properties of rocks is explored. The numerical approach is able to perform parallel simulation on large high-resolution micro-CT images. We show the importance of simulation directly on micro-CT images without reducing the resolution of rock micro-CT images. Further simulations are performed at Péclet regimes similar to sub-surface flow and the effect of flow rate on reactive transport is studied. This study illustrates the effect of inclusion of solid migration and the capability of simulation of reactive transport directly on high-resolution images and helps understand the reactive transport at the pore scale.

1. Introduction

Reaction at the pore scale has been attracting attention in both scientific and engineering disciplines. Main applications of reactive transport include enhanced oil recovery in petroleum engineering (Maheshwari et al., 2013; McLeod, 1984), carbon dioxide capture and storage (House et al., 2006; Obi and Blunt, 2006), recovery from unconventional resources of energy (Vengosh et al., 2014) and diagenetic processes studies (Lehmann et al., 2002). Apart from these applications in subsurface environment, reactive transport is also commonly involved in a wide range of chemical engineering and sciences. One example is the optimisation of catalyst. Crude oil flows through a pore network of catalyst. Chemical reactions including catalytic hydrodemetallation occur in the pore space of catalyst pellets (Keil and Rieckmann, 1994). Another example is the development of fuel cells (Chen et al., 2012; Ismagilov et al., 2000; Kjeang et al., 2009) where oxidants and fuels transport in the flow channels (Kjeang et al., 2009). They are mixed between the cathode and anode and chemical reactions occur on the electrode surface (Chen et al., 2012).

Numerical study of transport phenomena in porous media without reaction has been considered for more than a decade

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(Maier et al., 2000; Zhang and Kang, 2004; Mostaghimi and Mahani, 2010; Ovaysi and Piri, 2011; Mostaghimi et al., 2014, 2015). Maier et al. (2000) combined the random-walk particle-tracking method with the lattice Boltzmann method to simulate dispersion at different Péclet numbers. Their results show that the rates of longitudinal dispersion are much lower than previous published results. Zhang and Kang (2004) used the lattice Boltzmann method to investigate the solute transport in fractured porous media and quantified the interactions between a fracture and its host matrix. They found that the mass transfer constant is related to the matrix diffusivity and the square of the solid grain size in the porous medium. Ovaysi and Piri (2011) applied a moving particle semi-implicit method to simulate solute transport in disordered porous media and their results show that the incorporation of inertial forces into their model may decrease the predicted dispersion coefficients. Bijeljic et al. (2013) applied the continuous time random walk framework for simulating the non-reactive transport and flow in different classes of rocks to study the dispersion and solute transport in pore space. They related the propagators with the velocity distributions and provided suggestions for large scale simulations.

However, when reactions occur at the pore scale, the pore structure will change significantly and affect the fluid flow transport consequently. To understand the reaction processes at the pore scale, two approaches have been widely applied (Blunt et al., 2013). One approach is based on network modelling, which extracts a representative combination of pore nodes connected by throats from the images. Pore network modelling has been widely employed to study reactive flow in porous media and investigate the reaction-induced changes of transport properties (Algive et al., 2009, 2007, 2010; Raoof et al., 2012; Varloteaux et al., 2013a). Scaling effects of reaction rates were also investigated by analysing the concentration heterogeneity within the pore network models (Li et al., 2006, 2008; Meile and Tuncay, 2006). Pore network modelling is efficient in computing transport properties (Algive et al., 2009) but it cannot preserve complex porous structures of rocks (Varloteaux et al., 2013a) and is unable to resolve the local alteration of solute concentrations due to reaction.

A more deterministic approach is direct modelling which computes the flow properties directly on the porous media geometry. The lattice Boltzmann method as the most popular method of direct modelling (Blunt et al., 2013) has been applied to study reactive flow in porous media. A 2D lattice Boltzmann model has been established to study dissolution as well as precipitation at pore scale by Kang et al. (2003, 2002). Kang et al. (2006) extended their lattice Boltzmann model to multi-component fluids to perform the 2D study of reaction between the carbon dioxide saturated brine and numerically constructed porous media. Acharya et al. (2007) applied a 2D pore-scale model to test the application of classical transverse dispersion coefficients in reactive flow in different porous media structures. They combined lattice Boltzmann and finite volume methods to solve for flow field and reactive transport, respectively and found that reactant mixing can be determined by transverse dispersion coefficients in different geometrical properties. Willingham et al. (2008) applied the lattice Boltzmann method for calculating fluid flow and used a finite volume method for solving the reaction to predict the process of reaction in 2D and study the effect of pore structure on reactive flow. Their results demonstrated that the porous media geometry plays a significant role in reactant mixing and extent of chemical reaction. Yoon et al. (2012) presented a 2D lattice Boltzmann model for simulating the precipitation and dissolution of calcium carbonate and compared their results with microfluidic observations. Kang et al. (2014) employed their lattice Boltzmann reaction model to study the changes of hydrological properties resulted from dissolution and obtained the permeability and porosity relationship of a 2D model with a grid size of 256×256. They showed that the reaction regimes are closely related with Péclet and Damköhler regimes. Yoon et al. (2015) included biofilm dynamics into their numerical model based on the lattice Boltzmann method and compared their predictions with 2D microfluidic experiment results. They indicated that the incorporation of biofilm growth enables the numerical model to simulate the pore clogging as well as mineral precipitation.

Besides lattice Boltzmann method, conventional computational fluid dynamics methods such as finite difference and finite volume methods (Maheshwari et al., 2013; Manwart et al., 2002; Molins et al., 2012; Shabro et al., 2012) and smooth particle hydrodynamics methods (Edery et al., 2011; Sadhukhan et al., 2012; Tartakovsky and Meakin, 2006; Tartakovsky et al., 2007) have also been used to study the reaction in porous media. Maheshwari et al. (2013) applied a 3D pore scale model based on finite volume method to evaluate the wormhole induced by dissolution in carbonates. Their numerical model were limited to $150 \times 60 \times 60$. Pereira Nunes et al. (2016) applied a particle-based simulation method and successfully predicted the carbonate dissolution; due to computational efficiency they decreased the resolution of image and number of voxels from $922 \times 902 \times 911$ to $455 \times 451 \times 461$. Their results provided insight into pore-scale processes of dissolution in carbonates.

Direct modelling can accurately capture the alteration of pore structure and concentration in porous media due to reaction. Its main drawback is computational time especially for large high-resolution micro-CT images that may have sizes higher than $1000 \times 1000 \times 1000$. Thus, the domain size has become a limiting factor for direct simulation of reactive transport. For the same sample, a larger image size means a better resolution which can directly reflect the geometrical properties of the porous media and will also affect the prediction of reaction parameters (Li et al., 2006). To deal with larger images with better resolution, we develop a parallel scalable reaction simulator capable of dealing with available large high-resolution images without the need to upscale.

In addition, the movement of solid particles released due to dissolution with fluid flow also plays an important role in reaction process. It can result in pore clogging to block the flow in micro-channels and change the pore structure (Luquot et al., 2014; Wantanaphong et al., 2006). Experimental observations have reported its significant effect on petrophysical properties (Qajar et al., 2013). Besides, in low salinity enhanced oil recovery, the ion exchange processes can change the ion concentration (Austad et al., 2015; Yutkin et al., 2016) and result in fines migration. The movement of fine solid particles may lead to significant changes in petrophysical properties and oil recovery efficiency (Tang and Morrow, 1999). However, the literature in numerical modelling of reactive transport has commonly ignored this phenomenon.

This paper aims to introduce a scalable numerical framework to computationally model the complex reactive transport in porous media at the pore scale. The migration of solid particles released due to dissolution is incorporated into the numerical model. The numerical simulation combines lattice Boltzmann and finite volume methods and can be performed directly on micro-CT images. We compare dissolution results of our numerical simulations with dynamic imaging experiments published by Menke et al. (2015) and explain the effect of solid migration on porosity profiles as well as the permeability of the rock. We also explore the effect of the resolution of micro-CT images on numerical results. At the end, we conduct simulations on the same rock at the flow regime similar to the subsurface reservoir conditions.