

Pore shape evolution in different transport regimes: Single-pore scale simulations in carbonate rocks

Introduction

Calcite is the main mineral found in carbonate rocks, which form significant hydrocarbon reservoirs and subsurface repositories for CO₂ sequestration. Major processes that occur upon injection of CO₂ in carbonates are dissolution of calcite and modification of the pore-space structures. A number of pore network models acknowledge this dissolution-induced geometry change in the form of a relation between pore throat radius and the changing pore volume (Nogues et. al., 2013). However, this relation is considered to be independent of the transport and reaction regimes, in spite of observations at the continuum of different dissolution features being controlled by flow velocity and reaction rates (Luquot et. al., 2014). The choice of the relation between pore throat radius and pore body volume plugged into pore network models, impacts the extracted porosity-permeability relationship. That's why the investigation of this relation at pore-scale has motivated author for this research. Here we present a study on the shape of the dissolved pore in the context of varying flow velocity and reaction rates.

Model Description

To simulate the processes accompanied with the injection of CO₂ in a single pore of calcite, we have used COMSOL Multiphysics. The fluid velocity are within the range of creeping flow, so we solved stokes equation. The chemistry is defined by the coupled system of equilibrium reactions and kinetic reactions. The reactions related to the dissolution of CO₂ in water, formation of carbonic acid and dissociation of carbonic acid are assumed to be in the bulk and fast enough to be in equilibrium at each time step. The diffusion of produced ions from these reactions moves towards the surface of calcite and causes the dissolution of calcite. This dissolution is modeled with the nonlinear rate law which is a function of the rate with which species i.e. Ca and CO₃ are transported to and away from the CaCO₃ surface. This dissolution results into a moving boundary of calcite. The arbitrary-Lagrangian Eulerian (ALE) approach is used to track the solid-fluid interface at each time step. In this approach, all the governing equations are solved at the Eulerian grid and this grid is updated with time. The transport of bulk species is solved by the advection-diffusion-reaction equation. TO have a mass balance the flux conditions are defined from reacting surfaces to the bulk fluid.

Results

We used three average flow velocities as boundary condition, to cover diffusion dominated and advection dominated transport regimes. Each flow velocity yields a typical pore shape during the initial, transient stage of the simulation. We observed that this pore shape is a result of the competition between advection of reaction products along the pore length and diffusion-led transversal mixing. In the transient stage, these processes define a flow-regime specific pore shape that is subsequently observed to be quasi steady state. The diffusion-dominated flow is witnessed to produce the non-uniform shapes because the fluid becomes less reactive along the pore length. However, for the advection-dominated flow, the fast velocity keeps the fluid relatively more reactive towards the end of the pore length, thus yielding uniform pore shapes. To summarize, different transport regimes affect pore-shape evolution differently during the initial, transient stage of fluid

flow. The uniform or non-uniform pore shapes are inherited, remaining unaffected during quasi-steady state fluid flow (Figure 1).

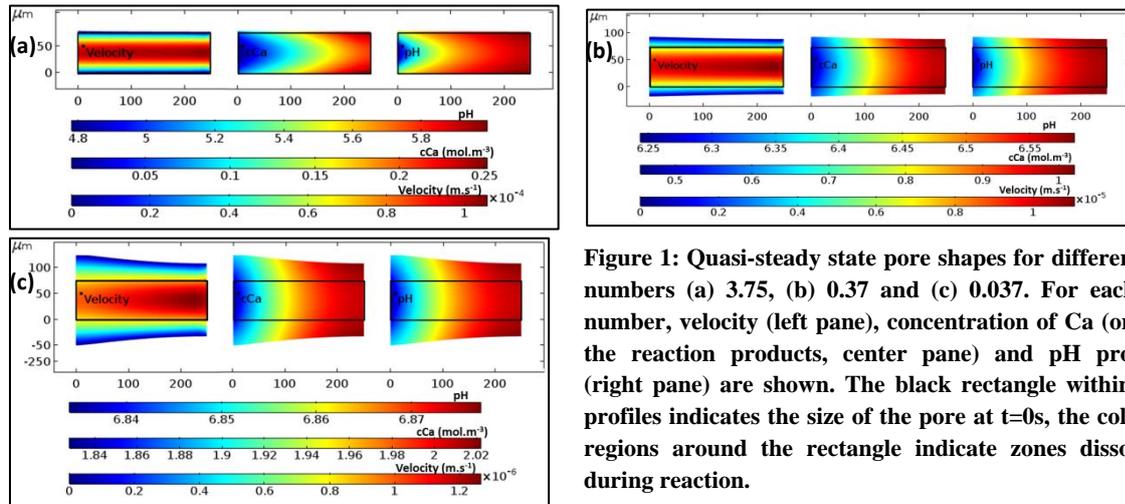


Figure 1: Quasi-steady state pore shapes for different Pe numbers (a) 3.75, (b) 0.37 and (c) 0.037. For each Pe number, velocity (left pane), concentration of Ca (one of the reaction products, center pane) and pH profiles (right pane) are shown. The black rectangle within the profiles indicates the size of the pore at t=0s, the colored regions around the rectangle indicate zones dissolved during reaction.

Since uniformity of a pore impacts the pressure drop profile along the pore, our observations at the transient-stage suggest that pore-shape and conductivity evolution cannot be assumed to be independent of flow regime (Figure 2).

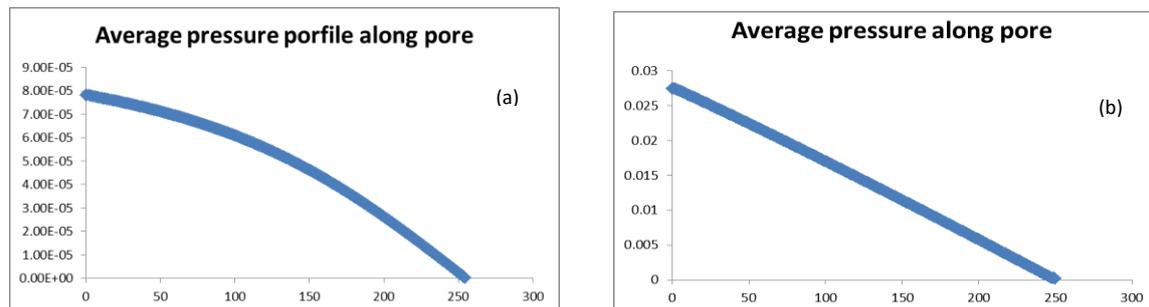


Figure 2: The average pressure profile along the pore for (a) Pe = 0.037 (diffusion dominated transport) and (b) Pe = 37.5 (advection dominated transport)

Future work involves validating these observations experimentally through microfluidic experiments. We have created a micromodel using calcite crystal by drilling a hole of size 0.4mm (diameter) and 0.7mm (length) using the laser beam. This channel can be seen in the con-focal microscopy. The next step is to establish the flow and track the position of the dissolving boundary, to validate the simulated results. And, implementation of these results in the form of the conductivity and volume change relation is also in process in such a way that our shape-factor information can be included in pore network modelling.

References

1. Noguees, Juan P., Fitts, Jeffrey P., Celia, Michael A. and Peters, Catherine A., Permeability evolution due to dissolution and precipitation of carbonates using reactive transport modeling in pore networks, *Water Resources Research*, **49(9)**, 6006–6021 (2013)
2. Luquot, L., Rodriguez, O. and Gouze, P. Experimental characterization of porosity structure and transport property changes in Limestone undergoing different dissolution regimes', *Transp Porous Med*, **101**, 507–532 (2014)