Pore-Scale Modeling of Permeability Evolution Due to Reactive Processes

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SUMMARY

Mineral precipitation in the pores near the wellbore and exsolution of biogenic gases can significantly reduce injection well performance in a bioremediation project. Although the typical size of a cleanup site is measured in hundreds of meters or even kilometers, the underlying geochemical and hydrologic processes in individual pores eventually define where and how fast the permeability will decline. Understanding these pore-scale mechanisms will help reduce the hindering effect of pore clogging and develop efficient well treatment procedures if the injectivity drops below a minimum tolerance level. Additionally, model-based correlations between porosity and gas saturation on the one hand, and permeability on the other are needed as input for realistic reservoir-scale numerical simulations.

We employ a sequential approach to model dynamic interaction between reactive transport and evolution of the pore space geometry. First, the flow field is evaluated by solving the Stokes equations. The result is passed as input data to a multicomponent reactive transport code, which computes dissolution and precipitation rates. The changes in the domain geometry caused by the reactive processes are tracked based on mass balance considerations. The flow field is re-evaluated after every domain update. Simulation of dissolution in simple pore geometries allows us to compute porosity-permeability and porosity-tortuosity changes as the pore structure evolves. Simulations of dissolution and precipitation of CaCO3 in simple pore geometries are used to examine the impact of pore structure evolution on permeability, tortuosity and average reaction rates. Three dimensional images of the porous media such as those collected by x-ray computed tomography are used as input data. Two-phase flow simulations evaluate the reduction of permeability by gas bubbles.

APPROACH

The pore-scale model describes the dynamic coupling between flow and reactive transport based on a sequential approach

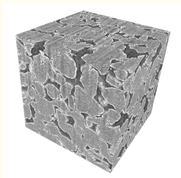
Stokes Flow

$$\begin{cases} \eta \nabla^2 v = \nabla p \\ \nabla \cdot v = 0 \end{cases}$$

geometry update

Reactive Transport

$$\begin{cases} \frac{\partial c_i}{\partial t} + v \nabla c_i - D \nabla^2 c_i - R_i = 0 \end{cases}$$



Geochemical System: Calcite Dissolution and Precipitation

The calcite dissolution rate is applied at the solid-fluid interface:

$$\left.\begin{aligned} CaCO_3(s) + H^+ &= Ca^{2+} + HCO_3^- \\ CaCO_3(s) + H_2CO_3^* &= Ca^{2+} + 2HCO_3^- \\ CaCO_3(s) &= Ca^{2+} + CO_3^{2-} \end{aligned}\right\} \quad \text{Rate} = (k_1 a_{H^+} + k_2 a_{H_2CO_3^*} + k_3)\left(1 - \frac{a_{H^+} a_{CO_3^{2-}}}{K_{eq}}\right)$$

Averaged Quantities

Permeability of the sediment in direction of flow calculated using the solution to the Stokes equations over the pore scale domain

$$k = \frac{Q^{tot} \mu L}{A_S \left(p^{out} - p^{in}\right)}$$

Tortuosity of the flow path calculated by solving Laplace equation over the pore scale domain (Johnson et al., 1987)

$$\alpha_\infty = \frac{\phi A \left|\psi_L\right|^2}{L \int \left|\mathbf{u}_p(\mathbf{r})\right|^2 dV}$$

Average reaction rate calculate as a mass balance between effluent and influent:

$$R_{avg} = \frac{\left(c_i^{out} - c_i^{in}\right) Q^{tot}}{\xi_{ij} A^{tot}}$$

SIMPLE PORE STRUCTURES

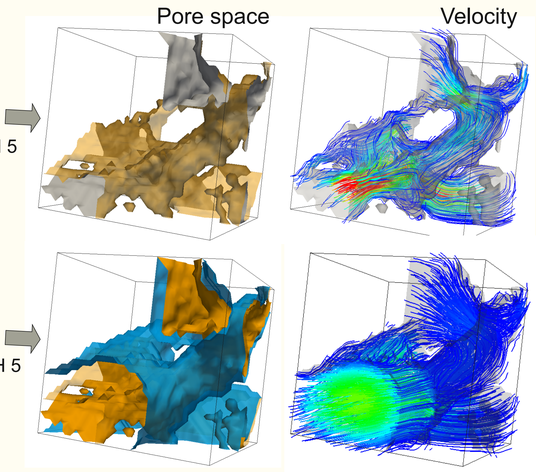
The effect of calcite dissolution on the pore space evolution (e.g. permeability, tortuosity) as a function of flow rates, and in turn, the effect on average reaction rates.

Initial pore space
Porosity = 0.144
30x30x30
Δx = 4.4 μm

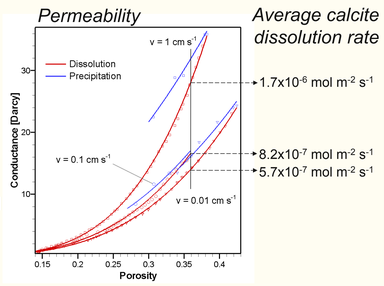
pH 5

Final pore space
Porosity = 0.382
30x30x30
Δx = 4.4 μm

pH 5



- As a result of dissolution, pores become wider and the flow paths becomes less tortuous.
- Conductance to flow increases faster than porosity
- Pore space evolution depends on the relative magnitude of flow and the reaction rate
- Different average reaction rates, calculated as a mass balance, show that at this scale the reaction is not transport-limited

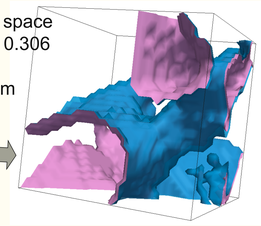


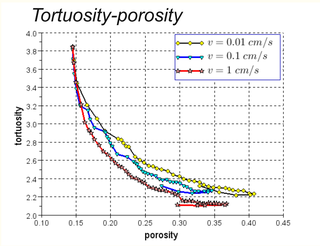
Pore space evolution due to calcite precipitation

- At low degrees of supersaturation, precipitation takes place via crystal growth on discrete surface features of the carbonate mineral surface and is modeled with a first order dependence on supersaturation.

Final pore space
Porosity = 0.306
30x30x30
Δx = 4.4 μm

pH 8.5

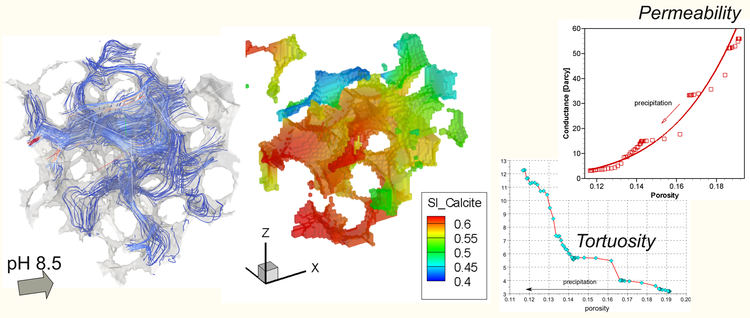




- The pore scale evolution due to heterogeneous precipitation on the pore wall starting from the end point of the dissolution experiment results in distinct porosity-permeability and porosity-tortuosity relationships.

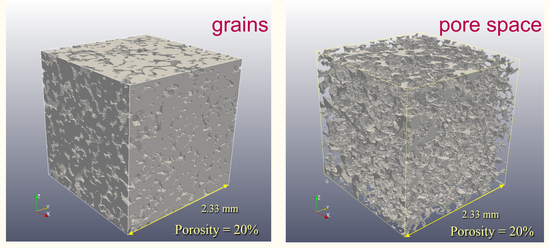
MULTIPORE STRUCTURES

Evolution due to calcite precipitation on a sphere pack

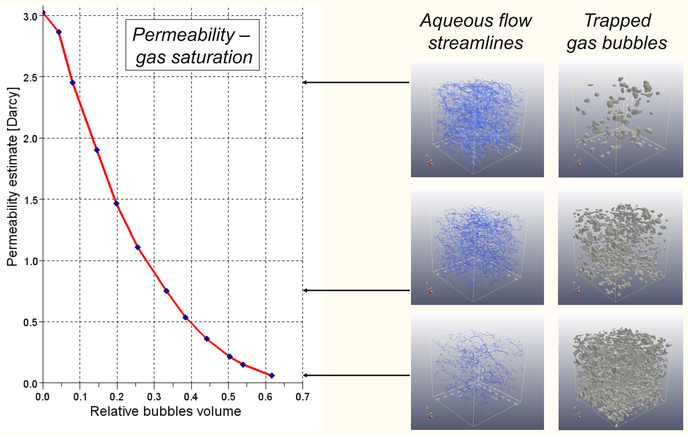


PERMEABILITY REDUCTION BY BUBBLES

The effect of the accumulation of biogenic methane or carbon dioxide on well injectivity is investigated with two-phase flow simulations that account for blocking of the flow paths by bubbles trapped in capillary equilibrium. The objective is to estimate the reduction of permeability by pore-scale simulations.



Bentheimer sandstone at 5.6 micron resolution



Capillary equilibrium fluid distribution was evaluated by the method of maximal inscribed spheres (Silin *et al.* 2011):

- Gas remains trapped for liquid saturations between 40 % and 100 %
- Even occupying small relative volume, the trapped bubbles significantly reduce the permeability to water.

CONCLUSIONS

Dissolution and precipitation reactions affect the pore space nonuniformly. As a result, a simple porosity-permeability correlation may be insufficient to describe the complexity of the reaction-induced pore space evolution. The relative magnitude of the reaction rate constants in relation to the flow rate affects the evolution of the permeability-porosity relationship. As a result of dissolution, pores become wider, flow paths become less tortuous, and permeability increases faster than porosity. For a given reaction rate constant, this process is more pronounced for faster flow rates. Hysteresis in the porosity-permeability evolution is observed in simulations of dissolution followed by precipitation.

Two-phase flow simulations are used to produce correlations between the relative volume of gas bubbles and permeability in capillary equilibrium.

REFERENCES

D. Johnson, J. Koplik, and R. Dashen (1987), Theory of dynamic permeability and tortuosity in fluid-saturated porous media, *Journal of Fluid Mechanics* 176, 379–402.

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Funding for this study was provided by the U.S. Department of Energy, BER Program Contract DE-AC020SCH11231 to the LBNL Sustainable Systems Scientific Focus Area