PORE-SCALE MODELING OF COUPLED HYDRO-GEOCHEMICAL PROCESSES

Cyprien Soulaine
Coupled hydro-geochimical processes

- Darcy scale = averaged equations with averaged properties (permeability, surface area...)
- How does the permeability evolves when the pore-structure changes due to the dissolution/precipitation?
- What is the surface area accessible to the acid component?
- What about multiphase reactive flows?
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The Darcy-Brinkman-Stokes\textsuperscript{1,2,3} equation allows a single domain formulation

\[ 0 = -\nabla \bar{p}_f + \frac{\mu_f}{\varepsilon_f} \nabla^2 \bar{v}_f - \mu_f \kappa^{-1} \bar{v}_f \]

Vanishes in the void space

Dominant in the porous region

\textsuperscript{1}Brinkman A Calculation of The Viscous Force Exerted by a Flowing Fluid on a Dense Swarm of Particles Appl. Sci. Res. (1947)
\textsuperscript{2}Neale and Nader Practical significance of Brinkman's extension of Darcy's law: coupled parallel flows within a channel and a bounding porous medium. (1974)
\textsuperscript{3}Soulaine and Tchelepi Micro-continuum approach for pore-scale simulation of subsurface processes Transport in Porous Media (2016)
- Matrix with low porosity, low permeability ≈ solid,
- Sharp fluid/solid interface,
- A no-slip boundary condition is automatically recovered at the fluid/solid interface\(^1\),
- The gradient of the porosity field is used to apply immersed reactive boundary conditions at the fluid/solid interface\(^2\),
- The volume fraction of solid is an unknown of the system that evolves with chemical reaction ("Volume-of-Solid" technique)\(^1\).


Reactive micromodels to investigate calcite dissolution

- Dissolution of a calcite crystal in a micro-channel (Sophie Roman, Wen Song and Tony Kovscek, Stanford University),
- Acquisition of a high resolution dataset to compare with numerical simulations.
Calcite dissolution: Simulation vs Experiment

experiment

simulation

Benchmark of pore-scale approaches with evolving fluid/solid interface

Molins et al. *Simulation of mineral dissolution at the pore scale with evolving fluid-solid interfaces: Review of approaches and benchmark problem set* (under review)
Upscaling of the pore-scale dissolution rate

\[ \text{Péclet number (Pe)} \]
\[ P_e = \frac{v_0 l_c}{D} = \frac{v_0 \sqrt{k}}{D} \]

\[ \text{Damkhöler number (Da)} \]
\[ Da = \frac{r l_c}{D} = \frac{r}{A_e D} \]

\[ \frac{A}{A_0} = 1 - \exp \left( -Pe^{-n} \left( \frac{Da}{Pe} \right)^{-m} \right) \]

Wormholing in reactive porous media

Wormholing in reactive porous media

- **Ramified wormholes**
- **Uniform dissolution**
- **One dominant wormhole**
- **Conical dissolution**
- **Compact dissolution**

(a) Graph showing the transition between different dissolution processes based on Pe and Da_l.

Multiphase micro-continuum model

Full Navier-Stokes approach

\[ \frac{\partial \phi S_l}{\partial t} + \nabla \cdot (\bar{v} S_l) + \nabla \cdot (\phi_r S_l (1 - S_l)) = 0 \]

\[ \frac{\rho}{\phi} \left( \frac{\partial \bar{v}}{\partial t} + \frac{\bar{v}}{\phi} \cdot \nabla \bar{v} \right) = -\nabla \bar{p} + \nabla \cdot \left( \frac{\mu}{\phi} \left( \nabla \bar{v} + t \nabla \bar{v} \right) \right) + F_c - \mu k^{-1} \bar{v} \]

Surface tension forces
Vanishes in the void space
Dominant in the porous region, depends on saturation through relative permeability

\[ k^{-1} = k_0^{-1} \left( \frac{k_{rl}}{\mu_l} + \frac{k_{rg}}{\mu_g} \right)^{-1} \]

Multiphase micro-continuum and surface tension forces

In the clear fluid regions,
- Sharp fluid/fluid interface,
- Curvature of the gas/liquid interface modeled by the Brackbill’s Continuum Surface Force (CSF)\(^1\)

\[
F_c = \sigma \nabla \cdot (\hat{n}_{lg}) \nabla S_l
\]

with
\[
\hat{n}_{lg} = \frac{\nabla S_l}{||\nabla S_l||}
\]

At the fluid/porous media interface, wettability condition described as an immersed boundary condition\(^2,4\)

\[
F_c = \sigma \nabla \cdot (\hat{n}_{lg}) \nabla S_l
\]

with
\[
\hat{n}_{lg} = \cos \theta \hat{n}_{wall} + \sin \theta \hat{t}_{wall}
\]

In the porous regions, surface tension forces modeled by capillary pressure\(^3\)

\[
F_c = \left( \frac{\partial p_c}{\partial S} \right) \nabla S_l
\]

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\(^1\)Brackbill et al. A continuum method for modeling surface tension Journal of Computational Physics, 1992, 100, 335 - 354


Interphase mass transfer

- Single-field formulation for the concentration of species in gas and liquid\textsuperscript{1,2,3},
- Continuum Species Transfer (CST) formulation\textsuperscript{1} to maintain the thermodynamics equilibrium at the fluid/fluid interface.

\textsuperscript{1}Haroun et al., \textit{Volume of fluid method for interfacial reactive mass transfer: Application to stable liquid film}, Chemical Engineering Science, 2010, 65, 2896 - 2909
\textsuperscript{3}Maes and Soulaine, \textit{A new compressive scheme to simulate species transfer across fluid interfaces using the Volume-Of-Fluid method}, Chemical Engineering Science, 2018
Simulation of multiphase reactive transport with moving boundary
Multiphase dissolution and wormholing

Single phase

Two phase


Conclusions

• Multi-scale analysis starting from the pore-scale,

• Development of a micro-continuum DBS framework with sub-grid models that includes multiphase effects,
  • Well-suited for image-based simulations with sub-voxel porosity,
  • Well-suited for simulating problems with moving boundaries at the pore-scale.

• The DBS framework is used to simulate mineral dissolution problem under single and two-phase flow,
  • We can reproduce experiments of a dissolving calcite crystal,
  • Identification of different dissolution regimes,
  • The presence of a second fluid phase impacts strongly the dissolution dynamics and limits the emergence of wormholes.

Post-doctoral position in multiphase reactive transport modeling
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Thank you for your attention!

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