The Role of Chemical Alteration in Arkosic Reservoirs

FEW0271

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Project Overview, 1/2

Does chemical alteration (negatively) impact CO_2 storage capacity in the Lower Mt. Simon sandstone?

This formation has attractive porosity and permeability, as well as abundant clay and feldspar mineralogy. Chemical alteration via CO_2 exposure is likely to enhance secondary clay formation, which may alter these injection properties.

Project objective: To quantify the role of chemical alteration on CO_2 injection and storage capacity in arkosic sandstone reservoirs, using

- detailed characterization of reservoir samples
- core-scale flow experiments at relevant (P, T, pCO_2) conditions
- reactive transport modeling and incorporation into reservoir simulator

Project Overview, 2/2

- Funding awarded August 2020 ($400k/y)
  - 3-yr project (August 2023)
  - pandemic, lab, hiring delays

- Project Participants
  - LLNL: experimental geoscientists; reactive transport, coupled chemo-mechanical modelers; reservoir code developers
  - Illinois State Geological Survey: Hongbo Shao, Steve Whittaker


Gabriela Davila


Yue Hao


Jaisree Iyer

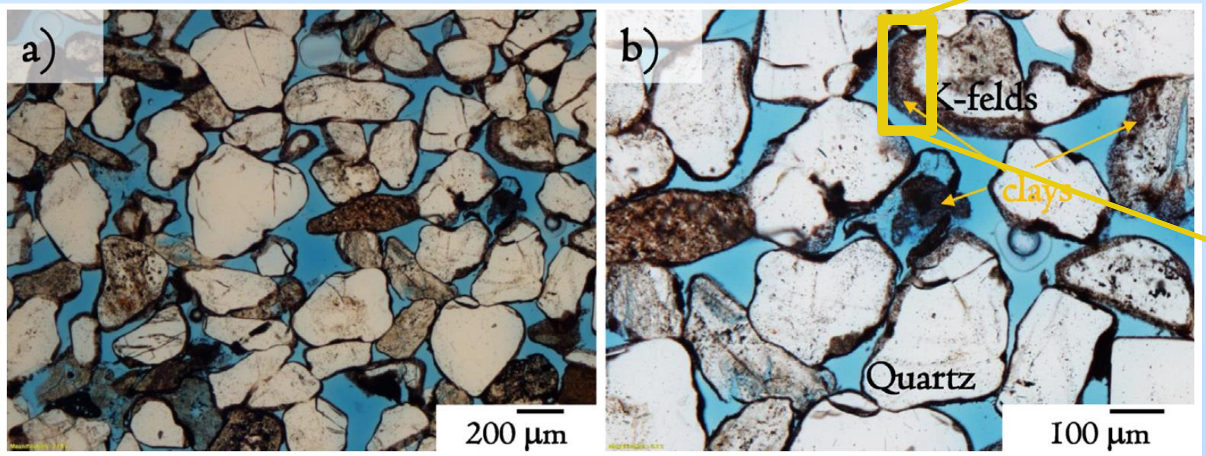

Matteo Cusini

Technology Background, 1/3

Chemical reactions among supercritical CO_2, brine, and the high surface area feldspars and clay coatings found in the Lower Mt. Simon pose an **important but poorly understood threat to CO_2 injection and long-term storage capacity.**

Completion of the work will yield a reactive transport model of this important reservoir and answer the question: does chemical alteration negatively impact CO_2 storage capacity in the Lower Mt. Simon formation?

Whittaker, ISGS

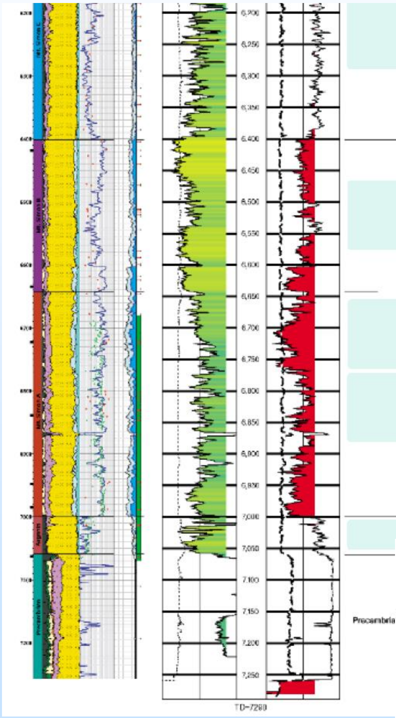


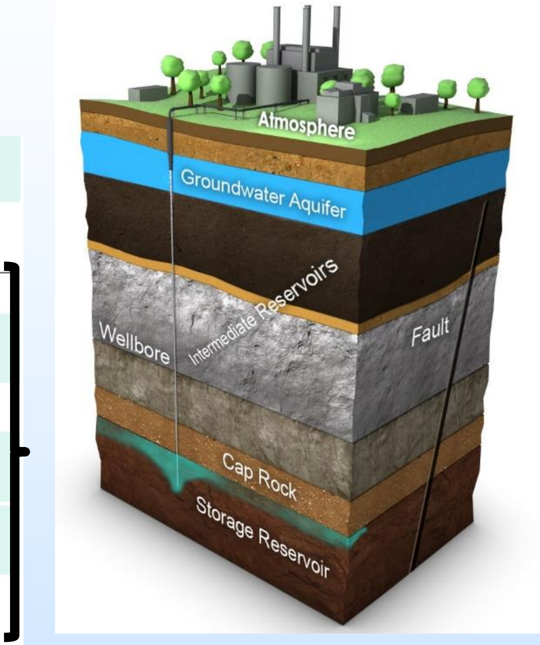
Davila et al., 2020

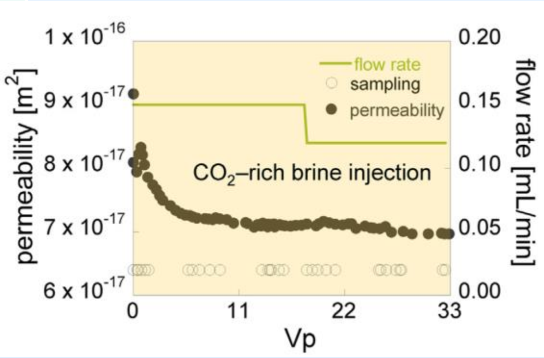
Technology Background, 2/3

This project meets the Carbon Storage Program’s goals to **address methods and tools that enable storage efficiency optimization**:

We will provide a process-based model accounting for observed changes to porosity and permeability, constrained by experiments on more common sandstone reservoir systems that experience a wider range of geochemical reaction (e.g., mineral dissolution, precipitation, and *in situ* alteration) during rock-scCO_2-brine interaction.





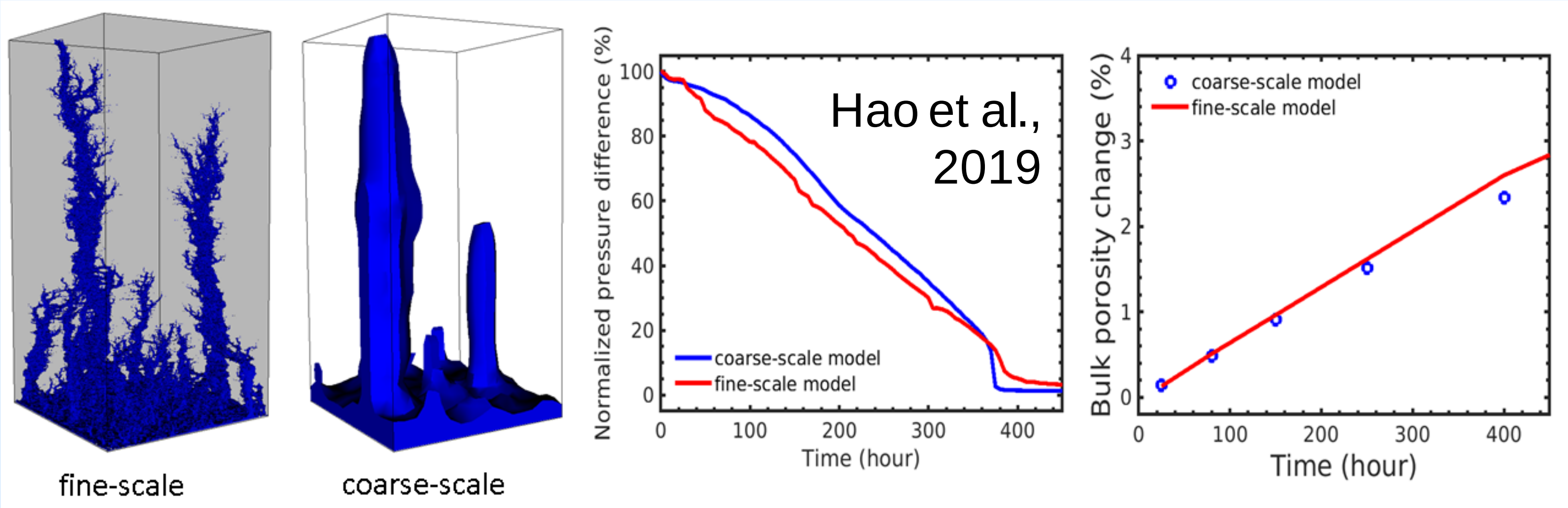


Davila et al., 2020

Technology Background, 3/3

This work builds heavily on techniques and workflows developed in **previously funded DOE Carbon Storage research** on carbonate reservoirs.

Upscaling core (cm-scale) observations to the meter-scale allowed us to examine scale dependence of key transport parameters and showed that we can correct for model resolution:



Hao et al., 2019

With such scaling, we can use much faster-running, coarser models and still be predictive. We will apply this methodology to derive relationships appropriate for sandstone reservoirs.

Technical Approach/Project Scope

Subtask 1.1: **Core-flood experiments on Lower Mt. Simon samples**

8(+) experiments, varying residence time/flowrate, both single-phase CO_2-saturated brine flow as well as multi-phase $scCO_2$ alternating flow

Subtask 1.2: **Measurement of chemical and mechanical alteration**

solution (major/trace elements, CEC) and solid (SEM, BET, NMR) chemistry analyses, pressure/permeability monitoring, non-destructive synchrotron-based imaging (XRCT) coupled with digital image analysis

Subtask 1.3: **Build reactive transport model**

preliminary geochemical and transport modeling, calibration of model against experimental results, investigation of kinetic reaction/surface area and porosity-permeability correlations, coupling of geochemical model with existing reservoir simulator

Lower Mt. Simon arkosic samples

6937 feet depth (2114 meters)

15-mm diameter ***horizontal*** subcores, 65mm length, core-flow experiments

mm-scale horizontal subcore, enhanced X-ray CT analysis

15-mm diameter ***vertical*** subcore, 65mm length, compare K_H/K_V permeability

rough-cuts from waste sections, thin/thick sectioning for microscopy, microprobe, nanoindentation, etc.

Preliminary model for reactivity

Conservation equation (advection, dispersion, reaction)

$$\frac{\partial(\phi C_j)}{\partial t} = \nabla\cdot\left(D\nabla C_j\right) - \nabla\left(v_D C_j\right) + R_j$$

$$R_j = -\sum_m \nu_{jm} R_m \qquad R_m = A_m \sum_{terms} k_{m,T}\, a_{H+}^{n_{H+}} \left(\prod_i a_i^{n_i}\right)\left(\left(\frac{IAP}{K_{eq}}\right)^{m_2} - 1\right)^{m_1}$$

Conceptual Model

3-D cylindrical core converted to 1-D symmetry

= Ci(in) + Ci(diss) - Ci(pp)

Condition Input
temperature
50.0
HCO_3^- 6.185e-1
pH charge
Na^+ 2.000e+0
Cl^- 2.0025e+0

Characterization data from:
Freiburg et al., 2016; Davila et al., 2020;
Davila, current results

Initial Conditions

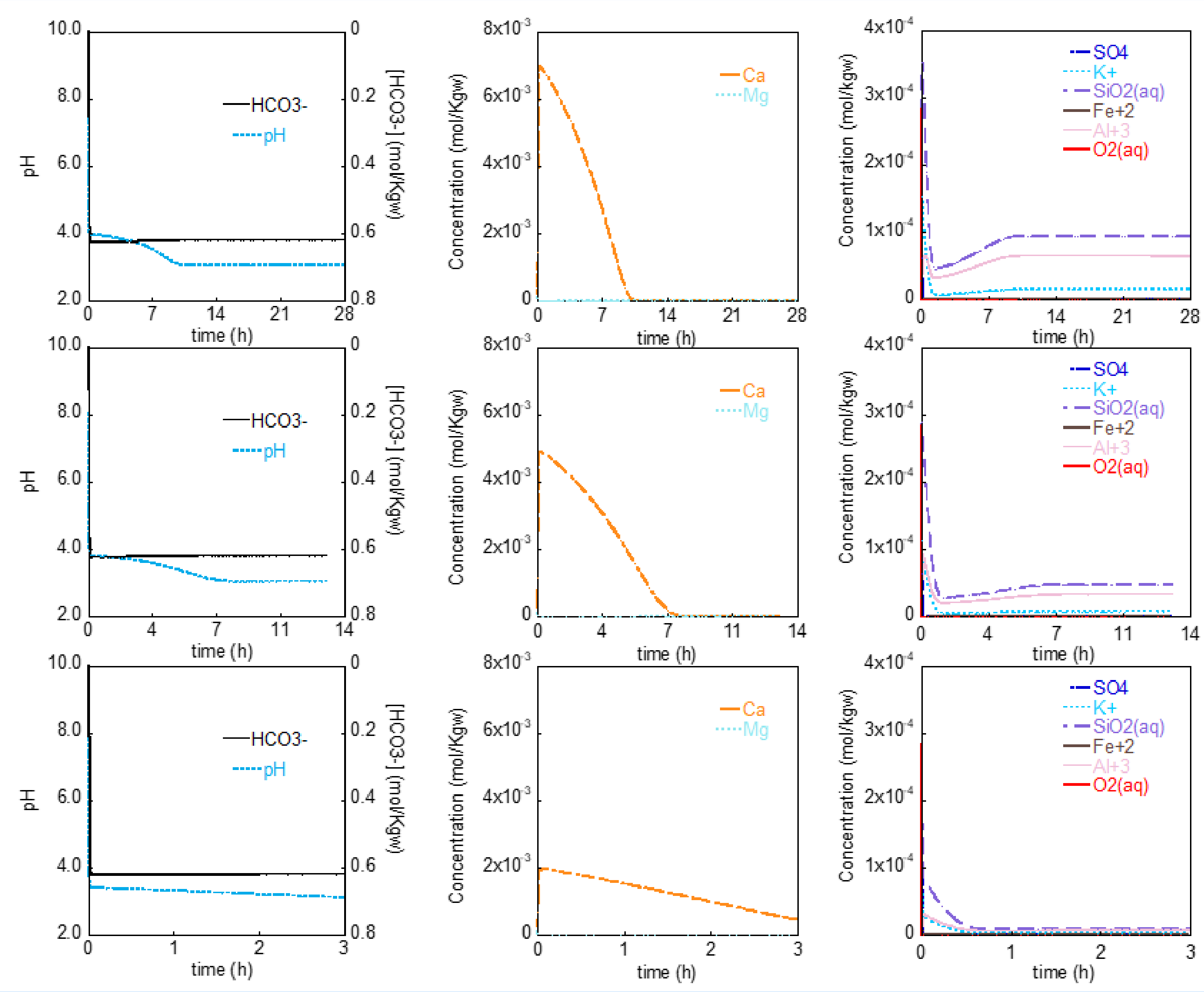
T = 50 ºC
P = 200 bar
Q_{fast} = 0.5 mL/min
Q_{med} = 0.1 mL/min
Q_{low} = 0.05 mL/min
ϕ = 0.179 ± 0.04
$k_{initial}$ = **allowed to vary**

Transport Parameters

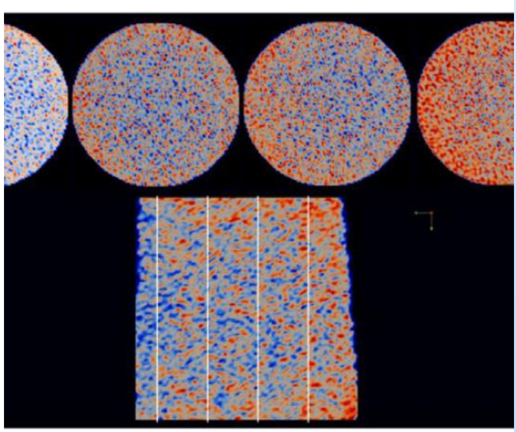
$D_{eff} = \phi^m D_o$
m = ~2.0 for sandstone rocks
initial $D_{eff} = 3 \times 10^{-13}\ m^2\ s^{-1}$

solution pH and porosity, preliminary results

fast → slow flowrate



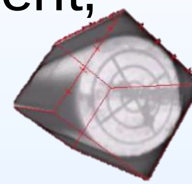
Redox-sensitive tracer metals may also be useful to track extent of reaction; e.g., Hongbo et al., 2020



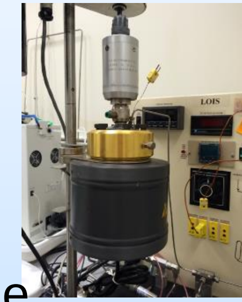
Davila et al., 2020

Additional characterization used to pinpoint key processes

- Changes to pore space/mineral distribution, grain displacement, initial sample heterogeneity – X-ray computed tomography



- Mineral association/proximity to pore space – scanning electron microscopy, microphotography



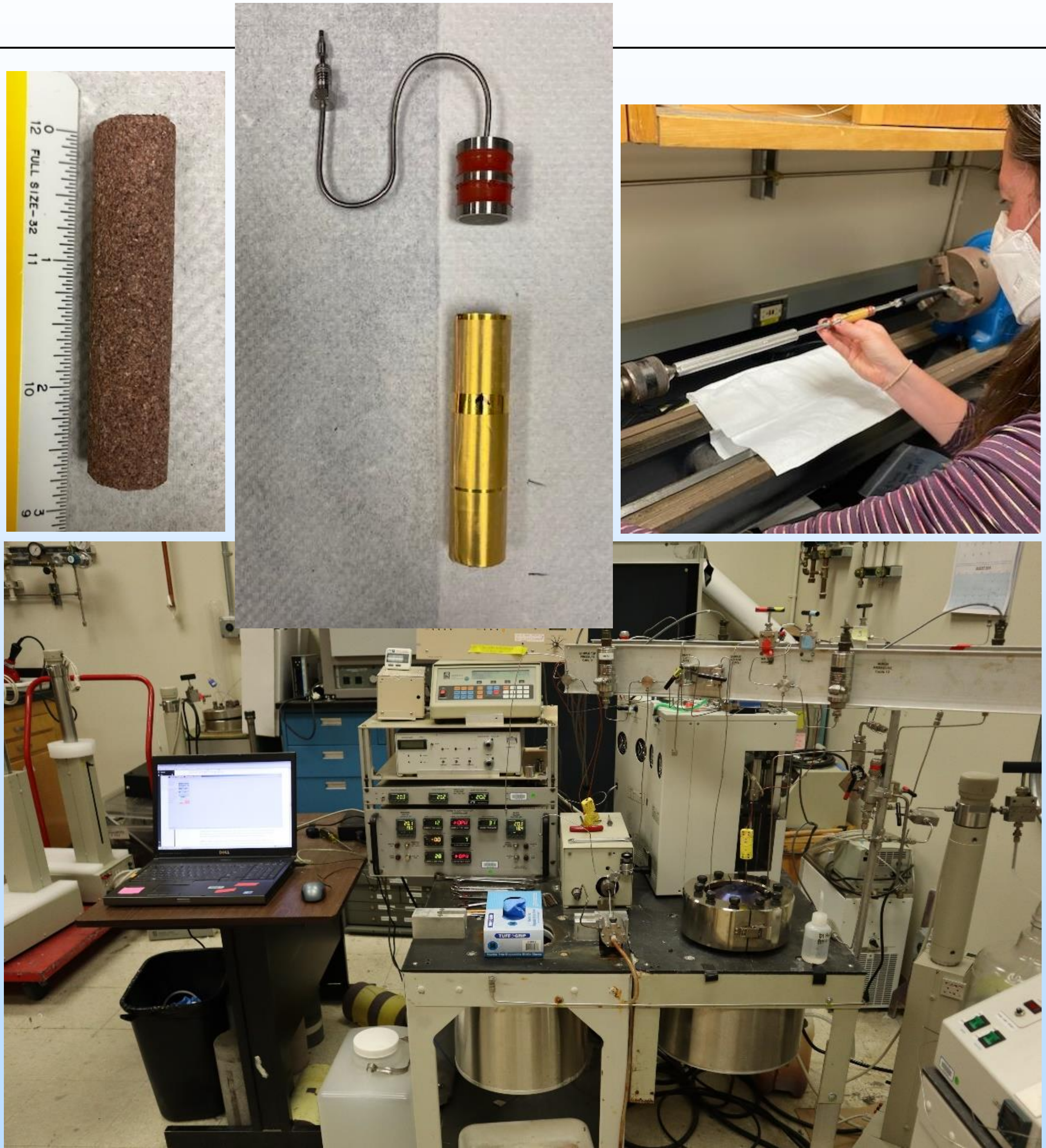
- Clay identification – X-Ray diffraction, long-term reaction, TEM
- Surface area – BET analysis, solid-state nuclear magnetic resonance

  Sanders et al., 2010, Measurement of reactive clay surface area using solid- state NMR of a probe molecule

- CO_2 and clay ion exchange effects – dialysis experiments

  Sakuma et al., 2022, Friction in clay-bearing faults increases with the ionic radius of interlayer cations

Single-phase experiments underway



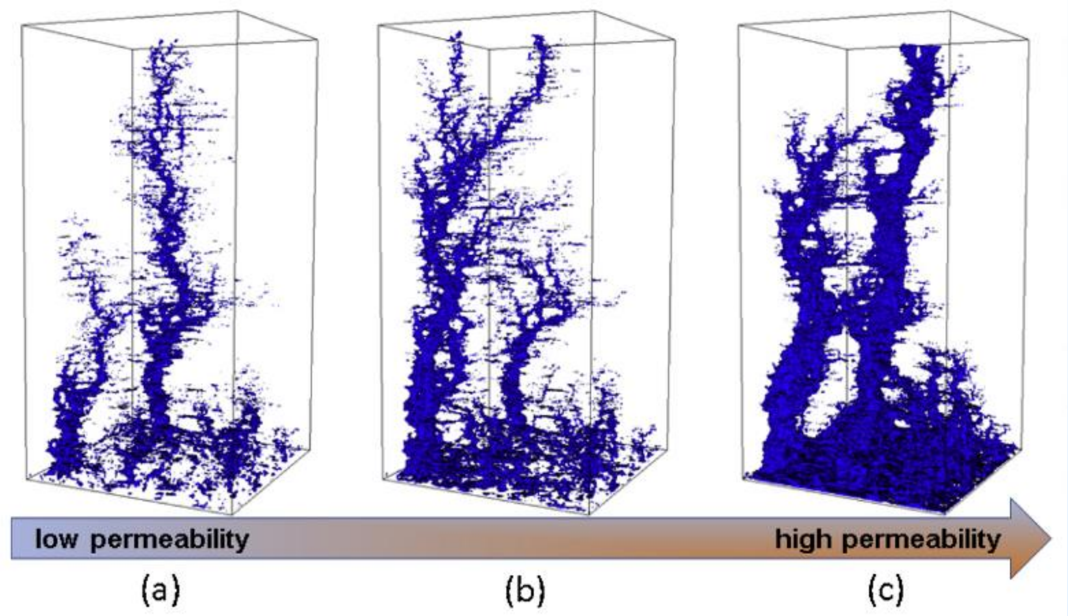
50°C, 56 bar pCO_2,
248 bar confining pressure,
single-phase flow, 3-30 days

5+ experiments before
changing to multi-phase flow

Porting reactive transport simulation capabilities to a new workflow

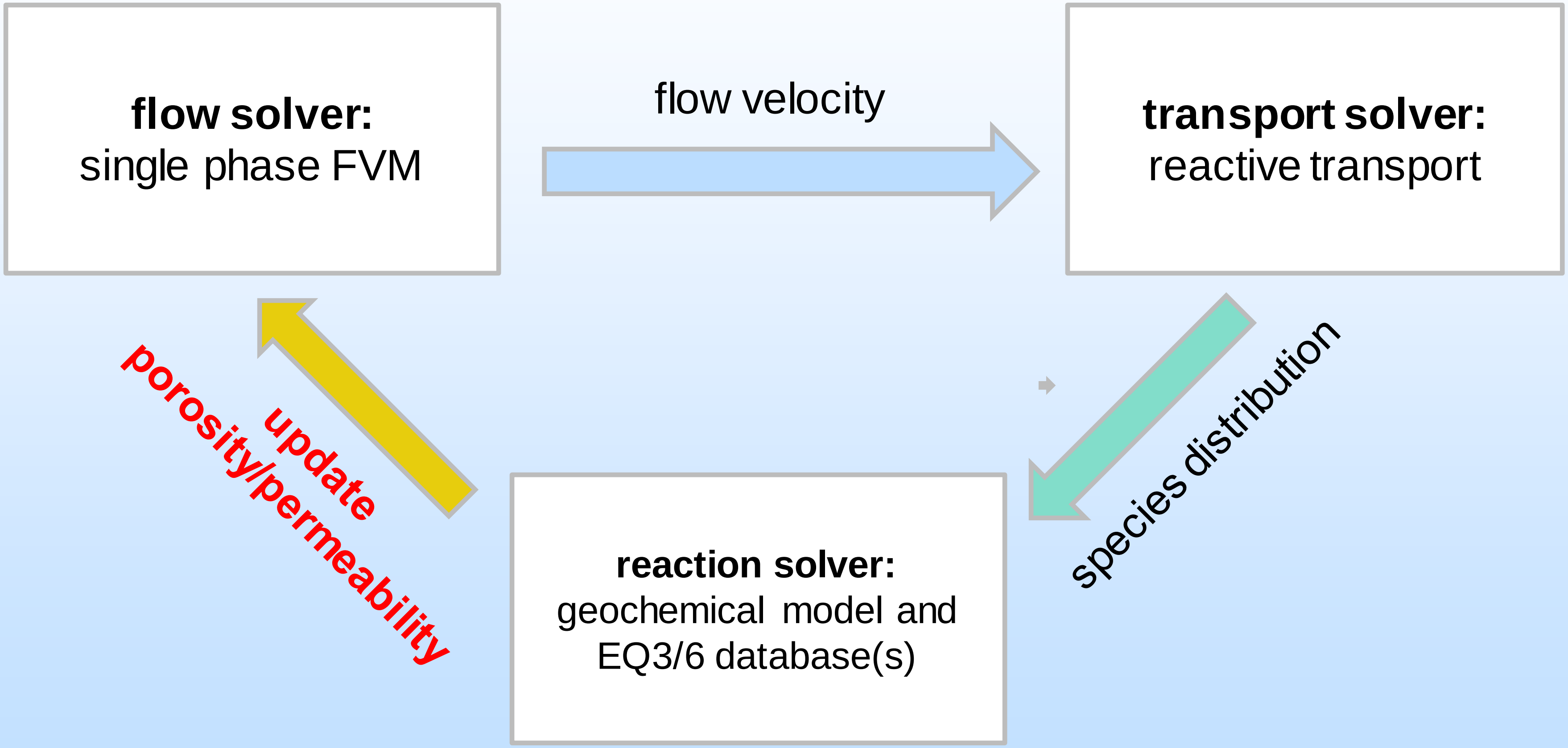
Hao et al., 2019

The original workflow utilized the reactive transport code, NUFT, which had been previously used for brute-force, high-resolution, meter-scale simulations of reactive transport in carbonate storage formation samples.



We can now leverage new developments within LLNL's multi-scale reservoir simulator, GEOSX, linking the geochemical solver EQ3/6's thermodynamic database with a flexible user input interface. This allows us to simulate flow, multi-species transport, and equilibrium aqueous speciation and reactions.

Coupling flow and reactive transport



flow solver:
single phase FVM

flow velocity

transport solver:
reactive transport

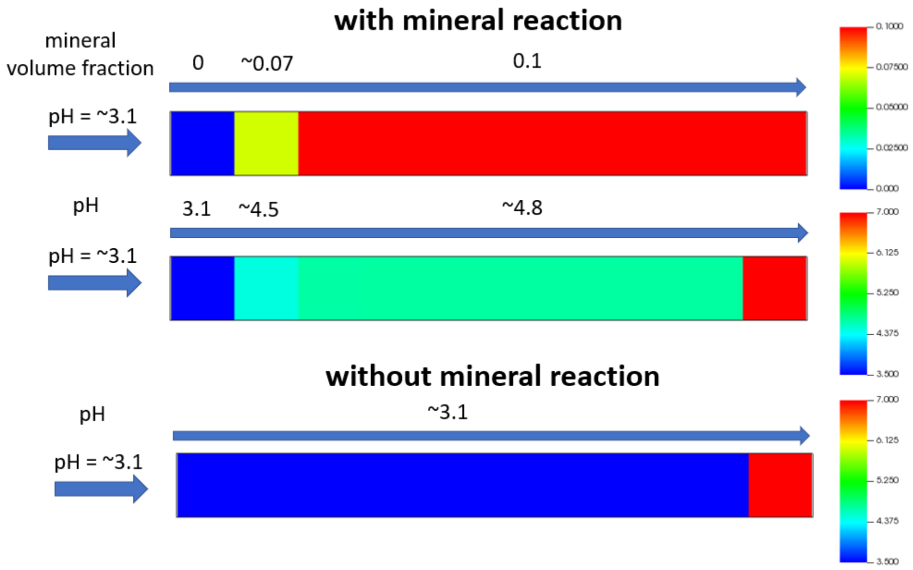
species distribution

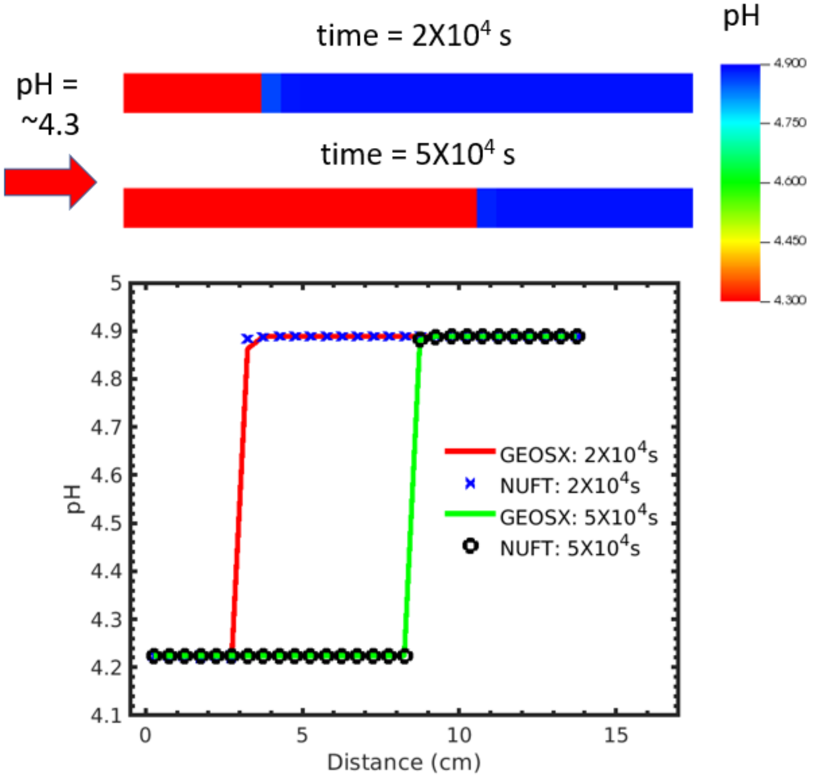
reaction solver:
geochemical model and
EQ3/6 database(s)

update
porosity/permeability

equilibrium reactions
kinetic (mineral) reactions

1-D validation of workflow against NUFT output

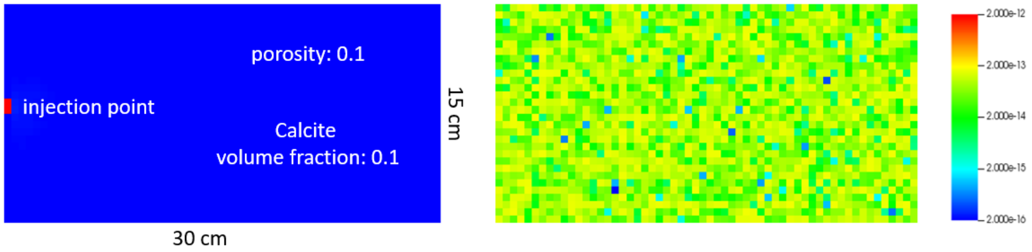




- This exercise repeated for a range of kinetic reaction rates for carbonate as well as arkosic scenarios.

2-D simulations: heterogenous sample, simplified chemistry

2D Simulation of Mineral Dissolution



model domain

initial permeability

- Extend to 3-D volume
- Refine model permeability domain with sample characterization statistics
- Test variable porosity-permeability forms (e.g., Sabo and Beckingham et al., 2021)
- Evaluate incongruent K-feldspar, clay kinetic reactions

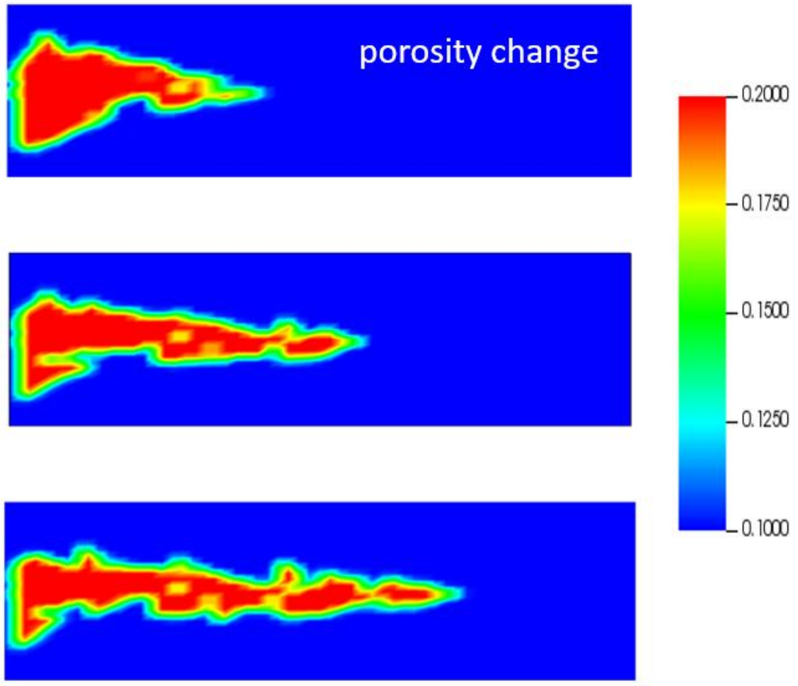
Time = 60 hours

n = 3

$$K_t = K_0\left(\frac{\phi_t}{\phi_0}\right)^n$$

n = 5

n = 7



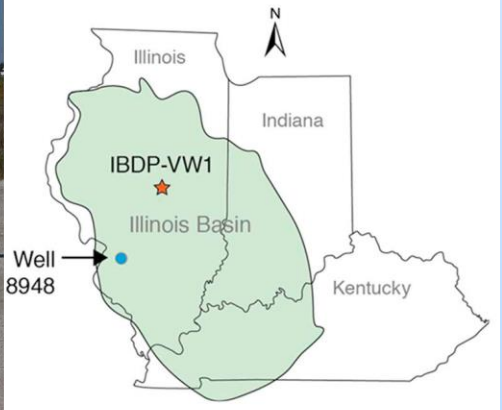
Accomplishments/Value

Key Accomplishments/Deliverables	Value Delivered
2020: Institutional upgrades to experimental equipment; posted postdoctoral position	- Leveraged institutional investments in laboratory facilities for upcoming work
2021: Down-selected from 64 candidates and hired staff; planning with ISGS and received core; brought technician on-site for sample prep and characterization; assessed kinetic data for clay formation, refined geochemical models; evaluated transport codes	- Hired an experienced scientist; preliminary modeling informed needed sampling procedures; gained statistics on sample variability; “shook-down” $scCO_2$ flow system
2022: Continued labspace upgrade; preliminary geochemical modeling for optimal chemical sampling; solid-phase characterization; single-phase core-flood experiments; submitted results to AGU Fall Meeting 2022; “road-testing” GEOSX with geochemical model	- Preliminary modeling should lower uncertainty in sample processing; first manuscript submission by end of calendar year; investment in simulation workflow will lower burden of upscaled simulations

A reactive transport model that describes the impact of CO_2-driven chemical alteration on feldspar-rich sandstone formations:

The CarbonSAFE program identified the Lower Mt. Simon sandstone for large-scale GCS based on exceptional porosity and permeability. CO_2 injection perturbs its chemical equilibrium, forming high-surface area clays, which may clog pores or change reservoir properties. We will deliver a reactive transport model of the formation that captures these processes.

Illinois Storage Corridor



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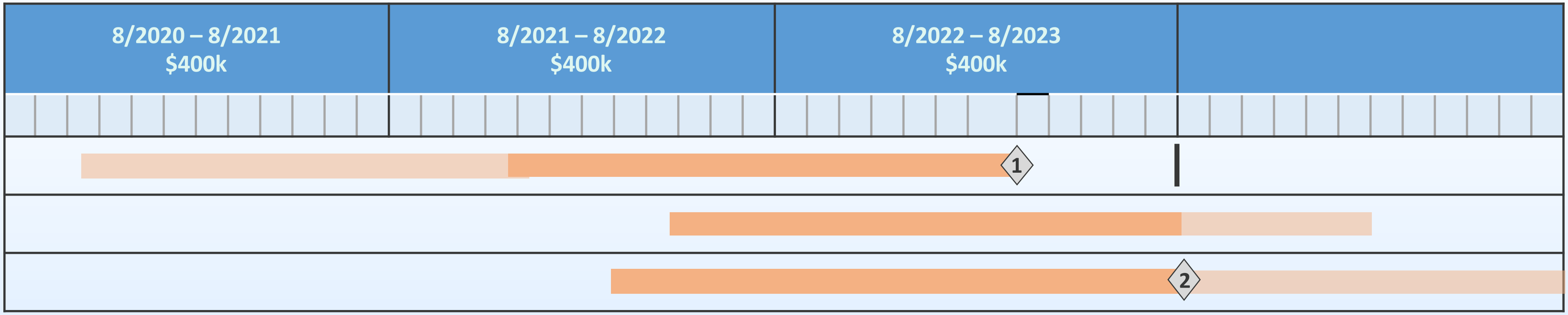
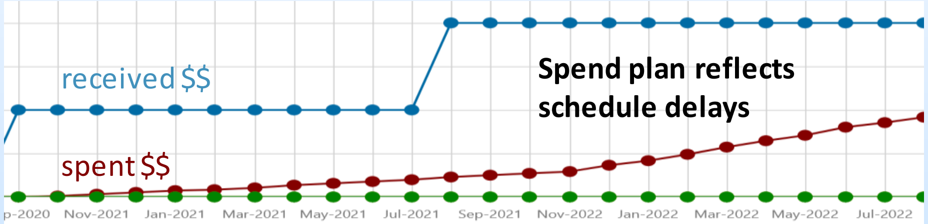
Progress





Spend plan reflects schedule delays

Milestones

1. Completion of core-flood experiments
2. Completion of chemo-mechanical-transport model for the Lower Mt. Simon sandstone

Milestone 1: Single-phase experiments ongoing: initial modeling predicts net porosity increase yet permeability decreases noted in all cases. Suite of experiments will be post-characterized and analyzed for presentation at AGU 2022. “Wet” $scCO_2$ experiments begin Dec 2022.

Milestone 2: Early progress made here, although note that final model calibration requires complete experimental datasets.