Evolution of hydrogeochemical properties during reactive flow-through experiments using CO_2-bearing solution

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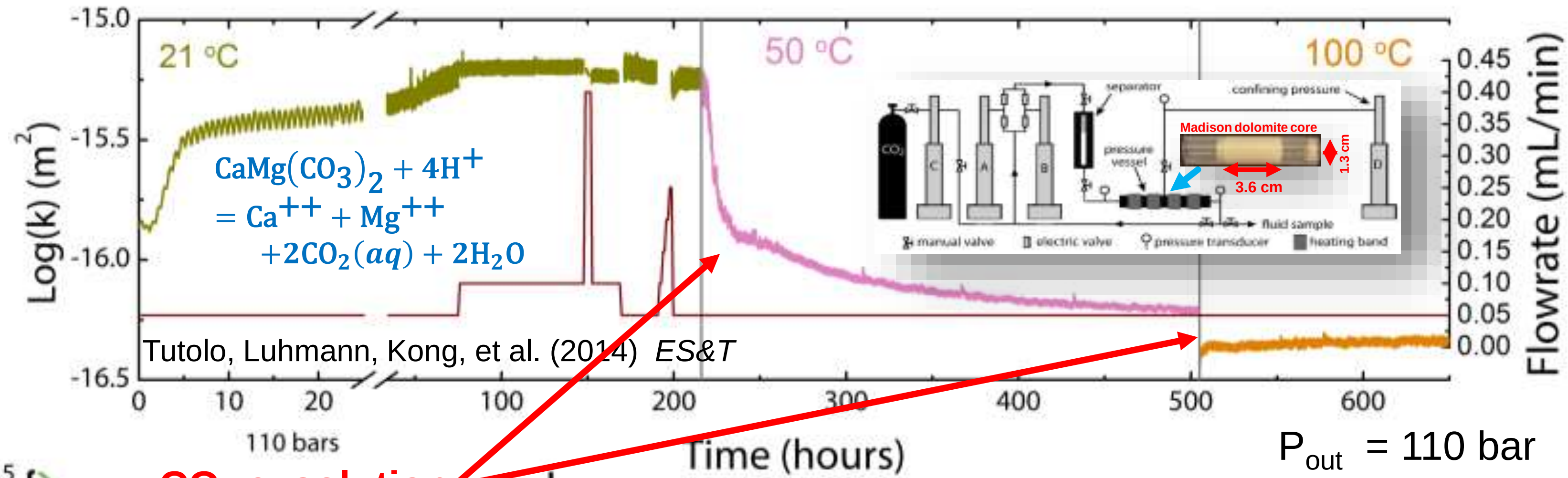
Outline

- Introduction
  Previous work and motivation
- Reactive flow-through experiments
  Experimental setup and results
- Numerical simulation
  Simulation setup and results
- Outlooks

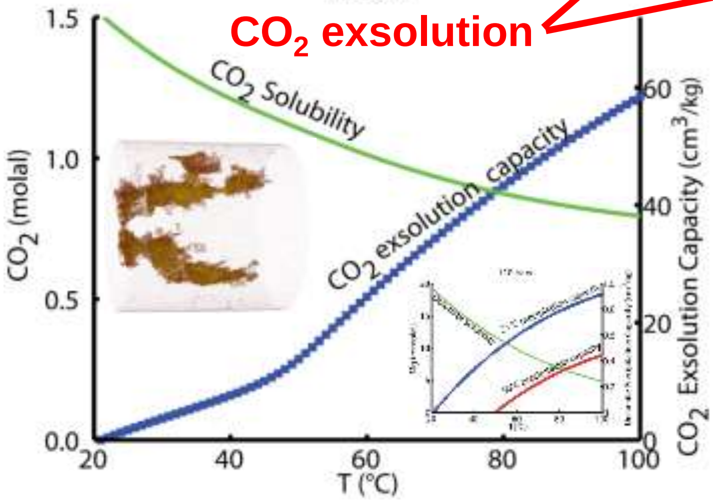
Precipitation due to temperature change



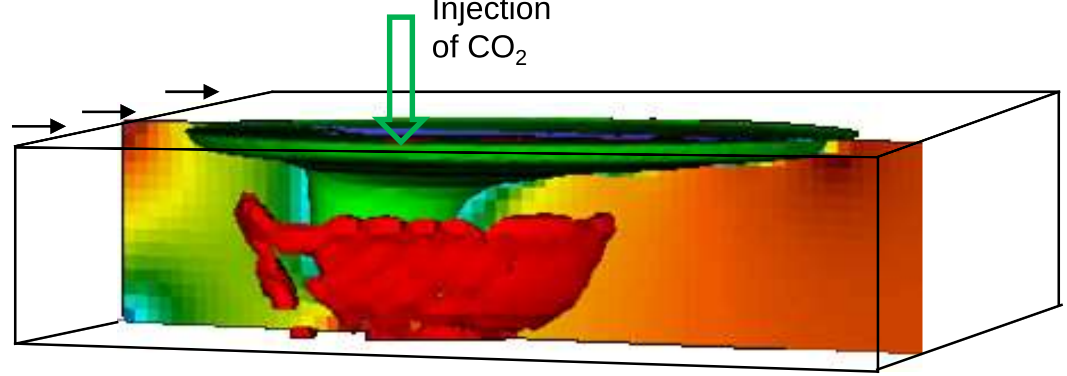
$CaMg(CO_3)_2 + 4H^+$
$= Ca^{++} + Mg^{++}$
$+2CO_2(aq) + 2H_2O$

Tutolo, Luhmann, Kong, et al. (2014) *ES&T*

CO_2 exsolution



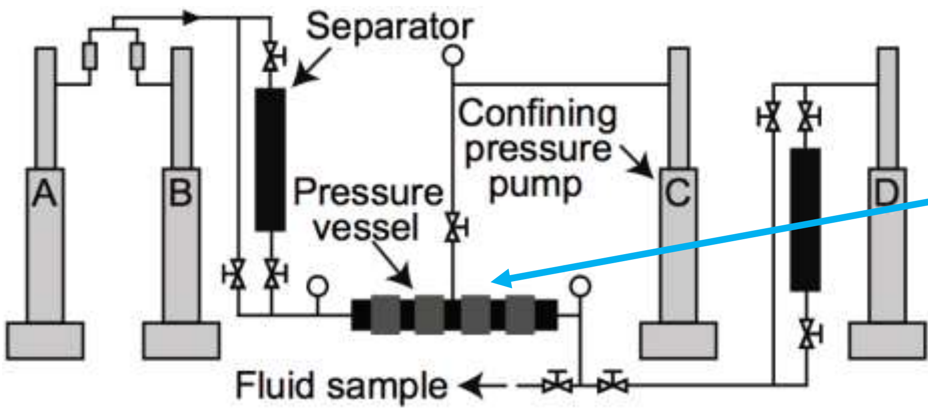
P_{out} = 110 bar
P_{conf} = 200 bar



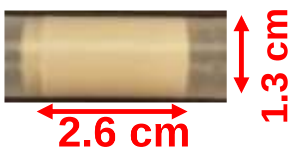
Tutolo, Kong, et al. (2014) *IJGGC*

Dolomite dissolution

Single-pass experiments: far-from equilibrium



Madison dolomite core

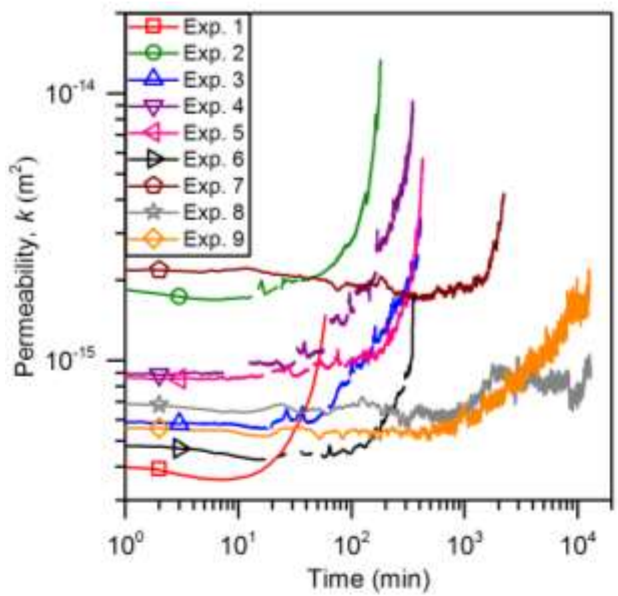


P_{conf} = 200 bar
P_{out} = 150 bar
T = 100 °C

Solution = 1 molal NaCl + 0.6 mol CO_2/kg

Luhmann, Kong, et al. (2014) Chem. Geol.

No.	Flowrate (ml/min)	Duration (min)	Pre-exp. mass (g)	Dissolved mass (g)	Initial permeability (m^2)	Final permeability (m^2)
1	1	61	8.213	0.146	3.7×10^{-16}	2.79×10^{-15}
2	0.316	180	7.921	0.114	1.31×10^{-15}	2.12×10^{-14}
3	0.1	408	8.037	0.112	5.7×10^{-16}	4.3×10^{-15}
4	0.1	350	8.012	0.094	7.7×10^{-16}	7.9×10^{-15}
5	0.1	427	8.001	0.118	7.5×10^{-16}	1.07×10^{-14}
6	0.1	367	7.833	0.12	4.5×10^{-16}	2.05×10^{-15}
7	0.0316	2,222	7.950	0.162	1.47×10^{-15}	3.8×10^{-15}
8	0.01	12,962	7.949	0.273	4.5×10^{-16}	1.32×10^{-15}
9	0.01	12,742	8.163	0.268	2.89×10^{-16}	1.32×10^{-15}



Dolomite dissolution

$Ca_{x=1.02}Mg_{y=0.98}(CO_3)_2$

Single-pass experiments: surface area v.s. porosity

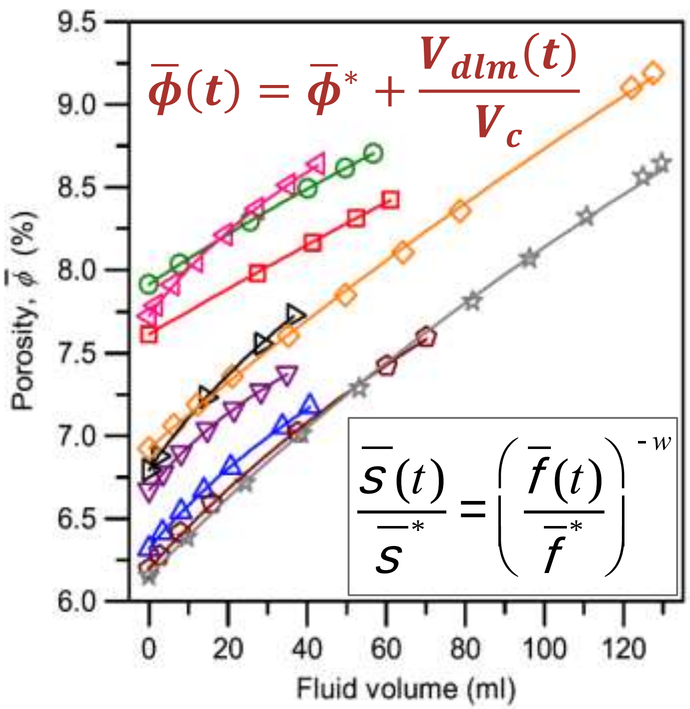
$$V_{dlm}(t) = 0.5 \upsilon Q_v \rho_f \int_{\tau=0}^{\tau=t} \left[x \Delta C_{\mathrm{Ca}}(\tau) + y \Delta C_{\mathrm{Mg}}(\tau) \right] d\tau$$

u : dolomite molar volume

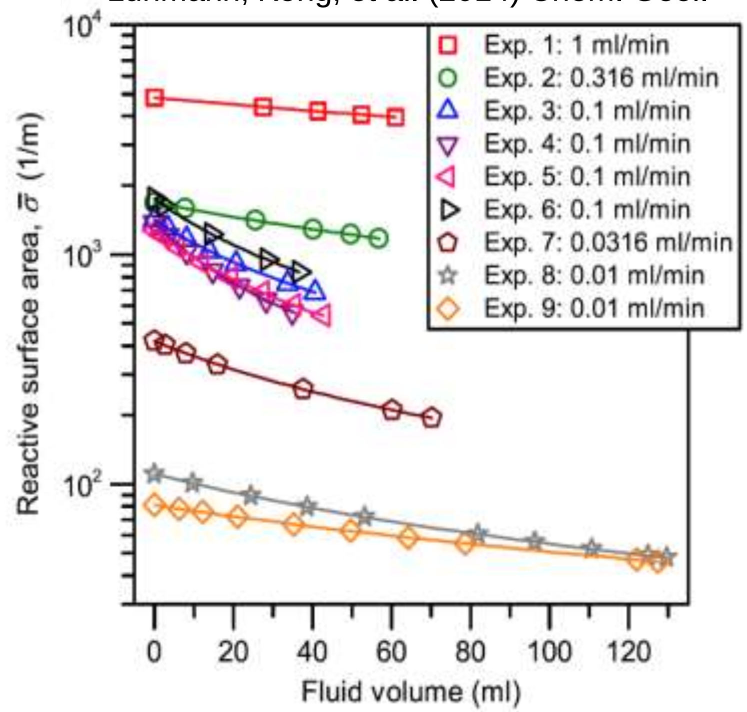
r_f : fluid density

ΔC_{Ca}: Ca concentration

ΔC_{Mg}: Mg concentration



Luhmann, Kong, et al. (2014) Chem. Geol.



$$\overline{R}(t) = \frac{1}{u\overline{f}(t)} \frac{\partial \overline{f}(t)}{\partial t} = \overline{r}\overline{s}(t)\left[1 - \frac{\overline{Q}}{K}\right]$$

$$\overline{f}(t) = \overline{f}^* \left[1 + tB(w)\right]^{1/w}$$

$$\overline{s}(t) = \frac{\overline{s}^*}{1 + tB(w)}$$

$$B(w) = wu\overline{r}\overline{s}^* \left[1 - \overline{Q}/K\right]$$

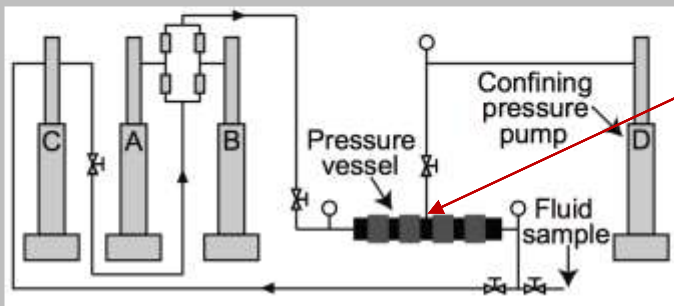
w: time-resolved dissolution efficiency.

Specific surface area determined by the BET method is at the order of 10^5 [1/m]

Initial porosity determined by X-ray Computed Tomography with a resolution of 8 um

Dissolution of K-feldspar-rich sandstone

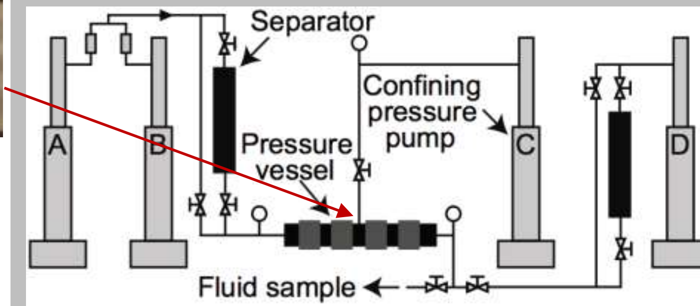
Tutolo, Luhmann, Kong, et al. (2015) *GCA*



Recycling (RC) experiment:
approaching equilibrium

Deionized water+ 0.86 mol CO_2/kg
Q=0.5 mL/min

P_{out} = 200 bar
P_{conf} = 240 bar
T = 150 °C



Single-pass (SP) experiments:
far-from equilibrium

0.94 mol NaCl/kg + 0.75 mol CO_2/kg
Q=0.1 mL/min

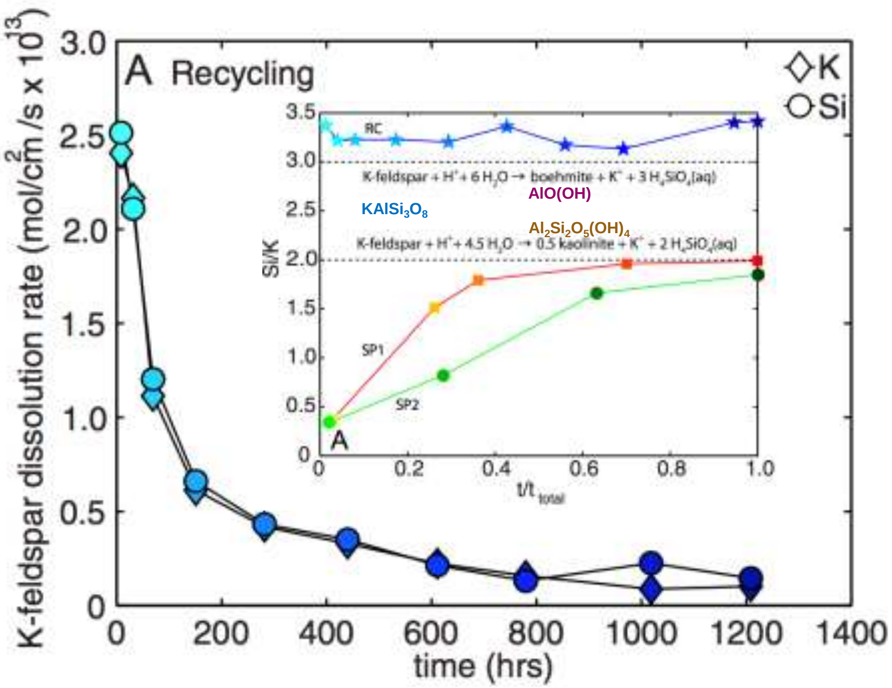
Summary of parameters measured on the three cores before and after the experiments.

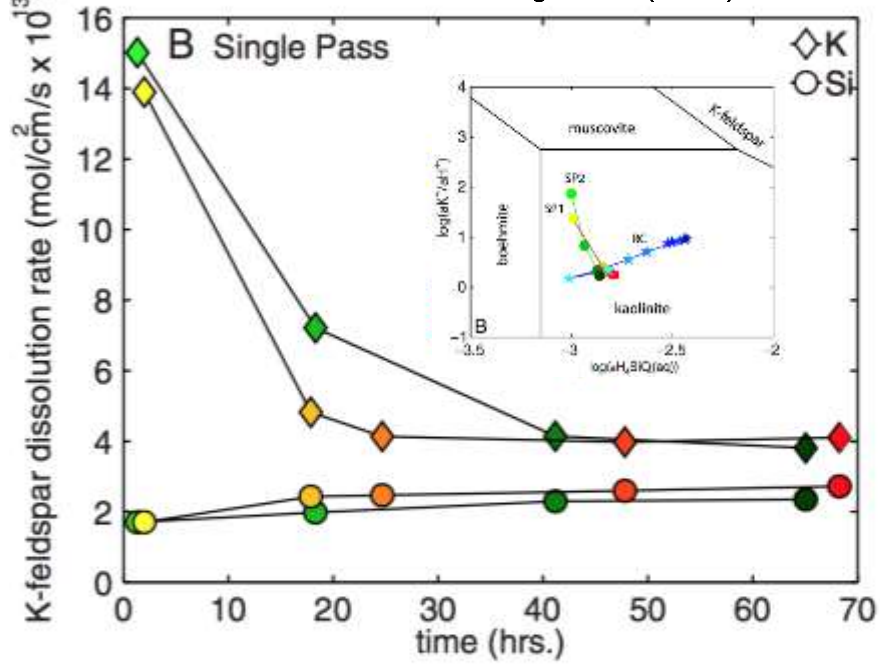
	Mass (g)			Permeability (10^{-14} m^2)		
Experiment	Pre	Post	Change (%)	Pre	Post	Change (%)
Recycling	5.632	5.509	−2.2	5.7	3.6	−37
Single-pass 1	–	–	–	2.3	1.6	−30
Single-pass 2	5.659	5.538	−2.1	4.7	3.5	−26

Reduction

Dissolution of K-feldspar-rich sandstone

Tutolo, Luhmann, Kong, et al. (2015) *GCA*

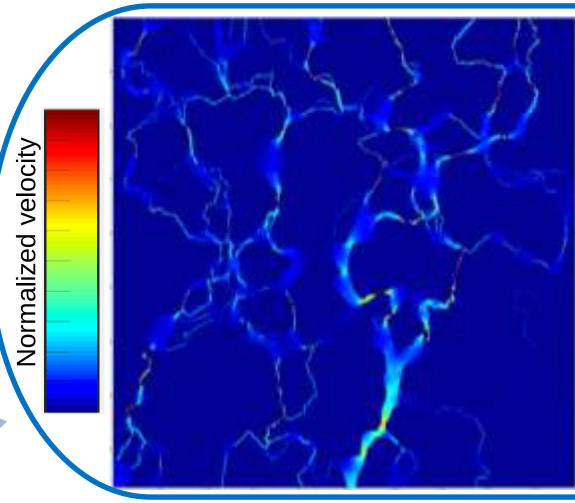




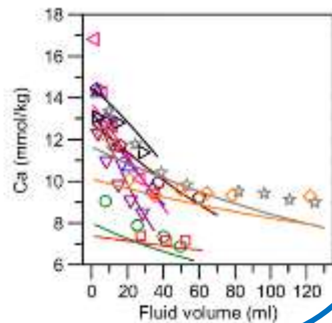
$$r = \frac{\Delta C Q}{\nu P S_{ksp}}$$

- ΔC: concentration change between inlet and outlet
- ν: stoichiometric number of the element in K-feldspar
- P: integer number of passes of fluid through the core
- S_{ksp}: surface area of K-feldspar
- Q: volumetric flow rate

Involved tasks in DESTRESS



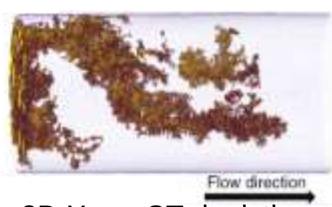
Task 4.4:
Simulation of the coupled processes



Task 6.5:
Reservoir management, operation and optimization of business

Reactive flow-through system

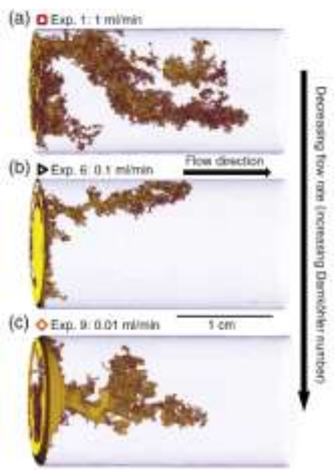
Task 6.1:
Chemical/acid stimulation performance prediction



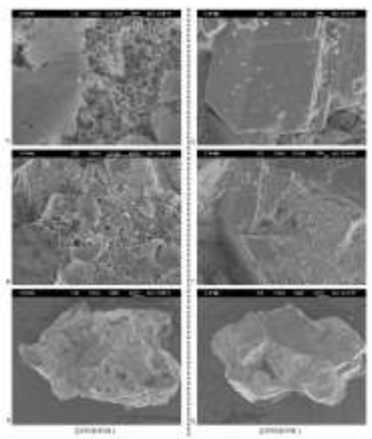
3D X-ray CT depicting mineral dissolution

Objectives

Mineral dissolution / precipitation



Luhmann, Kong, et al., 2014



Luhmann, Tutolo, Kong, et al., 2013

Property alterations

- Pore structure
- Pore size distribution
- Porosity
- Permeability
- Flow field
- Accessible surface area
- Reaction rates
- Reservoir productivity

Reactive flow-through experiments – Rock samples

35 μm-thick thin section

- SEM-BSE/EDS image
- Microscopy

Piece

- Hg porosimetry
- BET
- 3D micro-CT

Fine powder

- X-ray powder diffraction (XRD)

Depth: 981.0 m

Depth: 954.6 m

Core

- 3D micro-CT
- Porosity
- Flow-through experiments

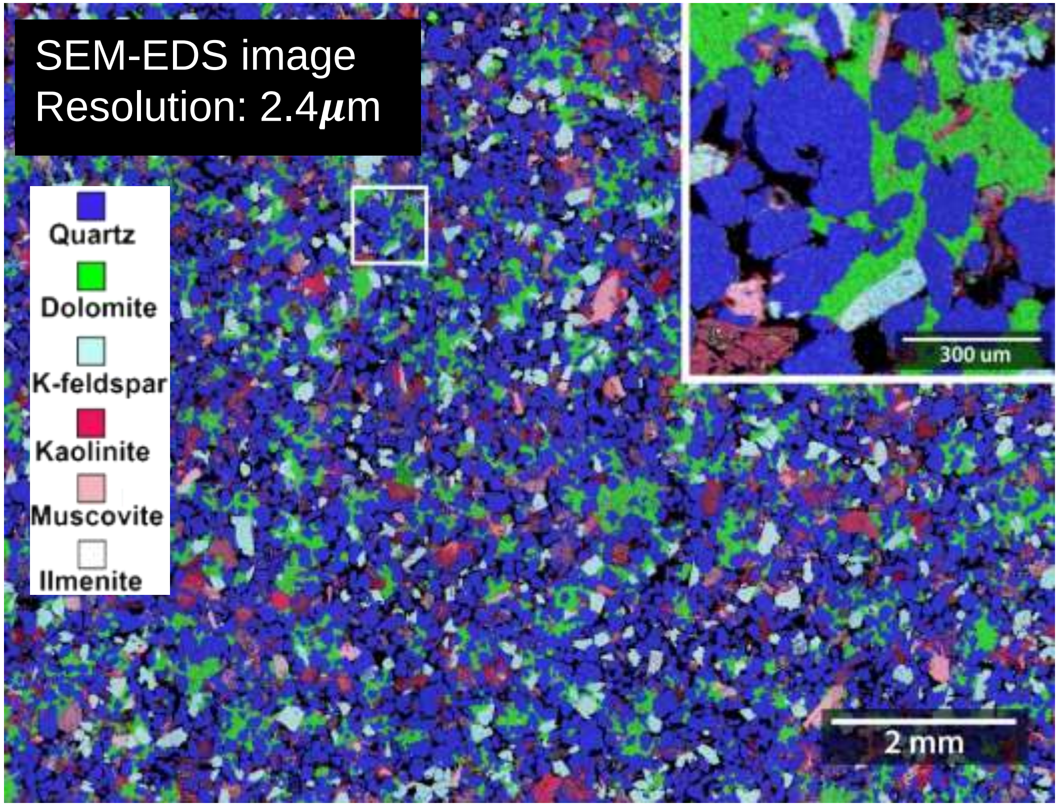
Fusion bead

- X-ray fluorescence (XRF)

BSE: Backscatter Electron Detector
EDS: Energy Dispersive X-ray Spectroscopy

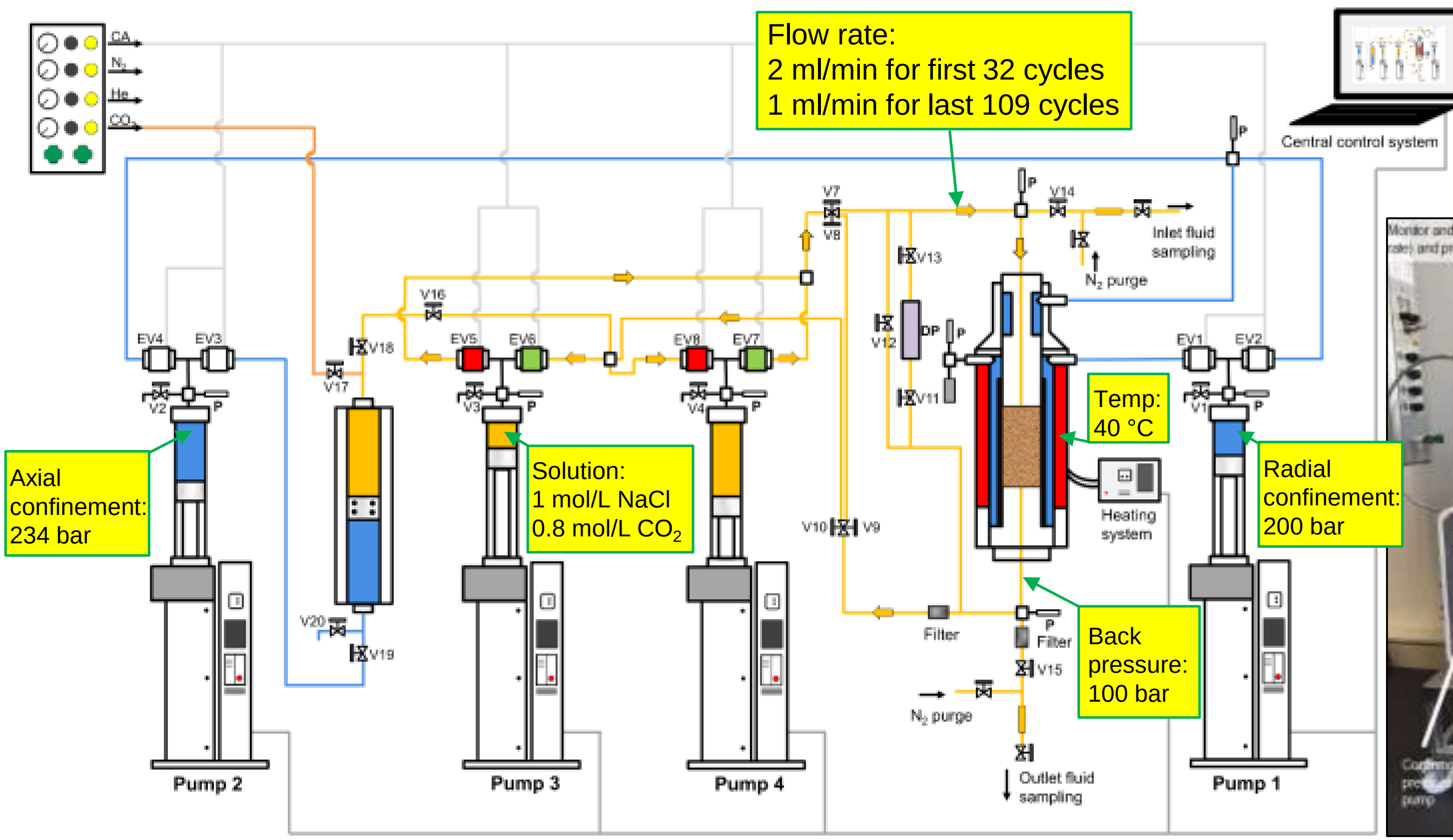
Rock composition

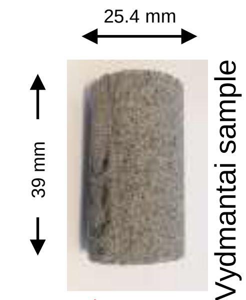
vol. %	Formula
45.53	SiO_2
12.22	$CaMg_{0.8}Fe_{0.2}(CO_3)_2$
9.93	$KAlSi_3O_8$
5.64	$Al_{1.9}Si_{2.1}O_5(OH)_4$
4.76	$K_{0.5}MgFe_{1.2}Al_{0.7}(AlSi_3O_8)(OH)_2$
0.27	$Fe_2Ti_5O_{12}$



EDS: Energy Dispersive X-ray Spectroscopy

Reactive flow-through experiments

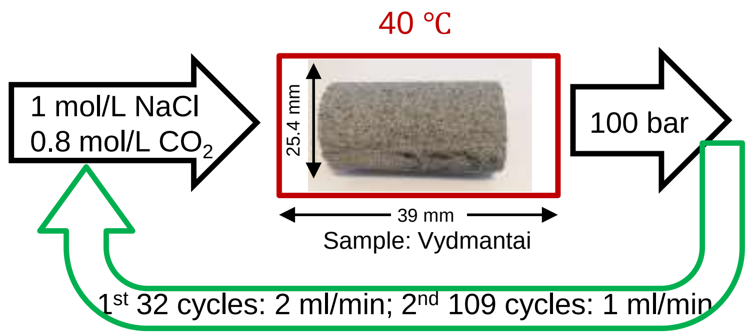


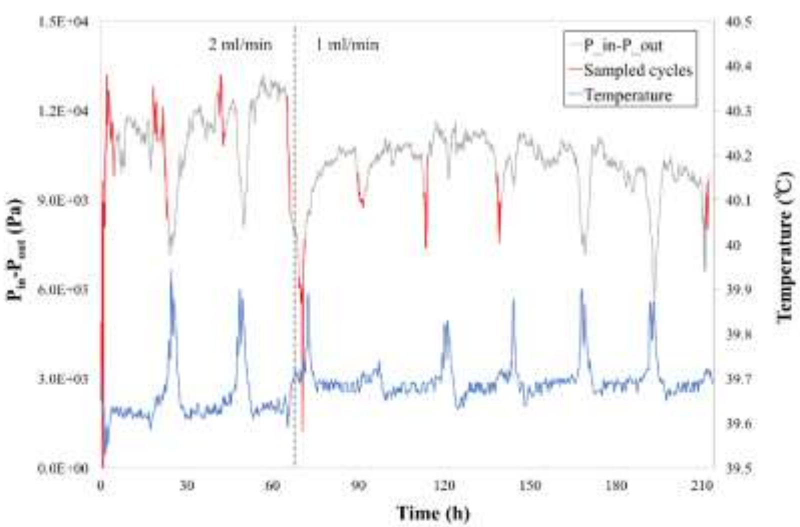


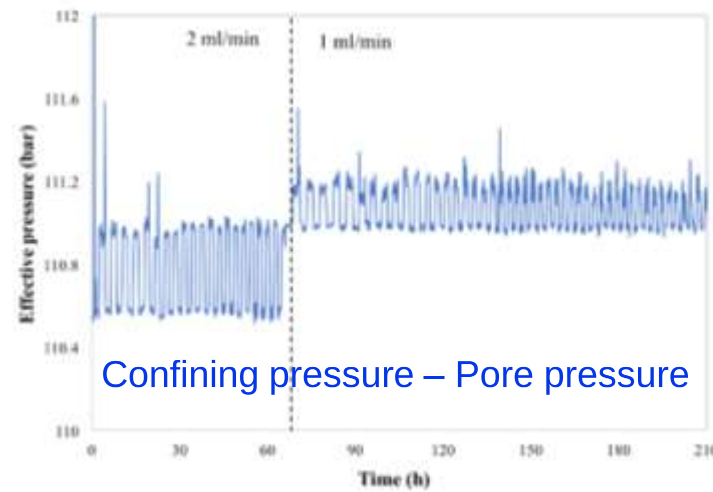
Recycling (RC) experiment: approaching equilibrium

Duration: 10 days
Total cycles: 141
Total recycled volume: 25.2L

Pressure measurements



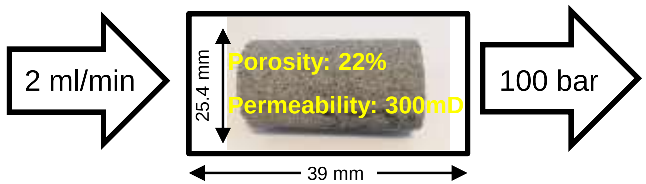




Numerical simulations with PFLOTRAN

40 ℃

Solution:
1 mol/L NaCl
0.8 mol/L CO_2



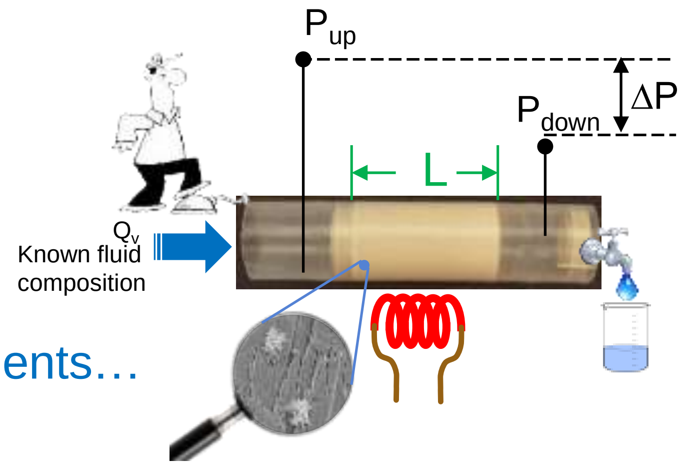
Sample: Vydmantai

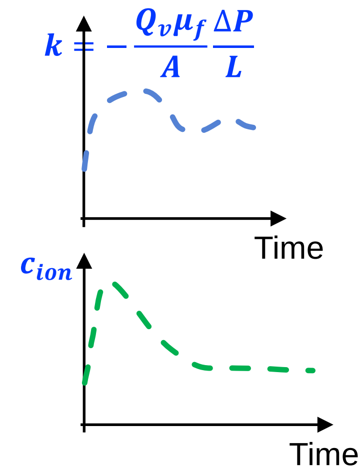
Mineral	Formula	Volume fraction (%)	Reactive surface area (cm^2/cm^3)	Rate constant ($mol/cm^2/s$) Palandri et al. (2004)
Quartz	SiO_2	46.0	1521.4	1.0×10^{-17}
Dolomite	$CaMg(CO_3)_2$	12.0	1328.7	3.0×10^{-8}
K-feldspar	$KAlSi_3O_8$	10.0	180.5	1.0×10^{-12}
Muscovite	$KAl_3Si_3O_{10}(OH)_2$	5.0	1310.0	1.0×10^{-16}
Kaolinite	$Al_2Si_2O_5(OH)_4$	5.0	26087.2	5.0×10^{-16}
Ilmenite	$FeTiO_3$	0.3	100.2	4.5×10^{-13}

Lab measurements

Outlooks

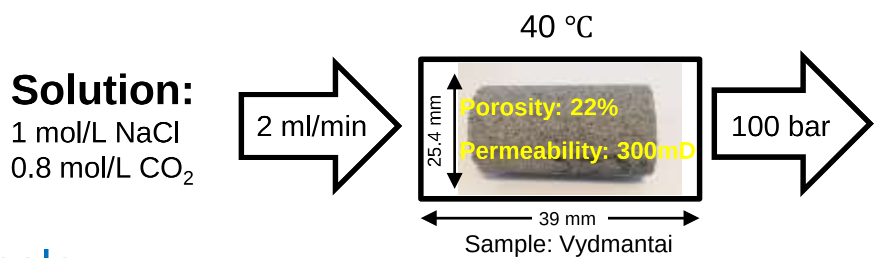
- Capability of flow-through experiments



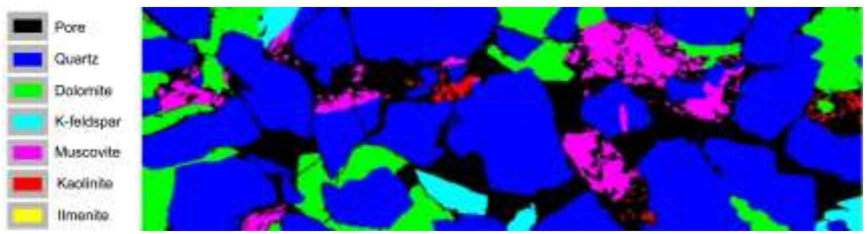


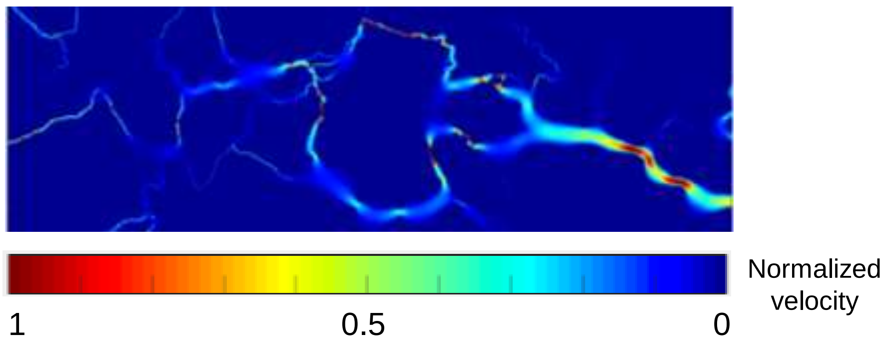
Beyond the experiments…

- Numerical challenges



2D and 3D pore-scale

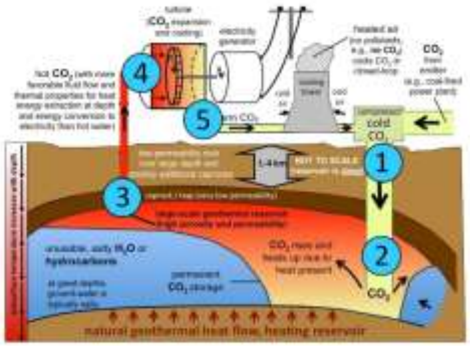




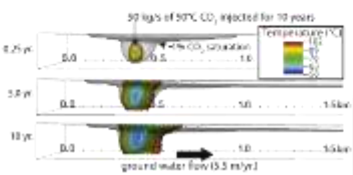
The Geothermal Energy and Geofluids Group

Numerics

CO2-plume geothermal



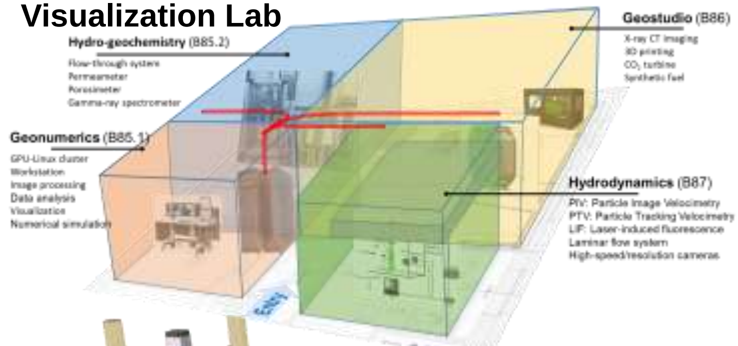
Multiphase-reactive flow



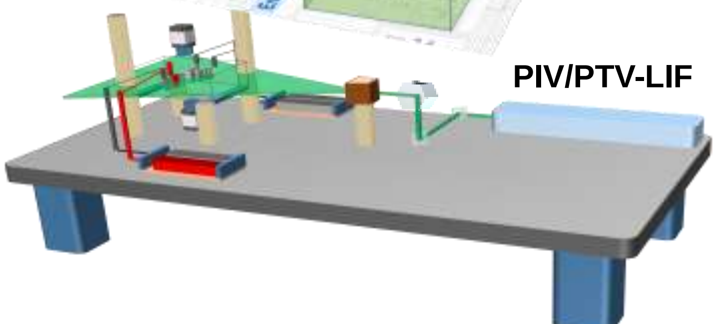
Reaktoro

Laboratory

The Geosystem Reactive Transport (GREAT) Visualization Lab

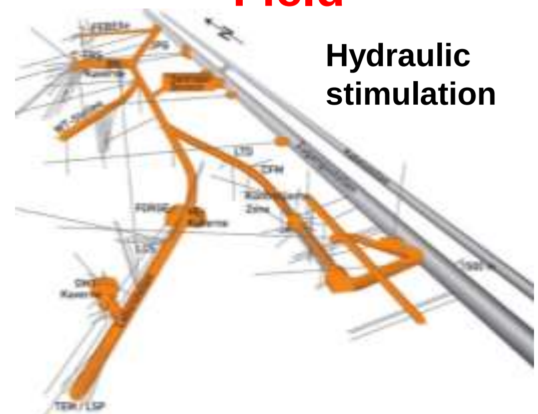


PIV/PTV-LIF

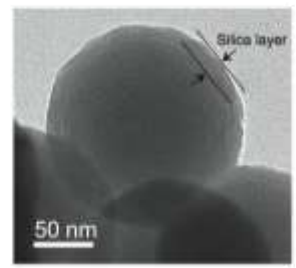


Field

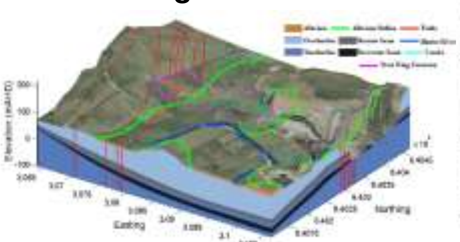
Hydraulic stimulation



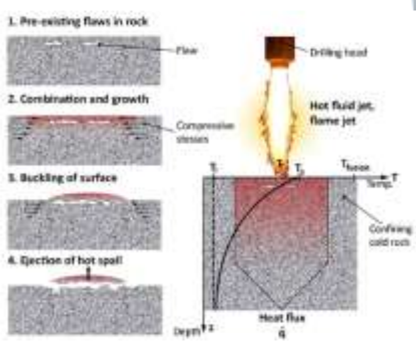
Reservoir characterization: Tracer test



Regional Groundwater Modeling



Spallation drilling



X-ray CT

Reactive system

Magnetotellurics (MT)