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Combining innovative experimental approaches and cross-scale reactive transport modelling for assessing coupled hydrogeochemical processes at interfaces in deep geological repositories for radioactive waste

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COMBINING INNOVATIVE EXPERIMENTAL APPROACHES AND CROSS-SCALE REACTIVE TRANSPORT MODELLING FOR **ASSESSING COUPLED HYDROGEOCHEMICAL PROCESSES AT INTERFACES** IN DEEP GEOLOGICAL REPOSITORIES FOR RADIOACTIVE WASTES

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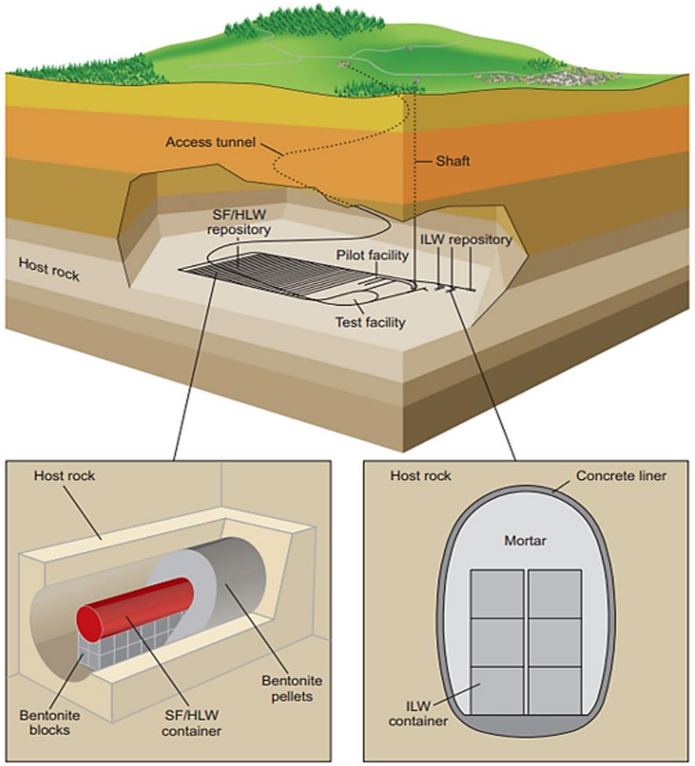
MOTIVATION

Understanding of dissolution/precipitation processes in porous media and coupling to transport properties is necessary for the assessment of the performance of engineered and natural barriers in nuclear waste repositories



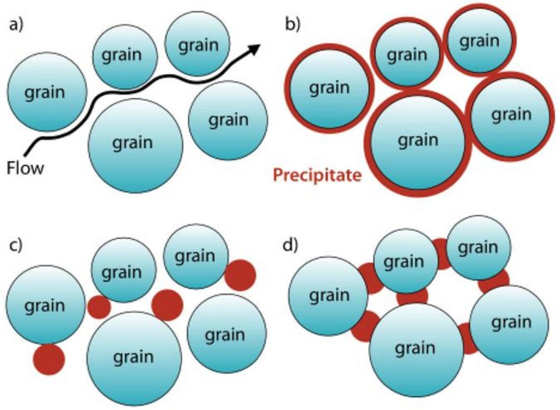
www.nagra.ch

- Reactive Transport Modelling can predict the fate of radionuclides in space & time
- Challenge: Description of the coupling between chemical processes and changes in material properties (e.g. porosity, permeability)

COUPLING OF POROSITY TO TRANSPORT PROPERTIES

The classical approach

dissolution/precipitation of minerals



porosity changes

permeability

Kozeny-Carman equation

diffusivity

Archie's law

classical approaches

Need of experimental benchmarks:

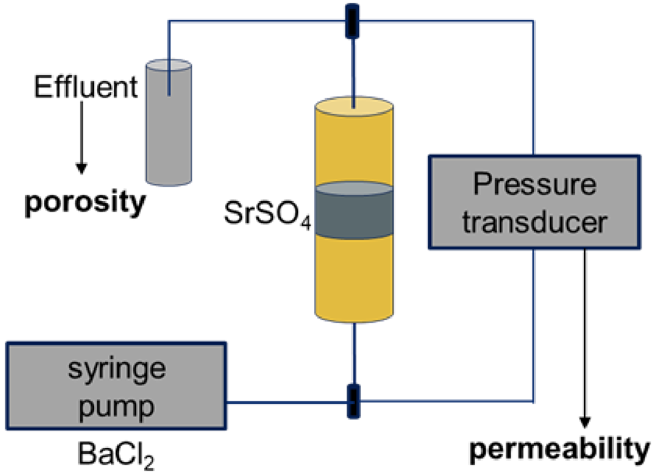
- to evaluate implementations in reactive transport models
- to build confidence in the predictions of reactive transport models

EXPERIMENTAL DESIGN & CONCEPT

Mineral precipitation and consequences on permeability

Investigation of the **effect of supersaturation** on precipitation processes in porous media

Flow through column



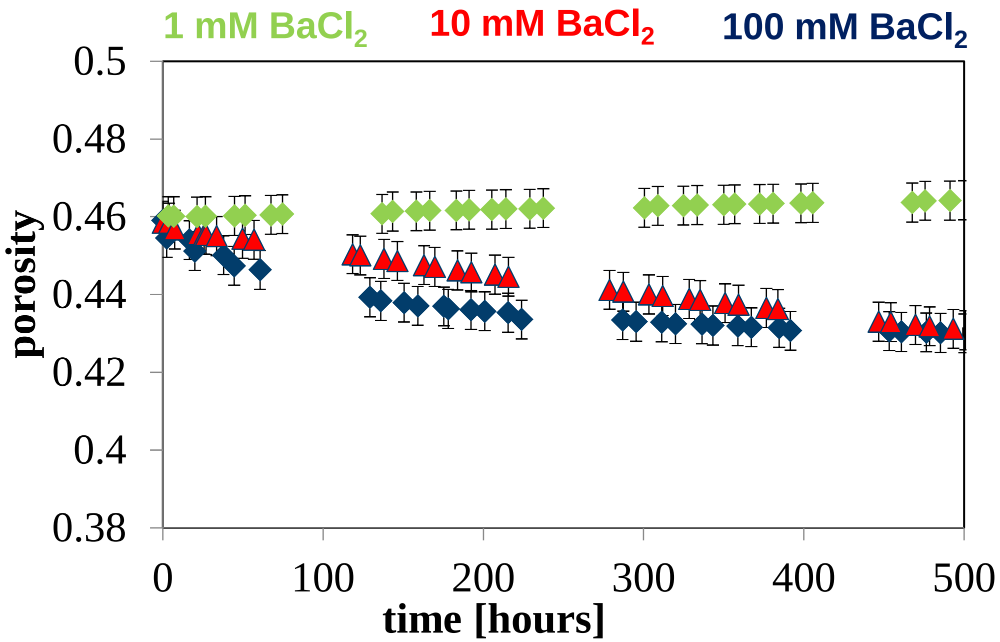
$$SrSO_{4(s)} + Ba^{2+}_{(aq)} \rightarrow Sr^{2+}_{(aq)} + BaSO_{4(s)}$$

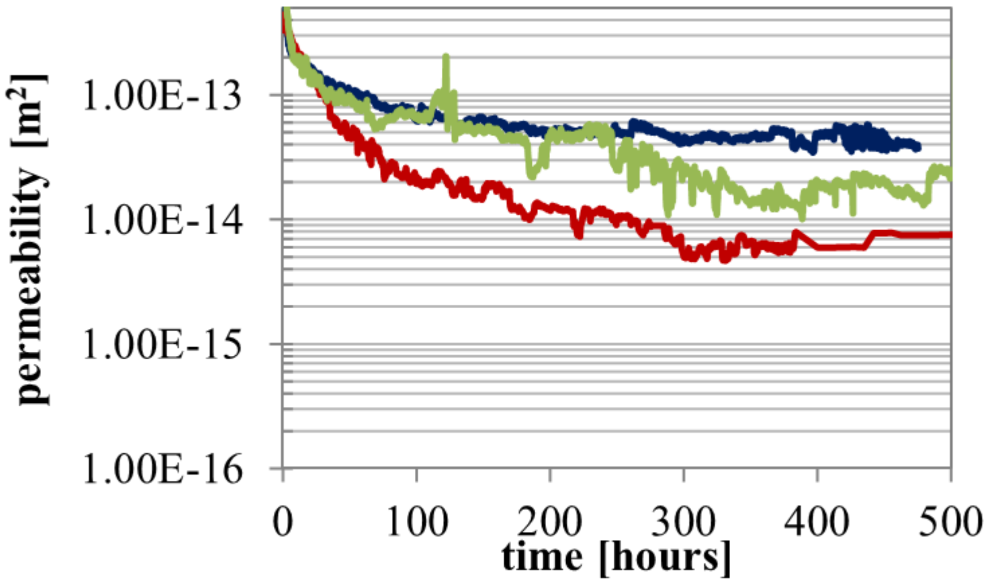
molar volume $BaSO_4$ > molar volume $SrSO_4$
→ **porosity & permeability decrease**
3 experiments: 100 mM Ba (exp. 1),
10 mM Ba (exp. 2), 1 mM Ba (exp. 3)

Nuclear Magnetic Resonance Imaging (MRI)

Assessing porosity and pore connectivity changes using high field MRI (B_0 = 4.7T)

POROSITY - PERMEABILITY CHANGES



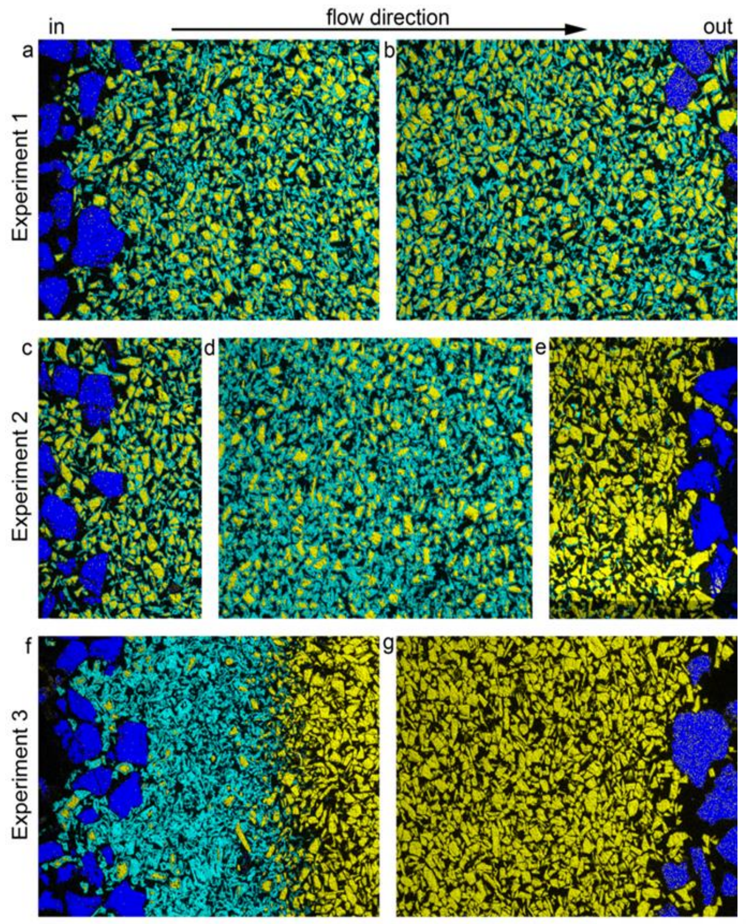


- In experiment 1 and 2, similar porosity, but permeability differs by 1 order of magnitude
- In experiment 3, porosity remains constant, but permeability decreases by 2 orders of magnitude

➢ Small changes in porosity cause significant changes in permeability

MICROSTRUCTURAL CHANGES

EDX elemental map of the reacted celestine zone with secondary barite overgrowth



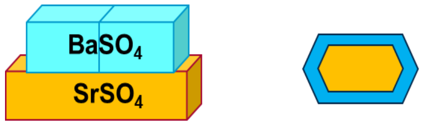
SI 3.97 - 3.94
100 mM $BaCl_2$

SI 3.82 - 0.344
10 mM $BaCl_2$

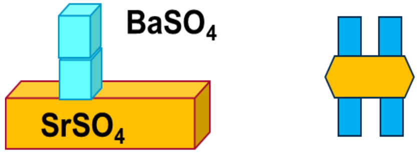
SI 3.35 - 0.277
1 mM $BaCl_2$

Saturation Index

- **100 mM Ba:** uniform $BaSO_4$ distribution



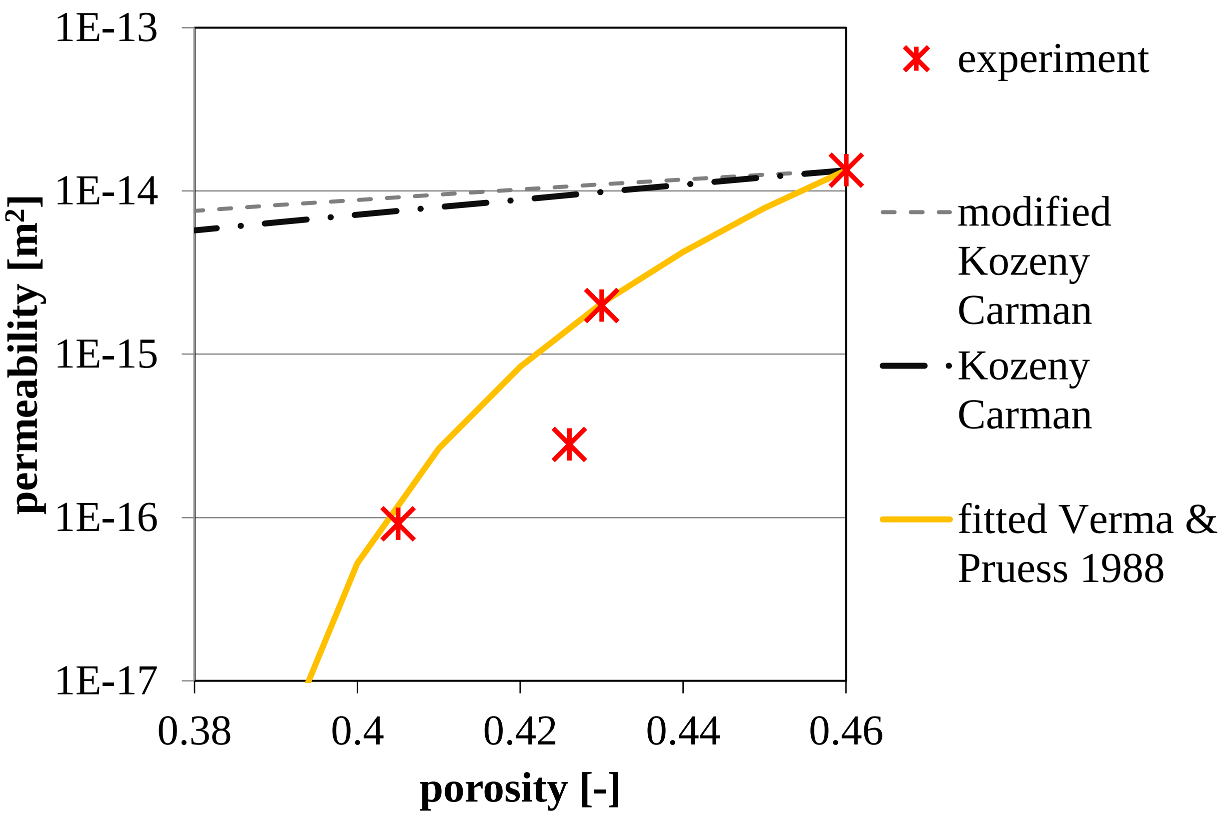
- **10 mM Ba:** non-uniform $BaSO_4$ distribution
- **1 mM Ba:** new pore architecture



➢ **Mineral growth mechanism influences the change in pore architecture and consequently permeability**

Poonoosamy *et al* 2020, Geochim Cosmochim Acta 270, 43–60.

EVALUATION OF POROSITY – PERMEABILITY COUPLING



- Simulations using modified Kozeny-Carman equation failed to predict permeability changes

$$k = k_0 \left(\frac{Ø}{Ø_0}\right)^3 \left(\frac{1-Ø_0}{1-Ø}\right)^2$$

- Alternative porosity - permeability relation (Verma & Pruess 1988) :

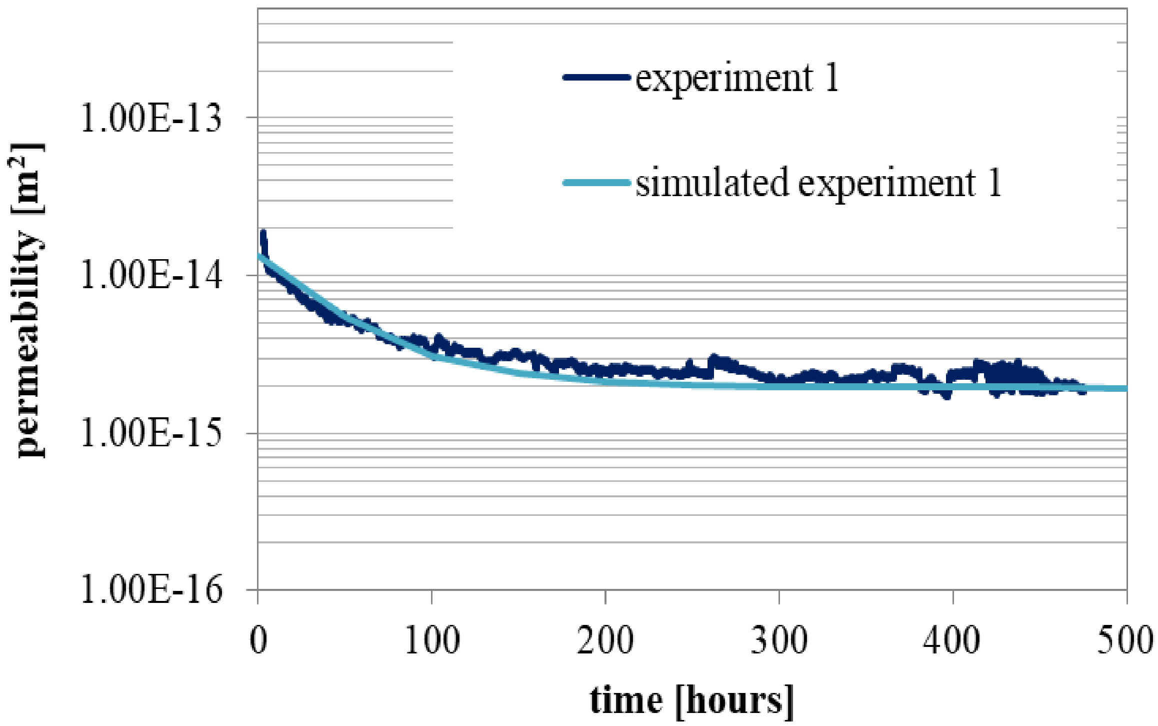
$$k = k_0 \left(\frac{Ø - Ø_{critical}}{Ø_0 - Ø_{critical}}\right)^n$$

k_0 initial permeability; $Ø_0$ initial porosity
$Ø_{critical}$ critical porosity
$1 \leq n \leq 6$, for $0.8\,Ø_0 \leq Ø_{critical} \leq 0.9Ø_0$
$Ø_{critical}$ and n set to 0.38 and 4 to match experimental data

Poonoosamy *et al* 2020, Geochim Cosmochim Acta 270, 43–60.

EVALUATION OF POROSITY – PERMEABILITY COUPLING

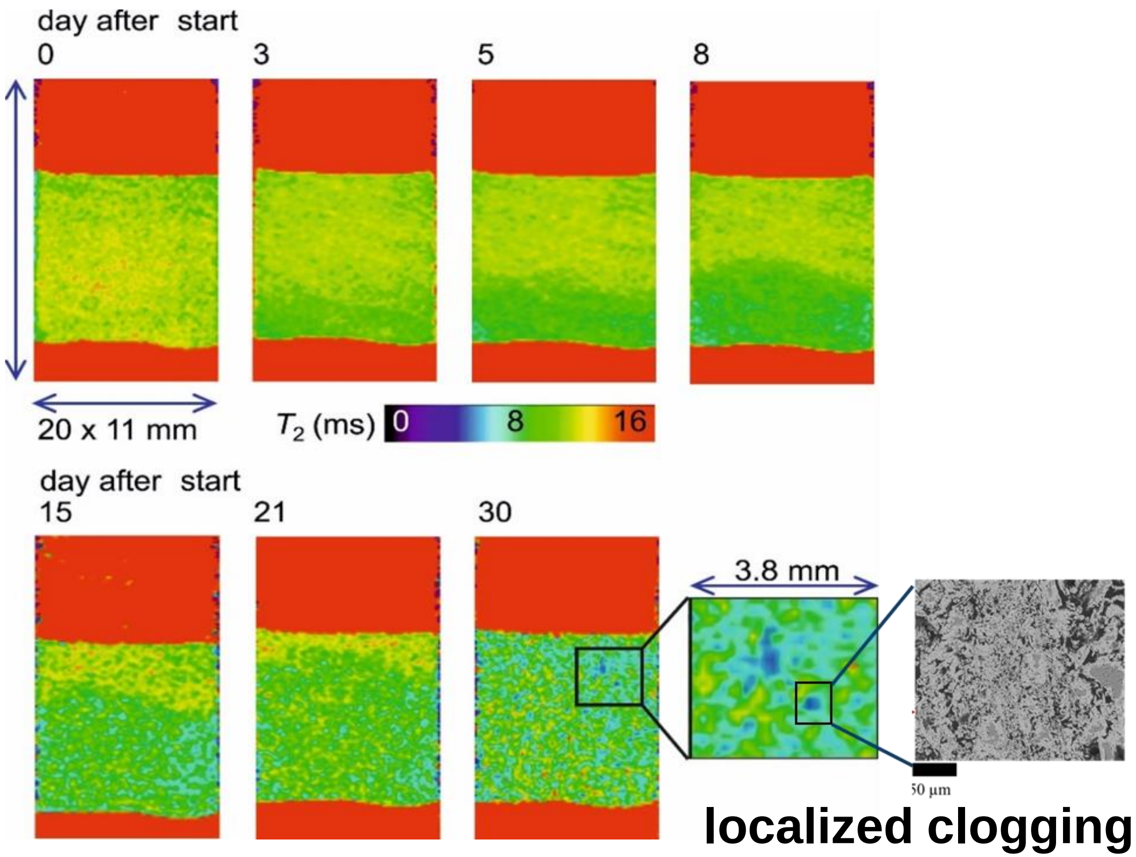
Reactive transport modelling using Open-GeoSys-GEMs



- Simulations using modified Kozeny-Carman equation failed to predict permeability changes
- More sophisticated porosity/permeability relationship involving a critical porosity required to describe permeability changes due to precipitation processes
- **What is the physical meaning of critical porosity?**

VISUALISATION OF POROSITY CHANGES BY MRI

Temporal evolution of porosity



localized clogging

- 2D relaxometry measurements show a decrease in porosity with time

- **Clogging zones**, need to be upscaled

→ **so-called critical porosity**

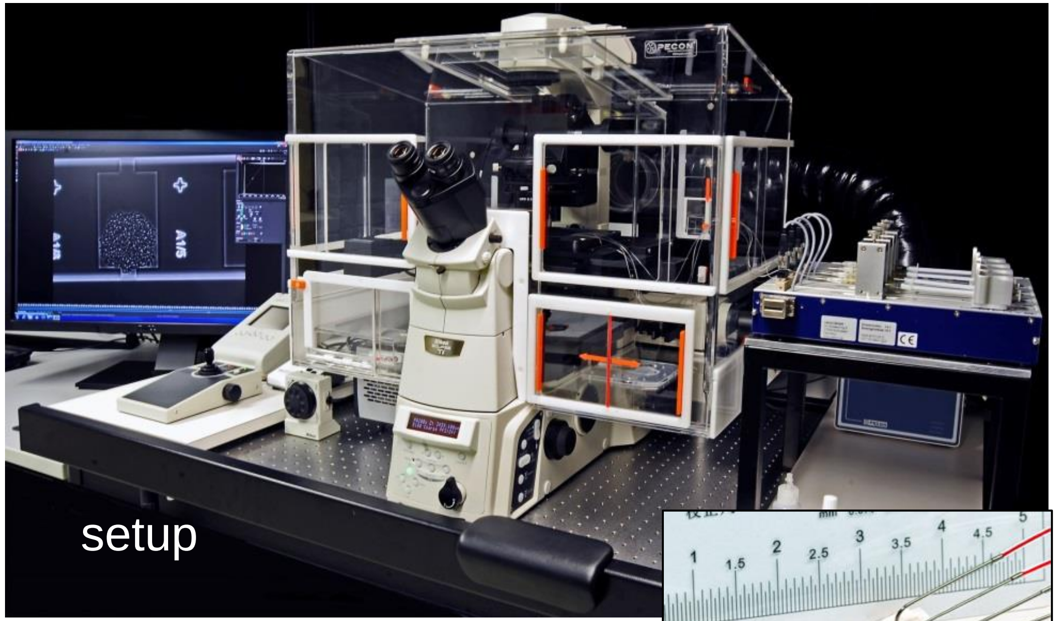
Can the concept behind the Verma and Pruess relationship be applied for a diffusive system?

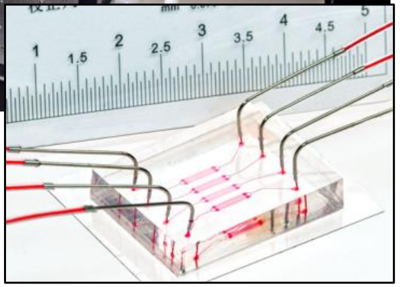
Poonoosamy *et al* 2020, Minerals 10, 226.

EXPERIMENTAL DESIGN & CONCEPT

Mineral precipitation and consequences on diffusivity

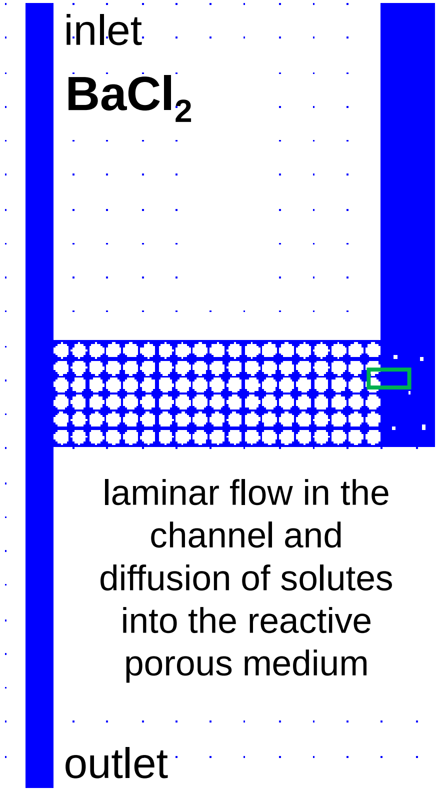
Microfluidic experiment



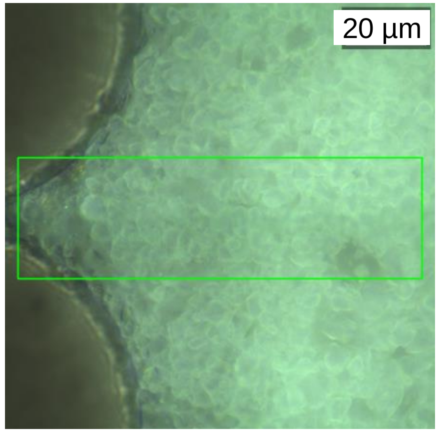


microreactor

Chip design



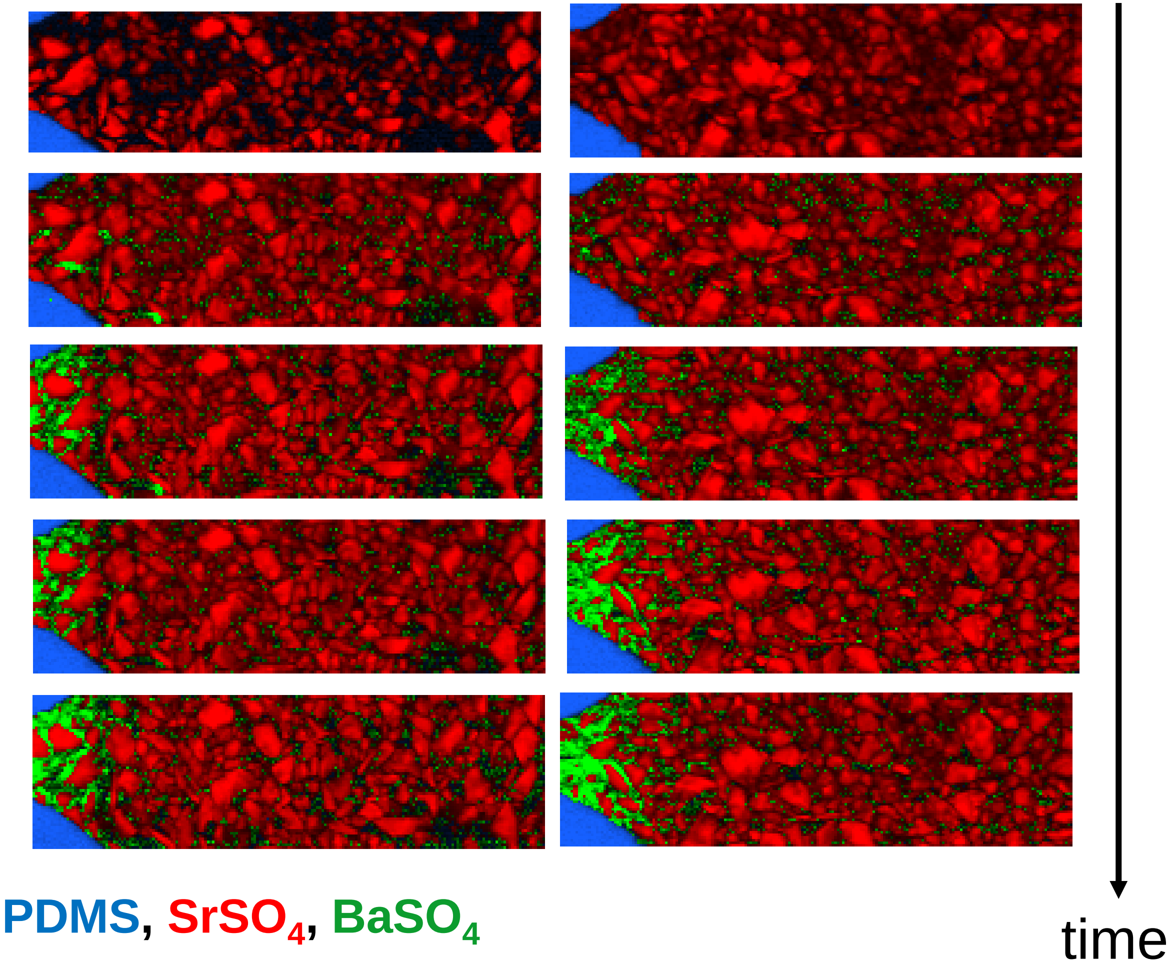
$$SrSO_{4(s)} + Ba^{2+}_{(aq)} \rightarrow Sr^{2+}_{(aq)} + BaSO_{4(s)}$$



reservoir of 800 µm × 450 µm × 1 µm filled with **$SrSO_4$** crystals of size 4-9 µm

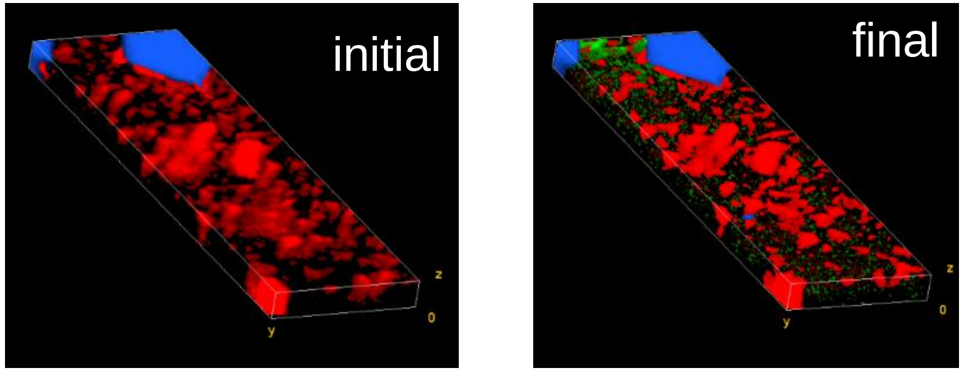
CHANGES IN PORE ARCHITECTURE

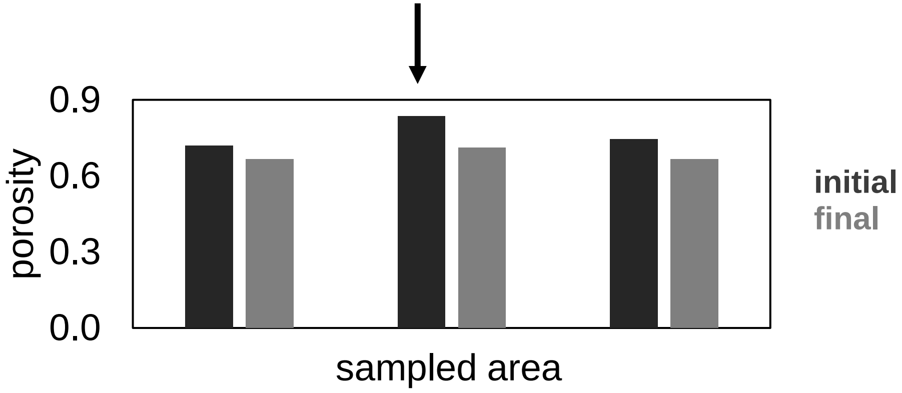
Raman imaging



PDMS, $SrSO_4$, $BaSO_4$

Raman tomographs

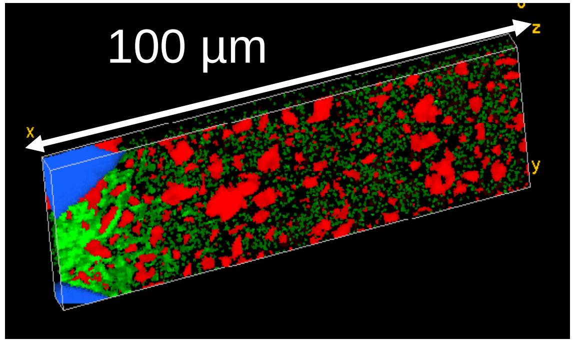




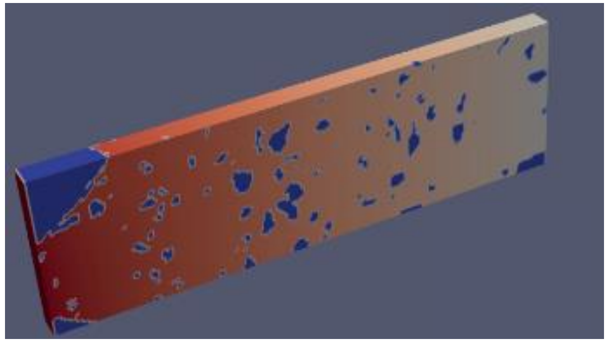
EFFECTIVE DIFFUSION COEFFICIENT

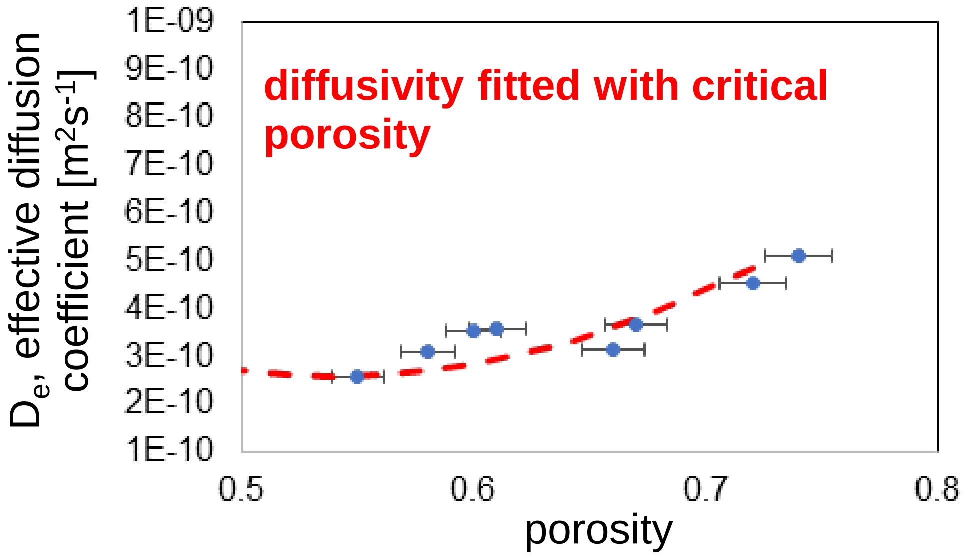
Pore scale modelling using Lattice Boltzmann method (ongoing)

Experiment



Pore scale modelling of tracer diffusion across the reacted porous medium to derive the effective diffusion coefficient D_e





- Derivation of a porosity – diffusivity relationship
- ★ Check poster of Mara Lönartz (11th November, 14:20h) for porosity – diffusivity relationships

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CONCLUSIONS

Experimental investigations show limitations of classical approaches to model hydrogeochemical processes associated with porosity decrease in porous media

Investigations at the pore scale are necessary to understand and quantify the effects of crystallization mechanisms on porosity changes

There is a need to develop process-based predictive models and mathematical relationships that account for small scale heterogeneity and integration into larger scale analysis (upscaling)

THANK YOU FOR YOUR KIND ATTENTION …

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