The influence of sedimentary heterogeneity on the diffusion of radionuclides in the sandy facies of Opalinus Clay at the geological scale

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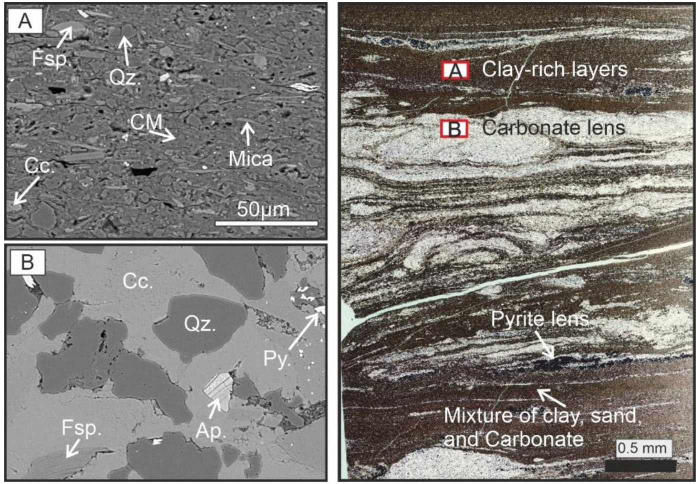
Motivation: Meaningful estimation of radionuclide migration in SF-OPA above the core scale requires a comprehensive study on the influence of sedimentary layers

Sandy facies of Opalinus Clay (SF-OPA)



Bollermann et al. (2022)

Heterogeneous structures at different scales:

- Pore network variability at the pore scale (nm to µm)
- Lamination, diagenetic minerals (cm to dm)
- Sedimentary layers with various subfacies (m and above)

→ Small-scale structural heterogeneities cause the differences in sedimentary and diagenetic facies in SF-OPA at larger scales

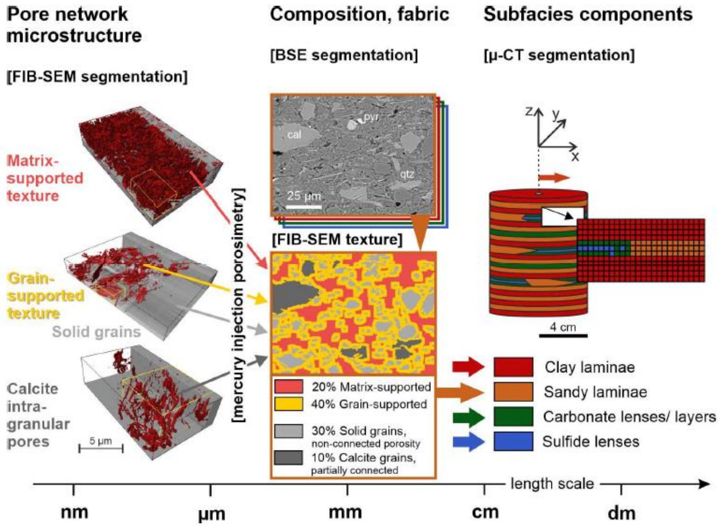
→ Small-scale heterogeneities can affect larger scales radionuclide migration in SF-OPA

⬇

- Improve understanding of heterogeneous diffusion in SF-OPA
- Enhance predictability of radionuclide migration in low-permeable host rocks.

Method: Field scale diffusive transport simulation with the quantified diffusivity based on digital rock physics and upscaling workflow

(1) Effective diffusivity quantification: Three steps digital rock physics in upscaling workflow



$$D_e = \frac{J \times L}{(C_{in} - C_{out})}$$

J is the mass flux through the domain under steady state

Upscaling workflow

Effective diffusivity: D_e

1. Pore network variability (nm~µm).
2. Compositional heterogeneity (nm~µm).
3. Subfacies variability (cm~dm).

(2) Field scale diffusive transport:

The general mass balance equation for the solute diffusion:

$$\frac{\partial C_{tot}}{\partial t} = \nabla \cdot (\mathbf{D_e} \cdot \nabla C), C_{tot} = (\emptyset + \rho_{bd} \cdot K_d) \cdot C.$$

where C (mol m^{-3}) is the concentration of aqueous solute and De (m^2 s^{-1}) is the effective diffusion coefficient tensor.

OpenGeoSys

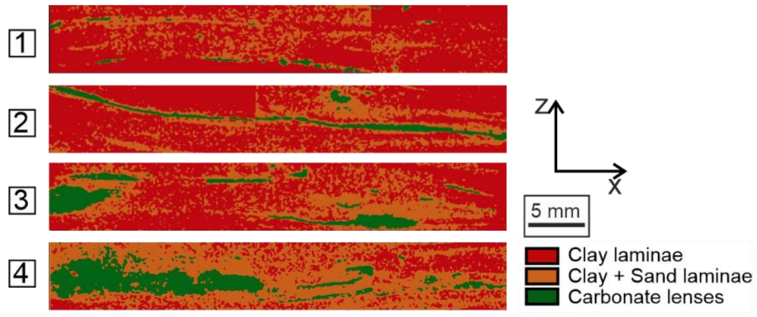
Lu et al. *Comput. Geosci*. 2022. 163

Bollermann et al. *Chem. Geol*. 2022. (In Review)

Yuan and Fischer. *Trans. Porous. Med.* 2021. 138, 245-264

Model setup: An endmember study based on specific sediment compositions

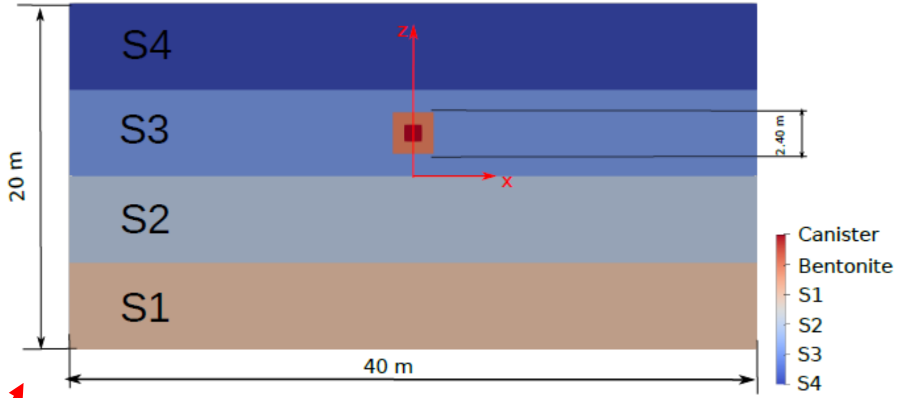
Four structures of SF-OPA with different sub-facies compositions.



Yuan and Fischer (2022)

Structure	S1	S2	S3	S4
Clay Laminae composition (%)	78.50	67.60	53.80	25.30
Clay+Sand composition (%)	20.00	26.90	36.40	55.00
Carbonate lenses composition (%)	1.50	5.50	9.80	19.70
Effective diffusivity in x direction ($\times 10^{-11} m^2/s$)	2.41	1.77	1.28	0.61
Effective diffusivity in z direction ($\times 10^{-11} m^2/s$)	0.77	0.43	0.43	0.18

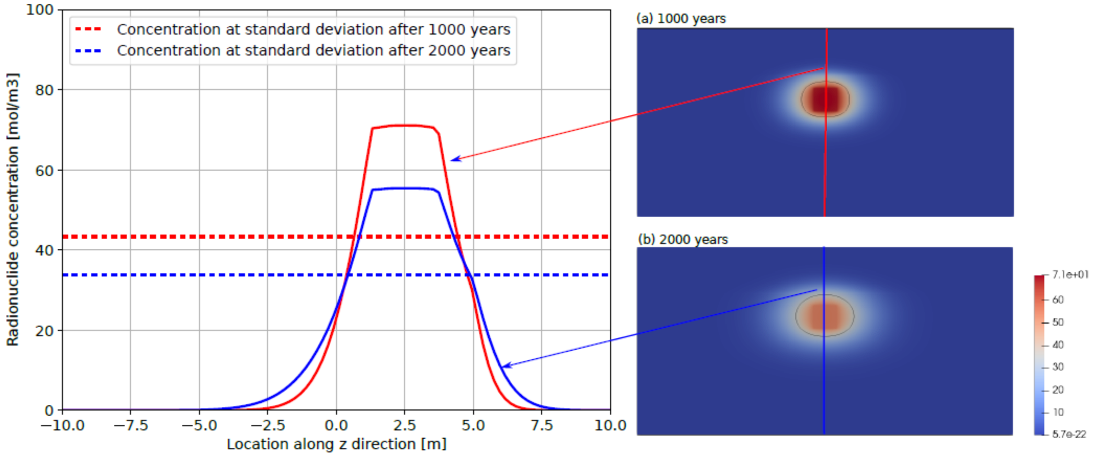
2D synthetic SF-OPA formation:

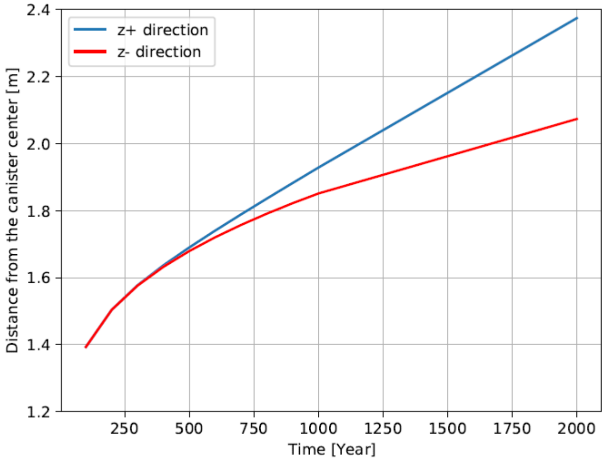


The four different structures in the SF-OPA formation:
With a low concentration of carbonate lenses (S1), lamination (S2), and a high concentration of carbonate lenses (S3 and S4).

- ✓ The initial concentration of the sodium (Na^+) tracer is 0.3 mol/L.
- ✓ All the boundaries are no-flux conditions.
- ✓ The total simulation time is 2000 years.

Results: Effects of specific sedimentary stratification on the diffusion front





(1) Sodium (Na^+) concentration profiles along z axis after 1000 years' (red line) and 2000 years' (blue line) diffusion. The radionuclide source is located in the position: z = 2.5 m.

- The highest concentration in the canister decreases from 0.71 to 0.55 mol/L.
- The diffusion front (61% of the maximum concentration) is located within S3 layer after 2000 years' diffusion.

(2) The displacements of diffusion front along z+ and z− direction versus diffusion time.

- The movement of the diffusion front along z+ direction is faster than that along z- direction, because the diffusivity of layer S4 is lower than it of layer S2.

Conclusion and outlook

1. A 2D structural model has been established to study the heterogeneous diffusion of radionuclide base on the effective diffusivity values upscaled from the smaller scales

2. In the field scale, the low-diffusivity neighboring layer around the canister layer will have better retardation effect for the radionuclide migration

Next step:

The sensitivity analysis will be focused on the impact of bedding angle and location of the canister on the spatio-temporal evolution of radionuclide concentrations and diffusion front movement.

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Thanks for your attention!

Reference

1. Catherine Feng Ji Yu. "Comparative study of convective and diffusive transport phenomena within the opalinus clay of Mont Terri." PhD diss., Aix-Marseille, 2017.
2. Grathwohl, P.: Diffusion in Natural Porous Media: Contaminant Transport, Sorption/Desorption and Dissolution Kinetics, 207, 1998.
3. Yuan, T. and Fischer, C.: Effective Diffusivity Prediction of Radionuclides in Clay Formations Using an Integrated Upscaling Workflow, Transport in Porous Media, 138, 245–264, 2021.
4. Yuan, T. and Fischer, C.: The influence of sedimentary and diagenetic heterogeneity on the radionuclide diffusion in the sandy facies of the Opalinus Clay at the core scale, Applied Geochemistry, In Review, 2022.
5. Lu, Renchao, et al. "A new operator-splitting finite element scheme for reactive transport modeling in saturated porous media." Computers & Geosciences 163 (2022): 105106.
6. Bollermann, T., Yuan, T., Kulenkampff, J., Stumpf, T., and Fischer, C.: Pore network and flow field analysis towards improved predictability of diffusive transport in argillaceous host rocks, Chemical Geology, In Review, 2022.