PAUL SCHERRER INSTITUT

Sergey V. Churakov :: Laboratory for Waste Management :: Paul Scherrer Institut

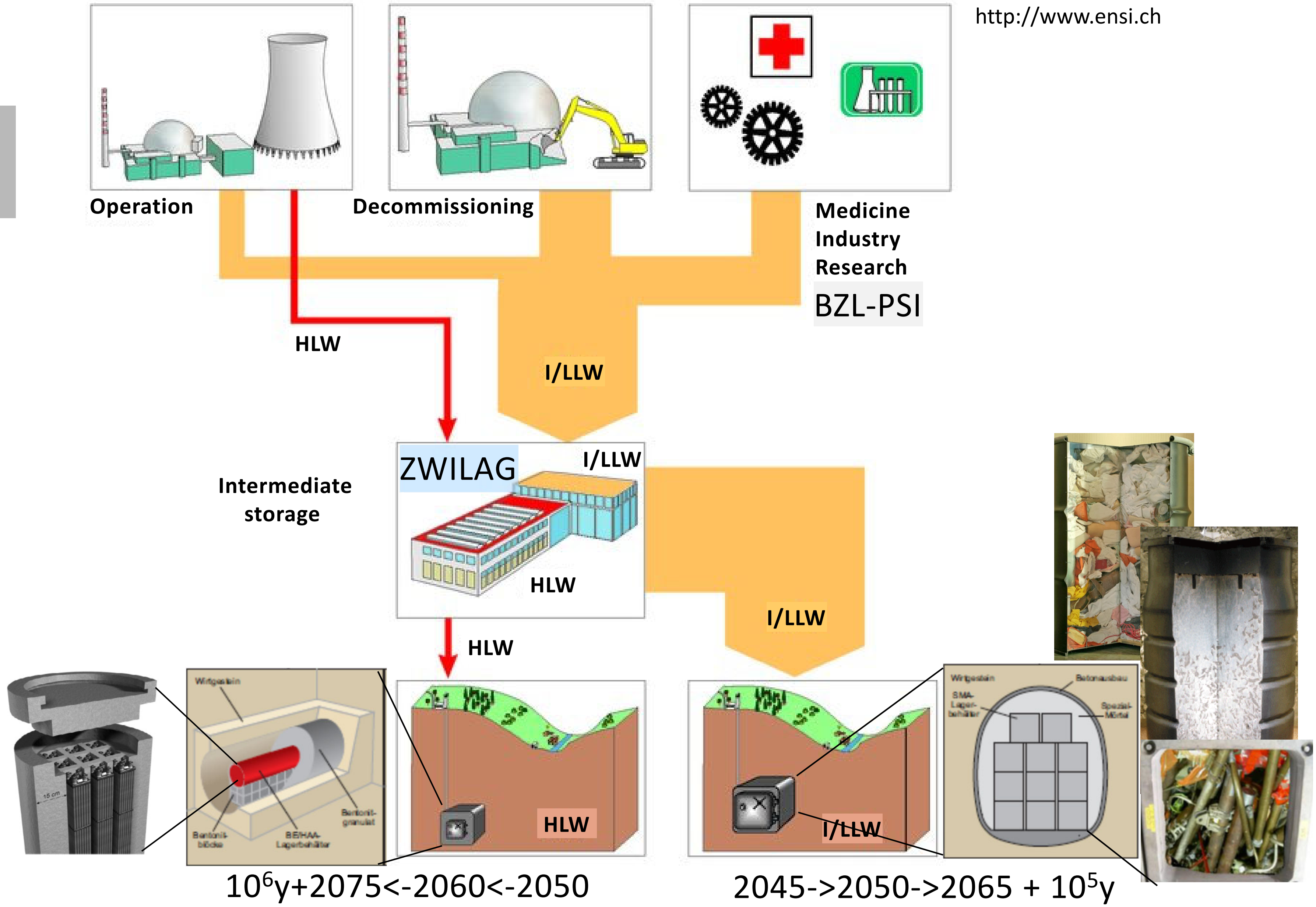
Fundamental and applied research on geochemistry of nuclear waste disposal in Switzerland

16th September, 2021 RWM RSO annual conference

Outline

- Radioactive waste in Switzerland
- Sectoral plan for Swiss Geological disposal
- Current focus of site selection process
- Laboratory for waste management (LES)
- Research on repository geochemistry
- Hot topics and needs for further research

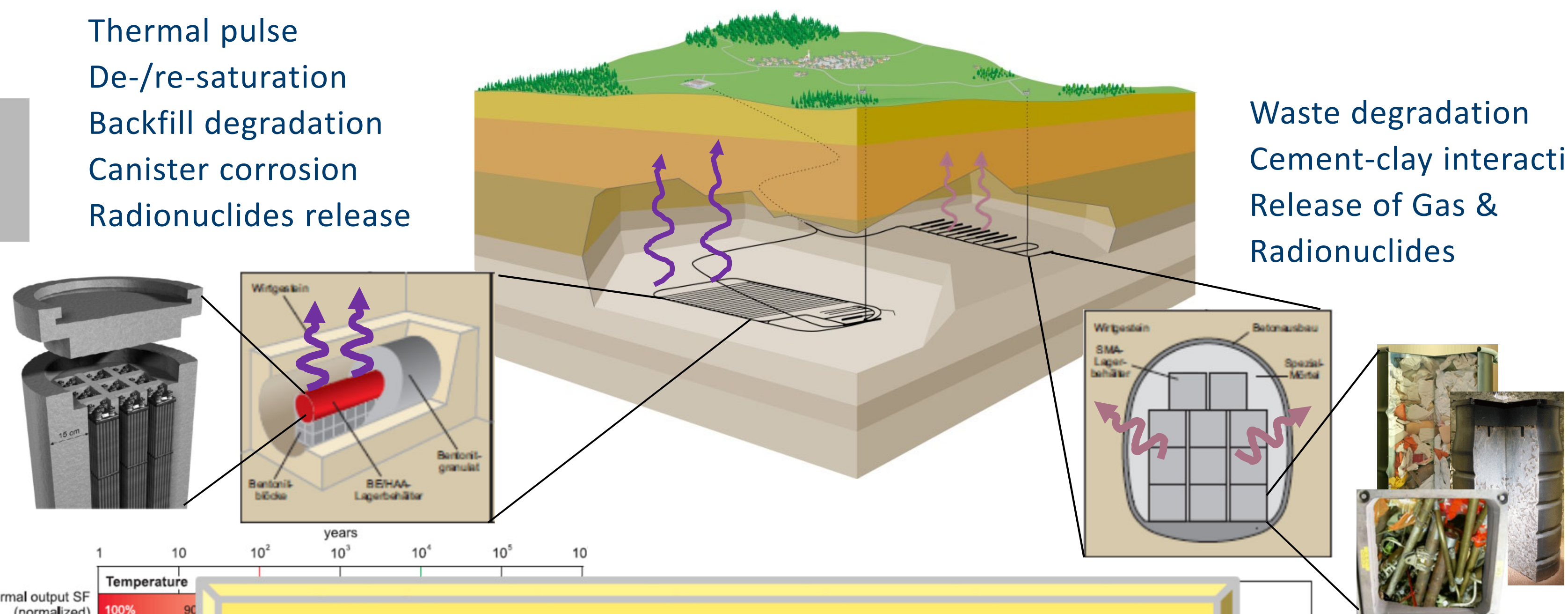
Origin of Nuclear Waste and Disposal Concept

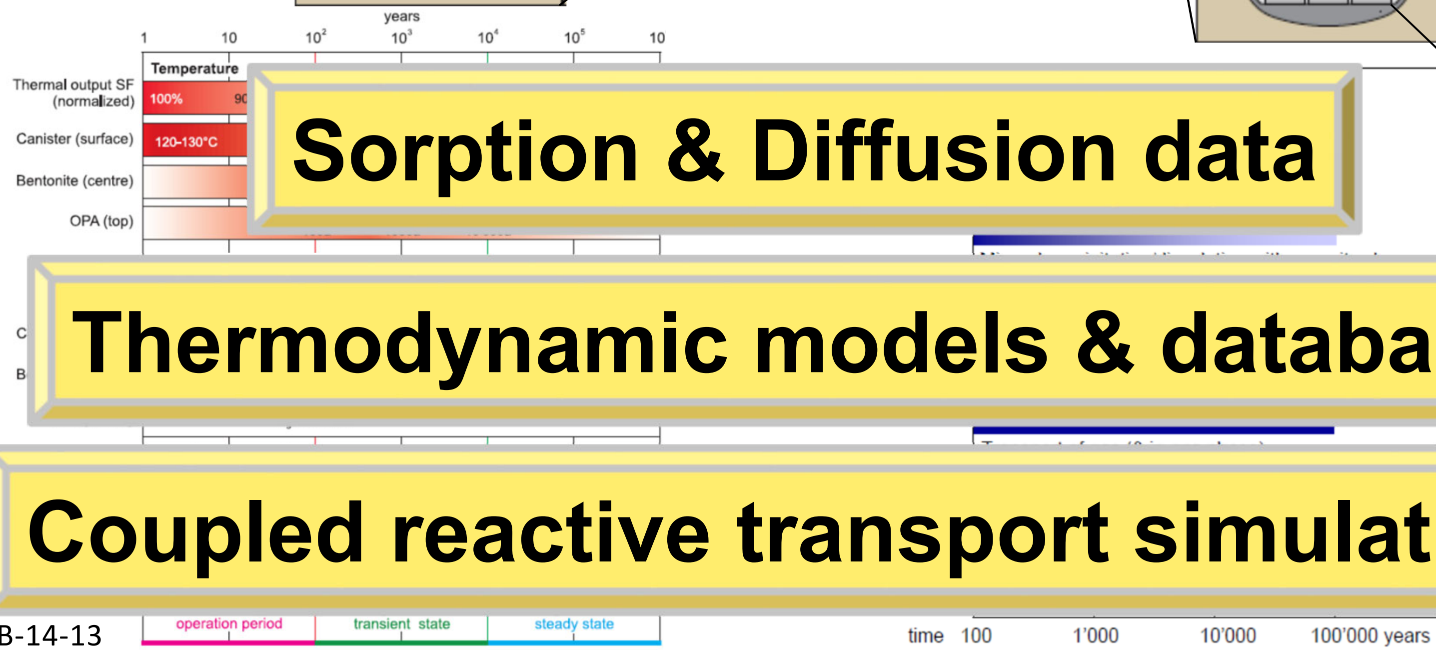


Coupled phenomena in repository systems

Thermal pulse
De-/re-saturation
Backfill degradation
Canister corrosion
Radionuclides release

Waste degradation
Cement-clay interaction
Release of Gas &
Radionuclides





Sorption & Diffusion data

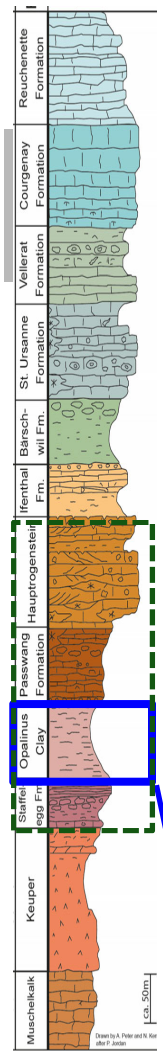
Thermodynamic models & databases

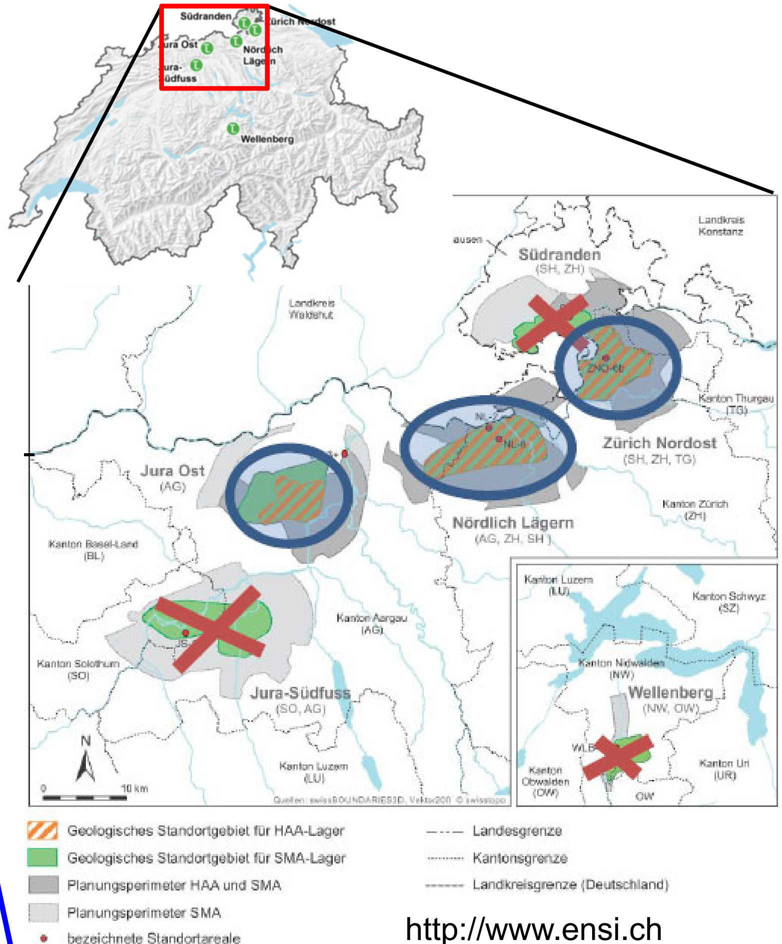
Coupled reactive transport simulations

NTB-14-13

NTB-14-11

Sectoral Plan and Legal Framewor1k





http://www.ensi.ch

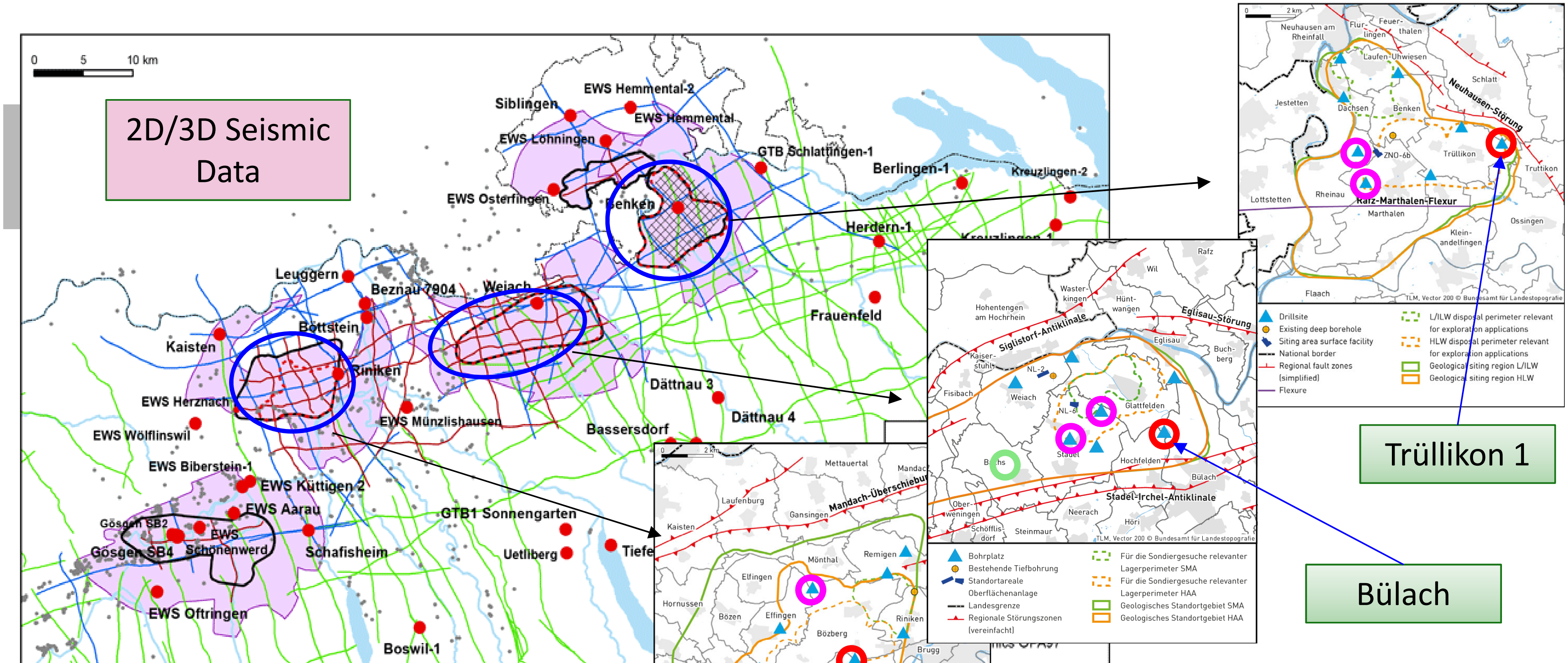
Opalinus Clay

- **Site selection process (SGT)** is coordinated by **Swiss Federal Office of Energy (BFE)**
- SGT is conducted by **National Cooperative for the Disposal of Radioactive Waste (Nagra)**
- Technical surveillance is conducted by **Swiss Federal Nuclear Safety Inspectorate (ENSI)**
- The approval is in competence of the **Swiss Federal Council (Bundesrat)**
- Bundesrat decision can be challenged by **Swiss National Referendum**

http://www.bfe.admin.ch

Stage 1		Stage 2 *		Stage 3			Public voting
3 HLW and 6 L/ILW sites		2(3) HLW and 2(3) L/ILW sites		1 HLW and 1 L/ILW sites		Public hearing	Pilot facility
Investigations	Review	Investigation	Review/consultation	Investigations	Lizensing	Review/consultation	Operation
2004 2008	2011	2012 2014	2015 2018	2019 2021	2022 2024	2025 2029	2030 2040 2060

Site exploration campaign 2019-2021



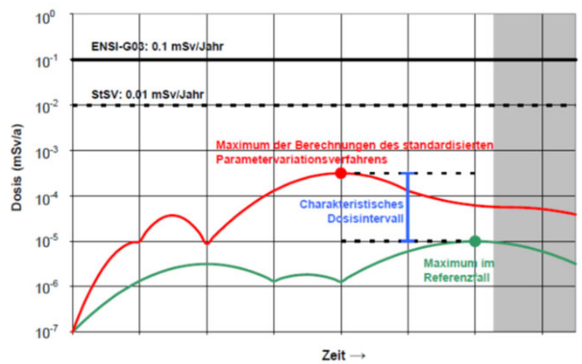
8 Vertical drillings

1 Inclined drilling

In total > 10 km long drilling interval

Importance of system understanding

- Underground exploration campaign of the host rocks at potential siting regions reveals non-negligible heterogeneities in terms of mineralogy, pore water chemistry and rocks petrophysical properties.
- *In situ* site specific geochemical conditions and the ones imposed in the laboratory during sample characterization are not identical due to various technical and methodological constraints.
- *In situ* condition in repository will evolve with the time due to the interaction between engineered barriers and the natural system evolution.
- The laboratory data need to be corrected for the deviation from *in situ* condition based on thermodynamic models. Same data and model are used for the model based prediction of the repository *in situ* conditions for timescale of several 100’000 years.



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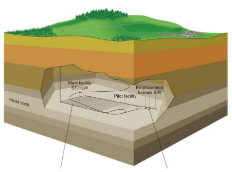
LES mission and vision

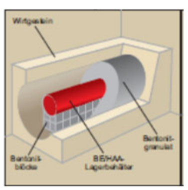
LES serves the **national needs, present and future**, in providing a **scientific basis for the safe disposal of radioactive waste**.

LES supports Nagra by providing **state-of-the-art synthesis reports and data for repository safety assessment** in the context of the national waste management programme.



LES carries out research in the areas of:

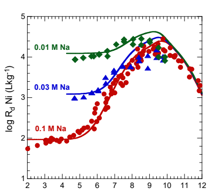
- **repository in situ conditions** and their evolution, and **repository induced effects** including both **modelling and experimental aspects**



- interface chemistry and transport of radionuclides in repository systems
- fundamental understanding of system behaviour for **long-term predictive modelling** and knowhow transfer.

LES maintains:

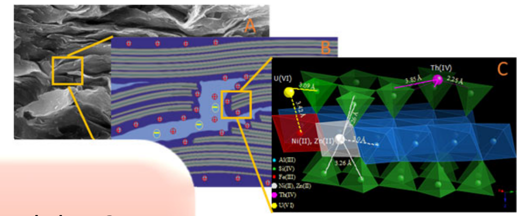
- **a proper balance between applied** and **fundamental research**
- **state-of-the-art expertise and knowledge** in strategic areas

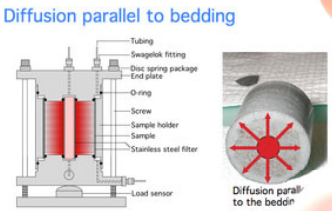


LES Long term vision:

Fully coupled THMC description of repository in situ conditions supported by multiscale modeling and experimental data at laboratory and field scale

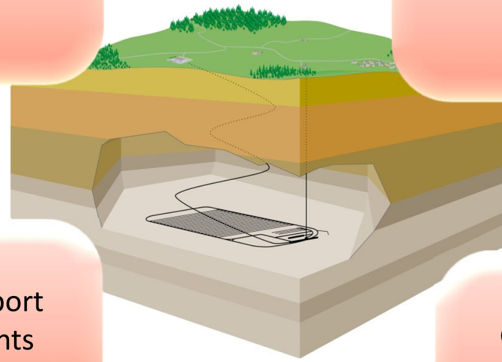
LES Research portfolio

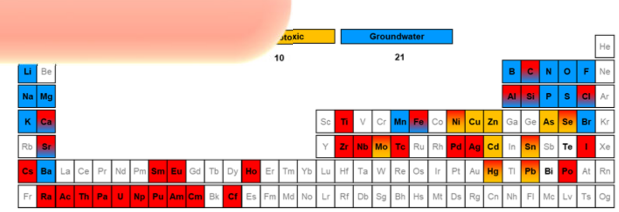




Diffusion studies at laboratory and field scale

Sorption of radionuclides & hazardous metals

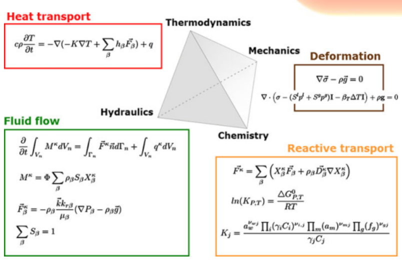


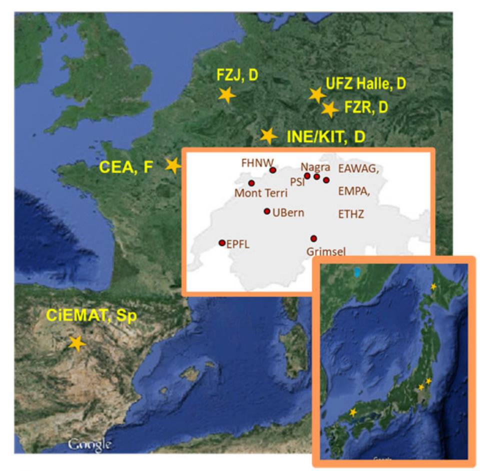


Multiscale reactive transport modelling and experiments

Geochemical modelling and thermodynamic databases

Coupled phenomena in repository system



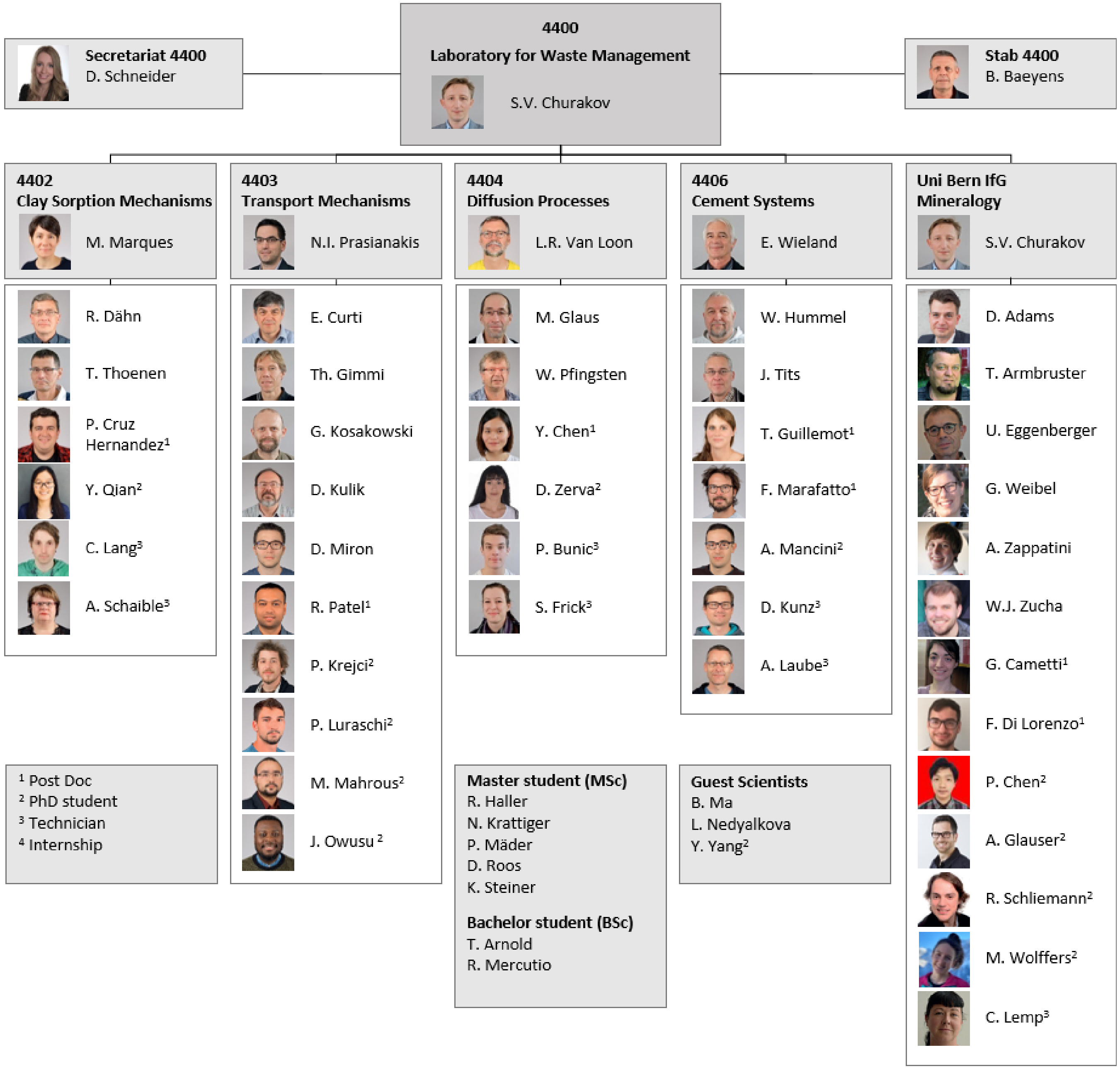


LES is :

- Demanded and reliable partner for Industry
- Key player in interactional research projects

nagra. FNSNF ANDRA swissnuclear HORIZON 2020 Mont Terri Project gsk ONDRAF NIRAS nanocem

LES-Organization



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SLS (PSI)

Hot Laboratory (PSI)

CSCS

SINQ (PSI)

Essential Infrastructure

Modeling Platform

GEMS SELEKTOR

OpenGeoSys

COMSOL©

XRD-Lab (UniBe)

Mont Terri and Grimsel URLs

Education platform

European joint Programming EURAD (HORIZON 2020/EURATOM)

- In 2018 SBFI has mandated PSI and Nagra to participate in the European Joint Programme (EJP) as Research Entity (RE) and Waste Management Organization (WMO), respectively.
- PSI/NES is the leading beneficiary and contact point for other RE participating as third linked parties from Switzerland (e.g. EMPA, ETHZ).

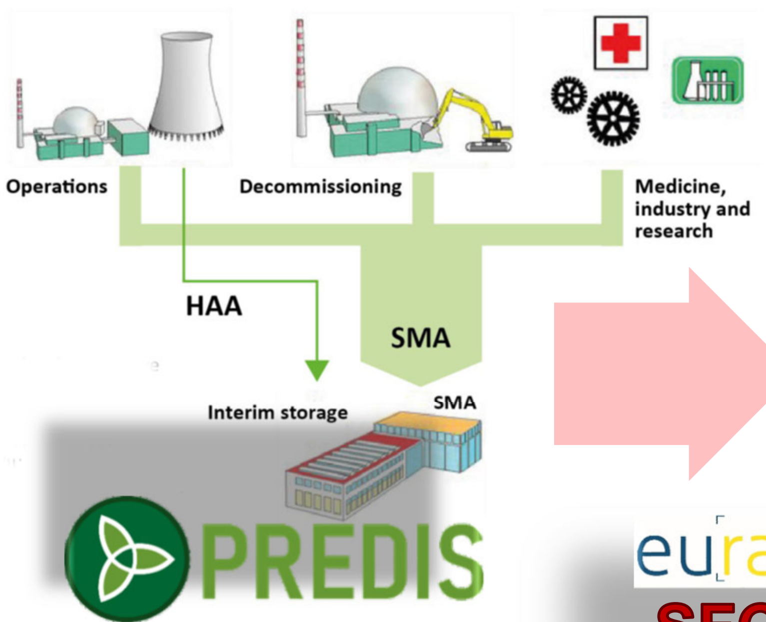
EURAD-1(2019-2024) budget:	59`871 KEUR
PSI budget (incl. EMPA):	2`343 KEUR
EU contribution:	50 %

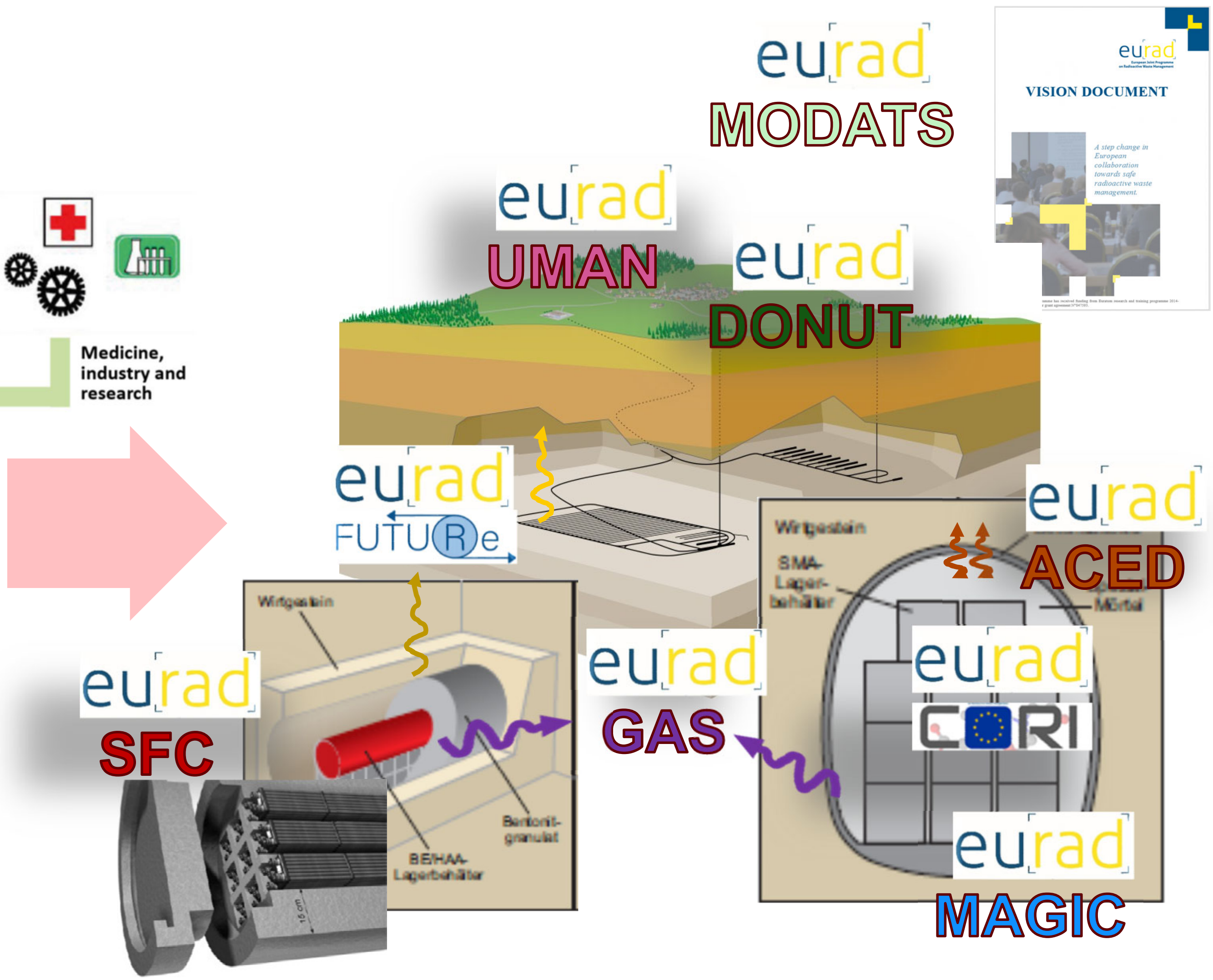
www.ejp-eurad.eu

European Joint Programme on Radioactive Waste Management

51 Mandated Organisations
54 Linked Third Parties
23 Countries
21 EU Member-States
2 Associated Countries

EURAD: *"… new era of efficient public RD&D funding in Europe…"*





LES/NES

Towards more realistic process modelling

PAUL SCHERRER INSTITUT PSI

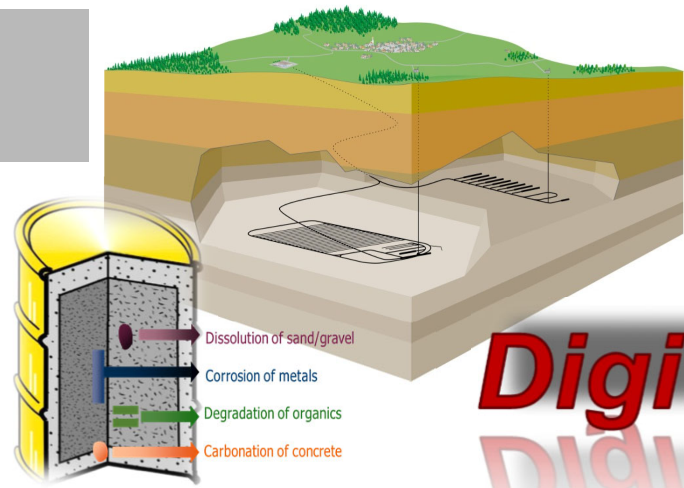
eurad MODATS

LES/NES nagra

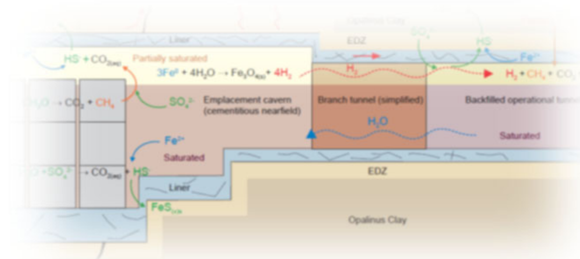
PREDIS

eurad DONUT

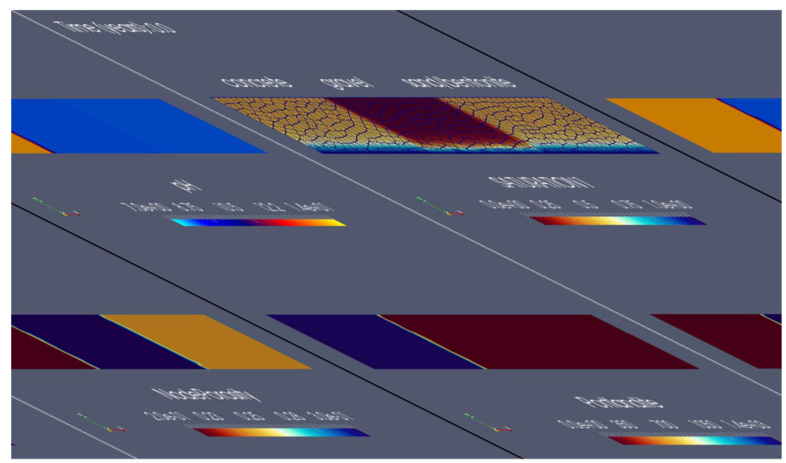
eurad ACED



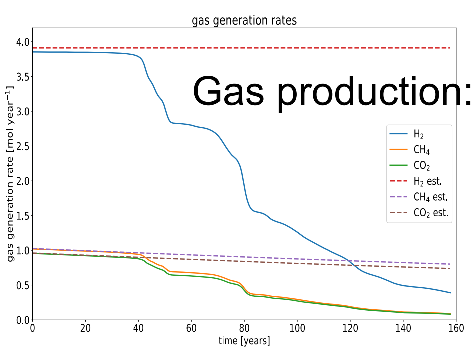
Digital Twin

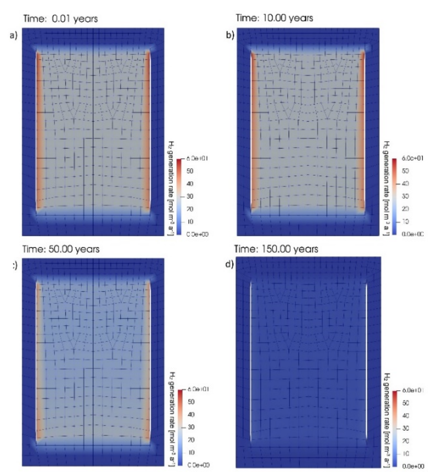


Repository in situ conditions



Gas production:

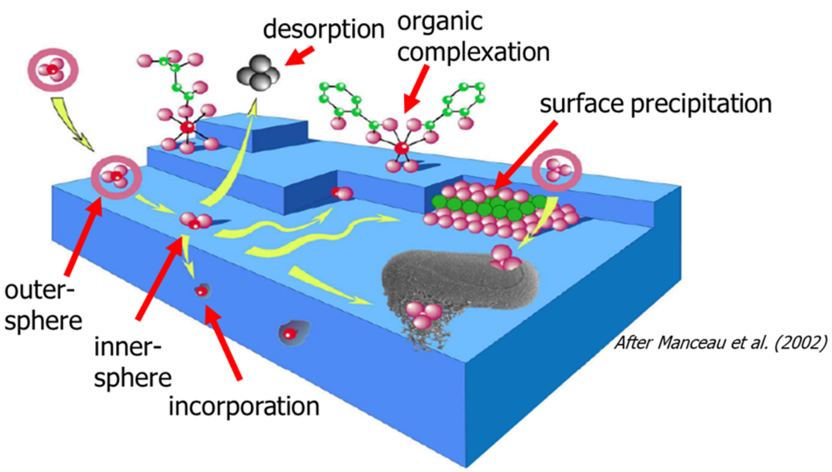


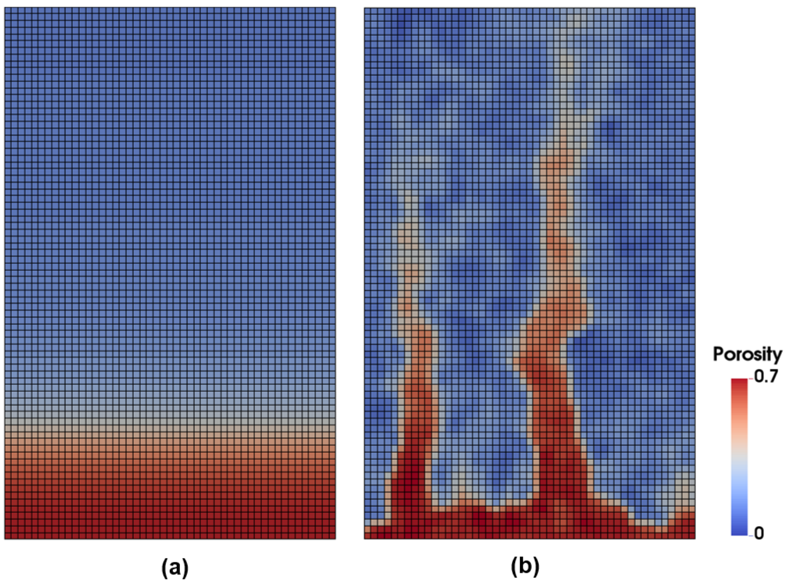


Huang, Shao, Wieland, Kolditz, Kosakowski (2021) www.nature.com/npjmatdeg 5, N4.

Hot research topics and strategic developments

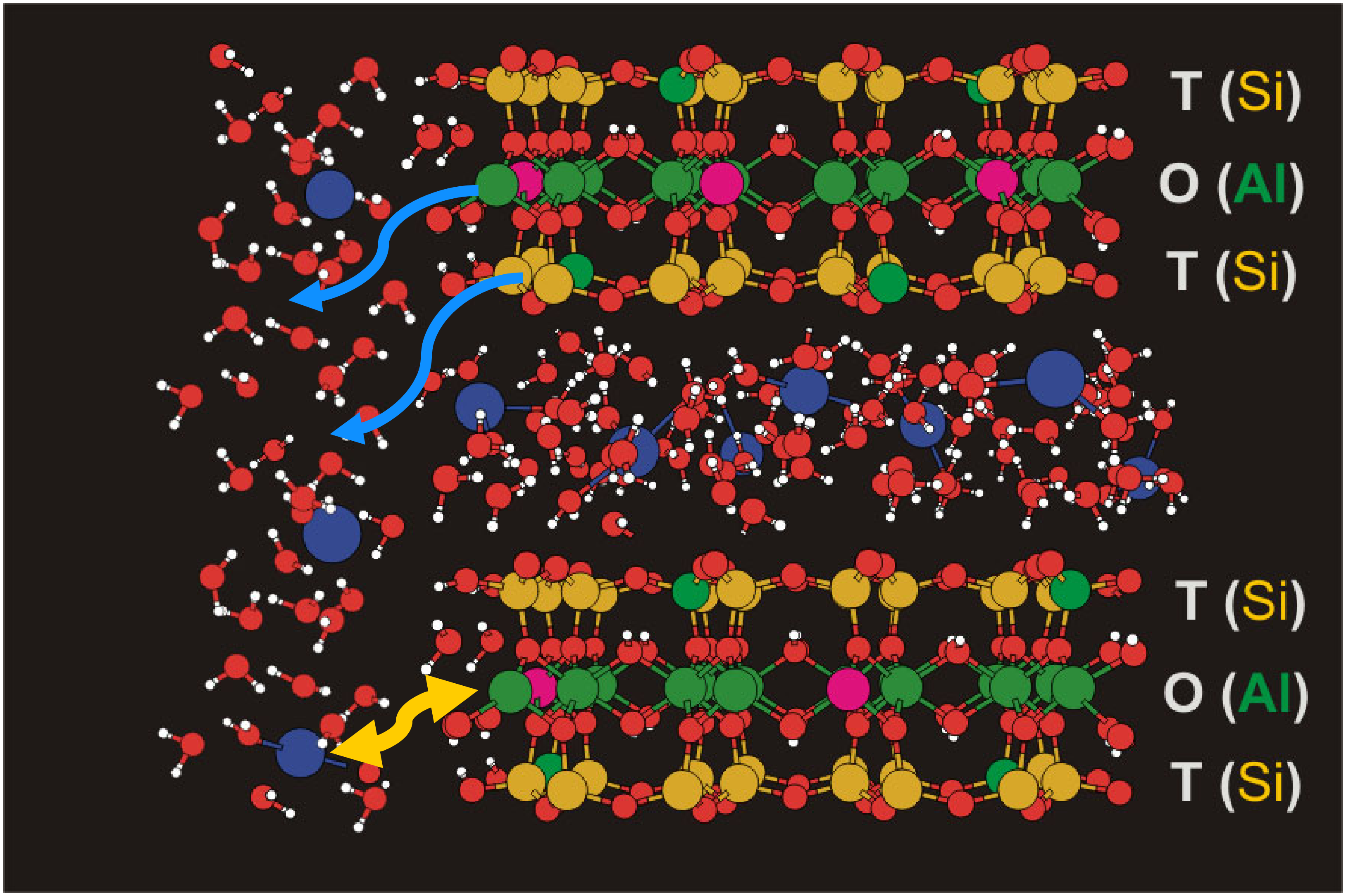
- Mechanistic understanding of radionuclide uptake mechanisms
- Coupled diffusion/advective transport phenomena and in reactive porous media
- Thermodynamic models and databases
- Frontiers of multiscale scale modelling





After Mahros at al. (2021) *(in prep.)*

Phyllosilicates reactivity

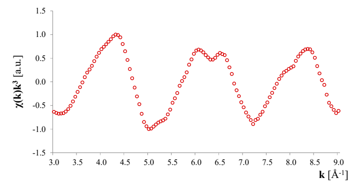


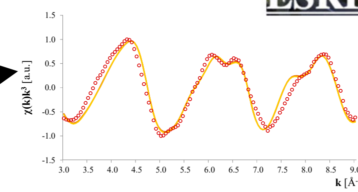
Churakov & Schliemann (2021) AIPEA

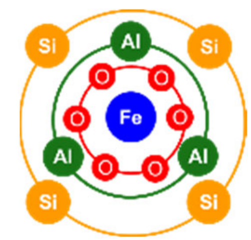
Spectoscopy meets molecular simulations

Conventional XAS data analysis

1) Measure of XAS spectrum
2) Fit shell model
3) Interatomic distances consistent with presumed structure?

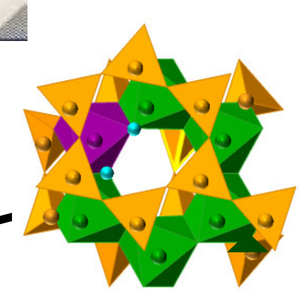


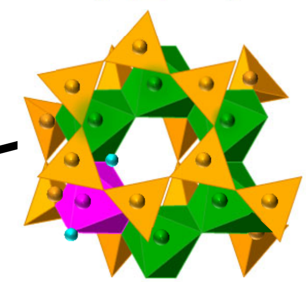


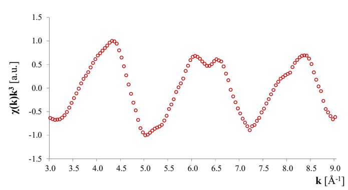


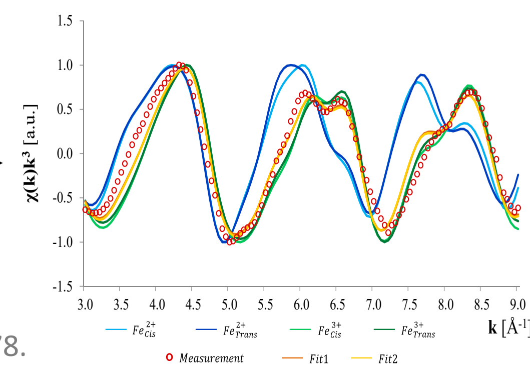
Computational XAS spectroscopy

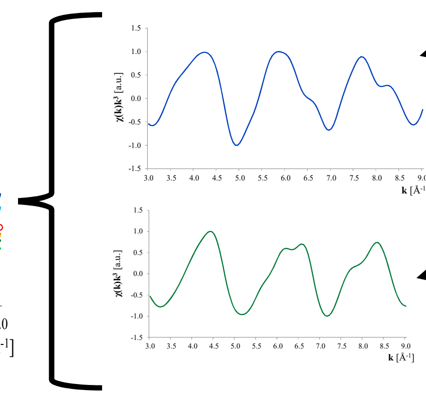
I) Measure of XAS spectrum
II) Molecular modelling of potential structures
III) Calculation of XAS spectra based on simulated structures
IV) Linear fit of calculated XAS spectra to measured ones







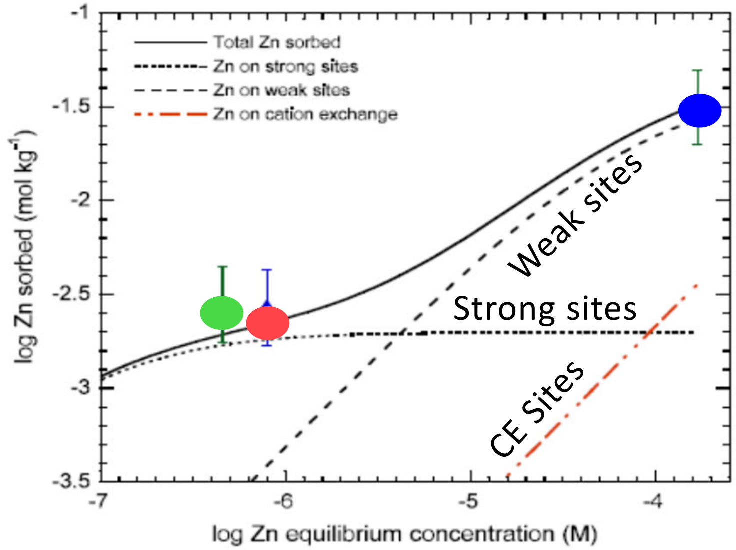


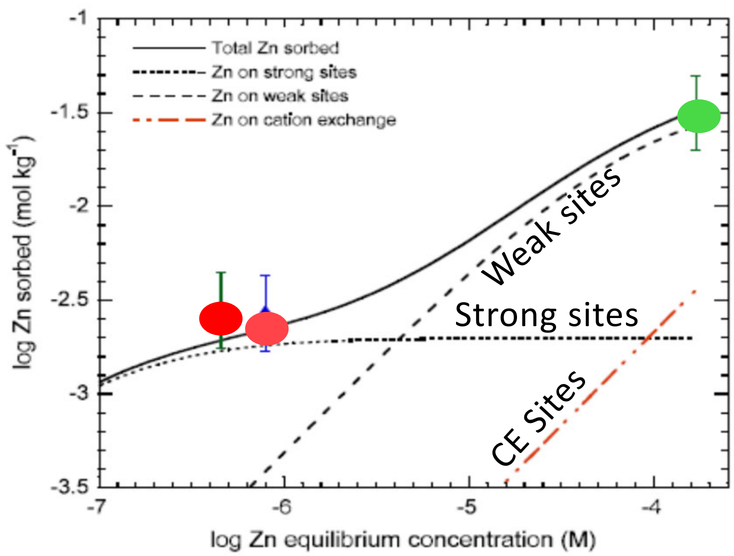


Churakov & Dähn (2013) ES&T, 47 6978.

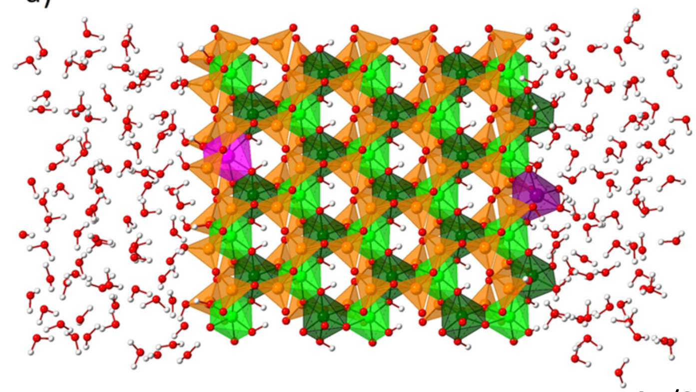
Me^{II} adsorption on Montmorillonite

Wet chemistry, spectroscopy & ab initio simulations

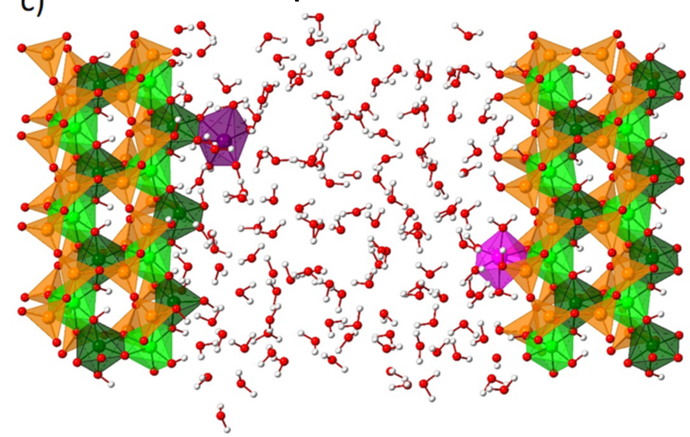




a) "Strong" Sorption sites Zn/$Fe^{2+/3+}$



c) "Weak" Sorption sites Zn/$Fe^{2+/3+}$

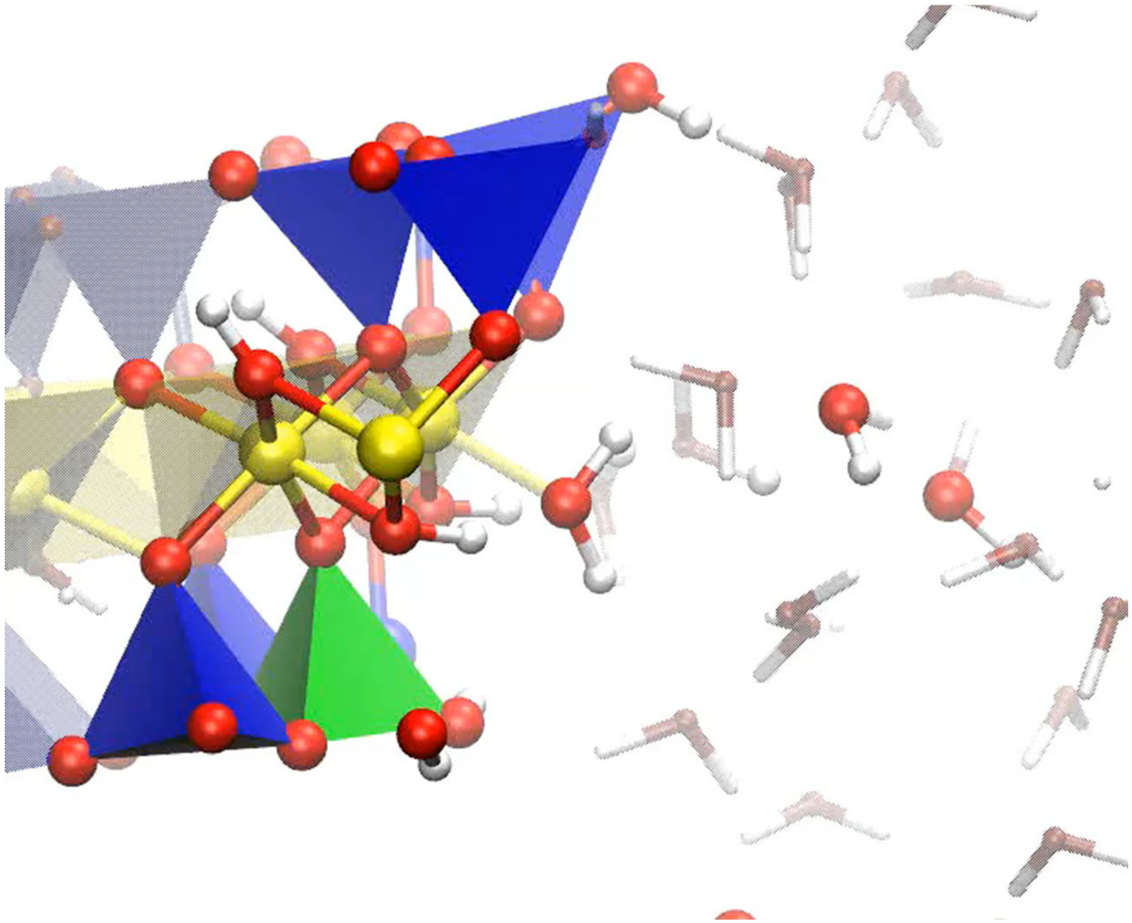


Spectroscopy and molecular modelling confirm existence of specific surface sites responsible for RN uptake.

Churakov & Dähn (2013); Keri et al. (2019) ES&T; Keri et al. (2017) ES&T

Clay edge dissolution mechanism

Ab initio molecular dynamics simulations



- Complex multistep reaction kinetics
- Concurrent reaction pathways
- Importance of solvent

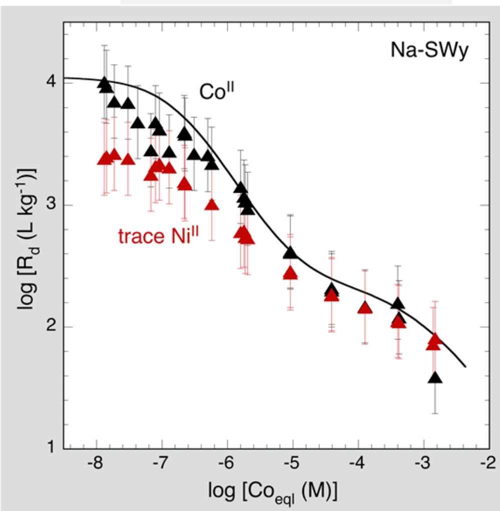
Schliemann & Churakov .(2021a,b) GCA.

Sorption competition in clay minerals

Sorption of **trace element** as function of increasing **blocking element** (binary systems). Metals involved Ni^{II}, Zn^{II}, Co^{II}, Fe^{II}, Pb^{II}, Eu^{III}, Am^{III}, Th^{IV}, Np^{V} and U^{VI}.

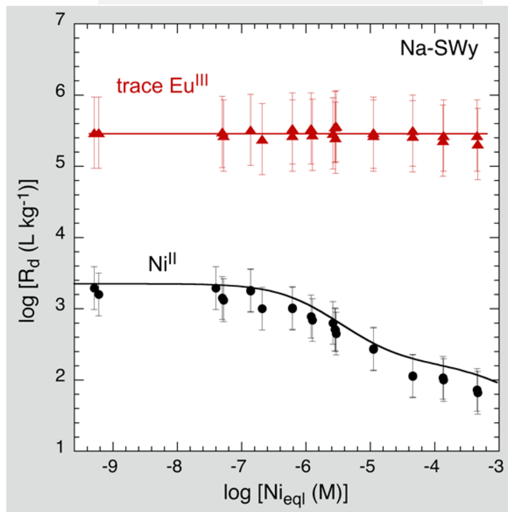
Me^{II} - Me^{II} Me^{III} - Me^{III}	competitive
Me^{III} - Me^{II} Me^{IV} - Me^{II} Me^{VI} - Me^{II}	not competitive
Me^{IV} - Me^{III} Me^{VI} - Me^{III}	not competitive
Me^{II} - Me^{III}	partially competitive

Competitive adsorption



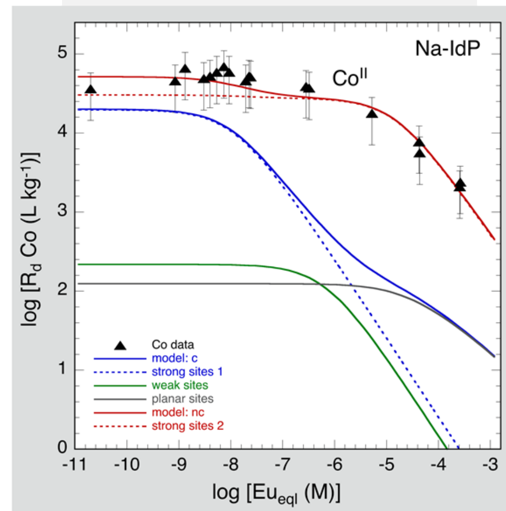
- Same strong sites.
- Sorption controlled by strength of SC constants.
- No need to change the model.

Non-competitive adsorption



- Different strong sites.
- Element specific strong sites need to be defined in the model

Partial-competitive adsorption



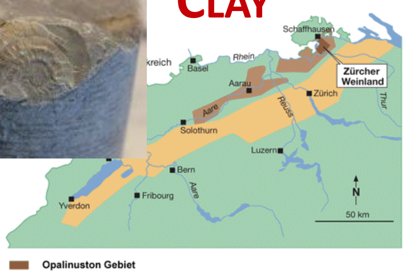
- Only partially same strong sites.
- Element specific subset of strong sites with different affinities and capacities need to be defined in the model.

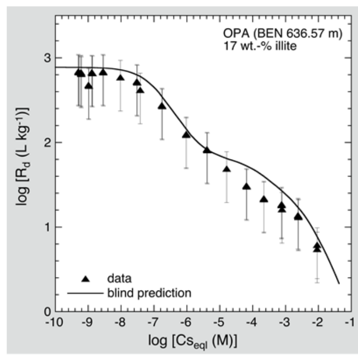
Marques Fernandes, M. and Baeyens, B. (2020) Competitive adsorption on illite and montmorillonite: Experimental and modelling investigations. Nagra Technical Report NTB 19-05.

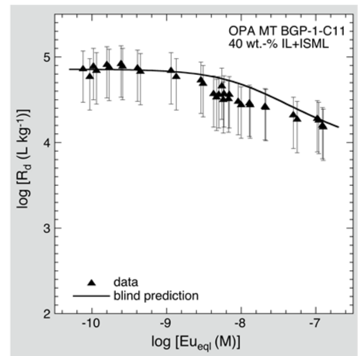
"Bottom-up" prediction of radionuclide uptake by argillaceous rocks

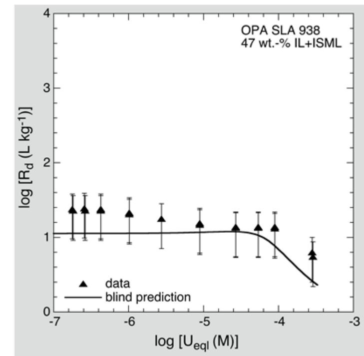
Bottom-up approach based on:
- clay content
- RN speciation in porewater
- RN-specific sorption model for illite/smectite

Opalinus Clay



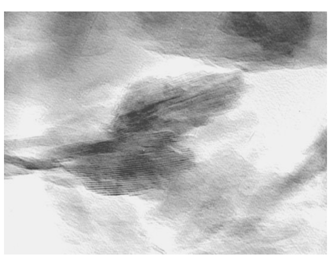




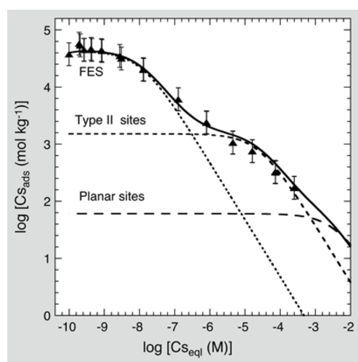


Bottom-up approach works

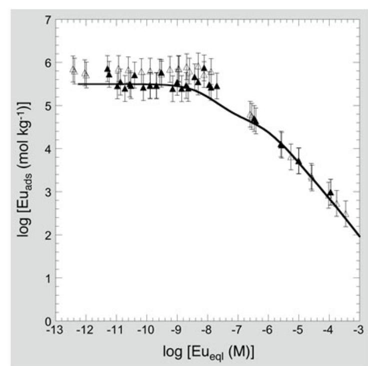
Illit



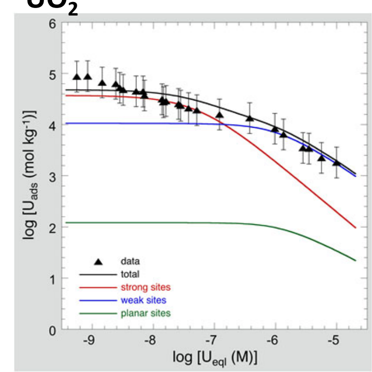
Cs



Eu

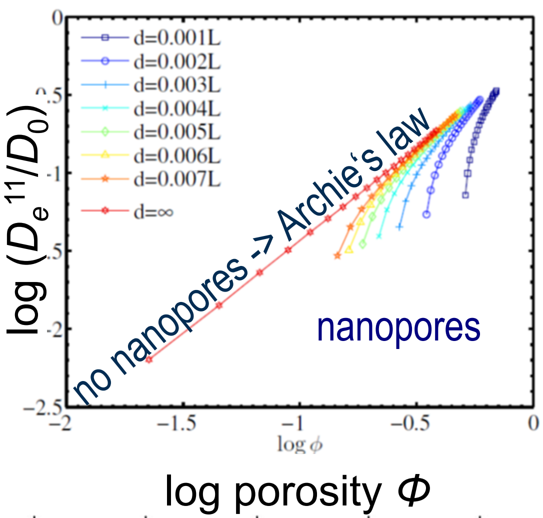


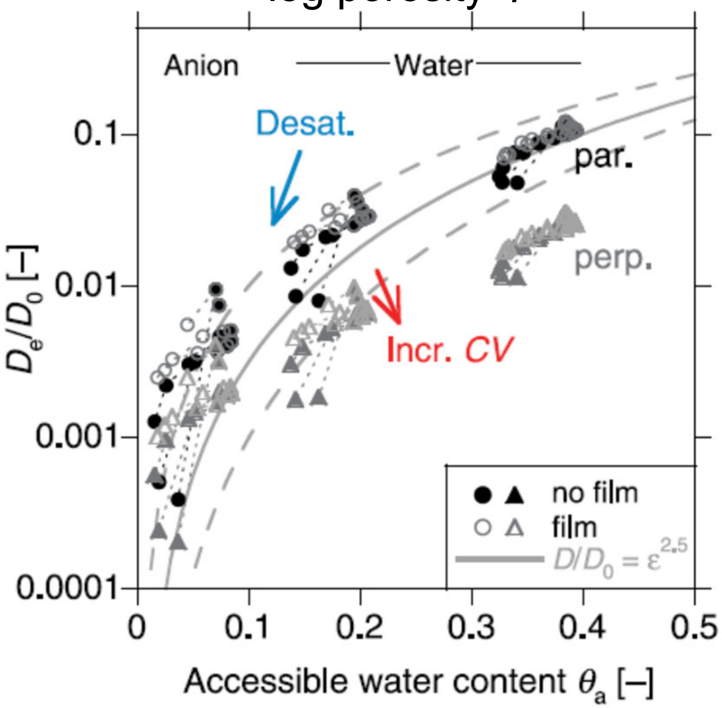
UO_2^{2+}

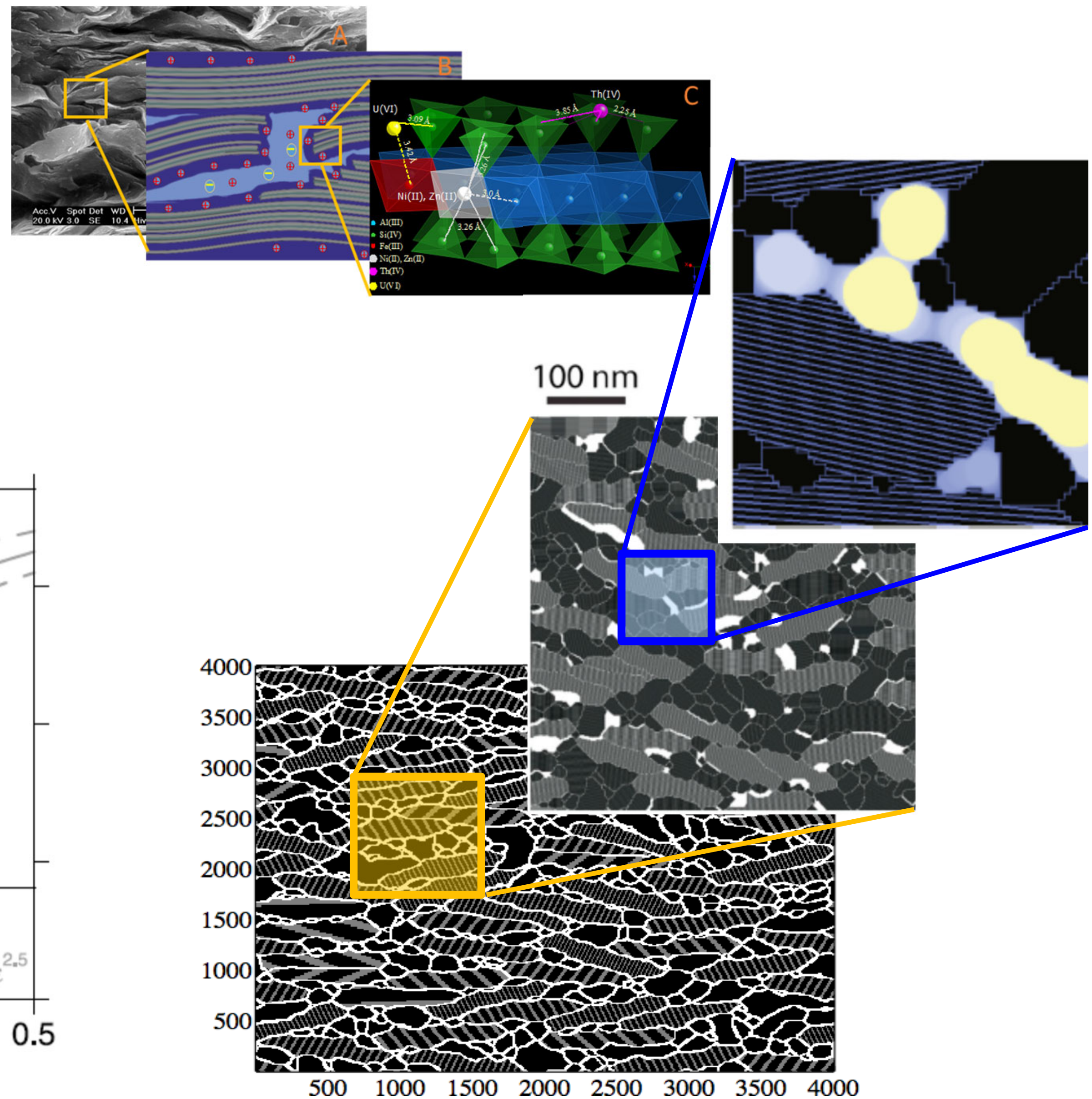


Marques Fernandes, M. and Baeyens, B. (2020) Competitive adsorption on illite and montmorillonite: Experimental and modelling investigations. Nagra Technical Report NTB 19-05.

Diffusion in partially saturated clays

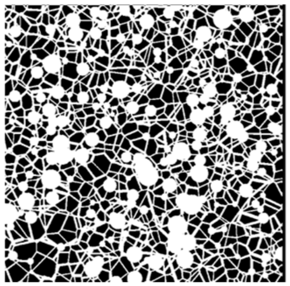




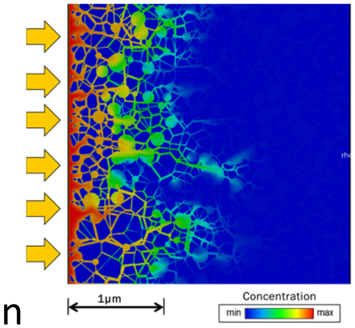


Tyagi et al. (2013) Adv. Water. Res; Churakov et al. (2014) Appl. Clay Sci.,; Gimmi & Churakov (2019) Appl. Clay Sci.

Transport phenomena in reactive porous media



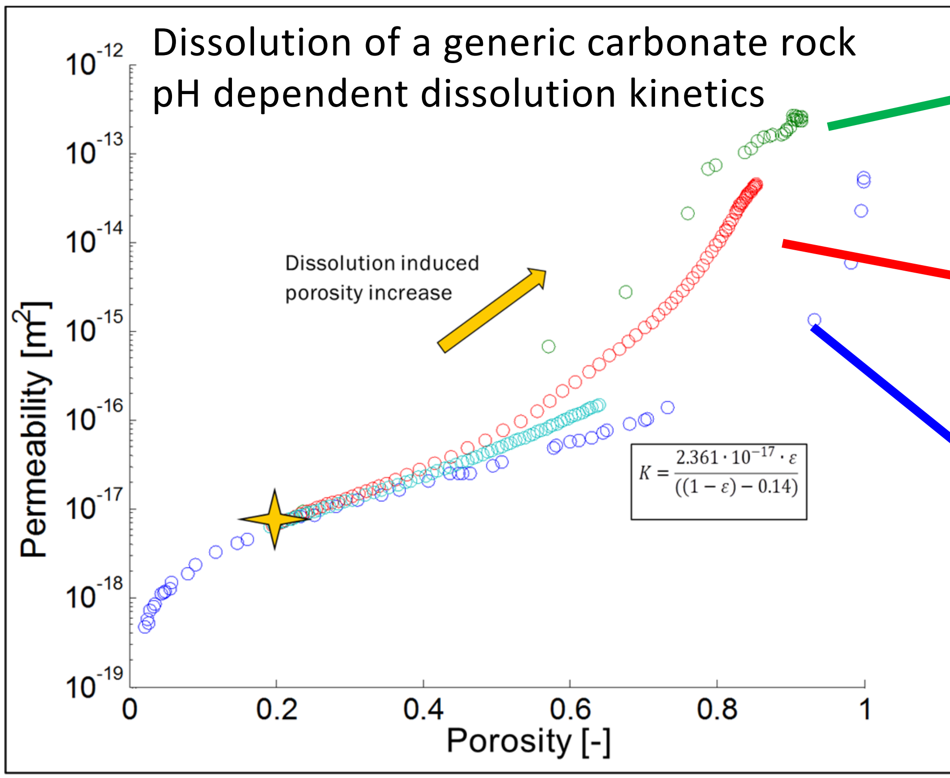
Voronoi tessellation



Evolution of porous medium (calcite matrix) due to reactive transport (dissolution)

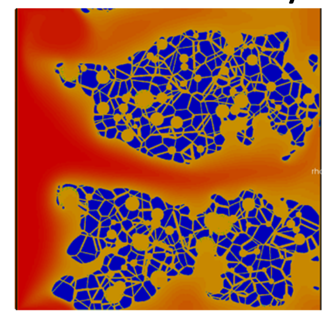
Porosity permeability relationships

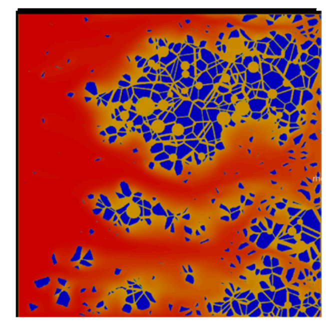
1) Pre-calculate
2) On-the-fly only where it is needed
3) Machine learning

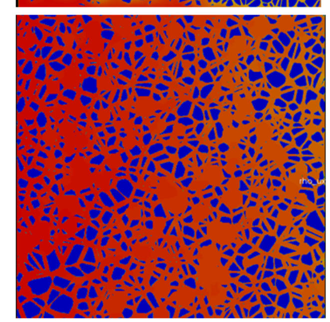


Prasianakis, et al. (2018) Geofluids, ID 9260603

Fast reactivity





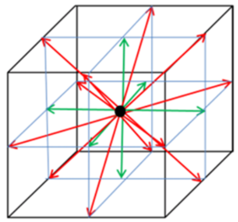


Slow reactivity

Digital Twin and Numerical diagnostics

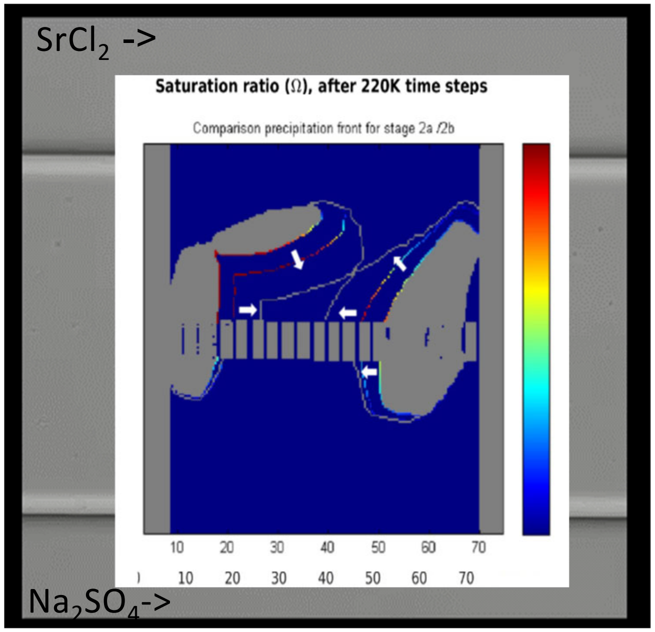
Augmented reality by **pore scale lattice Boltzmann modelling & diagnostics**.
Classical nucleation theory & heterogeneous reactions kinetic
Pore scale multicomponent transport

Injection of 10 mM $SrCl_2$ and 10 mM Na_2SO_4 → celestine precipitation, crystal growth



Layers of diagnostics

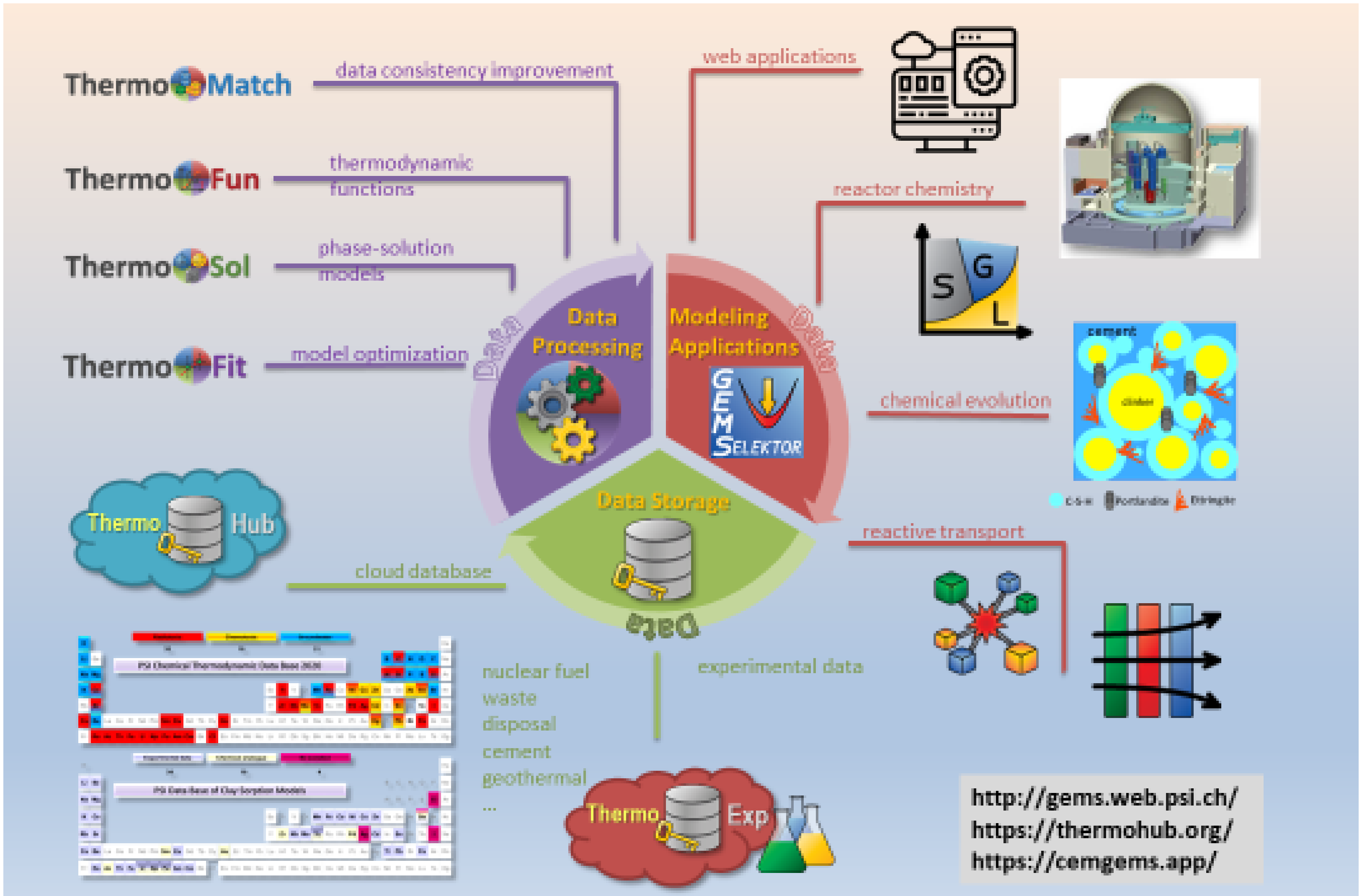
- Evolution of experiment (camera)
- Local flow-field and streamlines visualization (numerically calculated, experimentally verified).
- Spatial resolution of velocity field at different stages of the experiment (numerically calculated)
- Local species concentrations, saturation ratio (numerically calculated, interplay of advection/diffusion)
- Local precipitation rates at fluid-solid interface, prediction of directional differential growth (numerically calculated, color: precipitation rate)



$$SrCl_{2(aq)} + Na_2SO_{4(aq)} \rightarrow SrSO_{4(s)} + 2NaCl_{(aq)}$$

Poonoosamy, J., Westerwalbesloh, C., Deissmann, G., Mahrous, M., Curti, E., Churakov, S.V., Klinkenberg, M., Kohlheyer, D., Von Lieres, E., Bosbach, D., Prasianakis, N.I., A microfluidic experiment and pore scale modelling diagnostics for assessing mineral precipitation and dissolution in confined spaces, *Chemical Geology, 528, 5, 119264 (2019)*
Prasianakis, Churakov et al. Neural network based process coupling and parameter upscaling in reactive transport simulations, *Geochimica et Cosmochimica Acta. 291 126-143 (2020)*

Geochemical Toolkit:
All-in-One Databases & Models



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Software and Database
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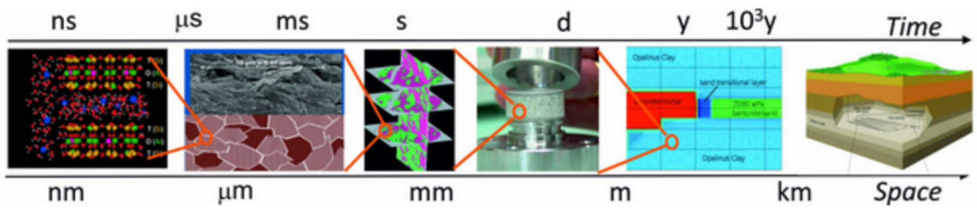

Laboratory for Waste Management (LES)

LES is the Swiss competence center for geochemistry and multiscale radionuclide and mass transport in argillaceous rocks and cement and their application to deep geological systems and Swiss radioactive waste repositories. LES offers attractive research projects at the bachelor, master, PhD, and postdoc levels in environmental sciences and nuclear engineering

Core Competences

- Geochemistry of repository systems
- Retention and migration of radionuclides
- Multiscale reactive transport in natural and engineered barrier systems
- Thermodynamic databases
- Geochemical education

Facilities, Tools and Infrastructure

- Synchrotron-based material characterization and neutron imaging
- Numerical simulations at atomistic, pore Lattice-Boltzmann, and continuum scales
- Geochemical and thermodynamic modeling tools
- State-of-the-art radiochemical laboratories

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Outlook

- **Mechanistic understanding** of **radionuclide uptake** needs further investigations combining traditional wet chemical methods, spectroscopy and theoretical modelling
- **Multiscale reactive transport phenomena** are extremely challenging and need further **development of upscaling strategies** and **numerical concepts for parameter transfer** between model and codes at different scales
- There is a large **potential** in **application of ML/AI algorithms** for the **model coupling** and **acceleration of geochemical modeling**
- **Experimental studies are indispensable for validation of numerical models**

Thank you for your attention!

Acknowledgments to Nagra for a longstanding partnership!