PAUL SCHERRER INSTITUT

PSI

UNIVERSITÄT BERN

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

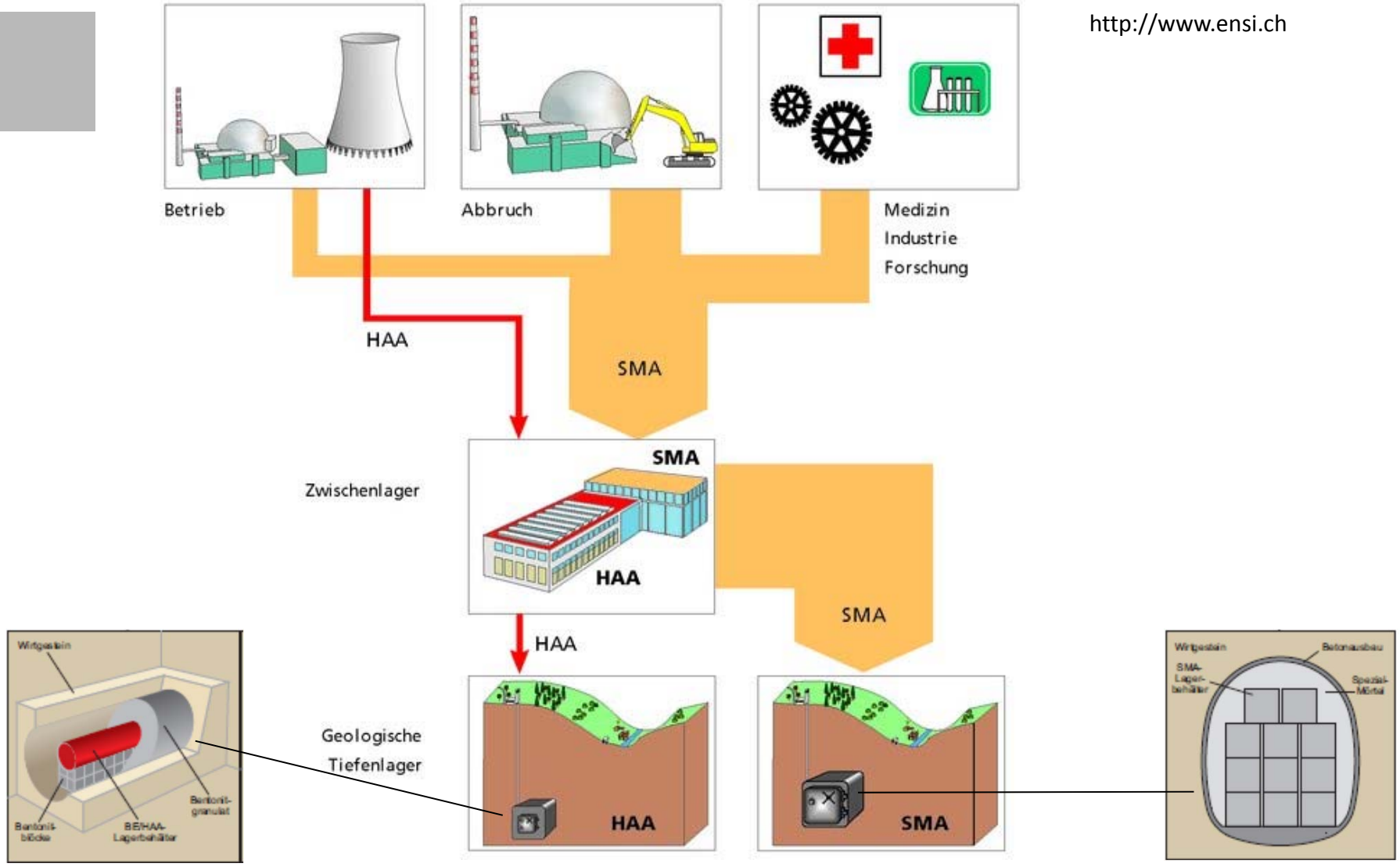
Labor für Endlagersicherheit

NES Kompetenzen und Highlights, Oktober 18, 2016, PSI

Outline

- Swiss waste disposal programme
- Laboratory for Waste Management (LES)
  - Mission
  - Organizational chart
  - Important infrastructure
- Selected research projects
  - Sorption of redox sensitive nuclides
  - ^{14}C speciation during corrosion of activated steel
  - Reactivity of technical barriers and material interfaces
  - Fundamental research on mineral surface chemistry and reactivity
- Summary

Origin of Nuclear Waste and Disposal Concepts



http://www.ensi.ch

Labor für Endlagersicherheit(LES): Mission

LES is a national center for geochemistry of waste disposal.

We provide:

- Scientific basis for the safe disposal of radioactive waste.
- State-of-the-art scientific documentation to support Nagra in Sectoral Plan.
- Geochemical and transport data sets for Nagra's safety assessment codes.

LES carries out a research programme in the following areas:

- Interfacial chemistry and transport of radionuclides in clay- and cement-based systems
- Thermodynamics and kinetics of retention in such systems.
- Reactive transport studies relevant to repository in situ conditions including both modelling and experimental aspects.

LES maintains knowledge in strategic areas for the needs of the Swiss waste disposal programme.

LES contributes to the education of young scientists in the field of geochemistry of geological waste disposal.

LES keep a proper balance between applied and basic research.

LES contribution to the Sectoral Plan stage 3

Maintain **state-of-the-art** functionality of key **models** and **datasets for safety analysis**, including sorption, diffusion and thermodynamics.

Fill **missing gaps in databases**:

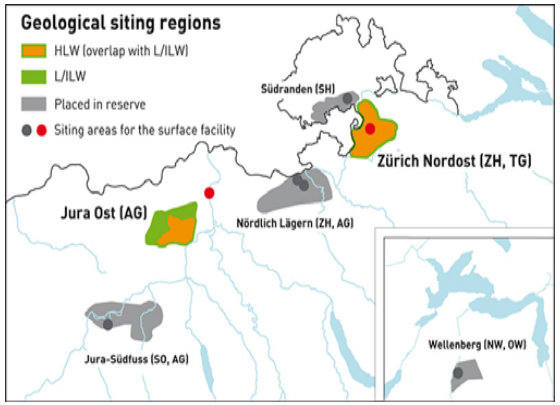
- **Redox sensitive elements** and justification of “chemical analog” arguments
- **Sorption competition / transferability** of data for compacted/disperse systems
- Chemistry of **dose determining radionuclides**

Geochemical evolution of in-situ repository conditions:

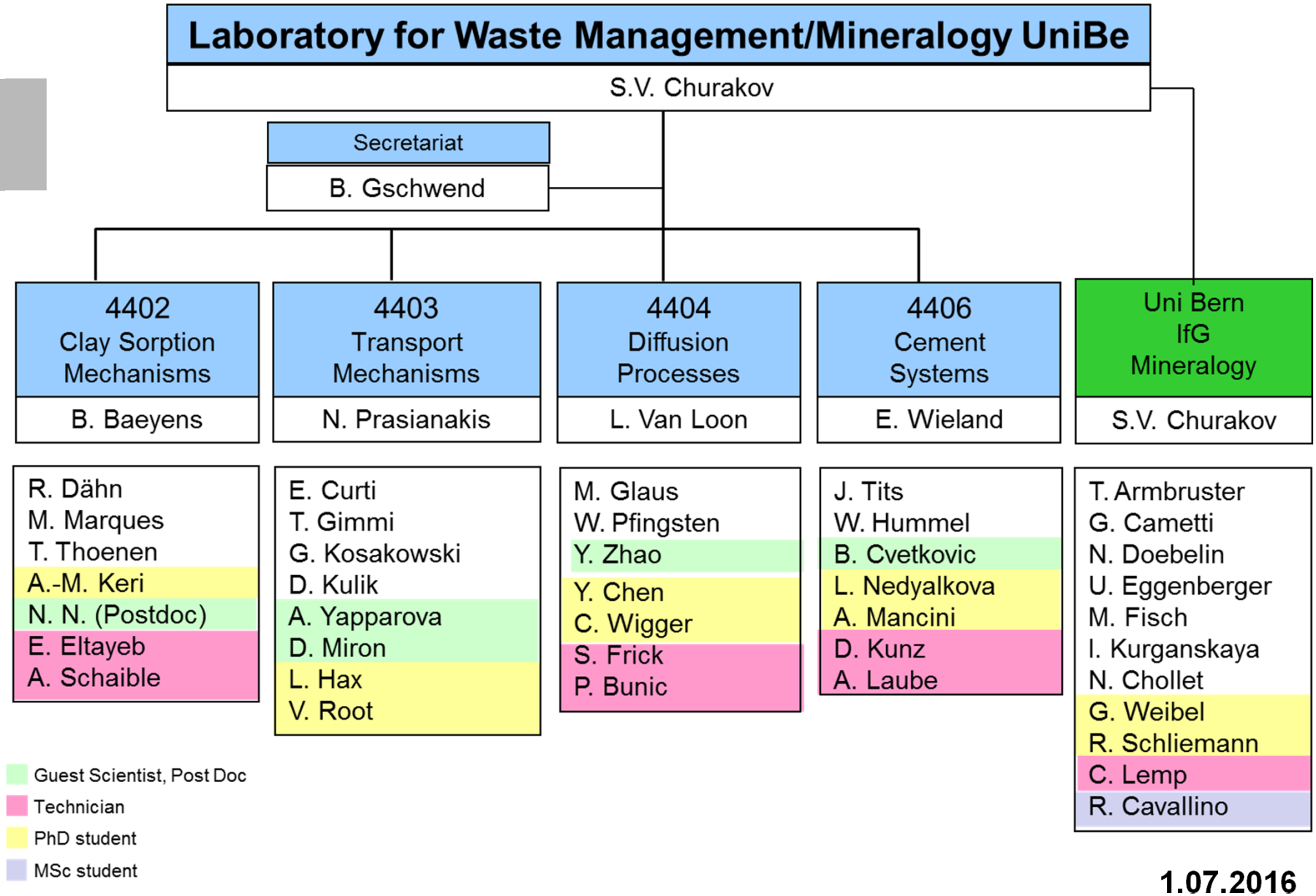
- **Reactivity of technical barriers** in the repository
- **Long-term evolution and safety function** of the multi-barrier system

Sample characterization from site specific field explorations

Scientific **documentation for the General License Application**

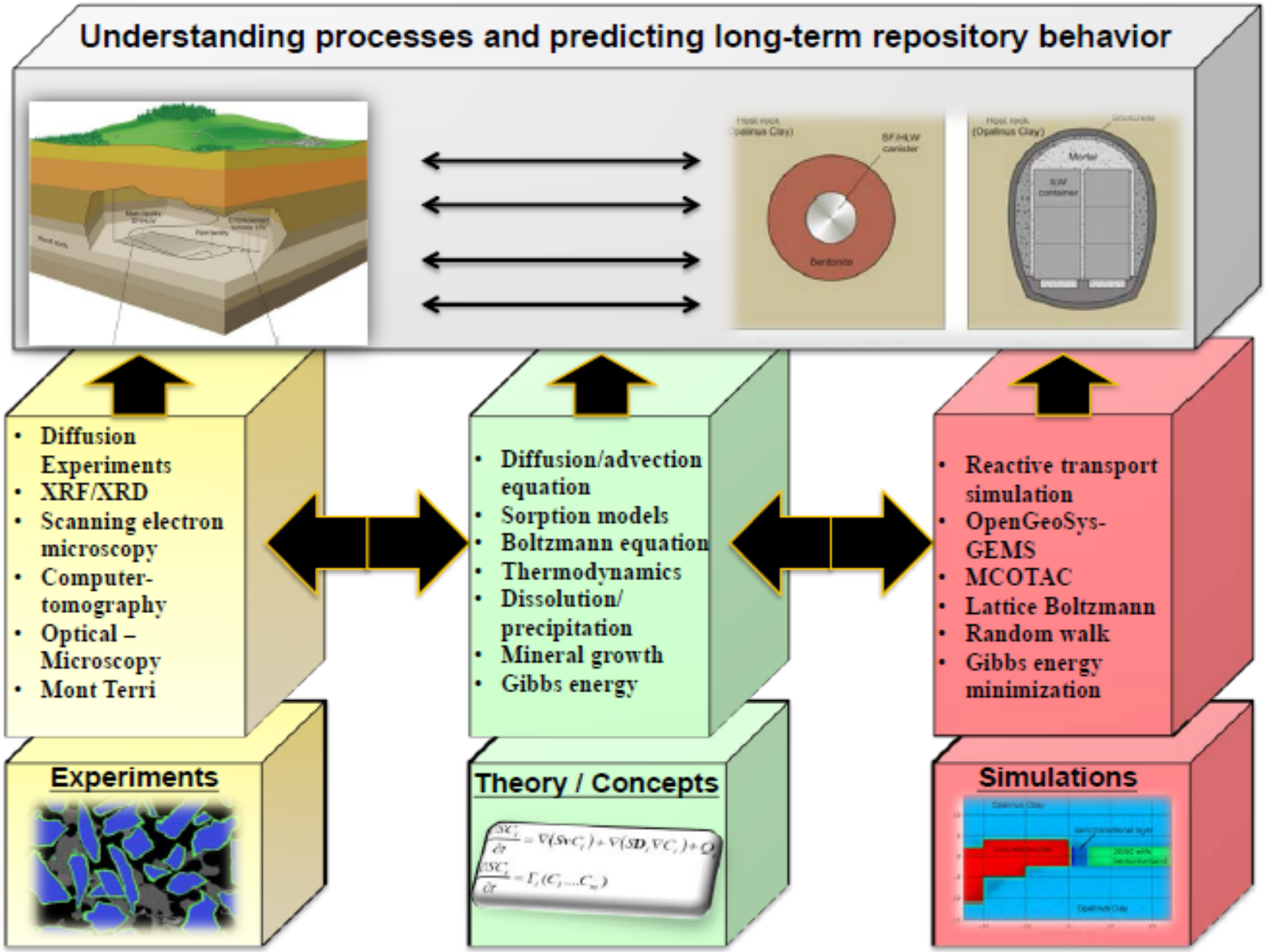


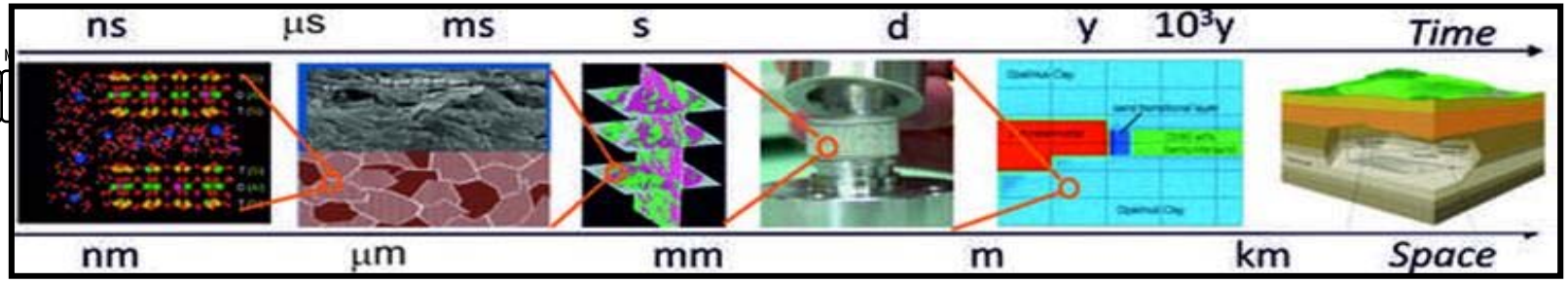
LES re-organization



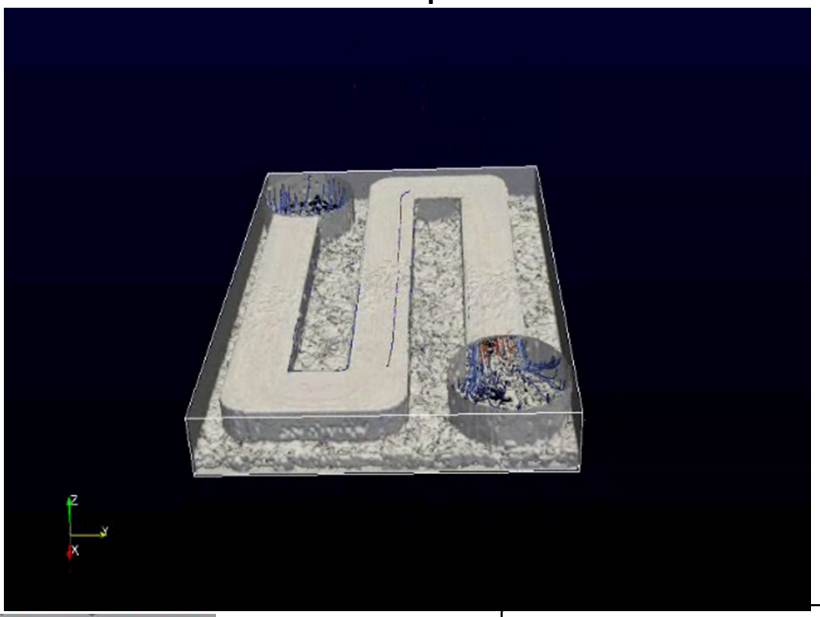
1.07.2016

Consolidation of modelling and experimental activities



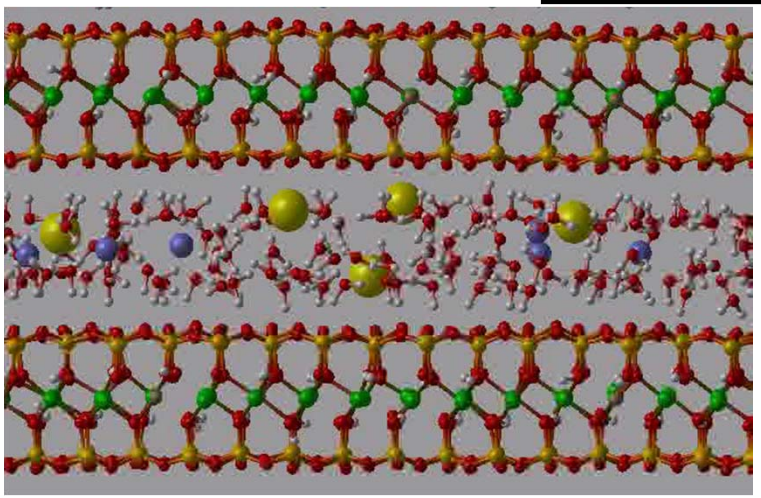


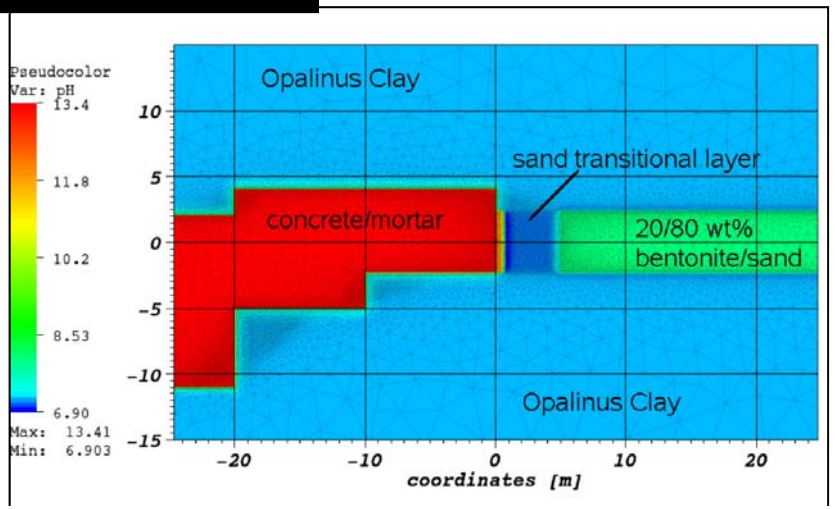
Pore scale transport simulations



Atomistic molecular simulations

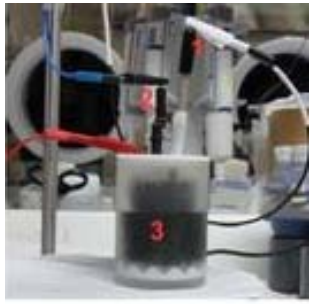
Field scale Reactive transport modelling

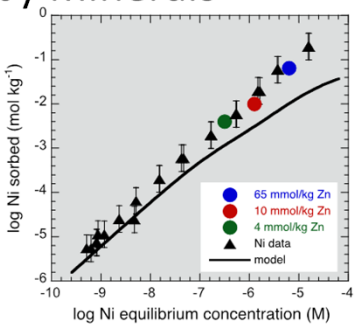


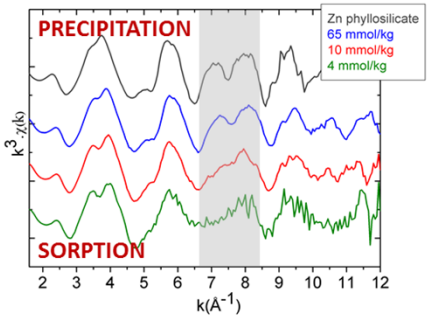


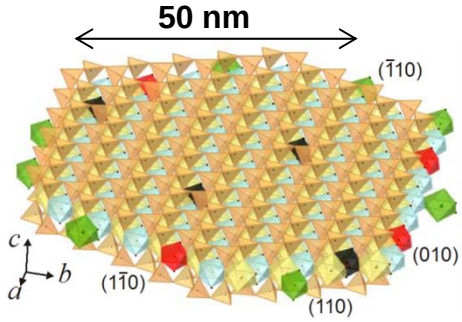
Core Competences

- Sorption measurements and model development for mechanistic understanding of contaminants retention by minerals

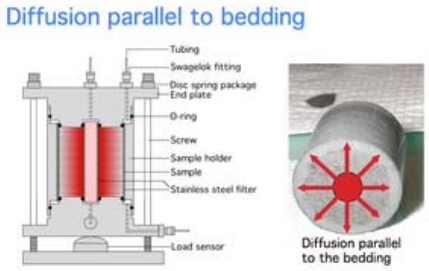


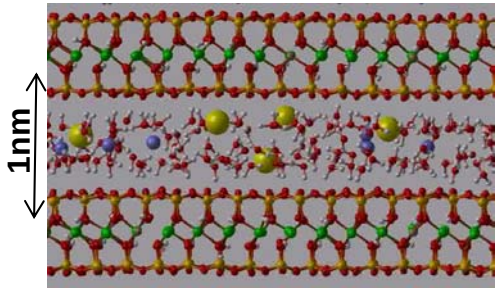


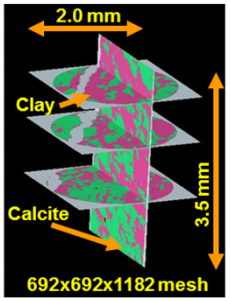


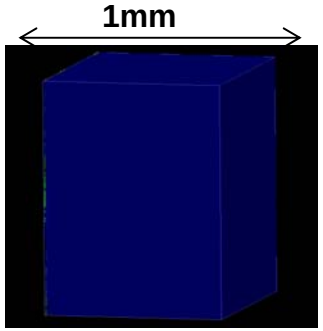


- Diffusion measurements and multi-scale transport simulations from an atomic level to a geological scale

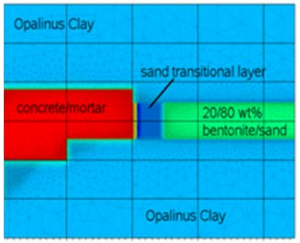


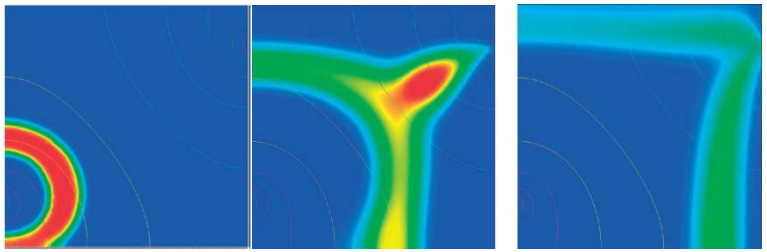


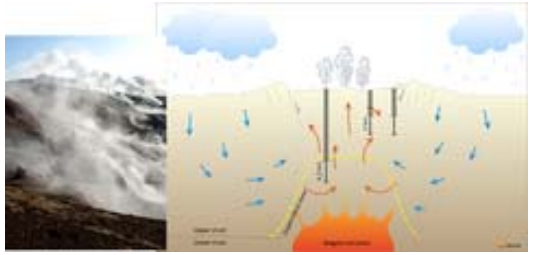




- Geochemical modelling of in situ conditions in energy-related subsurface systems (e.g. waste repositories, geothermal reservoirs, contaminated sites)





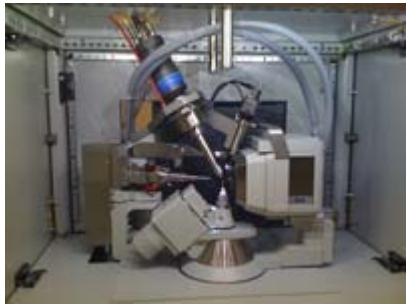


Important infrastructure

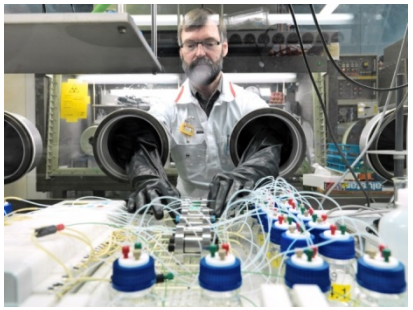
SLS (PSI)

SINQ (PSI)

XRD-Lab (UniBe)



Hot Laboratory (PSI)



CSCS

Mineralogy

UNIVERSITÄT BERN

PAUL SCHERRER INSTITUT

Geological Disposal Systems

Modeling Platform

GEM SELEKTOR

MCOTAC

B&B

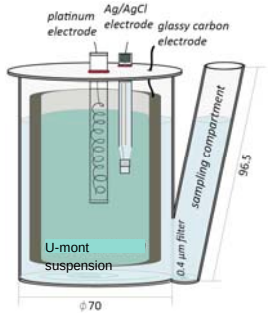
OpenGeoSys

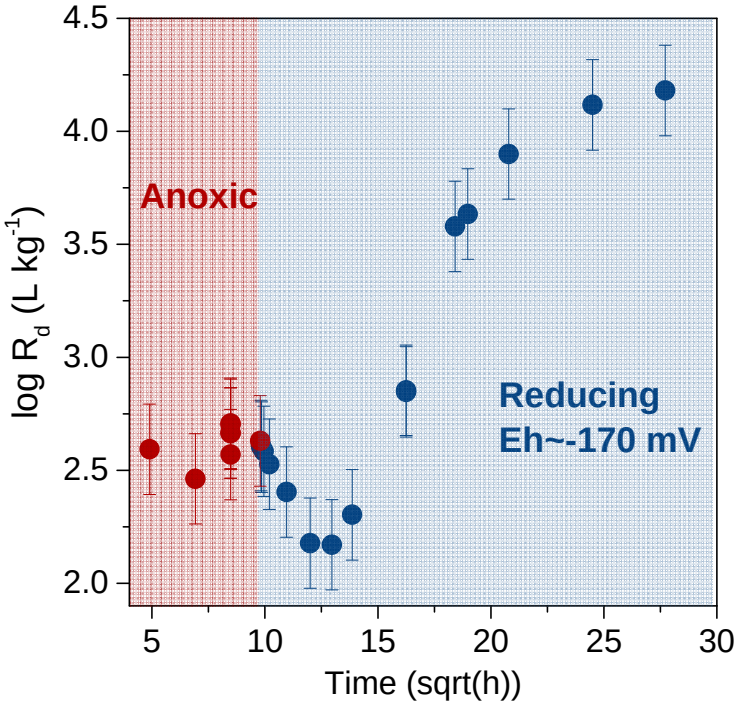
Mont Terri and Grimsel URLs

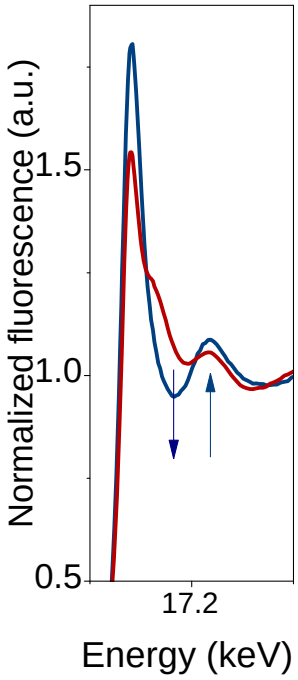
Education platform

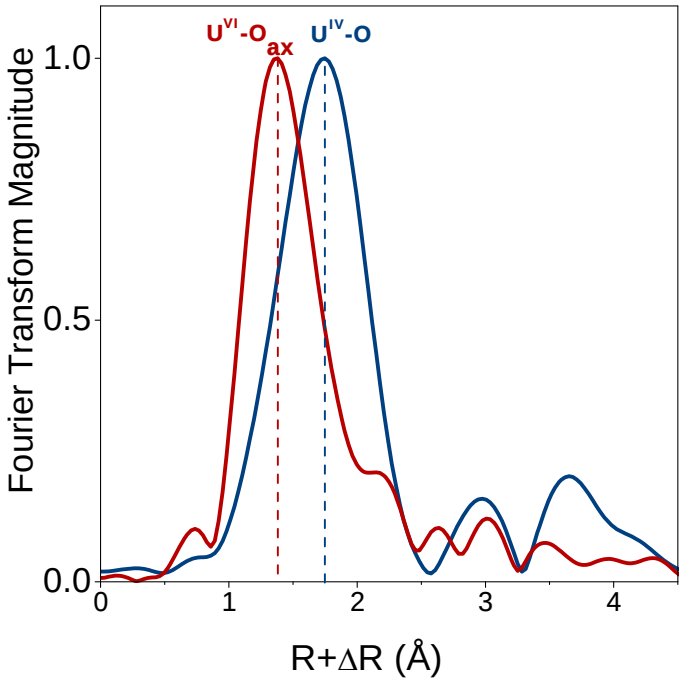
Redox studies on clays: uranyl sorption

U(VI) sorption on montmorillonite under anoxic conditions
U(IV) sorption under reducing conditions (in electrochemical cel)







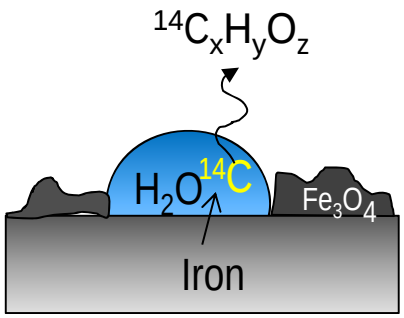


- Wet chemistry: Uranyl sorption increases under reducing conditions
- XAS corroborates the formation of U(IV) surface complexes under reducing conditions

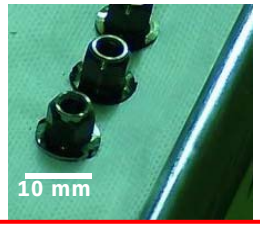
^{14}C speciation during corrosion of activated steel

Aim: Determination of the ^{14}C containing organic compounds formed during the anoxic corrosion of activated steel obtained from KKG in cement-type pore solution

^{14}C speciation

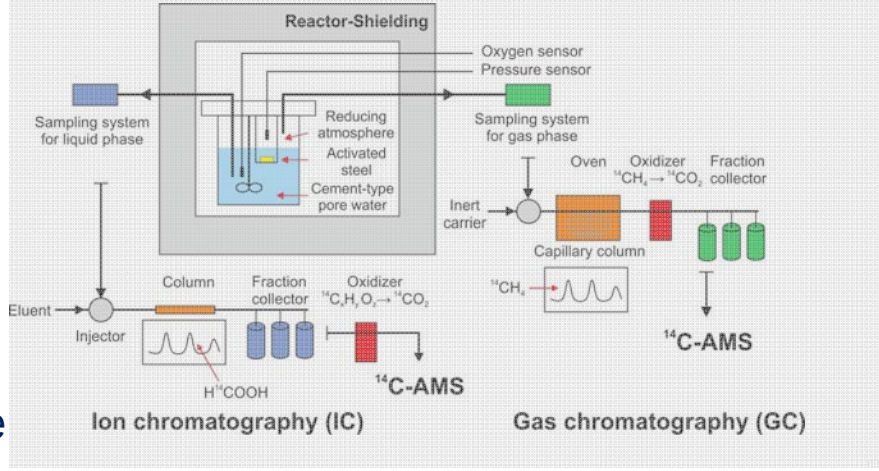


Activated steel nuts from KKG



Dose rate: ~ 30 mSv/h per gram material

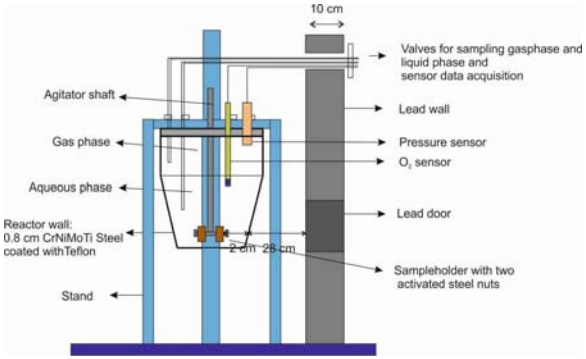
Experimental set-up of the corrosion experiment



- Development of the reactor system behind lead shielding due to high dose rate

Front view

Sketch



Sampling system for gas and liquid phase

gas samples

liquid samples

inside shielding

outside shielding

Corrosion experiment with activated steel nut segments was successfully started in May 2016. Regular sampling occurs, first results awaited.

Interaction of waste with engineered barriers

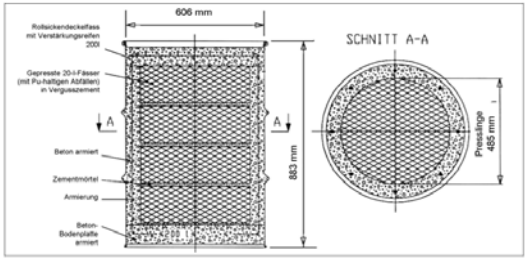
Aim: Experimental and modelling studies on the interaction of waste materials with cement paste as component of the engineered barrier

Examples:
- Thermodynamic modelling of the long-term chemical evolution of cement-stabilized waste packages
- Development of thermodynamic models of the iron-cement interaction

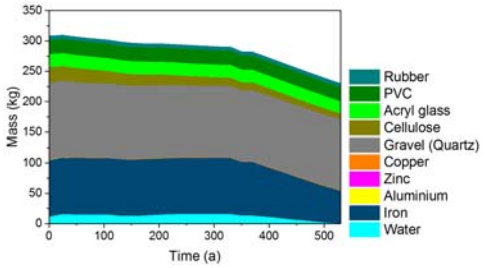
Thermodynamic modelling of waste packages

- Kinetically controlled degradation of waste materials
- Changes in the mineral composition of the cement barrier
- Influence on the porewater chemistry (e.g. pH)

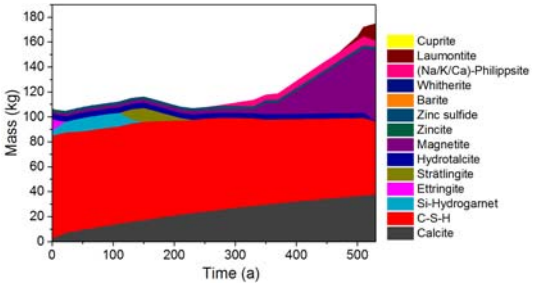
Waste package



Waste materials



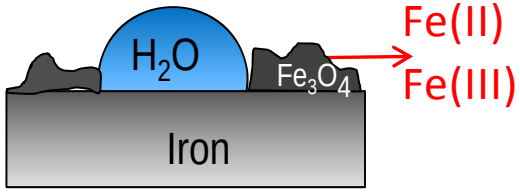
Mineral composition



Thermodynamic modelling of iron-cement interaction

- Interaction of Fe(II) with cement phases
- Identification of Fe and S species in anoxic cement paste using synchotron-based spectroscopic and diffraction methods

Iron corrosion



SA: Prediction of the long-term geochemical evolution of the chemical conditions and the heterogeneities in a cement-based near field

PAUL SCHERRER INSTITUT

Reactive transport simulation of Cement – Clay interaction

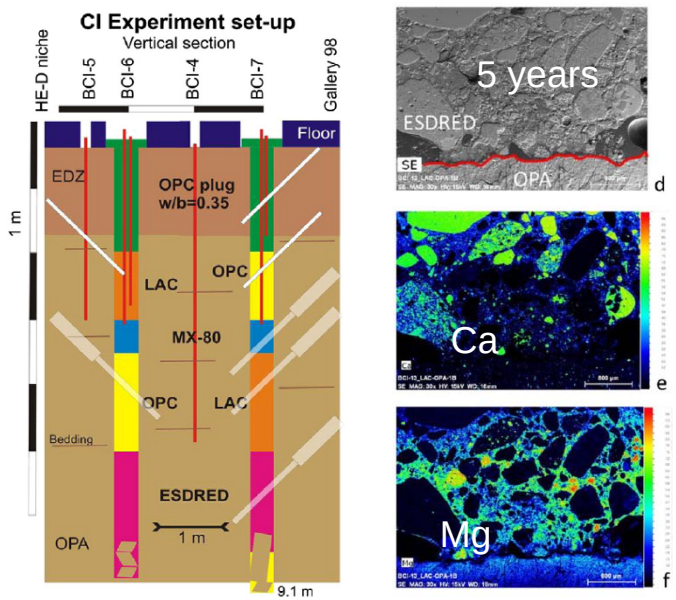
Cooperation with Mont Teri CI-experiment / Nagra / Horizon 2020 - CEBAMA

Aim: Analysis of evolution of material interfaces with help of numerical models

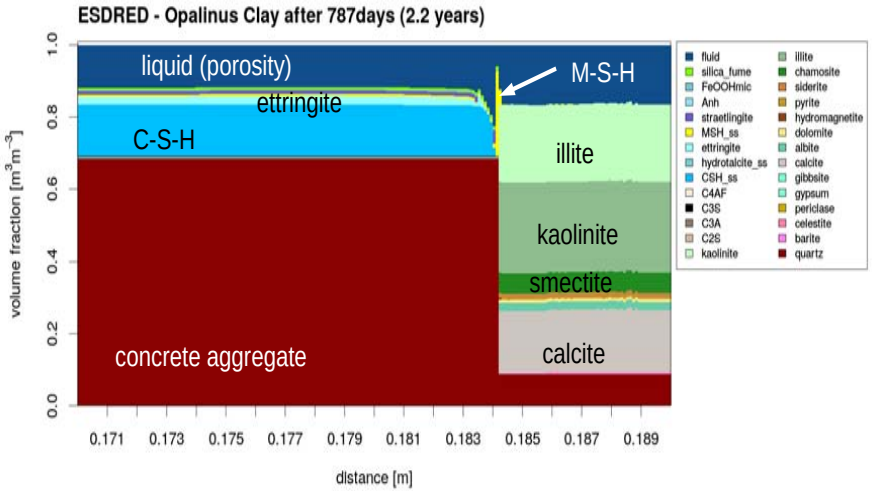
e.g. M-S-H phases (Magnesium replaces Calcium)

Challenges:

- Long term alteration/degradation of materials, involves competing processes on several spatial and temporal scales
- Complex chemistry such as localized dissolution and precipitation of minerals affects the macroscopic transport
- Introducing advanced concepts into numerical codes (e.g. electrochemical transport & chemical reactions, HORIZON 2020 project: CEBAMA PhD Hax Damiani)



Example: High resolution reactive transport simulations of concrete-clay interfaces from Mont Terri CI experiment

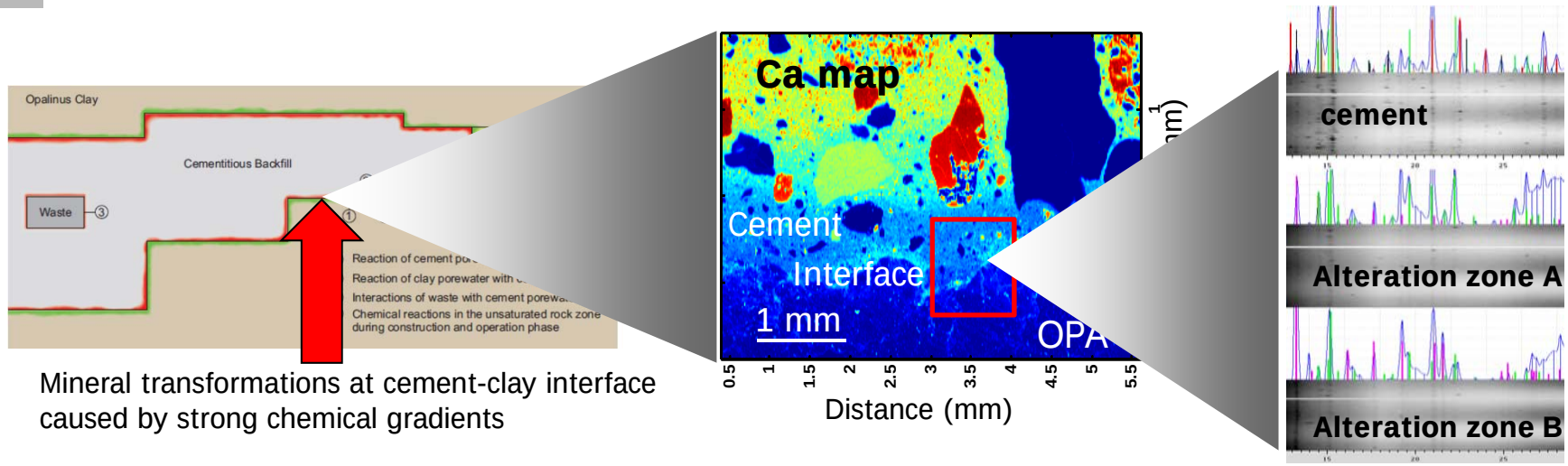


Micro-scale characterization at interfaces

Aim: Development of a synchrotron-based methodology to determine the mineral composition with micro-scale resolution

Example: Mineral composition at the cement-clay interface

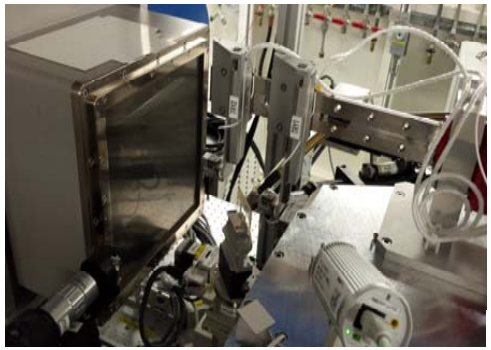
- Characterization of interfaces using micro-diffraction (micro-XRD)



Mineral transformations at cement-clay interface caused by strong chemical gradients

- Micro-XRD at microXAS@SLS using a 2 x 2 μm^2 beam and rotating the samples (thin sections)
- Identification and quantification of the minerals in the alteration zone between cement paste and clay (OPA)

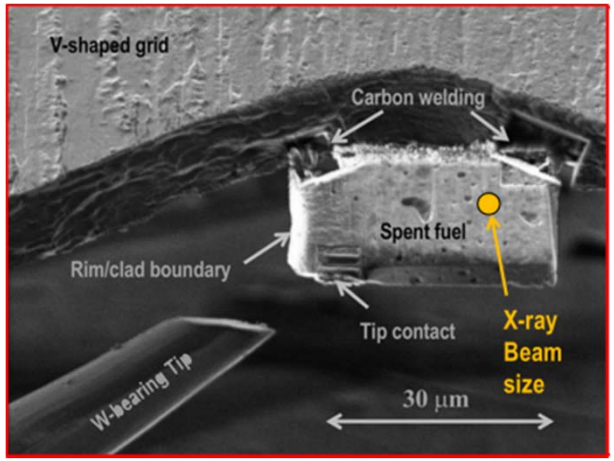
microXRD set-up



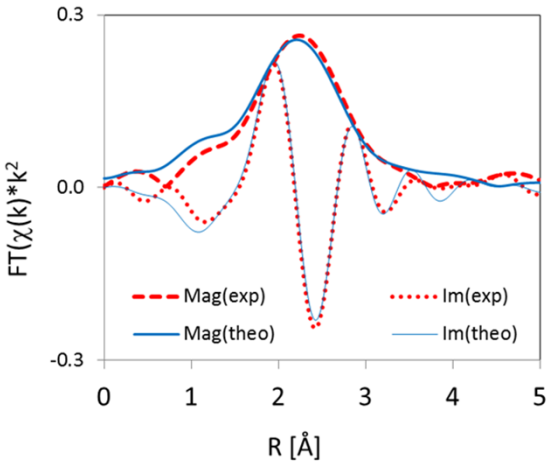
SA: Verification of results from geochemical modelling of the long-term interaction of cement paste and clay

The chemical state of ^{79}Se in spent nuclear fuel (LES/AHL/LRS/SYN – microXAS SLS)

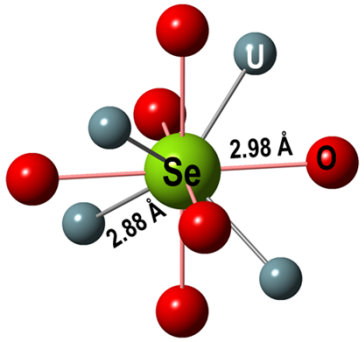
- Selenium originating from fission in light water reactors is tightly bound in the crystal lattice of UO_2
- Contrary to previous assumptions the safety-relevant radionuclide **^{79}Se** will be released at extremely low rates during aqueous corrosion of the waste in a deep-seated repository.
- Positive consequences for the safety assessment of high-level radioactive waste repository planned in Switzerland



SEM picture of a spent fuel sample prepared by Focused Ion Beam (FIB) milling.



EXAFS fitting

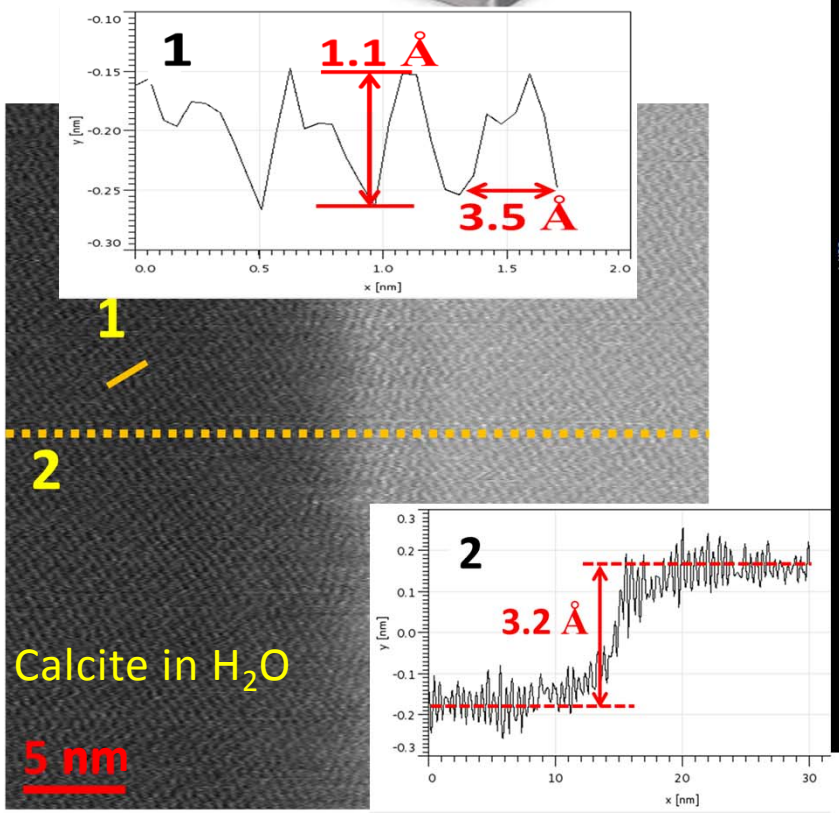


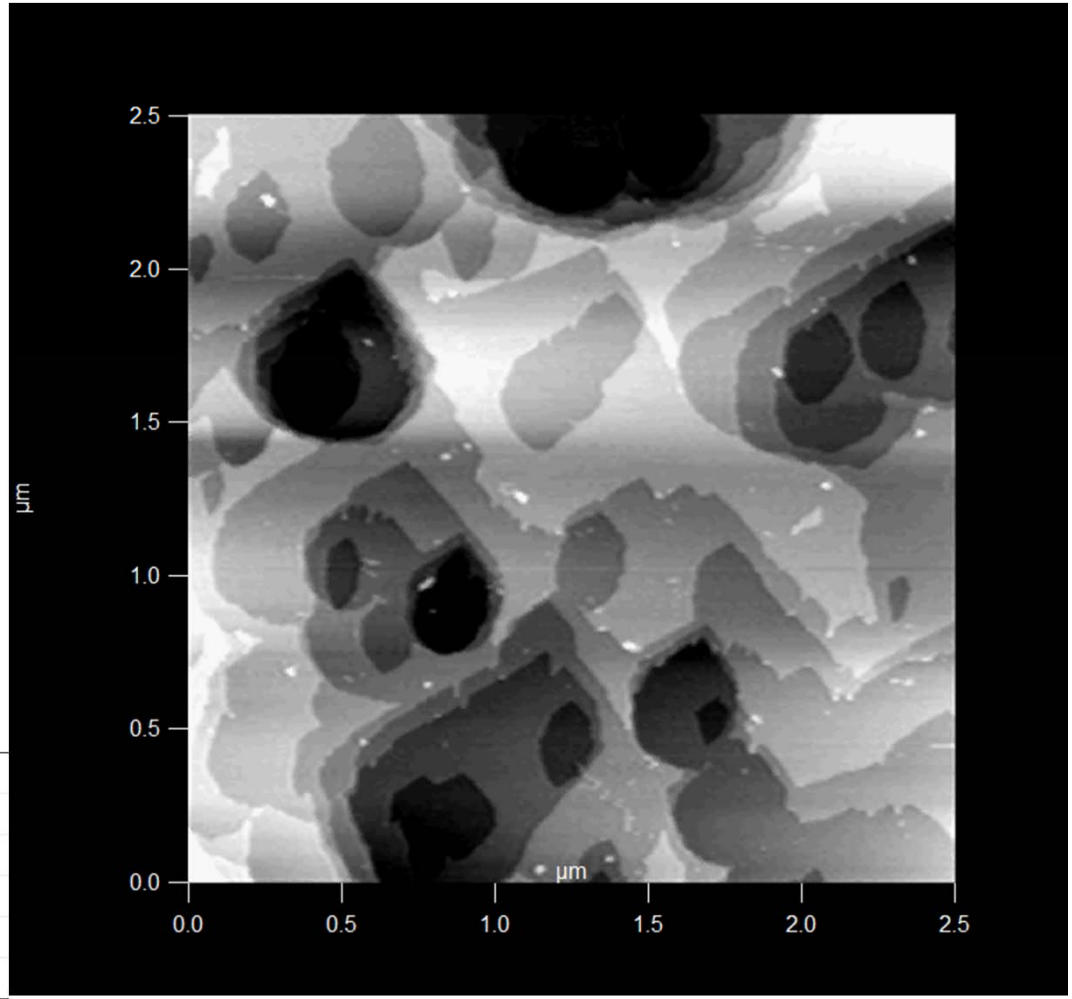
Optimized coordination environment of Se in UO_2

PAUL SCHERRER INSTITUT

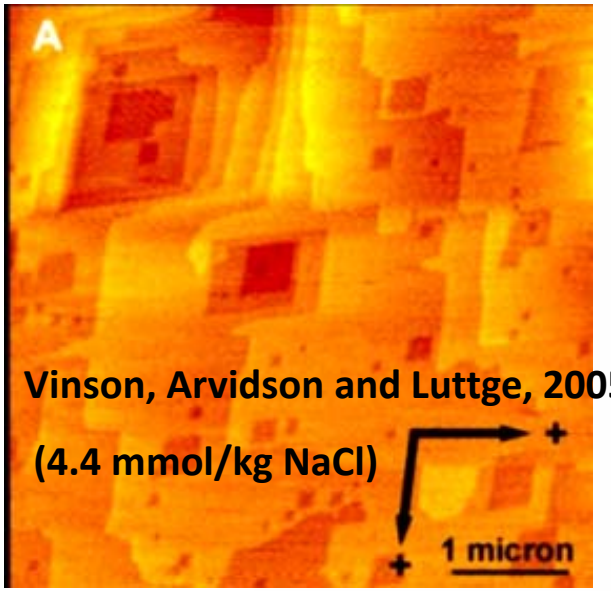
UNIVERSITÄT BERN

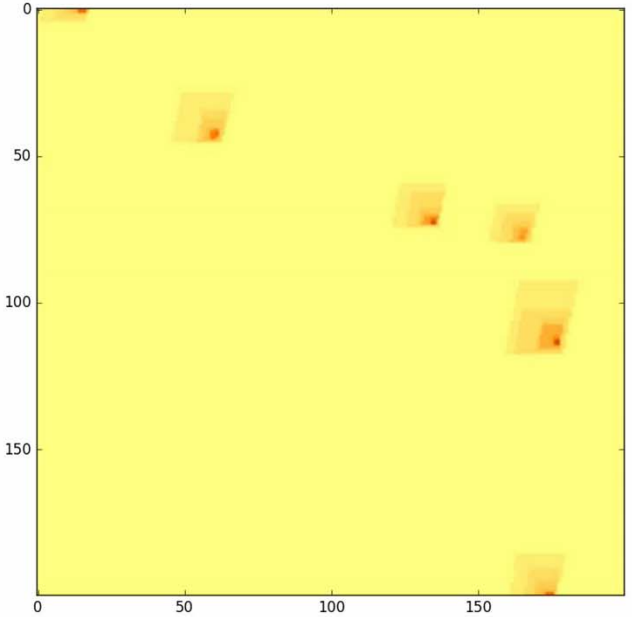
In situ observation and modelling of mineral kinetics

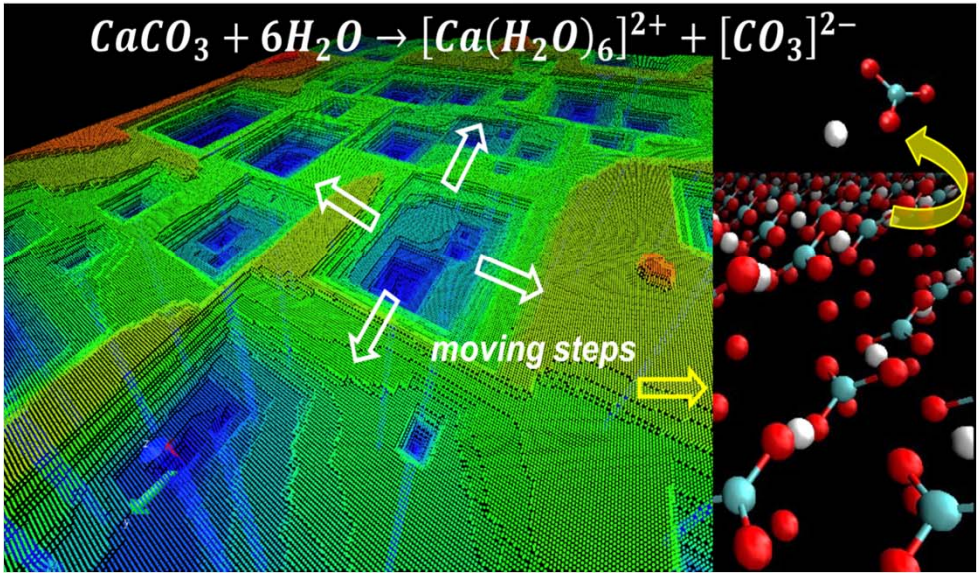


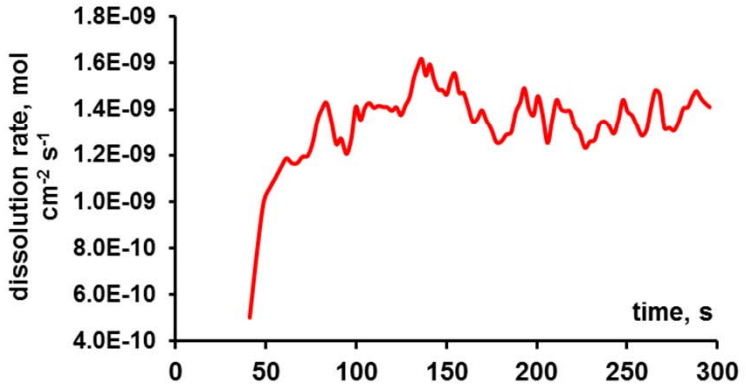


In situ observation and modelling of mineral kinetics









Kurganskaya (2016)

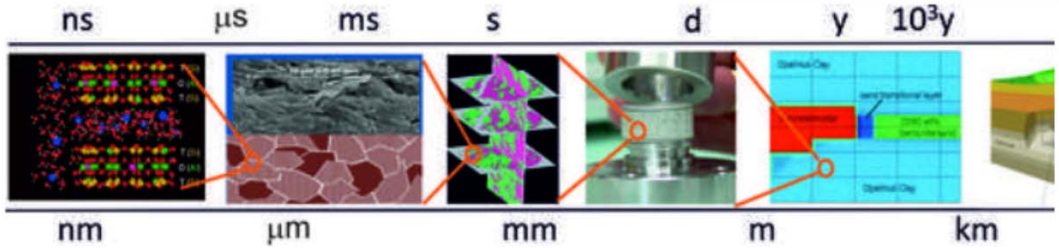
www.psi.ch/LES

LES

PSI Home » LES

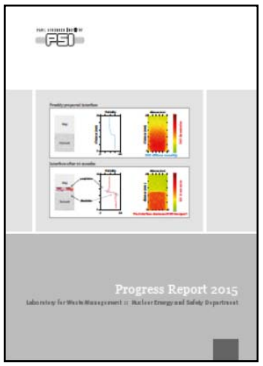
EDUCATION & JOBS EVENTS

- About LES
- Team
- Groups
- Projects
- Teaching and Education
- LES Events
- Software and Database
- Science Explained
- Scientific Highlights
- Publications
- Annual Reports



Laboratory for Waste Management (LES)

LES is the Swiss competence center for geochemistry and multi scale radionuclide and mass transport in argillaceous rocks and cement and their applications to deep geological systems and Swiss radioactive waste repositories.



PAUL SCHERRER INSTITUT

PSI

Wir schaffen Wissen – heute für morgen