PAUL SCHERRER INSTITUT PSI

UNIVERSITÄT BERN

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

Labor für Endlagersicherheit

PSI 24.10.2017

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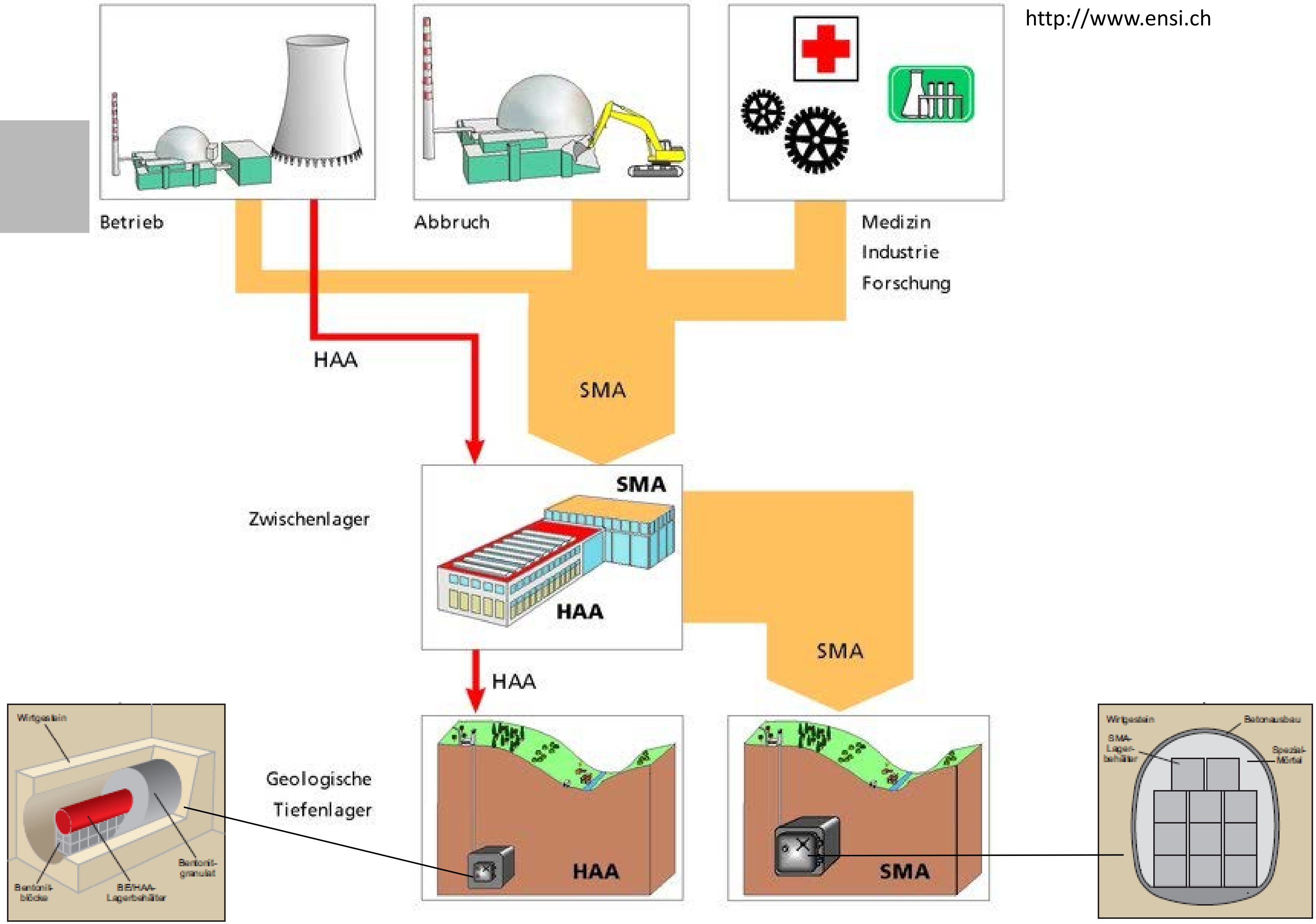
Outline

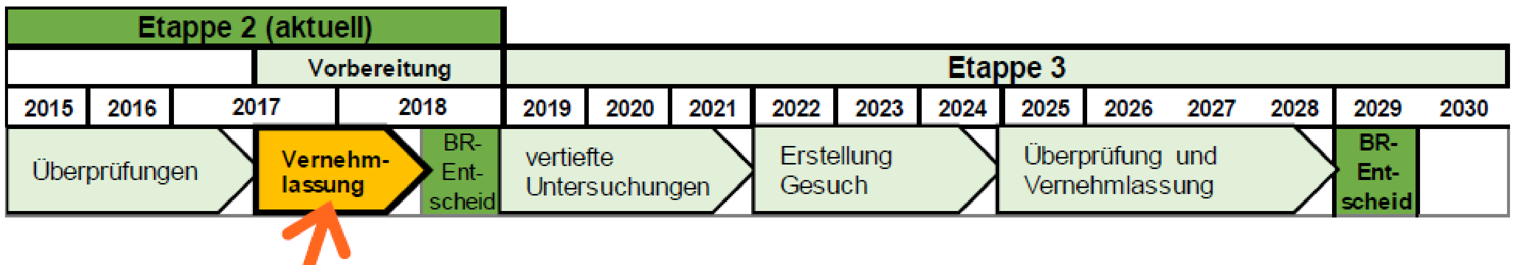
- Status Swiss waste disposal program
- Laboratory for Waste Management (LES)
  - Mission
  - Organizational chart
  - Important infrastructure and capabilities
  - External funding
- Selected research projects
  - Transport and retardation of dose determining nuclides
  - Reactivity of waste and material interfaces
  - Competitive sorption an transport in compacted systems
  - Thermodynamics at elevated temperatures
- Summary

Origin of Nuclear Waste and Disposal Concepts

http://www.ensi.ch



ABSCHLUSS ETAPPE 2, AUSBLICK ETAPPE 3



Vernehmlassung Etappe 2

- Ab Ende 2017, während 3 Monaten
- Offen für Kantone, Nachbarstaaten, Gemeinden, Organisationen, Bevölkerung
- Aufgelegte Dokumente: Ergebnisbericht (inkl. Objektblätter), Erläuterungs-bericht, Berichte der Nagra, alle Prüfungsberichte und Stellungnahmen

Sectoral plan

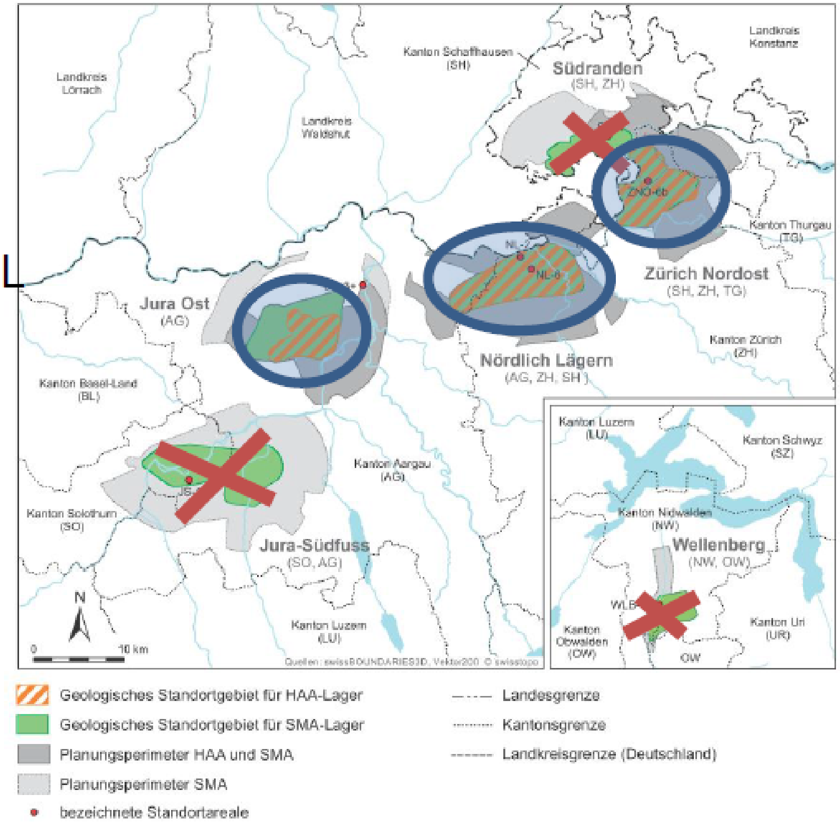
Gesamtbeurteilung ENSI

SMA

- NL hat keinen eindeutigen Nachteil
- Zurückstellung der Standortgebiete WLB, JS, SR wegen eindeutiger Nachteile im Vergleich zu ZNO, JO & NL

HAA

- kein HAA Standortgebiet hat einen eindeutigen Nachteil
- Bestehende Ungewissheiten sind in Etappe 3 SGT reduzierbar
- Kein SG wird zurückgestellt



LES Mission

LES (Labor für Endlagersicherheit) serves the **national needs, present and future**, in providing important parts of the **scientific basis for the safe disposal of radioactive waste**.

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

LES carries out a research programme in the areas of:

- **repository in situ conditions**, their evolution and **repository induced effects** including both **modelling and experimental aspects**.
- Interfacial chemistry and transport of radionuclides in repository systems
- fundamental understanding of system behavior for the **long term predictive modelling** and knowhow transfer.

LES maintains:

- **proper balance between applied** and **basic research**
- **the state-of-the-art expertise and knowledge** in strategic areas
- tight connections to the University of Bern to contributes to the education of young scientists in the field of geochemistry of geological waste disposal.

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**:

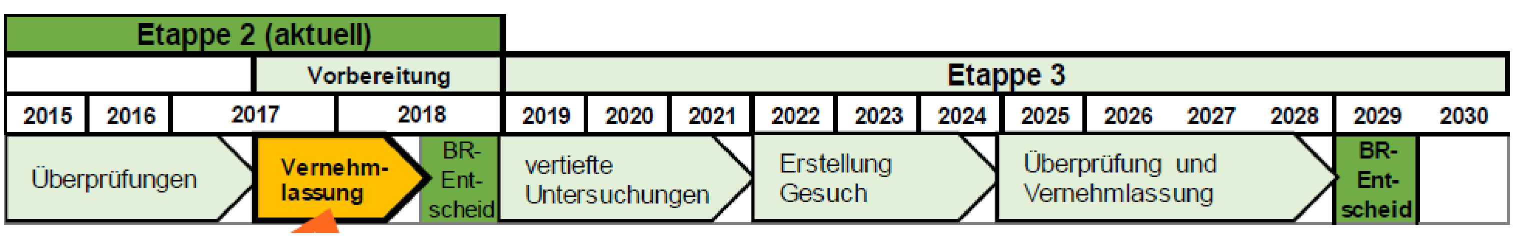
- **Redox sensitive elements** and justification of “chemical analog” arguments
- **Sorption competition / transferability** of data for compacted/disperse systems
- Chemistry of **dose determining radionuclides**

Geochemical evolution of in-situ repository conditions:

- **Production and transport of volatile species**
- **Reactivity of technical barriers** and **long term evolution of their safety function**

Sample characterization from site specific the field explorations

Scientific **documentation for the General License Application**



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LES-organization

Laboratory for Waste Management/Mineralogy UniBe

Churakov S.V.

Secretariat

Gschwend B.

4402 Clay Sorption Mechanisms

Baeyens B.

- Dähn R.
- Marques M.
- Thoenen T.
- Kéri A.-M.
- Wick S.
- Geng G.
- Marafatto F.
- Eltayeb E.
- Schaible A.
- Lösch H.

4403 Transport Mechanisms

Prasianakis N.

- Curti E.
- Gimmi T.
- Kosakowski G.
- Kulik D.
- Miron D.
- Nakarai K.
- Patel R.
- Yang G.
- Yang Y.
- Hax Damiani L.
- Krejci P.
- Luraschi P.
- N.N.
- Gatschet M.

4404 Diffusion Processes

Van Loon L.

- Glaus M.
- Pfingsten W.
- Frick S.
- Bunic P.

4406 Cement Systems

Wieland E.

- Tits J.
- Hummel W.
- Cvetkovic B.
- Geng G.
- Nedyalkova L.
- Mancini A.
- Kunz D.
- Laube A.

Uni Bern IfG Mineralogy

Churakov S.V.

- Adams D.
- Armbruster T.
- Cametti G.
- Doebelin N.
- Eggenberger U.
- Fisch M.
- Kurganskaya I.
- Weibel G.
- Schliemann R.
- Lemp C.

Legend:

- Guest Scientist, Post Doc
- Technician
- PhD student
- MSc student

Recent third party funded projects

- **EU-Horizion 2020:** *“DISCO: Spent fuel dissolution & chemistry in container Thermodynamic equilibria calculations in SF.”* E. Curti

- **EU-COFUND 2Y+1Y Postdoc :** “*Alkali-silica reaction in concrete*.”
- **EU-COFUND 2x2Y Postdoc :** “*Dissolution precipitation in porous media*.”

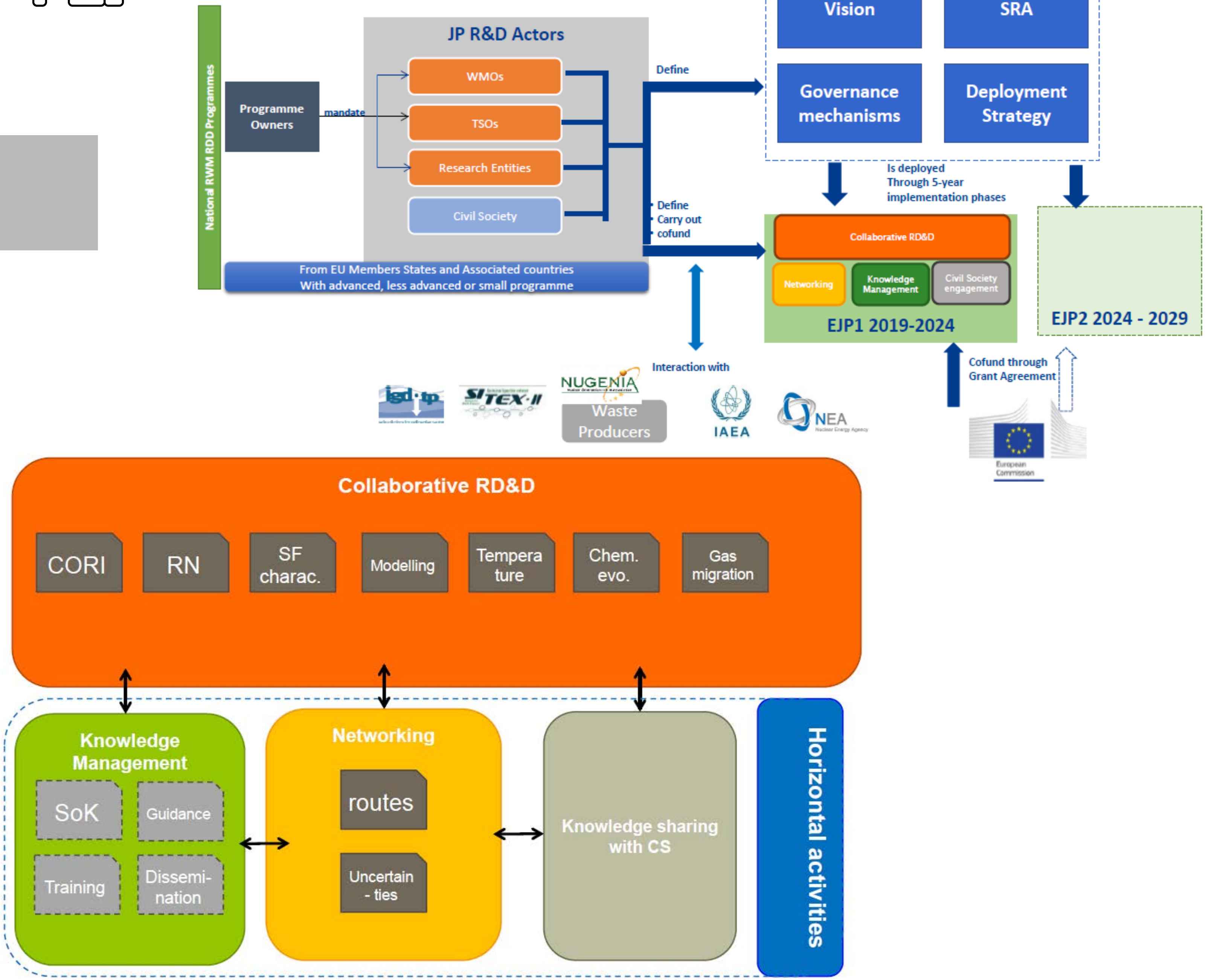
- **4Y SNF PhD**: “*Resolving dissolution precipitation processes in porous media: Pore scale lattice Boltzmann modelling combined with synchrotron based X - ray characterization*. “

- **3Y SNF PhD project :** “Transport of sorbed species in clays”

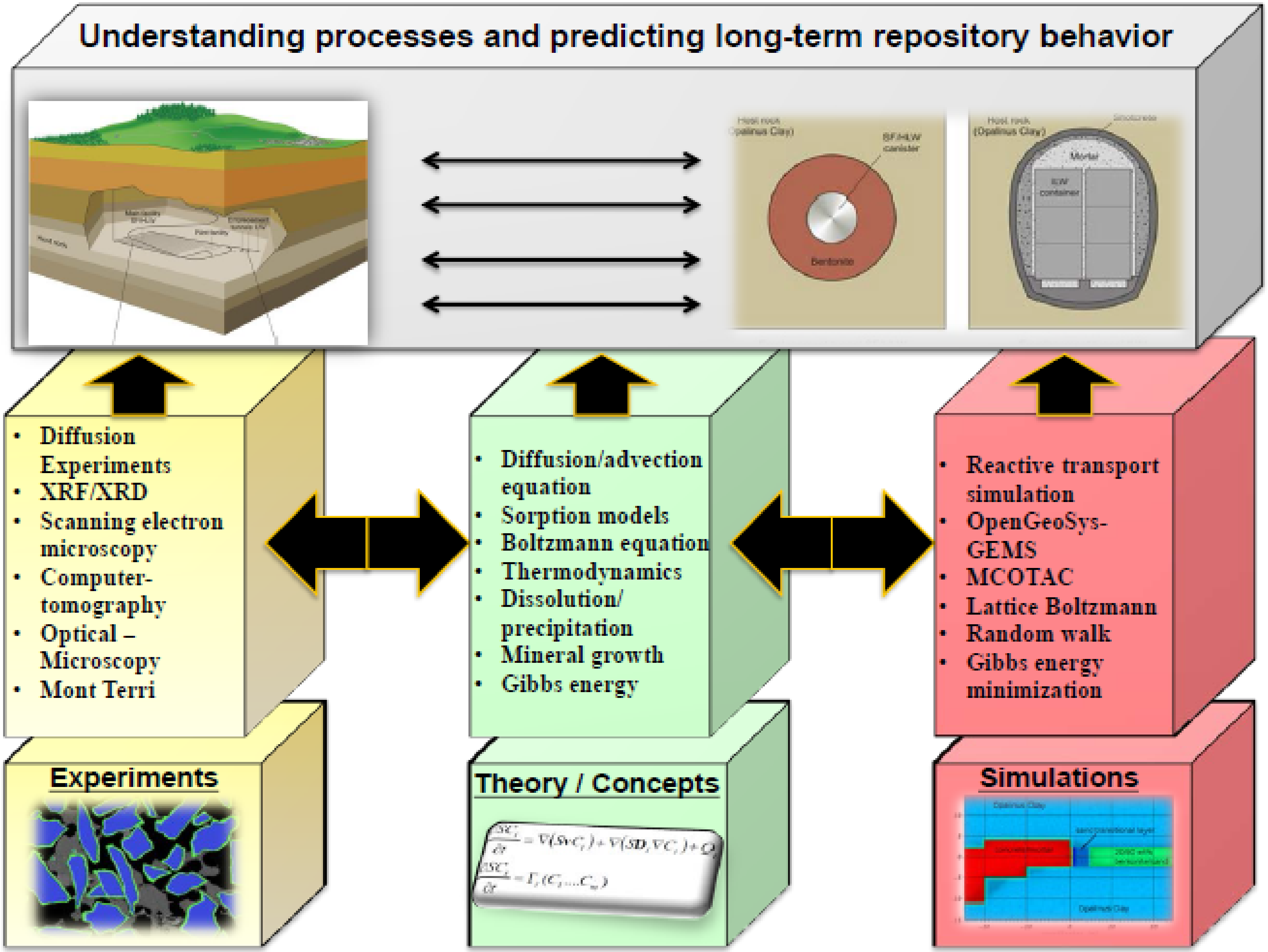
- **CROSS-PSI postdoc** “Cryo-microspectroscopy at the microXAS beamline for the investigation of redox- and radiation-sensitive samples

- **HPC projects at CSCS**: 400`000 Node/Hours (equiv. ~300KCHF)

EURATOM call: EJP1



Consolidation of modelling and experimental activities



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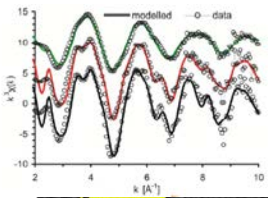
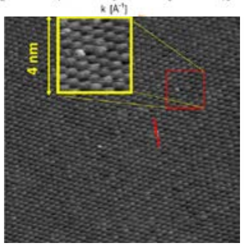
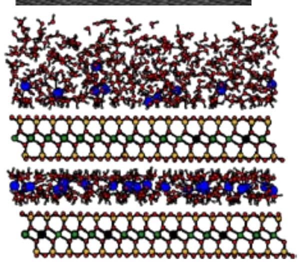
Mass transport in heterogeneous geochemical systems

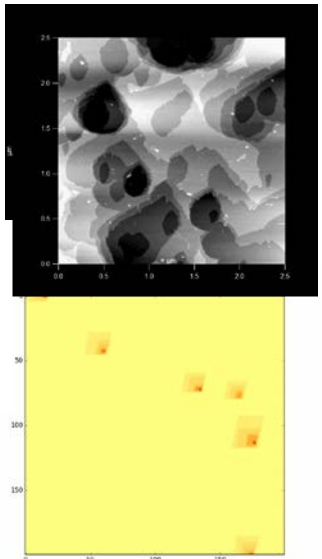
Down scaling

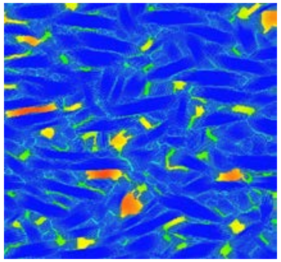
Atomic scale



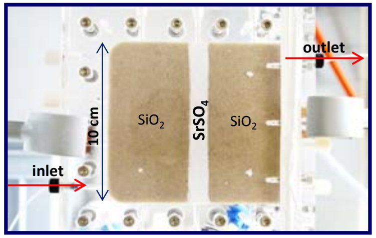


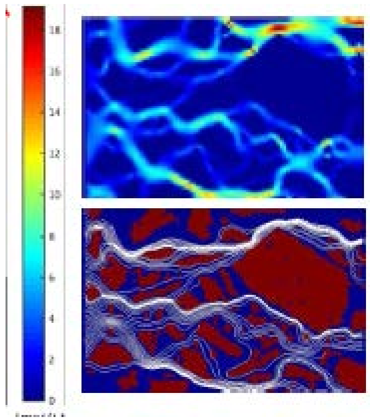






- Ions sorption and transport mechanisms at atomic scale





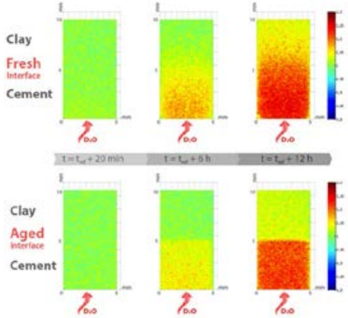
SCIENTIFIC REPORTS

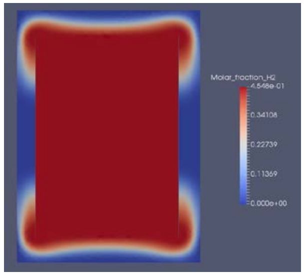
nature publishing group npg

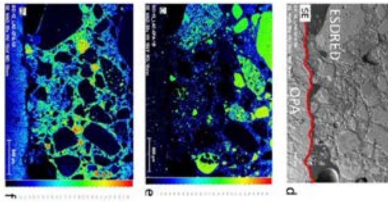
Scientific Reports

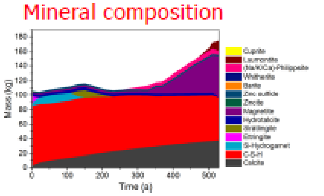
- www.nature.com/articles/s41598-017-14142-0
- Pore scale mass reactive mass transport
- Mechanistic description of mineral fluid interaction

- Characterization of material interfaces









- In situ characterization of materials interfaces
- reactive transport simulations and experiments

Field Scale

Up scaling

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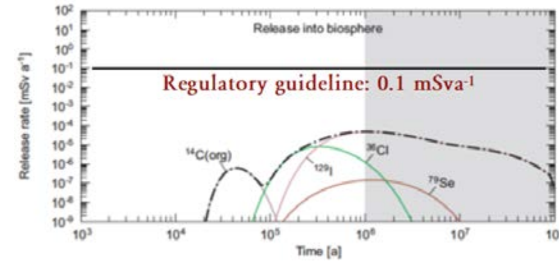
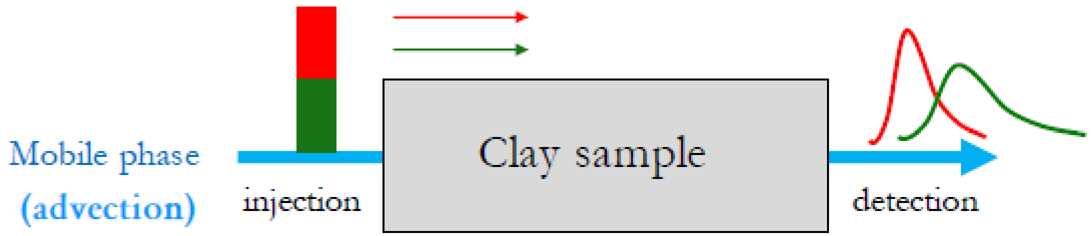
nagra

PhD thesis 2017

APPROVED



Setup: reference tracer+LMWOC



Can we determine the weak retardation?

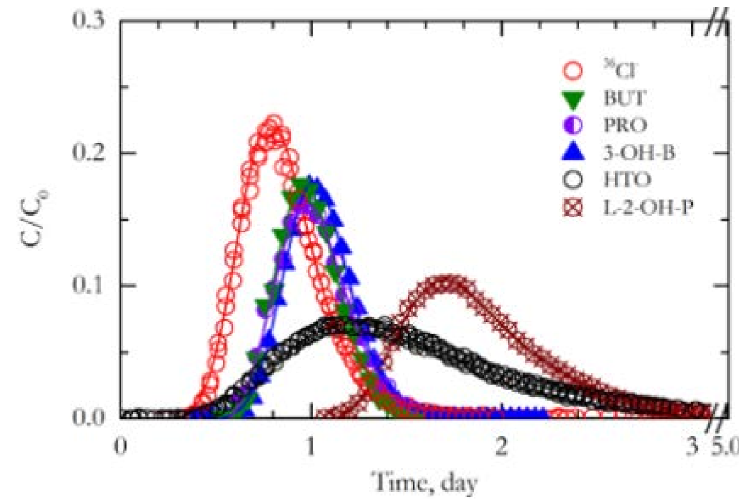
- Yes! (reproducibility, mass balance, sensitivity)

Do we understand the retardation?

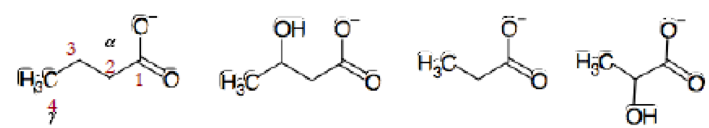
- To some extent! (anion exclusion, anion sorption)

Molecular level of structures?

- structure specific (functional group, aliphatic/hydroxylated, etc)
- clay specific (surface properties, etc)
- possible interactions



Reaction	
R1. $>AlOH_2^+ + Cl^- \longrightarrow >AlOH_2\text{-}Cl$	Ion pair
R2. $>AlOH + Cl^- \longrightarrow >AlOH...Cl^-$	H-bond
R3. $>AlOH + R\text{-}COO^- \longrightarrow >Al(OOC\text{-}R) + OH^-$	Anion exchange
R4. $>AlOH_2^+ + R\text{-}COO^- \longrightarrow >AlOH_2\text{-}(OOC\text{-}R)$	Ion pair
R5. $>AlOH + R\text{-}OH \longrightarrow >AlOH...OH\text{-}R$	H-bond

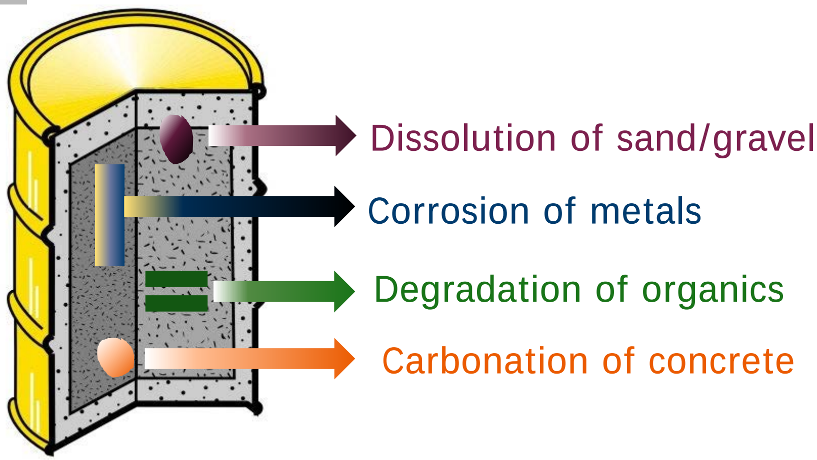


BUT 3-OH-B PRO 2-OH-P

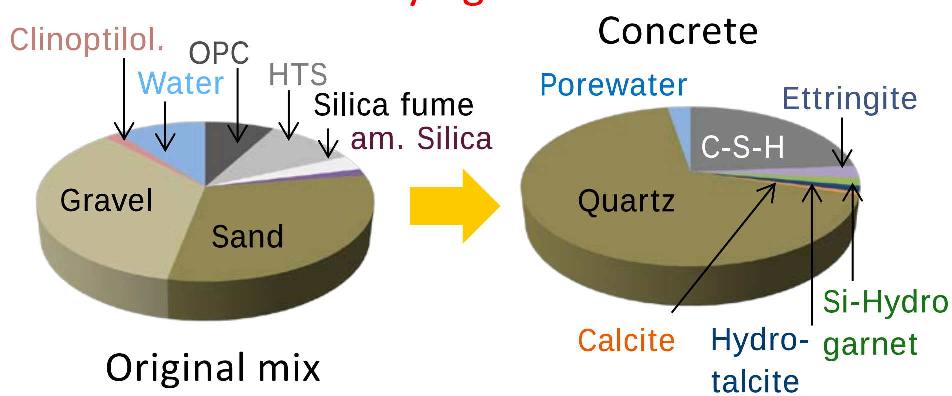
Interaction of waste with engineered barrier

Aim: Geochemical modelling of the temporal evolution of cement-stabilized waste sorts: Changes in the mineralogy of the conditioned waste and volume of waste package

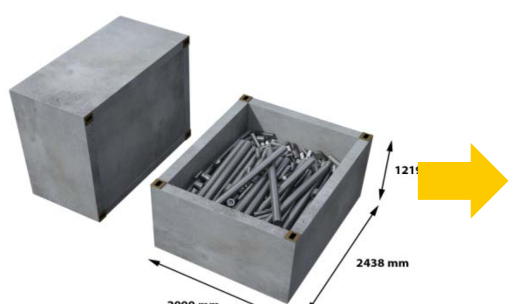
- ❑ Modelled waste sorts: three operational and two decommissioning waste sorts



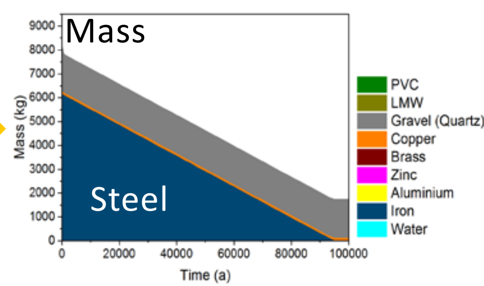
- ❑ Modelling of the initial composition of the solidifying concrete



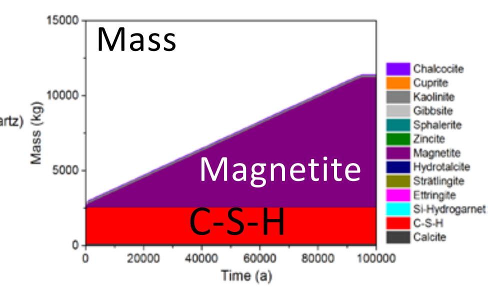
- ❑ Modelling of temporal evolution (waste materials, mineral composition, volume)



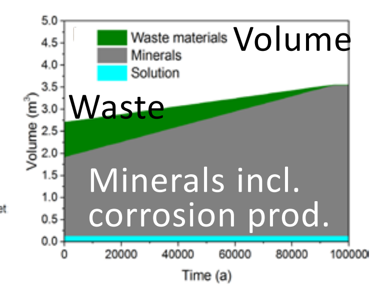
Decommissioning waste



Waste materials



Mineral composition

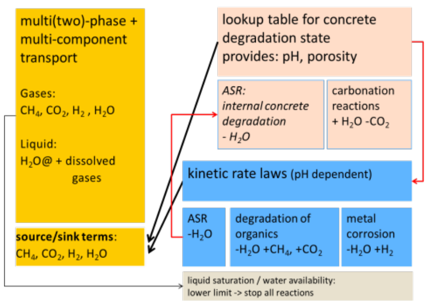


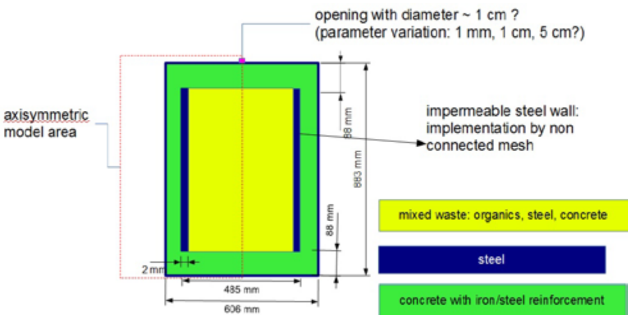
Volume

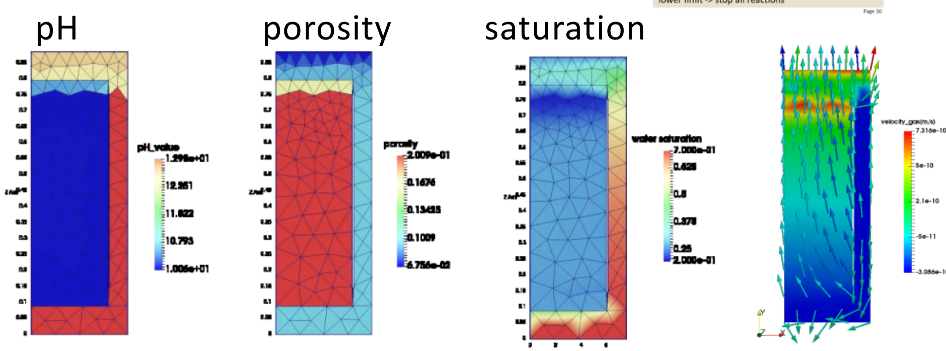
Influence of concrete degradation on gas- and water fluxes in a waste package

Aim: Modeling of interactions between chemical processes and multi-phase (gas/water) multi-component (several gases) in repository near-field

- Represent concrete degradation (carbonation and alkali-silica-reactions) by a look-up table that provides pH and source-sink terms for coupling with multi-phase transport codes
- pH dependent kinetic laws for metal corrosion







- **Application example:** Gas generation and humidity evolution in a waste package during 40 years of intermediate storage. (in cooperation with Helmholtz Institut für Umweltforschung UFZ)

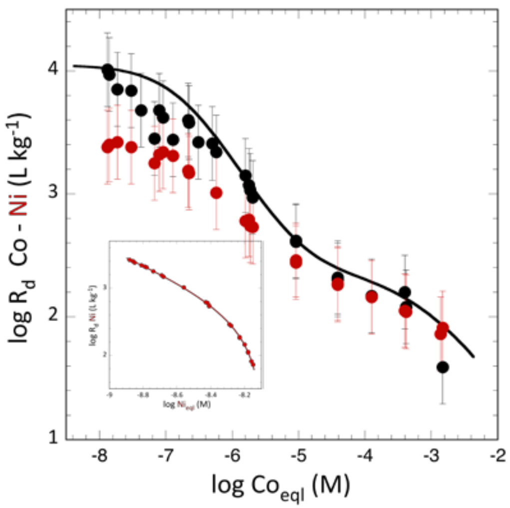
Competitive metal sorption on clay minerals

Motivaton: In a deep geological repository stable aqueous metals are present from different sources: backfill materials, the corrosion of steel canister, dissolution of the waste forms.

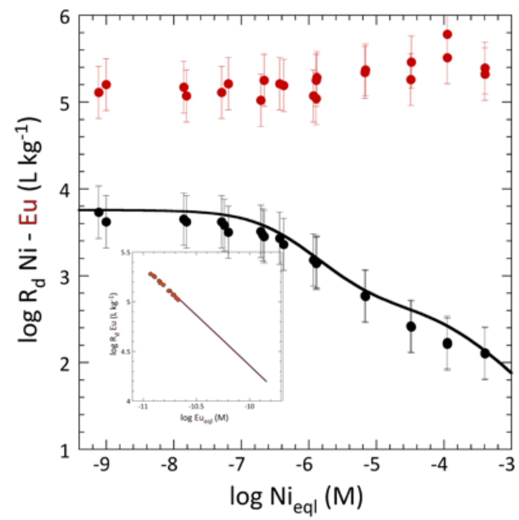
These metals can potentially compete with the released radionuclides for the available sorption sites and reduce their uptake on them.

Experimental:
Na-montmorillonite
Trace metal: Ni, Eu < 10^{-8} M
Blocking metal: Co, Ni: 10^{-8} – 10^{-3} M
pH = 7
Reaction time: 7 days

Results and Modelling



Ni^{II}-Co^{II} are competitive



Eu^{III}-Ni^{II} are non competitive

Conclusions:

- Metals with the same valence and hydrolysis behaviour compete: *e.g.* Ni^{II}-Co^{II} and Eu^{III}-Am^{III}
- Metals with different valence are non competitive: *e.g.* Eu^{III}-Ni^{II}; Th^{IV}-Ni^{II}; U^{VI}-Ni^{II}
- Competitive sorption can be quantified by sorption models (2SPNE SC/CE) and can be taken into consideration in the safety analysis of radioactive waste repositories.

NTB's Sorption Data Base

NAGRA NTB
October 2017

The Development of a Thermodynamic Sorption Data Base (TSDB) for Illite and the Application to Argillaceous Rocks
(in press)

M.H. Bradbury and B. Baeyens

NAGRA NTB
October 2017

The Development of a Thermodynamic Sorption Data Base (TSDB) for Montmorillonite and the Application to Bentonite
(in press)

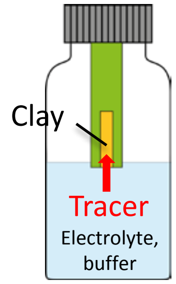
B. Baeyens and M.H. Bradbury

Effect of clay compaction on the transport properties of Eu(III) in illite

Aim

- To compare sorption data for Eu(III) in illite derived from diffusion experiments with those obtained in dispersed clay suspensions.
- To derive a speciation-based transport model for Eu(III) in compacted illite.

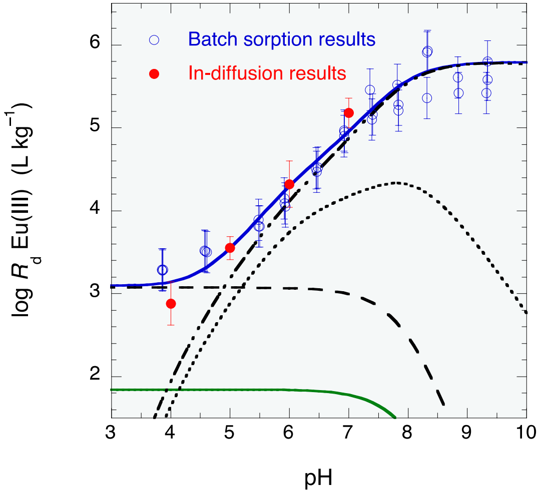
The in-diffusion technique



1. Equilibrate clay with electrolyte solution.
2. Add tracer (e.g. Eu(III)) to solution phase.
3. Monitor solution concentration of tracer as a function of time.
4. Measure tracer profile in clay at the end of the experiment.

=> Unambiguous information on sorption and diffusion properties

Results and simulations



- The results of sorption measurements of Eu(III) in compacted and dispersed illite are fully consistent.
- Sorption modelling (blue) of diffusion-derived data can be done using existing sorption models[a] (black lines, representing contributions from individual surface species).
- A minor mobile surface species (green) is additionally introduced to properly reflect the observed diffusion lengths in the in-diffusion measurements (not shown).

Conclusion

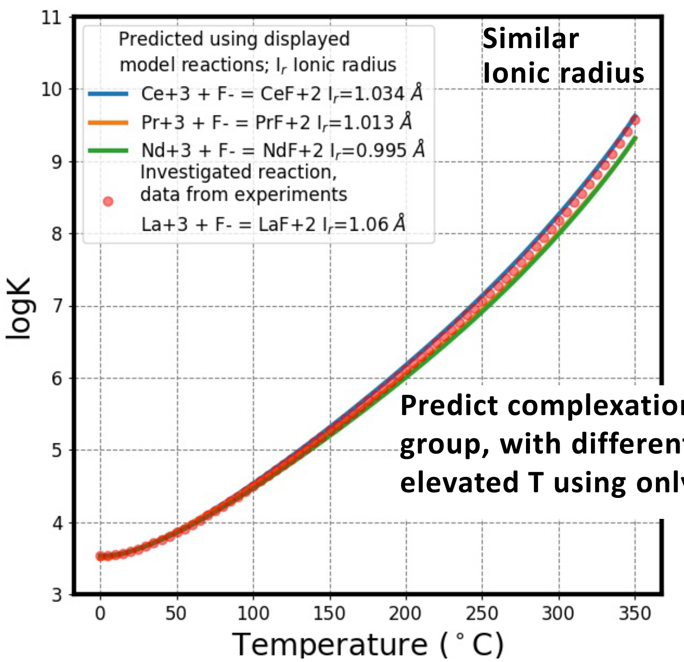
A speciation-based transport model for Eu(III) has been successfully tested for its ability to robustly predict the diffusion length as a function of broad parameter variations, such as pH, ionic strength, Eu concentration and competing species.

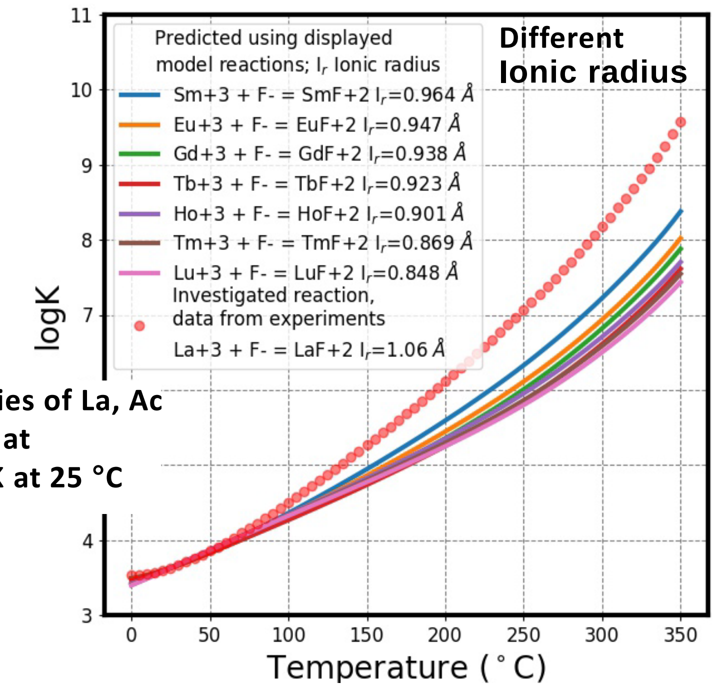
Predict thermodynamic properties at elevated temperature using *isocoulombic* reactions

(1) **Generate** chemical reactions from a given list of substances

(2) **Combine** investigated (unknown temperature effect) and model reactions (known temperature effect) into *isocoulombic* reactions (the same charge types on both sides of the reaction)

(3) **Investigate** which reactions better predict the properties of investigated reactions, knowing only the logK at 25 °C





Predict complexation properties of La, Ac group, with different ligands, at elevated T using only the logK at 25 °C

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www.psi.ch/les

Laboratory for Waste Management (LES)

- About LES
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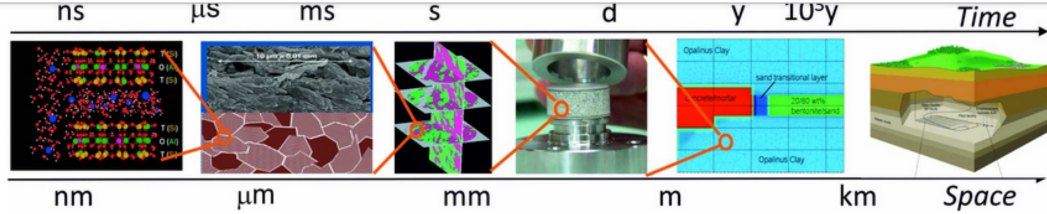
- Research Partners and Cooperations
- Teaching and Education
- LES Events
- Software and Database
- Scientific Highlights
- Hot New Papers
- Publications
- Annual Reports



Laboratory for Waste Management (LES)

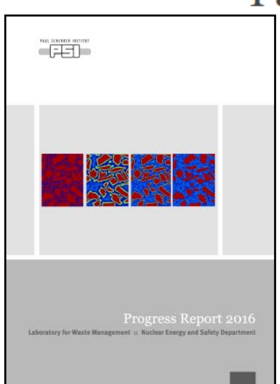
LES is the Swiss competence center for geochemistry and multiscale radionuclide and mass transport in argillaceous rocks and cement and their application to deep geological systems and Swiss radioactive waste repositories. LES offers attractive research projects at the bachelor, master, PhD, and postdoc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



b UNIVERSITÄT BERN

Contact

Paul Scherrer Institut
Laboratory for Waste Management
Prof. Dr. Sergey Churakov
5232 Villigen PSI

Secretary
Beatrice Gschwend
Telephone: +41 56 310 24 17
E-mail: beatrice.gschwend@psi.ch

Contact

University of Bern
Institute of Geological Sciences
Prof. Dr. Sergey Churakov
Baltzerstrasse 1+3
3012 Bern

Homepage NES

Nuclear Energy and Safety Research Division at PSI


Thank you for your attention