



Deliverable D5.3: Training material

WP-Future

The project leading to this application has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 847593.



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Executive Summary

Education and training event on radionuclide transport and retention took place on 17th November 2021. Several senior experts participating to Eurad have covered broad range of topics starting from RN transport and retention to uncertainty analysis in safety assessment calculations educational event which was primary targeting young scientists or researchers from other disciplines who would like to broaden their expertise.

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GEOLOGICAL DISPOSAL CONCEPTS

WP Future Education & Training Webinar

Norbert Maes (SCK CEN)

November 17, 2021

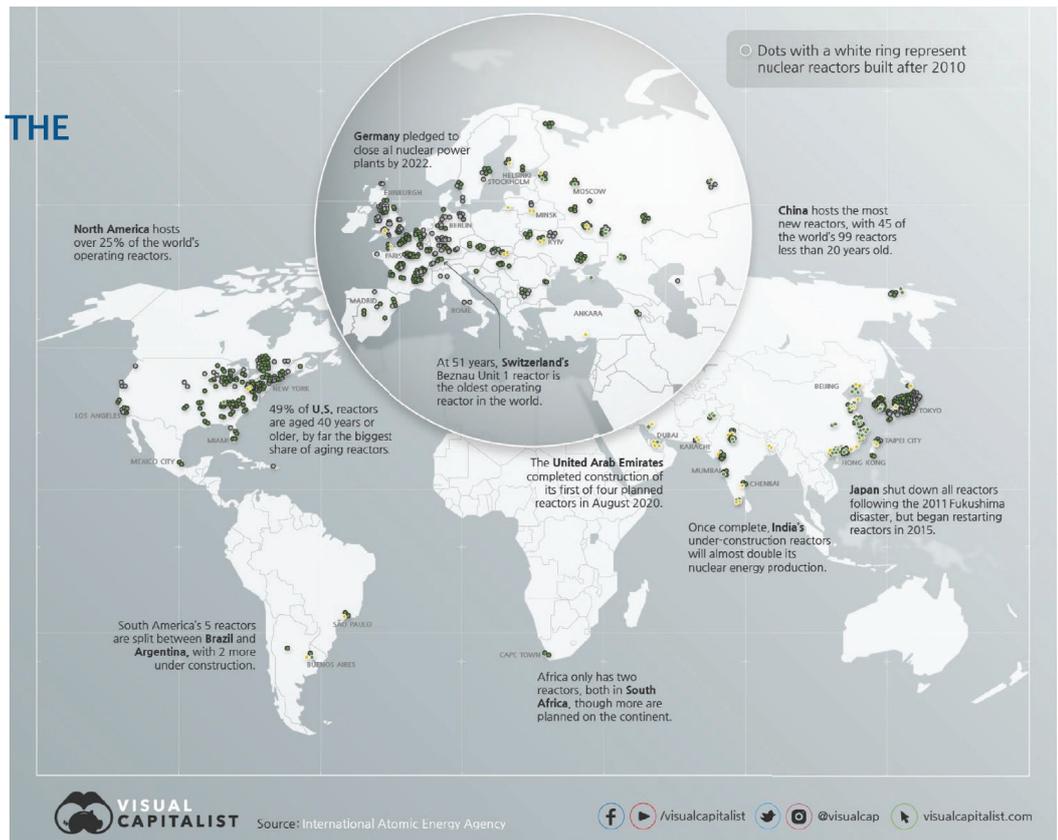
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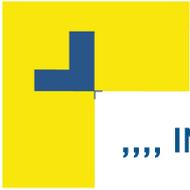
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EURAD WP FUTURE Education & Training webinar



NUCLEAR ENERGY IN THE WORLD

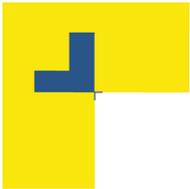
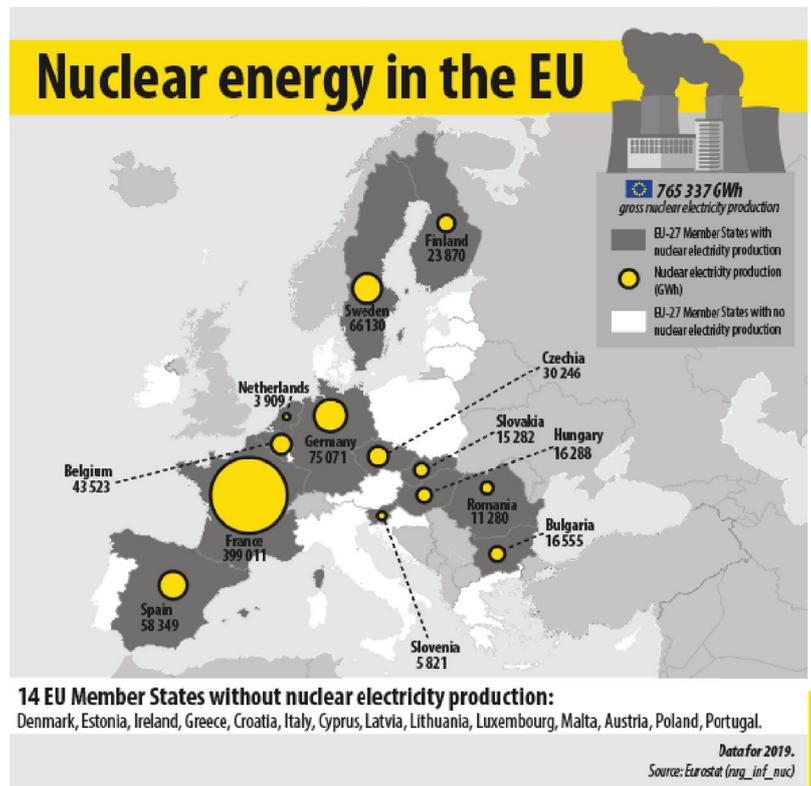




,,, IN EUROPE

Important Issue:

“How to dispose of this waste?”



FUTUR_e

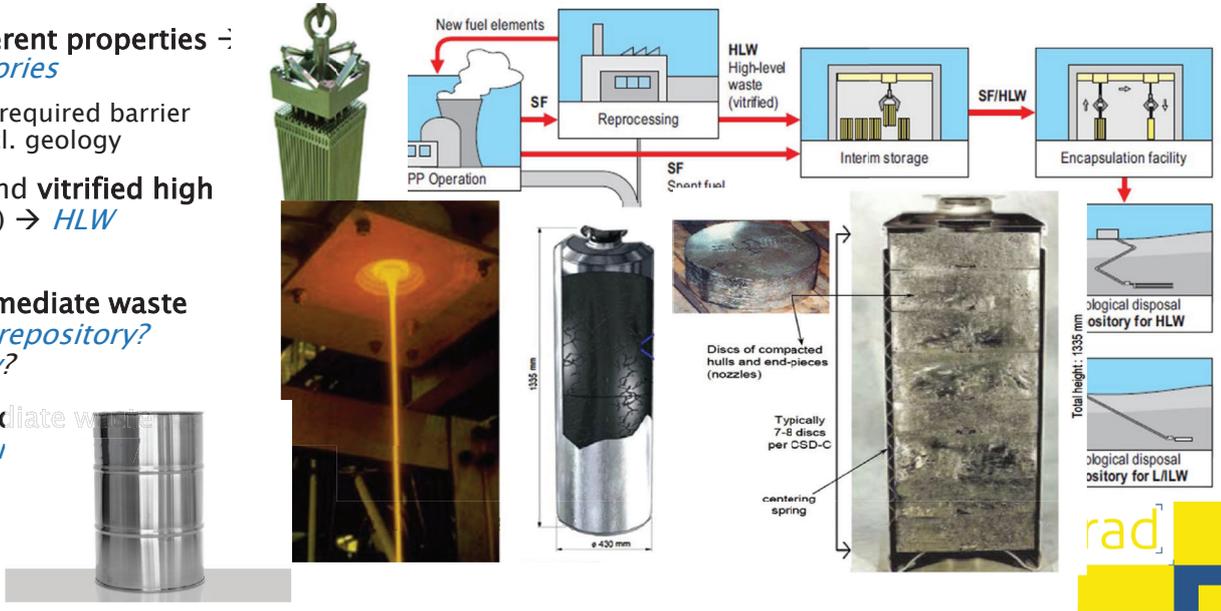
- Nuclear waste types vs. disposal requirements
- Disposal system
 - Safety functions
 - Multi-barrier principle
- Geological disposal
 - What, Why, History
 - Geological disposal concept in “Clay” host rocks
 - Geological disposal concept in “Crystalline” host rocks
 - Geological disposal concept in “Rock Salt” host rocks



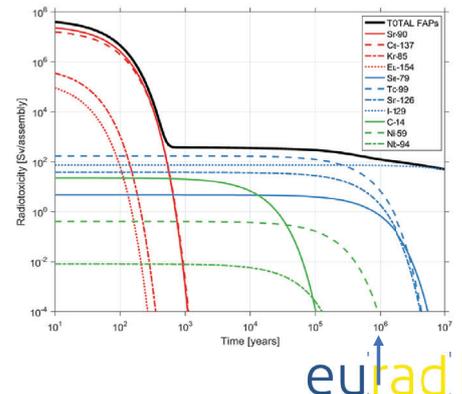
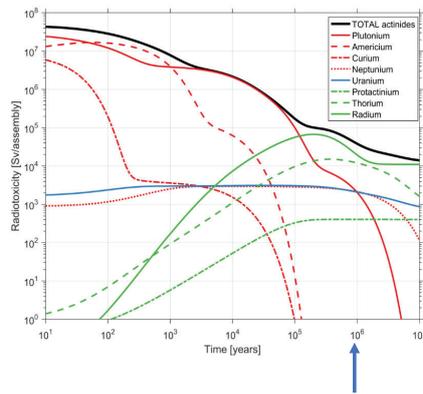
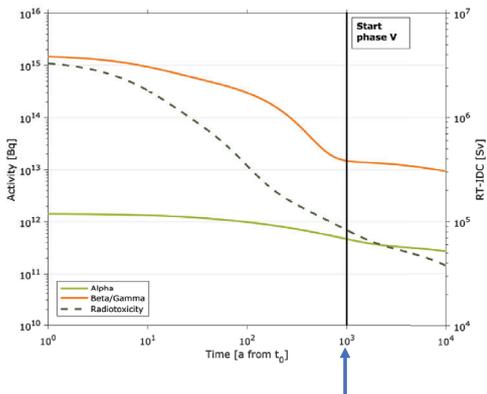
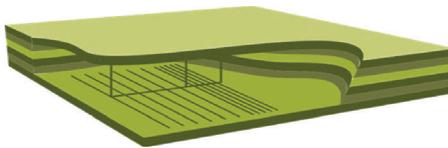


THE NUCLEAR FUEL CYCLE – DIFFERENT WASTE TYPES

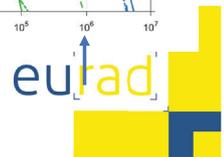
- Wastes with **different properties** → *different repositories*
 - differences in required barrier properties, incl. geology
- Spent Fuel (SF)** and **vitrified high level waste (HLW)** → *HLW repository*
- Long-lived intermediate waste (LL-ILW)** → *HLW repository? L/ILW-repository?*
- Low and intermediate waste (L/ILW)** → *L/ILW repository*

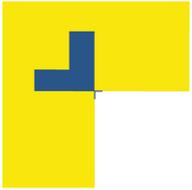


HAZARDS AND TIMESCALES – ex. L/ILW vs. SF&HLW



Source: E. Weetjens, SCK CEN



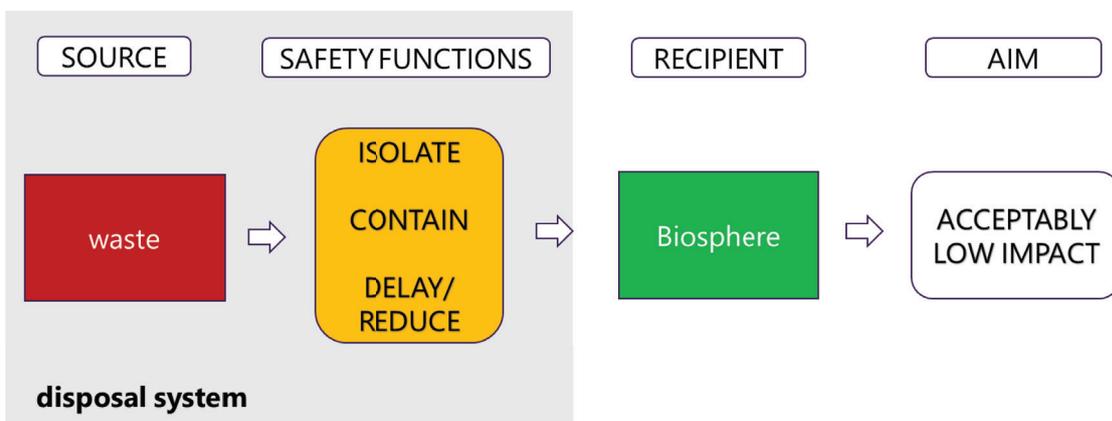


- Nuclear waste types vs. disposal requirements
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DISPOSAL SYSTEM

- Disposal options are designed to **contain** the waste by means of passive engineered and natural features and **isolate** it from the accessible biosphere to the extent necessitated by the associated hazard



DISPOSAL SYSTEM – SAFETY FUNCTIONS

• Isolation

- means design to keep the waste and its associated hazard apart from the accessible biosphere
- It also means design to minimise the influence of factors that could reduce the integrity of the disposal facility

How?

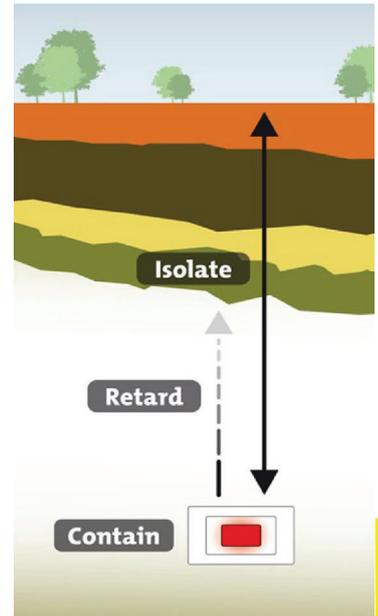
- *Selection of appropriate site and geology*
- *Emplacement at sufficient depth*

• Containment & Retardation

- Implies designing the disposal facility to avoid or minimise the release of radionuclides

How?

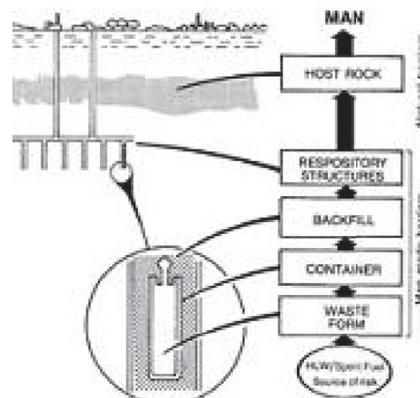
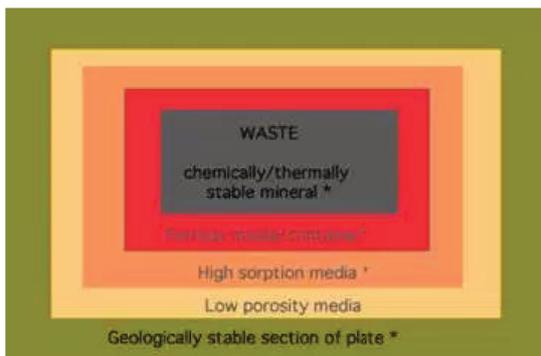
- *Stable and durable waste forms (slow release)*
- *(multiple) engineered barriers that provide **containment** and **retardation***
- *Natural barrier(s) that provide **retardation** (limit water flow, high sorption)*



DISPOSAL SYSTEM – MULTIPLE BARRIER PRINCIPLE

• Multiple barriers principle

- State of matter of the waste (e.g., nuclear fuel pellet)
- Final disposal waste package or canister (e.g. fuel element in container)
- Engineered barrier (e.g., bentonite clay)
- Geological barrier (the geosphere)



DISPOSAL SYSTEM – DEFENCE-IN-DEPTH (DID) PRINCIPLE

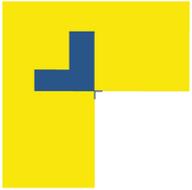
- design and optimise the disposal system according to the *defence-in-depth principle*:

→ The disposal system should be able to fulfill its safety functions despite uncertainties that may threaten their performance

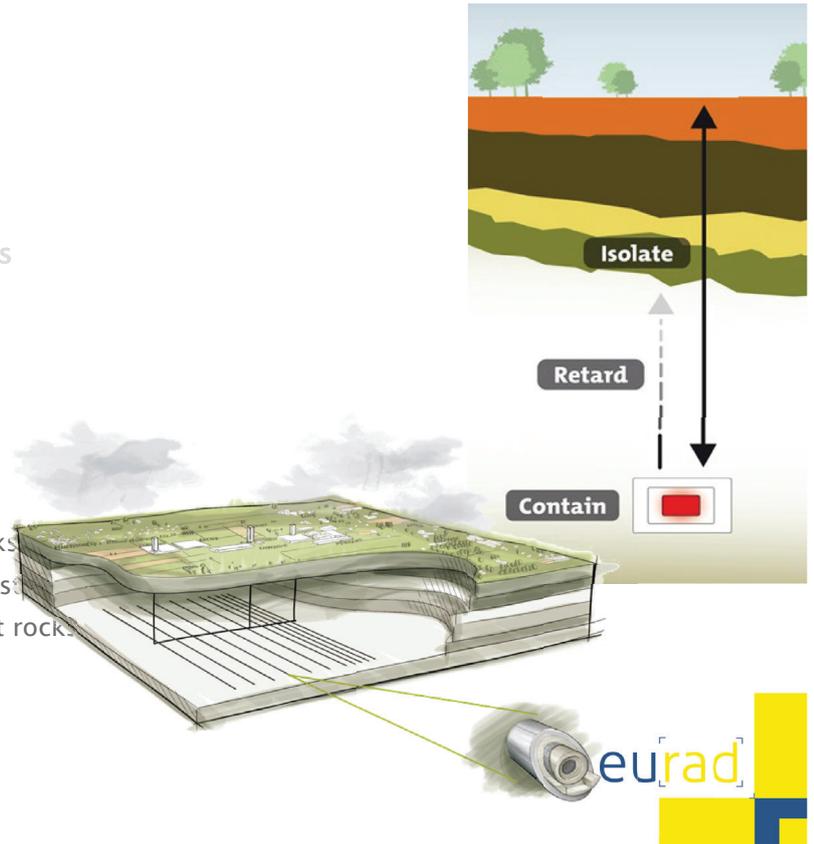
1. Prevention level: avoid uncertainties or their effect
2. Component level: robustness to maintain performance
3. System level: complementary and independent barriers
4. Control level: detect incidents and perform corrective measures

SELECTION OF A DISPOSAL OPTION

- Selection depends on many factors, both technical and administrative, such as:
 - Waste characteristics and inventory
 - The conditions of the country such as climatic conditions and site characteristics, availability of suitable host media
 - Radioactive waste management policy
 - Overall disposal strategy in the country (how many facilities)
 - National legislative and regulatory requirements
 - Political decisions
 - Social acceptance



- Nuclear waste types vs. disposal requirements
- Disposal system
 - Safety functions
 - Multi-barrier principle
- Deep Geological Disposal
 - What, Why, History
 - Geological disposal concept in “Clay” host rocks
 - Geological disposal concept in “Crystalline” host rocks
 - Geological disposal concept in “Rock Salt” host rocks



DEEP GEOLOGICAL DISPOSAL – WHAT?

- **Geological disposal = Burial at several hundred metres depth in stable rock environments**
 - Principle: the deep rock environment is stable and largely unaffected by environmental change for 100.000–1.000.000s years
- **For spent fuel, high-level waste and intermediate-level waste**
- **Provides isolation from man and environment**
- **Research is aimed at**
 - Designing repository systems that will provide good containment
 - Identifying the right types of geological environment
 - Evaluating how RNs will behave over long periods of time in the future





GEOLOGICAL DISPOSAL – WHY?

- Long time scales for decay
- **Ethics/sustainability/security**
 - Polluter pays (this generation pays)
 - Future generations may lack resources
 - Societal breakdown
 - Put beyond use



DEEP GEOLOGICAL DISPOSAL – A BIT OF HISTORY

- Earliest work from USA in 1950s and 1960s on deep salt formations
- EC commissioned work in 1975 to identify potentially suitable rock formations in Europe
 - Hard igneous and metamorphic rocks (granite, gneiss)
 - Clay-rich rocks
 - Salt formations
- Each country develops its own active R&D programme, depending on national geological conditions
- Basic R&D in the field and in the lab augmented by practical tests and (large-scale) experiments in specially constructed underground research facilities (URLs)
 - HADES, Mont-Terri, Bure, Aspö, Onkalo, Grimsel, Gorleben
- Practical implementation slow due to political and social problems

SEDIMENTARY ROCKS (E.G. CLAYS) AS HOST FORMATION

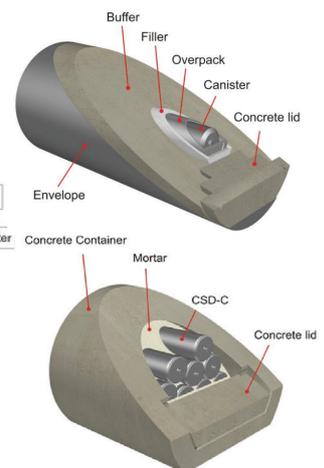
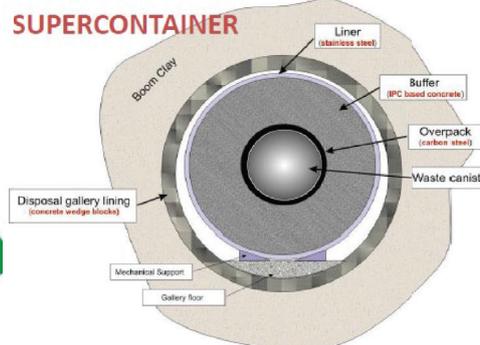
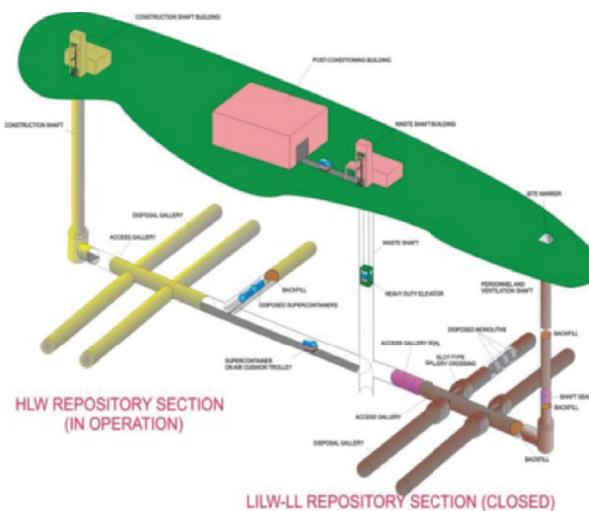
A wide spectrum of argillaceous media* are being considered as potential host rocks for deep geological disposal of radioactive waste, because of their favourable properties:

- thickness, continuity, relative homogeneity
- low hydraulic conductivity
- chemical buffering capacity
- plastic deformation and self-sealing of fractures
- geochemical characteristics that favour low solubility of radionuclides
- high sorption capacity

**from plastic, soft, poorly indurated clays to brittle, hard mudstones or shales*



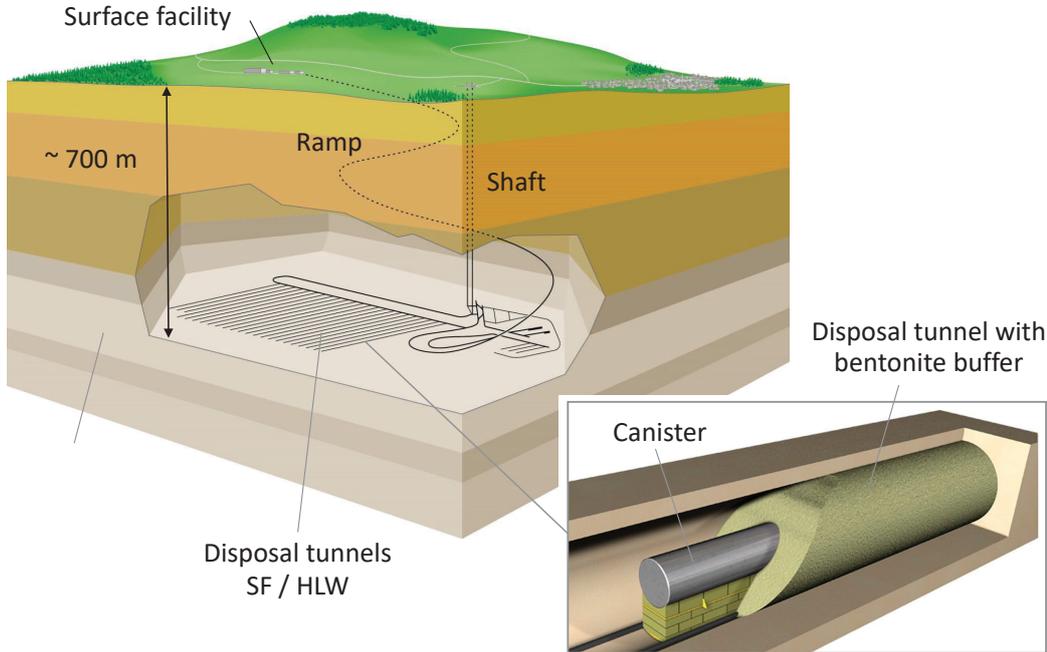
EXAMPLE OF A DISPOSAL CONCEPT IN POORLY INDURATED CLAYS (BOOM CLAY, B) – BY ONDRAF



- Belgian repository design:
 - CEC “contained environment concept” – concrete based “supercontainers”
 - Permanent shielding
 - ...but heavy
 - Shafts only, no ramps

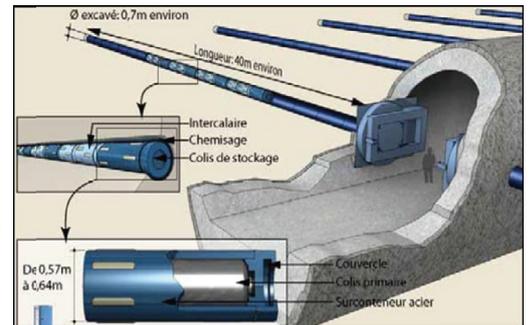
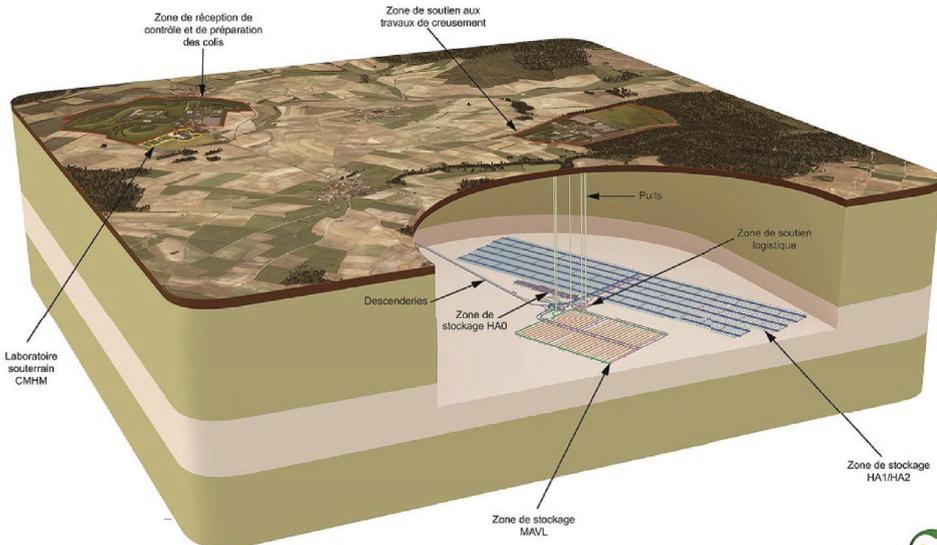


EXAMPLE OF REPOSITORY CONCEPT CLAYSTONE (OPALINUS CLAY ,CH) - BY NAGRA



EXAMPLE OF A REPOSITORY CONCEPT IN CLAYSTONE (CALLOVO-OXFORDIAN CLAY, F) - BY ANDRA - CIGEO PROJECT

Bloc diagramme 3D Cigéo



Echelle des ouvrages non respectée. Pendage des formations géologiques non représenté.





SAFETY OF A SYSTEM WITH CLAY HOST ROCKS

- **Difference of Importance** of individual **Barrier Elements** to achieve sufficient performance of the different **Safety Functions**

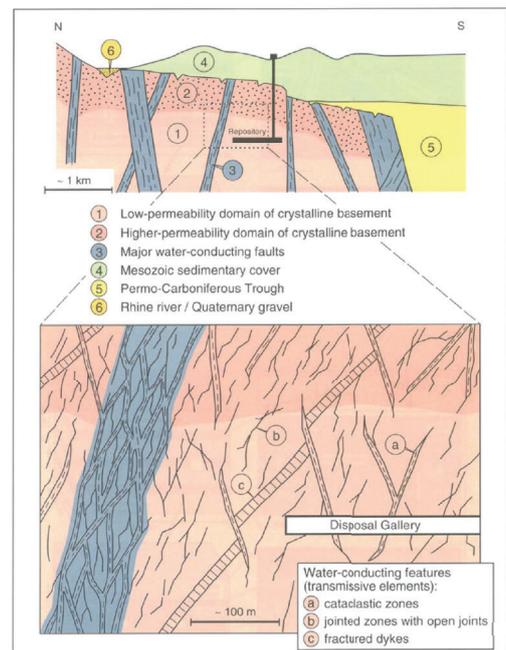
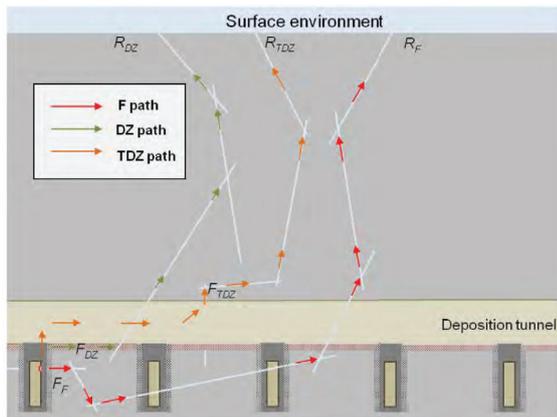
	Waste Matrix	Canister	Buffer/Seals	Host Rock	Geological situation
Immobilisation/ containment	** - ***	**			
Retention & Slow release	***		**	***	
Isolation & Stability			*	**	***

Adapted from P. Zuidema, NAGRA



MAGMATIC AND METAMORPHIC ROCKS (CRYSTALLINE ROCKS) AS HOST FORMATION

- Impermeable, Stable
- Brittle material
 - Fractures!
 - → RN pathways therefore: **Reliance on long-lived canister**



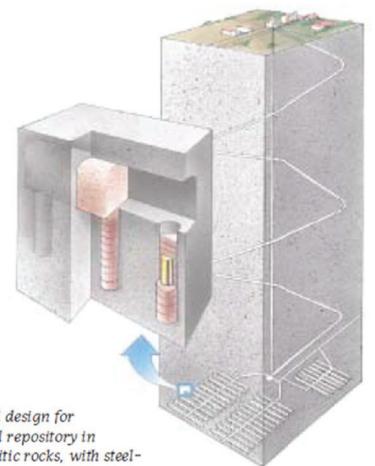
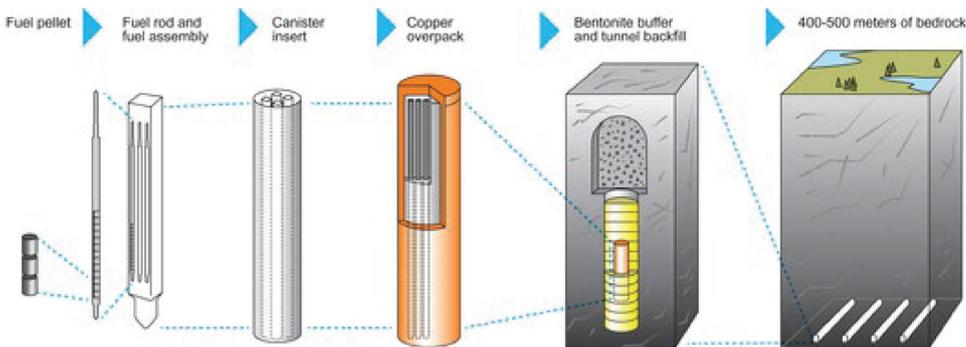


CRYSTALLINE ROCKS – COPPER CANISTERS

- Canister with copper shell – *lifetime up to 1 Mio. years*



REPOSITORY CONCEPT IN CRYSTALLINE ROCKS – KBS-3 CONCEPT USED IN SWEDEN AND FINLAND



Conceptual design for a spent fuel repository in hard, granitic rocks, with steel-copper containers surrounded by compacted bentonite clay (courtesy SKB).

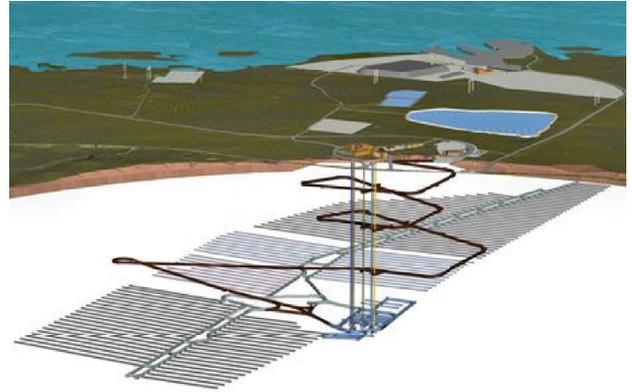
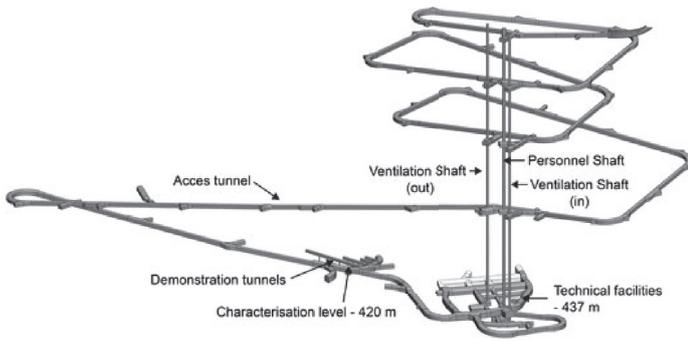
developed by SKB and implemented by POSIVA





DESIGN OF SF REPOSITORY IN FINLAND (OLKILUOTO) BY POSIVA OY – UNDER CONSTRUCTION

Onkalo rock characterisation facility



<http://www.intoeternitythemovie.com/>



SAFETY OF A SYSTEM WITH FRACTURED CRYSTALLINE ROCK

- **Difference of Importance** of individual **Barrier Elements** to achieve sufficient performance of the different **Safety Functions**

	Waste Matrix	Canister	Buffer/Seals	Host Rock	Geological situation
Immobilisation/containment	***	***			
Retention & Slow release	***		**	* - **	
Isolation & Stability			*	*	***

Adapted from P. Zuidema, NAGRA

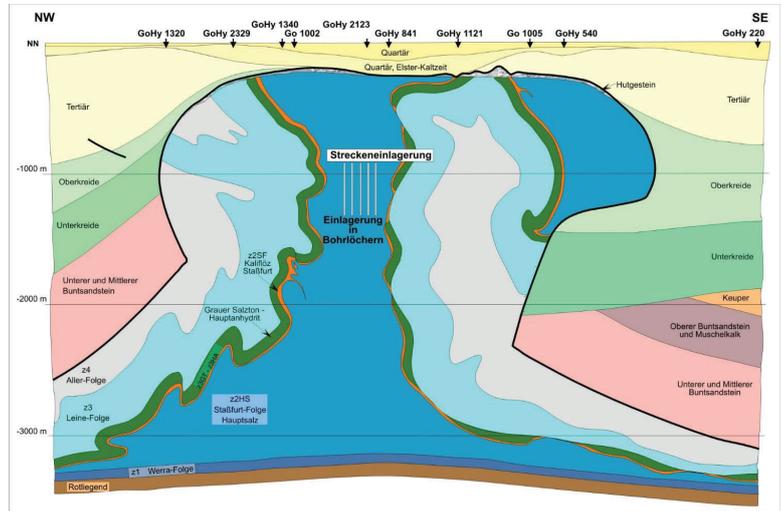


SALT ROCKS AS HOST FORMATION

- Dry environment for construction and underground operation
- High heat transfer
- Impermeable to groundwater
- Self-sealing (salt creep)



Example: Gorleben Salt Dome, Germany (Exploration site)



SAFETY OF A SYSTEM WITH ROCK SALT

- **Difference of Importance** of individual **Barrier Elements** to achieve sufficient performance of the different **Safety Functions**

	Waste Matrix	Canister	Buffer/Seals	Host Rock	Geological situation
Immobilisation/ containment	***	*		***	
Retention & Slow release	***				
Isolation & Stability			*	*	***

Adapted from P. Zuidema, NAGRA

CURRENT STATUS FOR SF/HLW REPOSITORIES IN EUROPE

- **Finland (crystalline rock):** construction licence granted (2015) – construction underway, operation licence application in preparation
- **Sweden (crystalline rock):** construction licence application submitted (2011), in final phase of discussion
- **France (sediments):** site selected, construction licence application in preparation, to be submitted in near future
- **Switzerland (sediments):** site selection is in advanced stage, 1 site is proposed for the submission of general licence application in ~ 2024
- **Germany (several host rocks):** site selection according to law in 2031 (with many steps in between)
- **Belgium (Sedimentary rocks):** design concept and Safety Case methodology was developed for Boom Clay as a potential host formation. But no decision host formation yet, requirement to explore other potential sedimentary rocks (from poorly indurated plastic clays – Boom Clay, Ypresian clays – to hard pelites, slates...) and to apply same methodology.
- **Czech Republic (crystalline rock):** 4 sites selected in 2020, final site and alternative will be selected based on SA in 2030.



SORPTION: Retention processes for Radionuclides

F. BRANDT, N. MAES, B. BAEYENS, M. MARQUES FERNANDES & M. KLINKENBERG

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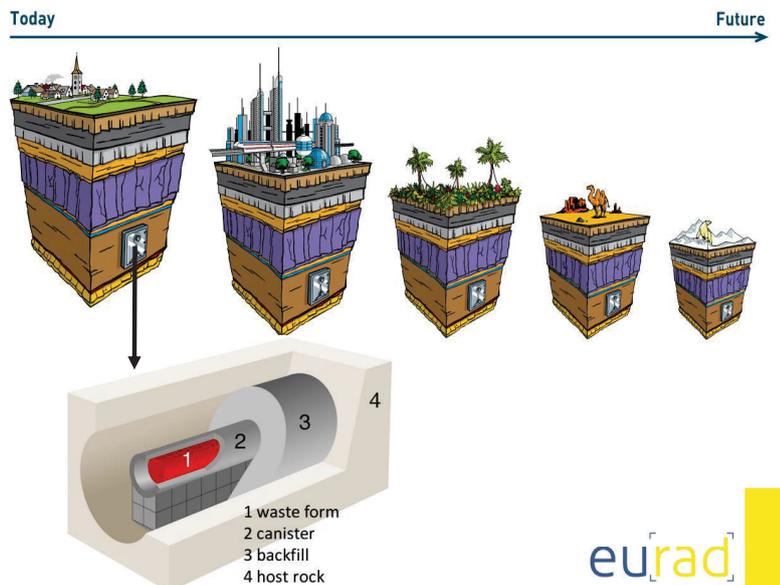
THE LONG-TERM SAFETY OF A DEEP GEOLOGICAL NUCLEAR WASTE REPOSITORY

Geochemical aspects

Aim: Prediction of the geochemical evolution of a nuclear waste repository system over geological time scales.

Including: the multi-barrier system of waste matrix, canisters, tunnel backfill, surrounding rock

- Required:** sound understanding of
- Radionuclide migration
 - Retention processes





DISPOSAL SYSTEM FOR HIGH LEVEL NUCLEAR WASTE

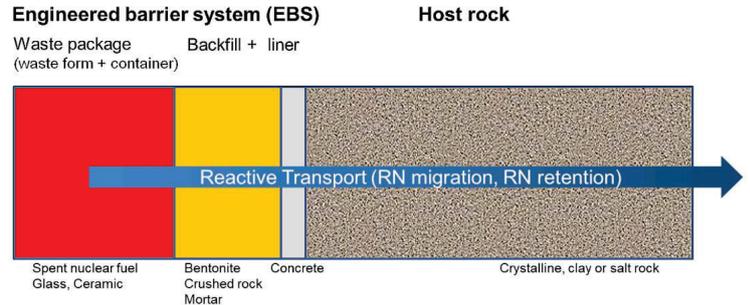
Complex multiphase system

In contact with water: geochemical gradients, especially at the interfaces of the different components of the disposal system

Secondary phase formation

- Radionuclide (RN) structural uptake
- RN adsorption

→ RN retention



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SORPTION

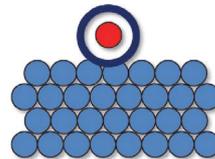
Adsorption processes

RN accumulated on a surface in a reversible way

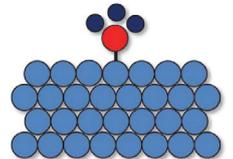
- Ion exchange
- Surface complexation

Structural uptake

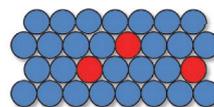
1. Electrostatic Adsorption



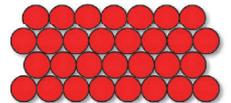
2. Chemical Adsorption



3. Coprecipitation/Solid Solution



4. Precipitation



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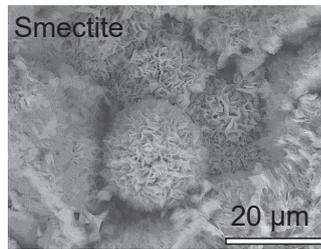
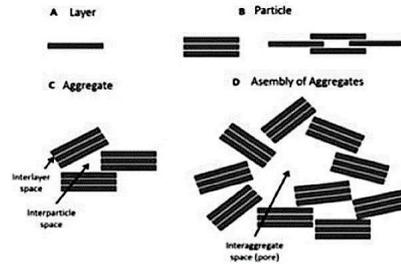
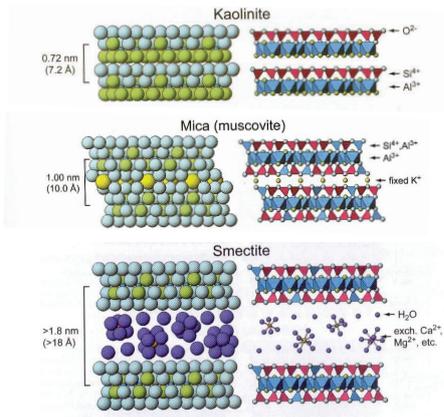


ADSORPTION ON CLAY MINERALS

Clay minerals

Layers of Si-O tetraheders with Al-O octaheders

Plate like structure forming stacks



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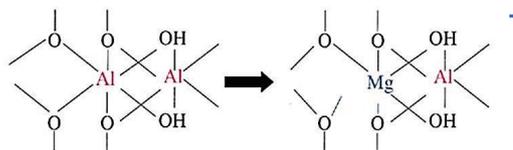


RN ADSORPTION ON CLAYS: EXCHANGEABLE CATIONS

Ion exchange

Ion exchange sites are typical for clay minerals due to:

- Substitution of Si or Al by an element of lower valency, Si(IV) by Al(III), Al(III) by Mg(II), a **permanent** negative charge is created.



- Neutralization by hydrated cations: Na⁺, Mg²⁺, Ca²⁺,... which form the **exchangeable cations** in the interlayers of the clay
- Ion exchange reaction: S-Na + K⁺ ⇌ S-K + Na⁺

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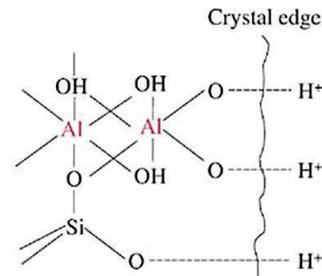
RN ADSORPTION ON CLAYS: SURFACE COMPLEXATION

Functional groups on mineral surfaces

Most important functional groups are -OH type groups:

- present on aluminosilicates (Al-OH & Si-OH)
- oxide surfaces (M-OH) and
- organics (carboxyl, carbonyl, phenolic) functional groups

Depending on the pH they are protonated, developing a positive charge or deprotonated, developing a negative charge



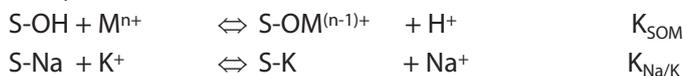
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RN ADSORPTION ON CLAYS - MODELING

Mechanistic interpretation: Surface complexation/ion exchange models

Based on interaction constants (K_{SOM} , $K_{Na/K}$) with the surface functional groups and ion exchange sites (thermodynamic basis)



→Valid for different chemical conditions

→Complex but robust – based on chemical understanding

Experimental data needed: Measuring adsorption as function of

- pH (edges)
- Electrolyte concentration
- RN concentration (isotherms)

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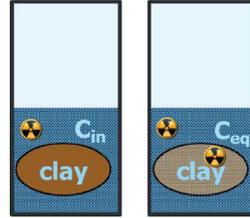
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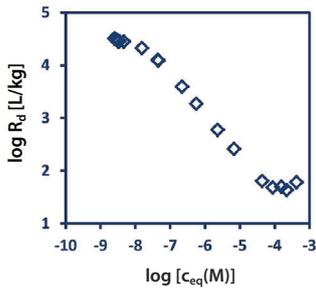
RN ADSORPTION ON CLAYS: BATCH EXPERIMENT

The solid-liquid distribution ratio R_d (K_d)

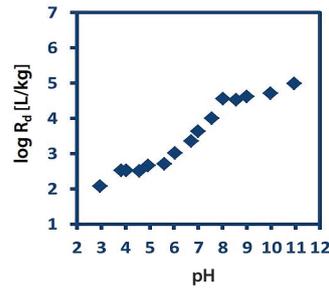
$$R_d = \frac{C_{solid}}{C_{liquid}} = \frac{C_{in} - C_{eq}}{C_{eq}} \frac{V}{M}$$



Adsorption isotherms (R_d vs. c_{eq})



Adsorption edges (R_d vs. pH)



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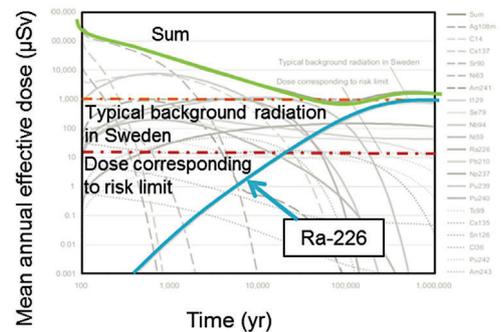
RN ADSORPTION IN NUCLEAR WASTE REPOSITORIES

The example of ^{226}Ra

In some scenarios, as a result of the ^{238}U decay chain ^{226}Ra dominates the dose during the late stages

Corrosion of spent fuel can lead to release and migration of ^{226}Ra into the near field of a nuclear waste repository

Adsorption on buffer materials or host rock (e.g. clay)?



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SKB, 2010



ADSORPTION ON CLAY MINERALS: EXAMPLE FROM EURAD-FUTURE (PSI - FZJ)

²²⁶Ra and Ba adsorption on montmorillonite (SWy)

Montmorillonite as model system for bentonite buffer

Scientific questions

- Quantitative understanding of ²²⁶Ra and Ba adsorption
- Derivation of a mechanistic adsorption model
- Is Ba a suitable analogue for ²²⁶Ra?
 - Use of Ba as carrier for the ²²⁶Ra tracer in adsorption isotherm

	Type of experiment	$[X^a]_{int}/c_{eq}$ (M) ^b	Ionic strength NaCl (M)	S/L (g L ⁻¹)	pH
¹³³ Ba	Edge	$3.3 \cdot 10^{-8} - 3.9 \cdot 10^{-8}$	0.03	2.91	3.3 - 9.3
¹³³ Ba	Edge	$4.3 \cdot 10^{-8} - 3.7 \cdot 10^{-8}$	0.3	2.96	2.6 - 10.6
²²⁶ Ra	Edge	$2.0 \cdot 10^{-7}$	0.02	2.21	5.5 - 10.2
²²⁶ Ra	Edge	$1.9 \cdot 10^{-8}$	0.14	8.33	5.6 - 10.2
²²⁶ Ra	Edge	$1.8 \cdot 10^{-8}$	0.3	13.95	5.7 - 9.9
¹³⁷ Ba/ ¹³³ Ba	Isotherm	$2.03 \cdot 10^{-9} - 1.8 \cdot 10^{-2}$	0.02	0.88 - 4.39	6.9 - 7.1
¹³⁷ Ba/ ²²⁶ Ra	Isotherm	$[^{137}\text{Ba}] 0.0 - 1.7 \cdot 10^{-2}$	0.02	0.84 - 4.29	6.5 - 7.0
	Isotherm	$[^{226}\text{Ra}] 4.3 \cdot 10^{-9} - 5.9 \cdot 10^{-9}$			

^aX=Ba or Ra; ^bFor edges initial concentrations and for isotherms equilibrium concentrations are given

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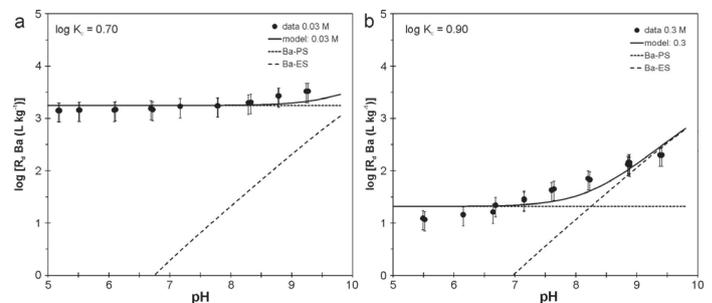


ADSORPTION OF Ba ON MONTMORILLONITE (I)

Ba pH adsorption edges

Adsorption mainly dominated by a **single cation exchange reaction** of Ba²⁺ with respect to Na⁺ on montmorillonite

For the more pronounced Ba uptake at high pH and IS, the **surface complexation of Ba** on edge sites modelled by one single surface complexation constant. This was fixed in all model calculations.



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Klinkenberg et al., 2021





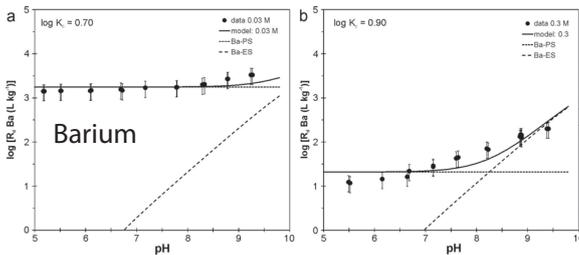
ADSORPTION OF ²²⁶Ra AND Ba ON MONTMORILLONITE

Ba and Ra pH adsorption edges: cation exchange

At low ionic strength: selectivity coefficients for Ba and Ra in good agreement ($\log K_c$ values vary between 0.70 and 0.84).

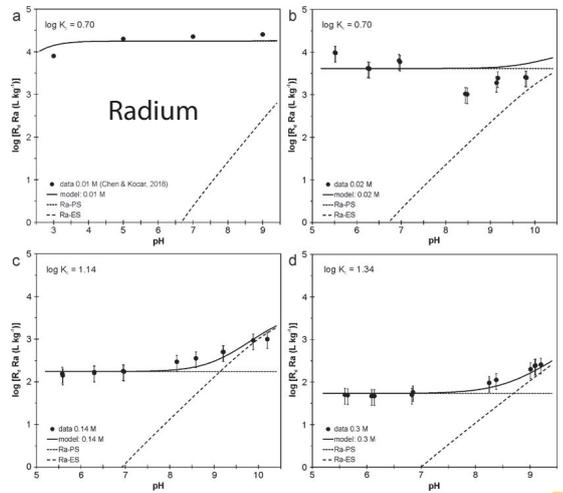
At high ionic strength, the K_c (Ba-Na) slightly higher compared to the low ionic strength data.

K_c (Ra-Na) exhibits a clear dependency on ionic strength



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ADSORPTION OF ²²⁶Ra AND Ba ON MONTMORILLONITE

Ba and Ra pH adsorption edges: cation exchange

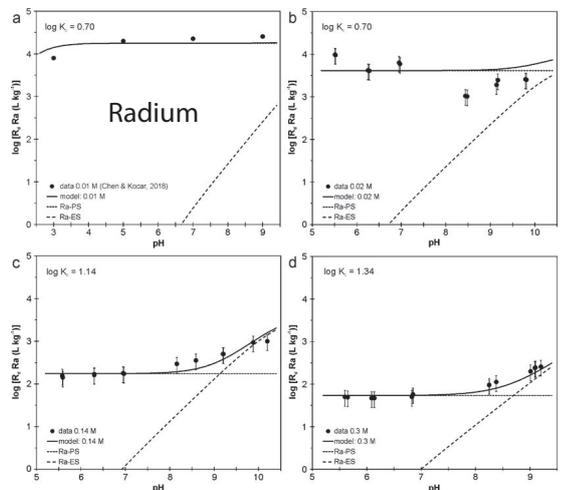
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Experiment	Kinetics		Edges				Isotherms
	0.02	0.1	0.01	0.02	0.14	0.3	
NaCl concentration (M)	0.02	0.1	0.01	0.02	0.14	0.3	0.02
Cation exchange reaction	$\log K_c$						
$2\text{Na-SWy} + \text{Ba}^{2+} \rightleftharpoons \text{Ba-SWy} + 2\text{Na}^+$	-	-	-	0.70	-	0.90	0.70
$2\text{Na-SWy} + \text{Ra}^{2+} \rightleftharpoons \text{Ra-SWy} + 2\text{Na}^+$	0.84	1.34	0.70	0.70	1.14	1.34	0.70

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ADSORPTION OF ²²⁶Ra AND Ba ON MONTMORILLONITE

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NaCl concentration (M)							0.02
Cation exchange reaction	log K_c						
$2\text{Na-SWy} + \text{Ba}^{2+} \rightleftharpoons \text{Ba-SWy} + 2\text{Na}^+$	-	-	-	0.70	-	0.90	0.70
$2\text{Na-SWy} + \text{Ra}^{2+} \rightleftharpoons \text{Ra-SWy} + 2\text{Na}^+$	0.84	1.34	0.70	0.70	1.14	1.34	0.70

and surface complexation

The contribution of surface complexation to the overall adsorption is the same for Ba and Ra and is independent of ionic strength.

Ra and Ba surface complexation can be described with an identical model

Site type	Capacity
$\equiv\text{S}^{\text{W}2}\text{OH}$	$4.0 \cdot 10^{-2} \text{ mol} \cdot \text{kg}^{-1}$
Surface complexation reactions	Constants
$\equiv\text{S}^{\text{W}2}\text{OH} + \text{H}^+ \rightleftharpoons \equiv\text{S}^{\text{W}2}\text{OH}_2^+$	$\log K^+ = 6.0$
$\equiv\text{S}^{\text{W}2}\text{OH} \rightleftharpoons \equiv\text{S}^{\text{W}2}\text{O}^- + \text{H}^+$	$\log K^- = -10.5$
$\equiv\text{S}^{\text{W}2}\text{OH} + \text{Ba}^{2+} \rightleftharpoons \equiv\text{S}^{\text{W}2}\text{OBa}^+ + \text{H}^+$	$\log K_i = -5.0$
$\equiv\text{S}^{\text{W}2}\text{OH} + \text{Ra}^{2+} \rightleftharpoons \equiv\text{S}^{\text{W}2}\text{ORa}^+ + \text{H}^+$	$\log K_i = -5.0$

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ADSORPTION OF ²²⁶Ra AND Ba ON MONTMORILLONITE

Ba and Ra isotherms

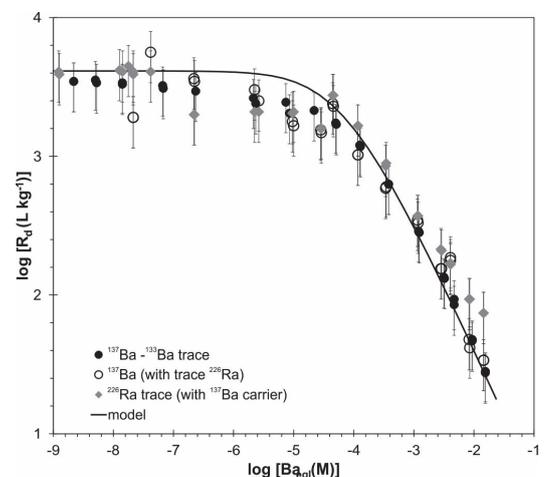
Very similar behaviour at pH = 7 and low ionic strength

Conclusions

Ba is good analogue for ²²⁶Ra regarding the adsorption on montmorillonite at ionic strengths < 0.1 M and pH < 8

Ra deviates in its behaviour at higher ionic strength.

A plausible explanation for this observation: larger ionic radius of Ra compared to Ba → favours its selectivity behaviour on Na-montmorillonite.



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SORPTION

Adsorption processes

Structural uptake

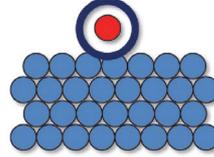
Solid-solutions: abundant in nature, leading to lower RN solubilities compared to pure phases.

Lacking thermodynamic data: so far limited applicability in HLW safety assessments

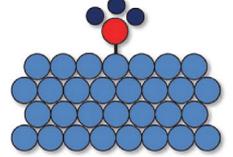
→ *conservatism in assessments?*



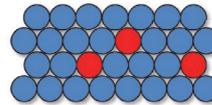
1. Electrostatic Adsorption



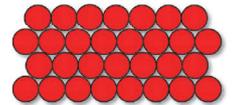
2. Chemical Adsorption



3. Coprecipitation/Solid Solution



4. Precipitation



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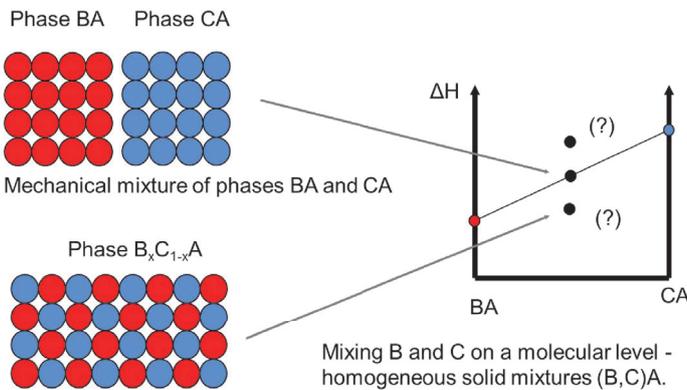
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WHAT IS A SOLID SOLUTION?

Example: Solid solution formation from the endmembers BA and CA

B, C = cations, A = anion



Solid solution

a single phase which exists over a range in chemical composition.

Most minerals in nature are solid solutions.

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THE STABILITY OF SOLID SOLUTIONS

Boundary conditions

Size difference

of the ions or atoms being mixed. If ionic radii differ by less than 15%, extensive or complete solid solution is often observed.

Flexibility of the crystal structure

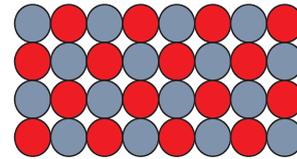
extend of solid solution depends on the ability of the structural framework to flex and accomodate differently-sized atoms

Ion charge

difference in bonding character, valence of ions being mixed, electron configuration effects



Phase $B_xC_{1-x}A$



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WHY ARE SOLID-SOLUTIONS IMPORTANT?!

..once again: the example of ^{226}Ra

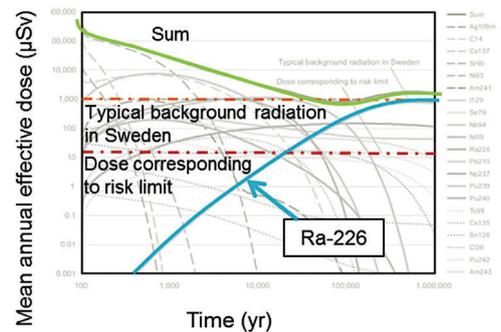
Relevance of the solid solution formation for the solubility:

Simplified example:

0.1 n NaCl background electrolyte, 25 °C

Solubility of pure RaSO_4 $2 \cdot 10^{-5}$ mol/L

Full equilibration of 0.5 g/L BaSO_4 to $(\text{Ba,Ra})\text{SO}_4$, $a_0 = 0$
final $c(\text{Ra})_{\text{eq}}$ $4 \cdot 10^{-8}$ mol/L



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SKB, 2010

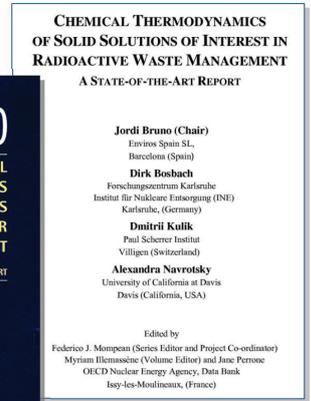
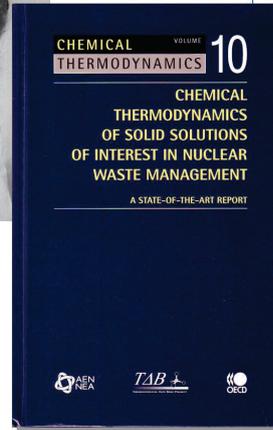




RADIUM, SULFATES AND SOLID SOLUTION FORMATION

.. the historical background

- 1898: Marie and Pierre Curie discover RaCl_2 – separated due to similar behaviour with Ba; pure Ra metal separated by M. Curie and A.-L. Debierne
- Lind et al. 1918: Solubility of RaSO_4
- Ground-breaking papers by Doerner and Hoskins (1925), Co-precipitation of Radium and Bariumsulfates; Doerner-Hoskins Partition Coefficient, Hahn (1926): Solubility of RaSO_4 ; Nikitin and Tolmatscheff (1936), solubility of RaSO_4
- Jucker and Treadwell (1954): Uptake of Ra in barite and celestite
- Weigel and Tinkl, (1967): Crystal structure of RaSO_4
- 1970s Solid solution-aqueous solution equilibria: Lippmann theory
- Langmuir and Riese (1985): Thermodynamics of Radium
- Zhu (2004): Binary Mixing Properties BaSO_4 - RaSO_4
- NEA (2007): solid solutions and radioactive waste management
- Bosbach et al. (2010), Curti et al. (2010): Recrystallisation of BaSO_4 in the presence of ^{226}Ra
- FZJ IEK-6: Vinograd et al., 2013, Klinkenberg et al., 2014, Brandt et al. 2015, Weber et al., 2016, Prieto et al., 2016, Weber et al. 2017, Vinograd et al., 2018a, Vinograd et al., 2018b, Klinkenberg et al., 2018, Bosbach et al., 2020, Brandt, et al. (2020)

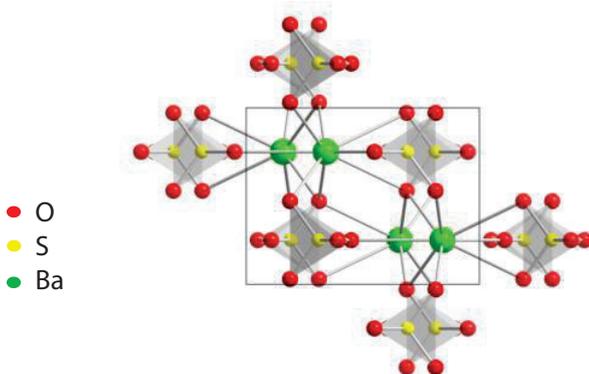


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THE ISOSTRUCTURAL SOLID SOLUTION



	RaSO_4	BaSO_4	SrSO_4
Cation radius CN 12 [Å]	1.70	1.61	1.44
a [Å]	9.16	8.87	8.36
b [Å]	5.56	5.54	5.35
c [Å]	7.30	7.14	6.87
Log Ksp	-10.26	-9.97	-6.63

- Orthorhombic

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Ra SOLUBILITY IN THE PRESENCE OF SOLID SOLUTIONS?

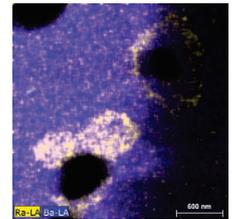
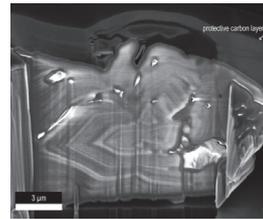
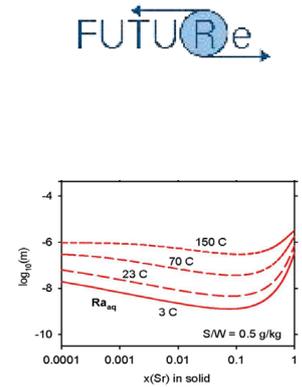
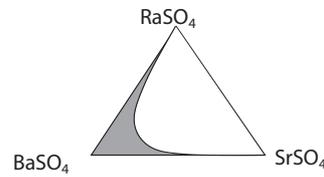
What do we need to know?

Prediction of the thermodynamic properties

- Molecular level **mixing model** → interaction parameters describing the ideality of the solid-solution
- Thermodynamic properties (**solubility**) of the end-members $BaSO_4$, $RaSO_4$, $SrSO_4$

Validation of the theoretical model and detailed process understanding of ^{226}Ra -uptake

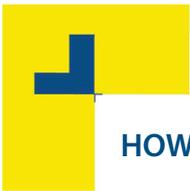
- Recrystallization experiments
- Microscopic analyses of the solid



eurad

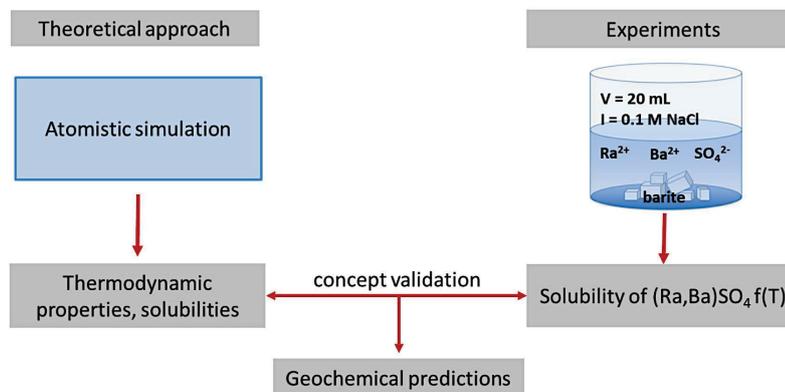
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HOW TO TACKLE THE QUESTION OF RA UPTAKE PREDICTION?

Approach



FUTURe

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HOW TO TACKLE THE QUESTION OF Ra UPTAKE PREDICTION?

The system Ra - Ba - SO₄ - H₂O

Solubility products BaSO₄ log(K_{SP}) = -9.97 to -9.98
 RaSO₄ log(K_{SP}) = -10.26 to -10.41

Relevant Ra-species Ra(s), Ra²⁺, RaOH⁺, RaCl⁺, RaCO₃(aq), RaCO₃(s)
 RaSO₄(aq), RaSO₄(s)

Interaction Parameters of the solid solution Ra_xBa_{1-x}SO₄
 $a_0 = 0.35$ (Zhu et al.) to 2.5 (Curti et al.)

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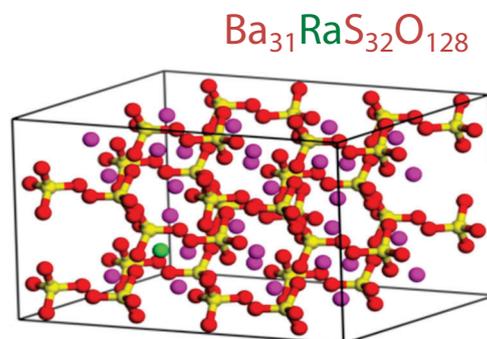
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THEORETICAL APPROACHES: INTERACTION PARAMETER

(Ba,Ra)SO₄ solid solution

Atomistic calculations, single defect approach
 DFT calculations with CASTEP
 Exchange-correlation functional of Wu & Cohen (2006)
 Ultra-soft OTFG pseudopotentials
 Plane wave cutoff 910 eV

a_0 (Ba,Ra)SO₄ = 1.0 ± 0.4



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Vinograd et al. 2014



RECRYSTALLIZATION EXPERIMENT

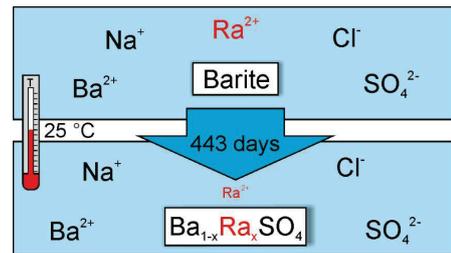
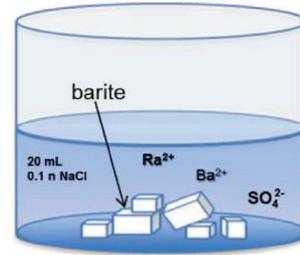
Why recrystallization and not co-precipitation?

Co-precipitation can lead to

- Kinetic entrapment during co-precipitation
- Chemical inhomogeneity of the solid phase

Careful at ultra trace RN concentration levels!

Re-crystallization experiments are carried out at close-to-equilibrium conditions between solid and aqueous solution to avoid these effects

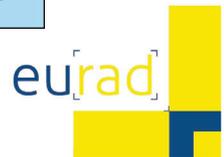
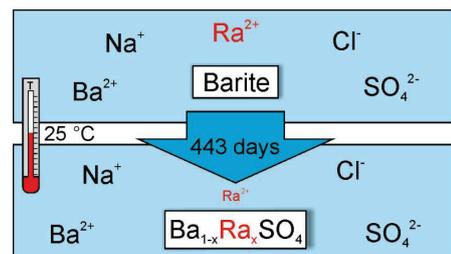
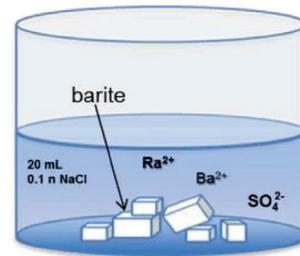
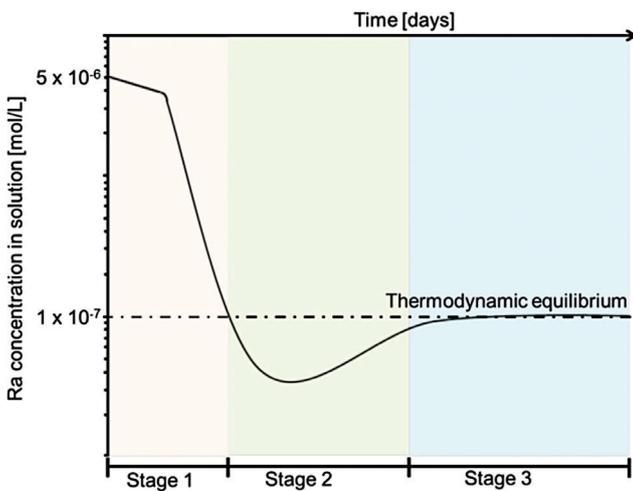


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RECRYSTALLIZATION EXPERIMENT



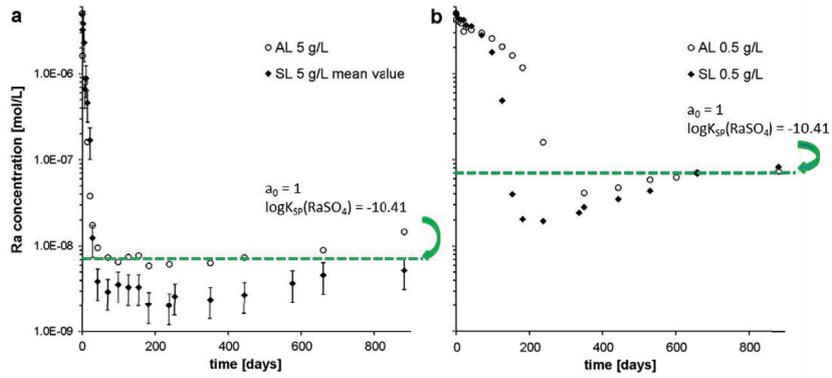
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Klinkenberg et al. ES&T, 2014



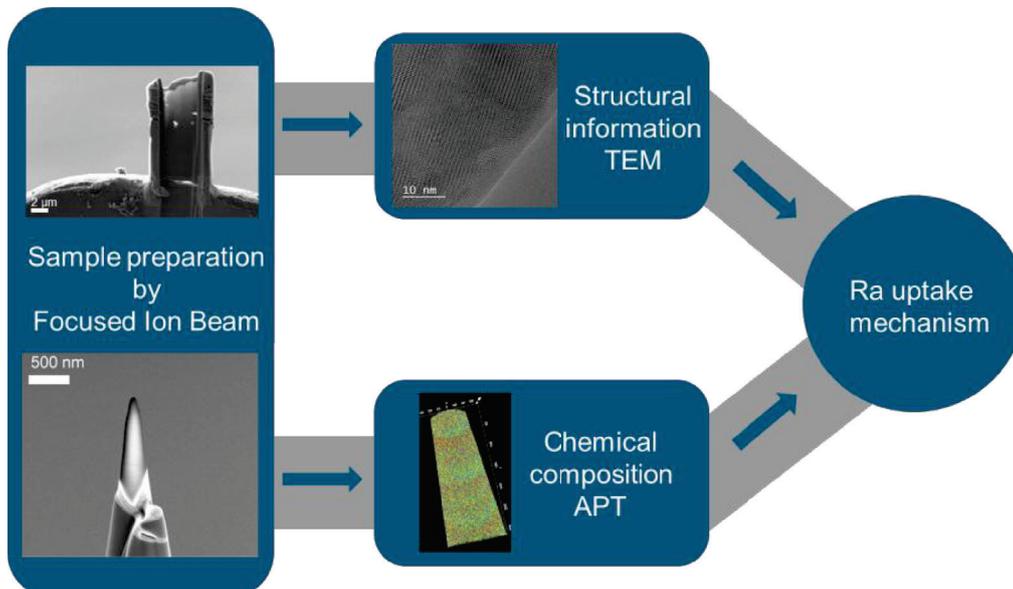
COMPARISON EXPERIMENT - PREDICTIONS



Excellent agreement with thermodynamic prediction at $a_0 = 1.0$ and $\log K_{sp}(\text{RaSO}_4) = -10.41$



THE UPTAKE PROCESS - MICROSCOPY

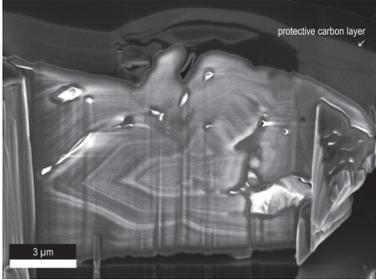




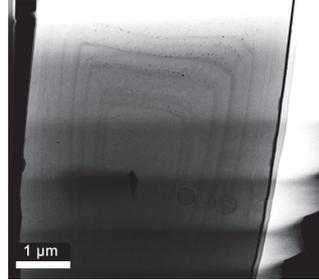
THE UPTAKE PROCESS - MICROSCOPY



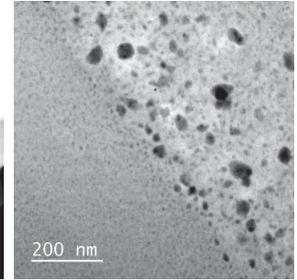
Electron Microscopy



FIB cross-section



HAADF STEM image



STEM image

SEM, TEM and STEM observation of the layered structure of barite

This structure consists of nano-scaled pores

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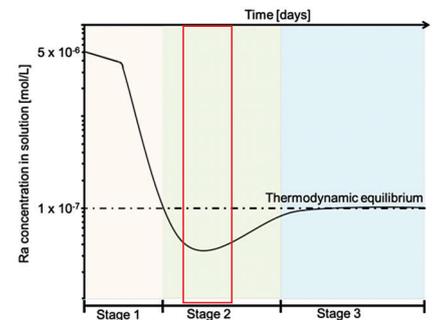
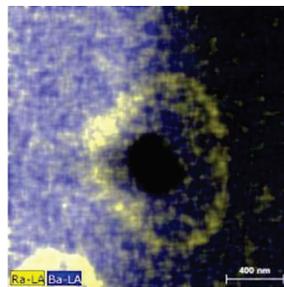
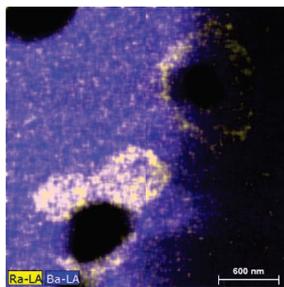
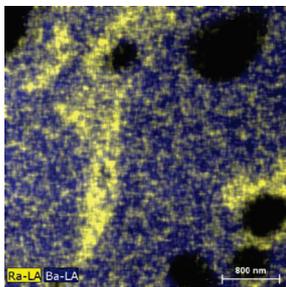
Weber et al., 2016



THE UPTAKE PROCESS - MICROSCOPY



Stage 2: STEM-EDX Mapping with ChemiSTEM and Super-EDX



At intermediate stages of the recrystallization of BaSO₄ to (Ba,Ra)SO₄:

The Ra distribution is not homogeneous, higher Ra concentrations occur near to pores

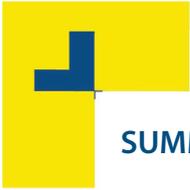
Pores are reactive sides → kinetics of Ra uptake depend on internal structure and surface of BaSO₄

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Weber et al., 2017





SUMMARY & CONCLUSIONS

Sorption of RN: adsorption and solid solution formation

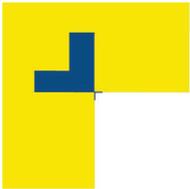
- The example of ²²⁶Ra shows that both mechanisms can be relevant and can be described by thermodynamic models
- ²²⁶Ra adsorption on clay: Ra behaves similar but not identical to Ba, can be modeled with existing surface complexation and cation exchange models
- ²²⁶Ra uptake in solid solutions can have a significant effect upon the solubility, depending on the total Ba and SO₄ inventory and uptake kinetics
- Not presented but also available as a solid solution model: the ternary system of BaSO₄-SrSO₄-RaSO₄ in solution

Open questions:

- Crystallization of the (Ba,Ra)SO₄ solid solution in confined spaces (EURAD-Future)
- Competition of adsorption and solid solution formation? ..to be investigated

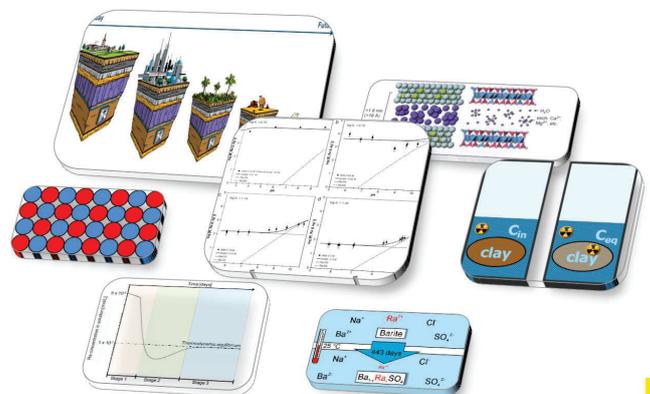
17.11.2021

Future 3rd Annual Meeting



THANK YOU VERY MUCH FOR YOUR ATTENTION....

QUESTIONS?



17.11.2021

Future 3rd Annual Meeting



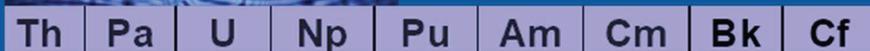
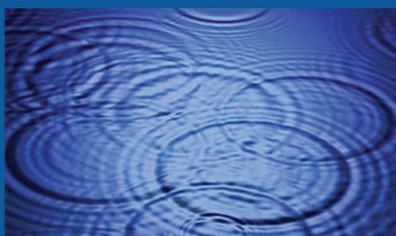
Interfacial redox reactions, X-ray absorption spectroscopy, and what they can do for the safety of radioactive-waste repositories

Andreas C. Scheinost

The Rossendorf Beamline @ ESRF, Grenoble, France

Institute of Resource Ecology @ HZDR, Dresden, Germany

Institute of Geological Sciences @ University of Berne, Switzerland



Andreas Scheinost scheinost@esrf.fr | www.hzdr.de

Nuclear waste

Sources



Inventory TBq (World)

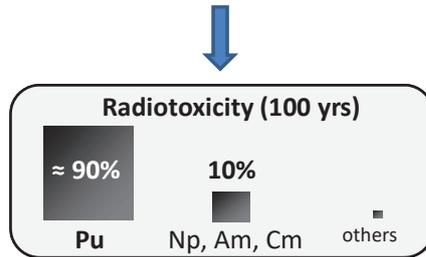
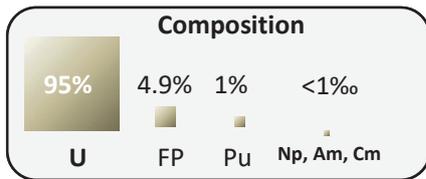
	Low/intermediate level	Spent fuel	Reprocessing
Power plants	1.2·10 ⁶	2.8·10 ¹⁰	4.2·10 ⁷
Institutional	7.0·10 ⁵		
Military	7.0·10 ⁵		3.1·10 ⁷

Source: IAEA-TECDOC-1591 (2007)

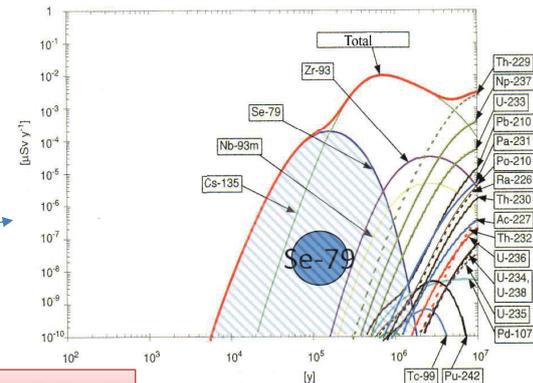
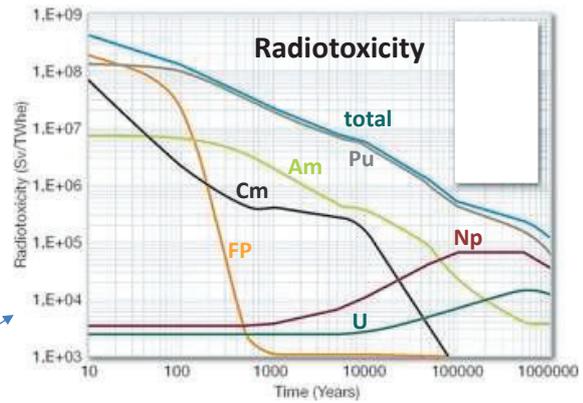
- Switzerland: 2%
- India: 2% (?)
- Germany: 3%
- Korea (Rep.): 4%
- UK: 5%
- France: 6%
- Japan: 7%
- Russia: 10% (?)
- Canada: 19%
- USA: 28%



Spent nuclear fuel



Radioactive dose released into the biosphere ???



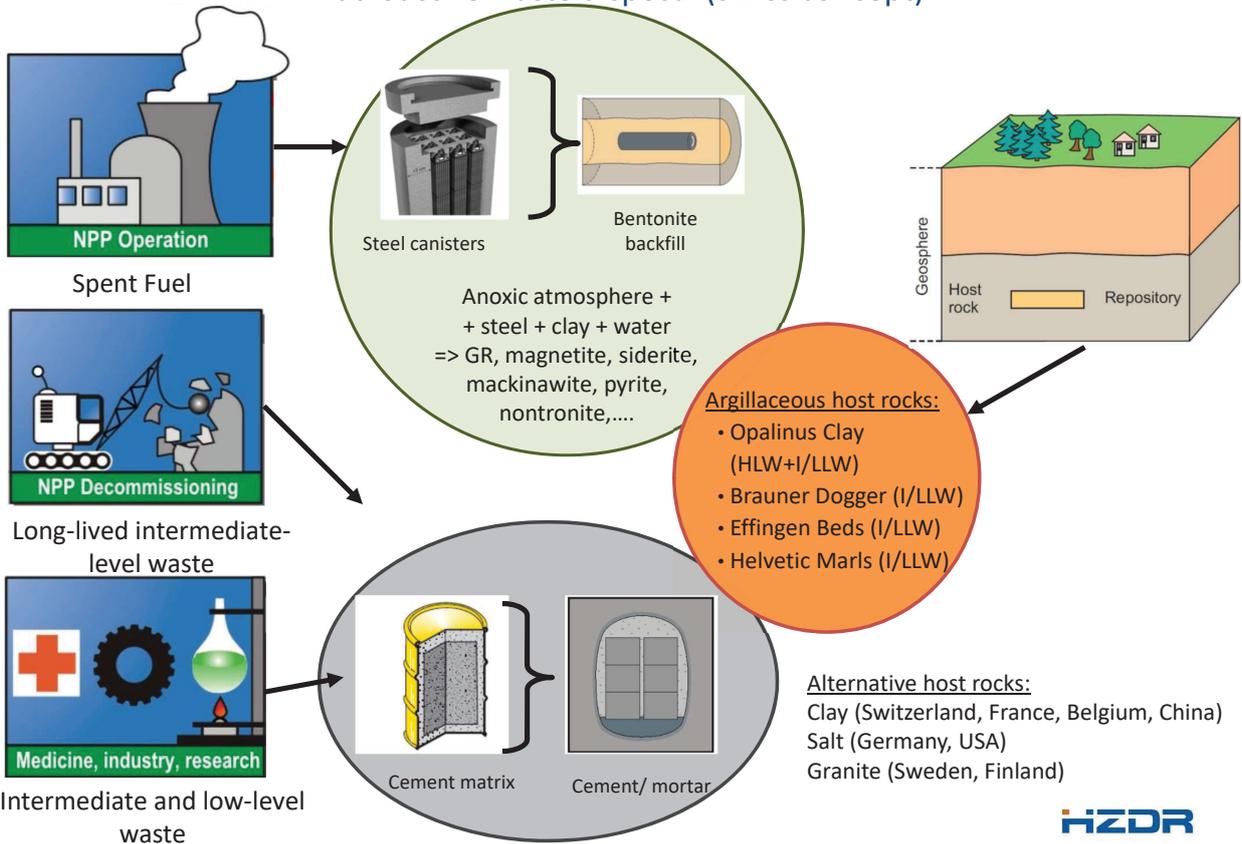
(geo)chemistry + reactive transport modeling!

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Radioactive waste disposal (Swiss concept)

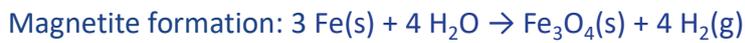
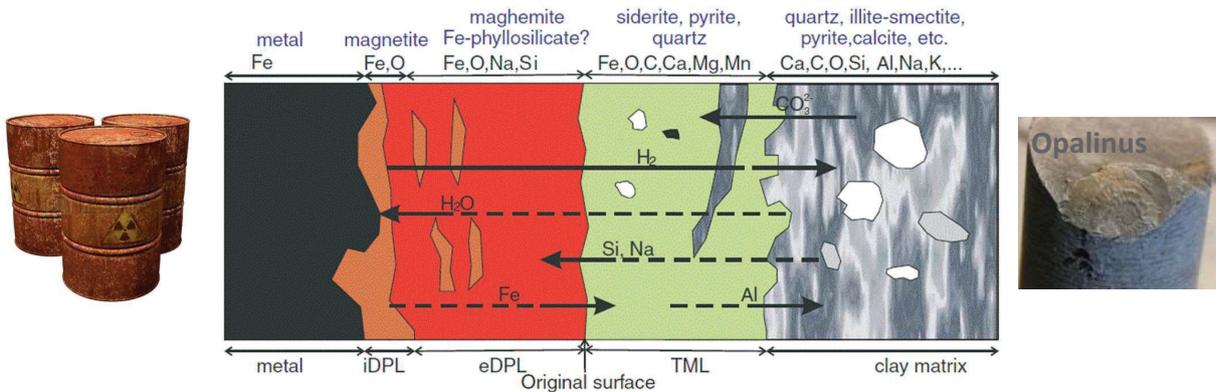


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Steel/clay interface at anoxic conditions



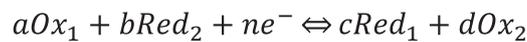
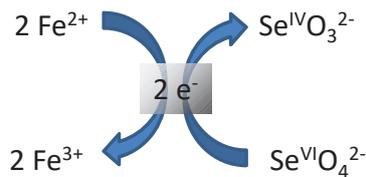
(Grambow et al. 1996, Schlegel et al. 2010)



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What is redox?



Nernst equation

$$E = E^0 + \frac{0.06}{n} \log \frac{[Ox_1]^a [Red_2]^b}{[Red_1]^c [Ox_2]^d}$$

P= 1atm, T° = 25°C

Equilibrium



Kinetics

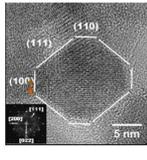
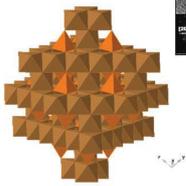
- Electron transfer
- Sorption
- Structural changes
- Precipitation/Dissolution
-



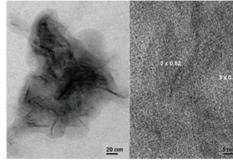
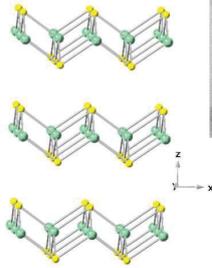
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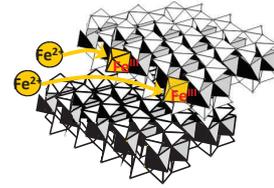
Fe^{II} water/mineral interfaces with significant reduction potential



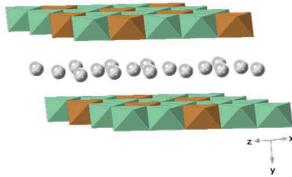
Magnetite (Fe^{II}Fe^{III}₂O₄)
 common in aquifers, corroding steel
 e⁻-conducting along Fe^{II}/Fe^{III} octahedral layers



Mackinawite (Fe^{II}S)
 metastable in sulfur-rich aquifers
 e⁻-conducting

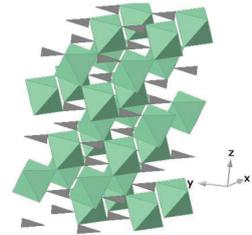


Montmorillonite with sorbed Fe²⁺ or structural Fe^{II}
 (backfill, aquifers)



Green rust (Fe^{II}Fe^{III}(OH)₂Cl)
 corroding steel, metastable in aquifers
 (semi)conducting along sheets

- Fast reduction at mineral/water interface:
- sorption = surface enrichment of redox partners (Fe and Se)
 - sorption = decreased redox potential because of ligand effect (Stumm 1992)
 - sorption = contact with electron conducting lattice (Becker et al. 2001)



Siderite (Fe^{II}CO₃)
 carbonate-rich aquifers
 e⁻ insulator

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Key radionuclides and their oxidation states/species

	Se	Tc		U	Np	Pu	Am
VII	 Selenate	 Pertechnetate					
VI				UO ₂ ²⁺ Phosphate	-	PuO ₂ ²⁺	-
V				UO ₂ ⁺	NpO ₂ ⁺	PuO ₂ ⁺	-
IV	 Selenite	TcO ₂		UO ₂	NpO ₂	PuO ₂	-
III				-	-	Pu ³⁺	Am ³⁺
0	Se ⁰						
-II	FeSe HSe ⁻						

Reduction

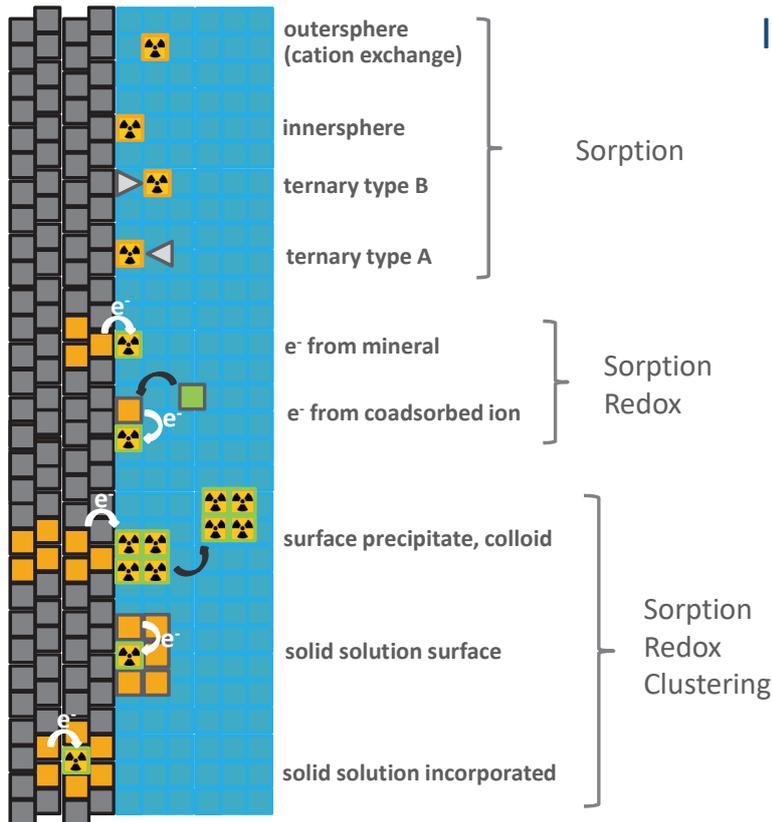
Sorption ⇒ Precipitation?

Aqua species ↔ solids
 Anions ↔ cations
 Transient ↔ thermodynamically stable states

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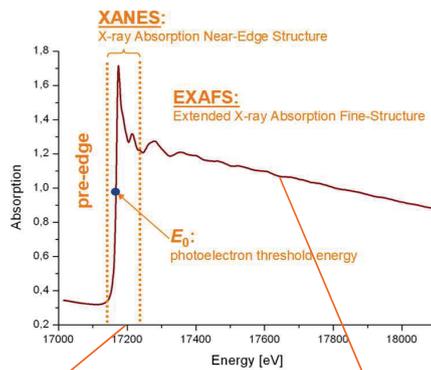
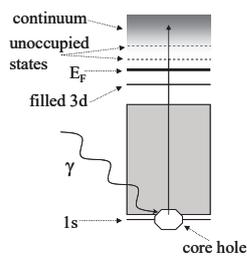
Mineral/Water Interfacial Processes



HZDR

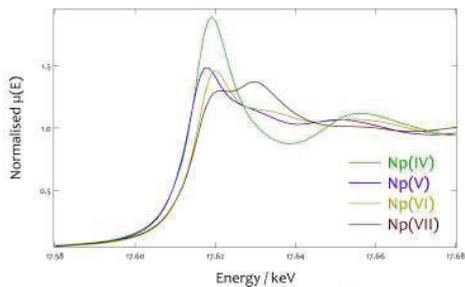
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X-ray absorption fine-structure spectroscopy

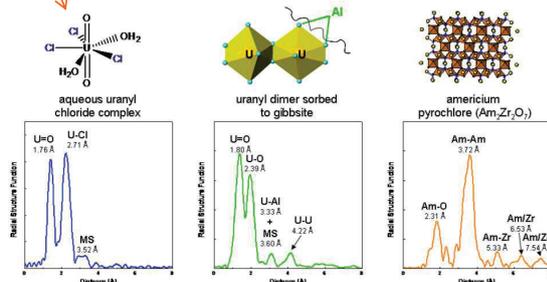


- XANES:**
- excitation of core-level electrons to unoccupied orbitals
 - identity and oxidation state of central atom

- EXAFS:**
- excitation of core-level electrons to continuum
 - Z ± 3, coordination number (CN) ± 25%, distance (r) ± 0.01 Å, disorder (DW) of atom neighbors



Np L_{III}-edge XANES spectra with different Np oxidation states

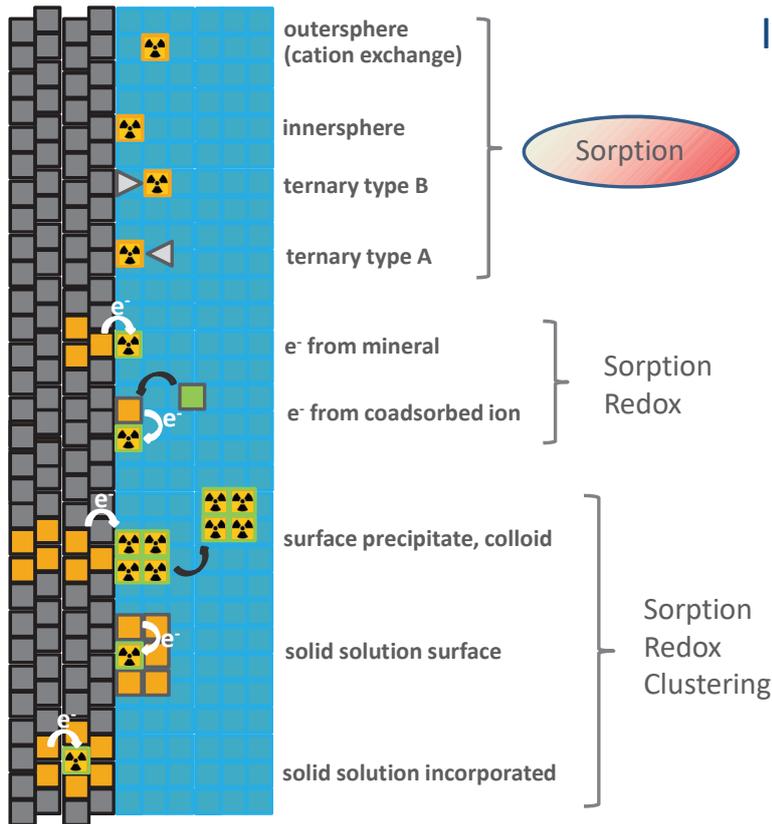


$$\chi(k)_{\text{exp}} = \sum_j N_j \text{Ampl}(k, r_j) e^{-2r_j^2 k^2} \sin(2kr_j + \phi_{\text{total}}(k, r_j))$$

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Mineral/Water Interfacial Processes



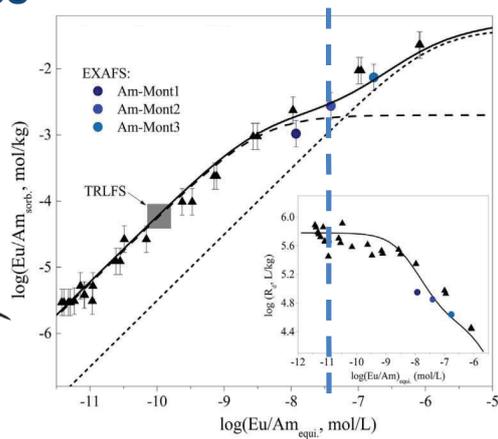
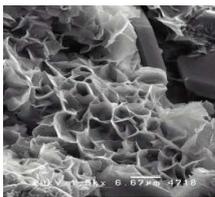
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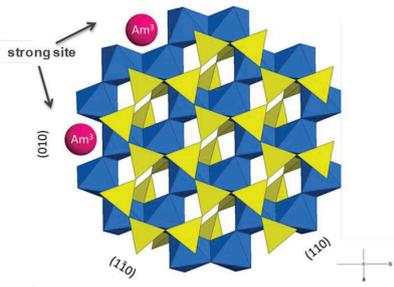
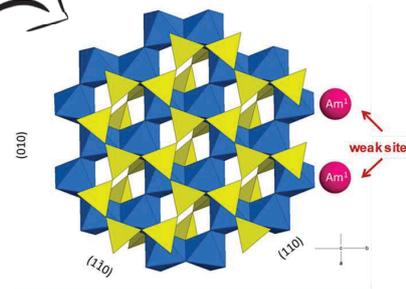
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⁹⁵Am
Americium
243(95)

Sorption of trivalent actinides onto montmorillonite Molecular-level understanding of macroscopic strong sites



weak sites



Sample	Coordination shell			Higher shells		
	CN	R [Å]	σ^2 [Å ²]	CN	R [Å]	σ^2 [Å ²]
CE	9.0 ^f O	2.47	0.0074	-	-	-
Strong	9.0 ^f O	2.46	0.0081	3.0 Al	3.24	0.0077
Weak	9.0 ^f O	2.46	0.0079	1.0 Al	3.20	0.0054

Marques Fernandes et al. 2016. *Water Research* 99, 74-82

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PSI

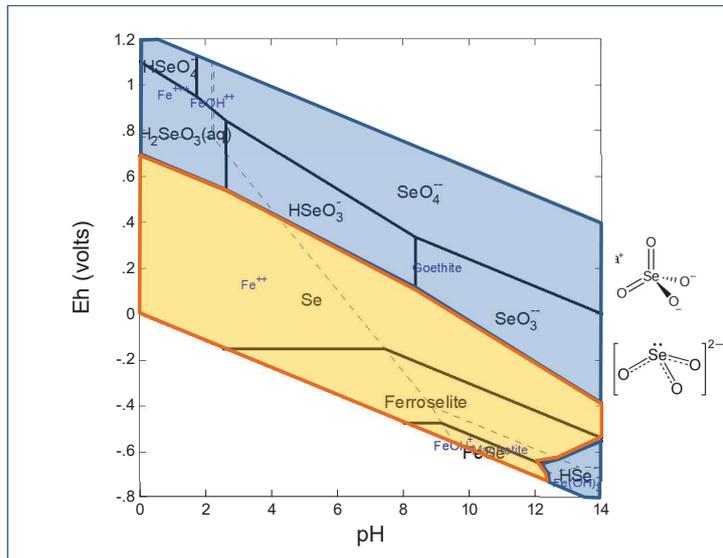
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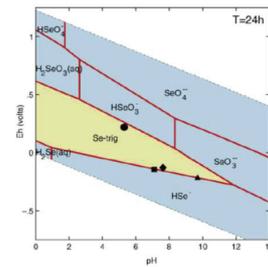
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Selenium

in presence of Fe

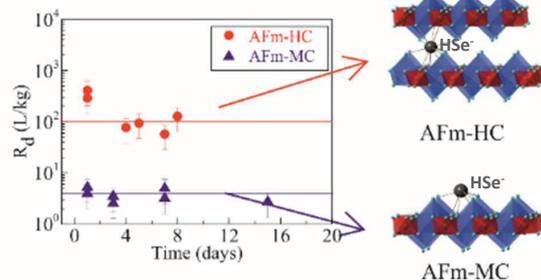
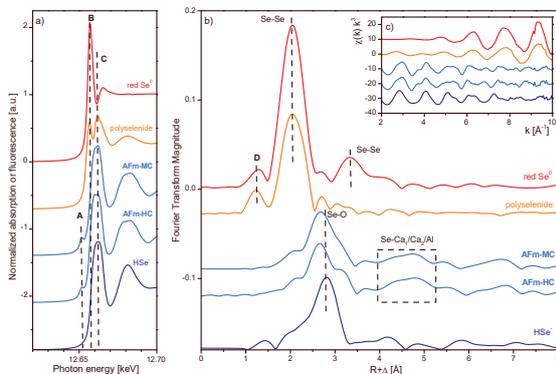


without Fe



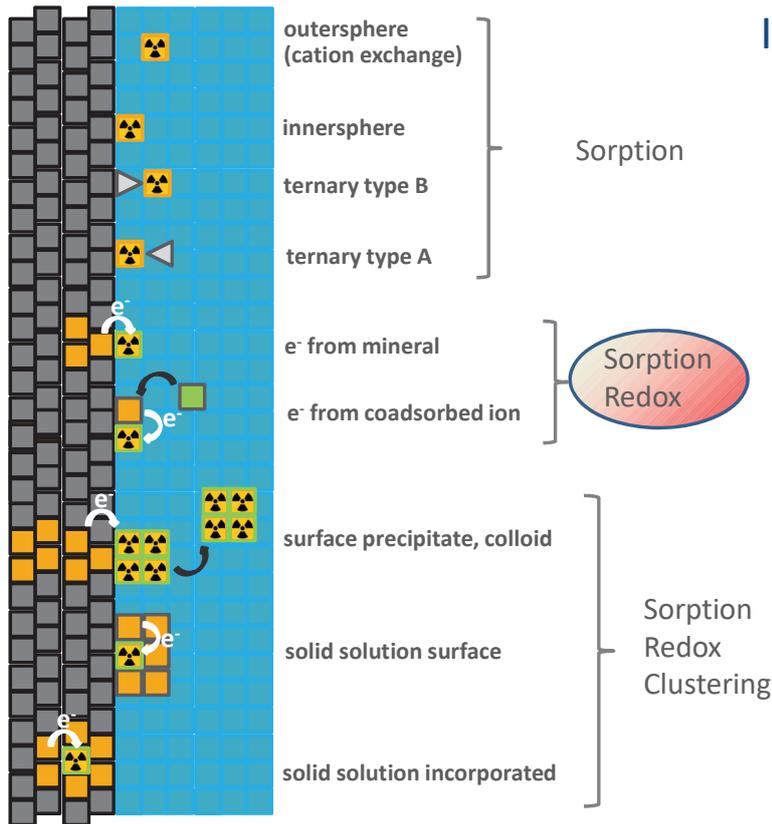
Retention of selenium by concrete barriers Structure and sorption of an elusive anion, HSe⁻

HSe⁻ is barely sorbed by most minerals, posing a significant risk for the safe disposal of this long-lived fission product. The cement barrier foreseen for intermediate-level waste repositories, however, contains AFm phases, which due to their anion exchanging properties may be able to retain HSe⁻.



HSe⁻ is significantly sorbed in the interlayers of AFm-HC, and also better protected against re-oxidation
HSe⁻ is lesser sorbed by AFm-MC due to the lesser accessibility of its interlayer space.

Mineral/Water Interfacial Processes

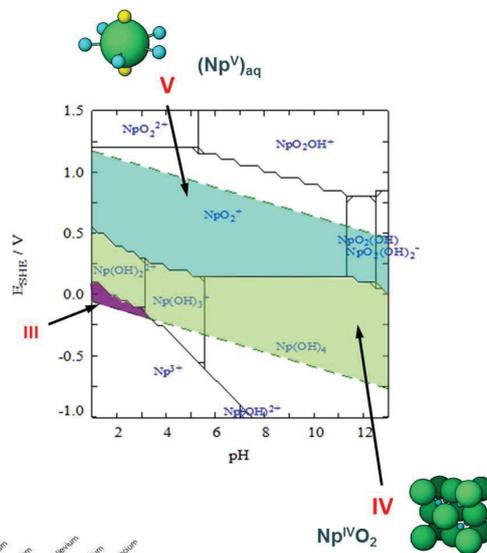


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93
Np
Neptunium
237.048

Neptunium

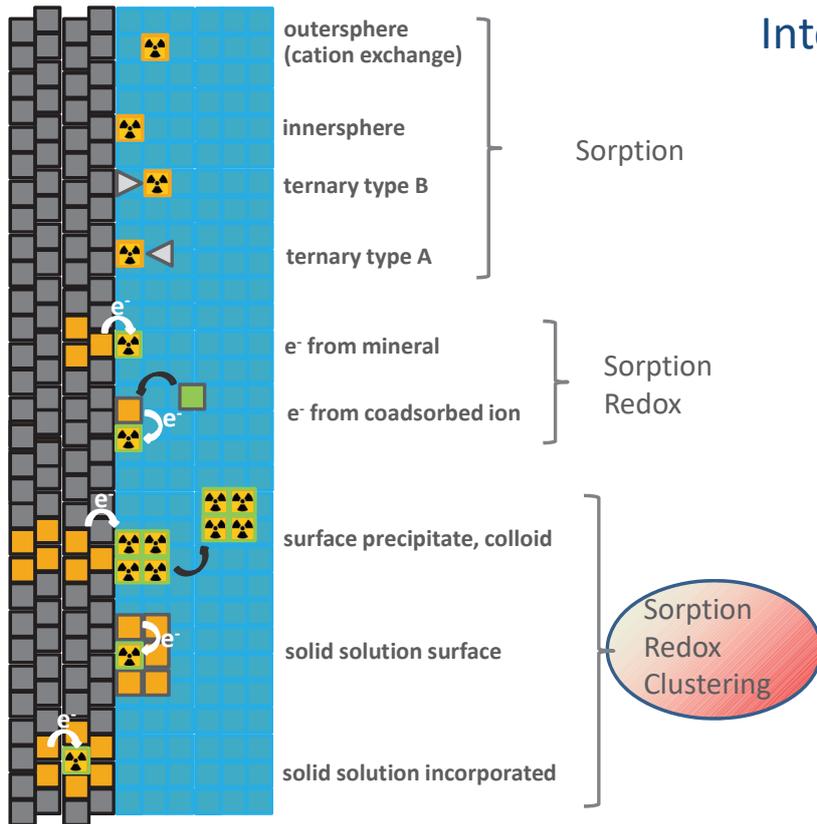


ACTINIDES												
89	90	91	92	93	94	95	96	97	98	99	100	101
Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No
(232)	(231)	(238)	(237)	(244)	(243)	(247)	(247)	(251)	(252)	(257)	(258)	(259)

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Mineral/Water Interfacial Processes

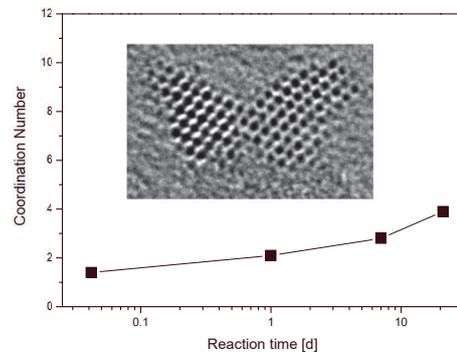
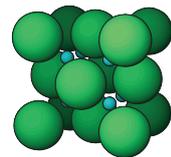
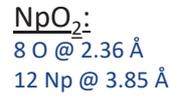
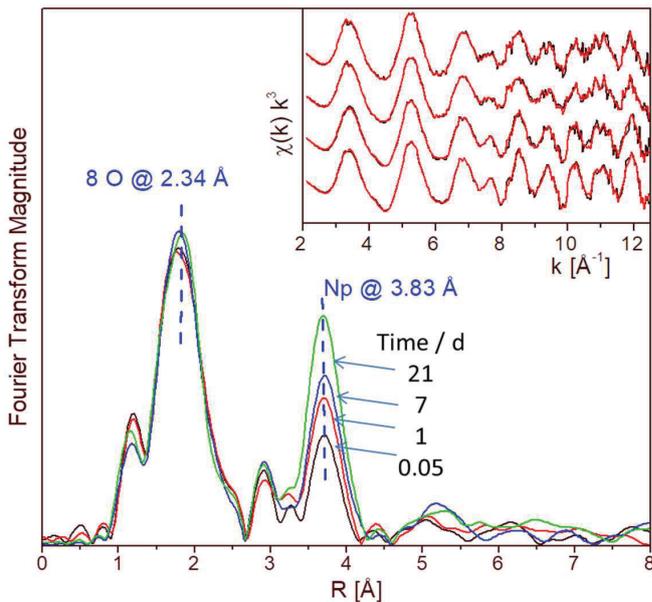


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⁹³
Np
Neptunium
237.045

Np(V) sorption/redox on siderite



- Complete reduction of Np^{V} to Np^{IV}
- Precipitation of NpO_2 -like phase, nanoparticles, which grow with time
- Siderite (e^- insulator, O-terminated) is able to promote the one-electron reduction from Np^{V} to Np^{IV} , but surface complexation is not strong enough to prevent NpO_2 precipitation



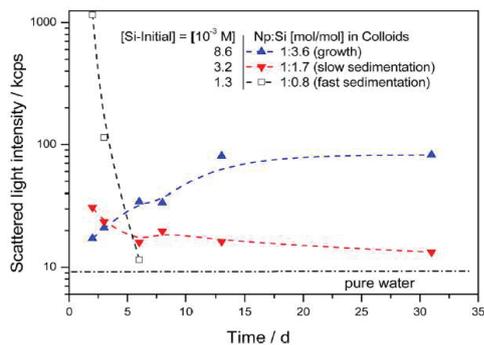
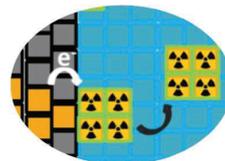
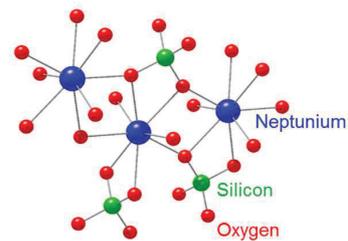
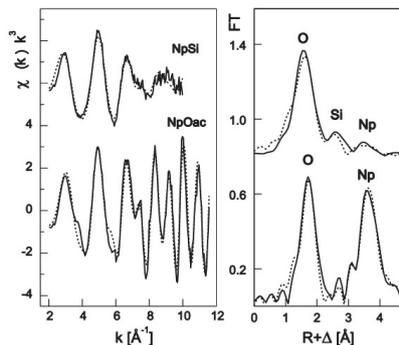
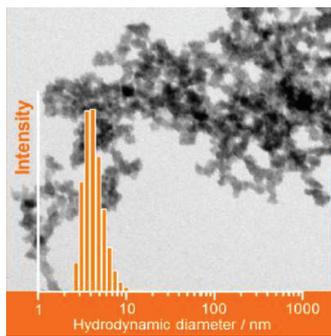
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A. C. Scheinost, R. Steudtner, R. Hübner, S. Weiss, F. Bok, *Environ. Sci. Technol.* **50**, 10413-10420 (2016).

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Formation of nanosized Np^{IV} silica colloids

Np^{IV} carbonate+silicate solutions, simulating Np release in alkaline near-field, which is diluted with neutral water in the far-field



- Amorphous NpO₂ structure with SiO₂
- Silanol groups provide negative surface charge
- Depending on Si concentration, stable colloidal suspension form
- Risk of Np migration with ground water

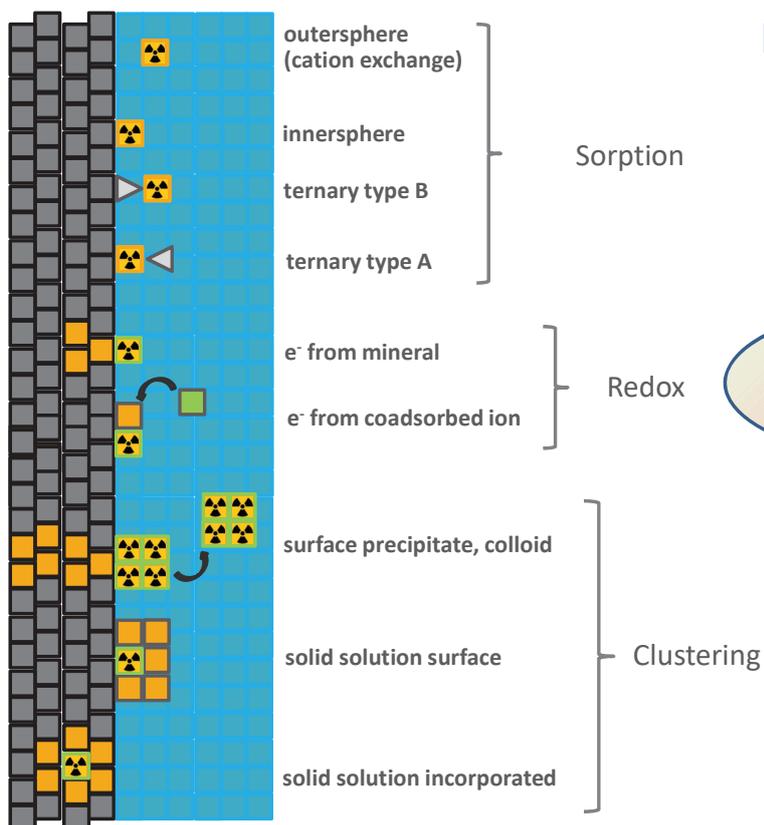
R. Husar et al., *Environ. Sci. Technol.* **49**, 665–671 (2015).



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Mineral/Water Interfacial Processes



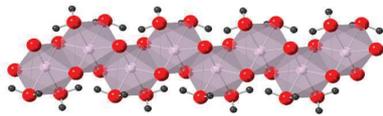
Often several processes in the same system....



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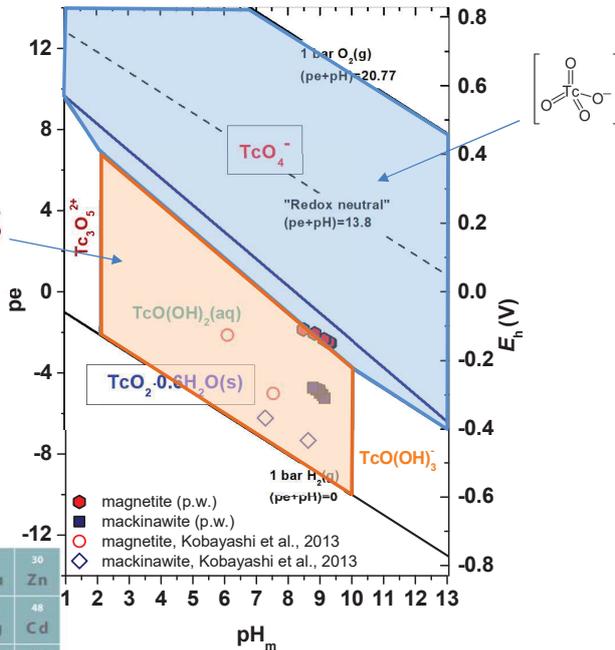
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Technetium

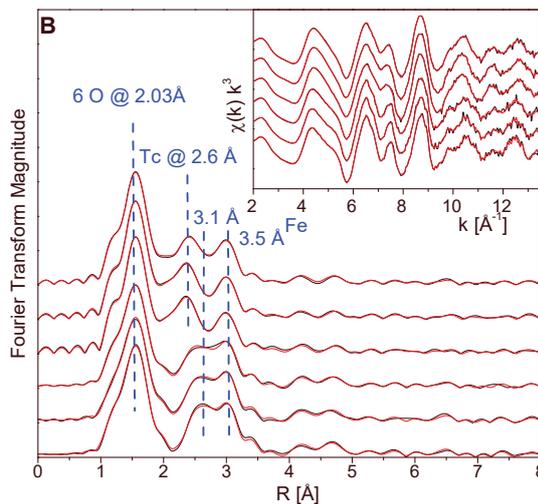
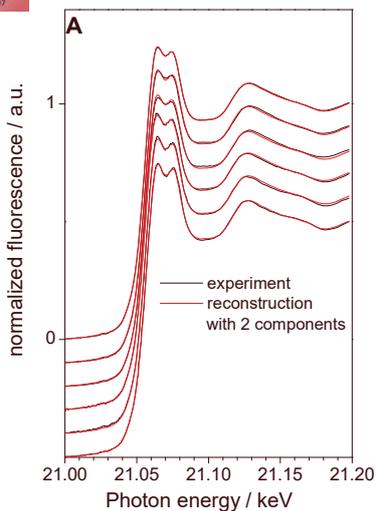


d block
(transition elements)

21	22	23	24	25	26	27	28	29	30
Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn
39	40	41	42	43	44	45	46	47	48
Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd
57	72	73	74	75	76	77	78	79	80
La	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg



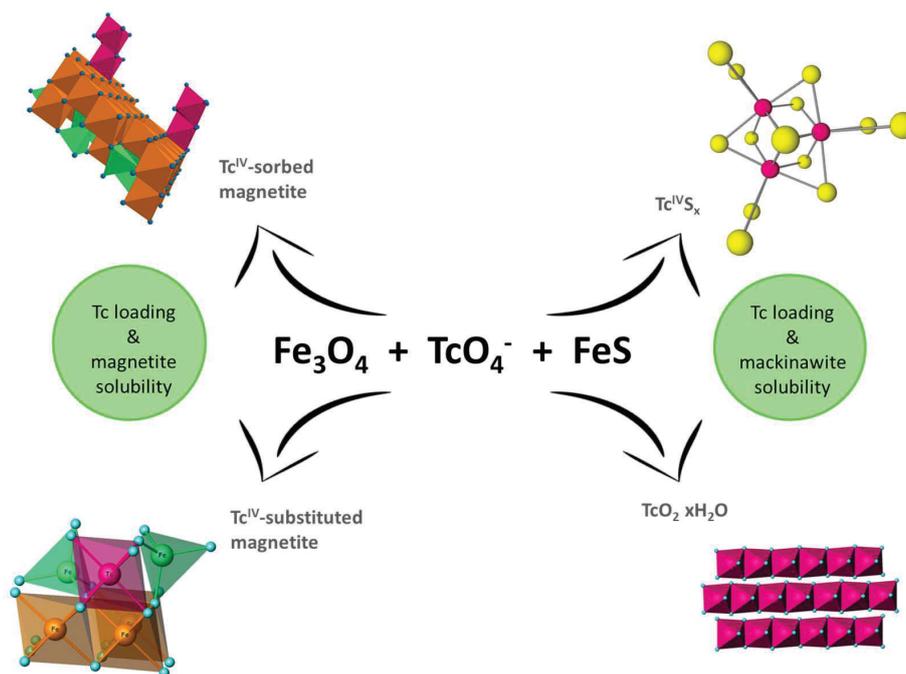
Tc sorption/redox on magnetite (Fe_3O_4)



$[Tc]_{end}: \sim 10^{-10} M$
 $pH_{end}: \sim 9$
 $pe_{end}: \sim -6$

$[Tc]_{ini}/M$	Tc loading/mg/kg
1: 2×10^{-4}	400
2: 2×10^{-4}	600
3: 2×10^{-4}	900
4: 2×10^{-5}	400
5: 2×10^{-5}	600
6: 2×10^{-5}	900

- Complete reduction of Tc^{VII} to Tc^{IV}
- Low $[Tc]_{ini}$: ES/CS coordination to FeO_6 -octahedra, partial incorporation into magnetite structure (\Leftrightarrow complete incorporation observed by Kobayashi et al. 2013 at lower pH 6.5)
- High $[Tc]_{ini}$: Sorption of $[TcO_2]$ -dimers in addition to structural incorporation of monomers
- Initial concentration explains previous contradictory results on Tc^{IV} speciation in reducing environments (Morris & coworkers, Zachara & coworkers,...)

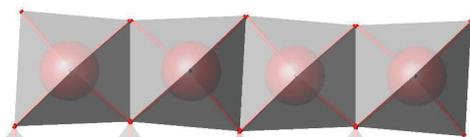
Tc^{VII} retention mechanisms by magnetite and mackinawite

E. Yalçıntaş, A. C. Scheinost, X. Gaona, M. Altmaier, *Dalton Trans.* **45**, 17874-17885 (2016).

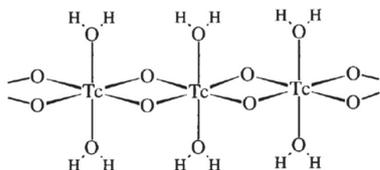
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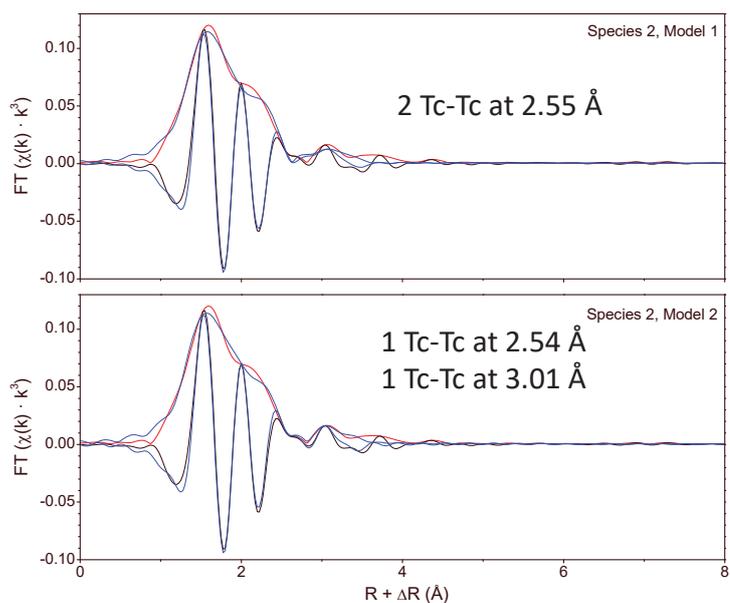
Even a simple structure like TcO₂ · xH₂O seems to be elusive.....

Alternating short (2.62 Å) “metallic”, and long (3.08 Å) oxidic Tc-Tc distances like in crystalline TcO₂ (Rodriguez et al. 2007).....



...or only short (2.57 Å) “metallic” Tc-Tc distances as derived by EXAFS for TcO₂ · xH₂O (Lukens et al. 2002)?

We can fit both models.....



Based on EXAFS alone decision is difficult.....can theory help?

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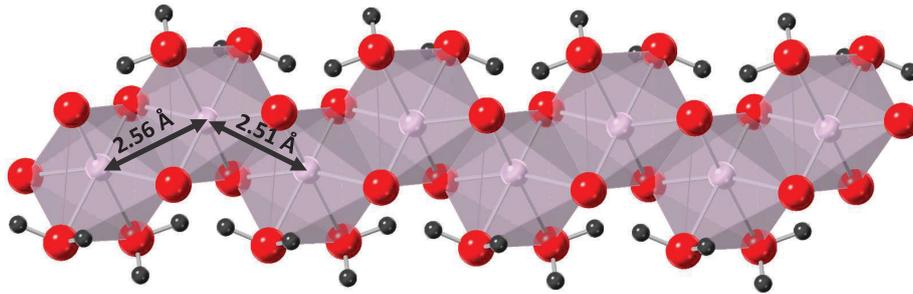
Structure of $TcO_2 \times 2H_2O$ chains: Atomistic calculations

Relative bonding energies (kJ/mol):

Structure	Termination	Rel. Energy
alpha	-H ₂ O	29.4
beta	-H ₂ O	0.0
gamma	-H ₂ O	190.3
alpha	-OH	73.3
beta	-OH	79.3
gamma	-OH	95.9

Interatomic distances (Å):

	DFT	EXAFS
4x Tc-O	1.96-2.01	2.01
2x Tc-OH ₂	2.23	2.47
2x Tc-Tc	2.51-2.56	2.55



Zigzag (β-type) chains with H₂O termination are energetically most stable, Tc-Tc distances consistent with EXAFS!

Geometry optimizations with PBE-D3/TZP(ae) and ZORA scalar relativistics

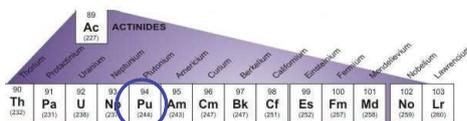
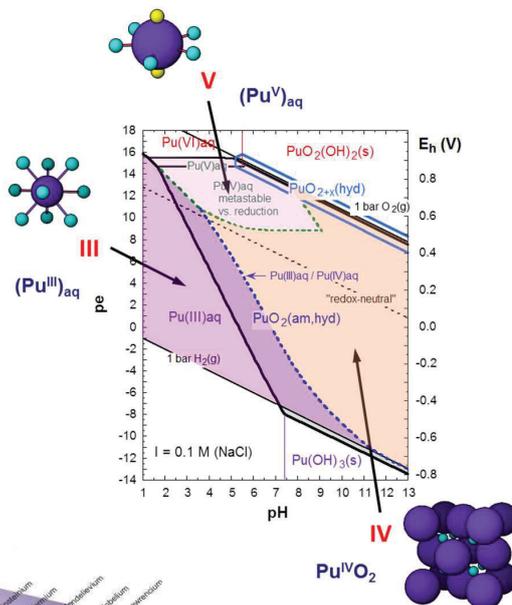
A. Faria Oliveira, A.B. Kuc, T. Heine, A. C. Scheinost, in preparation



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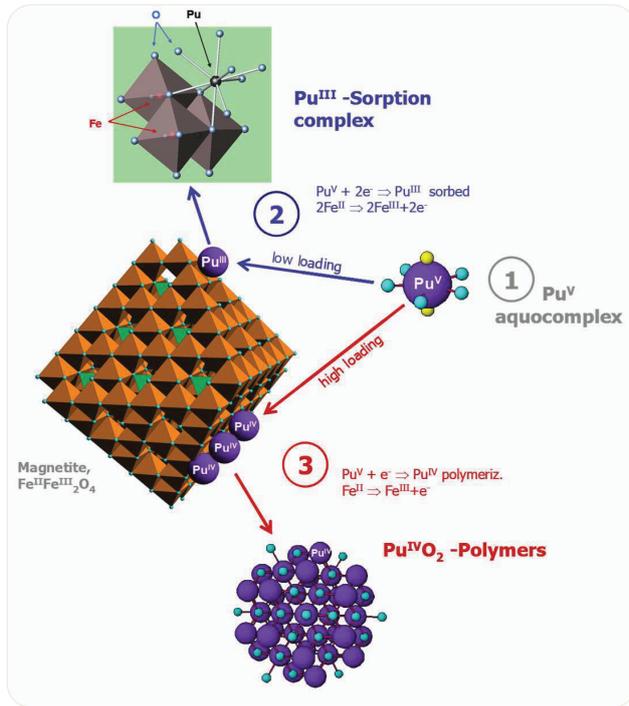
Plutonium



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Plutonium retention by magnetite {111} faces



R. Kirsch et al., *Environ. Sci. Technol.* **45**, 7267–7274 (2011)

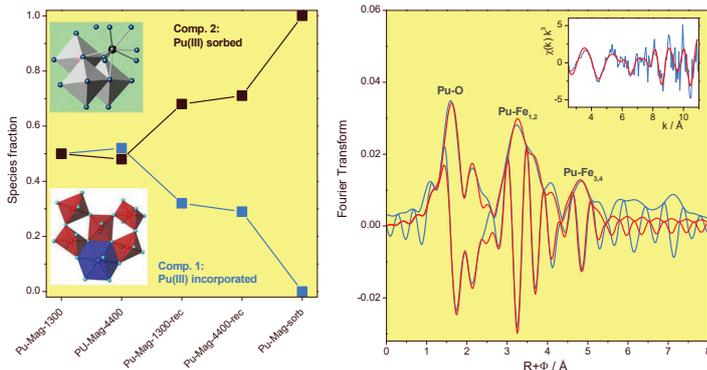
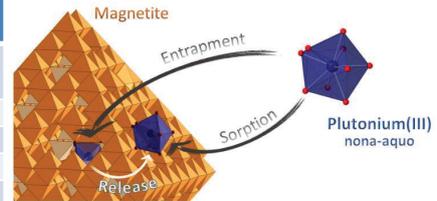


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- Mobile Pu(V) aquo complexes are sorbed and reduced to oxidation states 3 and 4
- At high surface loading, PuO₂ polymers form with low solubility (<10⁻¹⁰ M), but potential mobilization as colloids
- At low surface loading, we observed the formation of a trivalent, tri-legged Pu sorption complex with very low solubility (<10⁻¹⁰ M)
- Can Pu(III) also be incorporated by co-precipitating magnetite?

Plutonium retention by coprecipitating magnetite: Kinetic entrapment versus sorption

Paths	Component 1 Incorporated Pu(III)						Magnetite Oh site #		Component 2 Sorbed Pu(III)	
	CN	R/Å	σ ² /Å ²	ΔE ₀ /eV	χ _{res} /%	CN	R/Å	CN	R/Å	
Pu-O ₁	4.5	2.22	0.0089 ^a	8.4	11.1	6	2.06			
Pu-O ₂	3.8	2.45	0.0089 ^a					9	2.45	
Pu-Fe ₁	2.7	3.68	0.0032 ^b			6	2.95	3	3.54	
Pu-Fe ₂	1.9	3.93	0.0032 ^b			6	3.46	3	4.20	
Pu-Fe ₃	12*	5.17	0.0150 ^c			12	5.11			
Pu-Fe ₄	8*	5.47	0.0150 ^c			8	5.42			



T. Dumas et al., *ACS Earth Space Chem.* **3**, 2197-2206 (2019).

- Pu(III) can be incorporated by magnetite, by creating a pyrochlore-like local cluster within the magnetite structure
- But only up to 50%, and with time/recrystallization, this amount decreases to 33%
- Surface complexation seems to be thermodynamically more important
- Incorporation seems to proceed through kinetic entrapment



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EXAFS: Experiments with strict conservation of oxidation states

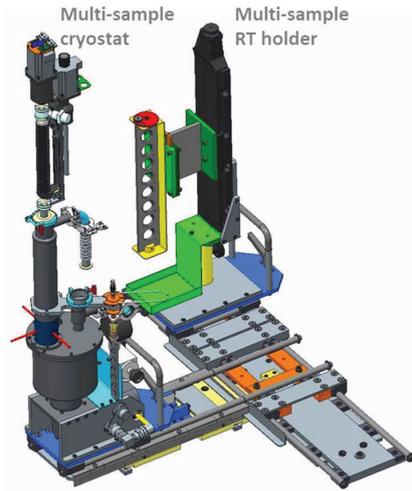


Sample preparation

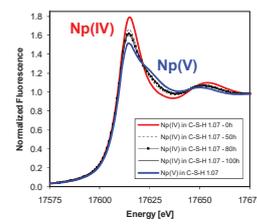
anoxic gloveboxes (<2 ppmv O₂)
flash-freezing in LN₂ (kinetics!)
gamma-spectroscopy on dry ice

Transport to ESRF

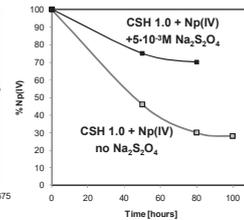
Special LN₂-Dewar with insert for RN samples
gamma-spectroscopy on dry ice



Np(IV) oxidation test:



(Courtesy X. Gaona, PSI)



X-ray absorption spectroscopy

Multisample Cryostat at 15 K (-258 °C)
special double-confinement sample holders (design CEA)

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The Rossendorf Beamline (ROBL-II) @ ESRF



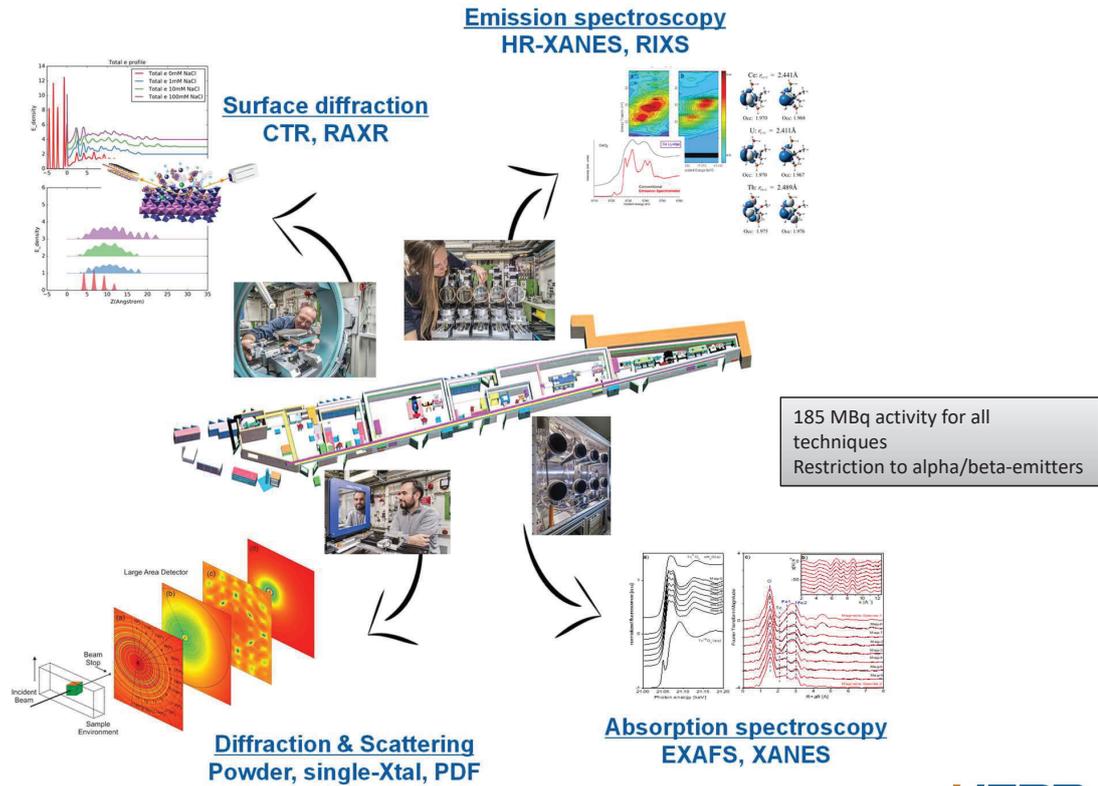
- ✓ Operated at ESRF (The European Synchrotron, Grenoble, France) by Helmholtz-Zentrum Dresden-Rossendorf since 1998
- ✓ Embedded within Helmholtz Association's (HGF) research program NUSAFE (Nuclear Waste Management)
- ✓ Dedicated to the chemistry of actinoides (Th, Pa, U, Np, Pu, Am, Cm, ..., Cf, ...) and fission products (Se, Tc, Sn, I, lanthanoides)
- ✓ X-ray absorption spectroscopy to study oxidation state (XANES) and local structure (EXAFS) of these elements
- ✓ 2016-2020 major upgrade (4.5 Mio EUR) of ROBL into a synchrotron toolbox for actinide research (ROBL-II)
- ✓ 2016-2020 major upgrade (150 Mio EUR) of ESRF into 4th generation synchrotron (Extremely Brilliant Source)

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ROBL-II: A synchrotron toolbox for actinide research



HZDR

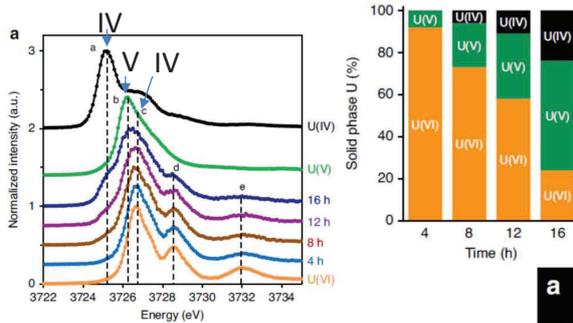
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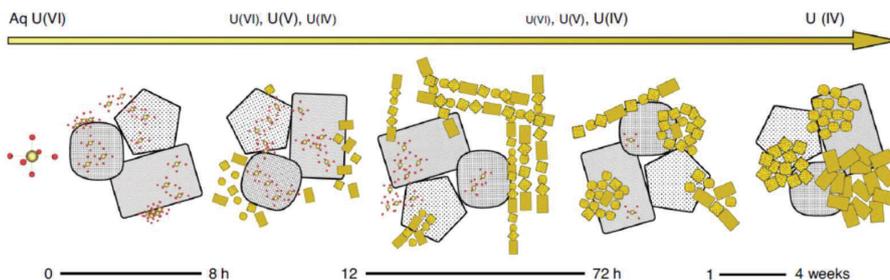
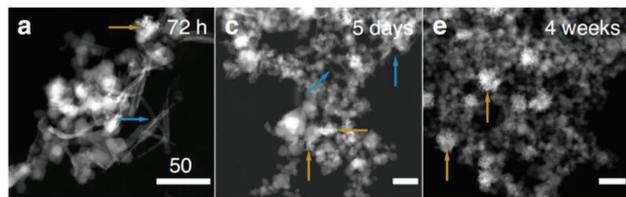
92

U
Uranium
238.029

U(VI) reduction by magnetite: U-M4 HERFD-XANES



- Formation of U(V) during transition from U(VI) to U(IV)
- UO_2 phase forms first as nanowires through oriented attachment of NPs...
- ...before collapsing into nanoclusters

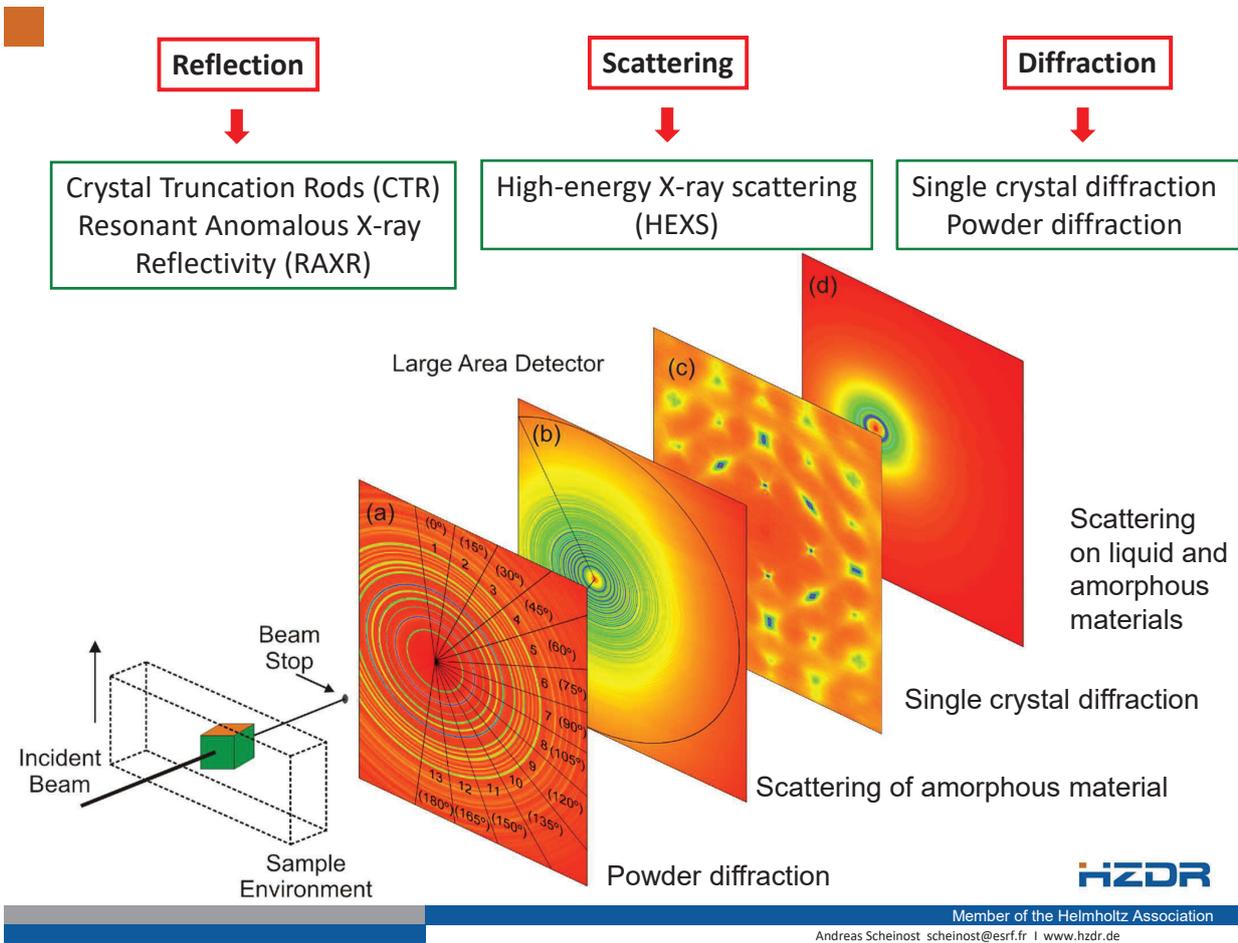


Z. Pan *et al.*, *Nature Communications* 11 (Aug 10, 2020).

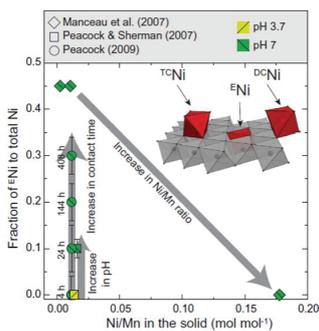
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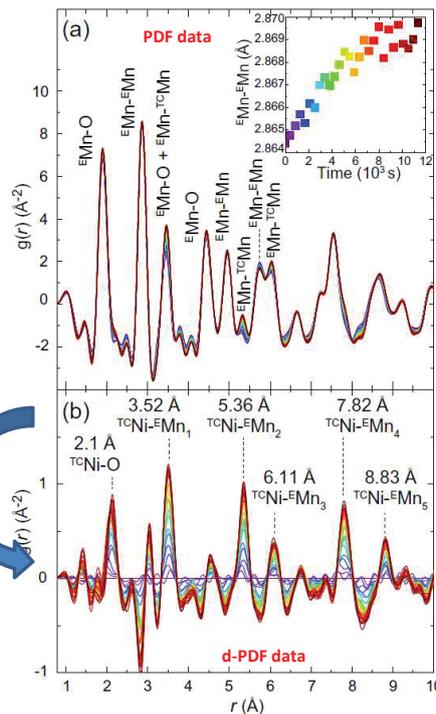
Andreas Scheinost scheinost@esrf.fr | www.hzdr.de



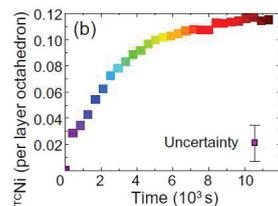
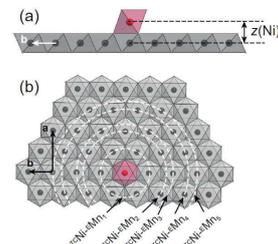
Time-resolved X-ray pair distribution function: Structural modification of vernadite ($\delta\text{-MnO}_2$) by Ni



- Complementary to XRD: allows for a detailed analysis of structural modifications, at the molecular scale
- PDF data are collected as a function of time. Upon subtraction of the initial data, allows probing any structural modifications (d-PDF analysis)
- For example, for clay minerals (figures: sorption by $\delta\text{-MnO}_2$): determination of the mechanisms of Ni adsorption
- Complementary to EXAFS: correlations can be observed up to several tens of nanometers, but (i) lower sensitivity than EXAFS and (ii) any modification of the sorbing phase will induce complexity in data analysis, because changes will be reflected in the d-PDF



All correlations observed in the d-PDF are consistent with Ni sorption above a vacancy



After Grangeon et al., Chemical Geology, 2017



**We are happy to
welcome you at
ROBL-II....**





WP5 - FUTURE: 3RD ANNUAL MEETING

Radionuclide transport in clay

17 November 2021 • Zoom • Martin Glaus



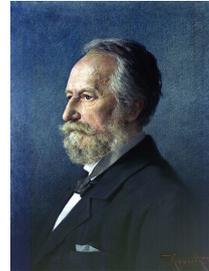
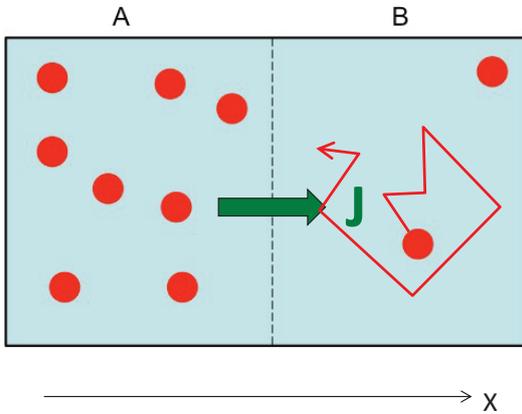
The project leading to this application has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement n° 847593.



OUTLINE

- **Diffusion processes:**
 - Concentration gradients (solution phase) as the driving force
- **Experimental methods and information gained**
 - Inert media: steady-state phase vs. transient phase
 - Charged clay media
- **Effects of surface charges in clays:**
 - Surface diffusion of cations
 - Anion exclusion
- **Diffusion parameters in charged clays**
 - Dependence on chemical conditions of solution phase (ionic strength, pH, competing ions)
- **Application in performance assessment:**
 - Bottom-up approach for diffusion
- **Conclusions**

DIFFUSION IN DILUTE BULK SOLUTIONS



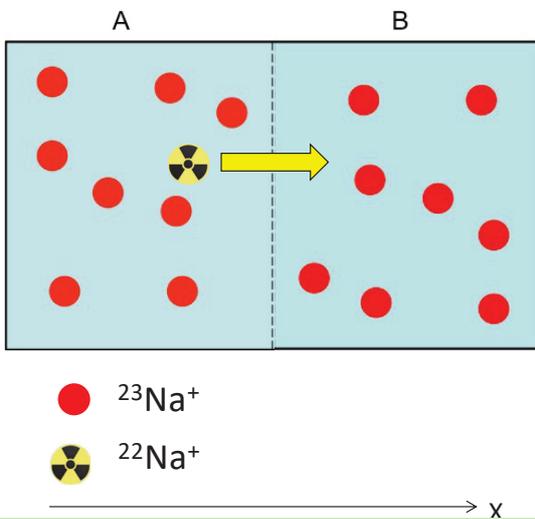
A. Fick (1829 – 1901)

Mass flux (J) ~ (e.g. concentration) gradients

$$J = -D \frac{\partial C}{\partial x} \quad D: \text{Diffusion coefficient}$$

Focus on concentration-driven diffusion in the following presentation!

TRACER DIFFUSION IN DILUTE BULK SOLUTIONS

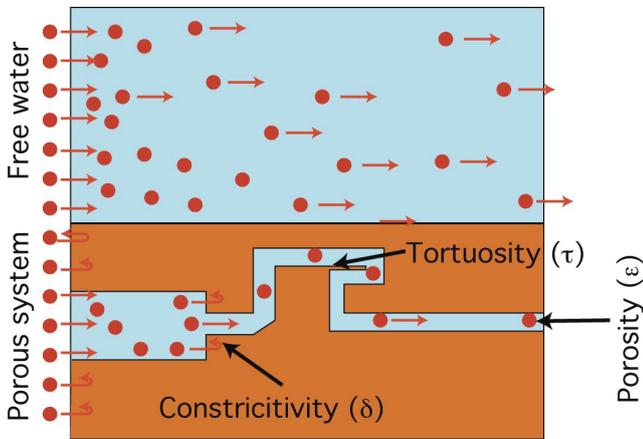


Mass flux (J) ~ (e.g. concentration) gradients

$$J_{\text{tracer}} = -D \frac{\partial C_{\text{tracer}}}{\partial x}$$

$$J(^{23}\text{Na}^+) = 0$$

DIFFUSION IN INERT POROUS MEDIA: 'PORE DIFFUSION MODEL'



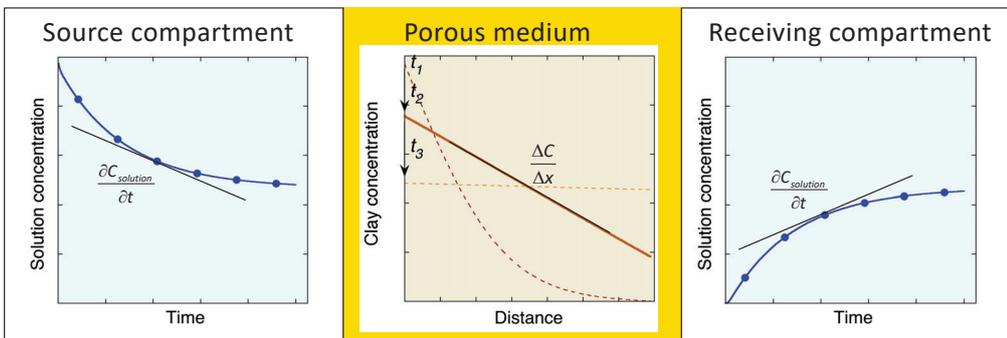
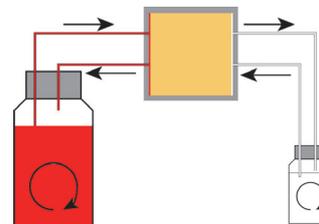
Effective diffusion coefficient D_e :

$$D_e = \varepsilon \frac{\delta}{\tau^2} D_0 = \frac{\varepsilon}{G} D_0 = \dots$$

- D_e : Effective diffusion coefficient
- D_0 : Bulk diffusion coefficient
- ε : Porosity → affecting cross section area
- δ : Constrictivity → affecting mobility
- τ : Tortuosity → affecting diffusion path (also termed as 'geometry factor' G)

Further parameter influences, see:
 Shackelford, C.D., Daniel, D.E. (1991) *J. Geotech. Eng. ASCE* 117, 467–484.

THROUGH DIFFUSION EXPERIMENT INERT TRACER



$$\frac{\partial C_{solution}}{\partial t} \Rightarrow J_{(t)}$$

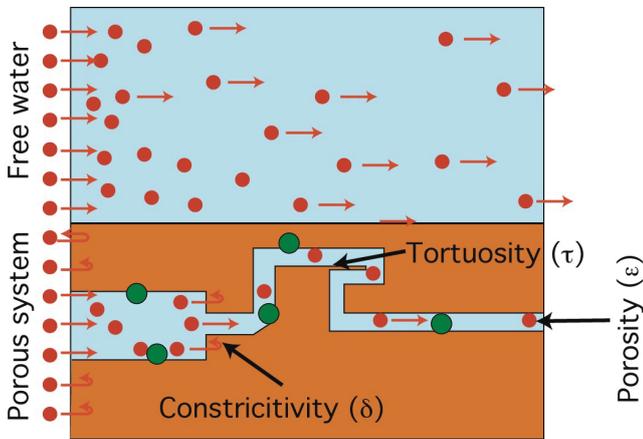
$$D_e = -J_{(t)} \left(\frac{\Delta C}{\Delta x} \right)_t^{-1}$$

"Steady-state (approx)": Flux(upstream) = Flux(downstream => linear profile in clay

Primary observables: Changes of mass/concentration (solution) as a function of time
 Concentration (gradient)/difference in clay

Derived quantities: Flux [mol/m²/s] (from time derivative of concentration in solution)
 D_e [m²/s] (from concentration gradient/differences in clay)

DIFFUSION IN INERT POROUS MEDIA



The rock capacity factor (α)

$$\alpha = \varepsilon + \rho_{bd} R_d$$

ε: Porosity

ρ_{bd}: Bulk dry density (kg m⁻³)

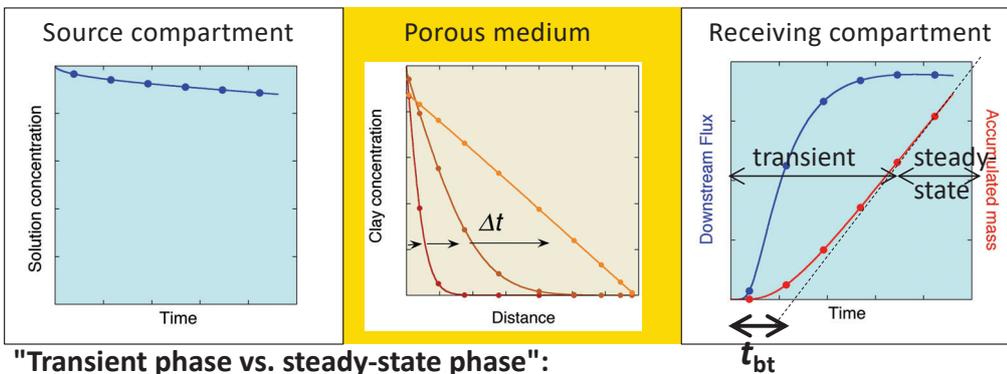
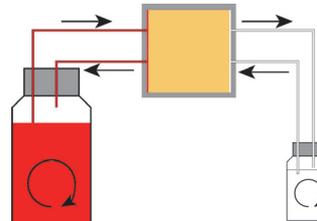
R_d: Sorption distribution coefficient (m³ kg⁻¹)

... is reflected by:

1. the breakthrough behaviour
2. The concentration ratio 'clay/aqueous phase'

$$\alpha = \frac{C_{tot}}{C_{aq}}$$

**THROUGH DIFFUSION EXPERIMENT
SORBING TRACER, C_{dsb} ≈ 0**



Fick's 2nd law:

$$\frac{\partial C}{\partial t} = D_a \frac{\partial^2 C}{\partial x^2} = \frac{D_e}{\alpha} \frac{\partial^2 C}{\partial x^2}$$

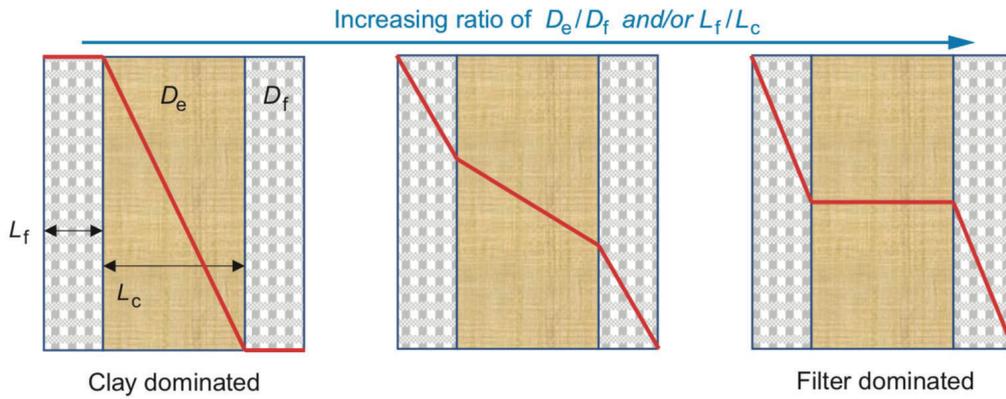
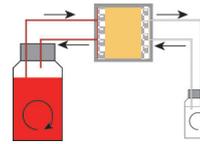
Primary observables: Breakthrough time (t_{bt})
Concentration ratio
Flux in steady-state phase

Derived quantities:

α
R_d
D_e



CONFINING FILTERS



$$D_{e,all} = \frac{D_{e,f}D_{e,c}(2l_f + l_c)}{2l_fD_{e,c} + l_cD_{e,f}}$$

Glaus et al (2015) J. Contam. Hydrol. 177-178, 239-248.

"Steady-state phase":

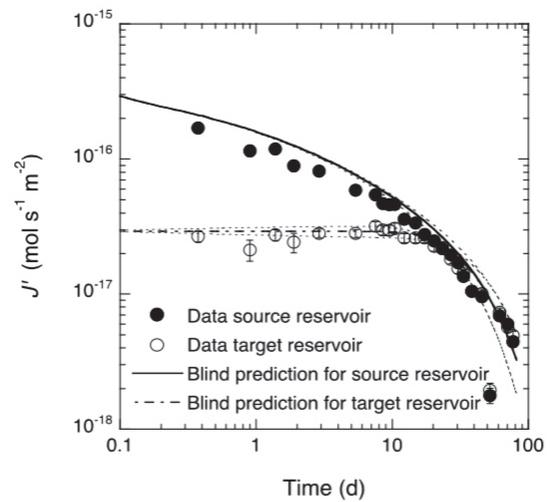
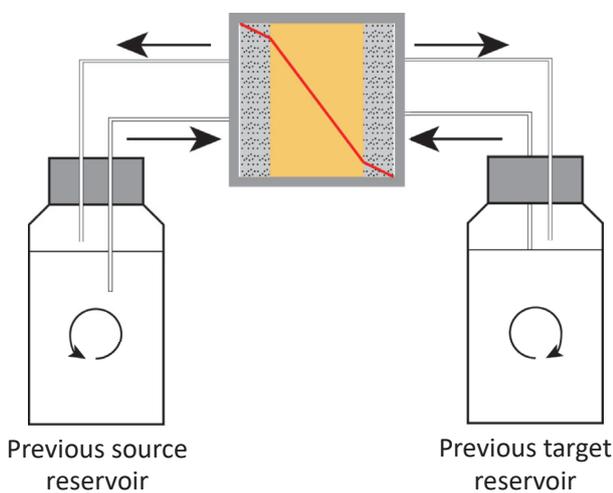
Primary observables: J_{all} (overall flux), ΔC_{all} (overall concentration difference)

Derived: ΔC_{clay} , $D_{e,c}$ (effective diffusion coefficient in clay)



OUT-DIFFUSION

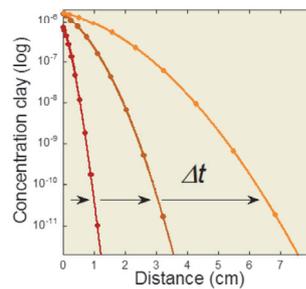
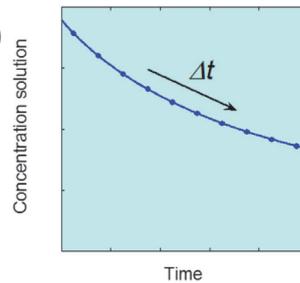
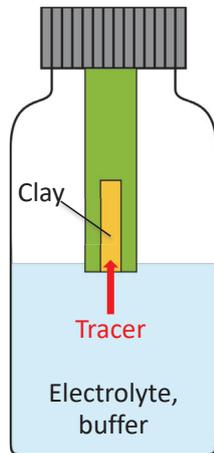
- Following through-diffusion
- Crux: Knowledge of initial conditions





IN-DIFFUSION

- Concentration depletion in reservoir ($0 - t$)
- Concentration profile (at time t)



- Sampling in solution
- Tracer concentration in reservoir (as a function of time)

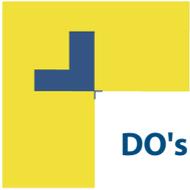
- Slicing of clay sample
- Tracer profile in clay (as a function of distance)



DO's AND DON'Ts

- **Setup:**
 - Use filter confinement only if required
 - Avoid advection (gradients in hydraulic head)
 - Materials (diffusion cell + filters): Contamination of solutions / uptake of solutes
- **Dimensions of diffusion geometry:**
 - Optimisation of geometry necessary (avoiding filter dominance)
 - Thickness \longleftrightarrow gradients, flux
 - Cross section area \longleftrightarrow flux
 - Volumes of clay and filter (\longrightarrow change mass transfer)
- **Time aspects:**
 - Diffusion length \longleftrightarrow apparent diffusion coefficient (D_a)
 - Through-diffusion: Duration 6x Break-through time
 - In-diffusion: Profile resolution
- **Cross-check of results:**
 - Outdiffusion, profile analysis

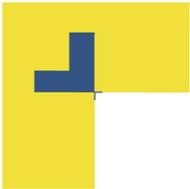
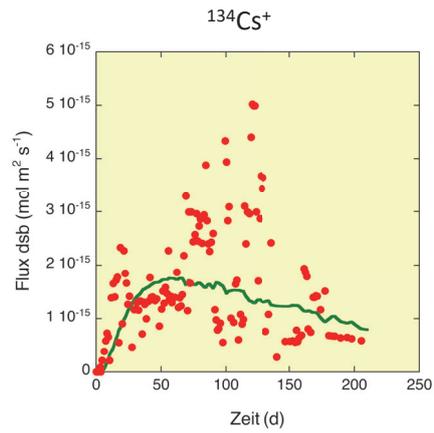
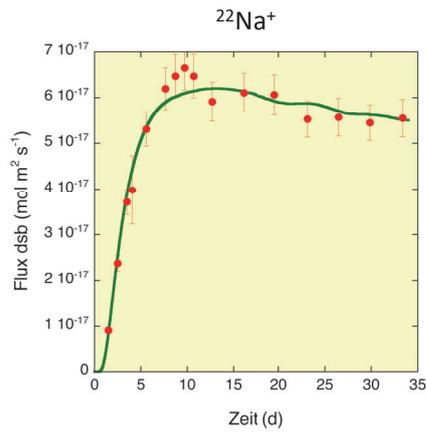




DO's AND DON'Ts

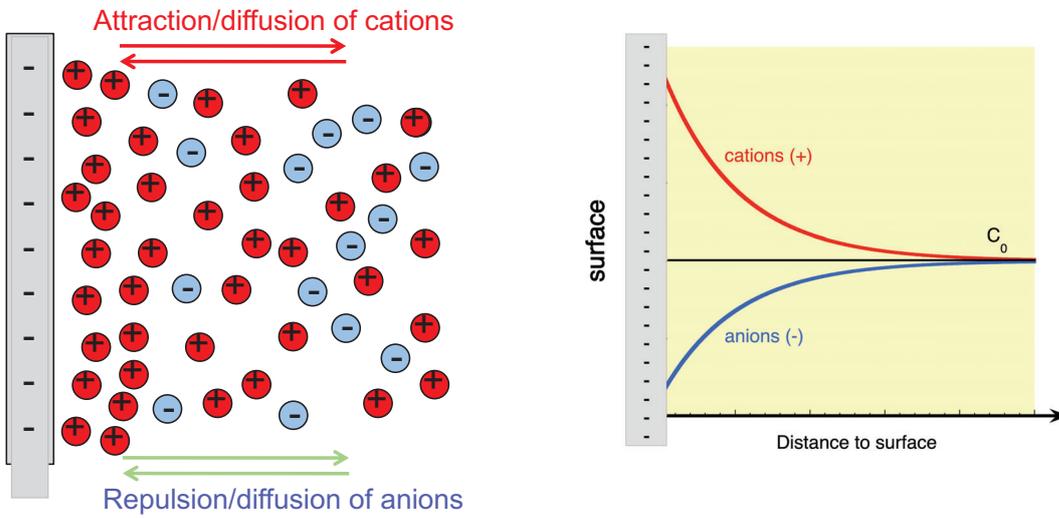
- **Through-diffusion:**

- Probing of boundaries (only!)
- Independent checks most valuable (out-diffusion, profile)
- Verification of modelling assumptions necessary



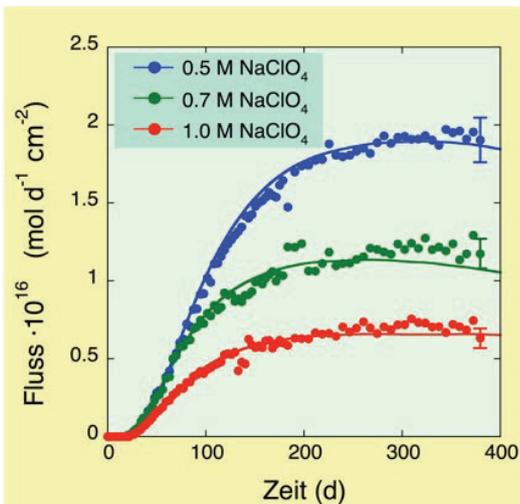
DIFFUSION PROCESSES IN CHARGED CLAYS: "SIMPLE CATIONS + ANIONS"

DIFFUSION IN CHARGED POROUS MEDIA



- ⇒ Larger fluxes of cations
- ⇒ Smaller fluxes of anions

DIFFUSION OF Sr^{2+} IN COMPACTED MONTMORILLONITE



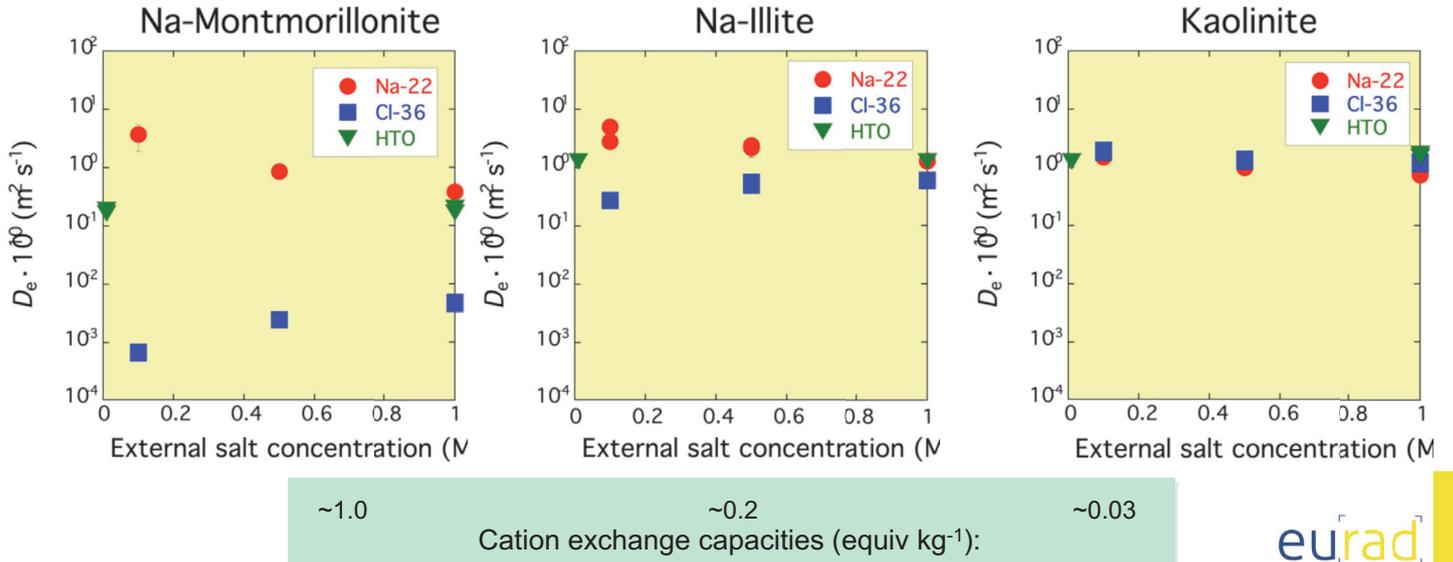
- Increasing flux with decreasing ionic strength
- ± same breakthrough times

- ⇒ D_e and R_d correlated (D_a ± same)
- ⇒ Cation-exchange species mobile?!
- ⇒ "Interlayer diffusion"

$$D_e = -J_{(t)} \left(\frac{\Delta C}{\Delta x} \right)_t^{-1}$$

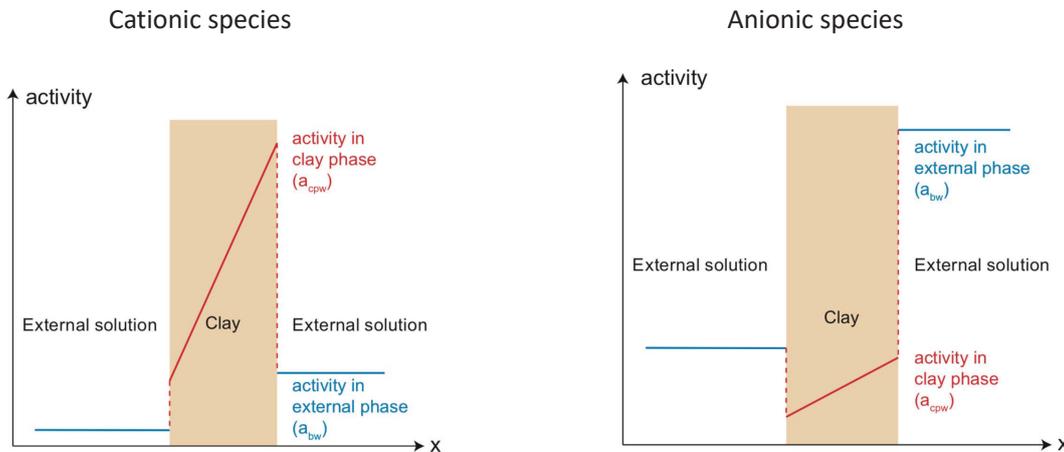
Glau et al. (2007). Environ. Sci. Technol. 41, 478–485.

DIFFUSION OF CATIONS AND ANIONS IN DIFFERENT CLAY TYPES



Glaus et al. (2010). Geochim. Cosmochim. Acta 74, 1999-2010.

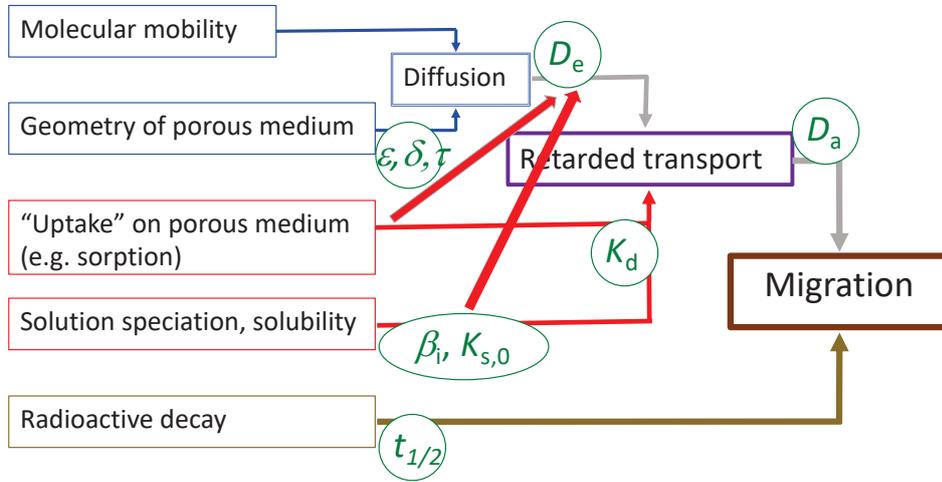
"EFFECTIVE CONCENTRATION GRADIENTS IN CLAY"



Flux related to concentration gradients in aqueous phase
 $\Rightarrow D_e$ depends on "chemical enrichment factors"

Glaus et al. (2017). PSI Bericht 17-08.

CONCLUSIONS: PARAMETER SCHEME FOR RETARDED TRANSPORT



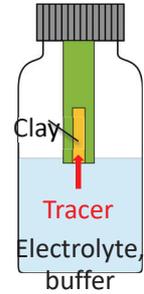
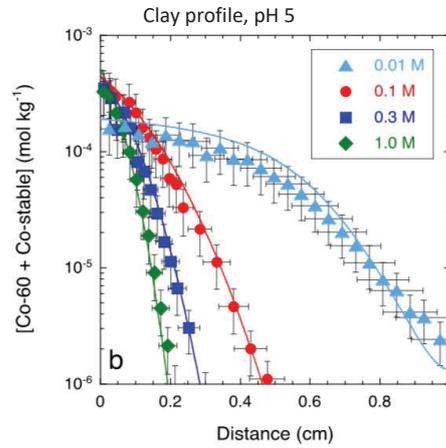
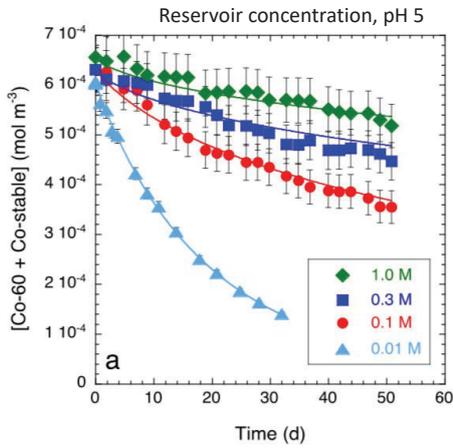
Fick's 2nd law (1-D)

$$\frac{\partial C}{\partial t} = D_a \frac{\partial^2 C}{\partial x^2}$$

$$D_a = \frac{D_e}{\varepsilon + \rho_{bd} K_d}$$

DIFFUSION PROCESSES IN CHARGED CLAYS: SURFACE COMPLEXING CATIONS

IN-DIFFUSION OF Co²⁺ IN COMPACTED ILLITE, pH 5 AND 9



- ± same boundary concentrations
- Increasing diffusion depth
- D_e (pH 5) = D_e (pH 9)

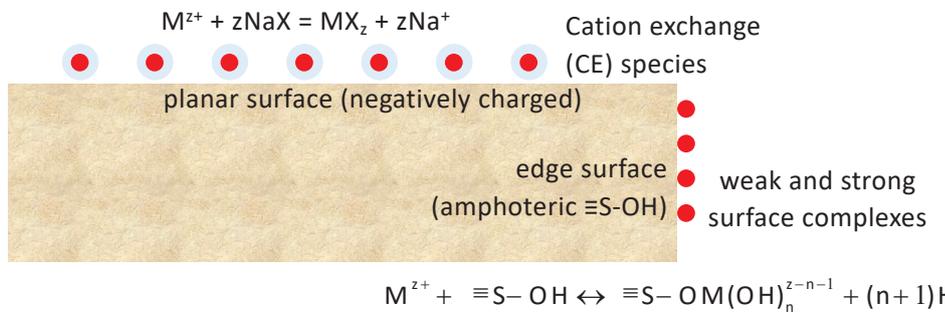
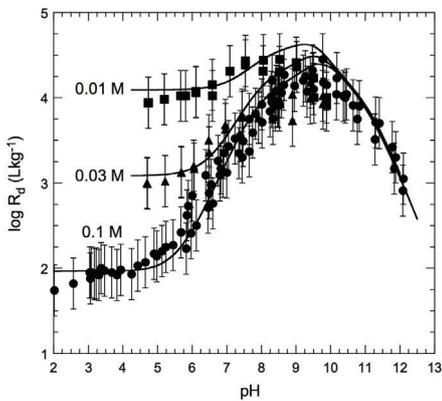
=> R_d ± same

=> D_e increasing with decreasing ionic strength

Glaus et al. (2015). Geochim. Cosmochim. Acta 165, 376-388.

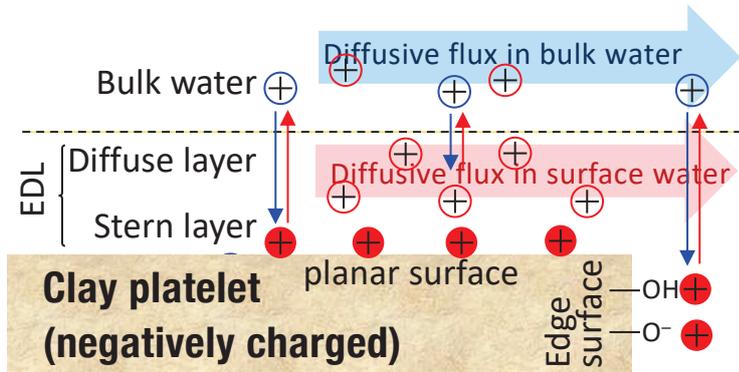
2SPNE SC/CE SORPTION MODEL

2 sites protolysis non-electrostatic surface complexation and cation exchange



Bradbury & Baeyens (2017). Nagra Technical Report NTB 17-13.

2SPNE SC/EDL DIFFUSION + SORPTION MODEL

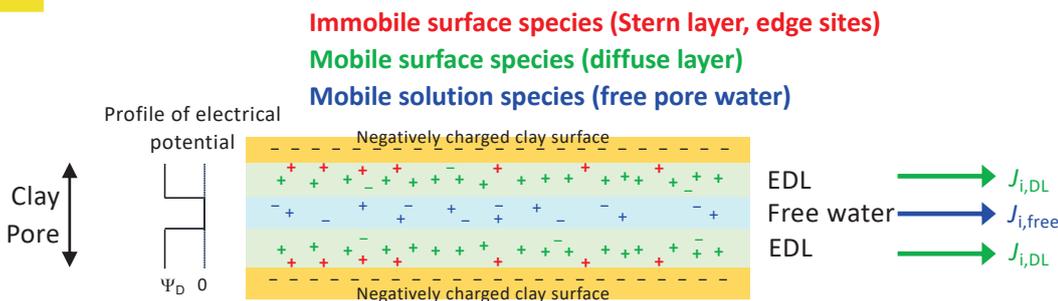


EDL = Electrical double layer

- Mobile species: Aqueous phase, Diffuse layer (open symbols)
- Immobile species: Stern layer species, Strong + weak sites (closed symbols)
- Red / blue circles: Different cationic species

Glaus et al. (2021). ACS Earth Space Chemistry 5, 2621-2625.

ENRICHMENT / DEPLETION FACTOR \leftrightarrow EFFECTIVE DIFFUSIVITY



$$D_e = \frac{\varepsilon}{G} D_w \left(f_{free} + f_{DL} \frac{C_{DL}}{C_{free}} q_{\eta} \right)$$

Flux contribution "free" (under f_{free})

Flux contribution "DL" (under $f_{DL} \frac{C_{DL}}{C_{free}} q_{\eta}$)

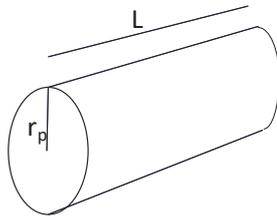
Concentration enrichment/depletion factor

- depending on pore water composition: ionic strength, pH, competing species, ligands
- depending on surface properties: Charge density, charge shielding

Appelo et al. (2010). Geochim. Cosmochim. Acta 74, 1201–1219

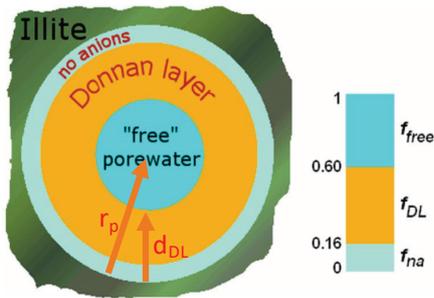
Glaus et al. (2015). Geochim. Cosmochim. Acta 165, 376-388.

PORE GEOMETRY IN EDL MODEL



Volume = $\pi r_p^2 L$
 Surface $\cong 2\pi r_p L$
 Volumetric-specific surface = $2/r_p$

=> Pore radius from specific surface area

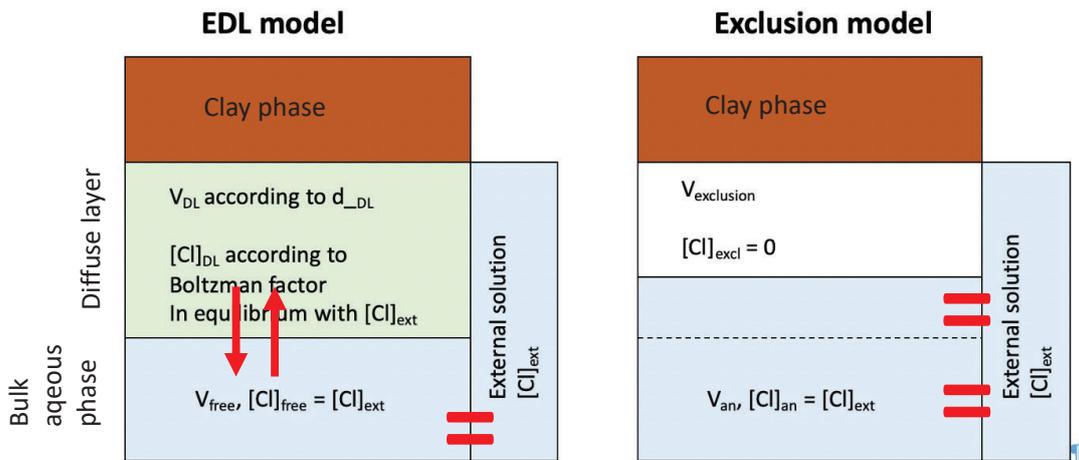


=> d_{DL} = Debye length(I) $\cdot n_{DL}$
 => f_{free} from "difference between pore radius and Donnan thickness d_{DL} "

Glaus et al. (2015). Geochim. Cosmochim. Acta 165, 376-388.

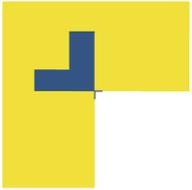


ANIONS: CONCENTRATION DEPLETION versus COMPLETE EXCLUSION



$R_d(Cl)$ ← "Translation" readily available by recalculation → Anion accessible porosity ... porosity fraction





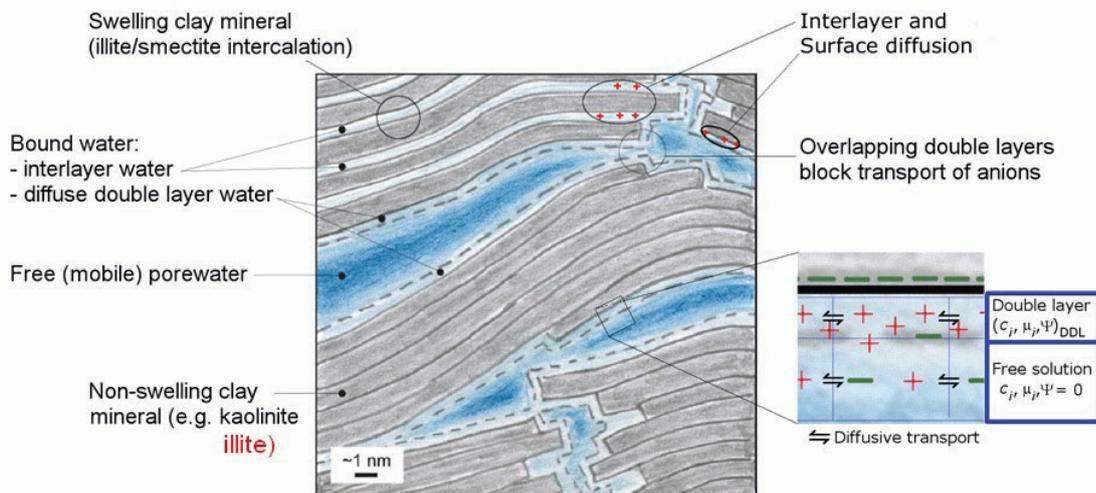
FROM CLAY MINERALS TO CLAY ROCKS



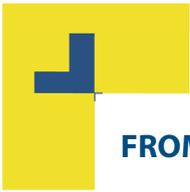
PORE STRUCTURE OF CLAYS

Neutral and positively charged species:
- total porosity

Negatively charged species:
- reduced porosity
- anion exclusion



Appelo et al. (2010). Geochim. Cosmochim. Acta **74**, 1201-1219.



FROM MODEL TO REAL SYSTEMS

- Pure mineral phases
- Single electrolytes
- Synthetic pore water
- Isotropic diffusion
- Equilibrium situation
- Representative nuclide



- Mineral assemblage, clay rock
- Equilibrium pore water

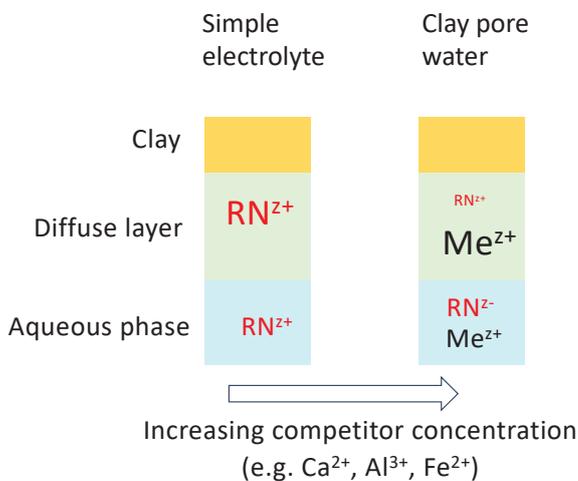


- Anisotropic diffusion
- Kinetic phenomena, long-term processes
- Periodic system of elements / species

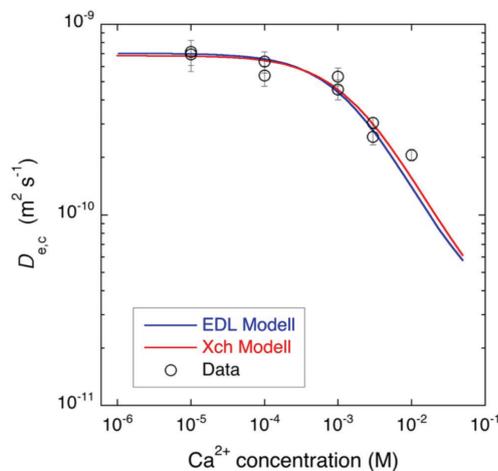
Bottom-up scheme in WP Future:
 Bottom-up I: from disperse minerals to compacted minerals
 Bottom-up II: from single minerals to clay rock



COMPETITION EFFECTS IN EDL DIFFUSION



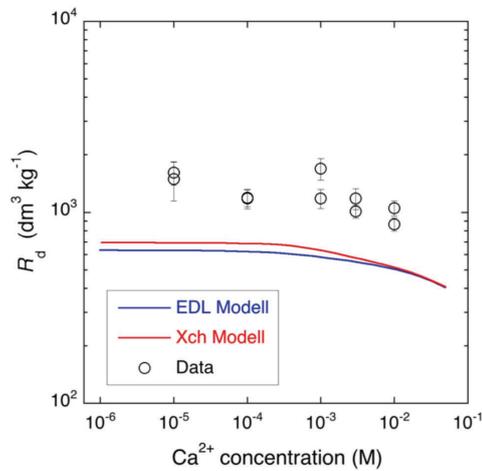
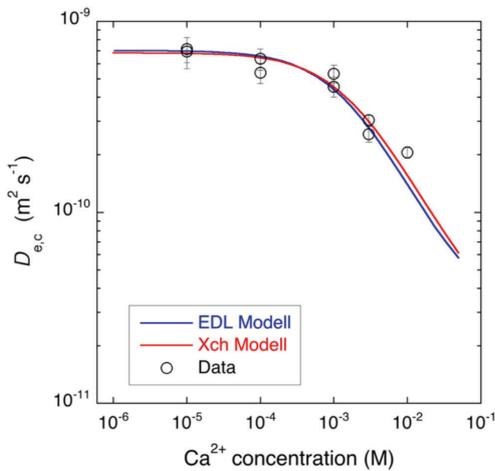
⁵⁷Co²⁺ diffusion in illite (pH 5, NaCl 0.1 M, 1700 kg m⁻³)



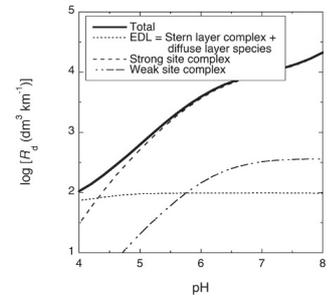
Glaus et al. (2021). ACS Earth Space Chemistry 5, 2621-2625.



CATION COMPETITION IN SINGLE MINERAL (ILLITE): DIFFUSION BEHAVIOUR OF $^{57}\text{Co}^{2+}$



$^{57}\text{Co}^{2+}$ diffusion in illite
(pH 5, 0.1 M, 1700 kg m^{-3})

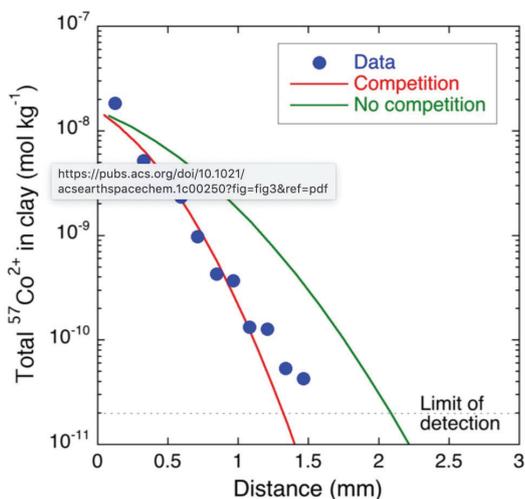


- Ca^{2+} competes with Co^{2+} in the double layer $\Rightarrow D_e$ depends on Ca^{2+} concentration
- No competition at edge surfaces (strong + weak sites) $\Rightarrow R_d$ almost not affected

Glaus et al. (2021). ACS Earth Space Chemistry 5, 2621-2625.

FROM ILLITE \rightarrow OPALINUS CLAY

Measured tracer profile + semi-blind prediction



Extrapolation to Opalinus Clay:

- Scaling of cation-exchange capacity
- Scaling of specific surface
- Slight adaptation of strong site complexes (taking into account competition with FeII)

Glaus et al. (2021). ACS Earth Space Chemistry 5, 2621-2625.

IMPACT ON STRONGLY SORBING RADIONUCLIDES

	Sorption (K_d)	Effective diffusivity (D_e)
Solution parameters		
Ionic strength	weak: only cation exchange	strong: Enrichment factor
pH	strong: edge species (strong, weak)	strong: hydrolysed RN's
Competing alkaline earths	weak: only cation exchange	strong: Enrichment factor
Competing transition metals	strong: edge species (strong, weak)	weak: Conc. too low
Ligands	strong: edge species (strong, weak)	strong: Enrichment factor
Surface parameters		
CEC	weak: only cation exchange	strong: Enrichment factor
Index cation	weak: only cation exchange	strong(?): Enrichment factor
Edge sites	strong	weak

CONCLUSIONS

- **Methods for diffusion measurements are well established:**
 - Pitfalls awaiting
 - Interpretation not straightforward in all respects
- **Use of effective diffusion coefficients implies:**
 - Careful evaluation of diffusion/retardation parameters necessary when deducing the properties of intact rock from ideal model systems
 - **Consistency required for sorption and diffusion models**
- **Simplifications for application in performance assessment are defensible, provided they are based on sufficient process understanding**
- **Open issues (technical/conceptual):**
 - Dissolution/precipitation phenomena in nanoscale pores
 - How to include solid-solution phenomena in 'retardation factors' and apparent diffusion coefficients (D_a)



ACKNOWLEDGMENTS

Our thanks go to

- Nagra for partial financial support
- L. Van Loon, S. Frick, P. Bunic, W. Müller
- C.A.J. Appelo (Phreeqc modelling)
- Eurad project organisation



WP FUTURE

Training materials

Transport processes in crystalline rock

*Václava Havlova (ÚJV Řež), Cornelius Fischer (HZDR), Ulrich Noseck (GRS),
Maria Siitari-Kauppi (Uni Helsinki)*



The project leading to this application has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement n° 847593.

17.11.2021

FUTURE WP, Annual meeting, 17.11.2021



CONTENT



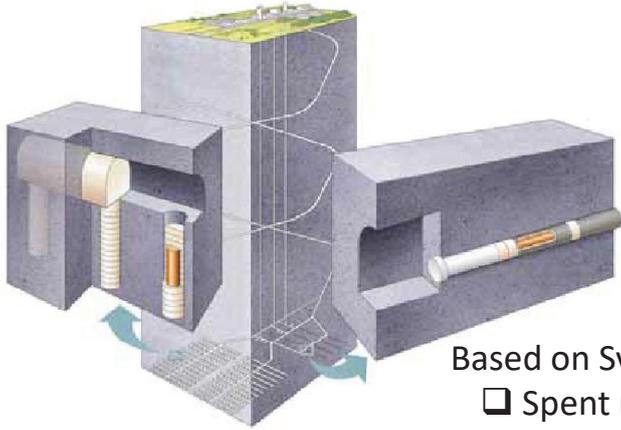
- 1. Introduction: Deep Geological Repository in Crystalline rock**
- 2. Crystalline rocks**
- 3. Main RN transport processes in crystalline rock**
 1. Advection and dispersion
 2. Retention
 - 2.1 Diffusion
 - 2.2 Sorption
- 4. Transport data used in safety assessment**

How to gain data?

 1. Laboratory studies
 2. In situ studies
 3. Natural analogues
- 5. Conclusions**

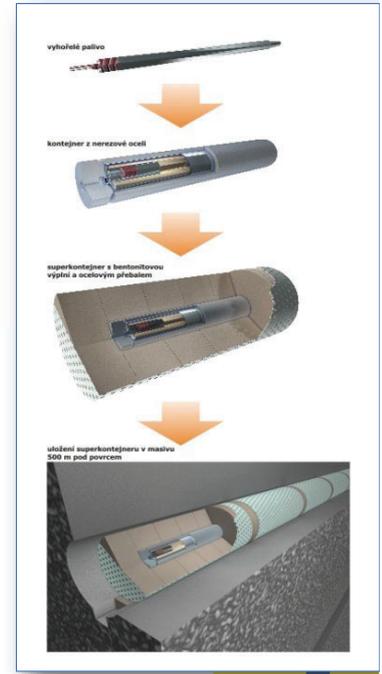


1. Introduction: Deep geological repository (DGR) in crystalline rocks

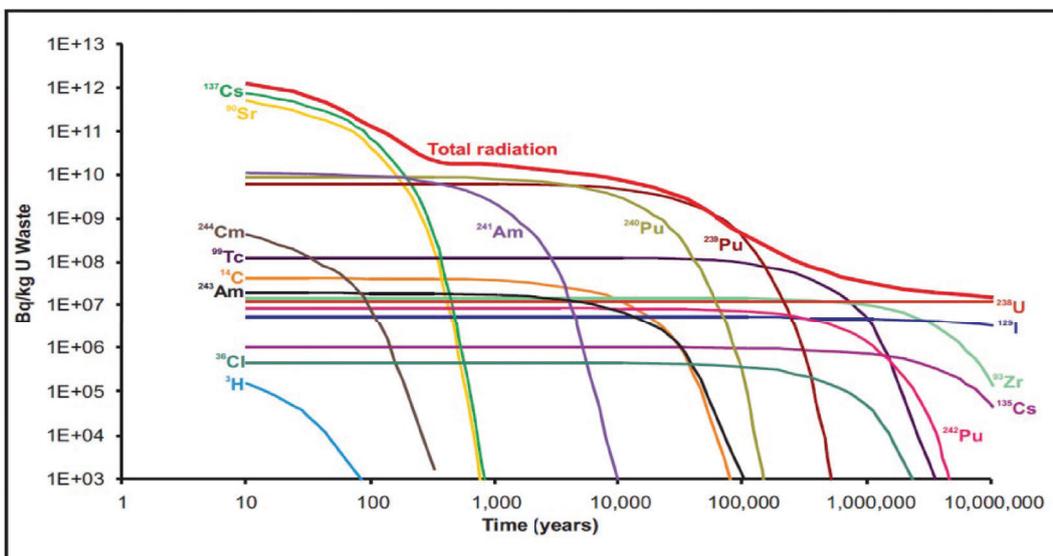


Based on Swedish KBS-3 concept

- Spent nuclear fuel
- carbon steel/ copper container
- bentonite
- crystalline host rock

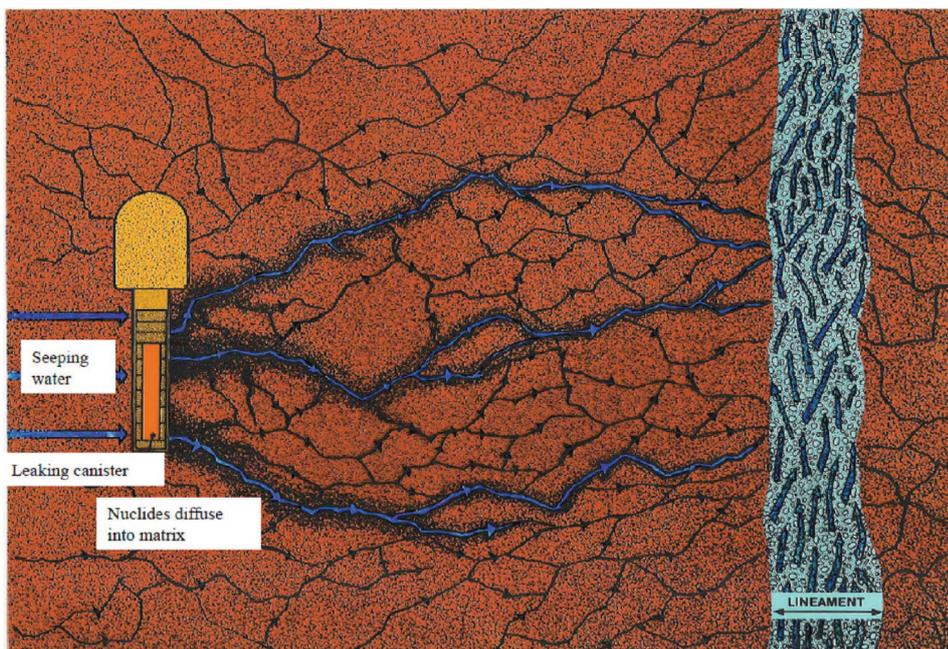


1. Introduction: Why we have to think over long time periods?



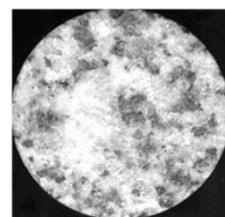
M. Herod, *GeoPoll: What should we do with radioactive waste?*, 2015.

1. DGR in crystalline rock

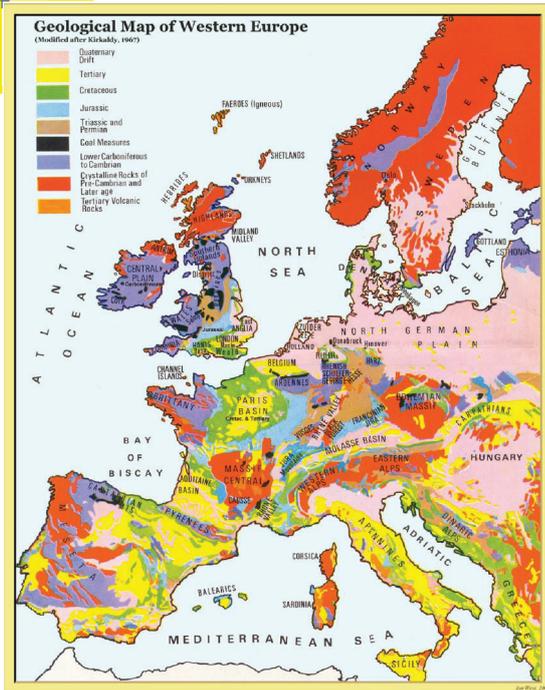


1. Crystalline host rock Safety functions

- **Isolation of the waste**
 - Protection of the technical barriers
 - Low water flow in / around near-field → Low release rates
 - Favourable chemical conditions
- **Transport barrier for radionuclides**
 - Long transport times
 - Radioactive decay
- **Dispersion / Dilution**
 - Diffusion / dispersion / dilution in space (3-D)
 - Dispersion in time (for peak release from near-field)



2. Crystalline massives in Europe



- Scandinavia (Sweden, Finland)
- Central Europe (Czech Republic, Germany, France)
- Spain
- Alpine countries

DGR in crystalline

- Sweden, Finland
- Czech Republic
- Slovakia (?), Spain (?), GB(?), Germany (?)



<https://earthymission.com/what-do-you-tread-on-in-different-parts-of-western-europe/>

2. Crystalline rocks

Magmatic (Igneous) rocks



Hrádek (CZ)

Äspö (SE)

- formed through the cooling and solidification of magma within the Crust
- composition: quartz, plagioclase, feldspar, mica, accessory minerals

Metamorphic rocks

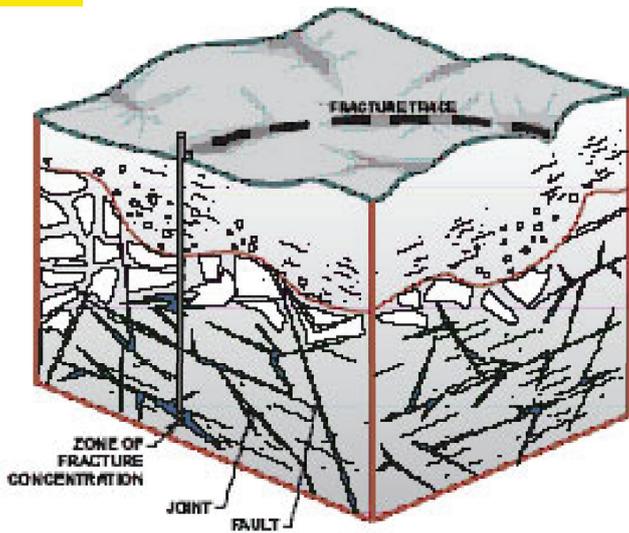


URL Bukov (Kráví hora)

- transformation of existing rock types. The original rock (protolith) is subjected to heat (temperatures greater than 150 to 200 °C) and pressure (150 megapascals (1,500 bar)) causing profound physical or chemical change
- composition: differs according to original rock
- Migmatites: similar composition to granites; amphibolites or granulites: differ



2. Fractured crystalline massives



Migmatite from Bukov URL (CZ) with calcite infill – FUTURE WP

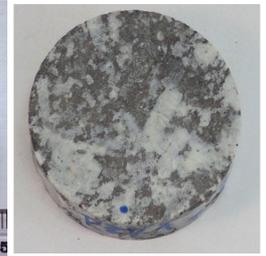
Rock Matrix



Äspö (SE)

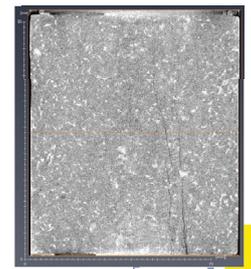


Grimsel (CH)



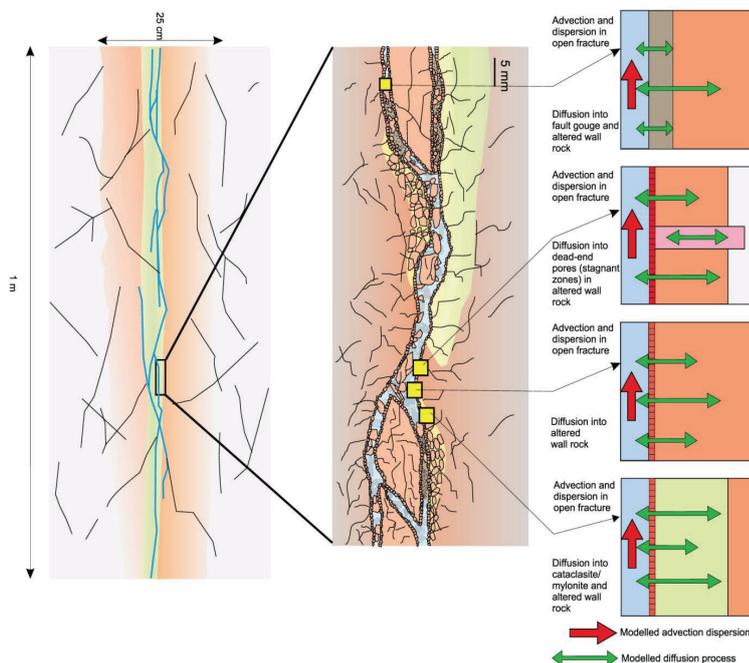
Horka (CZ)

Fissure



Open fracture (Bukov URL) FUTURE WP

3. Transport of RN in the crystalline host rock

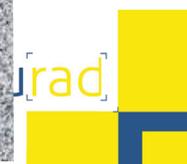
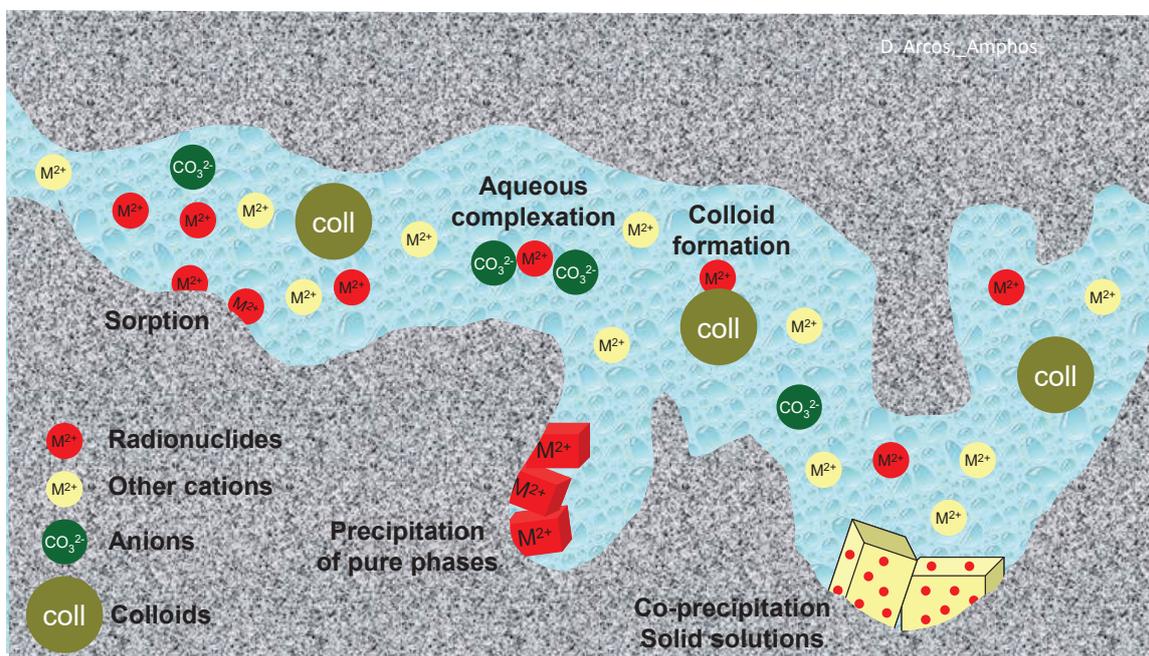


FRACTURE ⇒ (ADVECTION)

• **ROCK MATRIX** ⇒ **DIFUSION**

Advection – diffusion processes at the fractures (Poteri et al., 2002).

3. RN and colloids in the fracture



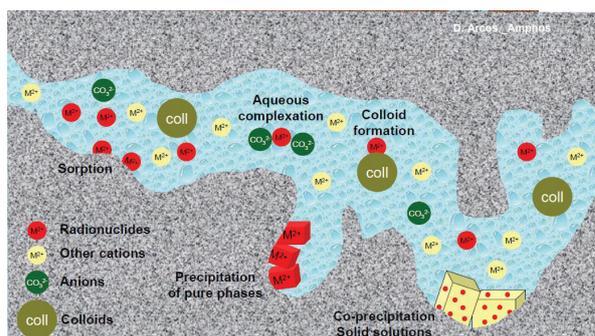
3. Factors influencing radionuclide migration in granitic rock environment

Tracer properties

- Atomic/molecule size
- Charge
- Chemical properties
- Redox behaviour
- Precipitation
- Complex formation
- Solubility

Rock properties

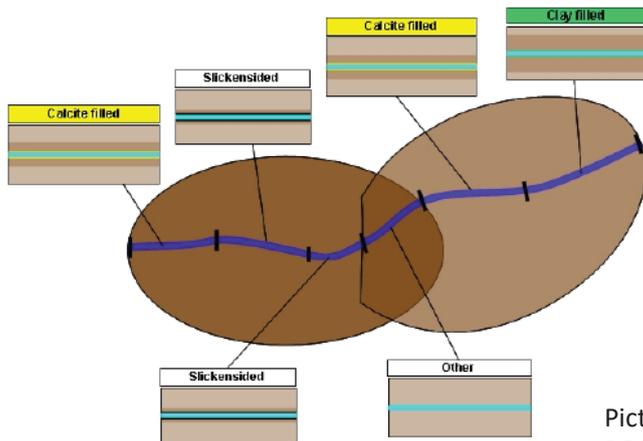
- Rock density
- Fracture density and length
- Transmissivity, strength distribution
- Porosity
- Mineralogical composition
- Hydraulic conductivity
- Hydrochemical parameters



3.1 Transport processes: Advection (Darcy's law)

Velocity of water movement in the direction of lower hydraulic gradient

Crystalline rocks: flow in fractures



$$v_D = -K \cdot \frac{\partial H}{\partial x}$$

v_D Darcy's velocity (m/s)

K hydraulic conductivity (m/s)

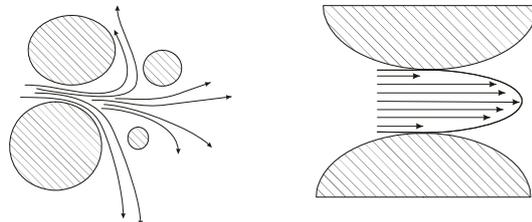
$\frac{\partial H}{\partial x}$ hydraulic gradient

Picture source: POSIVA



3.1 Transport processes: Advection

- Crystalline rocks: bulk groundwater movement in fractures
- Non-sorbing radionuclides: follow the velocity of the groundwater flow can be delayed due mechanical dispersion



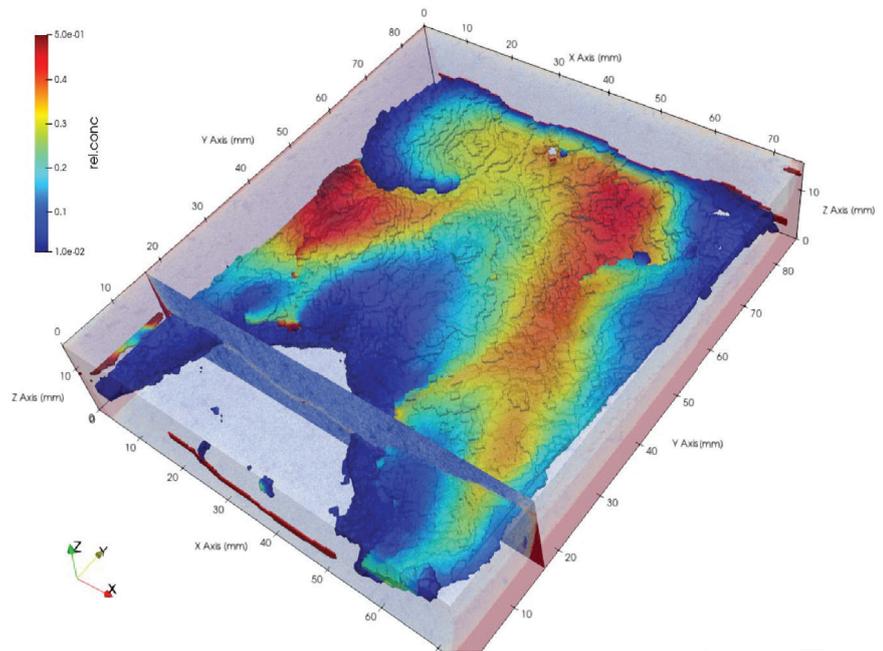
- Sorbing radionuclides: interaction with fracture walls → sorption → retention
Special case: interaction with colloids/organics in the GW → movement with flowing water
- **Important fracture property**: flow wetted surface, fracture aperture and infill



3.1 Transport processes: Advection

Flow wetted surface

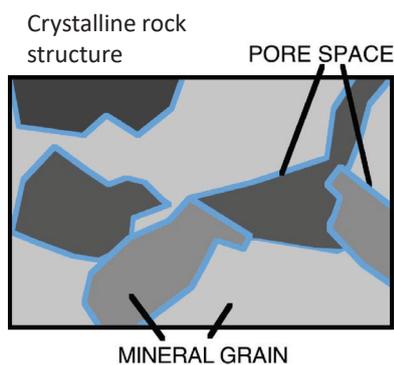
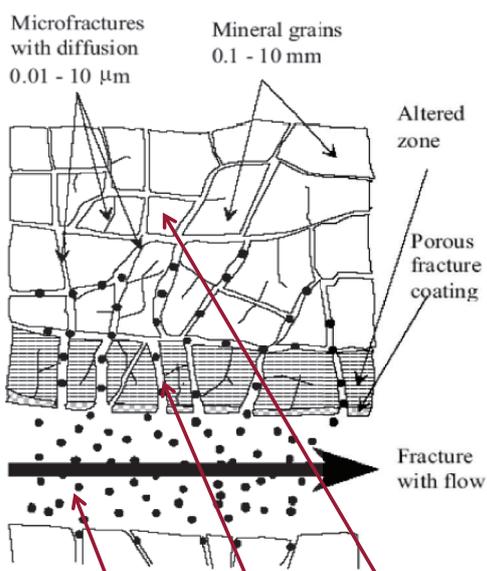
Fracture surface
Channelling



Project TH02030543, artificial fracture,;
MODFLOW modelling flow 1 ml.min⁻¹
T+1000s, KBr



3.2 Transport in rock matrix

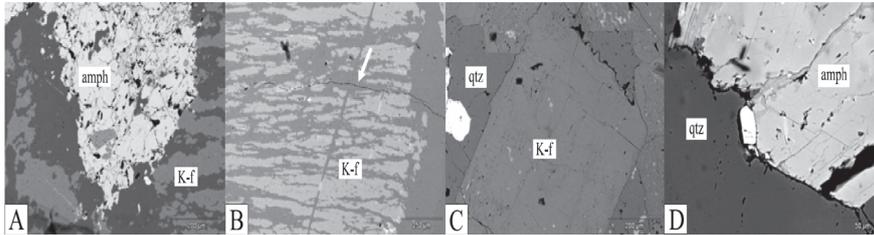


<https://wgnhs.uwex.edu/maps-data/data/rock-properties/understanding-porosity-density/>

$$\epsilon = \epsilon_f + \epsilon_t + \epsilon_r$$

- ϵ total porosity
- ϵ_t flow porosity (advection)
- ϵ_t effective porosity (diffusion)
- ϵ_r residual porosity (dead end pores)

3.2 Porosity and pore network

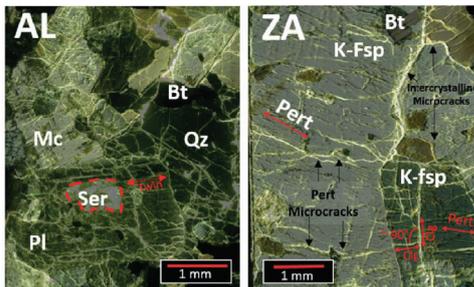


Porosity types (SEM) (Bongiolo et al., 2007): A – intragranulárn microcracks in amphibolite, B – transgranulárn mikrocracks in feldspar, C – continuous systém of intergranular fissures, D – intragranulárn fissures at amphibole and intergranulárn microfissure at the grain boundary

Connected pore network is assumed to be infinite in crystalline rock

Crystalline rock porosity: ~ 0,5% (undisturbed rock)

Freire – Liszta et al. (2015)



3.2 Porosity

$$\varepsilon = \varepsilon_f + \varepsilon_t + \varepsilon_r$$

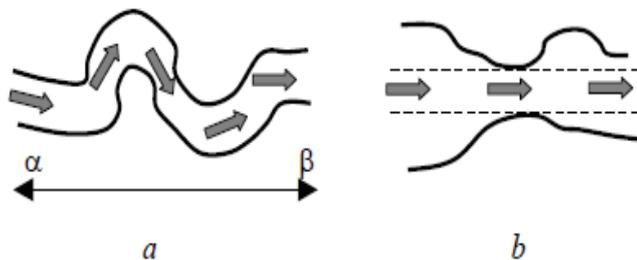
ε total porosity

ε_t flow porosity (advection)

ε_e effective porosity (diffusion)

ε_r residual porosity (dead end pores)

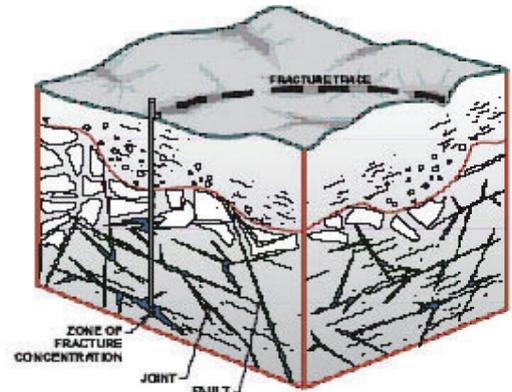
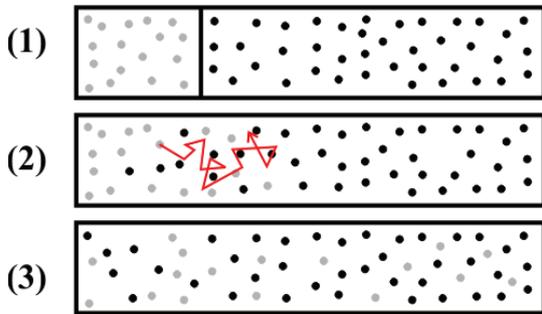
- Tortuosity (A)
- Constrictivity (B)



3.2 Diffusion process (M. Glaus presentation)

- Net transport of molecules from a region of higher concentration to one of lower concentration by random **Brownian** molecular motion (chemical gradient)

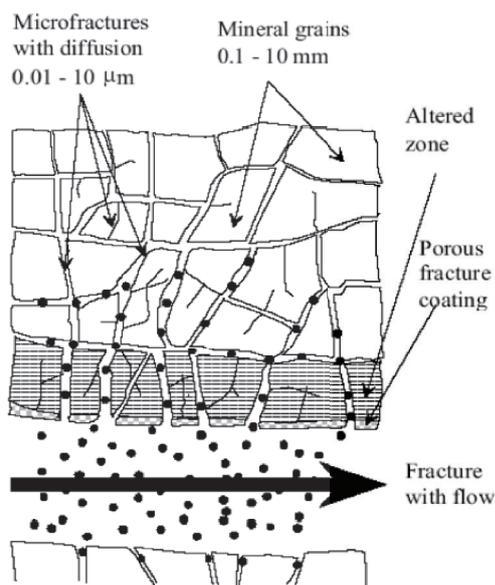
The result is a gradual mixing of molecules.



- Matrix (effective) porosity: 10-100 larger than flow porosity (I. Neretnieks)
- Important property: porosity and pore connectivity, pore infill



3.2 Diffusion in fractured rock environment



A conceptual model for radionuclide transport in a fracture coupled with matrix diffusion (reproduced from Rasilainen, 1997)

- I. Fick's law:

$$F_{i,x} = -D_x \left(\frac{\partial c_i}{\partial x} \right)_{y,z}$$

$F_{i,x}$ particle flux per cross-sectional area

D_x diffusion coefficient ($m^2 \cdot s^{-1}$)

c_i concentration of i (mol)

- I. Fick's law (t, y, z):

$$\frac{\partial c_i}{\partial t} = D_x \left(\frac{\partial^2 c_i}{\partial x^2} \right)_{y,z}$$



3.2 Diffusion in fractured rock environment

I. Pore diffusion coefficient $D_p = D_w \frac{\delta_D}{\tau^2}$

II. Effective diffusion coefficient (diffusion in pores)

$$D_e = \varepsilon_t D_p = \varepsilon_t D_w \frac{\delta_D}{\tau^2} = F_f D_w \quad F_f = \frac{D_e}{D_w}$$

I. Apparent diffusion coefficient (includes sorption)

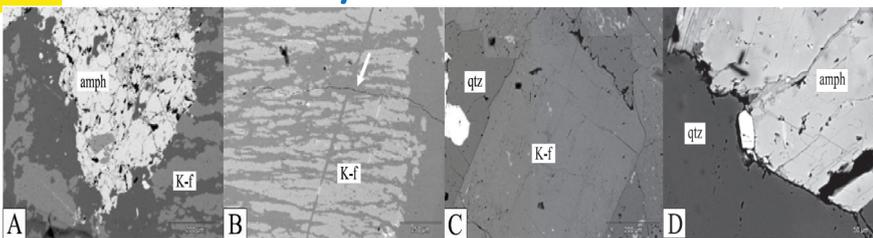
$$R = 1 + \rho \frac{(1 - \varepsilon_t) K_d}{\varepsilon_t}$$

$$D_a = \frac{D_p}{R} = \frac{D_e}{\alpha} = \frac{D_e}{\varepsilon_t + \rho K_d}$$

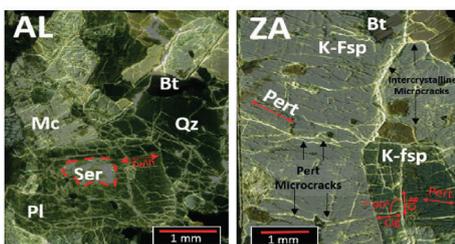
García-Gutiérrez et al. (2003)

D_w diffusion coefficient in free water
 F formation factor ($F = \varepsilon_t \delta_D / \tau^2$)
 K_d distribution coefficient (m³/kg)
 R retardation coefficient
 α rock capacity factor ($\alpha = \varepsilon_t + \rho K_d$)
 δ_D constrictivity
 ε_t flow porosity (advection)
 ε_t effective porosity (diffusion)
 ε_r residual porosity (dead end pores)
 ρ rock density
 τ tortuosity

3.2 Outerganular and interganular pores: Pathways to diffuse in the rock matrix



Porosity types (SEM) (Bongiolo et al., 2007): A – intragranular microcracks in amphibolite, B – transgranular microcracks in feldspar, C – continuous system of intergranular fissures, D – intragranular fissures at amphibole and intergranular microfissure at the grain boundary

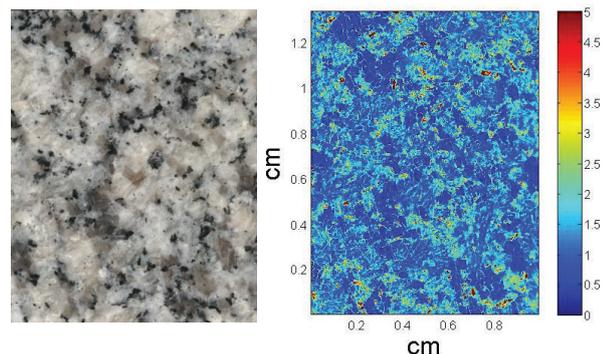


Freire – Liszta et al. (2015)



Äspö (SE)

Uni Helsinki Porosity determination in crystalline rock



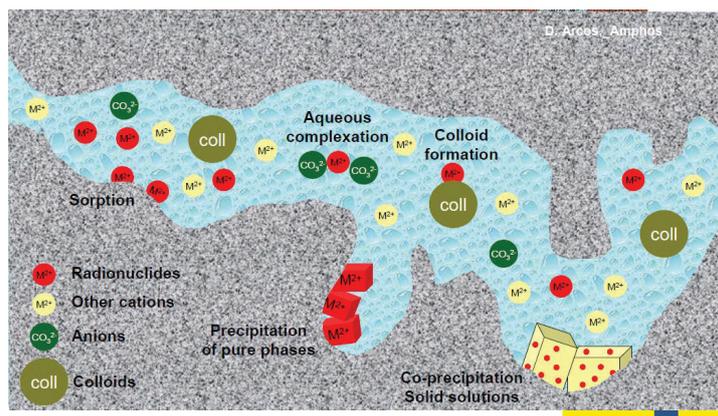
3.3 Retardation processes (F. Brandt presentation)

➤ Chemical retardation (Adsorption)

- Electrostatic adsorption (ionic exchange, surface complexation)
- Precipitation/co-precipitation
- Incorporation into mineral lattice

➤ Physical retardation

- Diffusion into dead-end pores
- Molecular filtration
- Ionic exclusion
- Reversion osmosis



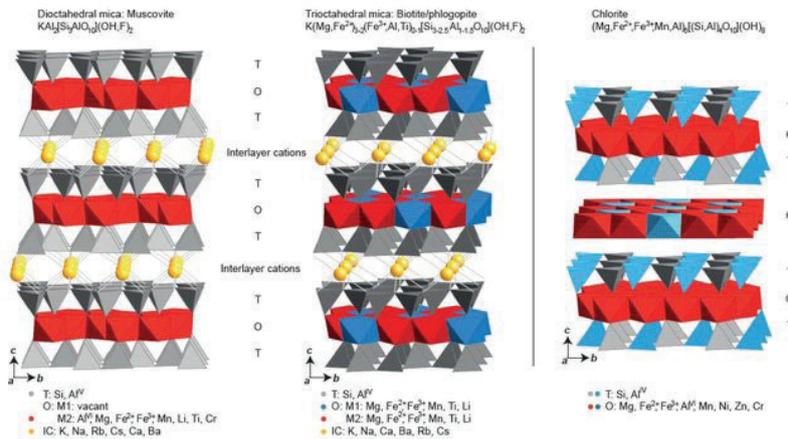
3.3 Retardation processes: Important properties

- content of rock forming minerals and secondary/fissure infill minerals with higher surface area and CEC
- porosity
- presence of organic matter and microbes (redox processes and precipitation)
- composition of groundwater
- concentration of RN in groundwater
- presence of competing ions (in groundwater)

3.3 Retardation processes – cont.

- Ionic exchange

Metal ions /Radionuclides contained in an aqueous solution are exchanged with ions contained in a solid material



Structure of muscovite, biotite and chlorite

https://www.researchgate.net/publication/260028005_Low-temperature_magnetic_anisotropy_in_micas_and_chlorite/figures?lo=1

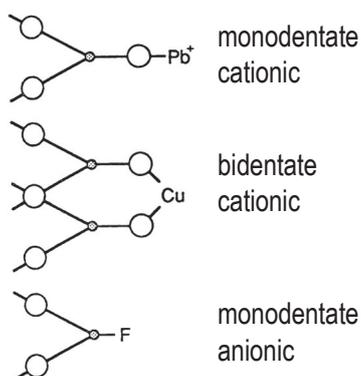


3.3 Retardation processes – cont.

- Surface complexation (specific adsorption)

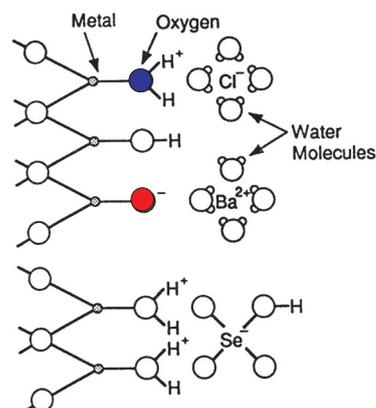
Complexes are formed between functional groups on the surface (OH⁻, CO₃²⁻, F⁻, PO₄³⁻) with species in the solution

Inner sphere complexes

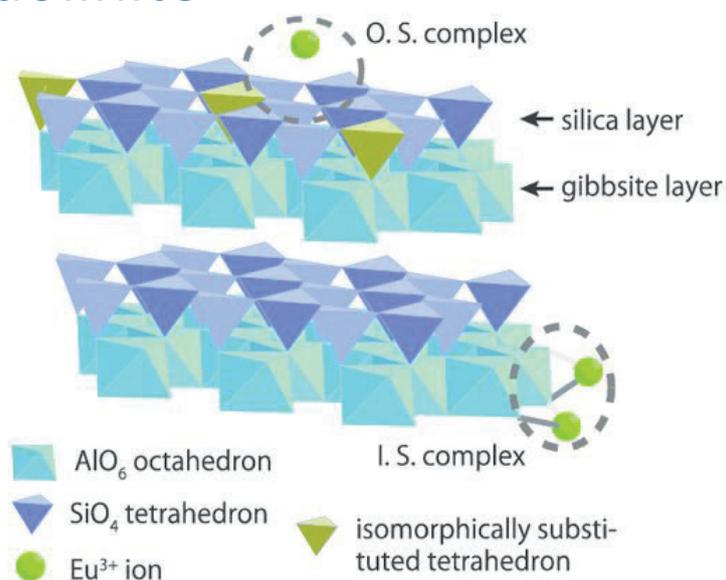


D. Arcos_Amphos21

Outer sphere complexes



Example: Surface complexation of Eu on kaolinite



i.s. – inner sphere complex
o.s. – outer sphere complex

Ishida et al. (2012):



3.3 Retardation processes – continuation

- **Precipitation/Co-precipitation**

- formation of pure RN phases
- formation of solid solutions via coprecipitation or recrystallization

- **Ionic exclusion**

Due to negative charge in mineral surfaces anions are repelled and cannot go through nanometer scale pores (I-, Cl-). Not as important as in clays (and bentonite) where large portion of the pores are in nanometer scale .

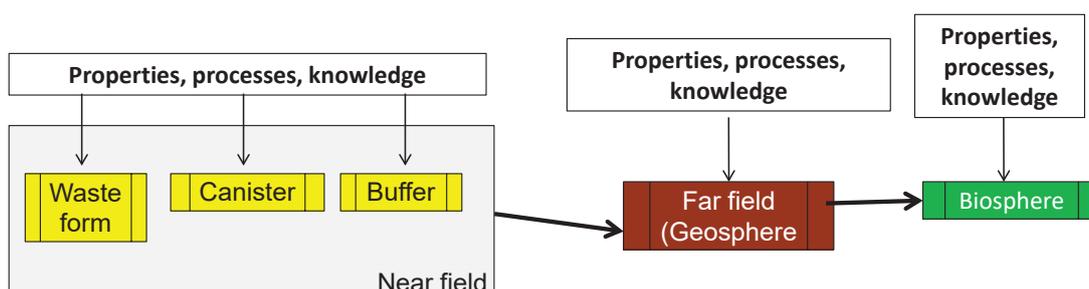
- **Molecular filtration**

Influence of pore size: Too big molecules/colloids binding radionuclides cannot migrate through too narrow pores. Tracers /radionuclides cannot even get concentrated due to this process.



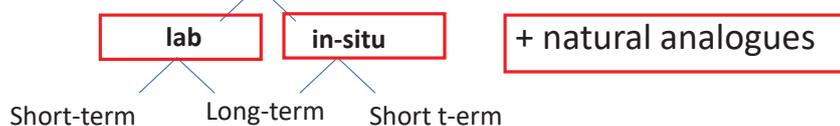
4. Transport data used in safety assessment

- **Safety assessment** is a key tool to prove that repository components fulfill their safety function
- Abstraction of real system into simplified form of conservative (less positive) model

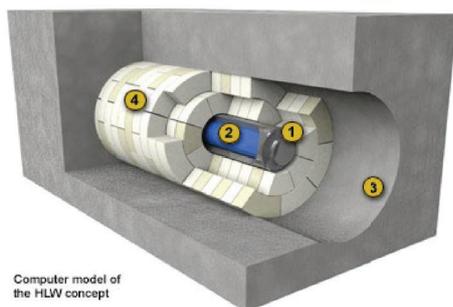


4. How to gain transport data?

Experimental activities



Example: Full-scale Engineered Barriers Experiment (FEBEX), GTS, Švýcarsko



Computer model of the HLW concept

- Long-term (GTS) experiment in 1:1 scale. Dismantling after 20 years of emplacement (1995 – 2016).

http://www.grimse.com/images/stories/videos/FEBEXDP_Grimse_Website.mp4

<http://www.grimse.com/gts-phase-v/febex/febex-i-introduction->

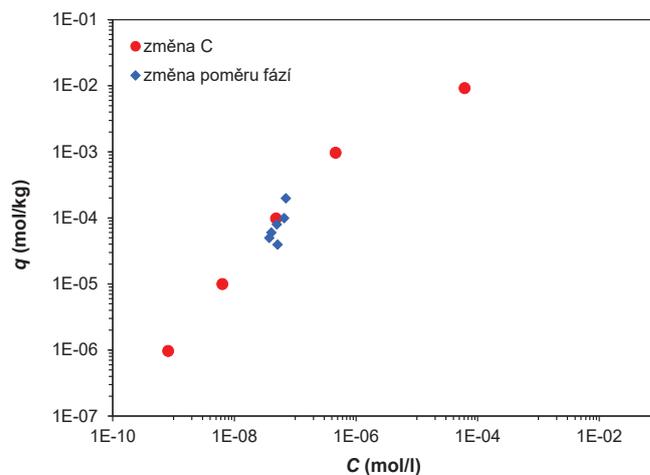
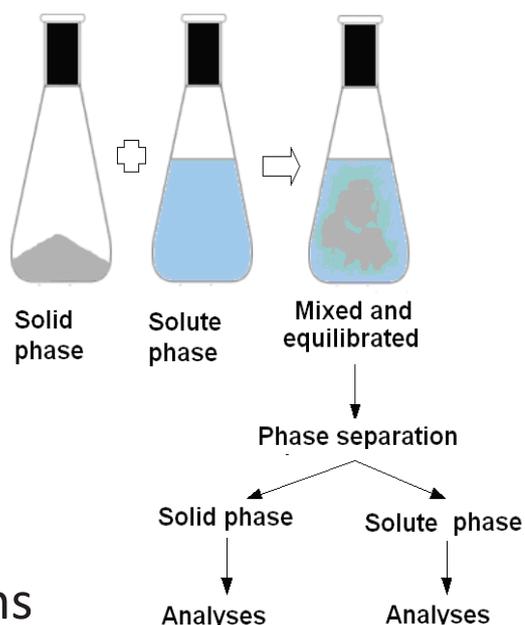
4. How to gain transport data?

- 4.1 Laboratory studies
 - 4.1a Sorption
 - 4.1b Diffusion
 - 4.1c Advection-retention
 - 4.1d Advanced transport studies
- 4.2 In situ studies
- 4.3 Natural analogues

45

4.1a Sorption data: Batch sorption experiments

Sorption: presentation by Felix Brand (FZJ) & Andreas Scheinost (HZDR)



Sorption of ^{63}Ni on migmatite (Bukov URL); Future WP

K_D ,
sorption
isotherms

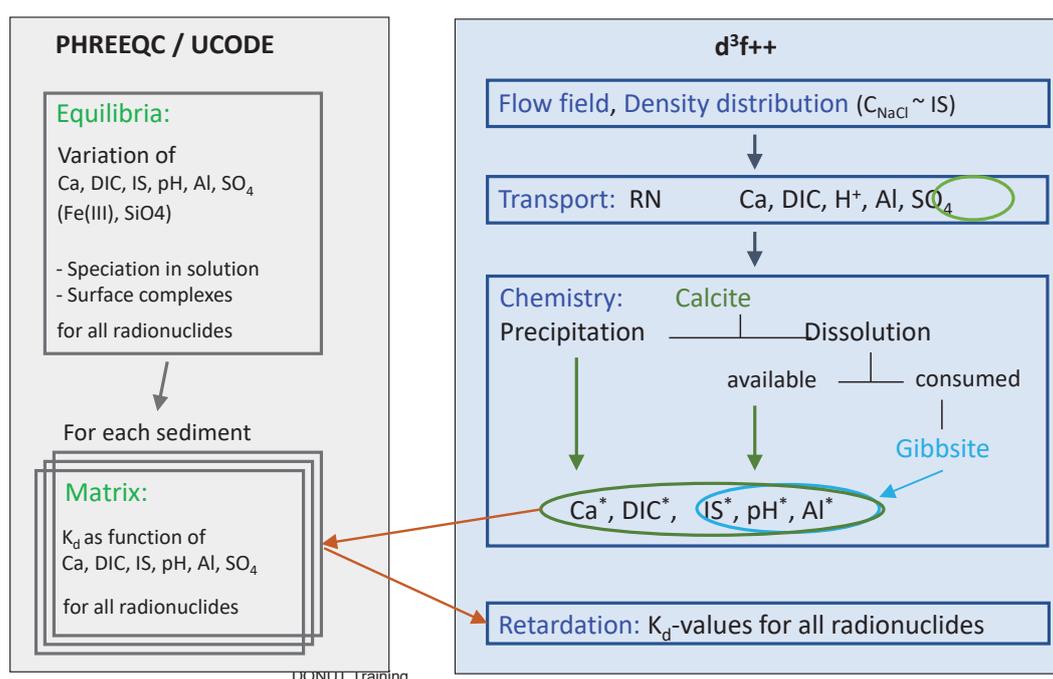
4.1a Representation of sorption in Safety assessment

- **Current safety assessment:** Reactive transport codes utilizing conservative K_d -concept with constant distribution coefficients (K_d -values)
- **But:** Changing geochemical conditions due to climatic changes (e.g. thawing permafrost, marine transgression) have a significant impact on sorption behavior



- Need to describe RN transport / sorption as a function of important geochemical / environmental parameters
- **New:** Coupling of “Smart K_d -concept” with transport program (here: d^{3f} – code, GRS)

4.1a Implementation of the smart K_d -concept



4.1a Alternative approach using the smart K_d -concept

Smart K_d -concept: One possibility to predict variations in sorption as a consequence of changing physicochemical conditions

- Based on mechanistic surface complexation models (SCM) & ion exchange (IX)

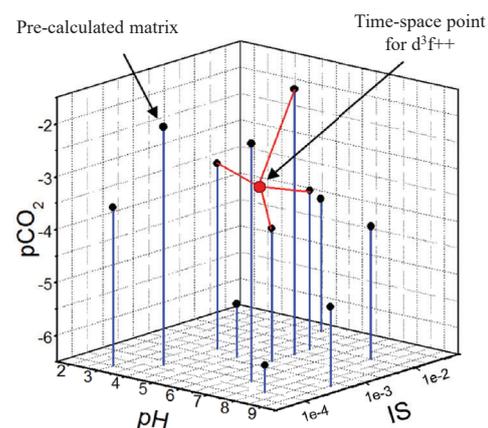
- Geochemical speciation code (PHREEQC) coupled with user interface (UCODE) and external statistical analysis (SIMLAB) to form one tool
- Strategy has numerous benefits:
 - Smart K_d -values can easily be computed for large numbers of environmental parameter combinations
 - Variable geochemistry is taken into account more realistically
 - Uncertainty Analysis and Global Sensitivity Analysis are accessible
 - Higher efficiency in computing time than in case of a direct coupling of PhreeqC with transport program



4.1a The Smart K_d -Concept



- Conceptual model for the consideration of temporally and spatially variable influencing parameters
- **A priori:** Compute multidimensional K_d -matrices for relevant sediments and relevant geochemical conditions (PHREEQC)
- **d^{3f++} :** Determine nearest neighbors for environmental parameters specific for each point in time and space
- Interpolation of appropriate smart K_d (red point) from pre-calculated multidimensional smart K_d -matrix
- Averaged value is transferred back to d^{3f++} for transport calculations



Example for a multidimensional matrix of smart K_d -values and search for nearest neighbors.

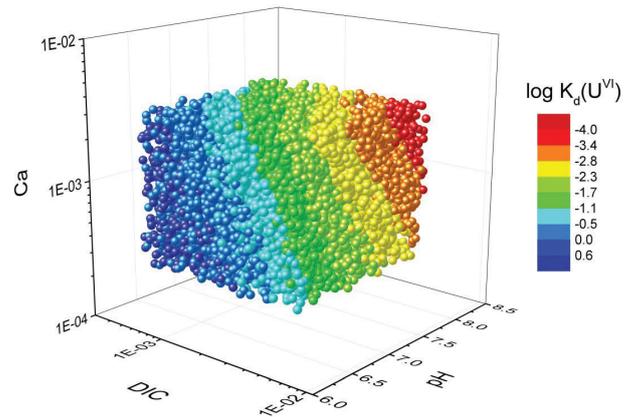


Higher efficiency in computing time than a direct coupling of PHREEQC with transport programs



4.1a The Smart K_d-Concept

- Conceptual model for the consideration of temporally and spatially variable influencing parameters
- **A priori:** Compute multidimensional K_d-matrices for relevant sediments and relevant geochemical conditions (PHREEQC)
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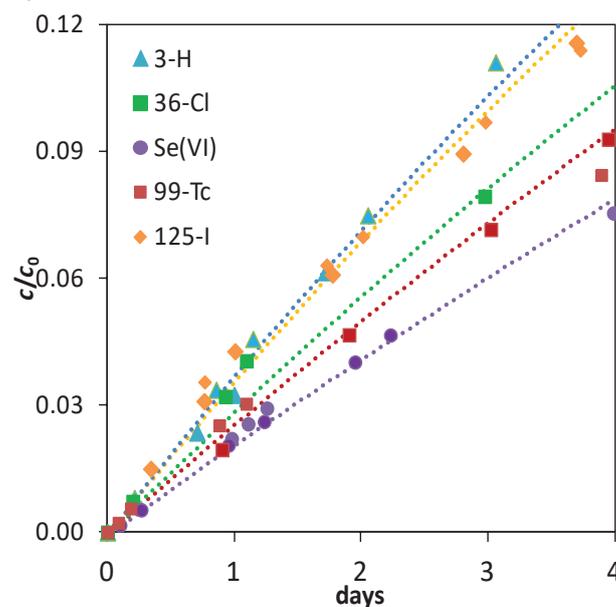
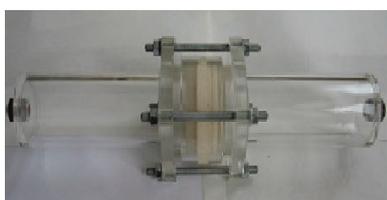
Concrete example: 3D-plot from 5D smart K_d-matrix for Uranium(VI) as a function of pH, calcium (Ca) and dissolved inorganic carbon (DIC).

➔ Higher efficiency in computing time than a direct coupling of PHREEQC with transport programs

4.1b Diffusion data: flow through (no flow) experiments

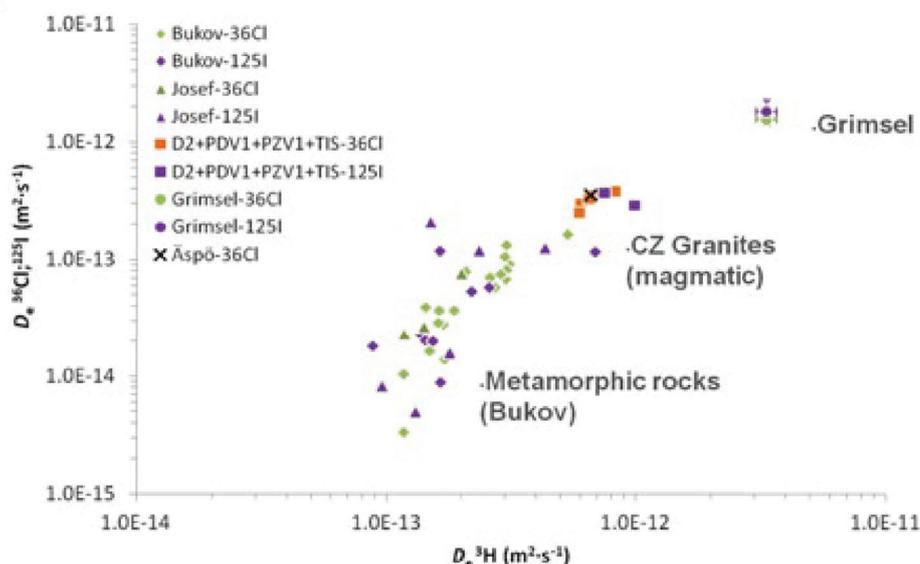


Diffusion cells for granite



Anion diffusion anions through Aare granite (LTD Phase III., project, E. Hofmanová)

4.1b Data example: Range of D_e in magmatic and metamorphic rocks



The results were gained within SURAO funded projects "Research and development support for DGR safety assessment" and „Long term diffusion Phase III.“ V. Havlova, Migration 2017, Barcelona, Spain

4.2 Diffusion porosity, effective diffusion coefficient, distribution coefficient

$$\varepsilon_d \quad D_e \quad K_d$$

- Porosity of granitic rock is less than **1 %**; altered & weathered zones adjacent to water conducting fractures are about **5-10 %**
- Diffusivities of ions and molecules in water are about **$0.5-2 \times 10^{-9} \text{m}^2/\text{s}$**
- Ions and molecules' diffusivities in granitic rock are about **$10^{-14} - 10^{-12} \text{m}^2/\text{s}$**
- Non sorbing radionuclide penetrates about **5 cm** in a year in granitic rock
- Strongly sorbing radionuclide penetrates less than **0.5 cm** in a year in granitic rock
- Distribution coefficient of I-129 is **$0.001 \text{ kg}/\text{m}^3$** and that of Cs-134 is **$0.5 \text{ kg}/\text{m}^3$**



4. 1b C-14-PMMA technique for heterogeneous pore structure characterization

$$\epsilon_d$$

Technique is based on intruding C-14-labelled methyl methacrylate (MMA) into a centimetre scale rock core.

The distribution of this radioactive tracer is then characterized using autoradiography. Quantitative porosity measurements are achieved with digital image analysis.

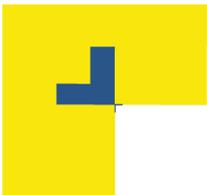
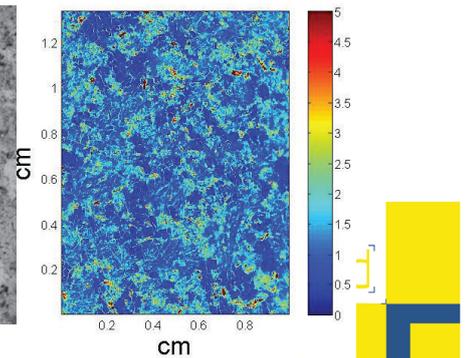
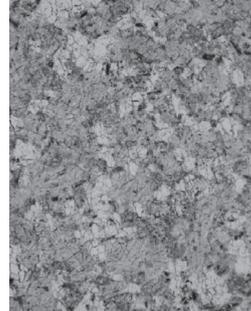
Impregnation



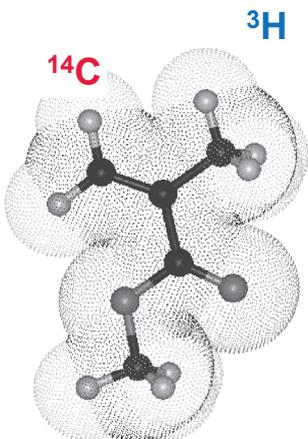
Polymerisation



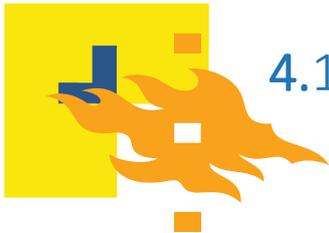
Autoradiography and Digital Image analysis



4.1b Steps in the method



- Drying of rock samples
- Impregnation of centimetric scale rock cores with C-14 labelled METHYLMETHACRYLATE (low viscosity) in vacuum
- Polymerisation of MMA into PMMA
- Autoradiography of sawn and polished rock surfaces
- Digital image processing of autoradiographs
- **Final result : Multiscale and Quantitative mapping of rock porosity!**



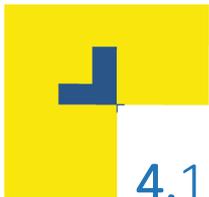
4.1b Autoradiography



Photograph of Grimsel granite sample



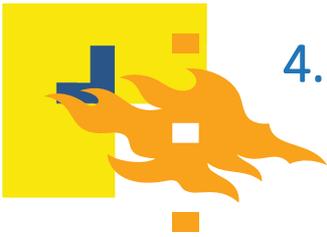
C-14-PMMA autoradiograph



4.1b Autoradiography ↔ Porosity

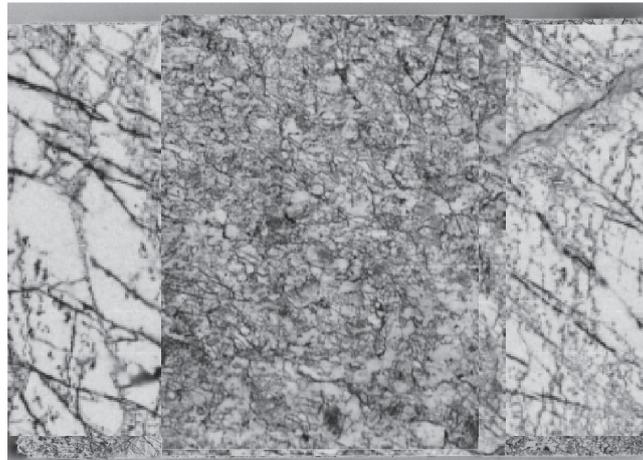
- Digitised grey levels of the autoradiograph are treated as intensities
- Conversion of grey levels to optical densities
- Conversion of optical densities to activities according to calibration sources and a correction factor
- **Initial tracer activity is considered to be diluted by the minerals at the scale of the pixel**





4.1b Autoradiography

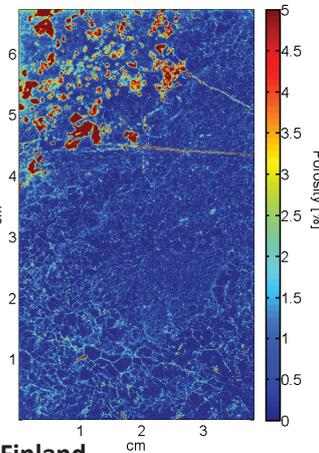
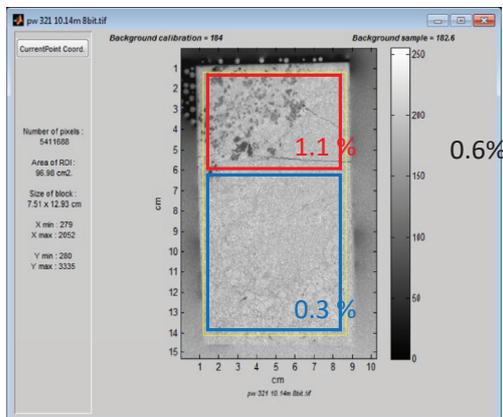
10 cm



Darker the shade – higher the porosity!

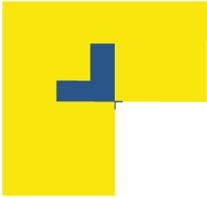


4.1b The amount of C-14-PMMA in the rock sample is converted to a porosity map



An example of metatextitic gneiss sample from Onkalo, Olkiluoto, Finland.





4.1c Advection-interaction: Column experiments

- Transport of radionuclides through crushed crystalline rock in presence of bentonite colloids in dynamic column arrangement
- Transport of radionuclides through crushed crystalline rock

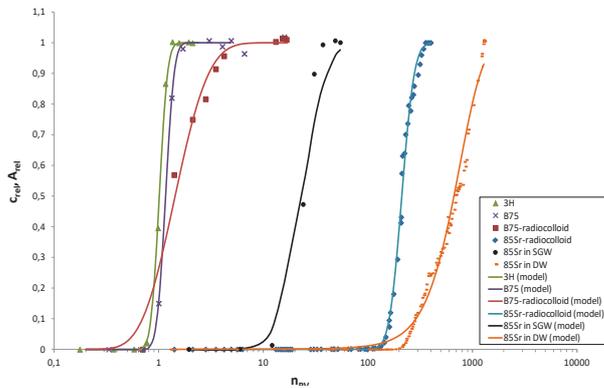
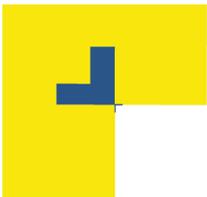


Fig. 1. The results breakthrough curves of different tracers through granite

Performed within FP EÜ project BELBAR



Fig. 2. Instrumental set-up of column experiments



4.1d Tomographic and surface-sensitive methods for transport analysis

Combination of methods

- (1) X-ray microtomography (μ CT)
Analysis of complex bundles of fracture geometries
- (2) Vertical Scanning Interferometry (VSI)
Analysis of surface topography with high spatial resolution
- (3) Positron Emission Tomography (PET)
Analysis of the flow field in fractures, validation of numerical flow field prediction

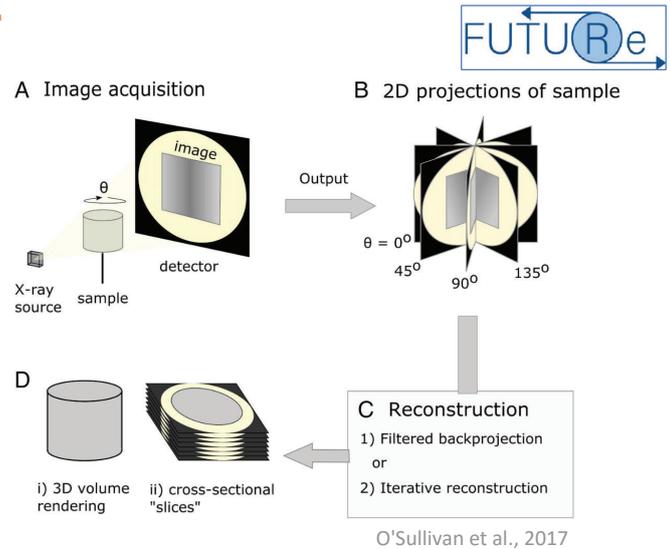
Motivation for this combination of methods

- Providing sufficient information for transport model geometry
 - Simplification vs. oversimplification: Quality of the transport model results
 - Validation of numerical results



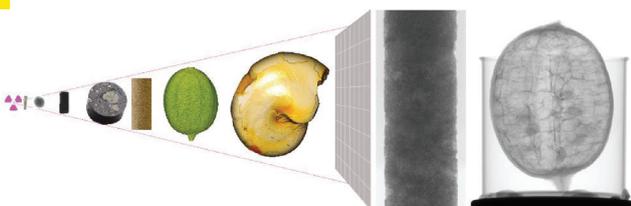
4.1d X-ray tomography: μ CT

- **X-rays are generated in an X-ray source**
 - are transmitted through the sample
 - are recorded by the X-ray detector as a 2D projection image
- **Sample is then rotated a fraction of a degree on the rotational stage**
 - Again: another X-ray projection image is taken
 - This step is repeated through a 180-degree turn (or sometimes 360 degrees, depending on sample type)
- **Spatially-resolved detection of the degree of attenuation of the incident X-rays**
 - Chemical composition
 - Material density (porosity, path length)
- **3D image of the absorptive power of the sample is computed from the 2D projections, so-called "reconstruction" process**
 - X-ray absorption is encoded as grey values in a stack of grey-scale images, Stack of (x, y) data, thickness: z

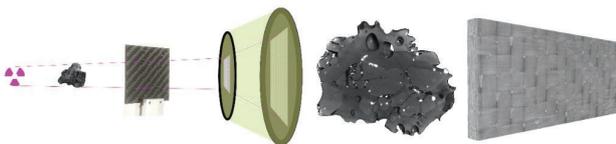


- Each dataset consists of voxels (volumetric pixels) containing volumetric information
 - Statistical analysis
 - Image analysis
 - Geometrical analysis, segmentation
- Slices can be analyzed, further processed into 3D models, made into movies, printed into 3D physical objects, ...

4.1d μ CT Spatial resolution vs. field-of-view (FOV)

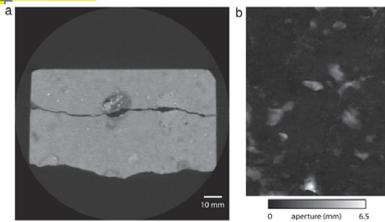


<https://www.ozeankruste.uni-bremen.de/index.php/de/labore/micro-ct-de/142-micro-ct-de.html>



- **Position of the sample within the beam cone**
- **Small sample, close to the X-ray source vs. large sample close to the detector**
 - Small sample (1 - 2 mm): voxel size of about 1 μ m
 - Large samples: voxel size of 20 - 40 μ m
- **Multi-focus setup (similar to synchrotron-based CT):**
 - magnification is achieved by a two-stage technique (\rightarrow green-colored cone)
 - beam path through the sample with very little divergence
 - large working distances, multiple objectives for magnification

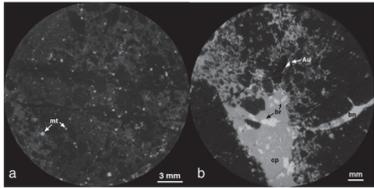
4.1d μ CT data reduction and interpretation



Ketcham et al., 2010

Fracture in a welded tuff sample

- (a) example slice, pumice clast at fracture surface
- (b) fracture aperture map

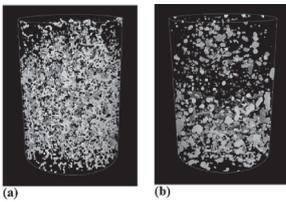


Kyle et al., 2008

Ore analysis

Main Grasberg intrusion breccia
magnetite (mt), chalcopyrite
(cp), bornite (bn) and gold (Au)

- Differentiating between metallic mineral grains with relatively small differences in density, e.g., bornite (5.1 g/cm^3) from chalcopyrite (4.2 g/cm^3), is relatively straightforward for isolated monomineralic grains or composites in a similar lower-density matrix
- Difficulties: typical intergrown ore minerals



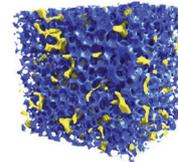
Singh et al., 2011

Fluid flow analysis (very limited opportunities!)

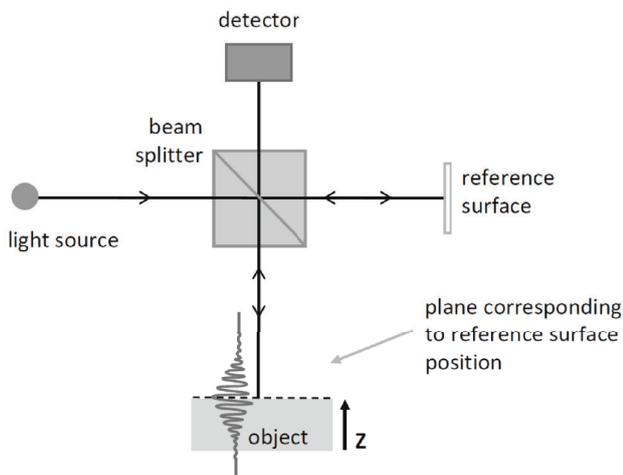
- Residual non-aqueous phase liquid (NAPL; organic) (yellow)
- water (blue) in bead pack (transparent)

d = 16.4 mm, h = 22.48 mm

- a) residual saturation
- b) after freeze-thaw cycle 10



4.1d Physical principle of interferometry: Height difference(s) derived from optical path difference analysis



Calculation of the envelope curve and envelope peak of each pixel:

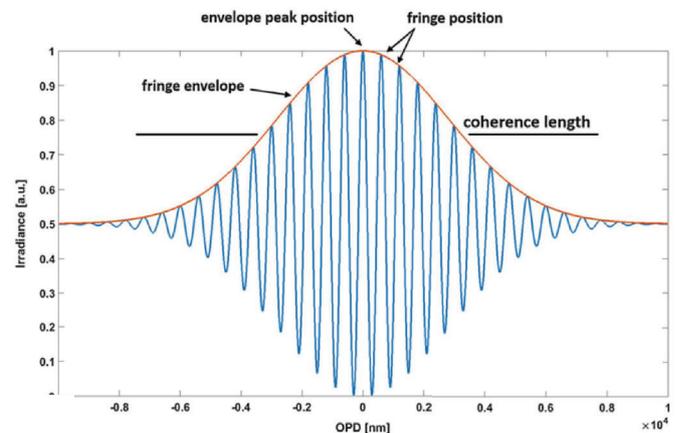
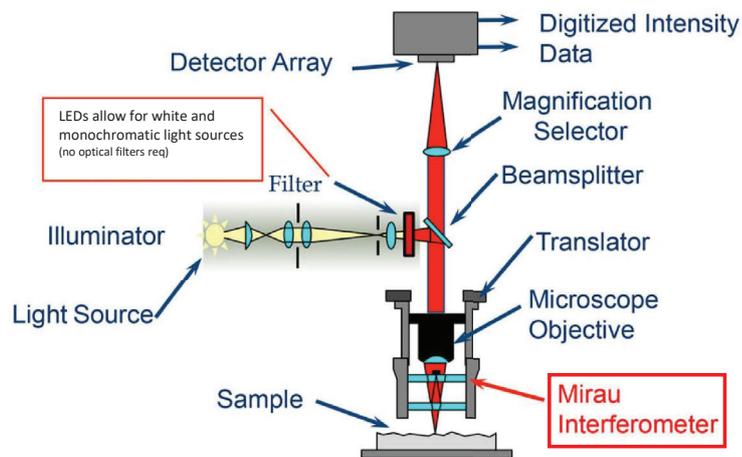


Fig. 5 Localization of interference fringes within the coherence length of white light

4.1d Simplified scheme of a vertical scanning interferometer (VSI)



Wyant (2013) Appl Optics



4.1d Sequences of surface topography analysis: → Rate map calculations recipe

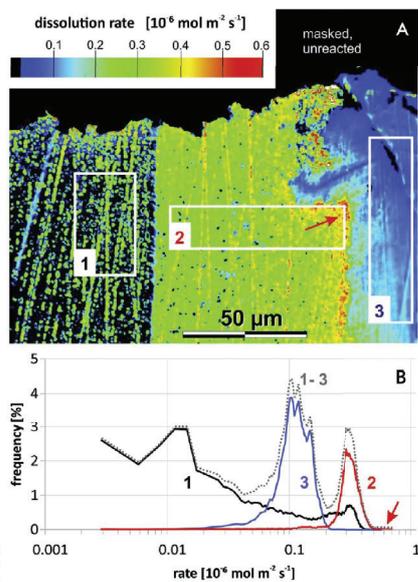


Fig. 2. Dissolution rate heterogeneity of polycrystalline calcite analyzed using (A) rate map visualization based on interferometry data and (B) rate spectra, based on three subsections of (A), each area is 1600 µm². The arrow indicates the highest reactivity at the grain boundary in-between grains 2 and 3.

Fischer & Lüttge (2017), EPSL

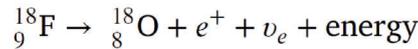
- You can analyze the topography and use it together with μ CT data
- Alternatively, you can proceed with the analysis of surface maps → which are a sequence of maps of a changing surface
 - Inert surface section: **height reference**
 - Calculation of **height difference** maps
 - Height difference (dz) per reaction time (dt) of each (x, y) map point contains information about the **height retreat velocity** (dz/dt)
 - material flux map is used to calculate the **rate map** by dividing each (dz/dt) value by the molar volume V_m
 - **Comparisons to mean reaction rates** are calculated by using the mean height retreat of all $dz(x, y)$ values



4.1d Positron Emission Tomography (PET)

• Radioactive decay of **positron-emitting radionuclides**: Positrons (e^+) and neutrinos (ν_e) are emitted

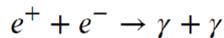
• Example: ^{18}F radionuclide



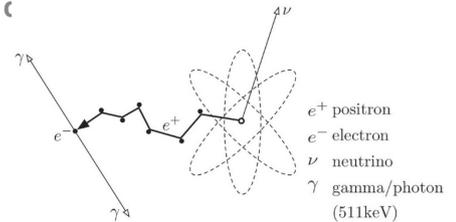
• Positrons travel through the surrounding material, loose energy, slow \dot{c}

• NOW: e^+ can combine with an electron (e^-)

• Annihilation event:



• Positron's **travel distance** $\sim 1 \text{ mm}$, this defines the spatial resolution



• Positron annihilation produces **two γ photons**, emitted in **opposite directions**

each with an energy of 511 keV, cf. special Theory of Relativity ($E = mc^2$), with $m = e^-$ rest mass



4.1d PET imaging background

• Detection of pairs of coincident photons

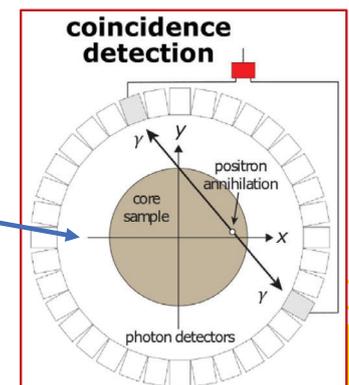
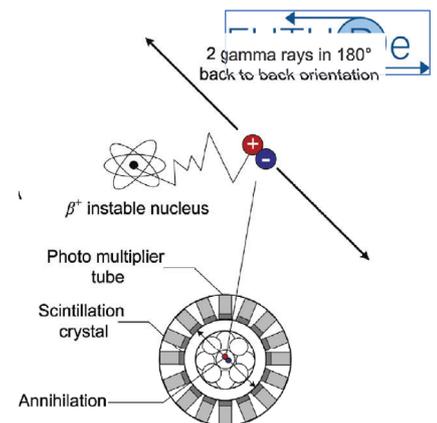
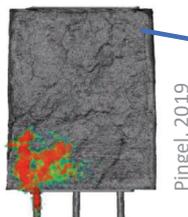
- Three-dimensional array of γ -photon detectors (= PET scanner)
- Individual inorganic scintillator crystals, emit visible light after being struck by a high-energy photon

• Entire three-dimensional volume of the sample is imaged simultaneously

• 'coincidence event': time between detection of two photons at two different detectors is less than defined time threshold (nanoseconds)

- line connecting the detectors: line-of-response (LOR)
- detectors have a finite surface area: volume-of-response (VOR)

Fracture flow imaging directly after the fluid is introduced into the left inlet



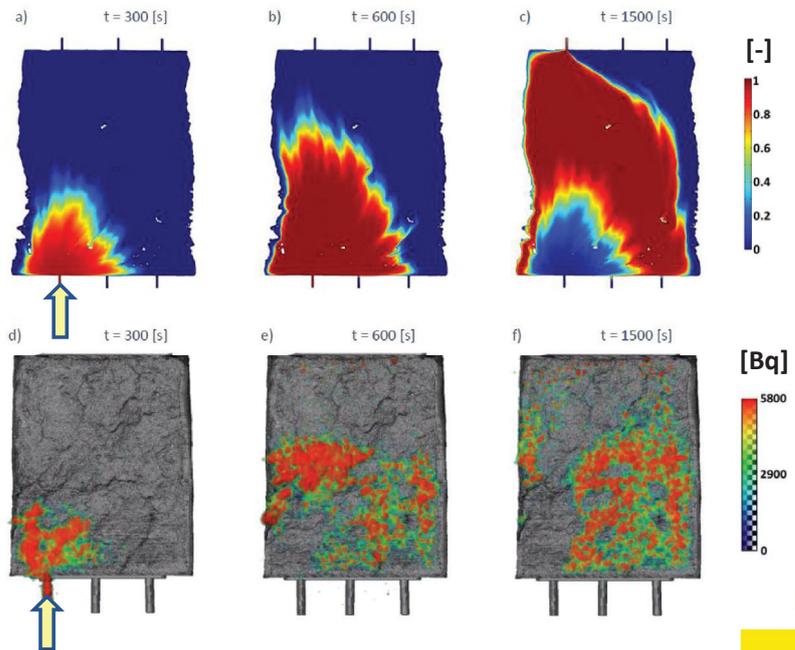
Zahasky et al., 2019

4.1d Application: Validation of simulation calculations

- (a)-(c) 2.5D **Transport model** (COMSOL) (Stoll et al., 2019)
- (d)-(f) corresponding time steps of a GeoPET experiment (Pingel, 2019)

Validation and conclusions:

- **Flow field analysis**, shape (both methods: a, d)
- Channeling (d, e)
- Increasing heterogeneity (f)



4.2. In situ studies

Focused on different aspects of DGR barrier safety functions



■ Laboratory experiments

- small scale, short term controlled conditions

■ In-situ experiments

- large scale, short term, less controlled conditions, closer to DGR

■ Natural analogues

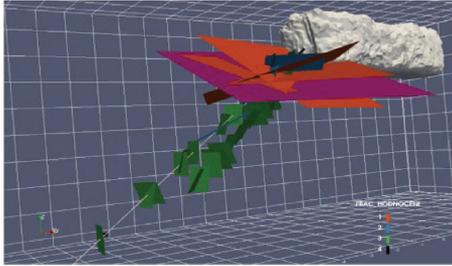
- large scale, long term, purely defined conditions



Ruprechtov natural analogue site

4.2 In-situ transport parameter determination FUTUR_e

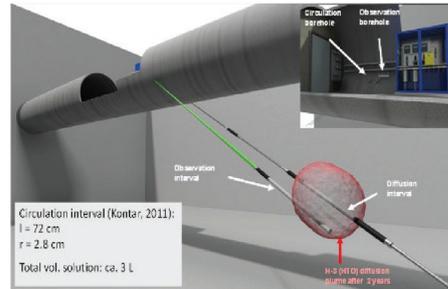
PAMIRE project:
HTO injection in the rock fractures systém (2018)
 URL Josef
 - Fracture properties, permeability



Fracture model in PAMIRE project -preliminary results Josef URL
 PAMIRE - <http://www.ujv.cz/cz/pamire>.
 TA04020986

Long term diffusion Phase III. project (2011 – 2018)

^3H , ^{22}Na , ^{133}Ba , ^{134}Cs and non-active Se(VI) ;
 Grimsel test site (GTS, CH)

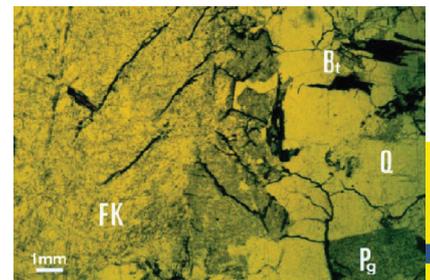


LTD Phase III. experiment concept. Figure NAGRA
 Long term diffusion Phase VI. project -
<http://www.grimsel.com/gts-phase-vi/ltd/ltd-introduction>



4.3 Natural analogues (www.naturalanalogues.com) FUTUR_e

- Identification of paleo-water pathways in crystalline rock can be done on the basis of natural processes (e.g. El Berrocal, Spain; Palmottu, Finland)
- Scale: m to X00 m
- Identification of paleo pathways due to identification of Fe oxide coating in fractures; El Berrocal, Spain; *Montoto and Mateos (2004)*.
- Migration of radionuclides through fracture (paleo-fissures in granite mapped using ^{235}U fission tracks)
 El Berrocal, Spain;
Montoto and Mateos (2004)

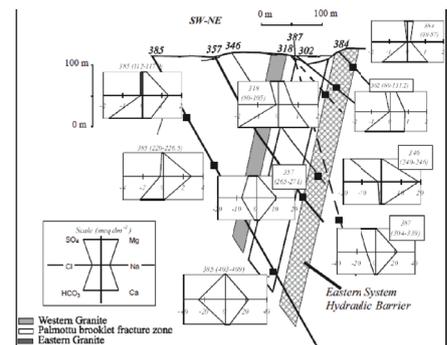
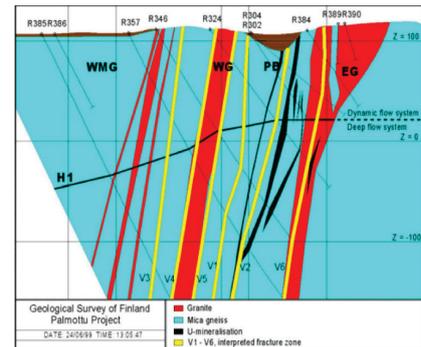


4.3 Natural analogue Palmottu

- Identification of large scale flow channels
- Hydraulic head measurements
- Tracer tests

- $^{234}\text{U}/^{238}\text{U}$ ratio
- Blind prediction modelling test etc.

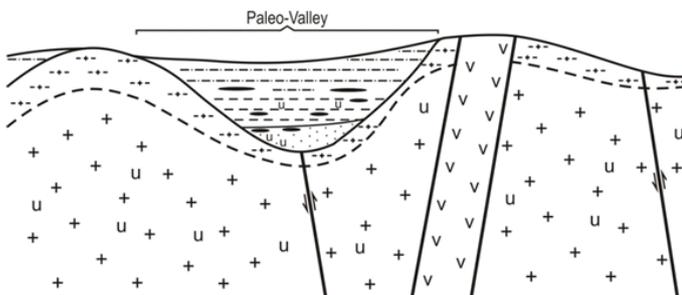
(Blomqvist et al., 2000, Rasilainen et al., 2001, Suksi et al., 2006, SKB TR-01-20)



4.3 Natural analog Ruprechtov (GRS - ÚJV – HU - GFZ - FZK INE - Krakow University)

(www.natural-analogues.com)

More than 10 year of hydrogeological monitoring and research of U migration in clays



Studies of U behaviour in the system of U – smectite clay – granite – Fe - S - microbes

<https://www.natural-analogues.com/the-natural-analogues/radionuclide-retardation-in-sediments-ruprechtov>

5. CONCLUSIONS

- crystalline rocks are important part of multibarrier systém of DGR
- their safety function has to be fulfilled in order to consider DGR safety
- evaluation of their transport properties enters safety assessment calculations
- Need to combine laboratory and in situ approaches; natural analogues are also usefull
- open questions remaining:
 - crystalline rock is heterogeneous multicomponent material (prediction of property on longer distance)
 - fissure property incorporation in SA models
 - upscaling

Thank you for you attention

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Aknowledgement

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- **EURAD EJP project** *The project leading to this application has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement n° 847593.*
- **SURAO, Czech Technology agency; EU projects BELBAR, CROCK**





UNCERTAINTY MANAGEMENT IN SAFETY ASSESSMENT AND ITS ROLE IN REPOSITORY DEVELOPMENT

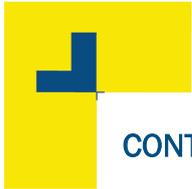
EURAD Education & Training webinar WP FUTURE
November 17, 2021 • Th. U. Kaempfer, Nagra, Switzerland



The project leading to this application has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement n° 847593.

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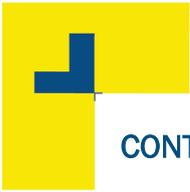
CONTENT

- **Iterative, step-wise repository development**
 - role of uncertainty management
 - safety oriented approach
- **Uncertainty management strategy**
 - types of uncertainty
 - options for dealing with uncertainty
- **Handling uncertainty in safety assessments**
 - reducing uncertainty in the assessment basis
 - assessing the impact of remaining uncertainty

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CONTENT – AND SOURCES

- **Iterative, step-wise repository development**
 - role of uncertainty management
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MATERIAL BASED ON

EURAD UMAN – THE WORK PACKAGE DEDICATED TO NETWORKING ON UNCERTAINTY MANAGEMENT

- inspired by discussions with many
- some slides by Frank Lemy, BeIV

NAGRA'S DISPOSAL PROGRAM – CURRENTLY AT THE 3RD AND LAST STAGE OF SITE SELECTION

- Nagra's approach on uncertainty management
- inspired by discussions and material by Nagra experts and contractors
- and personal experience



CONTENT – AND SOURCES



- **Nagra, Switzerland**
 - Since 2017
 - Section head safety analysis
 - Thomas.kaempfer@nagra.ch
- **AF-Consult (now AFRY)**
 - 2009-2017
 - Head groundwater protection and waste disposal
 - Projects for Andra France, BFE Germany, Nagra, etc.

- Research fellow at SLF Davos, Switzerland and ERDC CRREL, NH, USA
- PhD Computational Materials Science, EPFL Lausanne, Switzerland
- MS Mathematics, EPFL Lausanne, Switzerland

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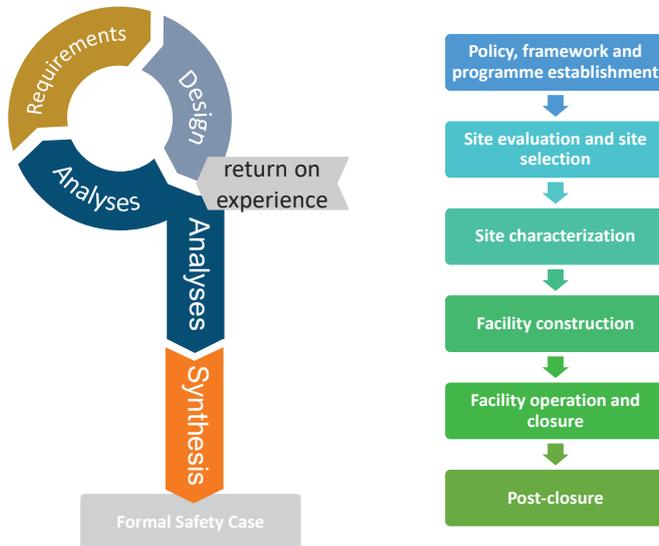
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STEP-WISE PROCESS OF IMPLEMENTING A DEEP GEOLOGICAL REPOSITORY



• Iterative process along phases

- requirements
 - design
 - analysis, including safety assessments
- For important milestones, a formal safety case is synthesized
- Uncertainty management must be adapted to the milestone & decision ahead

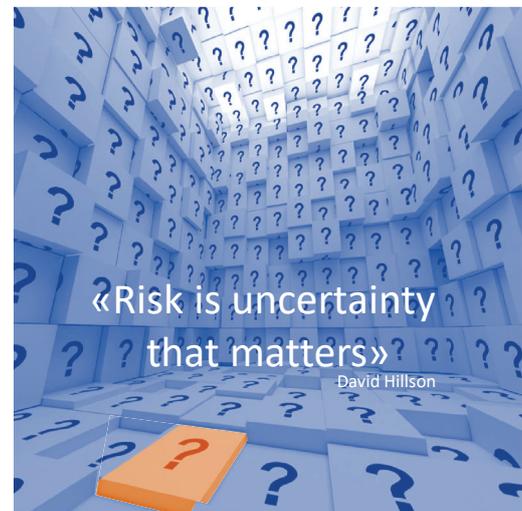
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UNCERTAINTY MANAGEMENT ADAPTED TO THE DECISION AHEAD

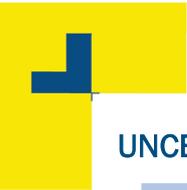
- We demonstrate that, despite remaining uncertainty
 - safety is given and
 - decisions are robust
- This requires a sound management of uncertainty
 - but it is neither required nor feasible to eliminate all uncertainty
- How to deal with an uncertainty "now"?
 - How can it be characterized now?
 - Must it (and can it in practice) be reduced, mitigated or avoided now?
 - Outlook: For future milestones, can it (and must it) be:
 - (better) characterized
 - reduced, mitigated, avoided
- Also to be assured: Robustness w.r. to «new» uncertainties



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UNCERTAINTY MANAGEMENT STRATEGY – HIGH-LEVEL PICTURE (1)

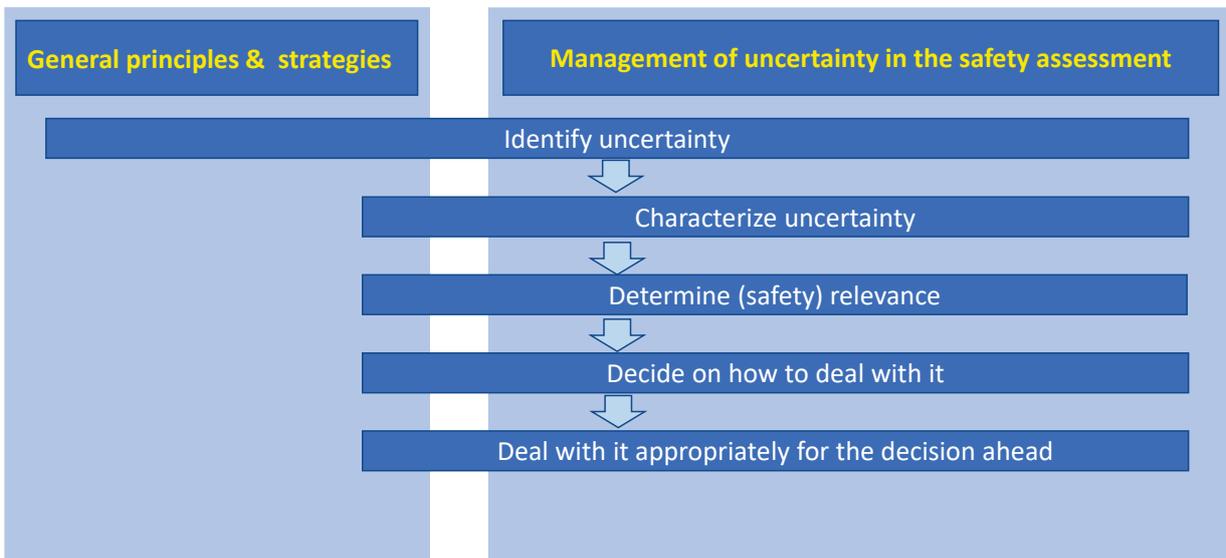


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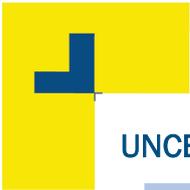
UNCERTAINTY MANAGEMENT STRATEGY – HIGH-LEVEL PICTURE (1)



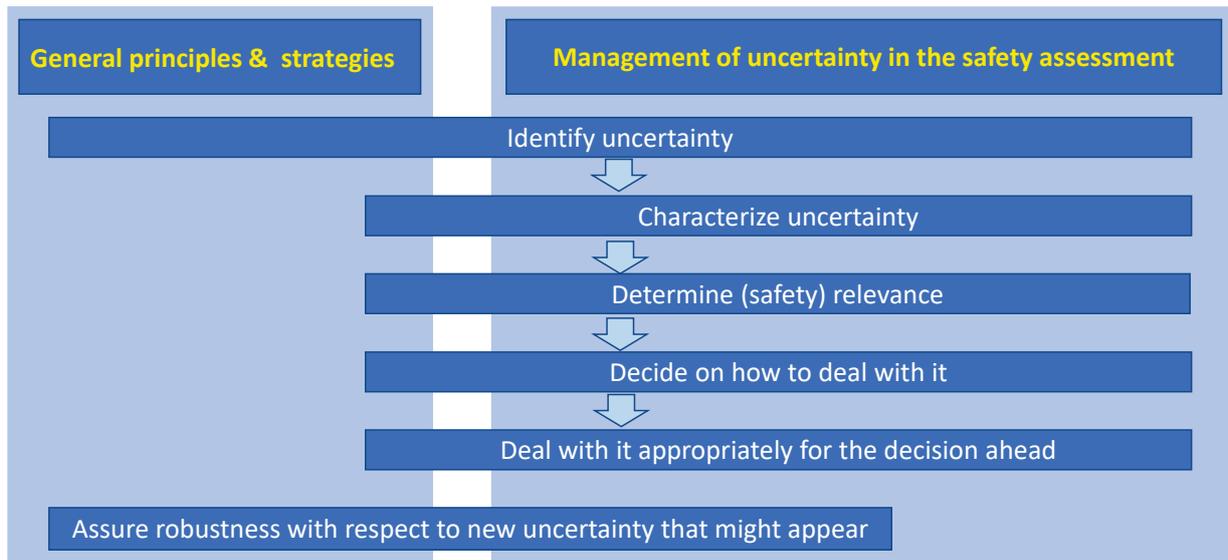
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UNCERTAINTY MANAGEMENT STRATEGY – HIGH-LEVEL PICTURE (1)

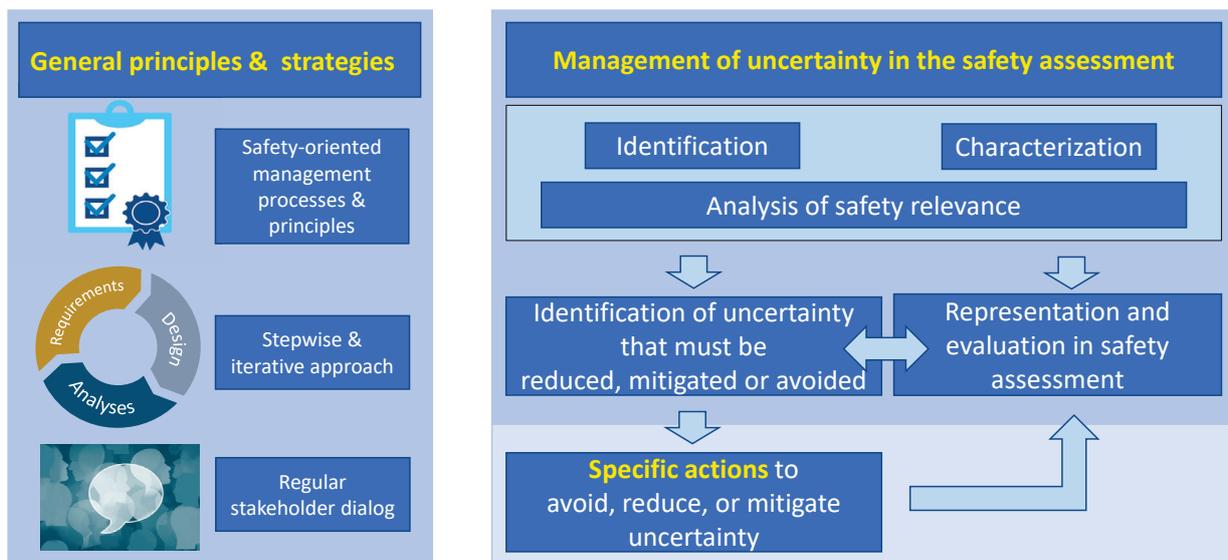


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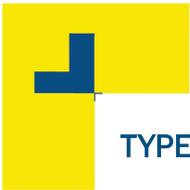
UNCERTAINTY MANAGEMENT STRATEGY – HIGH-LEVEL PICTURE (2)



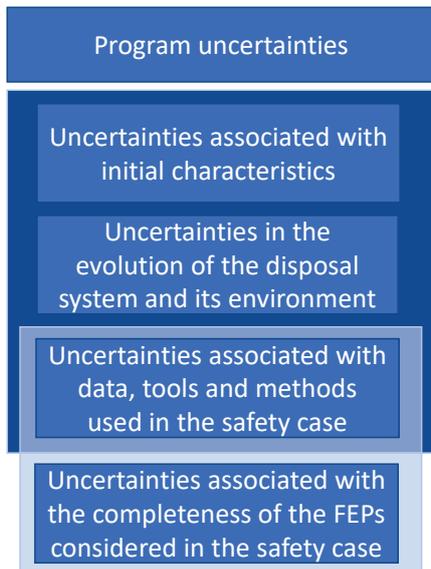
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TYPES OF UNCERTAINTY – A CLASSIFICATION SCHEME DEVELOPED IN WP UMAN



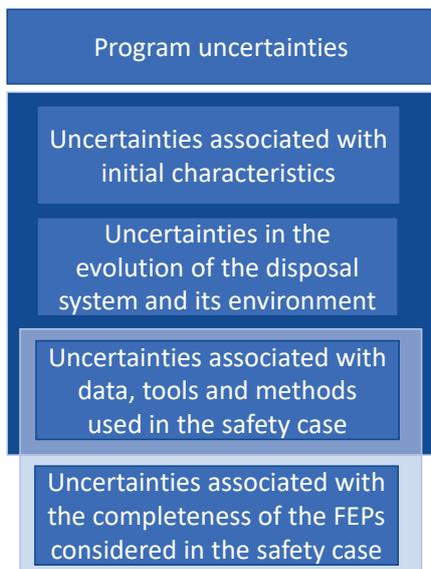
1. **associated with** the waste management **programme** and other prevailing **circumstances** (societal, resources, ...)
- associated with the disposal system**
2. **initial characteristics:** of the waste, site, and engineered components
3. **evolution:** including effects of events and processes that may affect the initial characteristics and long-term evolution (climatic, geologic,...)
- associated (mostly) with the assessment itself**
4. **data, tools and methods:** uncertainties related to methods (e.g., QA/AC), concepts (models), and parameters (data)
5. **completeness:** uncertainty arising when overlooking certain aspects relevant for safety

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PROPER MANAGEMENT OF UNCERTAINTY ASSURES SAFETY



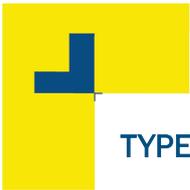
- a good (safety driven) process
- a good (safe) system
- a good (error free & complete) analysis

all needs to be demonstrated for a sound safety case

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TYPES OF UNCERTAINTY – BASED ON AVAILABILITY & USE OF KNOWLEDGE

	Information is available		Information is not available	
Level of information is used	(know knowns) 		known unknowns 	
Level of information is not used	unknown knowns 		unknown unknowns 	

Adapted from 2nd Joint Workshop IGSC/FSC, 9 October 2019, Perspectives on risk and uncertainty, A. Eckhardt

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UNCERTAINTY TABLE

5. Uncertainties associated with completeness

	Known Unknowns	Unknown/Ignored Knowns	Unknown Unknowns
1. Programme uncertainties	e.g. uncertainties in available disposal sites & stakeholder conditions,...	e.g. ignored lack of financial resources	e.g. unforeseen political instabilities
2. Uncertainties associated with initial characteristics	e.g. uncertainties in site characteristics	e.g. ignored safety-relevant site characteristics	e.g. unknown uncertainty in safety-relevant site characteristics
3. Uncertainties in the evolution of the disposal system & its environment	e.g. uncertainties in magnitude, time of occurrence or consequences of events & processes	e.g. ignored possible magnitudes of disturbing events (e.g. Fukushima)	e.g. unknown events & processes
4. Uncertainties associated with data, tools & methods	e.g. accuracy of measurements and modelling results,...	e.g. ignored mistakes in implementing safety-related activities (e.g. 2 nd WIPP incident)	e.g. unknown errors in modelling results

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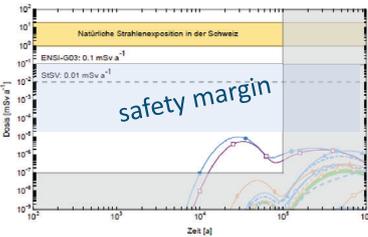




ELEMENTS OF AN UNCERTAINTY MANAGEMENT STRATEGY – MANAGING THE UNKNOWN (UN)KNOWN

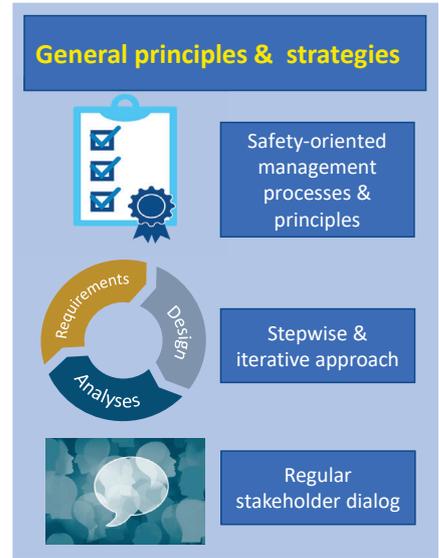


- Safety oriented processes and principles and an appropriate safety culture help to avoid «unknown/ignored knowns»
- Robustness and safety margins together with the iterative approach allow us to react to newly appearing «unknown unknowns»
- Regular stakeholder dialog enhances awareness and contributes significantly to a good process

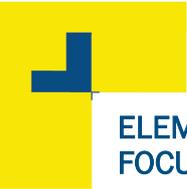


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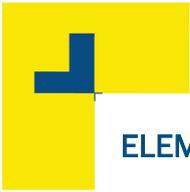
ELEMENTS OF AN UNCERTAINTY MANAGEMENT STRATEGY – FOCUSING ON KNOWN UNCERTAINTY

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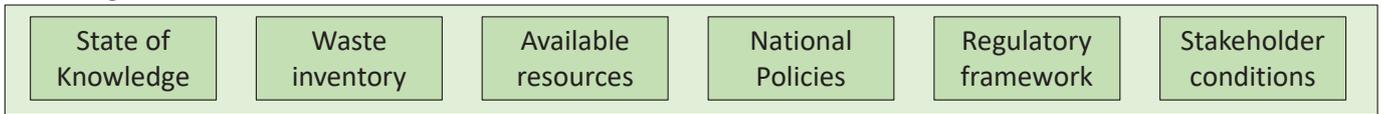
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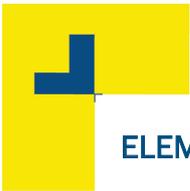
ELEMENTS OF AN UNCERTAINTY MANAGEMENT STRATEGY

Prevailing circumstances



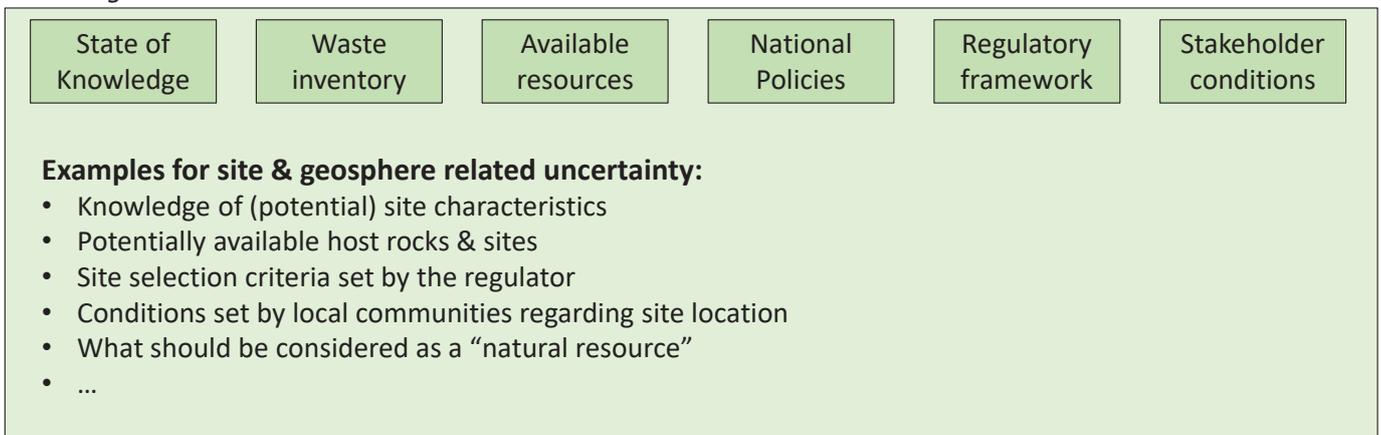
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ELEMENTS OF AN UNCERTAINTY MANAGEMENT STRATEGY

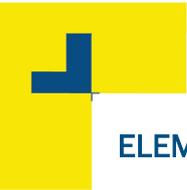
Prevailing circumstances



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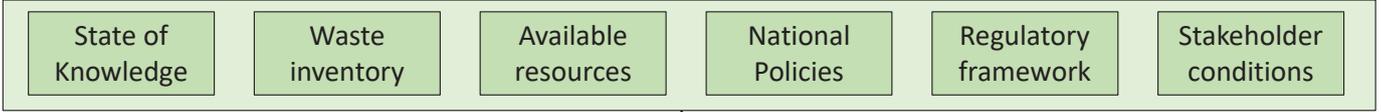
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ELEMENTS OF AN UNCERTAINTY MANAGEMENT STRATEGY

Prevailing circumstances

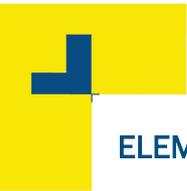
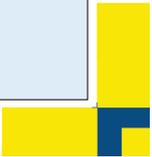


Programmatic activities



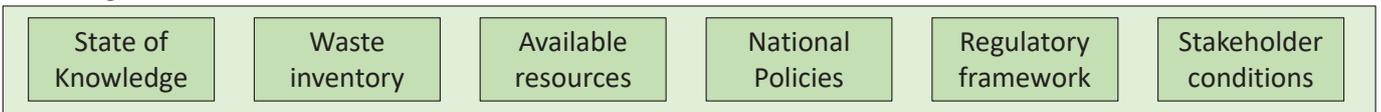
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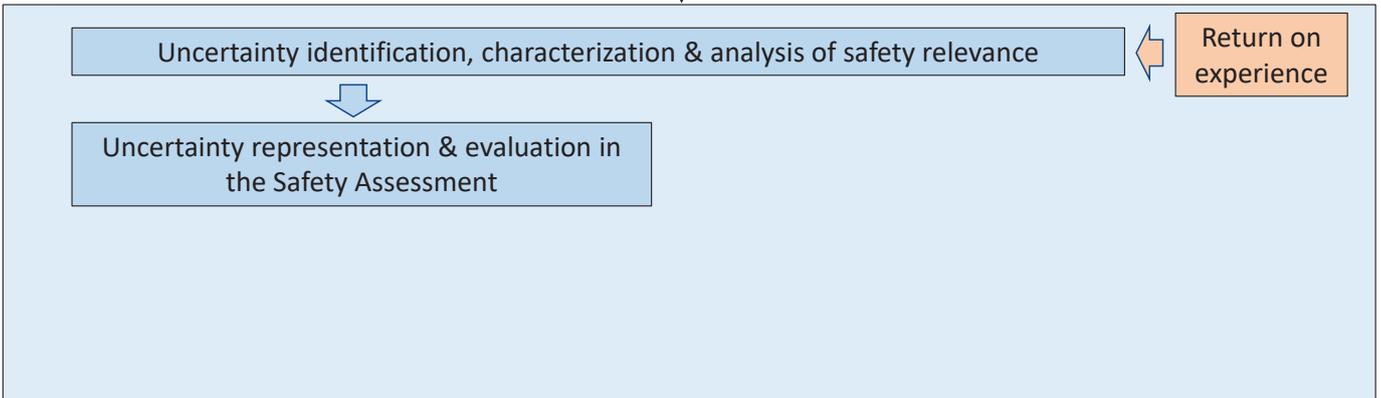


ELEMENTS OF AN UNCERTAINTY MANAGEMENT STRATEGY

Prevailing circumstances

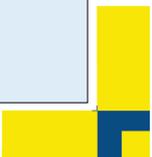


Programmatic activities



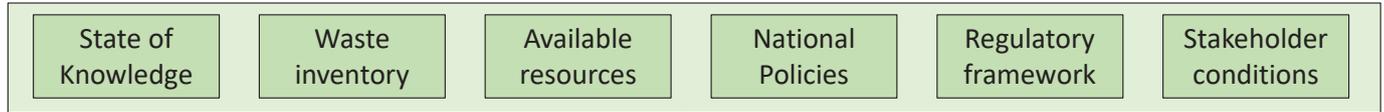
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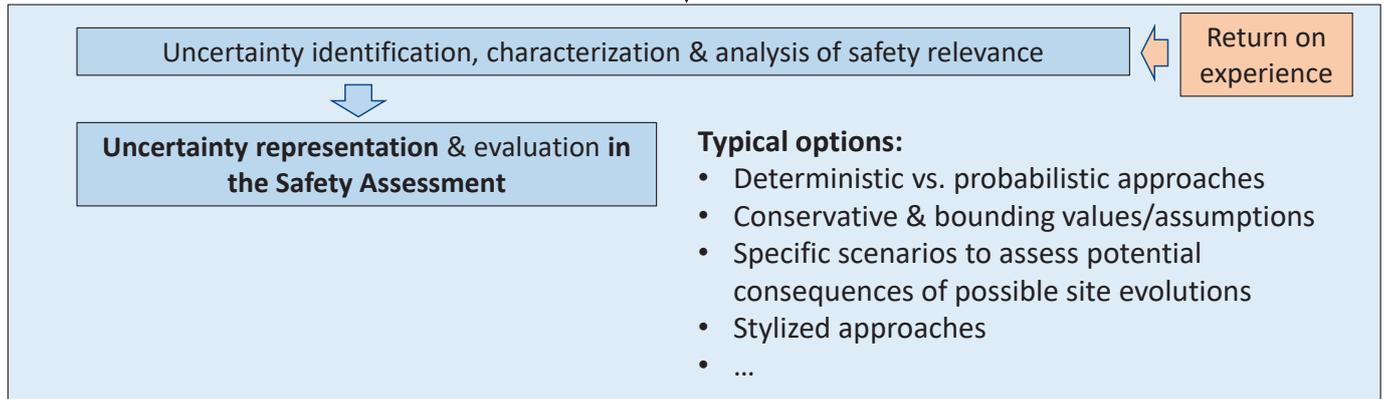


UNCERTAINTY REPRESENTATION IN THE SAFETY ASSESSMENT

Prevailing circumstances



Programmatic activities

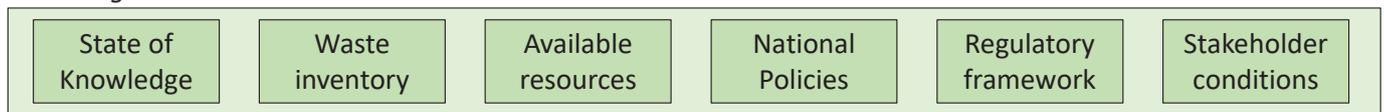


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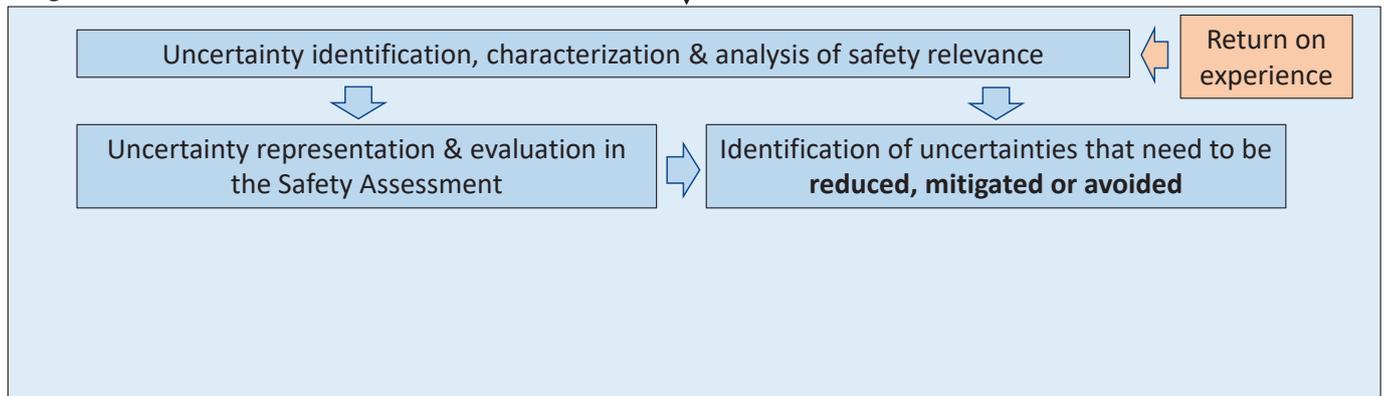
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ELEMENTS OF AN UNCERTAINTY MANAGEMENT STRATEGY

Prevailing circumstances



Programmatic activities

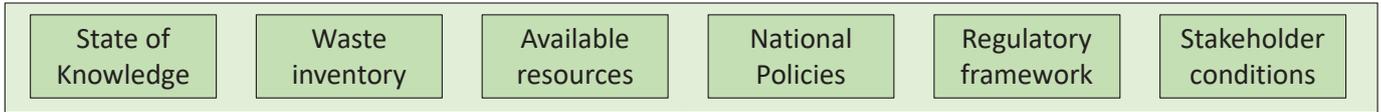


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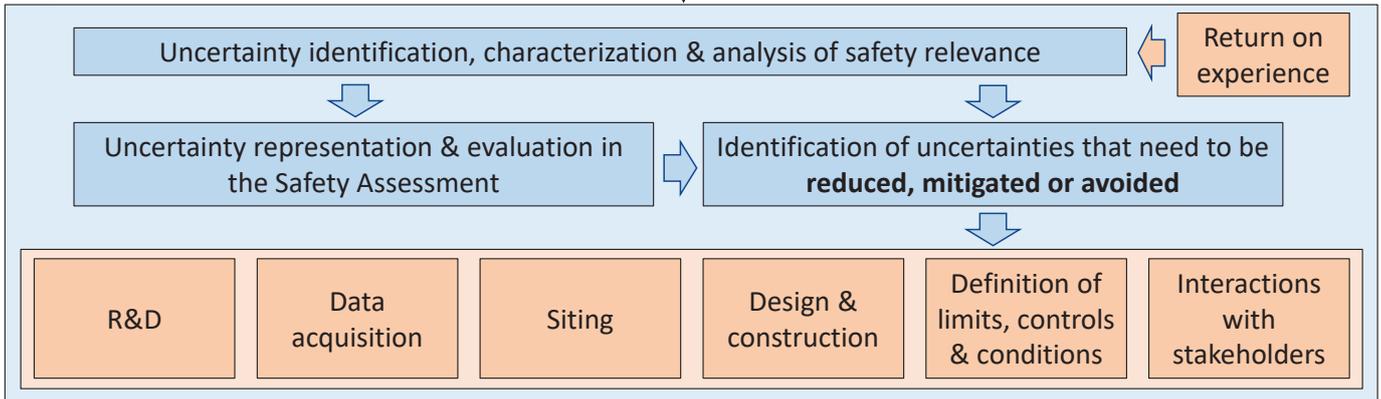
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ELEMENTS OF AN UNCERTAINTY MANAGEMENT STRATEGY

Prevailing circumstances



Programmatic activities

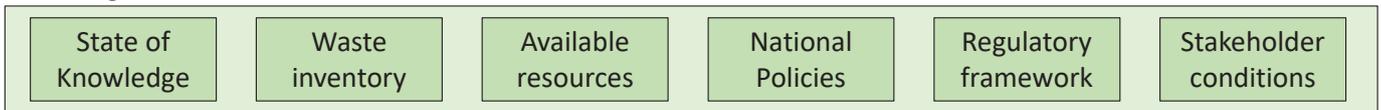


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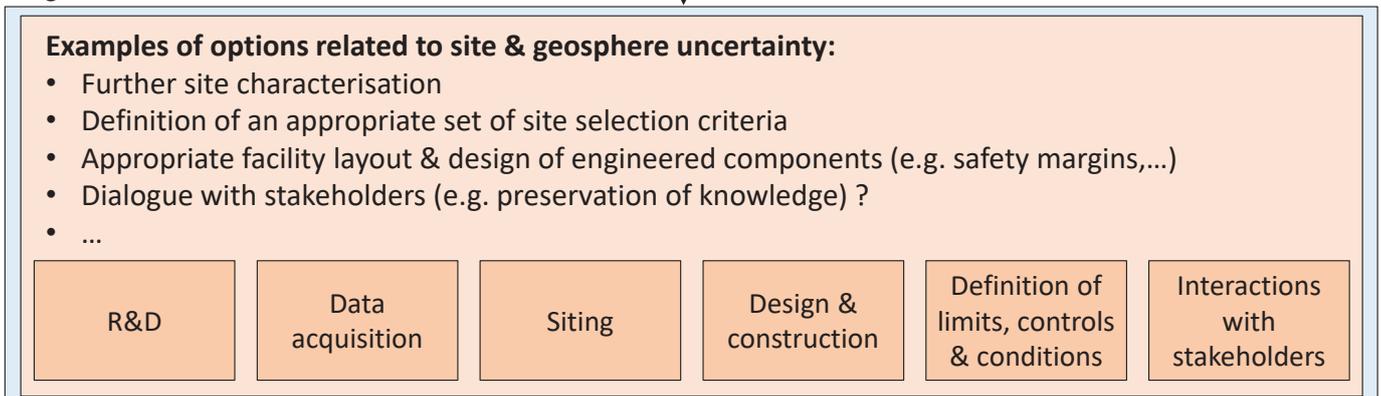
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OPTIONS FOR THE REDUCTION, MITIGATION OR AVOIDANCE

Prevailing circumstances



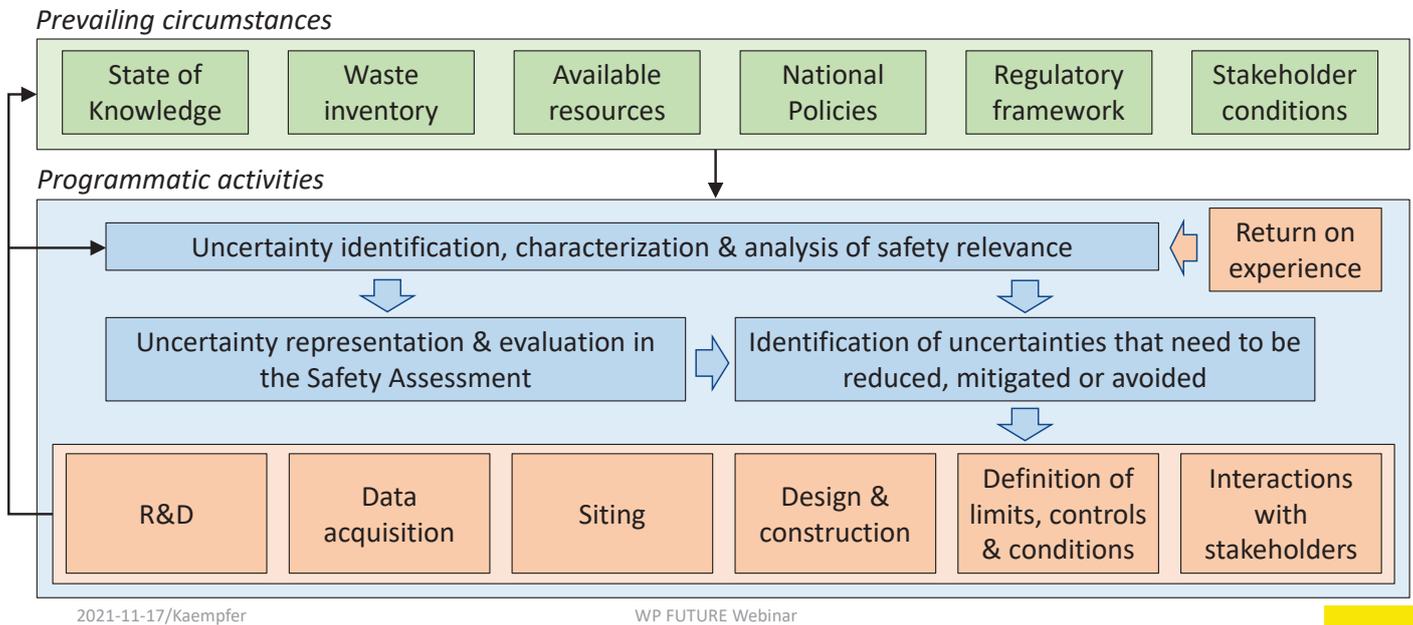
Programmatic activities



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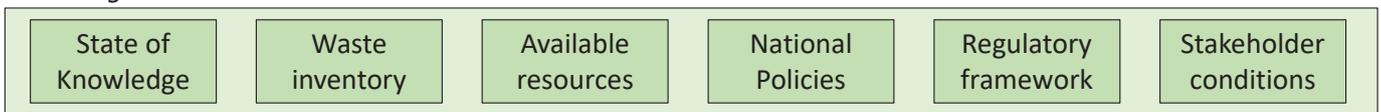
ITERATIVE NATURE OF AN UNCERTAINTY MANAGEMENT STRATEGY



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EXAMPLE – 3RD STAGE OF SITE SELECTION IN SWITZERLAND (1 – OVERVIEW)

Prevailing circumstances

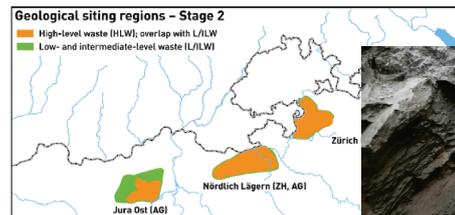


- Inventory: Spent fuel and high-level waste & low- / intermediate level waste
- Deep geological repository required by law
- Financial resources secured through funds (required by law)
- Sound regulatory framework, independent roles
- Most stakeholders accept the need for a repository and agree with the site selection process
- Advanced knowledge (disposal feasibility demonstrated 1988 / 2006)

EXAMPLE – 3RD STAGE OF SITE SELECTION IN SWITZERLAND (2 – STATUS)

- Site selection in three stages (Sachplan Geologische Tiefenlager)
 - safety driven site selection process
 - participatory process for e.g., sites for surface facility
- Result after 1st and 2nd stage / start of stage 3: Three siting regions to be further investigated
 - all with host-rock Opalinus Clay
 - all show high potential for safe repository project: Preliminary safety assessments demonstrate compliance with safety criteria and ample safety margin

Group of criteria	Criteria
1. Properties of host rock	1.1 Spatial extent 1.2 Hydraulic properties 1.3 Geochimie 1.4 Migration
2. Long-term stability	2.1 Durability 2.2 Erosion 2.3 Repository 2.4 Resources
3. Reliability of geological information	3.1 Characterization 3.2 Spatial 3.3 Temporal
4. Suitability for construction	4.1 Rock mass 4.2 Underground



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EXAMPLE – 3RD STAGE OF SITE SELECTION IN SWITZERLAND (3 – EXPECTATIONS)

- Last stage in site selection

3.1.3 Etappe 3: Standortwahl und Rahmenbewilligungsverfahren für SMA und HAA

Im letzten Schritt gilt es, die verbliebenen Standorte vertieft zu untersuchen und die standortspezifischen geologischen Kenntnisse falls nötig mittels erdwissenschaftlichen Untersuchungen (Seismik, Bohrungen) auf einen Stand zu bringen, der im Hinblick auf die Vorbereitung der Rahmenbewilligung einen vertieften Vergleich aus sicherheitstechnischer Sicht ermöglicht. Das Lagerprojekt wird unter Einbezug der Standortregion weiter konkretisiert und die sozioökonomischen Auswirkungen werden

- Reduction of uncertainty - if needed – by geological investigations (seismics, drilling) so as to allow for a more detailed safety based site comparison
 - geometric uncertainty (available space, thickness of host rock, faults, ...)
 - uncertainty of safety relevant properties (diffusion, permeability, ...)

- Safety case for the general license application

Für den gewählten Standort müssen genügend Kenntnisse vorliegen, um ein Rahmenbewilligungsgesuch einreichen zu können (Anhang IV). Etappe 3 leitet zum Rahmenbewilligungsverfahren und zur

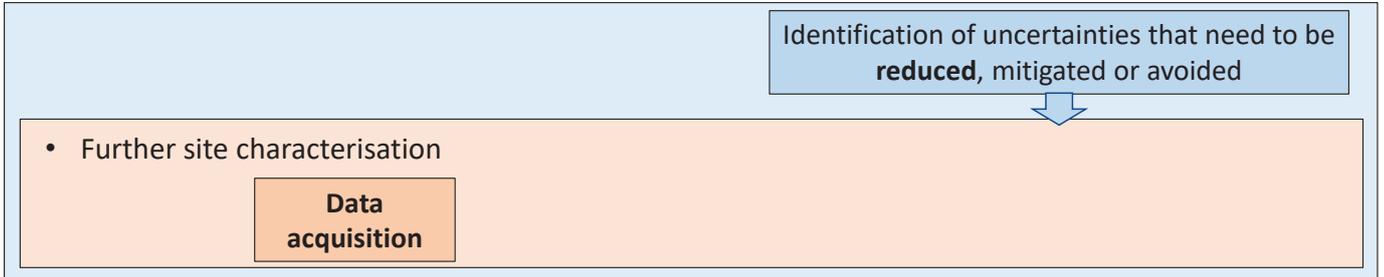
- Sufficient knowledge
- Confirmation, that remaining uncertainty does not compromise (post-closure) safety ... and also not technical feasibility, operational safety, ...

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EXAMPLE – 3RD STAGE OF SITE SELECTION IN SWITZERLAND (4A – REDUCTION OF UNCERTAINTY TO ALLOW FOR A SAFETY DRIVEN SITE SELECTION)

Programmatic activities



- Uncertainty in rock properties must be bound well enough as not to «mask» potential differences in candidate sites

→ Reduction of known uncertainty w.r. to rock properties (data uncertainty, e.g., diffusion)

→ Reduction of conceptual (model) uncertainty, e.g., diffusion ~ f(porosity)

→ Might lead to new knowledge (and new uncertainty)



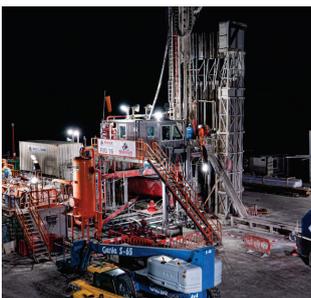
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EXAMPLE – 3RD STAGE OF SITE SELECTION IN SWITZERLAND (4B – REDUCTION OF UNCERTAINTY TO ALLOW FOR A SAFETY DRIVEN SITE SELECTION)

Targeted reduction of uncertainty → targeted data acquisition

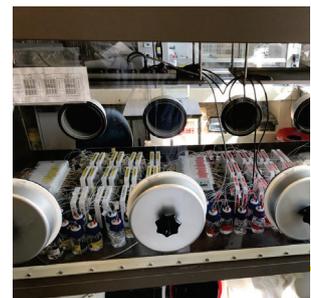
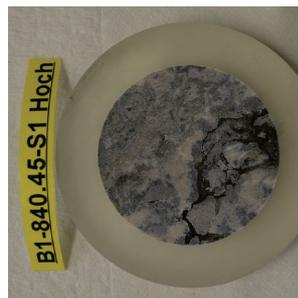
- Confirm (already ample) knowledge about host rock Opalinus Clay
 - systems understanding
 - diffusion data – is the Opalinus Clay equally favorable in all three sites?
- Enhance data set and knowledge regarding confining units (less clay rich)
 - broader uncertainty expected (higher variability)
 - are there significant differences between sites regarding barrier property?



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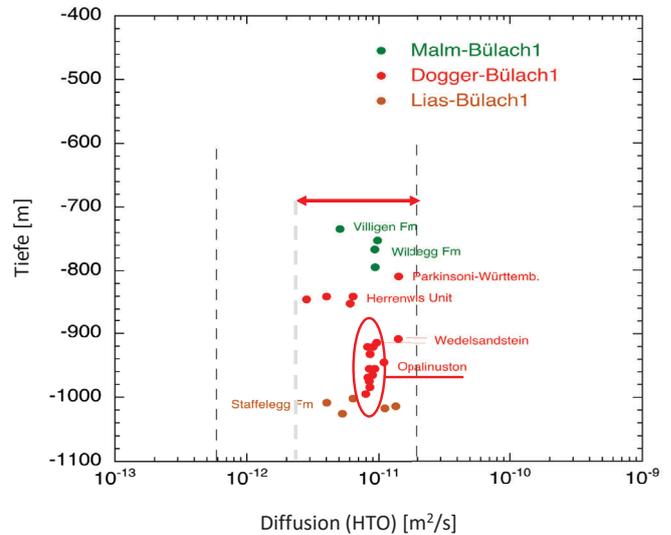
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EXAMPLE – 3RD STAGE OF SITE SELECTION IN SWITZERLAND (4C – REDUCTION OF UNCERTAINTY TO ALLOW FOR A SAFETY DRIVEN SITE SELECTION)

Intermediate results:

- Diffusive properties of Opalinus Clay confirmed
- Systems understanding enhanced
 - diffusion $\sim f(\text{clay content})$
 - $\sim f(\text{porosity, geometry factor})$
- Uncertainty bandwidth narrowed
 - but no significant difference between siting regions expected



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EXAMPLE – 3RD STAGE OF SITE SELECTION IN SWITZERLAND (5A – UNCERTAINTY REPRESENTATION IN SAFETY ASSESSMENT)

Programmatic activities

Uncertainty representation & evaluation in the Safety Assessment

Typical options:

- Deterministic vs. probabilistic approaches
- Conservative & bounding values/assumptions

- Remaining uncertainty does not undermine safety / a (safety) statement
- Safety margins are significant

Note: For a safety case, it must also be shown that the assessment is comprehensive (→ completeness)

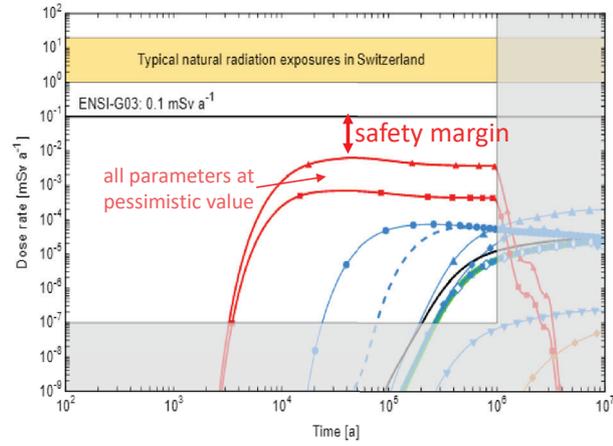
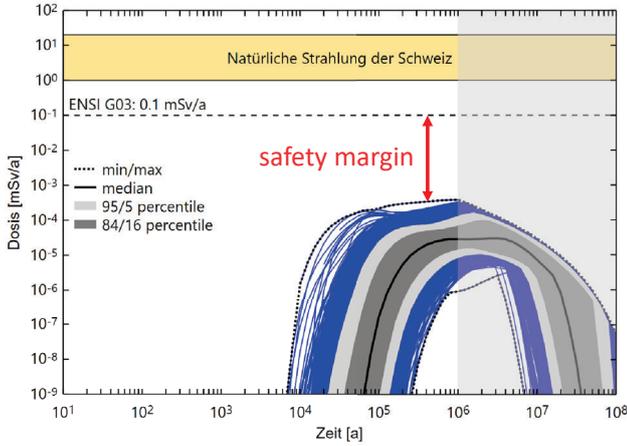
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EXAMPLE – 3RD STAGE OF SITE SELECTION IN SWITZERLAND (5B – UNCERTAINTY REPRESENTATION IN SAFETY ASSESSMENT)



Probabilistic and deterministic dose calculations (RN transport from waste to biosphere)

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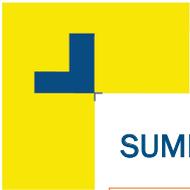


SUMMARY AND CONCLUSION

A systematic and safety oriented approach is essential for a high level of safety and the success of a disposal program – this includes a **systematic** and **safety oriented uncertainty management**

WP FUTURE Webinar





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A systematic and safety oriented approach is essential for a high level of safety and the success of a disposal program – this includes a **systematic** and **safety oriented uncertainty management**

a good (safety driven) process

a good (safe) system

a good (error free) analysis



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A systematic and safety oriented approach is essential for a high level of safety and the success of a disposal program – this includes a **systematic** and **safety oriented uncertainty management**

a good (safety driven) process

a good (safe) system

a good (error free) analysis



Safety-oriented management processes & principles



Stepwise & iterative approach



Regular stakeholder dialog

SUMMARY AND CONCLUSION

A systematic and safety oriented approach is essential for a high level of safety and the success of a disposal program – this includes a **systematic** and **safety oriented uncertainty management**

a good (safety driven) process



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a good (safe) system

Uncertainty management adapted to the milestone ahead

- is an uncertainty **safety relevant now**?
- how can it be characterized now?
- **must it** (and can it in practice) be **reduced, mitigated, avoided now**?
- **outlook:** For future milestones, can it (and must it) be
 - (better) characterized, reduced, mitigated, avoided
 - + robustness w.r. to «new» uncertainties
- For a safety case: Demonstrate safety under remaining uncertainty

a good (error free) analysis

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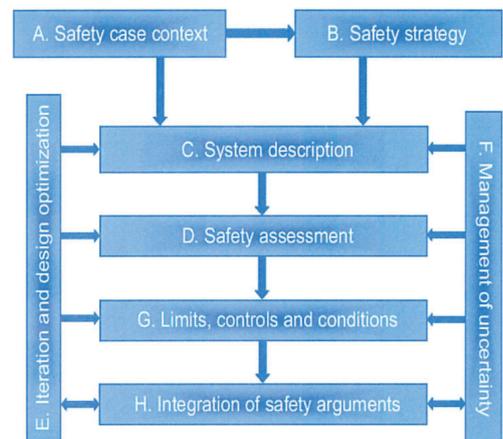
Implications of Safety case and Safety assessment progress for DGR and site selection development in Czech Republic

Lukáš Vondrovic
Head of RAW Repository Development Dept.
SÚRAO

20.06.2022

Safety case for radioactive waste disposal

- A collection of scientific, technical, managerial and administrative arguments and evidences in support of the safety of a disposal facility
- Safety assessment: systematic quantification of the exposure dose and comparison with dose criteria
- Safety case is developed in a step by step approach reflecting increasing technical knowledge and disposal concept maturity



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Legislative requirements for Safety case

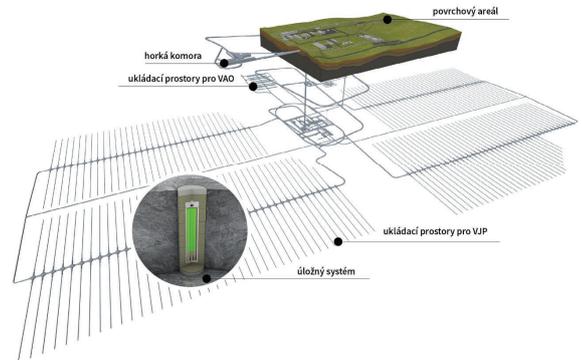
The annual effective dose to the representative person from the most exposed members will remain below 0.25 mSv under all possible features, events and processes that can occur during operational and postclosure period of the repository

The site must enable to prepare with good descriptibility and predictability

- Geological model
- Hydrogeological model
- Geomechanical model
- Geochemical model

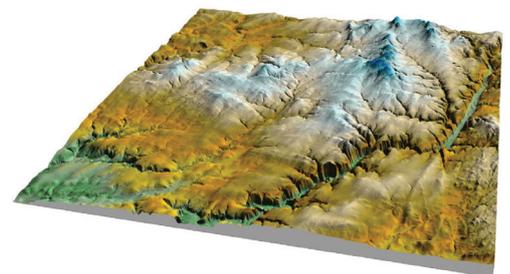
There is no geothermal energy source at site

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Legislative requirements for Safety case (example)

- depth extent and dimensions of the rock mass suitable for the siting of the deep geological repository and the distance from geological interfaces and tectonic faults that may act as pathways for the transport of radioactive substances,
- structural-geological properties of the rock environment in which the deep geological repository is to be located, including brittle and ductile tectonics,
- origin and expected development of the rock environment,
- suitability of the mechanical properties of the rocks in terms of ensuring the stability of the natural barrier of the deep geological repository,
- petrographic and mineralogical composition of the rock environment
- etc...

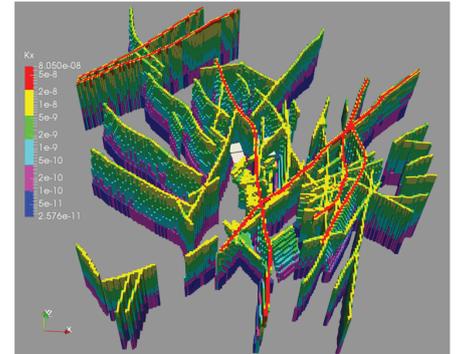


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Safety related requirements based on the analysis of safety functions „Isolate and Contain“ (IAEA SSR 5)

Capability to isolate radioactive waste from the environment

- Availability of homogenous rock domains in suitable depths several hundreds meters
- Geological stability (protection from external effect, eg. earthquakes, uplift, climatic changes)
- Good groundwater flow properties (based on hydrogeological models)
- Good transport properties
- Low probability of future human actions able to disturb repository



Capability to support EBS containment safety functions of

- Waste forms
- Waste packages
- Buffer, backfill and sealing materials

Describability and predictability of sites – necessity to understand site

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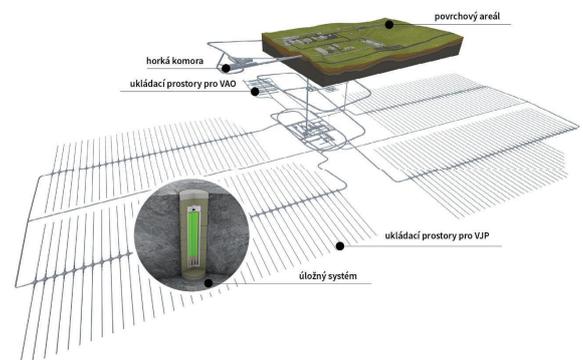
Components of DGR development in Czech Republic for safety case development

• Concept building

Development of disposal concept for conditions of crystalline host rock in Bohemian massif

• Site selection

Select final and backup site for DGR



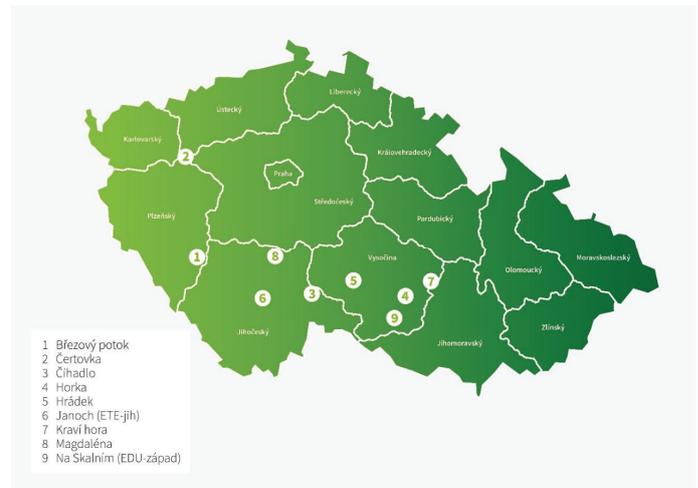
Case study: Shortlisting of number of sites 2019-2020

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Site selection process

Aim: Select final and backup site for DGR in 2030 by step-wise approach

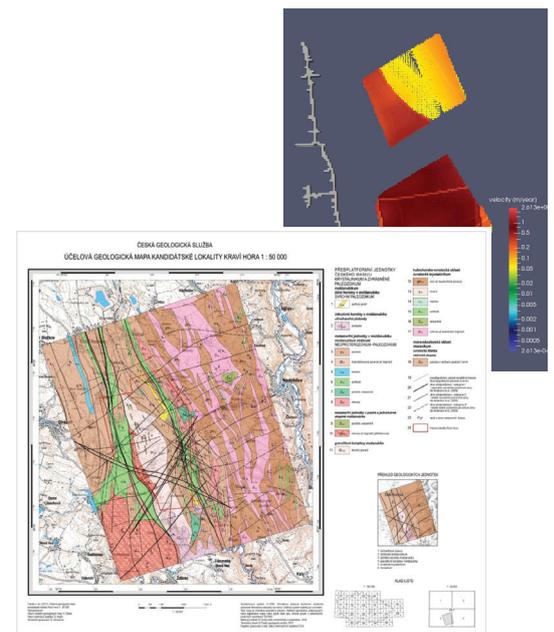
- 2019-2020: Site shortlisting 9 to 4 sites
- Preparation of demonstrational safety case
- Application of safety case based criteria
- Data from surface (any drillings)



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Kraví hora safety case

- To learn to prepare site specific safety assessment studies
- To get a deeper understanding of site specific problems
- To get feedback for further identification and specification of site specific features, events and processes
- Demonstration of capability of preparation of safety case

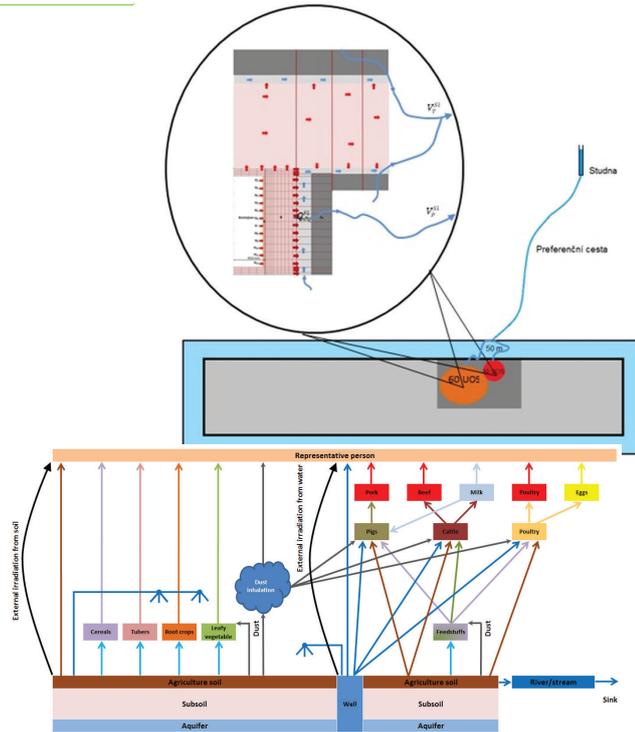


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Kraví hora safety case – demonstration of safety

- Safety function description
- FEP and scenarios development
- Disposal system description
- Site descriptive models creations
- Transport modelling from WDP to biosphere
- Dose estimation
- Uncertainty management

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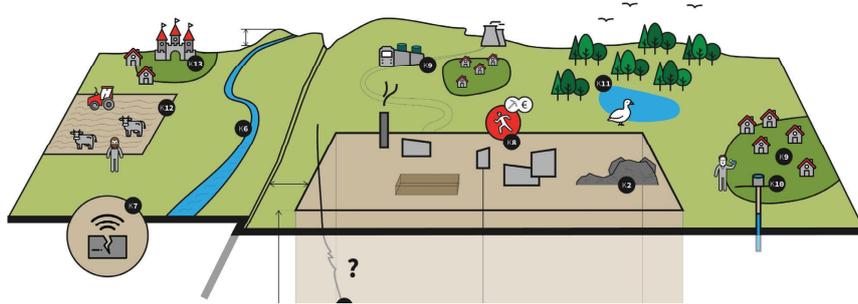
Site selection (key criteria)

- The siting characteristics of the individual localities must be compared in terms of the current stage of development of the DGR
- Characteristics that can be estimated based on current knowledge and that do not display mutual correlation
- Divided into partial indicators that represent the properties of specific sites
- Technical feasibility (C1, C2), Long term and operational safety (C3-C9), environmental assessment (C10-C13)
- 13 key criteria, 38 indicators



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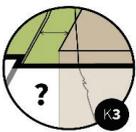
Site selection



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Long-term safety criteria

C3: Describability and predictability of the homogeneous blocks



The geological conditions must be such that they can be described to an extent that is accurate enough for the creation of 3D structural-geological models of the potential sites. This criterion included the definition of both the rock block considered for the construction of the underground part of the DGR and the geological conditions in the wider surroundings of the sites.

Indicators:

- degree of the brittle failure of the rock mass - fault structures
- degree of the brittle failure of the rock mass - fracture systems
- degree of ductile deformation

Used data: geophysical measurements, remote sensing, geological mapping

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Long-term safety criteria



C4 Variability of the geological properties

A degree of variability of the relevant geological properties that is so high that it does not allow for the preparation of reliable 3D geological, hydrogeological and geochemical models constituted one of the exclusion criteria. However, in this phase of the DGR site selection process (the reduction in the number of potential sites from 9 to 4), concerning which mostly surface geological research only has been conducted to date, this factor serves for site comparison purposes.

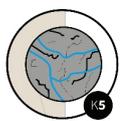
Indicators:

- the spatial variability of the rock environment
- the petrological variability of the rocks

Used data: geological models, surface geophysics, microstructural data

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Long-term safety criteria



C5 Water flow characteristics in the vicinity of the DGR and the transport characteristics

The assessment of the hydrogeological and transport characteristics of the site constitutes important input material for the safety assessment of the DGR. The most important way in which radionuclides may spread into the surrounding environment (biosphere) is generally considered to be their migration via groundwater flow.

Indicators:

- flow time from the DGR to drainage areas
- flow rate at the DGR level
- permeability in the DGR area
- the descending vertical flow component
- the maximum permeability of the fracture zones up to 500 m from the limits of the DGR
- specific flow in the DGR area
- dilution ratio
- **Used data: hydraulic models of sites**

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Long-term safety criteria



C6 Identification and location of drainage bases

Locations at which groundwater will potentially drain from the DGR (the so-called drainage base) influence the safety of the site. Groundwater flow is an important factor that influences the mobility of radionuclides in the rock environment. The DGR should be located in such a way that the transport pathways for radionuclides towards the drainage base(s) are as long as possible and the transport of radionuclides as slow as possible. Therefore, based on hydraulic models of the sites, indicators were defined that included various aspects of the anticipated radionuclide transport pathways from the repository level to the biosphere, i.e. to where water flows from the repository, to how many streams and the distance of the DGR from the nearest drainage base.

Indicators:

- the number of drainage streams
- representation of the drainage from the DGR area in the form of a single stream
- representation of the drainage from the DGR area in the form of a single catchment area,
- horizontal distance from the DGR site to the drainage site

Used data: hydraulic models of sites

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Long-term safety criteria



C7 Seismic and geodynamic stability

The geological structure of the area intended for the siting of the DGR must guarantee the stability of the facility for at least hundreds of thousands of years. According to Section 18, paragraph 2, g), i) and j) of Decree No. 378/2016 Coll. The following factors must be assessed: the occurrence of endogenous and exogenous phenomena (g) the anticipated development of the climate (i) and the vulnerability of the rock environment to long-term climate change (j).

Indicators:

- maximum horizontal seismic acceleration value
- altitude gradient
- percentage share of the relief area affected and deformed by recent cycles of reverse erosion and slope deformation
- occurrence of volcanic rocks of Paleogene to Holocene age and acids

Used data: climatic and seismic predictions, morphological analyses

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Long-term safety criteria



C8 Characteristics that might lead to intrusion into the DGR due to future human activities

According to international recommendations, intrusion into the repository as a consequence of future human activities may occur for two main reasons, i.e. either intrusion in order to use the disposed of waste as a secondary raw material or intrusion with the intention of the exploitation of other raw material resources in the area. With respect to the latter, it will, therefore, be important to prevent unintentional intrusion into the repository by humans following the potential loss of information on the existence of the facility.

Indicators:

- mineral deposit conditions at the site

Used data: geological databases, archives

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Operational safety criteria



C9 Phenomena that influence the spread of radioactive materials

With respect to the DGR, this criterion primarily concerns the assessment of the impact of a possible emergency situation regarding the so-called hot chamber in which spent nuclear fuel will be removed from storage and transport containers and inserted into waste disposal packages. The spread of radioactive materials could also occur in the event of an emergency during the transportation of the SNF from the storage facility to the DGR.

Indicators:

- distribution and density of the population and its development in terms of the spread of radioactive materials
- distance from a nuclear power plant

Used data: preliminary calculation of „collective dose“

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Implications for next stage

- Preparation of site specific safety cases
- Knowledge of site specific groundwater properties and hydrochemical zonation is essential
- Site specific knowledge of transport properties of host rock
- Using of data related with isolation part of repository (sorption, diffusion, Kd etc) – implication from FUTURE project

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General implications of safety assessment

- Disposal concept is important since the beginning of the process of siting
- Constructibility gains increasing importance in the later phases of site
- Geology data importance increases toward early stages and confirmation process
- Sensitivity analyses can provide feedback on what aspect of the site and design that are important

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Thank you for your attention
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