Karlsruhe Institute of Technology

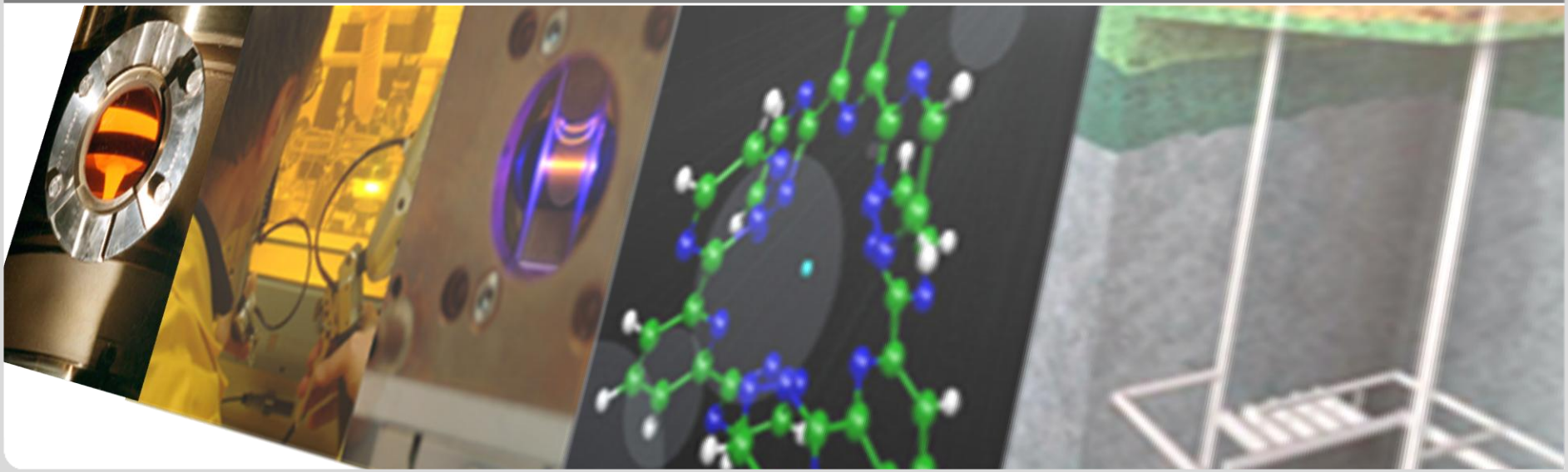
UNIVERSITÉ DE STRASBOURG

Visit to the Institute for Nuclear Waste Disposal (INE)

European Summer School Radiochemistry and Nuclear Instrumentation

Horst Geckeis (Email: horst.geckeis@kit.edu)

INSTITUT FÜR NUKLEARE ENTSORGUNG (INE)



European Summer School
Radiochemistry and nuclear instrumentation (low level radioactivity)

Visit at the Institute for Nuclear Waste Disposal (INE),
Karlsruhe Institute of Technology (KIT)

23 august 2017

Programme

09:00 h	**Arrival at KIT-Campus North entrance**	
09:15 h	**Welcome at INE and introduction**	*H. Geckeis*
09:30 h	**Research towards radioactive waste disposal at KIT-INE**	*H. Geckeis*
10:15 h	**Coffee break**	
10:30 h	**Ultratrace analysis of radionuclides by AMS**	*F. Quinto*
11:30 h	**Radionuclide speciation by laser spectroscopy**	*C. Garcia-Perez*
12:30 h	**Lunch**	
13:30 h	**Synchrotron based X-ray spectroscopic characterization of radionuclides and Visit of INE-Beamlines at ANKA (ANKA buiding)**	*T. Vitova*
15:00 h	**Visit of INE laboratories**	
	• Hot cells	*M. Herm*
	• Mass spectrometry	*F. Quinto*
	• Laser spectroscopy	*C. Garcia-Perez*
	• Geochemistry	*C. Joseph*
17:00 h	**End of visit**	

Topics

- What is KIT?
- Nuclear waste in Germany and worldwide
- Research at the Institute for Nuclear Waste Disposal

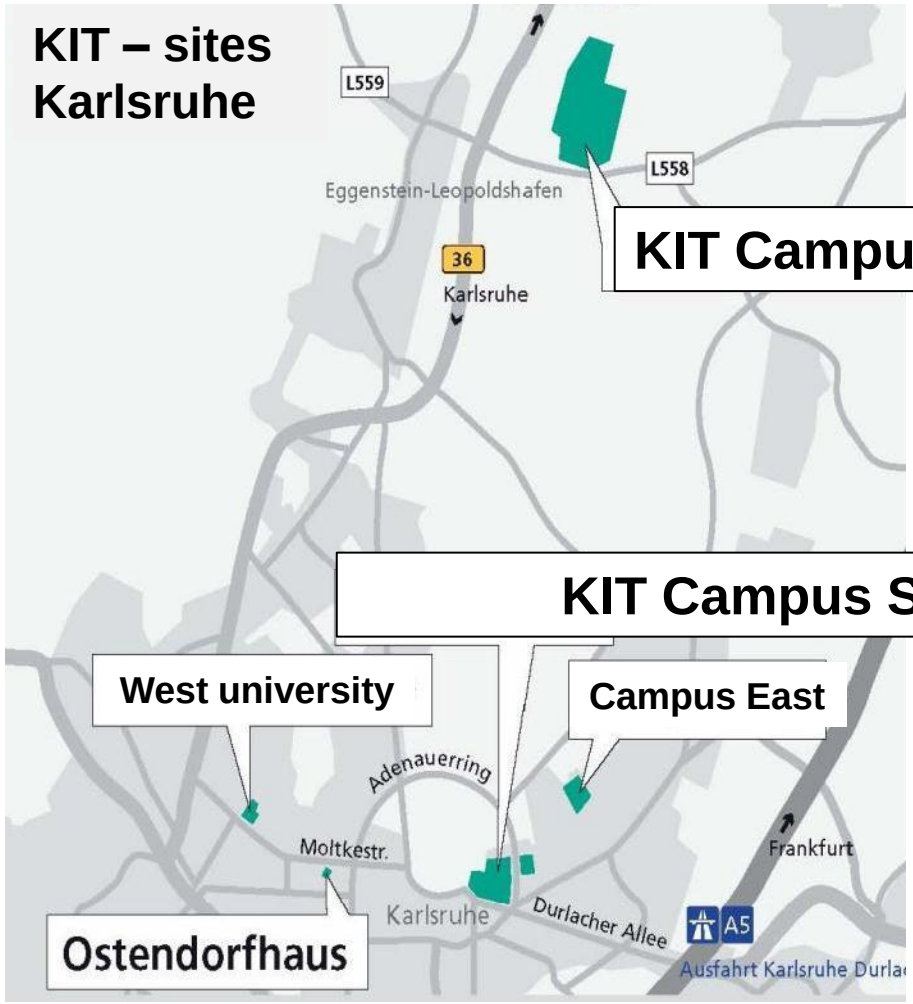
Karlsruhe Institute of Technology

KIT – The Research University in the Helmholtz Association

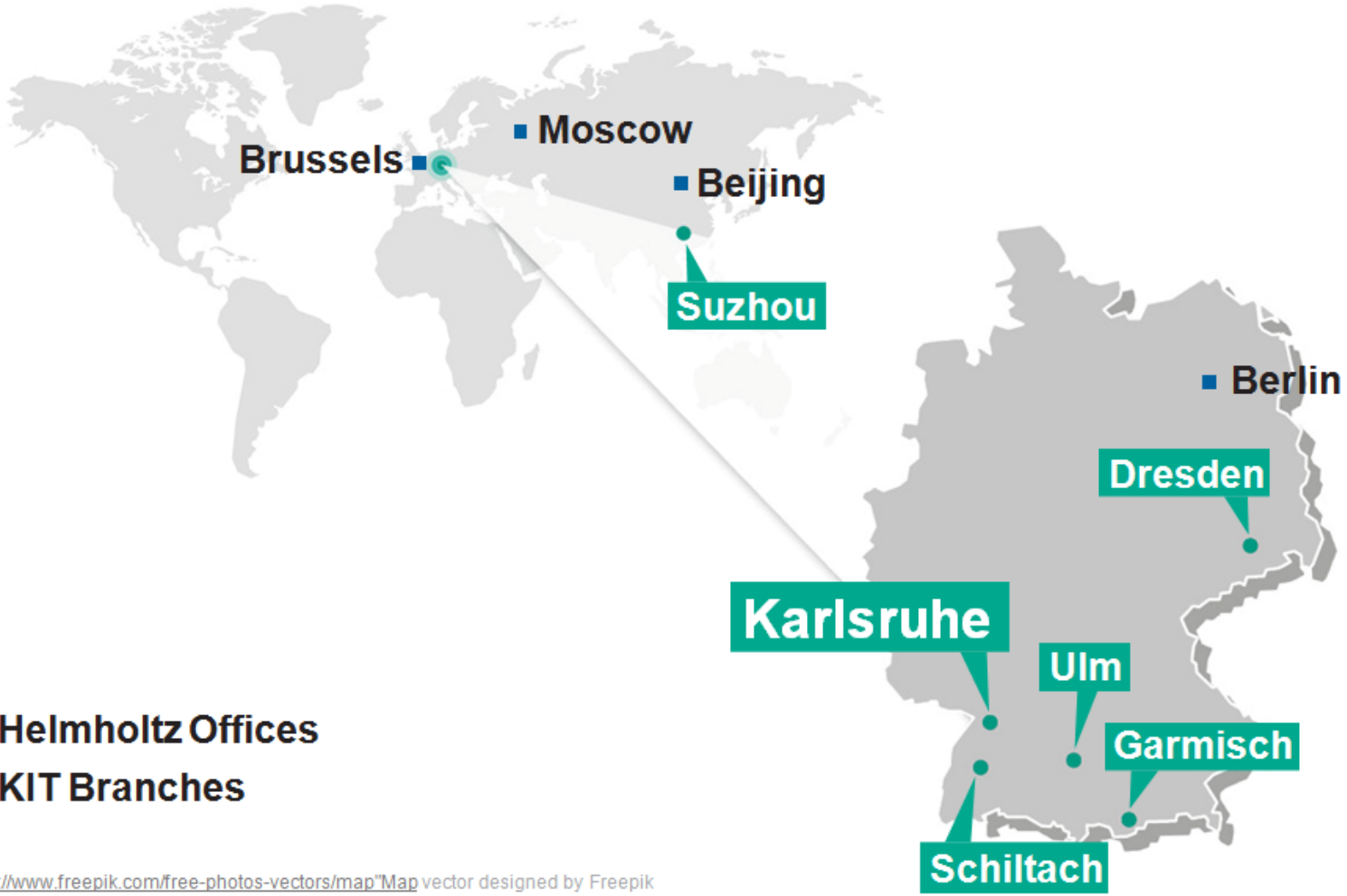
Status: January 2017

PRESIDENTIAL COMMITTEE

Two KIT sites



Branch Offices



- Helmholtz Offices
- KIT Branches

http://www.freepik.com/free-photos-vectors/map"Map vector designed by Freepik

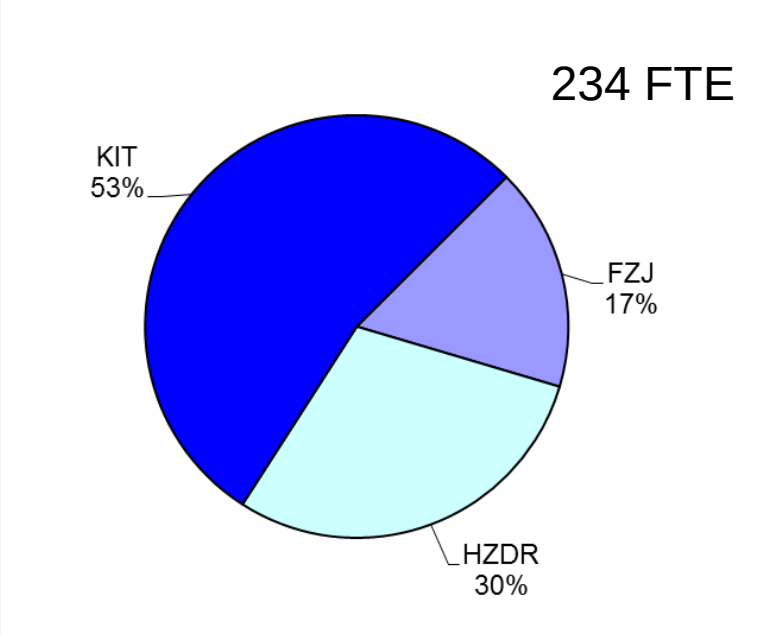
The research programme NUSAFE

Nuclear Waste Management and Safety as well as Radiation Research

HELMHOLTZ | ASSOCIATION

HELMHOLTZ | ASSOCIATION

NUSAFE resources:



67% Nuclear Waste Management

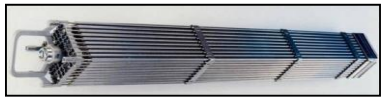
33% Reactor Safety

Topics

- What is KIT?
- Nuclear waste in Germany and worldwide
- Research at the Institute for Nuclear Waste Disposal

Nuclear waste in Germany

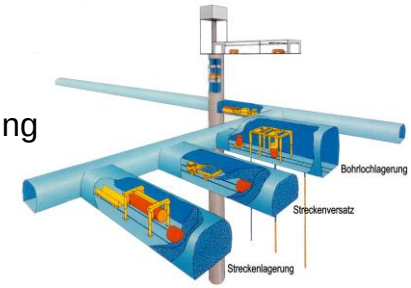
Irradiated
Nuclear fuel

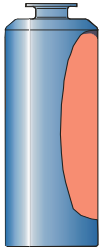


Vitrified waste
From reprocessing

Intermediate storage

Repository
For heat producing
Radioactive
Waste





Non-heat producing radio-
Active waste from
NPP and decommissioning

Non-heat producing radioactive waste
From medicin, research etc.

Intermediate storage

Repository
For non-heat
producing
Radioactive
Waste

Nuclear waste in Germany

Irradiated
Nuclear fuel

Vitrified waste
From reprocessing

Intermediate storage

Non-heat producing radio-
Active waste from
NPP and decommissioning

Non-heat producing radioactive waste
From medicin, research etc.

Intermediate storage

HEAT GENERATING WASTE:

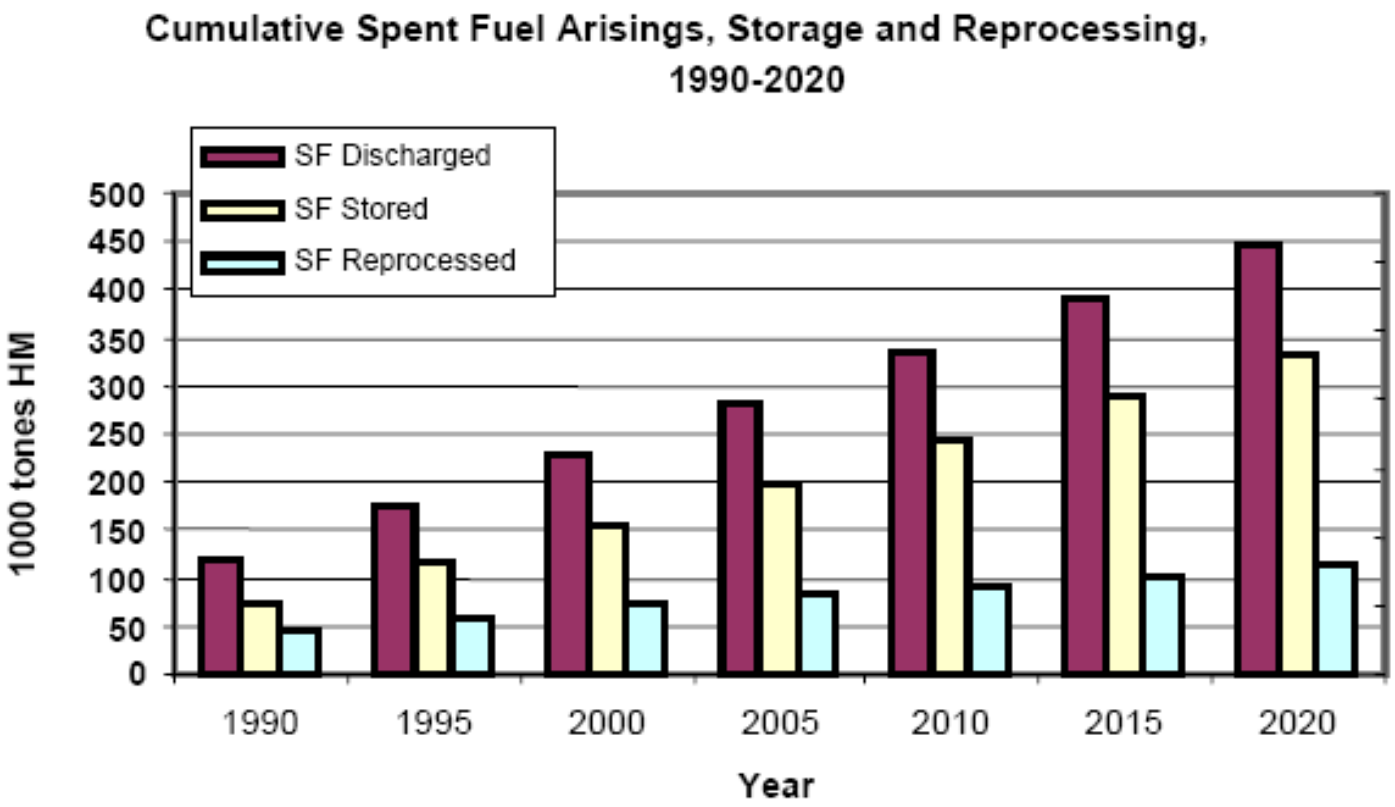
(10.550 tHM) NPP
21.000 m³
5.700 m³ Research/
Prototype
Reactors/
Pilot repr. Plant

1.400 m³ vitrified waste
(7.973 coquilles from 6.700 tSM repr. spent fuel)

WASTE WITH NEGLIGIBLE HEAT GENERATION:

~ 300.000 m³

Spent fuel arising world wide



Per year: 10.500 t spent nuclear fuel

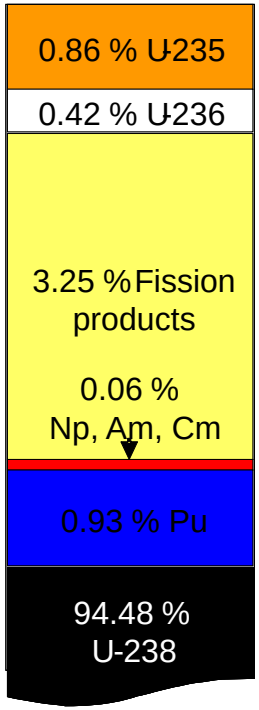
Cumulative until 2020: ~ 445.000 t spent nuclear fuel

~ 1/3 reprocessed ➔ Vitrification of remaining high level liquid waste

Fukuda et al. IAEA overview of global spent fuel storage, IAEA-CN-102/60

Content of high-level nuclear waste

spent fuel



PWR; Burn-up 33GWd/tU

Some long-lived fission and activation products

C-14	($t_{1/2}$ ~ 5.700 a)
Cl-36	($t_{1/2}$ ~ 300.000 a)
Se-79	($t_{1/2}$ ~ 360.000 a)
Tc-99	($t_{1/2}$ ~ 213.000 a)
I-129	($t_{1/2}$ ~ 15,6 Ma)

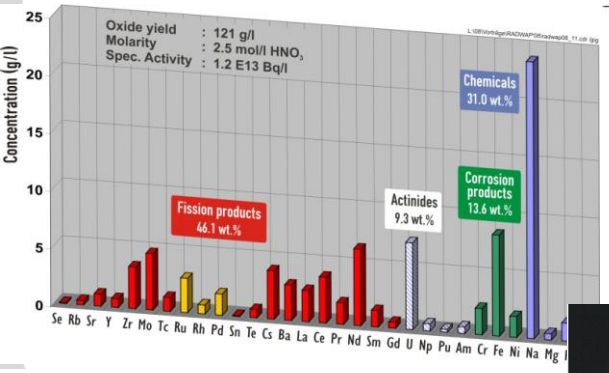
Fission products:
Cs-137, Sr-90 ($t_{1/2}$ ~ 30 a)

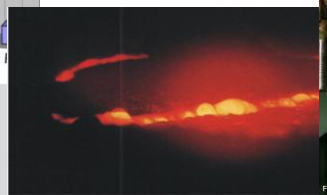
Important Transuranium elements:

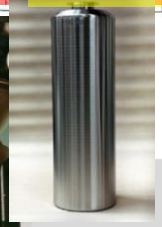
Pu-238 ($t_{1/2}$ ~ 88 a)
Pu-239 ($t_{1/2}$ ~ 24.110 a)

Am-241($t_{1/2}$ ~ 432 a)
Cm-243($t_{1/2}$ ~ 29 a)

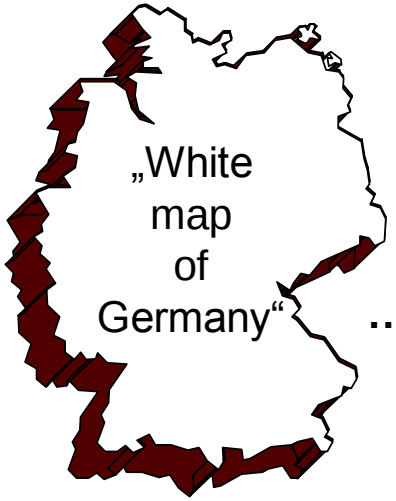
Np-237 ($t_{1/2}$ ~ 2.1 Ma)







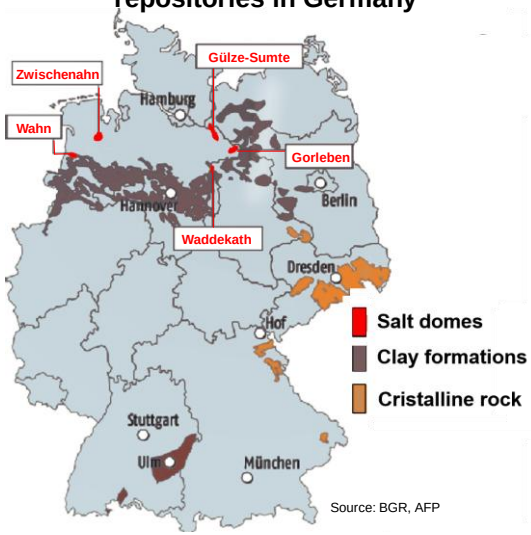
Vitrified liquid waste



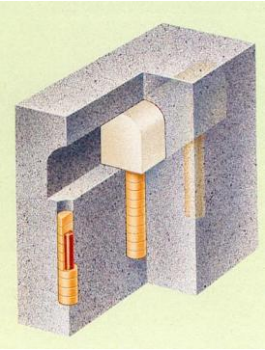
… is not white!

No host rock formation excluded

Potential host rocks for nuclear waste repositories in Germany

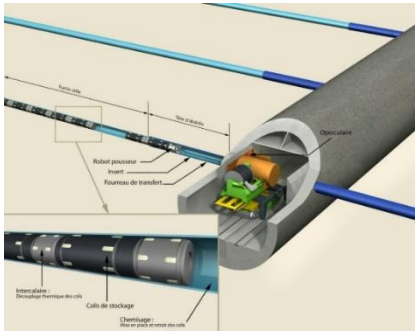


➔ Site selection does not only require a science based selection of host rocks but also a selection of repository concepts!

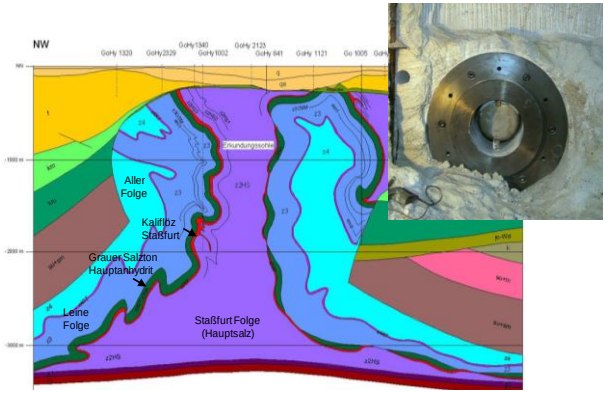


Copper canister for spent nuclear fuel

Crystalline rock



Argillaceous rock

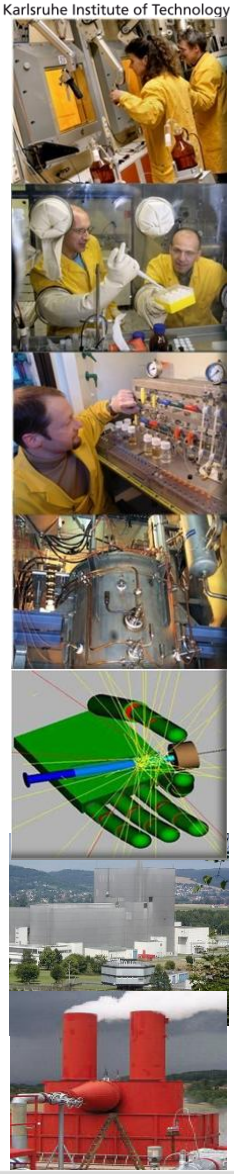


Rock salt

Topics

- What is KIT?
- Nuclear waste in Germany and worldwide
- Research at the Institute for Nuclear Waste Disposal

KIT – Institute for Nuclear Waste Disposal (INE)



Research and development activities at KIT-INE are an integral component of national provident research and encompass the following areas:

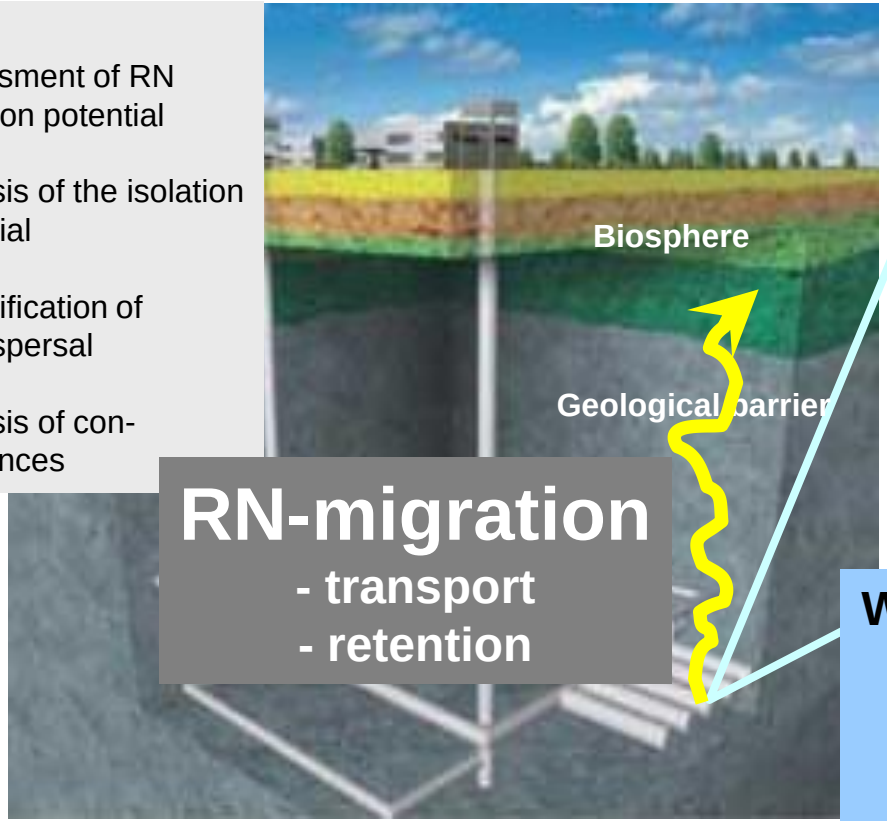
- **Long-term safety research for nuclear waste disposal**
- **Actinide coordination chemistry**
- **Decommissioning of nuclear facilities**
- **Geothermal energy**
- **Research for radiation protection**
- **(Immobilisation of high level radioactive waste by vitrification)**

NUSAFE

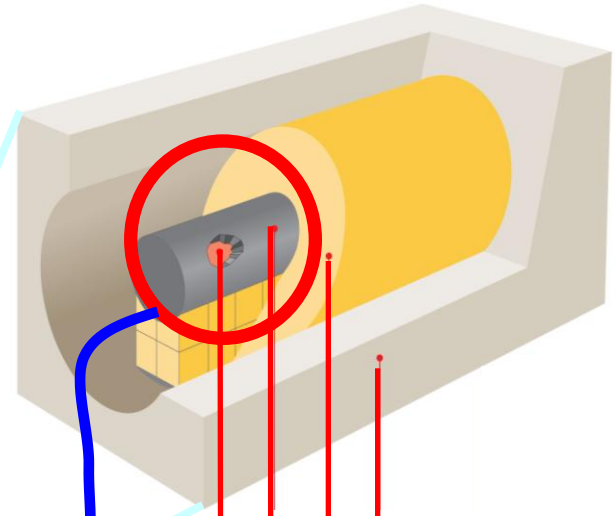
Processes in the multibarrier system

- Chemical and geochemical aspects -

- Assessment of RN retention potential
- Analysis of the isolation potential
- Quantification of RN dispersal
- Analysis of con-sequences



RN-migration
- transport
- retention



Water access:

RN-source (solubility)

Geological barrier

Technical barrier

Container

Waste form

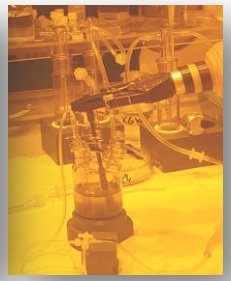
Source term:

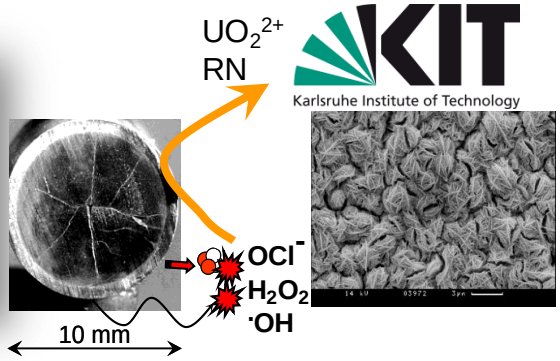
$$r = k_0 \times A_{\min} \times e^{-E_a/RT} a_{H^+}^{n_{H^+}} \; g(I) \prod_i a_i^{n_i} f(\Delta G_r)$$

Transport + retention:

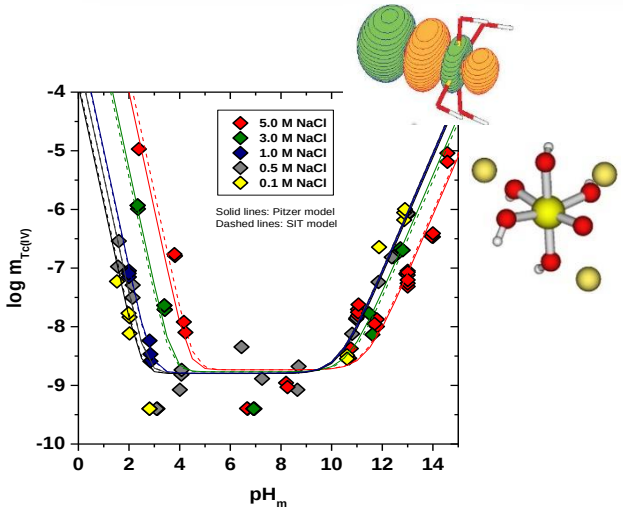
$$-u\frac{\partial c_i}{\partial t} + D\frac{\partial^2 c_i}{\partial x^2} - \frac{\rho}{\theta} K_d \frac{\partial c_i}{\partial t} = \frac{\partial c_i}{\partial t}$$

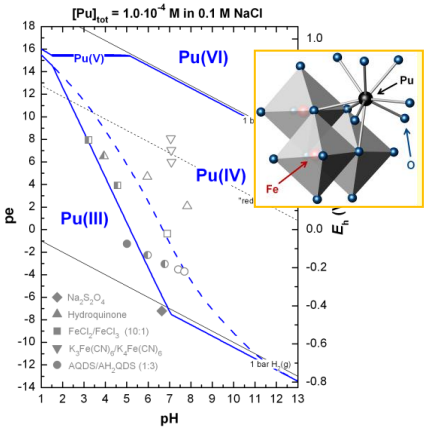
Nuclear waste form behavior
- spent nuclear fuel
- HLW glass
- Cementitious waste



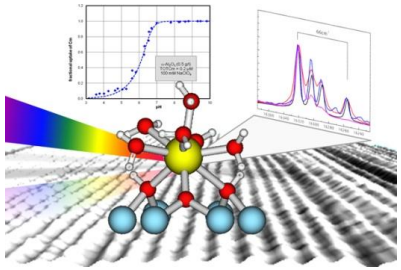


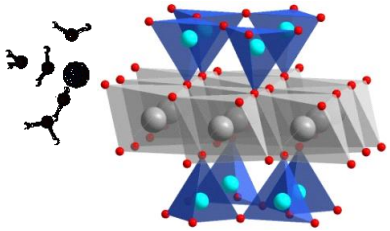
Aquatic chemistry
- Solubility
- Redox
- Complexation

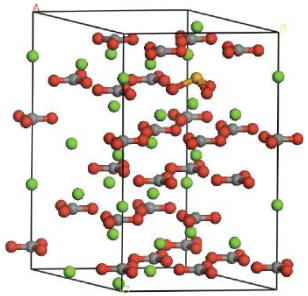




Solid/liquid interactions
- Sorption to mineral surfaces
- solid-solution formation
- Colloid formation

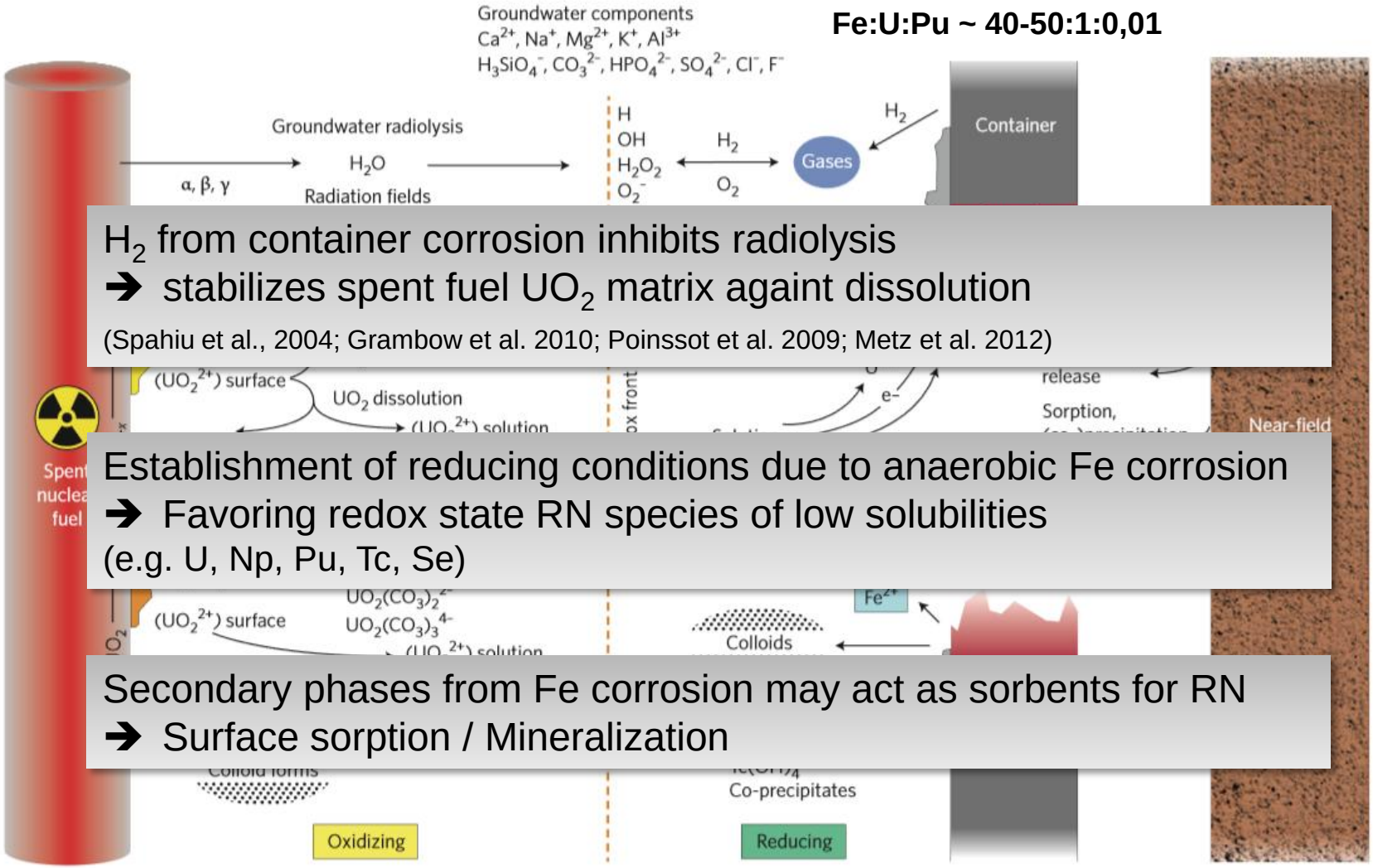






Processes in the near field of a repository for spent nuclear fuel

Molar ratio:
Fe:U:Pu ~ 40-50:1:0,01



H_2 from container corrosion inhibits radiolysis
➔ stabilizes spent fuel UO_2 matrix againt dissolution

(Spahiu et al., 2004; Grambow et al. 2010; Poinssot et al. 2009; Metz et al. 2012)

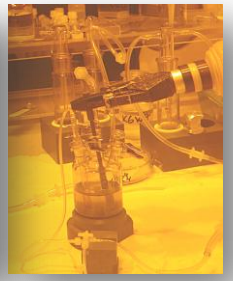
Establishment of reducing conditions due to anaerobic Fe corrosion
➔ Favoring redox state RN species of low solubilities
(e.g. U, Np, Pu, Tc, Se)

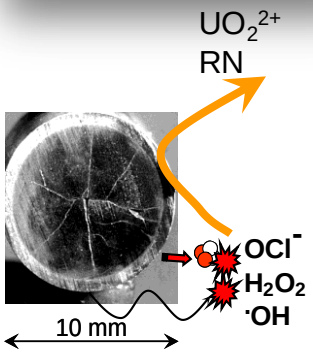
Secondary phases from Fe corrosion may act as sorbents for RN
➔ Surface sorption / Mineralization

R. Ewing, Nature Mat., 2015

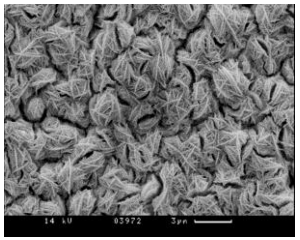
The role of the waste form:
- Spent nuclear fuel -

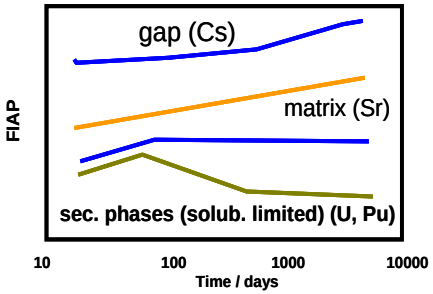
- Impact of radiolysis on fuel behavior in a repository
- Long term stability of fuel in the repository („H_2-effect“)
- „Instant release“ fraction
- New issue: degradation behavior of heat generating radioactive waste under extended interim storage conditions?

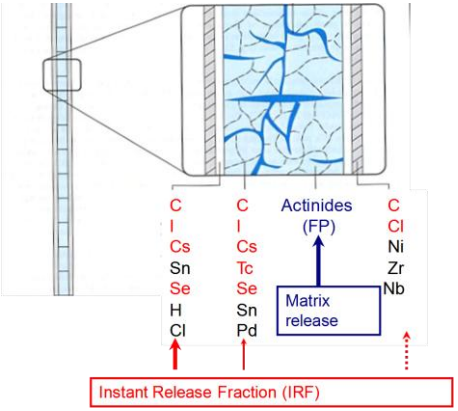


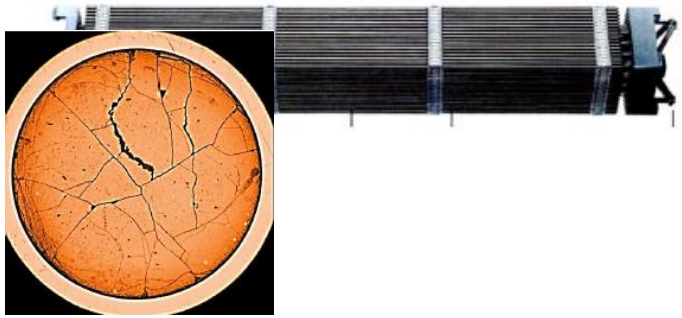


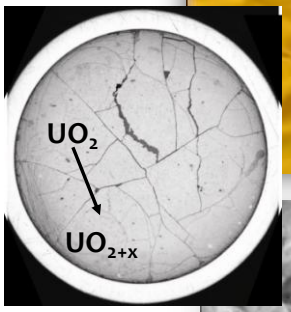
Fuel pellet

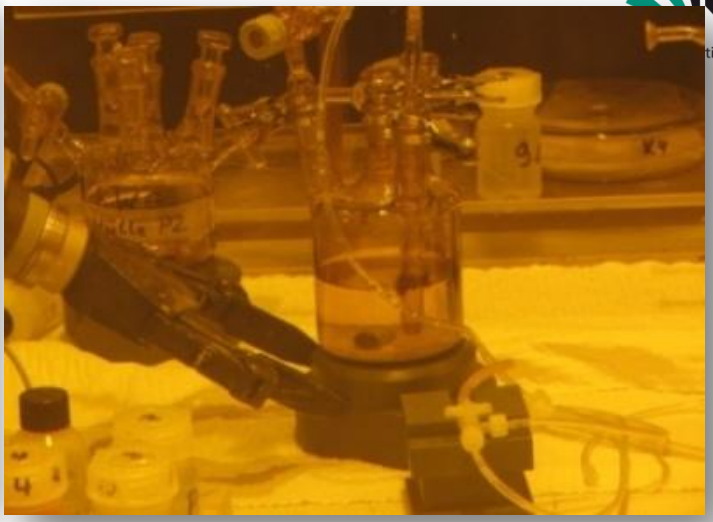


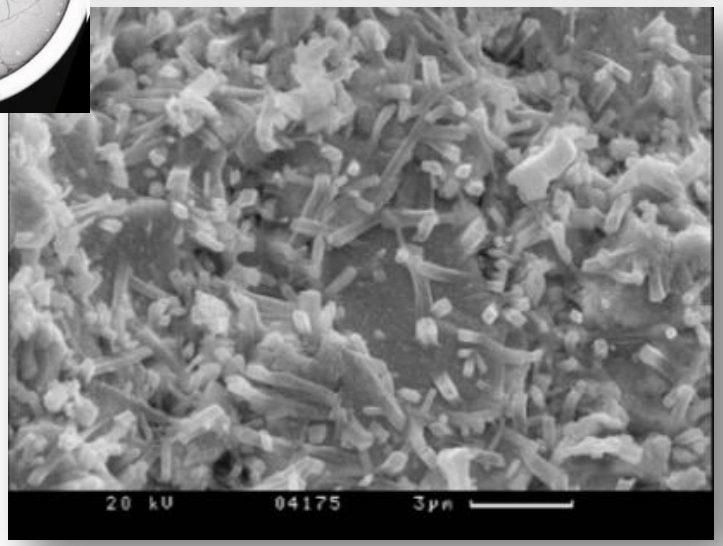


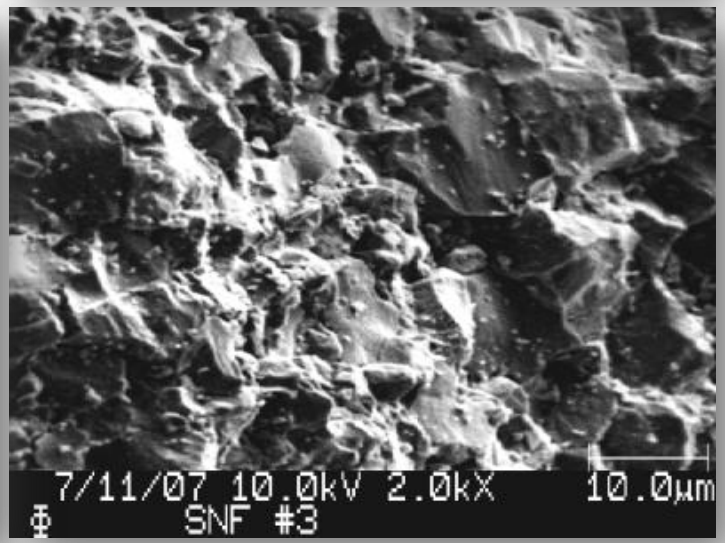










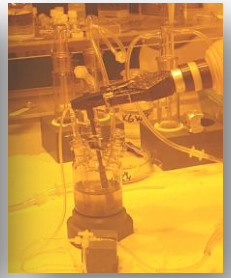


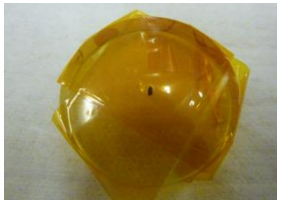
Anoxic corrosion of UOX fuel

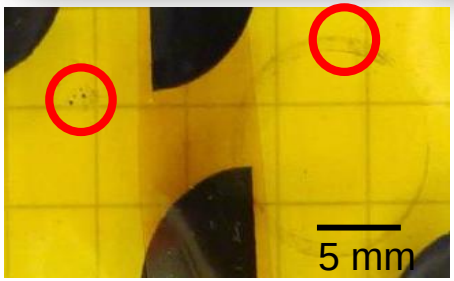
Anoxic corrosion of UOX fuel in presence of H_2

Source: Loida et al. (2007) MRS SBfNWM 30, vol. 985, 14-20; Metz et al. RCA, 2012

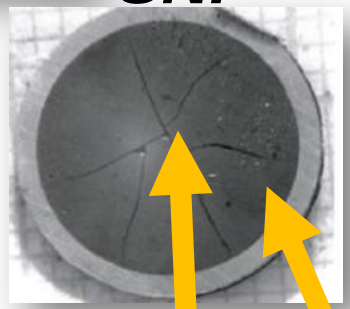
HR-XANES analysis of Uranium oxidation state in spent UO_2 fuel

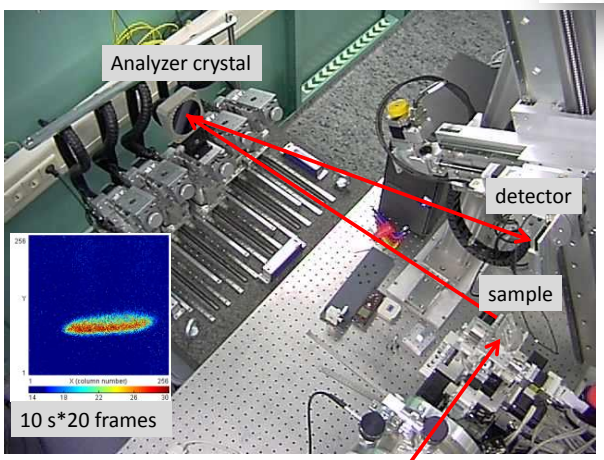






SNF



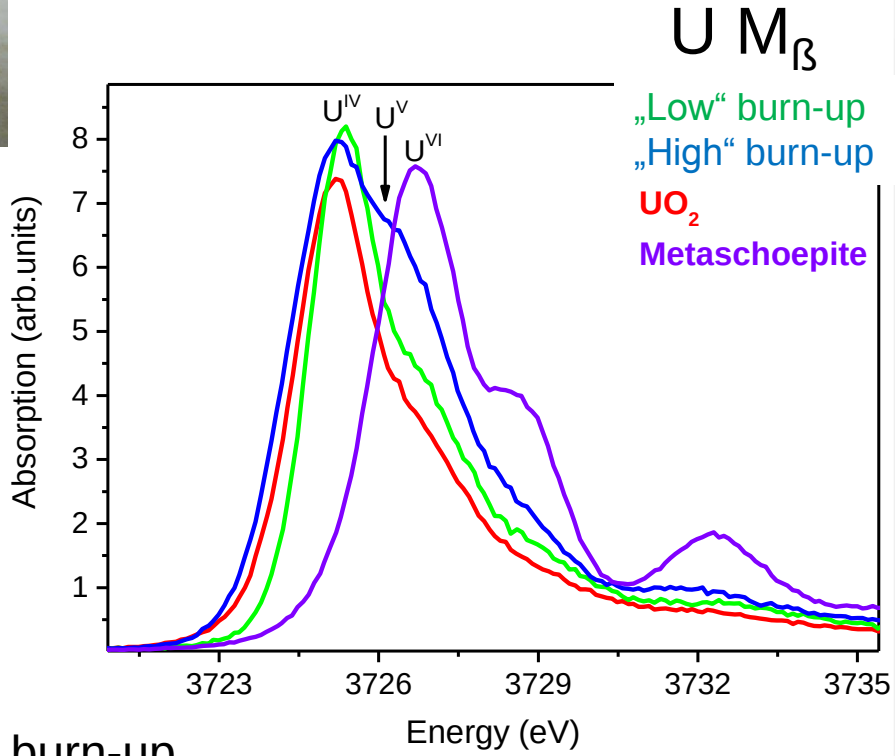


HR-XAS spectrometer at INE-Beamline

X-ray beam

„Low“ burn-up center

„High“ burn-up rim



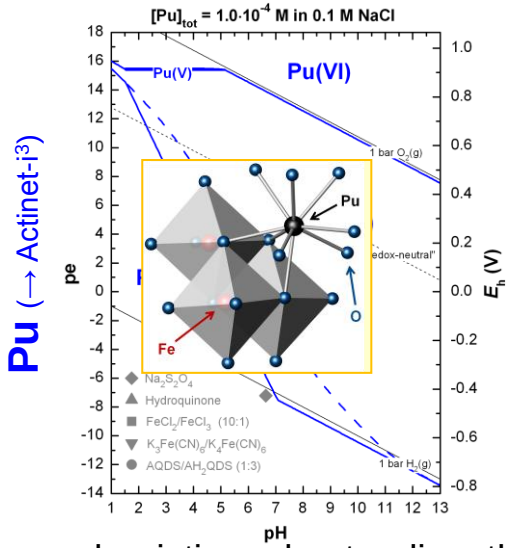
1st time:
Detection of U(V) in spent fuel

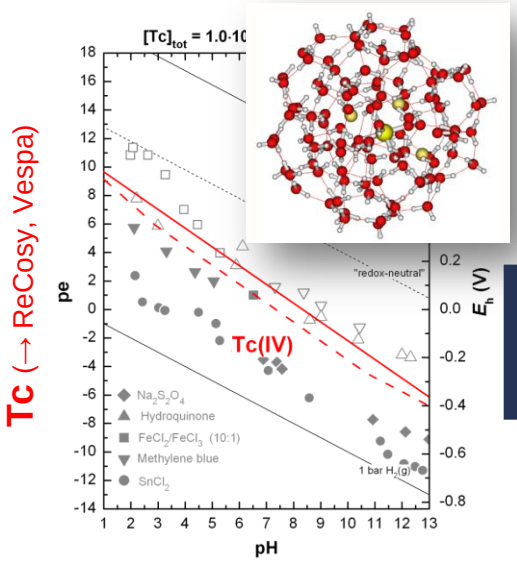
S. Bahl et al., to be submitted to Environ. Sci. Technol.

Karlsruhe Institute of Technology

Example: redox chemistry of actinides/fission products

Fundamental research





THEREDA

OECD NEA TAB Thermochemical Data Base Project

Reference data base projects

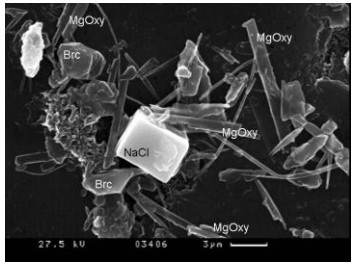
mechanistic understanding, thermodynamics, kinetics

(coop.: Kyoto University, KAIST, LANL - Carlsbad office)

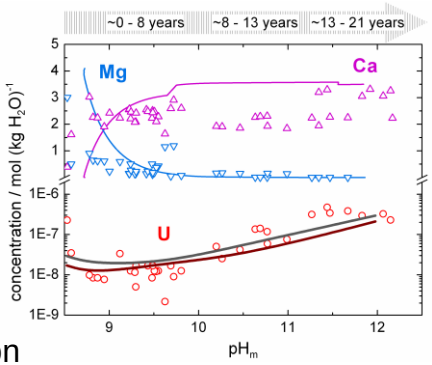
Example: radionuclide retention in cementitious phases

Applied research

1:1 scale experiments from the Asse mine



Secondary phase formation During cement corrosion

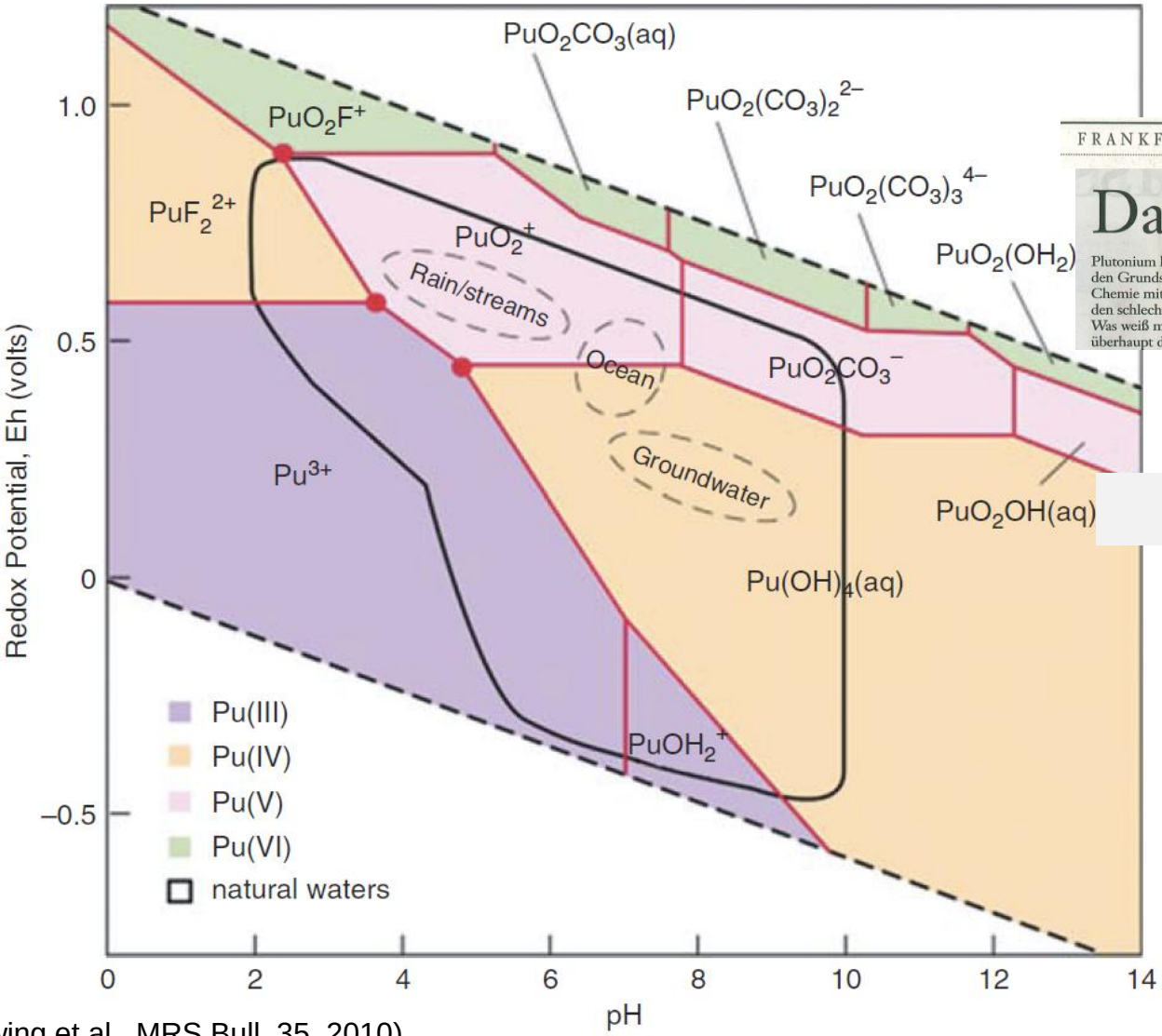


Geochemical description

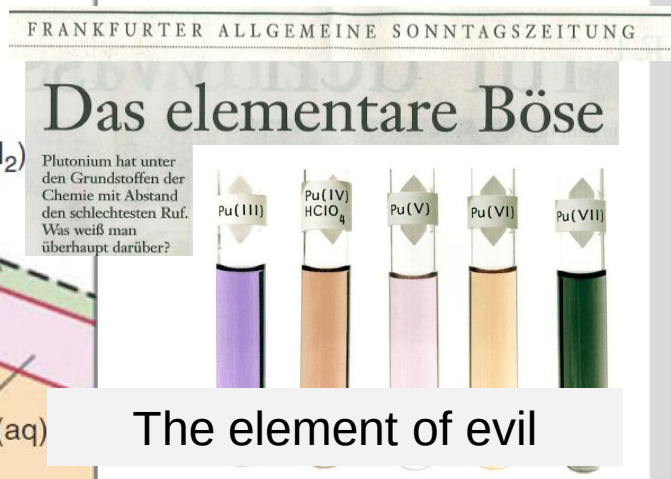
Öko-Institut e.V. Institut für angewandte Ökologie Institute for Applied Ecology

Bundesamt für Strahlenschutz

Redox chemistry of Plutonium



(Ewing et al., MRS Bull. 35, 2010)


FRANKFURTER ALLGEMEINE SONNTAGSZEITUNG

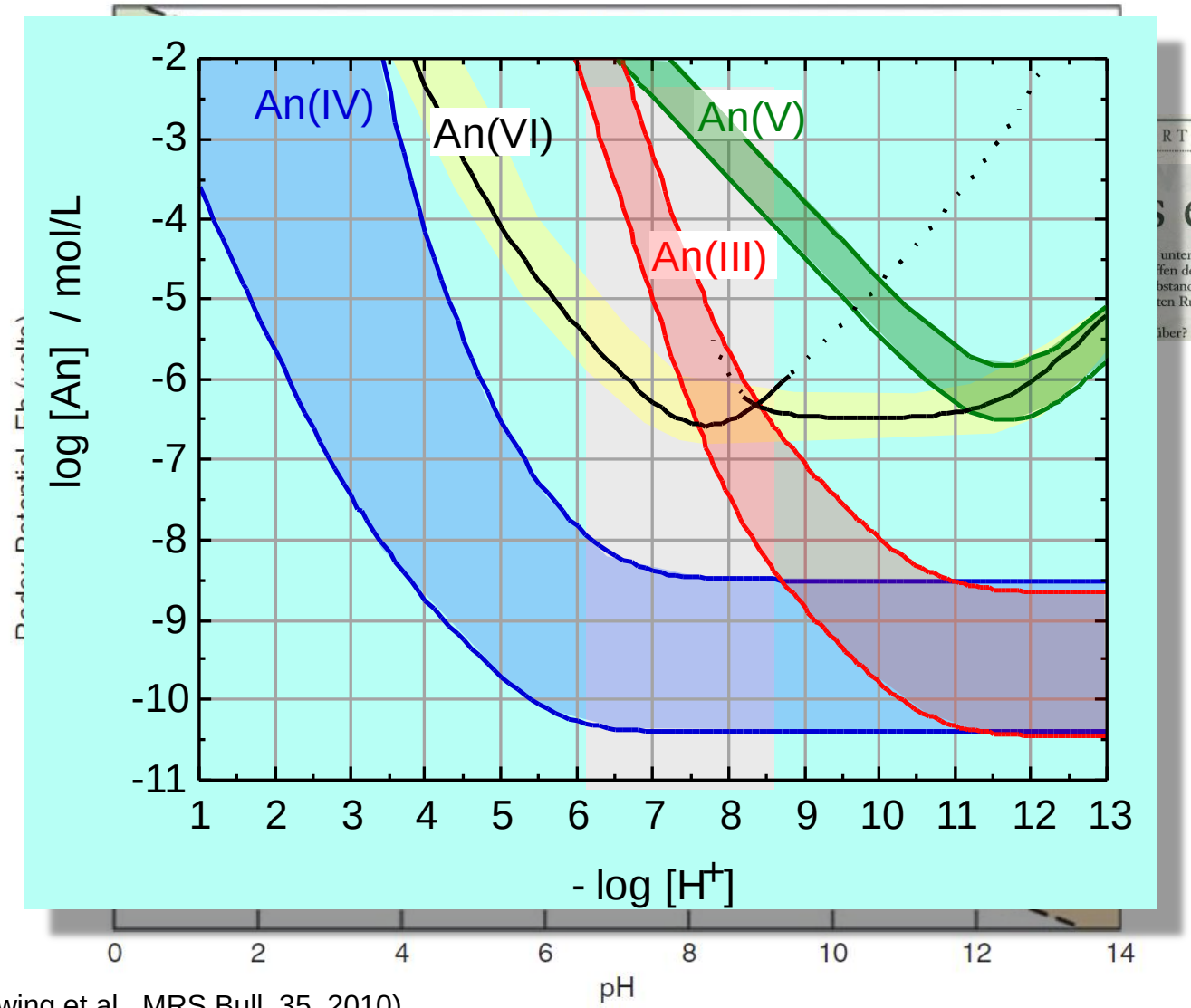
Das elementare Böse

Plutonium hat unter den Grundstoffen der Chemie mit Abstand den schlechtesten Ruf. Was weiß man überhaupt darüber?

The element of evil

Fellhauer, Altmaier, Fanghänel, 2010

Redox chemistry of Plutonium



(Ewing et al., MRS Bull. 35, 2010)

The element of evil

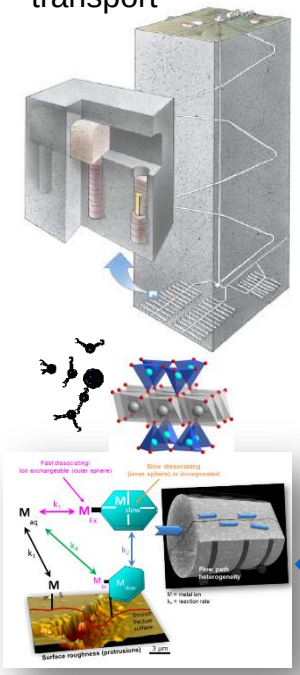
Fellhauer, Altmaier, Fanghänel, 2010

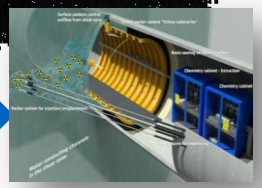
Host rock: crystalline

Impact of glacial meltwater onto the erosion of bentonite barriers in a repository in granite and onto the clay nanoparticle borne radionuclide transport

Host rock: clay rock

Diffusion and retention of radionuclides iin clay rock: Impact of Pore water composition (I, pH, Eh, CO_3^{2-}, T…)

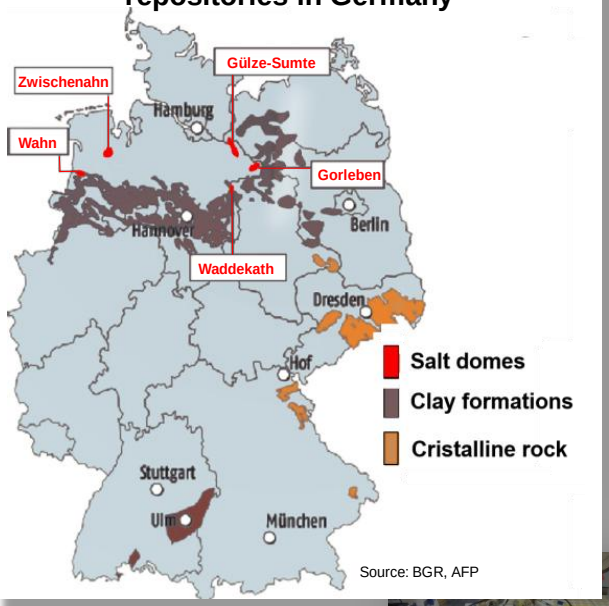




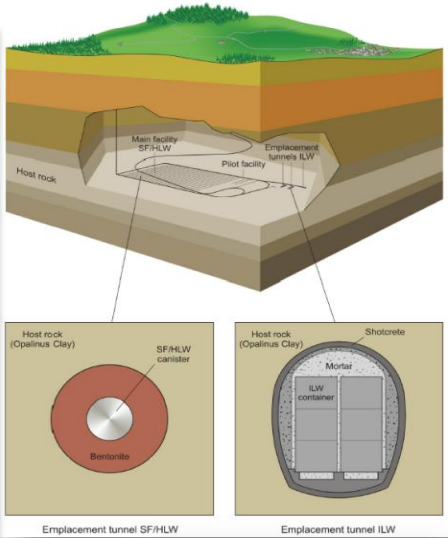
Laboratory

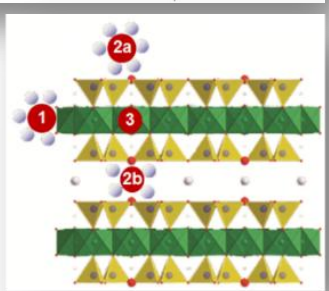
underground rock laboratory

Potential host rocks for nuclear waste repositories in Germany



Source: BGR, AFP





Sorption mechanism of cations on three-layer sheet silicates (i.e. montmorillonite).

KOLLORADOe

BELBaR

Bundesministerium für Wirtschaft und Technologie

US-DOE

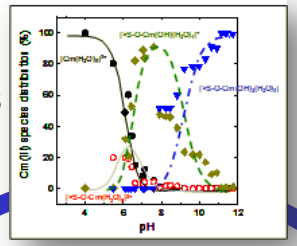
EURATOM

CatClay

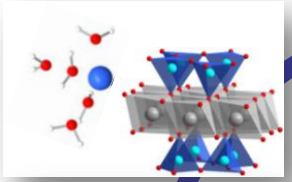
Comprehensive approach: from the nanoscale to the field

Example: Impact of glacial melt water induced erosion of bentonite barriers in a repository in crystalline rock

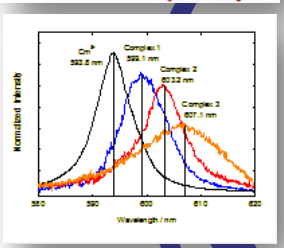
Geochemical speciation, Reaction kinetics



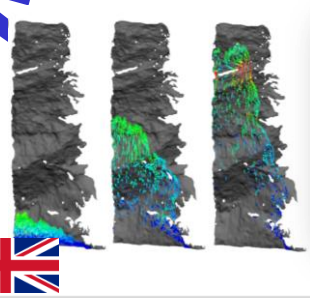
In-situ experiments (laboratory; underground rock laboratories - Grimsel, Äspö)
➔ **upscaling**

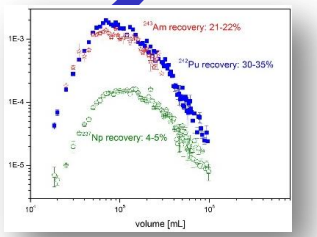


Spectroscopy, Quantum Chemistry



Radionuclide behaviour in the geosphere



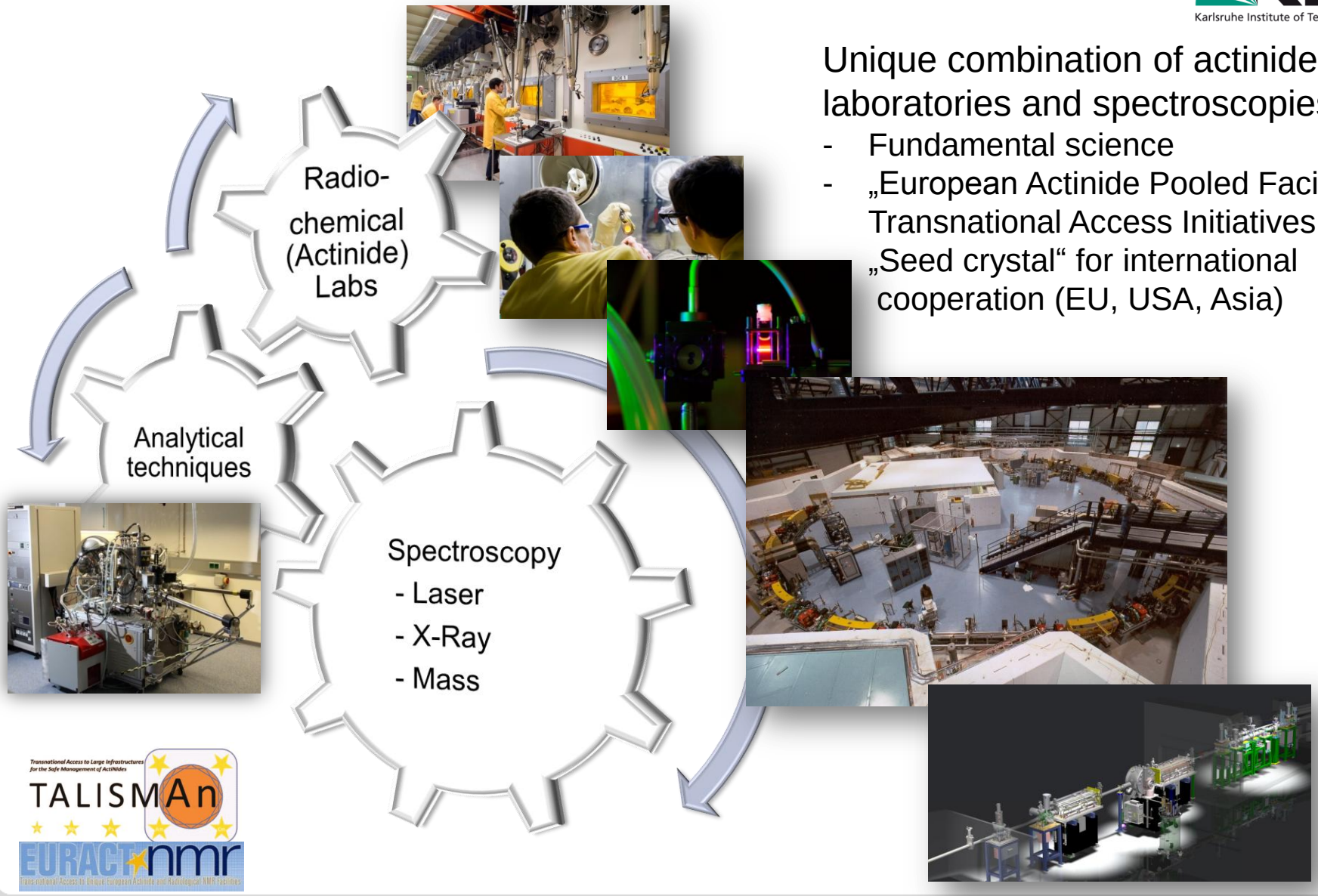


Simulation of radionuclide migration by reactive transport modeling

Bundesministerium für Wirtschaft und Technologie **KOLLORADOe**

EURATOM BELBaR CFM

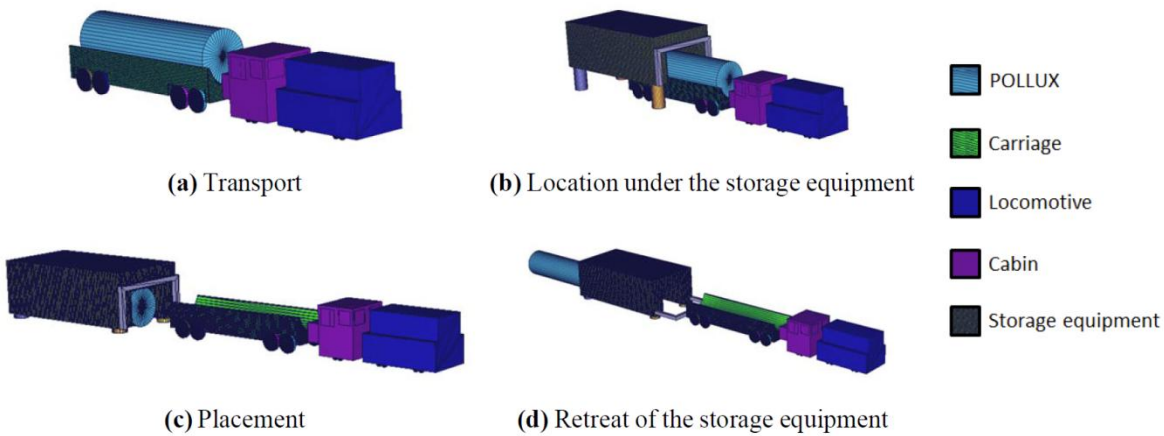
Laboratory platform



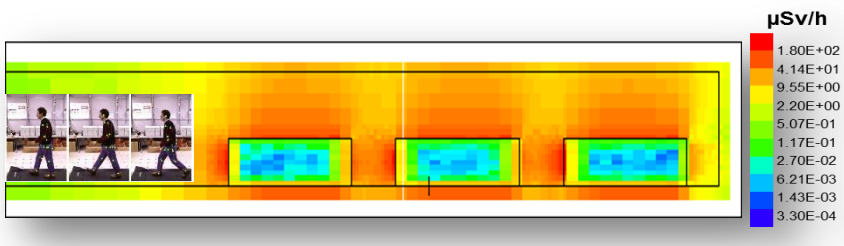
Unique combination of actinide laboratories and spectroscopies

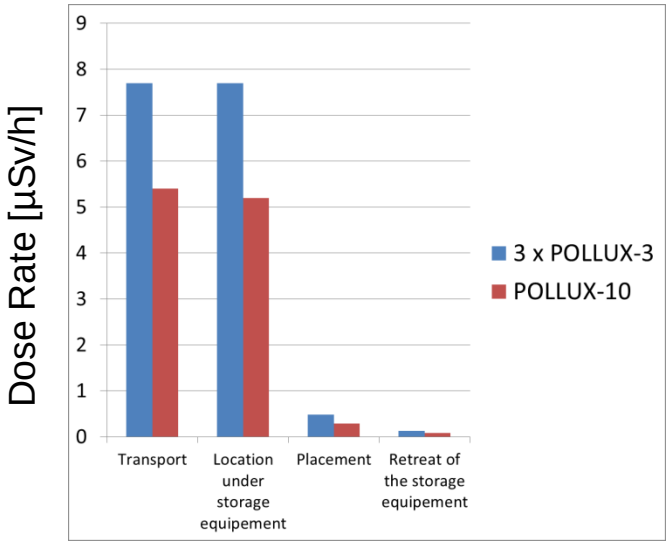
- Fundamental science
- „European Actinide Pooled Facility“ Transnational Access Initiatives „Seed crystal“ for international cooperation (EU, USA, Asia)

Investigation of POLLUX storage/disposal scenarios (rock salt and clay stone)



(a) Transport (b) Location under the storage equipment (c) Placement (d) Retreat of the storage equipment





<u>Comparison - a similar amount of SNF should be disposed:</u>

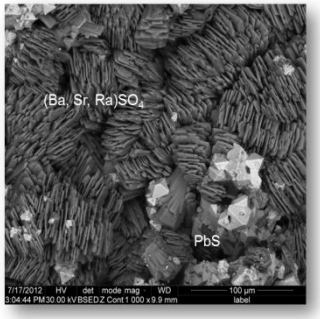
- Germany: 90% UOX + 10% MOX
- POLLUX-10 casks in **rock salt**
- POLLUX-3 casks in **claystone (temperature constraint)**

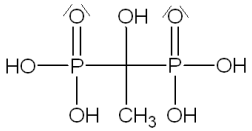
=> 1 POLLUX-10 vs. 3 POLLUX-3

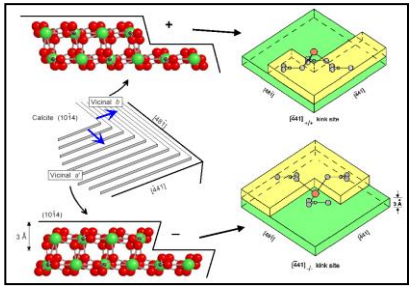
NORM residues from geothermal power plants

Data: Scheiber et al. (2012)

NORM residues C [Bq /g]	Dose rate [heat exchanger] (exemplarily)
up to 1,350 (Ra-226)	12 µSv/h in 1m distance
up to 1,100 (Pb-210)	> 34 µSv/h in 0.1m distance



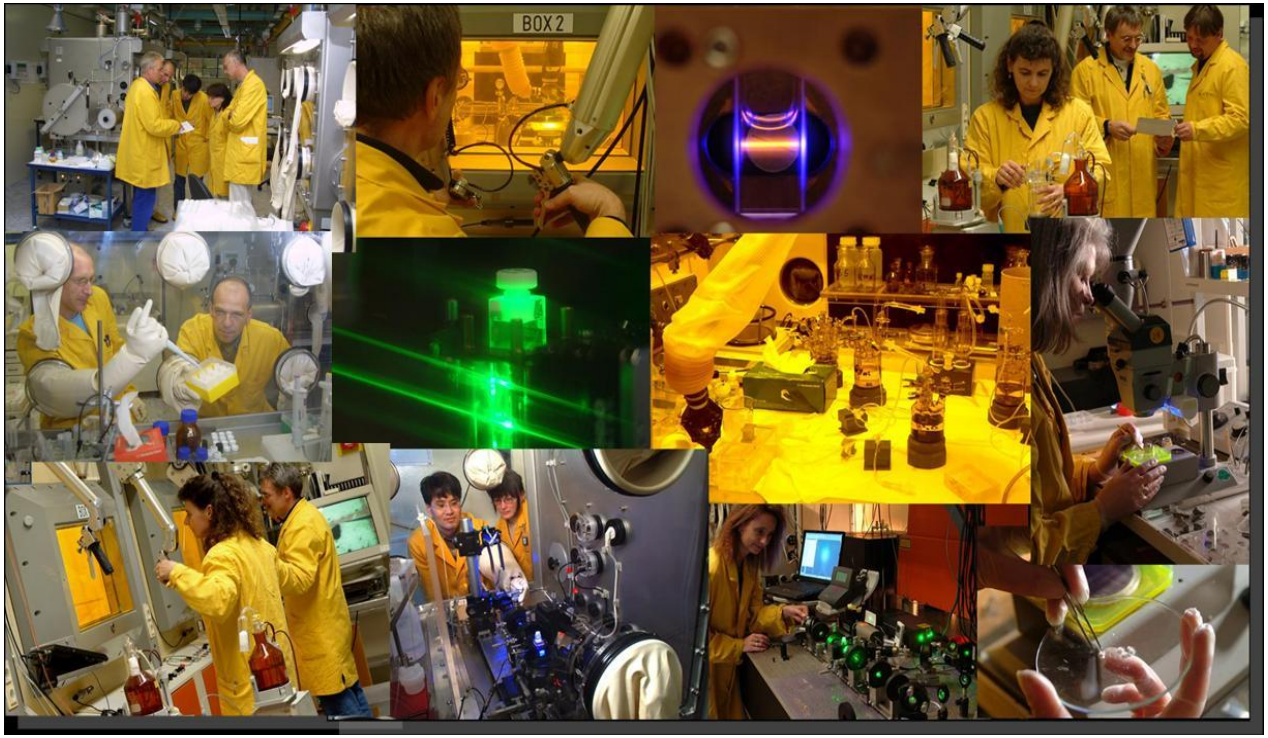




HEDP as crystal growth inhibitor
(**h**ydroxy**e**thylidene- **di**phosphonate, $C_2H_8O_7P_2$):

- Formation of scalings particularly in the cold part of the facility (ΔT 90° C)
- Primarily Ba/Sr-sulfate & PbS
- ^{226}Ra in Ba/Sr-Sulfate, ^{210}Pb PbS.
- **Development of Inhibitor concept to avoid scale formation**

KIT - Institute for Nuclear Waste Disposal (INE)



INE:

- employees: ~ 105
- Radiochemical laboratories
- Hot cell laboratories
- Glove boxes
- „State of the Art“-instrumentation

- Research is embedded in European and International projects and cooperations

- INE is engaged in training and education of students

- Close cooperation with various universities