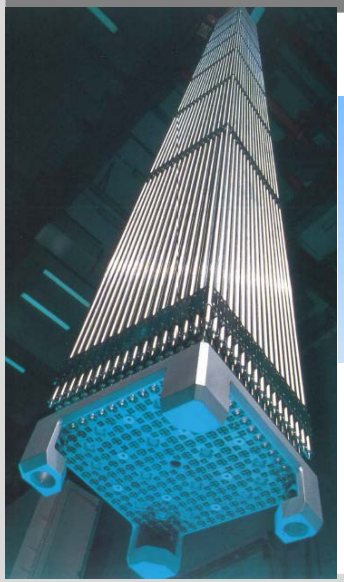
Karlsruhe Institute of Technology

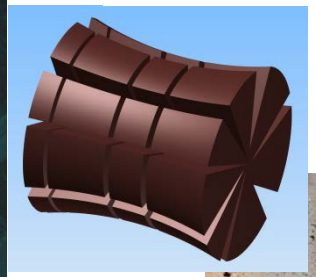
Remaining questions after FIRST-Nuclides

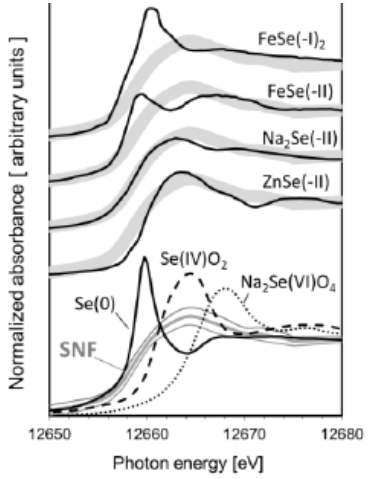
IGD-TP 6th Exchange Forum, November 3-4, 2015, London, UK

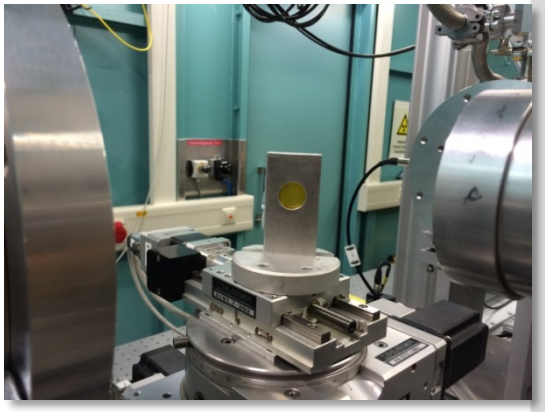
Bernhard Kienzler

INSTITUTE FOR NUCLEAR WASTE DISPOSAL (INE)









CP FIRST-Nuclides

- **Objective**: Quantification the rapid release of radionuclides from high burn-up used fuel after canister failure.
  - Relation of FGR to IRF for ^{129}I, ^{79}Se, ^{135}Cs, for high burn-up / lin. power rate ranges, full set of sample sizes, typical groundwater, aerobic to reducing conditions, quantification (speciation) of ^{14}C, Se
  - Modelling
  - Training, Education, Dissemination
- **Partners**: 10
- **Associated Groups**: 13
- **End-Users**: 6
- **Funding**: total: 4.74 Mio. €, EC contribution: 2.49 Mio. €
- **Duration**: 01.01.2012 – 31.12.2014

Outcome of CP FIRST-Nuclides

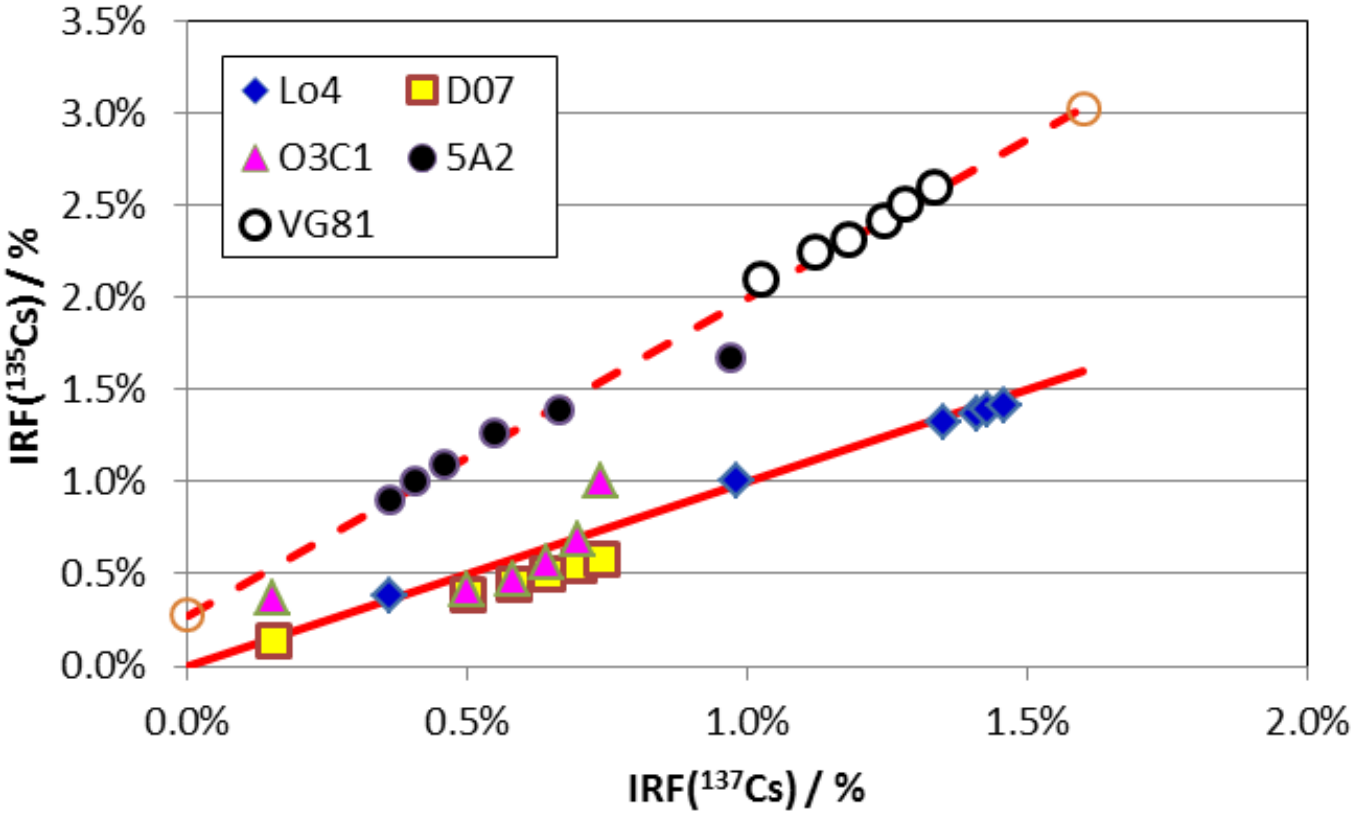
- Results from 12 different types of high burn-up UO2 LWR SNF.
  - IRFs at ~45 different time steps
  - up to 3 sample preparations
  - up to 20 isotopes.
- Huge investments for setup and implement the experiments / analytical tools and instruments
- Clearance by the utilities to publish the spent fuel data.

Contradictions in FIRST-Nuclides outcome

- Contradiction of measured ^{79}Se release with speciation of Se in the UO_2 matrix.
- Resolving open questions for some nuclides, such as of ^{135}Cs showing a higher release in comparison to ^{137}Cs.

➔ Keep the experimental set-up, samples and materials, technques and experienced staff.

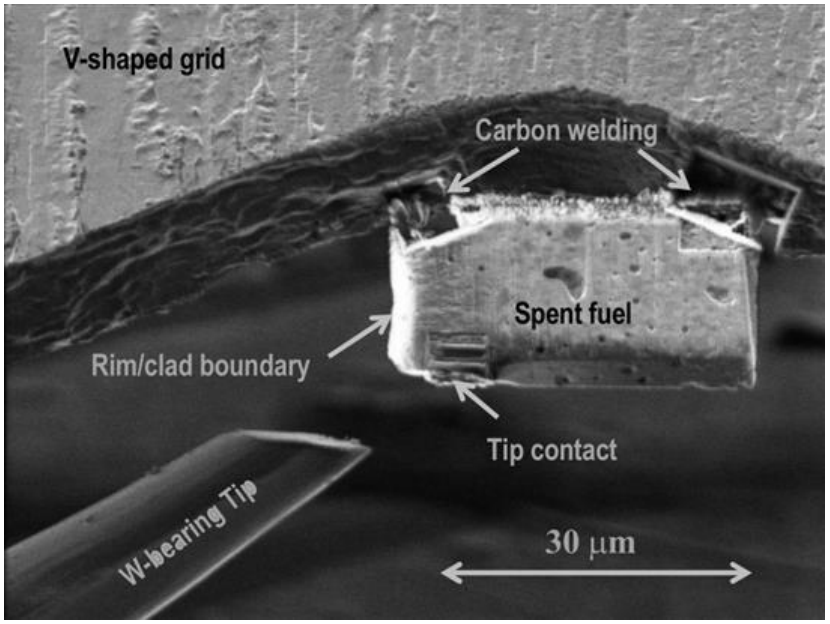
Cs release



PWR SNF: Higher ^{135}Cs release fraction in comparison to ^{137}Cs (by a factor of 1.7). The reason cannot be explained.

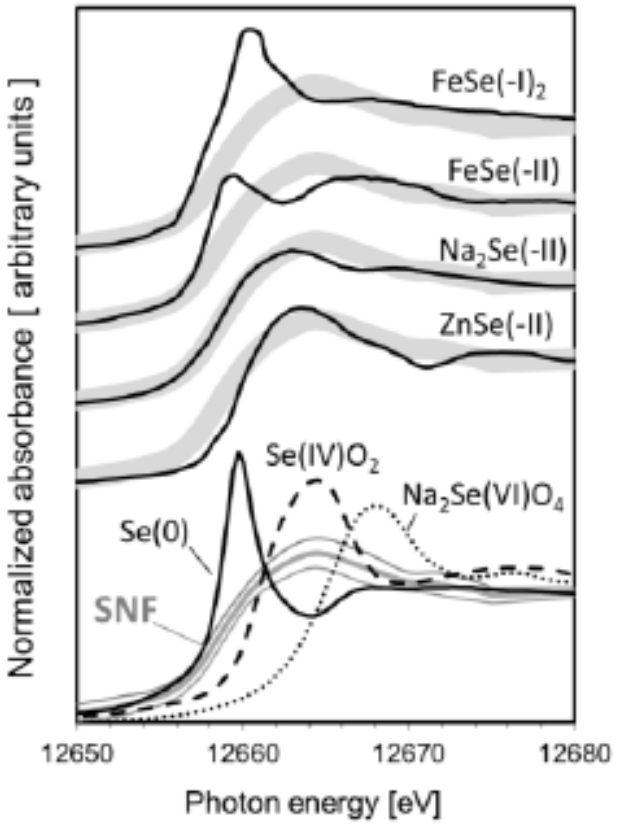
Element speciation in used fuel: Se

STUDSVIK & PSI: X-ray spectroscopic investigations on the chemical state of ^{79}Se in high-burnup UO_2 spent fuel



FIB Sample ("RIM") of Oskarshamn used fuel for XANES measurements

Swiss light source



E. Curti, et al., *Journal of Nuclear Materials, V*ol. 453, pp. 98-106, 2014.

Speciation of Se

PSI, Studsvik

Result: XANES measurements show either a

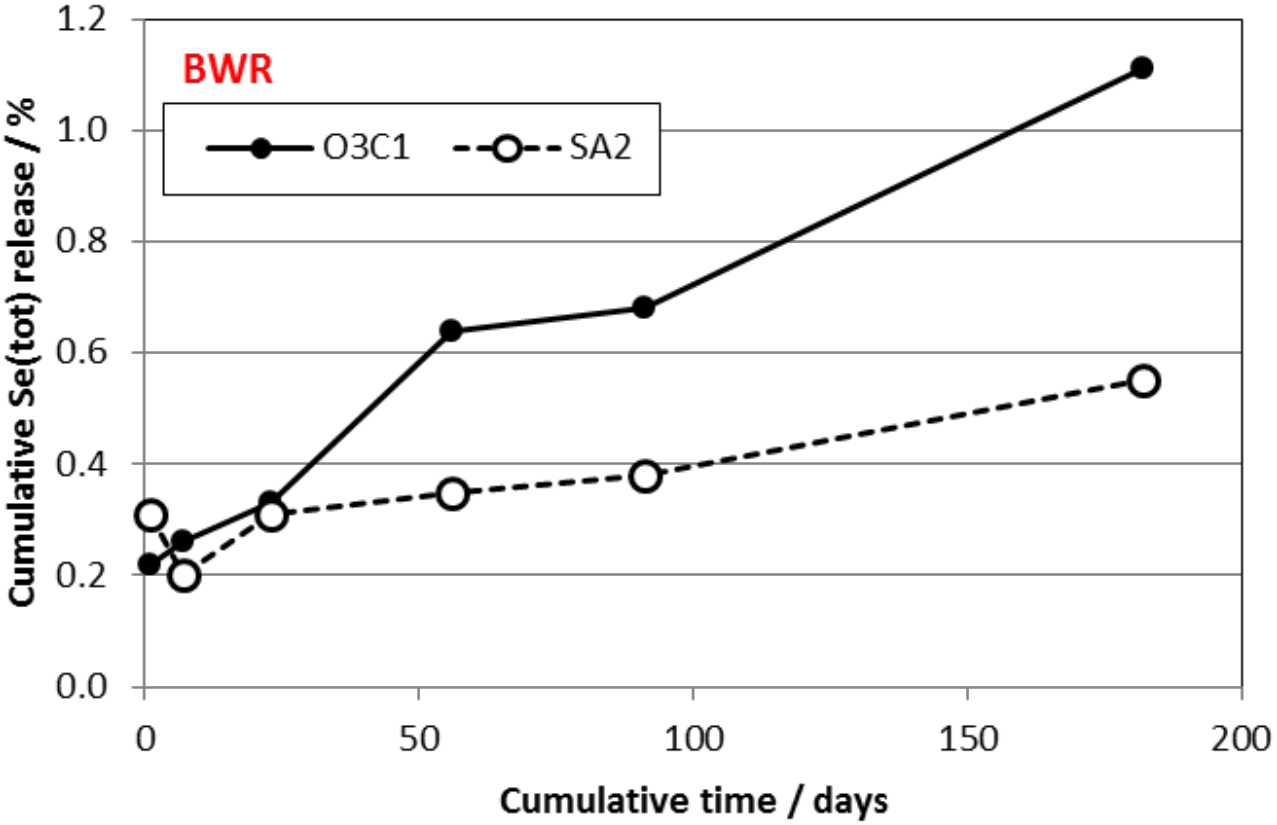
- mixture Se(0) and Se(IV) or
- pure Se(-II)

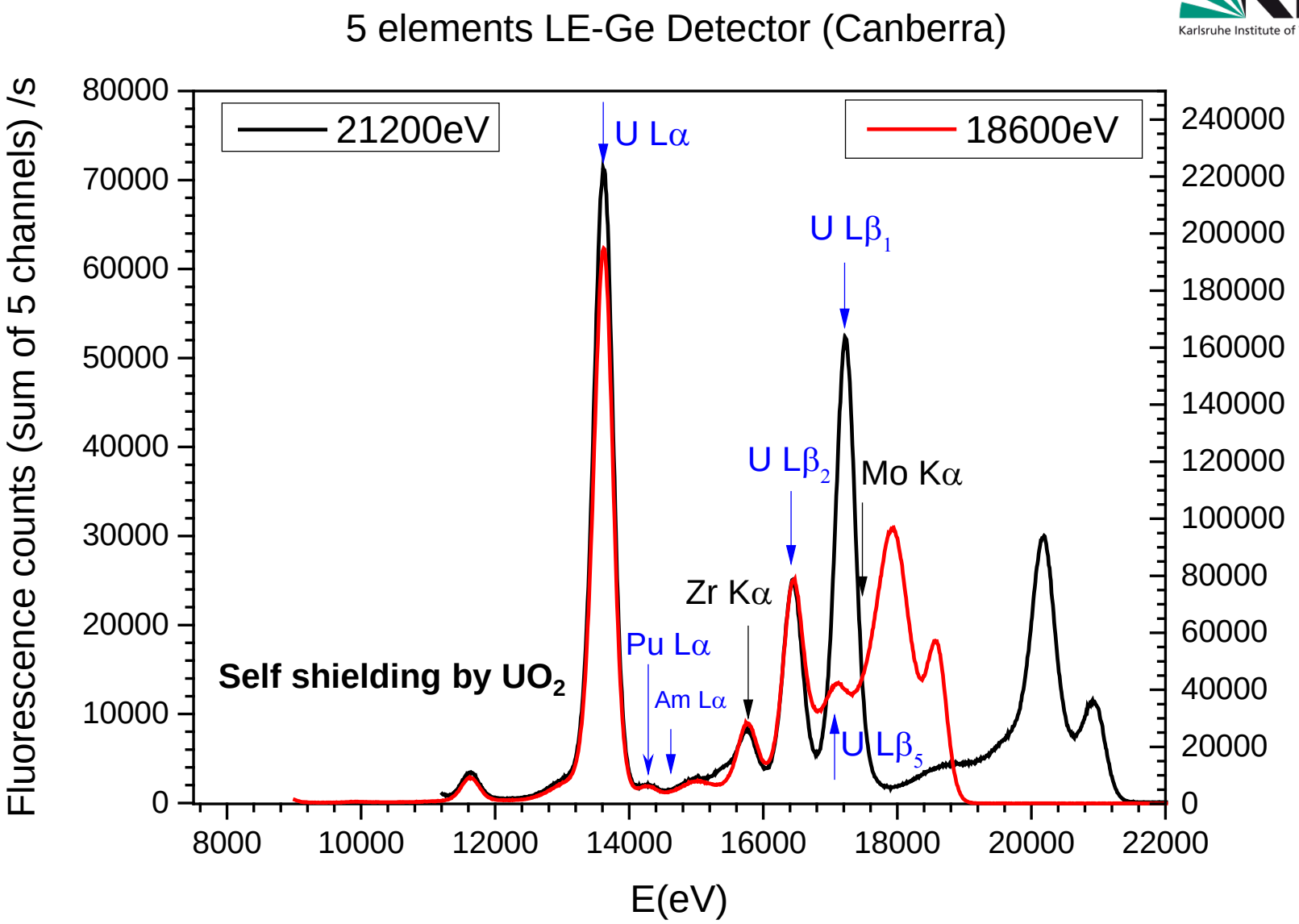
Combined crystallography and ab initio calculations indicate that Se (including ^{79}Se) is present in UO_2 SNF in a homogeneous chemical form:

➔ dispersed Se(-II) replacing oxygen sites in the UO_2 lattice.

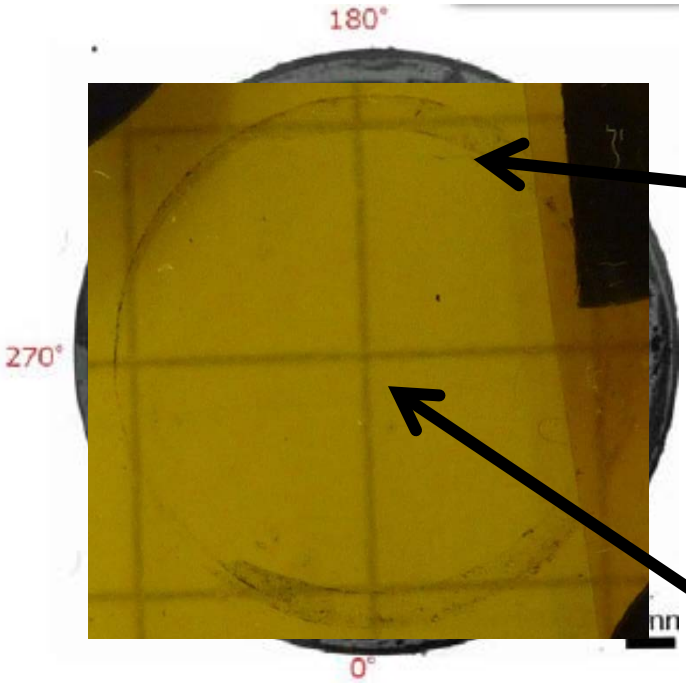
Curti E. et al. (2014) Selenium redox speciation and coordination in high-burnup UO_2 fuel: Consequences for the release of ^{79}Se in a deep underground repository. J. Nucl. Mat., DOI: 10.1016/j.jnucmat.2014.07.003.

IRF(Se_{tot}) release as function of time




5 elements LE-Ge Detector (Canberra)
21200eV
18600eV
U Lα
U Lβ1
U Lβ2
Mo Kα
Zr Kα
Pu Lα
Am Lα
U Lβ5
Self shielding by UO2
Fluorescence counts (sum of 5 channels) /s
E(eV)
0
10000
20000
30000
40000
50000
60000
70000
80000
0
20000
40000
60000
80000
100000
120000
140000
160000
180000
200000
220000
240000
8000
10000
12000
14000
16000
18000
20000
22000

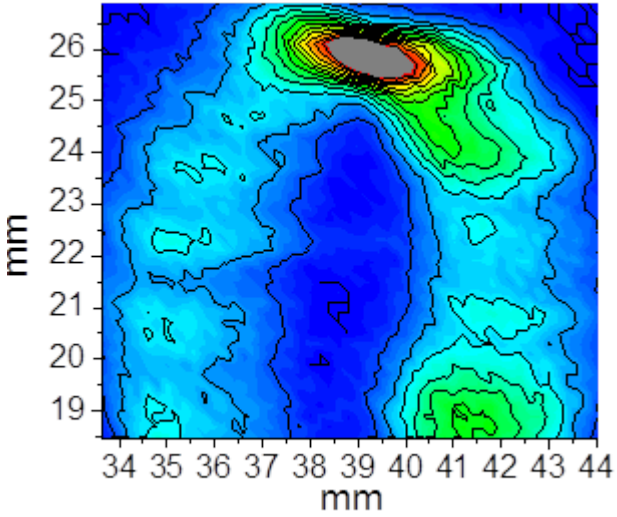
U M4 fluorescence mapping @ INE beamline (ANKA)



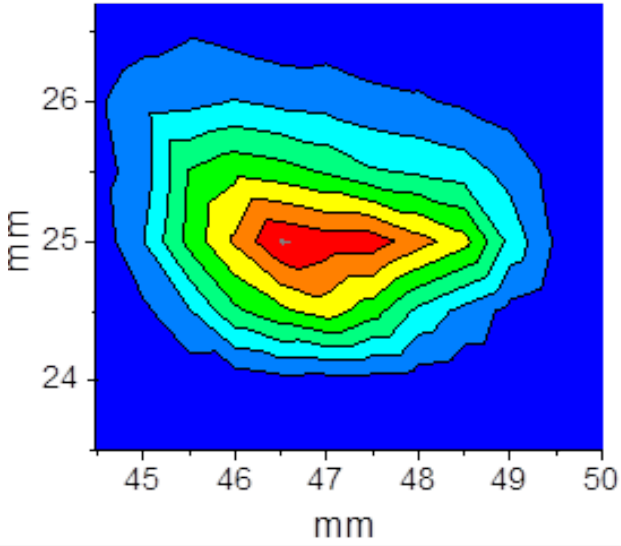
Print of a fuel pellet onto plastic

Rim

Core

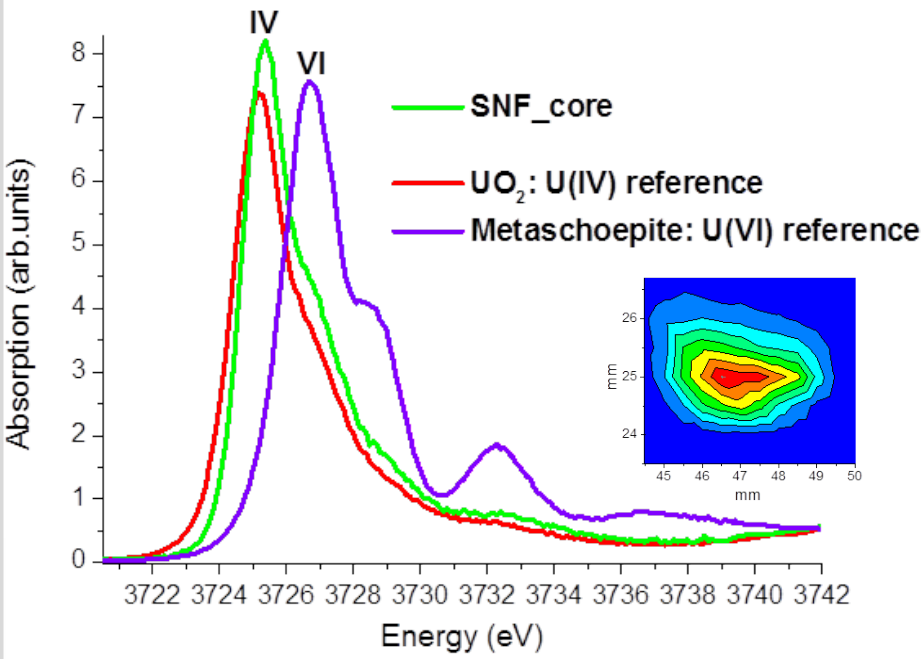


U M_{β} fluorescence maps



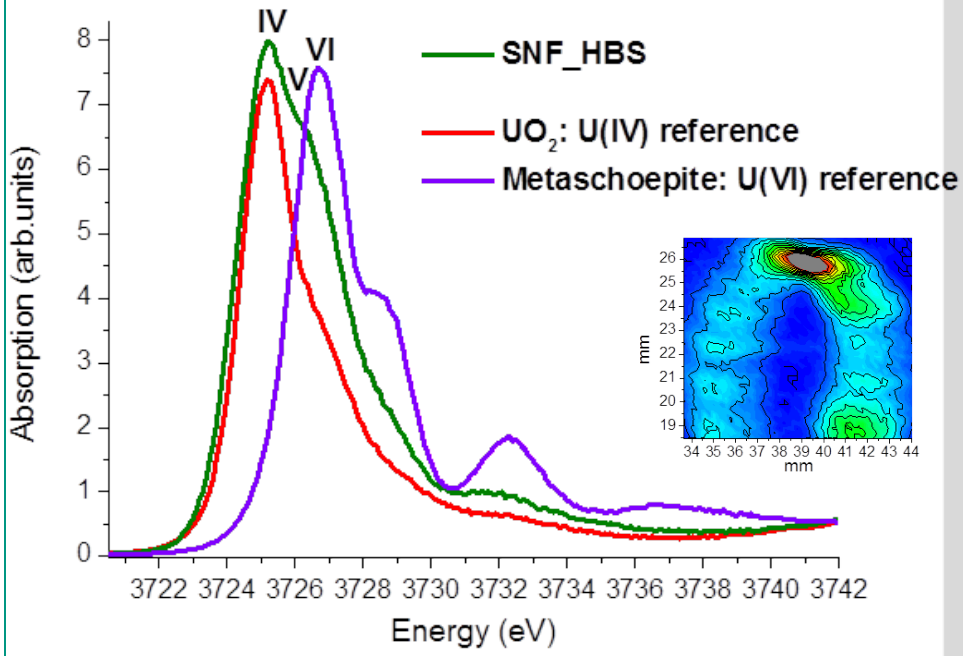
U M4 edge high energy resolution XANES

Core



- **Core:** U(IV) dominates,
  minor U(VI) contribution

Rim

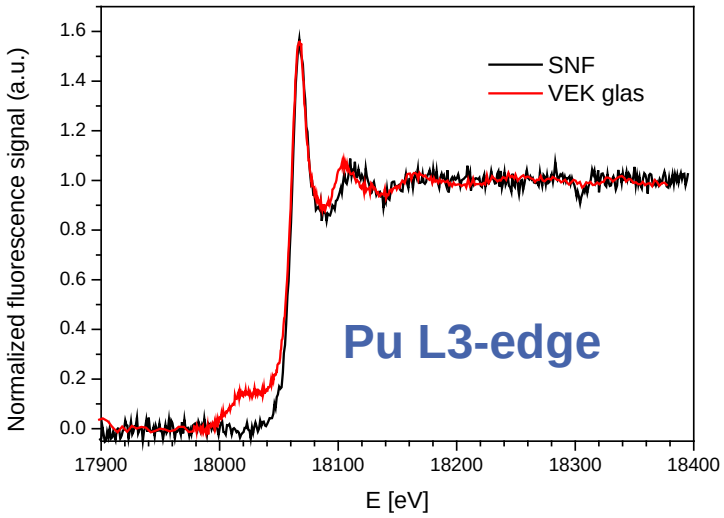


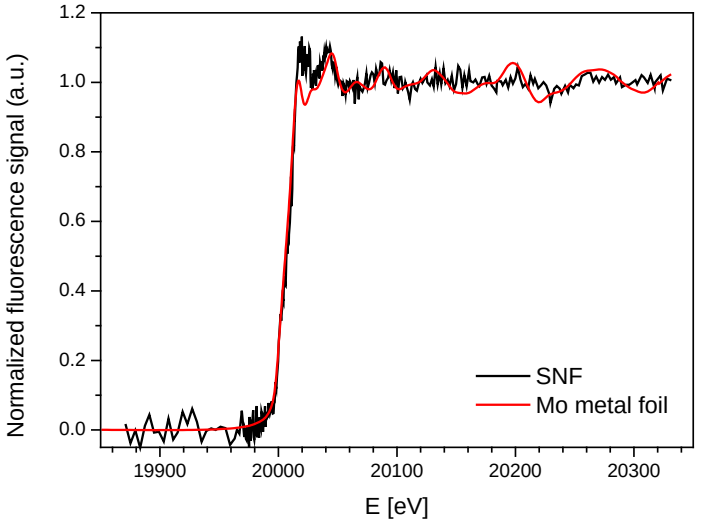
- **Rim:** U(IV) dominates,
  U(V) contribution likely

T. Vitova et al. 2015 in preparation

SNF Results: XANES measurements

Measurements up to now: **U L3**, **Pu L3**, **Am L3**, **Mo K**, **Tc K** - **Np L3** and **Cm L3** not measurable!





Plutonium:

- Pu present as Pu(IV)
- pre-edge feature?

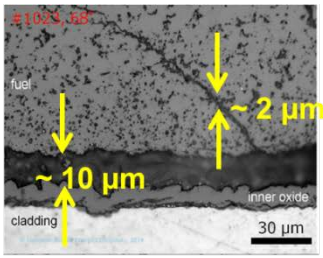
Molybdenum:

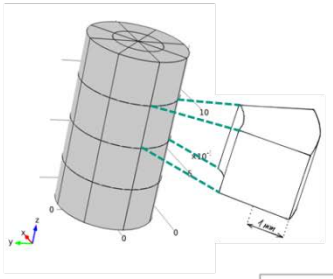
- Mo seems to be present in metallic Mo(0) state!

What‘s about MOX?

Modelling

Water saturation of a pellet and release function





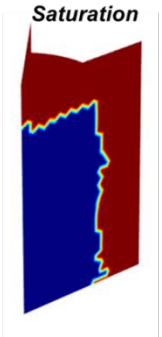
Variably-saturated flow model

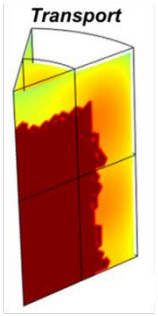
➔ Richards equation (Bear and Cheng, 2010), extension of Darcy's Law to unsaturated conditions.

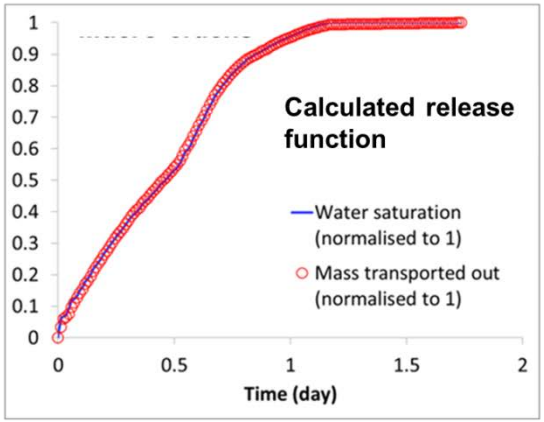
water retention / relative permeability:

➔ van Genuchten model (van Genuchten, 1980)

M. Pękala et al., AMPHOS21, Spain



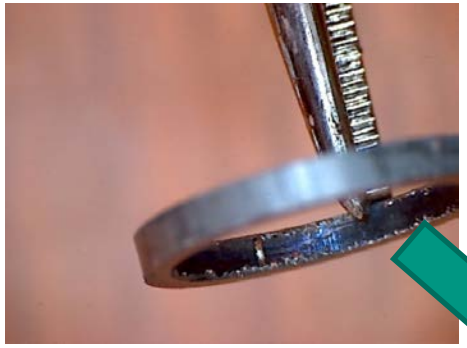


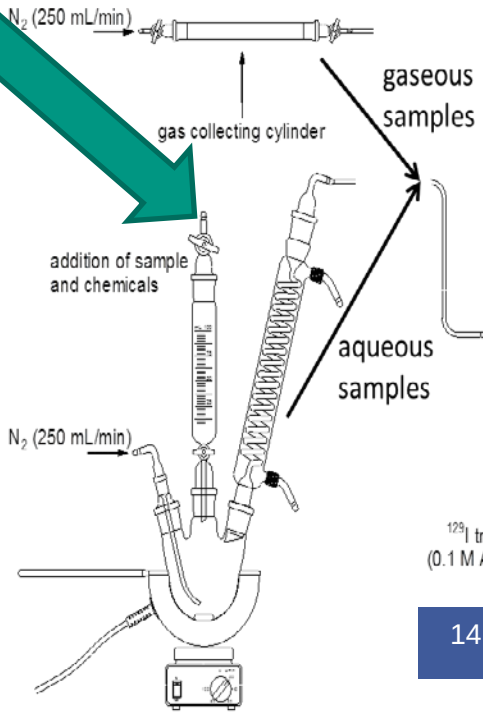


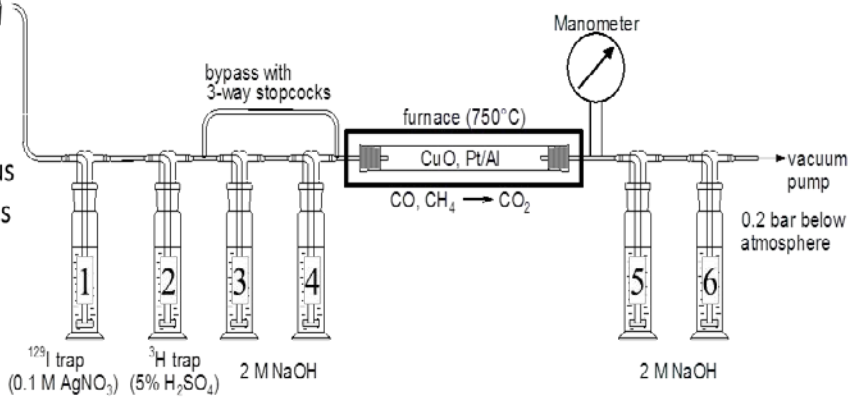
Relation to „Reactor Codes“

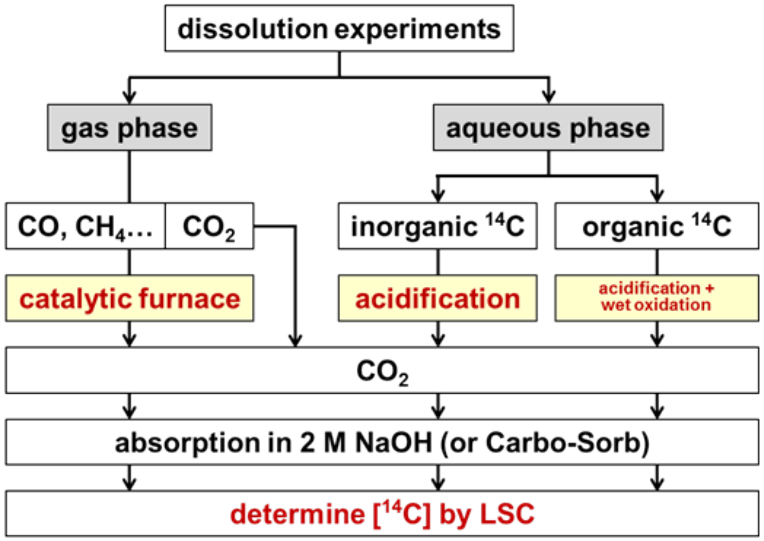
pending

Design for C-14 extraction and C-14 speciation









^{14}C extraction system

Summary and Conclusions

- … research heavily relies on the state-of-the-art hot laboratories, and highly encourages … to maintain hot laboratory capabilities and plan for future renewal.

Pellet: ~10^{10} Bq
γ Dose rate: ~4 Gy/h
β Dose rate: ~120 Gy/h
α Dose rate: ~700 Gy/h (surface)

Continuation

- Improved statistics for the IRF of additional fission products, especially under reducing conditions.
- One single leachant. **Competitive reactions** (e.g. Br^-)**?**
- **FGR** during leaching tests.
- In depth investigations of low concentrated but very relevant isotopes FPs ^{79}**Se** and **Pd**, activation products ^{36}**Cl** and ^{14}C.
- Clear correlations of the IRF with nuclear power plant operational parameters (power rates, temperatures, FGR).
- Improved basis for delineating the IRF from long-term radionuclide release.
- Data for additional types of samples including **MOX** fuel and doped fuels and dependence of IRF on the type and quantity of **dopants**)
- Modelling approaches closer related to reactor codes

KIT's contribution to a Spent Fuel project:

- MOX, PWR (1200 days), burn-up of 38 GWd/tHM
  Natural UO_2 enriched by 3.2% Pu
- Dissolution based radionuclide release from MOX
  - Reducing conditions
  - Sufficient experimental duration ➔ IRF and matrix dissolution
- Analysis of gas phase (fission gas release)
- Analysis of the leachates
  - Fission products: ^{90}Sr, ^{99}Tc, ^{129}I, ^{137}Cs,
  - Activation product ^{14}C
  - Actinides: ^{238}U, $^{239/240}Pu$, ^{241}Pu, ^{241}Am, ^{237}Np, (Reprocessed U: ^{236}U)
- ^{14}C analytics/speciation
- Efforts towards Se and Cl analytics.

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Thank you for your attention