Studsvik

Leaching of doped irradiated fuel under H_2 conditions

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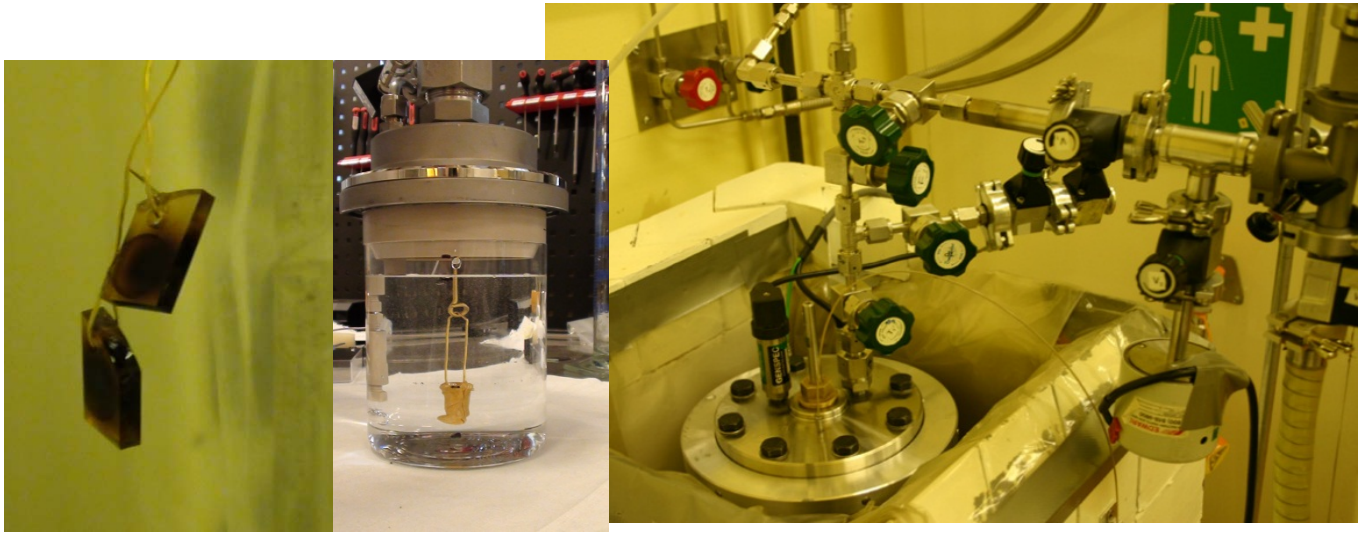
Outline

- Studsvik capabilities
- Background: Leaching of doped irradiated fuel in aerated conditions
- Suggested studies: Leaching of doped irradiated fuel under H_2 conditions

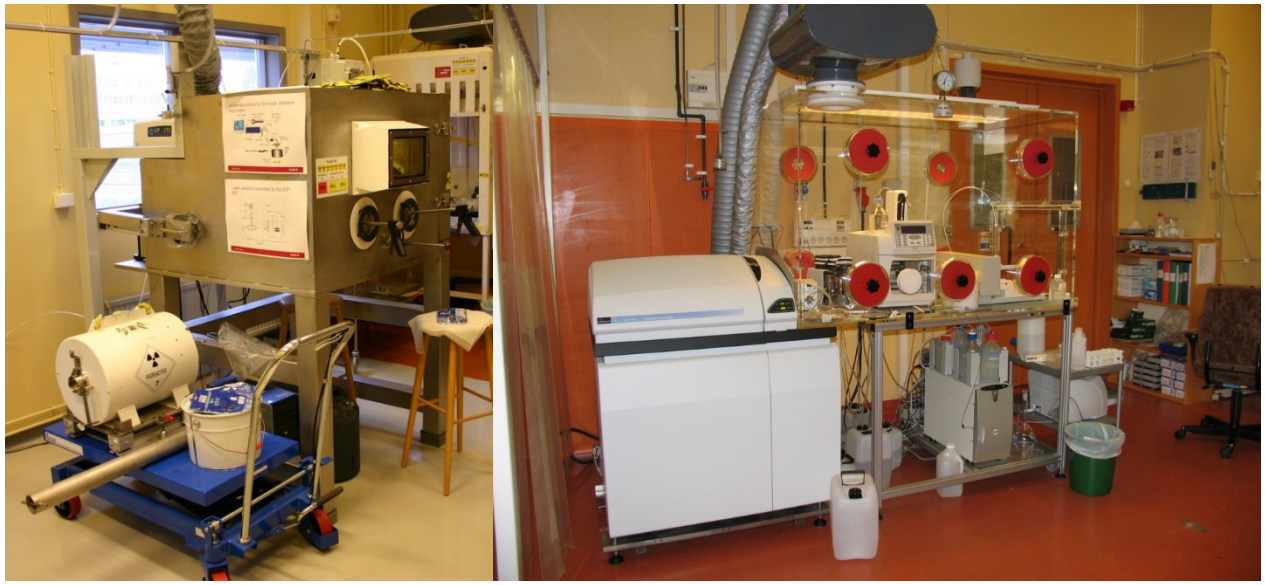
Studsvik Capabilities for deep repository studies

- Clean Hot Cell environment
- Spent fuel autoclaves
- Radiochemistry laboratory; Gloveboxes, Fuel mill



Studsvik Capabilities for deep repository studies

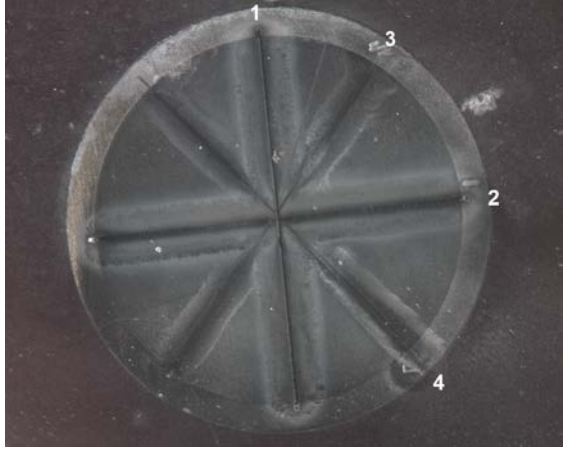
- Clean Hot Cell environment
- Spent fuel autoclaves
- Chemical analysis instruments, such as ICP-MS, for highly radioactive solutions and Gas-MS

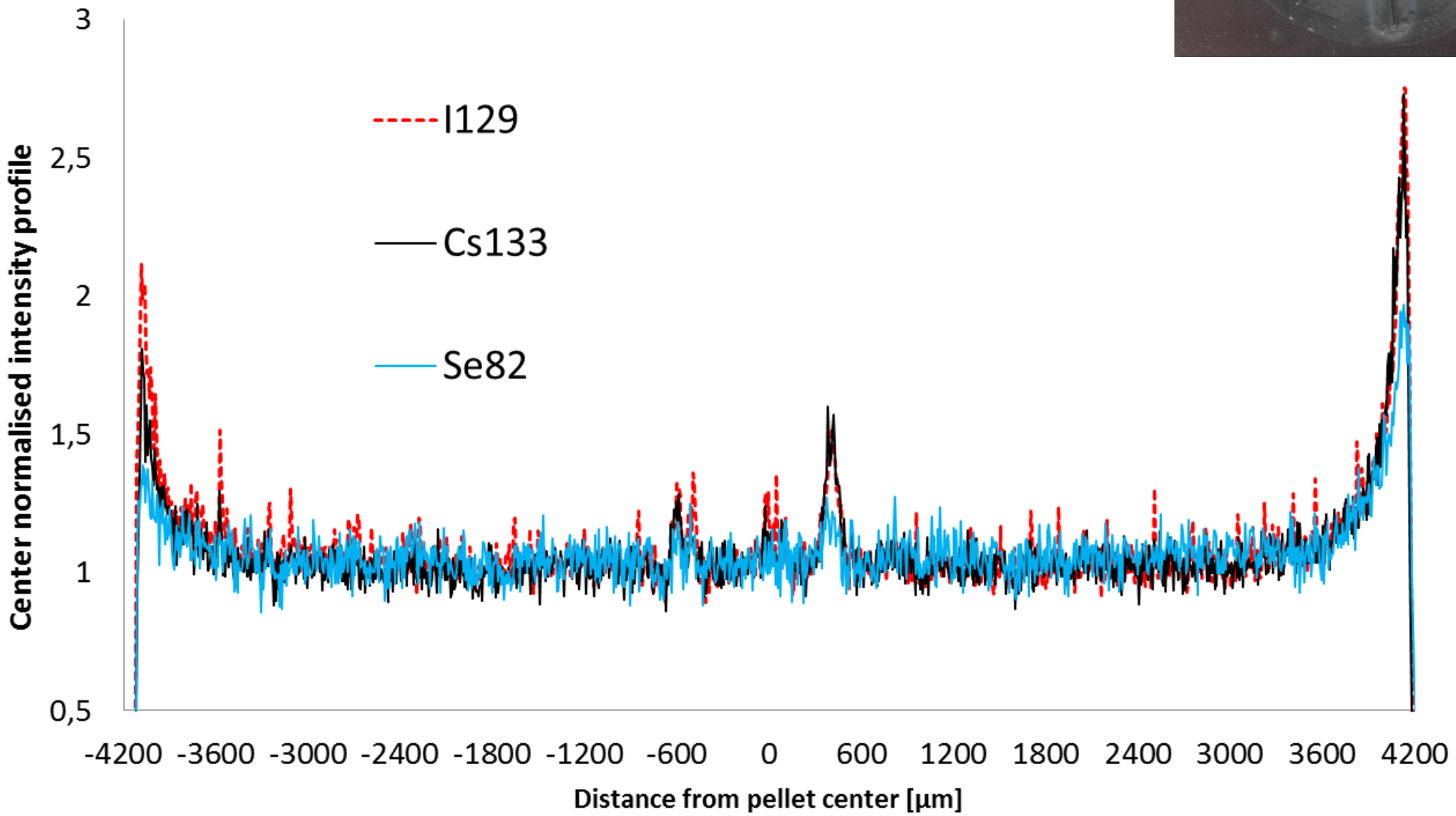


Studsvik Capabilities for deep repository studies

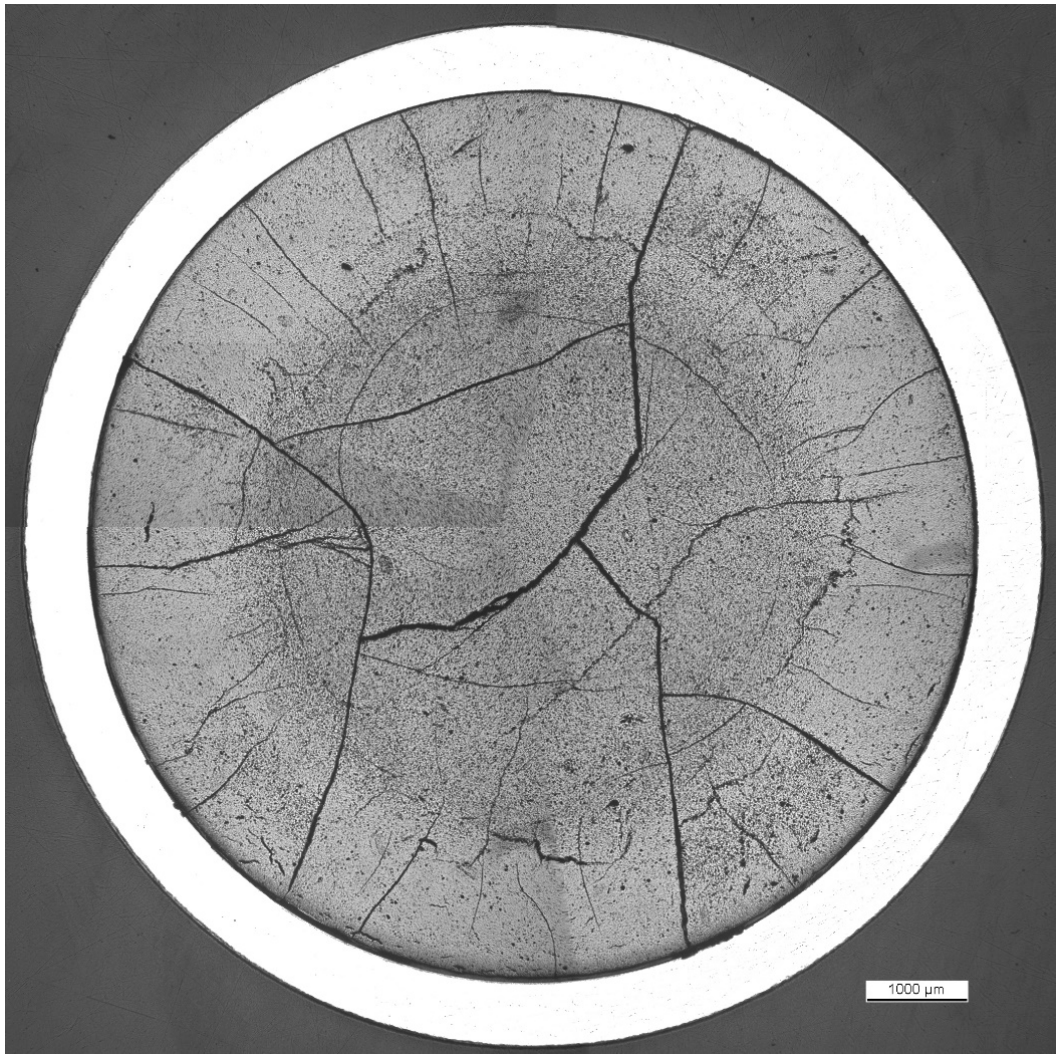
- Clean Hot Cell environment
- Spent fuel autoclaves
- Chemical analysis instruments, such as ICP-MS, for highly radioactive solutions
- Spent fuel characterisation techniques such as:
  - Laser ablation
  - SEM/WDS
  - FIB (TEM-sample preparation)

Radial I, Cs and Se profiles by LA-ICP-MS

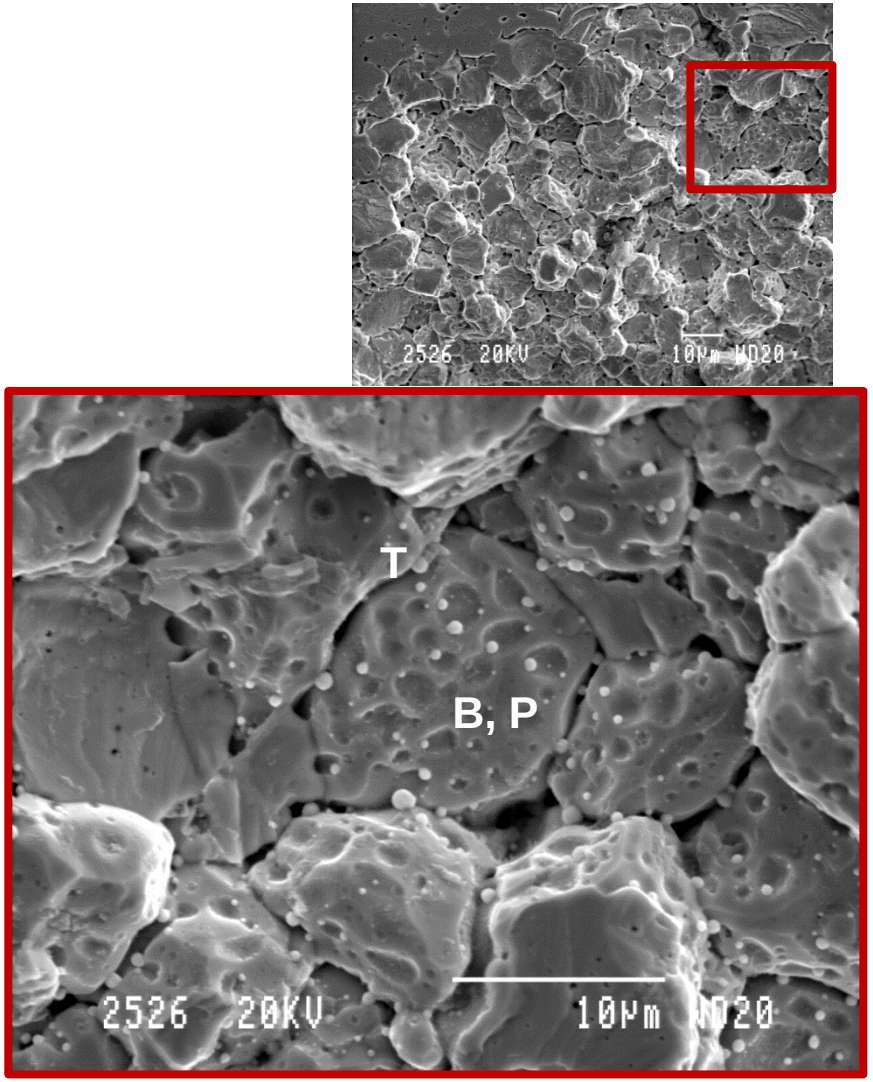




Courtesy of the Studsvik Cladding Integrity Project II (SCIP II)



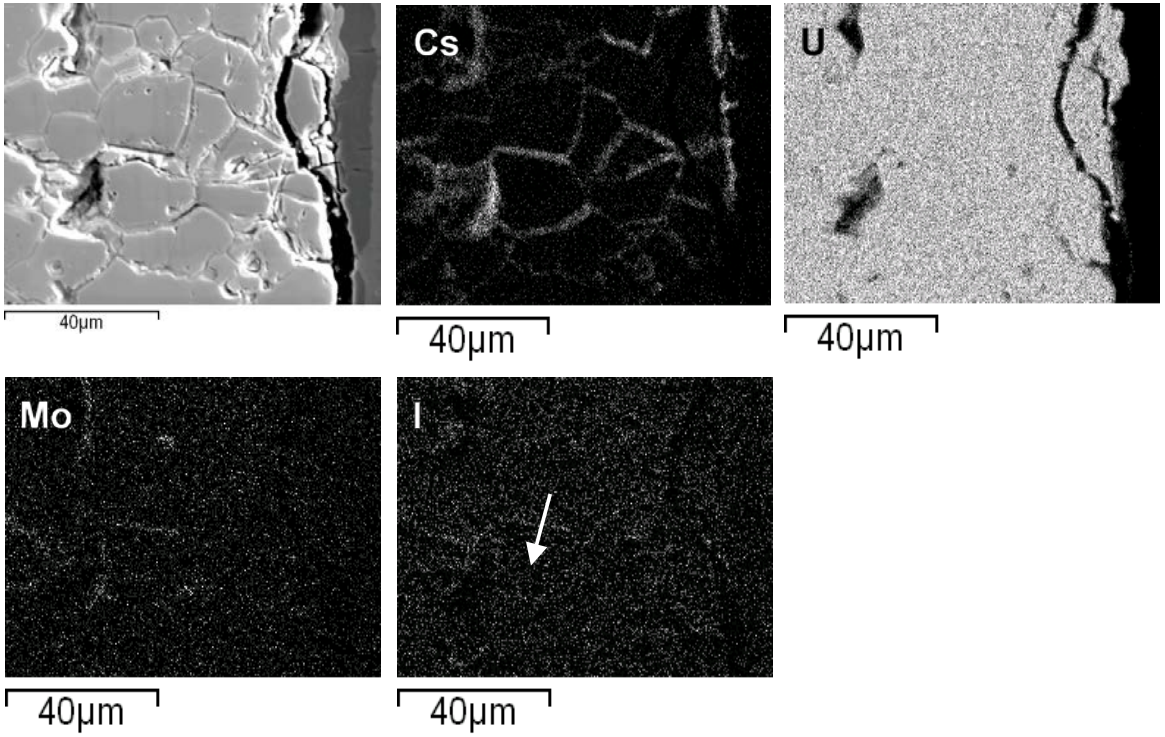
Cross section light optical micrograph of light water reactor UO_2 fuel irradiated at 40 kW/m.

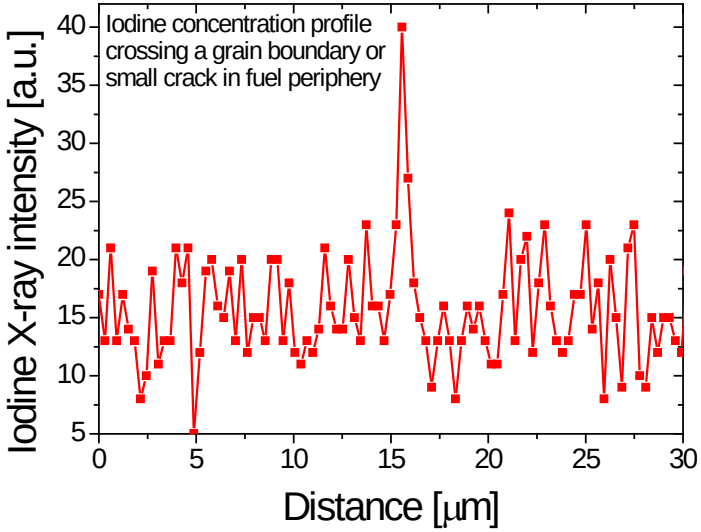


Scanning electron fractograph of light water reactor UO_2 fuel showing intragranular bubbles (B) and grain edge tunnels (T) and metallic precipitates (P).

SEM-WDS data

- I, Cs and Mo present in the grain boundaries/cracks close to the cladding





Leaching of doped fuel

- Trends in reactor operation:
  - Power uprates
  - Longer fuel cycles
  - Higher burn-up
- Additives/dopants:
  - Gd oxide
    - Burnable poison
    - Reduced thermal conductivity
  - Al/Cr oxides
    - Increase grain size
    - Reduce FGR
    - Reduced hyperstochiometry, reduced FP solubility[1]

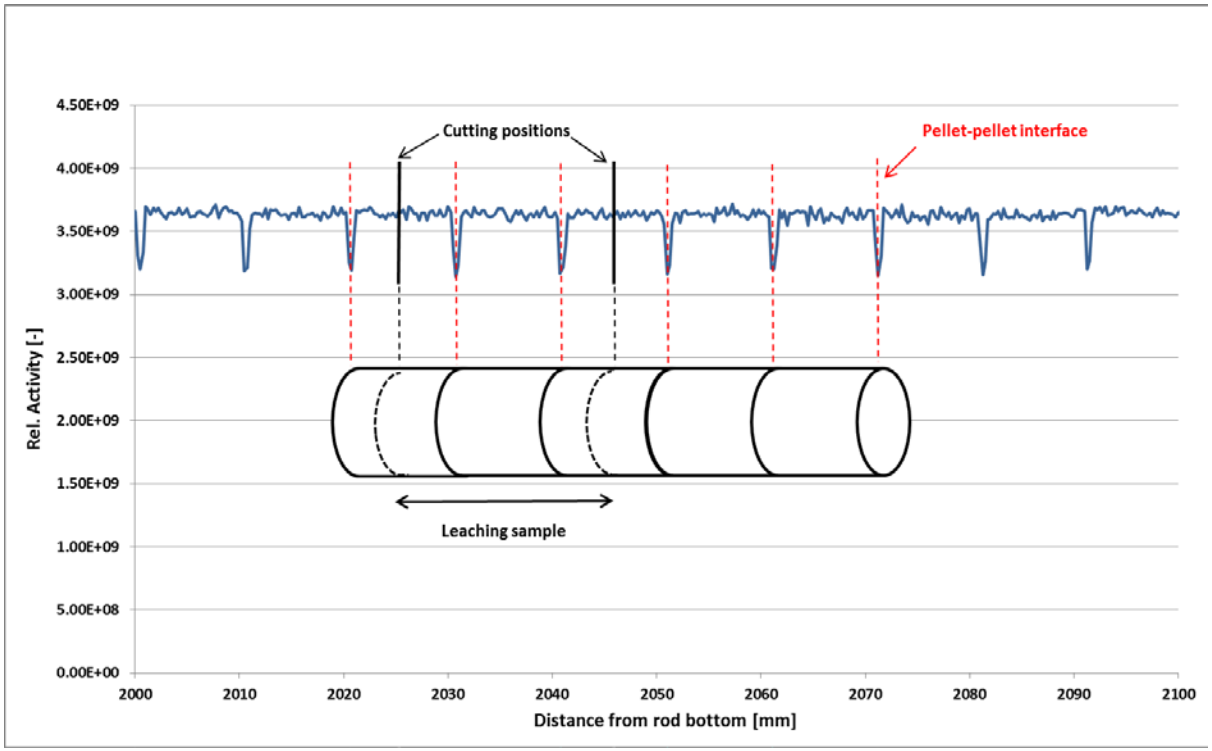
[1] *M. W. D. Cooper et al J. Nucl. Mater., 443 (2013) 236-241*

Leaching of doped fuel in air – sample selection

Sample name	Reactor type	Fuel type	FGR [%]	Calculated BU (rod average) [MWd/kg U]	Sample type
5A2	BWR	Std UO_2	~2.4	57.1	Fragments + cladding
C1	BWR	Al/Cr doped UO_2	~1.4	59.1	Fragments + cladding

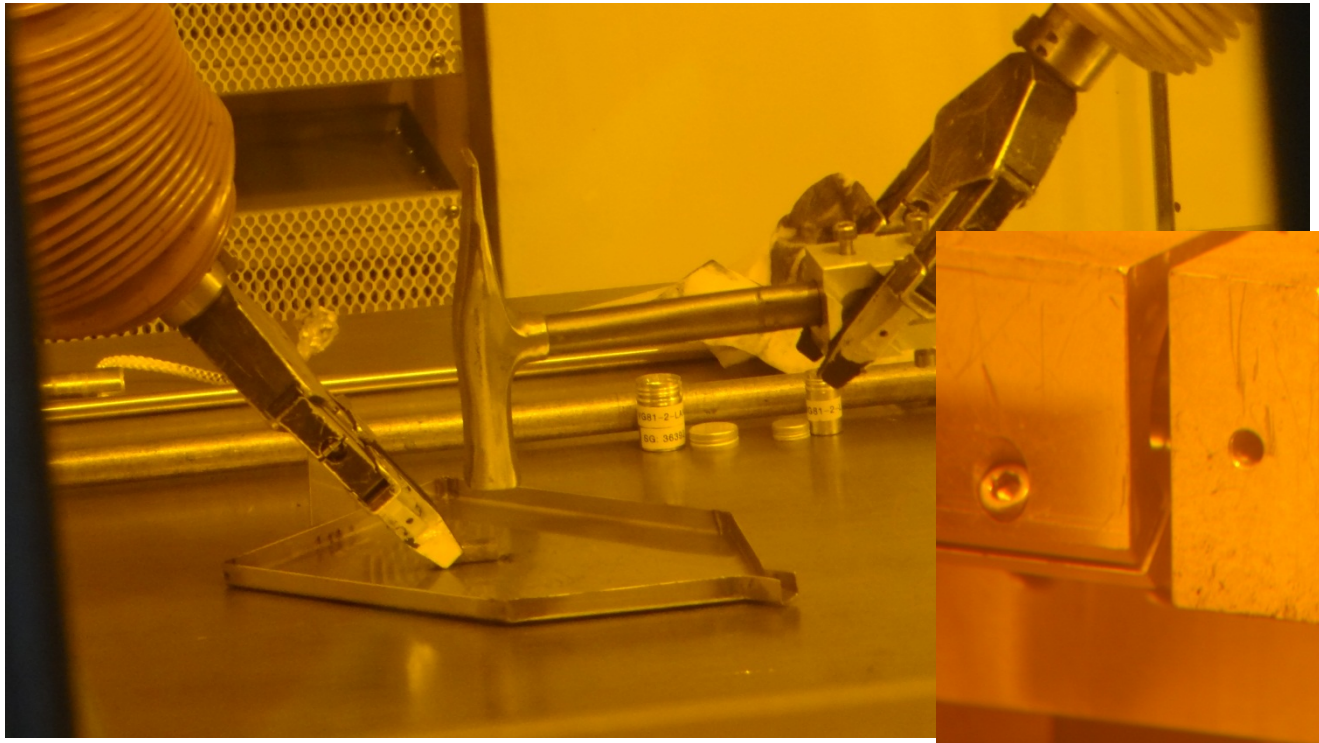
Sample compartments

Fragments + separated cladding



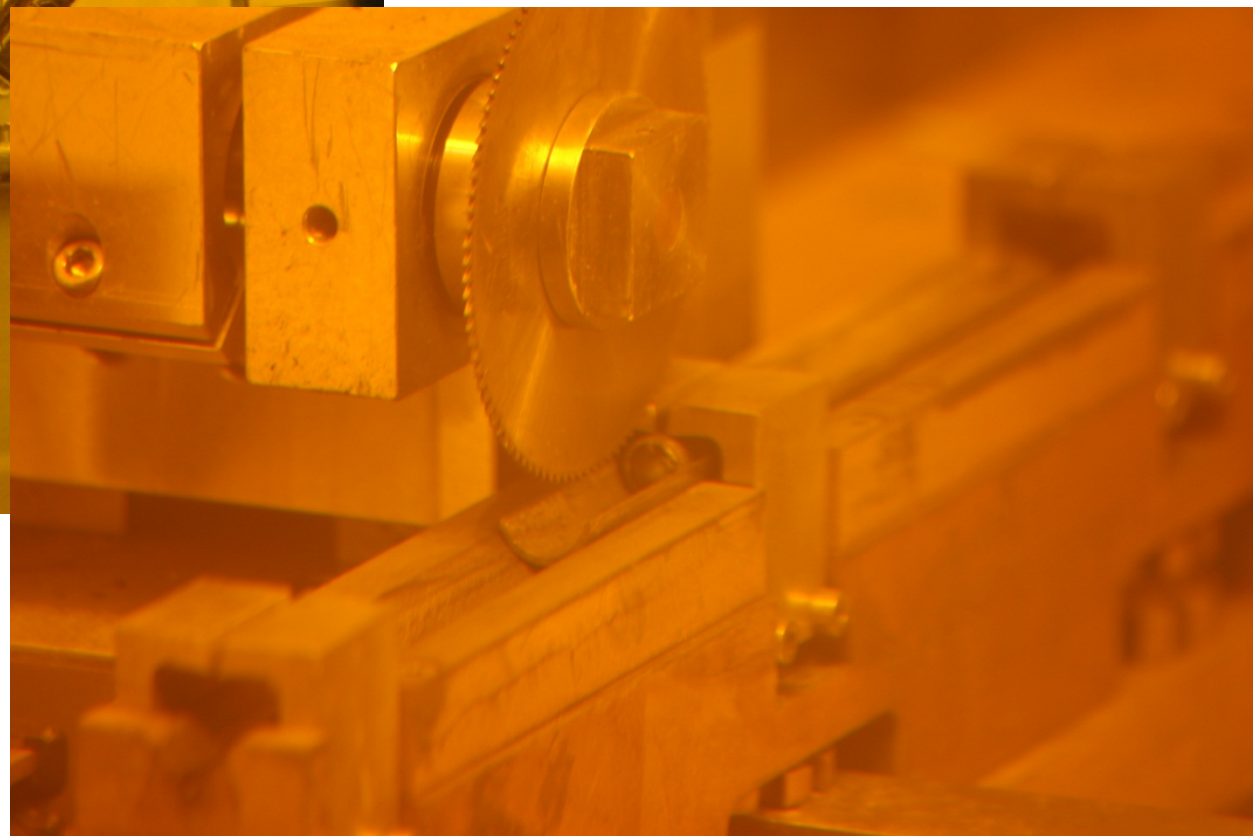
- Gamma scanning
- Cutting
- **Separation of cladding by axial cutting or compressing**
- Weighing

Cladding detachment



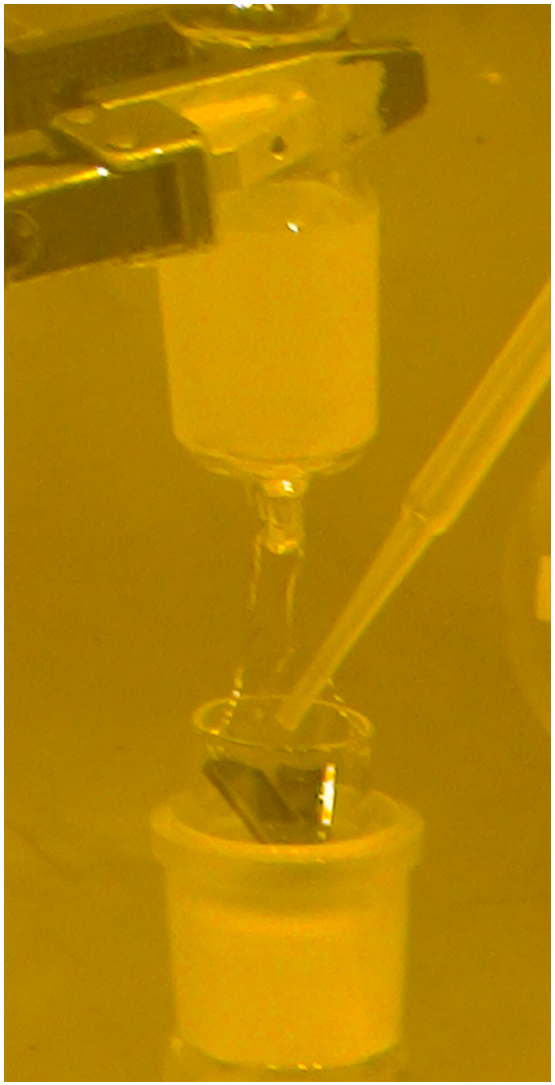
Crushing

Cutting



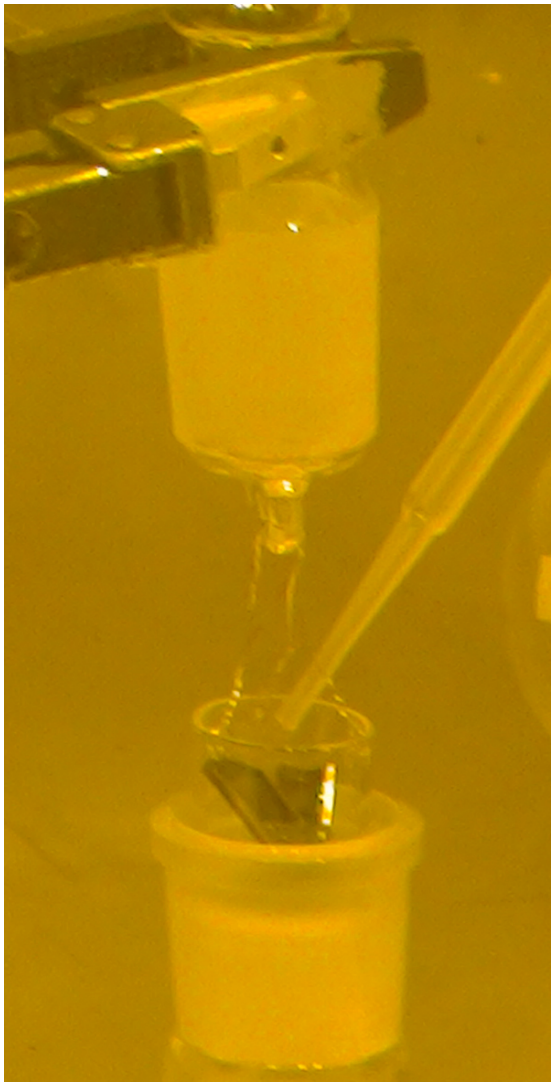
Experimental Set-up

- Performed in Hot Cell dedicated to leaching studies
- Fragments + cladding are placed in glass basket and wetted before immersion.
- Immersion in 200 ml leaching solution (10 mM NaCl + 2 mM $NaHCO_3$).

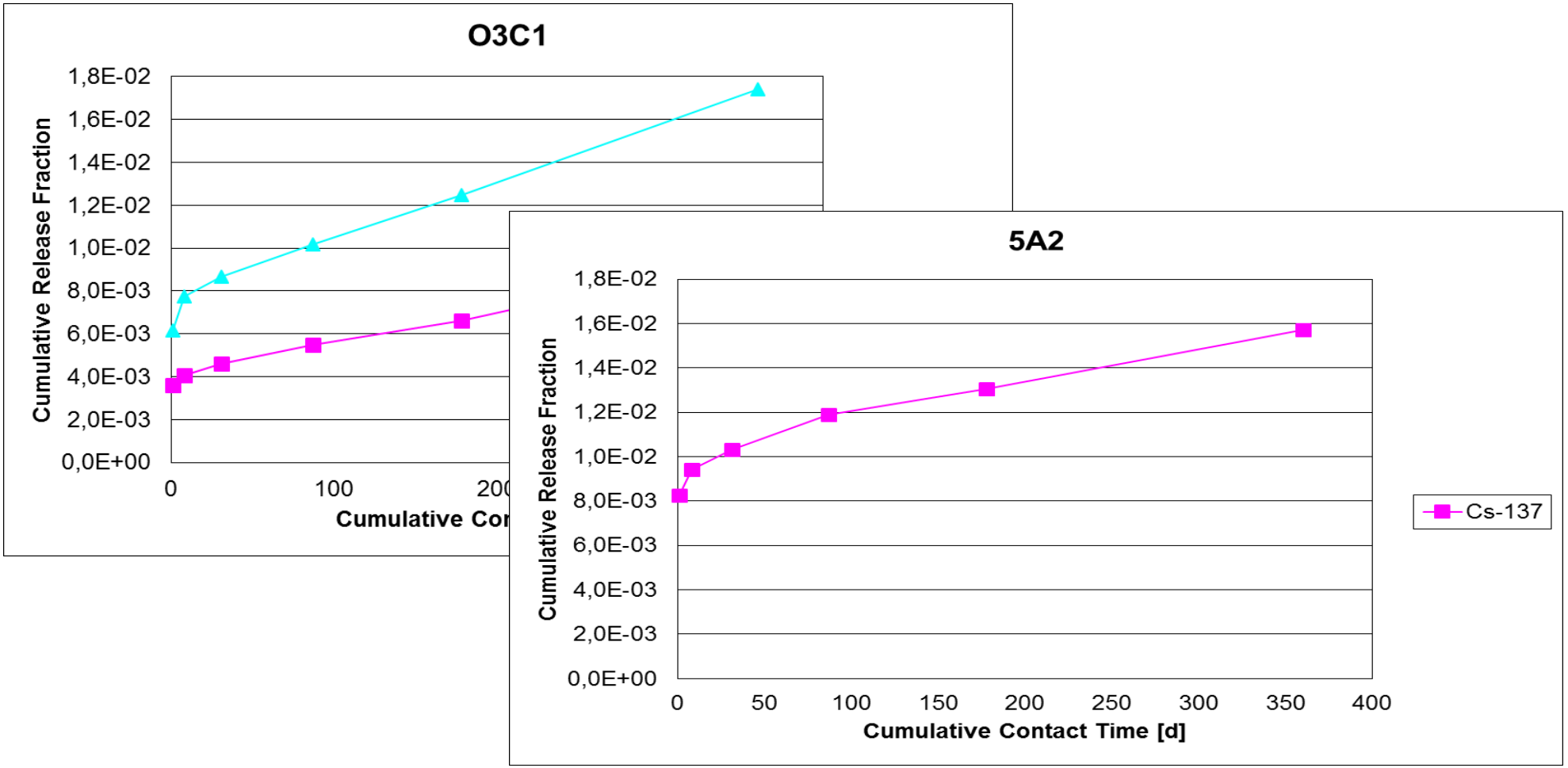


Experimental Set-up

- 1 year leaching
- 6 contact periods/ sampling points
- Fresh leaching solution at each sampling point.
- Analysis by ICP-MS



Cumulative release fractions C1 and 5A2



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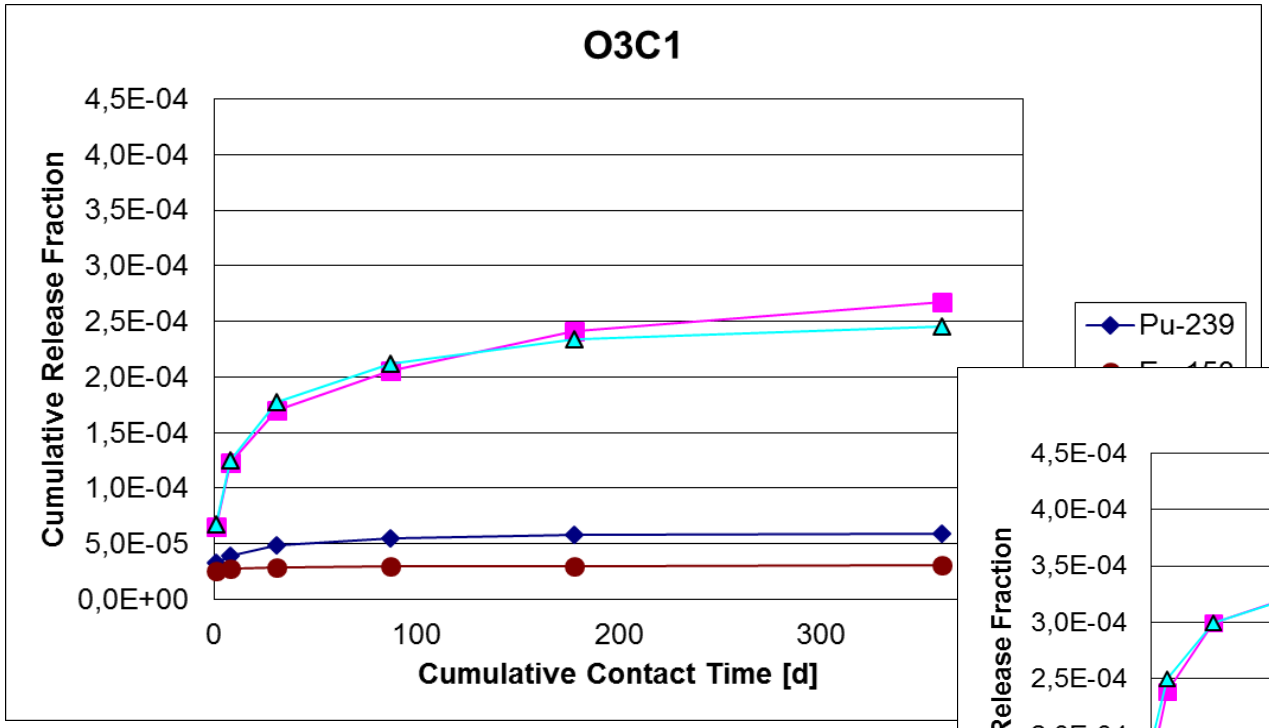
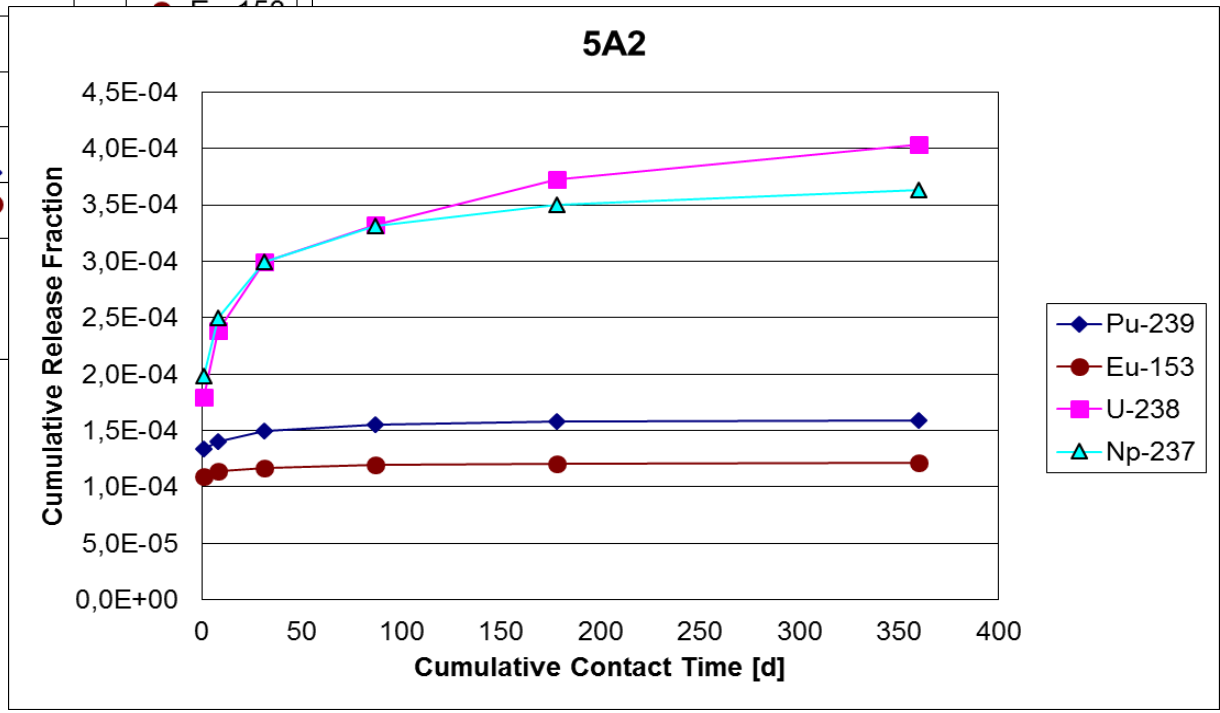




Nuclide release as percentage of ^{238}U release at 365 days (cumulative)

	5A2	C1
Low UO_2 solubility		
Cs-137	3894%	3627%
Tc-99	516%	1127%
Mo-100	1064%	2079%
Sr-90	245%	276%
Ba-138	115%	120%
Rb-85	785%	1128%
UO_2 matrix		
Lanthanides	30%	12%
Ce-140	31%	13%
Pr-141	32%	12%
Nd-144	30%	11%
Eu-153	30%	12%
Actinides		
U-238	100%	100%
Np-237	90%	92%
Pu-239	39%	22%

Cs-137 higher for 5A2, opposite for other nuclides. 5A2 (std) has higher instant release?

Lanthandies and actinides: Low release due to sorption. Small difference 5A2 vs C1 (ADOPT).

Nuclide release as fraction of ^{238}U release at 365 days (last contact period)

	5A2	C1
Cs-137	85	118
Tc-99	24	49
Mo-100	40	83
Sr-90	1,2	1,6
Ba-138	0,06	0,01
Rb-85	18	25

C1 (ADOPT) has higher release.

Cs-137 Initial phase (prev. slide): controlled by grain size/diffusion distance. C1 – larger grains = lower release.

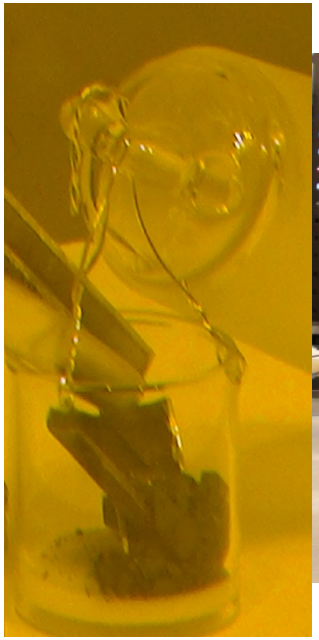
Other nuclides: matrix solubility? Reduced solubility caused by reduced hyperstochiometry in doped fuel.

Summary – leaching in air

- IRF Cs and I fuel fragments.
  - Cs release initially higher from 5A2 (due to smaller grains?)
  - Other nuclides higher from C1 (ADOPT) (due to lower matrix solubility?)
- Matrix dissolution:
  - C1 (ADOPT) generally slightly lower release than standard fuel.

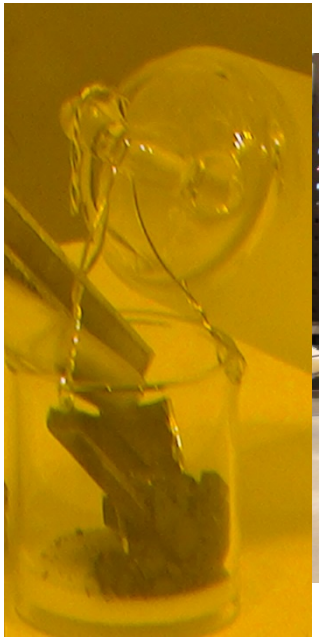
Suggested studies: Leaching of doped irradiated fuel under H_2 conditions

- Aim:
  - Study the effects of doping on matrix dissolution under deep repository conditions
  - Study the effect of doping on the influence of metallic particles on matrix dissolution



Base Scope

- 2 parallel autoclave experiments where 5A2 fuel and C1 fuel are tested under identical conditions
- Preferably chose pre-leached samples
- Leaching for 1-2 years under H_2 atmosphere and analyse the release of e.g. U, Pu, Np, Tc, Mo etc as a function of time.



Scope – Examples of “add on”

- Characterization of fuels (before/after) leaching using advanced microscopy to understand potential effects of doping
- Analysis of “additional” nuclides/elements e.g. Se, C-14
- Experiments in sealed containers to measure radiolysis products and couple with fuel dissolution and modelling

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