Evaluation of completed projects: FIRST Nuclides

Bernhard Kienzler, Karlsruhe Institute of Technology (KIT)

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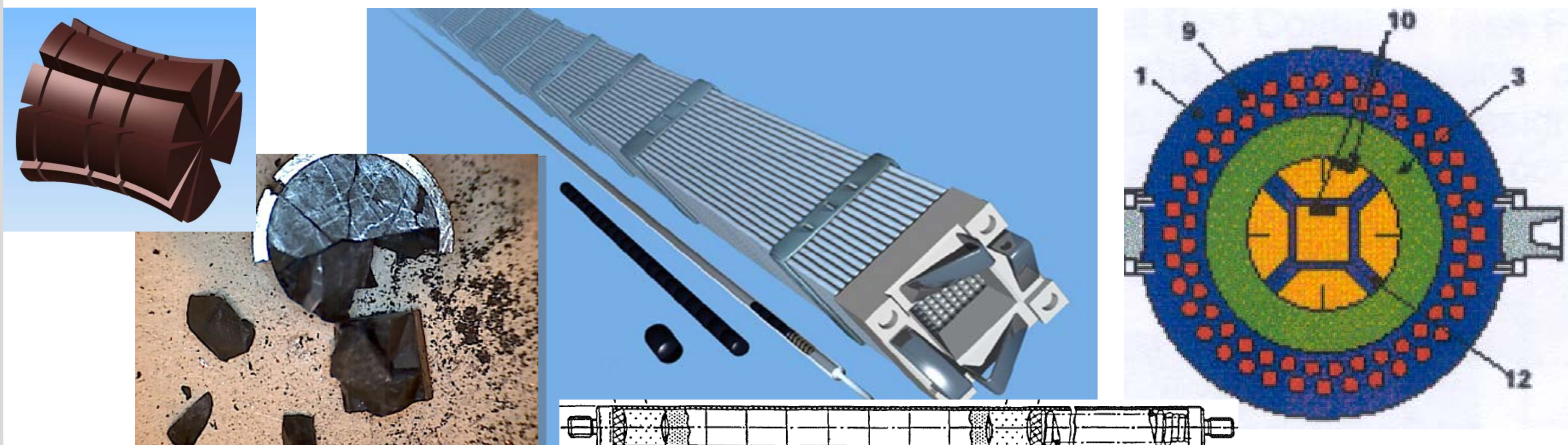
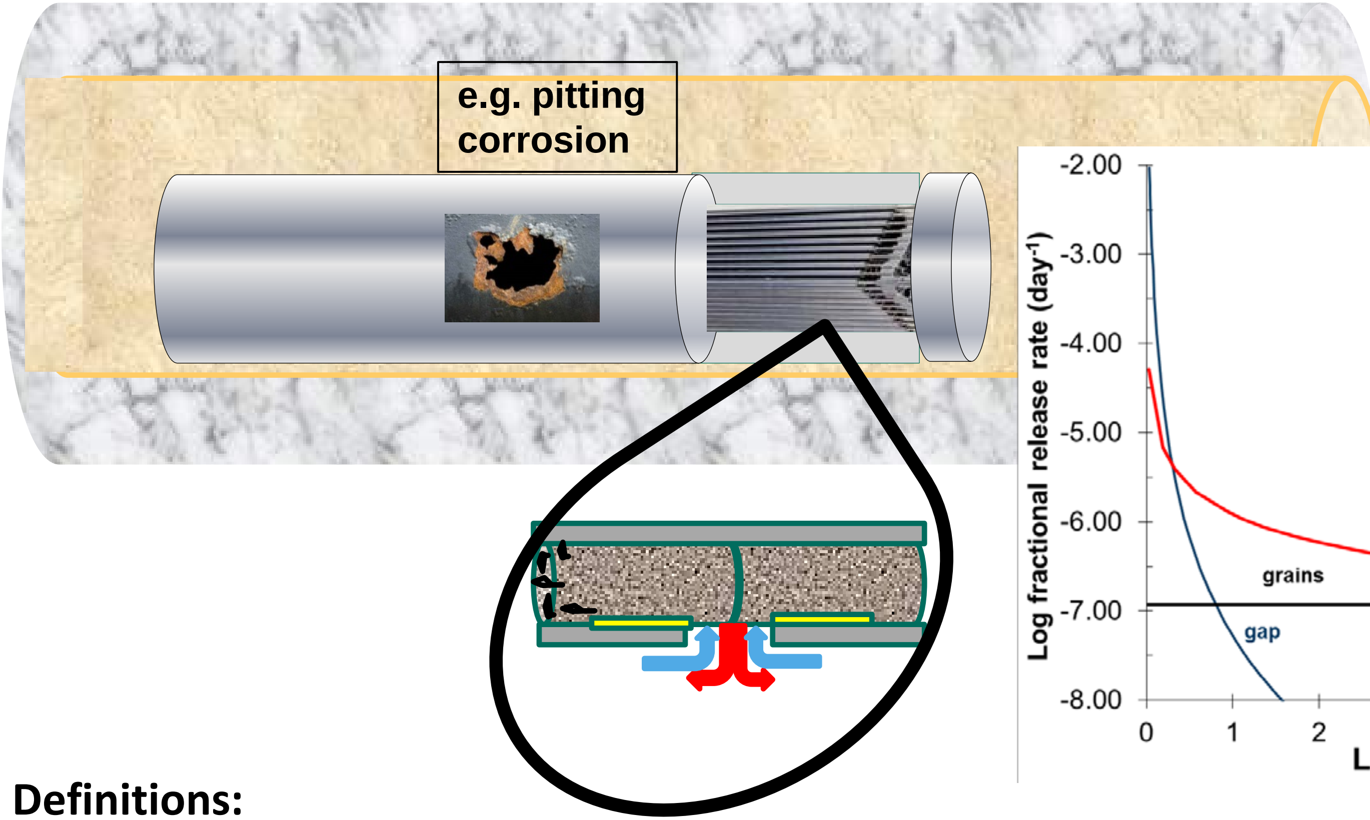
Institute for Nuclear Waste Disposal

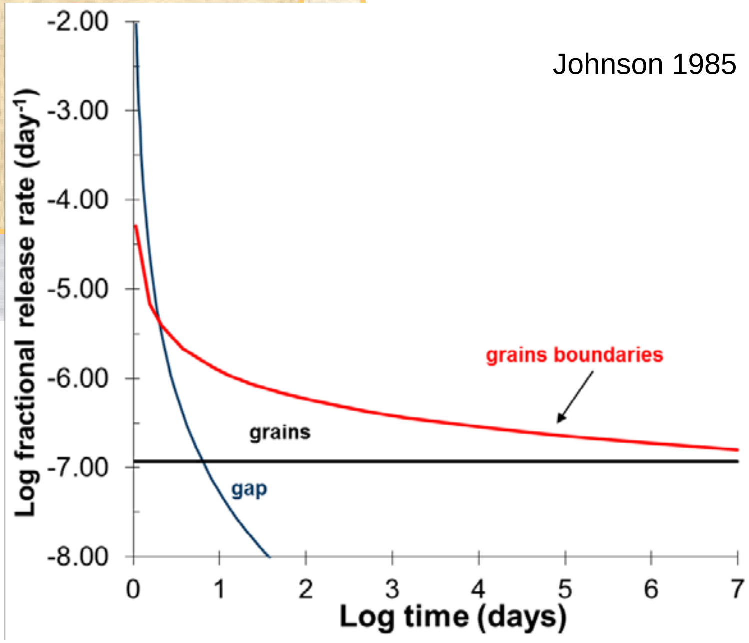


Content

- Relevance and definition of fast/rapid/instant release
- Collaborative Project FIRST-Nuclides:
  - Materials
  - Gas release
  - Dissolution based release
  - Modelling
  - Relations for estimation of IRF
- Element speciation in used fuel
- Summary, Conclusions, Outlook

Relevance and definition of fast/rapid/instant release





Definitions:

- **PA term:** „instant“ release
- **Experimental / lab related:** fast release release from different inventories:
  - gap inventory
  - grain boundary inventory
  - matrix contribution

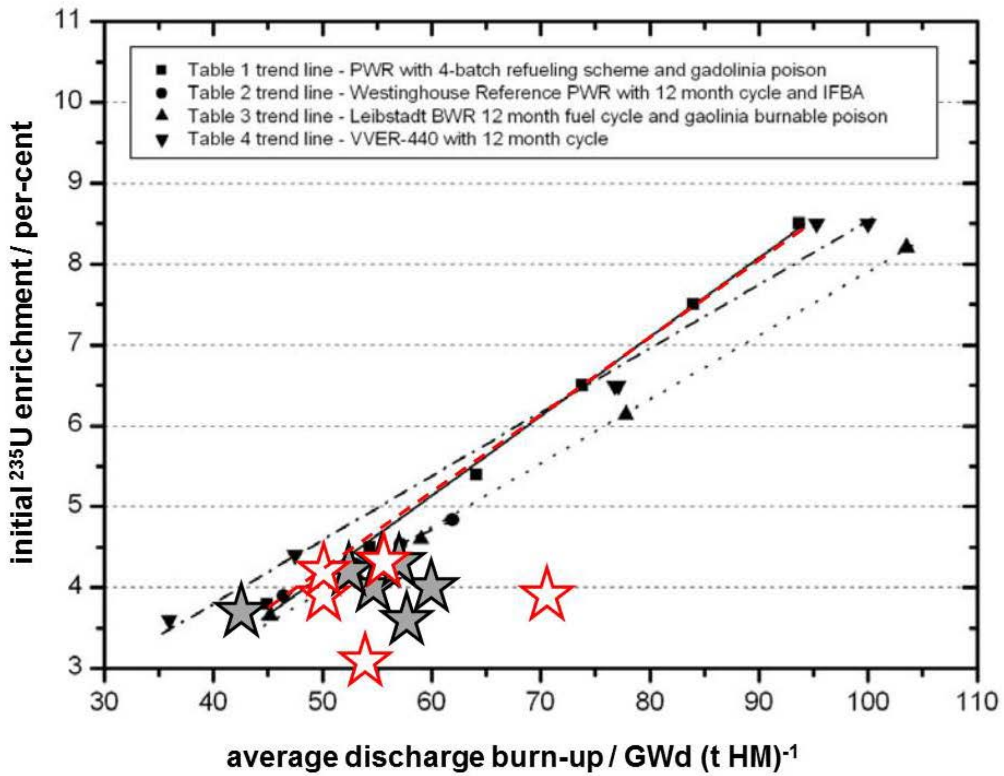
State-of-the-Art before FIRST-Nuclides

- Characterization of SNF: Rim properties, Xe, Cs diffusion
- Experimental investigations: > 40 publications evaluated
  - Lack of burn-up history data
- Distinguished papers
  - Poinssot et al. 2001, 2005, 2007, …
  - Ferry et al. 2003, 2007, …
  - Grambow et al. 2010
  - Johnson et al. 1985, 2004, 2005, 2012, …

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

Initial composition of UO_2 nuclear fuel (MOX excluded)

^{235}U enrichment



NEA Nuclear Science Committee (2006)

Additives

Criticality control: burnable poisson Gd_2O_3

- BWR fuel elements:
  1 to 18 doped rods per element
  ~ 1 to 7 wt.%.
- PWR fuel elements:
  2 and 12 doped rods per element
  ~1 and 7 wt. %.

Fission gas release: Grain size UO_2:

- Metal oxides: Cr_2O_3/Al_2O_3
  ~ 0.1 to 1 wt.%
  ➔ grain size 20-25 µm.
- Other compounds (sintering aids) allylhydridopolycarbosilane (AHPCS) .

CP FIRST-Nuclides

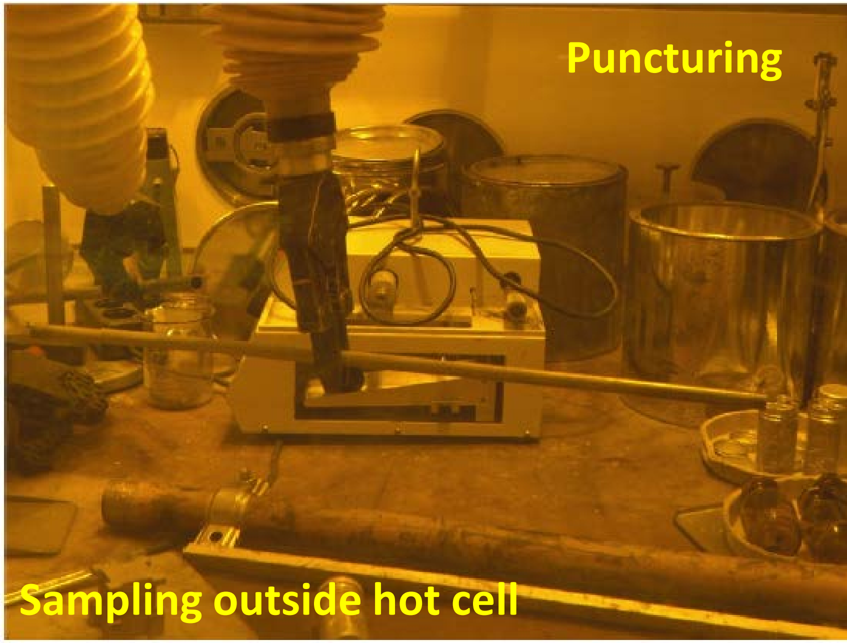
- **Objectives**: Quantification the rapid release of radionuclides from high burn-up used UO_2 fuel after canister failure.
  - Relation of FGR to IRF for ^{129}I, ^{79}Se, ^{135}Cs, for high burn-up / lin. power rate ranges, full set of sample sizes, typical groundwater, aerobic to reducing conditions, 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

Selected high burn-up UO_2 fuel samples

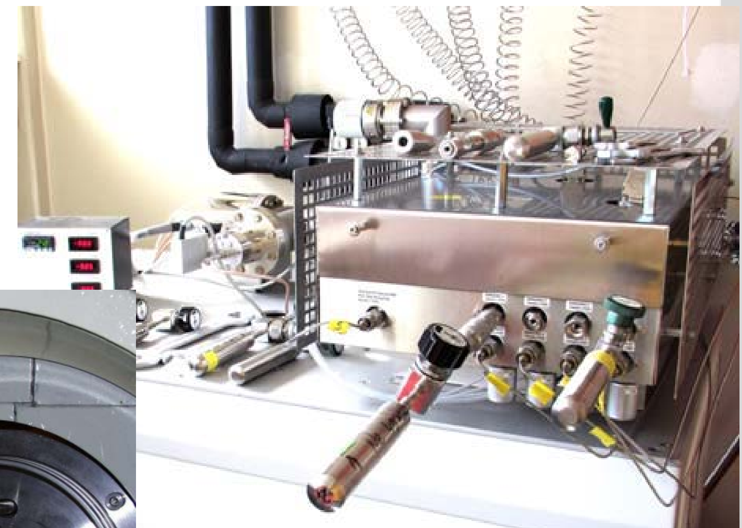
		PWR	BWR	THTR / VVER
Discharge		**1989 -2008**	**2005 – 2008**	
Pellet	Initial Enrichment	**3.80 – 4.94 %**	**3.30 -4.25 %**	2.4 -16.8%
Irradiation	Burn-up	**50.4 – 70.2 GWd/t**	**48.3 – 59.1 GWd/tU**	
	Cycles	2 - 14	5 – 7	
lin. power	average	**186 - 400 W/cm**	**143 - 290 W/cm**	130 – 228 W/cm
FGR		**4.2 – 13.2 %** (MOX 26.7 %)	**1.3 – 3.1 %**	
Dopants	(1 sample)	8% Gd	Cr_2O_3/Al_2O_3	

WP 2: Gas release and rim and grain boundary diffusion

KIT: Measurement of FGR of the rod used for the leaching experiments

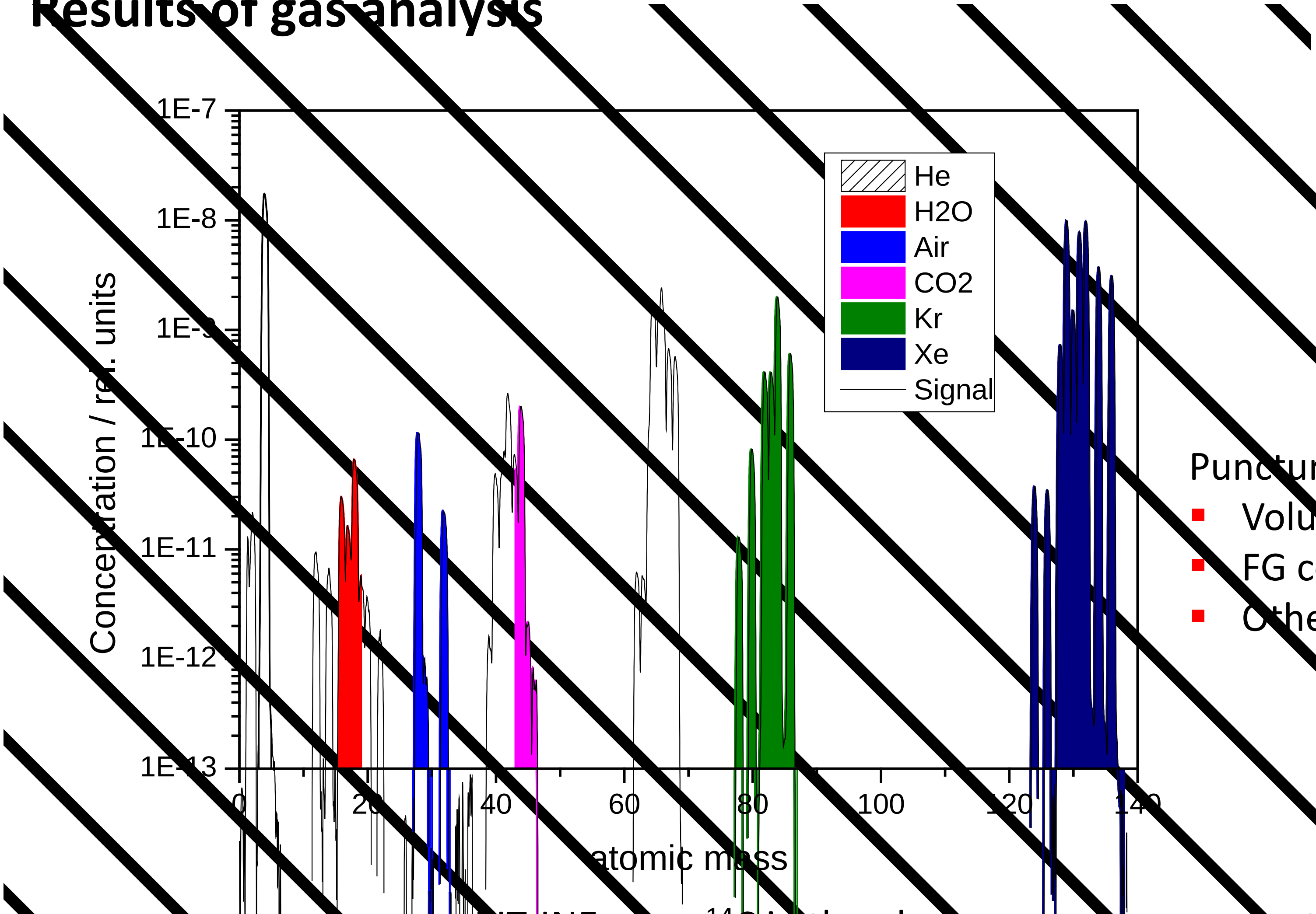


Analytics @ INE



Gas mass spectrometer

Results of gas analysis



Puncturing test:

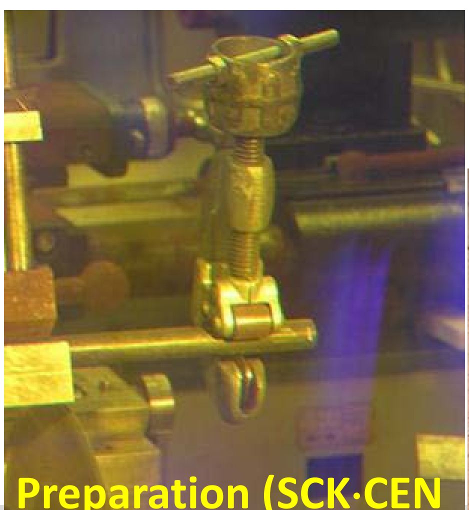
- Volume
- FG concentrations
- Other gas conc.

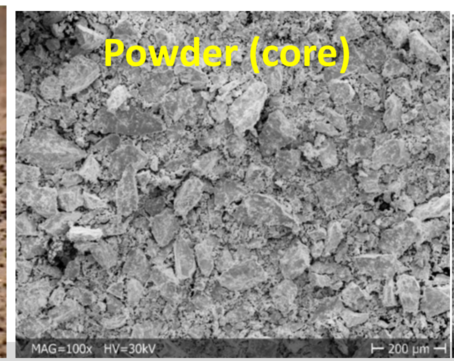
KIT-INE: ^{14}C in the plenum gas measurement:
Estimated ^{14}C mass in the rod: $4.3 \cdot 10^{-6}$ mol
➔ IRF($^{14}CO_2$) = 0.2 % of ^{14}C (gas phase)

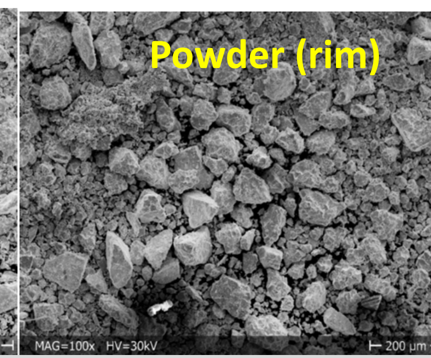
Sample preparation

- Experiments carried out using four kinds of samples:
  - bare fuel: piece of fuel and cladding with a defect.

  - Pellet: cladded (fuel + cladding) segment.
  - fragments: pieces or portions of fuel without cladding.
  - powder: fuel obtained after a complete mechanical process of decladding, drilling or milling and sieving

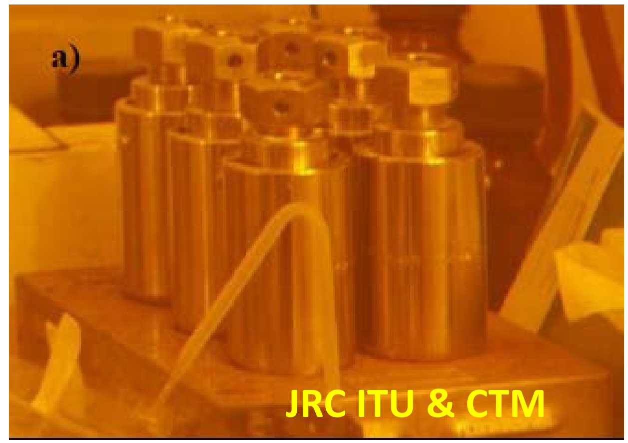


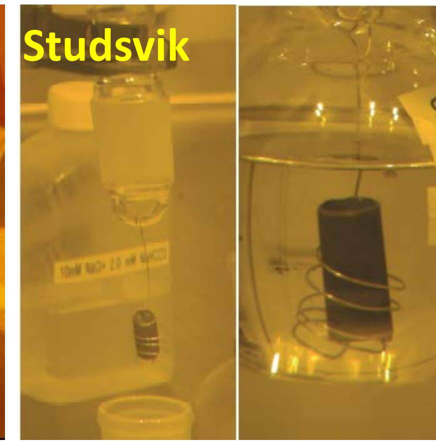




WP 3: Dissolution based release

- Dissolution based radionuclide release and to the extent possible the chemical speciation of the relevant isotopes.



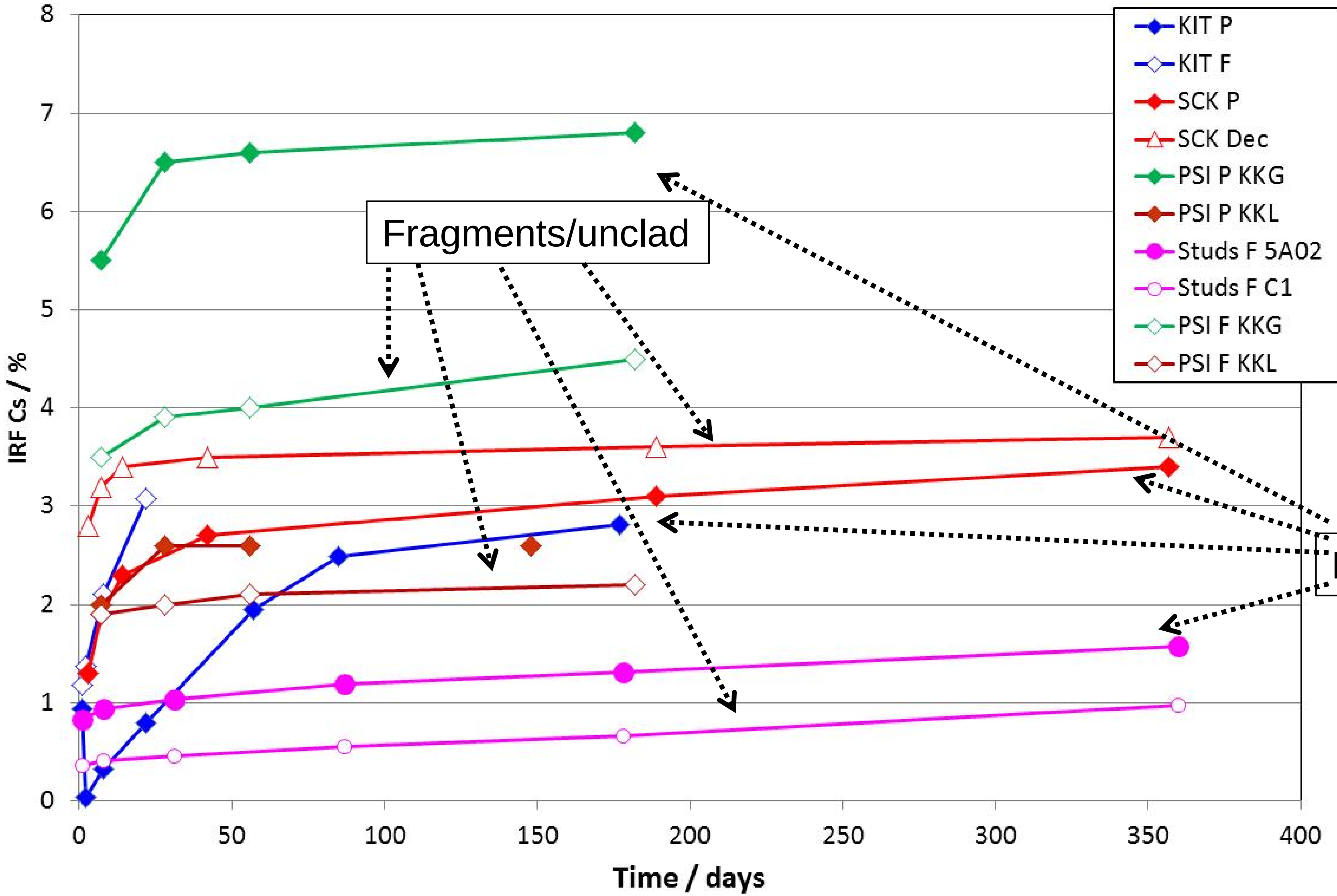


Leachants

	NaCl	$NaHCO_3^-$	pH	Conditions
PSI	19	1	8.5	anoxic / slightly oxidizing
KIT	19	1	8.9	Ar/H_2 (Eh = -116 mV)
ITU/CTM	19	1	7.4	Oxidizing (air)
SCK·CEN	19	1		anoxic / slightly oxidizing
Studsvik	10	2	8.1-8.2	Oxidizing (air)

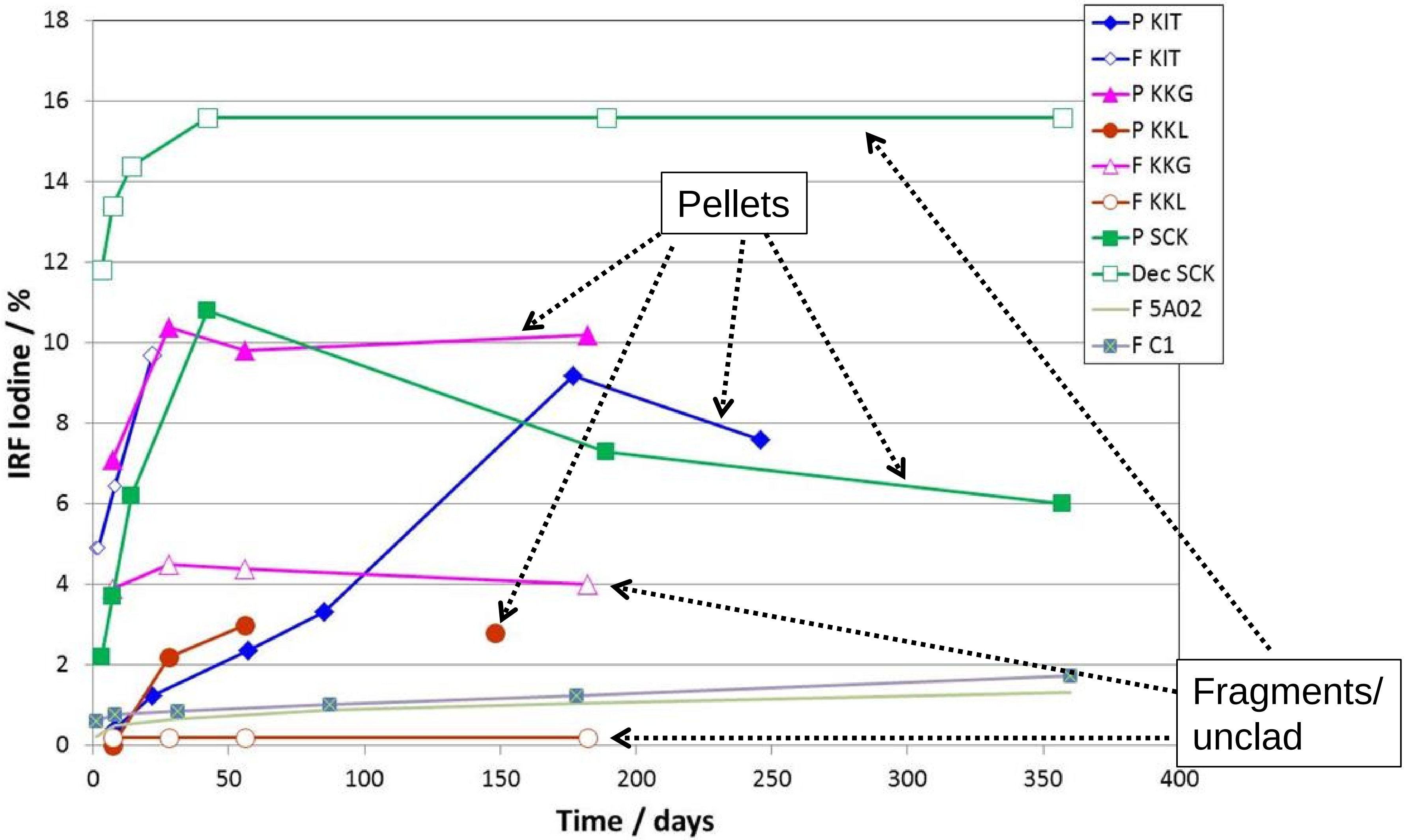
IRF$_{Cs}$ of different fuel / sample sizes

FIRST Nuclides



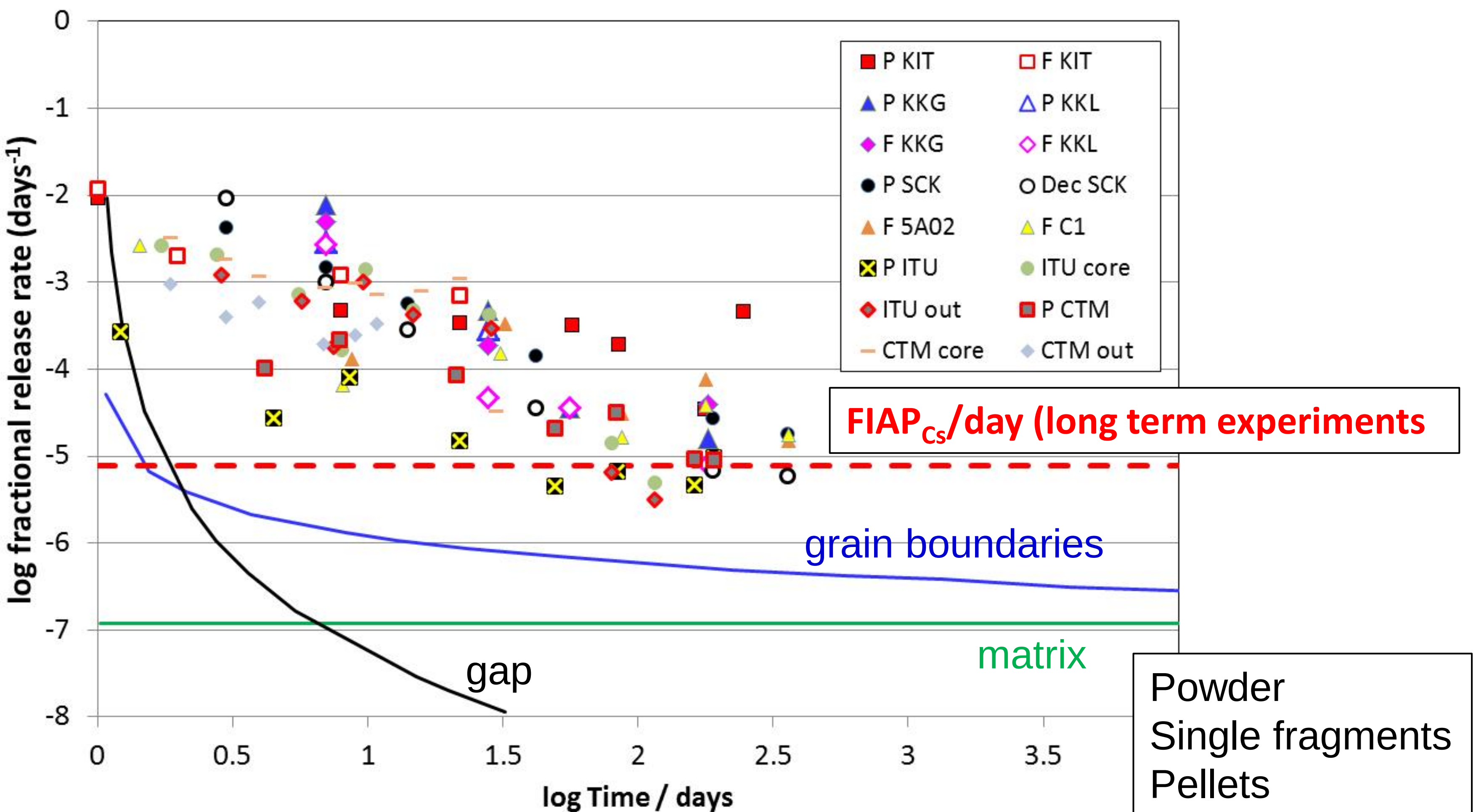
WP1 WP2 WP3 WP4 WP5

IRF$_{iodine}$ of different fuels / sample sizes



WP1 WP2 WP3 WP4 WP5

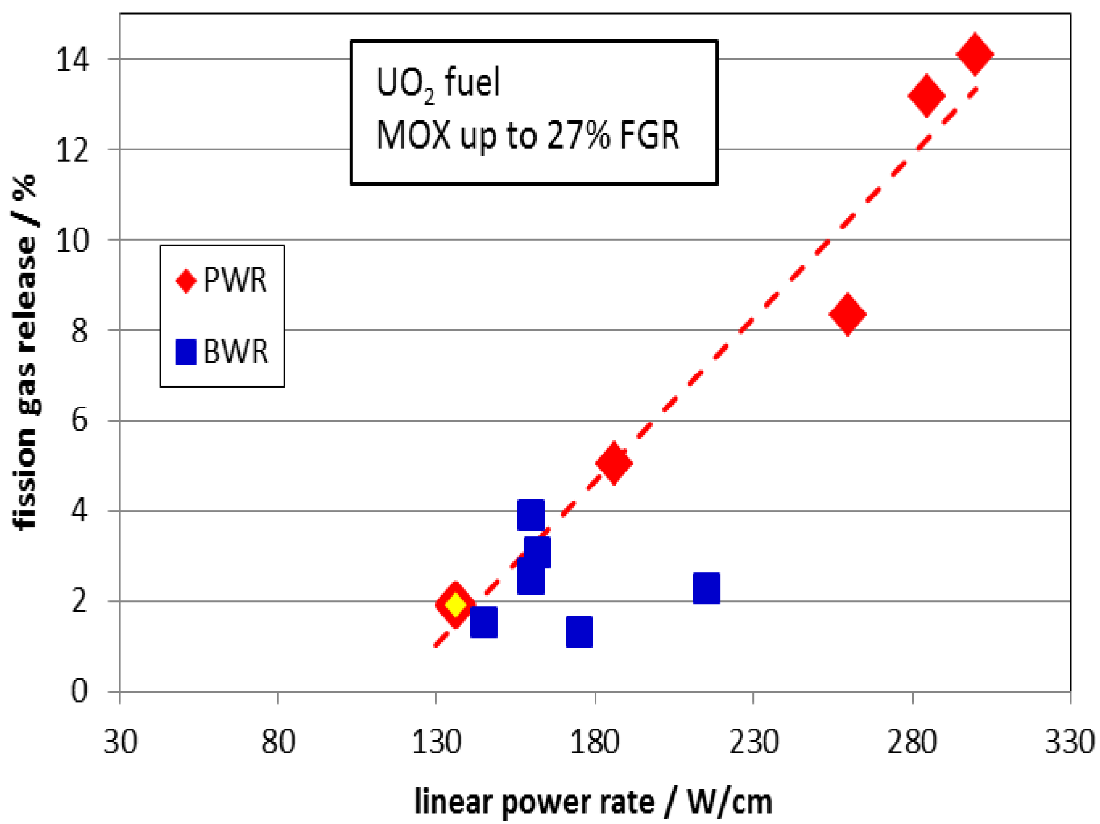
IRF Cs: Delineation from long-term release



Powder
Single fragments
Pellets
Slices of pellets

Relations

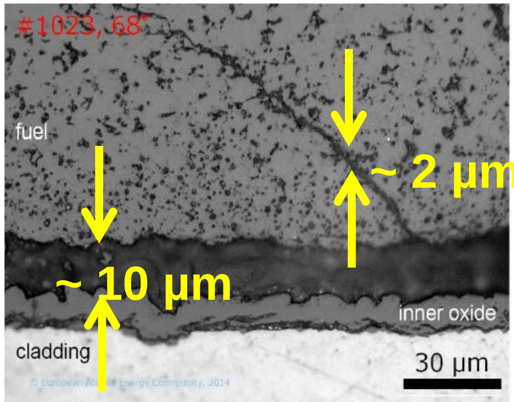
- Fission gas releas (temperature), le
- IRF(Cs) proportio
- IRF(I) proportiona
- IRF of Cs or I depe nature of the fuel
- Doping of UO_2 wi

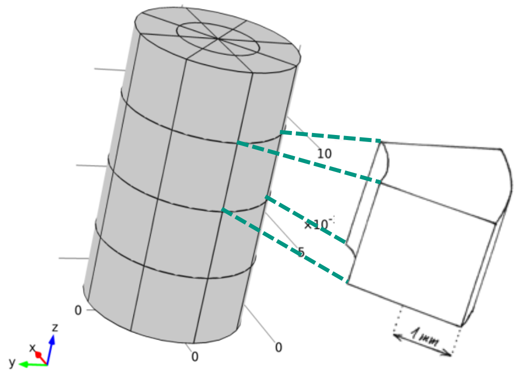


FGR ~ lin. Power rate

WP 4: Modelling

Water saturation of a pellet and release function

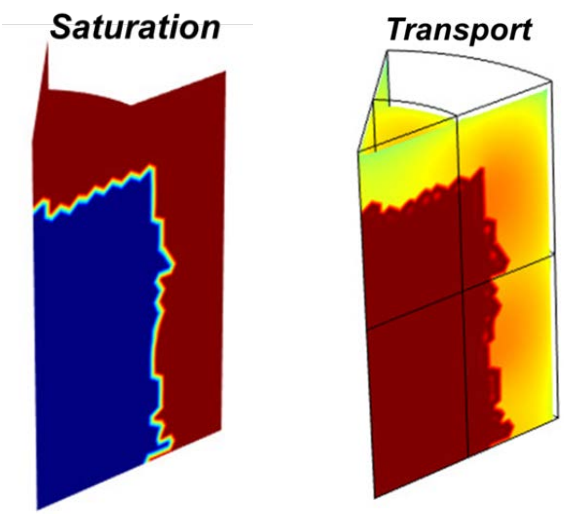


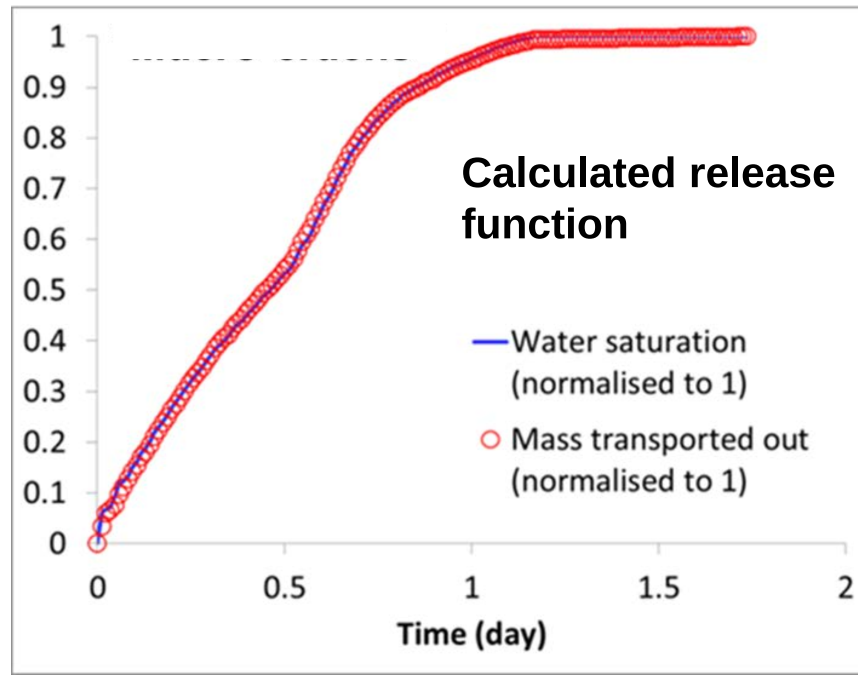


Variably-saturated flow model
- ➔ Richards equation (Bear and Cheng, 2010), extension of Darcy's Law to unsaturated conditions.

water retention / relative permeability:
- ➔ van Genuchten model (van Genuchten, 1980)

M. Pękala et al., AMPHOS21, Spain





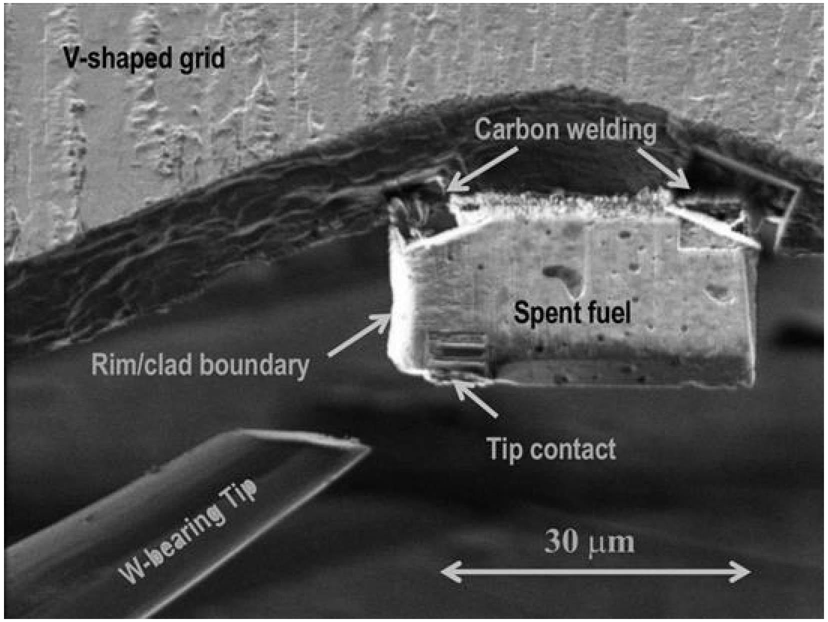
WP leader: Joan de Pablo (CTM/UPC)

IRF Database (Excel spreadsheet)

- Compilation of instant release data of radionuclides obtained by published studies over the last decades under different experimental conditions, different type of fuels, etc.
  - Detailed Info.
  - References
  - Detailed tables
- Database and Users-Guide available from the Coordinator of FIRST-Nuclides

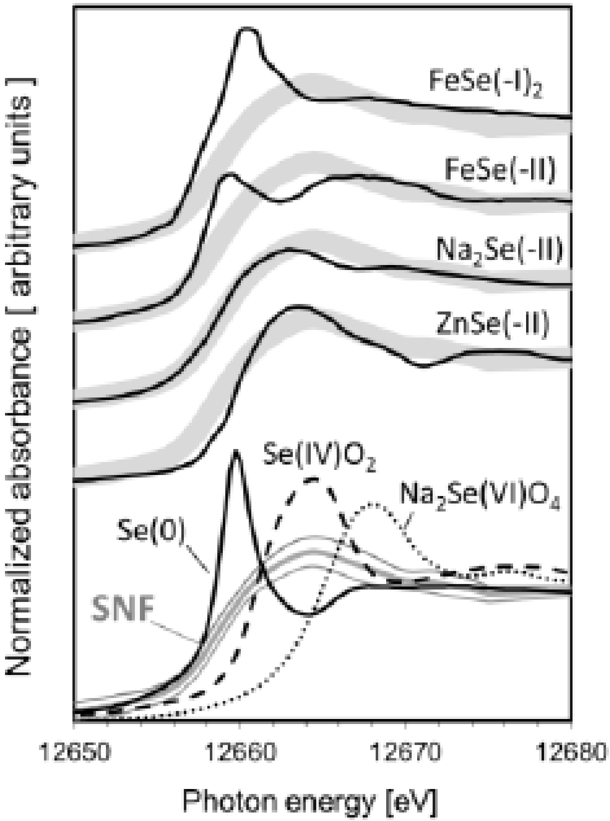
Element speciation in used fuel: Se

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



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.

End-User opinion

- Highly relevant for all WMO for direct disposal of spent nuclear fuel.
- IRF contributes substantially to the peak release after container breaching.
- Results important for PA because
  - … experimental determination for moderate and high burn-up UO_2 fuels, including doped fuels, ….
  - Improvement of analytical techniques for some difficult to measure radionuclides such as ^{14}C and ^{79}Se,
  - Insights into mechanisms related to fission product release
  - Data base for release of Cs and I from high burn-up fuel
  - Comprehensive comparisons of IRF with fission gas release (FGR)
  - … for estimation of IRF for the whole populations of fuel rods in a disposal.

Conclusions

- Successful project
- Scientifically
  - Improved understanding of “IRF“
  - Correlations between “reactor data” and IRF
  - ^{79}Se release & speciation in the UO_2 matrix.
- Publications available / in preparation
- Deliverables openly available (www.firstnuclides.eu)
- IRF database available
- Open issues still exist

Open issues

- IRF on the type and quantity of dopants.
- MOX and reprocessed U fuel
- IRF under reducing conditions.
- Tc behavior
- FGR measurement during leaching and correlation with in-pile data. Determination of FGR ?
- Quantification of ^{14}C and ^{36}Cl and understanding the impurity level ranges in fuels from different suppliers.
- Clarification of inconsistent results

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The research leading to these results has received funding from the European Atomic Energy Community's Seventh Framework Programme (FP7/2007-2011) under grant agreement no. 295722, the FIRST-Nuclides project.

Partners:

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End-Users:

ANDRA La maîtrise des déchets radioactifs · enresa · nagra aus verantwortung · Bundesamt für Strahlenschutz · ONDRAF/NIRAS · SKB Svensk Kärnbränslehantering AB

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