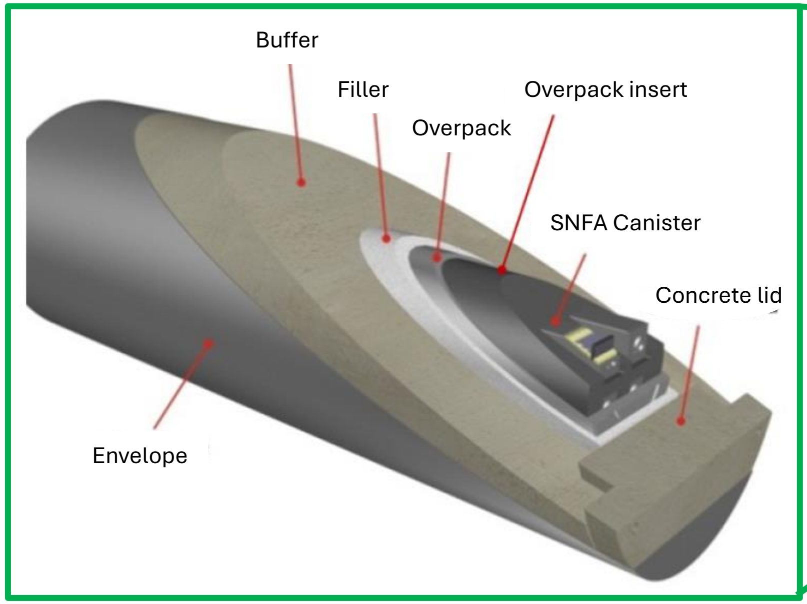
European Partnership
on Radioactive Waste Management

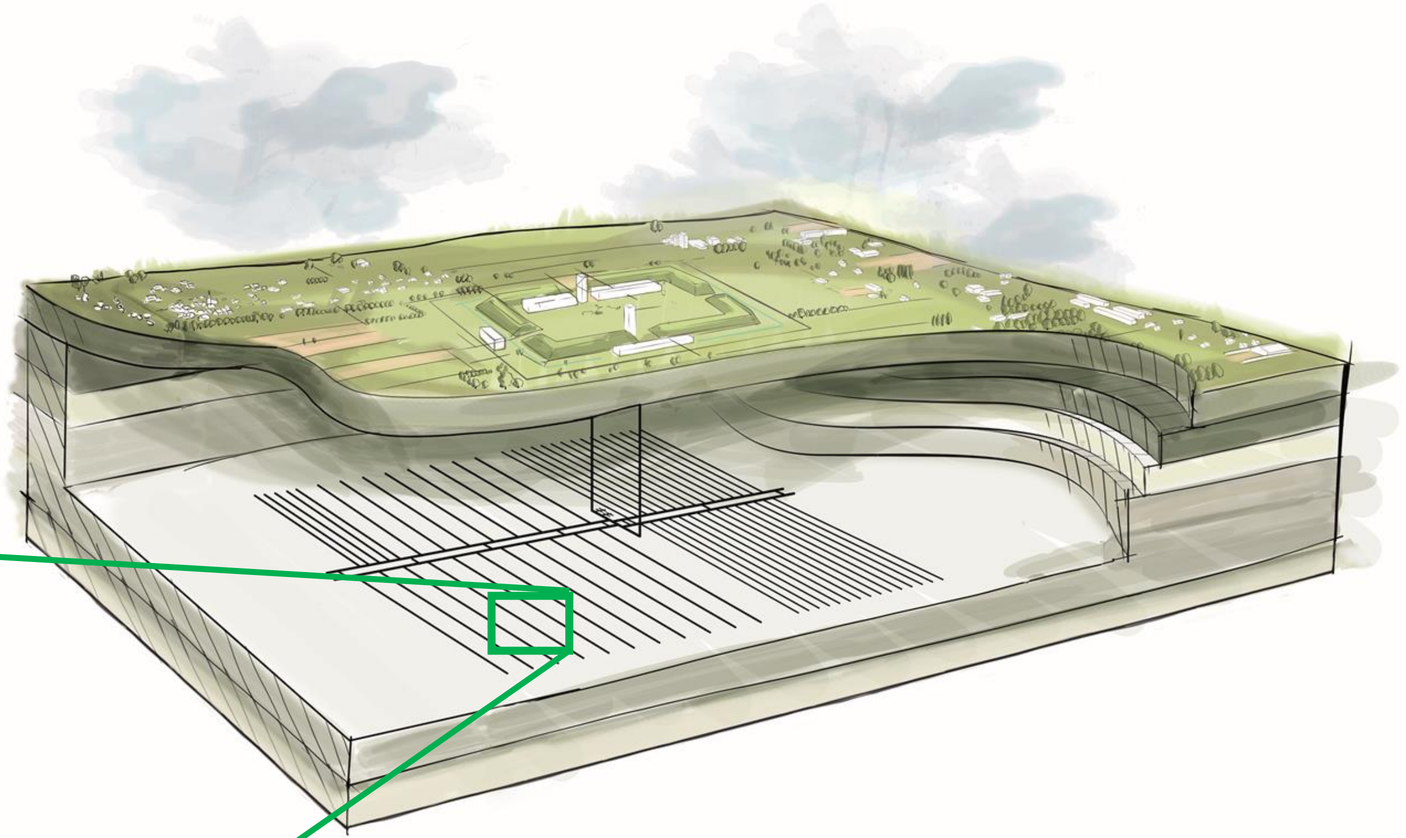
SPENT NUCLEAR FUEL LEACHING EXPERIMENTS TO INVESTIGATE RADIONUCLIDE RELEASE UNDER REPRESENTATIVE REPOSITORY CONDITIONS

SF-ALE program

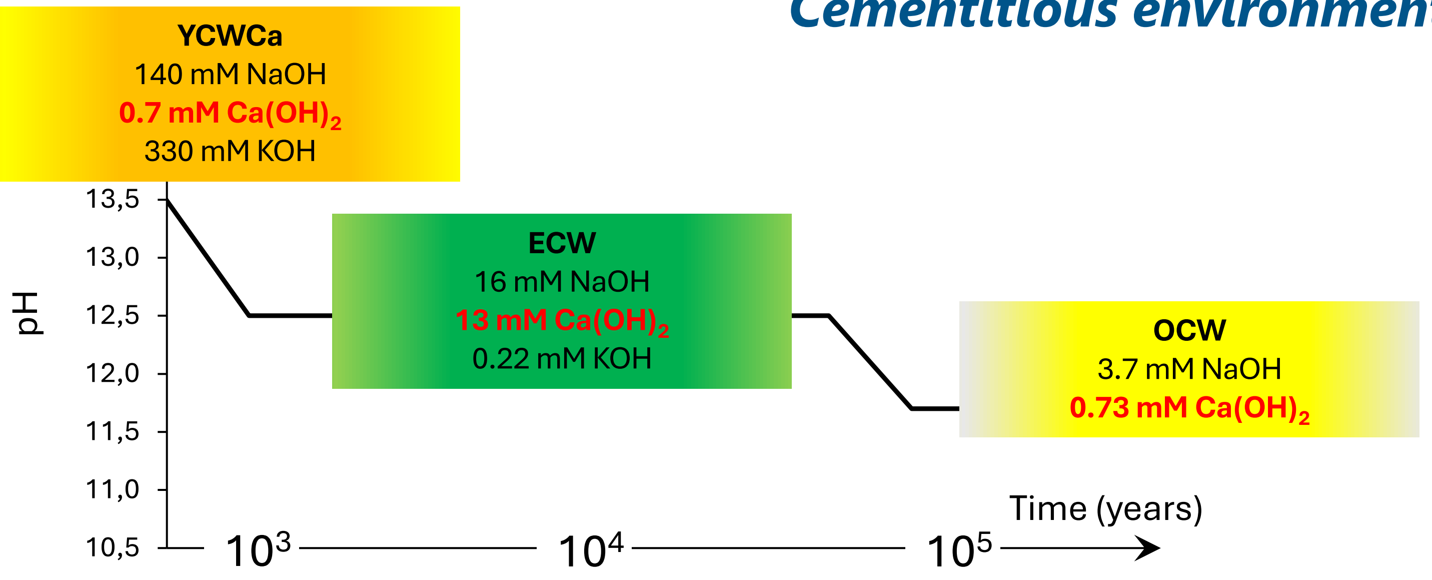
Co-funded by the European Union under Grant Agreement n° 101166718

Reference Belgian design for UOX irradiated fuel
Supercontainer design

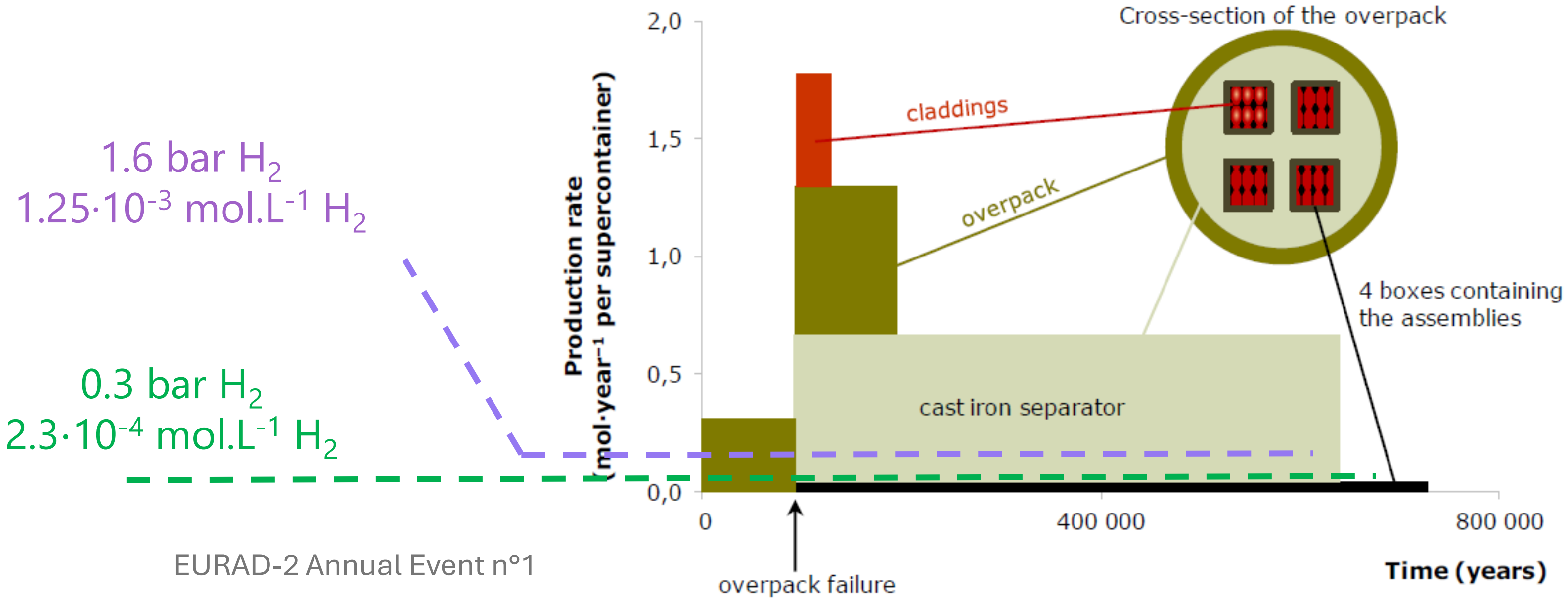




Boundary conditions
Cementitious environment (high pH) and H_2 production



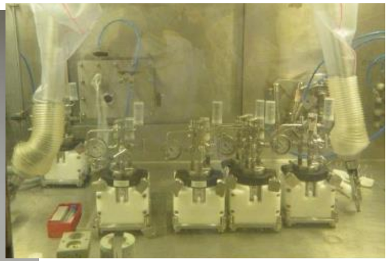
Estimation of the gas source term for spent fuel, vitrified high-level waste, compacted waste and MOSAIK waste, SCK•CEN-ER-162
(Best estimate source term: corrosion rate of 0.1 µm/y for carbon steel and 0.01 µm/y stainless steel)



Experimental planning

Phase I: Fast release of radionuclides

Phase II: Matrix dissolution

Liners & SF holders:

2018 – 2023: SF-ALE 1: Titanium

2023 – 2026(...): SF-ALE 2: PEEK

	Phase I						Phase II					
SF-ALE 1	0	5	21	82	271	586	733	762	908	1093	1273	1643
	Wash.	40 bar				4 bar	40 bar					

Sept. 2018 — *March 2023*

	Phase I						Phase II							
SF-ALE 2	0	5	30	100	200	300	305	380	530	670	850	1030	1210	1400
	Wash.	40 bar				Wash	40 bar							

Feb 2023

- ➢ After 5 days: renewal of the leaching solution and gas atmosphere
- ➢ Rinsing of liners with 1M HNO_3, contact time 7 days

Spent Fuel characteristics

sck cen

Reactor name	Tihange 1, Belgium
Reactor type	PWR
Initial fuel enrichment (^{235}U)	4.25%
Fabrication	Standard
Duration of irradiation (d)	997
Cooling time (y)	12
Number of cycles	2
Average burn-up ($MWd.kg^{-1}$)	50.5
Local burn-up ($MWd.kg^{-1}$)	54.6
Average Linear Power Density ($W.cm^{-1}$)	321
$FGR_{puncturing}$	14.1%

KIT Karlsruhe Institute of Technology

PWR Gösgen (Switzerland)
50.4 $GWd.t_{HM}^{-1}$, 260 $W.cm^{-1}$
$FGR_{puncturing}$: 8.3%

PWR Biblis-A (Germany)
46.9 $GWd.t_{HM}^{-1}$, 210 $W.cm^{-1}$
$FGR_{puncturing}$: 2.0%

(…)

JÜLICH Forschungszentrum

MOX
50, 52 , 29 $GWd.t_{HM}^{-1}$

eurad 2

Experimental conditions

	‡ Exp.	Atmosphere (40 bar)	Mol(H_2).L^{-1}	Leaching solution
SF-ALE 1	1.6H	4% H_2 / Ar	$1.25\cdot10^{-3}$	YCWCa
	0.3H	0.75% H_2 / Ar	$2.4\cdot10^{-4}$	YCWCa
	0.3H – Bic	4% H_2 / Ar	$2.4\cdot10^{-4}$	Bicarbonate
SF-ALE 2	0.3H – a	0.75% H_2 / Ar	$2.4\cdot10^{-4}$	YCWCa
	0.3H – b			
	Ar – a	Ar	--	
	Ar – b			

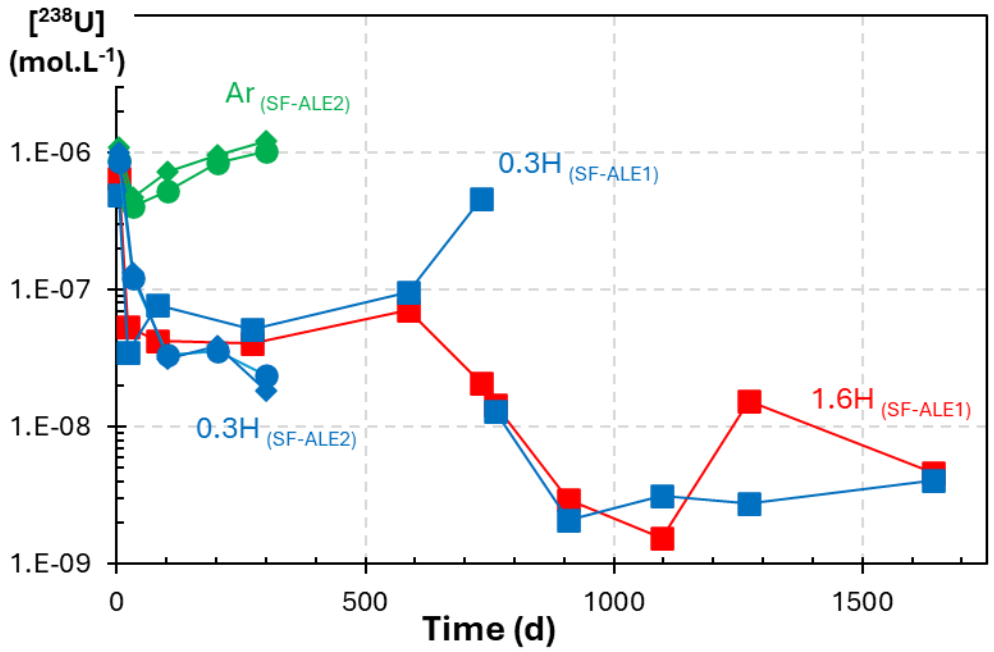
‡ ‘1.6H’ and ‘0.3H’ is the hydrogen partial pressure

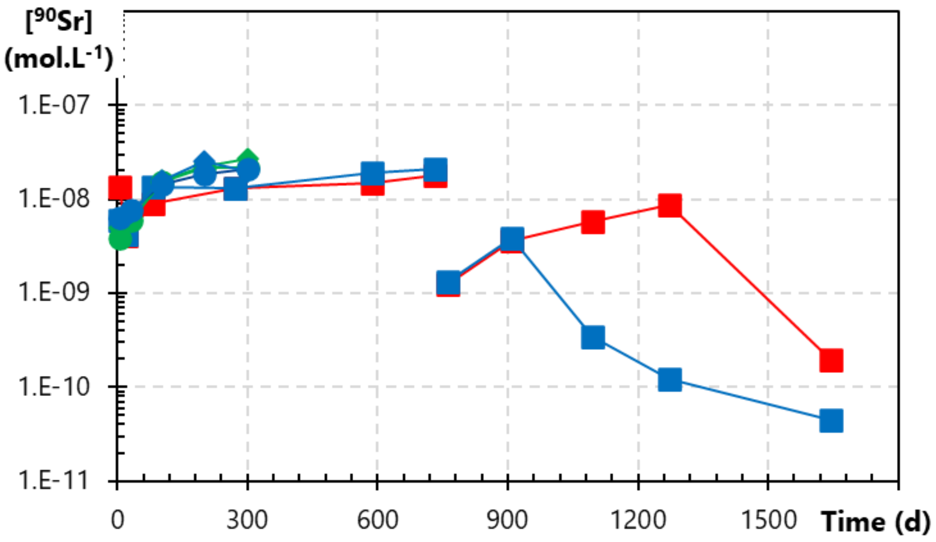
Mol.L^{-1}	[Na]	[Ca]	[K]	[Al]	[Si]	[CO_3^{2-}]	[Cl]	pH
YCWCa	$1.4\cdot10^{-1}$	* $3.8\cdot10^{-4}$	$3.7\cdot10^{-1}$	$6.0\cdot10^{-4}$	$3.0\cdot10^{-5}$	$3.0\cdot10^{-4}$		13.7
Bicarbonate	$2.0\cdot10^{-2}$					$1.1\cdot10^{-3}$	$1.9\cdot10^{-2}$	7.4 – 8.2

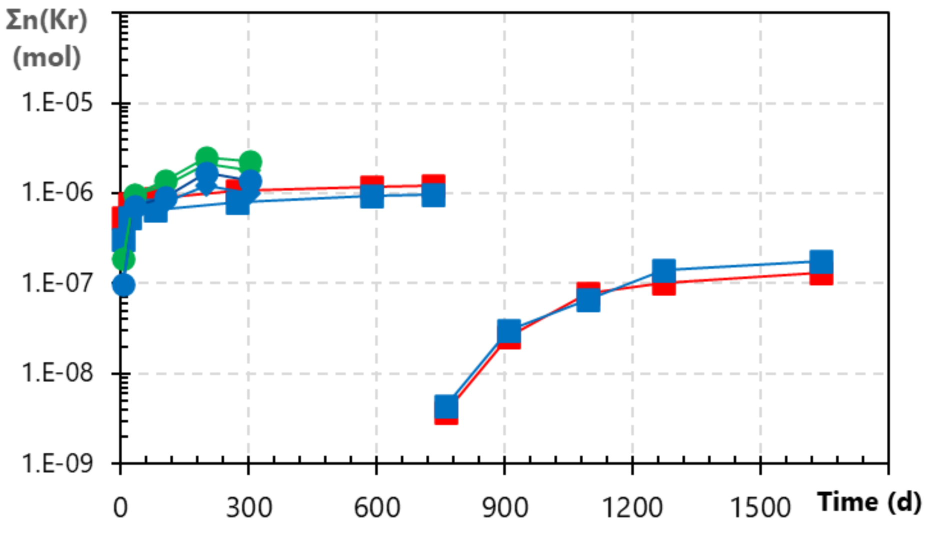
* Initial calculation: [Ca]= $7.0\cdot10^{-4}$ mol.L^{-1}

1.6H	0.3H	Ar

Radionuclides released







- Influence of the hydrogen
- Reproducibility of the experiments
  - SF-ALE 2 experiments
  - SF-ALE 1 & 2: 0.3H experiments
- To be compared with phase solubilities
  - e.g. UO_2(am, hyd)=$10^{-8.5\pm1}$ mol.L^{-1}

Fraction of the inventory released

Fraction of the Inventory in the Aqueous Phase

$$FIAP\ (i,t) = \frac{n_{i,aq}(t)}{n_{i,tot}} \times 100$$

$n_{i,aq}(t)$: measured moles of (i) released in solution at time (t)

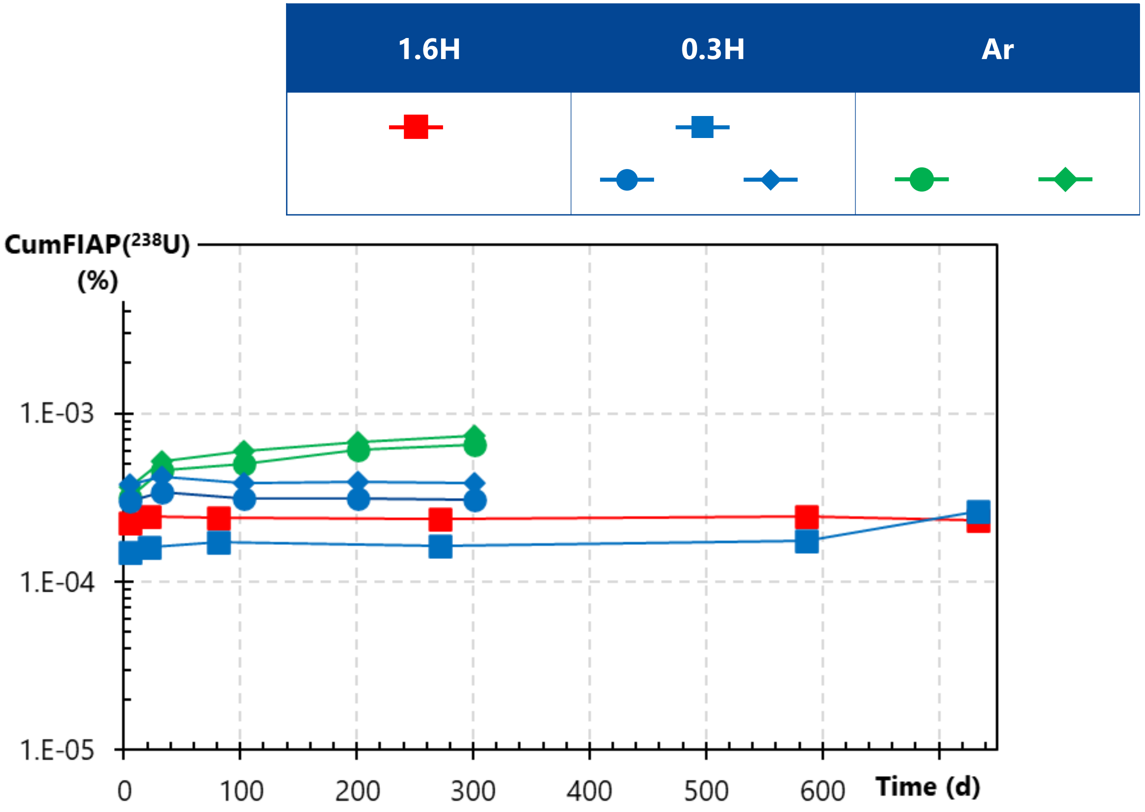
$n_{i,tot}$: total moles of (i) in the fuel sample

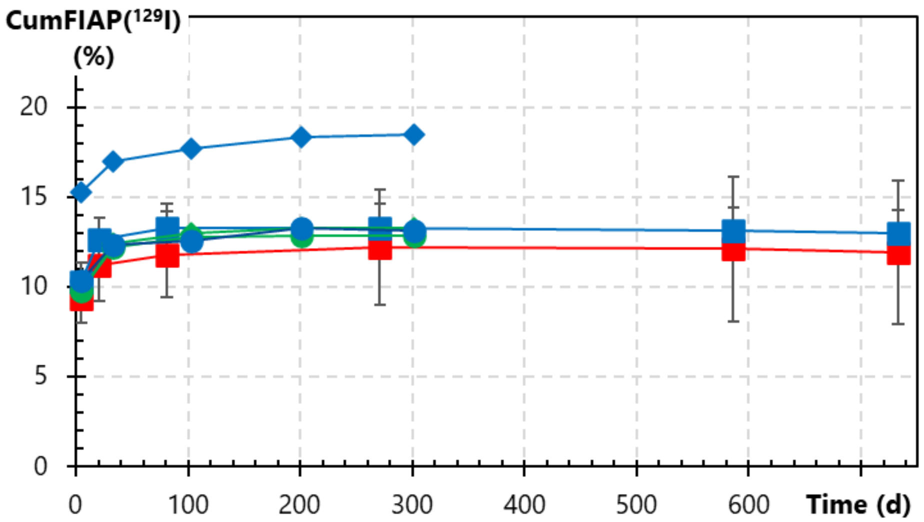
- Calculation of the isotopic inventory (e.g. Serpent 2)
- Radiochemical determination

$$CumFIAP = FIAP_{preleaching} + FIAP_{Phase\ I} + \dots$$

Limitations:

- Very low concentration/activity in solution (< measurable value)
- Difficult to measure radionuclides
- Spent fuel sample (size, shape,…)





Mennecart, Th. & al, MRS Advances 2025, https://doi.org/10.1557/s43580-025-01270-3

Accessible Fraction of the Inventory (AFI) = IRF$_{(Phase\ I)}$

$$AFI\ (i,t) = CumFIAP\ (i,t) - CumFIAP\ (U,t)$$

AFI is an estimation of the fraction of radionuclide (i) that is not incorporated in the UO_2 matrix, based on the release in solution measured ***in the leach tests***

AFI (%)	1.6H	0.3H	0.3H - Bic	Ar		0.3H		FGR 14.1%
Duration (d)		733				301		
^{129}I	11.9 ± 5.0	13.0 ± 5.4	13.8 ± 4.6	12.8 ± 5.3	13.3 ± 5.4	13.1 ± 5.4	*18.5 ± 7.6*	≈
^{137}Cs	2.8 ± 1.0	2.9 ± 0.9	2.6 ± 0.7	3.5 ± 0.9	3.4 ± 0.8	3.0 ± 0.7	3.4 ± 0.9	<60%
^{90}Sr	$(6.0 \pm 2.1) \cdot 10^{-3}$	$(3.9 \pm 1.3) \cdot 10^{-3}$	$(4.6 \pm 1.4) \cdot 10^{-2}$	$(2.0 \pm 2.0) \cdot 10^{-3}$	$(3.1 \pm 2.4) \cdot 10^{-3}$	$(5.7 \pm 2.6) \cdot 10^{-3}$	$(5.9 \pm 2.9) \cdot 10^{-3}$	
^{99}Tc	$(4.5 \pm 1.7) \cdot 10^{-3}$	$(1.9 \pm 0.7) \cdot 10^{-2}$	$(6.7 \pm 2.3) \cdot 10^{-2}$	$(1.0 \pm 0.2) \cdot 10^{-2}$	$(4.2 \pm 3.2) \cdot 10^{-2}$	$(4.6 \pm 4.8) \cdot 10^{-2}$	$(5.9 \pm 5.1) \cdot 10^{-2}$	

Fraction sorbed should be included in the calculation: underestimation of the AFI values

Conclusions and way forward

Long term leaching experiments of UOX fuel using autoclaves to investigate the release of the fission gases and a selection of fission products. The experiments are performed in duplicate to increase the confidence in the results.

- ➢ Good reproducibility of the experiments in similar conditions, even if some differences appeared likely attributed to the SNF samples themselves
- ➢ Provide relevant information when disseminate the results: (Spent) Nuclear Fuel properties
- ➢ Representativeness of the experimental conditions – WP17 CSFD
- ➢ Need for improvement of the analytical methods (e.g. DTM)
- ➢ Modelling approach (high pH, complex system): potential collaboration task 6 with WP18-DITUSC

Thanks to...

... and to you for your attention

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This study was carried out in the framework of a public-public cooperation between SCK CEN and ONDRAF/NIRAS.

This cooperation concerns scientific research and technical-scientific support for the safe management of radioactive waste on short-, middle- and long-term, including training and education on this subject.

With this partnership, ONDRAF/NIRAS and SCK CEN want to strengthen and consolidate their collaboration within strategic research domains of the Belgian national programme for radioactive waste management. In this way, a robust framework is created with a view on increased efficiency and long-term continuity of knowledge and expertise within the Belgian programme.

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Belgian Nuclear Research Centre

ONDRAF/NIRAS

SCK CEN
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Studiecentrum voor Kernenergie
Centre d'Etude de l'Energie Nucléaire

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