

Spent Fuel leaching in repository relevant conditions: Influence of H₂ concentration and pH

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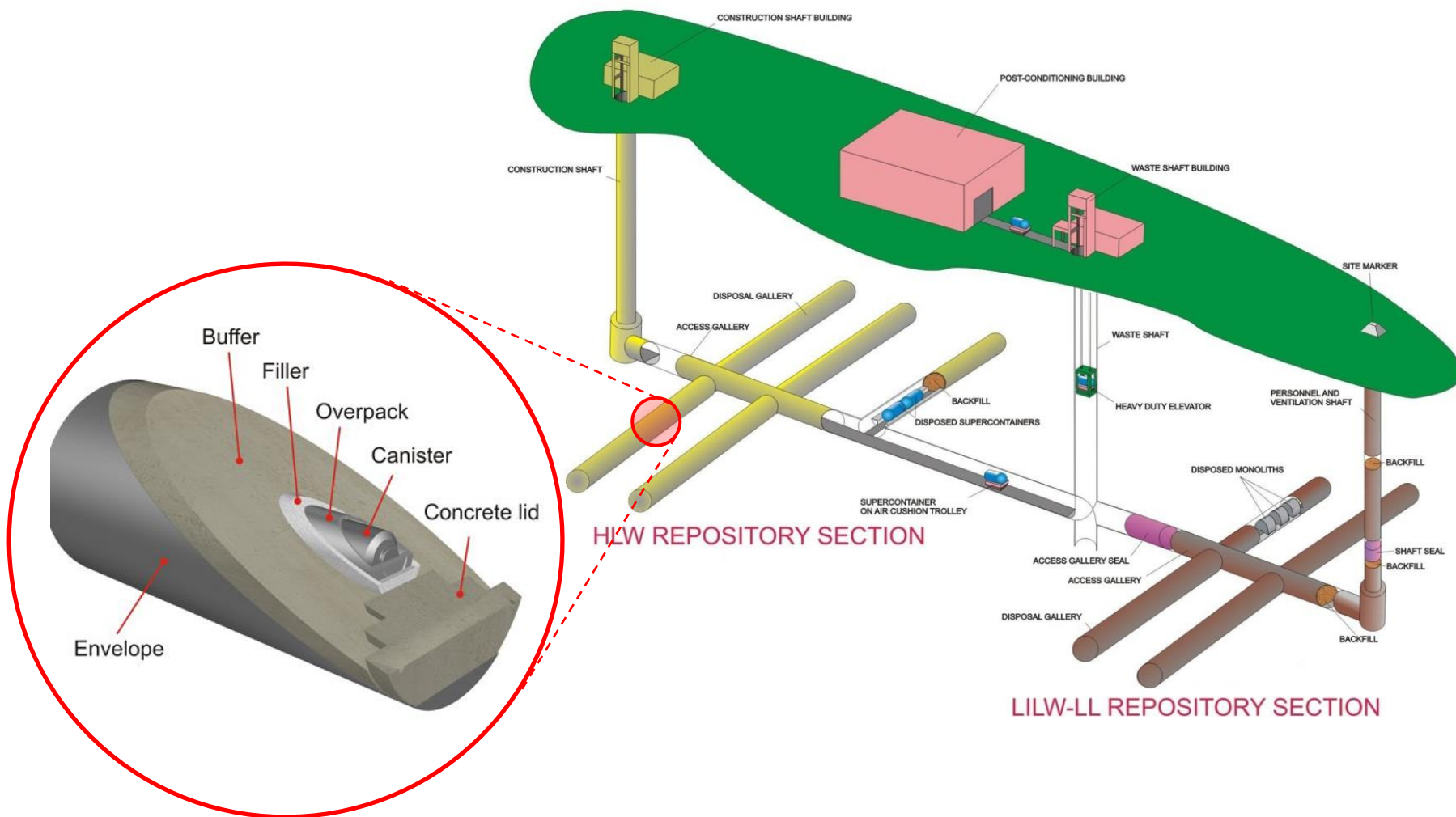
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Spent Fuel Workshop
Ghent, 14th – 15th November 2019

Reference Belgian design (Supercontainer) for UOX irradiated fuel



Boundary conditions: cementitious environment (high pH) and H₂ production

YCW

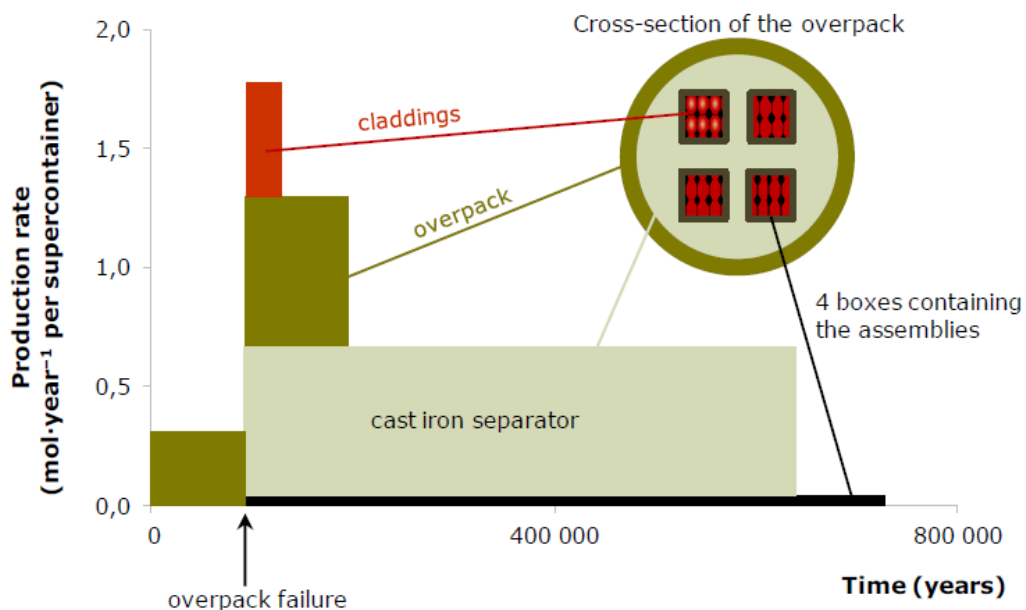
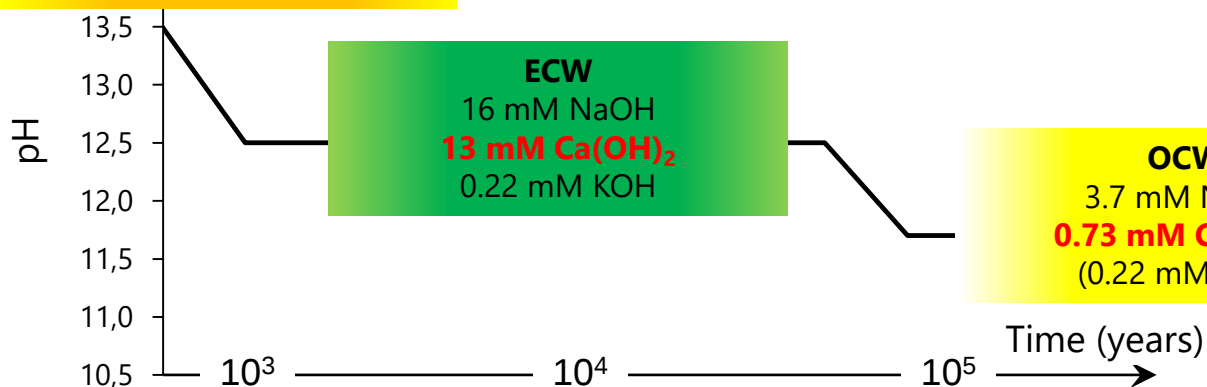
140 mM NaOH
0.65 mM Ca(OH)₂
330 mM KOH

ECW

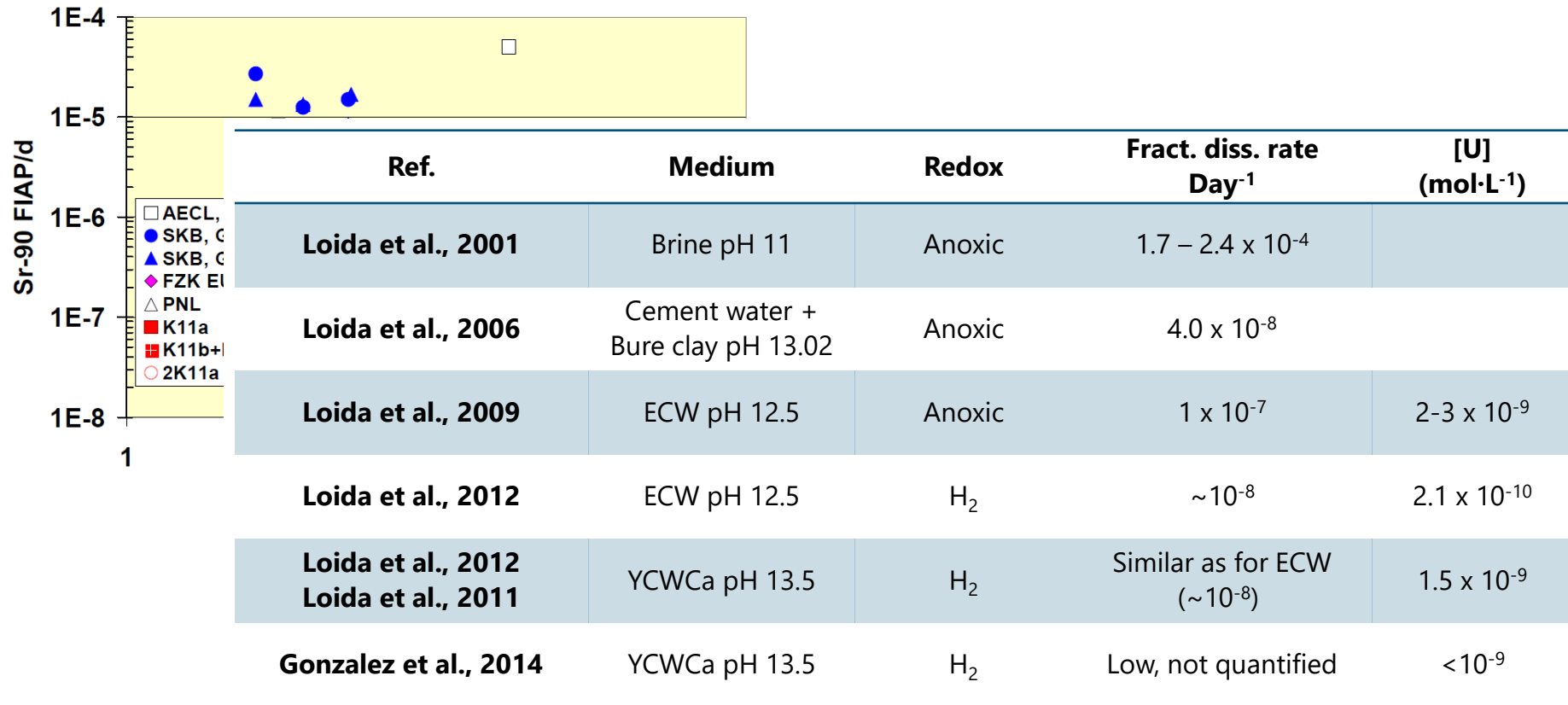
16 mM NaOH
13 mM Ca(OH)₂
0.22 mM KOH

OCW

3.7 mM NaOH
0.73 mM Ca(OH)₂
(0.22 mM KOH)



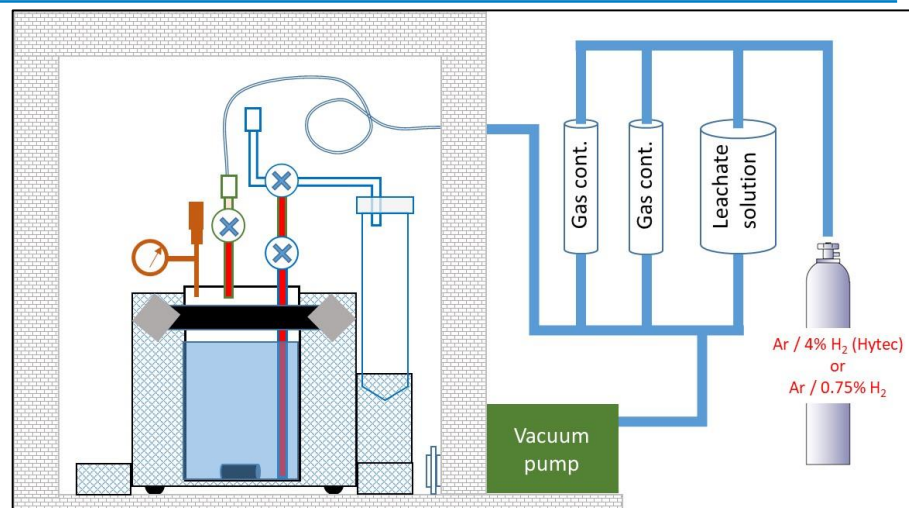
Previous spent fuel experiments showed no unfavorable high pH effect



- Previous tests at KIT/INE in ECW and YCWCa with Gösgen PWR fuel, without H₂ or with ~high H₂ pressure
- Current tests with lower H₂ pressure and with Tihange PWR fuel at SCK•CEN

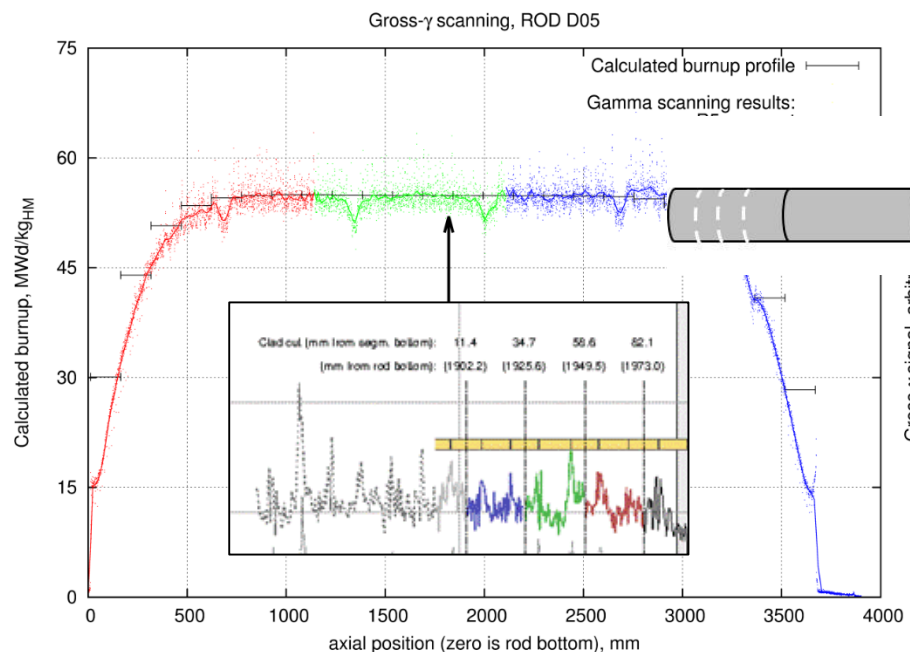
Spent Fuel - Autoclave Leach Experiment (SF-ALE): Experimental setup

FIRST Nuclides

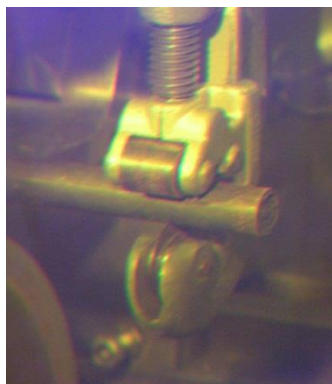


Exp.	Leaching solution	Atmosphere (40 bar)
1	YCWCa (pH 13.5)	4% H ₂ / Ar
2	YCWCa (pH 13.5)	0.75% H ₂ / Ar
3	Bicarbonate solution (pH 7.4)	0.75% H ₂ / Ar

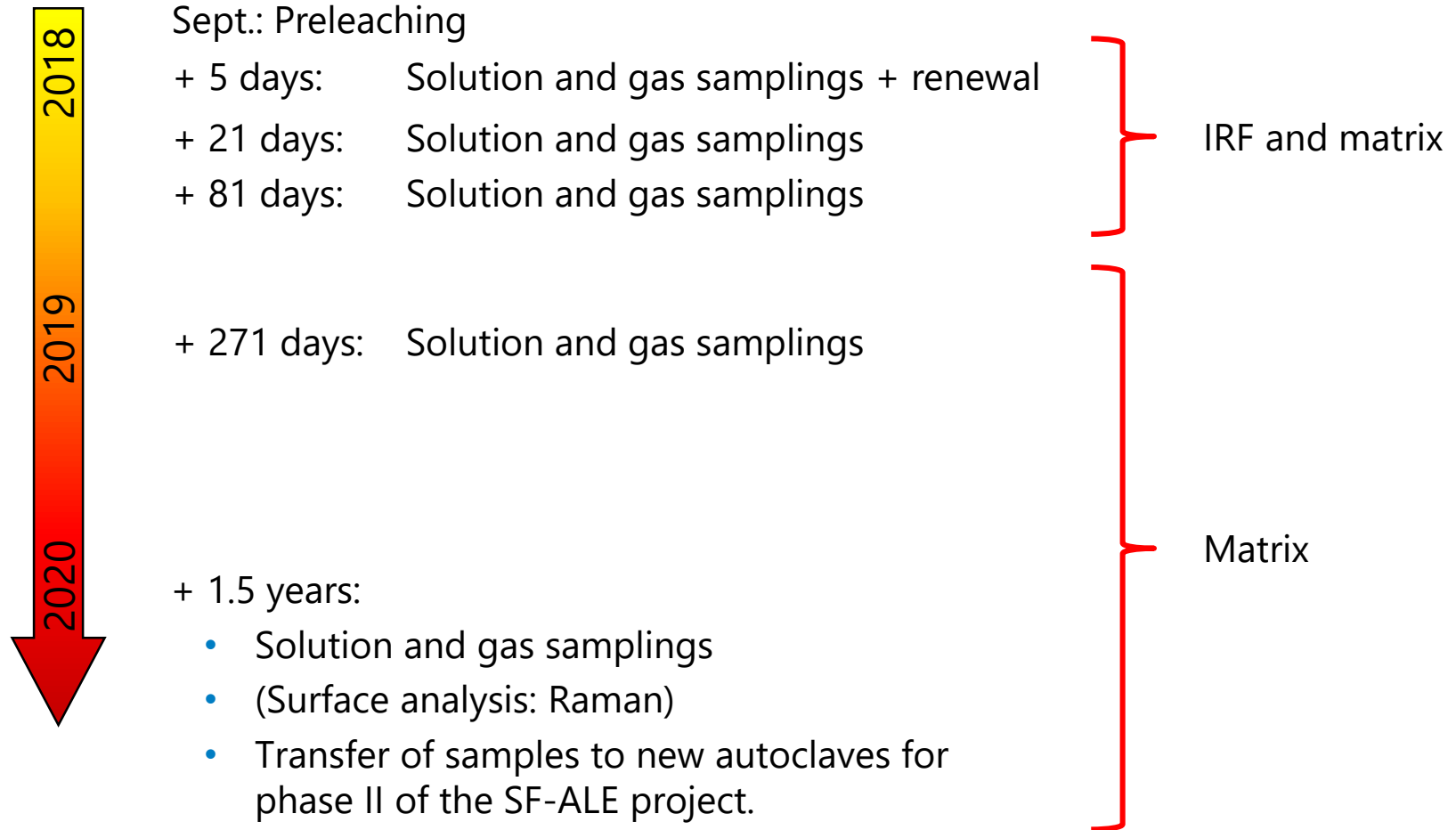
Spent Fuel - Autoclave Leach Experiment (SF-ALE): Sample preparation



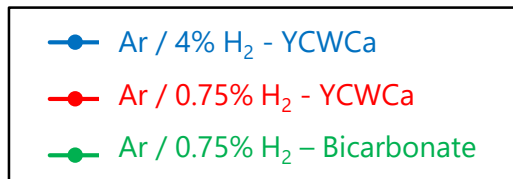
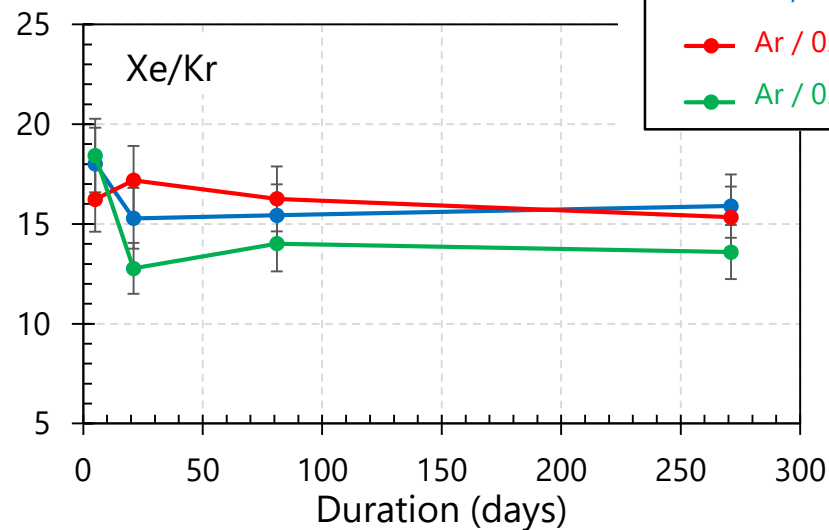
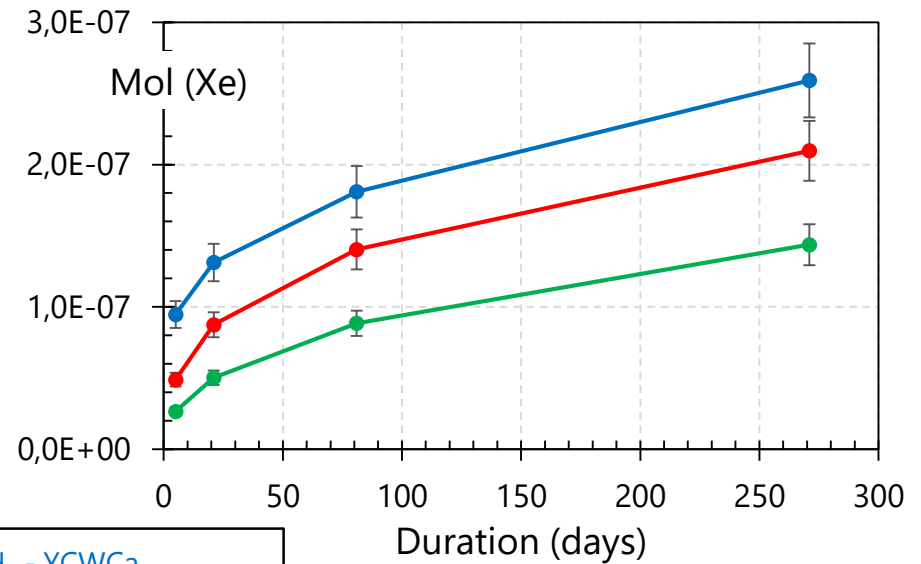
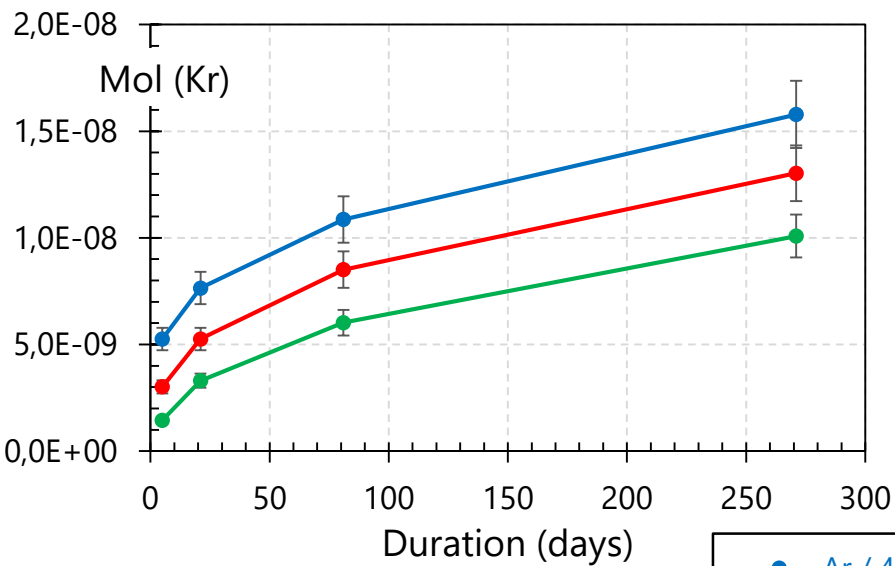
- In hot cell under air atmosphere
- Dry cutting



- Tihange 1 (PWR – 15x15 fuel assemblies)
- UO₂ fuel (4.25% ²³⁵U) – M5 cladding
- 2 irradiation cycles of 18 months
- Linear power: > 300 W.cm⁻¹
(1st cycle: >350 W.cm⁻¹; 2nd cycle~250 W.cm⁻¹)
- Local burnup: 54.6 MWd/kg_{HM}
- Fission Gas: 14.1 % measured
11% calculated

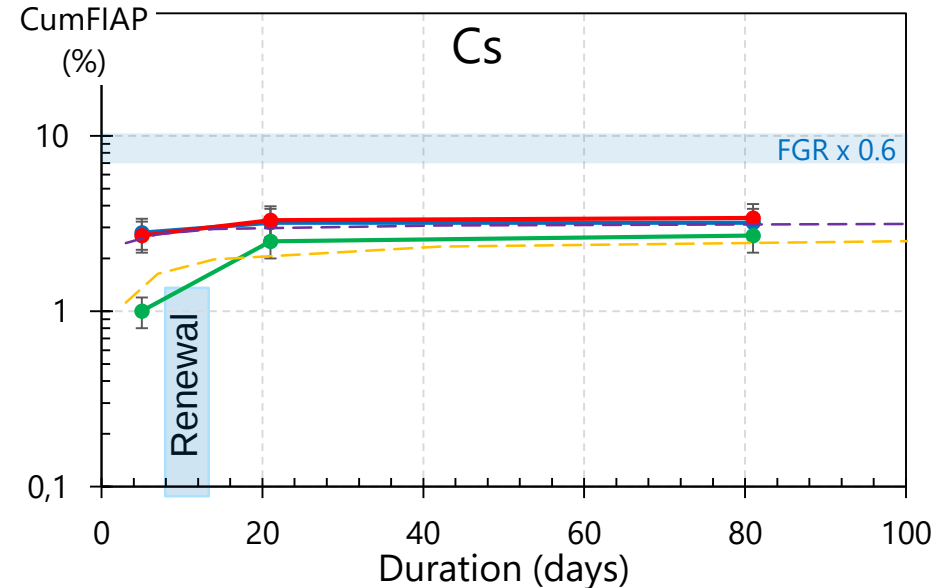
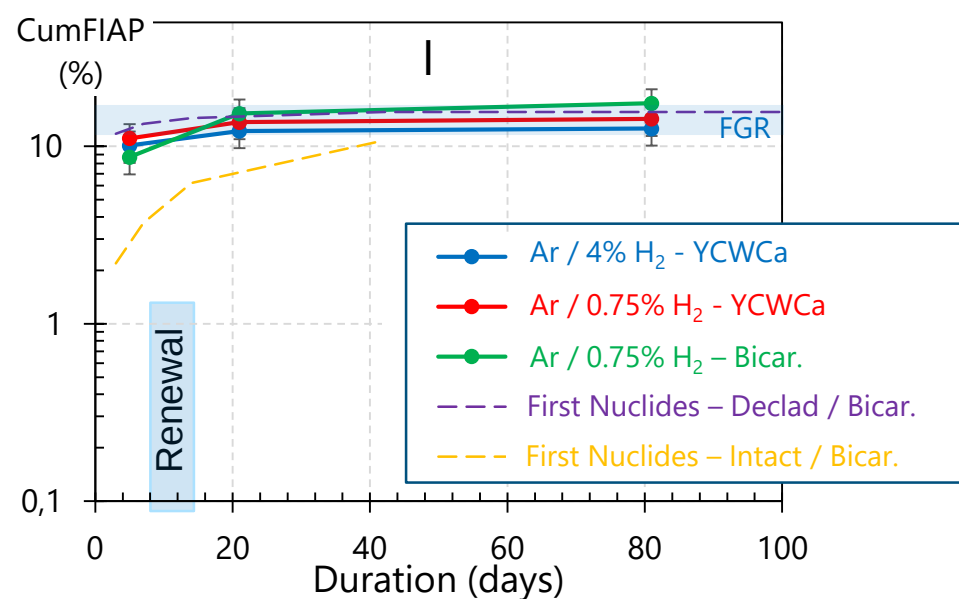


Gas sampling – Kr and Xe evolution



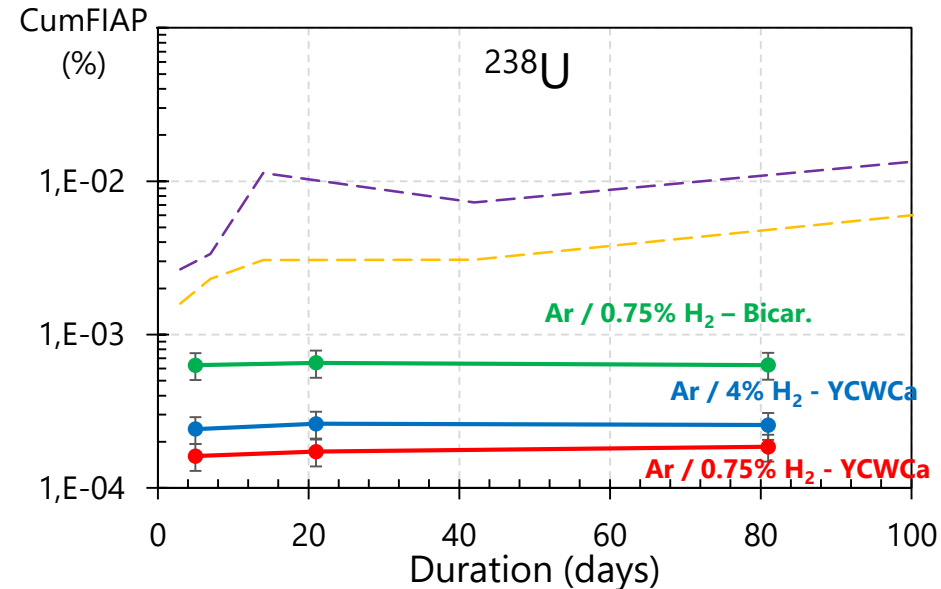
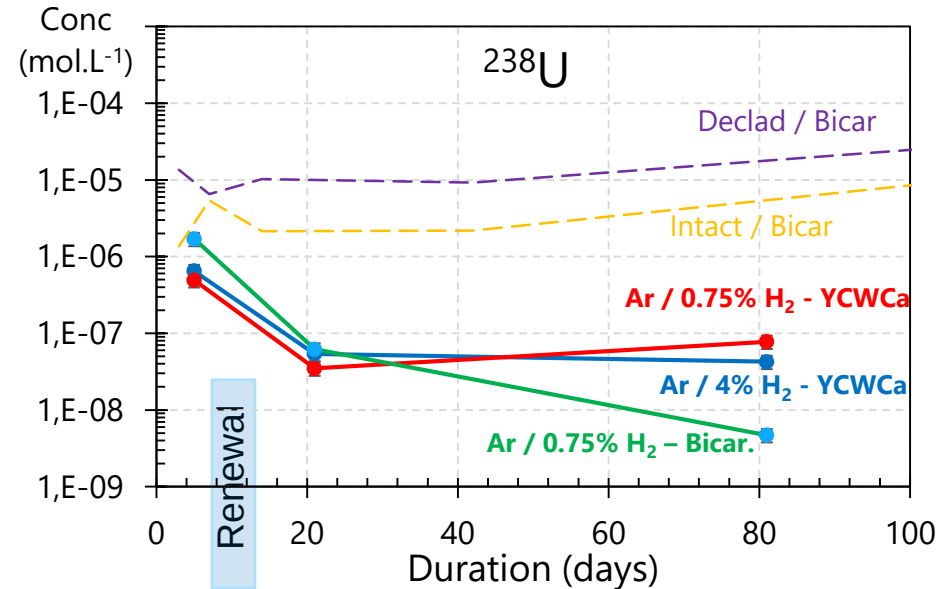
- Continuous release in solution
- Xe/Kr = 15
(Around 9 for a standard UO₂ fuel)

Comparison Cs and I with the FGR



- The pH or presence of H₂ doesn't affect the I and Cs release
- For Iodine, the CumFIAP is close to the FGR, around 14 % measured (11% calculated)
- For Caesium, the CumFIAP is lower than 60% FGR (often taken as expected theoretical maximum)

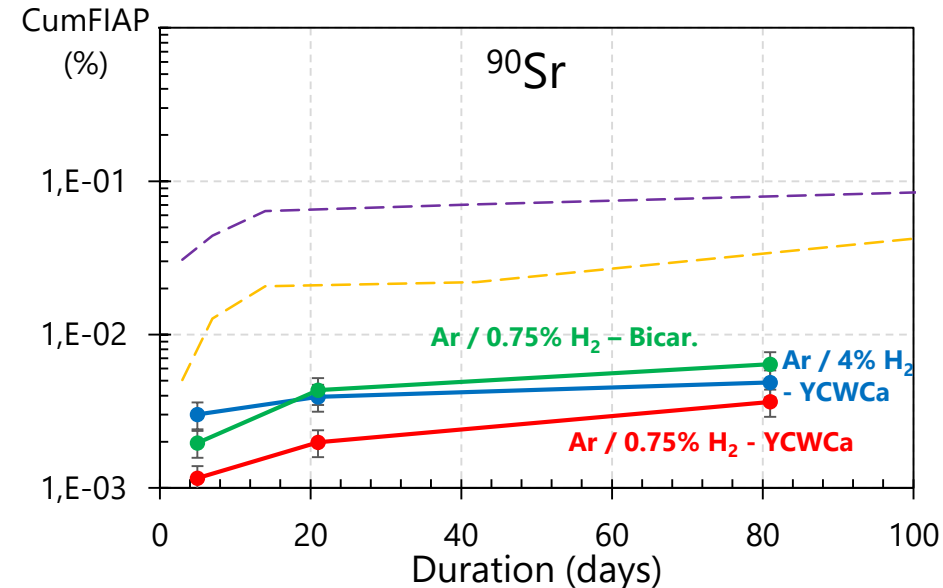
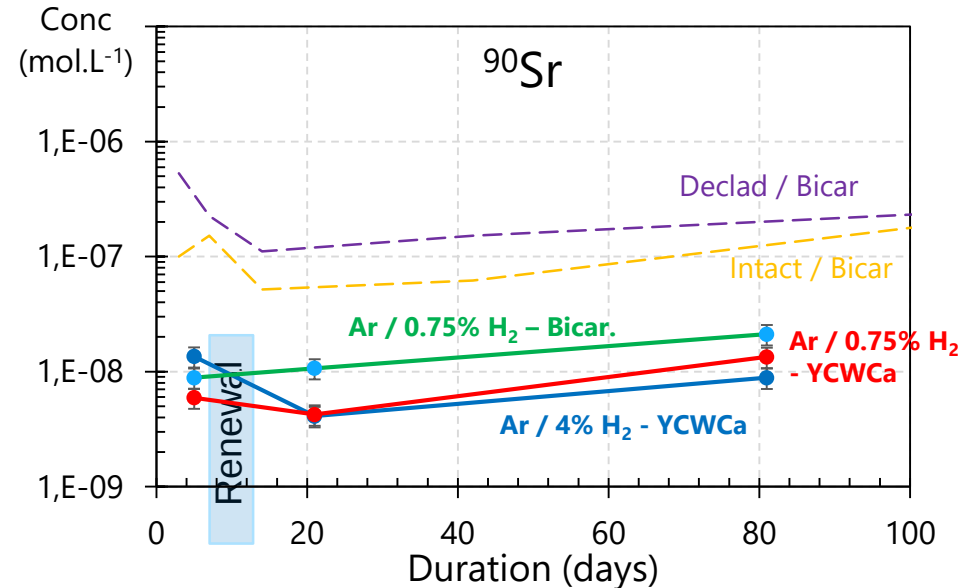
Matrix dissolution - Evolution of U and Pu



- High initial release of uranium (dissolution of the oxidized surface layer)
- Stabilization of the uranium concentration ($3 \times 10^{-8} - 8 \times 10^{-8} \text{ mol.L}^{-1}$)
- [U] are 3 orders of magnitude lower than those in experiments without H₂
- All [Pu] are below the detection limit of the analytical technique ($< 10^{-9} \text{ mol.L}^{-1}$), whereas they were in the range $2 \times 10^{-8} - 2 \times 10^{-7} \text{ mol.L}^{-1}$ in absence of H₂ gas
- Indication of the tightness of the autoclaves

FRR	(year ⁻¹)
4%H ₂ - YCWCa	3.1×10^{-6}
0.75%H ₂ - YCWCa	2.2×10^{-6}
0.75%H ₂ - Bicar	$< 7.7 \times 10^{-6}$

Sr evolution



- No fast release in the first 5 days, compared to I and Cs
- Slight increase of the concentration whatever the experimental conditions
- 1 order of magnitude lower with H₂ than without H₂
- 1 order of magnitude higher than CumFIAP of uranium

FRR	(year ⁻¹)
4%H ₂ - YCWCa	5.9 x 10 ⁻⁵
0.75%H ₂ - YCWCa	4.4 x 10 ⁻⁵
0.75%H ₂ - Bicar	7.8 x 10 ⁻⁵

- **Continuous release of the fission gases** during the leach experiment (cfr. tests by KIT/INE for First Nuclides)
- **Little effect of test medium** (pH, carbonate concentration) and **H₂ pressure** on Iodine and Cesium release
 - CumFIAP(I) $\approx$ FGR, CumFIAP(Cs) < 0.6 FGR
- **With H₂**, uranium concentrations are **2 or 3 orders of magnitude lower than without H₂**
 - Decreasing the H₂ pressure from 1.6 bar to 0.3 bar has no significant effect
- The trends should be confirmed by the longer durations.

Thanks to...



... and to you for your attention