Neutron irradiation of graphite in fuel salt

ORNL MSR workshop 22 July 2022

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NRG

Nuclear. For life.

Contents

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NRG program overview

- **Sponsored by the Dutch Ministry of Economic Affairs as part of a broader Nuclear Energy R&D program**

- **In collaborations with JRC, TU Delft and CV Rez**

- **Program objective: contribute to MSR technology development and realisation**
  - Obtain **operational experience**
    - salt handling, liquid fuel irradiation
  - **Qualify** materials and fuels
    - Temperature, Radiation, Corrosion
  - Study **fission product behavior** (normal and accident conditions)
  - Tackle **waste issues**
  - Work towards **Integral Demonstration**

Ministry of Economic Affairs of the Netherlands

JRC EUROPEAN COMMISSION

TU Delft Technische Universiteit Delft

Centrum výzkumu Řež s.r.o.
Research Centre Řež

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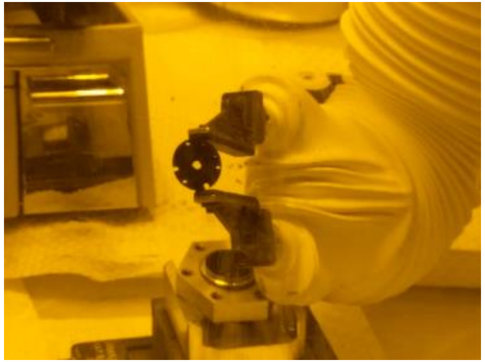
Graphite / fuel salt activities

- **Neutron irradiation of fuel salt in graphite crucibles (SALIENT-01)**
- **Development graphite impregnation set-up**
- **Other activities within the MSR program**
  - Neutron irradiation of alloys under inert and fuel salt conditions
  - Gamma irradiation of fuel salts
  - Modelling > SPECTRA / CFD



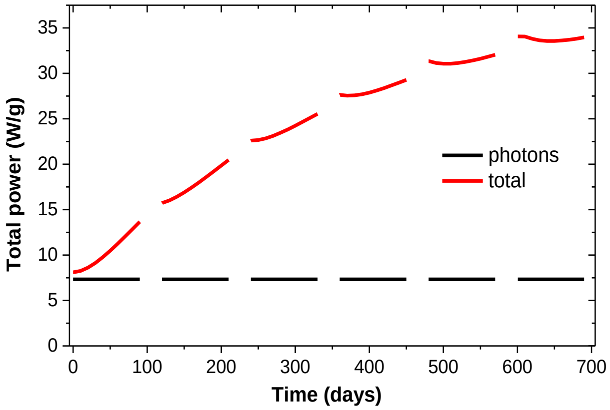
SALIENT-01 objectives

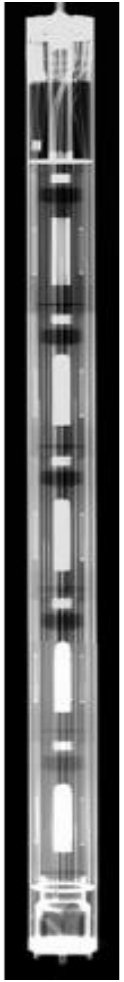
- Confirm claims of good fission product retention in the salt
- Obtain size distributions for noble metal particles
- Assess interactions between fuel salt and fine-grained nuclear graphite, as well as possible uptake of fission products by the graphite
- ‘Stepping stone’ to more complex irradiations with e.g.: salt buffering, redox control, fission gas removal through helium bubbling, and salt flow

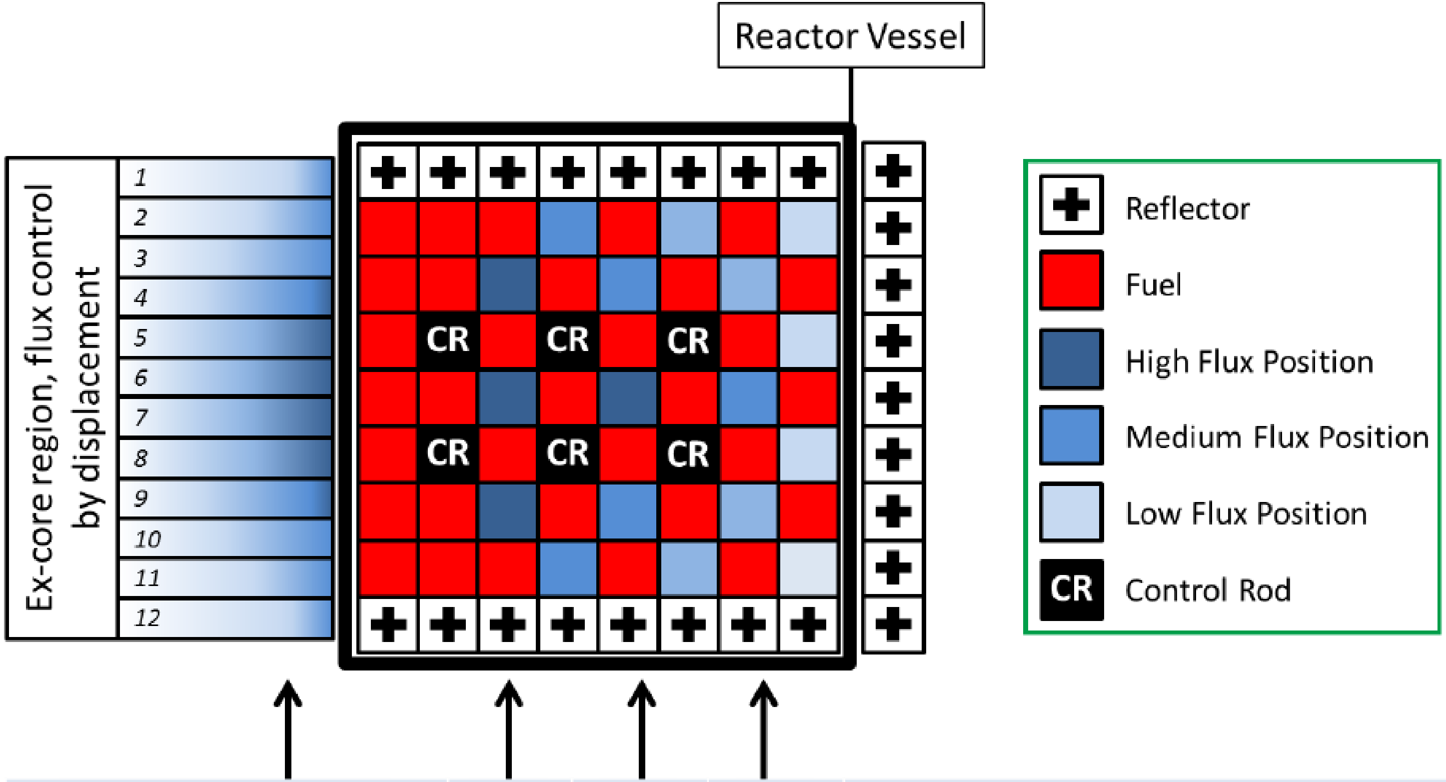
See also: Hania et al, Nuclear Engineering and Design 375 (2021) 111094

SALIENT-01 Design

- Irradiation of 78LiF-22ThF_4 salt
- 5 open capsules fabricated from nuclear-grade graphite (4 loaded) in containment with He-Ne mixture
- Fuel power rises during irradiation due to production of U-233
- Fixed crucible temperature (~600 °C) actively maintained during irradiation





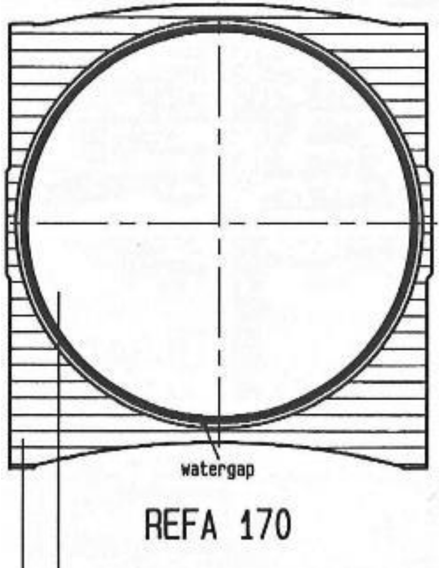


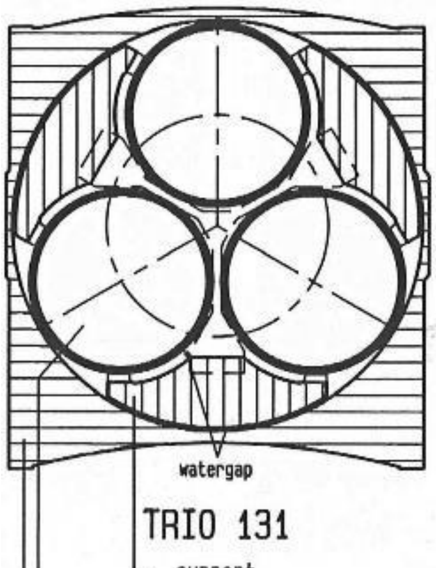
0-3, location dependent	5-8	3-5	1-3	Material DPA rate (DPA/year)
0-700, displacement controlled	n/a	500-700	300-400	Linear heat rate (W/cm fresh LWR fuel)

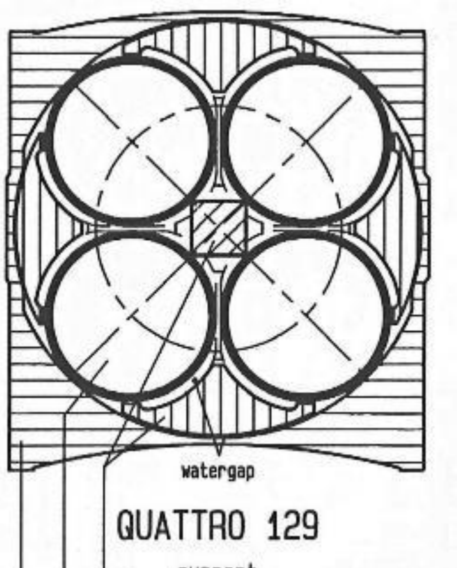
The stable and constant flux profile in each irradiation position is a unique HFR feature

HFR Standard Irradiation Rigs

- Outside is water-cooled, inside gas swept (mixtures of helium, neon, nitrogen)
- Instrumentation throughputs
- Customisation possible
- SALIENT-01 in TRIO facility







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Graphite crucibles

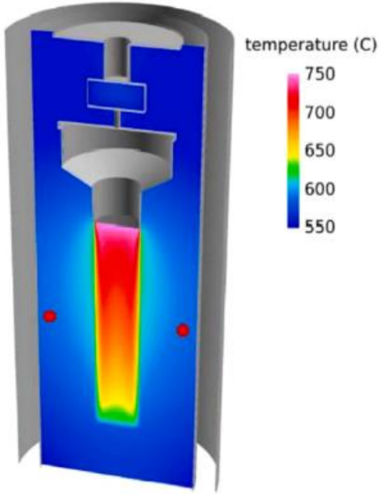
- 2 fine graphite types
- Diameter 20 mm / length 80 mm
- 3 monitorsets to confirm received dose and burn-up
- Holes in circumference for gas lines and thermocouples
- 1 crucible with Ni inner liner to compare deposition on graphite and metal
- Received dose ~4 DPA, linear shrinkage < 2 vol% expected

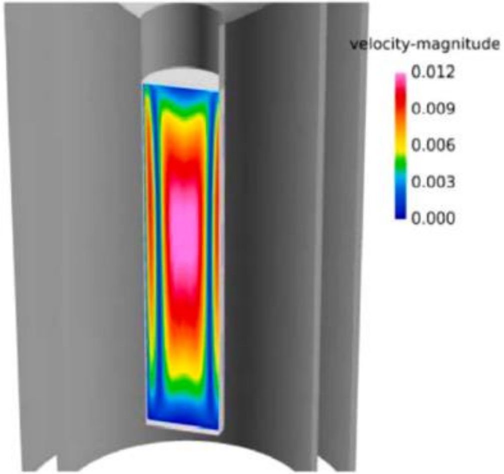
Crucible nr. (bottom to top)	Graphite type	Sample weight (g)	Th-232 wt (g)	Column height* (mm)	Salt column diameter (mm)
L1	PCIB	7.1293	4.1331	36.4	7.6
L2	T-950	6.1344	3.5564	36.4	7.0
L3	PCIB	6.2791	3.6403	36.6	7.0
L4	T-950	7.1743	4.1592	37.1	7.8
L5	PCIB	–	–		–

* Determined at room temperature.

Salt temperature and velocity

- CFD analysis performed to assess salt temperatures and flow velocities
- Assumed salt power of 23 W/g and thermocouple temperature of 590°C
- Temperature variation on surface of crucible ~40°C, flow velocity 12 ± 1 mm/s (upwards in centre and downwards on crucible wall)





SALIENT-01 vs generic MSR conditions

Representative features

- Fine grained graphite in contact with fuel salt
- Irradiation controlled to typical MSR operating temperature
- Build-up of fission products
- Shrinking of graphite with irradiation

Experimental limitations

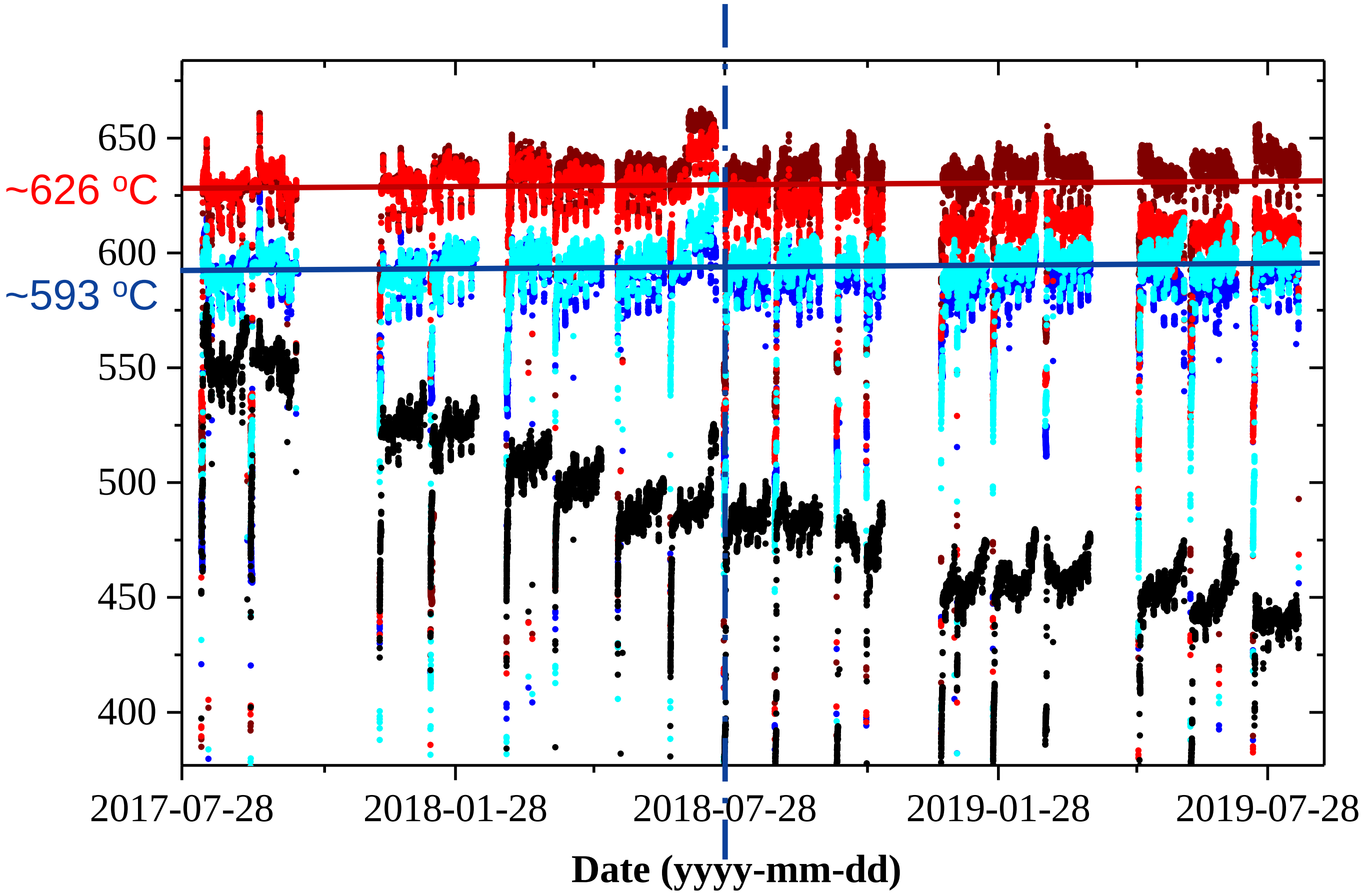
- Limited to ~4 DPA received by the graphite
- Limited to one salt type (78LiF-22ThF_4) with limited flow (up to ~1 cm/s)
- Shutdowns between irradiation cycles with low temperature conditions, potentially allowing fluorine gas production by radiolysis (but no direct effects witnessed with irradiation)
- No redox control

Irradiation (August 2017 – August 2019, 17 cycles)

Measured graphite wall temperatures over 508 full power days
Active temperature regulation of the salt-bearing capsules.

SALIENT-01 was moved to a lower-flux position following cycle 8

Estimated burn-up 1.5-2% FIHMA

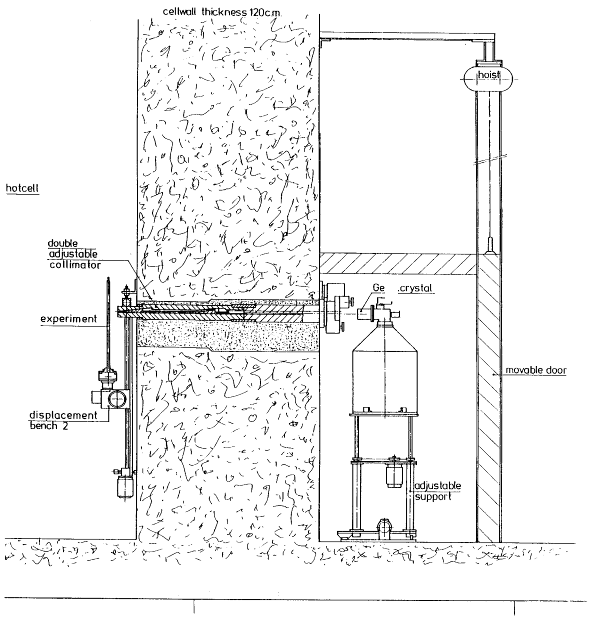


Post-irradiation characterisation activities

- Gamma spectroscopy
- Gas analysis by mass spectrometry
- Scanning Electron Microscopy (EDS / WDS) graphite crucibles
- Transmission Electron Microscopy
- Analysis of dose monitor sets
- CFD analysis to determine salt temperature distribution during irradiation using measured burn-up and temperatures

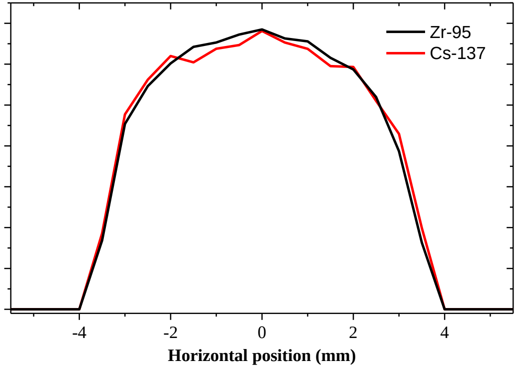
Gamma spectrometry set-up

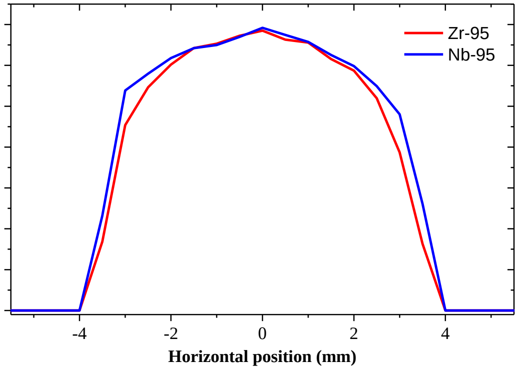
- In-cell displacement bench to place objects for collimator
- HPGe crystal measures the incident gamma rays

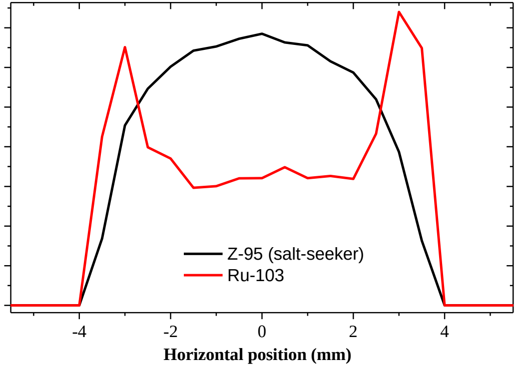


Results gamma spectroscopy

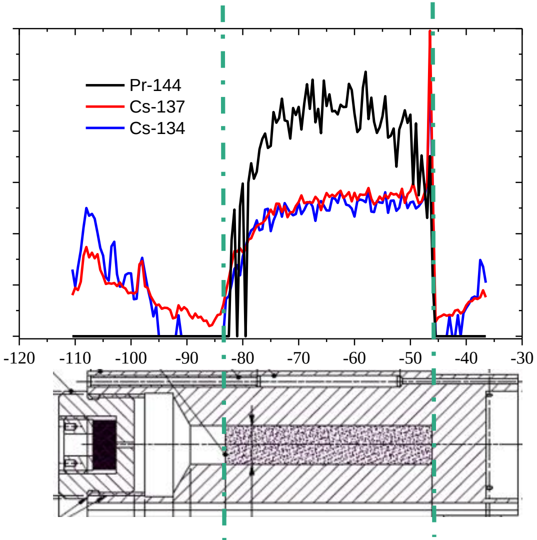
Radial scan
(ID crucible 8mm)

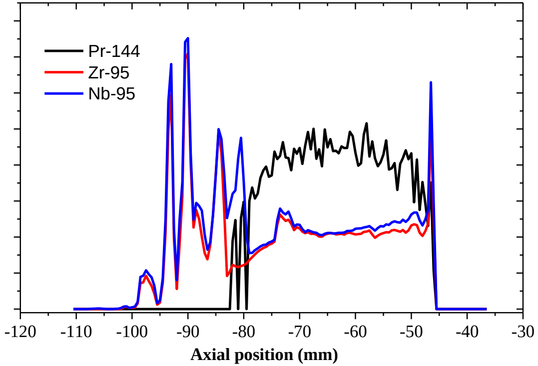


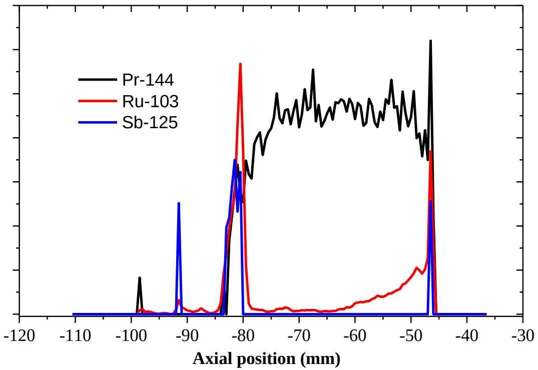




Axial scan
(L crucible 80mm)



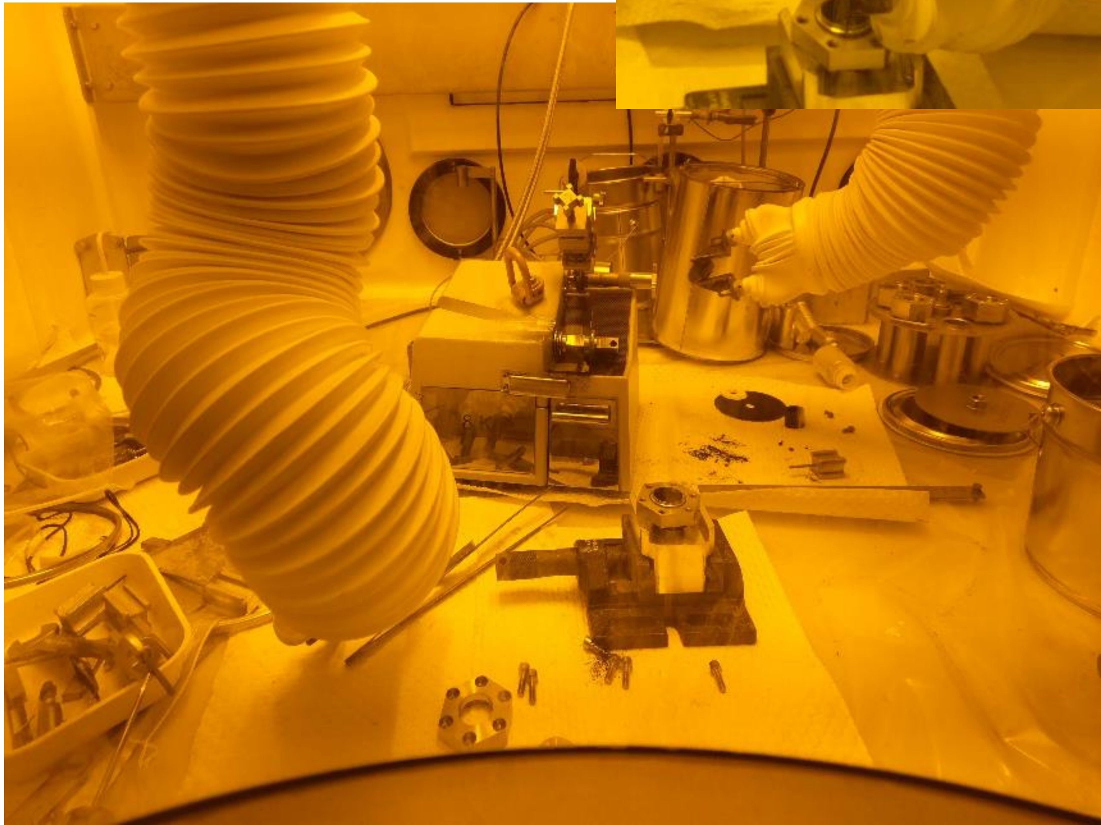




Dismantling of graphite crucibles in glovebox hot cell

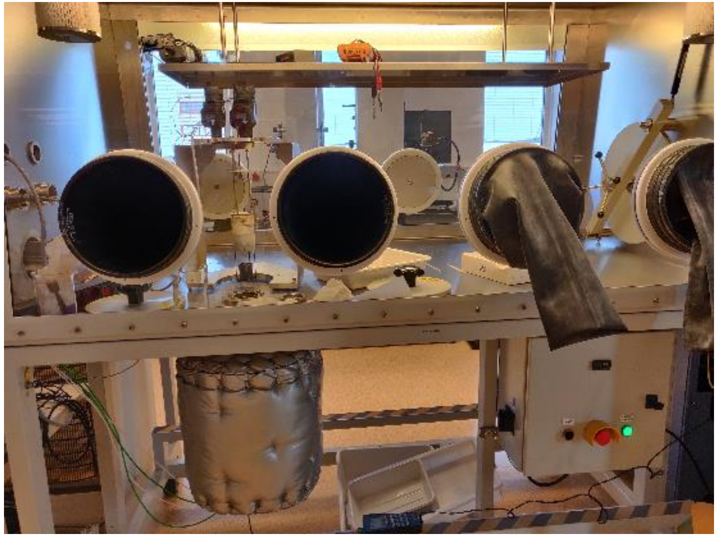
- Alpha hot cell, nitrogen-flushed (O_2 <0.4%, H_2O <200 ppm) for
  - dismantling
  - aqueous chemistry/sampling
    - ICP-MS / ICP-OES
    - Gamma spectrometric analysis
  - high-temperature oven testing (oven in test phase)

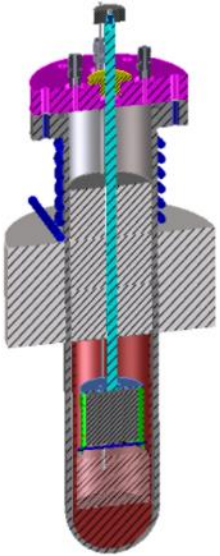
- Alpha hot cell, nitrogen-flushed (O_2 ~1%) for electron microscopy (SEM/EDS/WDS/EBSD)



Development of graphite impregnation set-up

- Set-up placed in glove box for handling of fuel salts and (irradiated) graphite
- Salt bath with Ø50 mm for three cylindrical specimens
- Covers MSR temperature / pressure conditions (max. 800ºC / 8 bar)
- Vacuum and Argon shielding gas
- Design of glove-box impregnation system completed, fabrication in progress

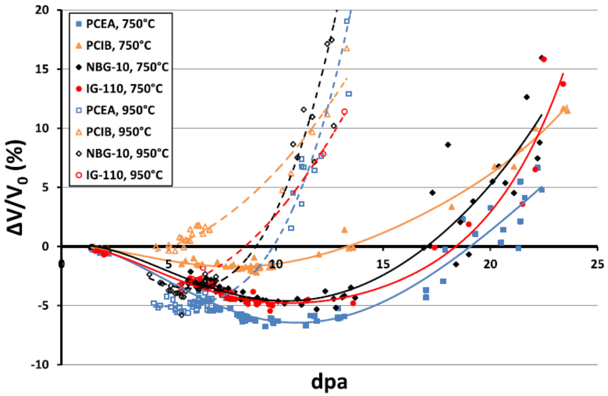


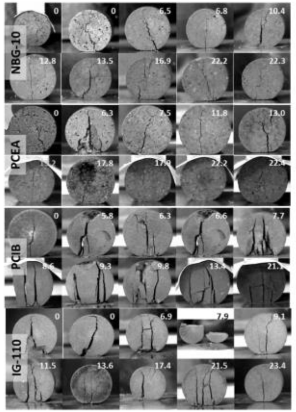


Impregnation set-up measurement plan

- **Impregnation of LiF-ThF_4 into PCIB graphite to compare / complement with SALIENT-01**
  - Virgin graphite
  - Irradiated graphite from SALIENT-01 (and possibly INNOGRAPH) @ 4-5 dpa
  - Determining effect of impregnation time / specimen size
  - Comparison with post-irradiation data from SALIENT-01 (operating at 2.5-3.5 bar, TBD)
- **Compare different virgin and irradiated graphites from INNOGRAPH archive**

See also: Comparison of irradiation behaviour of HTR graphite grades (Heijna, 2017)





Conclusions and next steps

Conclusions

- Successful irradiation of fuel salts in graphite crucibles
- First PIE results available give information on distribution and retention of fission elements

Next steps

- Continue (technically challenging) dismantling
- Preparation of samples for in-cell microscopy (light microscopy and SEM/EDS/WDS)
- Finalise fabrication of impregnation set-up and initiate testing of graphites