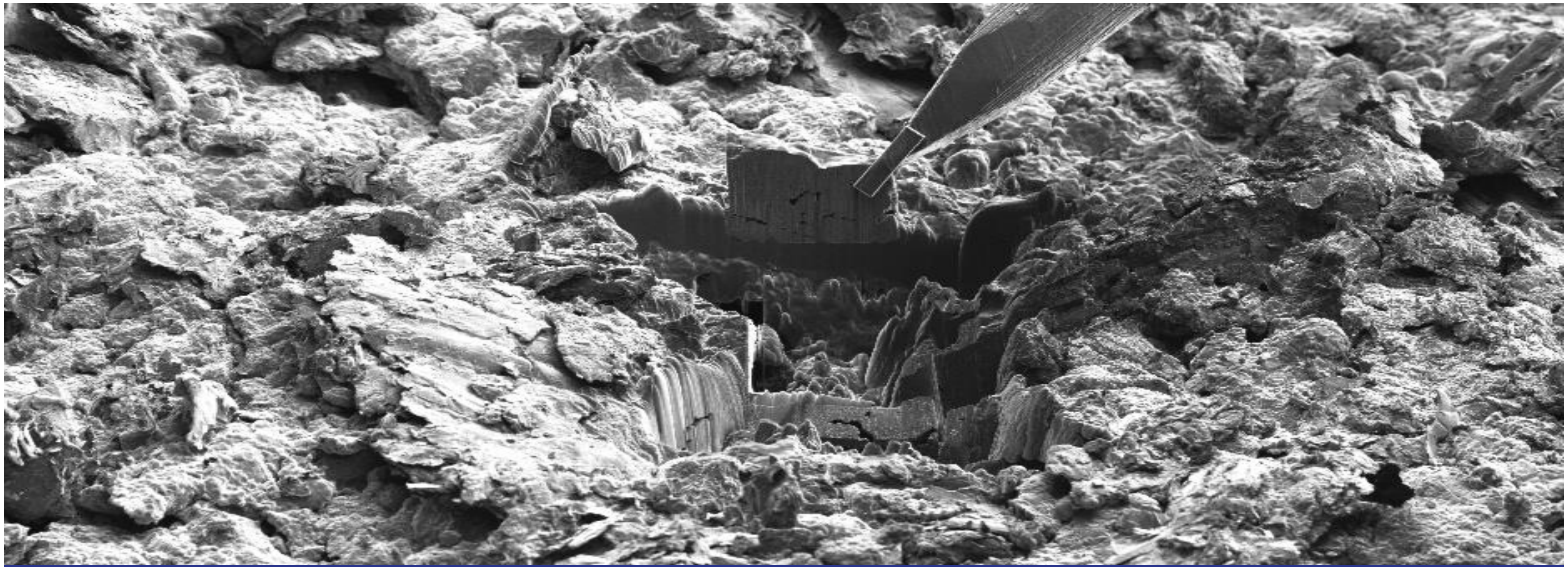


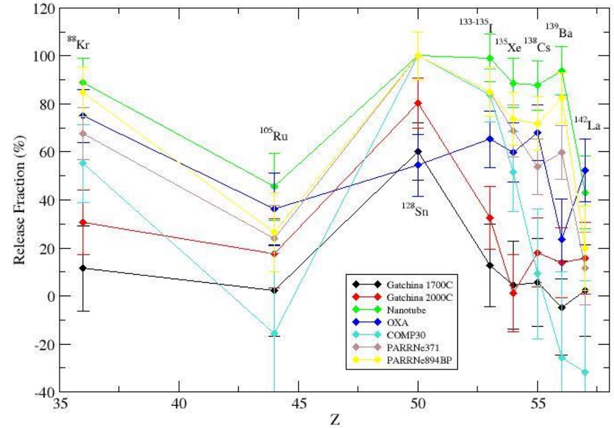
Bye Bye ActILab

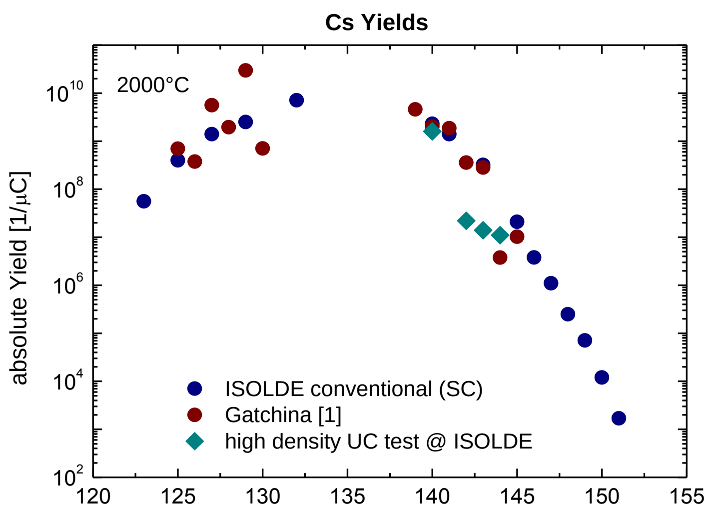
The CERN ActILab Highlights

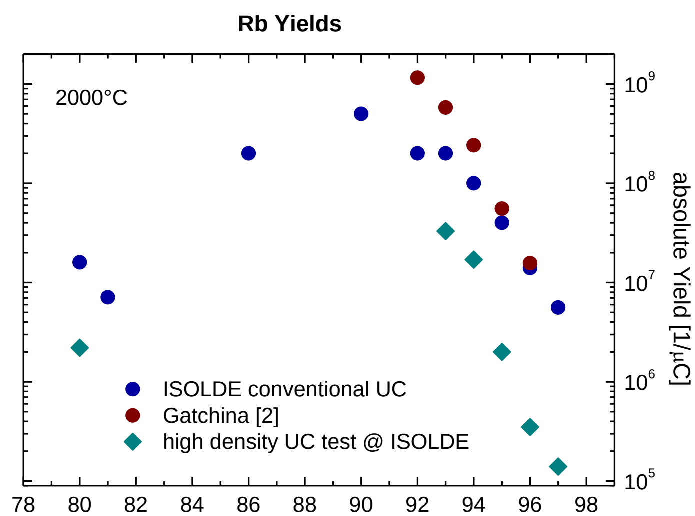
Alexander Gottberg (CERN)

Nominal Density Uranium Carbide (Task 4)

Release fractions from Rabbit samples, ALTO 2013



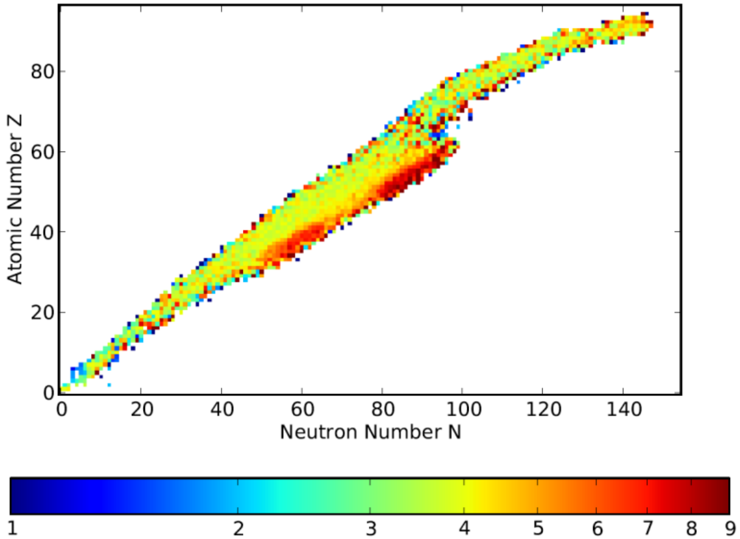




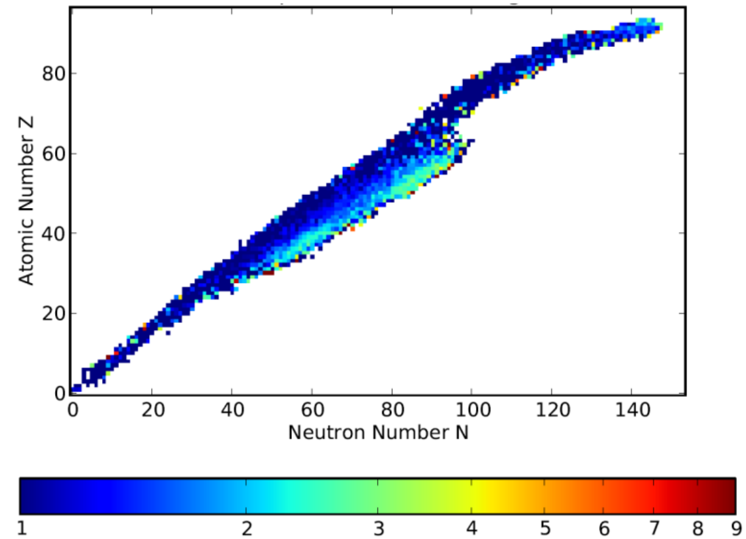
[1] V. N. Panteleev, et al., Eur. Phys. J. A **42**, 495-501 (2009)
[2] V. N. Panteleev, EMIS-15, June 25, (2007)

Nominal Density Uranium Carbide (Task 4)

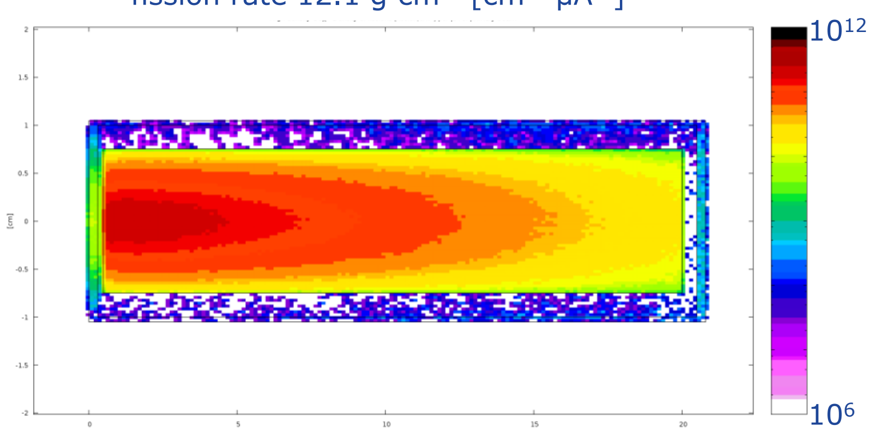
Production yield ratio HD : LD in first pellet



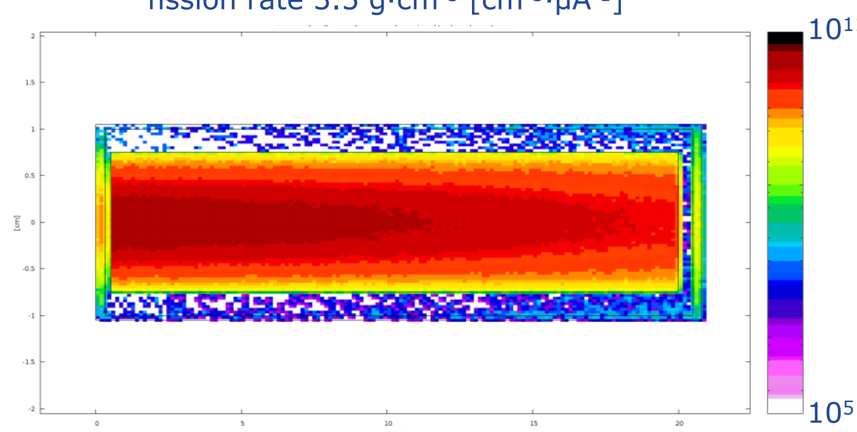
Production yield ratio HD : LD in last pellet



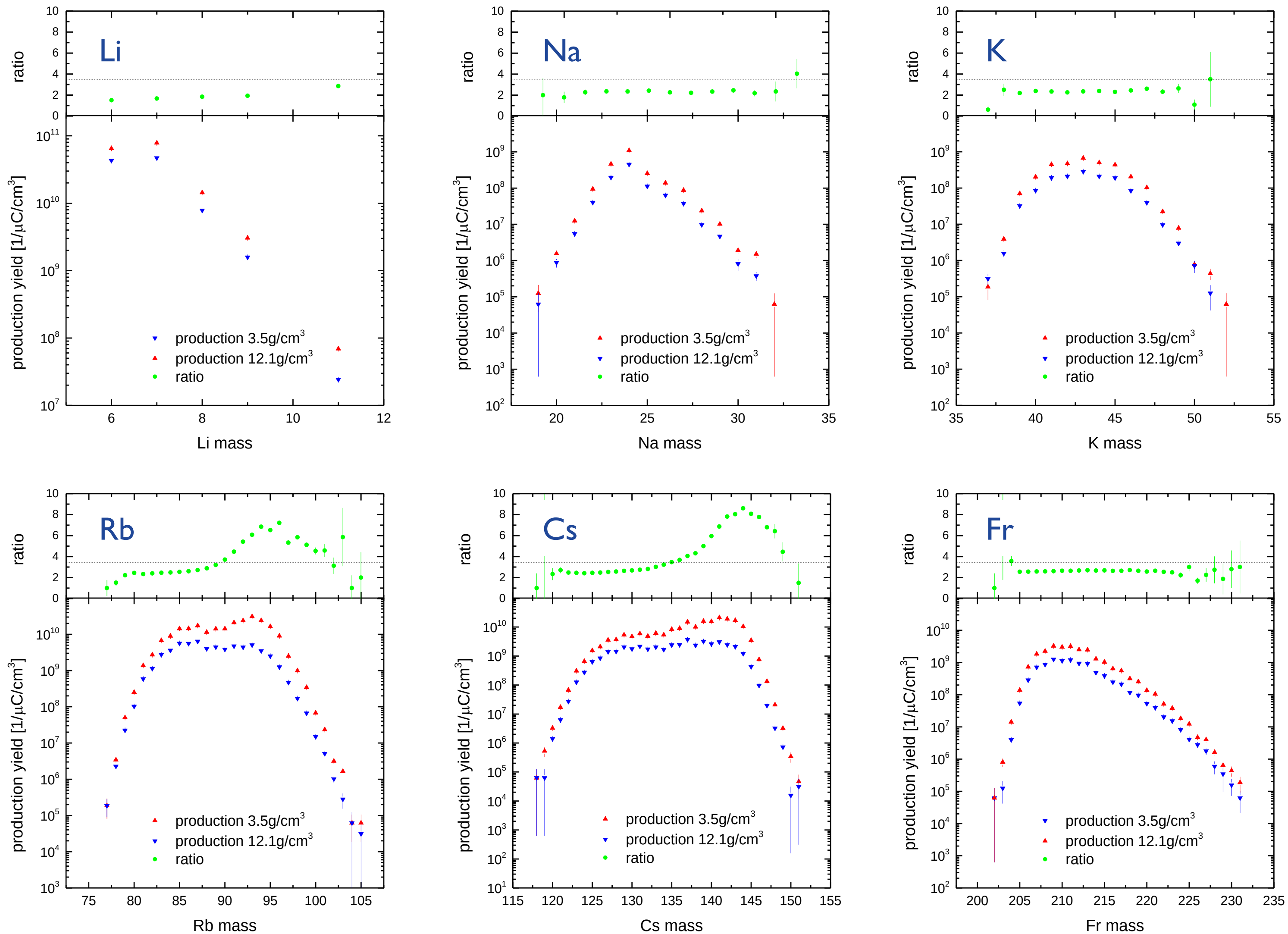
fission rate 12.1 g·cm^{-3} [cm^{-3}·μA^{-1}]



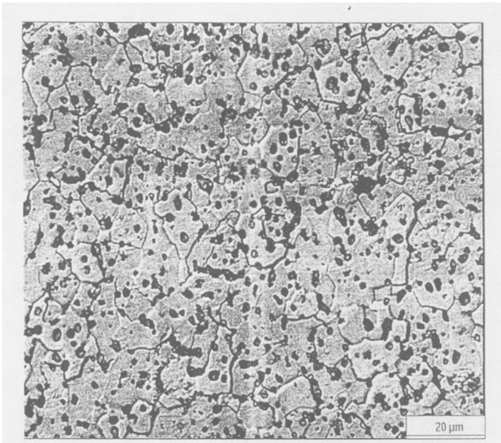
fission rate 3.5 g·cm^{-3} [cm^{-3}·μA^{-1}]



Nominal Density Uranium Carbide (Task 2 + 4)

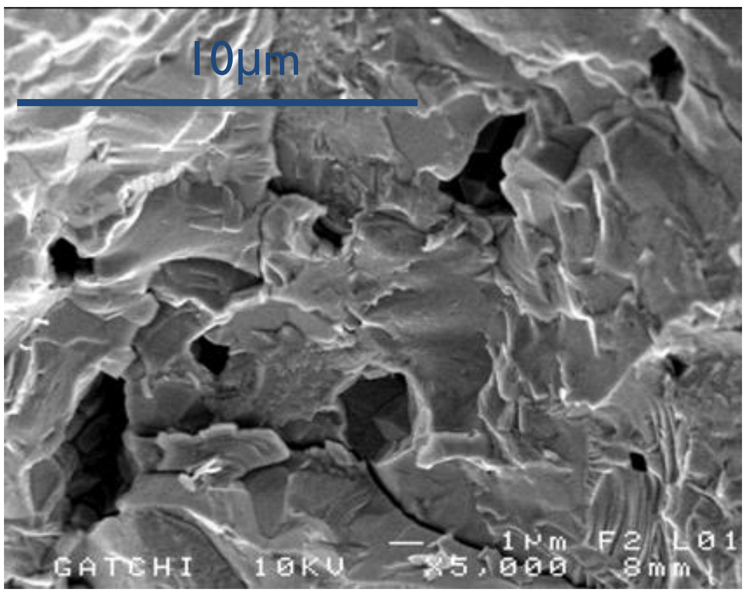


Nominal Density Uranium Carbide (Task 2 + 4)

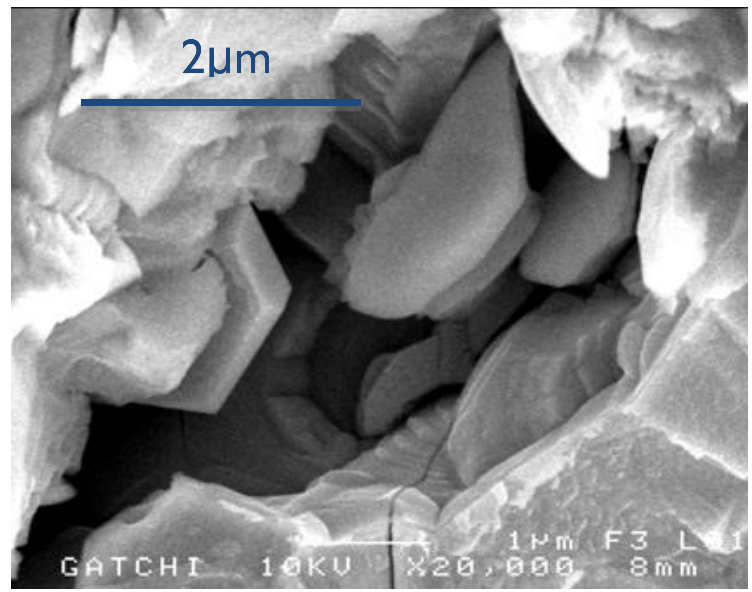


Porosity from picnometry in IPNO (tbc): less than 1%
Geometrical porosity: 0%

But microscopic morphology more complex than anticipated



zoom on the porosities, SE mode



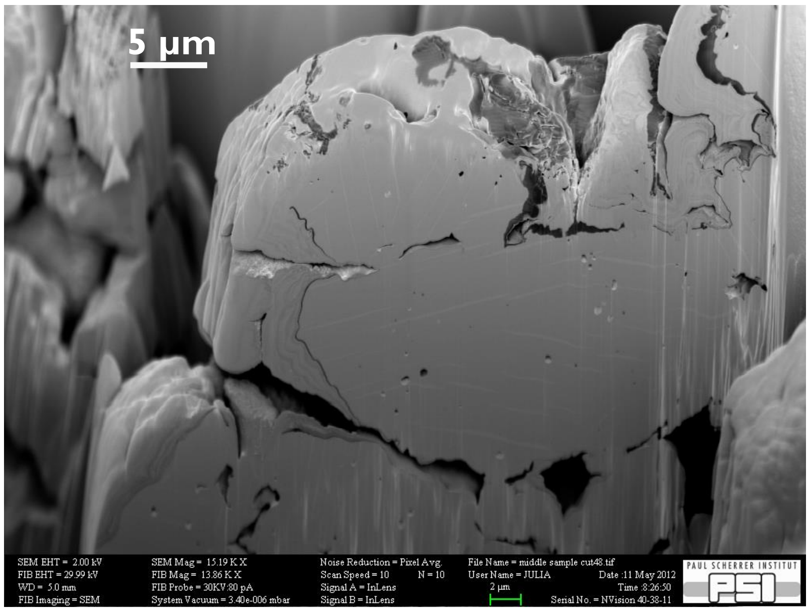
in a pore, on the fracture, SE mode

Sandrine Tusseau-Nenez (IPNO)

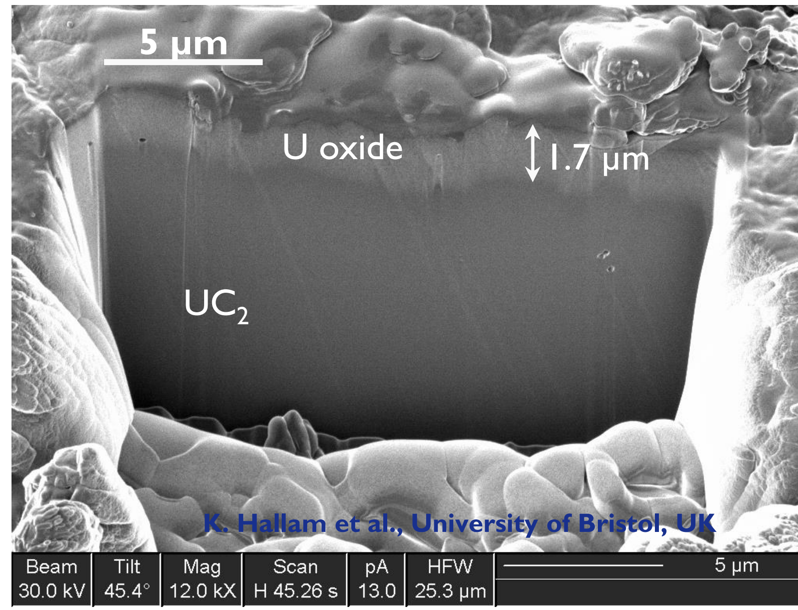
Characterization of UC_X
Task 2

Oxidation on Microscopic Level

As prepared (stored under argon)

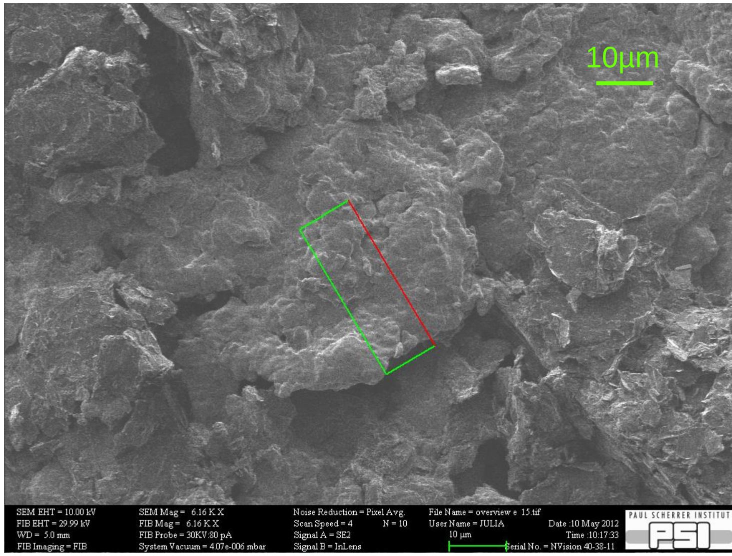


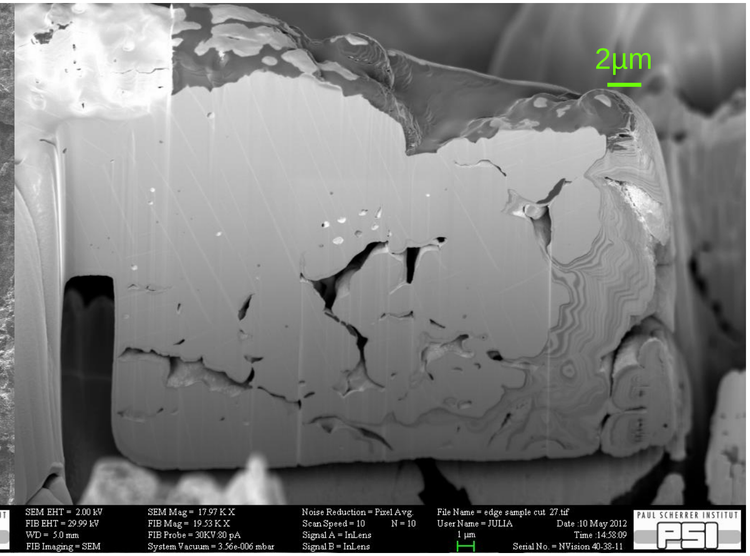
Stored in air



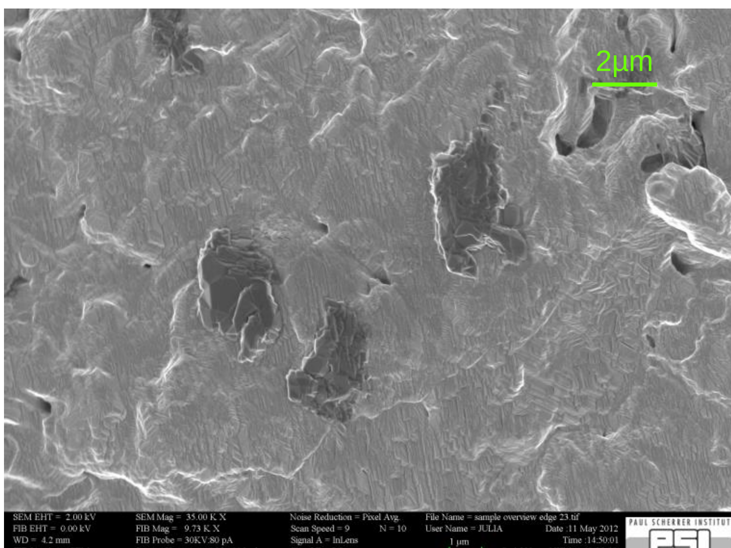
Oxidation process without thermodynamic activation is slow (in conventional UC_x) and follows a surface to bulk mode.

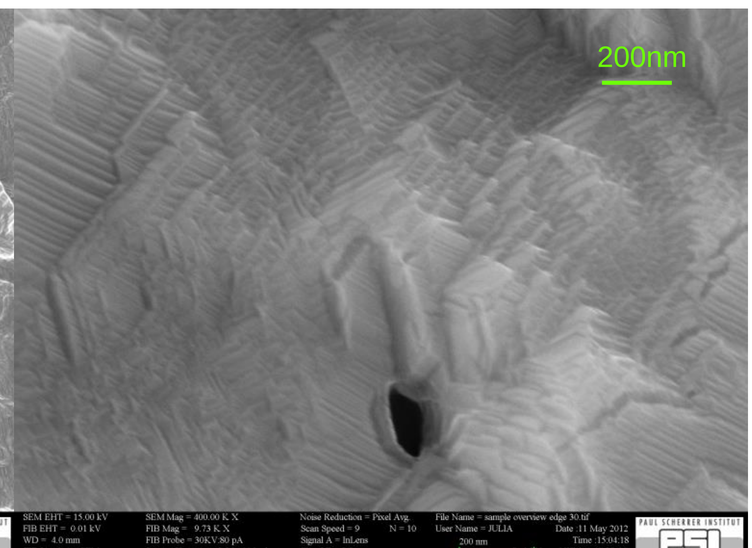
Material Investigation at PSI/SLS (non irradiated)





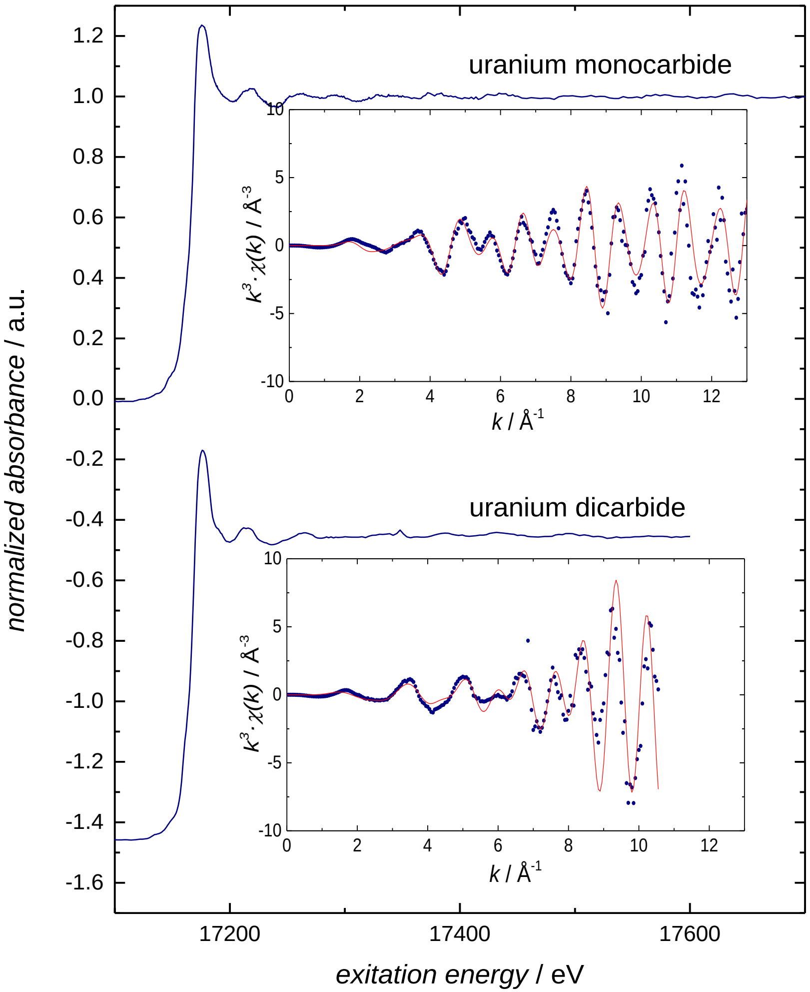
as prepared (≤1850°C)



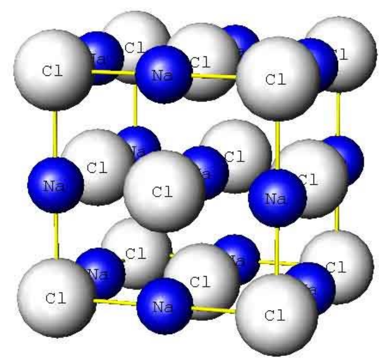


as operated (2100°C for 5 days)

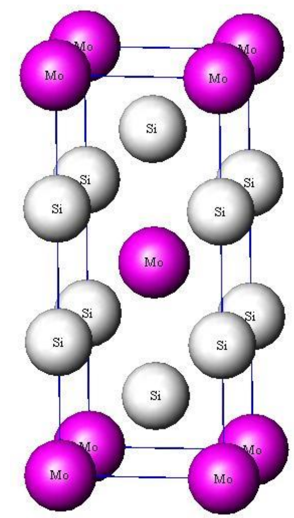
EXAFS for indirect identification of uranium valence at PSI/SLS



- Valence-induced shift of adsorption edge is very small and hart to resolve
- Change in lattice are causing major changes in EXAFS oscillatory features



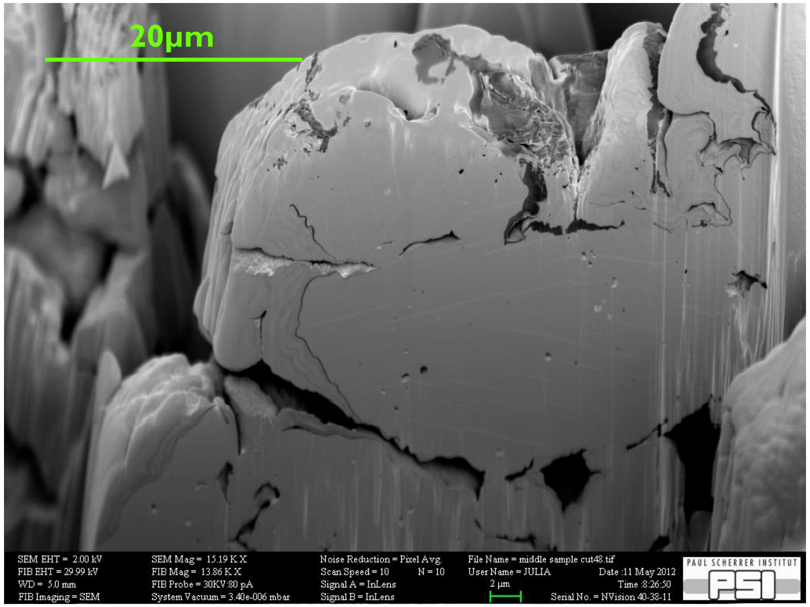
UC (Fm-3m) as NaCl

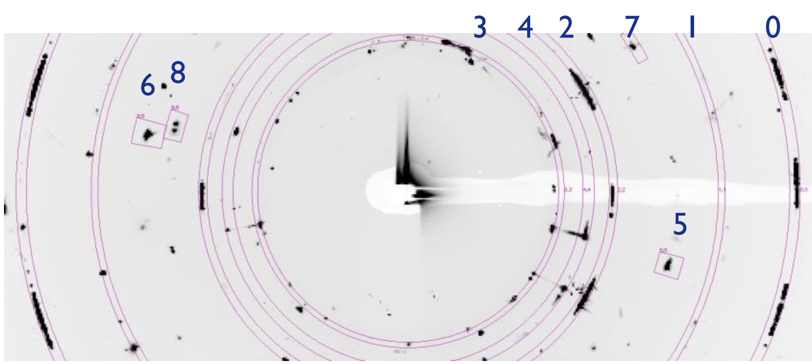


UC_2 (I4/mmm) as $MoSi_2$

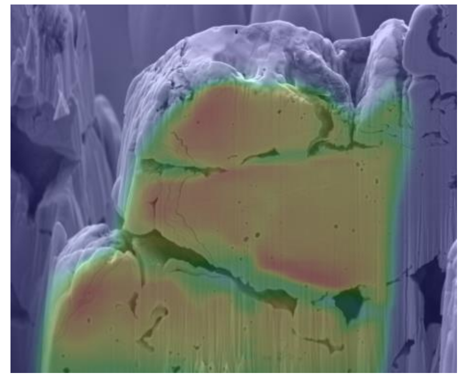
Material Investigation at PSI/SLS (non irradiated)

SEM Uranium

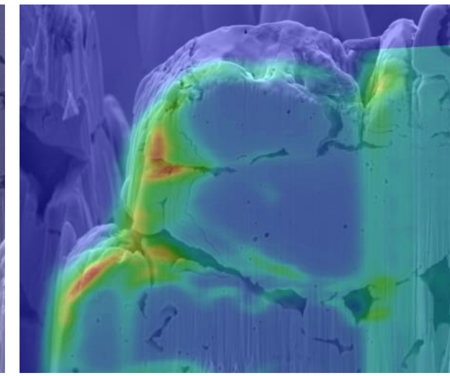




μ-spot X-ray fluorescence mapping

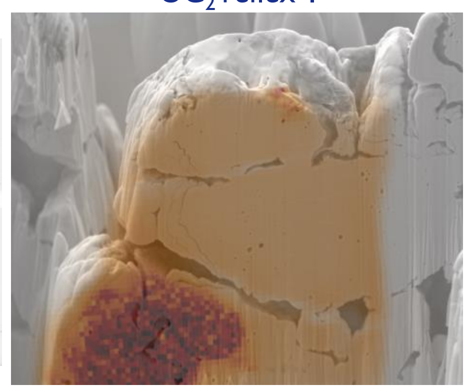


Uranium

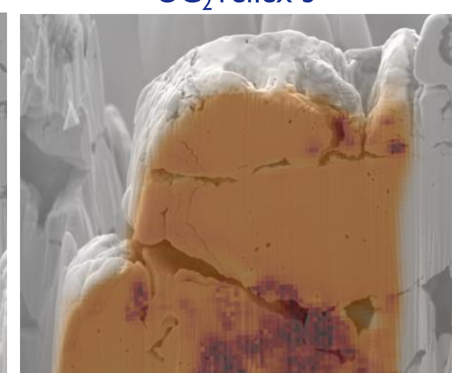


Gallium

UC_2 reflex 1

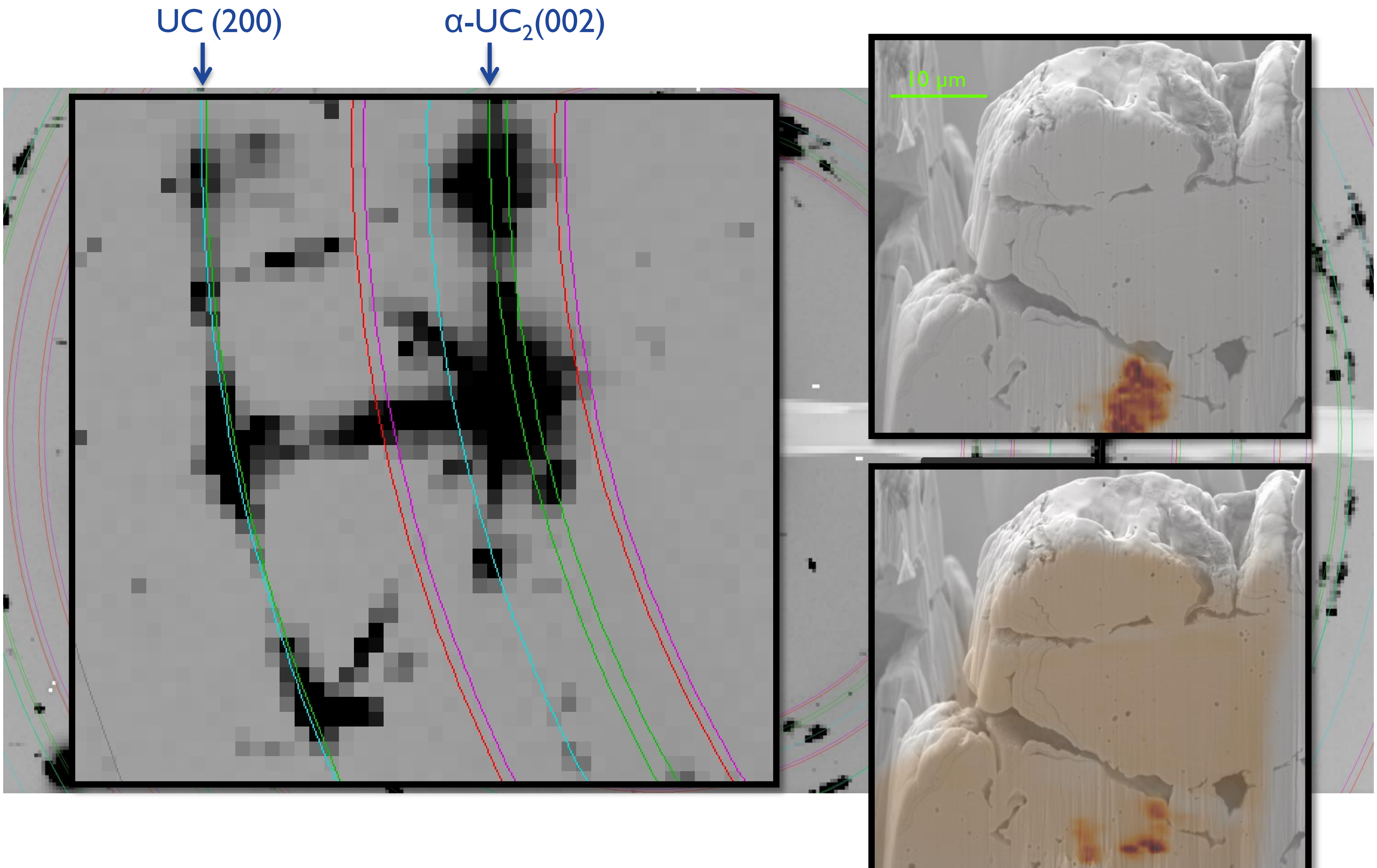


UC_2 reflex 3

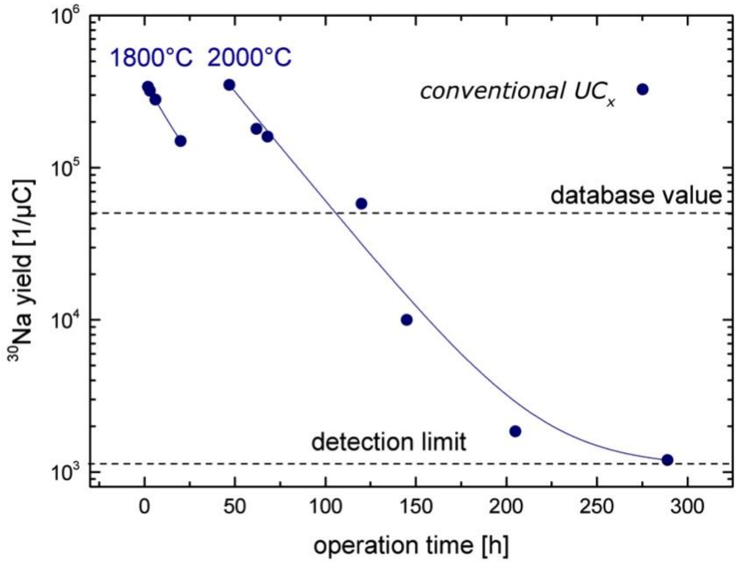


μ-spot X-ray diffraction

Why is UC_x such successful target material?



UC_x Diffusion Study



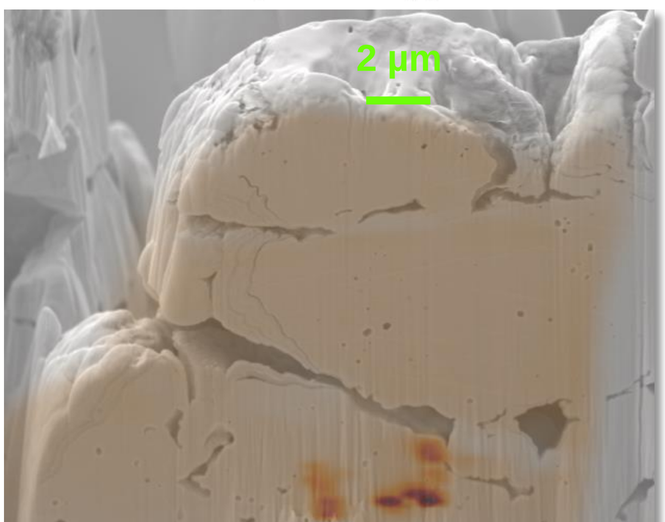
For pure diffusion and short half life:

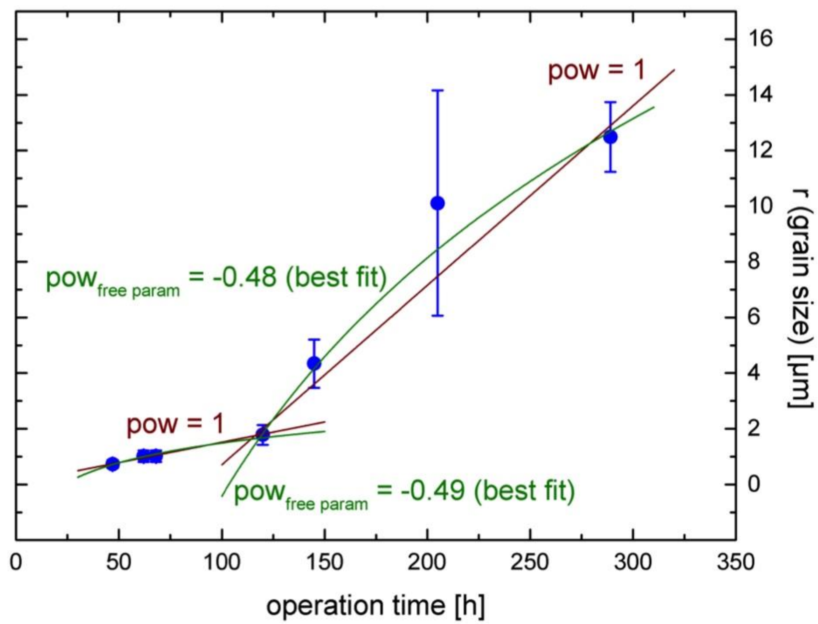
$$\varepsilon_{rel}(T_{1/2}) = \frac{3\left(\sqrt{\pi^2 \lambda/\mu_0}\right)\coth\sqrt{\pi^2 \lambda/\mu_0} - 1}{\pi^2\left(\lambda/\mu_0\right)}$$

$$\mu_0 = \pi^2 \frac{D}{r^2}$$

R. Kirchner, NIM B, **B70**, 186-199 (1992)

→ r^2 / D,
diffusion constant D(T) from initial particle size measurements: D(2300 K) = $6 \cdot 10^{-11}$ cm^2/s



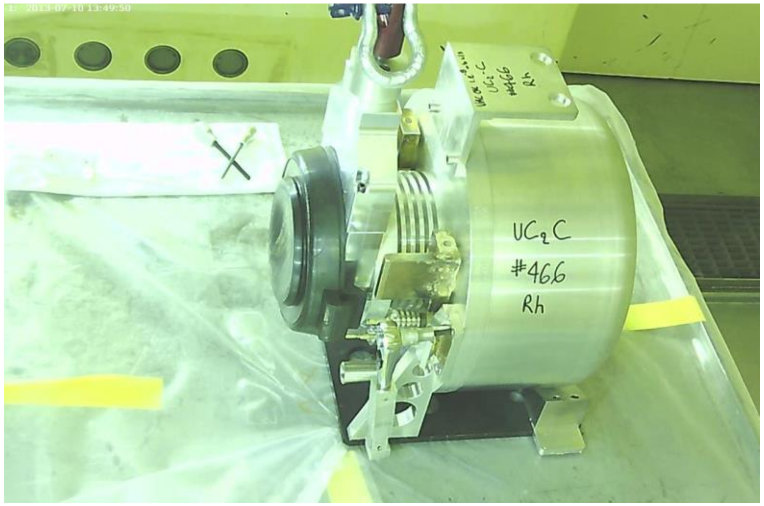


Post-Irradiation Studies
Task 3

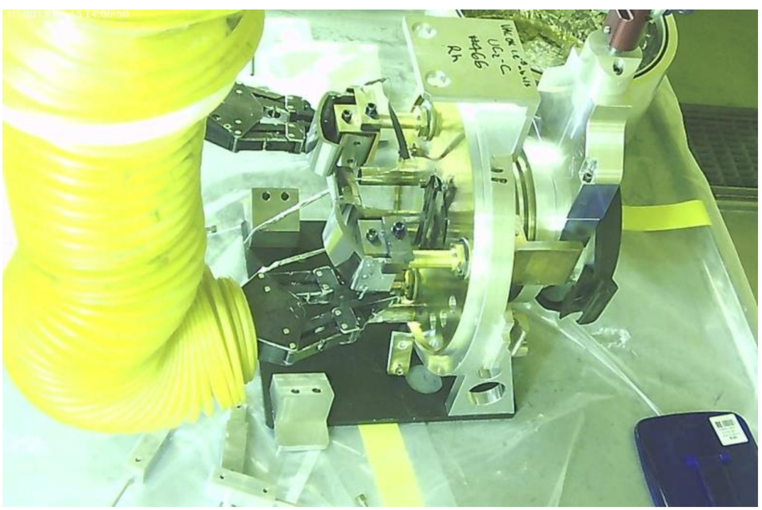
Irradiated UC_x Target Unit 466

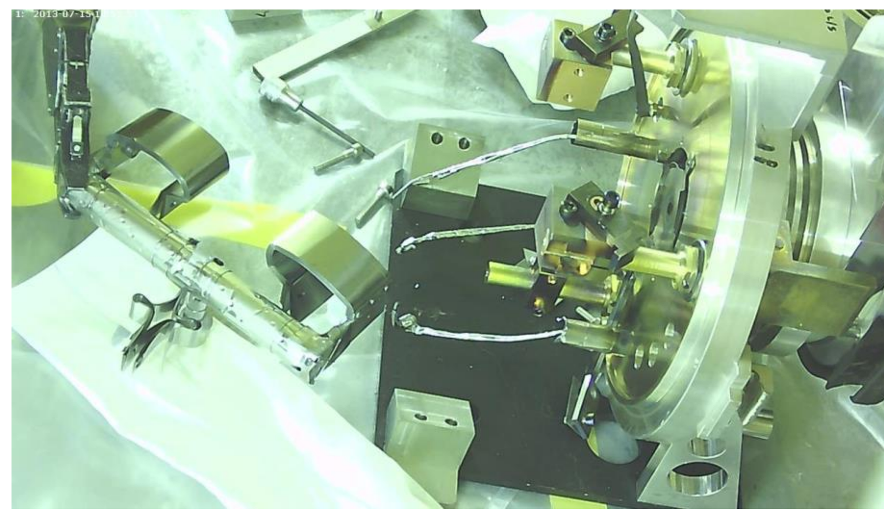
- Irradiation in October + November 2011 on both front ends
- total of $8.8 \cdot 10^{18}$ protons of 1.4 GeV
- Road transport in January 2013 from CERN to Paul Scherrer Institute (270 km)
- 4 mSv/h on contact with parcel

Dismantling UC_x Target Unit 466



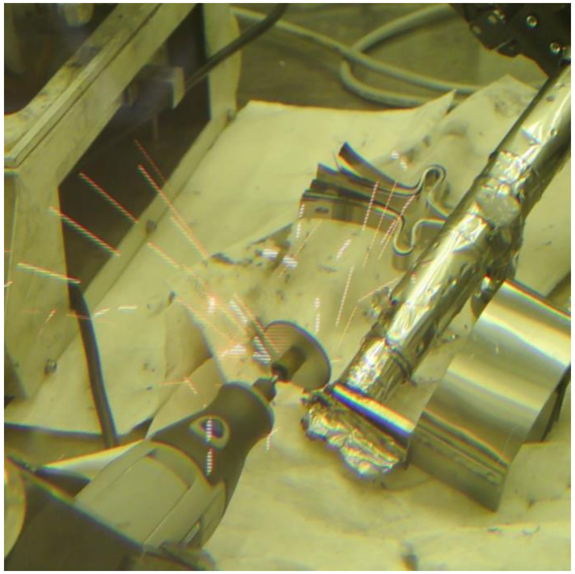
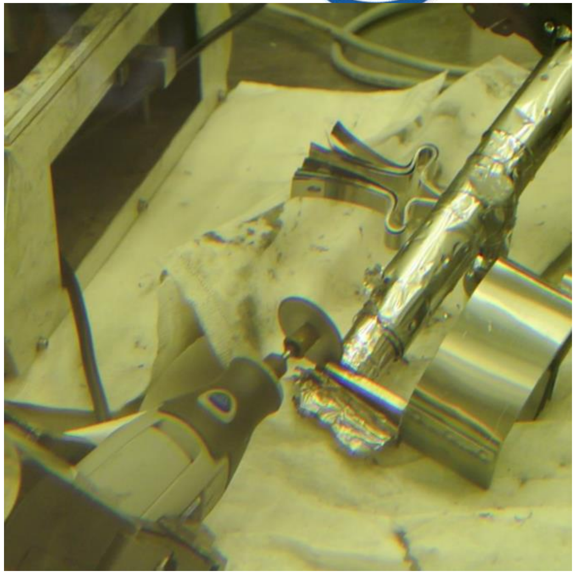
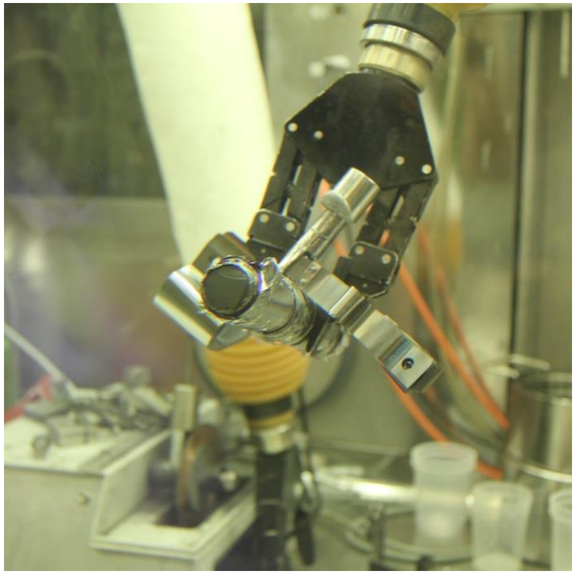
- Opening target vessel in hot cell chain in air (6 mSv/h on contact with Al beam window)
- Extraction of tantalum container (19 mSv/h on contact with Ta proton beam window)
- Sealing of ion source and mass marker outlet with epoxy glue to prevent oxidation of carbide material

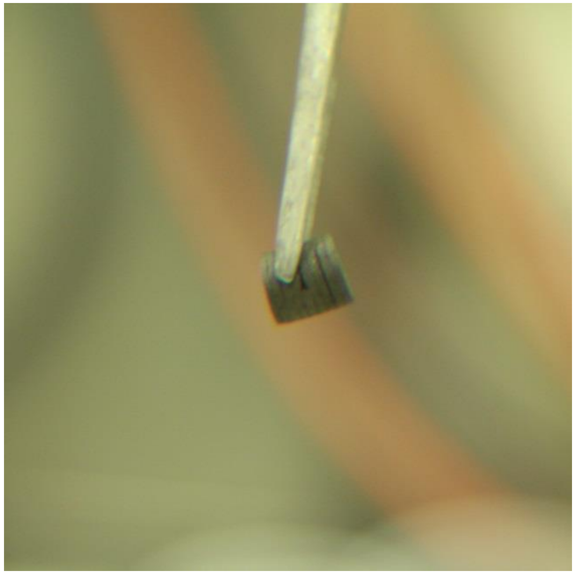




Opening Ta Container of UC_x Target Unit 466

- Transfer of Ta container with UC_X into inert-gas hot cell
- Cutting of sealed container
- Extraction of UC_x for further investigations (500 µSv/h on contact with single pellet)

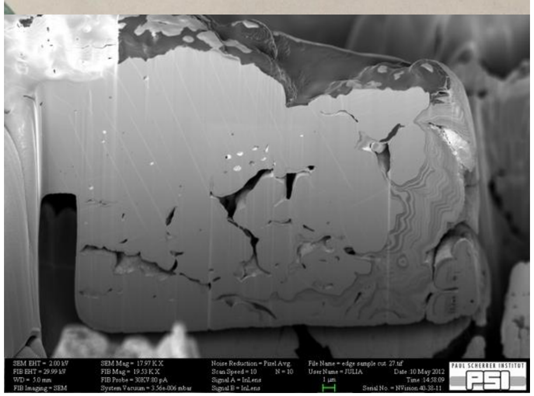


UC_x Post-Irradiation Studies at PSI

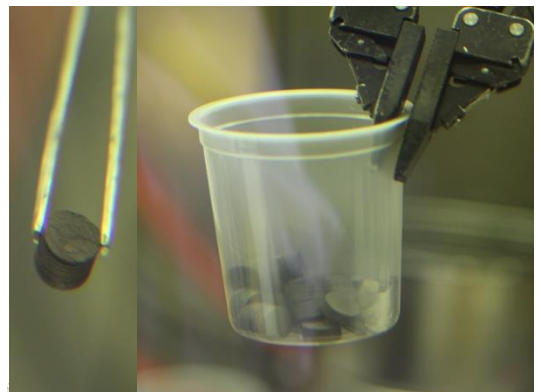
Post-irradiation analysis:

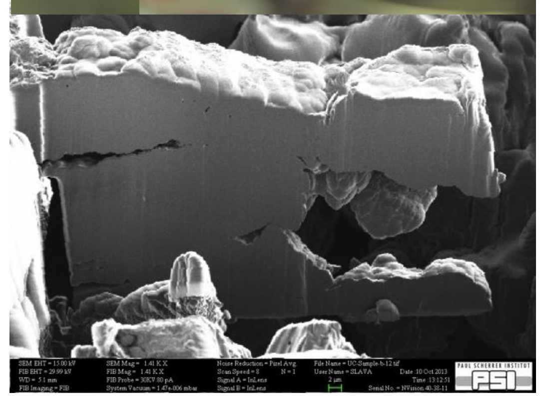
- Pellets appear macroscopically unchanged
- Microscopic evolution of pore distribution and grain size under irradiation observed
- Synchrotron data only partially analyzed

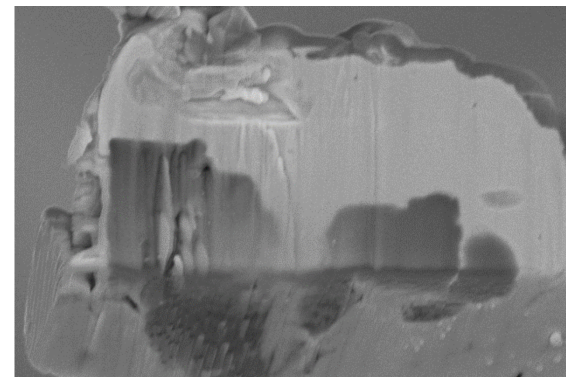
before irradiation

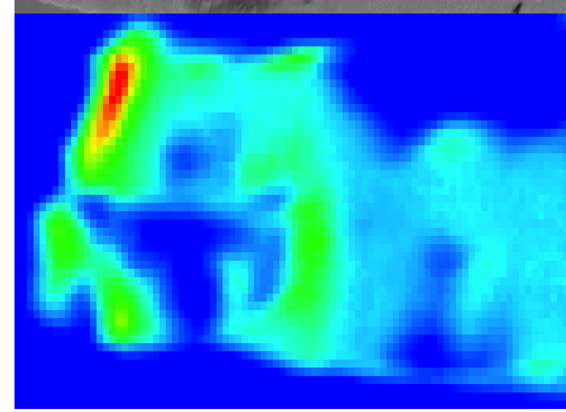


after irradiation

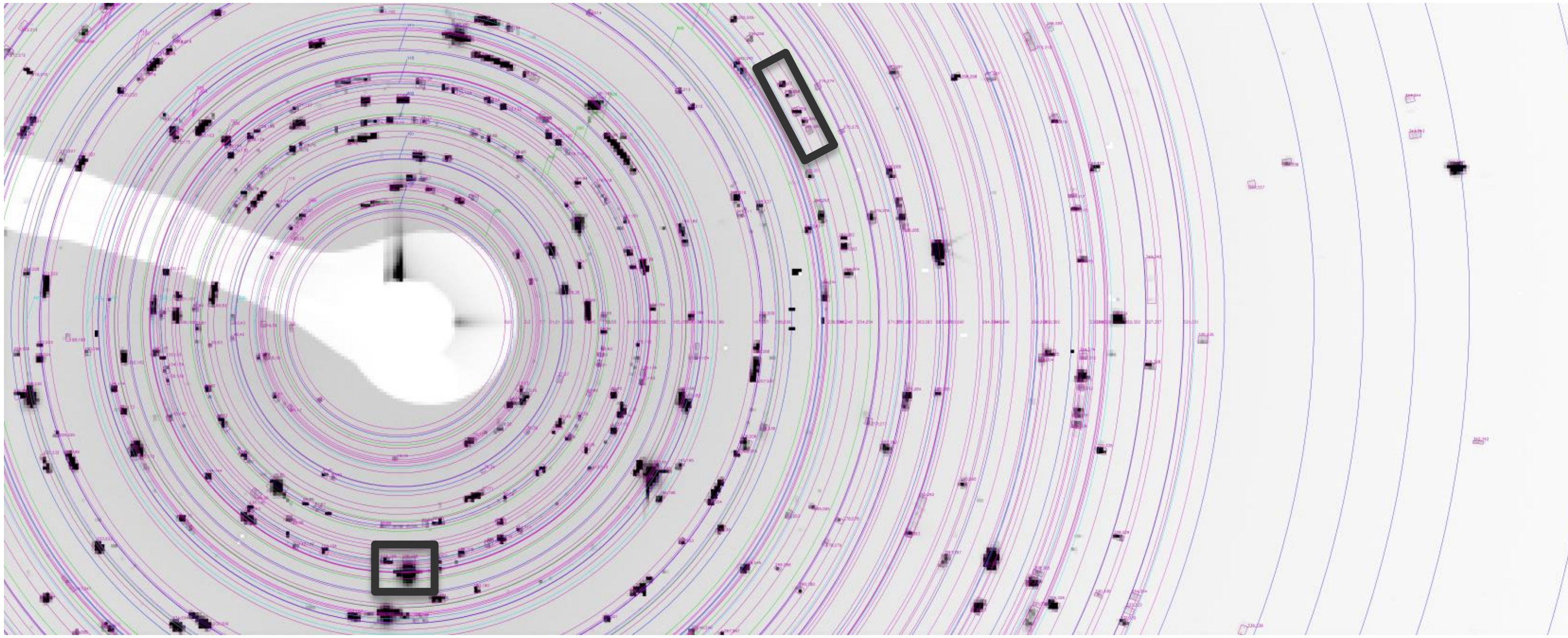


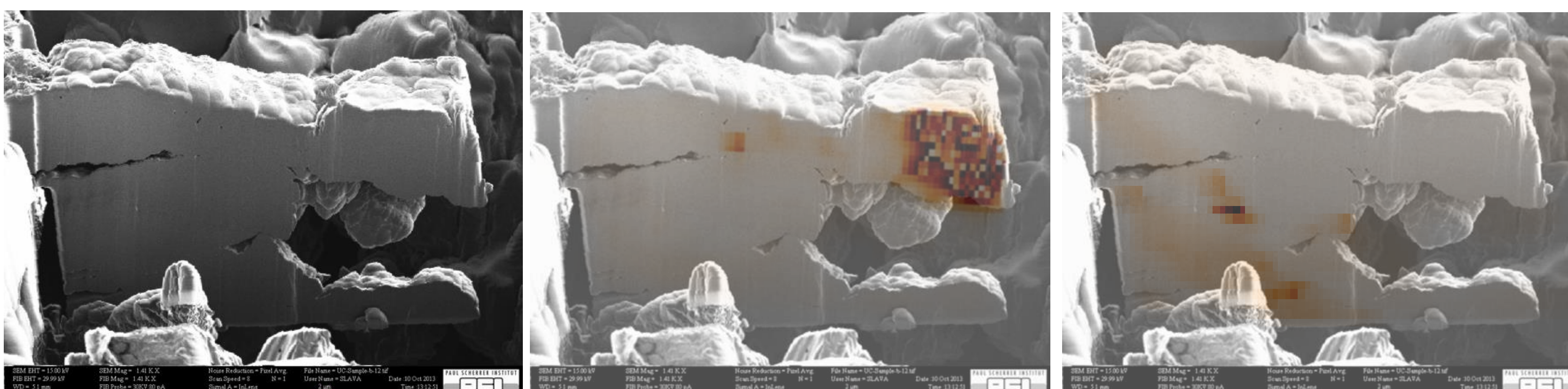






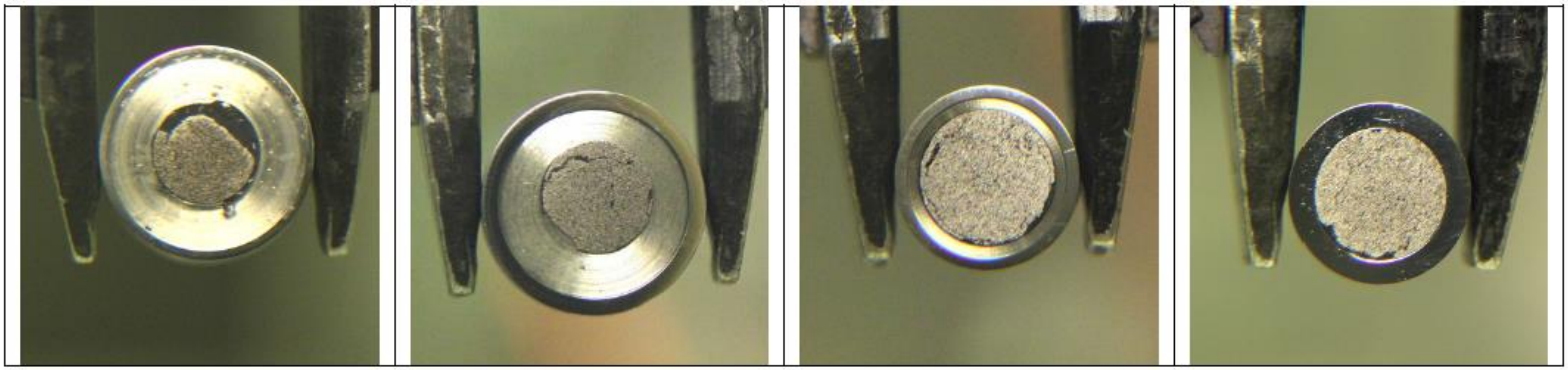
UC_x Post-Irradiation Studies





Electron Probe Micro Analysis at PSI

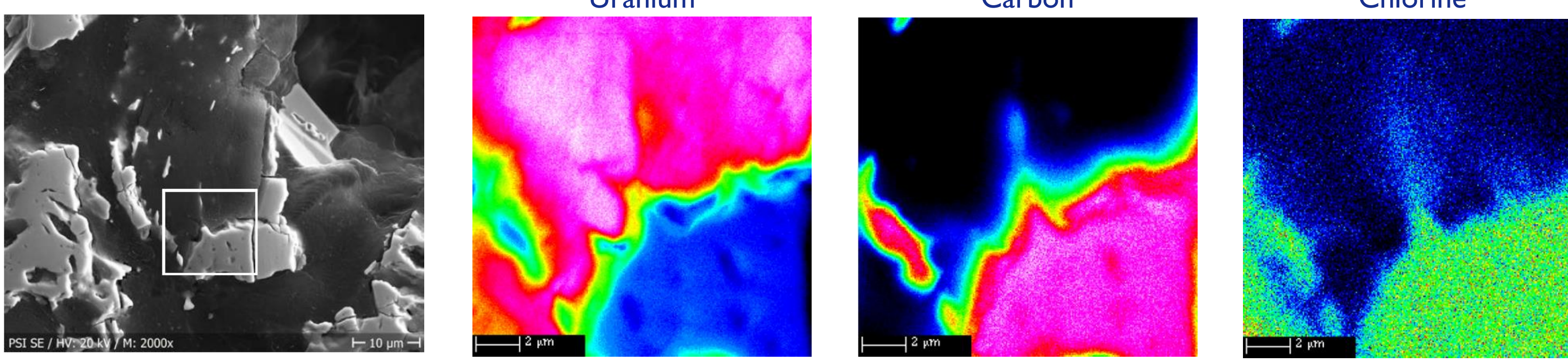
Preparation (polishing) of samples in nitrogen atmosphere



Non-irradiated reference | Proton beam entrance | From container center | Proton beam exit

- Extensive EPMA data set still under analysis
- Fission product concentration below detection limit
- Confirmation of zones with varying carbon concentration causing UC_2-UC phase competition

Uranium | Carbon | Chlorine



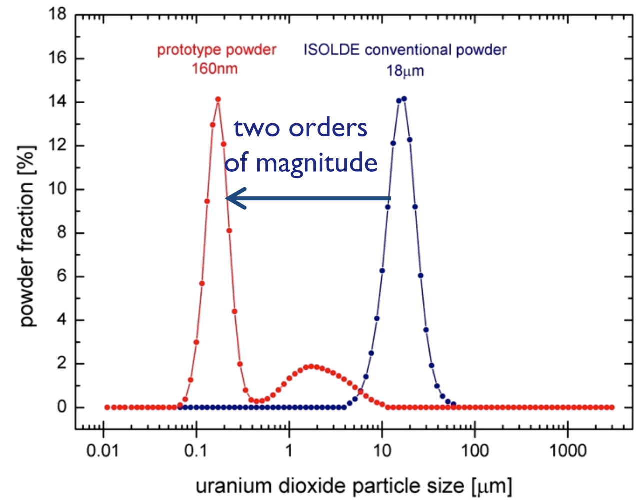
Material Synthesis
Task 1

Composing a Recipe for Nano-Grained UO_2

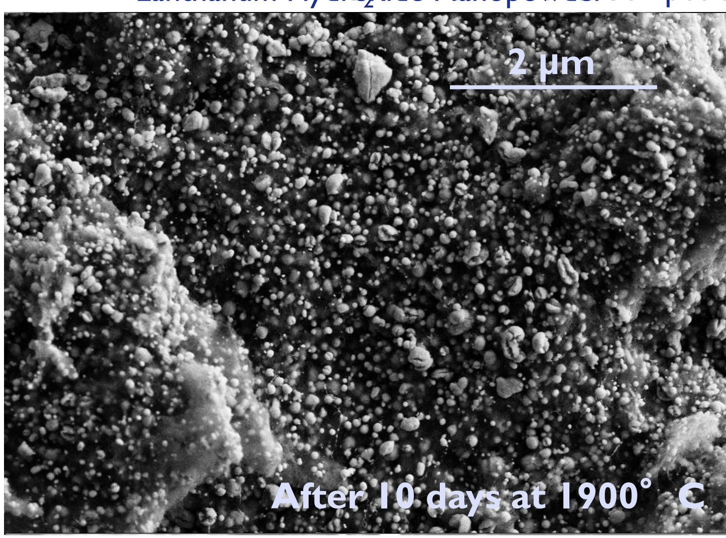
Synthesis of de-novo designed uranium carbide matrixes:

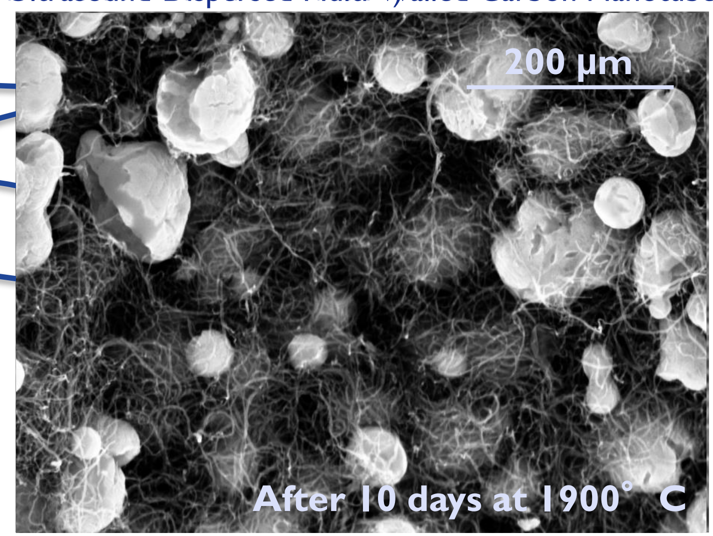
Different microstructures, densities, grain sizes, crystal structures tested → tailor-made matrix:

- Suspension grinding of UO_2 powder to 160 nm average particle size
- Wet-mixing with multi-walled carbon nanotubes
- Ultrasound drying of mixture and pressing to 1.6 g/cm^3 pellets
- Fast reactive sintering to mixed uranium carbide in carbon nanotube matrix



Lanthanum Hydroxide Nanopowder / UC_2-MWCNT Nano Composite (After Carbo-Thermal Reduction) / Ultrasound-Dispersed Multi-Walled Carbon Nanotubes





Microstructure investigations of UC_2-MWCNT nano composite currently in preparation

Some few Material Characteristics

	ISOLDE conventional				2014 Nano				UC HD
	graphite	UO_2	UO_2 + 6C pellet	UC_2 + 2C pellet	graphite CNT	UO_2	UO_2 + 5C pellet	UC + 2C pellet	UC pellet
particle size [µm]	28	16			0.01 x 10	0.21			
SSA [m^2/g]	3.6	4.9	5.2	0.1	287	16	30	221	2.20
diameter [mm]			14	12.3			15	14.1	13.2
thickness [mm]			1.6	1.45			1.04		1.00
density [g/cm^3]			4.50	3.67			1.89	*1.95**	13.20
reduced U fraction				0.999				*≈0.95**	1.000
UC_2 phase fraction				0.295				0.149*	0.941
C phase fraction			0.309	0.147			0.190	0.082*	0.059
UO_2 phase fraction			0.222	0.000			0.136	0.009*	0.000
porosity			0.469	0.558			0.674	0.760*	0.000
total target mass [g]			110	94			51.49	39.63	361.8
				100.00%				42%	414.07%

* Property needs to be confirmed

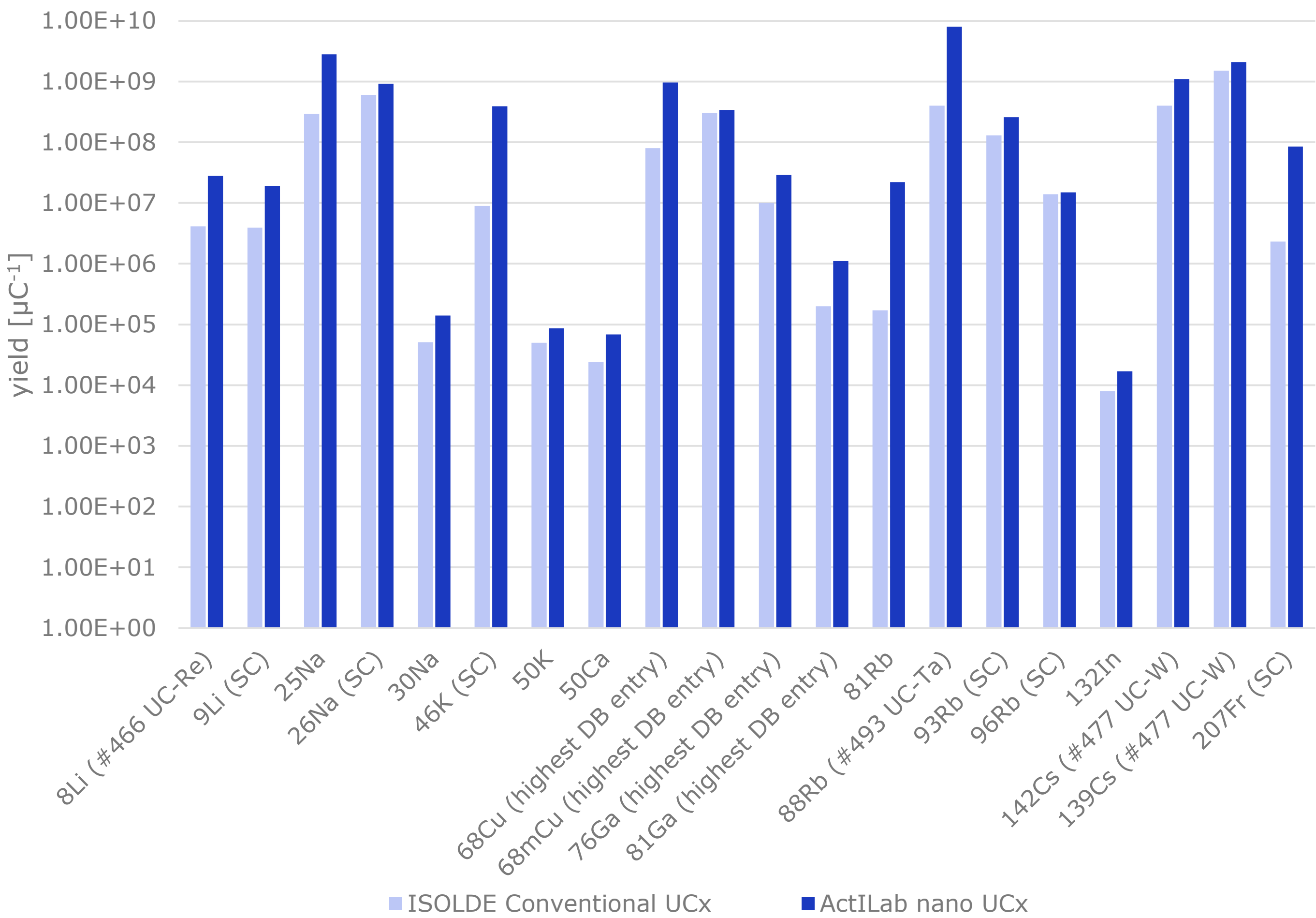
UO_2 + 6(5)C pellets

UC_2 + 2C pellets

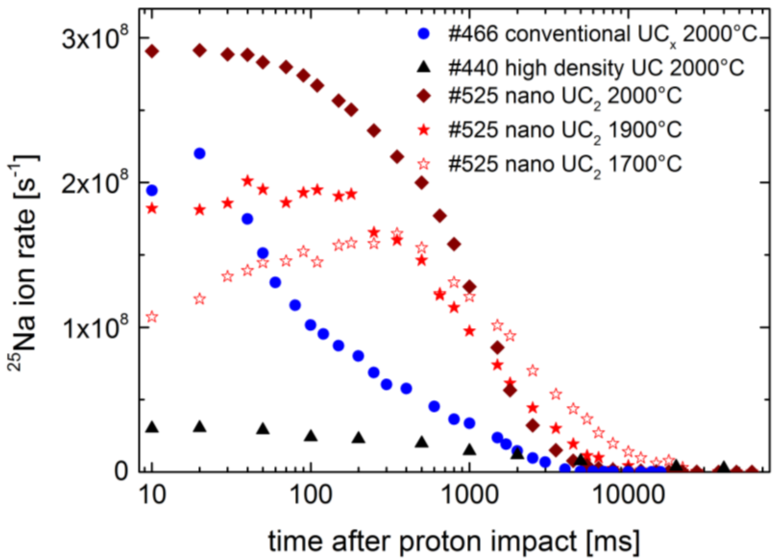
Online Tests at ISOLDE
Task 4

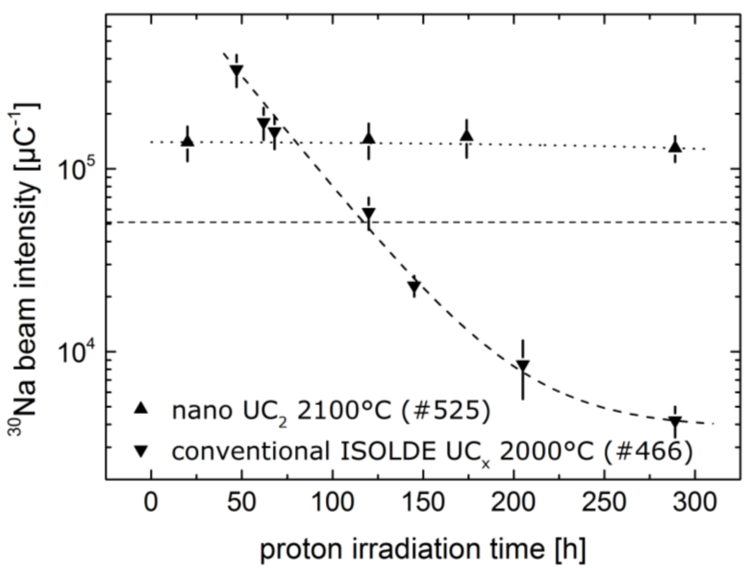
unit #498 on HRS in 2012 – difficult, but ^{11}Be and ^{30}Na yield evolution studied
unit #525 on GPS in 2014 – tests successful

ENSAR-ActILab: Nano Uranium Carbide at ISOLDE



Nano Uranium Carbide Online at ISOLDE





isotope	half life	yield [μC^{-1}]
^{8}Li[1]	840 ms	$2.8\cdot10^{7}$
^{9}Li[1]	178 ms	$1.9\cdot10^{7}$
^{25}Na[1,5]	59.6 s	$2.8\cdot10^{9}$
^{26}Na[1,5]	1.07 s	$9.2\cdot10^{8}$
^{30}Na[2]	48 ms	$1.4\cdot10^{5}$
^{46}K[1,4]	115 s	$3.9\cdot10^{8}$
^{50}K[1,2,4]	472 ms	$8.6\cdot10^{4}$
^{50}Ca[2]	13.9 s	$6.8\cdot10^{4}$
^{41}Sc[1,5]	596 ms	$1.1\cdot10^{5}$
^{68}Cu[2,7]	30 s	$9.6\cdot10^{8}$
^{68m}Cu[1,7]	3.8 min	$3.4\cdot10^{8}$
^{76}Ga[2]	32.6 s	$2.9\cdot10^{7}$
^{81}Ga[2]	1.22 s	$1.1\cdot10^{6}$
^{81m}Rb[2]	30.3 min	$2.2\cdot10^{7}$
^{88}Rb[1,4]	17.8 min	$8.0\cdot10^{9}$
^{93}Rb[1,4]	5.84 s	$2.6\cdot10^{8}$
^{96}Rb[1,5]	199 ms	$1.5\cdot10^{7}$
^{114}Ag[2,8]	4.5 s	$1.4\cdot10^{2}$
^{116m}Ag[2,8]	8.2 s	$9.0\cdot10^{2}$
^{117}Ag[2,8]	72.8 s	$1.6\cdot10^{3}$
^{118}Ag[2,8]	3.7 s	$6.2\cdot10^{4}$
^{120}Ag[2,8]	1.17 s	$2.3\cdot10^{4}$
^{122}Ag[2,8]	0.52 s	$\leq1.3\cdot10^{3}$
^{132}In[1,2]	0.20 s	$1.7\cdot10^{4}$
^{139}Cs[1,4]	9.3 min	$2.6\cdot10^{9}$
^{142}Cs[1]	1.68 s	$1.1\cdot10^{9}$
^{148}Cs[1,4]	158 ms	$1.7\cdot10^{4}$
^{207}Fr[3]	0.148 s	$8.5\cdot10^{7}$
^{230}Fr[1,4]	0.596 s	$7.1\cdot10^{5}$
A = 225[6]		$1.1\cdot10^{9}$

[1] assessed through β detection with release curve integration

[2] assessed through γ detection

[3] assessed with scintillator, assuming 50% α detection efficiency

[4] beam composition assessed through γ spectroscopy

[5] beam composition assessed through half-life measurement

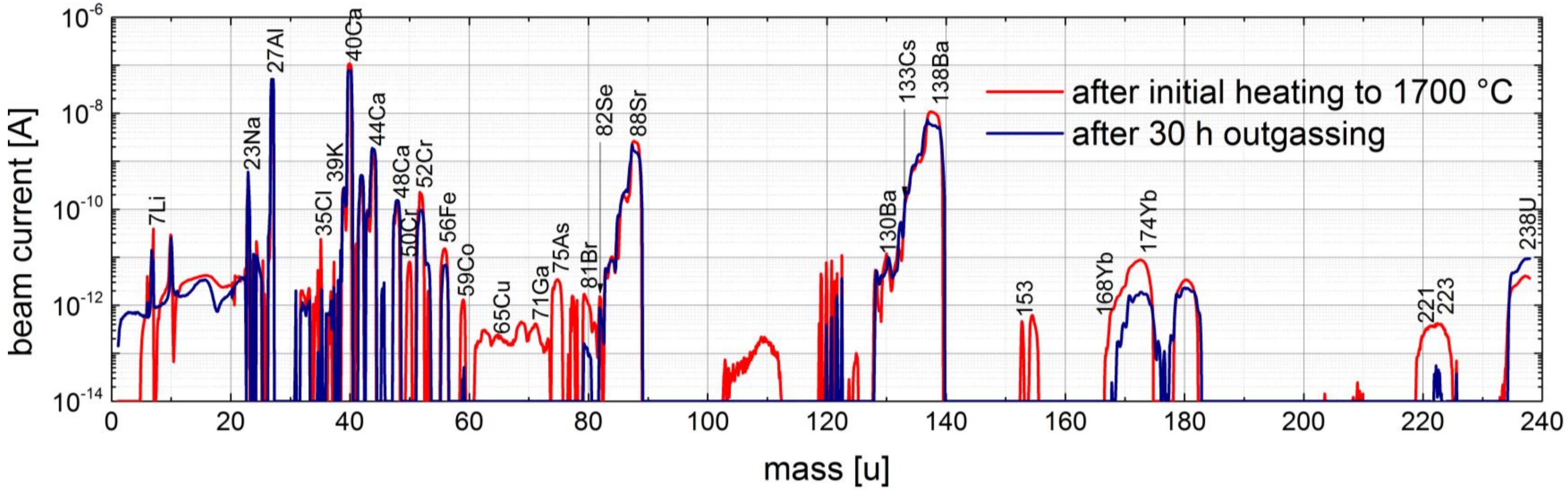
[6] faraday cup measurement

[7] resonant laser ionized

[8] extracted in its 2+ state

[9] assuming 100% branching into investigated γ transition

Nano Uranium Carbide Online at ISOLDE



New material:

- Seems increases isotope yield of most investigated elements (Li, Na, K, Ca, Cu, Ga, Rb, In, Ra, Fr)
- reduced ageing effects (reduction of yield over time)
- reduces actinide waste by 60%
- chemically unstable
- Resource-intense to synthesize (additional development needed)
- Rich in synthesis-induced contaminations

What has ActILab brought us? (my perspective)

- A healthy collaboration between several European institutes leading to results that where out of reach before
- Paving the way for easier access to infrastructure between the labs
- Blowing breaches into Europe's shipment channels for actinide materials
- Extension of collaborative effort into other materials (Julien Guillot)
- Deep insights to conventional UC_X properties, dynamics, ageing mechanisms
- At least one new and promising target material
- LOTS OF FUN