Unité mixte de recherche
CNRS-IN2P3
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http://ipnwww.in2p3.fr/

ACTILAB

Development of UCX targets in the ACTILAB project

- The context of the project
- Highlights on the main achievements
- Final step and outlook

C. Lau *et al.*, ECOS-EURISOL joint Town Meeting, October 2014

Context of the Project

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RNB intensities mainly depends on the target performances

UC_X target high performances are crucial for next generation facilities:

ISOLDE SPES exotic beams for science Spiral2 EURISOL

→ Better understanding material properties ⇔ release kinetics

+ Studying new synthesis techniques

Task #1: Synthesis of new actinide targets

Task #2: Characterization of new actinide targets

Task #3: Actinide targets properties after irradiation

Task #4: Online Tests of Actinide Targets

Project submitted with a budget request of 460 K€,

but finally initiated with 336 K€.

CERN	GANIL	INFN	IPNO	PSI
113 K€	38 K€	72 K€	55 K€	58 K€

Investigation of Standard UC_x

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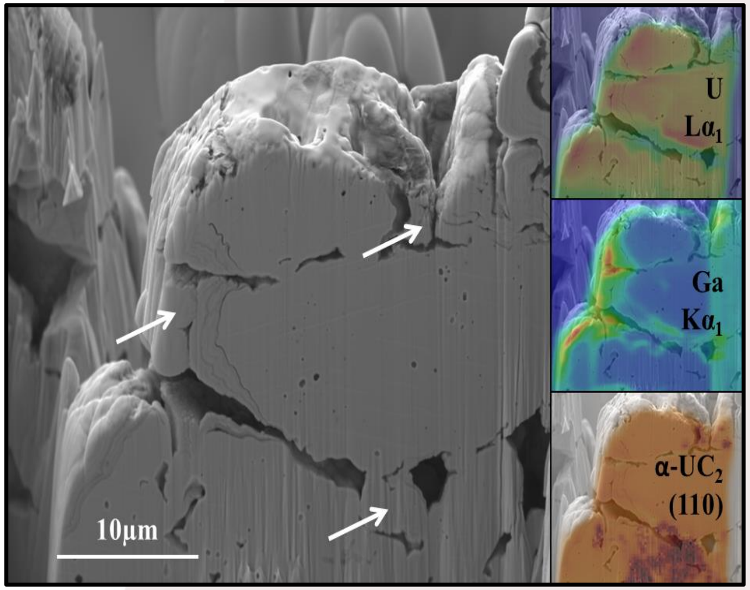
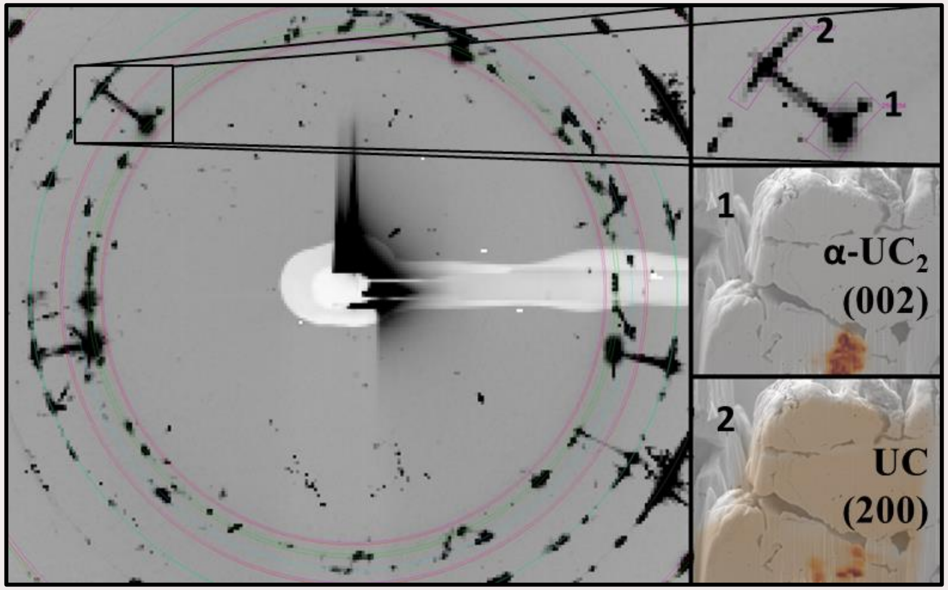
Characterization of conventional UC_x using synchrotron-based micro-beam analysis:

Microscopic morphology – buried porosity & chemistry



Kinetic stabilization by sub-microscopic UC – UC_2 phase competition

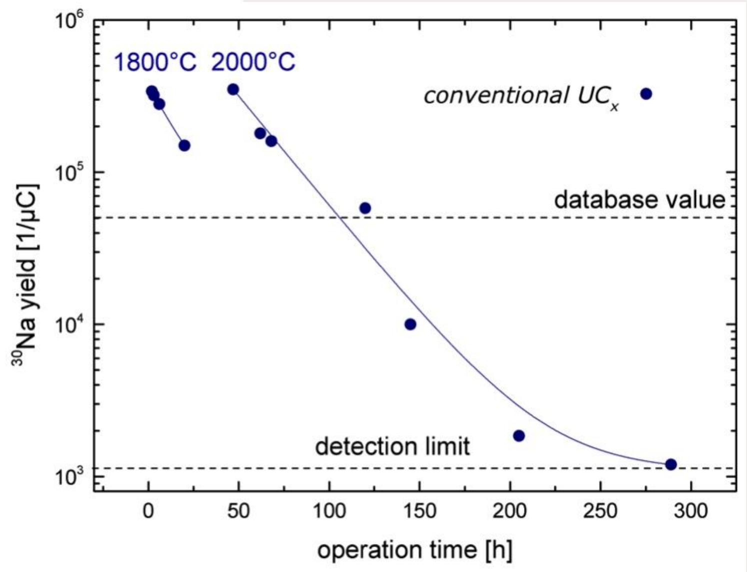


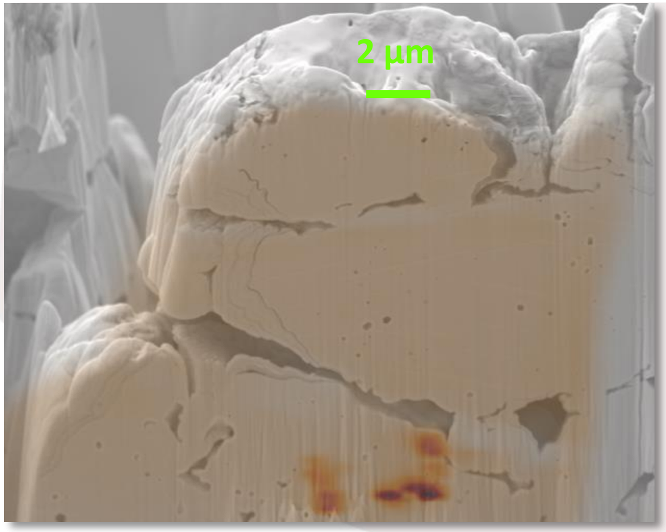
- Grain size of material is smaller than previously estimated;
- Global phase transition from α-UC_2+UC+5C towards β-UC_2+2C observed at 2100°C

- Phase competition between UC and α-UC_2 as yet missing explanation of performance and durability of this material's microstructure over long time irradiation at very high temperatures

A. Gottberg, et al., submitted

Confirmation of Hindered Grain Growth





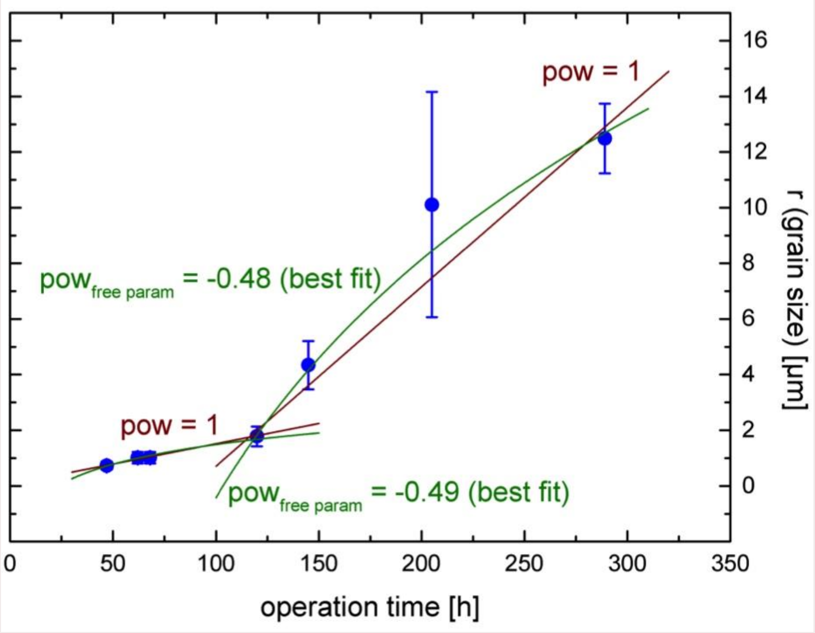
For pure diffusion and short half life:

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

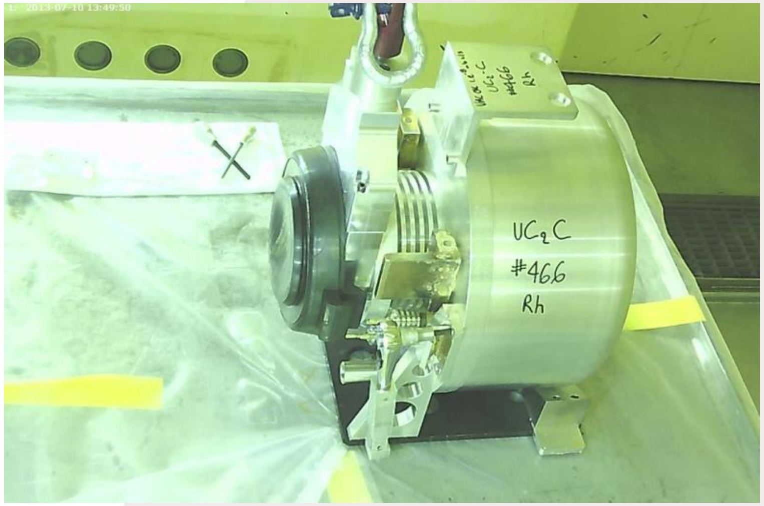
$$\mu_0 = \pi^2 \frac{D}{r^2}$$

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

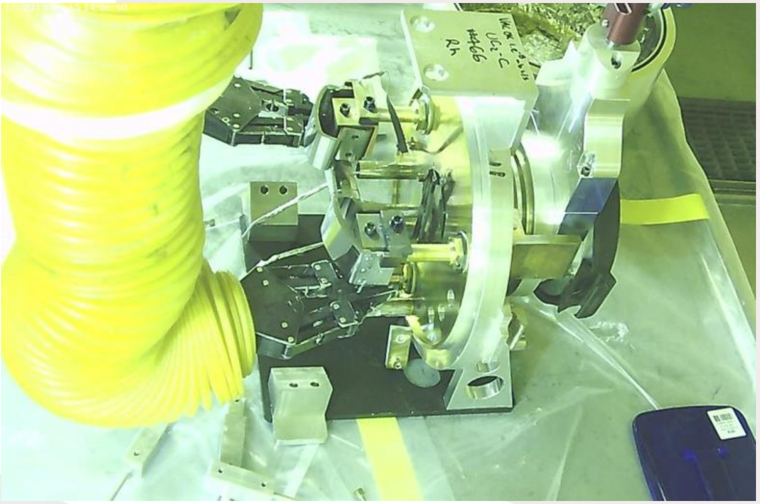


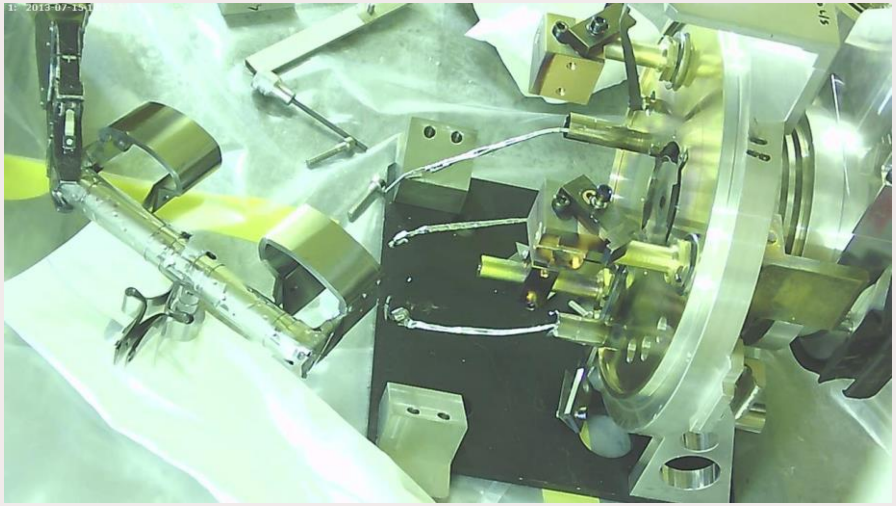
Dismantling Target Unit at PSI



UC_X target unit used for ^{30}Na yields:

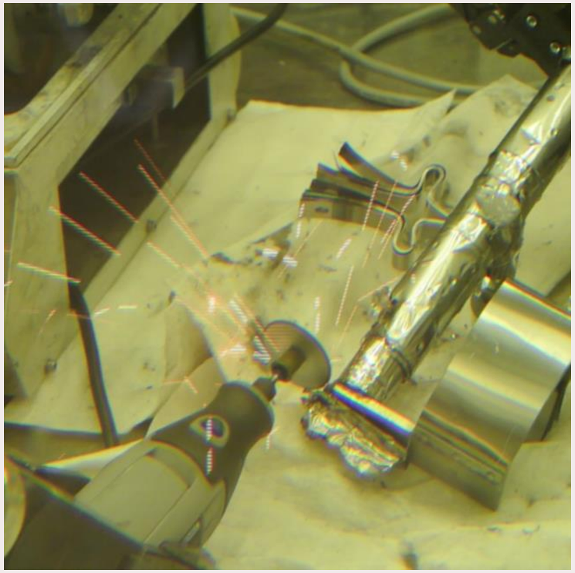
* Opening target vessel in a hot cell chain in air (6 mSv/h on contact with Al beam window)
* Extraction of tantalum container (19 mSv/h on contact with Ta proton beam window)
* Sealing of ion source and mass marker outlet with epoxy glue to prevent oxidation of carbide material

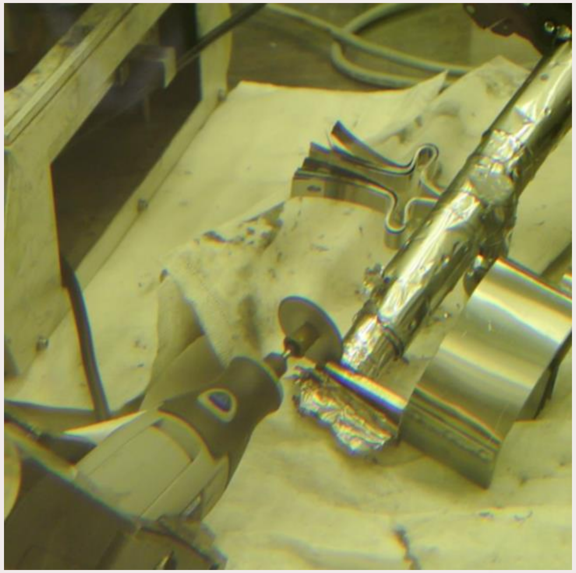


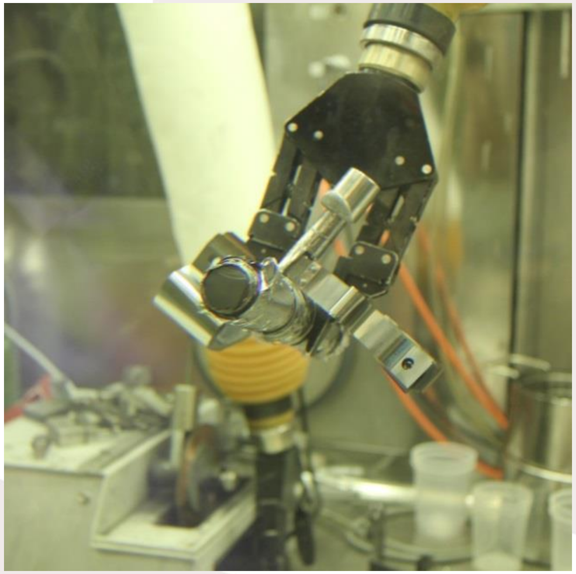


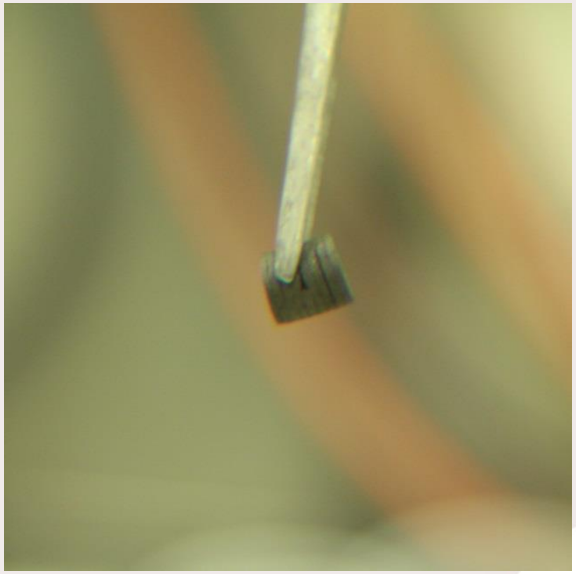
Dismantling Target Unit at PSI (2/2)

- 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)
- Pellets appear macroscopically unchanged









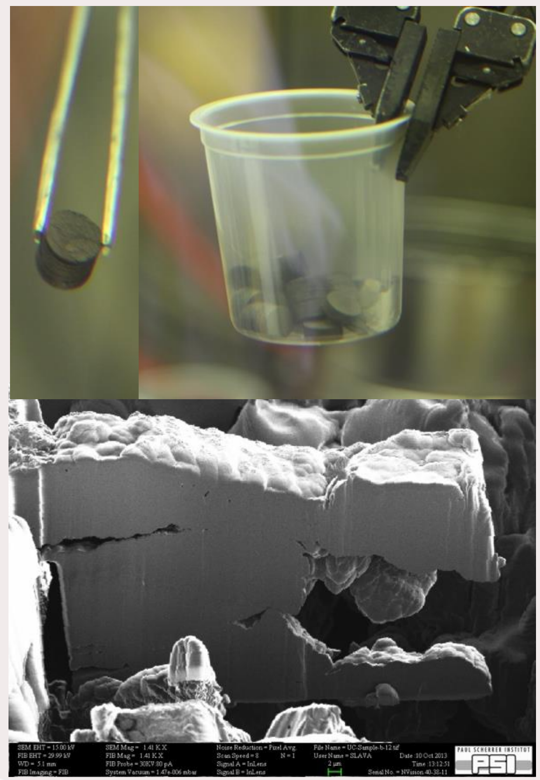
UC_x Post-Irradiation Study at PSI

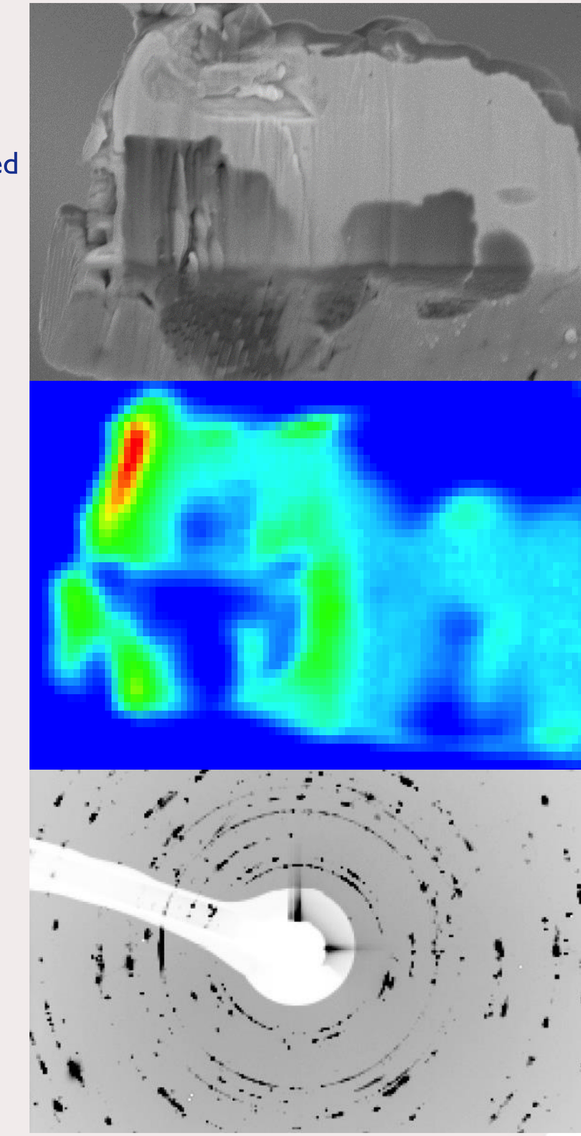
Post-irradiation analysis:

- Pellets appear macroscopically unchanged
- Microscopic evolution of pore distribution and grain size under irradiation observed
- Further results of synchrotron investigations under analysis

before irradiation

after irradiation





Electron Probe Micro Analysis at PSI

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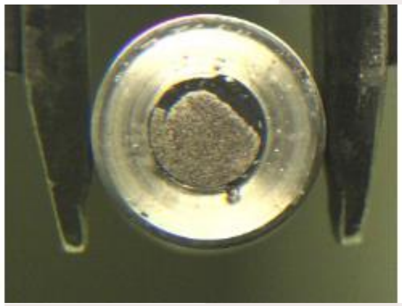
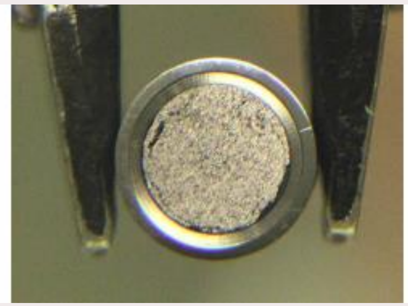
Preparation (polishing) of samples in nitrogen atmosphere



Non-irradiated reference

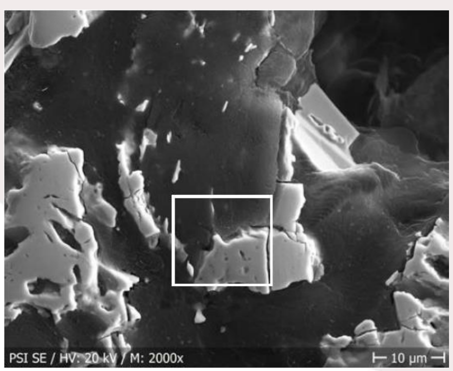
Proton beam entrance



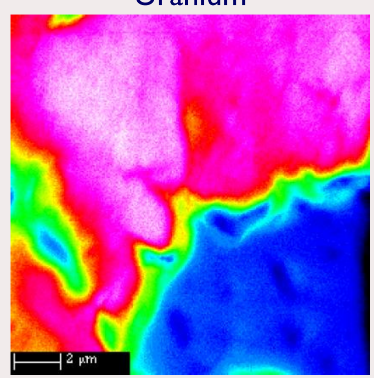
From container center

Proton beam exit

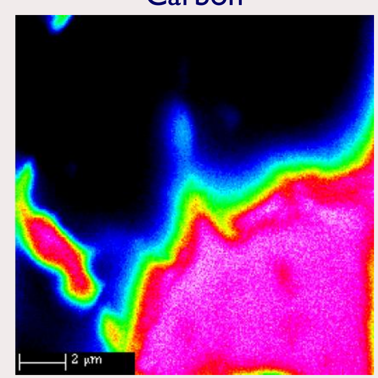
- Extensive EPMA data set still under analysis
- Confirmation of zones with varying carbon concentration causing UC_2-UC phase competition



Uranium



Carbon

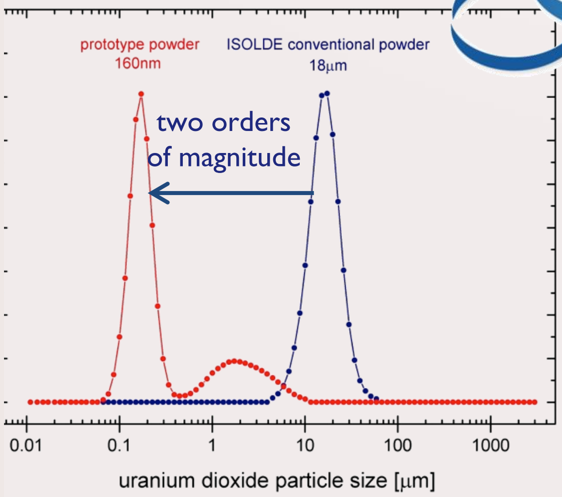


Composing a Recipe for Nano-Grained UO_2

Synthesis of de-novo designed uranium carbide matrixes:

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

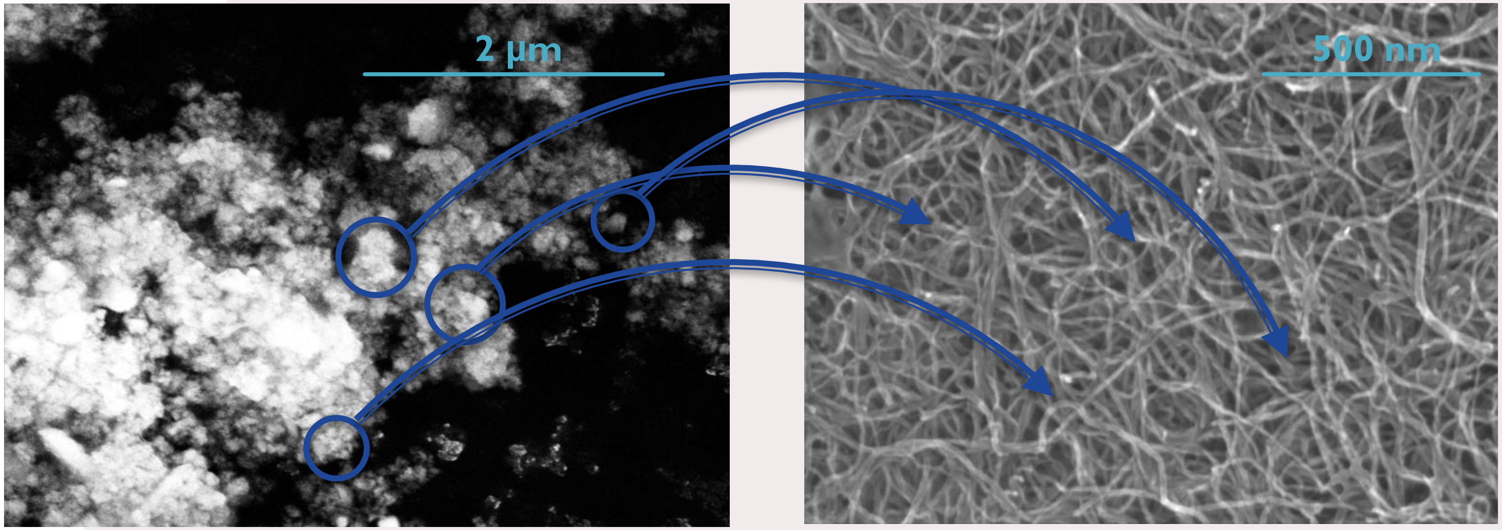
* Suspension grinding of UO_2 powder to 160 nm average particle size
* Wet-mixing with multi-walled carbon nanotubes
* Ultrasound drying of mixture and pressing to 1.6 g/cm^3 pellets
* Fast reactive sintering to mixed uranium carbide in carbon nanotube matrix



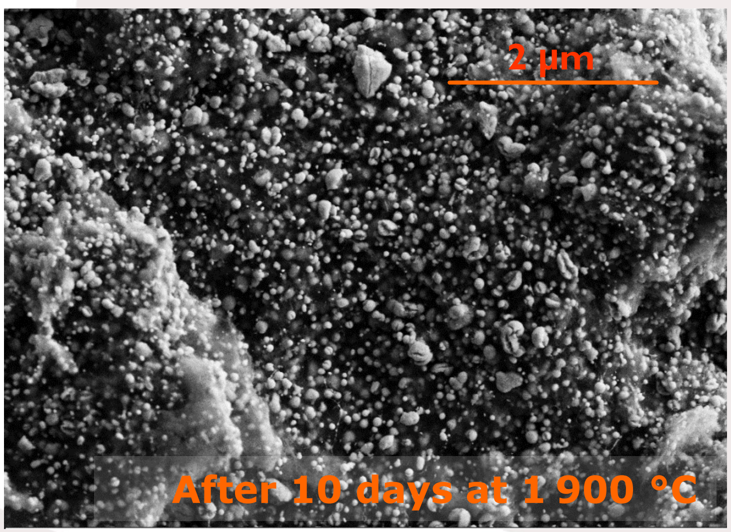
Microstructure of UC_2-MWCNT nano composite currently under investigation…

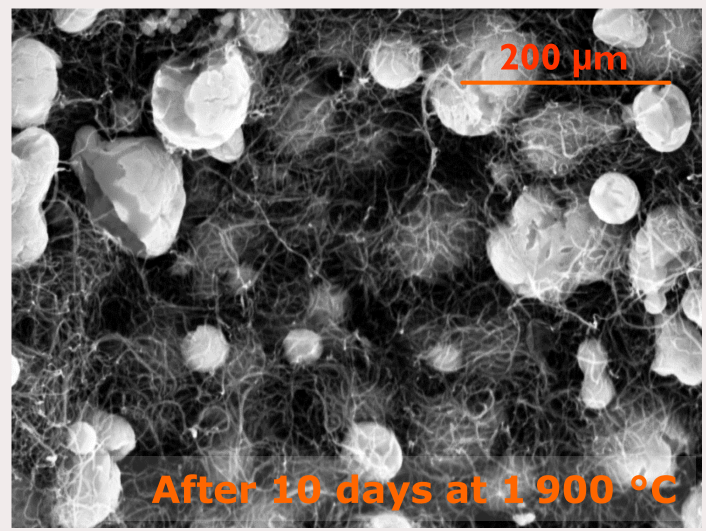
Investigation also initiated on **La** , a kind of chemical analogue of **U** (Cf. next slide)

Lanthanum Hydroxide Nanopowder — Ultrasound-Dispersed Multi-Walled Carbon Nanotubes



LaC$_2$-MWCNT Nano Composite (After Carbo-Thermal Reduction)





1st On-Line Results

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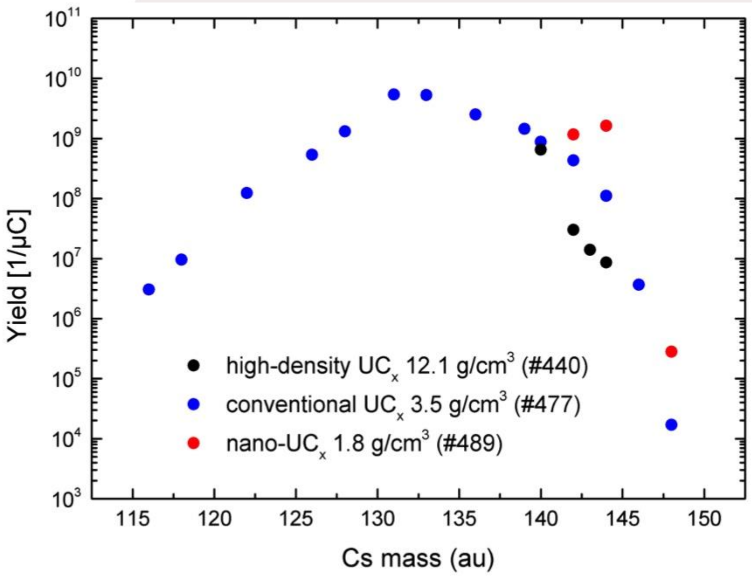
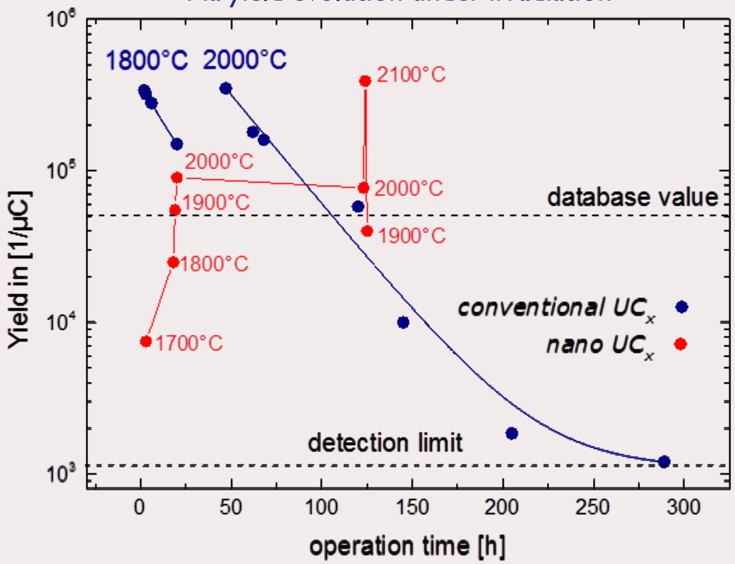
- ^{11}Be: 6. 10^7 ion/µC (gain of one order of magnitude)
- Record yields for Cs isotopes
- Structure seems preserved over time and temperature (at least >100 h)…



^{30}Na yield evolution under irradiation



in progress…

The release experiment at ALTO

Accélérateur Linéaire et Tandem à Orsay

IPN Institut de Physique Nucléaire Orsay

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1st tests: B. Hy et *al.*, Nucl. Instr. Meth. B 288 (2012) 34.

: fission products (FPs)

Deuterons
27 MeV
20 nA

Graphite converter

P1 P2

Neutrons

20 min irradiation
30 min « cooling »

Detector 1

Initiale activity
P1 and P2
γ meas.

Detector 2

Furnace
1700°C/30 min
10^{-5} mbar
Cooling 10 min→70°C

Room T

Detector 1

Final activity
P1 and P2
γ meas.

Detector 2

Previous exp.

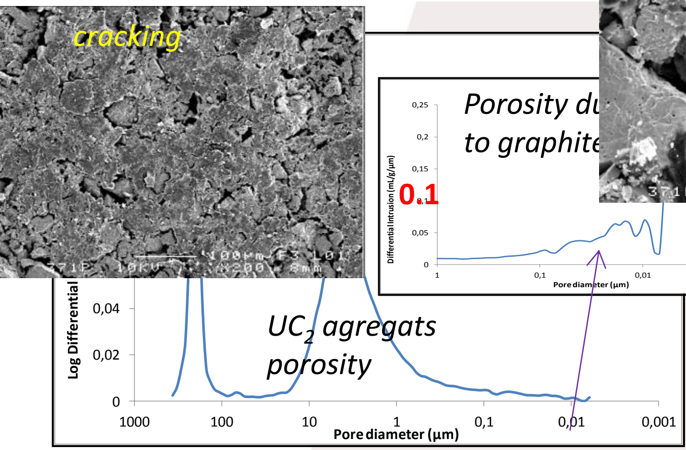
With new furnace

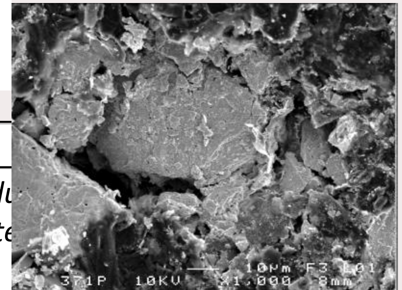
Correlation between microstructure and γ-spectroscopy measurements after irradiation and heating

Microstructure and release

PARRNe :

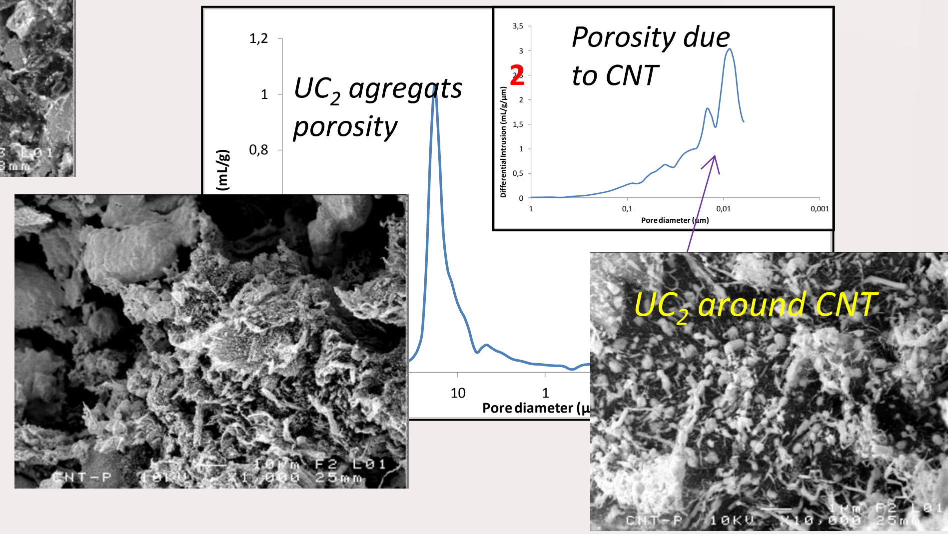
ρ_{eff}= 8.1 g.cm^{-3} and P_{open}= 32 %



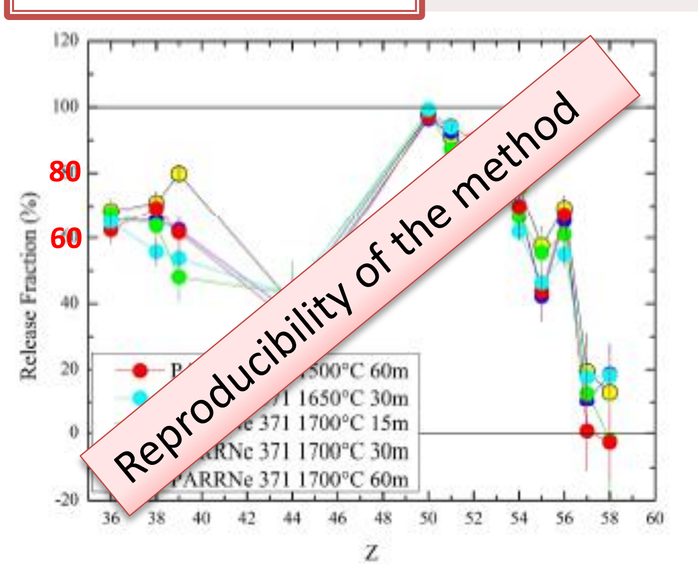


CNT (C nanotubes):

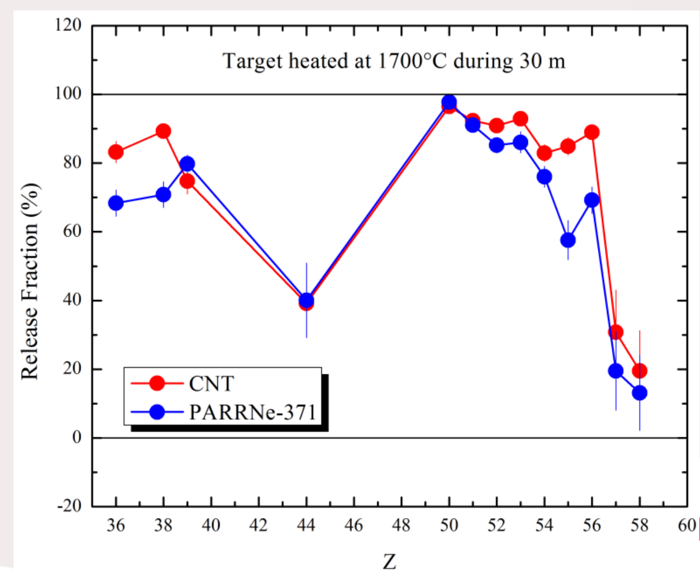
ρ_{eff}= 8.5 g.cm^{-3} and P_{open}= 59 %



Release measurements:



Higher open porosity with small pores
⇨
Higher release



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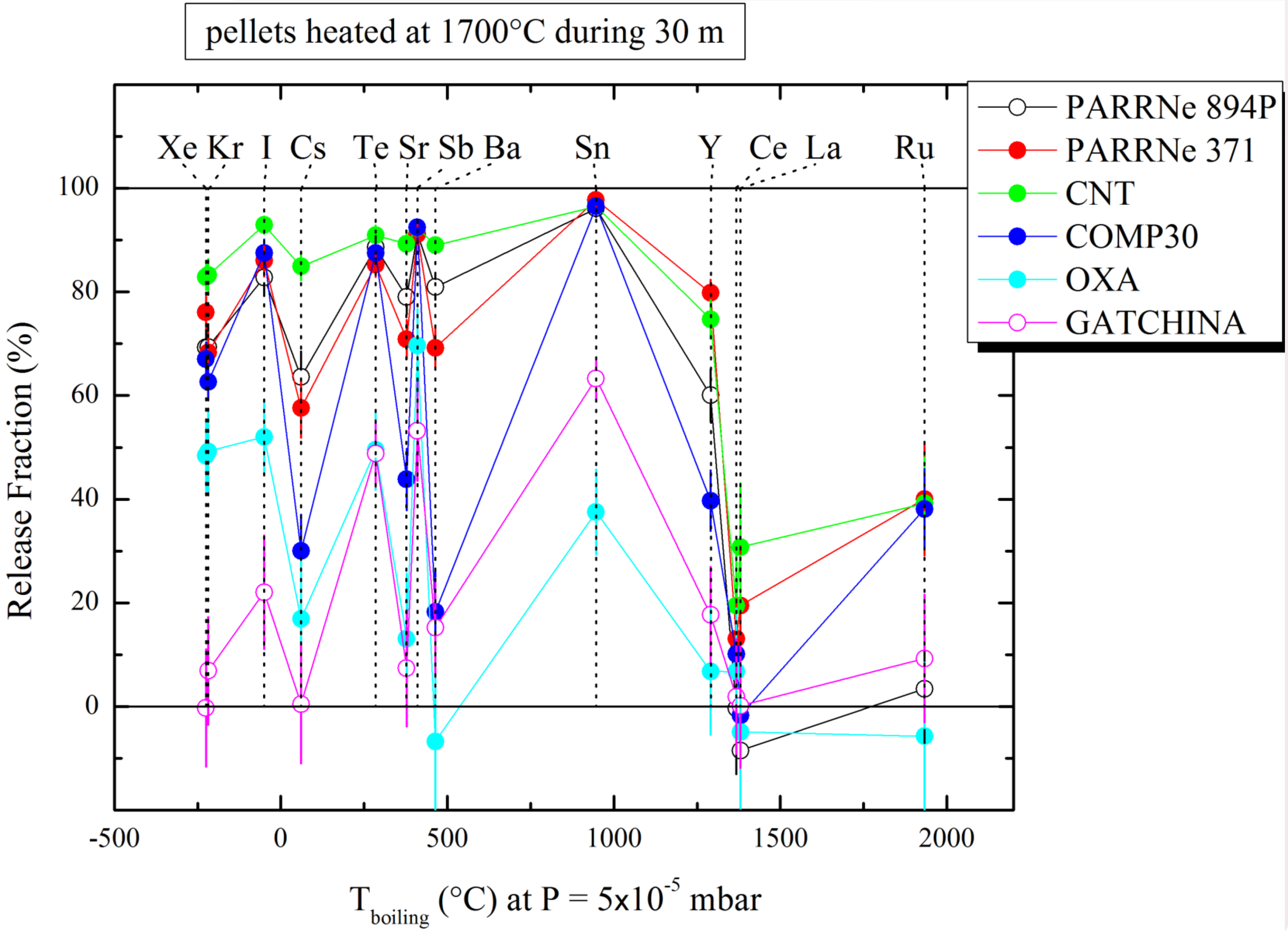
pellets heated at 1700°C during 30 m
Xe Kr I Cs Te Sr Sb Ba Sn Y Ce La Ru
PARRNe 894P
PARRNe 371
CNT
COMP30
OXA
GATCHINA
Release Fraction (%)
100
80
60
40
20
0
-500
0
500
1000
1500
2000
T boiling (°C) at P = 5x10^-5 mbar

Thermal Conductivity & Emissivity

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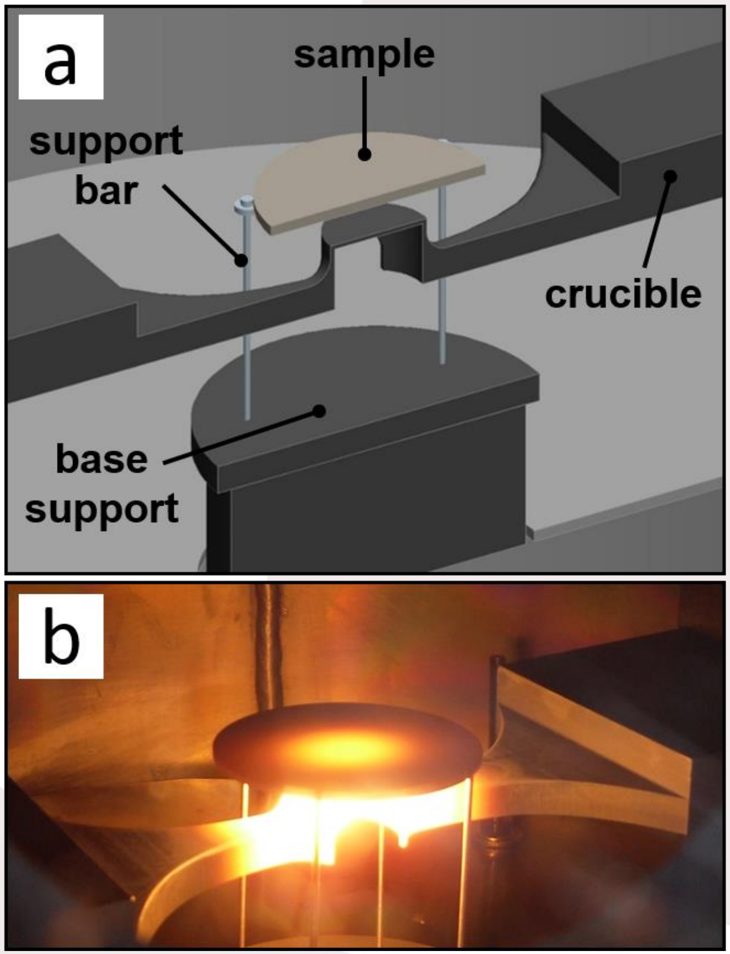
SPES exotic beams for science

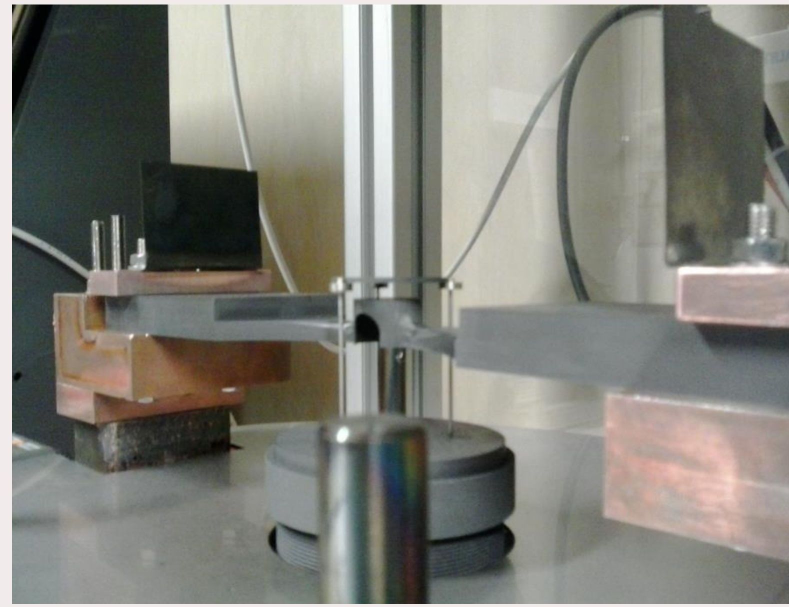
Setup for thermal conductivity measurements

Original setup – INFN/LNL



M. Manzolaro et *al.*, Rev. Sci. Instr. 84 (2013) 054902.

Newly developed setup (Padova)



allowing measurements for samples of smaller sizes (down to ∅ ~30 mm)

Thermal Conductivity of UC_x

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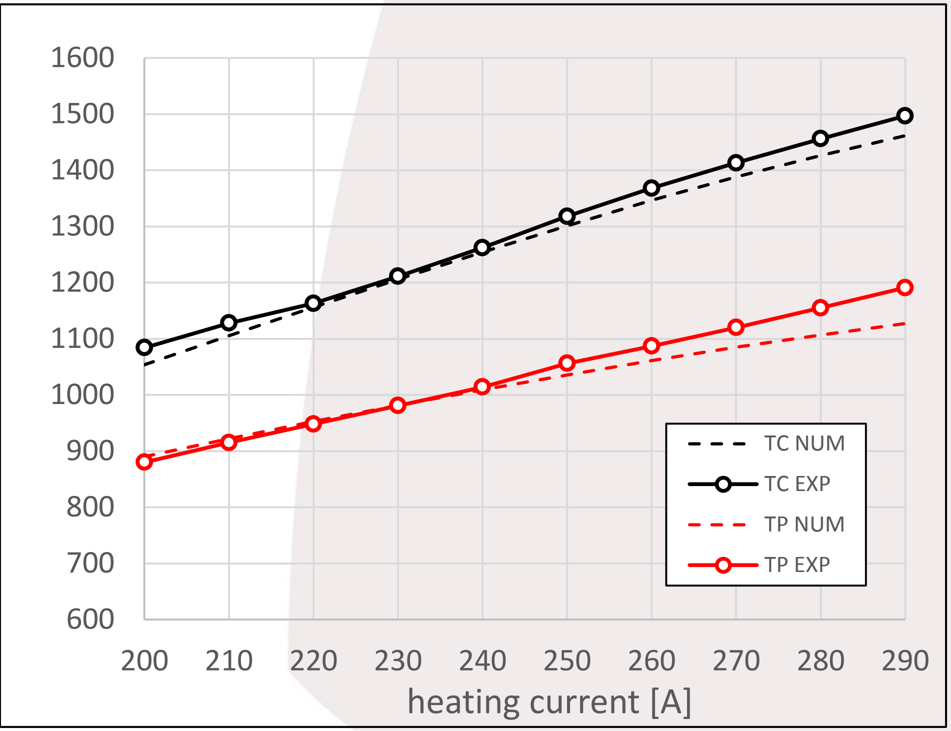
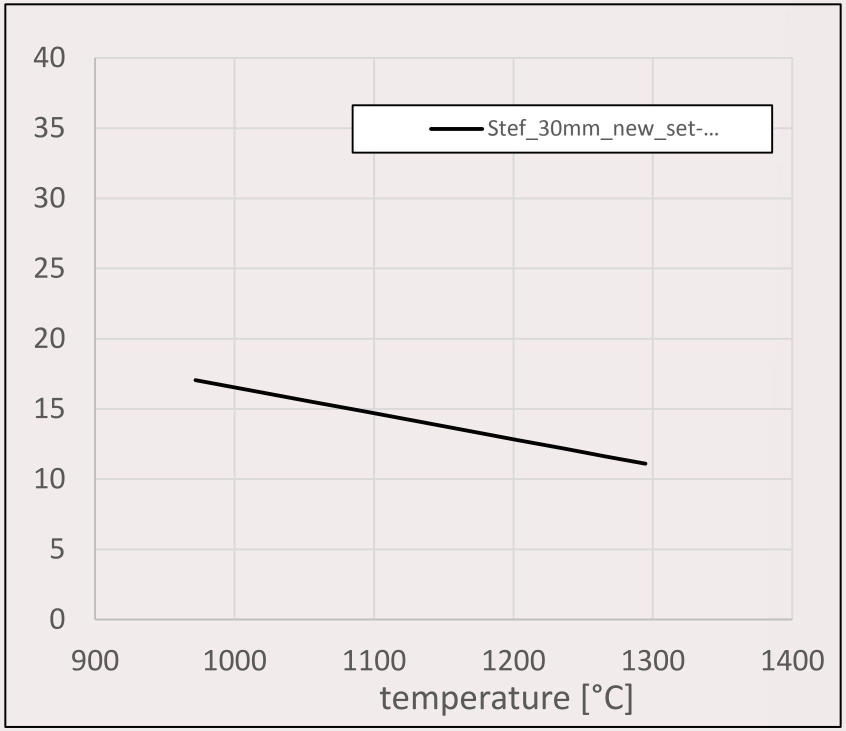
SPES exotic beams for science

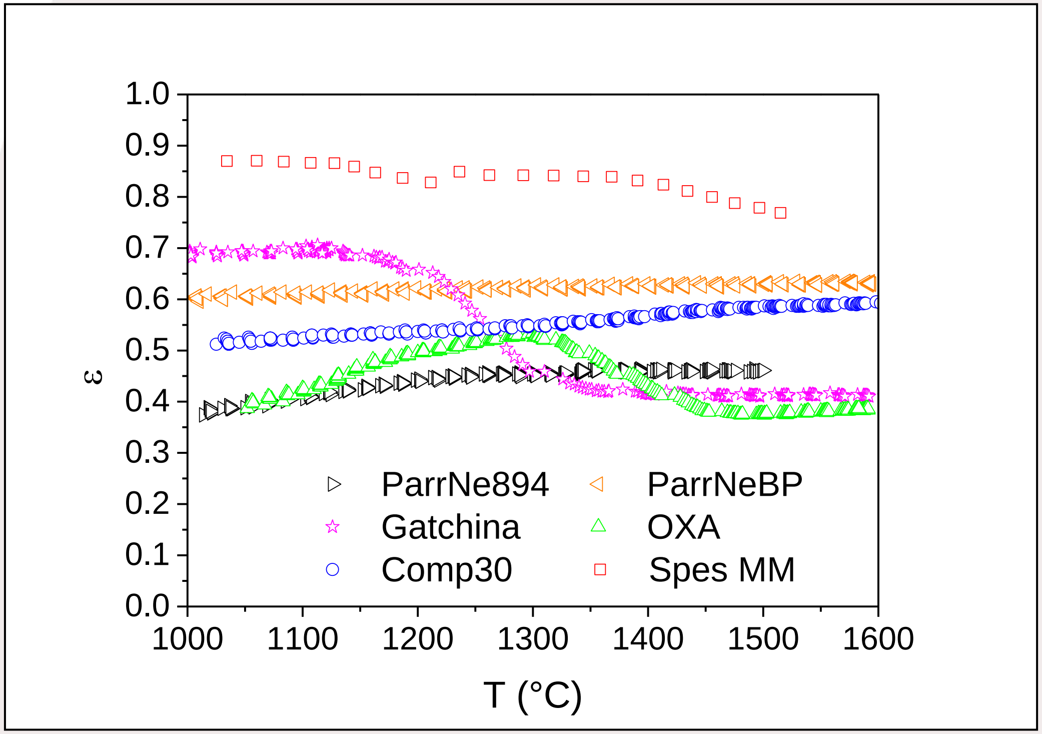
Temperature [°C] measurements vs. numerical data



Inverse analysis gives Thermal conductivity [W/m°C]



Thermal Emissivity of UC_x



pellet	mass (g)	main phase	mass of U (g)	diameter (mm)	thickness (mm)
GATCHINA	1.80	UC	1.71	13.2	1.0
PARRNe894	0.82	UC_2	0.74	13.0	1.9
OXA	0.61	UC	0.70	7.4	1.9
PARRNeBP	0.87	UC_2	0.79	12.6	1.5
COMP30	0.68	UC_2	0.62	8.3	2.5
SPES MM	3.51	UC_2	2.92	28.9	1.4

Final step and Outlook

The goals of ACTILAB project are close to be completed within ENSAR extension:

* Last measurements requested for the project are about to be completed (post-irradiation analysis, emissivity measurements etc.).
* A final on-line experiment at ALTO (IPNO) or ISOLDE (CERN).
* Going toward nano structured porous UC_X …
* Articles have been submitted or will be submitted soon.
* Samples of UCX have been shipped between CERN, INFN, IPNO and PSI.

in further details:

* PIE of nano-UCX + Determination of phase dynamics.
* Developing the new synthesis procedures, characterization and on-line tests.
* Target recycling (reprocessing).