Development of new neutron-rich radioactive beams

Study of the experimental timeline to build a UC_x target

NESTER Group- ALTO Collaboration

Graduate school PHENIICS
2014-2017 (2ème année)

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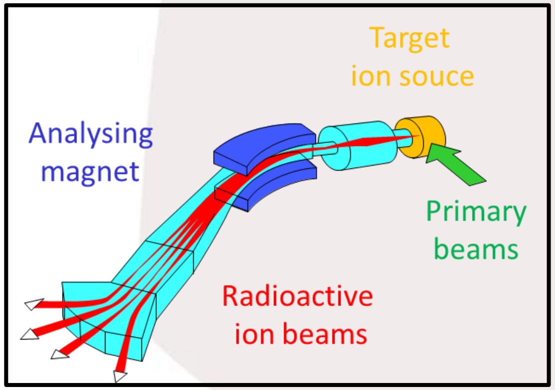
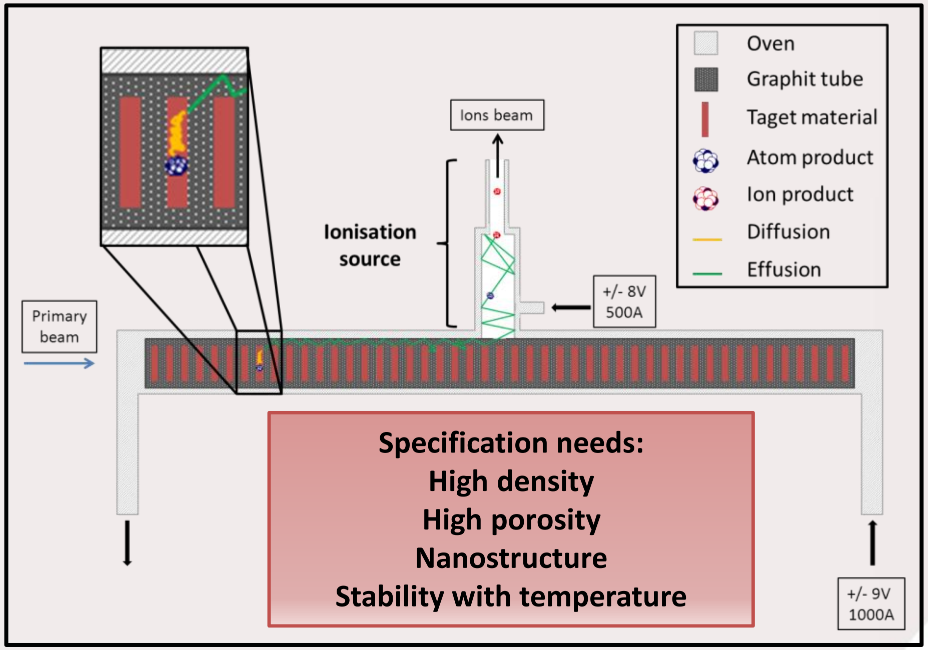
CONTEXT

- Strong demand of exotic beams for the nuclear structure study by β decay
- A method of production of radioactive beams: ISOL technique (Isotope Separation On-Line)



$$I = I_P . \sigma . N . \varepsilon_r . \varepsilon_{ion} . \varepsilon_{tr}$$

I_P : intensity of the incident particle beam from the accelerator
σ : cross section of interest isotopes
ε_r : release efficiency of element of the target to the ion source
ε_{ion} : ionization efficiency of this element
ε_{tr} : transport efficiency of the separator

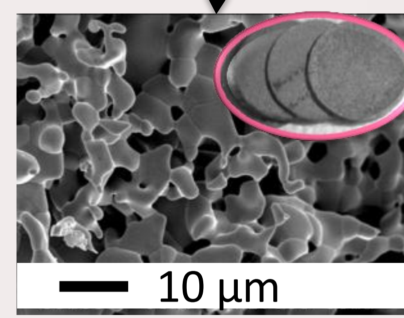


Specification needs:
High density
High porosity
Nanostructure
Stability with temperature

TIMELINE TO BUILD A UCX TARGET

Synthesis of UC_X target

Mixing between Uranium powder and carbon powder → Pressing ~ 500 MPa → Green Pellet → Carburization: 1800°C at 10^{-3} Pa → carburized pellet



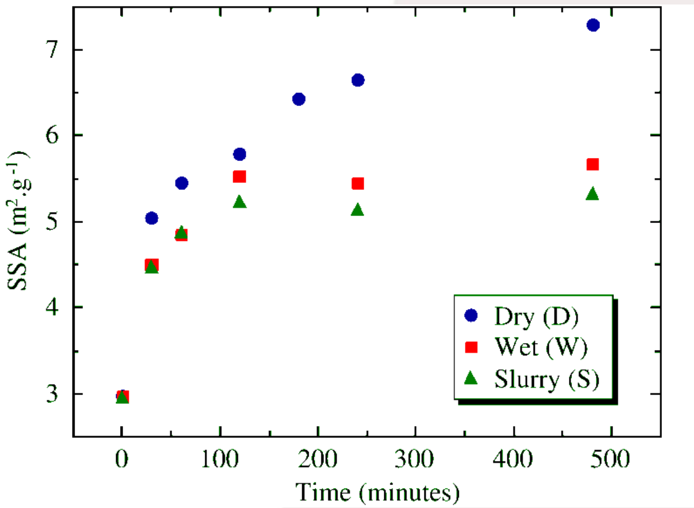
Reaction of carburization: $UO_{2(s)} + 6C_{(s)} \rightarrow UC_{2(s)} + 2C_{(s)} + 2CO_{(g)}$

PROCESS IMPROVMENT

Questions addressed:

- What is the optimal grinding time? (Slide 5)
- What is the pressure to be applied on the pellets and its holding time? (Slide 6)
- What is the optimum thickness of the pellets? (In progress - Slide 7)
- What are the precursors to choose? (Slide 8-11)
    - Carbon sources (carbon black, MWCNT, graphite, graphene)
    - Uranium oxide or oxalate
    - Optimize the structure of the pellet to be stable at high temperature
- How fast should be done sintering? (In progress)

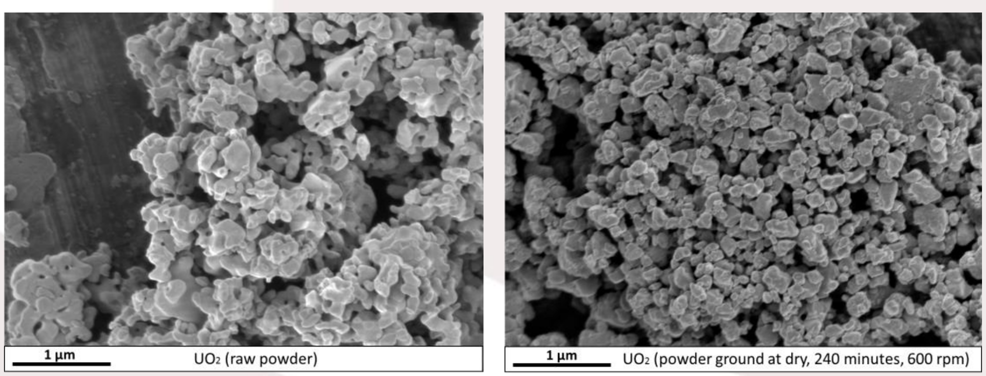
Study of uranium oxide milling: (Guillot *et al*; NIMB, Vol 374, 1 May 2016, P 116–120)



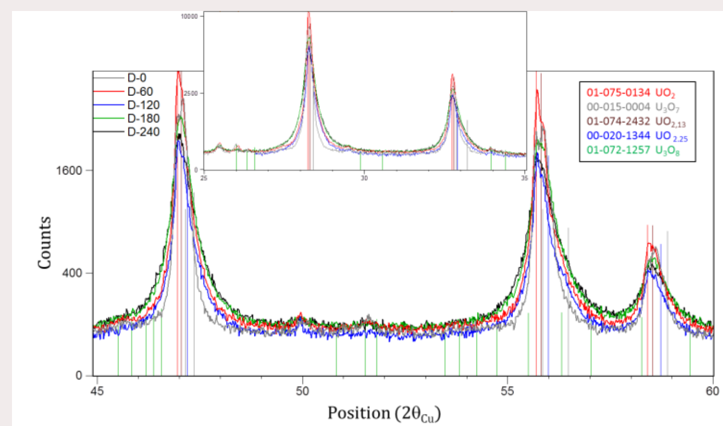
Specific surface area evolution, for the 3 types of samples, as a function of the milling time. (Error bars ± 0,10 $m^2.g^{-1}$)

Model	Phase	Crystallographic data	D-0	D-240	W-240	S-240
UO_{2+x}	UO_2	cell parameter(Å)	5.4576 ± 0.0001	5.4579 ± 0.0002	5.4597 ± 0.0004	5.4608 ± 0.0009
		quantity (wt%)	50.8 ± 0.7	23.6 ± 0.7	16.7 ± 0.8	44.3 ± 0.5
		cristallite size (nm)	170 ± 4	221 ± 25	200◆ ± 0	214 ± 7
		microstrain	0.00074 ± 0.00002	0.00183 ± 0.00005	0.00048 ± 0.00001	0.00068 ± 0.00001
		x	0.14	0.13	0.11	0.10
UO_{2+x}	U_4O_9	cell parameter(Å)	5.4389 ± 0.0002	5.4467 ± 0.0004	5.4463 ± 0.0003	5.4484 ± 0.0003
		quantity (wt%)	39.3 ± 0.8	45.6 ± 1.1	53.5 ± 1.0	33.2 ± 0.5
		cristallite size (nm)	249 ± 8	106 ± 11	200◆ ± 0	200◆ ± 0
		microstrain	0.00100 ± 0.00001	0.00580 ± 0.00008	0.00224 ± 0.00004	0.00422 ± 0.00004
		x	0.34*	0.25	0.26	0.24
b-U_4O_9	U_3O_7	cell parameter(Å)	5.4170 ± 0.0000	5.4383 ± 0.0011	5.4395 ± 0.0005	5.4357 ± 0.0008
		quantity (wt%)	9.8 ± 0.4	30.8 ± 1.0	29.7 ± 0.6	22.5 ± 0.4
		cristallite size (nm)	39 ± 2	11.0 ± 0.5	25.2 ± 0.9	10.9 ± 0.3
		microstrain	0.00008 ± 0.0006	0.0102 ± 0.0004	0.00512 ± 0.00009	0.0034 ± 0.0005
		x	0.57	0.34	0.33	0.37

Table : Rietveld refinements using Maud software and Delft isotropic model.
**: hyperstoichiometric U_3O_7 phase.* ◆ : fixed crystallite size for a convergent refinement.



SEM observation of raw and 240 minutes milled powder

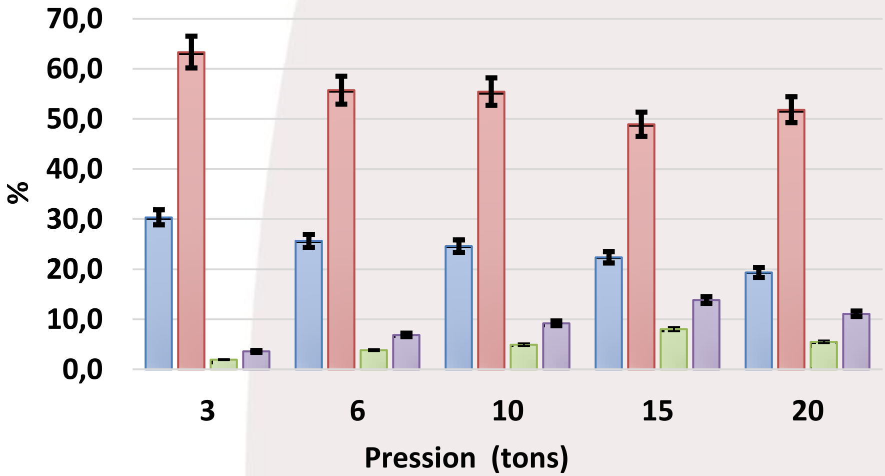


XRD phase identification for raw and powders ground at different times.
Vertical lines correspond to ICDD patterns.

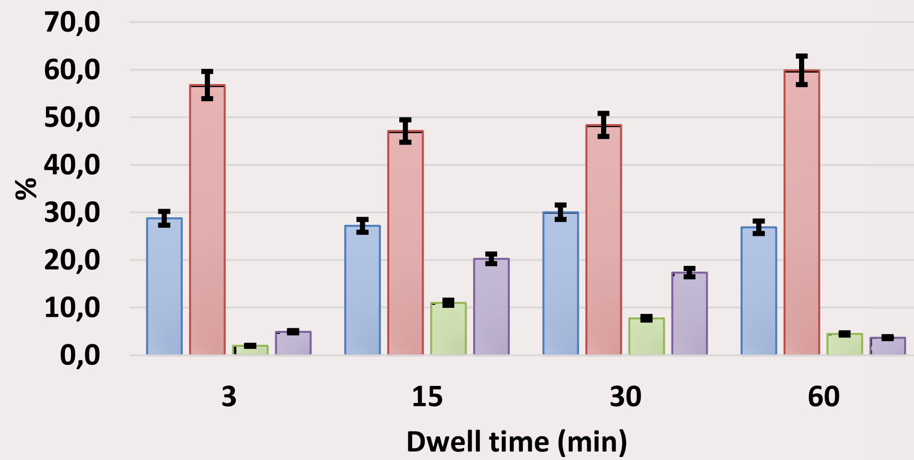
PRESSING

Study of the influence of pressing on the open porosity:

Variation pression with 3 minutes of dwell time



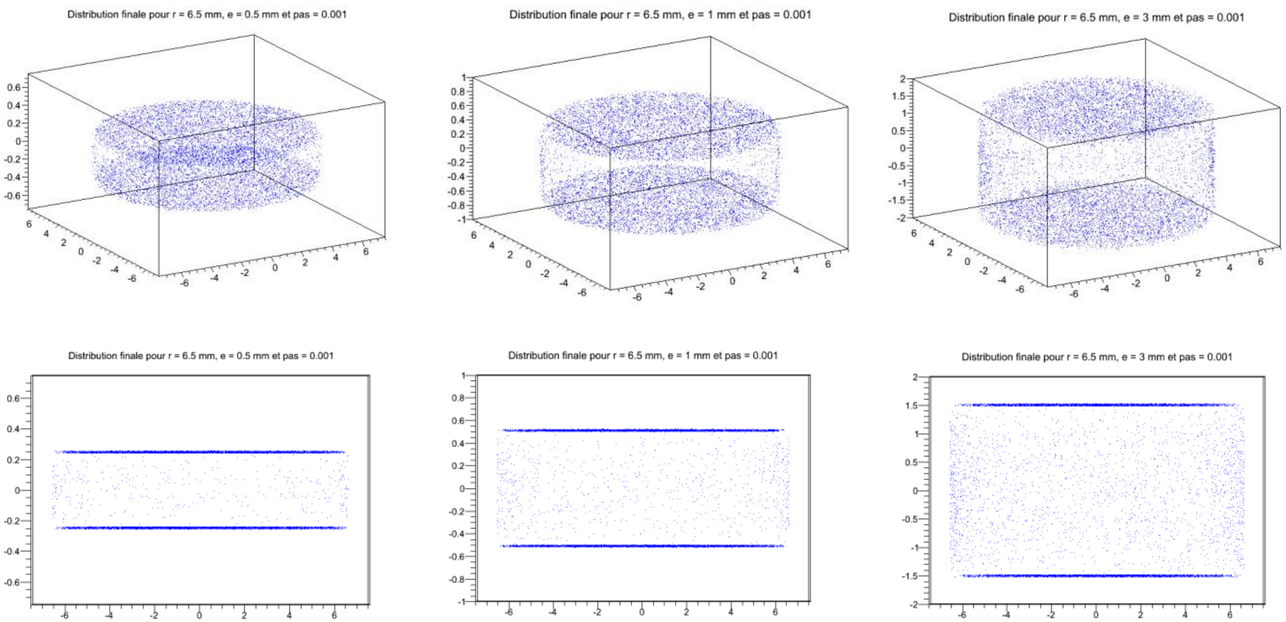
Variation dwell time with the same pression (3 tons)

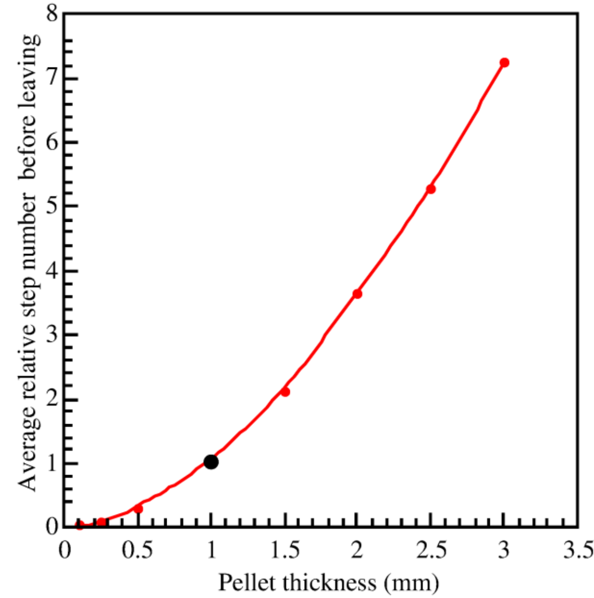


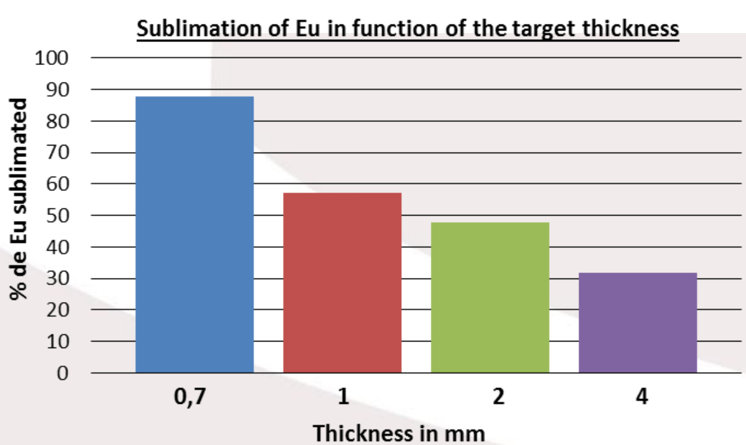
- Open porosity before sintering
- Open porosity after sintering
- Close porosity before sintering
- Close porosity after sintering

- low-tonnage improve open porosity
- No influence on the dwelling time

THICKNESS



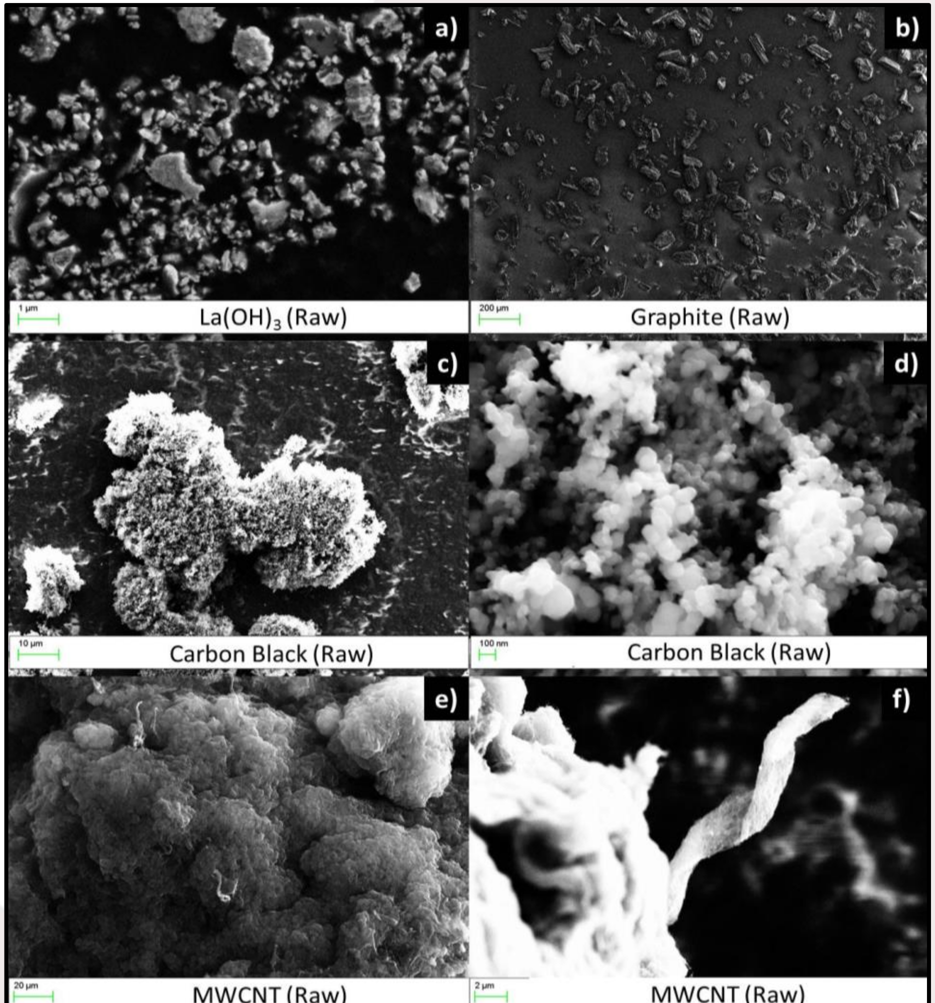




In order to correlate the thickness with the release of targets, graphite pellets have been pressed to different thicknesses and doped with stable Europium to simulate fission products. The mixture was composed 10g of graphite and 50 mg of Europium. These pellets have been heated (1200°C during 4h) and weighted in order to determine the quantity of europium get out.

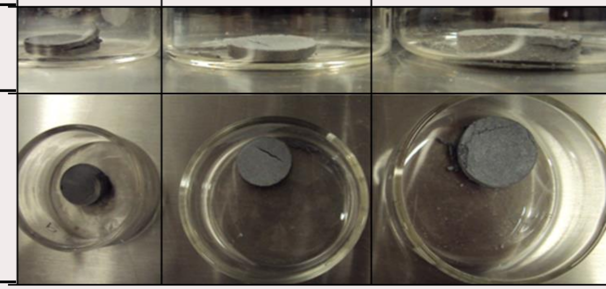
CARBON PRECURSORS

carbon sources used:



SEM pictures realised on Sigma-Zeiss

Exposition time in the air	2h	6h	12h
Side view			
Top view			



Strong hygroscopicity of lanthanum oxide
Choice to start with $La(OH)_3$
Strong degassing during carburization could induce additional porosity

Raw powder	Specific surface area (m²/g)	G_{BET} (nm)
Lanthanum hydroxide	11	122
Graphite	2	2000
Carbon nanotubes	292	36
Carbon black	37	83

SSA values determined on a Quantachrome 2002e

- **Agglomeration** of lanthanum hydroxide, carbon black and MWCNT fibers
- **Nanometric size** carbon black and MWCNTs
- **Micrometric size** of graphite and lanthanum hydroxide

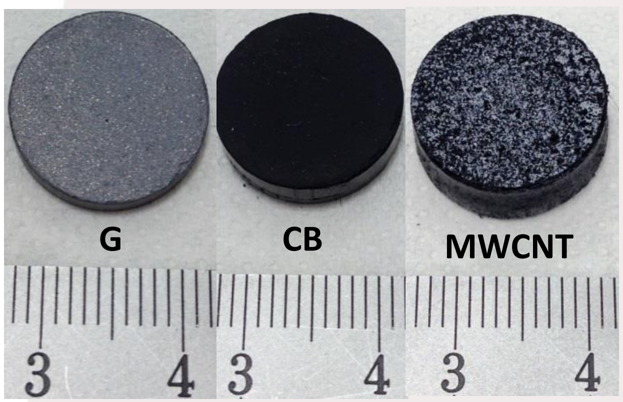
Internship M2, Julien Guillot,
Supervisor Alexander Gottberg CERN 2014

CARBON PRECURSORS

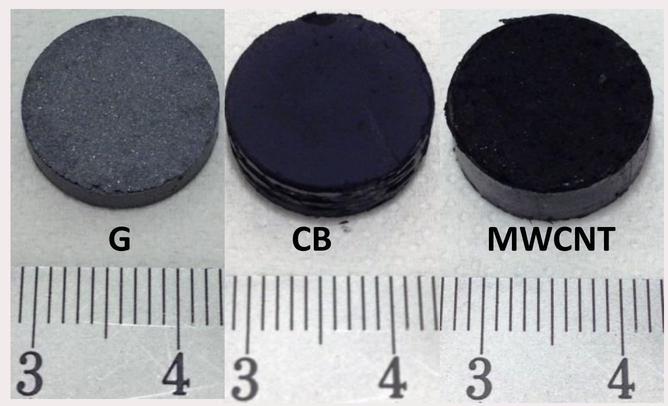
Homogeneity of the mixture

2 $La(OH)_3$ + 11 carbon pellets

pressed at 6 tons (1 ton for carbon black) for 1 min



The raw powder of $La(OH)_3$ mixed with various carbon sources in an agate mortar



The ground powder of $La(OH)_3$ mixed with various carbon sources using stirring and ultrasound

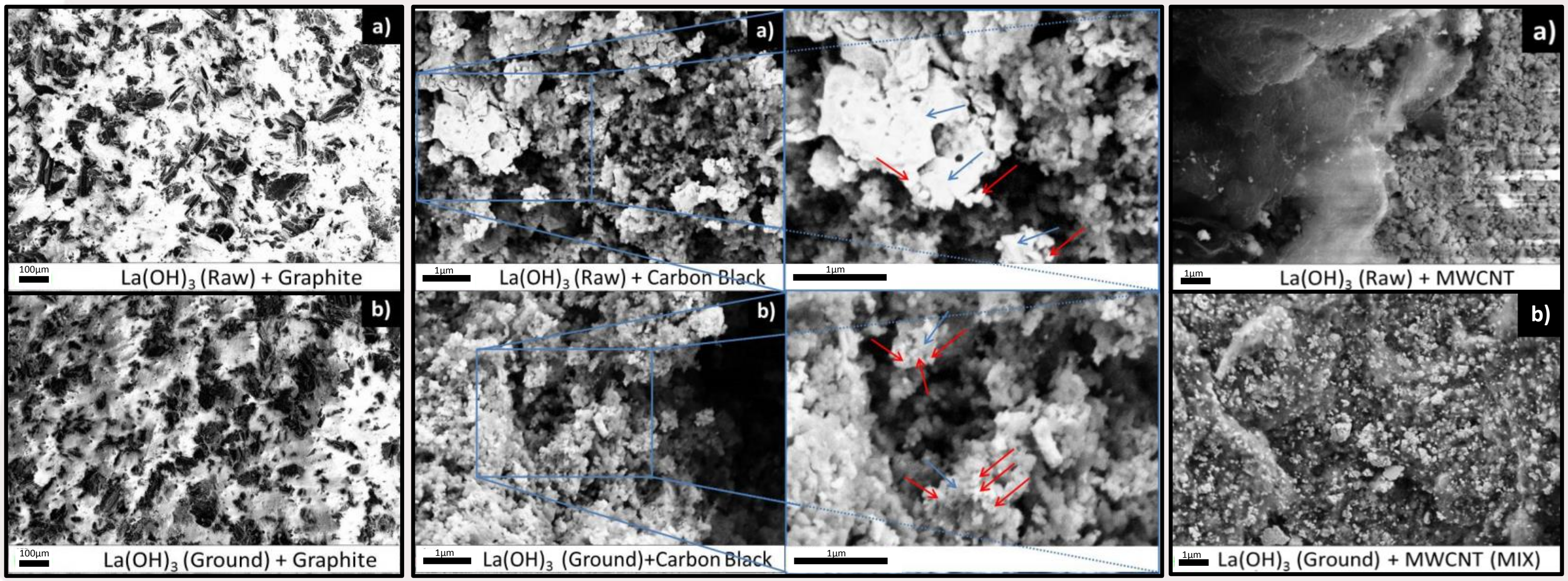
CARBON PERCURSORS

Homogeneity of the mixture

- SEM analysis of 2 $La(OH)_3$ + 11 carbon pellets

Conventional protocol

New protocol

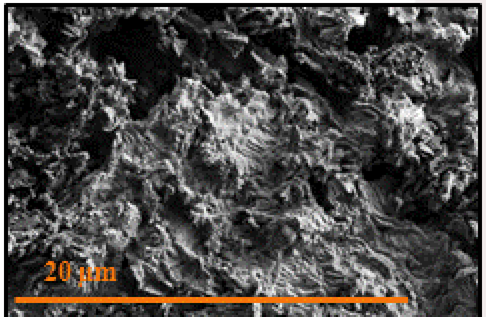


SEM pictures realised on Sigma-Zeiss

- **Loss of $La(OH)_3$ particle-size advantage after grinding due to probable fast particle growth in nano-$La(OH)_3$ agglomerates at high temperature**
- **Best mixture with wet mixing + ultrasound + + stirring for the carbon sources (CB and MWCNT)**
- **Importance of nanosized carbon precursors for good homogenization in the pellet**

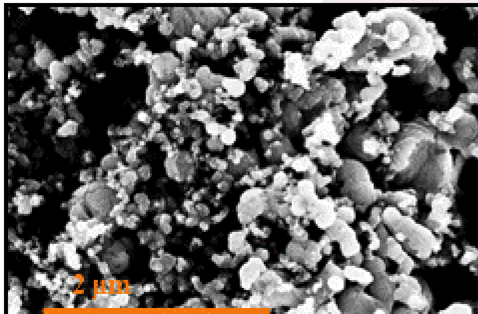
CARBON PRECURSORS

Test of the sintering: (Guillot *et al*; in preparation in NIMB 2016)

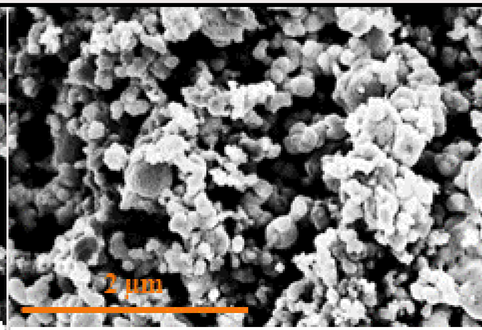


LaC_2 + 4C (Graphite)

Techniques	BET		He pycnometry		XRD	
	SSA ($m^2.g^{-1}$)		Open porosity (%)		crystallites size (nm)	
	green	carburized	green	carburized	green	carburized
Mix graphite	16	0,428	32	63	23	55
Mix carbon black	28	12	52	-	23	37
Mix MWCNT	96	49	83	83	23	26



LaC_2 + 4C (Carbon Black)

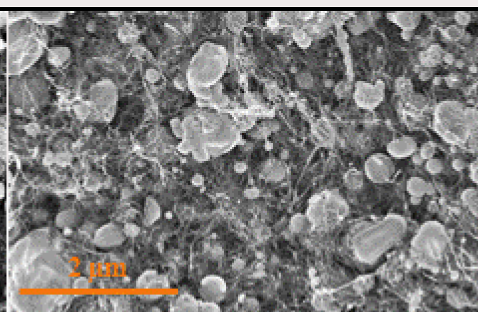


LaC_2 + 4C (Carbon Black) 3days

LaC_2 + 4C (Carbon Black) 3days

The mixture with carbon nanotubes powder has a good stability at high temperature.
After 6 days sinterring the LaC_2 grains are always nanometrics.

LaC2 + 4C (MWCNT) (no dwell time)



LaC_2 + 4C (MWCNT) 3 days

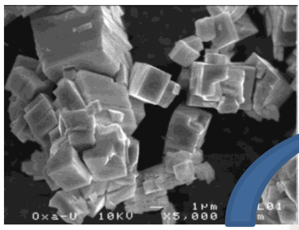
LaC_2 + 4C (MWCNT) 6days

Dwell time	BET
	SSA (m^2/g)
none	49
3 days	60
6 days	59

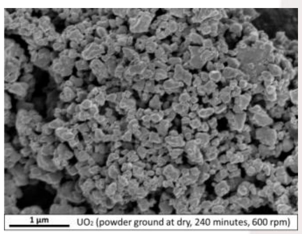
SEM pictures realised on Sigma-Zeiss

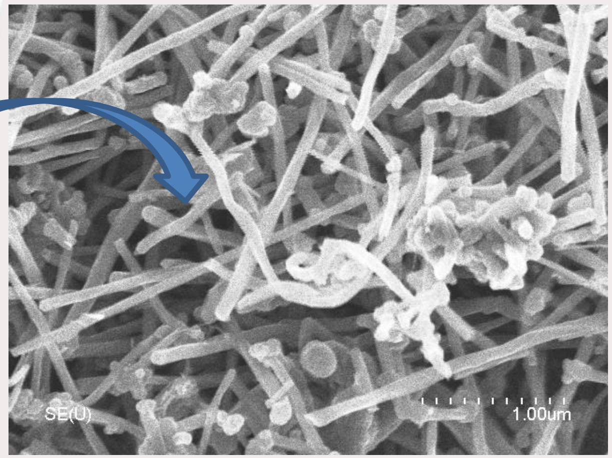
WAYS TO EXPLORE AND ALTO EXPERIMENT

Targets of uranium oxalate or oxide with carbon nanotubes by optimized mixing



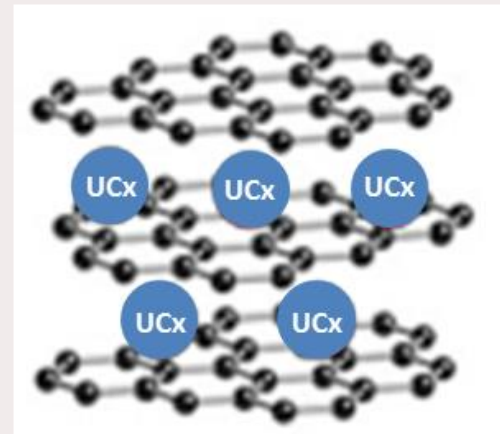
Uranium oxalate made at IPNO





Carbon nanotubes

Exfoliation of graphite sheets and inserting UCx particles



Different samples :

- UO_2ground + MWCNT (new protocol)
- UO_2 ground + graphene
- $(UO_2)_2(C_2O_4)_3$ + graphite (conventional)
- $(UO_2)_2(C_2O_4)_3$ + MWCNT (new protocol)
- Conventional PARRNe
- Sintering test on all samples
- Different carbon ratio on conventional PARRNe

Irradiations of different synthesized samples

→ TANDEM (MAY 2016)

To correlate the structure of the pellets and the FP release properties

Thank you for your attention