ActILab closing meeting (WP8, FP7 ENSAR)

Contribution IPNO

R&D on UCx TARGET

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ACTILAB JRA

OBJECTIVES OF THE TARGET R&D

Nowadays

The most widely used ISOL targets = **uranium carbide + graphite (UC_X), mostly UC_2**

→ ISOLDE, ALTO, TRIUMF, SPES, RISP project

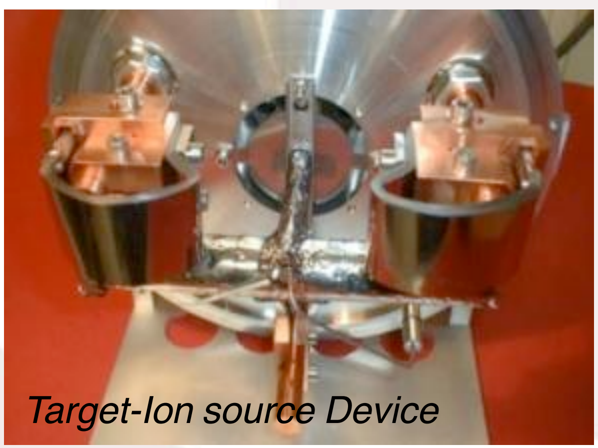
Concentration of ^{238}U ~ **3 g/cm^3**

Operate at temperatures ranging from **2 000 °C to 2 200 °C**

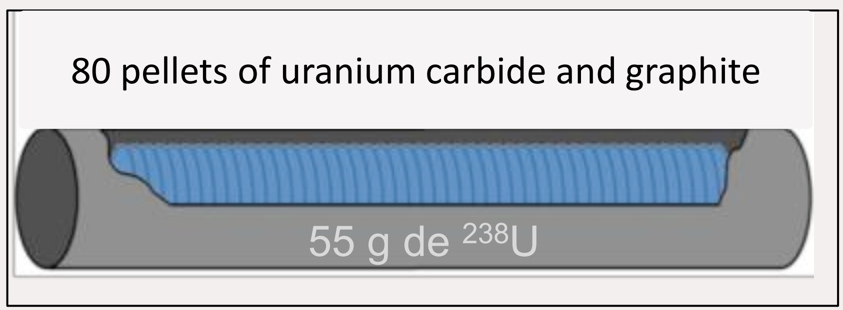
Today : ALTO, 10^{11} fissions / sec, inaugurated in May

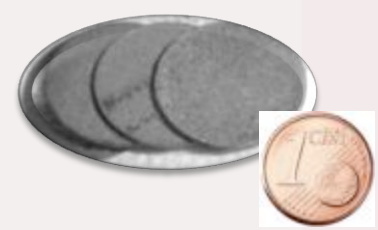
Tomorrow : **SPIRAL2**, 10^{13} fissions / sec

Future : **EURISOL**, 10^{15} fissions / sec



Target-Ion source Device





Carburization

$$UO_2 + 6C \rightarrow x\,UC_2 + (1-x)\,UC + (3-x)\,C + 2CO_{(g)}$$

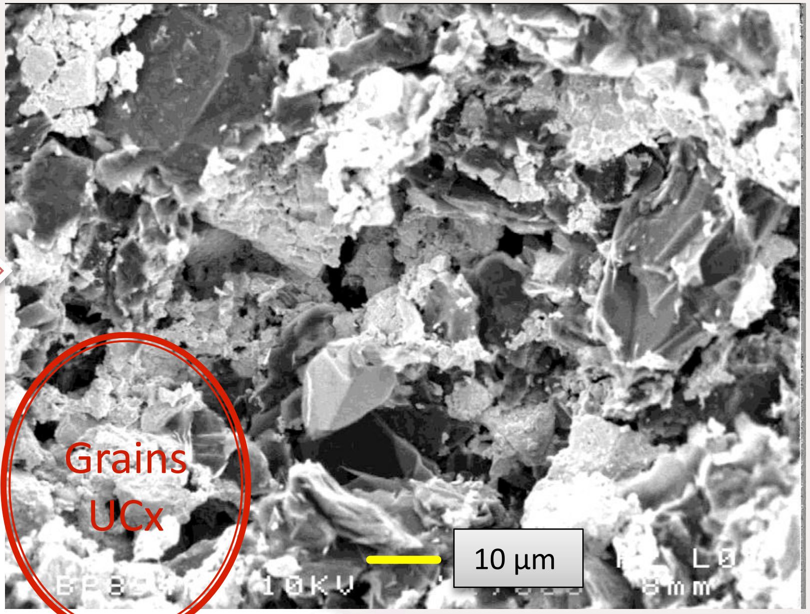
HOW TO INCREASE RIB?

Intense beam → improvement of the FPs releases

High production rate of FPs ⇨ **Increasing uranium density**

Favour the FPs releases, particularly crucial for the short-lived species ⇨
Controlling the porosity
Reducing the thickness of pellets or the UC_x grain size

Uranium Photofission
2000°C / 10^{-5} mbar



Mass spectra → RIB
And in particular short-lived species

- Diffusion through the UCx grains
- Effusion via porosity
- Low volatility of studied elements

OBJECTIVES OF THE TARGET R&D

To get an in-depth understanding of the properties of the material as a function of the nature and the proportion of the reactants and of the method of synthesis.

To investigate correlations between the release efficiencies and the physicochemical properties of the samples.

How ?

Study of the key parameters of UCx pellets syntheses

Systematic characterizations to identify the stabilized phases, porosity and microstructure

Systematic γ-spectrometry measurements of pellets irradiation and heating at high temperature (T ≥ 1700 °C)

What have we developed to do that?

A method to simulate irradiation tests at ISOL facilities and to measure the release properties of pellets after irradiation and after heating

Only 2 pellets instead of a complete target

First results published in November 2012 by
B. Hy et al., Nuclear Instruments and Methods in Physics Research B 288 (2012) 34 – 41

CONCLUSIONS OF 1ST RESULTS

These **two preliminary release experiments** allowed us to **validate the experimental protocol** in order to qualify definitely and study the “ideal” sample for the common operating conditions (2000 °C in vacuum).

Improvements were necessary

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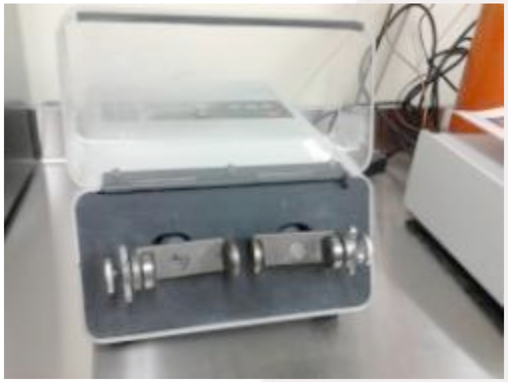
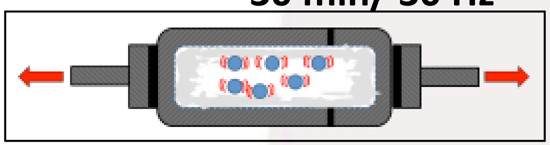
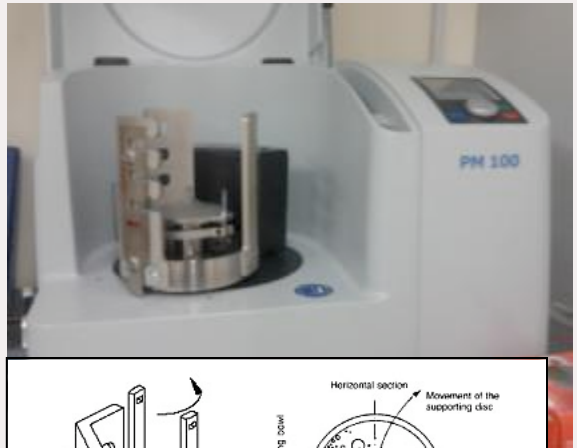
REPRODUCIBILITY – GRINDING, NEW WAY

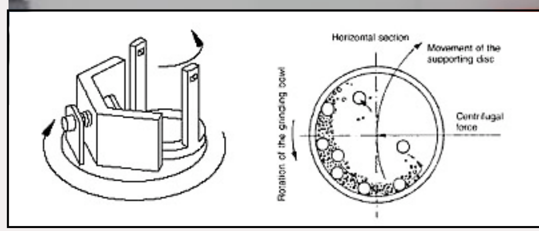
- Grinding with planetary miller :



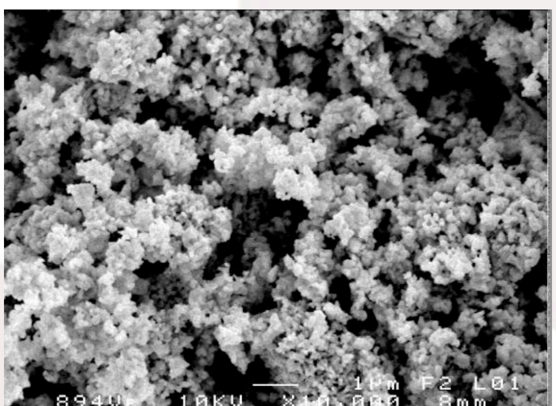
30 min/ 30 Hz

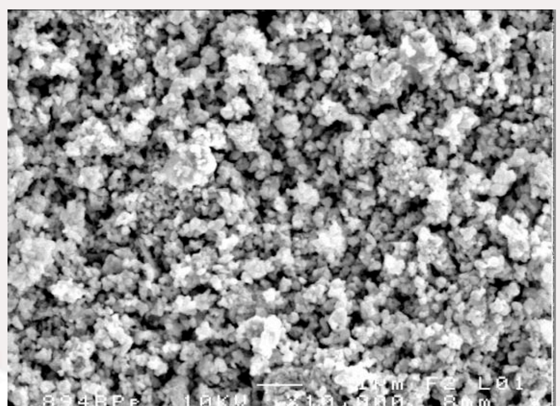






Improving the homogeneity in shape and grain size





La_2O_3 powder, 3 mm diameter balls, 30 min of grinding

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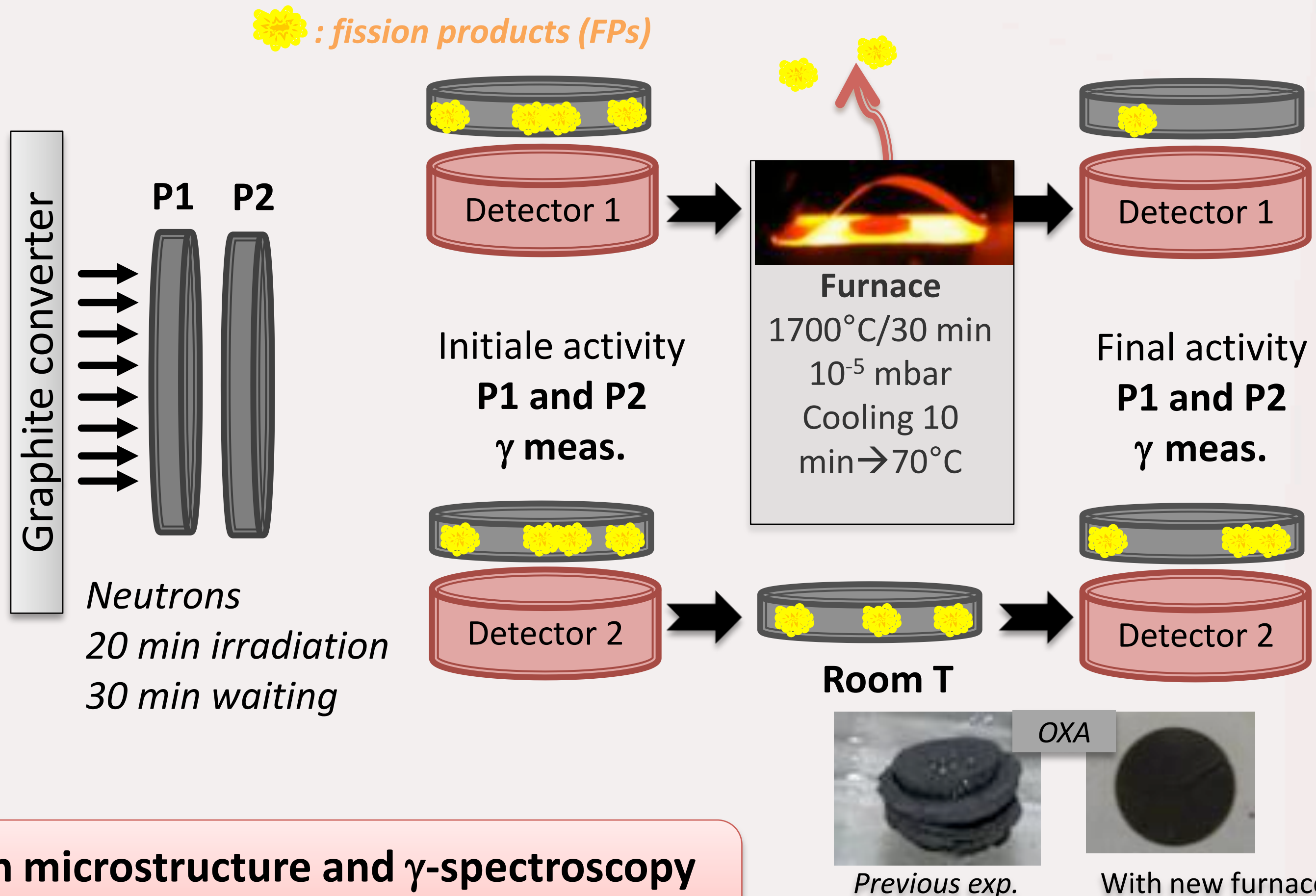
ÉCOLE POLYTECHNIQUE ParisTech
ICMMO
LSI

ON-LINE IRRADIATION TESTS : IMPROVEMENTS

1st tests : Hy et al., NIMB, 288 (2012) 34–41

Deuterons
27 MeV
20 nA
ALTO



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

SAMPLES WITH DIFFERENT MICROSTRUCTURES

typically 1-2mm thickness and 10-20mm diameter

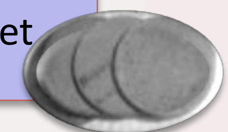
Sample	Uranium	Carbon	ratio C/U	Milling of uranium powder	Carburization
OXA	IPNO oxalate	Graphite	3	No	16h at 1770° C
COMP30	IPNO oxalate	Graphite +30 wt % of microfibres	3	No	16h at 1770° C
PARRNe 371	natural UO_2 depleted 0.3% - ref. MN371	graphite	6	Mixer miller PM100 RETSCH	16h at 1770° C
PARRNe 894	natural UO_2 depleted 0.25% - ref. MN894	graphite	6	Mixer	16h at 1770° C
PARRNe 894 BP	natural UO_2 depleted 0.25% - ref. MN894	graphite	6	Planetary miller PM200 RETSCH	16h at 17700C
CNT	Westinghouse UO_2	Carbone nanotubes	6	No	20 min at 1700° C

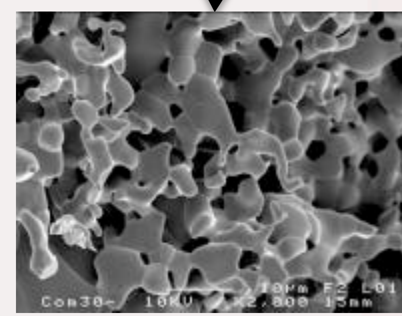
CHARACTERISAZATION OF THE MICROSTRUCTURE

Study of the microstructures of the pellets ⇔ FPs release ⇨ reproducibility

Multi-parameter study : powder grain size/shape, ways of pressing and sintering ⇔ porosity - structure - microstructure

U + C powders mixing: 3 ways of synthesis+ planetary miller → Pressing 3 to 6 GPa → Green pellet → Carburization: Reactive sintering @ 1800°C → Sintered pellet





XRD, He pycnometry, Hg porosimetry, BET et laser granolumetry @ IPN

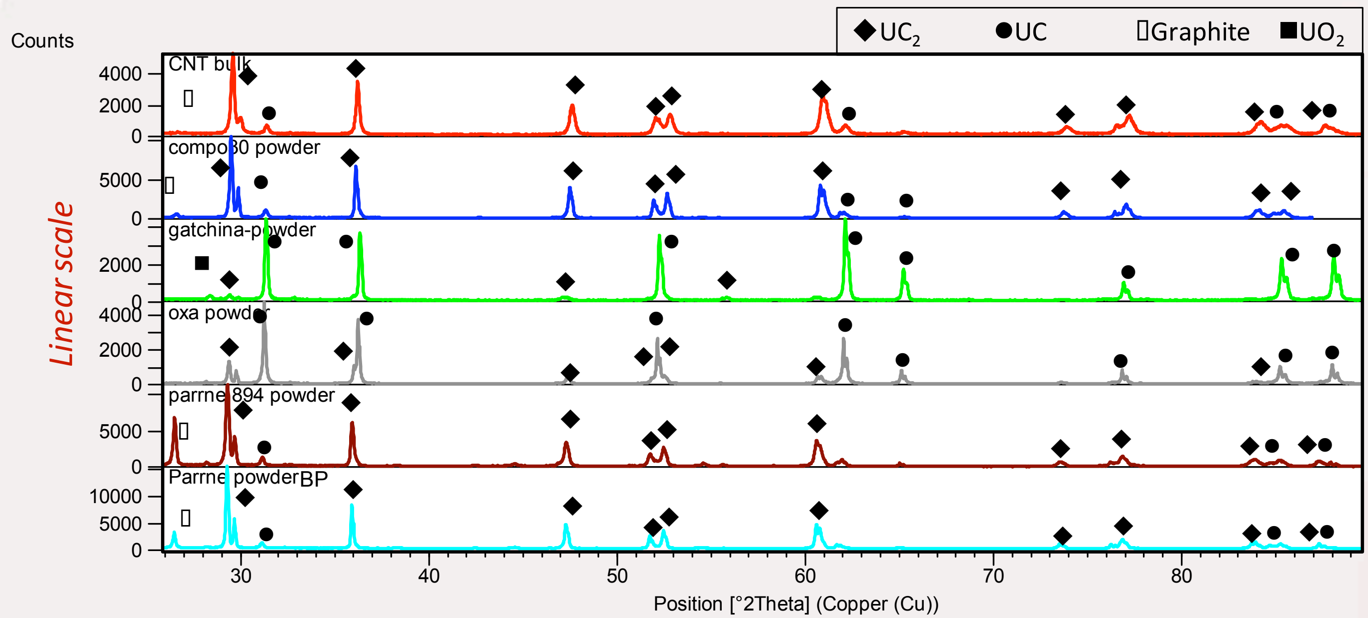
SEM/EDS on radioactive samples @ Sciences Chimiques de Rennes

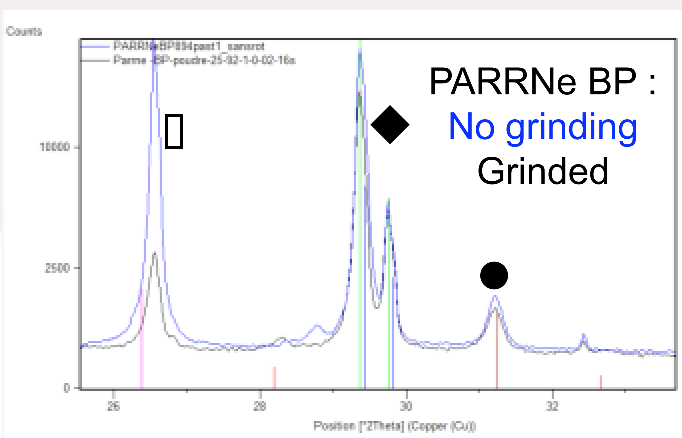
SEM/EDS on non radioactive samples (La_2O_3 etc...) @ ICMMO

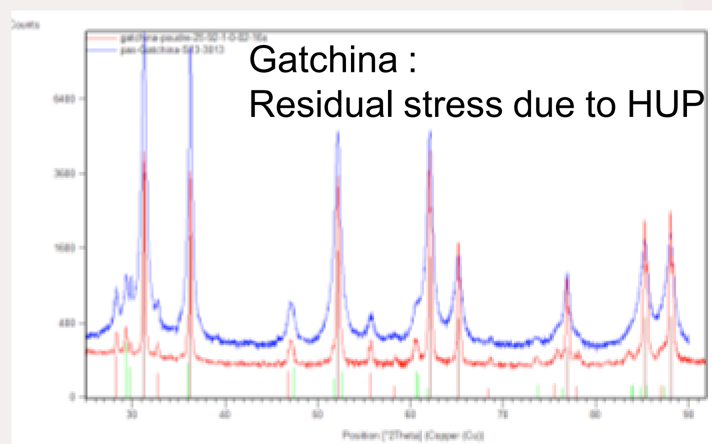
XRF and XRD high resolution @ LISA ITODYS

Université de Rennes 1 · Université Paris Sud · Université Paris Diderot

XRD : PHASE IDENTIFICATION







Need of grinding of pellet (agate mortar)
Pyrophoricity

XRD : QUANTITATIVE PHASE ANALYSIS

1. Need of grinding the pellet
2. Quantitative phase analysis possible to compare relative quantities of UC and UC2.
3. Quantitative phase analysis including graphite difficult due to texture and absorption. Not recommanded

Sample	Phase and their relative proportion (wt %)
OXA	UC / UC_2 and 70.5 / 29.5
COMP30	UC / UC_2 and 8.6 / 91.4
PARRNe 371	UC / UC_2 and 10.6 / 89.4
PARRNe 894	UC / UC_2 and 10.9 / 89.1
PARRNe 894 BP	UC / UC_2 and 5.8 / 94.2
CNT	UC / UC_2 and 14.7 / 85.3

Carburization equations should be rewritten as:

OXA: $U(C_2O_4)_2{,}2H_2O + 3C \rightarrow x\,UC + (1-x)\,UC_2 + (1+x)\,C + 4\,CO_{2\,(g)} + 2H_2O_{(g)}$
Gatchina: $U + (1+x)\,C \rightarrow (1-x)\,UC + x\,UC_2$
COMP30 : $U(C_2O_4)_2{,}2H_2O + 4C \rightarrow (1-x)\,UC_2 + x\,UC + (2+x)\,C + 4CO_{2(g)} + 2H_2O_{(g)}$
Others : $UO_2 + 6C \rightarrow x\,UC_2 + (1-x)\,UC + (3-x)\,C + 2CO_{(g)}$

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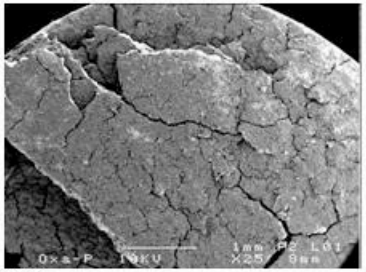
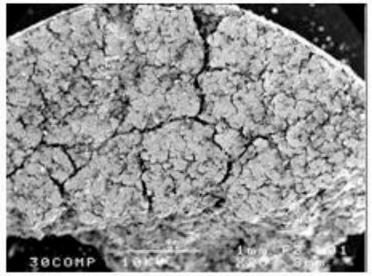
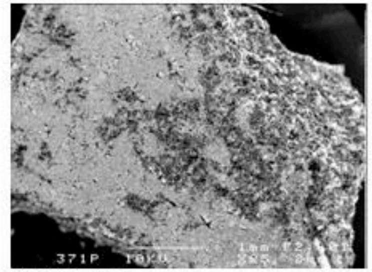
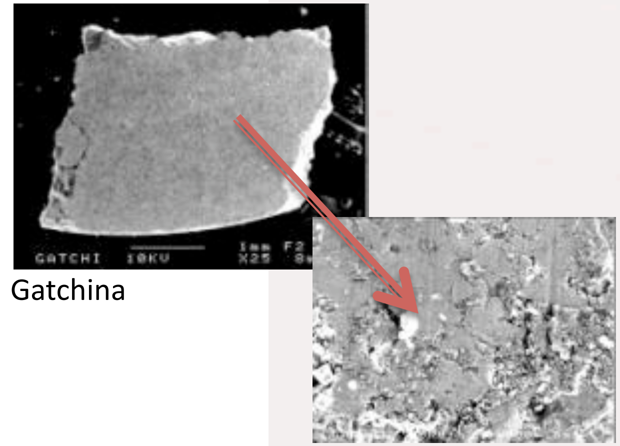
MICROSTRUCTURES BY SEM AND HE PYCNOMETRY

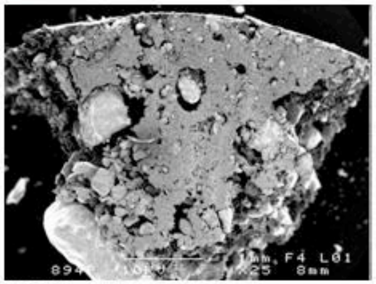
surface

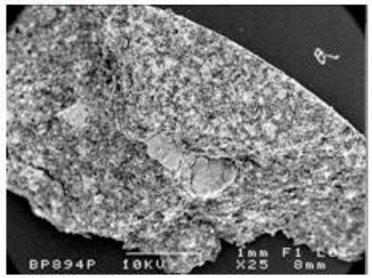

OXA

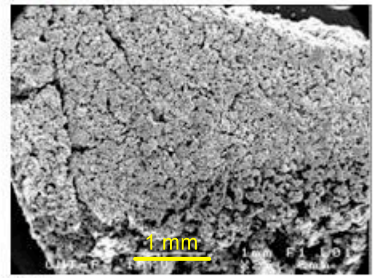

COMP30


PARRNe 371


Gatchina

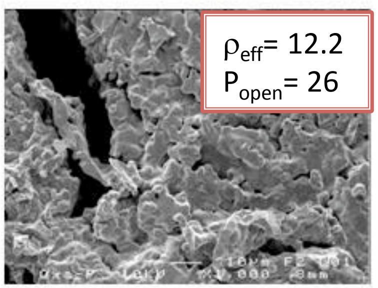

PARRNe 894


PARRNe BP

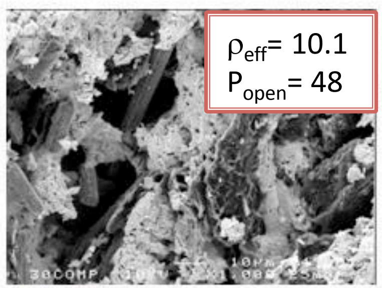


CNT

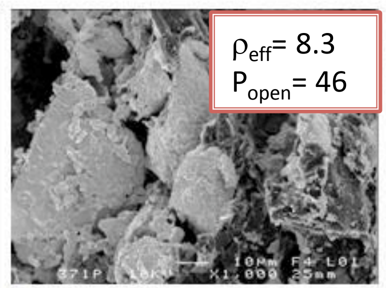
fracture



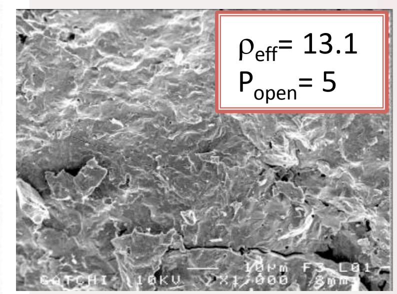
OXA



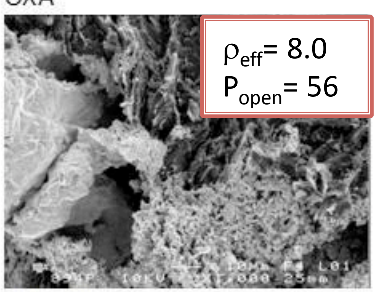
COMP30



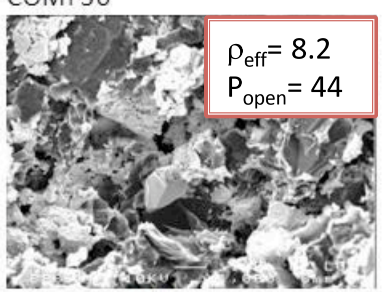
PARRNe 371



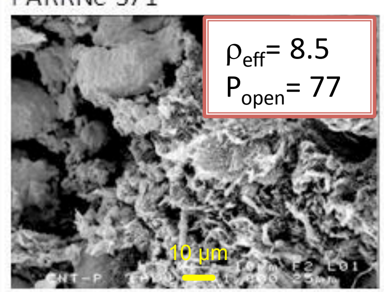
Gatchina



PARRNe 894



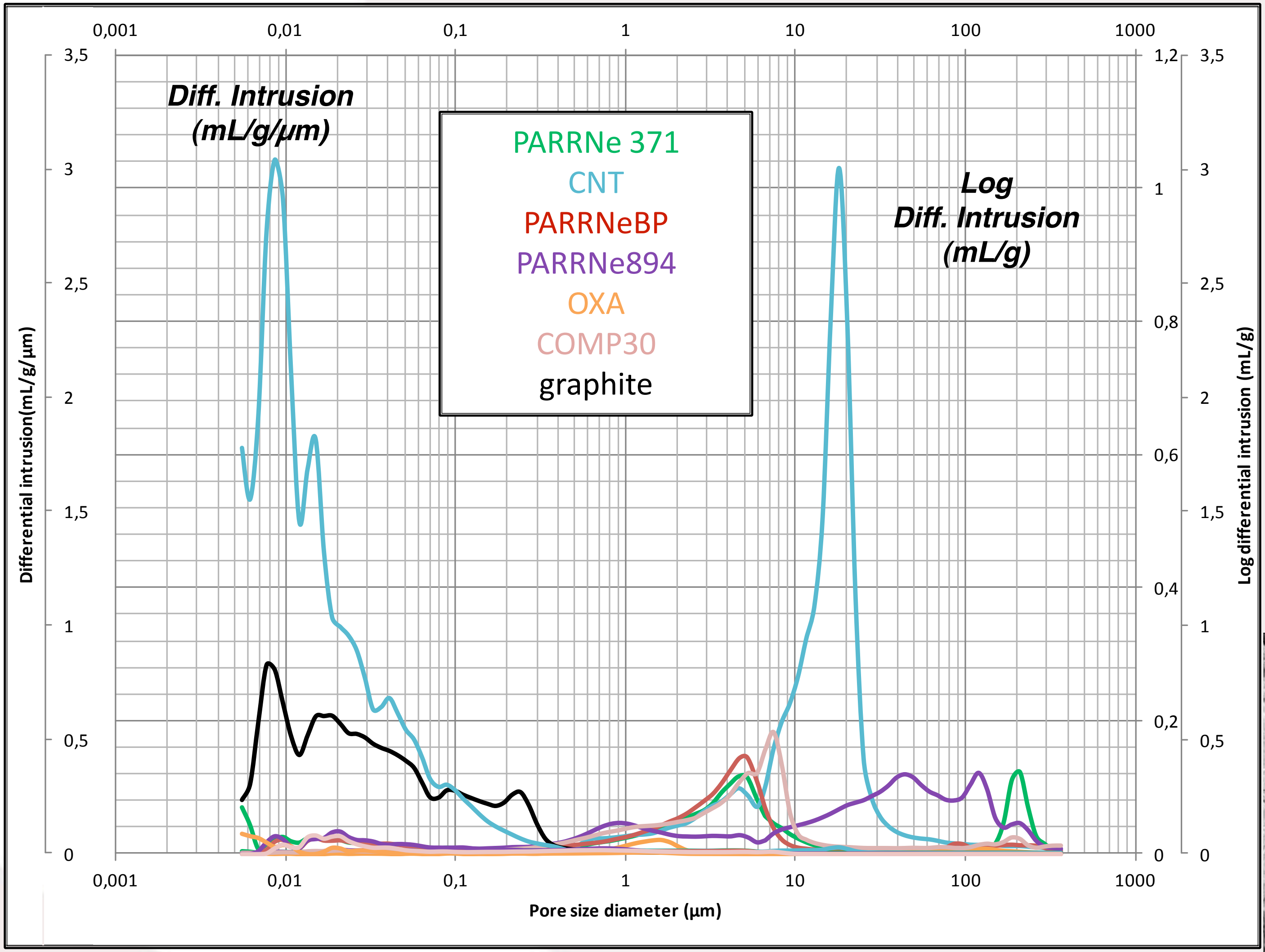
PARRNe 894BP



CNT

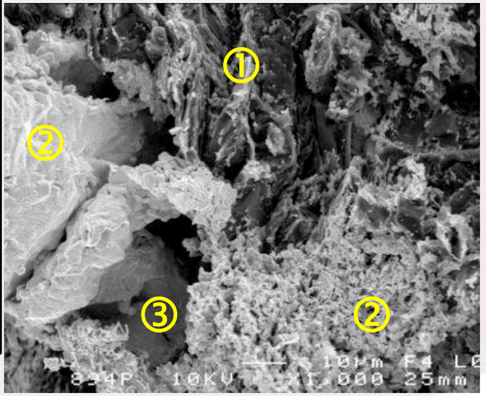
He pycnometry:
ρ_{eff} : $g.cm^{-3}$
P_{open}: %

PORE SIZE DISTRIBUTIONS



Hg porosimetry : Necessary to obtain a quantitative pore size distribution

PARRNe894 :



① Graphite or CNT

② UCx agglomerates

③ Very large pores

CONCLUSIONS ON PHYSICOCHEMICAL CHARACTERIZATIONS

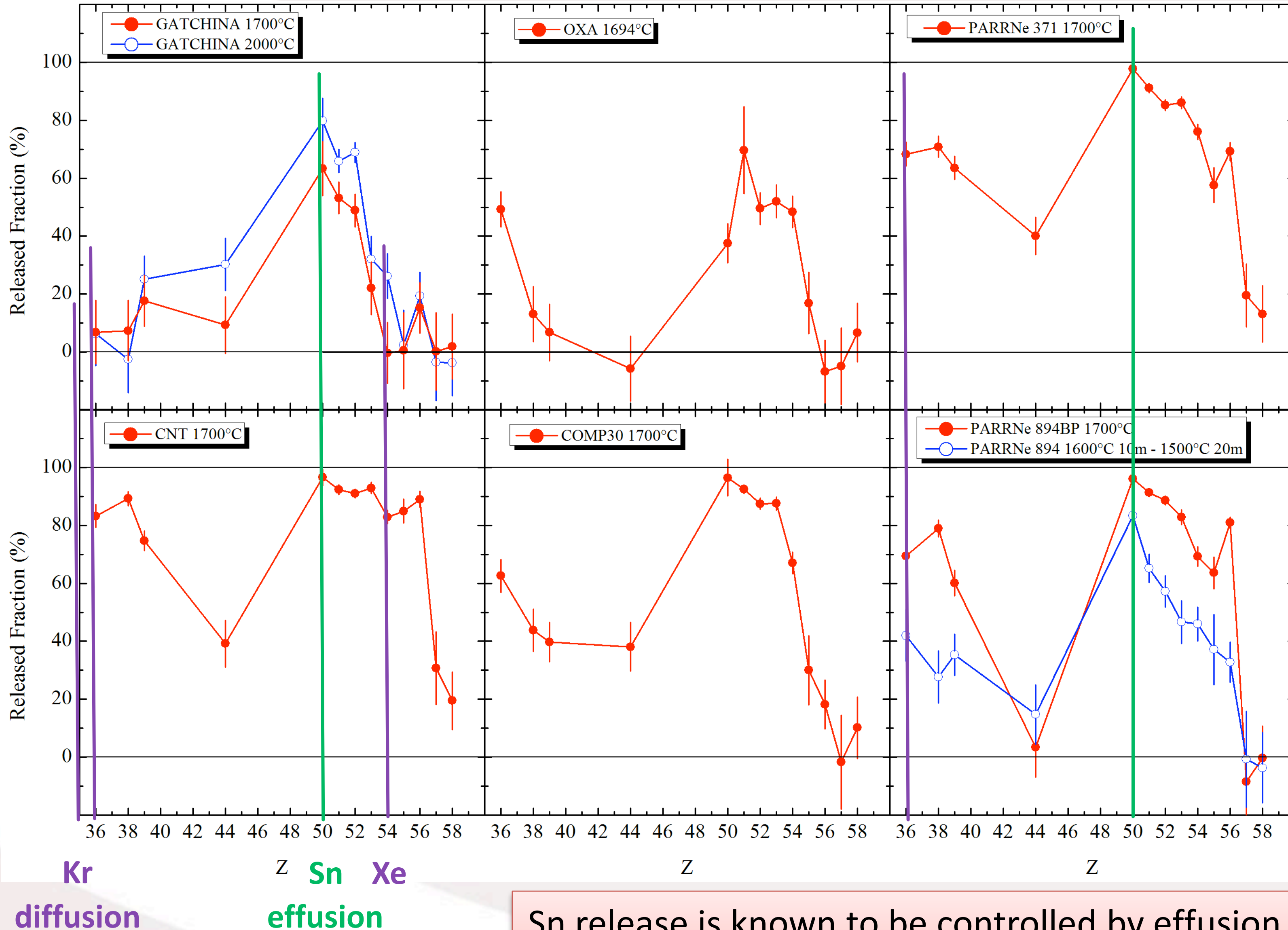
The grinding of uranium dioxide improves the homogeneity of the microstructure after carburization. Of course, the shrinkage is increased by 20% but the open porosity is still high. The carburization of grinded powder tends to stabilize mostly UC_2.

Using uranium oxalate instead of uranium dioxide favors degassing during carburization but the closed porosity obtained is the highest.

Using microfibers favors the formation of very large pores (cracks on surface) but the average open porosity is not increased in comparison with the samples made of graphite only.

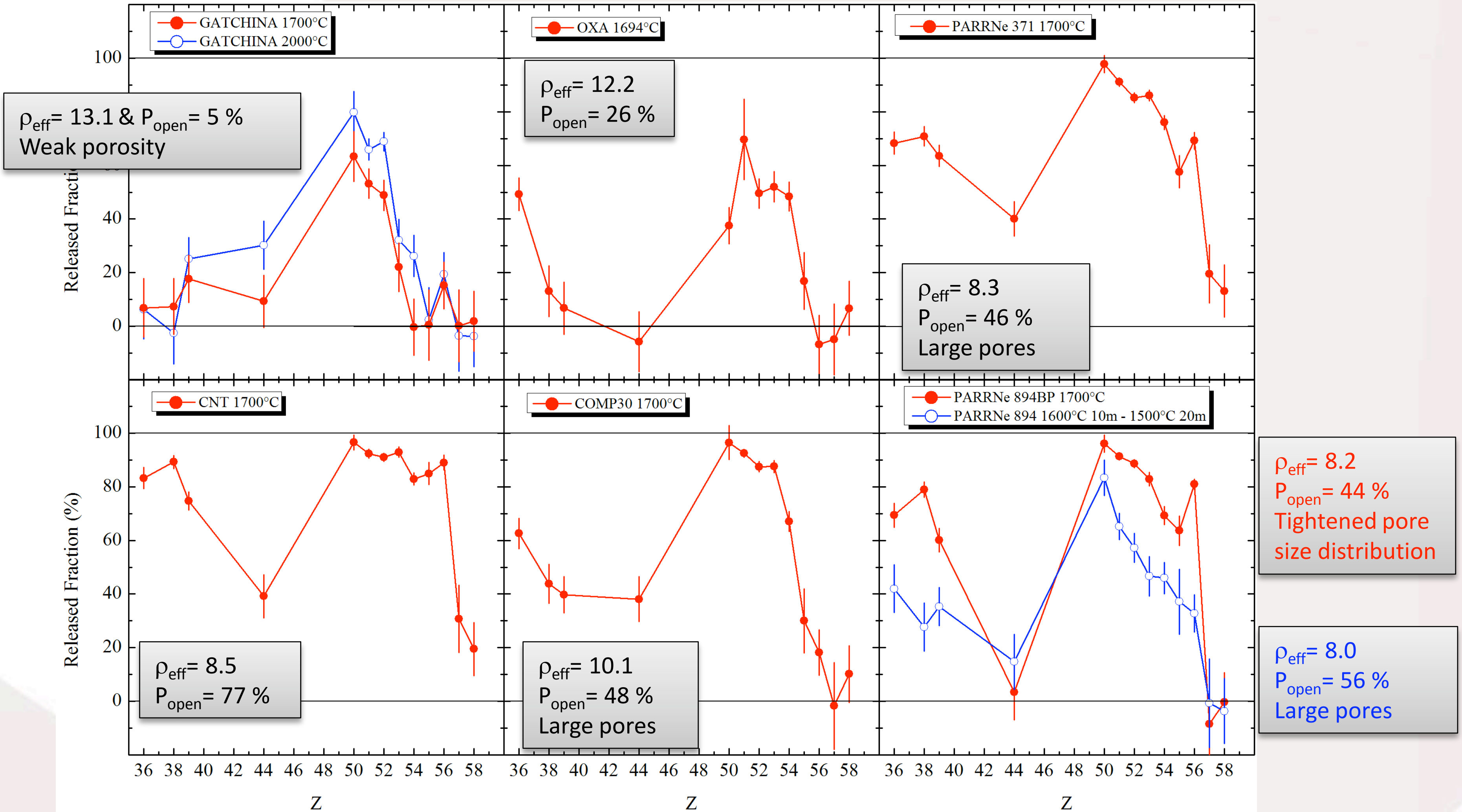
The use of CNT improves the open porosity. The microstructure of the pellet is more homogeneous with a limited grain growth. Nevertheless, it should be reminded that the carburization took place only 20 minutes, instead of 17 hours for the other samples.

RELEASE FRACTIONS OF SAMPLES

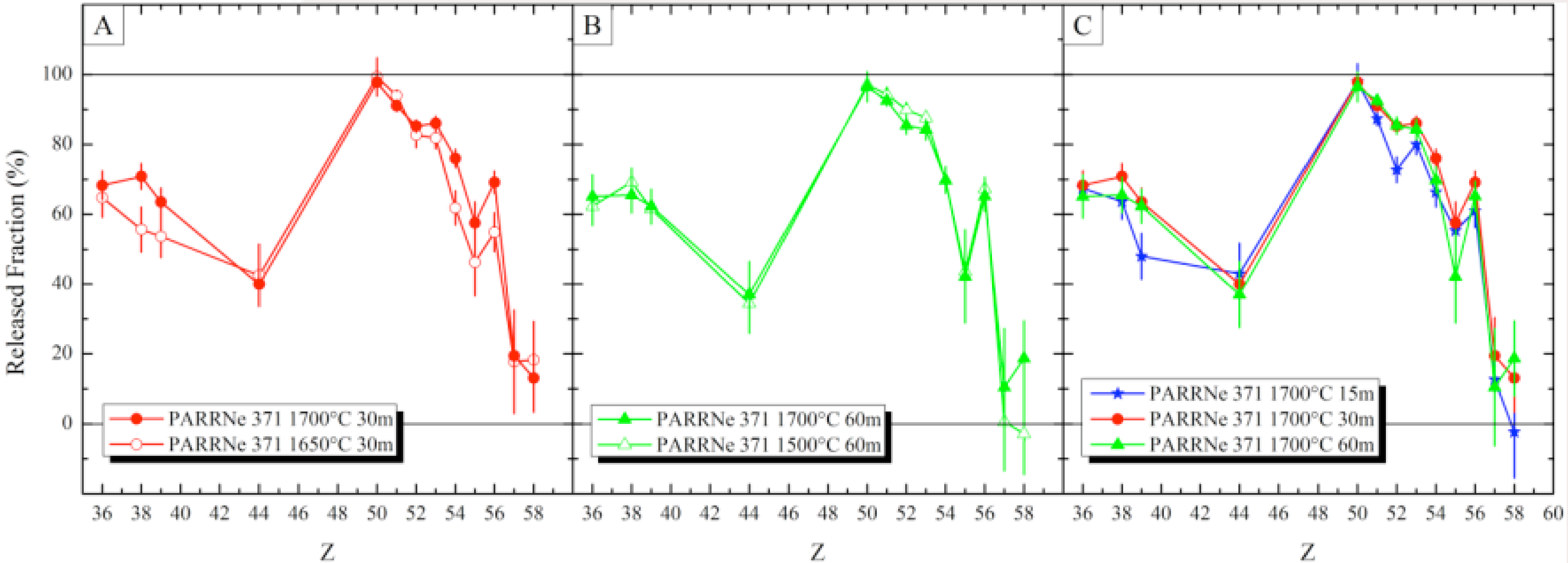


Sn release is known to be controlled by effusion whereas Kr and Xe by diffusion

CORRELATION MICROSTRUCTURE AND FPS RELEASES



RELEASE PARAMETERS & REPRODUCIBILITY

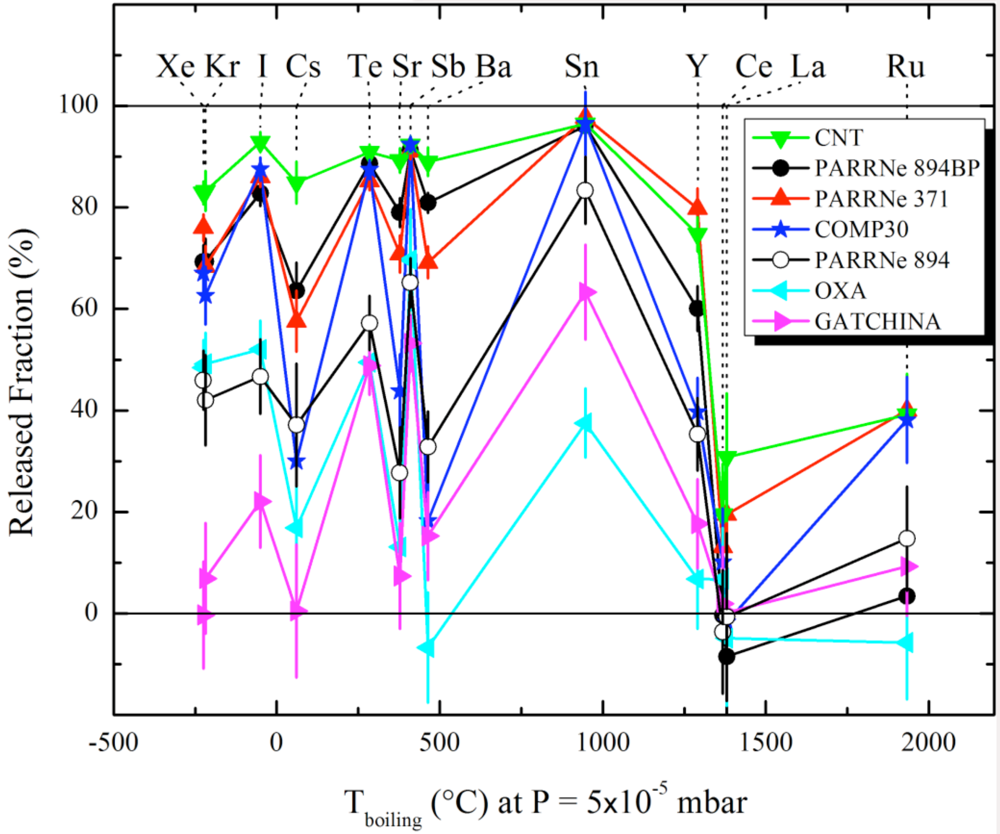


Influence of :

- Heating temperature
- Heating time

→ **Reproducibility in our measurements**

RELEASE FRACTIONS VS BOILING TEMPERATURE



Classifications of samples :

CNT

PARRNe 894BP

PARRNe 371

COMP30

PARRNe 894

OXA

GATCHINA

Ce, La and Ru are poorly released by all the samples.

Ce and La →resistance to release attributed to the chemical analogy of Ln with U

Ru → due to the heating temperature used (1700 ° C) lower than the Ru boiling T

.

CONCLUSIONS

Heating :

Validation of the on-line tests (irradiation, heating @ 1700°C and 2000°C under secondary vacuum

↳ Good mechanical stability of the pellets

Release efficiencies :

Validation of the γ-spectrometry measurements

Physico-chemical characterizations:

Clearly, diffusion is strongly correlated with the open porosity but not only

↳Best candidates : CNT & PARNNeBP (now used @ALTO)

① High porosity

②Tightened pore size distribution

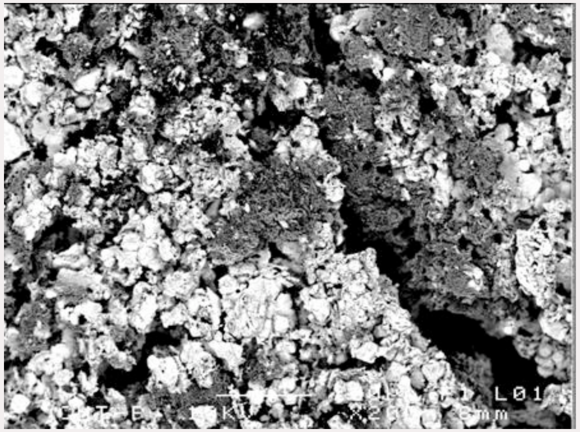
③Homogeneity of the grain size and the microstructure

CONCLUSION AND PERSPECTIVES

ALTO RIB in 2015, June

Test of CERN CNT target prepared as conventional PARRNe, i.e. without a optimized mixing of UO_2 and CNT before carburization

Physico-chemical characterizations are in progress



@ IPNO

PHD work on nanostructured UCx targets and production of refractory lanthanide beams