# STUC Project :
# **ST**udies of **U**ranium **C**ompounds

## Matthieu Lebois

**Project member's list:**

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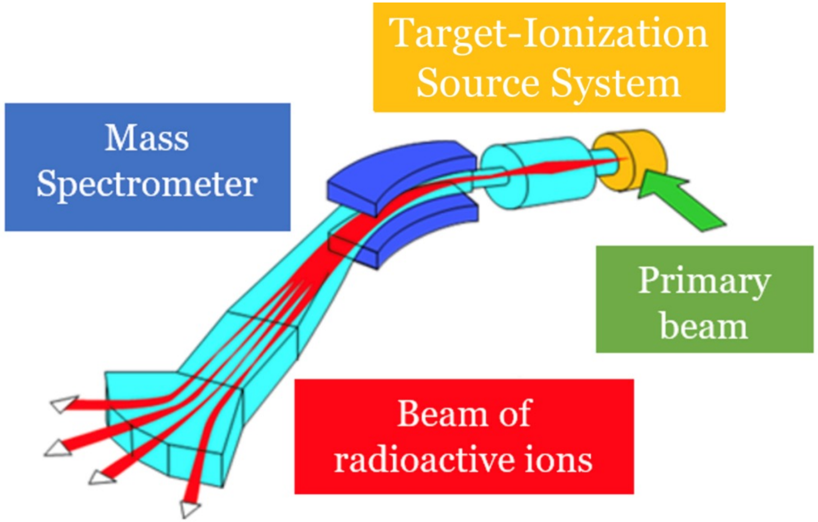
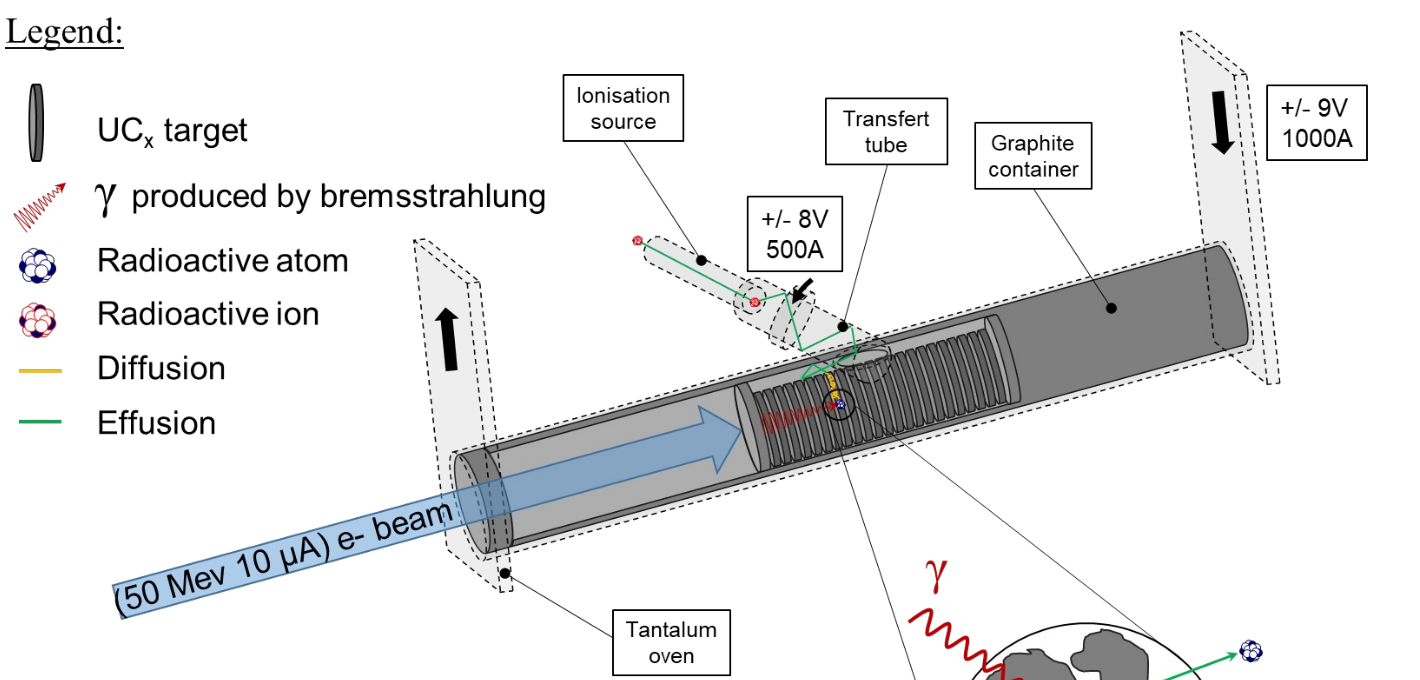
# Context

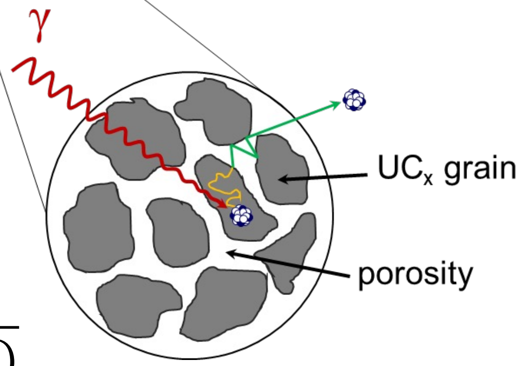
**CONTEXT**

- Strong demand from nuclear physics for exotic beams to study nuclear structure by β-decay
- Production of radioactive beams at ALTO: ISOL (Isotope Separation On-Line) method

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



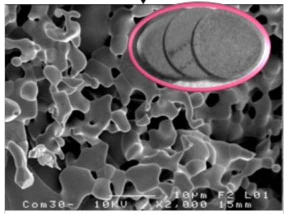

$$\boldsymbol{\varepsilon_r = \varepsilon_{(diff \rightarrow eff)}}$$

$$\varepsilon_{diff} \rightarrow C\sqrt{\frac{D}{\lambda . a^2}} \qquad \varepsilon_{eff} \sim \frac{1}{1 + \lambda n(t_{vol} + t_{coll})}$$

# How to build a target ? What caracterization?

- **Synthesis of $UC_x$ target:** $UO_2 + 6C \rightarrow (1-x)UC + xUC_2 + (3-x)C + 2CO_{(g)}$

Mixing of uranium and carbon precursor powders → Pressing ~ 220 MPa → Green Pellet → Carburization : 1800°C at $10^{-3}$ Pa → Carburized Pellet

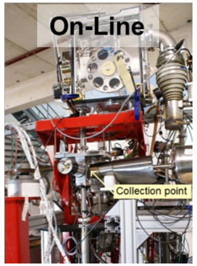



- **Techniques used for the physico-chemical characterization of $UC_x$ targets :**

XRD | SEM | BET | He Pycnometry | Hg Porosimetry

- XRD: Analysis of phases
- SEM, BET: Grain and aggregate size
- He Pycnometry, Hg Porosimetry: Proportion and distribution of porosities

- **Equipment for measuring released fractions (off-line) and production (on-line) :**

# Study on the influence of the microstructure

| N° sample | Sample name | |
|---|---|---|
| 1 | $UO_2$ ground+CNT PM | ● ● ● ● ○ |
| 2 | $UO_2$ ground+CNT UM | ● ● ● ● ○ |
| 3 | $UO_2$ ground+graphene | ● ● ● ● ○ |
| 4 | OXA+graphite PM | ● ● ● ● ○ |
| 5 | OXA ground+CNT UM | ● ● ● ● ○ |
| 6 | OXA+CNT UM | ● ● ● ● ○ |
| 7 | PARRNe BP894 | ● ● ● ● ○ |
| 8 | PARRNe BP897 PM | ● ● ● ● ○ |
| 9 | PARRNe BP897 PM 12d | ● ● ● ● ● |
| 10 | $UO_2$ ground+CNT PM 12d | ● ● ● ● ● |
| 11 | $UO_2$ ground+CNT UM 12d | ● ● ● ● ● |
| 12 | $UO_2$ ground+graphene 12d | ● ● ● ● ● |
| 13 | $UO_2$ ground+CNT-5mol UM | ● ● ● ● ○ |
| 14 | $UO_2$ ground+CNT-7mol UM | ● ● ● ● ○ |

Uranium precursor:

- Oxyde d'uranium
- Oxalate d'uranium

Carbone precursor:

- Graphite
- Graphene
- CNT

Molar ratio C/U

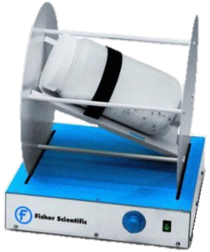
- 5
- 6
- 7

12-day heating after carburation :

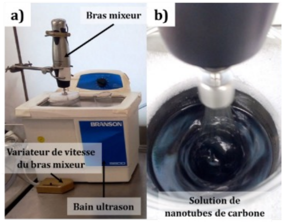
- Yes
- ○ No

Precursor powder mixing:

- Robin™ Powder Mixer



- Ultrasonic liquid mixing



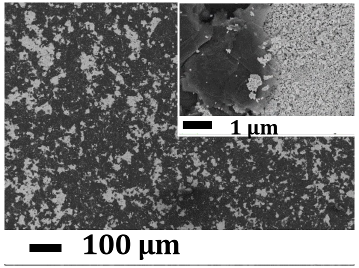

- Graphene blender

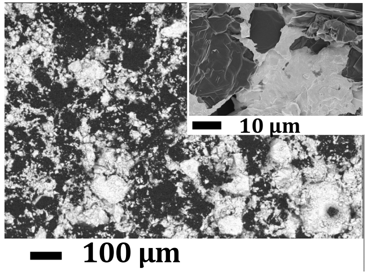
J. Guillot *et al.* NIMB 2018, 2019, 2020

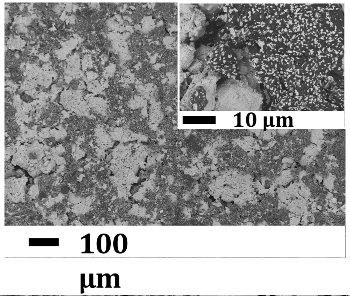
# Study on the influence of the microstructure

Green pellet | Carburized pellet | Carburized pellet + 12-day heating

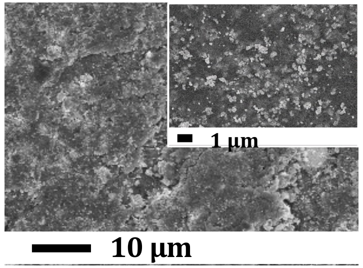
Powder Mixer

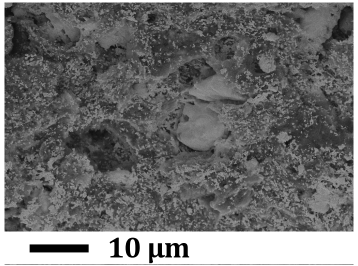


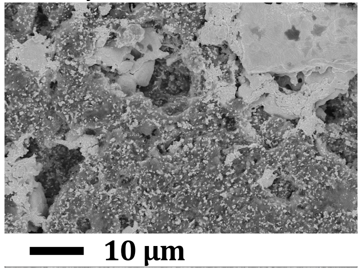



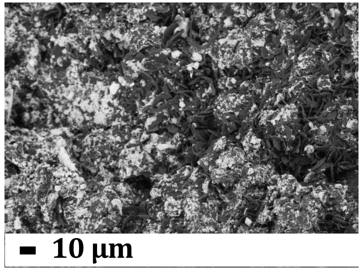

Ultrasonic liquid mixing

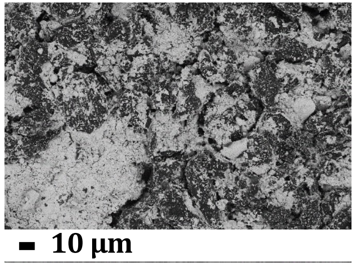


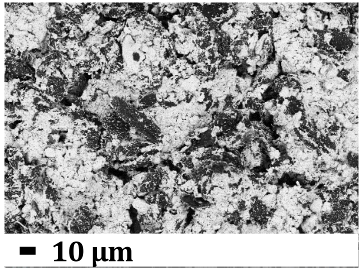




Graphene blender

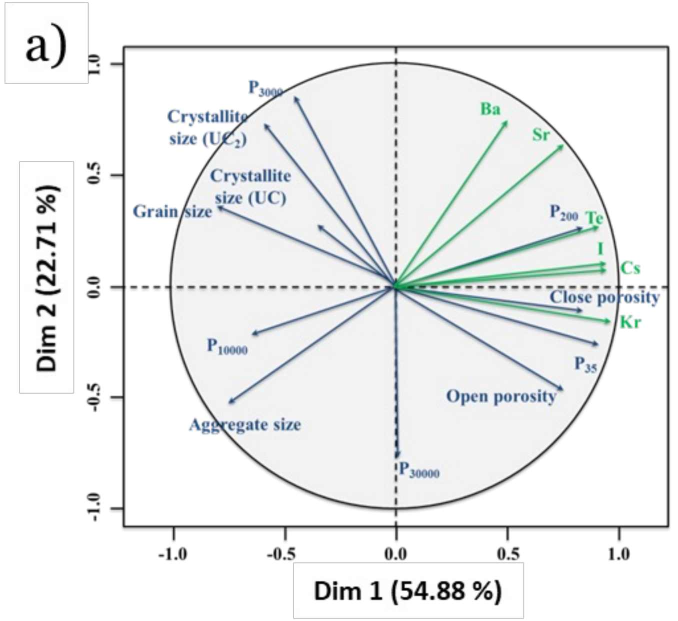


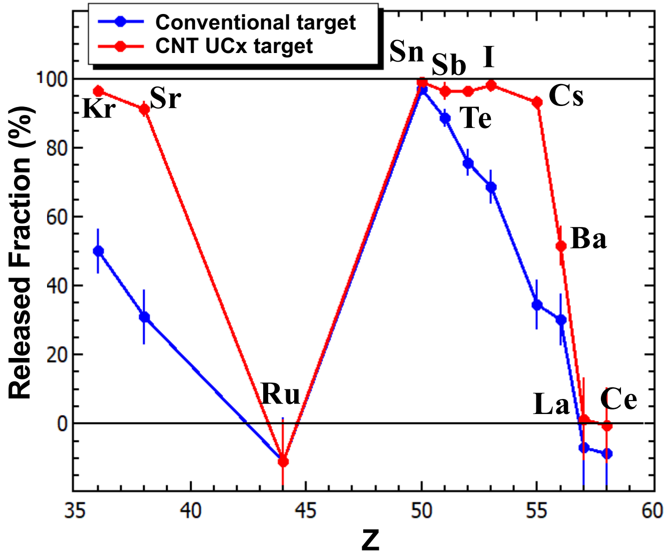




J. Guillot *et al.* NIMB 2018, 2019, 2020

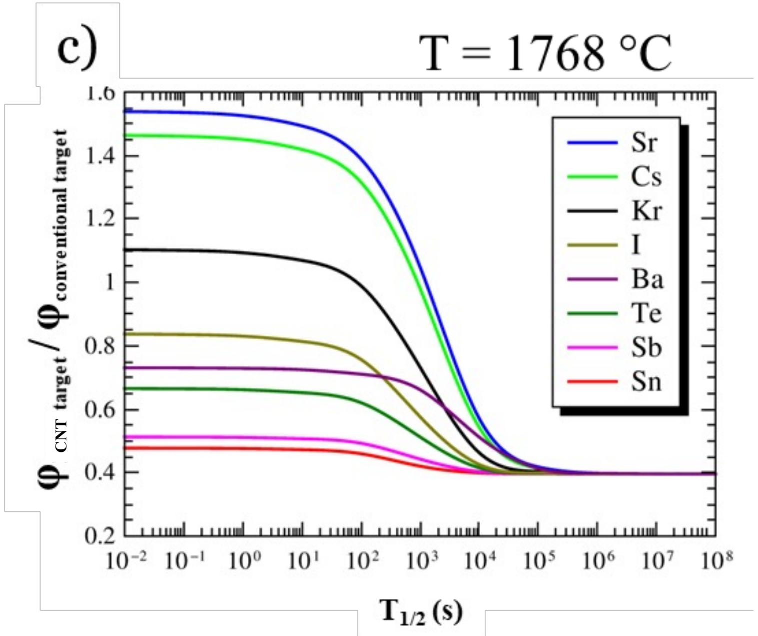
# Study on the influence of the microstructure




a) Correlations between release fractions (in green) and target properties (in blue)




b) Released fraction comparison between conventional target (mostly $UC_2$) and an R&D target made with CNT

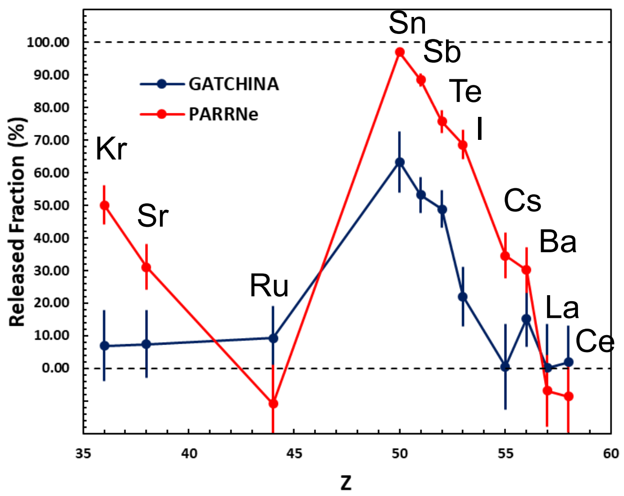



c) Comparison of fission products released between R&D target and conventional target

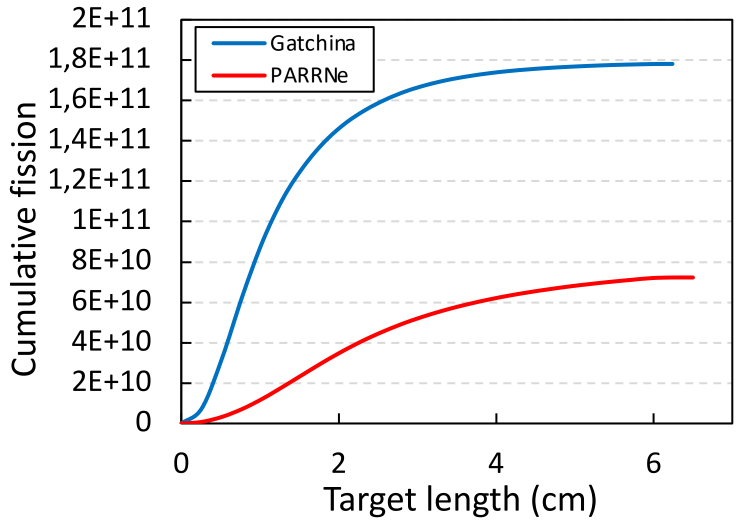
**Loss of production is compensated by improved release (Sr, Cs and Kr)**

J. Guillot *et al.* NIMB 2018, 2019, 2020

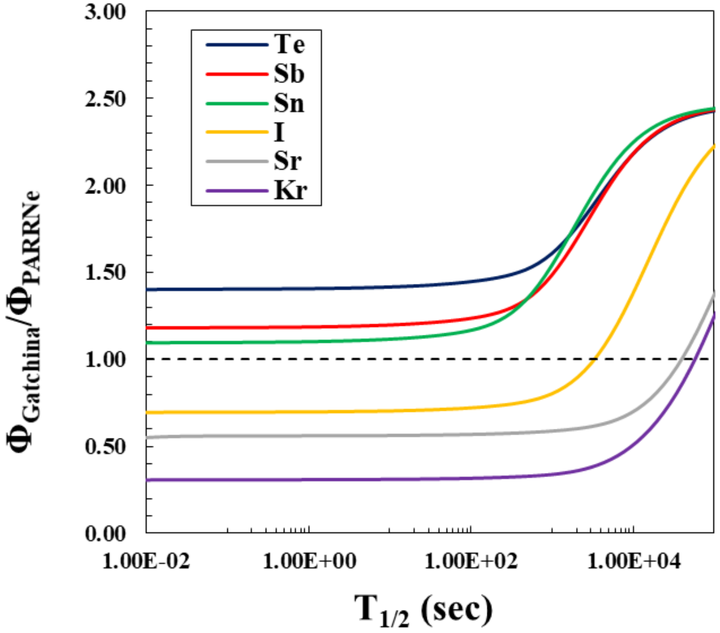
# Study on the influence of the microstructure




Released Fractions from:
PARRNe pellet (Ø=13 mm, th=1.7 mm) heated to 1768°C
Gatchina pellet (Ø=13.2 mm, th=1 mm) heated to 1700°C.




Number of fissions per second cumulated along the PARRNe and Gatchina targets and normalized for a 10 µA electron beam




Estimation of the production ratio between a Gatchina target and a PARRNe target

Physico-chemical characteristics obtained by XRD and Helium pycnometry

| | | Gatchina | PARRNe |
|---|---|---|---|
| Quantity of phases (%) | $UO_2$ | 4.5 | - |
| | UC | 86.9 | 5 |
| | $UC_2$ | 8.6 | 87 |
| | C | - | 8 |
| Apparent density ($g/cm^3$) | | 12.4 | 3.82 |
| Porosity (%) | Open | 5 | 51 |
| | Close | 2 | 5 |

The loss in releases is compensated by improved production (Sn, Te and Sb)

**S. Tusseau-Nenez *et al.*** Nuclear Instruments and Methods in Physics Research B 370 (2016) 19–31

# STUC PROJET: STudy of Uranium Compounds

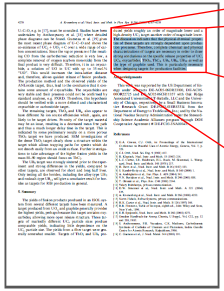
| Elements | Release process | Target caracteristics |
|---|---|---|
| Sr | - | Low density<br>Small grain<br>Open pores |
| Cs | Diffusion[1] | |
| Kr | Diffusion[2] | |
| I | Diffusion/Effusion[2] | |
| Te | - | High density<br>Open pore (?) |
| Sb | - | |
| Sn | Effusion[2] | |

**1: F. Hosni** *et al.* NIM B 247 (2006) 205–209

**2: B. Roussière** ***et al.*** NIM B 246 (2006) 288–296

Table 1
List of the targets

| Target | $UC_2$:C | $UC_2$ particle size (μm) | Density (g/cm³) | Thickness (cm) | Production method |
|---|---|---|---|---|---|
| ANL-oxide | 1:2 | – | 2.61 | 0.15 | $UO_2 + C$ |
| ANL 200 | 1:8 | <250 | 5.65 | 0.076 | CERAC $UC_x$ |
| ANL 325 | 1:3 | <43 | 5.24 | 0.072 | $U_{met} + C$ |
| ANL 400 | 1:3 | <37 | 5.49 | 0.077 | $U_{met} + C$ |
| $ThO_2$ | – | – | ~7 | – | Commercial |
| $UB_4$ | – | – | 2.1 | – | $UCl_4 + MgB_2$ |
| Refrac | 1:0.2 | – | 10.97 | 0.1138 | $U_{met}$ + C via $UH_3$ |



duced yields roughly an order of magnitude lower and a high-density $UC_2$ target another order of magnitude lower. The discussion indicates that the physical-chemical properties of these targets are strongly dependent upon production processes. Therefore, complete chemical and physical characterizations of targets are necessary in order to draw strong conclusions on the specific release properties of UC, $UC_2$, oxycarbides, $ThO_2$, $ThC_2$, $UB_2$, $UB_4$, $UB_{12}$ as well as the type of graphite used. This is particularly necessary when designing targets for production facilities.

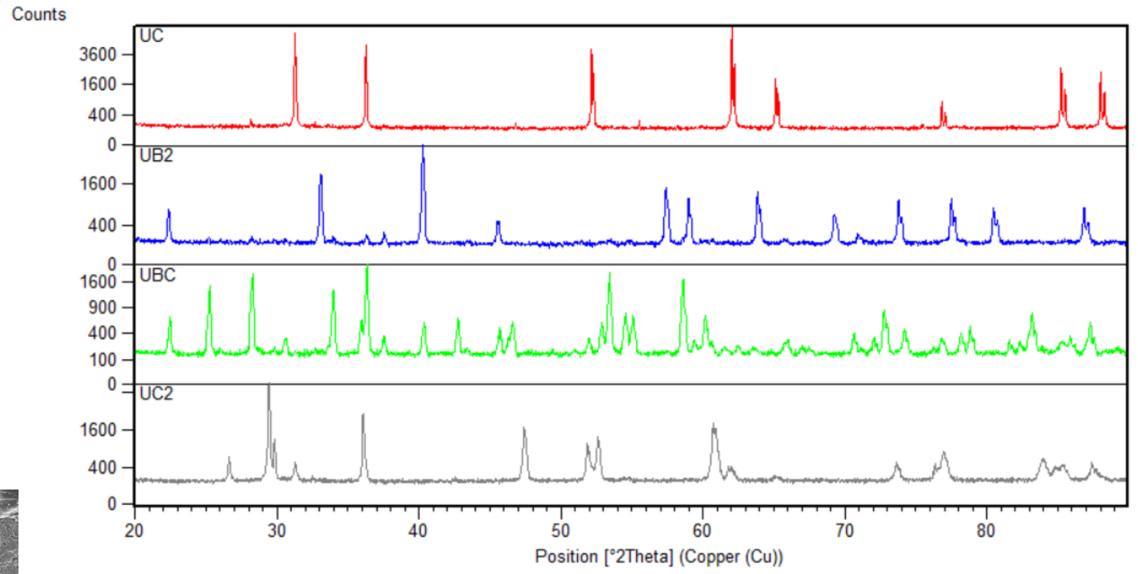
**A. Kronenberg** ***et al.*** Nucl. Instr. and Meth. in Phys. Res. B 266 (2008) 4267–4270

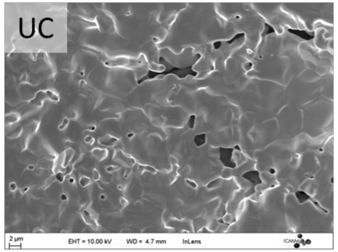
**Synthesis of various uranium compounds (UC, UBC, $UB_2$ and $UC_2$) :**
(**Aim:** To study the influence of uranium alloy density on the release of fission products.)

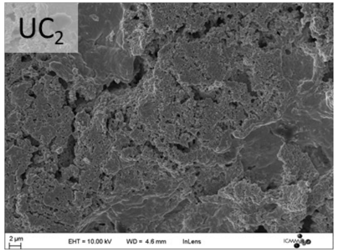
- If we add porosity to a very dense target, does this improve release ?
- Is there a difference in release if we use uranium compounds with theoretical densities for UC, UBC, $UB_2$ and $UC_2$?
- Is there any influence of the chemical environment ?

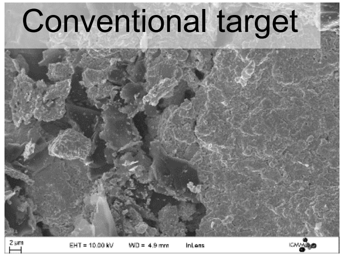
# STUC PROJET: STudy of Uranium Compounds

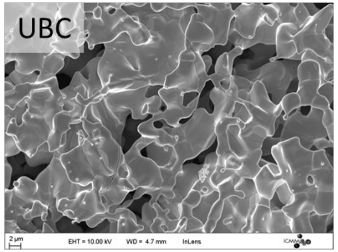
- $UO_{2\,(s)} + 4C_{(s)} \longrightarrow UC_{2\,(s)} + 2CO_{(g)}$
- $UO_{2\,(s)} + 3C_{(s)} \longrightarrow UC_{(s)} + 2CO_{(g)}$
- $UO_{2\,(s)} + 3C_{(s)} \longrightarrow UC_{(s)} + 2CO_{(g)}$
  $UC_{(s)} + BN_{(s)} \longrightarrow UBC_{(s)} + N_{(g)}$
- $2UO_{2\,(s)} + 3C + B_4C_{(s)} \longrightarrow 2UB_{2\,(s)} + 4CO_{(g)}$

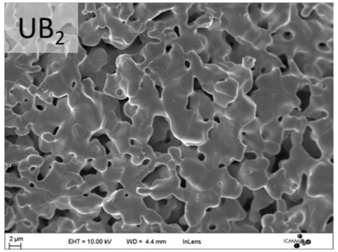


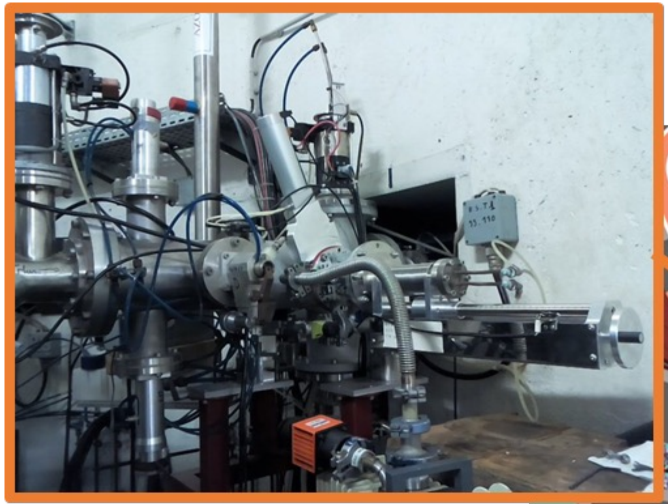



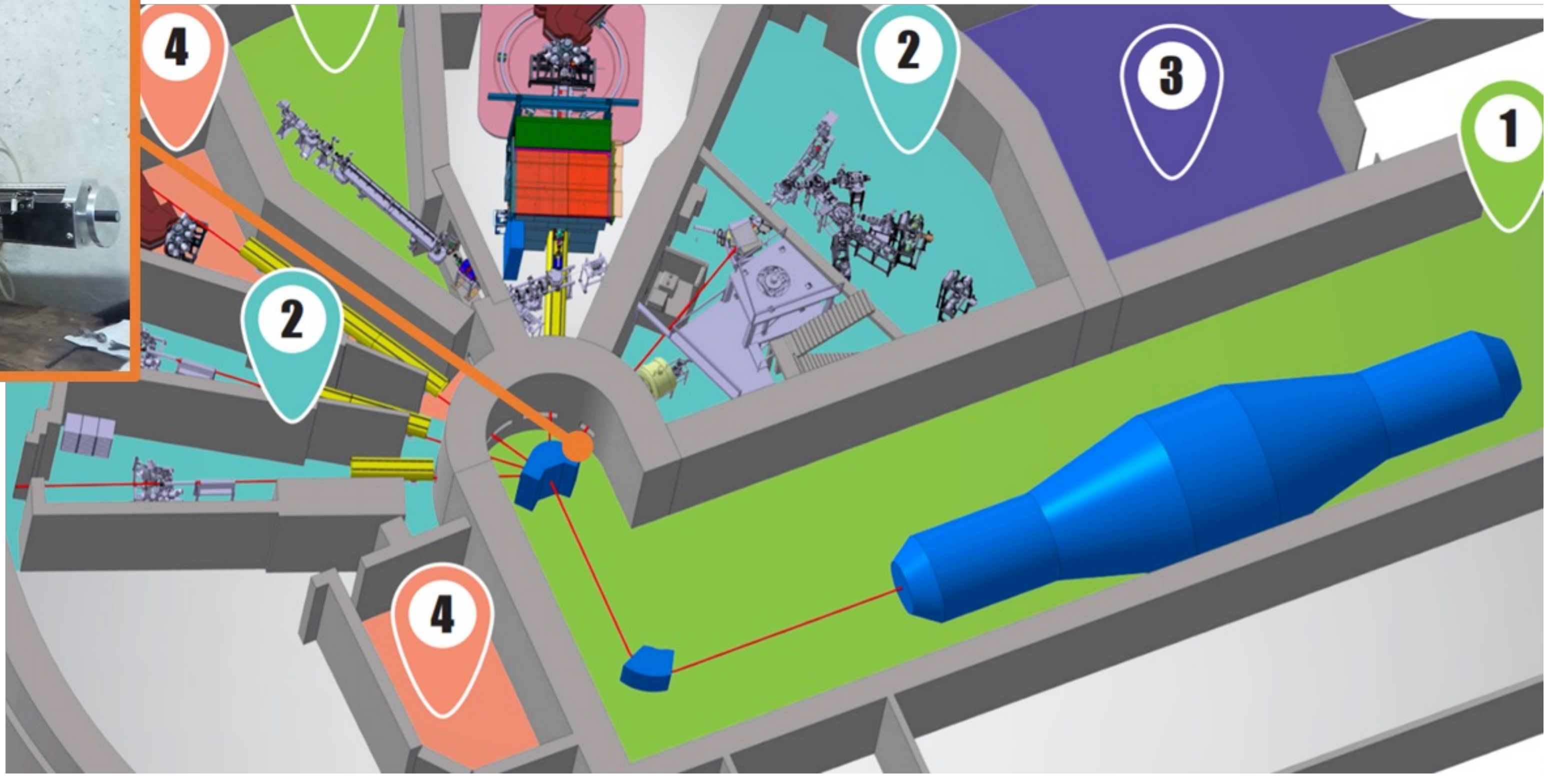



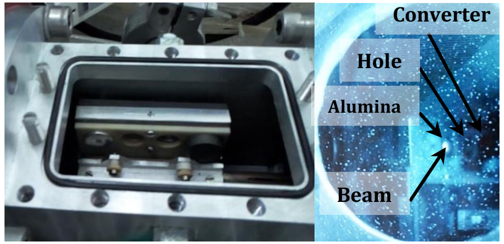

| Target | Apparent density | Porosity (%) | | SSA | Open pore size distribution (%) | |
|---|---|---|---|---|---|---|
| | (g/cm$^3$) | open | close | (m$^2$/g) | <10 µm | >150 µm |
| Conventional target ($UC_2$ + 2C) | 4.5 | 44 | 1 | 0.4490 | 75 | 25 |
| UC | 8.16 | 39 | 1 | 0.0763 | 94 | 6 |
| $UB_2$ | 5.78 | 53 | 2 | 0.1032 | 80 | 20 |
| UBC | 6.93 | 42 | 1 | 0.0496 | 100 | 0 |
| $UC_2$ | 5.86 | 46 | 3 | 0.3965 | 88 | 12 |




4
2
3
1
2
4

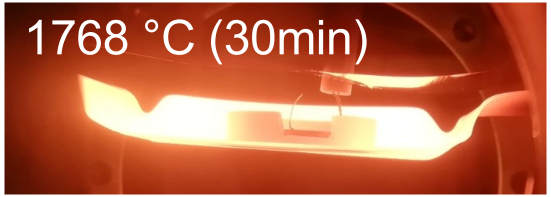
# STUC PROJET: STudy of Uranium Compounds

Irradiation:

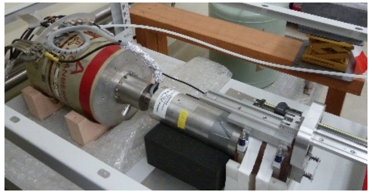



1st measurement :

Heating:



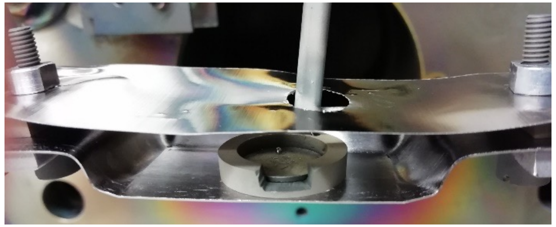

2nd measurement :

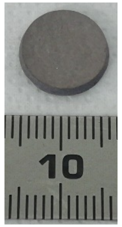


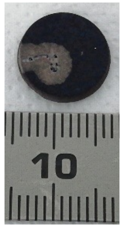
Irradiation conditions :

- Beam $^2H$
- Energy 26 MeV
- Intensity 20 nA
- Time of irradiation 20 min

$$R = \frac{I_{P1}}{I_{P2}}$$





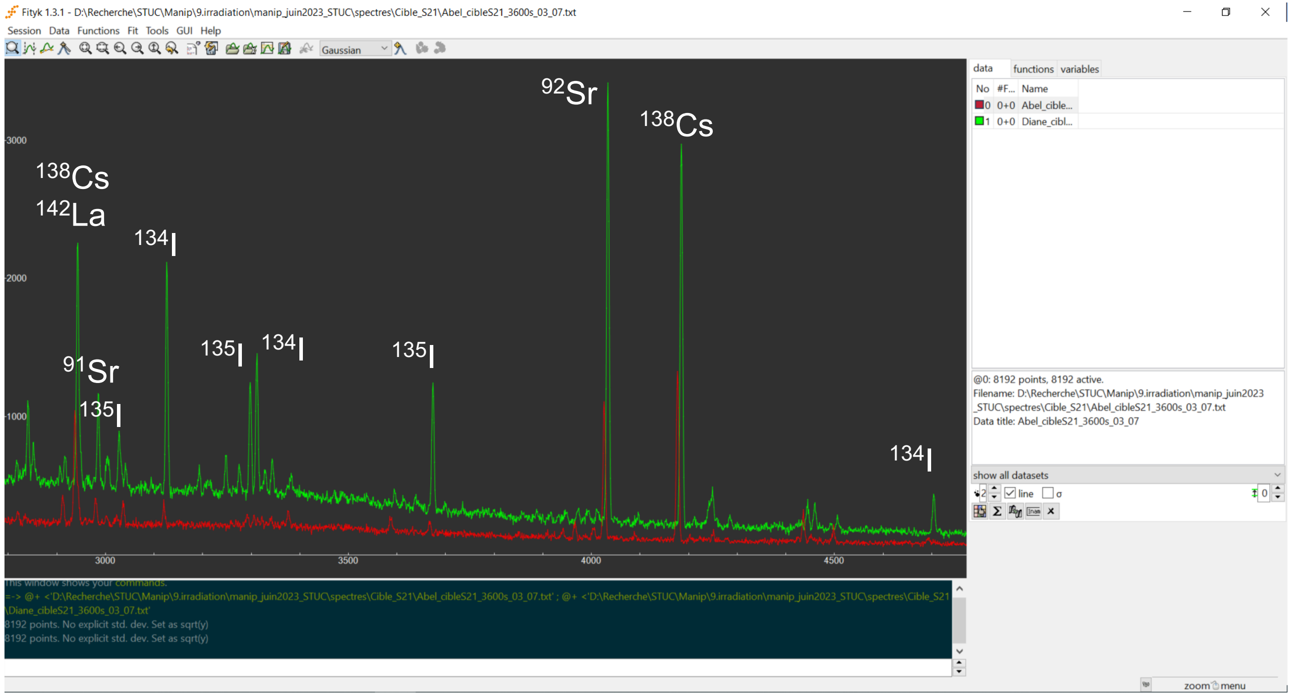



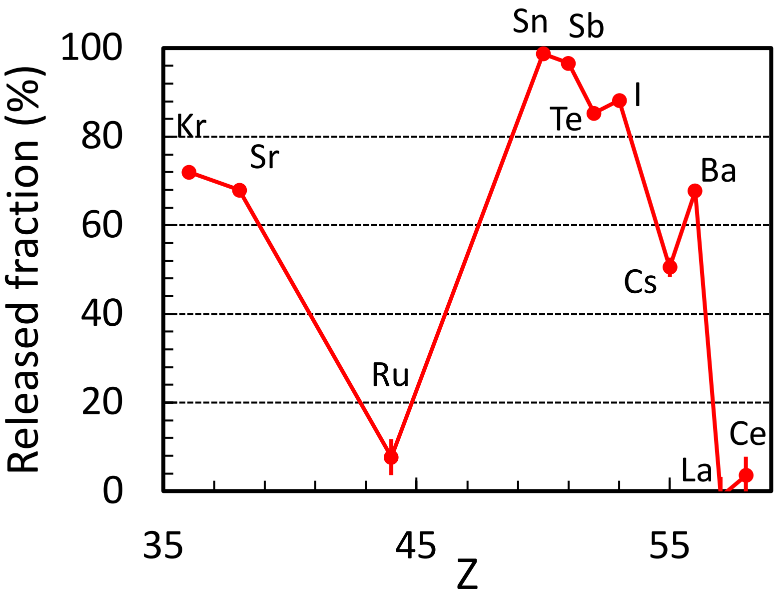

Reaction between $UB_2$ and Pt
→ thermocouple

$$RF = 100\left(1 - \frac{I_{heated}}{I_{unheated}}\right) \quad with \quad I_{unheated} = I'_{P2} \times R \quad and \quad I_{heated} = I'_{P1}$$

# STUC PROJET: STudy of Uranium Compounds

Experiment performed in July 2023, analysis in progress !




Validated reproducibility conditions

Conclusion: there are no universal targets, but one target for each element.

The aim of this R&D project is to answer the following questions:

- If we add porosity to a very dense target, does this improve release ?
- Is there a difference in release if we use uranium compounds with theoretical densities for UC, UBC, $UB_2$ and $UC_2$?
- Is there any influence of the chemical environment ?

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# Thank you for your attention