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# TISD activities during LS1

Update from last report

22 Oct 2013

T. Stora

# Reminder

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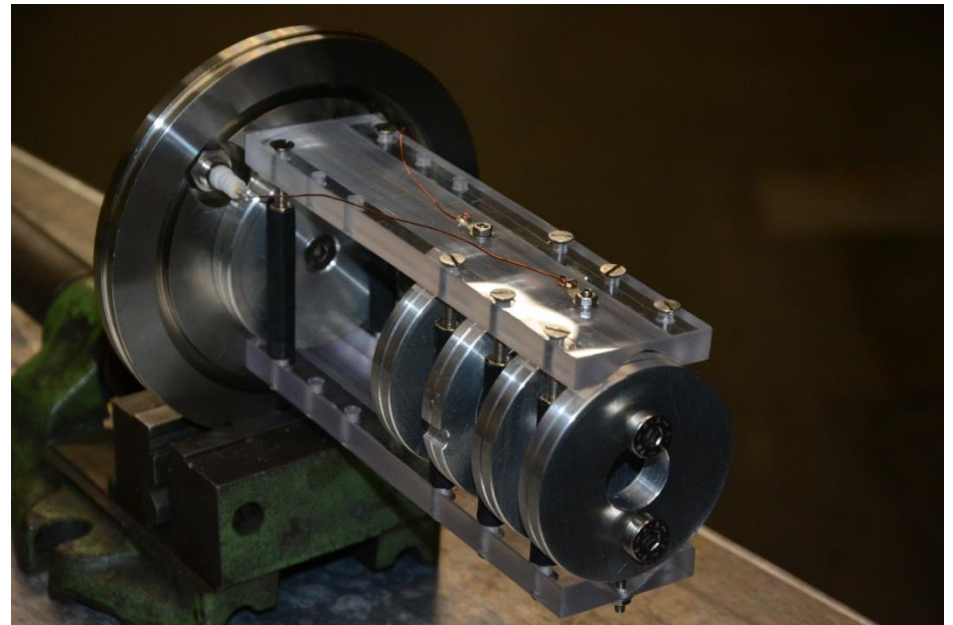
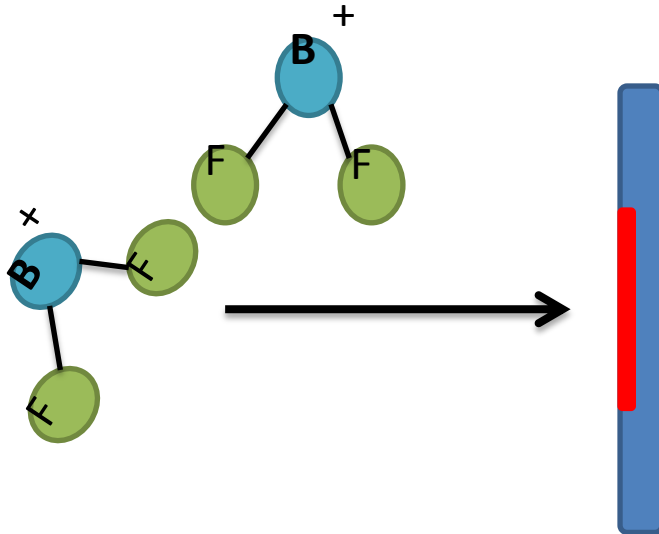
- 8B
- 9C
- 37K
- LIEBE (LBE loop)
- ActILab ENSAR (nanoUCx)
- Ba beams
- Neutron converter
- (RILIS in VADIS) molecular beams

## ISOLDE

- Diffusion studies of boron in possible target materials
- Implantation of  $^{10}\text{B}$  as  $^{10}\text{BF}_2$  into pressed pills of  $\text{Al}_2\text{O}_3$ , CNT,  $\text{MgF}_{2[1]}$ ;
- HV=70kV-80kV

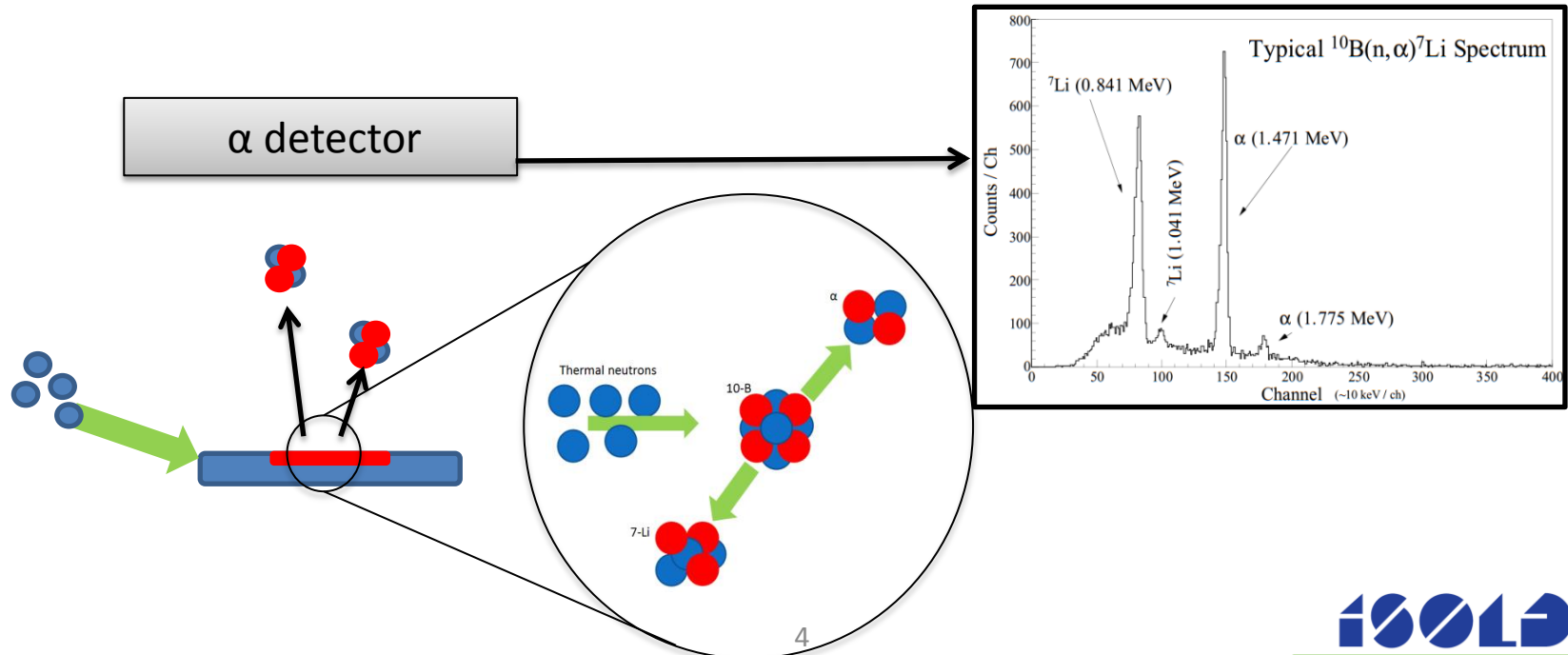
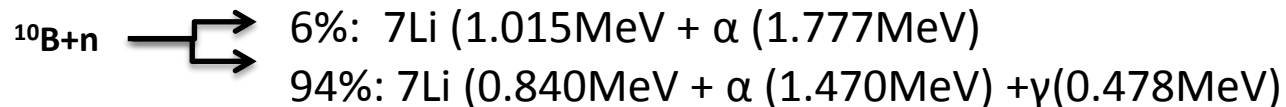
## SARAF

- Characterisation of neutron beam
- Implantation of  $^{10}\text{B}$  as  $^{10}\text{BF}_2$  into Al ( $200\mu\text{g}/\text{cm}^2$ ) foil,  $r_{\text{coll}}=1.5\text{mm}$ ,
- HV=32kV
- $n_{\text{BF}_2^+}=4*10^{16}$

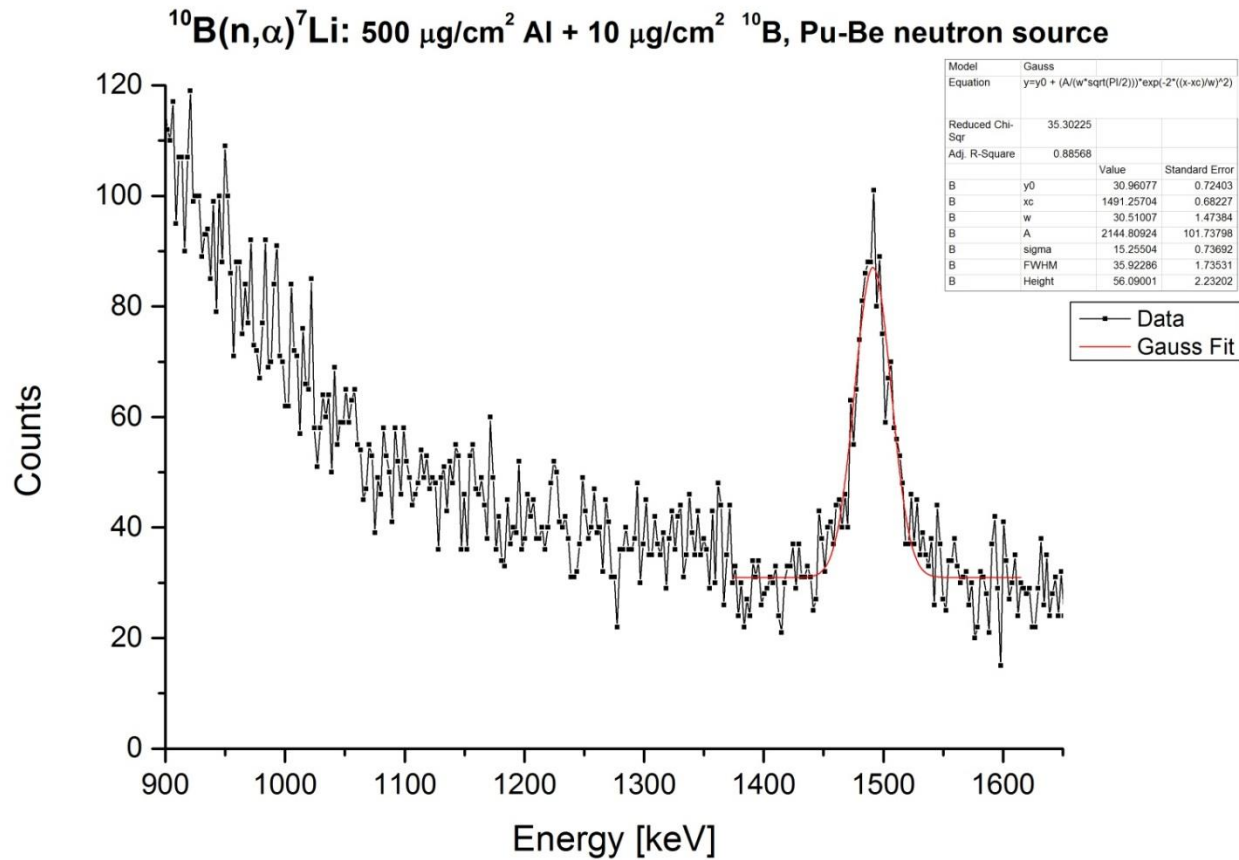


## ● Principle

- Irradiation of sample with thermal neutrons:
- For diffusion measurements: Pu-Be Source  $1.1 \cdot 10^8$  n/s @  $4\pi$ , (before moderation)
- Neutron capture cross section of  $^{10}\text{B}$ :  $\sigma_{\text{th}} = 3840$  barn
- Detection of produced  $^7\text{Li}$  and  $\alpha$  from  $^{10}\text{B}(n,\alpha)^7\text{Li}$



- Measurement with calibration sample, done with Pu-Be source

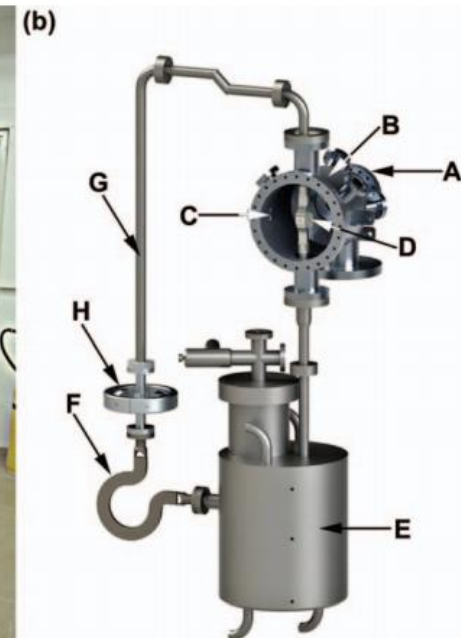
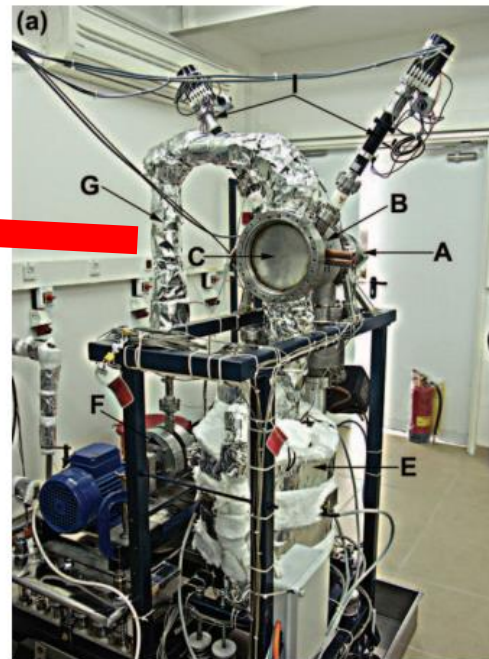


## ● SARAF@Soreq

- Located near Yavne, Israel
- Proton beam ( $E=1.91\text{MeV}$ ) from Soreq Applied Research Accelerator Facility (SARAF) hits liquid lithium target (LiLiT),  $T\sim 200^\circ\text{C}$
- Production of 10-100keV neutrons:  ${}^7\text{Li}(p,n){}^7\text{Be}$
- Irradiation of  ${}^{10}\text{B}$  sample with fast neutrons
- Coincidence detection of produced  ${}^7\text{Li}$  and  $\alpha$  from  ${}^{10}\text{B}(n,\alpha){}^7\text{Li}$

Rev. Sci. Instrum. **84**, 123507 (2013)

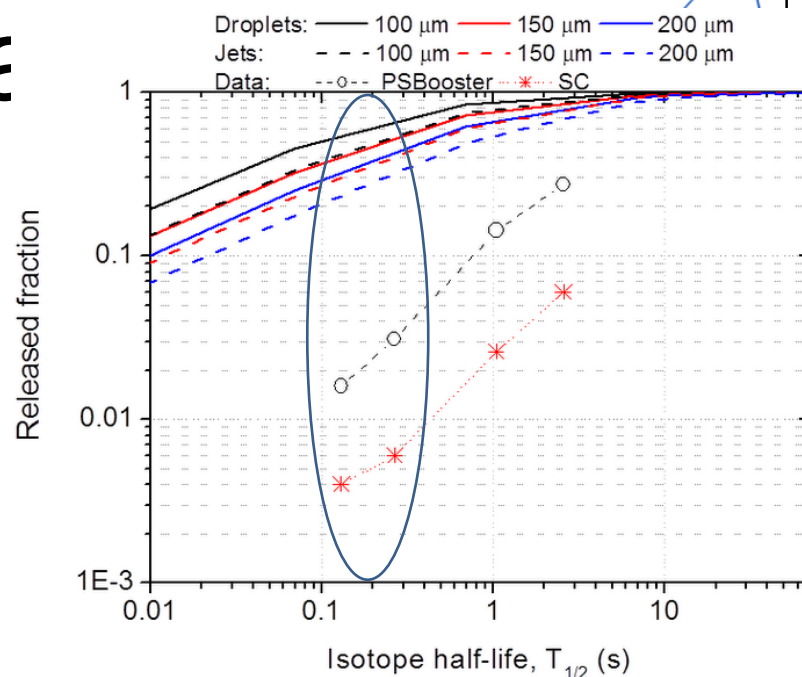
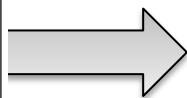
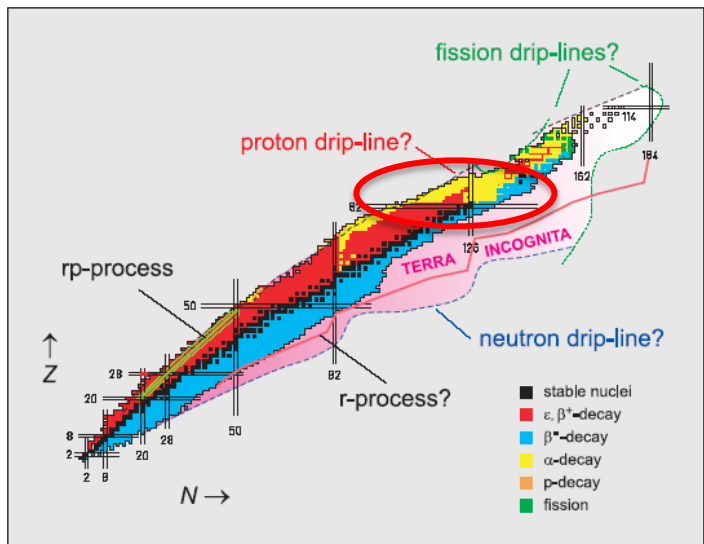
Rev. Sci. Instrum. **84**, 123507 (2013)



Validation of the conceptual design of EURISOL direct target by prototyping at Cern-ISOLDE

## Back to edit $M\alpha$

Motivation: Improvement of yields of short-lived Hg isotopes

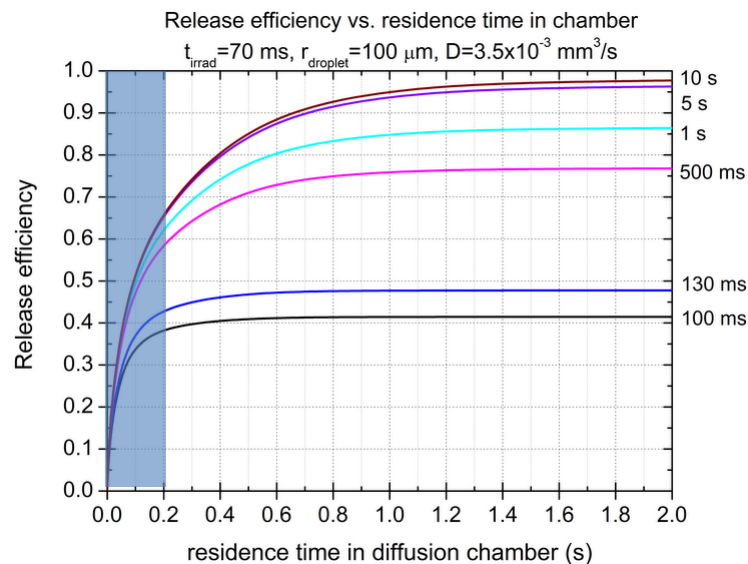


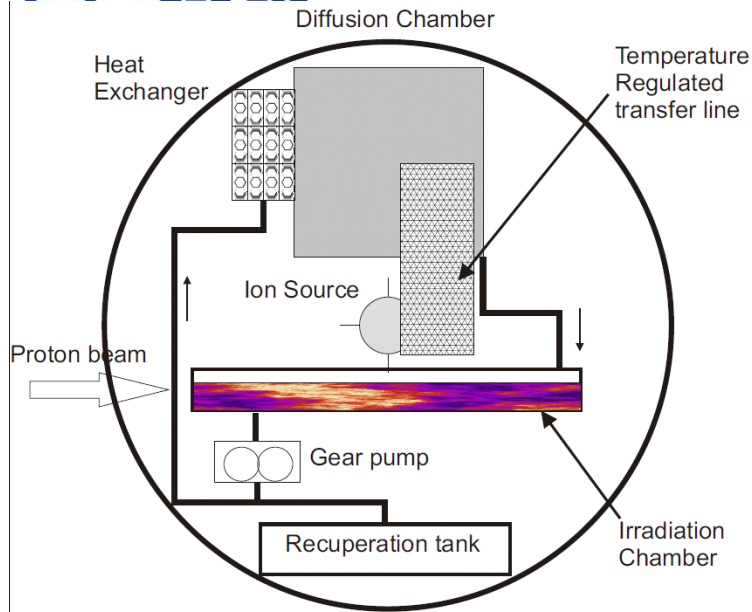
**Diffusion:** model from Fujioka et al. (NIM 186 (1981) 409)  
Data: Lettry et al. (NIM B 126 (1997) 170)

Total time till the post acceleration: < 300 ms.



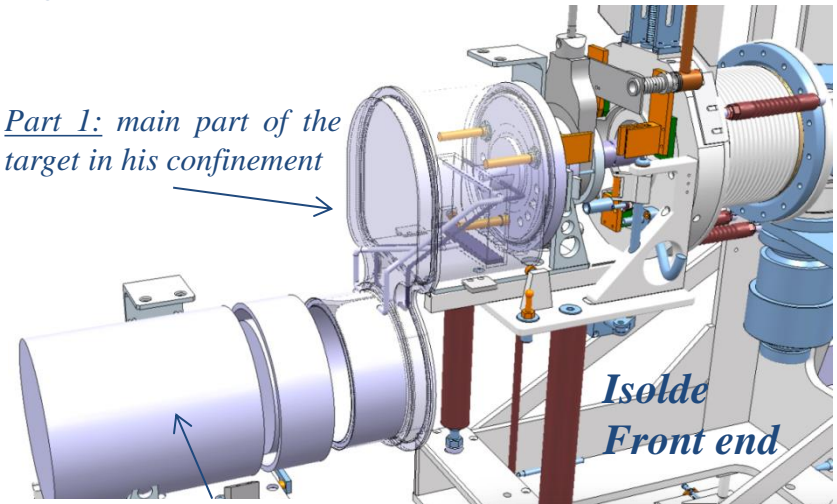
Diffusion of isotope when the LBE is on droplets shape which require the use of a grid, a diffusion chamber and a pump to recirculate the liquid.





**Development:** integration of a liquid loop target, working at high temperature (from 200 deg C up to 600 deg C) in the Cern-Isolde environment.

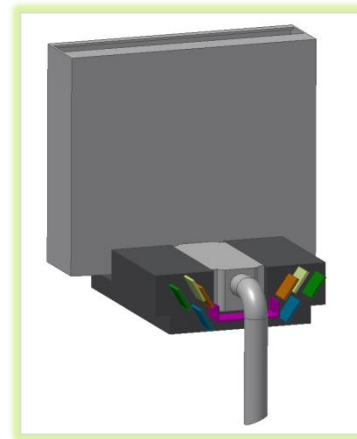
*Part 1: main part of the target in his confinement*



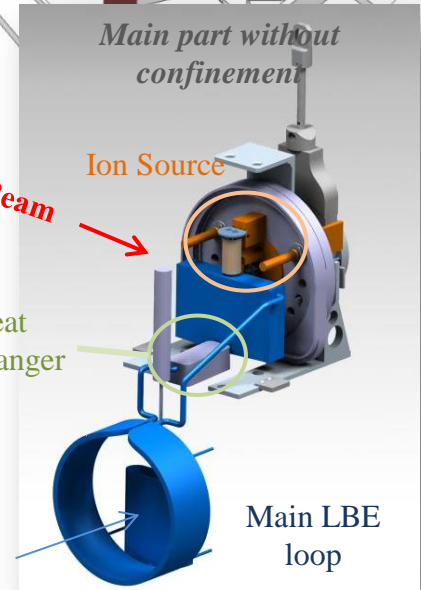
*Part 2: pump and engine to recirculate the liquid*

**Main challenges:**

- High power deposited on target to be evacuated (HEX to dimension at various T)
- Recirculation of liquid through a compatible LBE pump
- High temperature of use (up to 600 deg C)
- Integration in already existing (relatively small) environment
- Monitoring and control

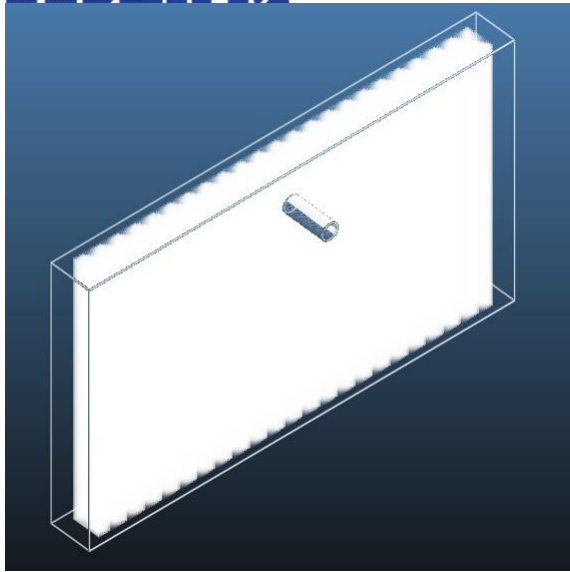


Feeder tank

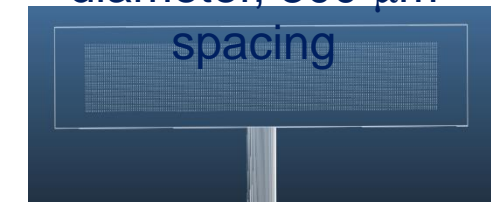




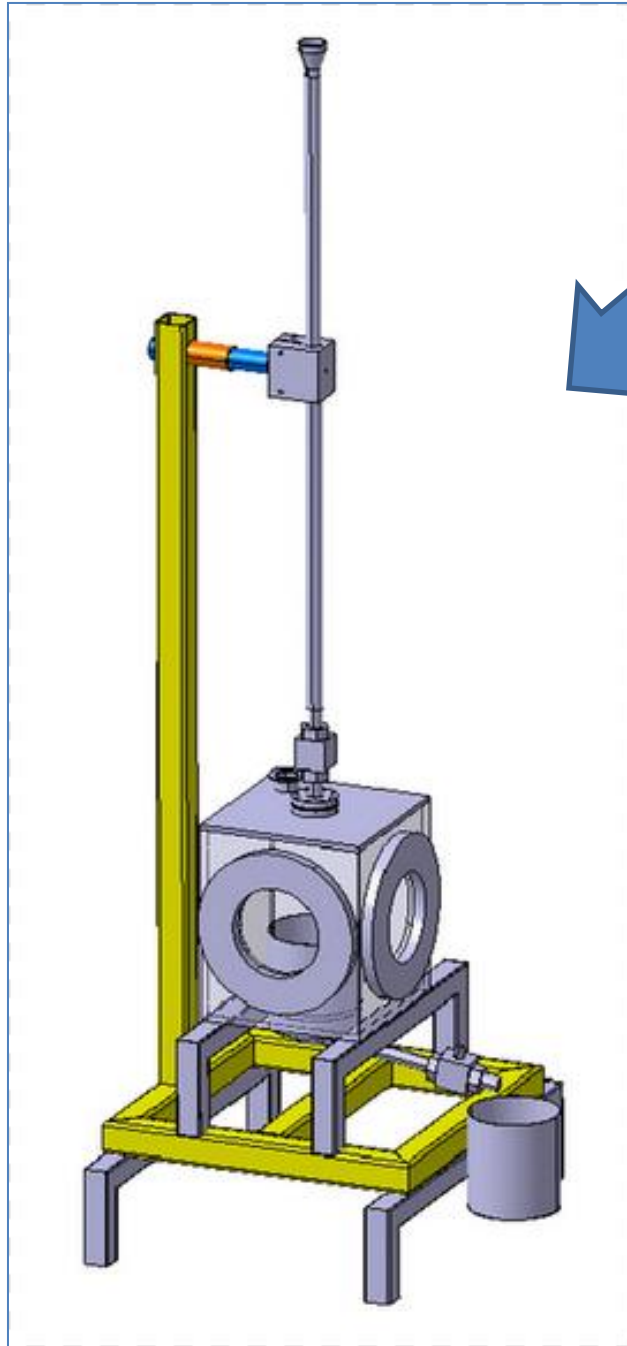
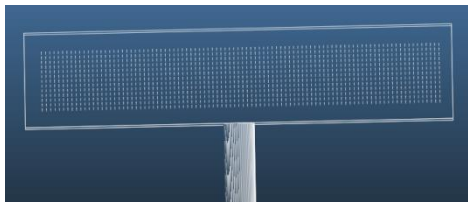
Model: Shower inside diffusion chamber



Grid: 200 $\mu$ m pore diameter, 300  $\mu$ m spacing



Grid: 200 $\mu$ m pore diameter, 1 mm spacing



Test stand to investigate shower formation in vacuum

Tests ongoing

LBE shower in air @ISOLDE-Jan 2014



# nanoTiC - motivation

High Melting point – 3160 °C

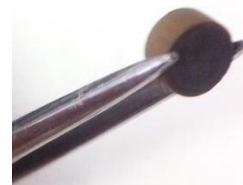
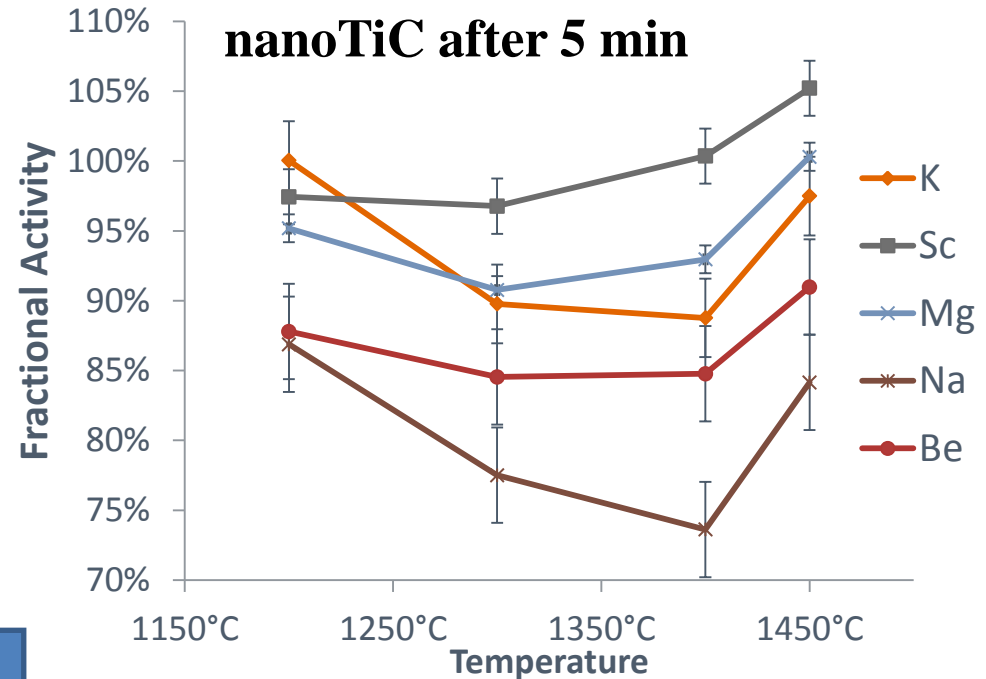
- Sintering studies done, shows that TiC can maintain a submicron structure at least until 1500°C
- Operation temperature has to be at least 1500°C (Ca, K, Sc beams)



- Avoid oxidation of TiC
- Improve nanostructure stability => Essential for good release
- TRIUMF shows release of Ca and K from TiC



## Release studies

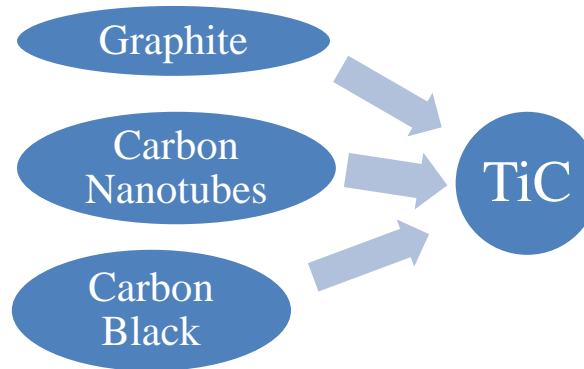


Fractional activity rise at 1450°C is attributed to an oxide layer in the samples at higher sintering temperatures.

# nanoTiC – ongoing studies

How to hinder sintering of TiC?

- Doping
- **Add (a higher melting point) second phase**
- ~~Lower temperature and time~~



- Study the effect of the different carbon polymorphs
- Study the volume ratio of carbon



- Isopropanol
- Polymeric dispersant



- Attrition mill
- Zirconia milling elements



- Drying
- Deagglomerate
- Pressing

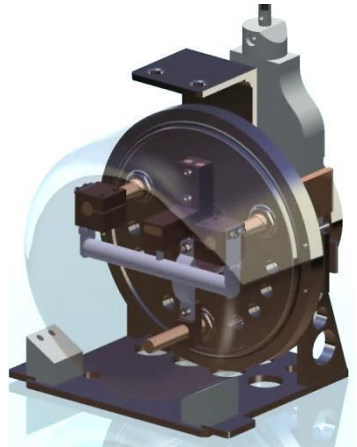
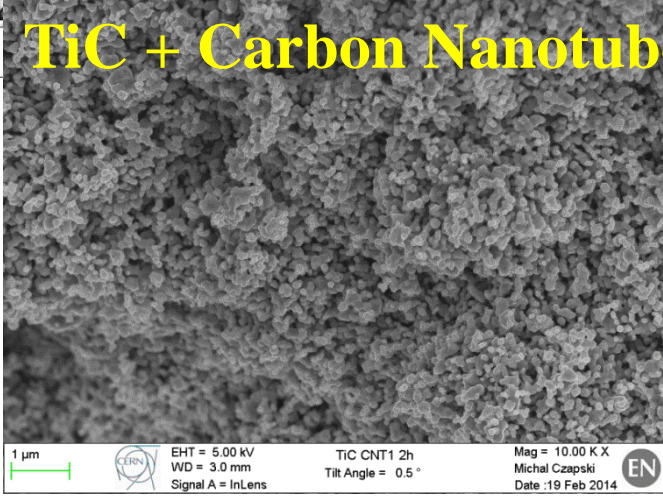
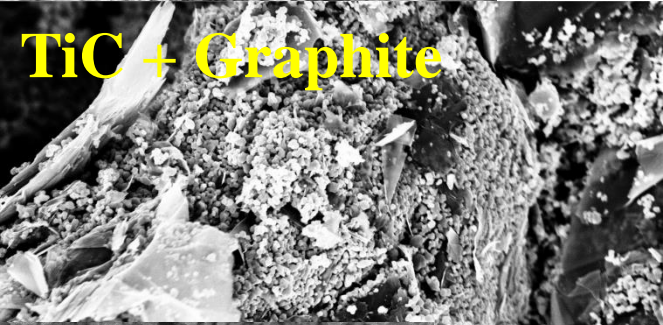
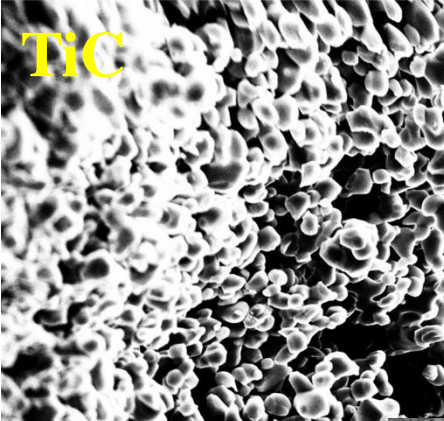
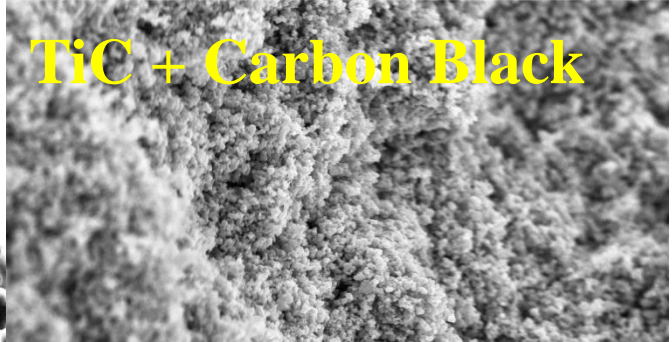


- 1500°C for 2 and 10h
- Vacuum



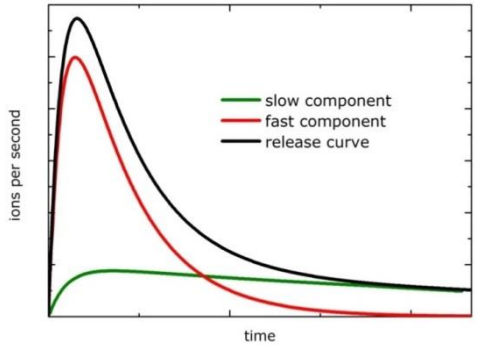
- Morphology
- Density
- Porosity
- Specific surface area

# nanoTiC – results and future studies



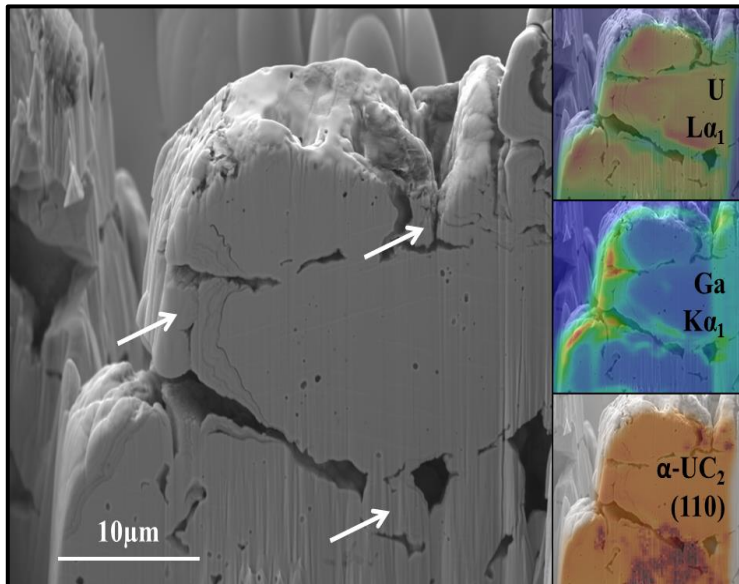
Future work:

- Release studies
- Target prototype
- Modeling

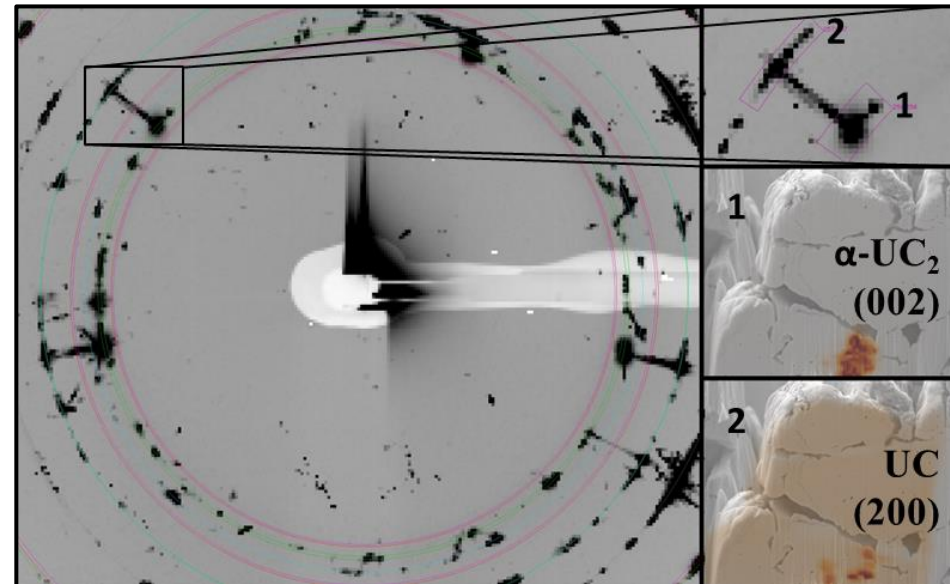


## Characterization of conventional UC<sub>x</sub> using synchrotron-based micro-beam analysis:

Microscopic morphology – buried porosity & chemistry



Kinetic stabilization by sub-microscopic UC – UC<sub>2</sub> phase competition



- Grain size of material is smaller than previously estimated; global phase transition observed at 2100°C
- Phase competition between UC and α-UC<sub>2</sub> as yet missing explanation of performance and durability of this material

A. Gottberg, et al., submitted

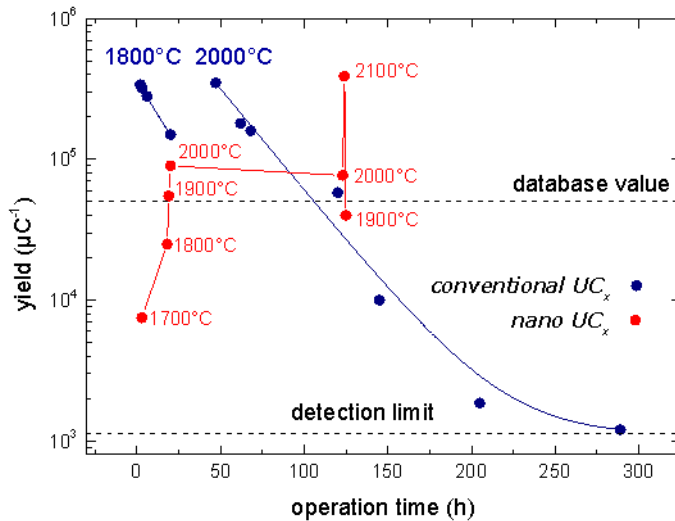
# ActILab-ENSAR

Online tests and synthesis of de-novo designed uranium carbide matrixes:

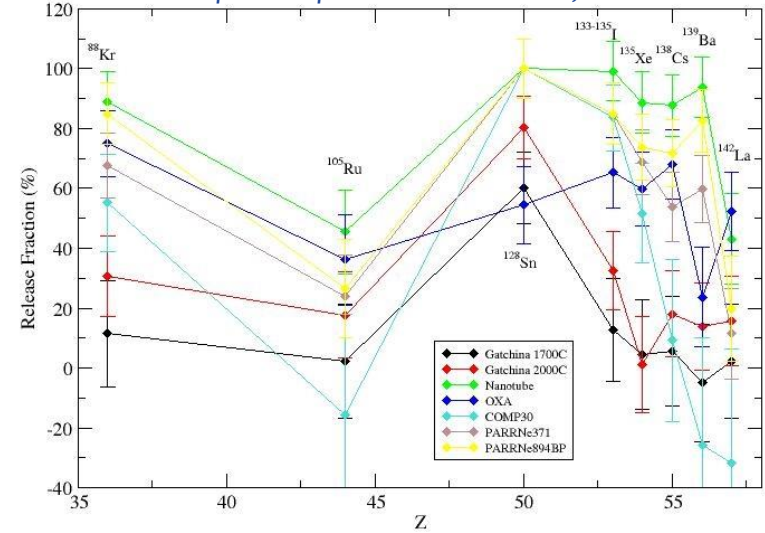
Different microstructures, densities, grain sizes, crystal structures tested → tailor-made matrix:

- Suspension grinding of  $\text{UO}_2$  powder to 160 nm average particle size
- Ultrasound-assisted mixing with multi-walled carbon nanotubes
- Pressing to 1.6  $\text{g/cm}^3$  pellets
- Optimized reactive sintering to mixed uranium carbide inside carbon nanotube matrix

$^{30}\text{Na}$  beam intensity evolution



Release fractions from RaBIT irradiations, ALTO 2013



Ch. Lau, et al., in preparation

ISOLDE online tests:

Despite major technical difficulties (RFQ, tape station, separator):

- record yield of  $^{11}\text{Be}$ :  $6.0 \times 10^7 / \mu\text{C}$ , database:  $7 \times 10^6 / \mu\text{C}$  (confirmed)
- Structure appears to be widely conserved over time and temperature

Repeat tests at ISOLDE and at ALTO within ActILab in 2014

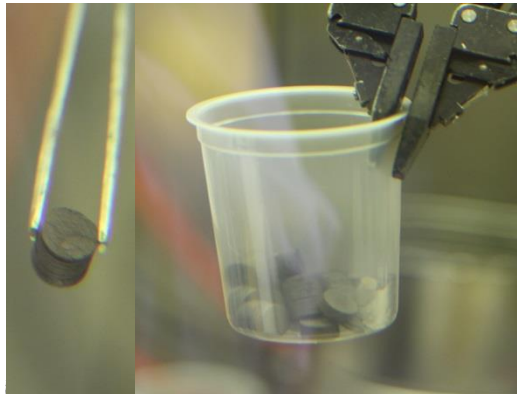
## Post-irradiation analysis:

- First analysis of an irradiated UC<sub>x</sub> target in the history of ISOLDE (important to understand ageing processes and for upcoming waste campaign)
- Opening target unit #466 ( $8.8 \cdot 10^{18}$  protons in 2011) in inert-gas hot-cell at PSI, 19 mSv/h on contact with Ta beam window

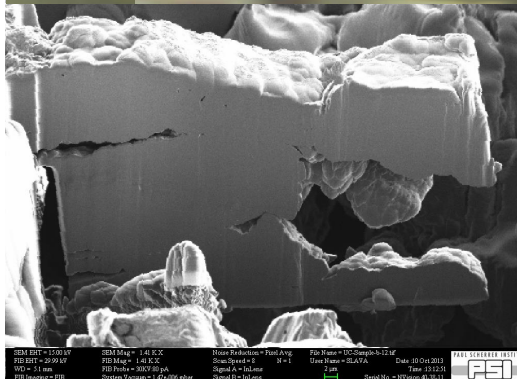
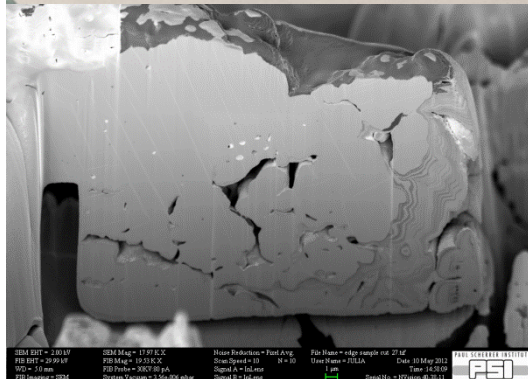
before irradiation



after irradiation



- Pellets appear macroscopically unchanged
- Microscopic evolution of pore distribution and grain size under irradiation observable
- 500  $\mu$ Sv/h on contact with single pellet



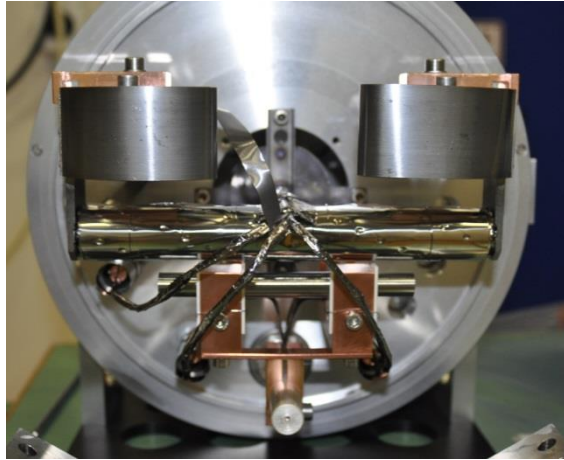
- Results of synchrotron investigations under analysis
- Ceramography and electron microprobe analysis scheduled for April
- Perform full suite of investigations on nano UC

SEM EHT = 15.00 kV SEM Mag = 17.071 X X Vision Database = Post-Act. File Name = edge\_rough\_001\_27.tif  
 FIB Mag = 29.94 kV FIB Mag = 19.53 X X Size Speed = 10 N = 10 User Name = JILIA Date = 10 May 2012 File Path = 00000000000000000000  
 FIB = 1.00 mm FIB Probe = 300 V pA Signal = InLens Scan Rate = 100 Lines/Sec  
 FIB Image = 2224

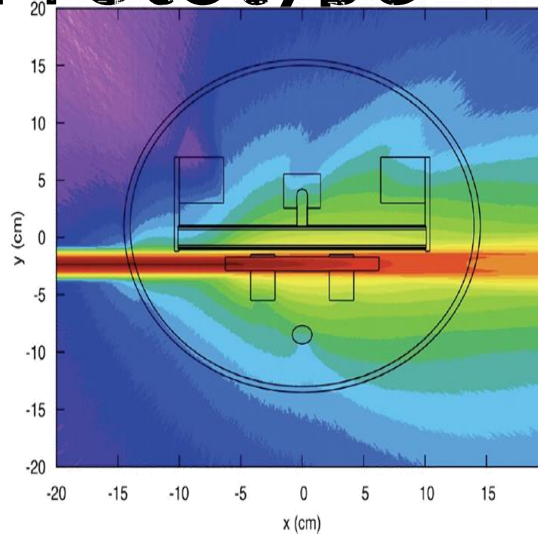
SEM EHT = 15.00 kV SEM Mag = 1.01 X X Vision Database = Post-Act. File Name = UC000000011.tif  
 FIB Mag = 29.94 kV FIB Mag = 1.41 X X Size Speed = 4 N = 1 User Name = BLAVA Date = 10 Oct 2012 File Path = 00000000000000000000  
 FIB = 1.00 mm FIB Probe = 300 V pA Signal = InLens Scan Rate = 1.41 Lines/Sec  
 FIB Image = 818

# Proton-to-Neutron Converter

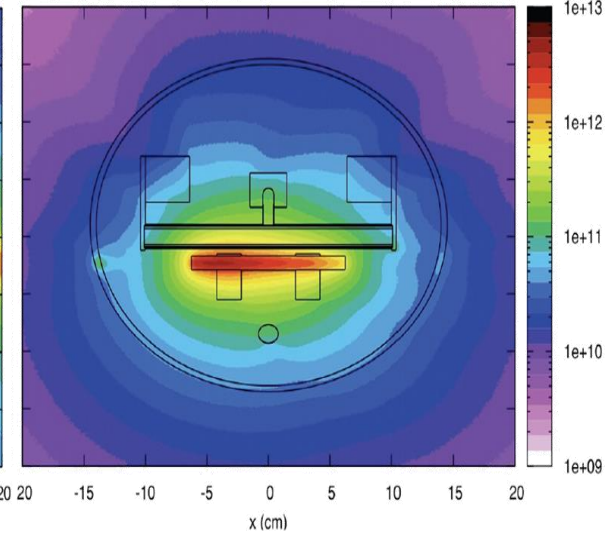
current design



proton fluence ( $p/cm^2/\mu C$ )



neutron fluence ( $n/cm^2/\mu C$ )

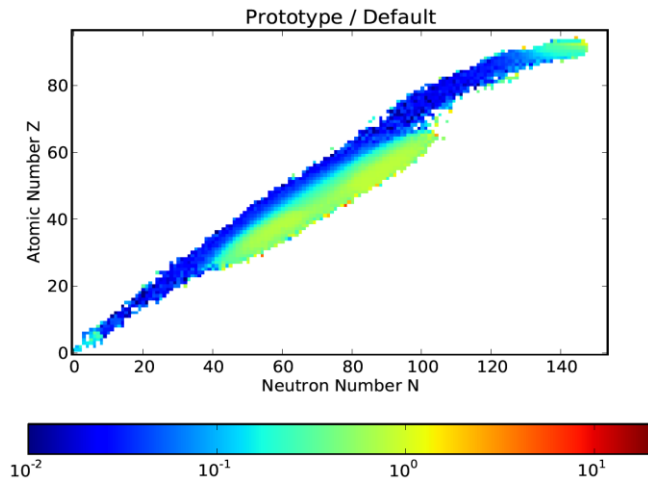


ISOLDE compatible  
geometry optimization



max high-energy neutrons  
min proton  
in  $UC_x$

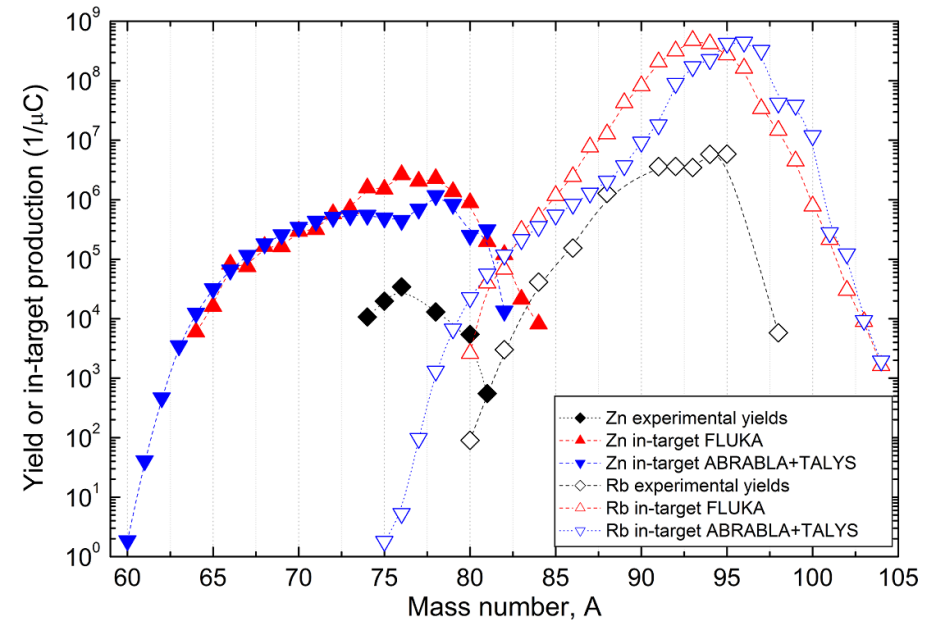
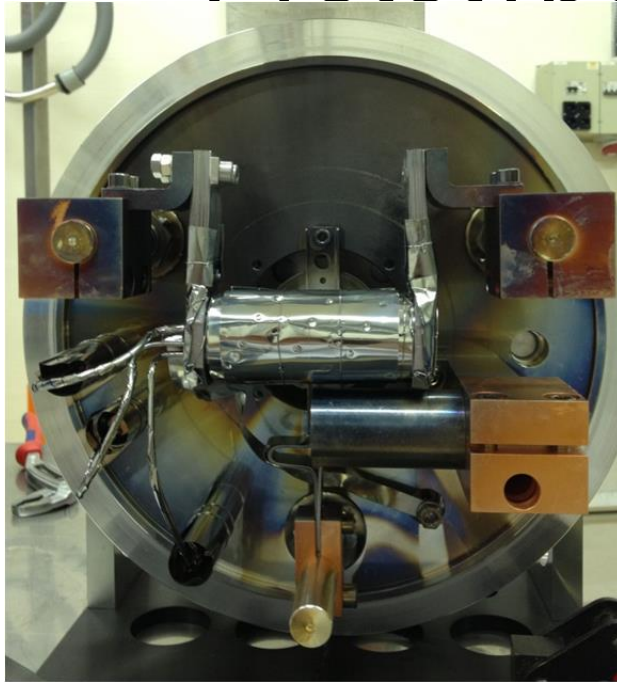
Operated in  
October 2012



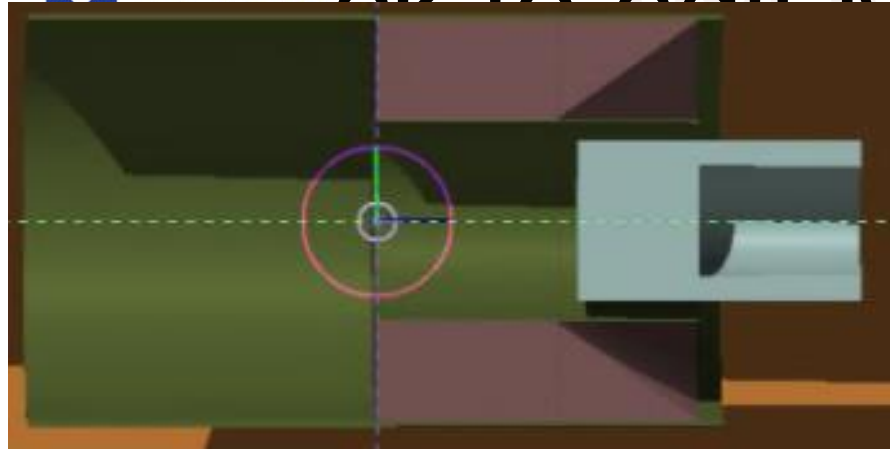
R. Luis et al., *European Physics Journal A*, 48 (2012) 90  
A. Gottberg et al. submitted to *Nucl. Instr. Meth. B* (2014)



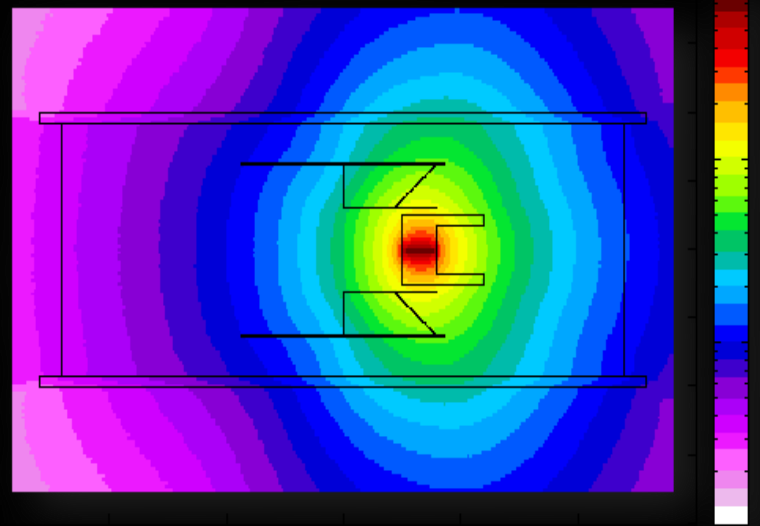
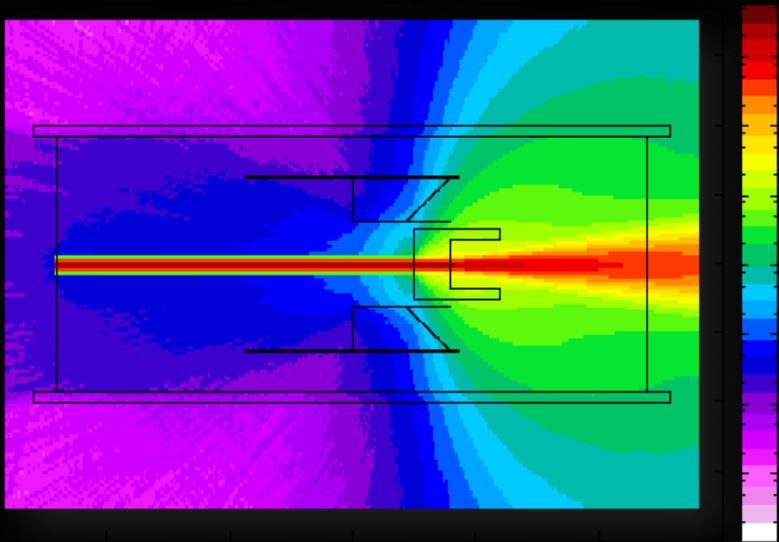
# Proton-to-Neutron Converter Prototype Online Tests



- Strong suppression of contaminations compared to conventional converter confirmed (10<sup>4</sup> for <sup>80</sup>Rb)
- Experimental ratio of n-rich species to contamination improved (200 for <sup>80</sup>Zn/<sup>80</sup>Rb)
- Poor extraction efficiencies due to low operational temperatures (caused by structural failure)

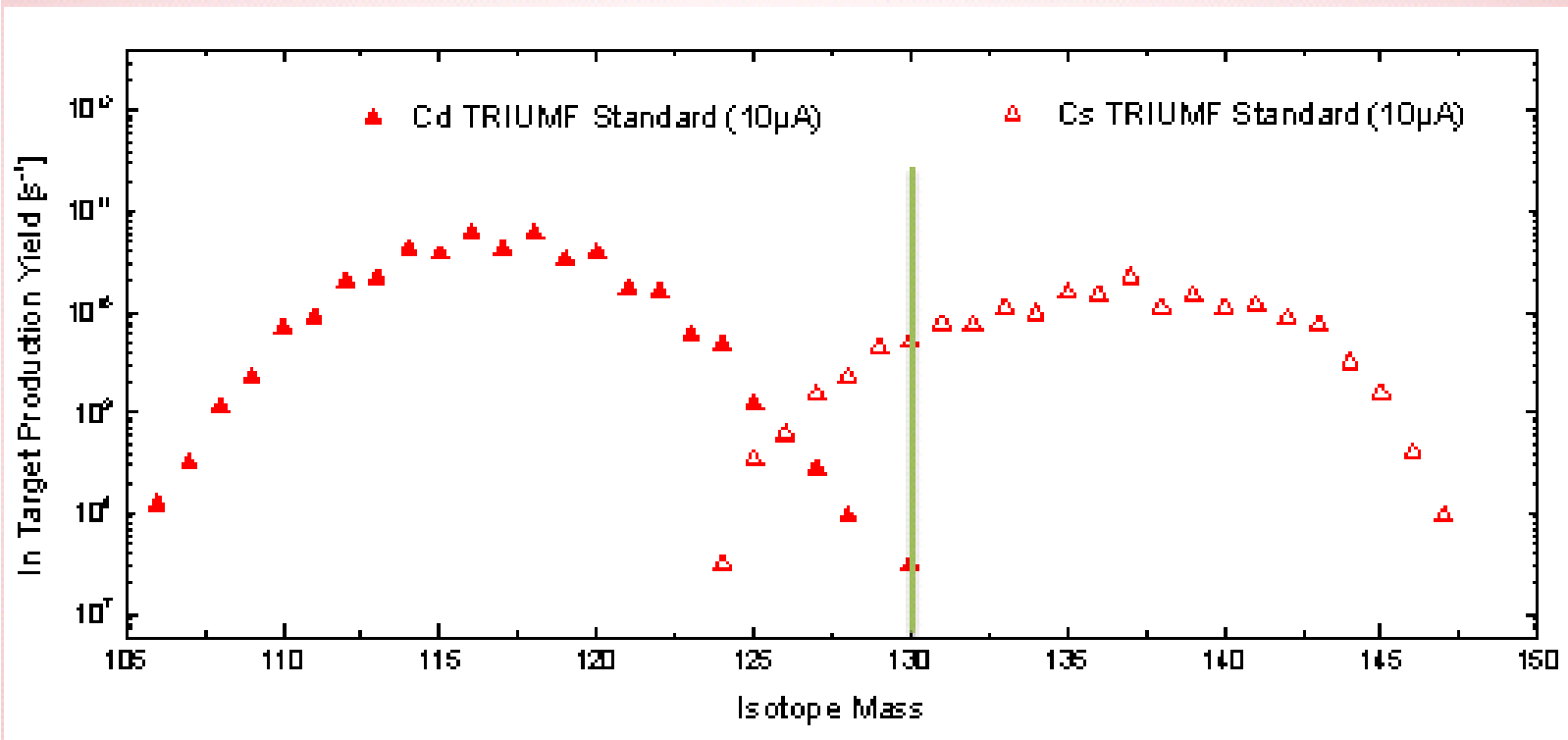


- Beam Energy: 500 MeV;
- Beam Intensity: 100μA;
- FWHM: 3mm;
- Particles per second: 6.242 E14;
- UCx density: 3.63g/cm<sup>3</sup>



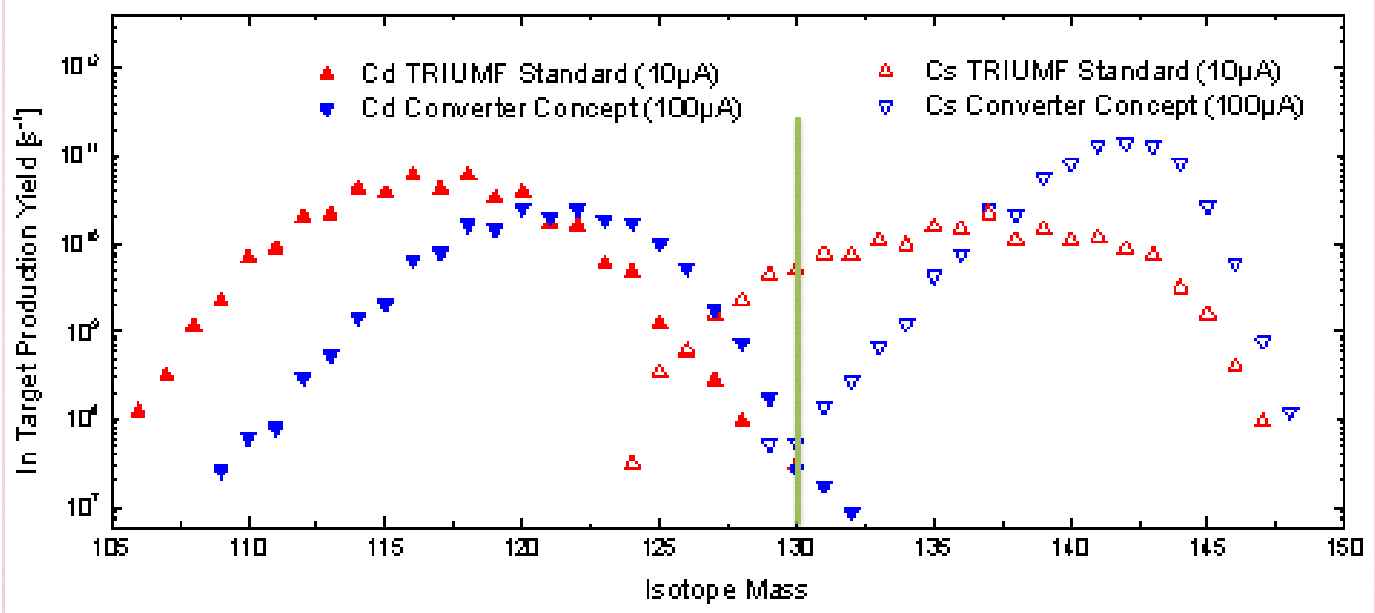
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# \* Production Comparison



# Click to edit Master title style

## \* Production Comparison



# Preliminary Thermal Analysis

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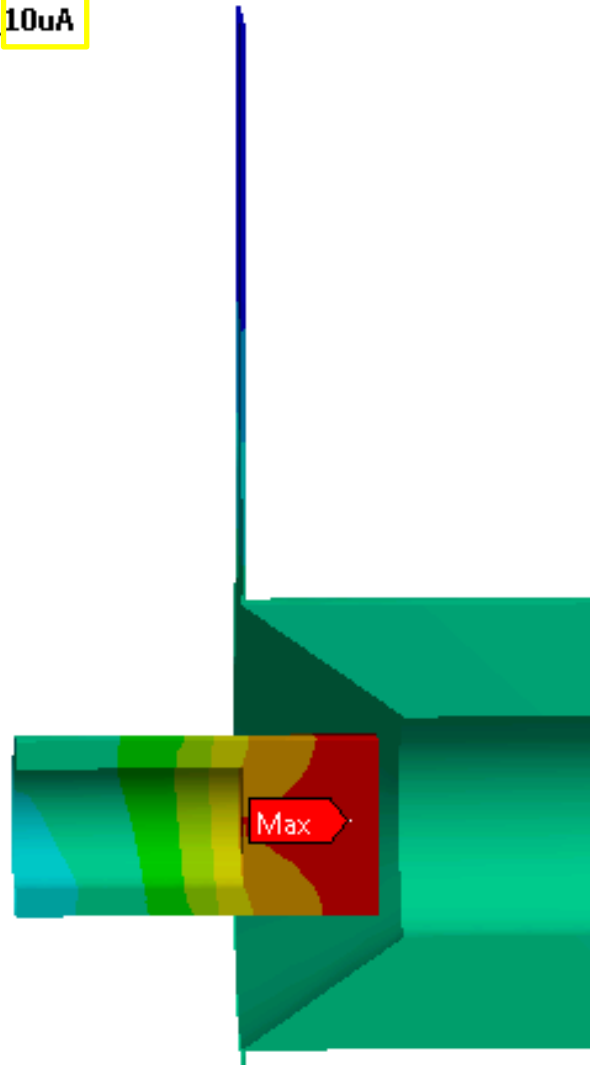
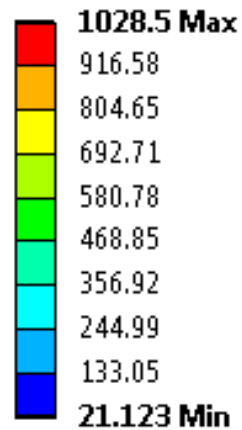
B: New enclosure\_10uA

Temperature

Type: Temperature

Unit: °C

Time: 1



# Synthesis of lanthanum carbide pellets

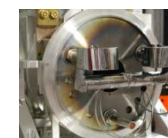
Work in IPNO: Use  $\text{LaC}_x$  as chemical homologue to  $\text{UC}_x$

	$\text{LaC}_2$	$\text{UC}_2$
Theoretical density ( $\text{g/cm}^3$ )	5.20	11.28
Crystal structure	HT $\beta$ : FCC	HT $\beta$ : FCC
	LT $\alpha$ : Tetragonal	LT $\alpha$ : Tetragonal
Lattice parameter	$\beta$ : $a_0 = 6.02 \text{ \AA}$	$\beta$ : $a_0 = 5.49 \text{ \AA}$
	$\alpha$ : $a = 4.00 \text{ \AA}$ , $c = 6.58 \text{ \AA}$	$\alpha$ : $a = 3.52 \text{ \AA}$ , $c = 5.99 \text{ \AA}$
$\alpha \rightarrow \beta$ $T_{\text{transition}}$ ( $^{\circ}\text{C}$ )	1060	1777

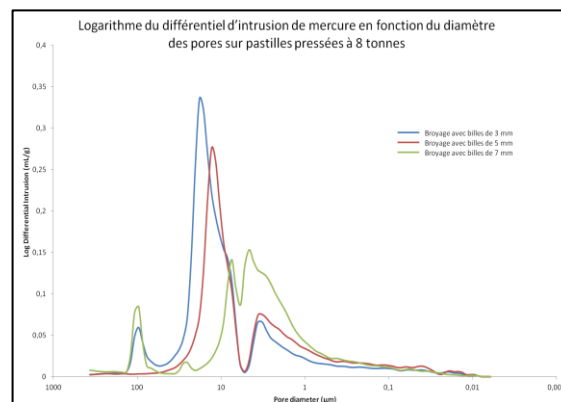
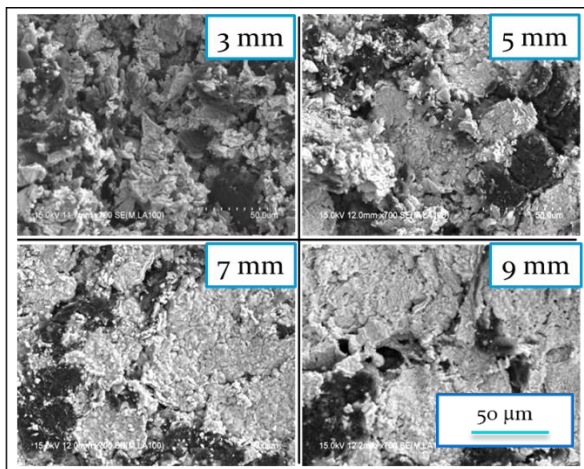
$\text{La}_2\text{O}_3$  is very hygroscopic and quickly turns into  $\text{La}(\text{OH})_3$

Choice to start with graphite and  $\text{La}(\text{OH})_3$  :

- lanthanum hydroxide stable in the air
- strong degassing during sintering = porosity  $\uparrow$



He pycnometry  
SEM  
XRD  
Hg porosimetry

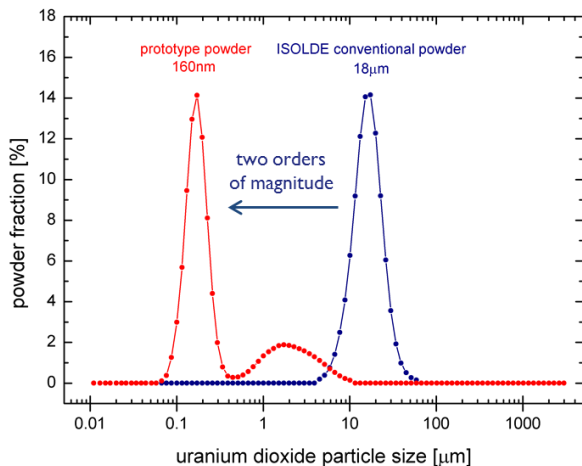


Smaller beads produce:

- Finer powder
- Larger pores

SEM Hitachi modèle S-4800 (LSI- Palaiseau). Observation backscattered electrons

## ○ Grinding with planetary mill :

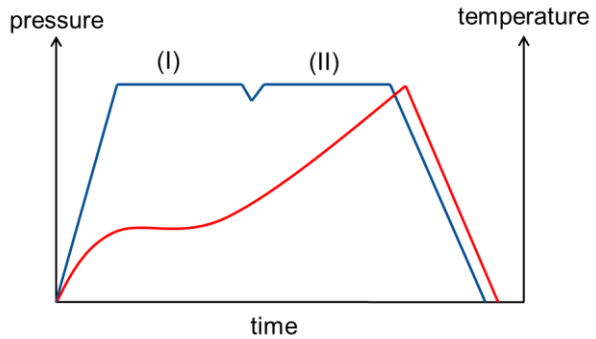
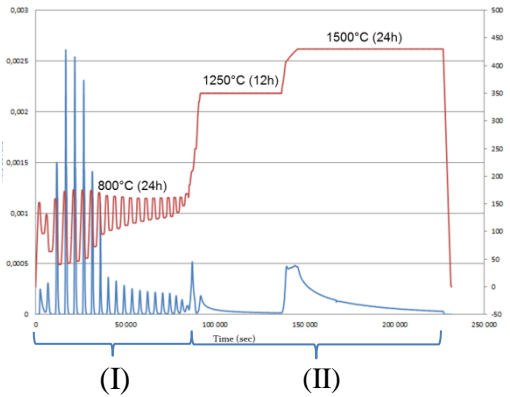
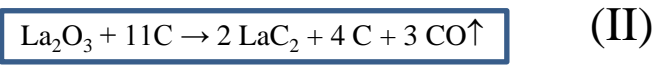


ISOLDE experience with nanoUCx shall be translated to LaCx. Milled powders would allow to produce smaller grains by two orders of magnitude. Smaller grains together with carbon nanotubes can be engineered for better preservation of microstructure at high temperature. Having small grains of the target increases the efficiency of diffusion of the isotope out of the target material.

This program can be used to manufacture LaC<sub>2</sub> targets.

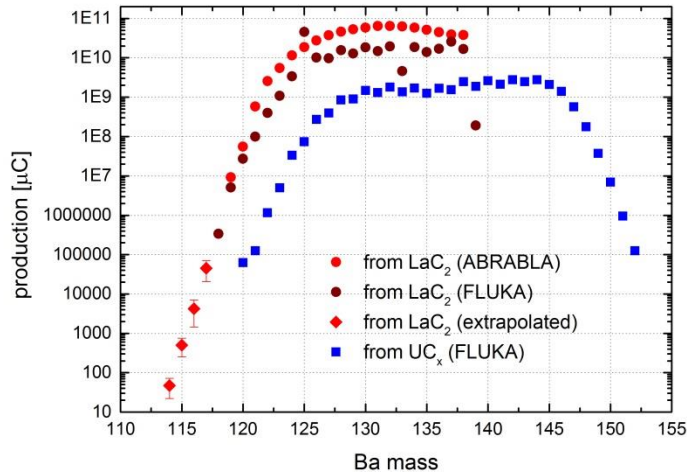
- AIMS:**
- Reducing grain size
  - Improving homogeneity in shape and grain size

## ○ Reactions during sintering:



- Choice of new outgassing program
- reduces the dwell time at high temperature
  - limit sintering responsible for the decrease of the porosity
  - limit the change in the microstructure (grain growth ...)

# Choice of LaC<sub>2</sub> for the production of i.e. neutron-deficient Barium beam



FLUKA simulations of the production of Ba isotopes from LaC<sub>2</sub> targets at ISOLDE

- neutron-deficient isotopes of Ba (Cs, Xe, etc.) isotopes LaC<sub>2</sub> shows higher production cross-sections than UC<sub>x</sub>
- For Ba with A<120 the production decreases rapidly and good extraction efficiencies are necessary for decent isotope beams

LaC<sub>2</sub> is a better candidate than UC<sub>2</sub>

## Purchase of material:

Start with La(OH)<sub>3</sub> :

- lanthanum hydroxide stable in the air
- grinding in isopropanol
- strong degassing during sintering = porosity↑

Start with carbon nanotubes :

- Size
- Thermostability
- Diffusion

Grinding equipment (Jar+Beads) in Tungsten Carbide:

- high density
- high hardness and high abrasion resistance
- high melting point (2900°C> work temperature ~ 2000°C)



