CERN

Present and Future pulsed proton beams at ISOLDE : Impact on target design and facility performance

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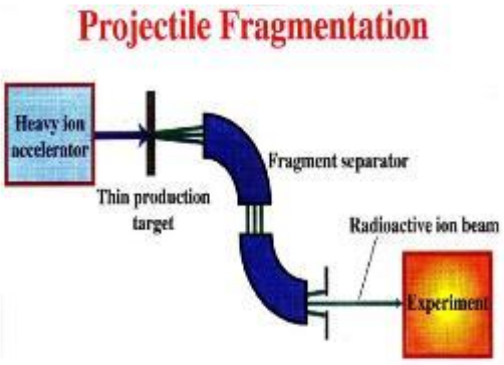
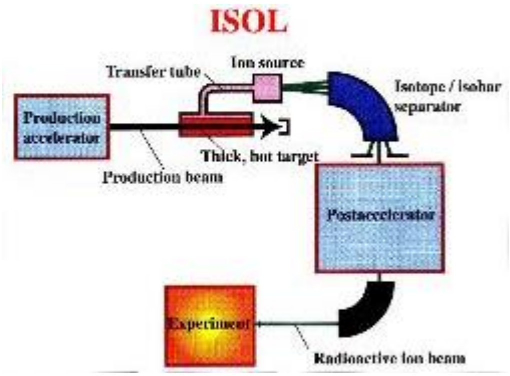
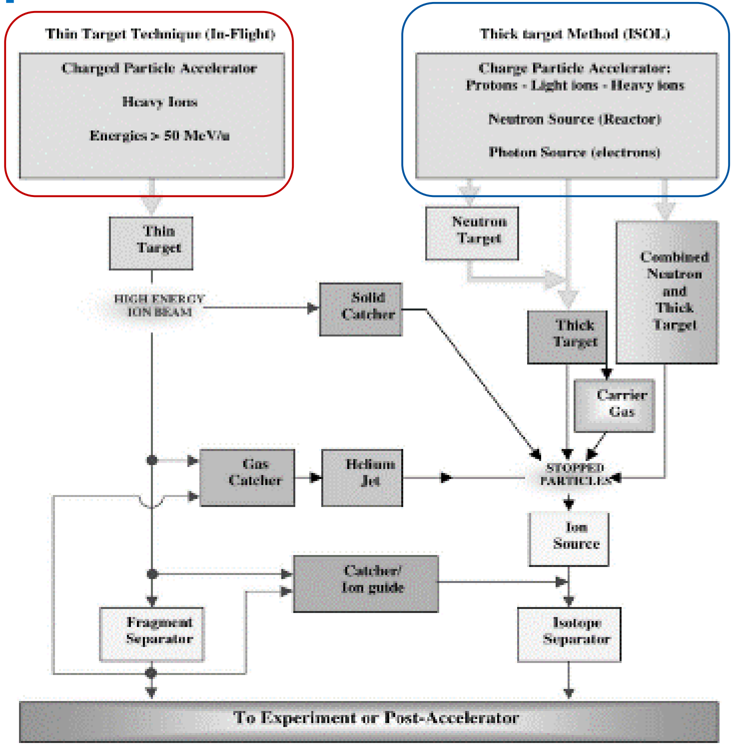
HiRadMat international conference – 8-12 July 2019

The main ingredients for isotope beam production : An accelerator for production + mass purification



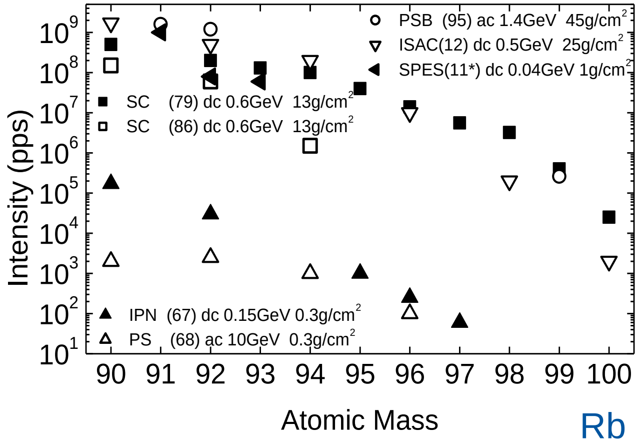




“Quality criterias” for an ISOL facility

Figure of merit of a given radioactive ion beam facility:

- **Diversity of available beams**.
- **Beam intensity (secondary ions/ primary beam μC).**
- Beam quality, for instance purity, time structure and emittance.
- **Facility down-time**.
- **Stability of beam intensity over time**.

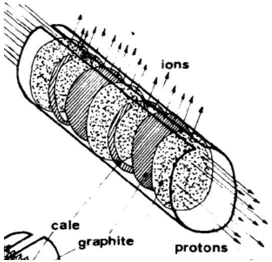


NIMB 317 (2013): 402-410

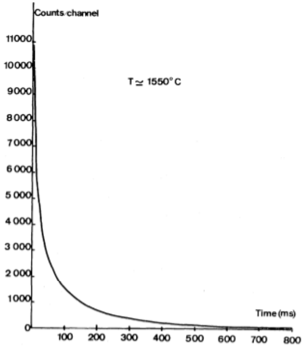
1^{st} Targets used at CERN-PS for alkali metals (p 10-24 GeV)

Target preparation:
5cm long, 6mm diameter.
36x 70µm C, 1-10µm (1-8mg/cm^2) U compound, 100µm gap: tot 0.3g/cm^2 U
Operated at ca 1500°C

$UO_2(NO_3)_2.6(H_2O)$ layer, converted to UO_3 at 200°C
Heated further to obtain U_3O_8 / UC / UC_2 / oxycarbide



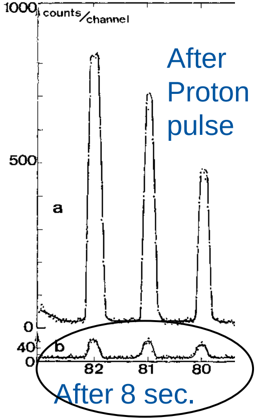
R. Klapish et al.
(UCx at CERN-PS&IPNO/CSNSM, 1967)



Na from Ir/C target

Fission
(*10.5GeV p on ThCx*)
Rb release

Phys Rev Lett, 1968

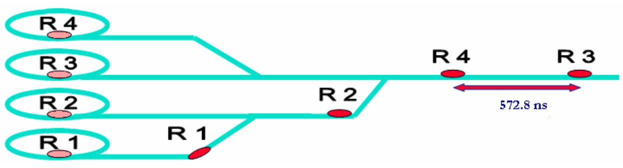


Proton beam for ISOLDE

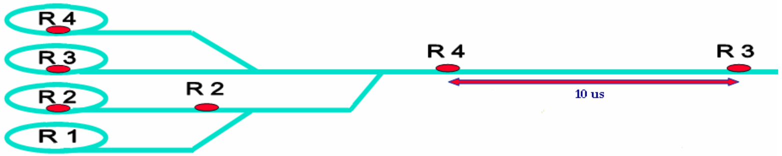
CERN Proton Synchrotron Booster

1.4 GeV protons → ISOLDE targets

- Normal beam
  - 4 bunches.
  - 230 ns bunch width.
  - 573 ns bunch spacing.
  - 1.2 s repetition rate.
  - most targets.

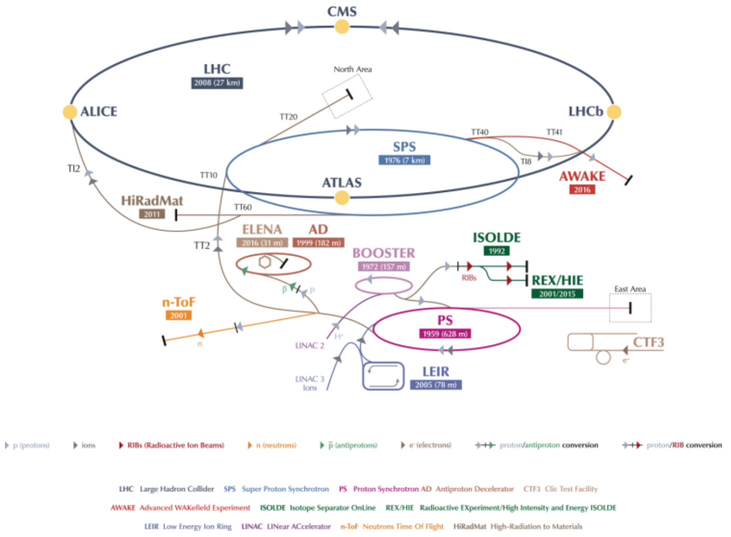


- Staggered beam
  - 3 bunches.
  - 230 ns bunch width.
  - 10 □s bunch spacing. 16µs
  - 1.2 s repetition rate.
  - liquid metal targets.

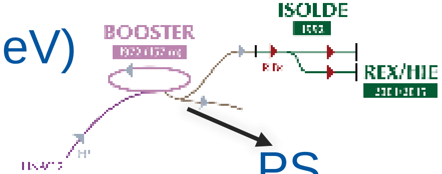


σ=2-3.5mm

	Current	Power
Average	1.92 □A	2.7 kW
Bunch	8.36 A	11.7 GW



LIU (2GeV)



Linac 4

PS

Beams under discussion : 2 GeV, 6-10kW

How to deal with a pulsed 2GeV 6-10kW beam on ISOL target(s) ?

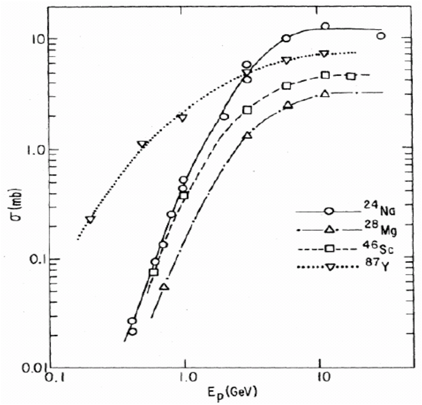
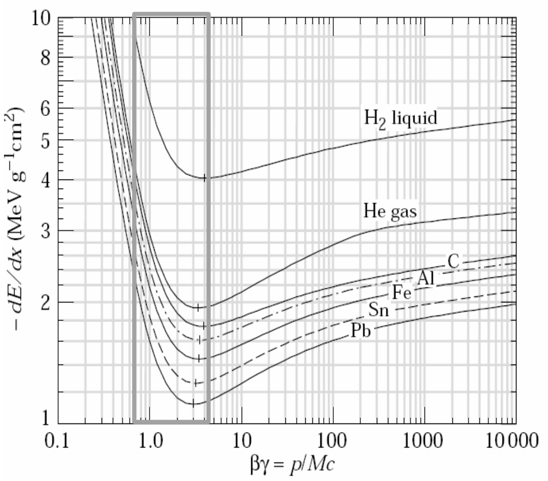
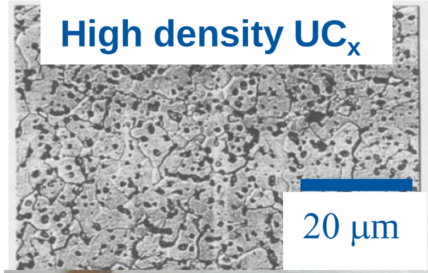
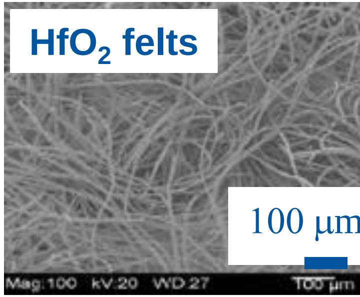
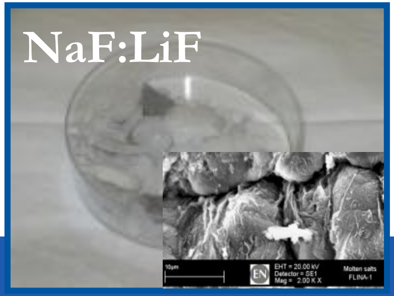


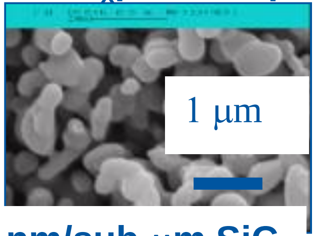
Figure 3: Measured experimental cross-section for different elements by interaction of protons of 200 MeV to 30 GeV energies with 197-Au ([4] and references therein). It is clearly seen that cross-section increases up to 10 GeV proton energy.

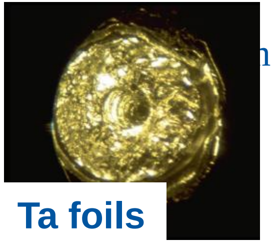








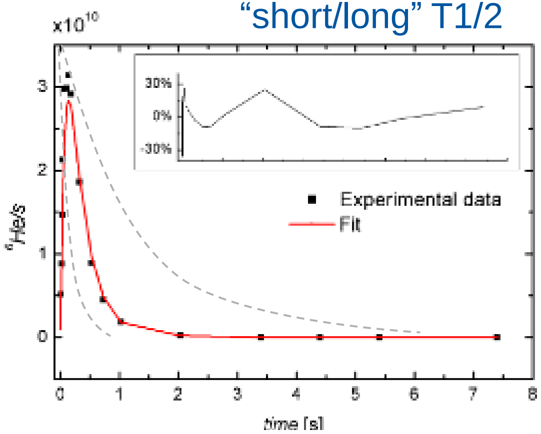




Isotope release : an analytical function

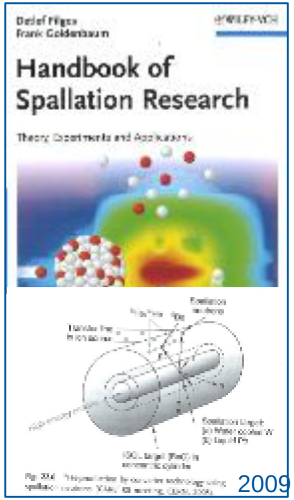
$$p(t) \propto p_{eff}(t) \star p_{diff}(t)$$

Isotope with
“short/long” T1/2



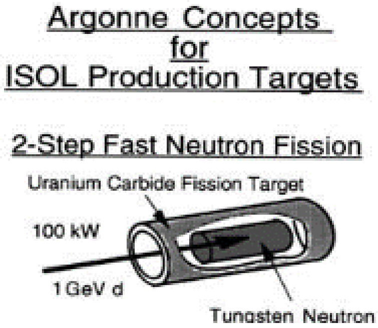
Temperature [°C]	t_{eff1} [ms]	t_{eff2} [ms]	t_{diff} [ms]	Release efficiency [%]	^{6}He production (N_0)
700	5.5	32	320	59	$2.7\ 10^{10}$
800	5.6	28	150	71	$2.6\ 10^{10}$
1000	4.7	28	1600	51	$4.1\ 10^{10}$
1130	3.3	27	190	79	$3.1\ 10^{10}$
1400	1.8	24	270	82	$2.9\ 10^{10}$

Eur. Phys. Lett. 98, 32001 (2012) & Europhysics news

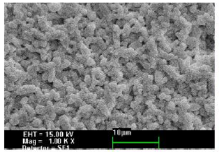


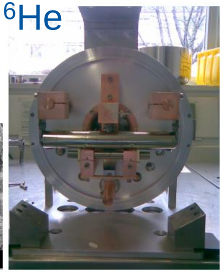
2009

J. Nolen (1995/2002)



^{9}Be (n,α) ^{6}He



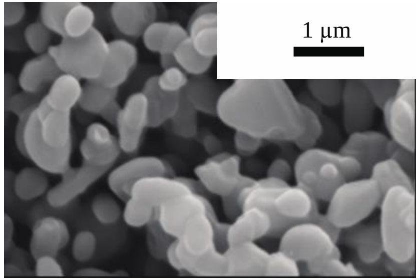


Beam induced grain growth

1^{st} submicron targets operated for fast isotope release (Mg)

High purity α-SiC porous target (63%), Saint Gobain Recherche

Before irradiation



Diffusion limited release:

$$\varepsilon_{diff} = \frac{3}{\pi}\sqrt{\frac{\mu}{\lambda}}, \mu = \frac{\pi^2 D}{r^2} \qquad \lambda << \mu$$

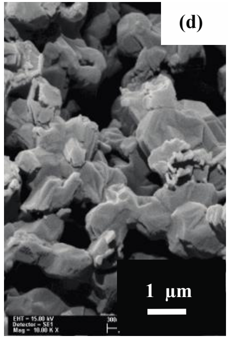
Sandrina Fernandes (EPFL, PSI)

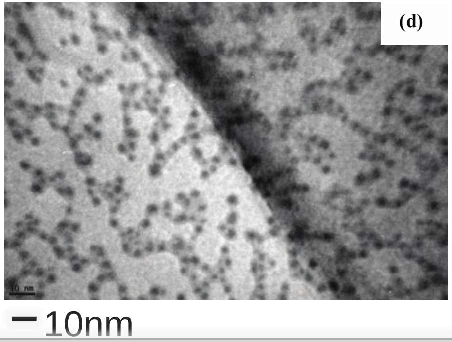
CERN-THESIS-2010-170
03/12/2010

After irradiation

Unit SiC334, operated at 1600C for 5days, ~ 3e17poT

Diam 14mm





Beam induced grain growth : dependance of position in pellet

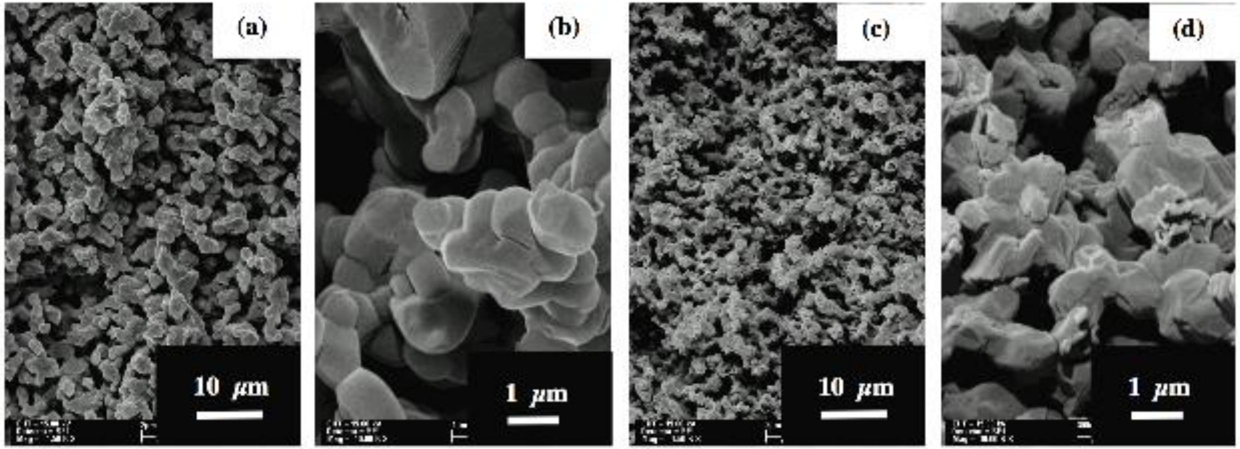
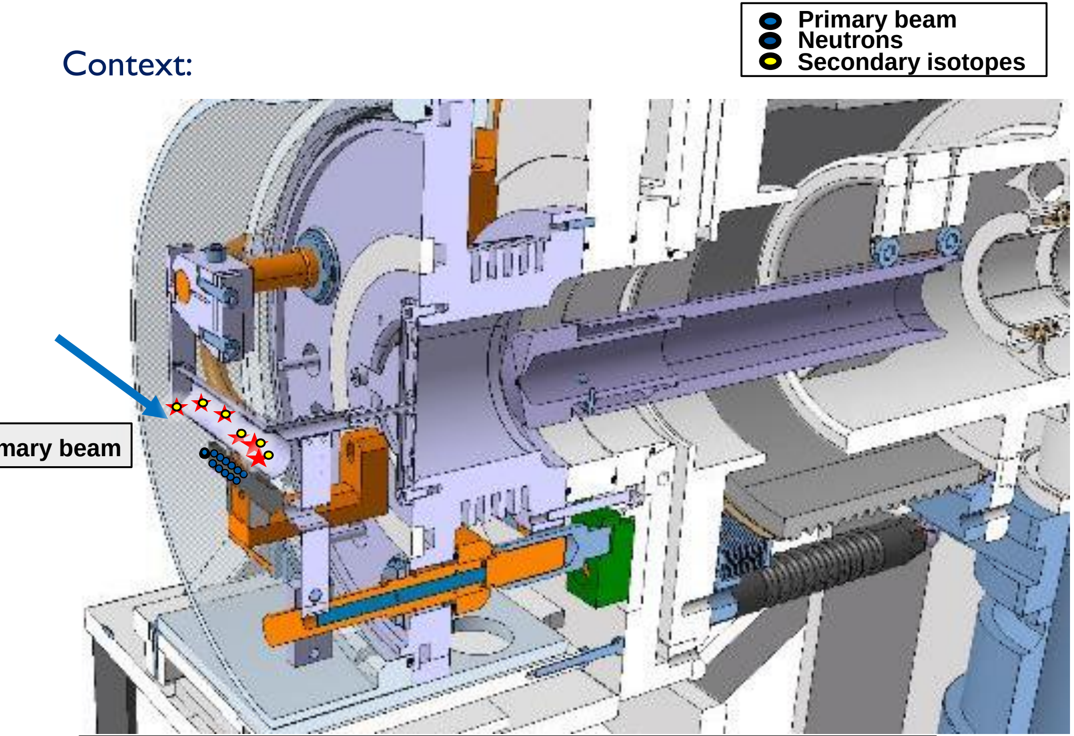


Fig. 3.16: *SEM analysis in the cross-section of the fractured pellet 40-41-42: a) center of the pellet, b) higher mag., c) region near the edge and d) higher magnification.*

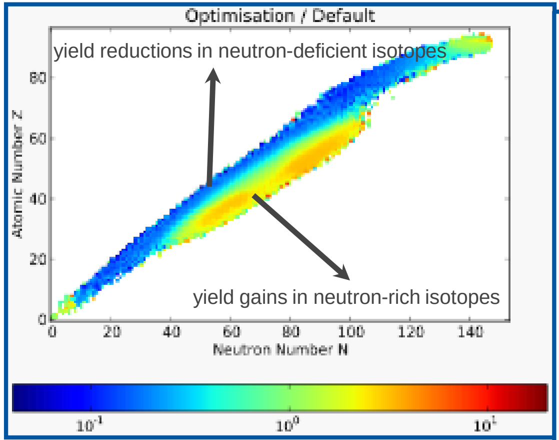
Neutron spallation source – "classical"

Context:

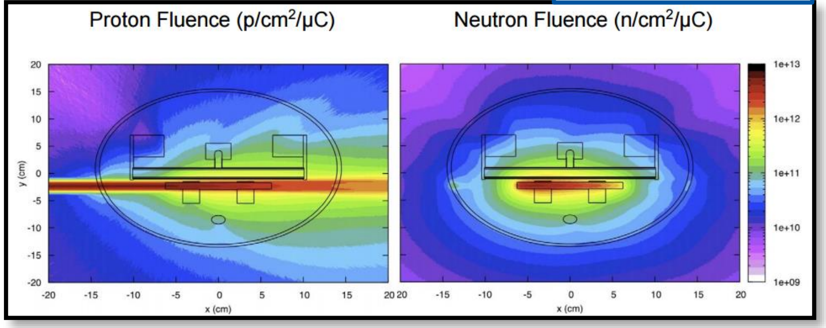


Secondary reaction created from neutron impacting onto target material

Secondary isotopes diffuse/effuse out of the material toward ion source

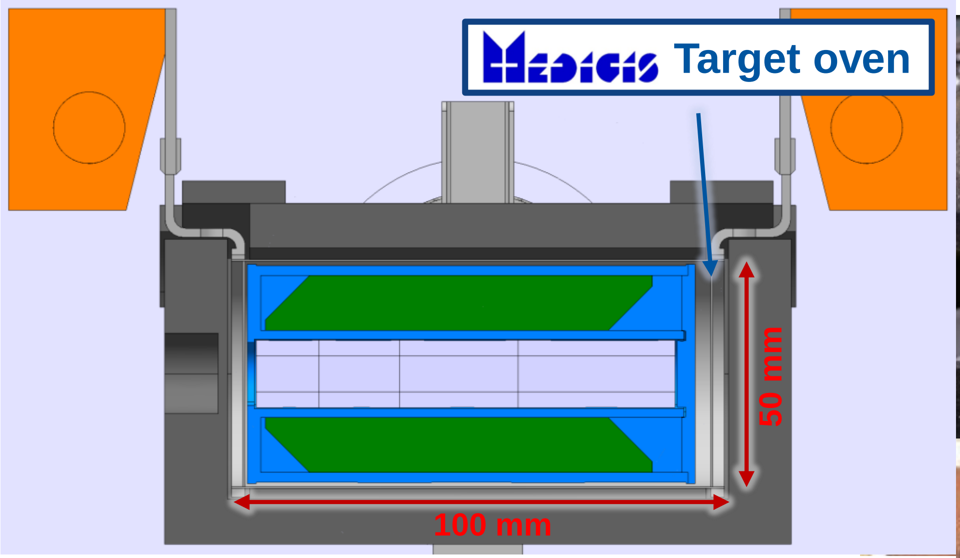


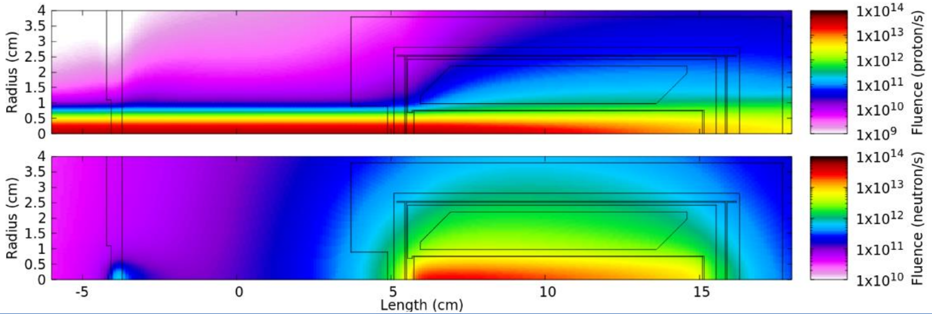
- Less contaminant from the neutron-deficient side -> beam more pure
- BUT current design not yet optimized on proton & neutron fluence



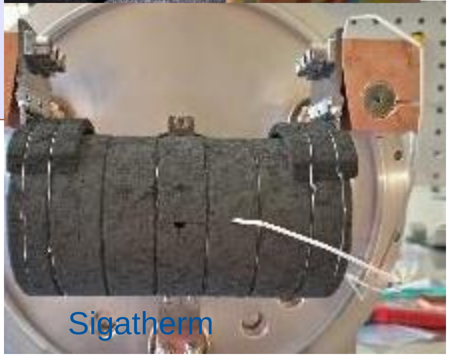
EPJA 48.6 (2012): 90.

Details on the prototype



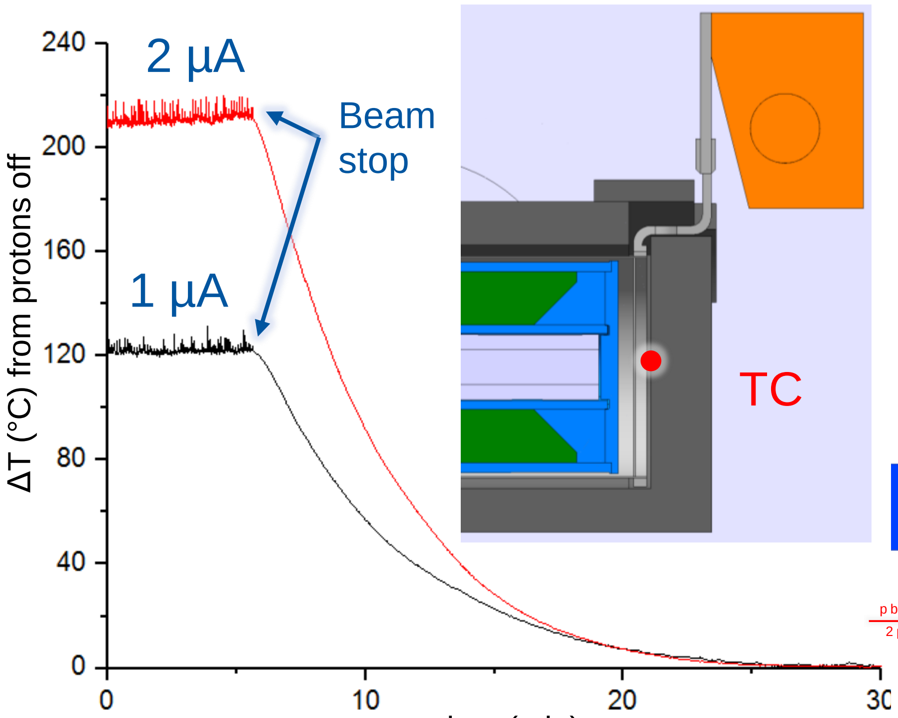


After (2200 °C – 16 h) – no change

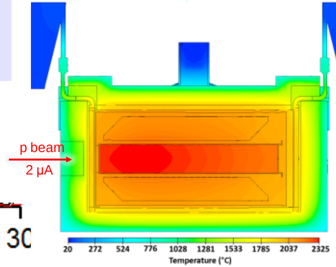


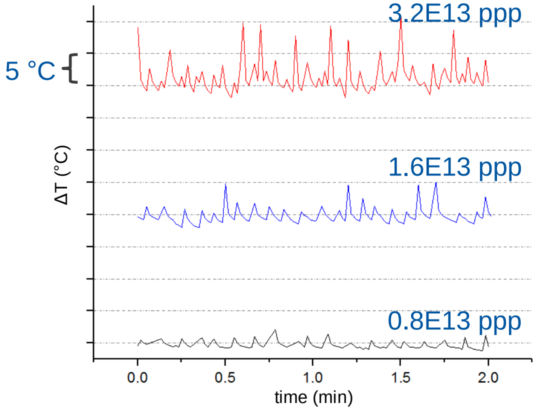
https://doi.org/10.1016/j.nimb.2019.04.060

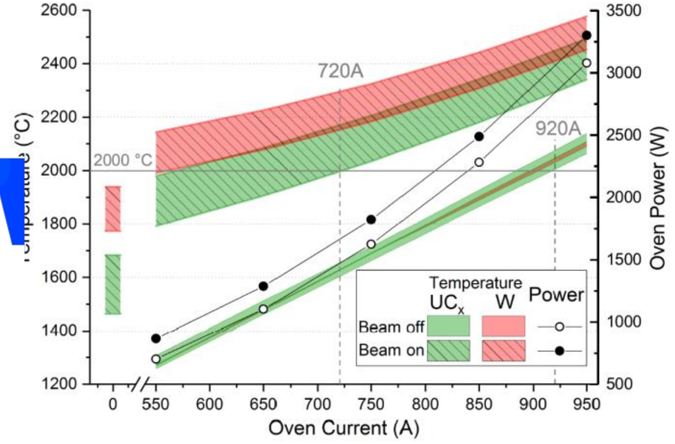
Online thermal data and simulations



J.P. Ramos, et al., to be published.

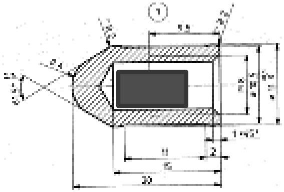






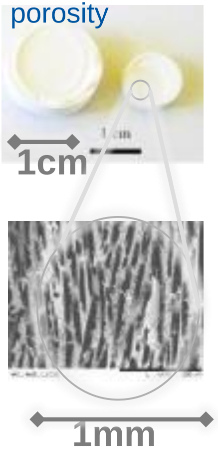
Study on porous targets with PSB and SPS beams

8 samples: (pellets Ø 2 cm x 2 cm) – 4 SiC & 4 Al_2O_3



beam: NORMGPS – 1.4 GeV, 3.2×10^{13}/pulse (2.4 μs/1.2s, 3-4 bunches), σ = 2.3
RaBIT + passive irradiation on target unit

Al_2O_3 with uniaxial porosity



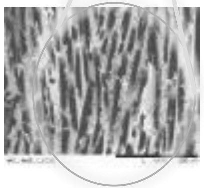
beam: SPS – 450 GeV, 4.9×10^{13}/pulse (7.2 μs/18s, 3-4 bunches), σ = 2.0
Max. cycles = 100
Setup - 8 samples in a row

NIMB317 (2013): 385-388.

Characterization of the target irradiation

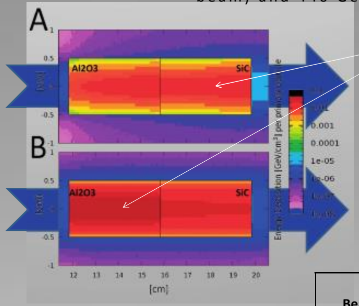
Al_2O_3 with uniaxial porosity

1cm



1mm

In order to predict if selected microstructures will survive the impact of a proton beam the study of mechanical integrity under irradiation with protons of 1.4 GeV (ISOLDE beam) and 440 GeV (HiRadMat beam) was performed.



Energy deposition spectra. A) HiRadMat. B) ISOLDE.

Maximum of deposited energy (E_{dep})
Region of the biggest thermal shock (σ)

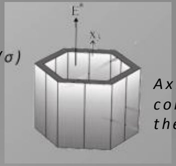
$$\sigma = \Delta T \, E^{*}_{young} \, \alpha$$

$$\Delta T = \frac{E_{dep}}{\rho^{*} C_p}$$

$$E^{*}_{young} = \frac{\rho^{*}}{\rho} E_{young}$$

$$\rho^{*} = \rho \, (1 - porosity)$$

$$\sigma^{*}_{f} = C \, \sigma_{fs} \left(\frac{\rho^{*}}{\rho}\right)^{3/2}$$



Axial compression of the walls [7]

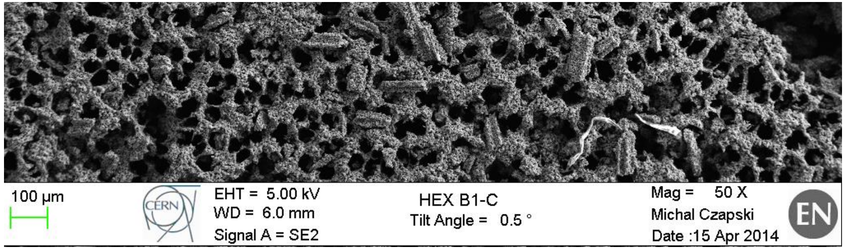
Beam	Energy *GeV*	Protons per pulse *x10^12*	Sigma *mm*	Repetition rate s
ISOLDE	1.4	30	3.5	1.2
HiRadMat	440	8	2.0	40

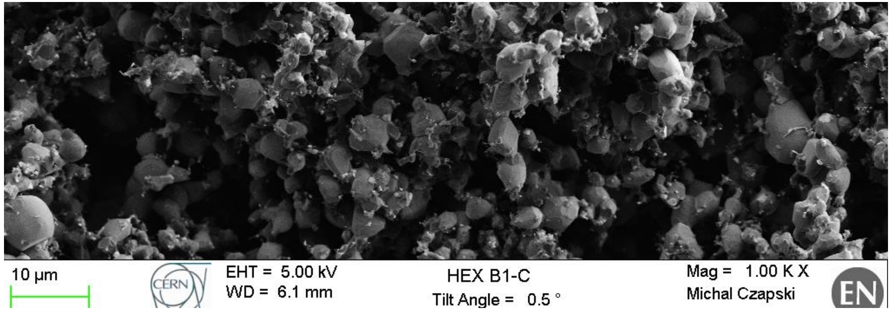
Powder	Beam	σ *[Mpa]*	σ^{*}_{f} *[MPa]*
Ceralox®	HiRadMat	31	276
	ISOLDE	23	
Hexoloy®	HiRadMat	19	184
	ISOLDE	15	
Crystar®	HiRadMat	21	60
	ISOLDE	16	

NIMB317 (2013): 385-388.

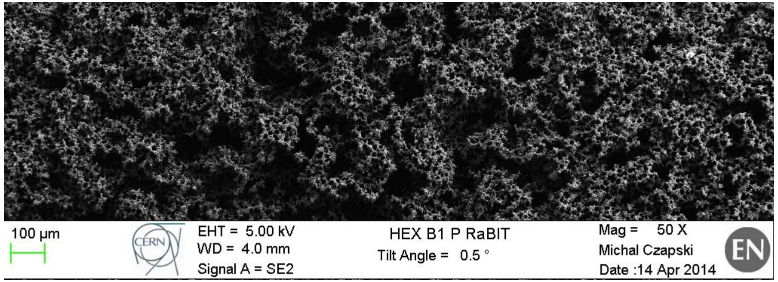
Comparison of microstructure – hexaloy SiC

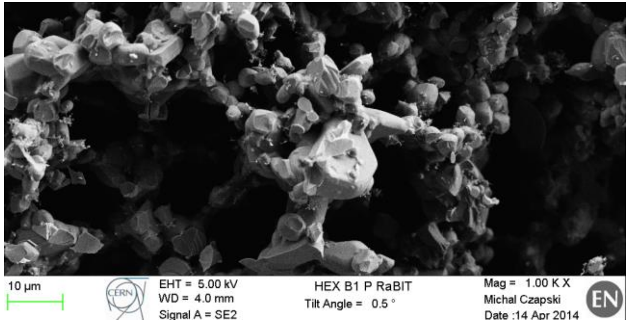
Before irradiation



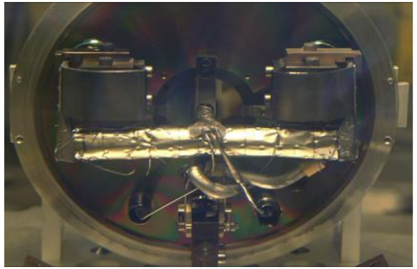


Post irradiation

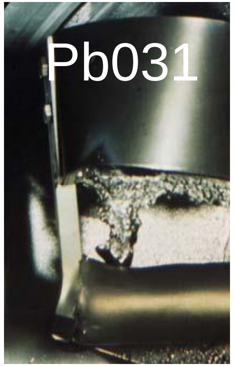




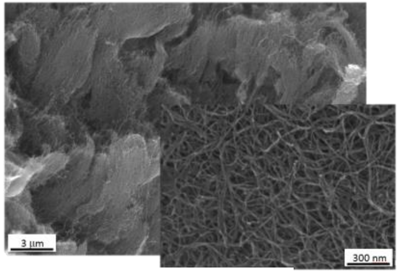
Some Issues to be addressed/checked with new beam parameters



Plastic deformation seen on Tantalum spallation neutron Source

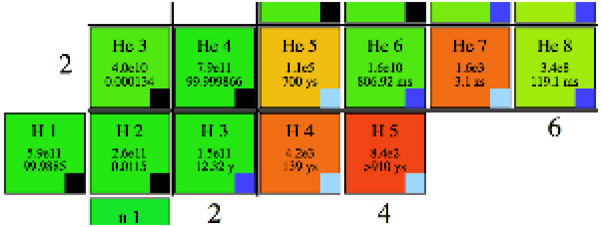


Welding rupture in window of molten lead targets (not an issue observed with solid targets at >2000C)



MWCNT

Porous target stability



Production of H and He (from 1 appm to 20appm in UCx) And its impact
We remain in the sub-dpa range

Outlook

- Possible proton beam upgrades can lead to important improvements in the output of the ISOLDE facility

- In the list of criteria :
  - beam intensity
  - beam stability
  - facility downtime

  are directly linked to target design which can cope, or not, with the new parameters

- Some investigations at the HiRadMat facility could help validate the most critical parts

Reserve

Acknowledgements :
D. Leimbach, J. Ballof, F. B. Pamies, E. Barbero, B. Crepieux, V. Samothrakis, T. Giles, S. Warren, B. Marsh, K. Chrysalidis, S. Wilkins, C. Granados, M. Mongeot, J. Karthein, D. Houngbo, L. Popescu, M. Dierckx, L. Egoriti, A. Gottberg, M. Ballan, S. Marzari, G. Neyens, K. Johnston, A. Dorsival, A.P. Bernardes, S. Sgobba, R.Luis, S. Cimino, D. Urffer, C. Tardivat

Reserve

Beam intensity and target temperature

RIB intensity [s^{-1} µA^{-1}]

Prim. Part. beam Intensity [s^{-1} µA^{-1}]

Avogadro

Diffus.+Effus. Efficiency

Beam transport Efficiency

$$I = \int \sigma(E)\,\Phi(E,x)\,\rho(x)\,N^{\#}/A\,dx\,\varepsilon_{\mathrm{diff+eff}}\,\varepsilon_{\mathrm{ion}}\,\varepsilon_{\mathrm{optics}}$$

Cross section [cm^2]

Target density [g cm^{-3}]

Target Atomic Mass [g]

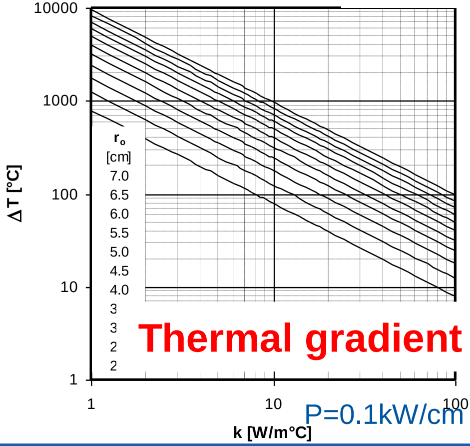
Ionization Efficiency

Energy deposition [MeV g^{-1}cm^2]

$$-\frac{dE}{\rho dx} \propto Z/A$$

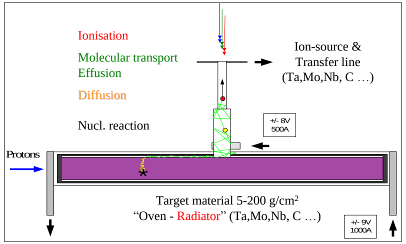
T=(1200-2200°C) + ΔT

Oxide Carbide Metal graphite



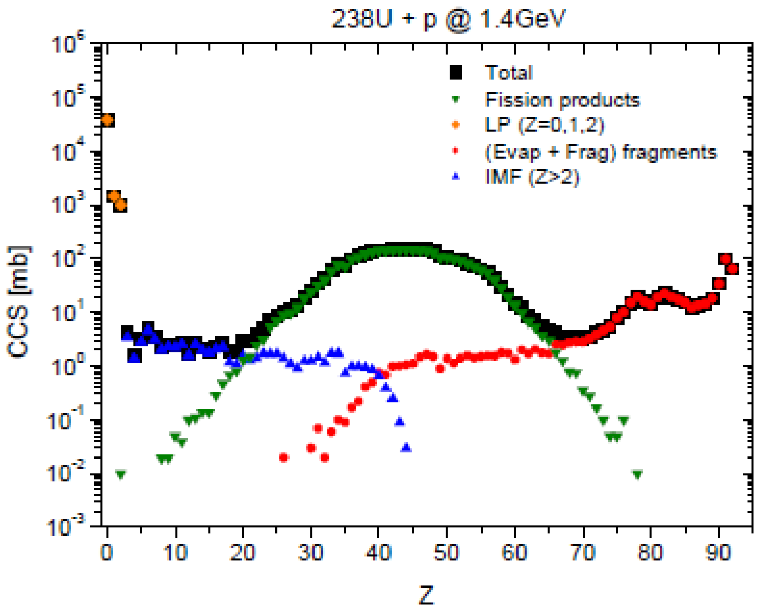
Thermal gradient

P=0.1kW/cm



Release time [s]

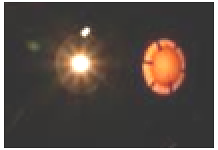
$\tau_0 \sim V^y \exp(1/T)$

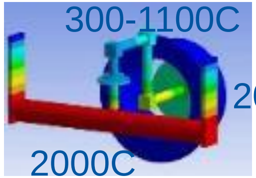

238U + p @ 1.4GeV
Total
Fission products
LP (Z=0,1,2)
(Evap + Frag) fragments
IMF (Z>2)
CCS [mb]
Z

From production to beam formation

Target (Nb/ZrO_2
by reactive brazing);
Operation at 1400C

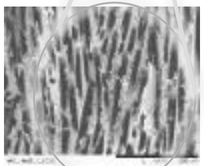
EURISOL-DS Final Report,
J. Cornell Ed, GANIL (2009)





E. Bouquerel, et al. "Beam purification by selective trapping in the transfer line of an ISOL target unit." NIMB 266.19 (2008): 4298-4302.

Al_2O_3 with
uniaxial
porosity



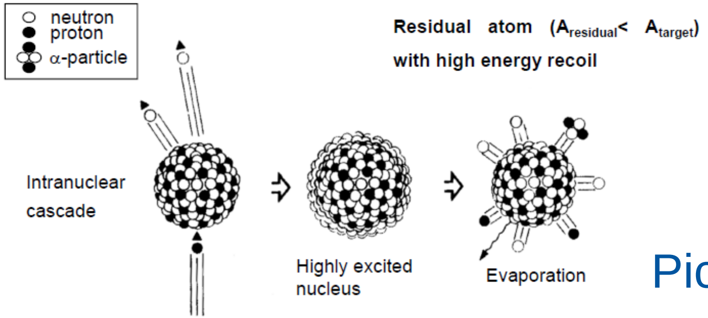
M. Czapski, et al. "Porous silicon carbide and aluminum oxide with unidirectional open porosity as model target materials for radioisotope beam production." NIMB317 (2013): 385-388.

Beam interaction with target

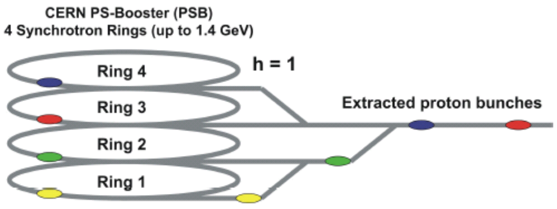
Nuclear transmutations

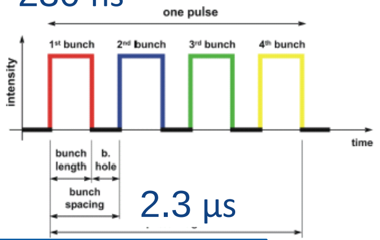
Production of impurities (He, H)

10^{-16} — 10^{-13} - 10^{-8} — 10^{-6} Time [s]

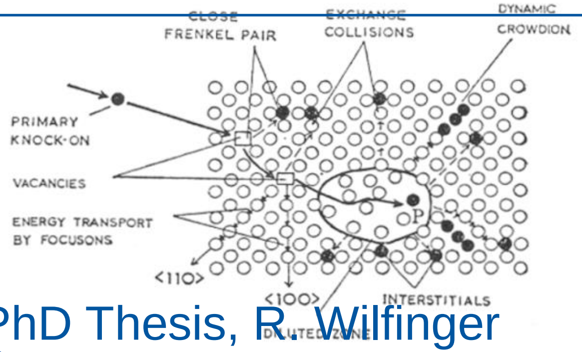


Beam power deposition – pulsed beams!





Thermomechanical stresses and shockwaves



Pictures from PhD Thesis, R. Wilfinger

Target Materials

120 Materials (possibly more) were tested and/or used as ISOL targets!

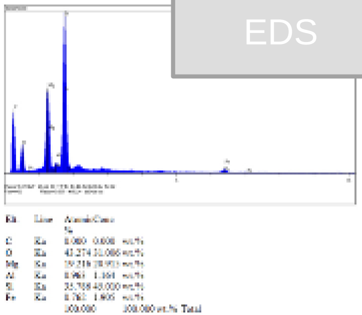
							Oxides				
Carbon Based	AlC_2	B_4C	C(gr)	**C (MWCNT)**	CaC_2	CmC_x	**Al_2O_3**	B_2O_3	BaO	**BeO**	
	GdC_x	**LaC_2**	ScC_2	**SiC**	TaC_x	ThC_2	**CaO**	CeO_2	Cr_2O_3	**HfO_2**	
	TiC	**UC_2**	VC	ZrC	Cm	Hf	La_2O_3	MgO	**NiO**	SrO	
	Ir	Ir/C	Ta/Ir/W	Mo	Nb	Os	Ta_2O_3	**ThO_2**	**TiO_2**	UO_2	
	Pu	Pt/C	Re	Re/C	Ru	Ru/C	Si layers	**Y_2O_3**	**ZrO_2**	ThO2/Ta	
Solid Metals	Sn/C	**Ta**	Ta/W	Ti	Th	Th/Ta	AlN	BaB_6	$BaZrO_3$	$TiO_2.(H_2O)_x$	First Materials
	Th/Nb	U	U/C	V	W	Zr	BN	Ca-zeolite	CaB_6	$ZrO_2.(H_2O)_x$	
	Au	Ag	Bi	Cd	Ce	Ce_3S_4	$Ce(OH)_4$	CaF_2	CeB_6	$CeO_2.(H_2O)_x$	
	Er:Cu	Ge	Gd:Cu	Hg	La	La:(Th/Si/Sc)	CeS	LuF_3	Na-zeolite	$ThO_2.(H_2O)_x$	
	La:(Y,Gd,Lu)	**NaF:LiF**	$NaF:ZrF_4$	Nd	Ni	Pr	Ta_5Si_3	Hf_5Ge_3	Hf_5Si_3	Sr stearate	
	Pt:B	Sc:La	Sn	Tb	TeO_2:KCl:LiCl	ThF_4:LiF	Hf_5Sn_3	Ta_5Si_3	Tl-zeolite	Ba stearate	
	Pb	**Pb:Bi**	Y:La	U	U:Cr	Zn	$Th(OH)_4$	Zr_5Ge_3	Zr_5Si_3	$TeCl_4$	
	Molten						Others				

- In squares – currently used at ISOLDE
- **Underlined and Bold** – had some kind of material development

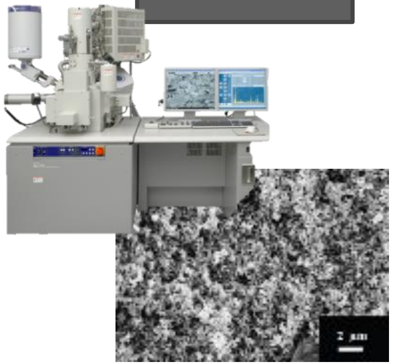
Post-irradiation study

at EN/MME-MM

EDS



SEM



Irradiation assisted stress corrosion cracking

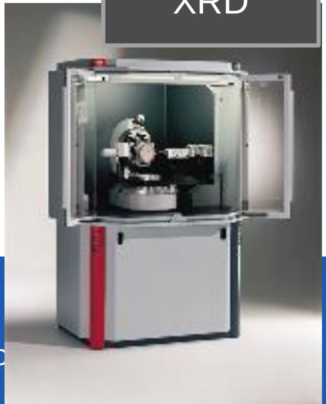
Microcompositional effects:

diffusion and segregation of impurities

Microstructural changes

- recrystalisation,
- phase transformation
- grain size change
- pore shrinkage
- grain coarsing

XRD



HiRadMat assessment

Expected Isotope production in the entire load of SiC per 10^{16} protons		Activity *MBq*	after one year *MBq*	Authoriz-ation limit *MBq*	H_{10} at 1 m after one year *mSv/h*	$H_{0,07}$ at 10 cm after one year *mSv/h*	$H_{0,07}$ at 10 cm after one year *mSv/h /pellet*	Expected dose equivalent $H_{0,07}$ at 10 cm for manipulation time of 1 pellet (1 min of operation) *µSv*
Na-22 (e,b⁺, γ)	4.25E+12	**0.09**	**0.07**	3.00	**2.36E-05**	0.14	0.02	**1.49E-05**
Be-7 (e, γ)	1.29E+13	**1.94**	**0.002**	100	**1.96E-08**	2.45E-06	3.06E-07	**2.55E-10**

Expected Isotope production in the entire load of Al_2O_3 per 10^{16} protons		Activity *MBq*	after one year *MBq*	Authoriz-ation limit *MBq*	H_{10} at 1 m after one year *mSv/h*	$H_{0,07}$ at 10 cm after one year *mSv/h*	$H_{0,07}$ at 10 cm after one year *mSv/h /pellet*	Expected dose equivalent $H_{0,07}$ at 10 cm for manipulation time of 1 pellet (1 min of operation) *µSv*
Na-22 (e,b⁺, γ)	6.28E+12	**3.87**	**0.12**	3.00	**4.26E-05**	0.26	0.03	**6.72E-05**
Be-7 (e, γ)	**1.86E+13**	**2.80**	**0.12**	100	**9.94E-07**	1.24E-04	1.55E-05	**3.23E-08**