Post-irradiation analysis of refractory metal foils irradiated at high temperature for EURISOL 100 kW targets

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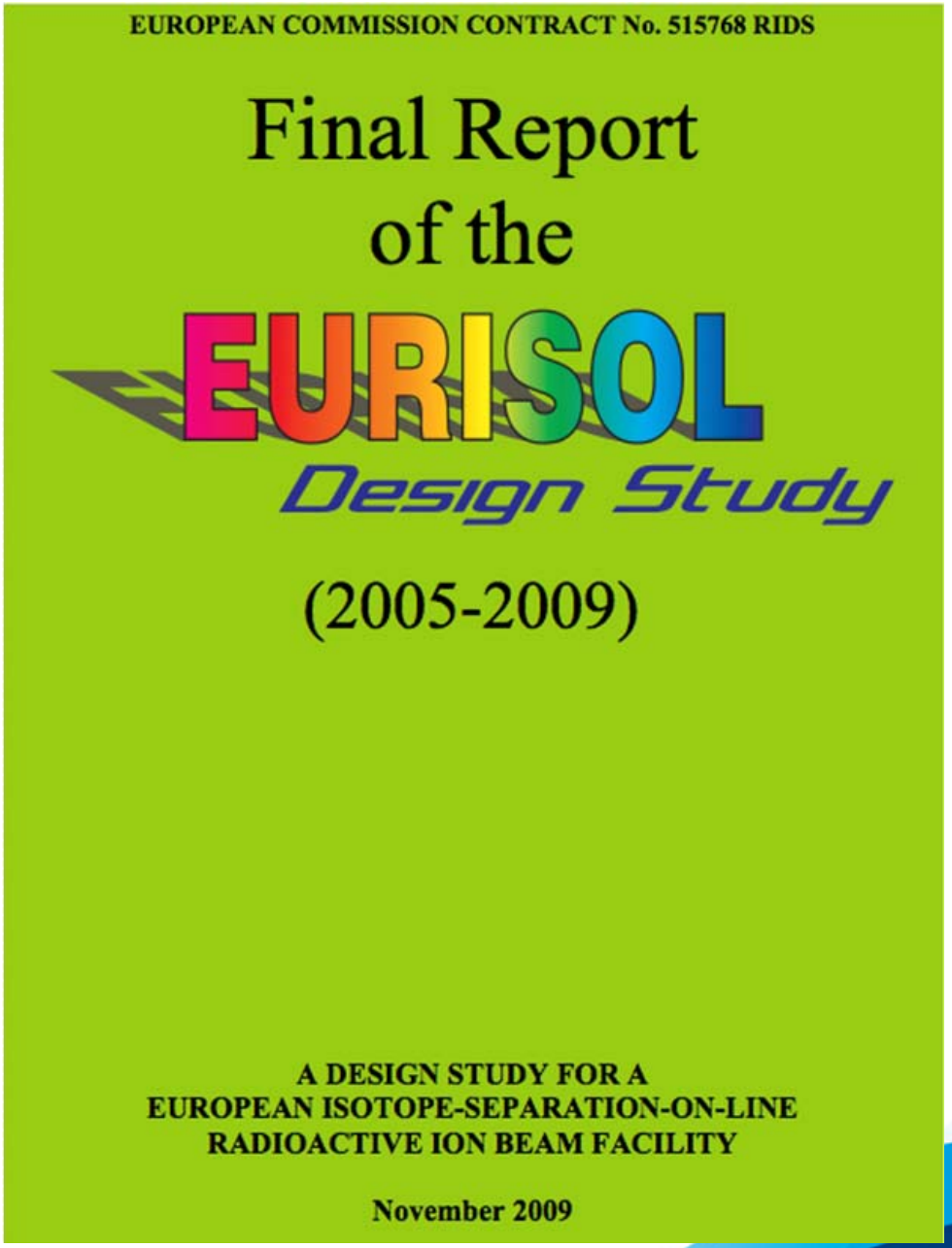
Outline

- The EURISOL Design Study
- TARPIPE motivation
- Pre-irradiation: samples/simulations/controls
- Irradiation
- Dismantling
- Post irradiation analysis

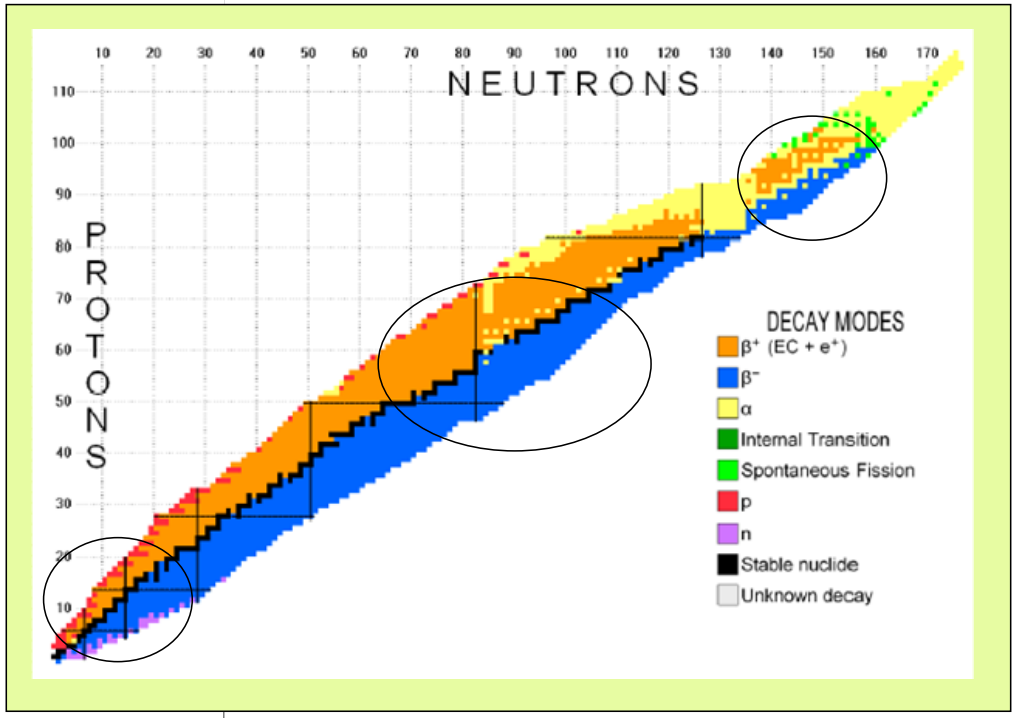
EUROPEAN SPALLATION SOURCE

PAUL SCHERRER INSTITUT

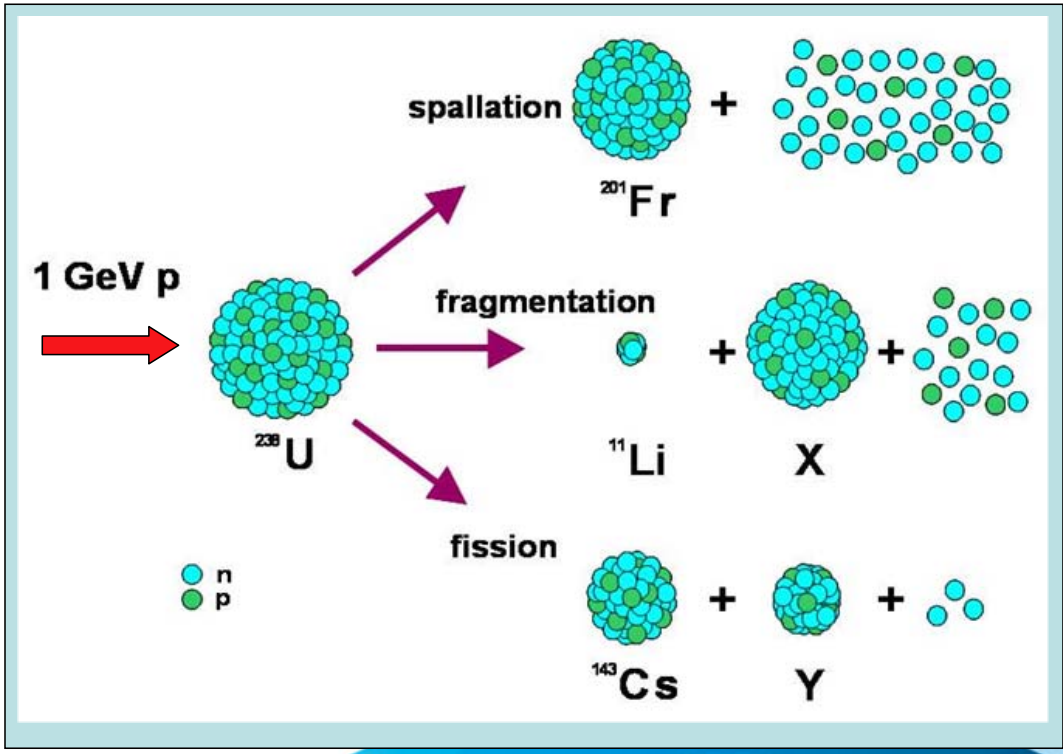
EURISOL Design Study



Radioactive Isotope Production



e.g. ISOLDE (1963-2004)
~ 750 RIBs produced



Relevant RIB Target Properties

- Density
- Porosity
- Thermal conductivity
- Heat capacity
- Emissivity
- Chemical reactivity
- Vapour pressure
- Ageing

- Energy deposition
- Isotope production
- Heat transfer
- In-target diffusion
- Effusion towards ion source

TARPIPE: Ageing under irradiation at high temperatures

Release Properties Temperature Dependence

Release efficiency

$$Y(T_{1/2}) = \int_{t=0}^{\infty} p(t) e^{-\lambda t} dt$$

Decay survival term

Delay function=differentiation of fractional release F(t)

Fractional release

$$F(t) = \int_{t'=0}^{t} p(t') dt' = I_{out}(t) / I_{in} \eta_i \eta_t$$

Where η_i and η_t are transmission and ionisation efficiencies

Pure diffusion

$$p_\mu(t) = \frac{8\mu_0}{\pi^2} \sum_{n=0}^{\infty} e^{-\mu_n t}$$

$$\mu_n = \mu_0 (2n+1)^2 \quad \mu_0 = \pi^2 D / d^2 \quad D = D_0 \exp(-E_A / kT)$$

μ_0 is the delay parameter, D_0 the diffusion constant and E_a the activation energy

Pure effusion

$$p_\upsilon(t) = \upsilon e^{-\upsilon t}$$

$$\tau_\upsilon^{del} = 1/\upsilon = \chi(\tau_a + \tau_f)$$

- mean number of collisions, χ
- mean sticking time, τ_a, per wall collision, depends essentially on temperature and adsoption enthalpy ΔH_a of surface of enclosure
- mean flight time, τ_f, between two wall collisions

Diffusion + effusion

$$p(t) = \int_{t'=0}^{t} p_\mu(t') p_\upsilon(t - t') dt'$$

Assume diffusion and effusion take places in subsequent intervals [0, t'] and [t',t].

Limitations

Diffusion-limited:
- Liquid metal targets (Pb, Sn)

Effusion-limited:
- surface adsorption for refractory elements
- intrinsic delay for very short half-lives

Preference: operation at uniformly high temperatures

TARPIPE Test Jig

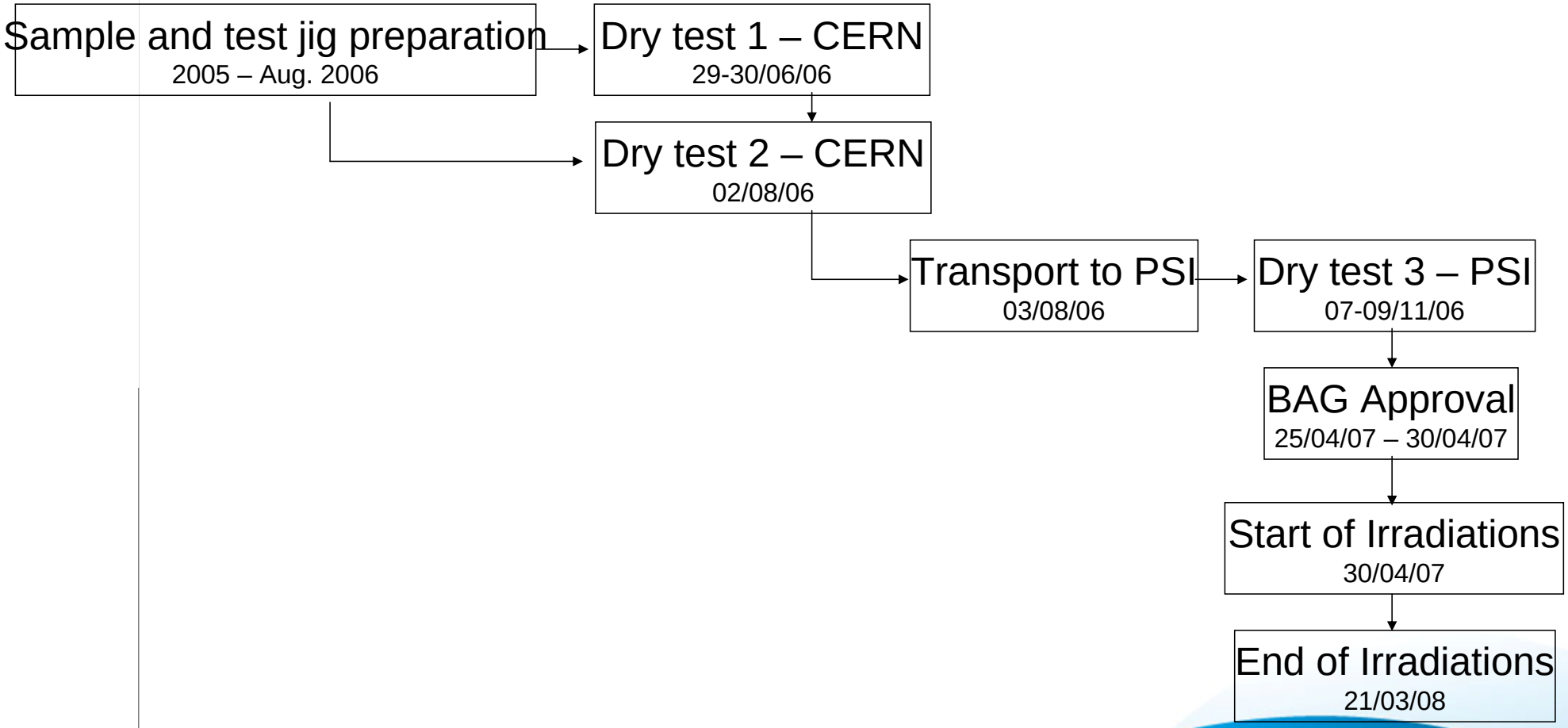
Adapted from LiSoR

Level	Container	Content
1	Mo	Oxides
2	Mo	Oxides + Metals
3	Graphite	Carbides
4	Mo	Metals

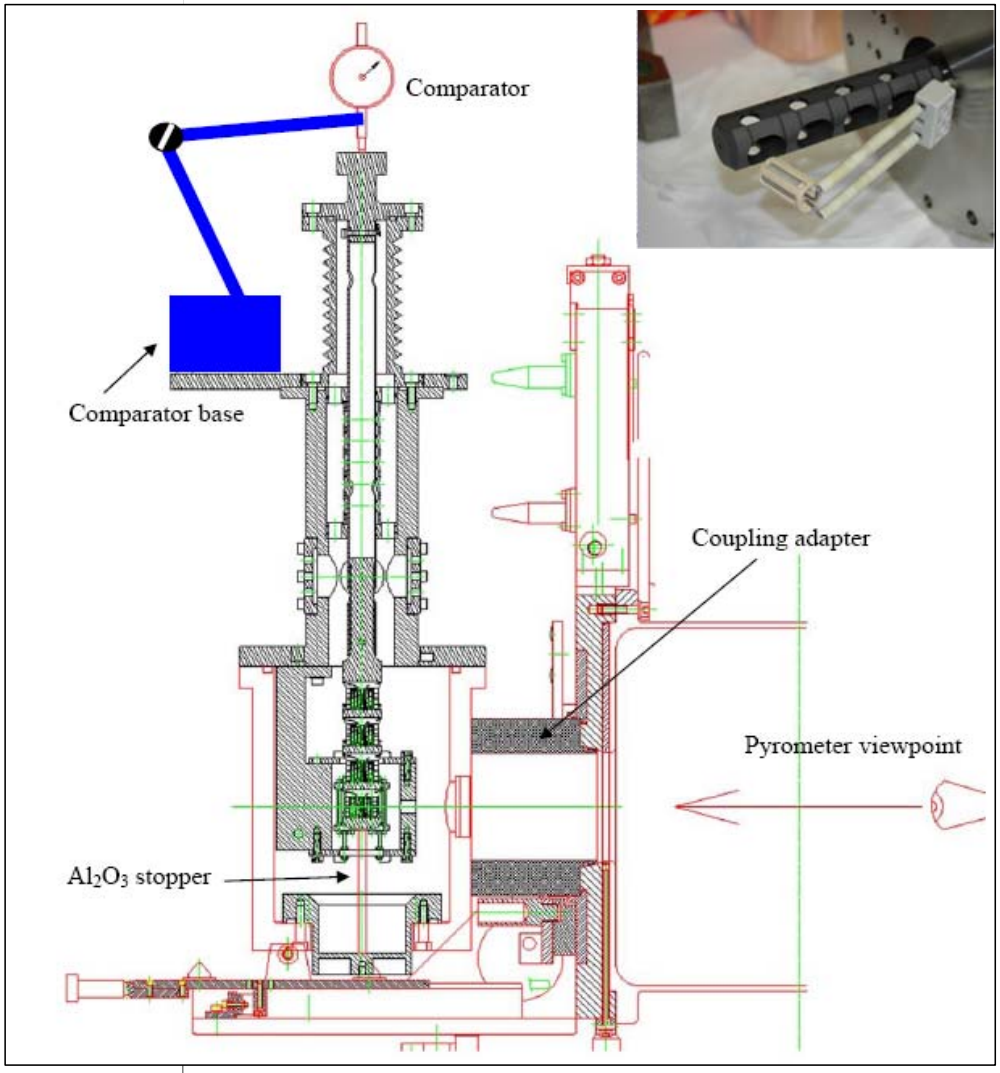
TARPIPE Samples

Material	Description	Melting point [°C]	Density [g.cm^{-3}]
Al_2O_3	SG foam	2054	1.03
Al_2O_3	SG foam	2054	0.42
Al_2O_3	Baikowski p.p.	2054	2.32
Al_2O_3	SG transparent	2054	3.92
MgO	CERN standard	2852	2.64
Y_2O_3	Zircar Zirconia felt	2690	0.32
CeO_2	Zircar Zirconia felt	2400	0.41
HfO_2	Zircar Zirconia felt	2774	0.63
TaO_2	Leico weave satin	–	1.24
ZrO_2	Zircar Zirconia felt	2677	0.37
SiC	SG foam	2700	0.37
SiC	SG CRYSTAR	2700	2.70
C	Carbon nanotubes	3527	0.32
SiC + C	SG SiC p.p. + carbon nanotubes	–	0.58
C	Nanotube fibres	3527	–
Mo, Nb, Re, Ta, W	Ref. Metal foils	2477 – 3410	8.58 – 21.04
Hf, Ir, Rh, Ti, Zr	Metal foils	1668 – 2410	4.54 – 22.4

TARPIPE Schedule



Dry Tests: CERN

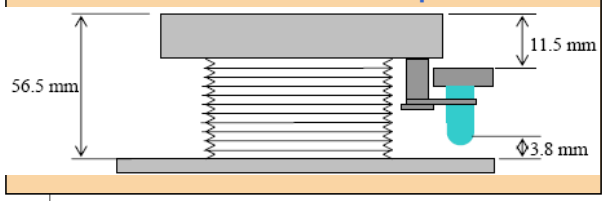


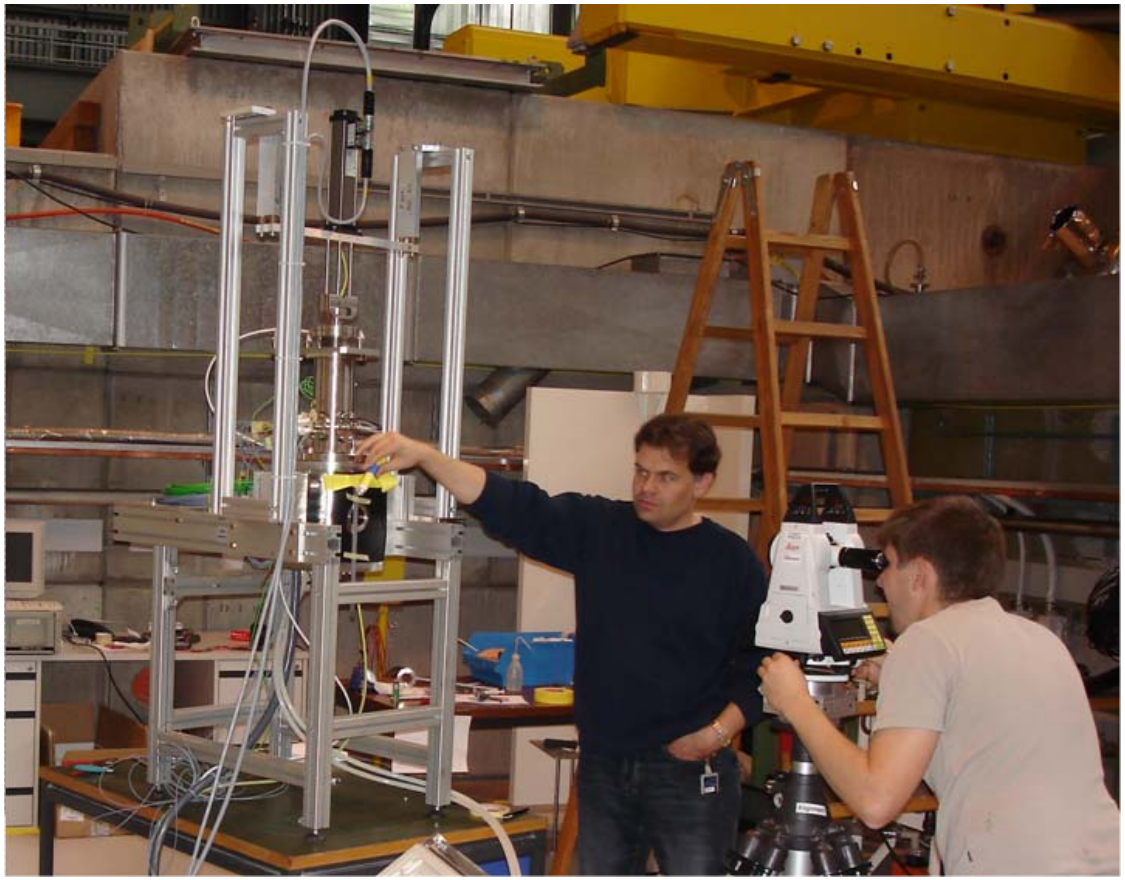
- **Dilatation of sample rod**
  - Max. (L1) = 2.5 mm
  - T_{graph} = 788 °C
  - T_{oven} = 1246 ° C
- **Water cooling**
  - 600 l/hr
  - ΔT = 0.6 °C
  - 430 W

Dry Tests: PSI

- Connectivity
- Leak/vacuum
- Thermocouples
- Oven
- Container position

Level 1 reference position





Power Supply				Level 1		Level 2		Level 3		Level 4		Graphite [°C]	Beam dump [°C]	Water in [°C]	Feedthrough [°C]
P [W]	I [A]	V_PS [V]	V_R [V]	Front [°C]	Back [°C]	Front [°C]	Back [°C]	Front [°C]	Back [°C]	Front [°C]	Back [°C]				
145	19.05	9	7.6	234	231	245	247	339		380	388	222	28	17.5	
269	24	12.85	11.2	325.7	317.4	352	353	478		542	544	309	34.6	17.5	70
344	26.5	15	13	363	353	397	400	537		610	615	342	40	17.5	83

Simulation Method 1/3

S. Marzari

- **3D modelisation in Autodesk Mechanical Desktop of a simplified geometry**

- **Transfer of this geometry to Ansys Workbench 10.0**

- **Completion of the geometry in the Design Modeler of Ansys (volumic surfaces)**

- **Two-step simulation (for each of the 4 levels)**
  - Step 1 for determination of the boundary conditions and structural deformations
  - Step 2 for determination of the temperatures inside the pills

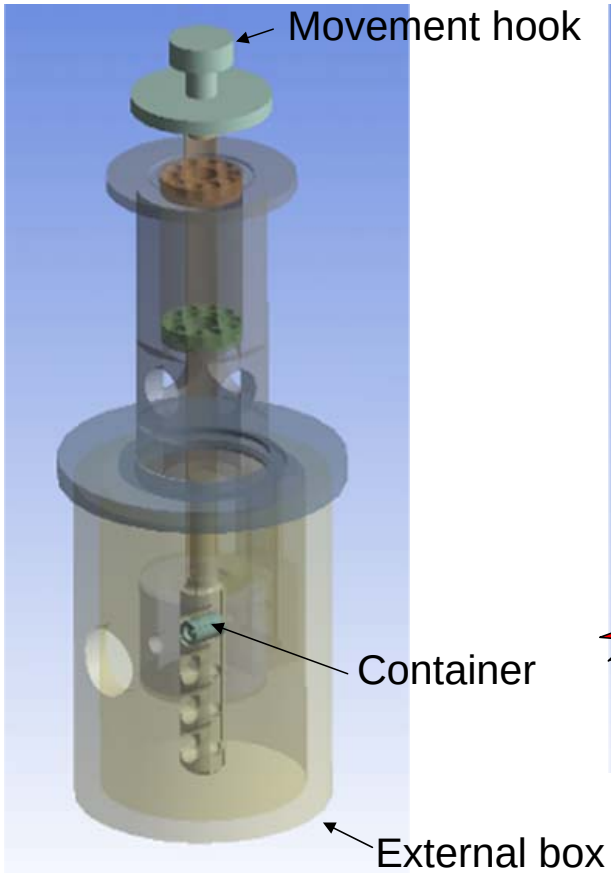
- **Optimisation**
  - Iteration between Ansys and Fluka
  - Material choice
  - Power optimisation

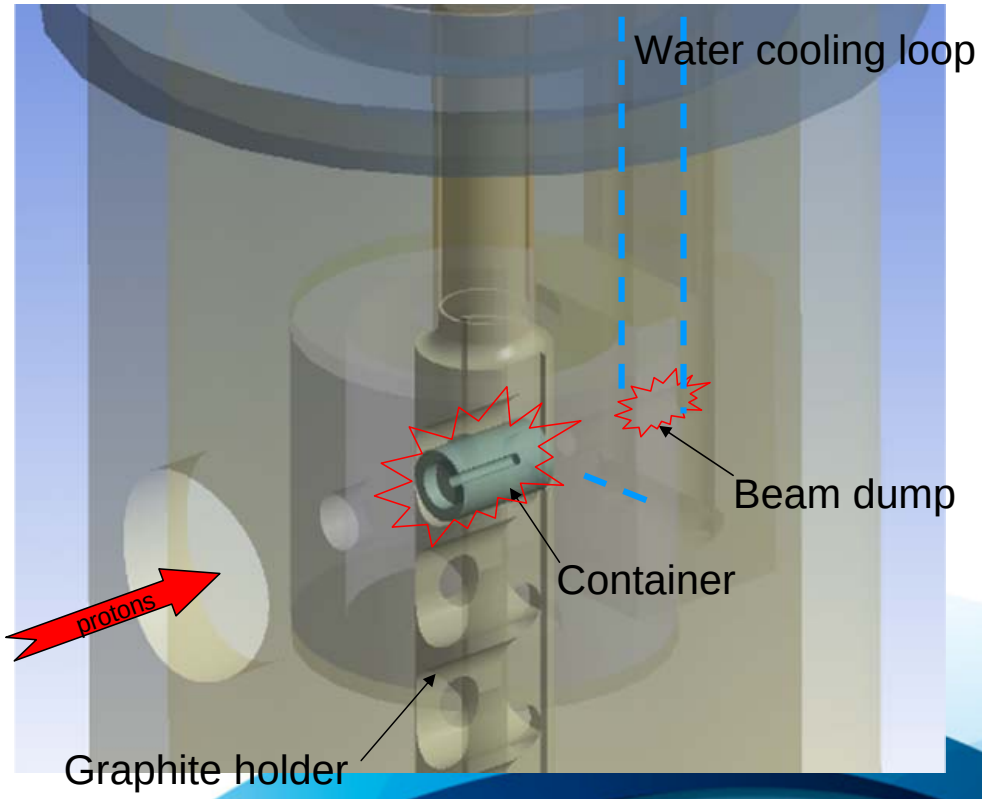
Simulation Method 2/3

S. Marzari

Step 1 simulation :

- Simplified internal shape of the container
- Used for definition of the boundary conditions for the detailed model (step 2)
- Determination of the dilatation movement of the hook
- Simplified geometry used outside the container (material, water cooling, radiation, convection etc…)



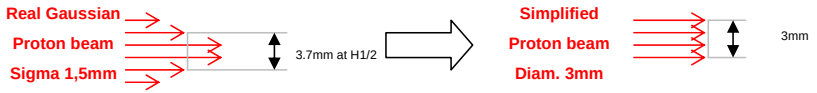


Simulation Method 3/3

S. Marzari

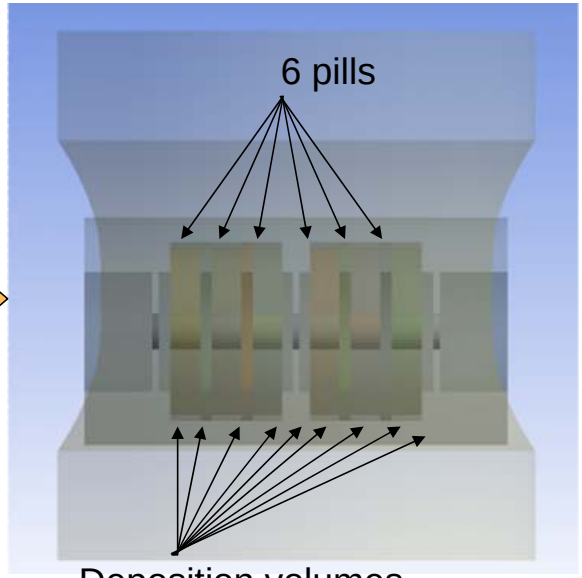
Step 2 simulation :

- Detailed internal shape of the container
- Used for determination of the temperatures reached for each pill or foil
- Real geometry used (materials, radiation, etc…)
- Assumptions:
  - uniform deposition on diam.3mm in the center of the pill or foil



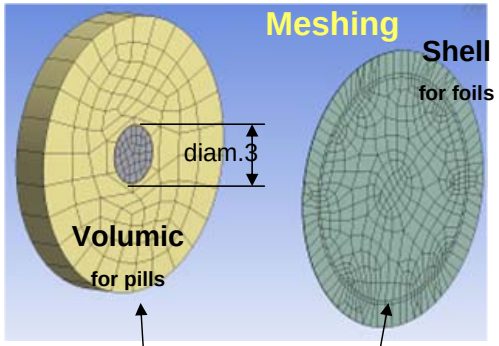
  - good thermal contact between pills, container and spacers,
  - no contact between container and graphite holder (only radiation)

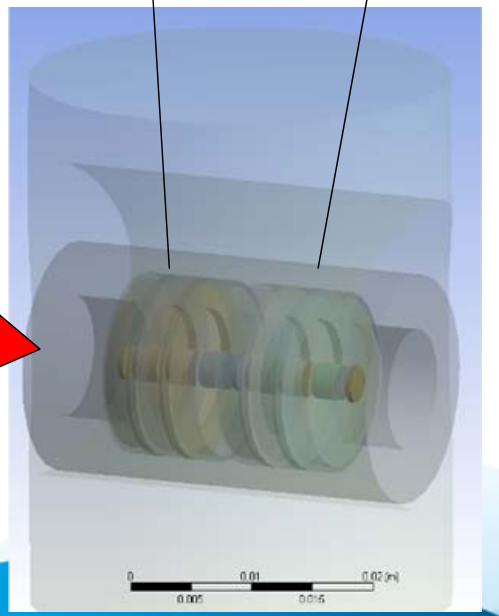
simplification



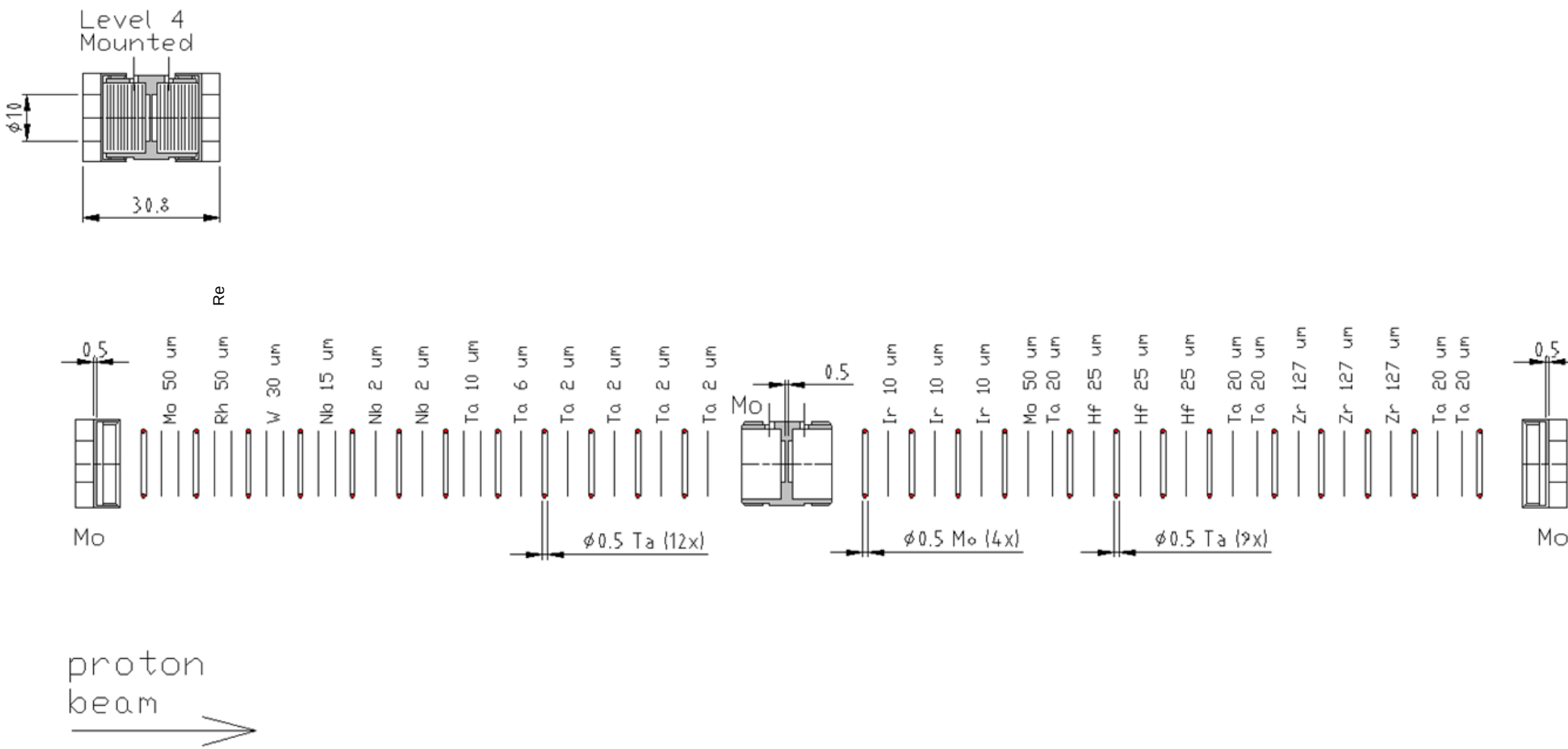
Deposition volumes
(pills or foils and walls)

protons





Level 4 Samples Simulation Input



Level 4 Simulation Data

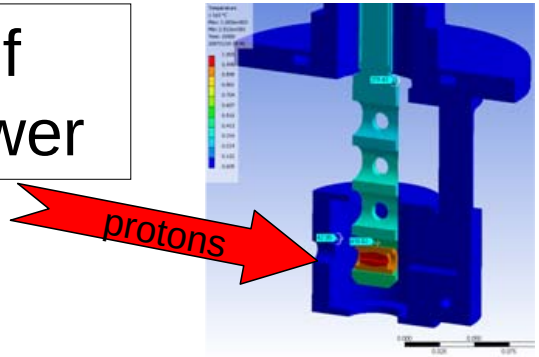
Environment:

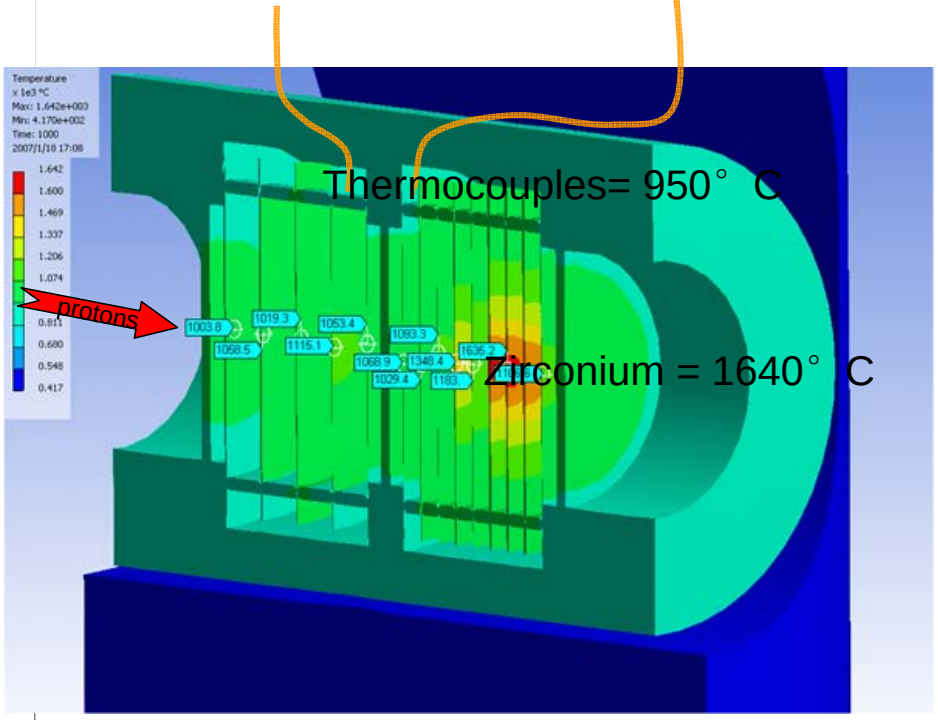
- Water cooling 38 ° C
- Power deposited in the container 97 W
- Power deposited in the beam dump 263 W

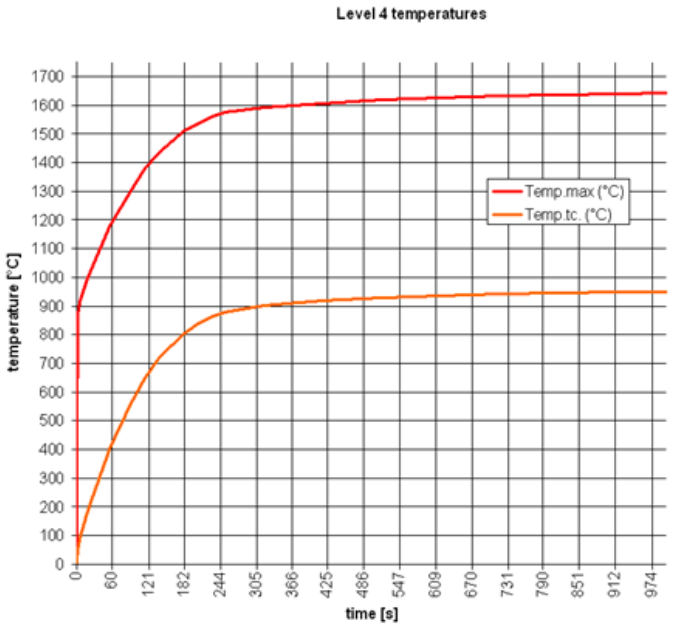
Results:

- Temperature of the surrounding graphite holder = 417° C
- Temperature of the surrounding beam dump = 42° C
- External hook temperature stabilized after 10000 sec.
- Temperature of the thermocouple graphite = 279° C
- Hook dilatation to be compensated = 0.85 mm

=> 20% of nominal power







Level 4 materials parameters & results

Total deposited power in the container =97 W, beam power 360 W (20%) calculated by FLUKA

Foil n°	Material	Thickness [mm]	Density [kg/m3]	Th. conductivity [W/mK]	Heat capacity [J/kgK]	Emissivity [-]	Heat dep. [W]	Temp. Results [° C]
wall	Mo	0.5	10200	142/105/88 (20/1000/1500° C)	250/310/440 (20/1000/1500° C)	0.3	16.3	1005 (2620)
1	Mo	0.100 (50+50um)	10200	142/105/88 (20/1000/1500° C)	250/310/440 (20/1000/1500° C)	0.3	3.2	1004 (2620)
2	Rh	0.100 (50+50um)	12400	150	244	0.15	6.3	1059 (1965)
3	W	0.060 (30+30um)	19300	170	133	0.18	2.8	1020 (3370)
4	Nb	0.034 (15+15+2+2um)	8570	53.7	268	0.15	1.1	1115 (2468)
5	Ta	0.034 (10+10+6+2+2+2+2 um)	16600	54.4/ 74 (20/1527° C)	140	0.2	1.2	1055 (3000)
wall	Mo	0.5	10200	142/105/88 (20/1000/1500° C)	250/310/440 (20/1000/1500° C)	0.3	14.5	1014 (2620)
6	Ir	0.030 (10+10+10um)	22400	147	133	0.3	2.4	1069 (2410)
7	Mo	0.050	10200	142/105/88 (20/1000/1500° C)	250/310/440 (20/1000/1500° C)	0.3	1.6	1029 (2620)
8	Ta	0.020	16600	54.4/ 74 (20/1527° C)	140	0.2	0.7	1094 (3000)

Level 4 materials parameters & results

Foil n°	Material (+density)	Thickness [mm]	Density [kg/m3]	Th. conductivity [W/mK]	Heat capacity [J/kgK]	Emissivity [-]	Heat dep. [W]	Temp. Results [° C]
9	Hf	0.075 (24+25+25um)	13100	23	146	0.25	3.2	1348 (2227)
10	Ta	0.040 (20+20um)	16600	54.4/ 74 (20/1527° C)	140	0.2	1.8	1183 (3000)
11	Zr	0.127	6490	22.7	281	0.3	8.3	1615 (1852)
12	Zr	0.127	6490	22.7	281	0.3	8.3	1642 (1852)
13	Zr	0.127	6490	22.7	281	0.3	8.3	1605 (1852)
14	Ta	0.040 (20+20um)	16600	54.4/ 74 (20/1527° C)	140	0.2	1.5	1106 (3000)
wall	Mo	0.5	10200	142/105/88 (20/1000/1500° C)	250/310/440 (20/1000/1500° C)	0.3	15.8	1014 (2620)

Beam Readiness

Safety thresholds determined from simulations for controls/interlocks

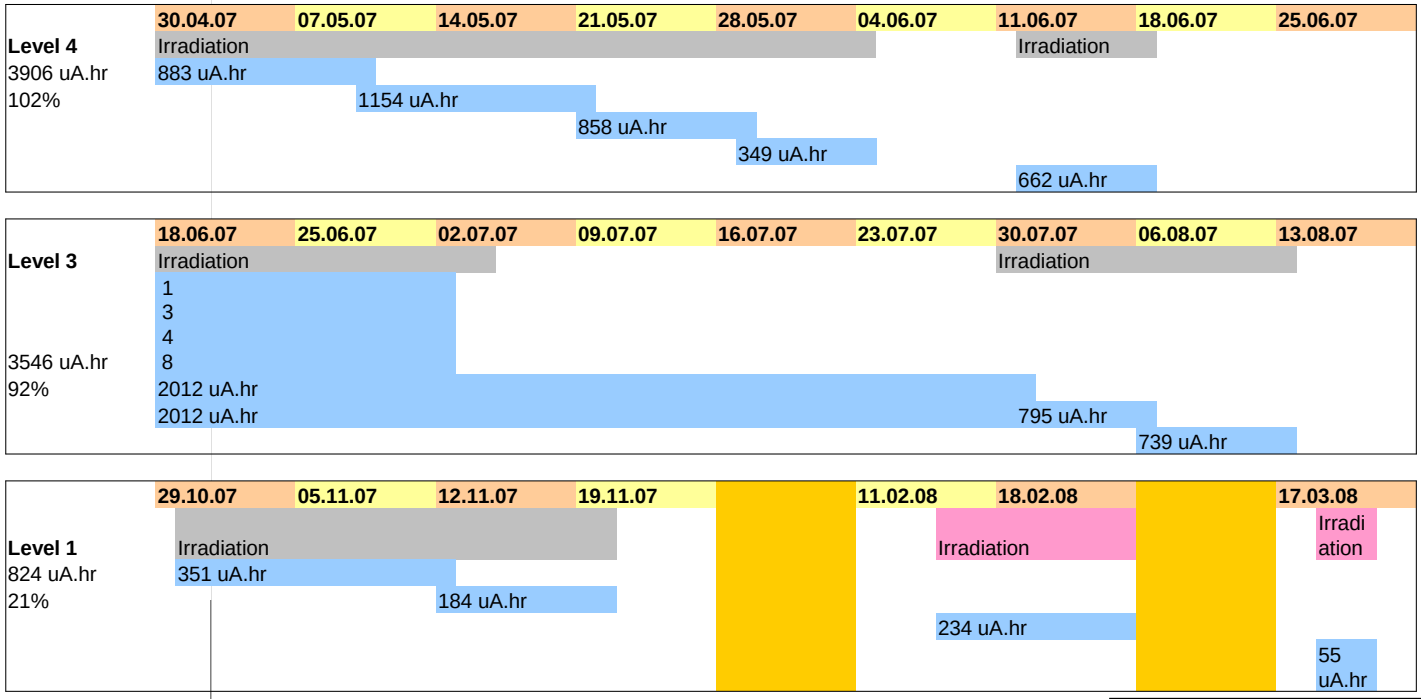
Level	Beam			Target container		
	I [μA]	P [W]	% of nominal (nom = 30 μA)	P_{dep} [W]	Hot spot [°C]	T.c. value [°C]
1	1.25	90	5	53	1432	786
2	3.75	270	15	52	1476	745
3	10	720	40	153	1630	820
4	5	360	20	97	1640	950

Documentation for B.A.G. (Swiss Federal Office of Public Health)

Referenz/Aktenzeichen:
Ihr Zeichen:
Unser Zeichen: DMU / SNI
Liebefeld, 25. April 2007

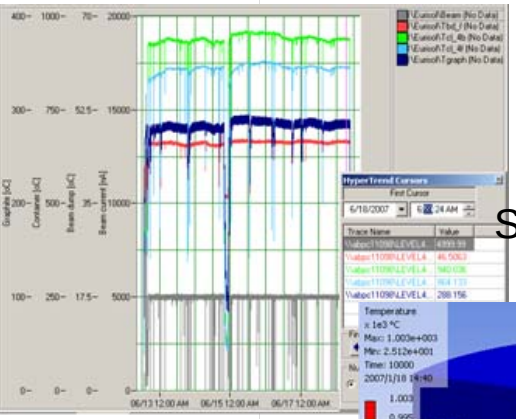
EURISOL: Strahlfreigabe

Irradiation Data

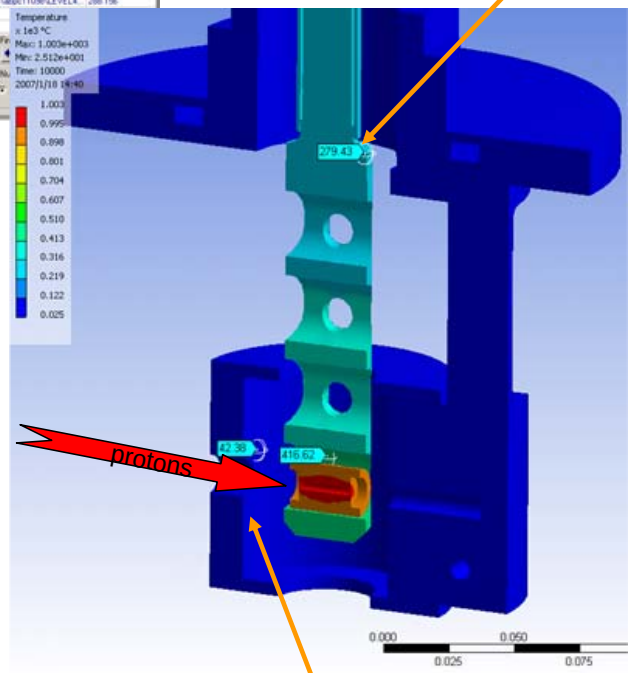


Irradiation days in 2007	< 96 days
Maximum beam current used	6.5 μA (level 3)
Highest bunker gamma level	5.7 $mSv.hr^{-1}$ (level 3)
Post-irrad. bunker gamma level [07.12.07]	4.5 $mSv.hr^{-1}$
Highest vacuum reading	1.7×10^{-5} mbar (level 1)
Highest graphite temperature	320°C (level 1)

Level 4: Online vs. Simulations

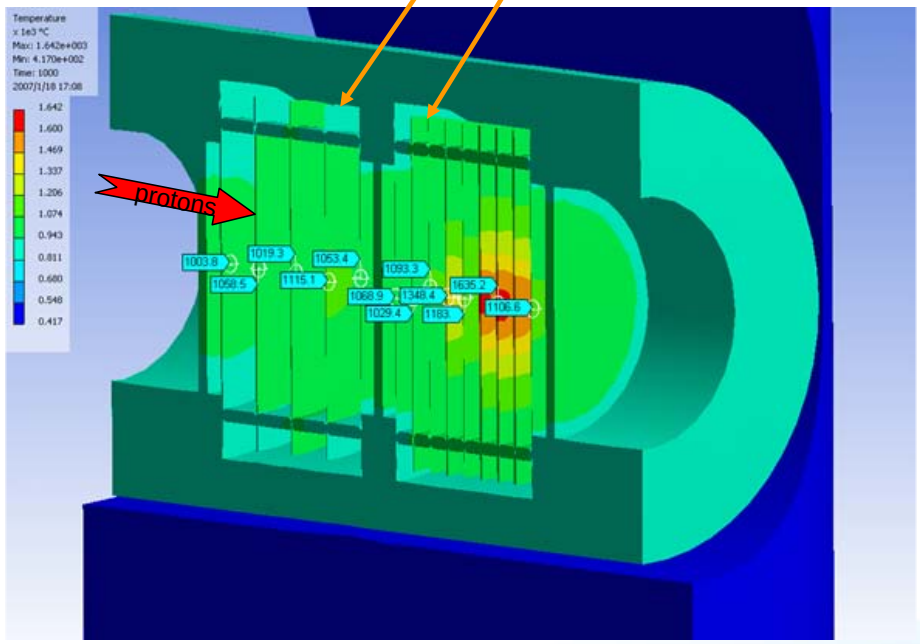


Simulated 279° C – Measured 275° C



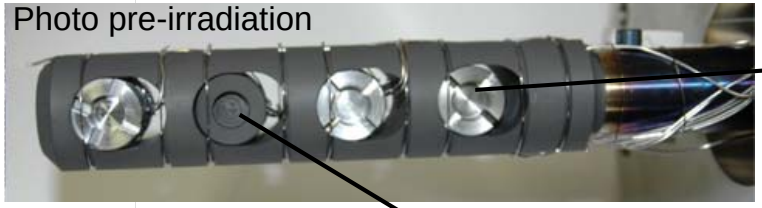
Simulated 42° C – Measured 48° C

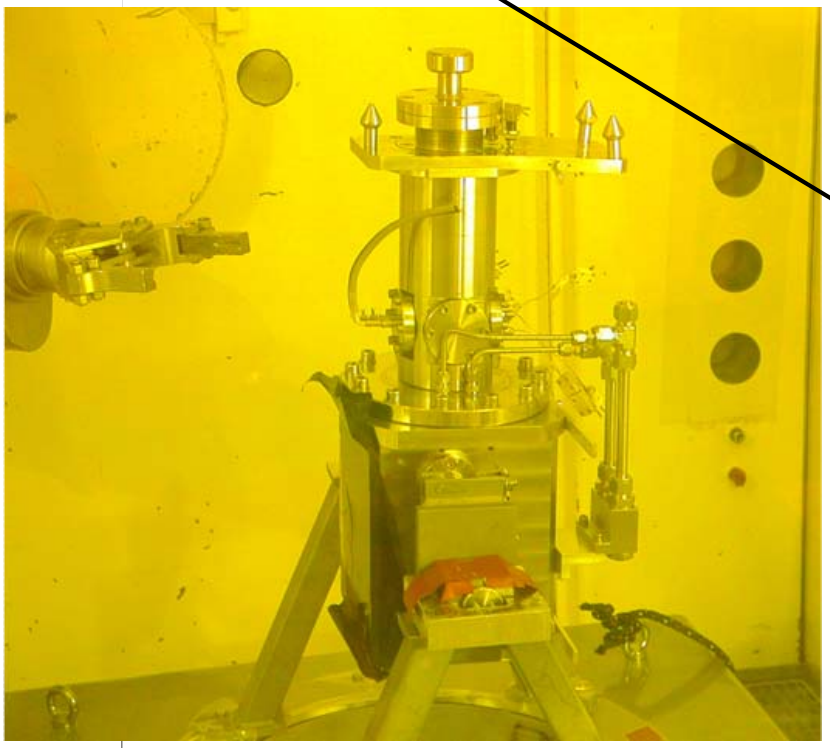
Simulated 950° C – Measured 870° C & 930° C

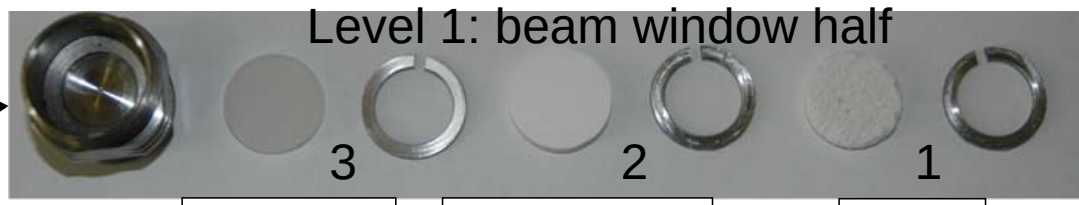


Post-Irradiation Phase

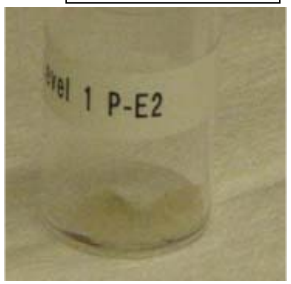
Photo pre-irradiation

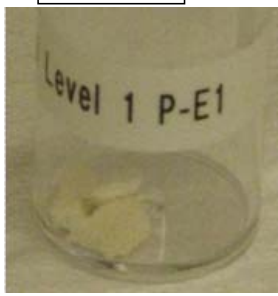


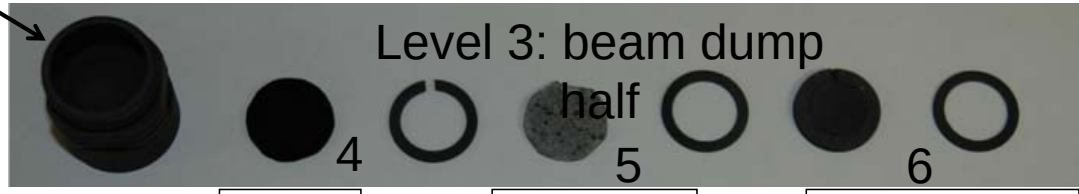




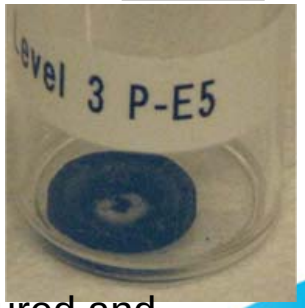
Al_2O_3 transparent | Al_2O_3 pressed powders | Al_2O_3 foam

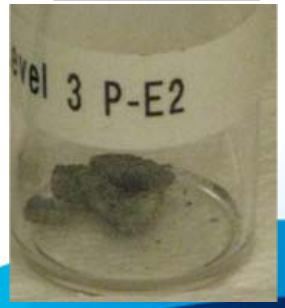


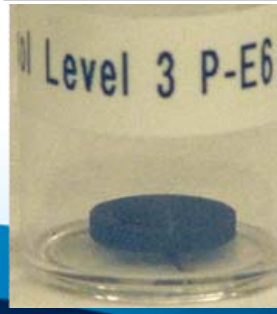




Nanotubes | Nanotubes + SiC | Nanofibres in C capsule

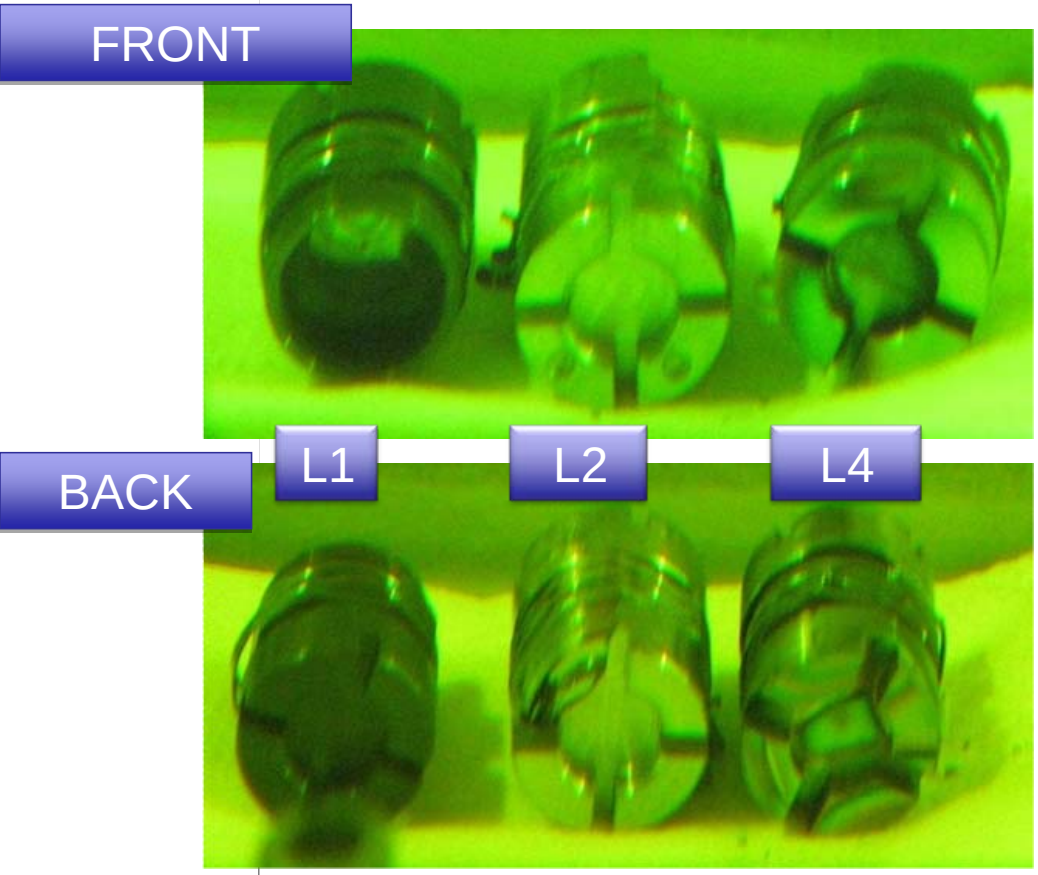




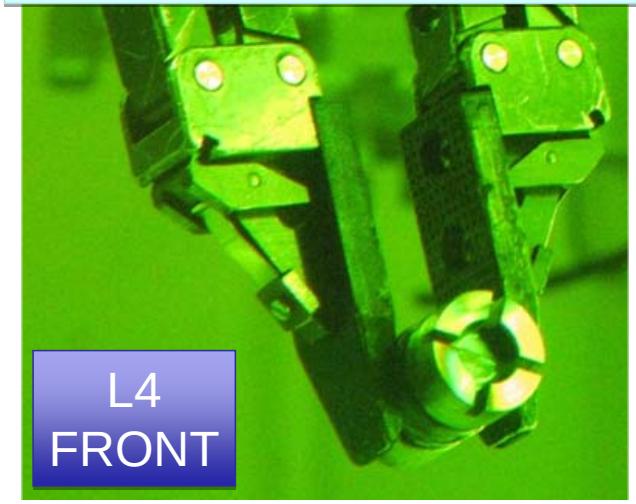


S. Fernandes et al. , Microstructure evolution of nanostructured and submicrometric porous refractory ceramics induced by a continuous high-energy proton beam, Proceedings of E-MRS 2010 Spring Meeting.

Level 4: Sample Identification



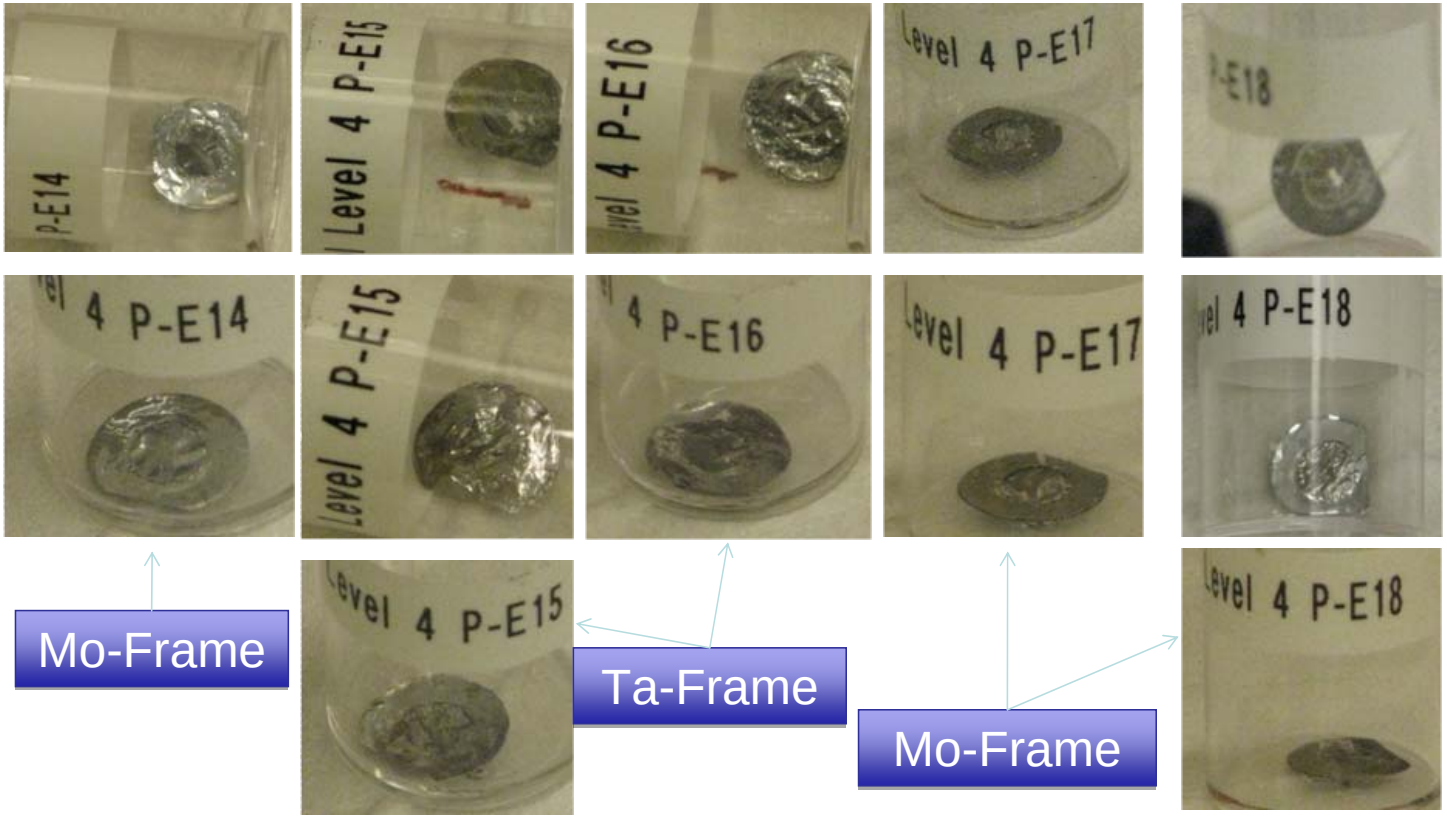
- Level 4 samples initially identified back-to-front
  - Either the samples were loaded incorrectly at CERN pre-irradiation.
  - Or what was thought to be the front-half of Level4 was actually the back-half when the samples were extracted post-irradiation in the PSI hot-cell.
  - Samples Identified with combination of visual inspection, gamma spectra, weight information and initial SEM/EDX data.



Photos by V. Boutellier

Tantalum foils: 2-6 μm

During assembly, fragile foils were spot-welded to supporting Mo or Ta frames



6 μm 2 μm 2 μm 2 μm 2 μm

L4 Gamma Spectra: Different frames

^{98}Mo frame

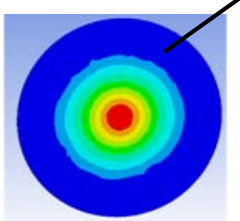
P-E14	Co-58	1.45E+04	34
	Y-88	5.69E+06	2.6
	Zr-88	1.21E+06	4.5
	Nb-95	3.09E+04	17.3
	Tc-95m	3.28E+05	2.5
	Hf-172	5.88E+04	47.4
	Lu-172	2.63E+05	5.8
	Lu-173	5.46E+06	2.5
	Os-182	6.77E+07	6.1
	Re-182	2.07E+09	10.1
	Re-182m	9.03E+05	9.2
P-E17	Y-88	5.31E+06	2.6
	Zr-88	1.14E+06	4.5
	Nb-95	2.66E+04	23.3
	Tc-95m	3.05E+05	2.4
	Lu-173	1.74E+06	3.1
	Os-182	3.34E+07	13.6
	Re-182	6.93E+08	18.8
	Re-182m	2.63E+05	27.9
P-E18	Y-88	5.28E+06	2.6
	Zr-88	1.12E+06	4.5
	Nb-95	3.45E+04	18.1
	Tc-95m	2.95E+05	2.5
	Lu-173	1.41E+06	3.3
	Os-182	1.03E+07	31.4
	Re-182	1.14E+09	12.8

^{181}Ta frame

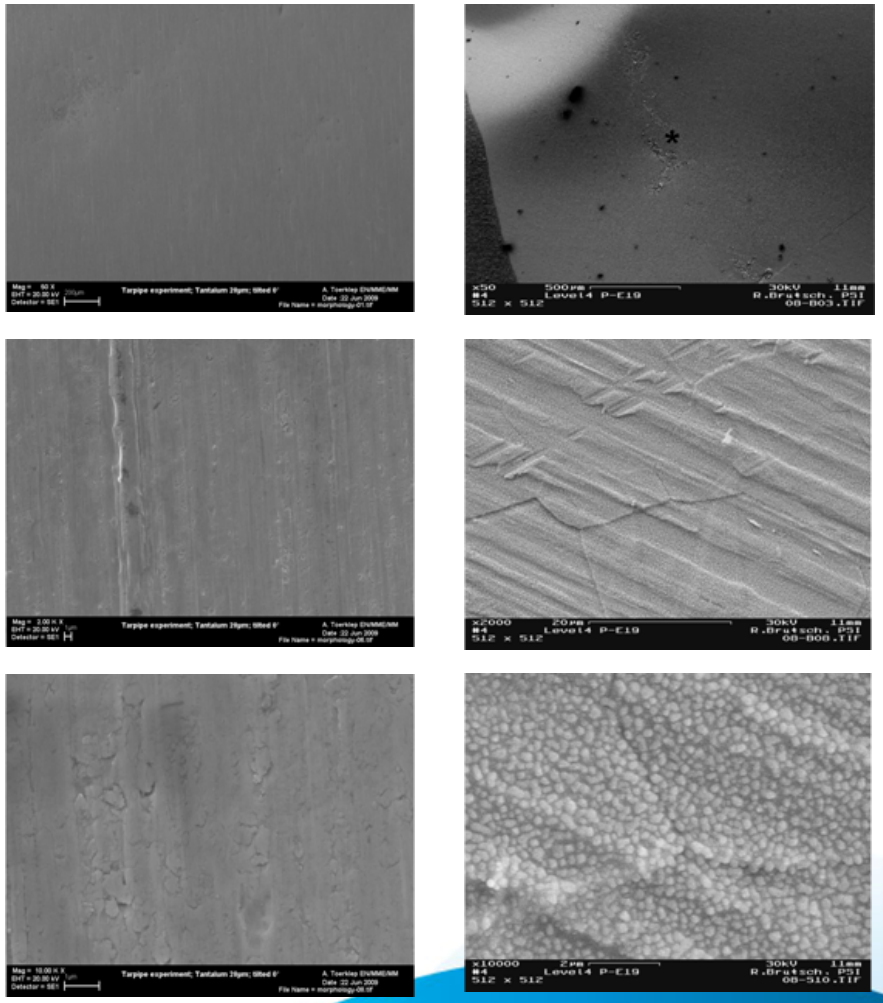
P-E16	Na-22	9.31E+03	5.3
	Co-57	4.74E+03	23.3
	Co-58	2.02E+03	58.2
	Y-88	1.95E+04	3.8
	Zr-88	4.02E+03	16.3
	Gd-153	2.84E+04	16
	Hf-172	6.64E+04	9.7
	Lu-172	1.08E+05	2
	Lu-173	5.40E+06	2.2
	Lu-177m	6.51E+03	14.8
	Os-182	1.32E+06	1.9
	Re-182	2.83E+06	3.2
	Re-182m	3.06E+05	2.2
	Ta-182	8.39E+05	1.6
	Ir-184	1.18E+10	4.5
	Pt-191	4.07E+04	26.2
	Tl-197	5.19E+09	43.4
P-E15	Na-22	8.04E+03	5.9
	Co-58	3.18E+03	41.9
	Y-88	8.54E+03	5.4
	Hf-172	7.78E+04	10.5
	Lu-172	1.01E+05	1.9
	Lu-173	6.07E+06	2.2
	Lu-177m	1.34E+04	15.9
	Hf-182m	1.19E+06	15.6
	Os-182	3.12E+05	1.9
	Re-182	2.05E+05	3.5
	Re-182m	2.02E+05	2.2
	Ta-182	8.62E+05	1.6
	Ir-184	2.14E+05	4.6
	Pt-191	1.99E+04	36.7
	Tl-197	7.57E+04	26.1

Tantalum samples

- Tantalum foils, 99.9% from Goodfellow
- Thickness 20 μm ± 20%
- Impurities (ppm max.) Al: 5, Ca: 2, Co: 1, Cu: 2, Ti: 20, V: 5, Cr: 5, Mn: 2, Fe: 30, Ni: 3, Cu:1, Zn:1, Sn: 2, W: 100, Zr:10, Si: 10, Nb < 500, Na: 10, Mo:100 and Mg: 5.
- Temperature during irradiation: 920° C.
- Recrystallisation occurred during irradiation (was not observed on reference sample heated for 2 hrs at 1100° C).

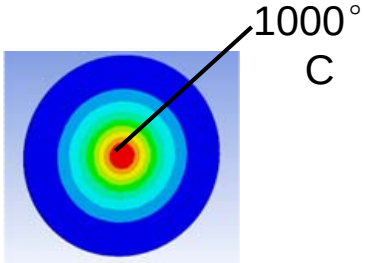


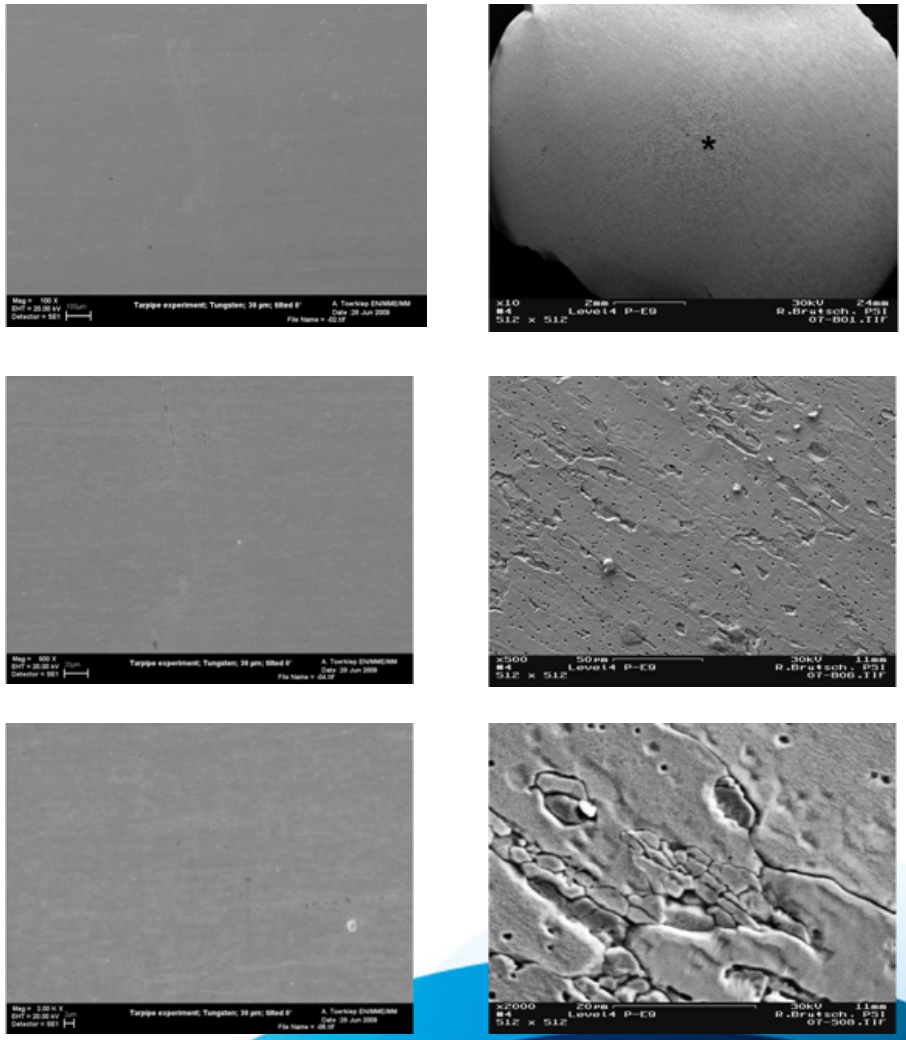
Before Irradiation | After Irradiation



Tungsten Samples

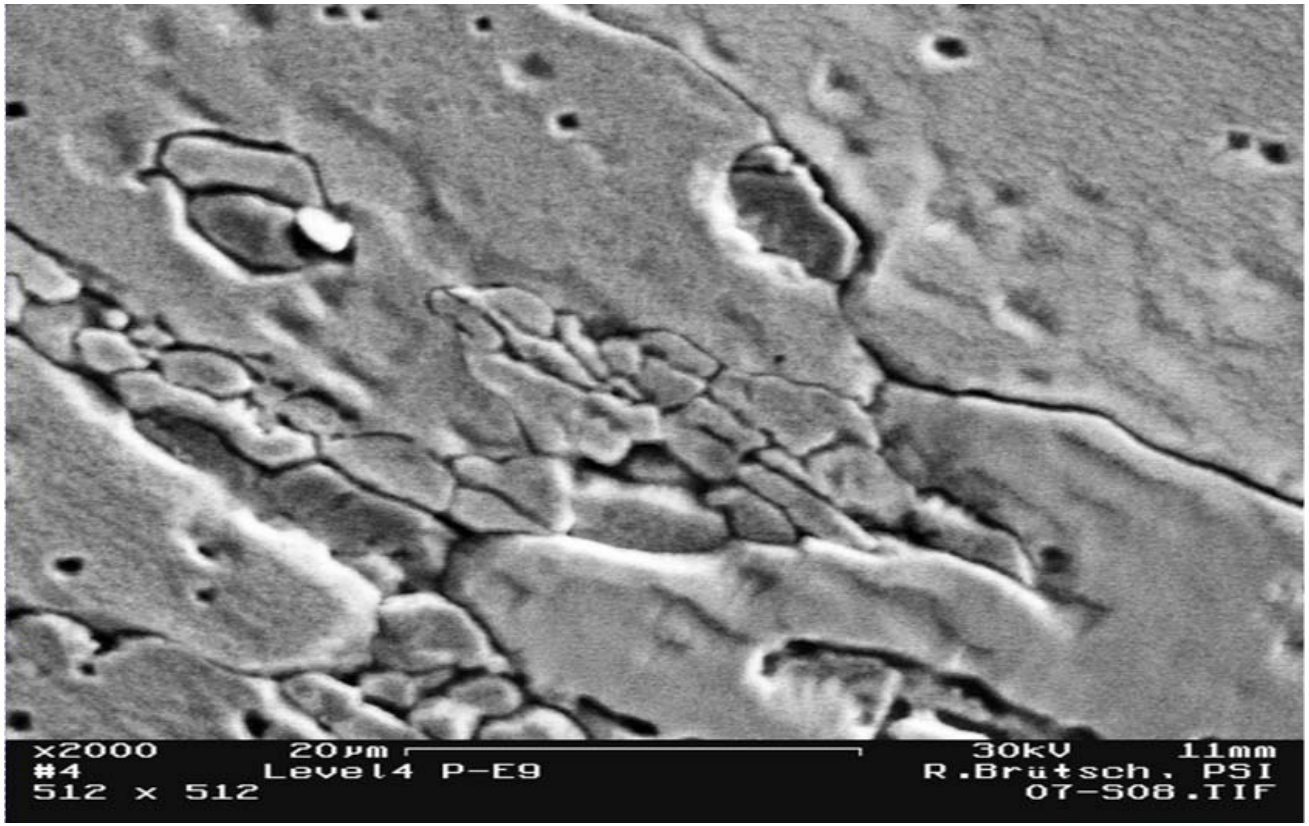
- Tungsten foils, 99.97% from Plansee
- Thickness 30 μm ± 20%
- Impurities (ppm max.) Ag: 10, Al:15, As: 5, Ba: 5, Ca: 5, Cd:5, Co: 10, Cr:20, Cu:10, Fe:30, K: 10, Mg: 5, Mn: 5, Na: 10, Nb: 10, Ni: 5, Pb: 5, Ta: 20, Ti: 5, Zn: 5, Zr: 5 and Mo: 100.
- Pronounced fibrous structure due to orientation of the grains from rolling.
- Temperature during irradiation: 1000° C.





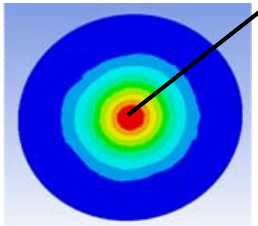
Tungsten Samples

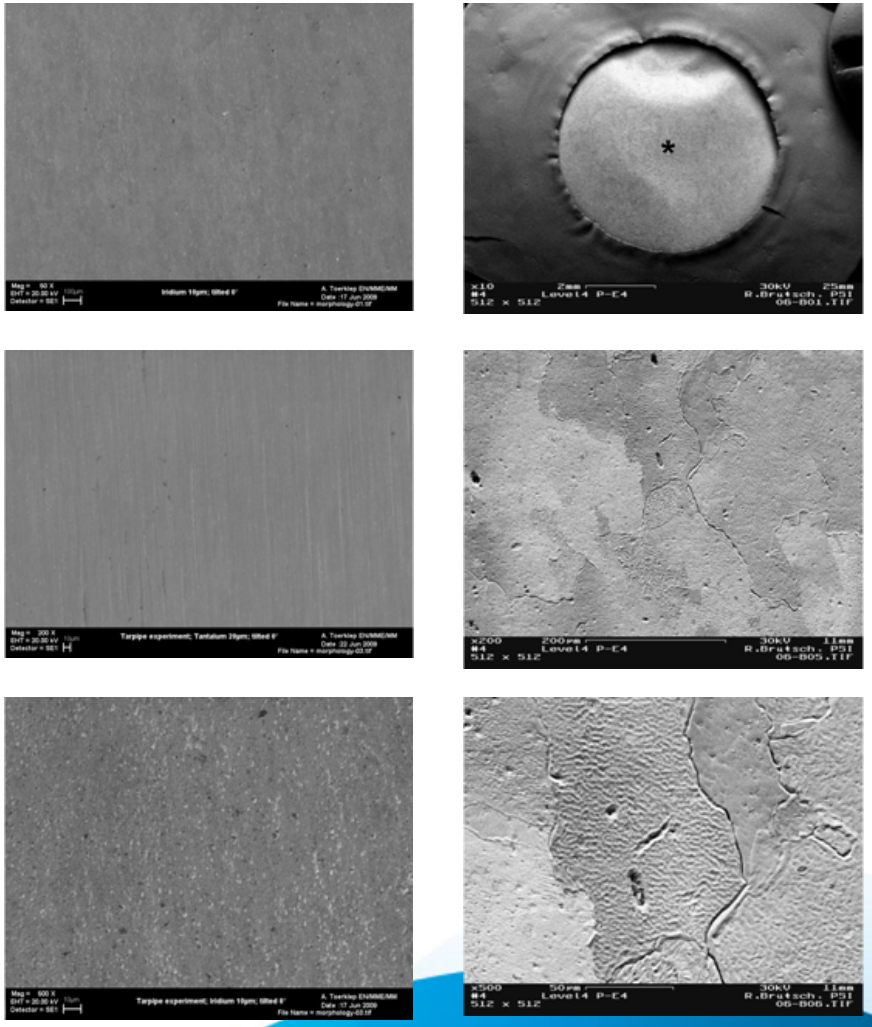
- Formation of 1 – 2 μm voids, grain growth and beginning of recrystallisation by seeding (small grains either in middle or at boundaries).
- Recrystallisation temperature of as received sample is 1150° C, lower under vacuum (- 50° C).
- Partial recrystallisation observed on non-irradiated sample at 1100° C for 2 hrs at P < 5 10^{-5} mbar.



Iridium Samples

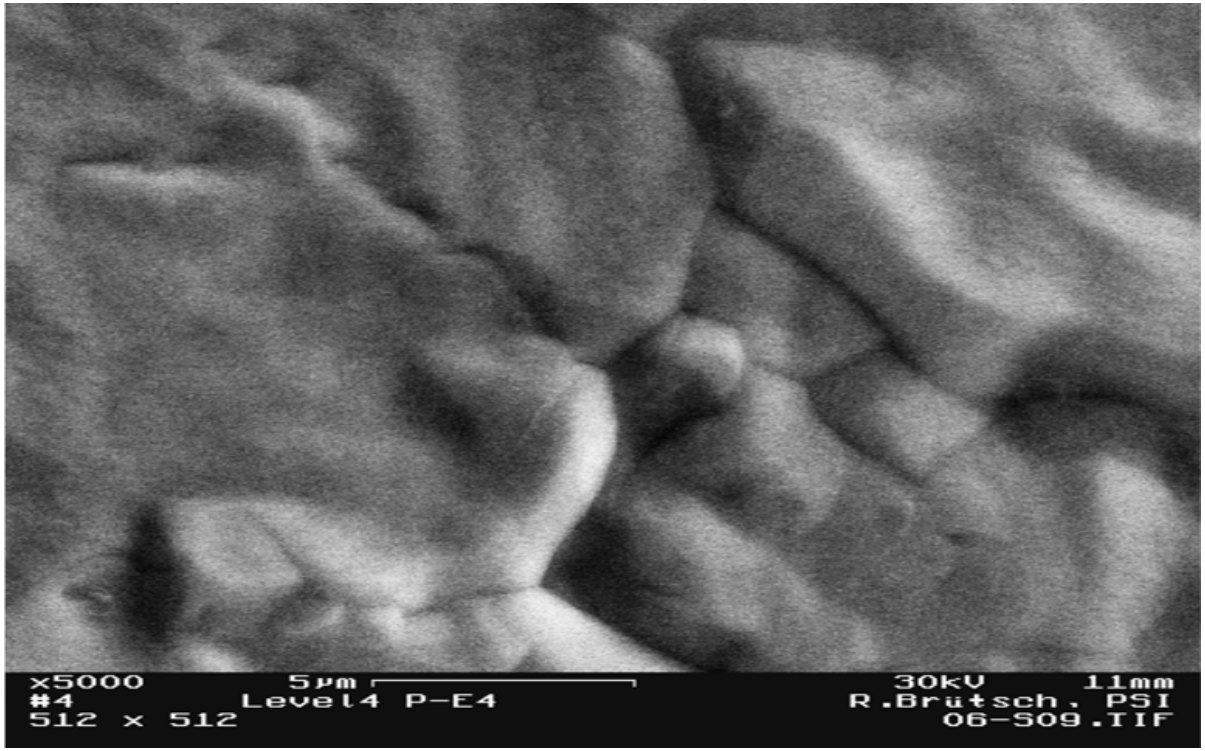
- Three samples L4P-E2,3,4 showed similar behaviour.
- Iridium foils, 99.9% from Goodfellow
- Thickness 10 μm ± 20%
- Impurities (ppm max.) Al: 5, Mg:1, Ba: 10, Ca: 30, Cu: 1, Fe: 30, Ni: 3, Cu:1, Si: 20, Na: 20, Mg: 1, Pb: 30, Pd:20, Rh: 200, Ru: 5.
- Temperature during irradiation: 1050° C.





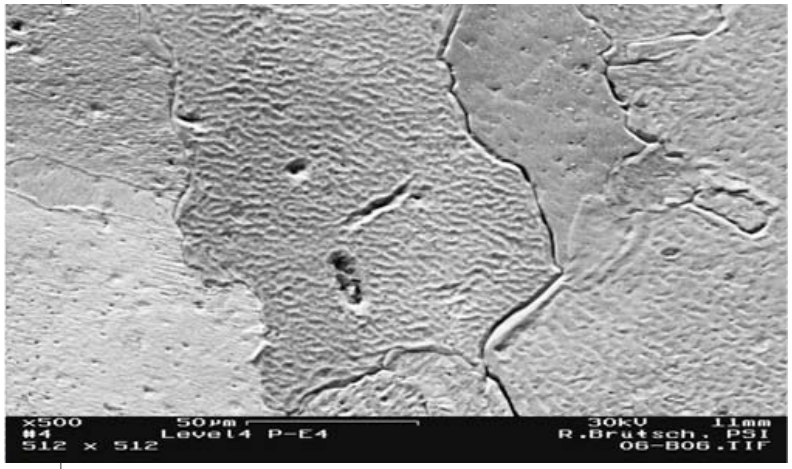
Iridium Samples

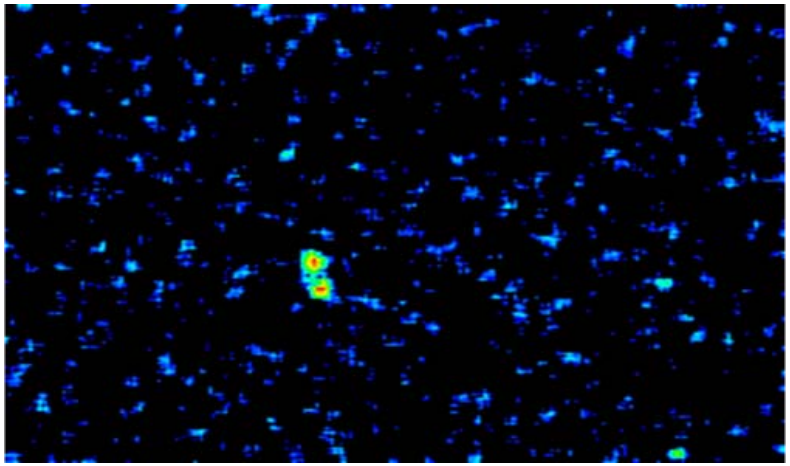
> Grain growth and increase in thickness at grain boundaries show recrystallisation during irradiation at high temperature.



Iridium Samples

> Oxygen inclusion observed





Summary

- Oxide, carbide and metal foil samples irradiated at high temperature to observe microstructure evolution through TARPIPE irradiation programme.
- Results on the ceramics are the subject of an earlier paper presented at the European Materials Research Society Spring Meeting 2010.
- Foils:
  - Ta, W and Ir operated at temperatures where creep deformation noticeable (30% of M.P. 905° C, 1027° C, 740° C).
  - Tantalum: Visible holes in 2 μm foils, grain growth ~ 20 μm under irradiation at high temperature (> 900° C). Places a limit on the minimum foil thickness.
  - Tungsten: Formation of 1-2 μm microvoids, recrystallisation by seeding under irradiation at high temperature (> 1000° C).
  - Iridium: grain growth and increase of thickness at grain boundaries.

Proton fluences (backup)

Parameter	Units	All	Zone 1	Zone 2	Zone 3	Zone 4	Zone 5
Level 4							
Level 4 current	protons/s	3.12E+13					
Irradiation time	hr	781					
Integrated protons	protons	8.78E+19					
Total protons/s	protons/s		2.13E+13	8.48E+12	1.34E+12	8.24E+10	1.87E+09
Proton flux	protons/cm^2/s		4.71E+14	6.25E+13	5.91E+12	2.60E+11	4.60E+09
	protons/cm^2		1.32E+21	1.76E+20	1.66E+19	7.32E+17	1.29E+16
Integrated protons	protons		5.99E+19	2.39E+19	3.76E+18	2.32E+17	5.27E+15