First Workshop on Actinide Target Development

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RADIATION PROTECTION ISSUES WITH ACTINIDE TARGETS AT ISOLDE

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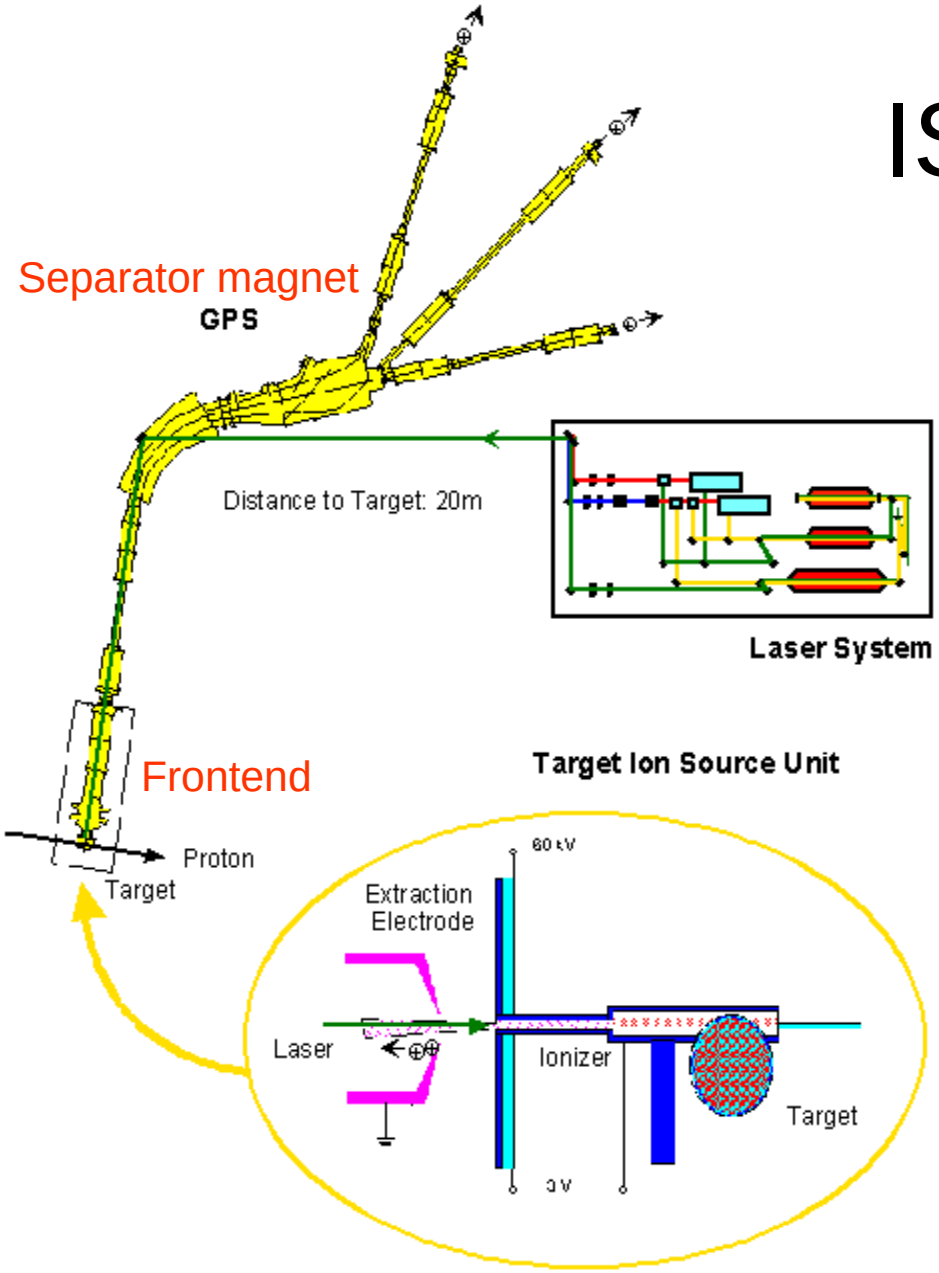
ISOLDE operation



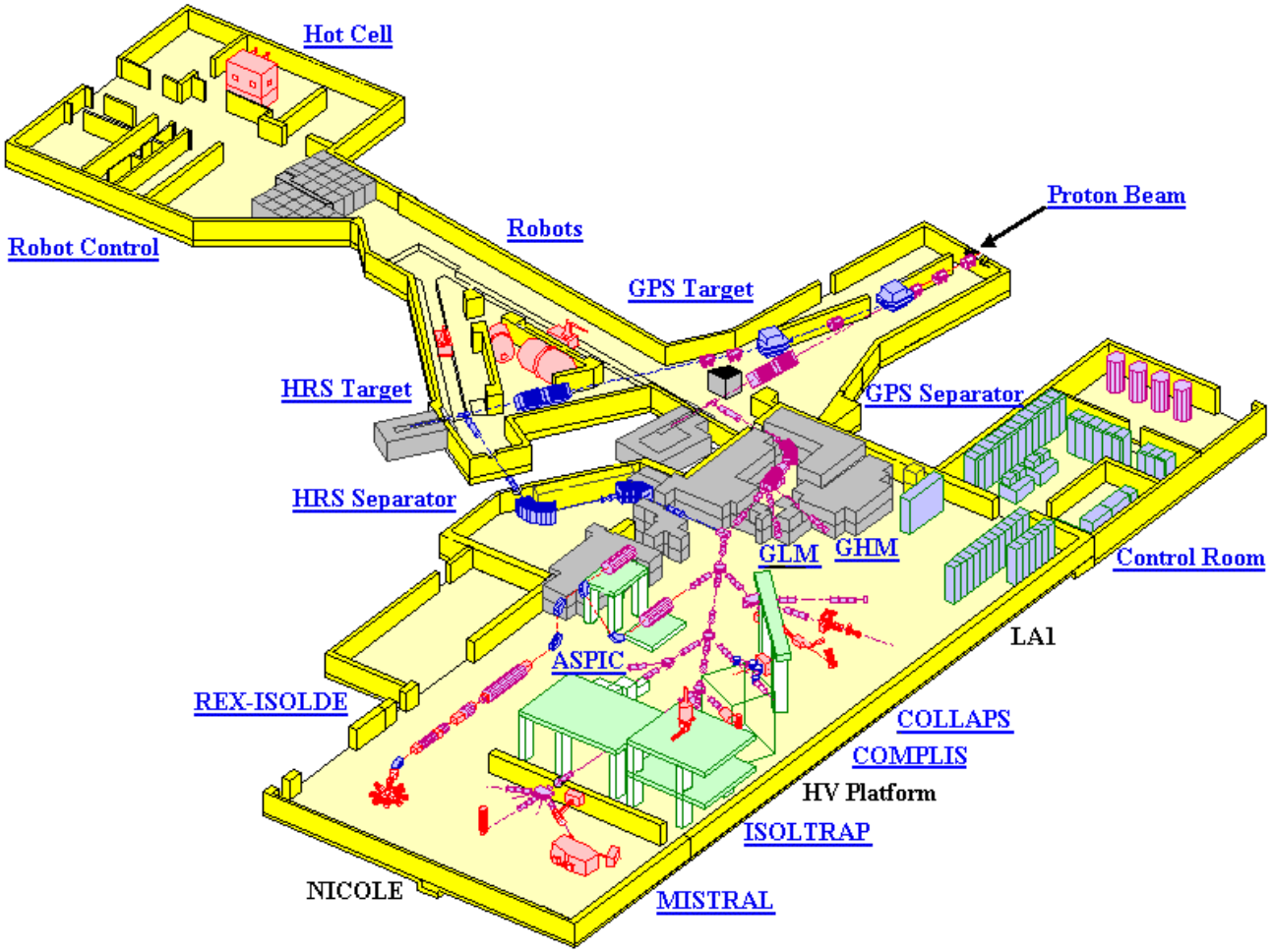
- Bombardment of heavy targets with 2 μA protons, *E*=1.4 GeV
- Production of radioisotopes by spallation, fragmentation or fission
- Ionisation of isotopes
- Mass separation
- Contamination of the vacuum system

Controlled areas

Access control, safety chain with interlocks « beamcuts »

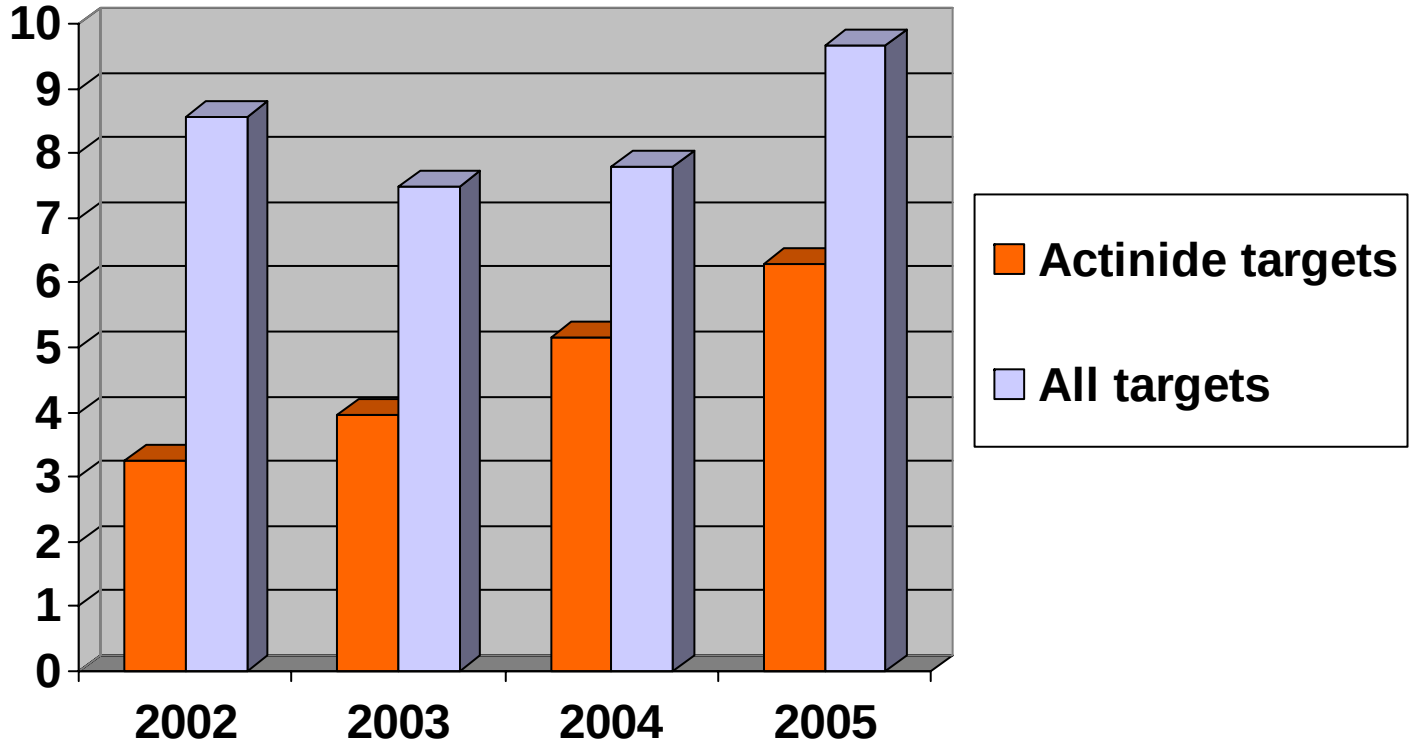
- Target area
  - Primary zone = forbidden area closed by a shielded door (120 tons) during operation (Monitor « BLM » at 2.3 m from target indicates 12 Gy/h for U-runs…)
  - Highly radioactive area during shutdown period (induced activity in faraday cages still > 2 mSv/h after 3 months of decay)
- Separators area
  - Primary zone = forbidden area during operation (ambient gamma doserate exceeds 100 mSv/h during U-runs)
  - Limited stay area during shutdown period (between 10 uSv/h and 2 mSv/h)
- Experimental hall
  - Simple controlled area (< 10 uSv/h) except on-line isotopes collections points (limited stay area)
- Laboratories
  - Class A work sector for unsealed radioactive sources handling (actinide targets production, autopsy, vacuum and beam diagnostic maintenance, offline operations…)

Facility layout



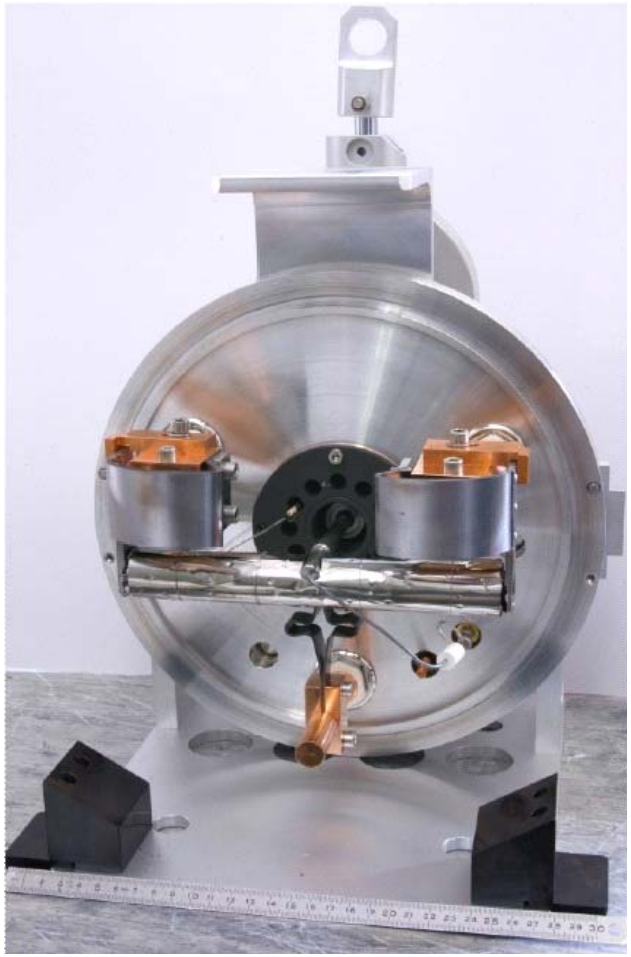
Protons and targets

Yearly total protons ($1^{E}19$) taken on both separators



Actinide target production

Risk of incorporation



Handling of UO_2 and ThO_2 (powder) requires a license and a class A work sector (use of unsealed radioactive sources exceeding 10000 times the limit of authorization)

Concentration (Bq/m^3) and relative exposure ($Bq \cdot h/m^3$) must be accurately measured and compared to thresholds of the Swiss Ordinance (dosimetry)

Measurement of incorporation must be done if the threshold of relative exposure is exceeded

150 $Bq \cdot h/m^3$ for U-238 (for 1 year)

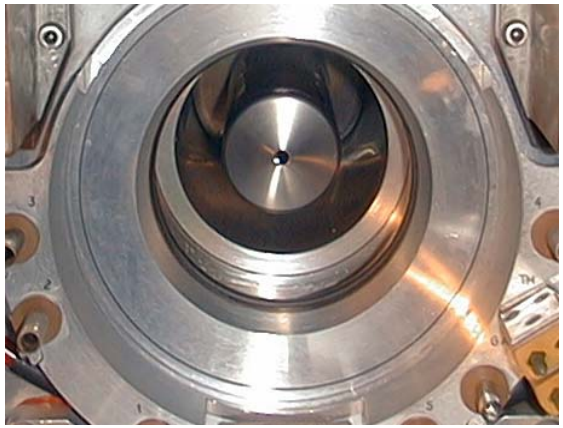
All steps in production (mix with C, pills and carburization) involve the risk of incorporation and require protective clothes, work methods and a close radiation protection survey

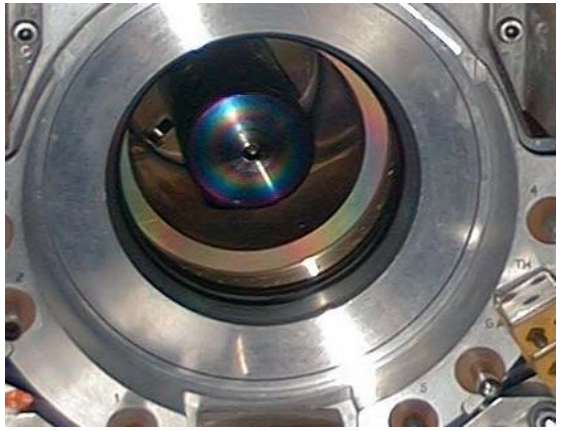
Migration of the radioactive contamination

- Interior of targets: refractory elements
- On the extraction electrode
- Focal plane of separator: not-selected isotopes (protected by removable covers).
- Around the slit of the switchyards and the beam diagnostic equipments
- Noble gases: virtually everywhere in the vacuum system by diffusion

Contamination of the extraction electrode

α-emitters and β/γ doserates





- Identified α-activity measured on the extraction electrode after 2 years of radioactive cooling

Isotope	$T_{1/2}$ (years)	Electrode (Bq)	Smear of 1 cm^2 on the electrode (Bq)
Gd-148	74.6	~ 100 000	~ 40
Po-208	2.9	~ 100 000	~ 92
Po-209	102	-	-
Po-210	0.38	~ 250 000	~ 230
Ac-227	21.77	~ 5	~ 4
Ra/Th-228	5.75	~ 10	~ 10

- Very huge level of contamination implanted around the hole (>>> Ti activation products)
  - **$H_{0.07}$ up to 7 Sv/h @ contact**
  - **H*(10) up to 20 mSv/h @ contact**

Measurements carried out 100 days after the run…

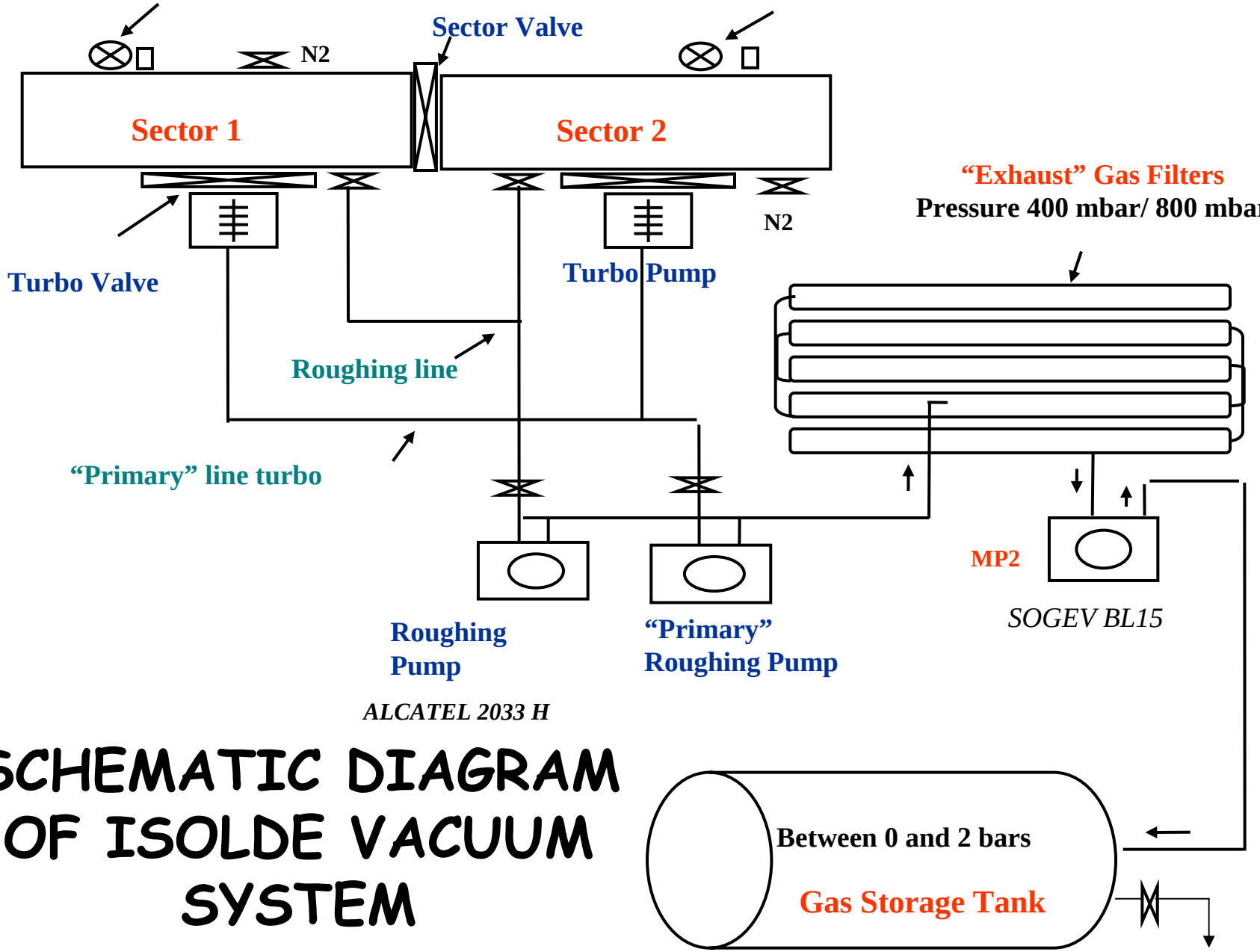
Isolde vacuum system

- A high vacuum system
- Pressure range between 1 and 10 μPa
- Use of *OIL ROUGHING PUMPS* and *TURBOMOLECULAR PUMPS*
- Use *EPDM* and *VITON O-RINGS* (NO METAL SEAL)
- No BAKE-OUT system
- Entirely controlled automatically by a SIEMENS PLC system

Roughing pumps and gas filters

- **Isotopes going out of the target without being ionized will:**
  - **Become solid (temperature loss or radioactive decay) and be deposited on vacuum chambers and won't pass through the barrier of the turbo molecular pumps**
  - **Stay in gaseous state and be pumped by the turbo molecular pumps. Some inert gases like Radon may reach the experiments in the hall → contamination check for any vacuum chamber opening!**

SCHEMATIC DIAGRAM OF ISOLDE VACUUM SYSTEM



Contamination of the vacuum equipments

- Volatile elements can be pumped and contaminate the vacuum system
- There they may decay into non-volatile elements and attach to walls

  $\Rightarrow$ Contamination found in

  – Turbomolecular pumps

  – Roughing pumps oil

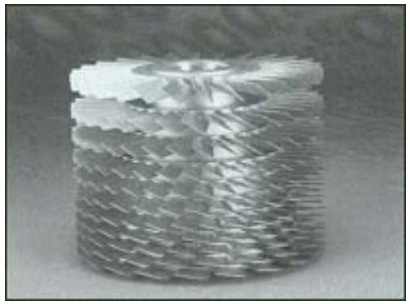
  – Gas storage tanks

Contamination of turbo molecular pumps

- Frontend TPs ensure the elimination of most of the radioactive gases leaving the target (~95%)
- Due to the pressure gradient (from Pa to µPa), gases reduce in volume and move to the last compressor blades and outlet. Some of them decay and become solids → deposition but vacuum not sufficient enough to ensure implantation after that the pump set back to normal pressure

Spreading contamination!

- Turbo pump is a physical barrier for particles in solid state



Smear on outlet of Frontend TP after 2 years of decay

Isotope	$T_{1/2}$ (y)	A (Bq)
Po-208	2.9	770
Po-209	102	40
Po-210	0.38	430
Ac-227	21.77	<0.1
Ra/Th-228	5.75	<0.1

Contamination of roughing pumps (1)

Oil specific activity in Bq/g

Measurement	Isotope	$T_{1/2}$	GPS Frontend (MP4A)	HRS Frontend (MP4B)	GPS Séparator (MP6A)	HRS Séparator (MP6B)	Experiments (MP8A)
Gamma spectrometry	Se-75	120 d	= **9.12E+03**	= **3.16E+03**	= **3.35E+02**	= **5.80E+01**	= **3.51E+00**
	Te-121m	154 d	= **4.87E+03**	= **9.79E+02**	= **1.38E+02**	= **1.21E+01**	= **1.30E+00**
	Te-123m	120 d	= **6.33E+03**	= **1.35E+03**	= **1.79E+02**	= **1.51E+00**	= **1.62E+00**
	Sb-124	60 d	= **7.33E+03**	= **1.66E+03**	= **1.00E+02**	= **1.70E+01**	= **1.10E+00**
	Sb-125	2.77 y	= **3.15E+03**	= **8.57E+02**	= **1.12E+02**	= **1.31E+01**	= **1.26E+00**
	I-125	60 d	= **4.11E+04**	= **1.47E+04**	= **9.49E+03**	= **6.17E+02**	= **2.02E+02**
	Cs-137	30.15 y	= **1.99E+03**	< 6.91E+01	= **9.05E+01**	= **2.67E+00**	= **1.47E+00**
	Hg-194	520 y	< 2.72E+02	< 1.04E+02	< 6.58E+00	< 1.03E+00	= **4.51E-01**
	Au-195	186 y	= **1.18E+04**	= **4.59E+02**	= **2.97E+01**	= **2.36E+00**	= **6.03E+00**
	Bi-207	31.55 y	= **1.07E+03**	= **9.36E+01**	= **1.27E+02**	= **1.28E+01**	= **2.68E+00**
Proportional counter	Alpha total		# **3.37E+04**	# **6.93E+03**	# **3.08E+03**	-	-
	Beta total		= **1.00E+05**	= **1.79E+04**	= **4.02E+03**	-	-

Contamination of roughing pumps (2)

Non volatiles Isotopes found in roughing pump oil

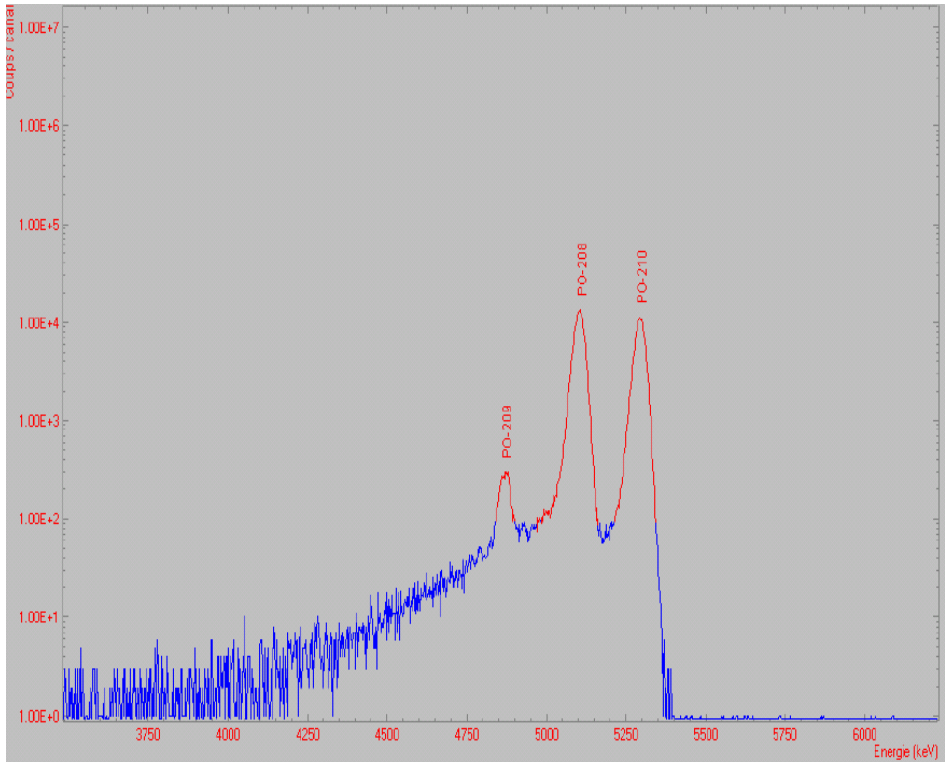
Volatile parent	Isotope	L_A /MBq	Contamination of one pump (3.6 l) /MBq
^{137}Xe	^{137}Cs	0.7	8.0
$^{195,195m}Hg$	^{195}Au	4.0	65.0
$^{207,211}Rn$, $^{207,211}At$	^{207}Bi	2.0	5.6
$^{208,212}Rn$,^{208}At	^{208}Po	0.002	40.0
^{209}Rn, ^{209}At	^{209}Po	0.002	1.0
^{210}Rn, ^{210}At	^{210}Po	0.002	36.6

Contamination of roughing pumps (3)

Alpha spectrometry

- α spectrometry on contaminated oil carried out by EIG with chemical treatment on the sample
  - Mineralization and electro-deposition
  - Chemical treatment efficiency checked with α/β spectrometry by liquid scintillation
- Results show ^{208,209,210}Po and fit CERN measurements except for ^{209}Po due to bad resolution spectrum and large tale (α spectrometry with very thin deposit ~1 mg)

Isotope	$T_{1/2}$	EIG Bq/g	CERN Bq/g
Po-208	**2.89 y**	1700	1700
Po-209	**102 y**	90	-
Po-210	**138.4 d**	15	210



pure α-emitters difficult to analyze, especially in oil (auto absorption)

Contamination of gas tanks

- 2 balloons (3 and 5 m^3, pressure between 10 and 2000 hPa allows a storage capacity of 24 m^3) emptied once/year
- Release with very slow flow rate 0.5 m^3/h within the ventilation rate of 10320 m^3/h → this ensure a second filtration and a high dilution factor that leave us far below the authorized release limits
- Measurements techniques:
  - ✓ Millipore filter and active charcoal (^{125}I, ^{68}Ge, ^{75}Se)
  - ✓ Differential ionisation chamber (^{3}H equivalent)
  - ✓ Gas sample for noble gases by γ-spectrometry ($^{42}Ar/K$, ^{85}Kr, $^{127,129m,131m,133}Xe$)

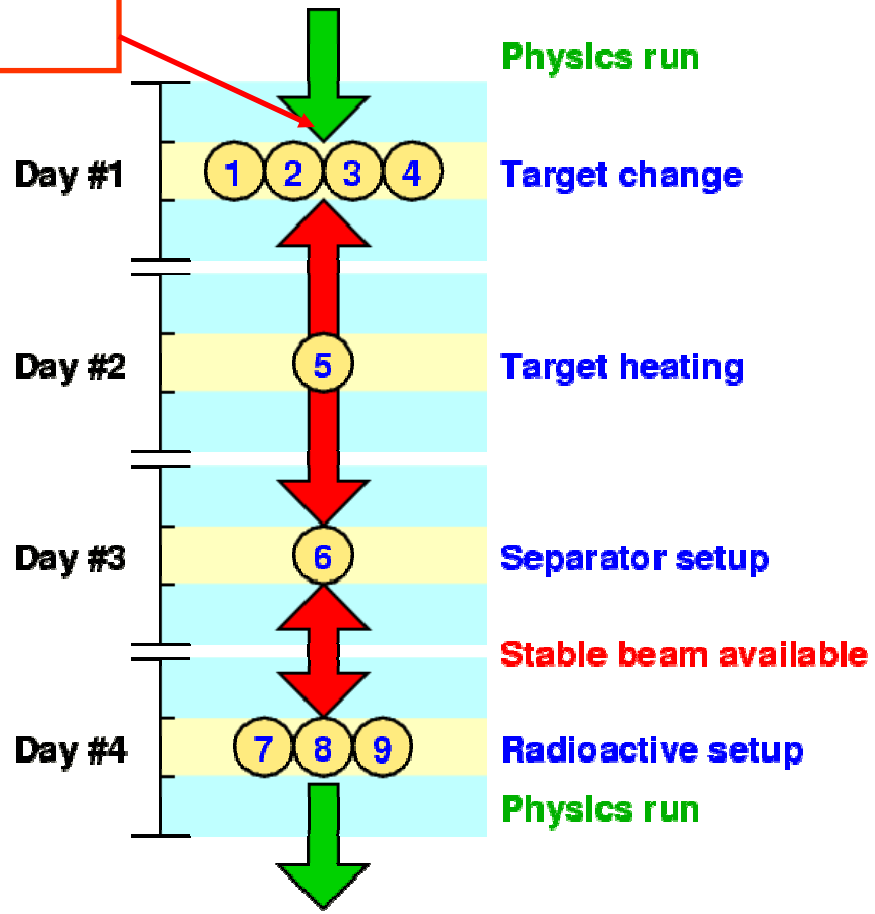
No α-emitters detected (and no γ-emitters daughters) during tanks release!

Acknowledgement: oil of roughing pumps behaves as an efficient filter but involves very delicate handling for maintenance work…

Target change: timing

Add delay of "Radioactive Cooling" for UCx and ThCx targets!

	Action	Allow
1	Cool old target	2 hr
2	Frontend to atmosphere and ventilation on access mode	1 hr
3	Put new target in zone	1 hr
4	Swap targets (robot)	2 hr
5	Pump / heat / outgas target	~40 hr
6	Setup source and separator	8 hr
7	Proton scan	2 hr
8	Yield check	5 hr
9	User consultation & beam handover	1 hr



UCx and ThCx target change

Radioactive cooling

Cooling period before target change

Irradiation mode	Delay
Protons taken on UCx or ThCx container (spallation)	**72 hr**
Protons taken on W-converter (fission)	**36 hr**

Reasons:

✓ automatic dust cap cover is not reliable

✓ Feedback from previous releases and container failures

Target	Radiologically important contamination	Characteristic half-life
U, Th	**α-emitters ^{227}Ac, ^{226,228}Ra, ^{208,209,210}Po … fission products**	**20 y, 1600 y, 5 y, 22 y various**
Pb	^{194}Hg/Au	520 a
Ta	Lanthanides (α): ^{148}Gd, ^{150}Gd	75a, 1.8E6 a
Other in Ta-container	mainly as for Ta	

UCx and ThCx target change

Access in target area and measurements

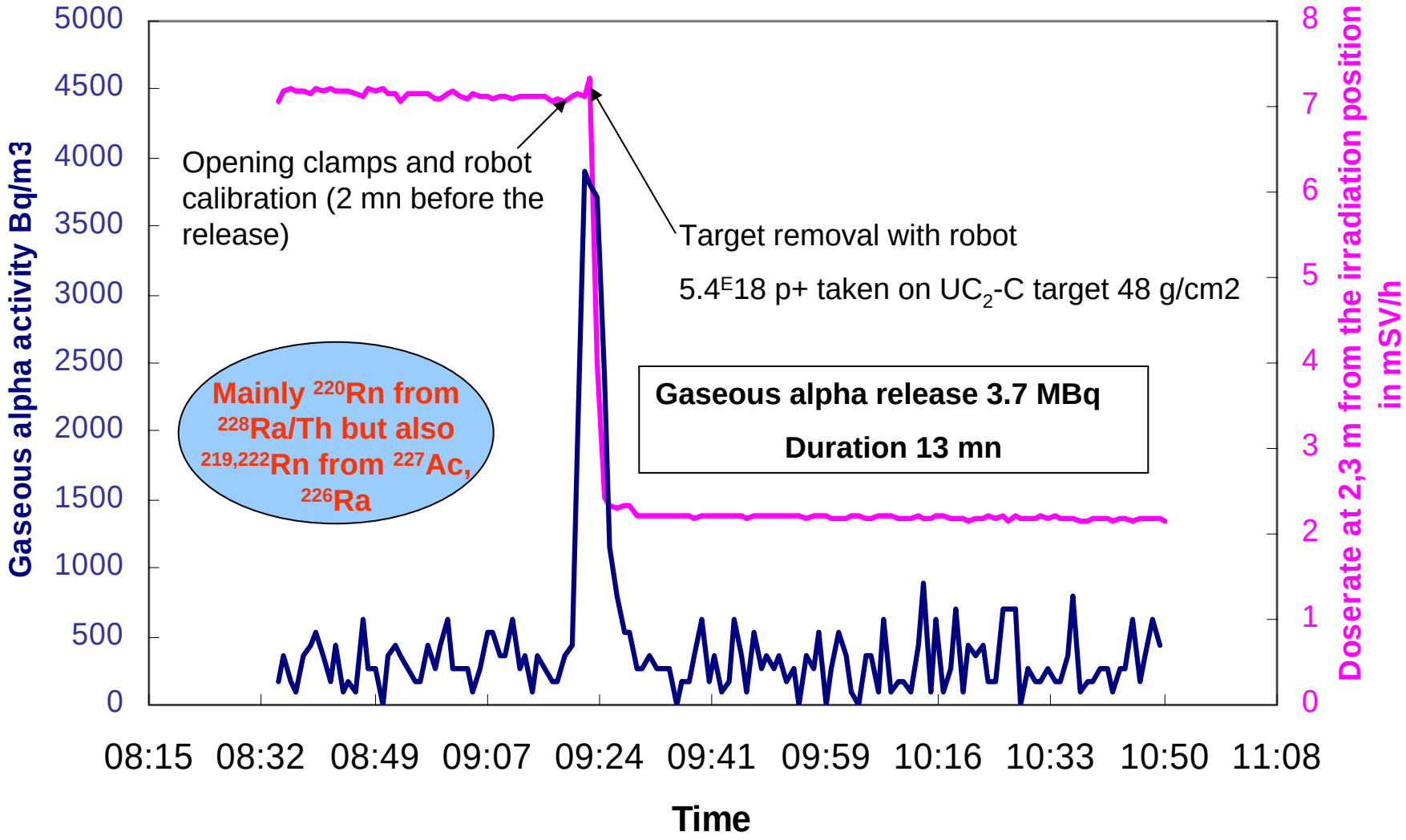
- **Ventilation on access mode, 30 mn for 3 air renewal in target area**
- **New aerosol filter on environmental station for short half lives measurement**
- **Access to put new target on robot exchange point**
- **Opening clamps → release measurements start**
- **Target handling (robot)**
- **Counting aerosol filter (sample ~1 hr) but please avoid target change on Friday evening! $T_{1/2}$(Pb-212)=10.6 h…**

Ventilation flows (m^3/h)			
Extraction type	**Flow capacity**	**Beam mode**	**Access mode**
Target area	12000	7200	10320
Laboratories	6000	5280	5280
Magnets	1200	1200	1200
Faraday Cages	500	500	500
Total	**19700**	**14180**	**17300**

- *Aerosol filter: millipore + active charcoal (conservative Eff. 50%)*
- *sampling/stack flow ratio: 0.0015*
- *Special integral program for environmental radiation monitors (acquisition data each mn)*
- *Gamma spectrometry, alpha and beta total on aerosol filter*
- *Lucas cell (ZnS) for gaseous alpha*

Typical gaseous alpha release for UC_2-C target change

Lucas cell measurement



Typical release assessment

Cooling delay (d)	Target Id	P^{+} $1^{E}18$ Separator	Rn-220 in KBq	Pb-212 In kBq	I-124 In kBq	I-126 In kBq	I-131 In kBq
3	UC_2-C #246	2.9 GPS	2900	38	175	110	600
3	UC_2-C #250	7.8 GPS	1500	240	300	220	1060
3	UC_2-C #252	1.7 GPS	600	125	235	155	600
3	UC_2-C #255	5.4 GPS	3700	380	35	30	140
3	UC_2-C #214	2.2 GPS	<100	300	42	140	230
6	UC_2-C #259	11.8 GPS	600	75	100	110	630
5	UC_2-C #261	5.74 HRS	<100	30	23	40	150
9	UC_2-C #214	2.1 GPS	<100	2	0.4	8	13

Unfortunately the link between cooling days, protons and activity is not very clear!

Isolde radioactive emissions during 2005

Isolde facility: radioactive emissions in Bq

Isotope	Total	RV	FRV
Short-lived	6.76E+12	5.90E+14	1.10E-02
Total beta	2.84E+05	1.30E+10	2.20E-05
Tritium	3.78E+07	3.50E+15	1.10E-08
Total alpha	0.00E+00	4.70E+11	0.00E+00
Be-7	1.89E+06	1.10E+13	1.70E-07
Na-22	1.65E+02	2.50E+10	6.60E-09
Zn-65	2.68E+01	2.20E+11	1.20E-10
Se-72	4.54E+04	1.10E+12	4.10E-08
As-74	5.78E+03	1.40E+12	4.10E-09
Se-75	7.27E+04	4.90E+11	1.50E-07
Te-121	5.08E+03	2.70E+12	1.90E-09
Te-121m	3.74E+01	1.30E+10	2.90E-09
Te-123m	1.47E+02	1.30E+10	1.10E-08
I-124	9.64E+04	1.10E+12	8.80E-08
Sb-124	6.72E+01	2.40E+11	2.80E-10
I-126	1.72E+05	3.60E+11	4.80E-07
I-131	4.04E+05	5.10E+11	7.90E-07

RV *reference value (0.2 mSv for a member of the public from the critical group)*

FRV *fraction of the reference value*

Gases tanks: emissons in Bq

Isotope	Total	RV	FRV
Tritium	3.17E+10	3.50E+15	9.10E-06
Ar-42	3.00E+05	4.10E+18	7.30E-14
Kr-85	2.09E+07	4.10E+18	5.10E-12
Xe-127	1.92E+07	1.70E+15	1.10E-08

Actinide targets change: emissions in Bq

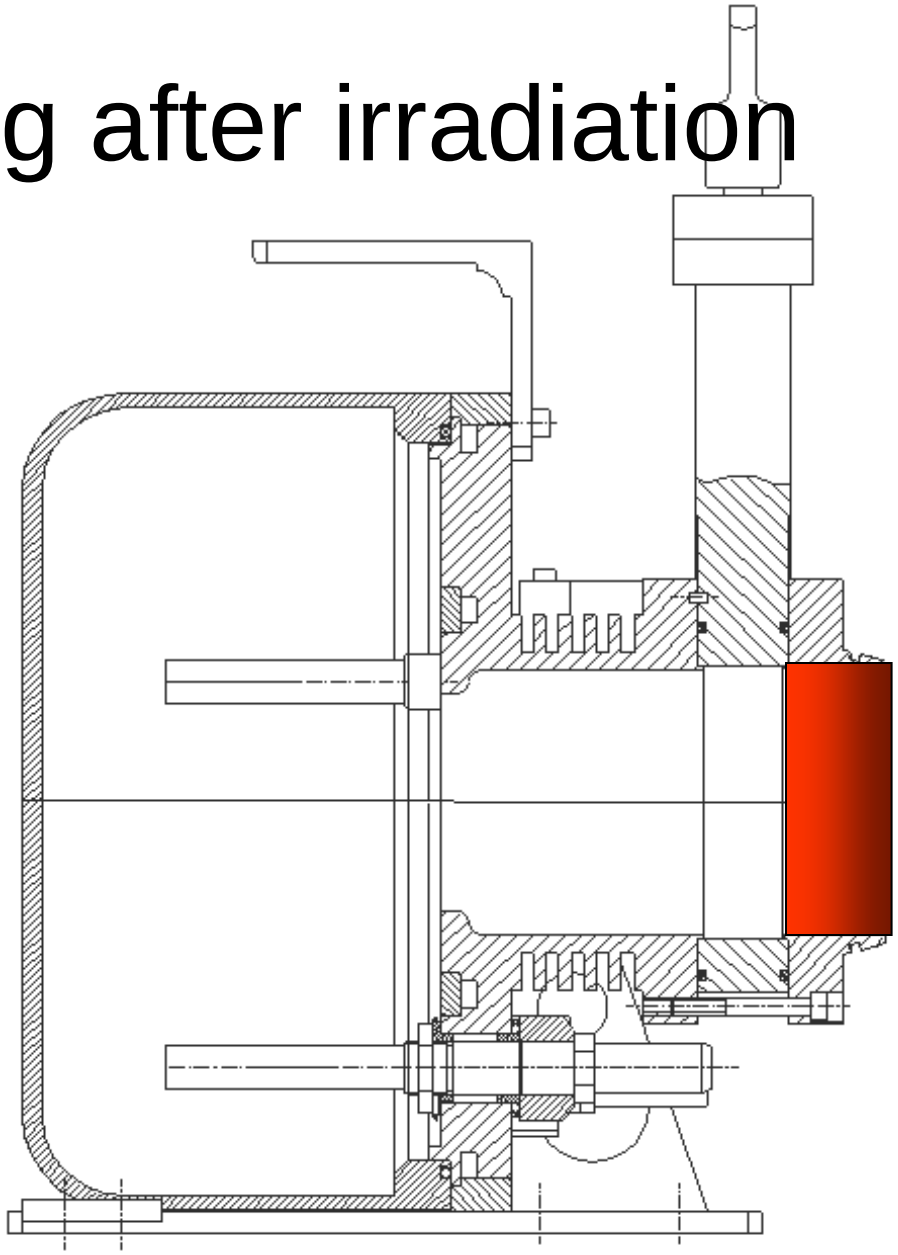
Isotope	Total	RV	FRV
I-124	7.05E+05	1.10E+12	6.40E-07
I-126	1.09E+06	3.60E+11	3.00E-06
I-131	3.61E+06	5.10E+11	7.10E-06
Rn-220	1.17E+07	4.70E+11	2.50E-05
Pb-212	1.02E+05	4.70E+11	2.20E-07

Target outgasing after irradiation

A weak point: remaining contamination on the connecting part. Automatic dust cap cover malfunctions lead to skip this operation and to avoid robot failures.

Dust caps are placed manually during the yearly targets transport (radioactive cooling of a few months)

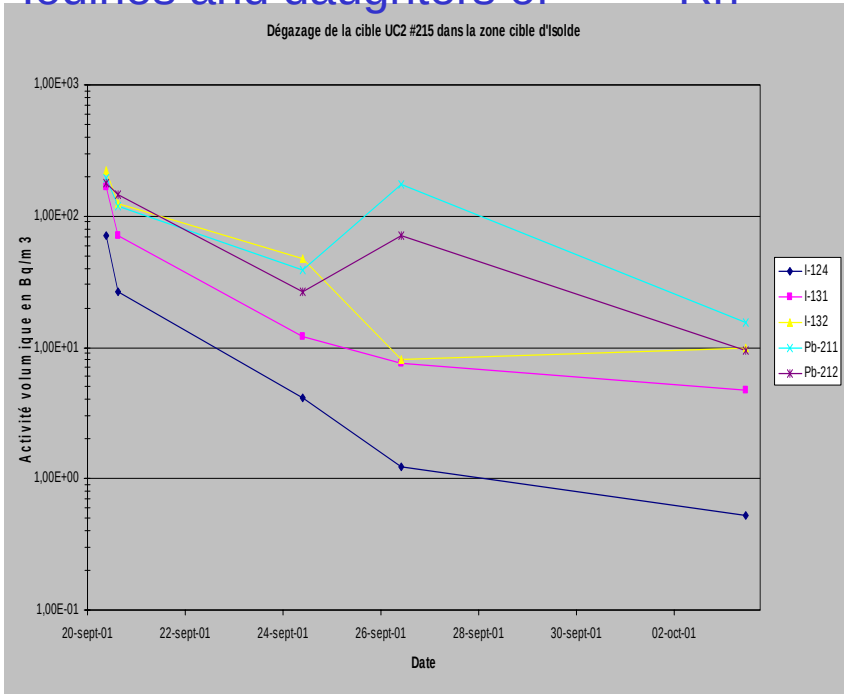
Target storage in ISR5 tunnel



Target UC_2-C #215 outgassing

Radioactive contamination from the connecting part presents a minor but significant risk of incorporation

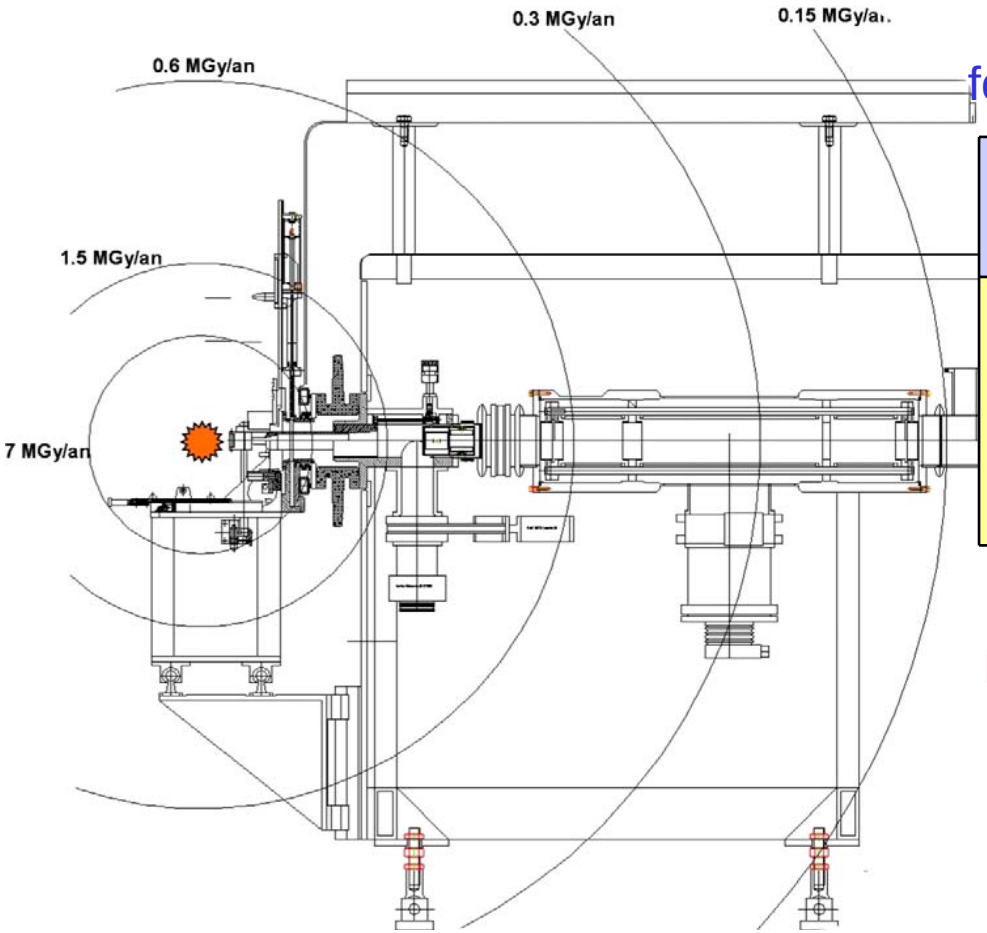
Iodines and daughters of $^{219,220}Rn$



Radio isotopes	Activity Bq/m_3	CA value	Dose factor Sv/Bq	Effective dose in µSv
I-124	7.10E+01	0.1	6.3E-09	0.8
I-131	1.65E+02	0.2	1.1E-08	3.4
I-132	2.23E+02	0.0	2.0E-10	0.1
Pb-211	1.96E+02	0.2	5.6E-09	2
Pb-212	1.80E+02	0.6	3.3E-08	11

Intervention of 1.5 hr in target area: 20 man*µSv by inhalation (negligible related to external exposure)

Integrated absorbed dose



Comparison of absorbed dose normalized for 1E18 p+ between light and heavy targets

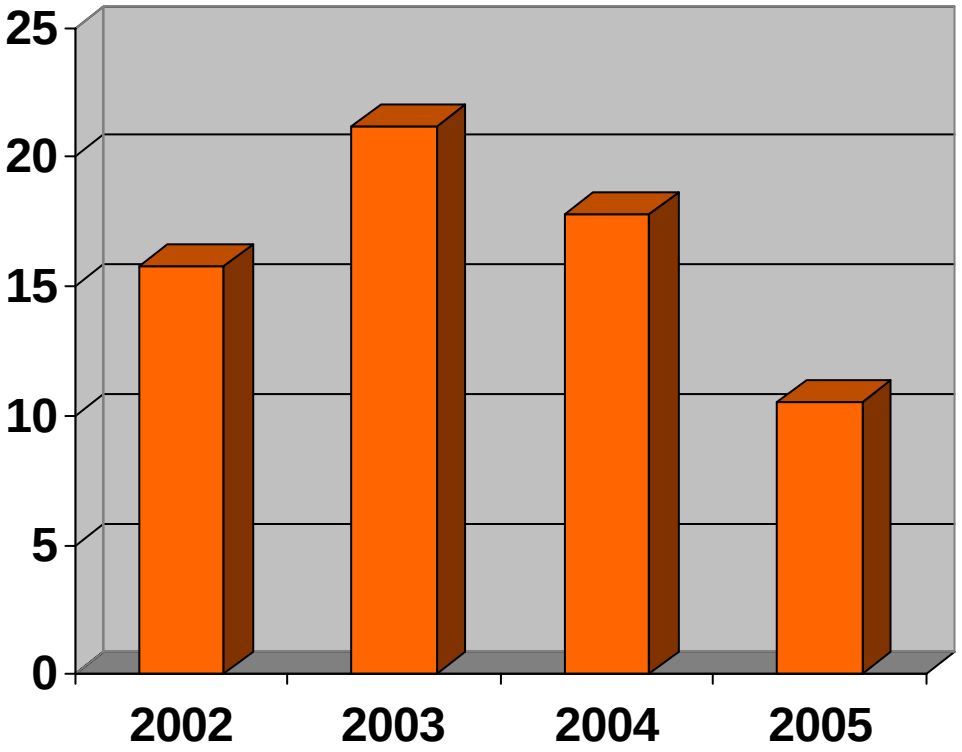
Material	Thickness (g/cm2)	Mean free path	Protons 1E18	HV room neutron integrated dose (mSv)	Relative absorbed dose (Gy)
Ti foil	19	0.26	0.62	2.48	331
Ta foil	26	0.25	2.43	1.93	247
ZrO_2	4	0.15	3.36	1.58	279
UC_2-C	47	0.52	3.35	5.07	475
UC_2-C	48	0.53	0.23	6.83	478

Higher neutrons flux with UCx targets →

- ✓ increase of induced activity
- ✓ therefore increase of external exposure and collective dose

Collective dose received at Isolde

Operational records (man·mSv)

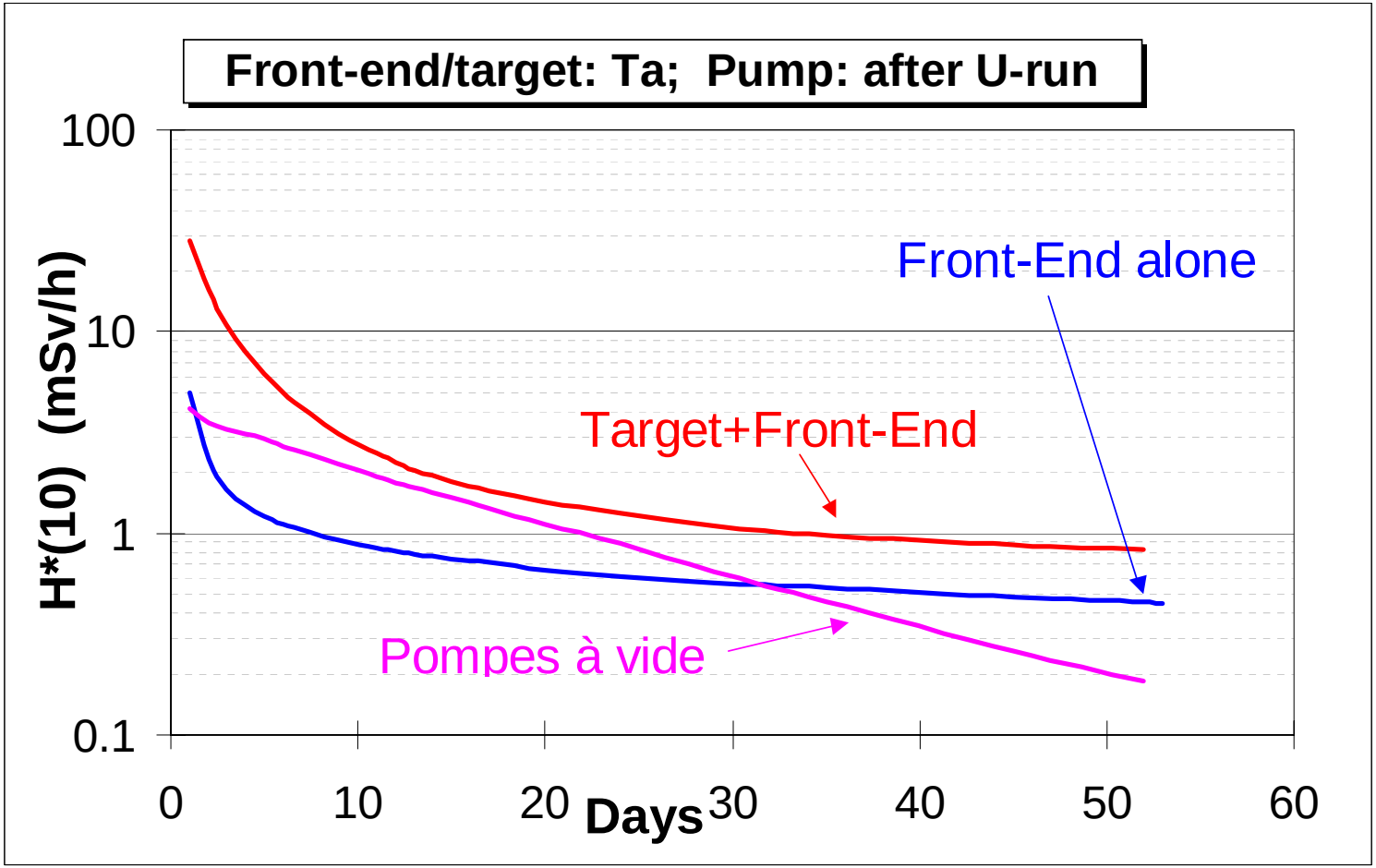


~ 95% received in the target area

~ 70% due to maintenance and reparation of the frontends

Such equipments MUST be designed in order to reduce the collective dose (easy plug connections, spare parts available, ergonomy, etc…)

Gamma-doserate from a roughing oil pump (mainly ^{206}Po/Bi, ^{205}Bi)



Radioactive waste

Project of ISOLDE targets elimination

- CERN presented to PSI a project including:
  - a computer code (Fluka) for visualization of radionuclides produced during irradiation and after their decay
  - Preliminary measurements and radionuclides inventory carried out on some representative targets
- The acceptance of the ISOLDE targets at PSI is conditioned to the main following requirements:
  - Radionuclide inventory for α β γ emitters for each target (with $T_{1/2}$>2y)
  - Separation of α-emitters to satisfy the limit in a content for the envisaged waste type (1 MBq α for 200 l drum)
  - Separation of aluminium parts
- Calculation methods shall be submitted for validation to NAGRA. If required, α spectrometry shall be performed to complete the radionuclide inventory
- Only storage and no project today for others radioactive waste (extraction electrodes, pumps, oil, etc…)

Actinides targets

Conclusions

- Enlarged opportunities for physics but big challenges for radiation safety!
  - Multitude of produced isotopes makes analysis and assessment difficult (pure α-emitters, ~150 γ-rays)
  - Radiation hazards increased with:
    - Highly radiotoxic contamination which requires an assessment of internal exposure
    - Higher induced activity and collective dose

- More constraints and costs for radioactive waste management. “Temporary” storage for radioactive decay is not sufficient… At ISOLDE, there is no storage place anymore and the 2006 irradiated units are coming soon. The future starts today.