EURISOL

EURISOL Town meeting - Lisbon - 18 October 2012

Hg target - Safety point

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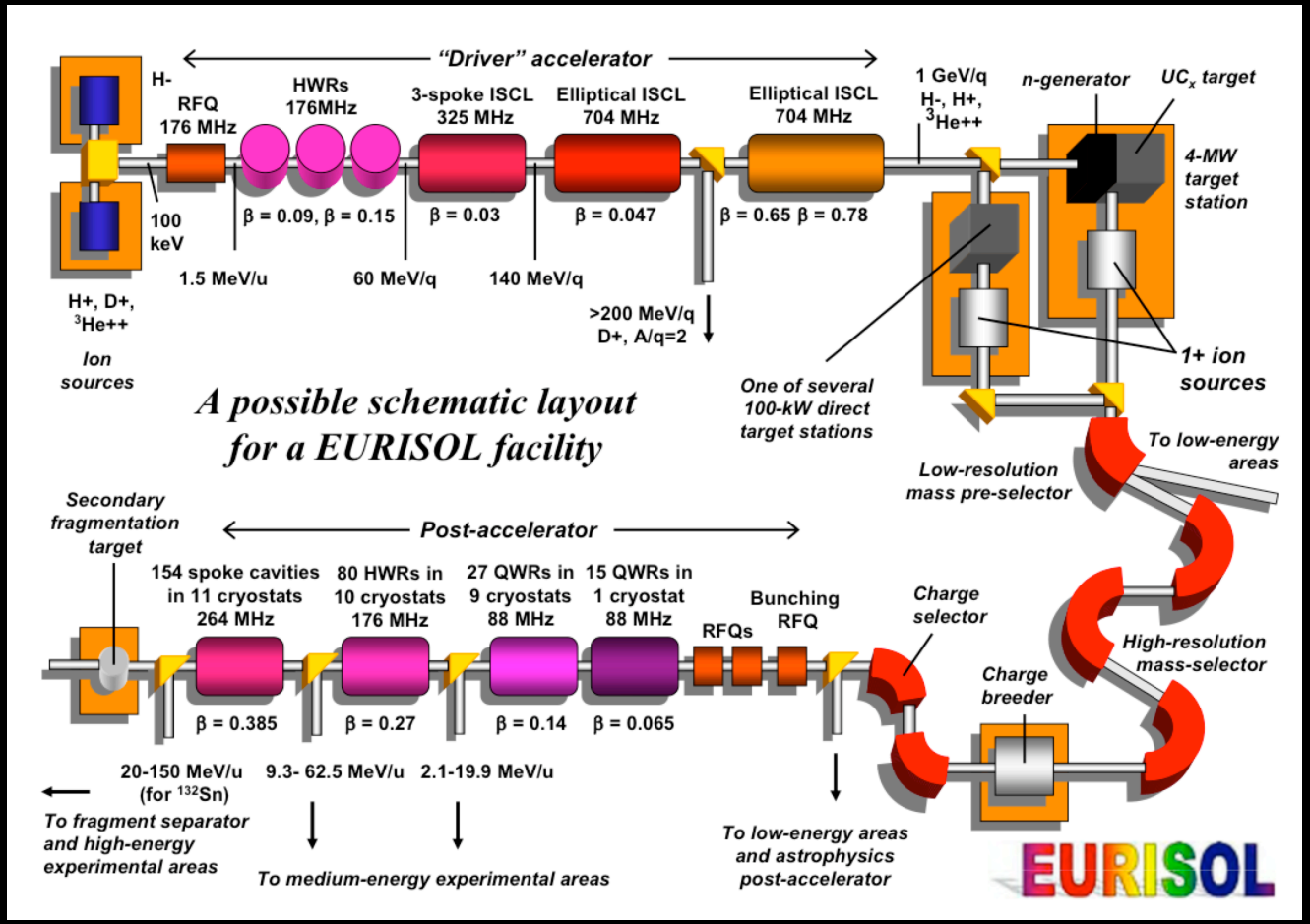
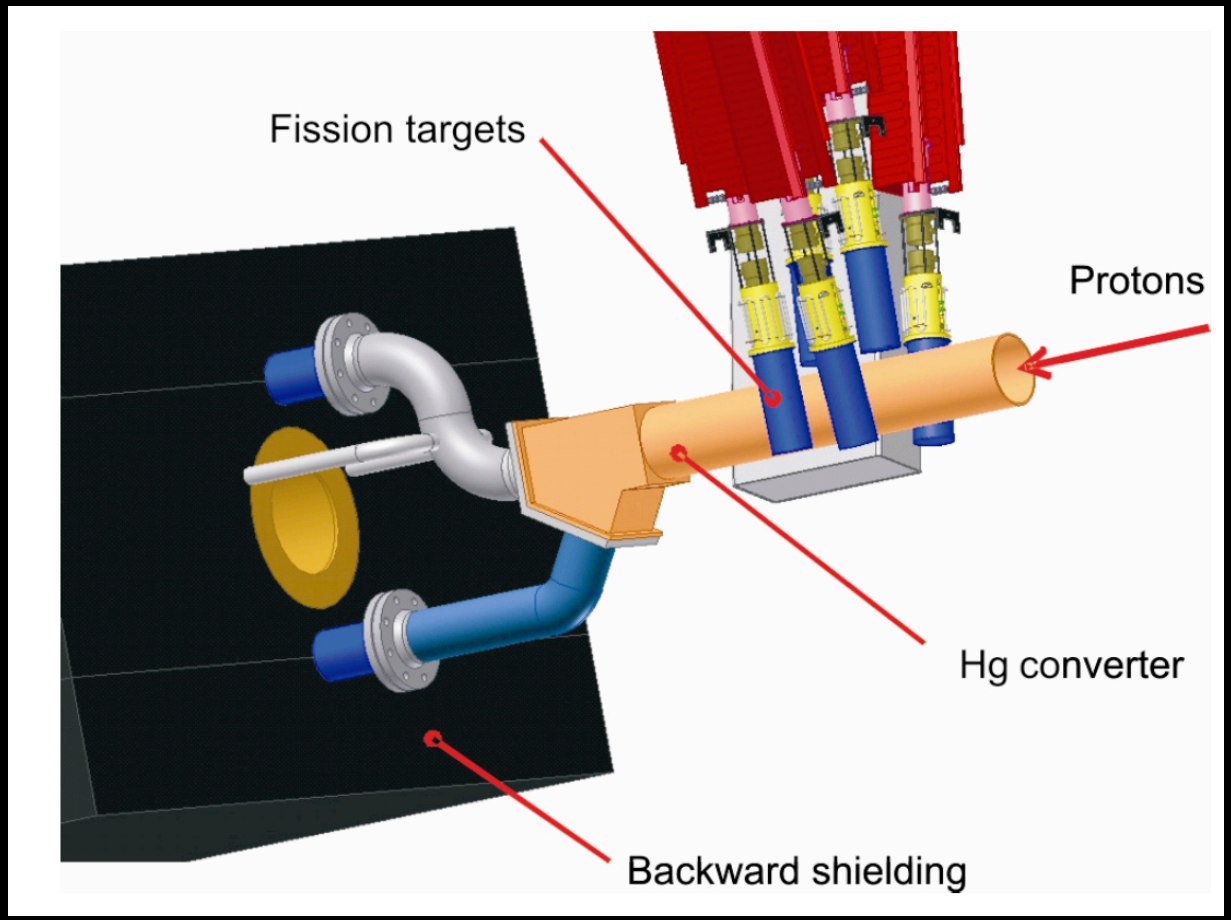
Irfu
CEA - Saclay

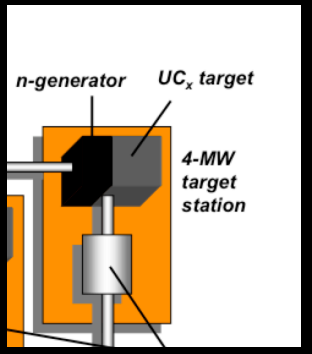
Hg target - Safety point

« a talk on radioactive inventory and Hg target disposal. In particular a discussion on the feasibility of using a Hg target from the safety and regulatory point of view would be much appreciated. »

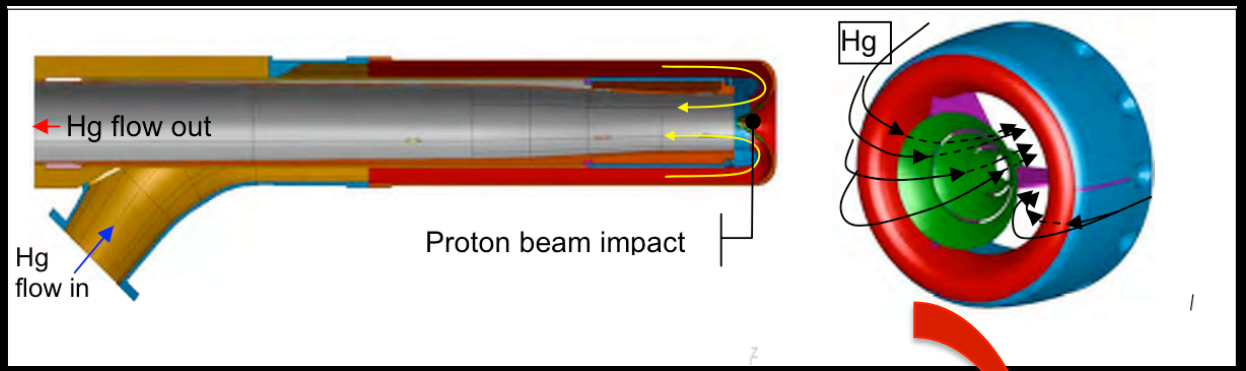
… Tentative summary from EURISOL-DS and literature

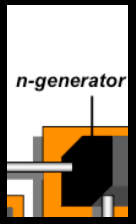

"Driver" accelerator
H-
RFQ
176 MHz
HWRs
176MHz
3-spoke ISCL
325 MHz
Elliptical ISCL
704 MHz
Elliptical ISCL
704 MHz
1 GeV/q
H-, H+,
3He++
n-generator
UCx target
4-MW
target
station
100
keV
β = 0.09, β = 0.15
β = 0.03
β = 0.047
β = 0.65 β = 0.78
1.5 MeV/u
60 MeV/q
140 MeV/q
>200 MeV/q
D+, A/q=2
H+, D+,
3He++
Ion
sources
A possible schematic layout
for a EURISOL facility
One of several
100-kW direct
target stations
1+ ion
sources
To low-energy
areas
Low-resolution
mass pre-selector
Secondary
fragmentation
target
Post-accelerator
154 spoke cavities
in 11 cryostats
264 MHz
80 HWRs in
10 cryostats
176 MHz
27 QWRs in
9 cryostats
88 MHz
15 QWRs in
1 cryostat
88 MHz
RFQs
Bunching
RFQ
Charge
selector
High-resolution
mass-selector
Charge
breeder
β = 0.385
β = 0.27
β = 0.14
β = 0.065
20-150 MeV/u
(for 132Sn)
9.3- 62.5 MeV/u
2.1-19.9 MeV/u
To fragment separator
and high-energy
experimental areas
To medium-energy experimental areas
To low-energy areas
and astrophysics
post-accelerator
EURISOL

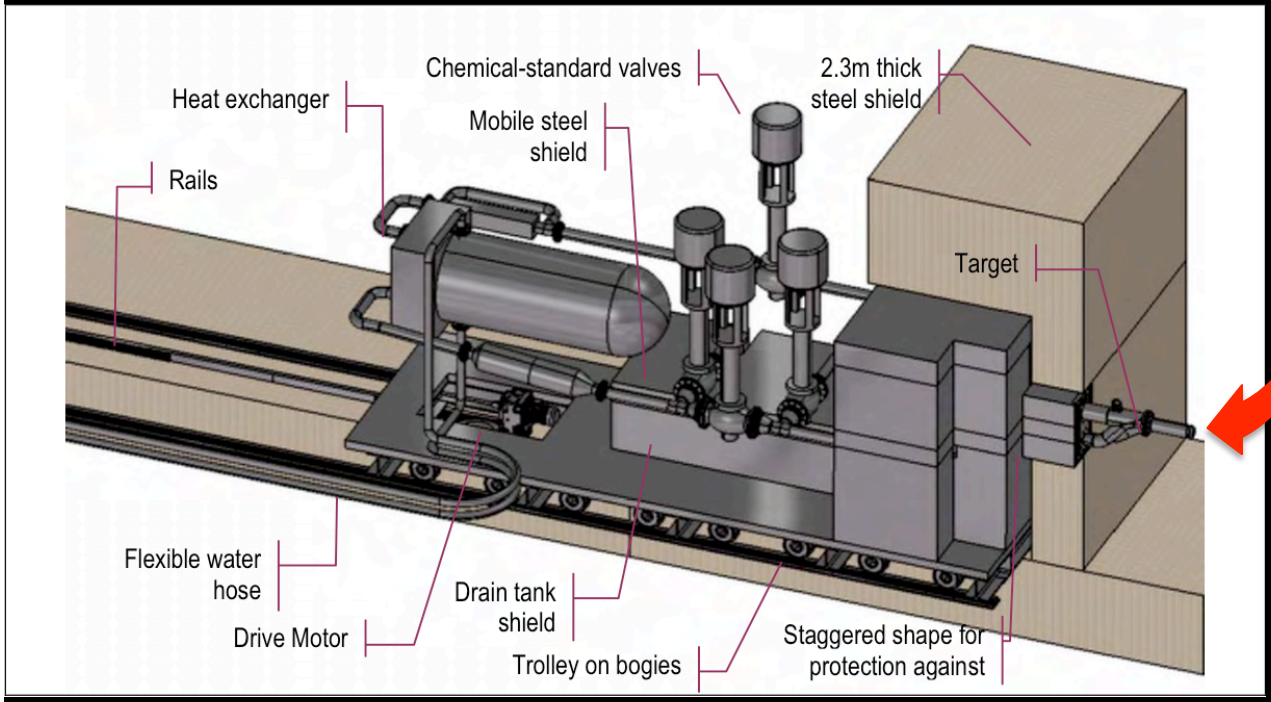

Fission targets
Protons
Hg converter
Backward shielding


n-generator
UCx target
4-MW
target
station

MMW view







Converter length: 45 cm
Converter radius: 15 cm
Beam power: 4 MW
Hg temperature: < 260 °C

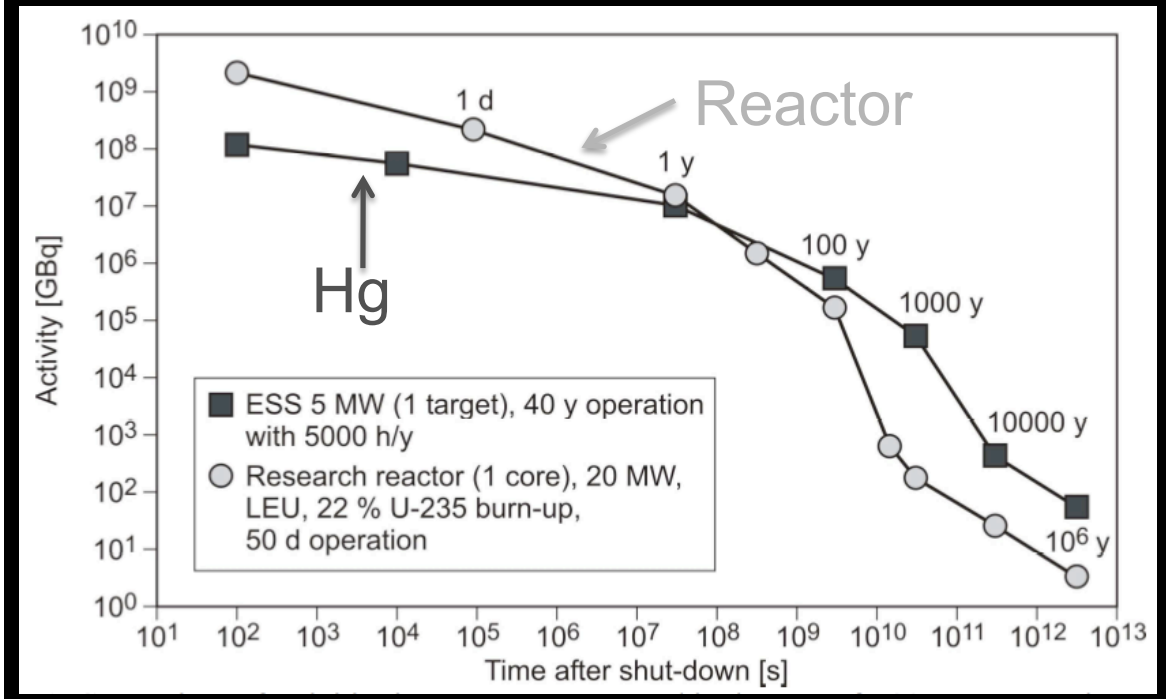
- Other places: SNS (2006) and JSNS (2008)
- Other possibilities: W (ESS) and Pb-Bi (MegaPie – Myrrha)

4 points addressed

- Radionuclide inventory / Radioactivity / Dose
- Operation / Hot cell
- Decommissioning / Disposal
- Legislation

Radionuclide inventory / Radioactivity / Dose

Total activity compared to research reactor

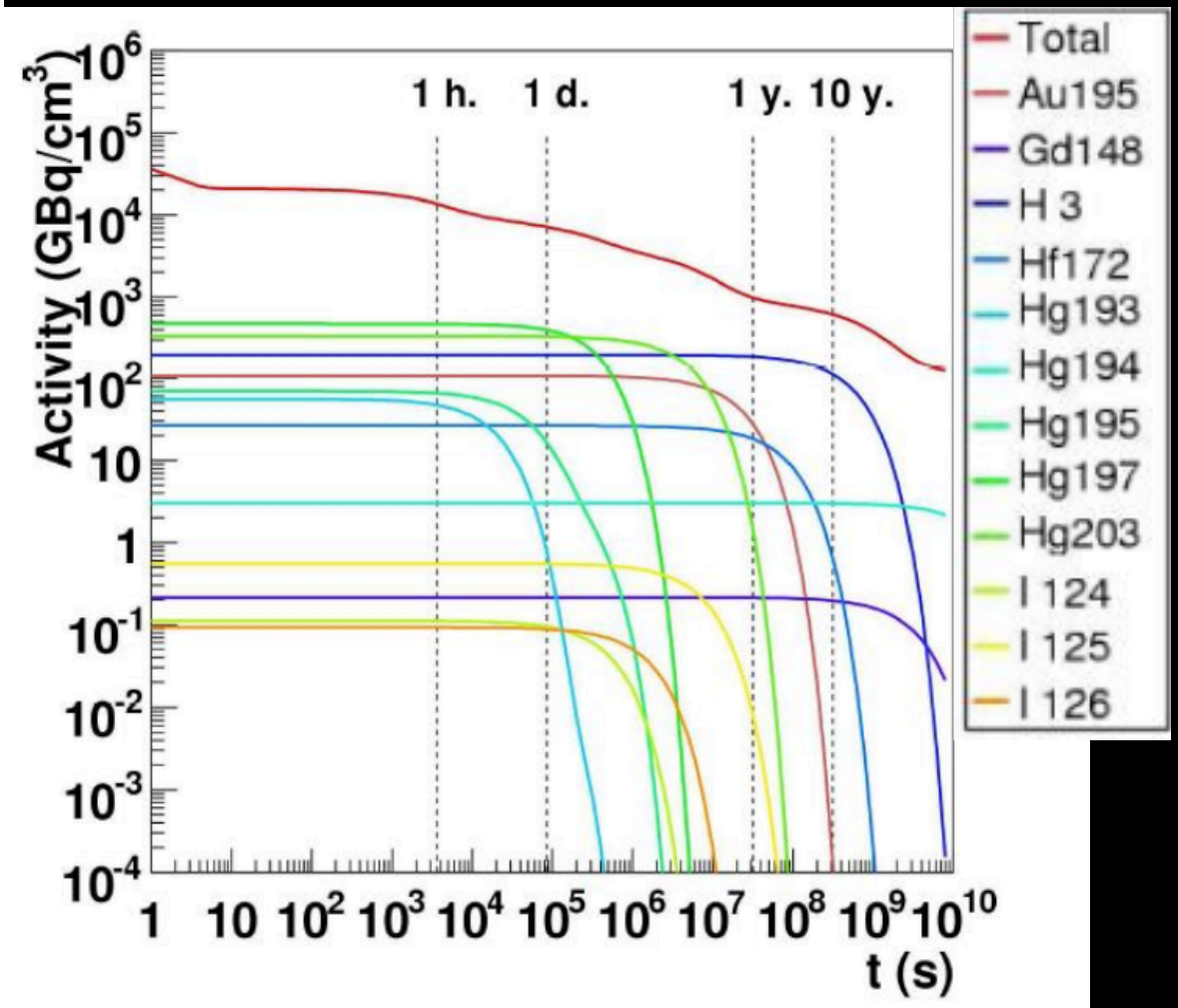


. Same order of magnitude:
1-10 MCi

. Shape different

. 1000 y after shut-down:
Hg > Research Reactor

Radionuclide inventory / Radioactivity / Dose



Main contributors to radiological consequences

- ^{148}Gd (α emitter)
- Tritium
- Hg (esp. ^{194}Hg)
- Iodine (volatile)

Activity in Hg target (40 y operation)

Radionuclide inventory / Radioactivity / Dose

- ^{148}Gd (α emitter)

NEW (not in research reactor)

inhalation

Less in Pb-Bi
More in W

Radionuclide inventory / Radioactivity / Dose

Now, calculation results reliable (in-target)

BUT Tritium missing (JSNS measurement)!

→ Chemistry (Hydrogen in Hg)… ?

inhalation/ingestion

- Tritium

Radionuclide inventory / Radioactivity / Dose

Hg radionuclide can't be separated

^{194}Hg (512y) → big issue in Hg disposal!

γ-groundshine

- Hg (esp. ^{194}Hg)

Radionuclide inventory / Radioactivity / Dose

Volatile (in operation)

ingestion

But Iodide formation (HgI_2) → less volatile

- Iodine (volatile)

Radionuclide inventory / Radioactivity / Dose

Consequences of high radioactivity

- Remote handling (Hot cell)
- Hg Purification?
- Off-gas process system

Operation / Hot cell

No leak acceptable (conventional and radiological toxicity)

Except in Hot cell (maintenance openings → leaks)

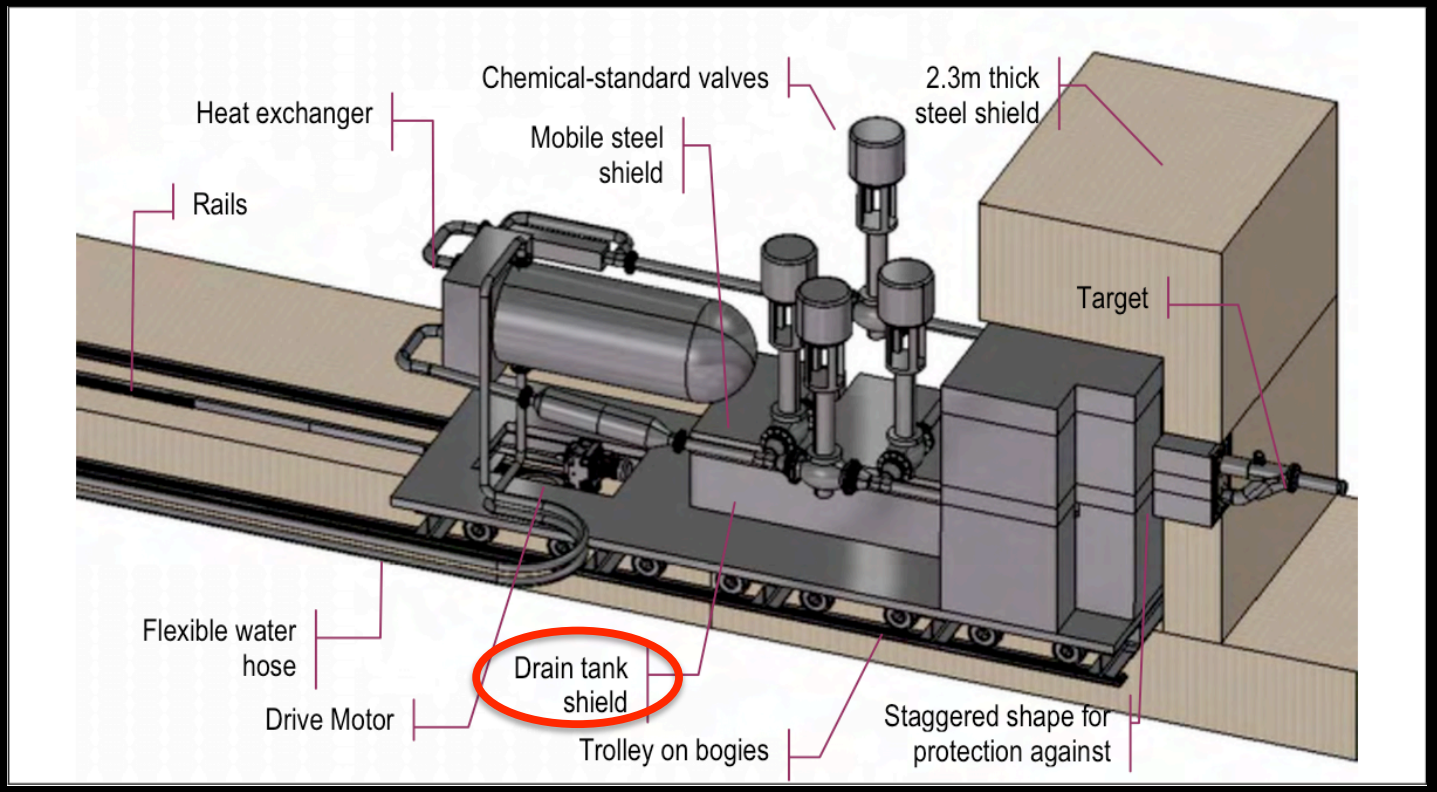
THEN fully remote handling! → large → costly

(SNS: 4 m x 30 m)

Operation / Hot cell

How to decrease radioactivity/dose?

→ drain Hg to a shielded tank



Operation / Hot cell

How to decrease radioactivity/dose?

→ drain Hg to a shielded tank

Unfortunately Spallation Products adhere to the pipe!!!

JSNS measurements:

→ Dose rates increased after draining Hg

Operation / Hot cell

How to decrease radioactivity/dose?

→ drain Hg to a shielded tank

Unfortunately Spallation Products adhere to the pipe!!!

JSNS measurements:

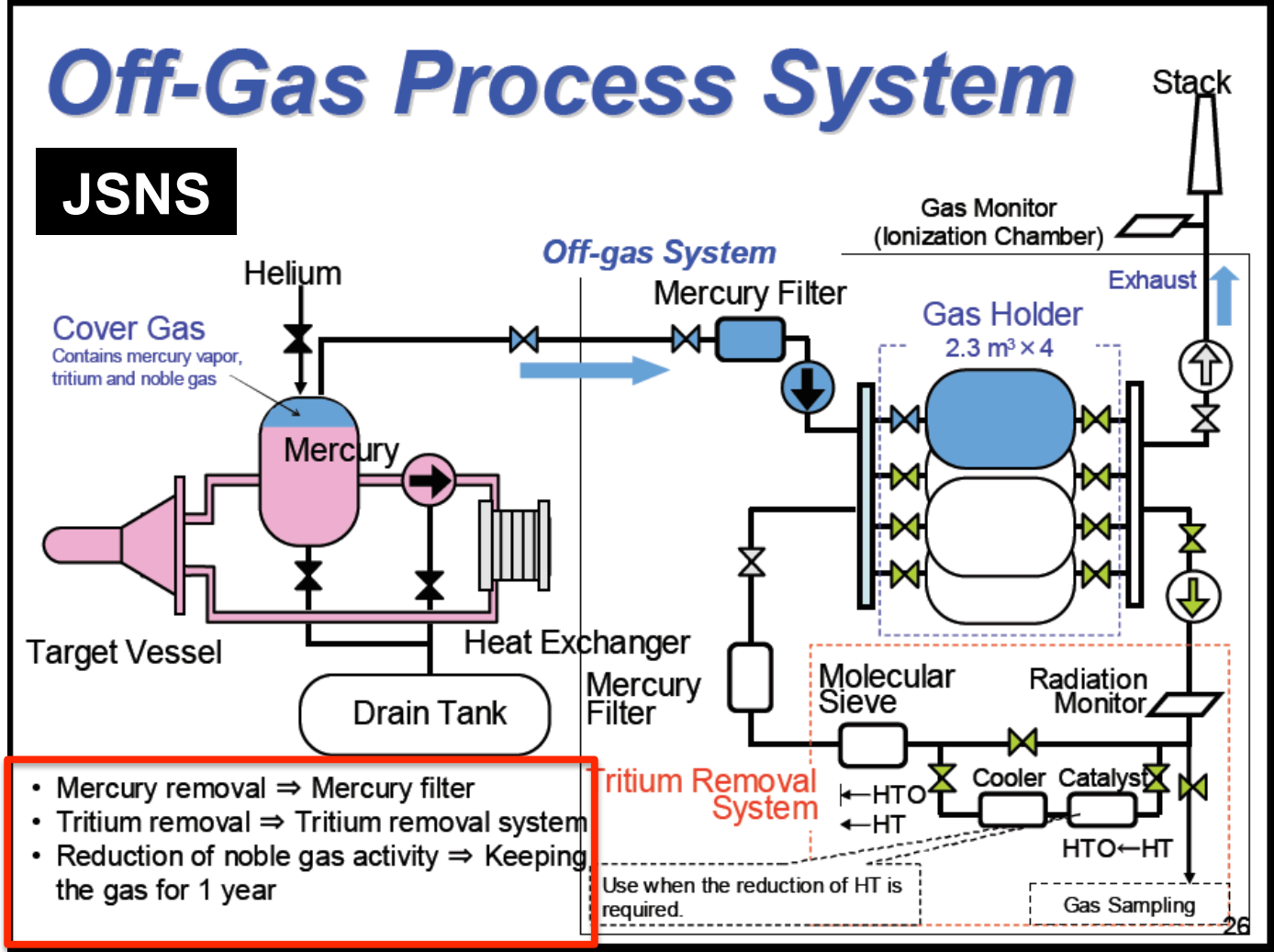
→ Dose rates increased after draining Hg

So, if purification methods has already been studied,
cleaning the loop seems to be necessary!

Operation / Hot cell

Management of the gases

Operation / Hot cell



cover gas:
Hg + tritium + noble gas

Exhausting gas safely needs:
. filters
. time (decay)

Slide from Y. Kasugai (JAEA)

Operation / Hot cell

Estimation of Off-gas Radioactivity

Gas Products	Half-life	Estimation		Regulation for the radioactivity concentration of exhaust gas [Bq/cm³]
		Total Activity of off-gas = *A* (Calculation in 1 MW and 5000 h operation) [Bq]	Radioactivity Concentration of the exhaust gas = $A/3.5\times10^{14}$ cm³/3 months (averaged for 3 months) [Bq/cm³]	
T	12.3 y	5×10^{13}	0.14	5×10^{-3} (HTO) 70 (HT)
^{37}Ar	35 d	1.1×10^{11}	3×10^{-4}	7×10^{2}
^{39}Ar	269 y	4×10^{8}	1.1×10^{-6}	0.2
^{42}Ar	32.9 y	1.1×10^{9}	4×10^{-6}	0.2
^{81}Kr	2×10^{5} y	6×10^{6}	1.8×10^{-8}	0.1
^{85}Kr	10.8 y	1.2×10^{10}	4×10^{-5}	0.1
^{127}Xe	36.4 d	1.2×10^{13}	3×10^{-2}	3×10^{-3}
^{194}Hg	520 y	9×10^{3}	2.6×10^{-11}	3×10^{-6}
^{203}Hg	46.6 d	1.1×10^{8}	3×10^{-7}	2×10^{-5}

- Concentrations of HTO component of tritium and ^{127}Xe exceeds the regulation in our estimation. => Off-gas process is required.

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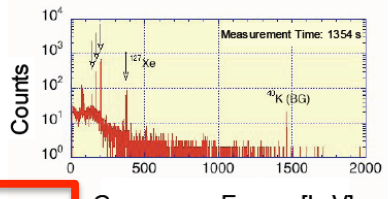
Confirmation of the Estimation

- After the beam operation with 17 kWh and 3-month cooling
- Off-gas sampling
- Gamma-ray Spectroscopy
  - Only ^{127}Xe was observed
  - Measurement : 4×10^{6} Bq (=E)
  - Calculation : 3.7×10^{7} Bq (=C)
  - C/E = ~10



- Tritium Measurement
  - Total: 3×10^{4} Bq (Detected HTO only, not HT)
  - Calculation: 3×10^{8} Bq
  - C/E = 10^{4} *=> Tritium Missing*

- There are large discrepancies between the calculation and measurement for radioactivity of tritium.
  - Based on the measurement, we do need to operate the tritium process loop.
- In order to investigate the "tritium missing issue", we have to consider the chemical behavior of hydrogen in mercury.

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Slides from Y. Kasugai (JAEA)

Decommissioning / Disposal

- Chemical toxicity must not be neglected, but Radiotoxicity dominates

- After shut-down, radioactivity = ~research reactor

Time	Hg-target 5 MW	Research Reactor 20 MW
10 y	5.E6	2.E8
1000 y	1.E5	2.E4
100000 y	1.5E2	1.E3

- Specificities
  - ^{194}Hg (512y)
    → purification inefficient
  - NO radioactive liquid in repository (EU)

→ Solidification is required!

Decommissioning / Disposal

- Solidification
- Main risk : solubility in water
- γ-irradiation ($^{194}Hg \rightarrow {}^{194}Au$ (strong γ emitter))

Decommissioning / Disposal

- Candidates:
  - 2 inorganic compounds: HgS – HgSe
  - 1 amalgam with Ag

- Experiment (stabilibily in diverse “water” and γ-irradiation)

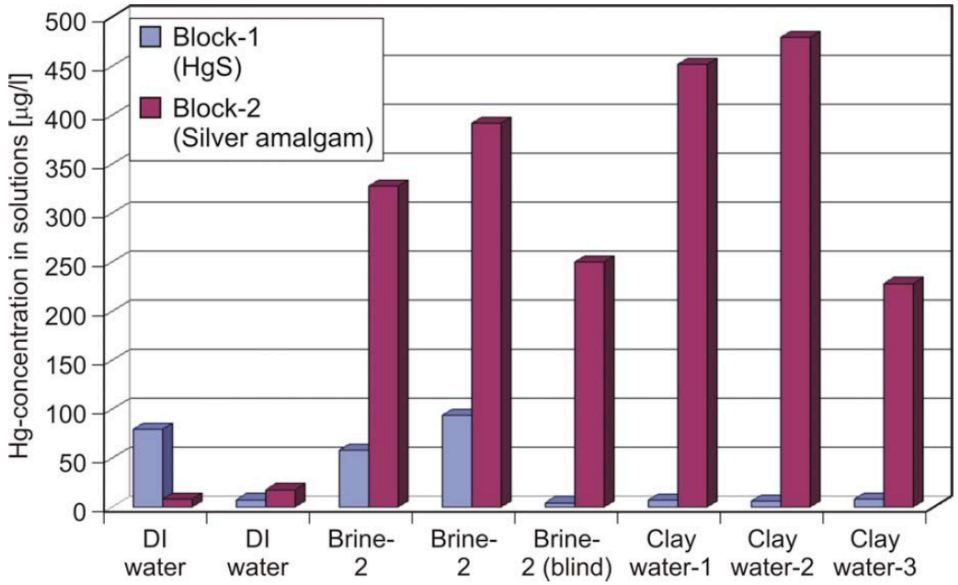


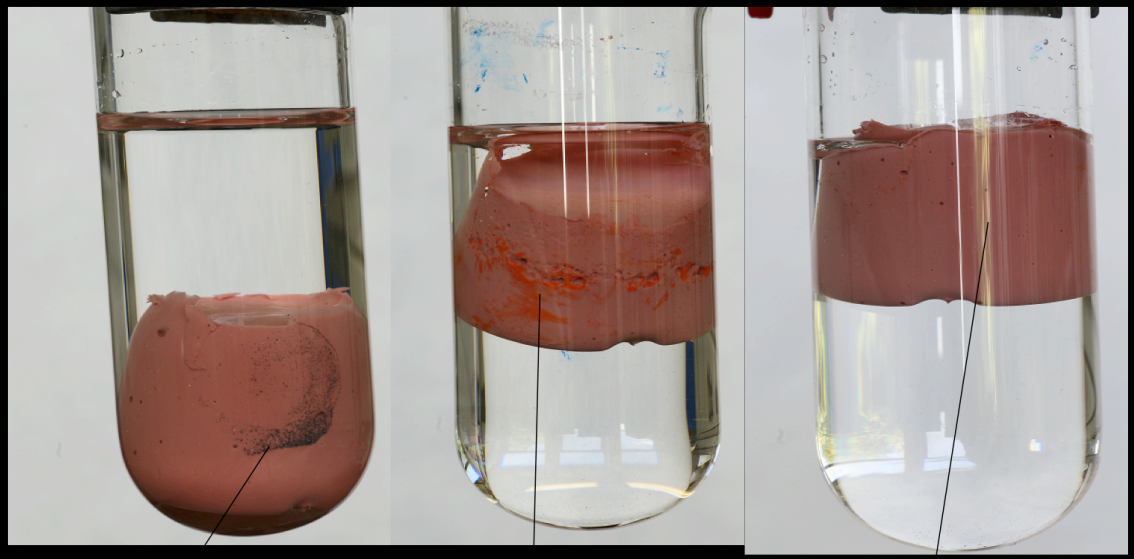
Fig. 3-1: Mercury concentration in solution after 2 months treatment of HgS and Ag-amalgam under γ-irradiation in diverse aqueous environments

- HgS better
(HgSe toxic and expensive)

- Radiolysis important

Decommissioning / Disposal

- Embedding HgS in a matrix increases its stability (final disposal)
  - Cement → better, but inhomogeneous distribution (Hg)
  - Polysiloxane → promising candidate, especially with double layer



Polysiloxane +HgSe(single coated)

Polysiloxane+HgS (single coated)

Double coated HgS sample

Decommissioning / Disposal

Solidification seems possible , but

- Up to now, only first step
  - lab scale experiments
  - γ-irradiation must be done with polysiloxane
- Large effort compared to Pb-Bi

Decommissioning / Disposal

SNS: "Disposal options are VERY limited"

Van Graves (talk at CERN (2008))

JSNS: "final decommissioning plan is not fixed yet"

Meigo-san (private communication)

SO, no experience from abroad!

Legislation

In EU, safety considerations are based on 3 categories (occurrence in one year)

- Normal operation and frequent abnormal events or 'incidents' ($>10^{-2}$ y^{-1}),
- Design basis accidents, DBA (10^{-2} y^{-1} - $5 \cdot 10^{-6}$ y^{-1})[1],

- Design extension accidents, DEA, 'hypothetical' events ($<5 \cdot 10^{-6}$ y^{-1})[1].

… and, over the last 20 years,
lower frequency limit of DBA is continuing decreasing (10^{-6}/y is expected)!

Legislation

Conventional Toxicity

- EU directive Seveso-II (toxicity of inactive Hg) doesn't apply for Eurisol
  (Masse of Hg < 50 t)
  But probably conventional toxicity should be considered in accident analyses

- Accidental Hg release:
  Chemical plants safety guidelines apply (slightly ≠ acc. countries)

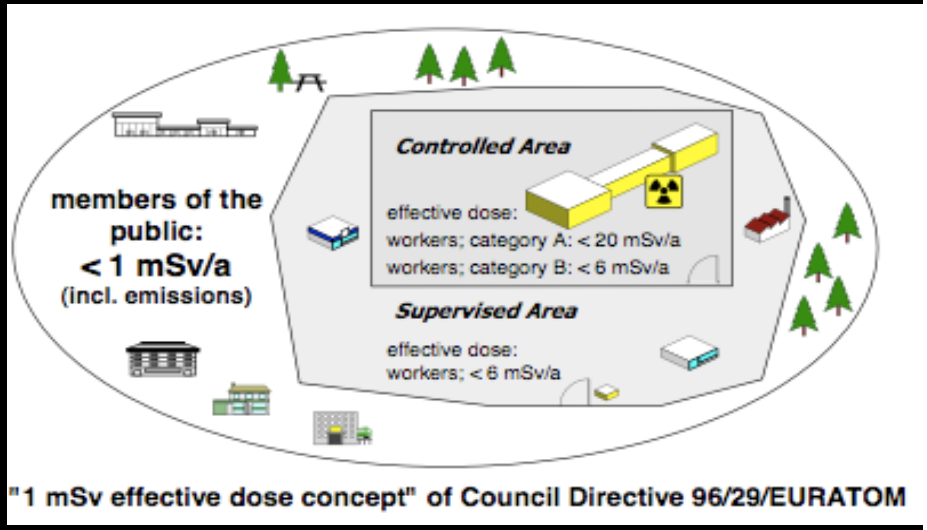
Potable water	EU	0.001 mg/l
	Germany	0.001 mg/l
	Japan	0.0005 mg/l
	USA	0.002 mg/l 2 ppb (EPA, FDA)
Air at work place	France	0.1 mg/m³ inorganic, skin, long-term limit (8 h)
	United Kingdom	0.05 mg/m³ long-term exposure limit (8 h) 0.15 mg/m³ short-term exposure limit (15 min)
	Germany	0.1 mg/m³ elemental and inorganic Hg 0.01 mg/m³ organic Hg 200 µg/l urine 50 µg/l blood (*)
	Sweden	NGV 0.05 mg/m³, skin
	Japan	0.05 mg/m³ elemental and inorganic Hg 0.01 mg/m³ organic Hg
	USA	0.01 mg/m³ alkyl compounds (organic Hg, 8 h time-weighted average) 0.1 mg/m³ organic Hg (OSHA) 0.05 mg/m³ met. Hg vapor for 8-hr shift and 40 h work weeks (OSHA)

Environmental Standards concerning mercury in different countries

Legislation

Radiological aspects

- Normal Operation → EURATOM directive 96/29



"1 mSv effective dose concept" of Council Directive 96/29/EURATOM

- BUT, formal licensing procedures ≠ acc. countries
  - UK, Germany, Sweden: EURISOL ≠ non-nuclear facility (but if fissiles appear…)
  - France, Belgium, Italy: EURISOL = non-nuclear facility

Legislation

Main conclusions

- Legislation is time and space dependent for radiological aspects (DBA)
  (EU standardization in the future?)

- MMW spallation target: innovative facility
  → formal licensing/authorisation process not completely established in EU

Site proposers should examine if detailed legal basis exists

Conclusion

Main point: Radioactive Hg disposal!

. Could be feasible, but more studies needed
. Expensive

Others points:

. Already in operation (SNS – JSNS)
. (EU) Legislation to be studied in each proposed site
. Comparison: Hg / Pb-Bi (- W?) should be done

CEA - Saclay

EURISOL

Thank you!