Historic experience, lessons learned and future strategies in handling large quantities of Ra-226 in hot cell facilities

2024-05-30

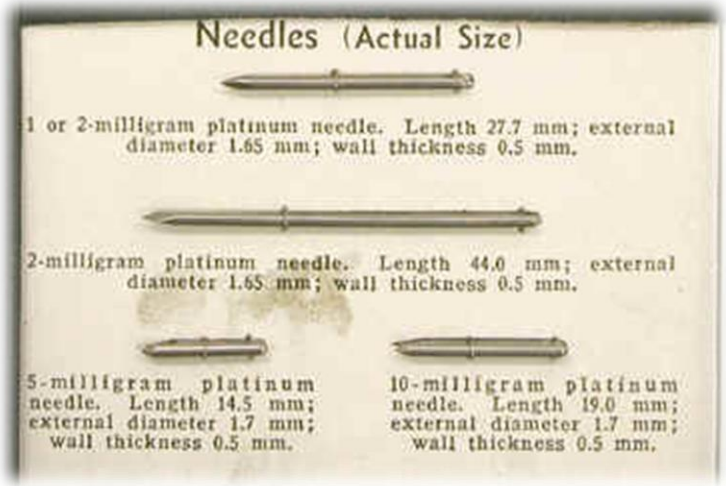
PANTERA

Radium's discovery in 1898 : Marie Skłodowska-Curie

'All medicine that relies on radioactivity – on irradiating people – goes back to Marie Curie.'
Dr. Spencer Weart, former director of the Center for History of Physics

Radium's historic use

- Radium's 2 main real uses were
  - An ingredient in luminescent paint, used for dials in military and civil applications, such as watches, dials in airplanes, exit signs, etc.
  - A source of radiation in cancer treatment
    - external radiation source (curietherapy) more powerful than X-rays (sometimes even the radium daughter radon-222 was collected as an even more potent source of radiation)
    - internal irradiation source (brachytherapy) in the form of needles introduced in the patient's body, in or near the tumor.

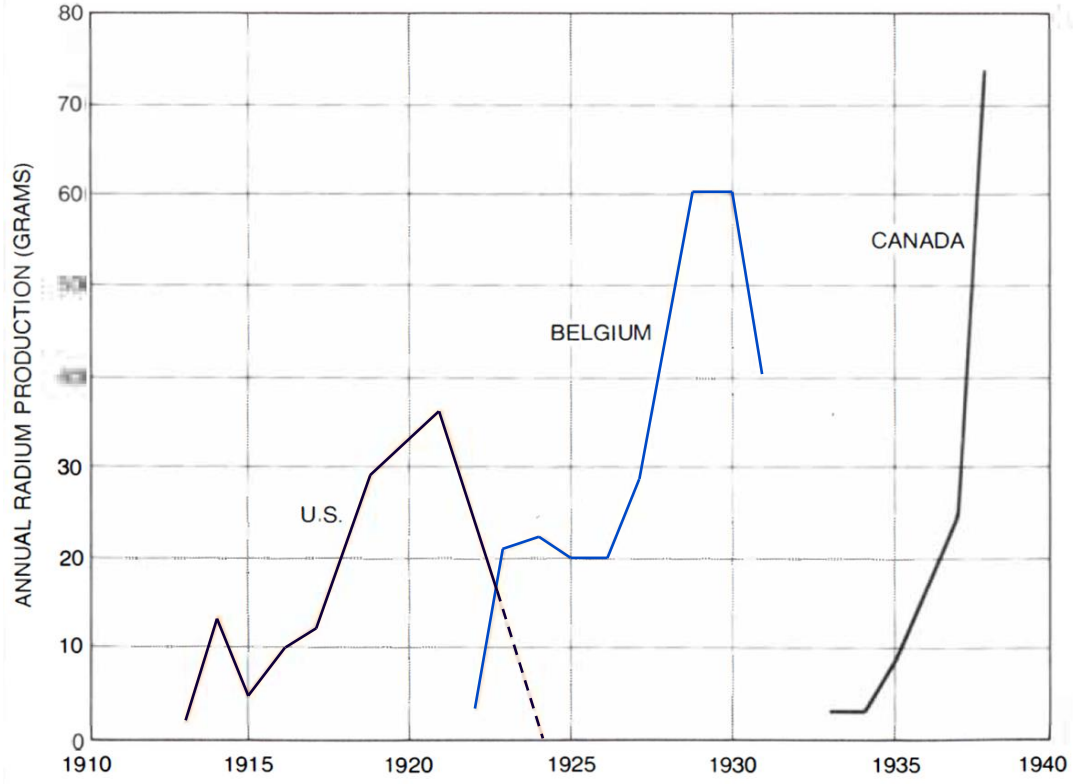


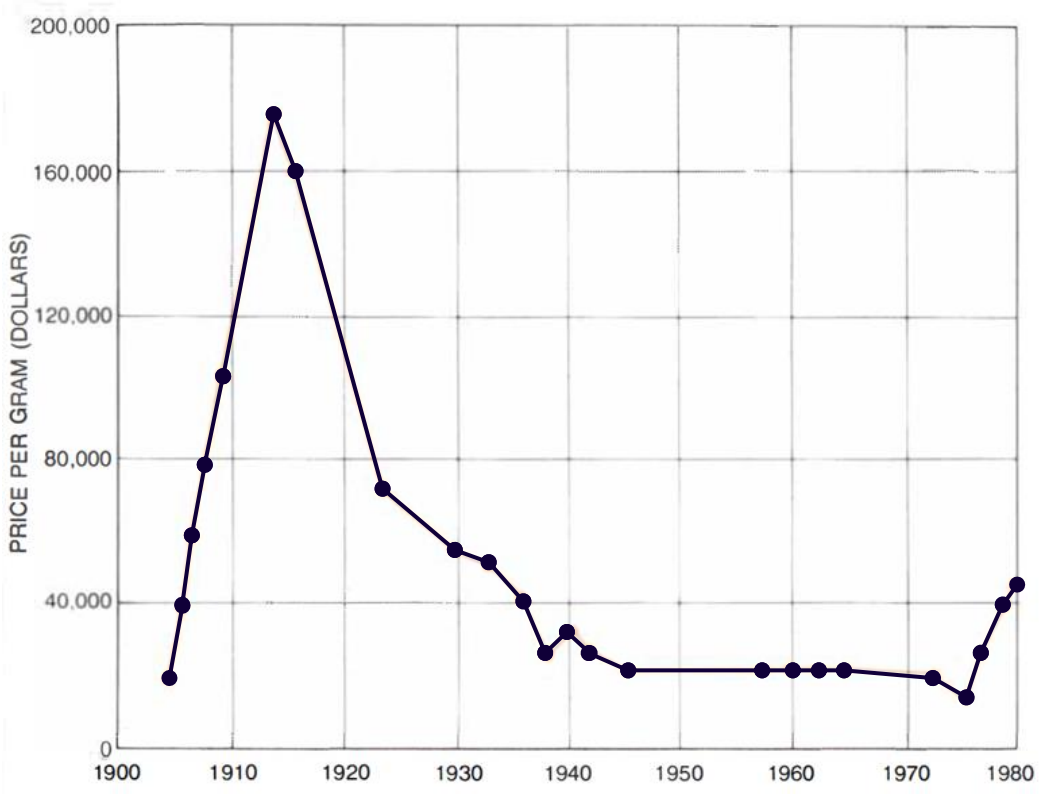
Radium in Belgium…

- Shinkolobwe Uranium mine, Congo
- Radium factory Union Minière, Olen

Radium supply throughout history

- Radium's availability increased from the early 1920's from the Joachimsthal and Colorado mines to the 1930's from the Union Minière Shinkilobwe mines in Belgian Congo (up to 1960) and after 1935 the Canadian Great Bear Lake mines





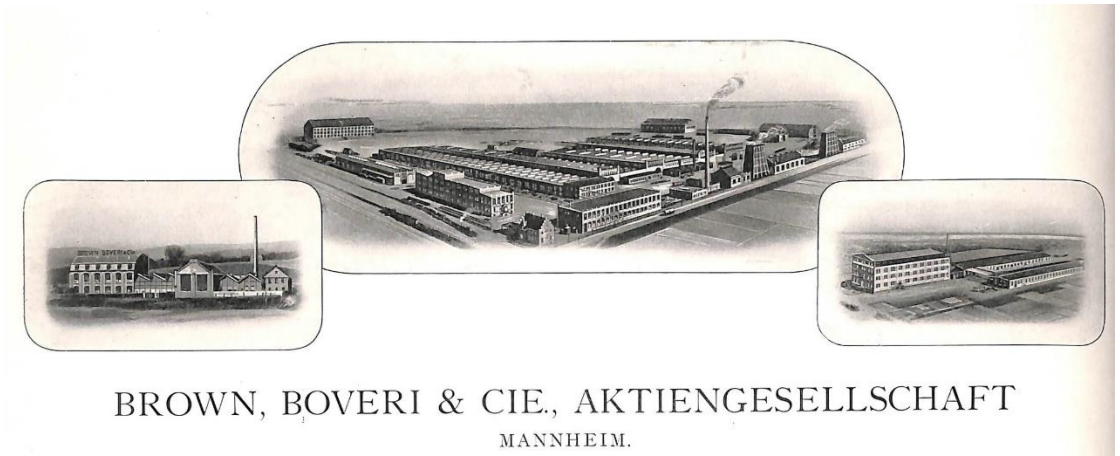
Start of a commercial project

UNION MINIÈRE DU HAUT KATANGA
DÉPARTEMENT RADIUM

SCK·CEN
STUDIECENTRUM VOOR KERNENERGIE
CENTRE D'ETUDE DE L'ENERGIE NUCLEAIRE

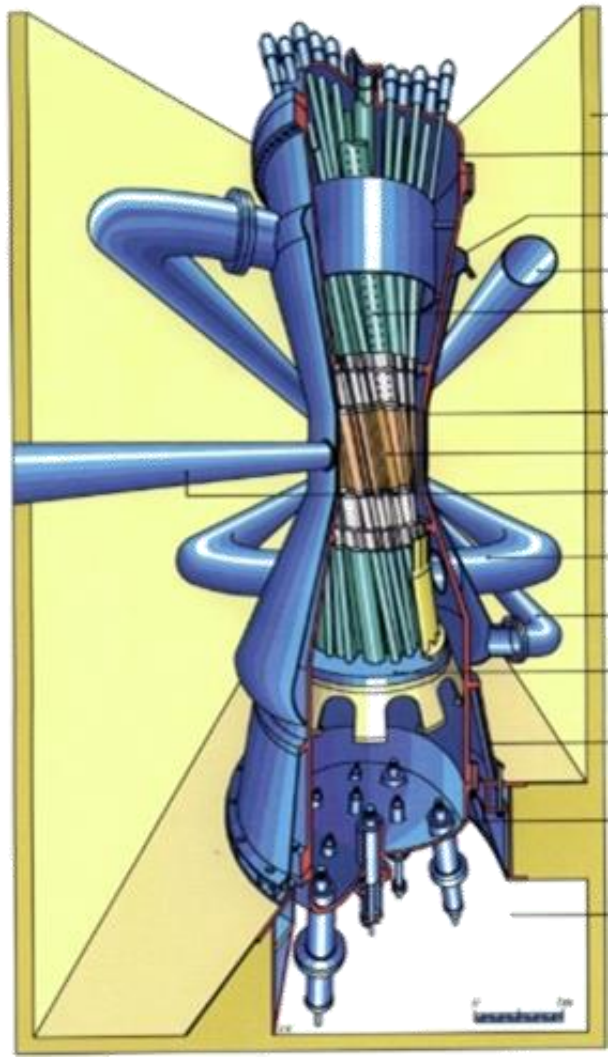
>200g (200 Ci) of pure Ra-226

Production of 20 g Ac-227 and pellet production



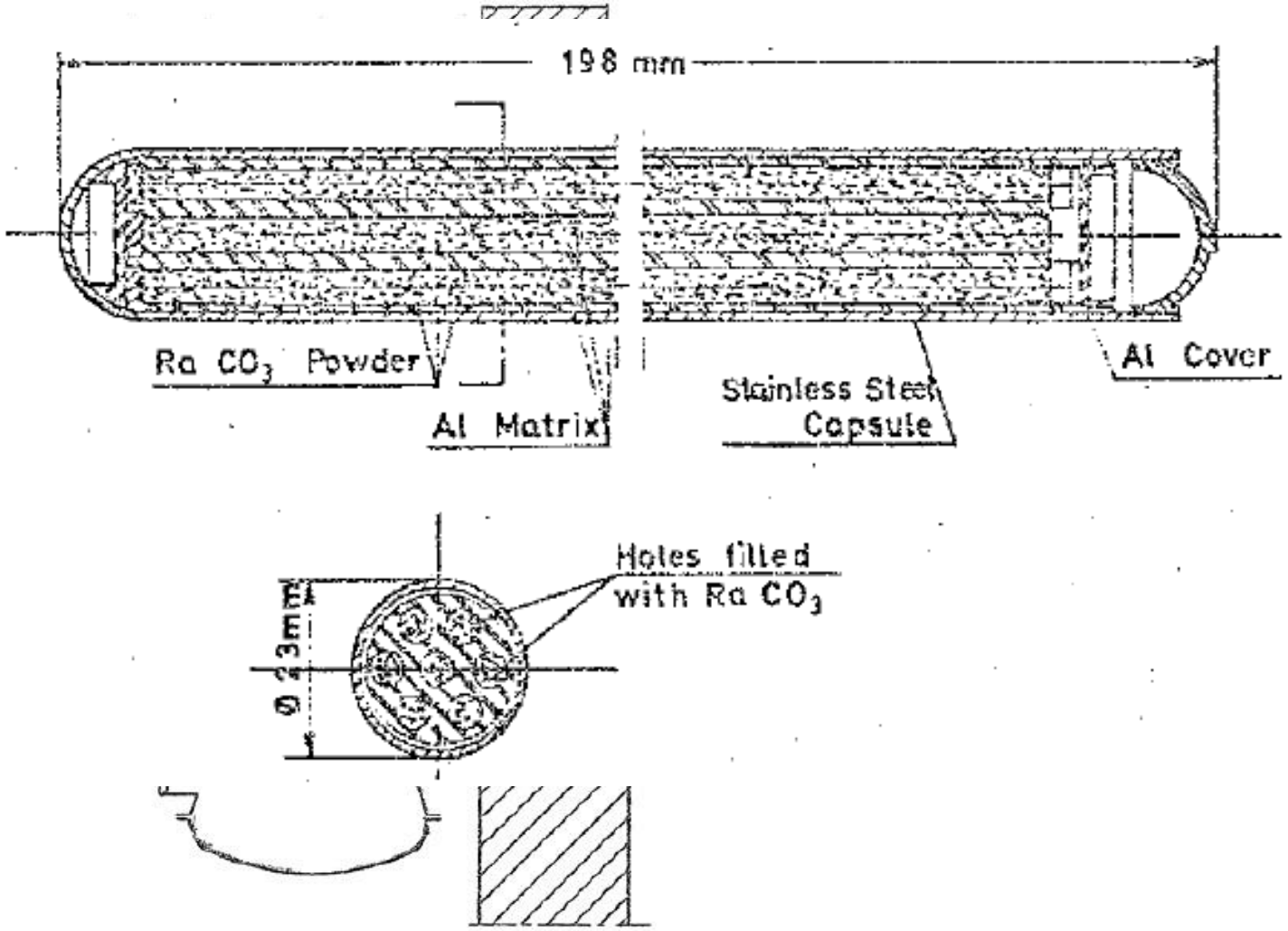
Construction of the Radioisotope Thermoelectric Generator (RTG)

Neutron irradiation of Ra-226



Th 226 30.70 m α 6.337, 6.234... γ 111, (242 131...), e⁻	Th 227 18.697 d α 6.038, 5.978 5.757... γ 236, 50, 256... σ_f 200	Th 228 1.9125 a α 5.423, 5.340... γ 84, (216...), e⁻ O20 σ 120, σ_f < 0.3	Th 229 7.0 μs / 7920 a IT (0.008) e⁻ α? α 4.845, 4.901 4.815... γ 194, 86, 211 31..., e⁻ σ 62.8, σ_f 30.8
Ac 225 9.920 d α 5.830, 5.793 5.732..., C14 γ 100, (150, 188 63...), e⁻	Ac 226 29.37 h β⁻ 0.9, 1.1 ε, α 5.40 γ 230, 158, 254 186...	Ac 227 21.772 a β⁻ 0.04... γ (38,...), e⁻ α 4.953, 4.941... γ (100, 160...), e⁻ σ 880, σ_f < 3.5E-4	Ac 228 6.15 h β⁻ 1.2, 2.1... γ 911, 969, 338 965...
Ra 224 3.6319 d α 5.6854 5.4486... γ 241..., C14 σ 12.0	Ra 225 14.9 d β⁻ 0.3, 0.4 γ 40, e⁻	Ra 226 1600 a α 4.7843 4.601... γ 186..., C14 σ 12.8, σ_f <5E-5	Ra 227 42.2 m β⁻ 1.3... γ 27, 300, 303 284, 330..., e⁻

$RaCO_3$ irradiation in BR2



[Baetsle et al. Power from Radioisotopes, 1972]]

Over 50 years ago...

S.C.K./C.E.N.
Scheikunde
S71/40/2113/4/DH/jv/73

Produktie Ac-Th uit Ra

	Nr caps	Plaats in BR2	Cyclus-duur	^{226}Ra in Ci	Ac in Ci	Th in Ci	Omzettings-coefficient $\frac{Ac}{Ra}$Ci	$\frac{Th}{Ra}$Ci	Datum in BR2
1969...	1	E 330	1	25,2	8,0	11,0	0,32	0,44	27/11
	2	G 300	1	± 30	10,0	12,5	0,33	0,42	20/12
1970...	3	G 300	1	± 24					

1000Ci hotcell

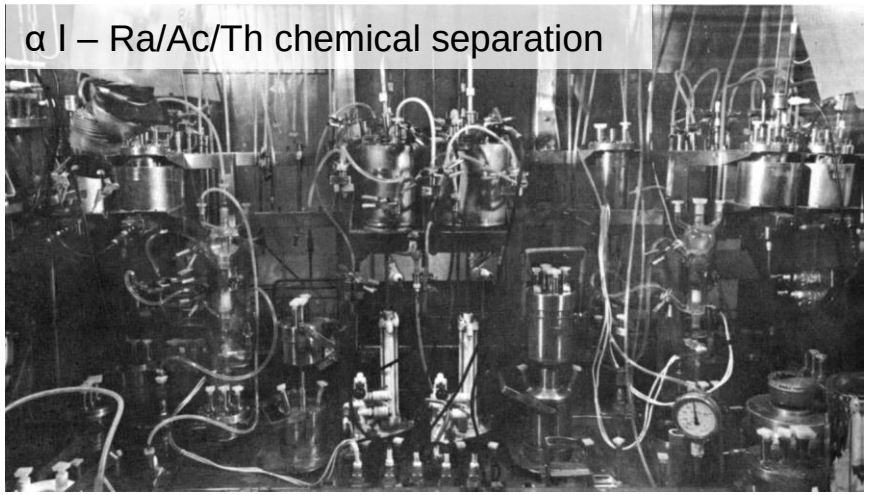
Shielding of the αI, αII and αIII boxes for :

- Ra/Ac/Th separation
- Ra recycling
- Ra capsule welding

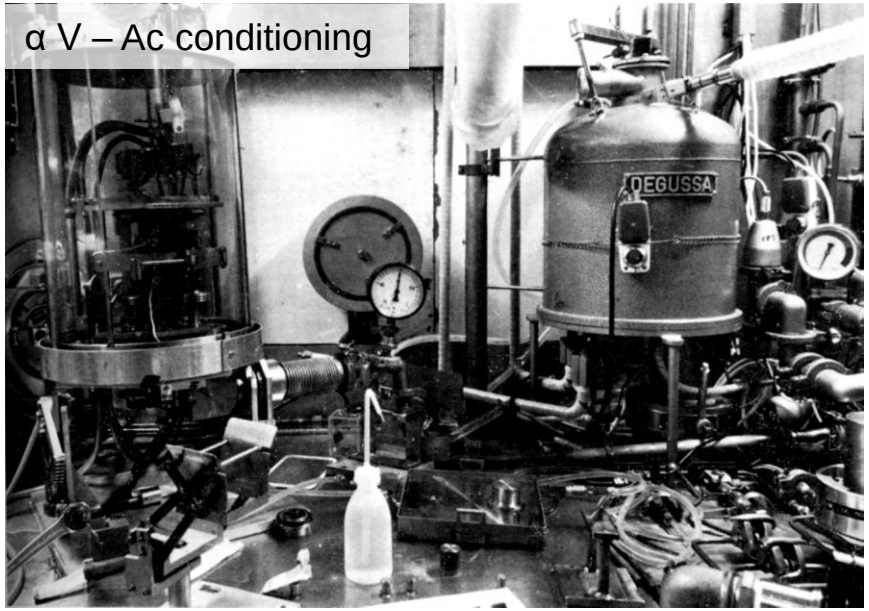
[Annual Scientific Report 1969]

Th-Ra-Ac separation…

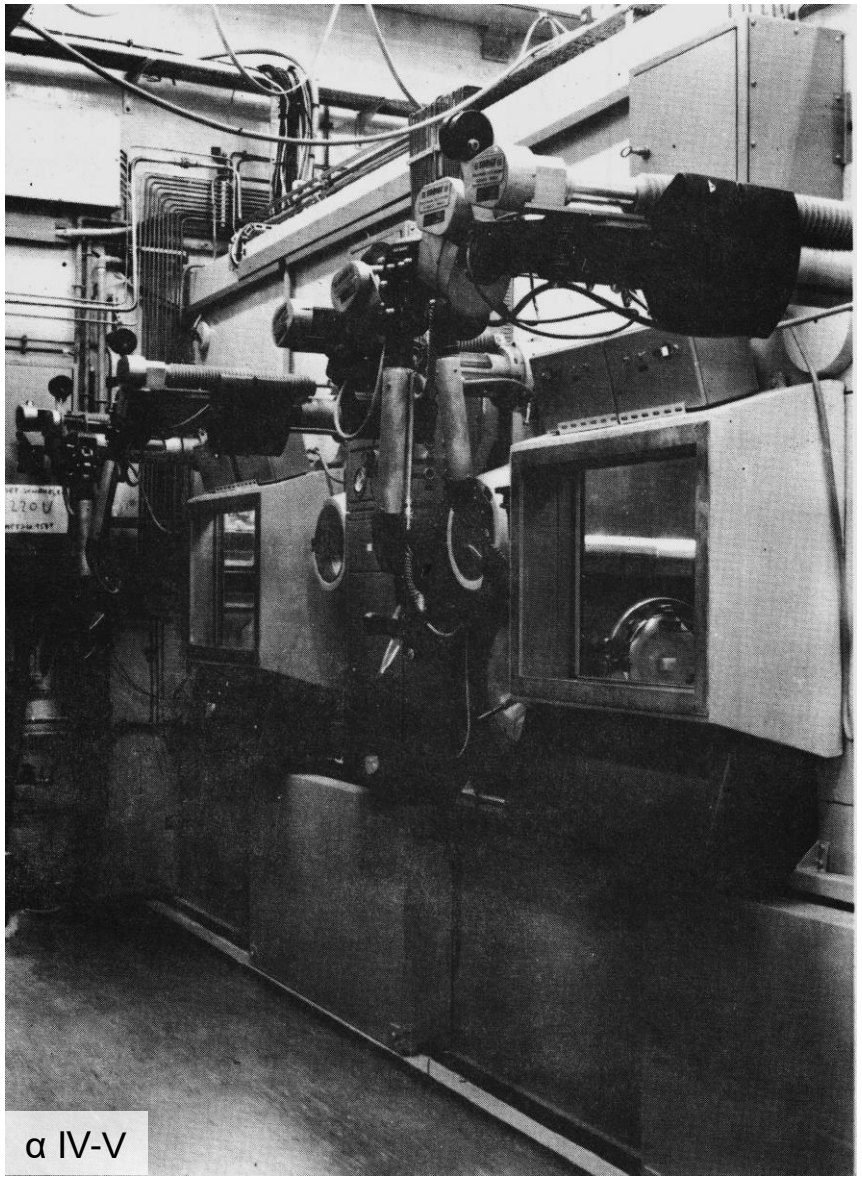
α I – Ra/Ac/Th chemical separation



α V – Ac conditioning

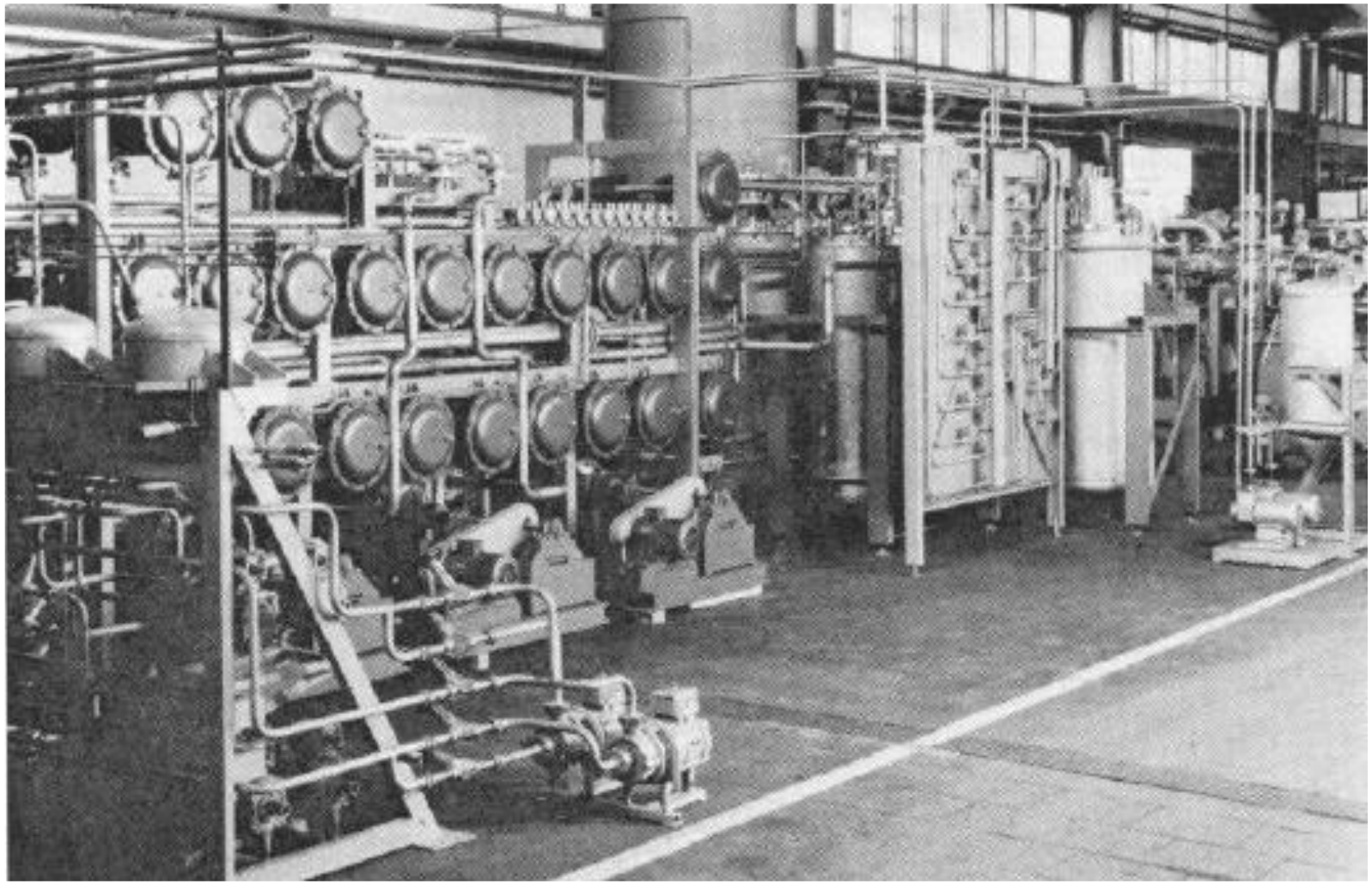


[Annual Scientific Report 1969]

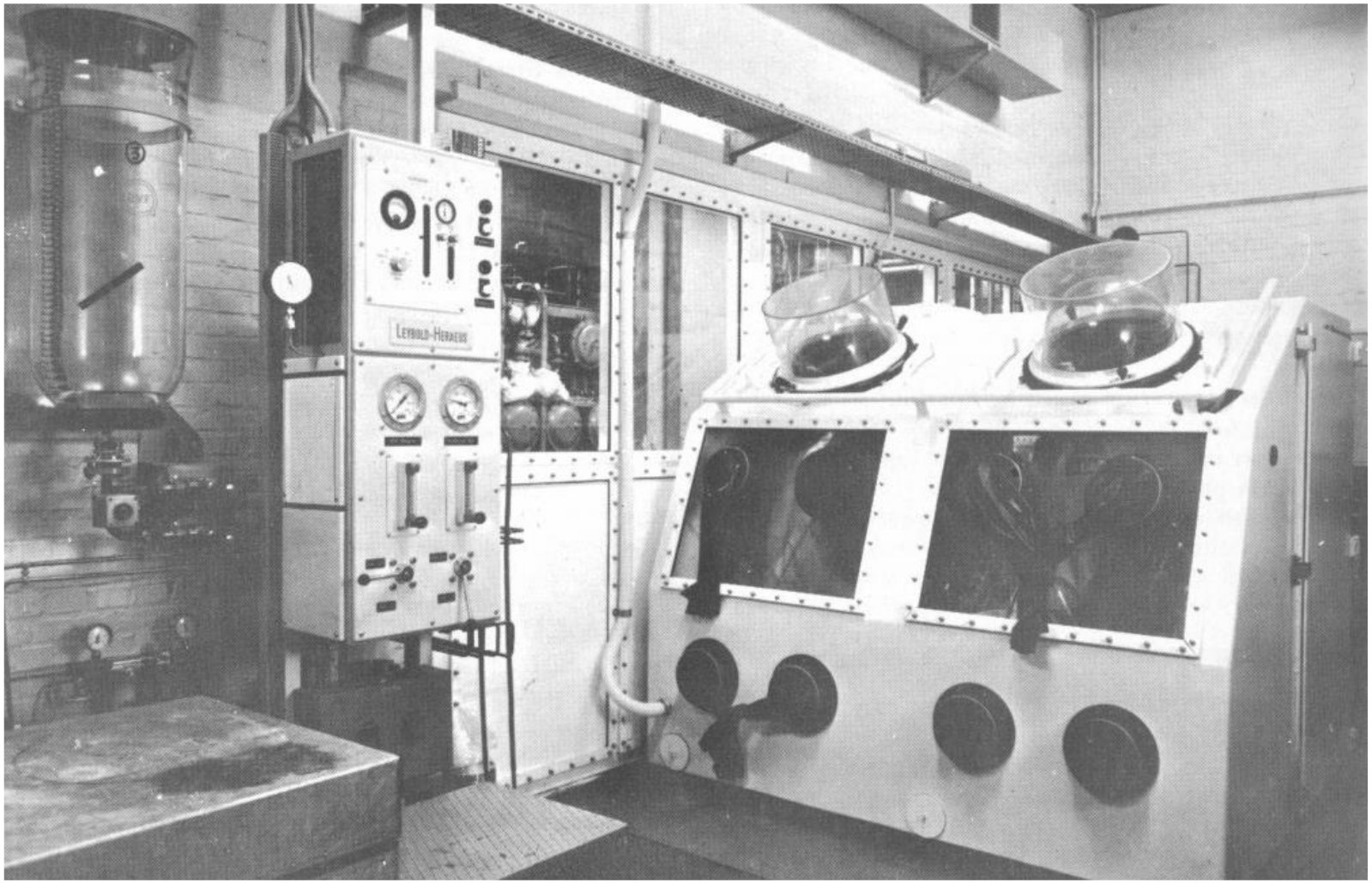


α IV-V

Radon Trapping Unit (Rn-222)

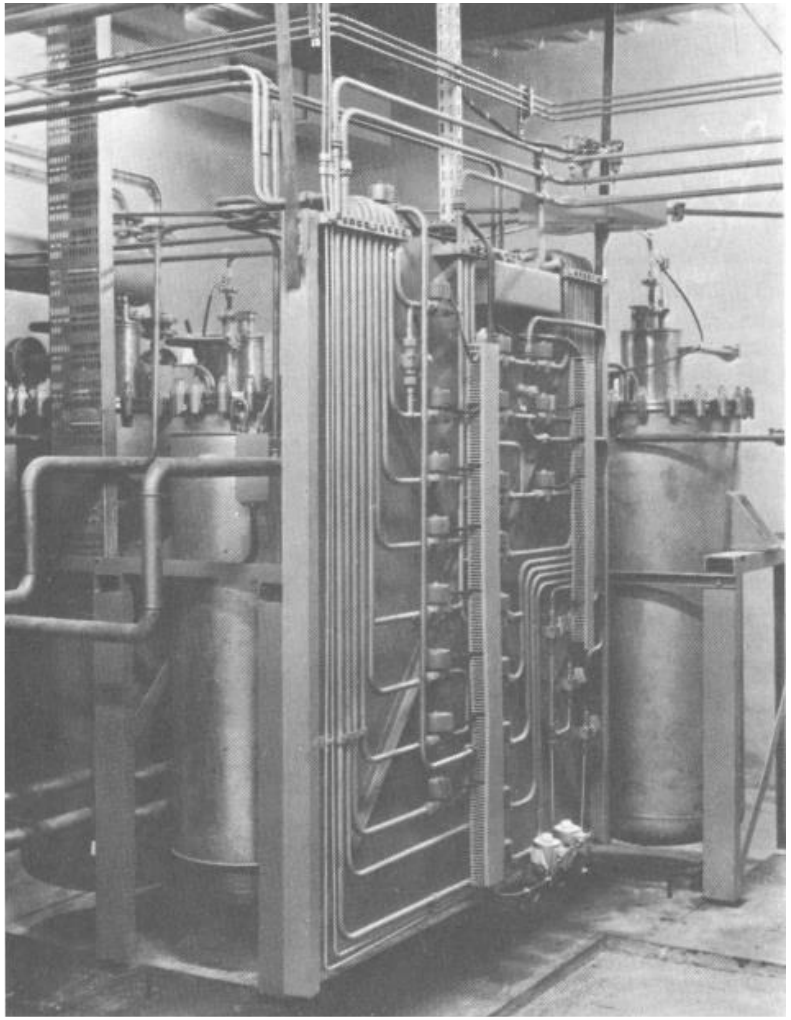


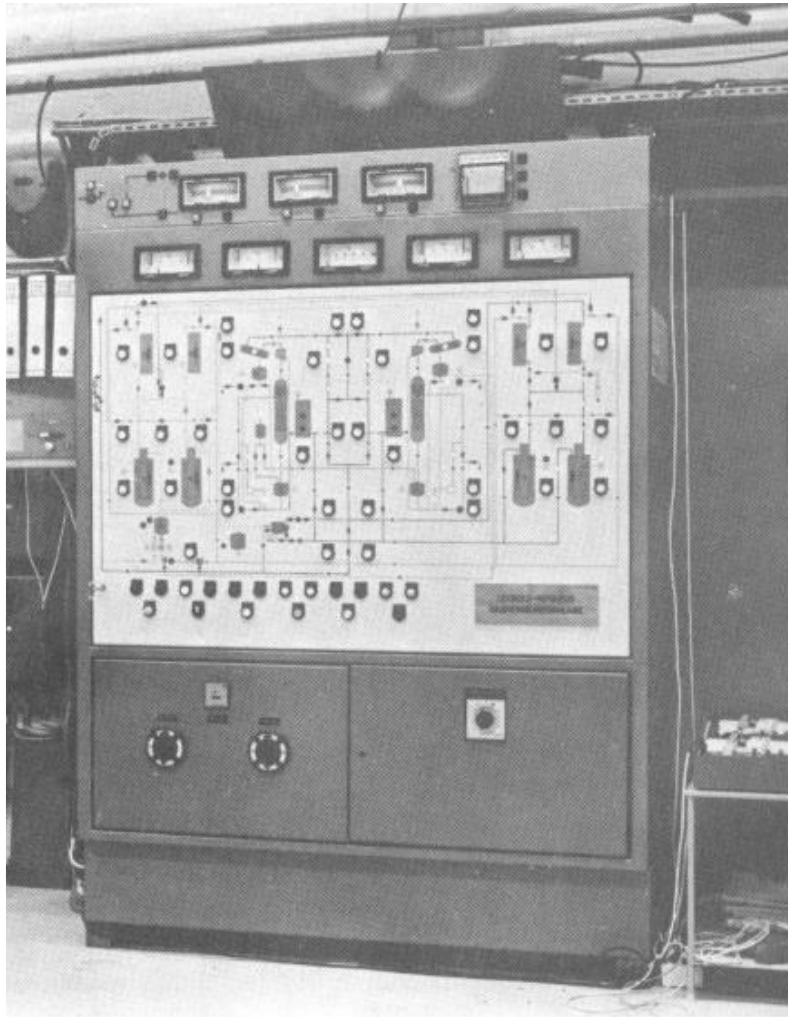
Radon Trapping Unit (Rn-222)



[Annual Scientific Report 1969]

Radon Trapping Unit (Rn-222)





[Annual Scientific Report 1969]

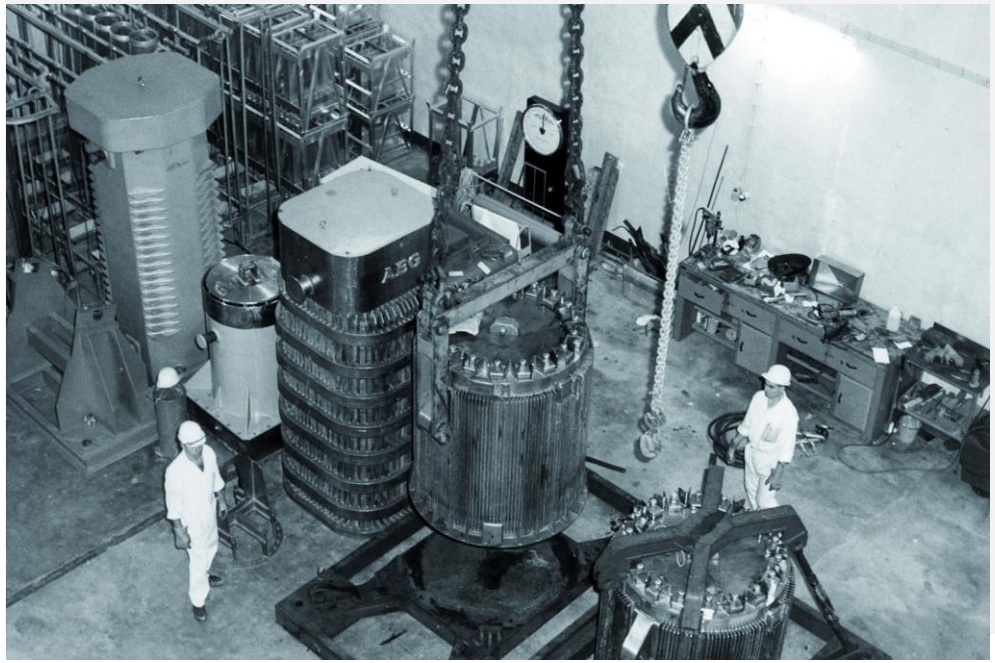
1974…Stop of production

- Production of Ac-227 stopped because of:
  - Low transmutation yield of Ra to Ac
  - Corrosion in cells αI + αII
  - Ac_2O_3 pellet production issues
  - Economic crisis
- All Ac-227, Th-228 and Ra-226 stored at BR1 in sealed capsules
- Waste stored

PUBLIC

Belgoprocess

- Founded after EUROCHEMIC stopped reprocessing operations
  - **Today ~ 500 employees in Dessel & Mol site**
  - **Subsidiary of ONDRAF/NIRAS**
- Core activities: waste management & decommissioning

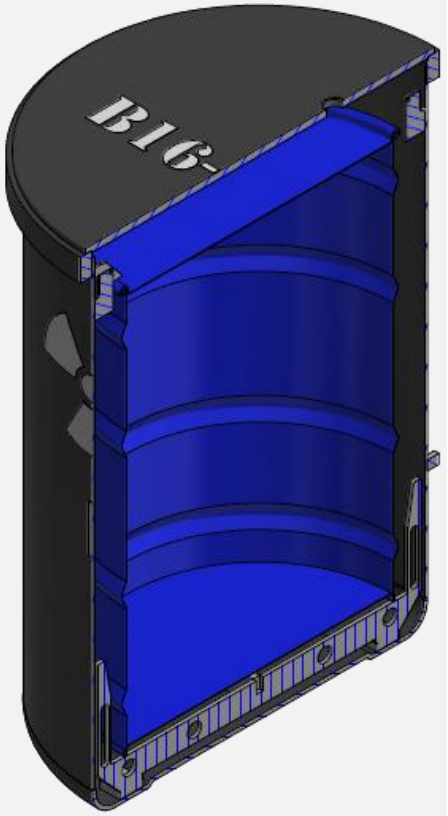


Origin of radium waste

- **Rich history**
  - Decommissioning of the Union Minière radium factory
  - Actinium program at SCK-CEN
  - Decommissioning of radium contaminated waste treatment facilities
- **Low Level Solid Waste**
  - max 2 mSv/hr / 4 GBq
  - 4000 drums
- **Intermediate Level Solid Waste**
  - Ra/Ac-227 process waste (metal, glass, plastic)
  - 100 mg up to 20 g Ra per package
  - 3-400 shielded packages
- **Sources**
  - Wide range: 10 mg needles – 10 gram capsules

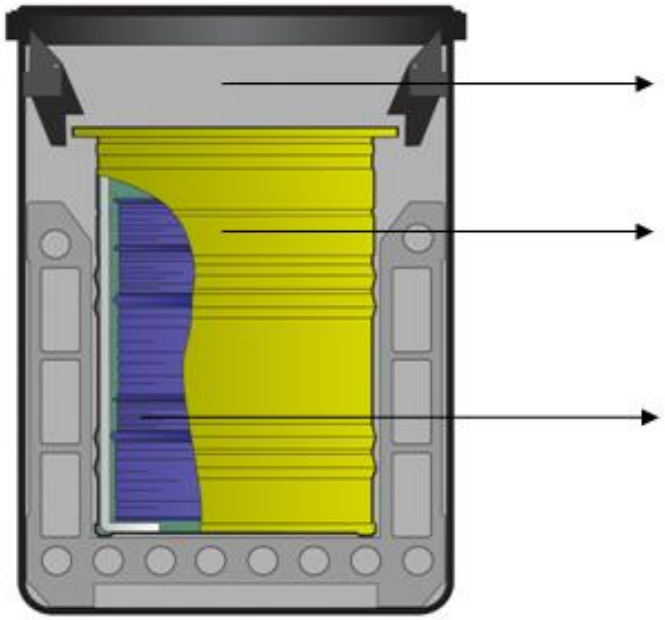
Processing of radium waste

- **Low Level Solid Waste**
  - Sorting, shredding and packing in PE/EVOH bags → radon management
  - Organic fraction → incineration (feed < 400 MBq/m³ - 10 mg Ra/m³)
  - Inorganic fraction → super-compaction & encapsulation
  - Interim storage



Processing of radium waste

- **Intermediate Level Solid Waste**
  - Remote handling
  - Compaction & immobilization in mortar
  - Robotic welding station
- **Sources**
  - Collection in shielded waste packages
  - Welding and immobilization



Challenges when handling of radium waste

- **PPEs → continuous radon monitoring in all critical areas**
  - > 300 Bq/m³ → full face mask (P3 or AC) + Tyvek
  - > 3000 Bq/m³ → overpressure suit + mask
  - Nose swap – whole body counting
- **Dosimetry**
  - Significant dose rates – good work preparation/training for high risk entries
  - ALARA: time & distance, time & distance…
  - Monitoring: body, extremities, eye & tele-dosimetry
- **Radon**
  - High renewal rate → avoid accumulation of daughter products
  - Keep Ra and TRU operations strictly/physically separated
  - Atmospheric conditions during discharge

Challenges when handling of radium waste

- **Hydrogen buildup**
  - Opening of old (waste) packages:
    - Pressure buildup cannot be excluded – examples at Belgoprocess
    - Remote drilling, venting and gas measurement before hands-on operations
  - New waste packages:
    - anaerobic corrosion / radiolysis → exclude water/organics

New Applications…

A valuable radioisotope for cancer treatment

ACPIL, a challenging project with strict conditions

Ac 227
21.772 a
β^- 0.04…
γ (38,…), e^-
α 4.953, 4.941…
γ (100, 160…), e^-
σ 880, σ_f < 3.5E-4

[Highlights 2015]

New opportunities in cancer treatment

Valuable capsules with thorium for Targeted Alpha Therapy

Th 228	Th 229	
1.9125 a	7.0 µs	7920 a
α 5.423, 5.340…	IT	α 4.845, 4.901
γ 84, (216…), e^-	(0.008)	4.815…
O20	e^-	γ 194, 86, 211
σ 120, σ_f < 0.3	α?	31…, e^-
		σ 62.8, σ_f 30.8

The past can hold v… 1970s…
SCK•CEN produce…
the rare research ir…
a scarce and pror…
project is picking…
contribution to c…

Thorium-229 (Th-…
atomic clocks, fr…
isotopes actiniu…
a great deal of…
encouraging, h…
of Th-229 is…
demonstratin…
in the historical sourc…
to initiate R&D into radiopharmaceu…
specifically interested in the decay product Ac-…
for direct applications and for the creation of a generator for…
Bi-213.

ELSEVIER

Applied Radiation and Isotopes 120 (2017) 40–44

Contents lists available at ScienceDirect

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journal homepage: www.elsevier.com/locate/apradiso

Thorium-229 quantified in historical Thorium-228 capsules

Sven Boden[a], Koen Vints[a], Simone Cagno[a,*], Dominic Maertens[a], Karen Van Hecke[a], Thomas Cardinaels[a,b]

[a] Belgian Nuclear Research Centre (SCK•CEN), Boeretang 200, B-2400 Mol, Belgium
[b] KU Leuven, Department of Chemistry, Celestijnenlaan 200F, P.O. Box 2404, B-3001 Heverlee, Belgium

CrossMark

[Highlights 2016]

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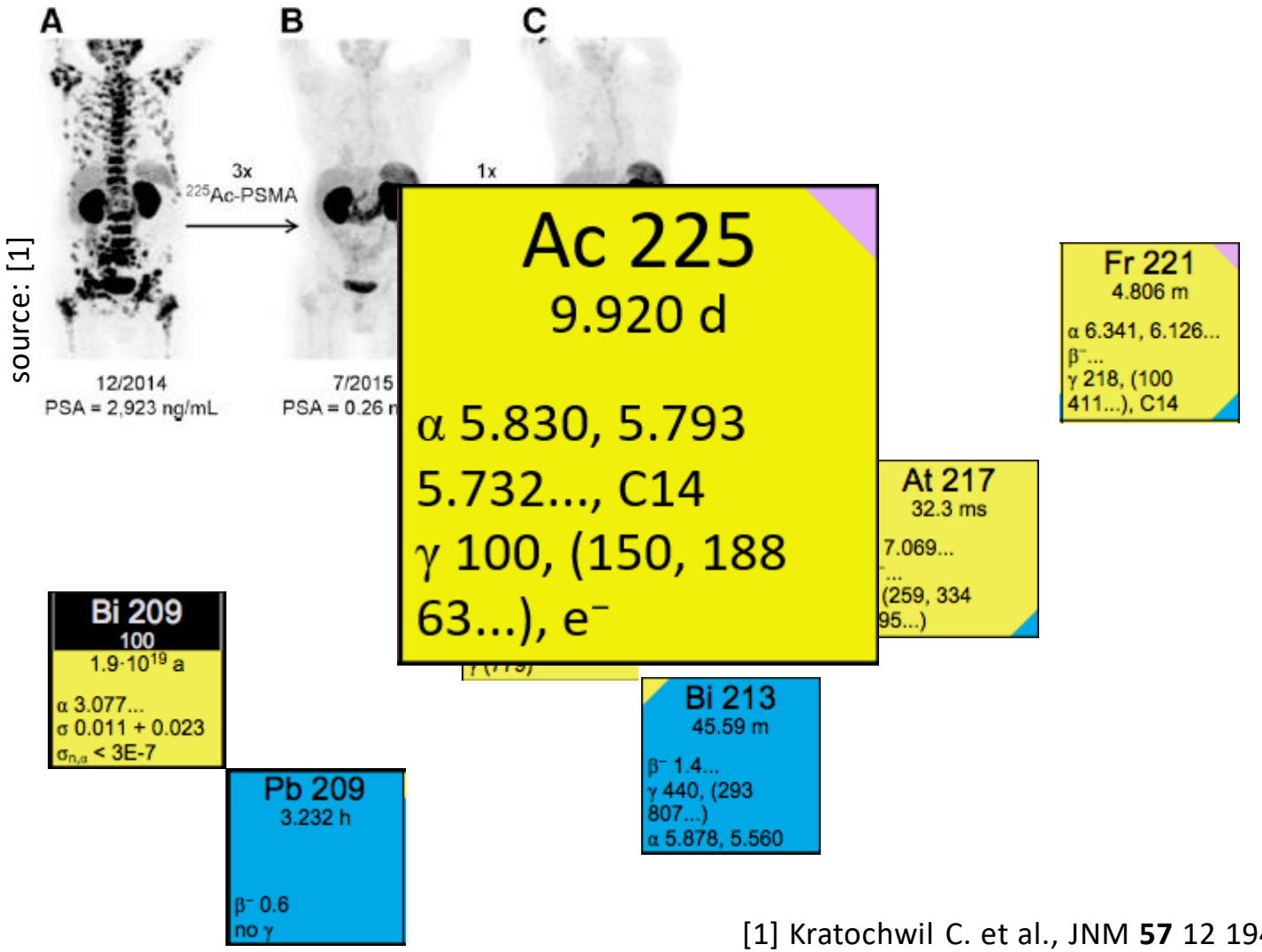
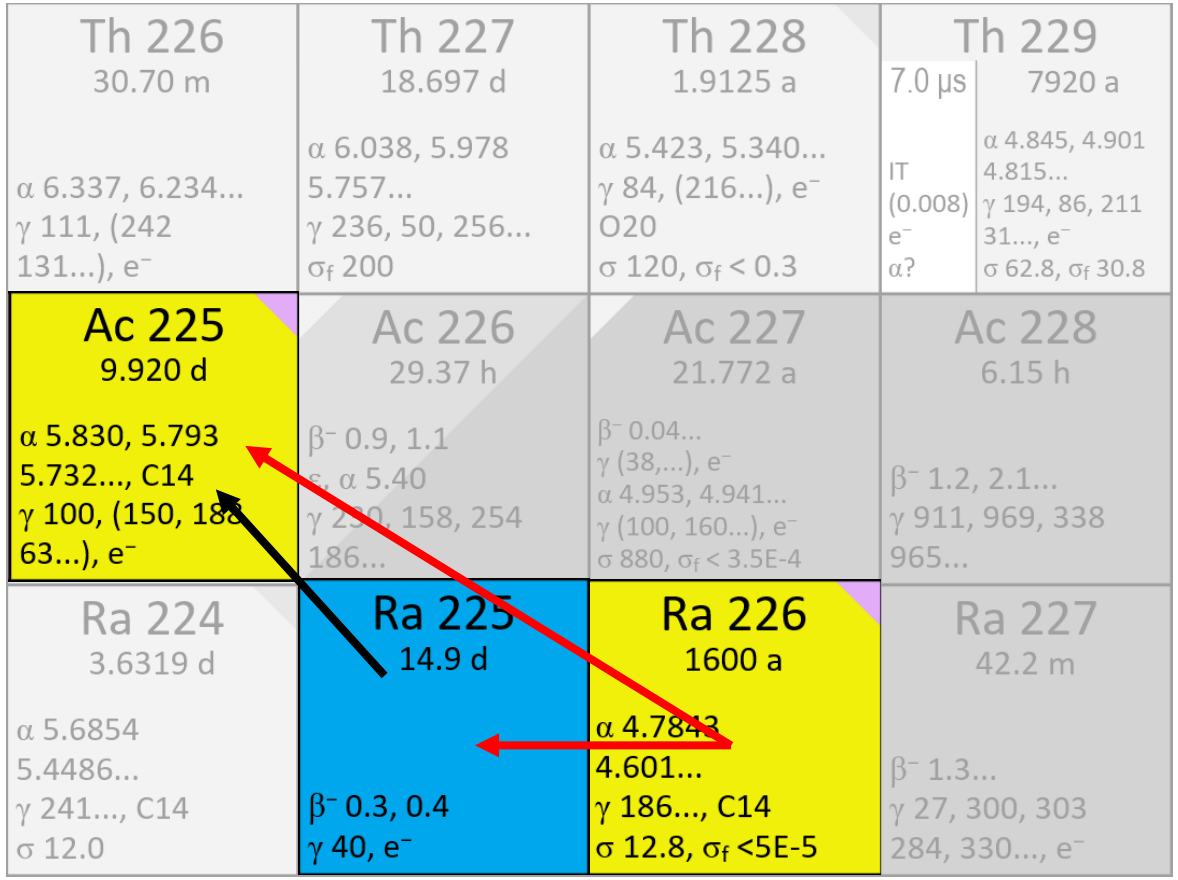
PanTera's target: ^{225}Ac



[1] Kratochwil C. et al., JNM **57** 12 1941 (2016)

Ac-225 by irradiation of Ra-226, but not with neutrons

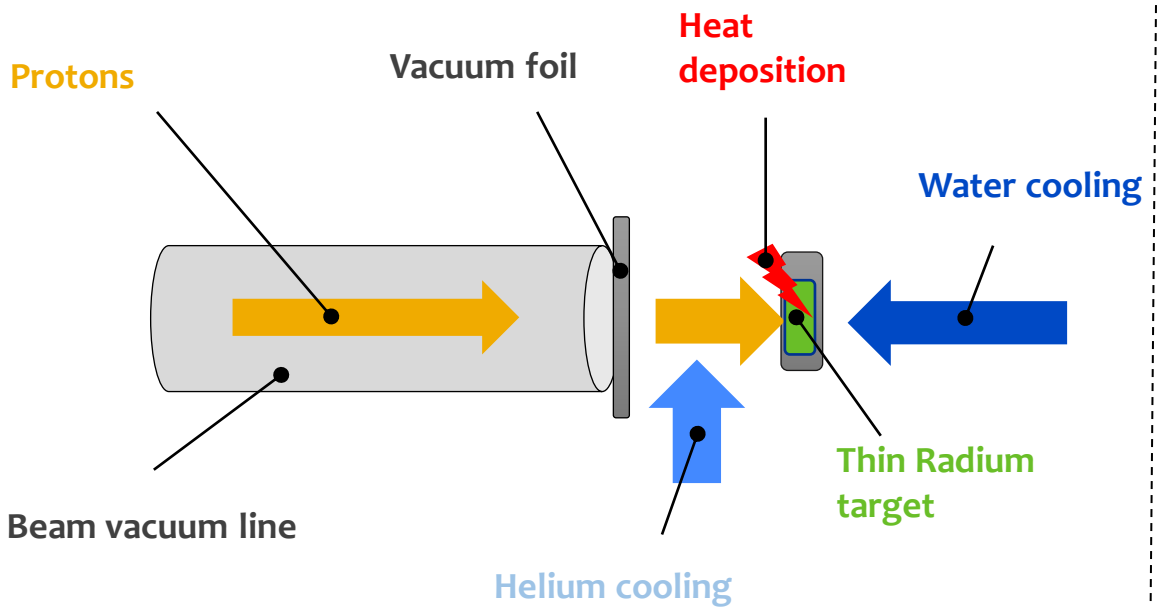


direct (p,2n)

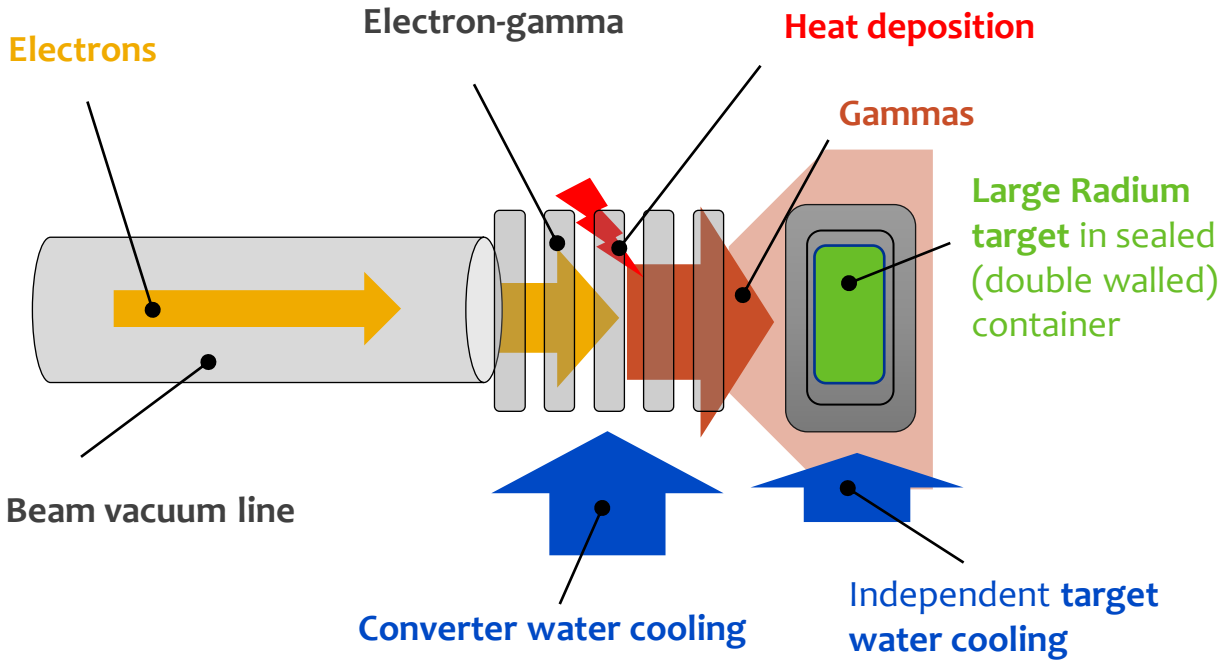
indirect (γ,n)

^{225}Ac – Proton vs. Gamma route: safety first

PROTON ROUTE



GAMMA ROUTE



−	Radium containment	+
−	Power deposition	+
−	Cooling	+
−	Failure impact	+

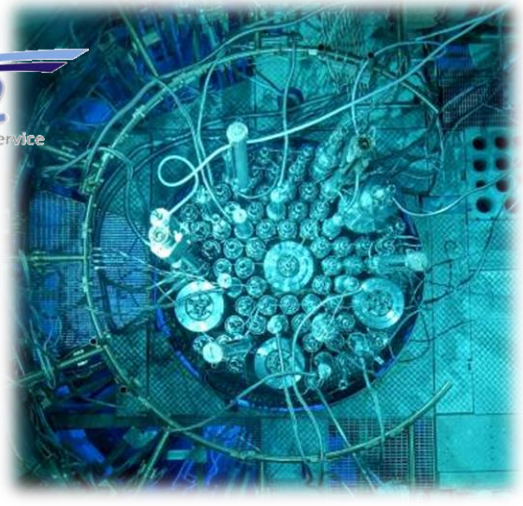
PUBLIC

PANTERA

A BETTER FIGHT FOR LIFE

BR 2 @ your service

PUBLIC



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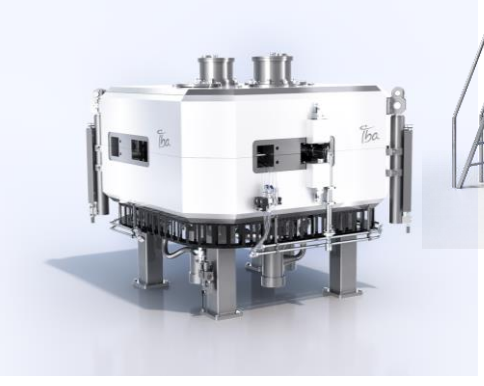
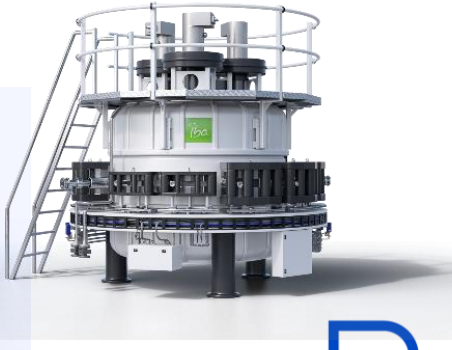
iba



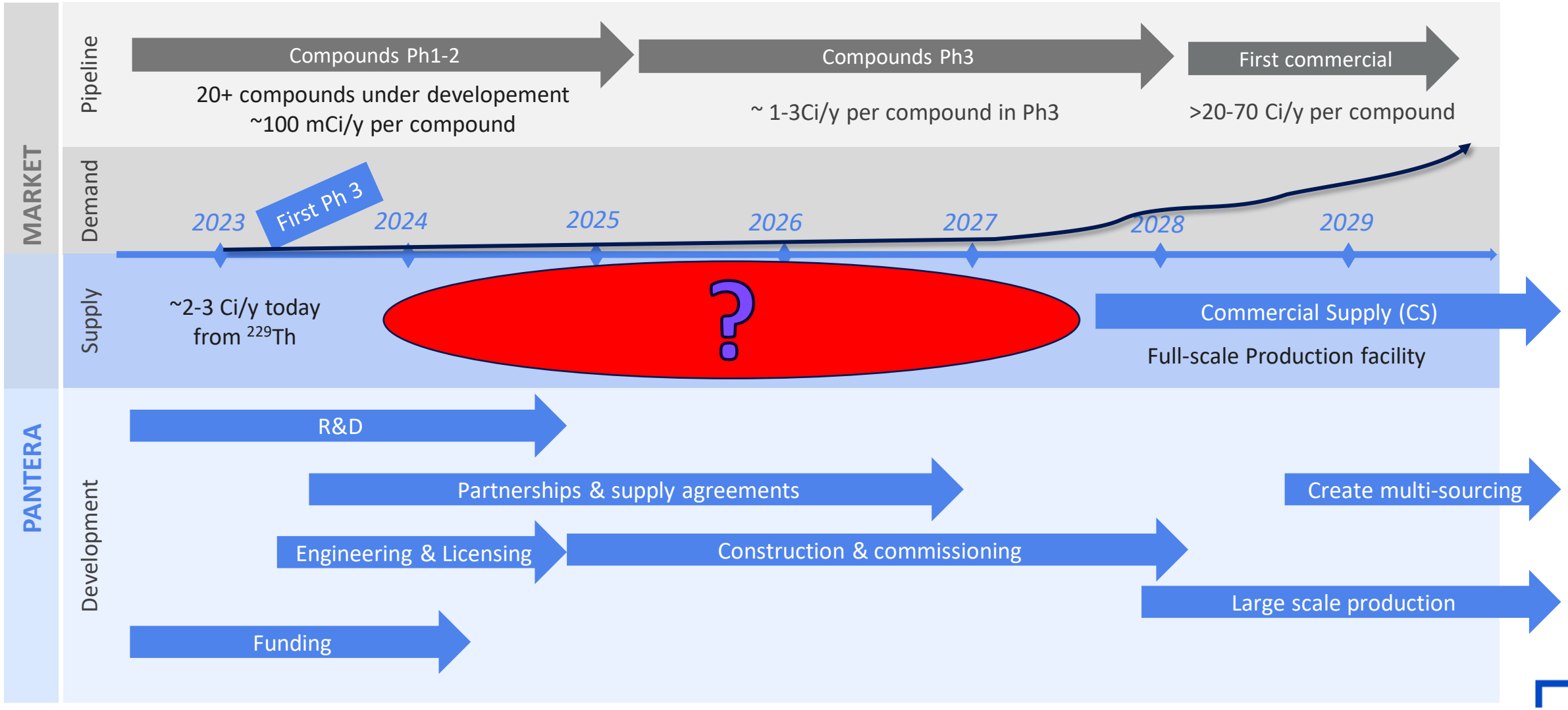


PanTera movie

https://youtu.be/sPhyZc9korc

https://pantera-life.com/

PanTera's Timeline



TerraPower Isotopes & PanTera announce partnership

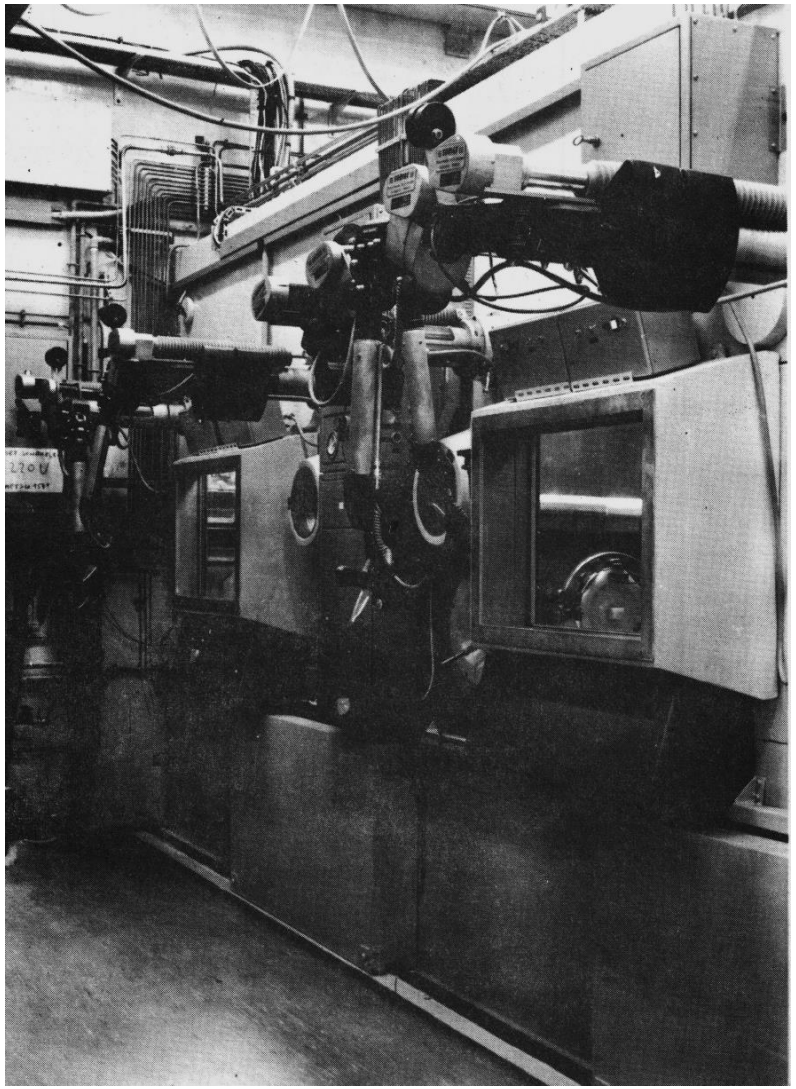
TERRAPOWER ISOTOPES

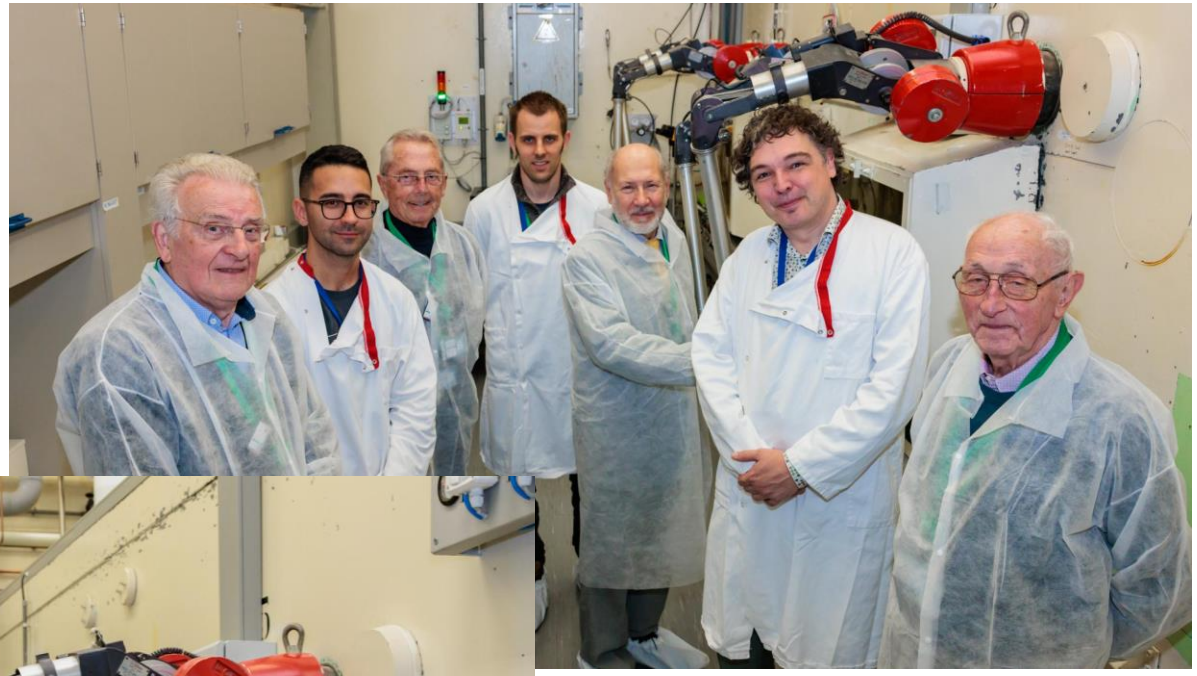
PANTERA

join forces to increase availability of Actinium-225

- PanTera will use TerraPower Isotopes's Thorium and generator technology to **increase near-term production in Europe** to support development & clinical trials
- TerraPower Isotopes will use PanTera's technology to ensure **large-scale supply is available in the US** for commercial phase

Th-Ra-Ac separation…





Setting up a Thorium route in Belgium

PANTERA

Thorium generator

- Two dedicated existing hotcells
- Thorium handling capabilities
- Radon trap (Rn-220)

Internal transport

Actinium final purification

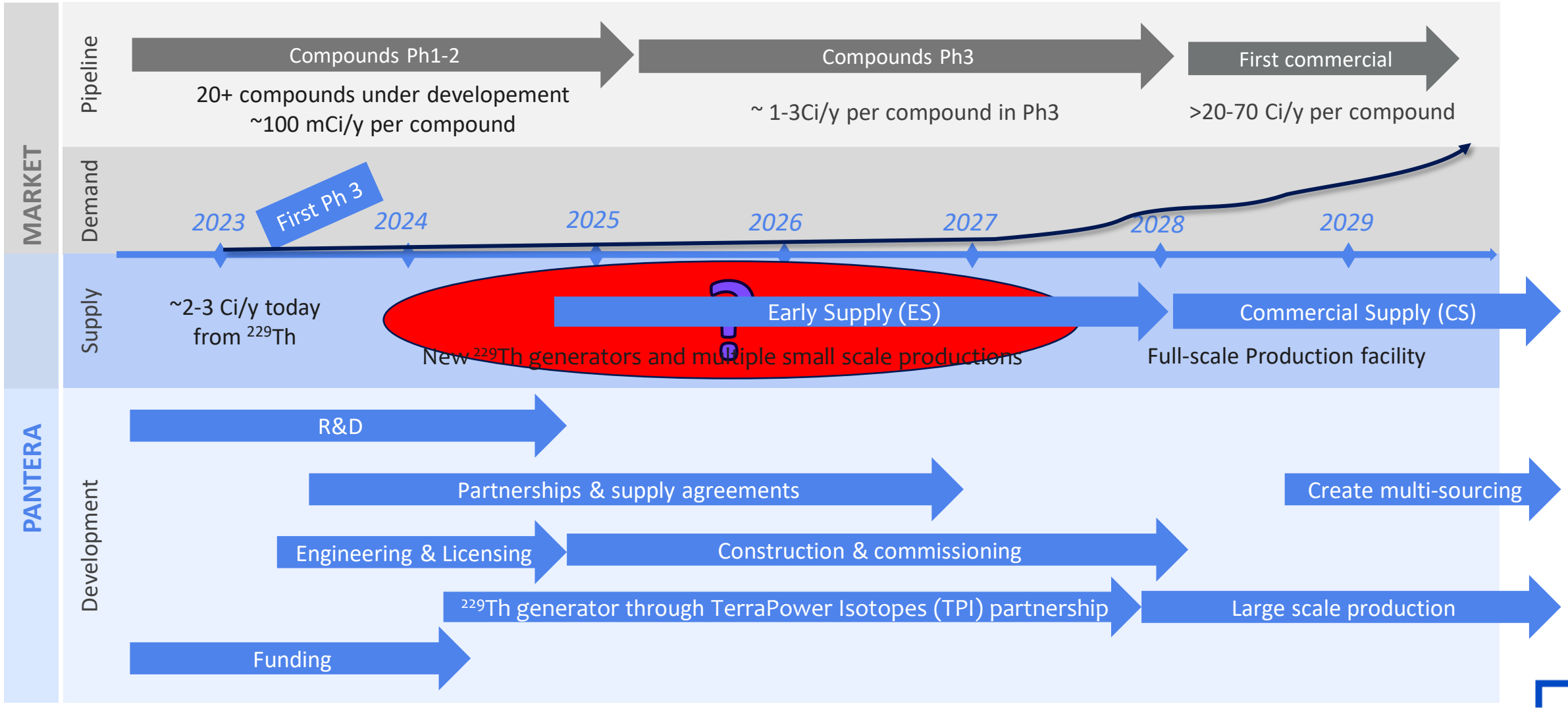
- Clean room environment with hotcell
- Last step purification
- Dispensing
- Transportation

External transport

- GMP-grade material
- Type-A package

sck cen

PanTera's Timeline



PanTera’s building – artist impressions

A Better Fight for Life by providing the know-how, services, material and infrastructure needed to **supply the rarest drugs** in the world, enabling the application of nuclear radiation as a dependable source of hope, as a precision tool for medicine which **attacks the disease**, not the patient

- PanTera