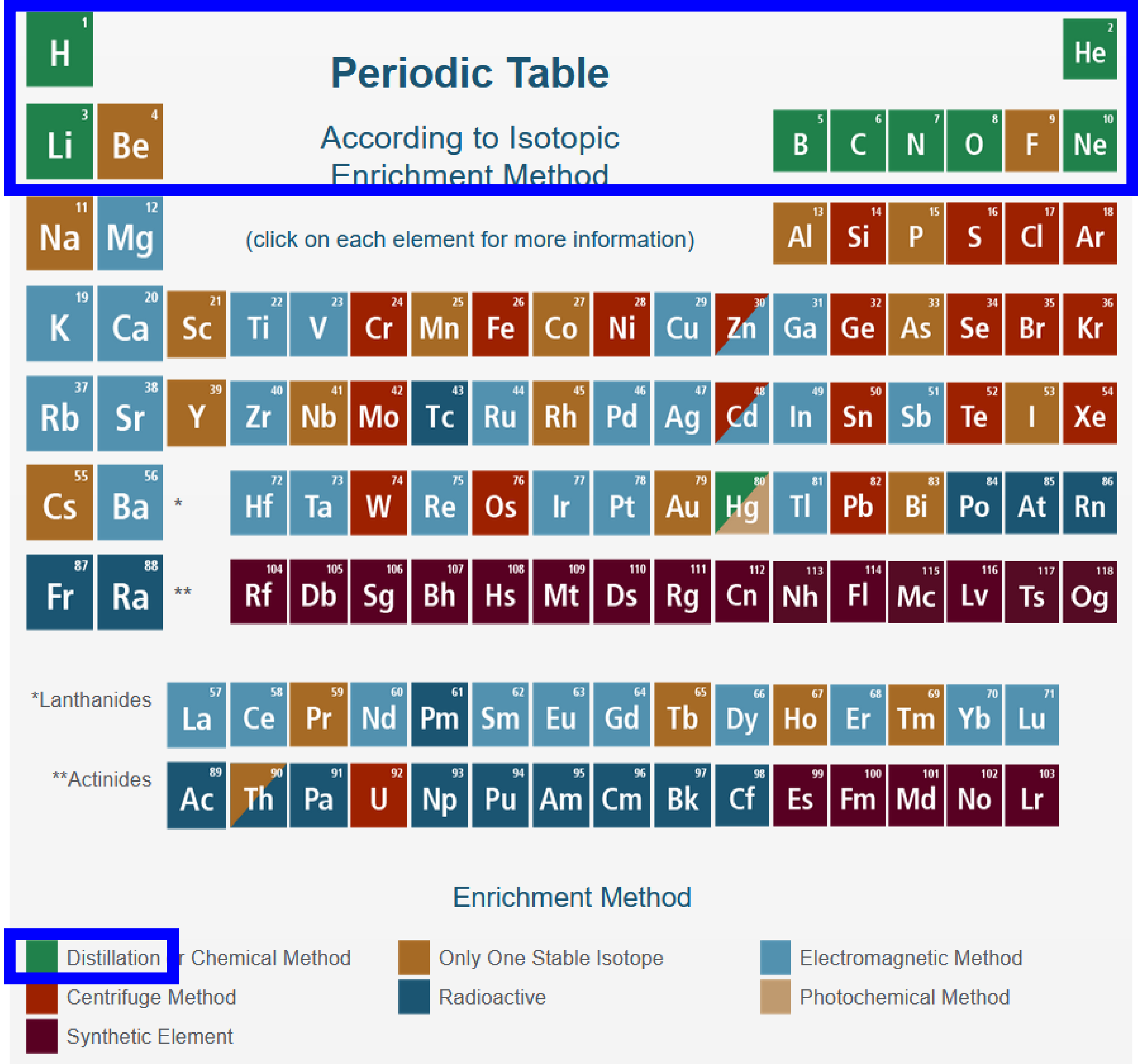
Mass separation of stable and radioactive lanthanide isotopes

Ulli Köster
Institut Laue-Langevin & UGA
Grenoble, France

Radiolanthanides Workshop, 3-5 September 2024

Disclaimer

No relation to the Tinner family
nor their customers.



Periodic Table

According to Isotopic Enrichment Method

(click on each element for more information)

Enrichment Method

- Distillation r Chemical Method
- Centrifuge Method
- Synthetic Element
- Only One Stable Isotope
- Radioactive
- Electromagnetic Method
- Photochemical Method

ISOFLEX

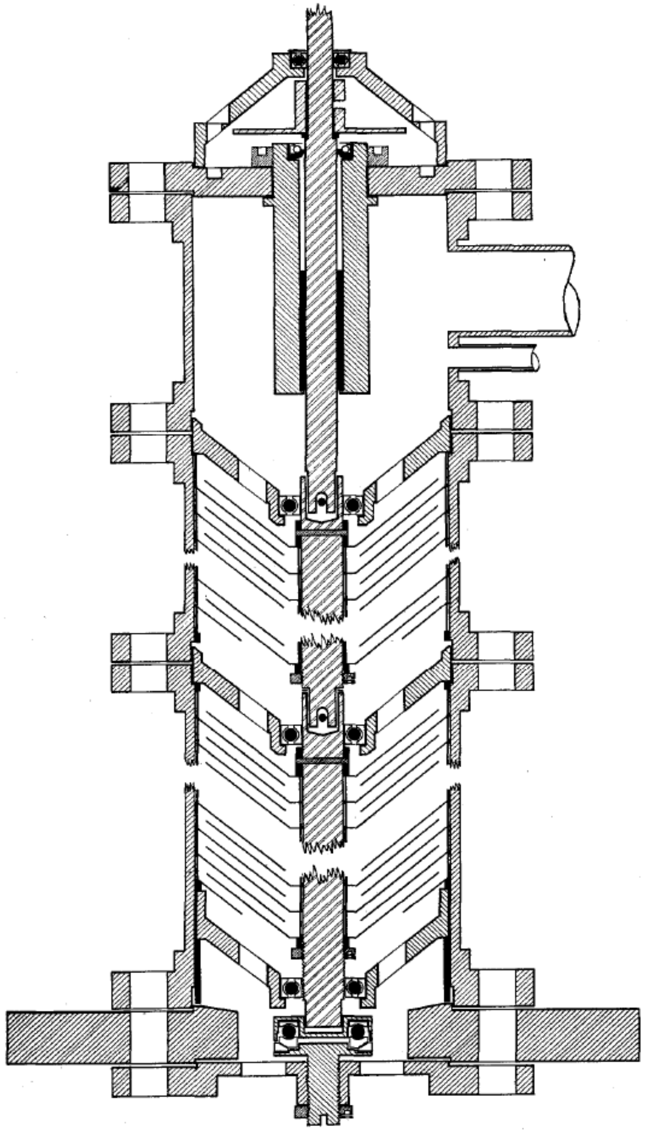
Isotopes for Science, Medicine and Industry

Distillation method

boiling points

$H_2{}^{16}O$ 100.0 °C

$D_2{}^{16}O$ 101.4 °C



G.N. Lewis and R.E. Cornell, JACS 1933;55:2179.

J.R. Huffman and H.C. Urey, Indus & Eng Chem 1937;29:483.

It's time to raise a glass (of heavy water) to a longer life

Elixir of life: 'Heavy' water could increase your lifespan by 10 years, say scientists

Dr de Grey, a 'bio-gerontologist' who leads the Methuselah Foundation, a charity which aims for 'the defeat of age-related disease and the indefinite extension of the healthy human lifespan', said the research was 'extremely promising'.

MailOnline

Update

TRENDS in Biotechnology Vol.25 No.9

Research Focus

Heavy isotopes to avert ageing

Vadim V. Demidov

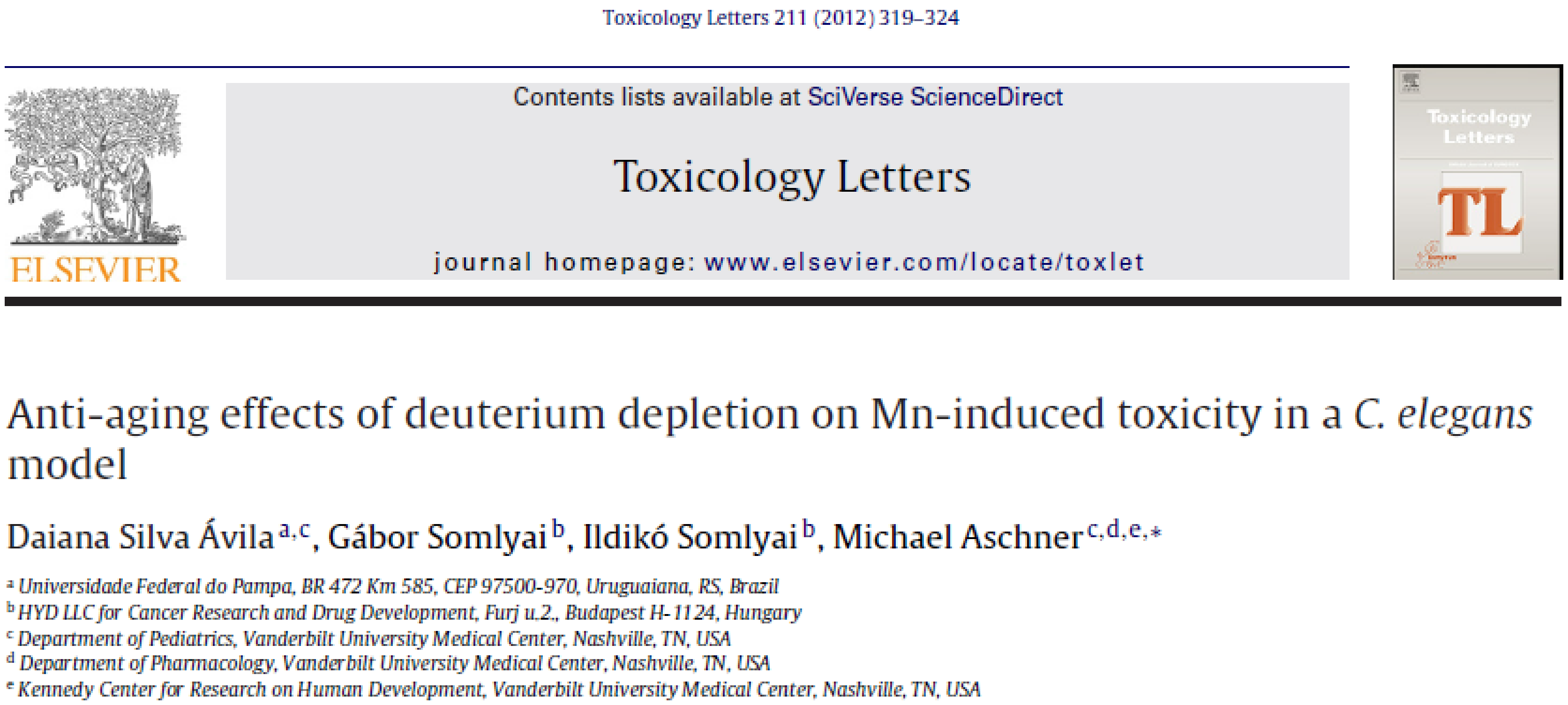
Center for Advanced Biotechnology, Boston University, 36 Cummington S

'Heavy water' could help us live longer

Deuterium depleted water

QLARIVIA®
DEUTERIUM DEPLETED WATER

BIOHACKER SUMMIT
OFFICIAL LONGEVITY WATER SPONSOR



Toxicology Letters 211 (2012) 319–324

Contents lists available at SciVerse ScienceDirect

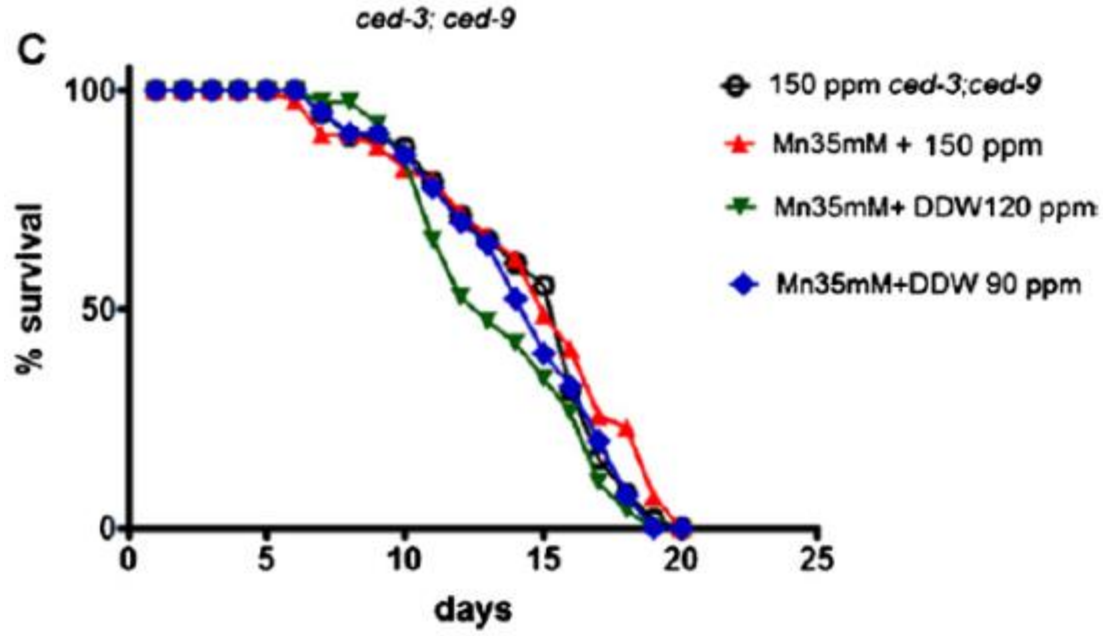
Toxicology Letters

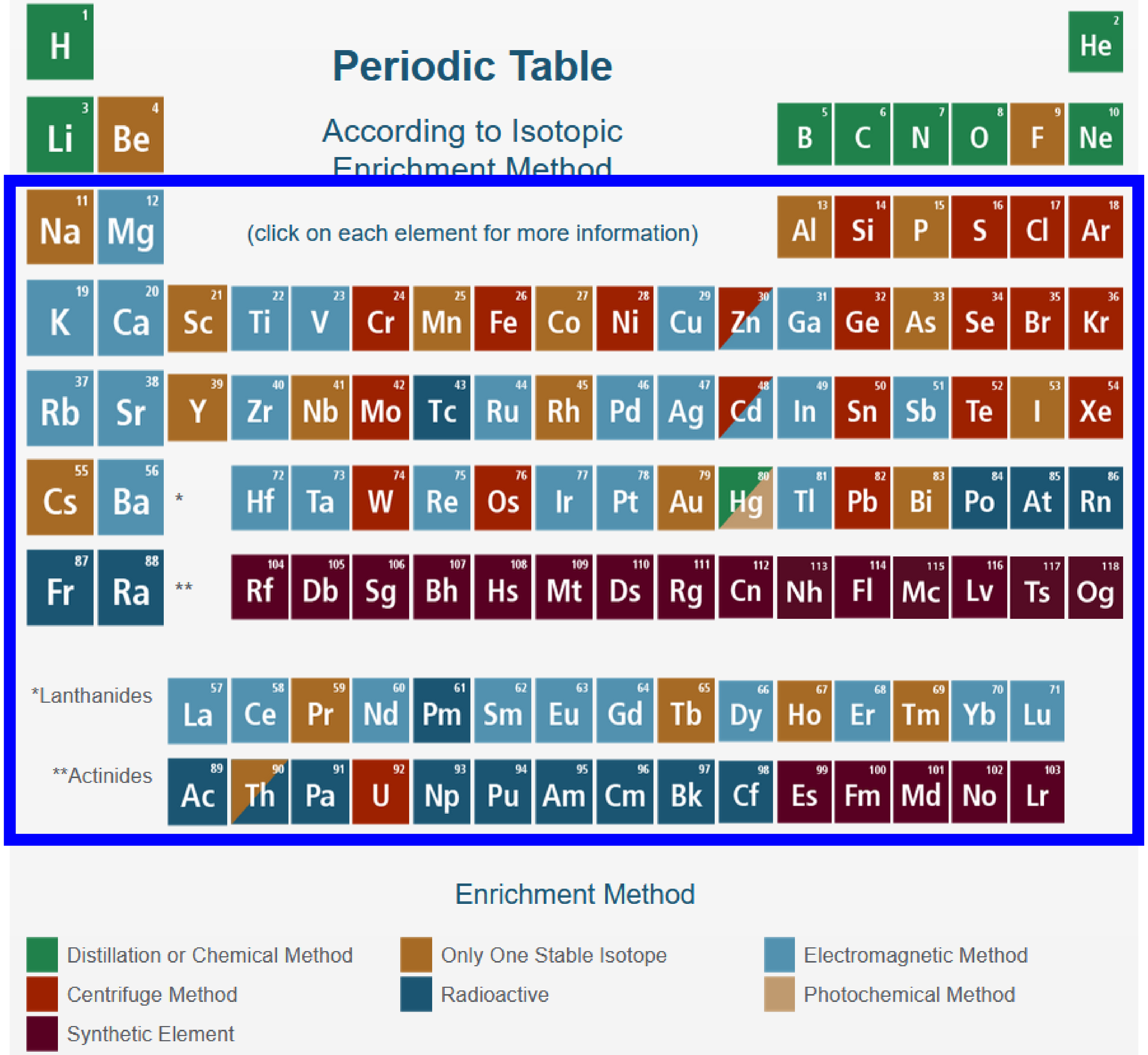
journal homepage: www.elsevier.com/locate/toxlet

Anti-aging effects of deuterium depletion on Mn-induced toxicity in a *C. elegans* model

Daiana Silva Ávila[a,c], Gábor Somlyai[b], Ildikó Somlyai[b], Michael Aschner[c,d,e,*]

[a] Universidade Federal do Pampa, BR 472 Km 585, CEP 97500-970, Uruguaiana, RS, Brazil
[b] HYD LLC for Cancer Research and Drug Development, Furj u.2., Budapest H-1124, Hungary
[c] Department of Pediatrics, Vanderbilt University Medical Center, Nashville, TN, USA
[d] Department of Pharmacology, Vanderbilt University Medical Center, Nashville, TN, USA
[e] Kennedy Center for Research on Human Development, Vanderbilt University Medical Center, Nashville, TN, USA





Periodic Table

According to Isotopic Enrichment Method

(click on each element for more information)

1 H																	2 He
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
55 Cs	56 Ba	*	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
87 Fr	88 Ra	**	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Cn	113 Nh	114 Fl	115 Mc	116 Lv	117 Ts	118 Og
*Lanthanides		57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu	
**Actinides		89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr	

Enrichment Method

🟩 Distillation or Chemical Method
🟫 Only One Stable Isotope
🟦 Electromagnetic Method
🟥 Centrifuge Method
■ Radioactive
▨ Photochemical Method
■ Synthetic Element

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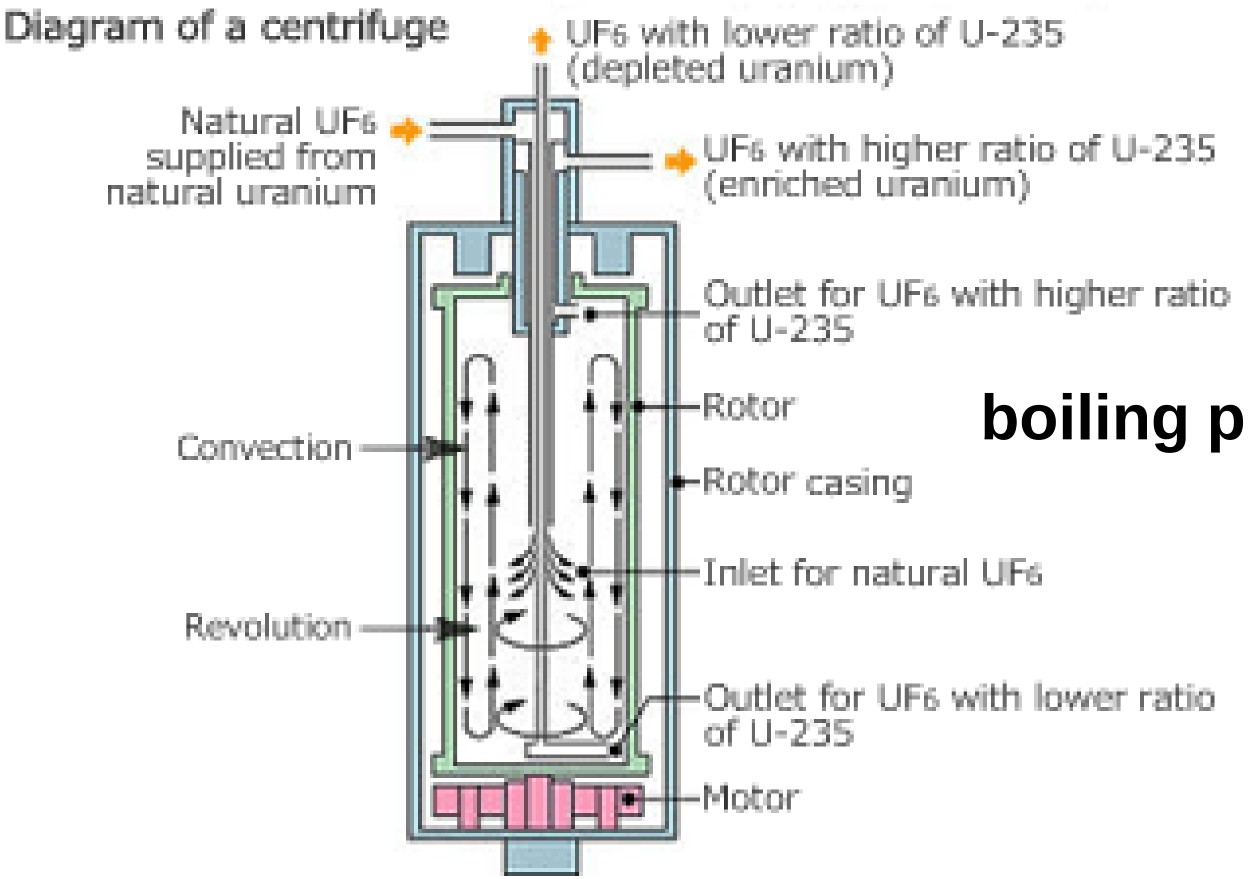
Isotope Separation by centrifugation



boiling point: UF_6 56 □C

ideal elementary separation factor (negligible back-pressure): $\boldsymbol{\alpha} = \mathbf{exp[(M_2 - M_1)}\ \Omega^2 r^2 / 2RT)$

for $^{235,238}UF_6$ as function of peripheral speed $v = \Omega r$ ($T = 310$ K):

v (m/s)	400	500	600	700
α	1.098	1.15	1.23	1.33

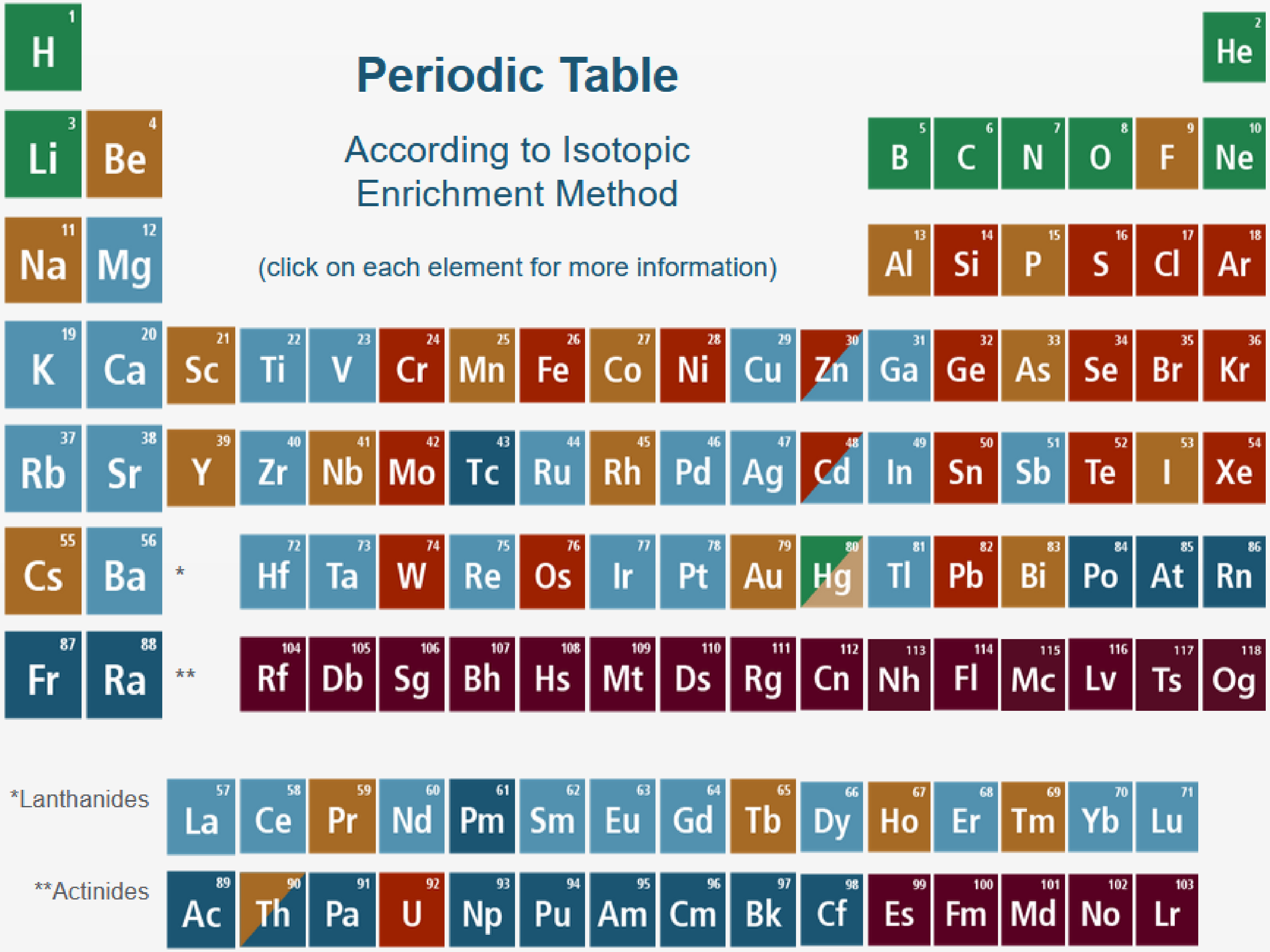
v is limited by the material strength of the wall !

Centrifuge facilities

Periodic Table

According to Isotopic Enrichment Method

(click on each element for more information)



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11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
55 Cs	56 Ba	*	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
87 Fr	88 Ra	**	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Cn	113 Nh	114 Fl	115 Mc	116 Lv	117 Ts	118 Og
*Lanthanides		57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu	
**Actinides		89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr	

Enrichment Method

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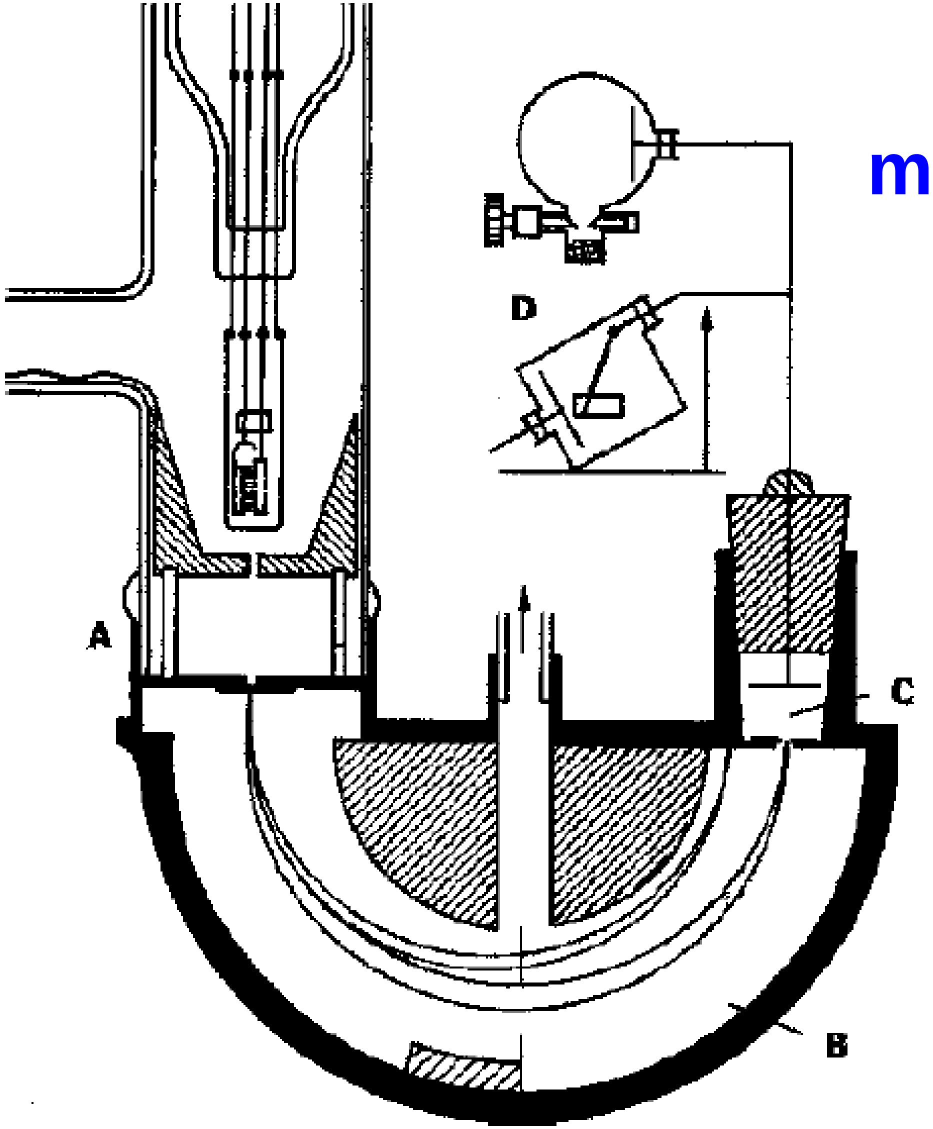
ISOFLEX

Isotopes for Science, Medicine and Industry

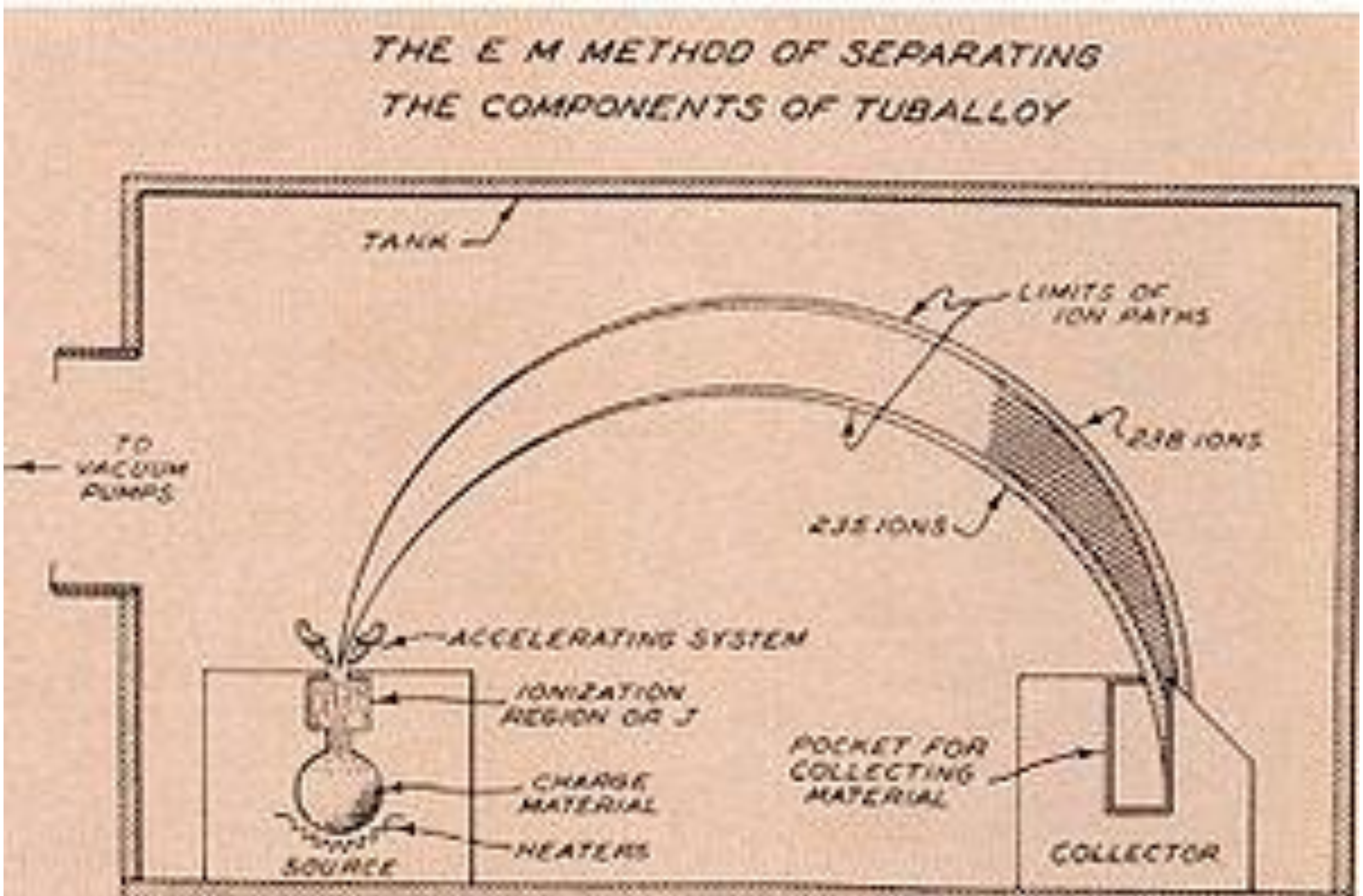
1918: Dempster 180 degree spectrometer



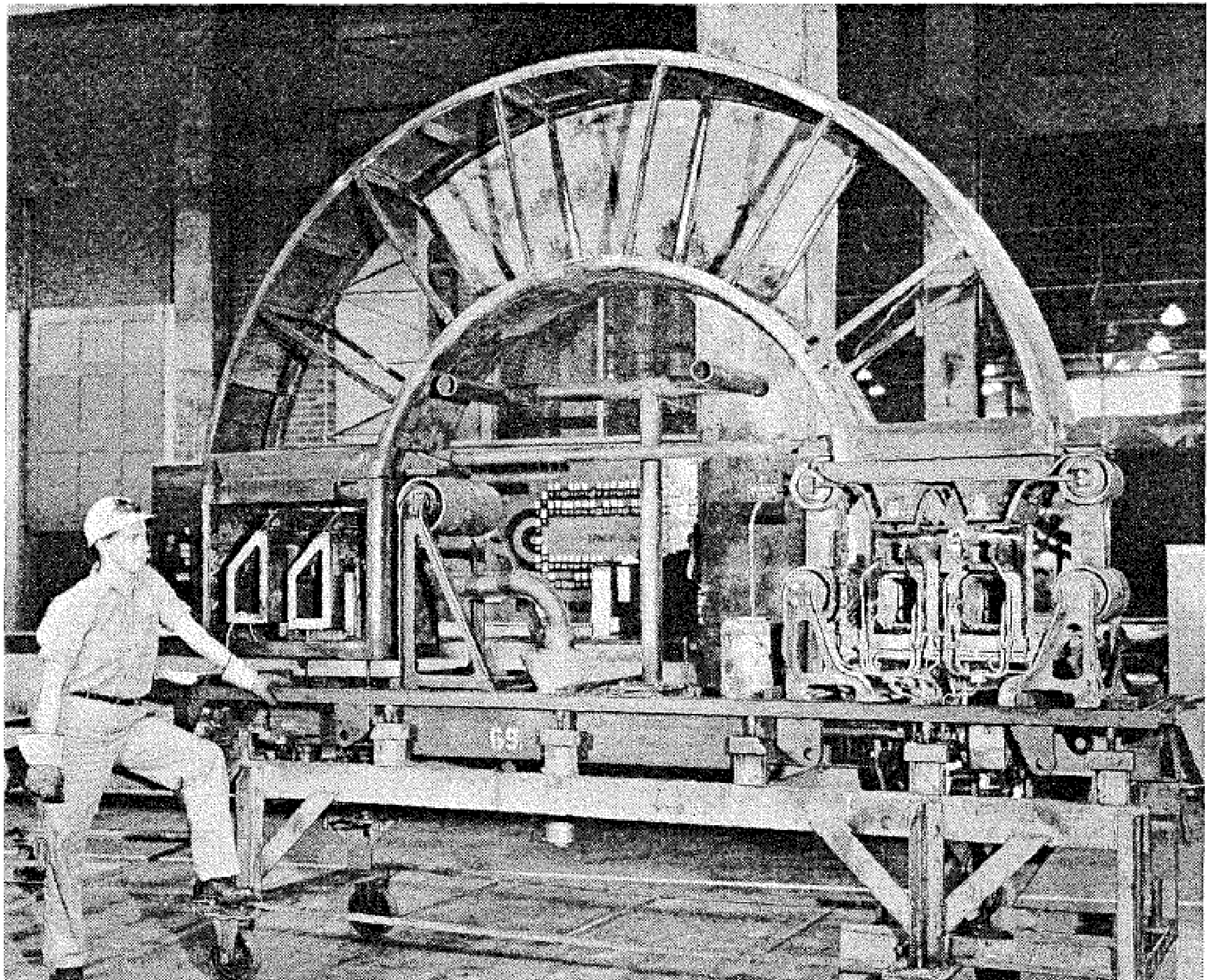
$m/q = (B^2 r^2) / 2 U$

1920: discovery of isotopes in Li, Mg, K, Ca, Zn

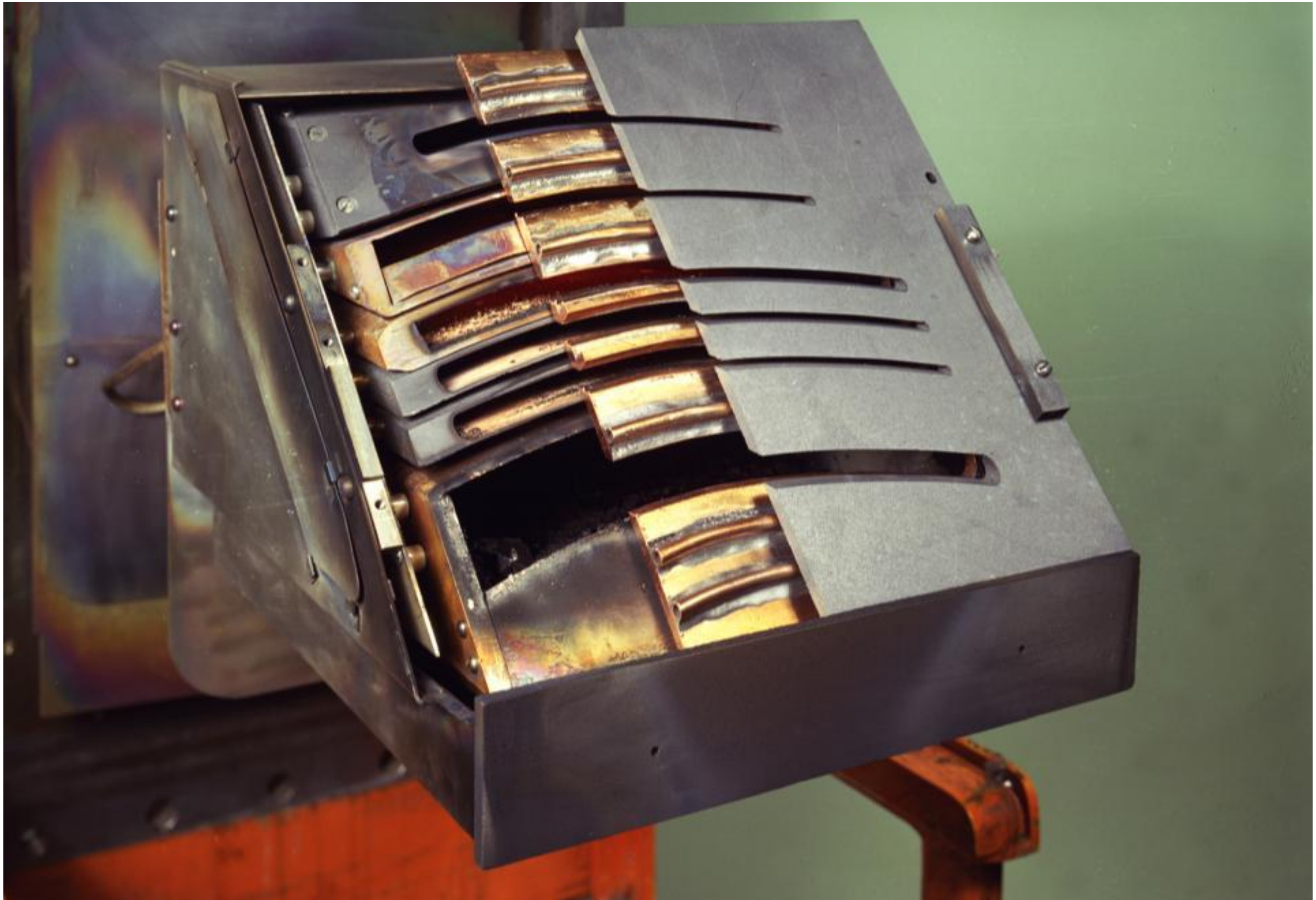
Calutron 1942: electromagnetic isotope separation



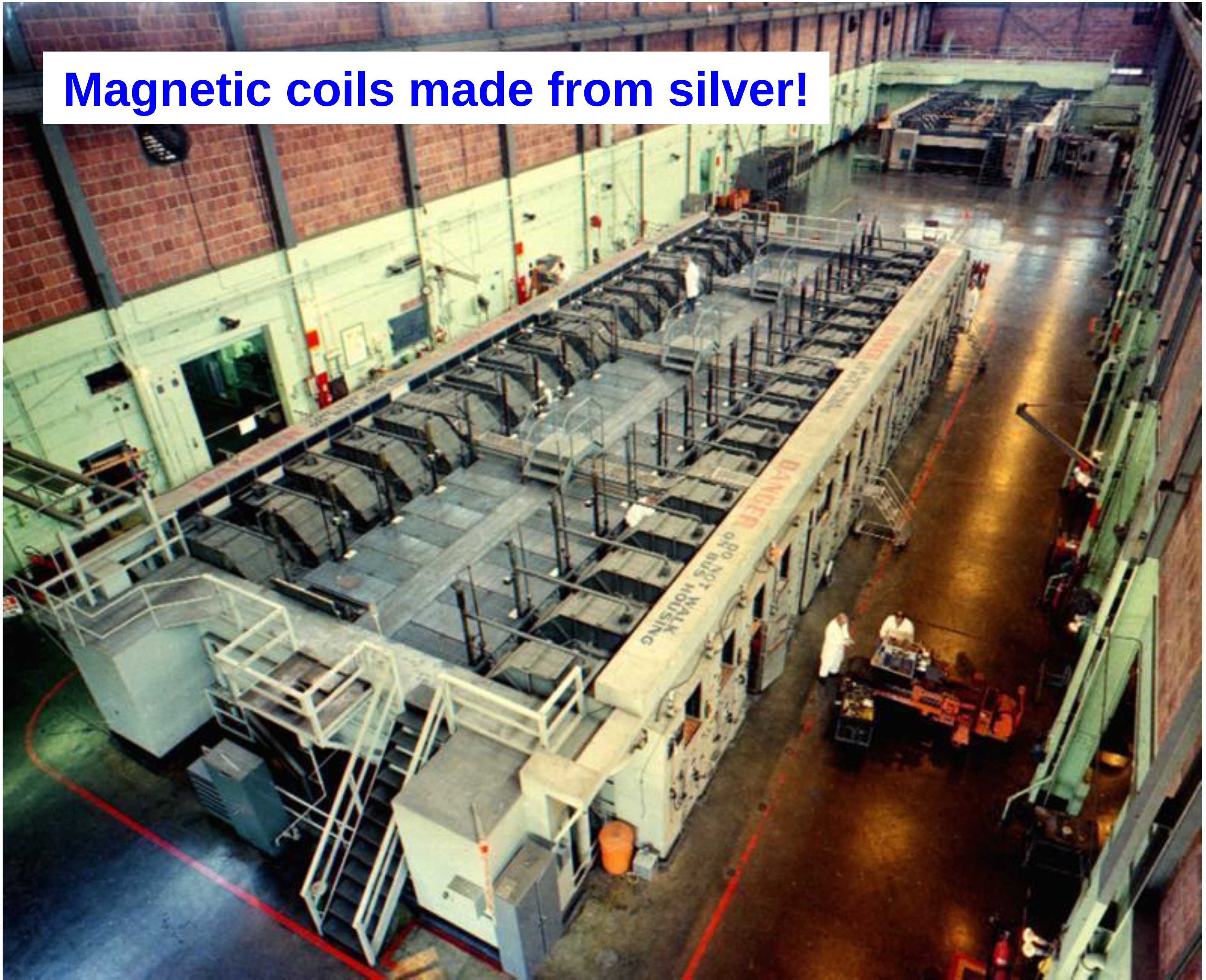
Calutron tanks

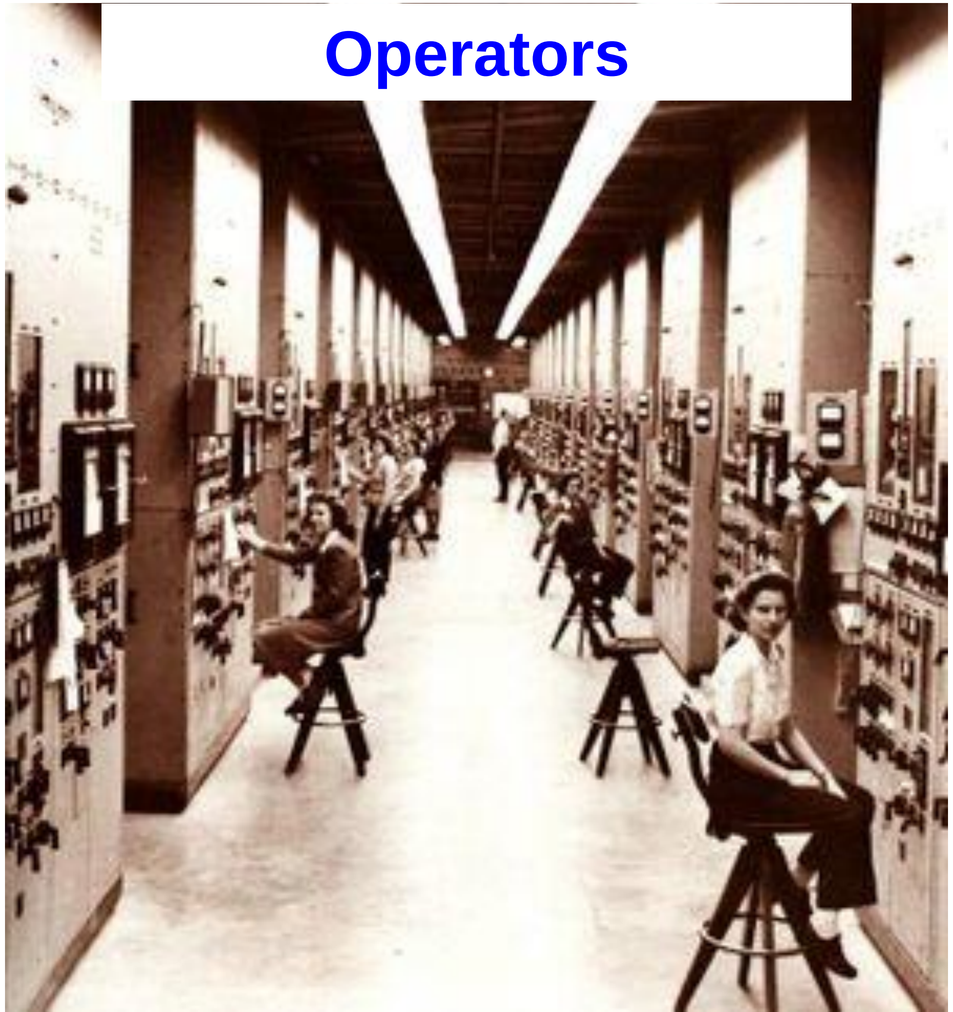


Collector plates of a Calutron



1945: large scale electromagnetic isotope separation

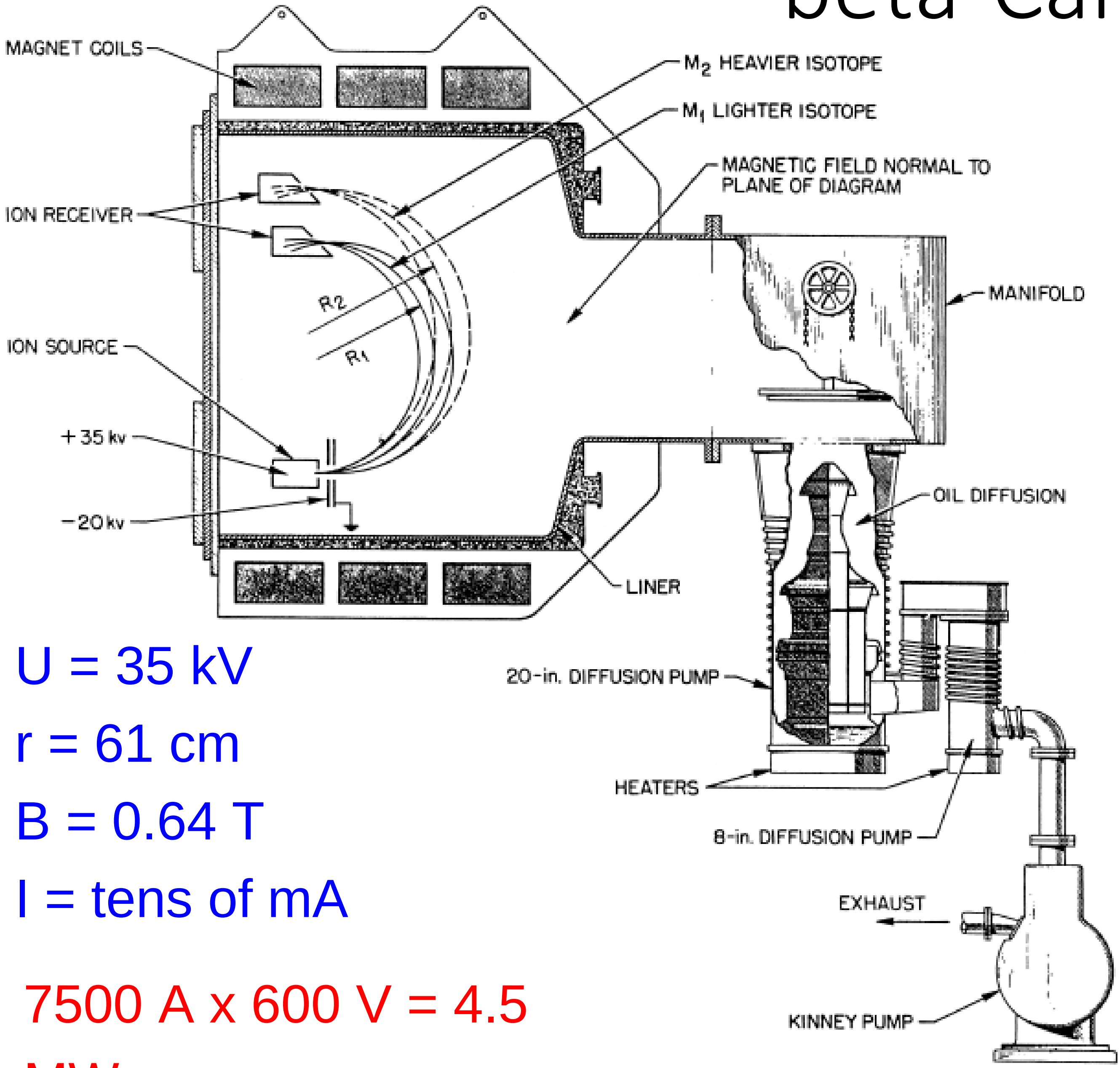

Magnetic coils made from silver!


Operators

>1945: dismantling of most calutrons

13300 t of silver loan returned (present value 12 G$)

beta-Calutron



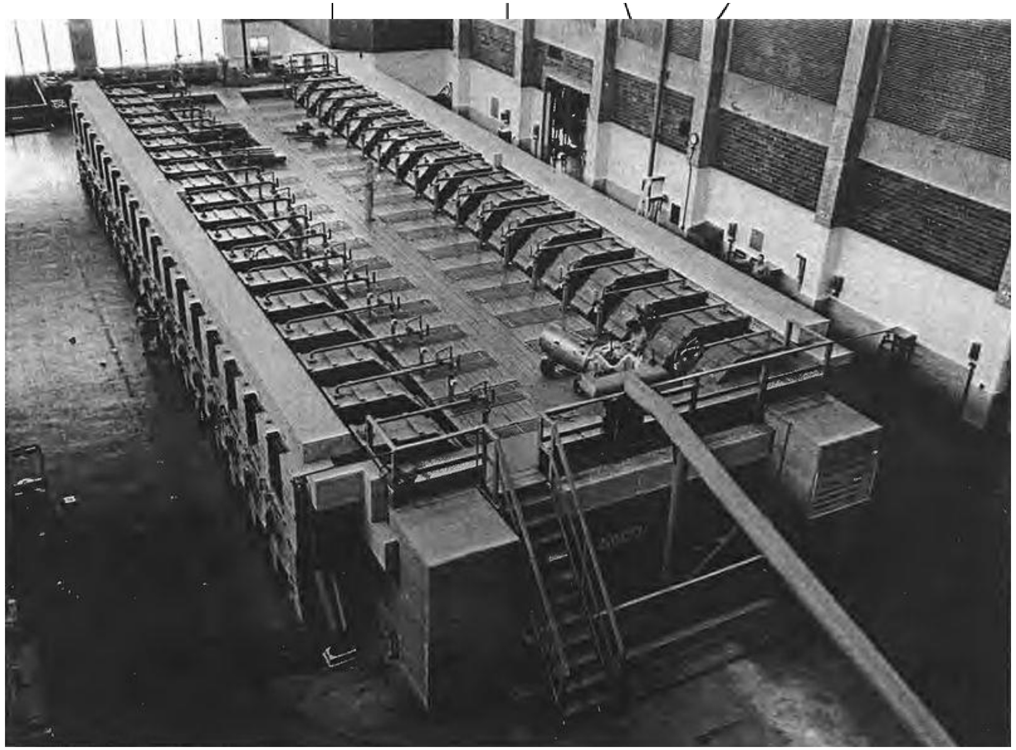
Daily production per tank:

160 mg ^{176}Yb

200 mg ^{160}Gd

110 mg ^{155}Gd

4 mg ^{152}Gd



$U = 35$ kV

$r = 61$ cm

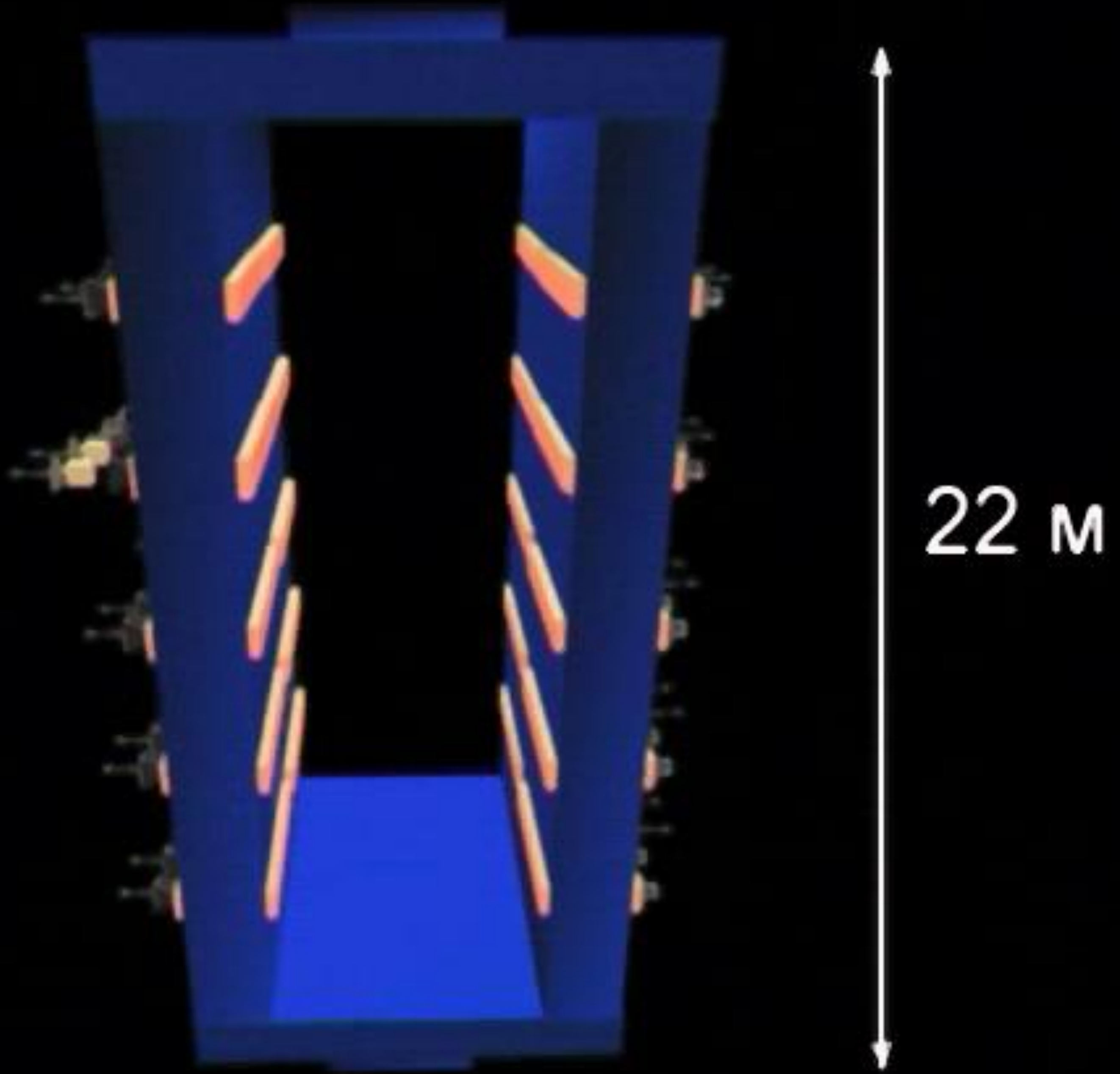
$B = 0.64$ T

I = tens of mA

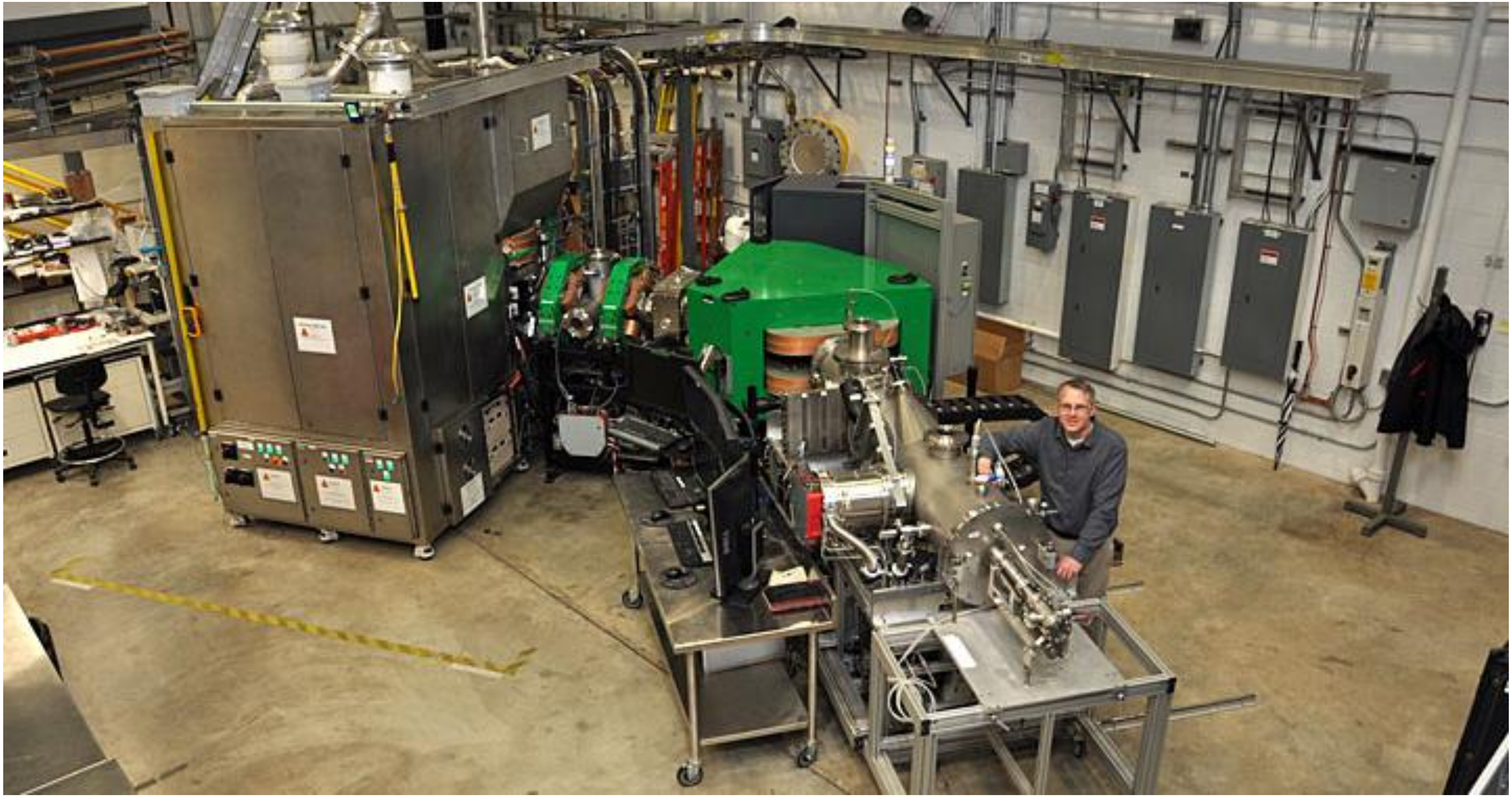
7500 A x 600 V = 4.5 MW

AL&AK Yergey, J Am Soc Mass Spectrometry

Russian EMIS

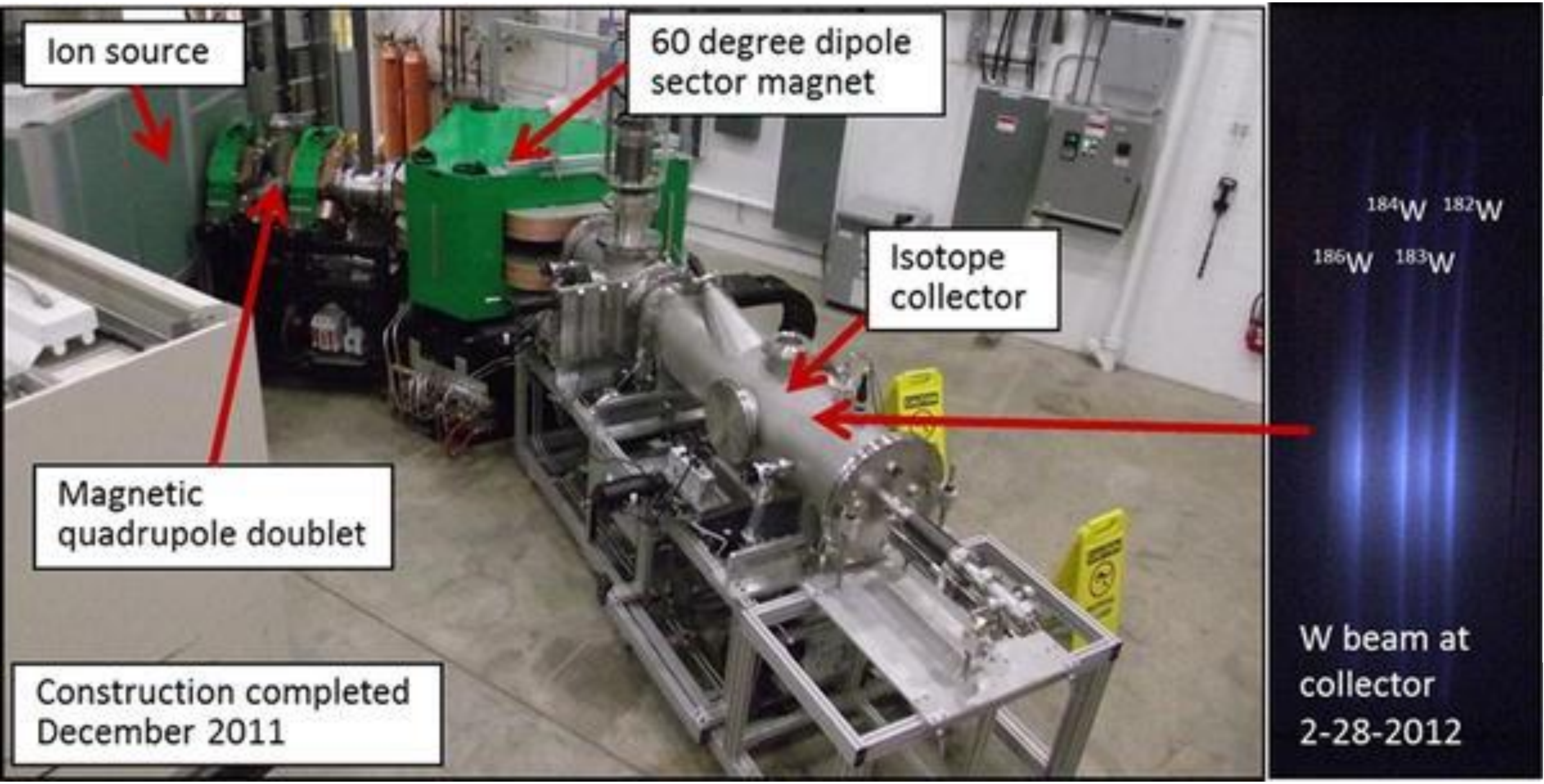

22 м

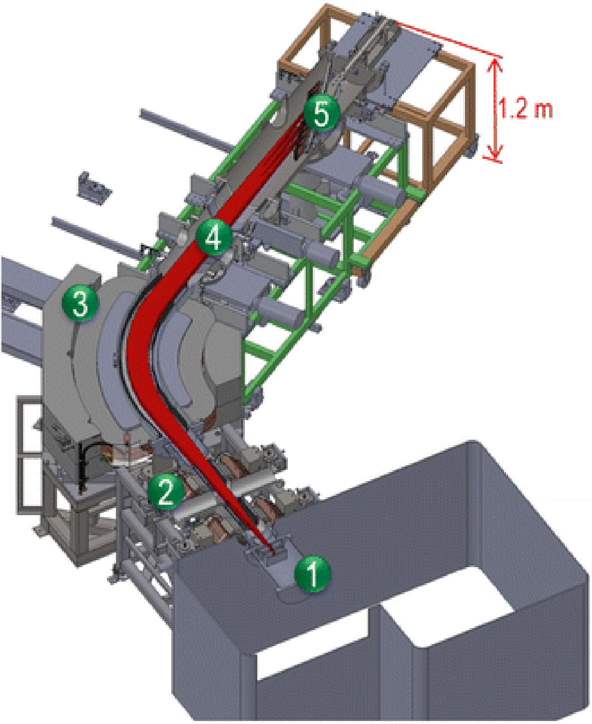
New DOE EMIS



B Egle, J Radioanal Nucl Chem 2014;299:995.

New DOE EMIS





Poster by Stuart Warren et al.

B Egle, J Radioanal Nucl Chem 2014;299:995.

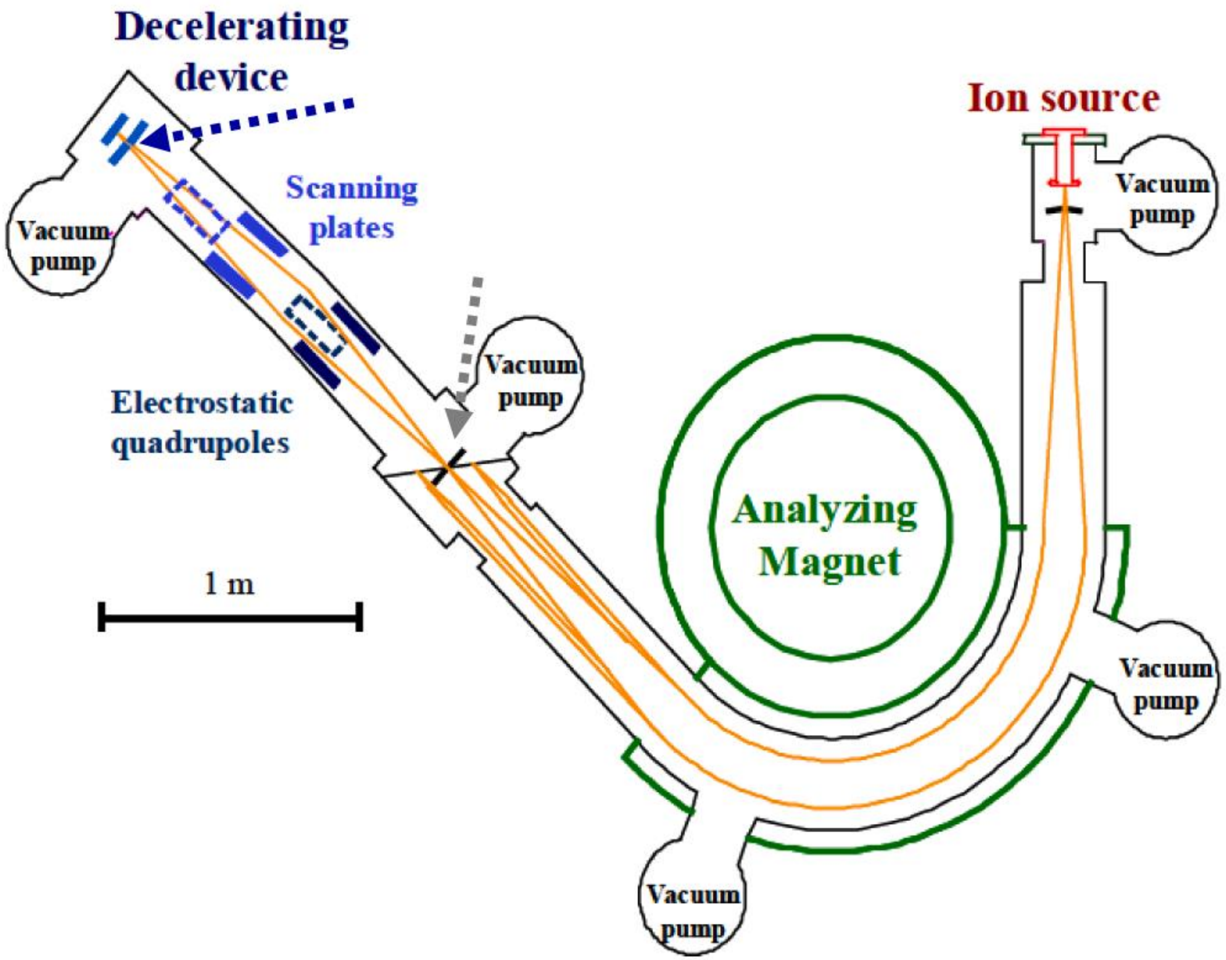
SIDONIE mass separator

IJCLab
Irène Joliot-Curie
Laboratoire de Physique des 2 Infinis

$^{157}Gd/^{158}Gd = 0.047(5)\%$

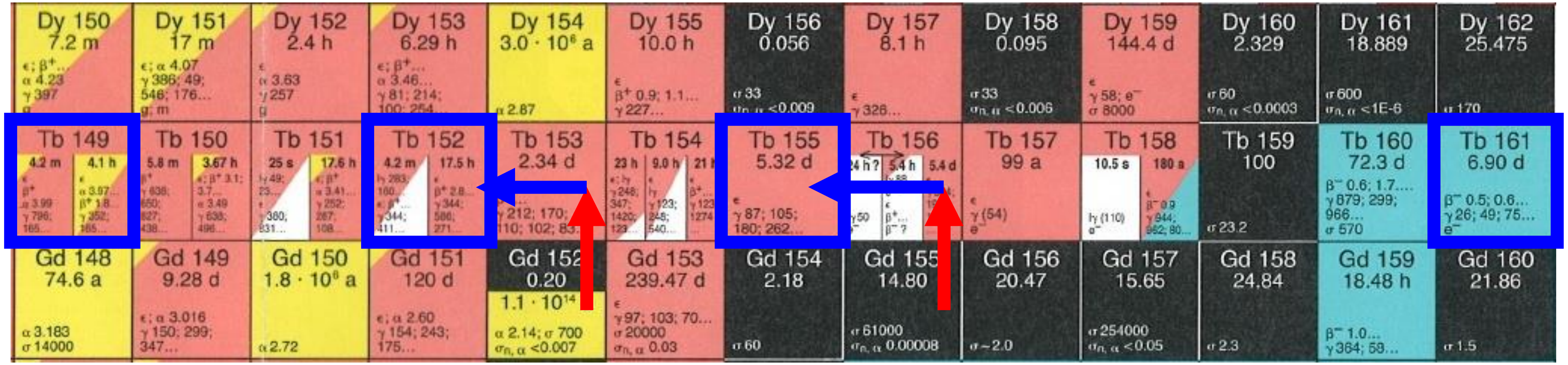
U.K. et al., Nucl Instr Meth B 2020,463:111.

Poster by Morgane Bouteculet et al.



J. Camplan et al., Nucl. Instr. Meth. 84 (1970) 37.
K. Alexandre et al., Nucl. Instr. Meth. 84 (1970) 45.
C.O. Bacri et al., Nucl. Instr. Meth. B 406 (2017) 48.

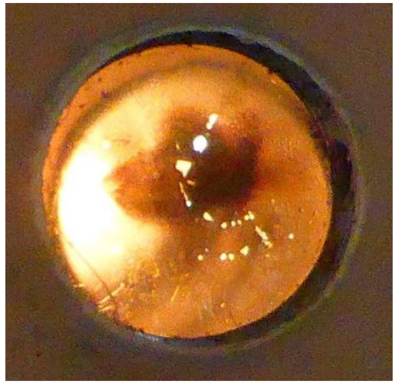
^{152,155}Gd(p,n)^{152,155}Tb production



^{152}Gd(p,n)^{152}Tb □12 MeV p

^{155}Gd(p,n)^{155}Tb □12 MeV p

>99.7% pure ^{152}Tb produced

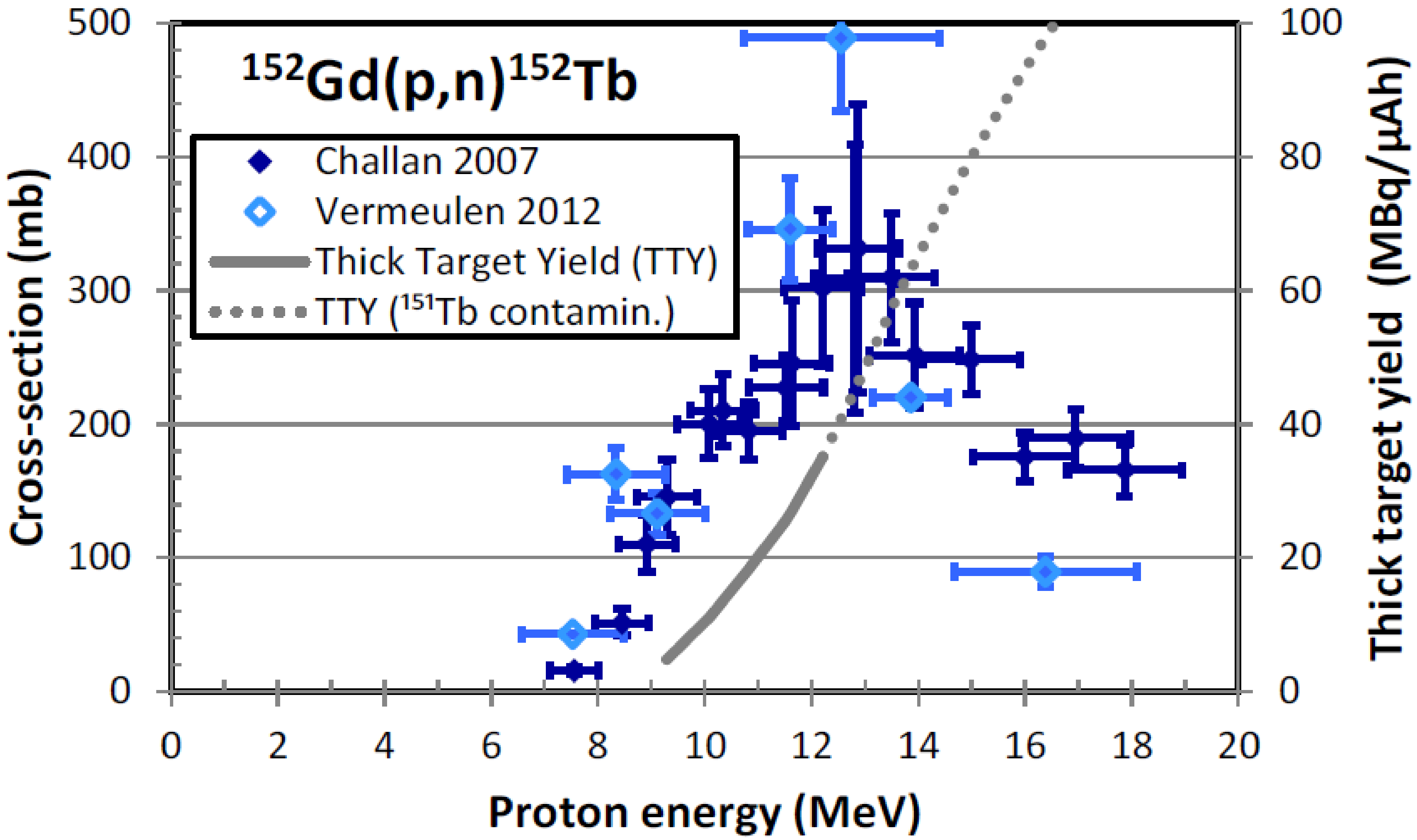


MLL
Maier-Leibnitz-Laboratorium
für Kern-, Teilchen- & Beschleunigerphysik

Poster by Morgane Bouteculet et al.

U.K. et al., Nucl Instr Meth B 2020,463:111.

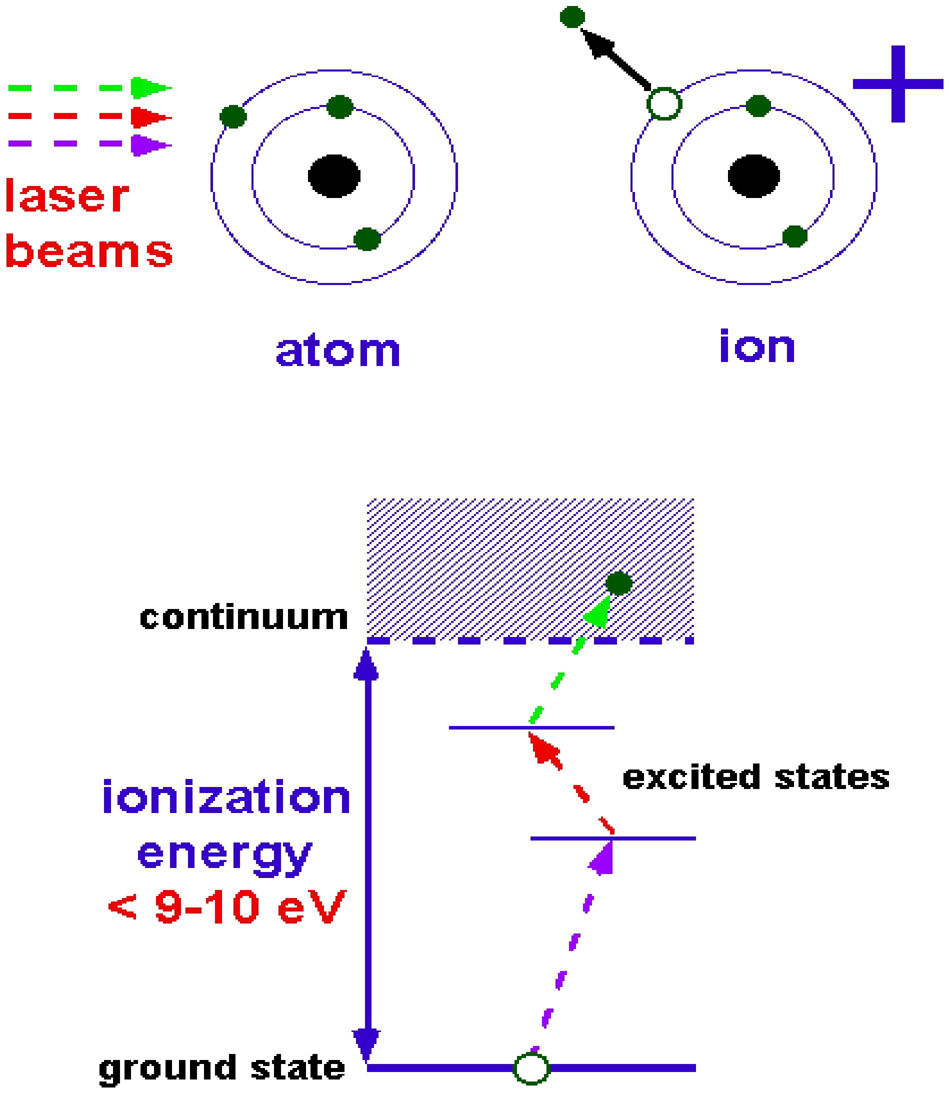
^{152}Tb production from ^{152}Gd targets



12 MeV protons on 150 mg/cm^2 ^{152}Gd target □30 MBq/µAh

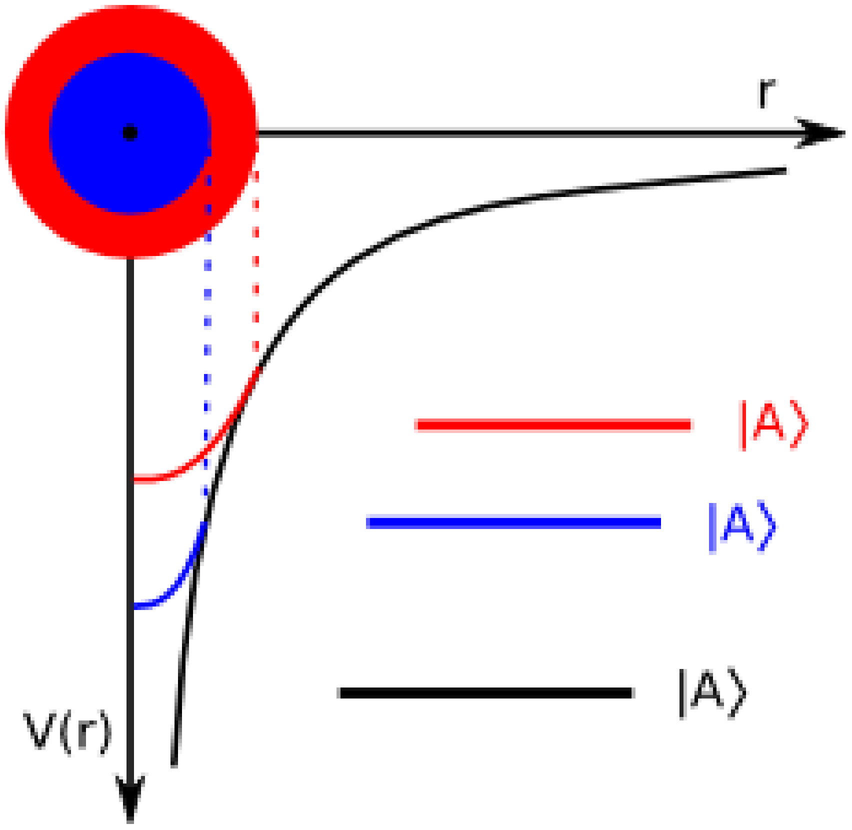
100 µA for 12 h: **20-30 GBq at EOI** □**100 patients imaged**

Laser Ionization

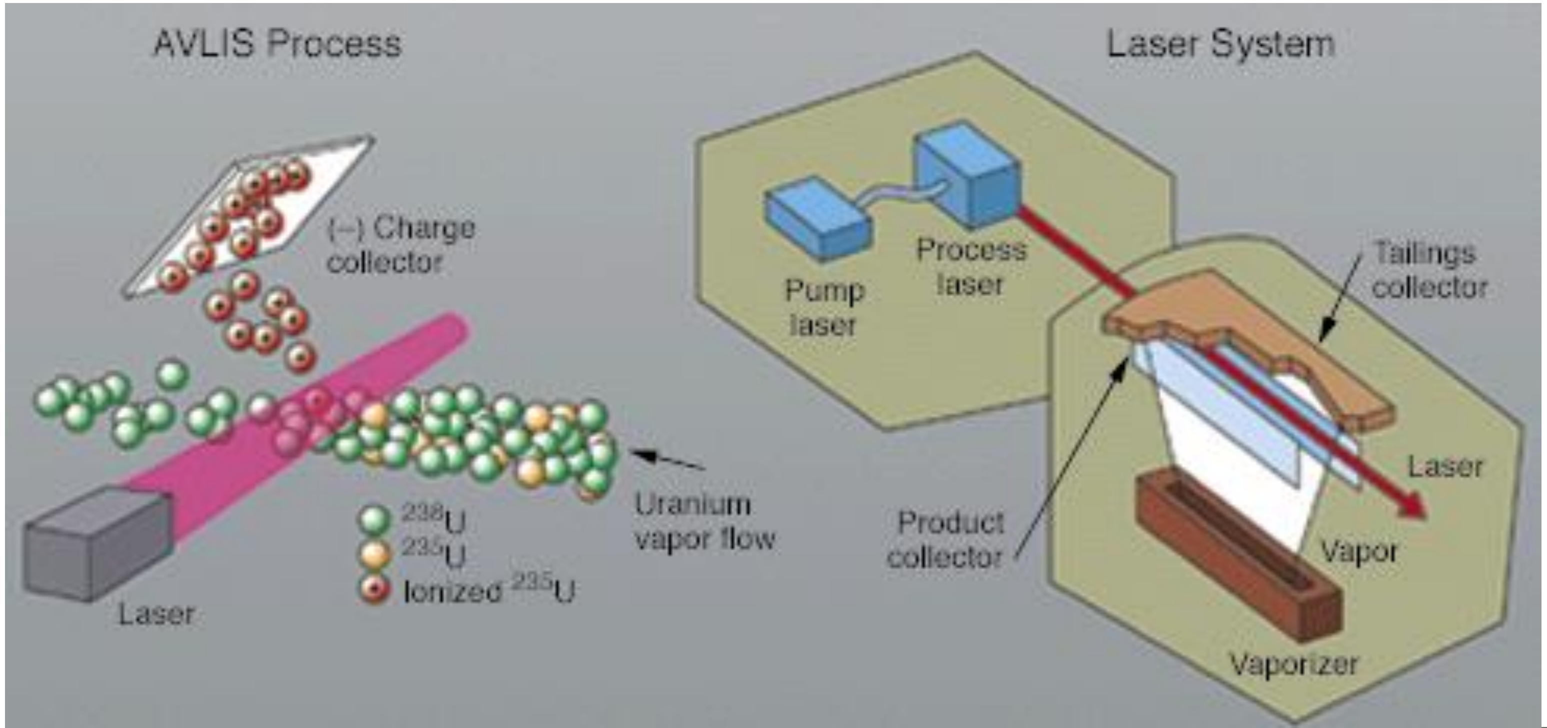


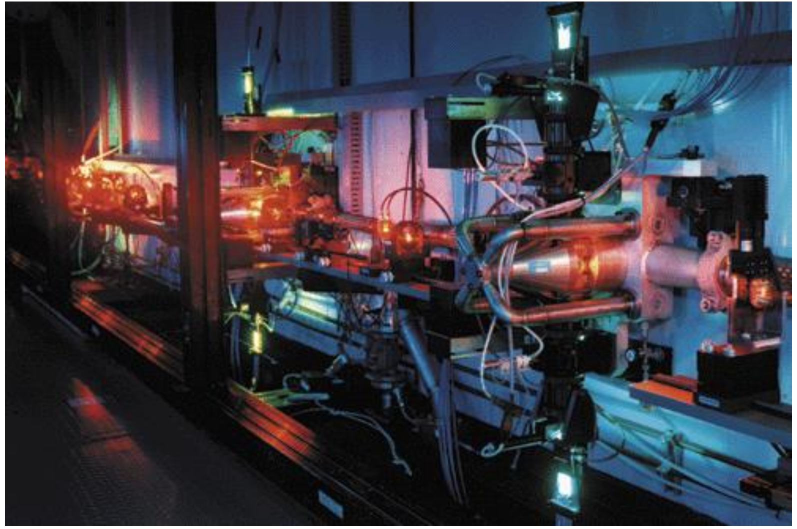
FIELD SHIFT

due to different charge radii



AVLIS (Atomic Vapor Laser Isotope Separation)





3-step resonant laser ionization of ytterbium

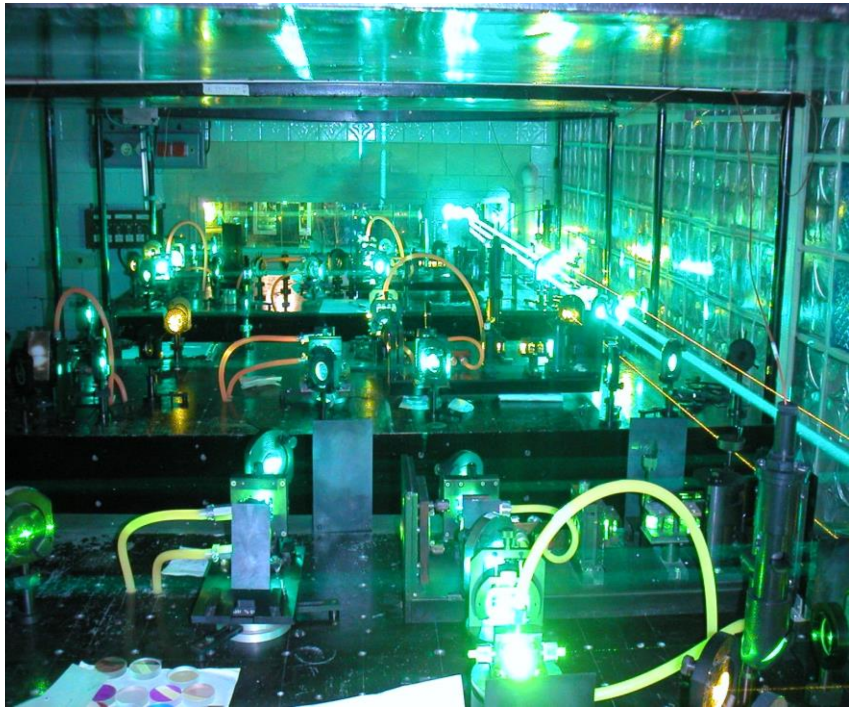
Channel	Wavelength, nm	Dye	Power, W	Spectr.band, MHz	Pulse width, ns
1	555	R110	5	500	15
2	581	R6G	5	500	15
3	582	R6G	20	$3 \cdot 10^4$	20

Pumping: power 120 – 130W, λ = 510nm, f = 10kHz, τ = 20ns.

Output of the system: 3 g/ year

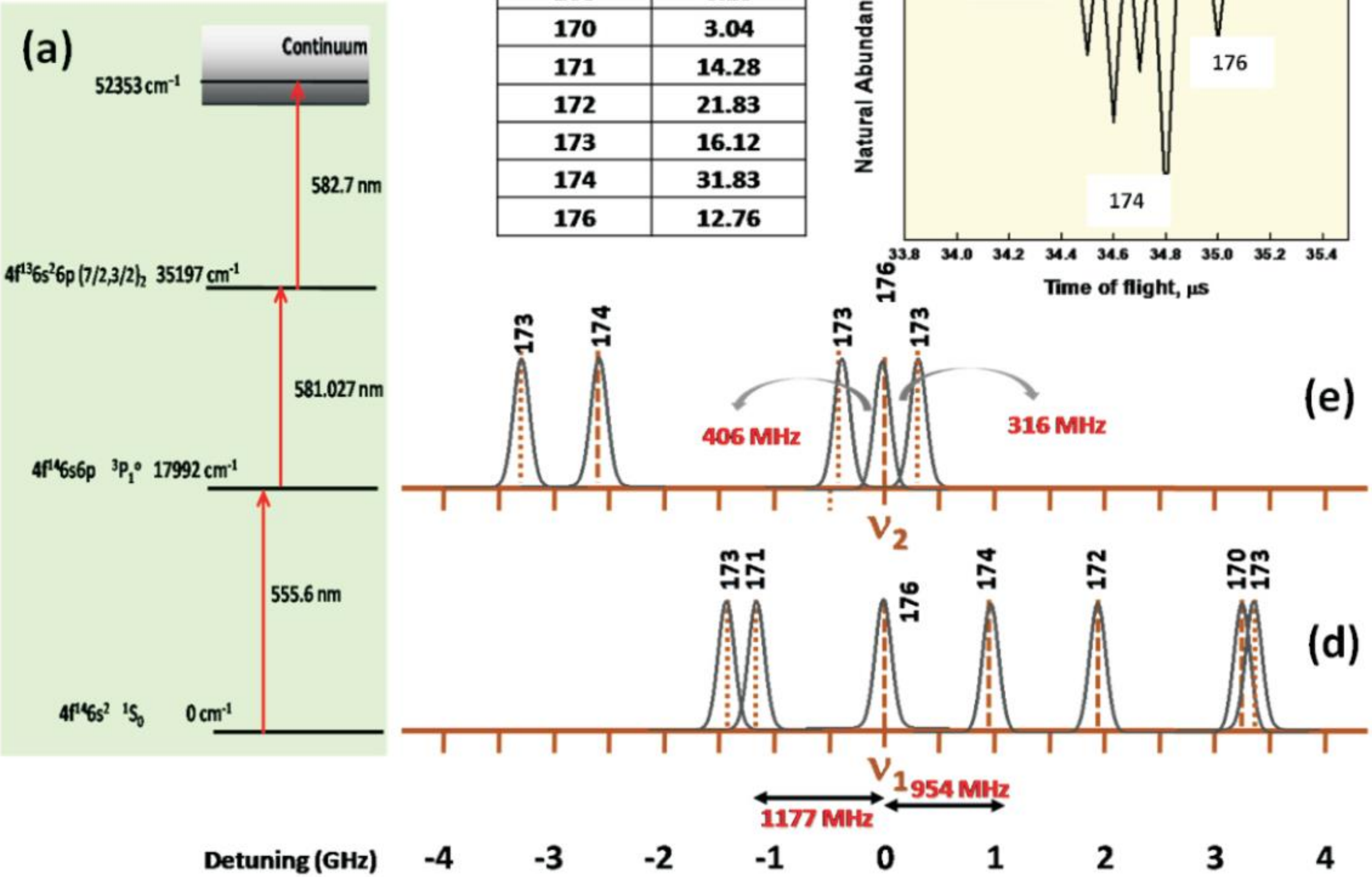
Final isotope content:

- Yb – 168 – 20.21% (0.14 % natural Yb)
- Yb – 170 – 2.36%
- Yb – 171 – 18.38%
- Yb – 172 – 15.45%
- Yb – 173 – 12.1%
- Yb – 175 – 22.38%
- Yb – 176 – 9.12%



S. Akulinichev et al., INR Troitsk, ICTR-PHE 2012.

^{176}Yb Selective Photoionization Ladder



Yb Isotopes amu	Abundance %
168	0.13
170	3.04
171	14.28
172	21.83
173	16.12
174	31.83
176	12.76

Enrichment of
5-7 mg/h ^{176}Yb

∼50 g/year

A.D. Rath et al., BARC Newsletter, March-April 2022.

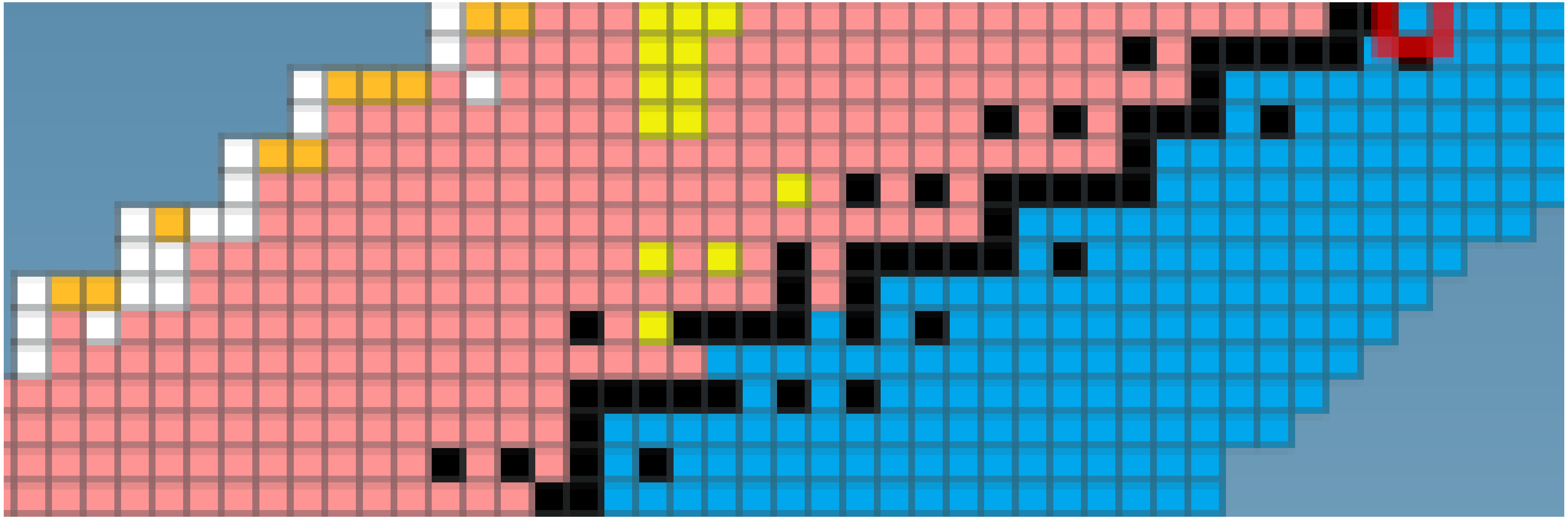
Mass separation of stable lanthanide isotopes

57	58	59	60	61	62	63	64	65	66	67	68	69	70	71
La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu

Electromagnetic isotope separation (EMIS) highest SF!

Atomic vapor laser isotope separation (AVLIS)

Mass separation of radioactive lanthanide isotopes

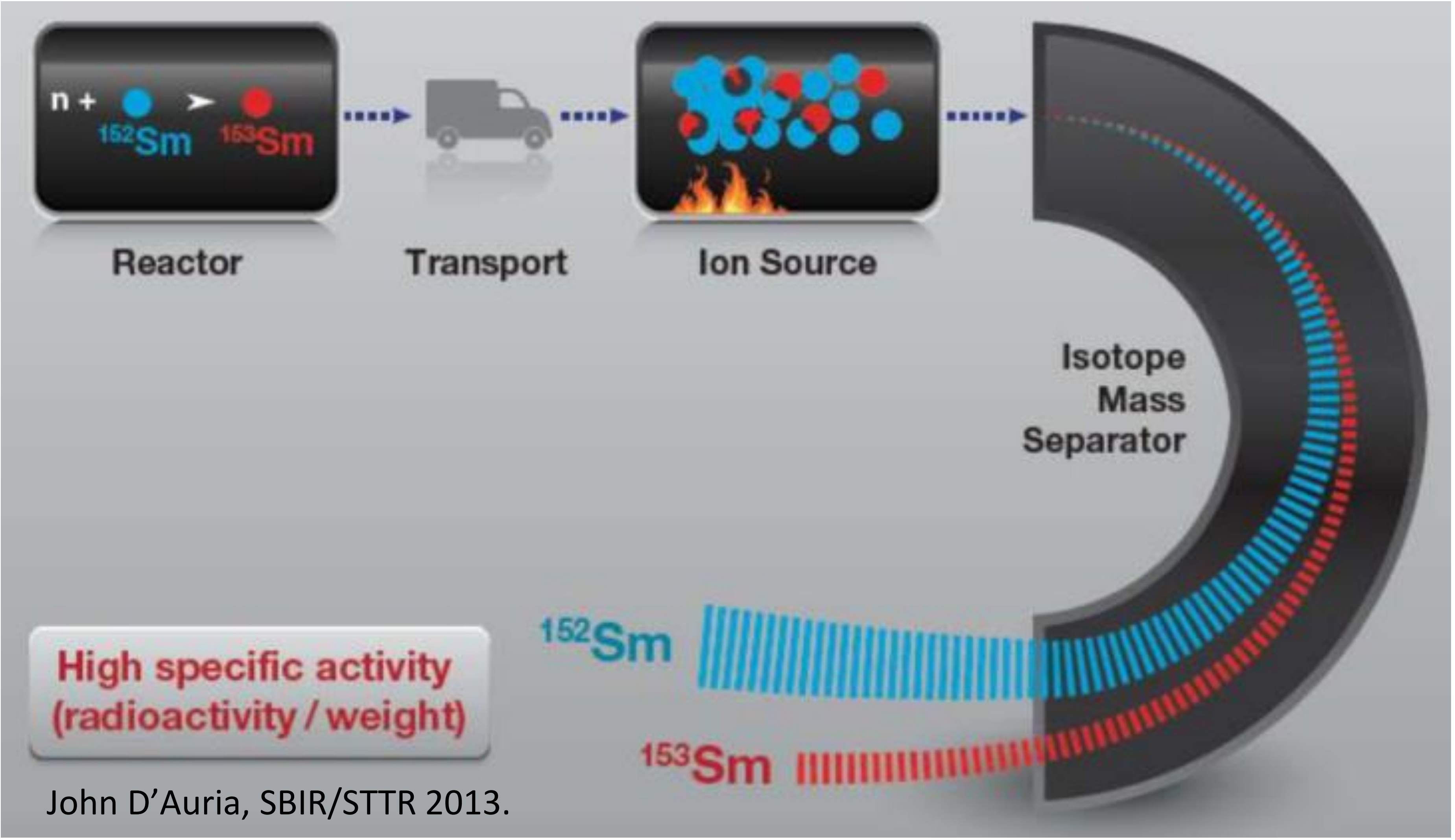


Electromagnetic isotope separation (EMIS) highest SF!

Atomic vapor laser isotope separation (AVLIS)

Resonant laser ionization + EMIS

“Upgrade” of c.a. to n.c.a. by mass separation



John D’Auria, SBIR/STTR 2013.

TRIUMF

Oak Ridge
National Laboratory

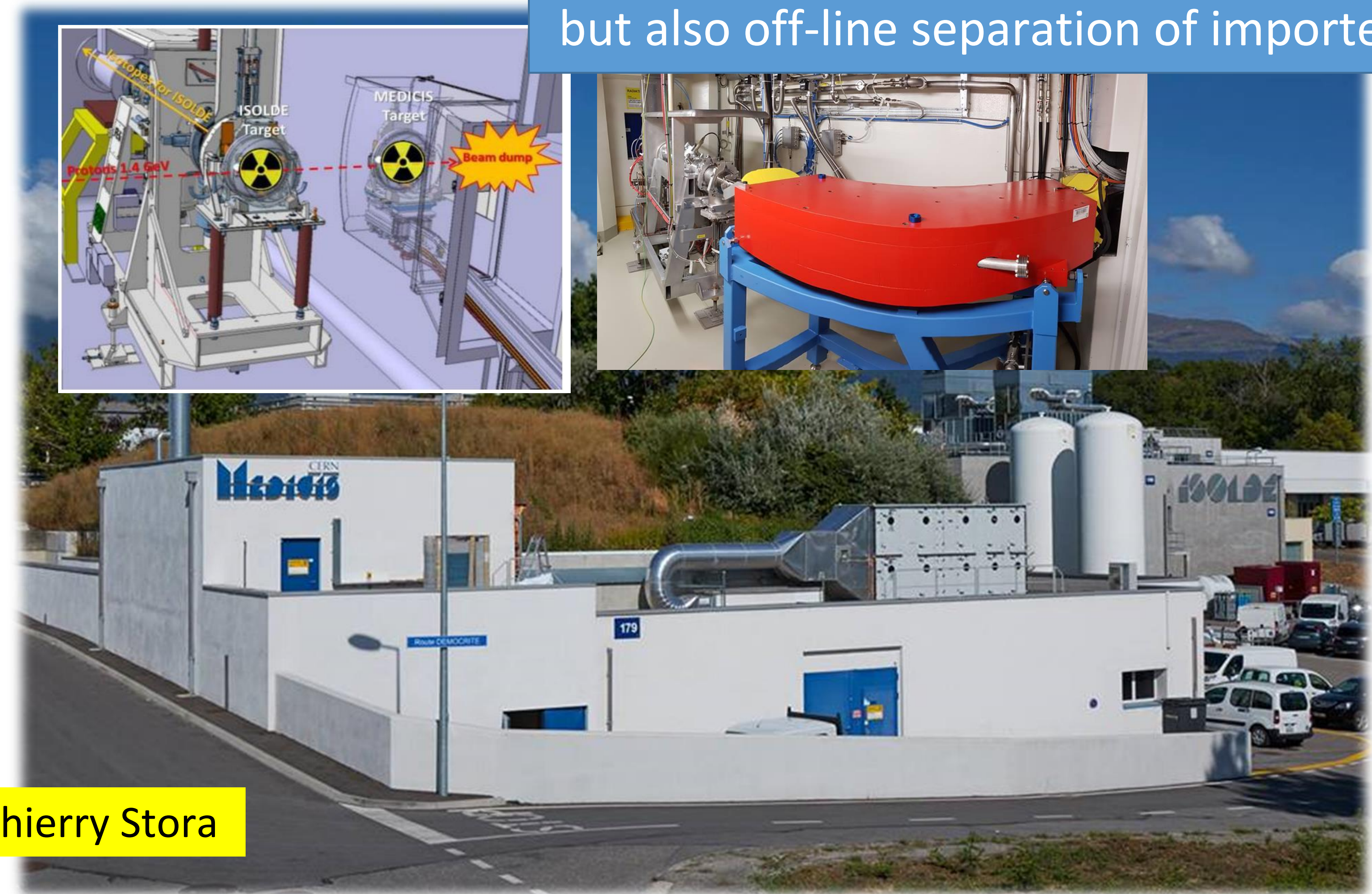
CERN

MEDICIS

J.M. D’Auria et al., Rev Sci Instrum 2013;84:034705.
R. Formento et al., Nucl Instrum Meth B 2020;463:468.
Z. Talip et al., Appl Radiat Isot 2021;176:109823.

a very useful beam dump !

but also off-line separation of imported activity

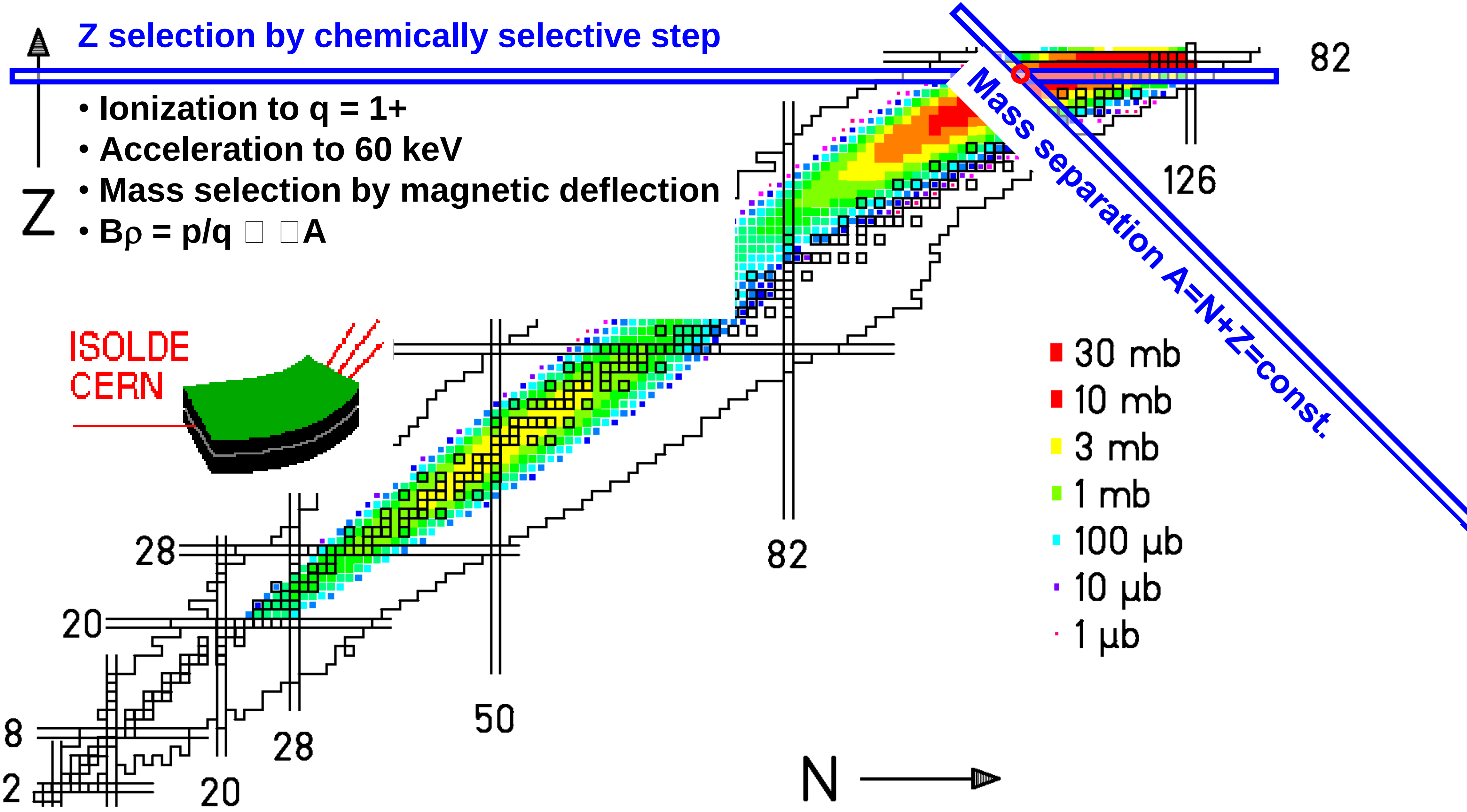


Talk by Thierry Stora

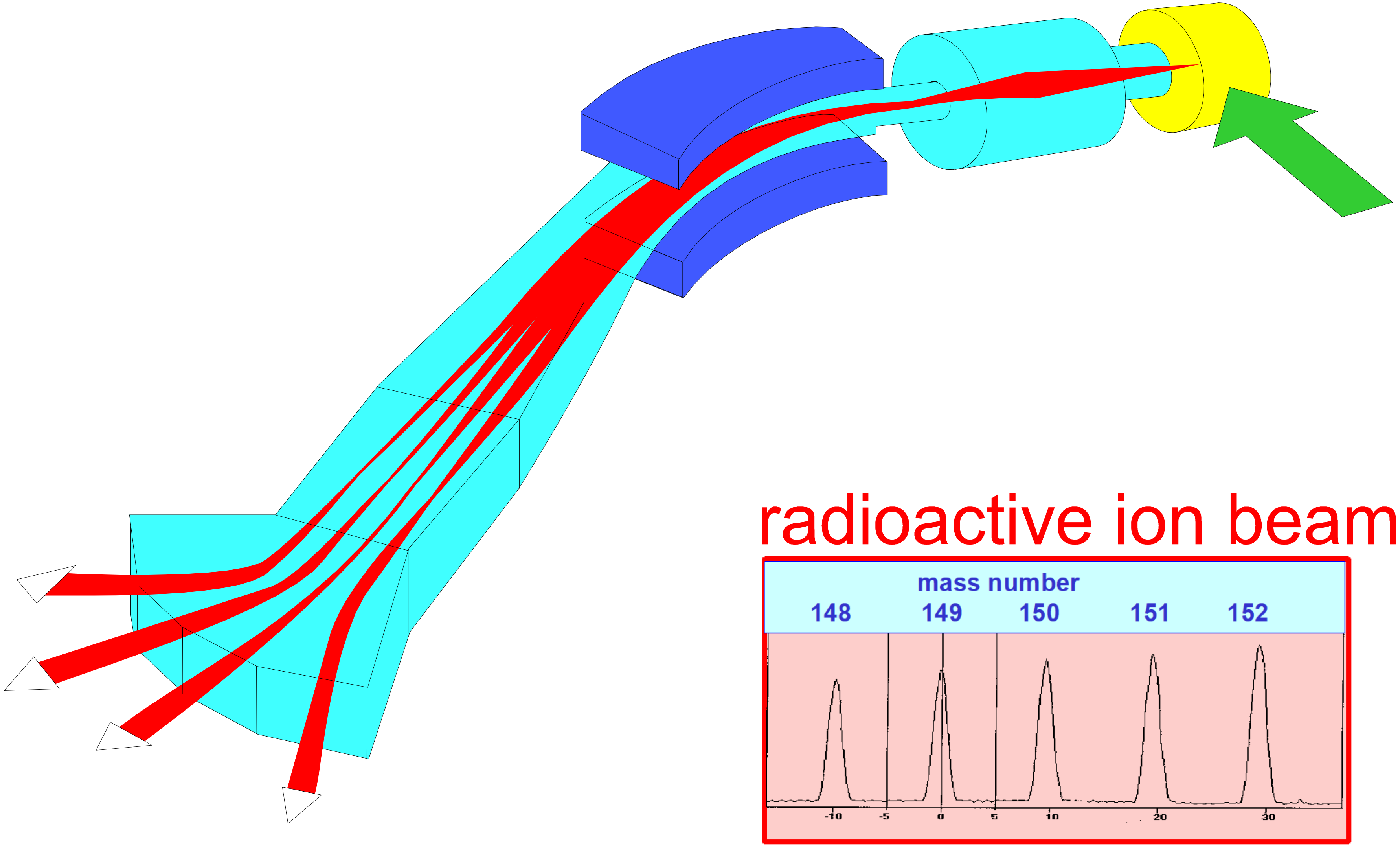
Isotope selection with the ISOL method

Z selection by chemically selective step

- **Ionization to q = 1+**
- **Acceleration to 60 keV**
- **Mass selection by magnetic deflection**
- **Bρ = p/q □ □A**

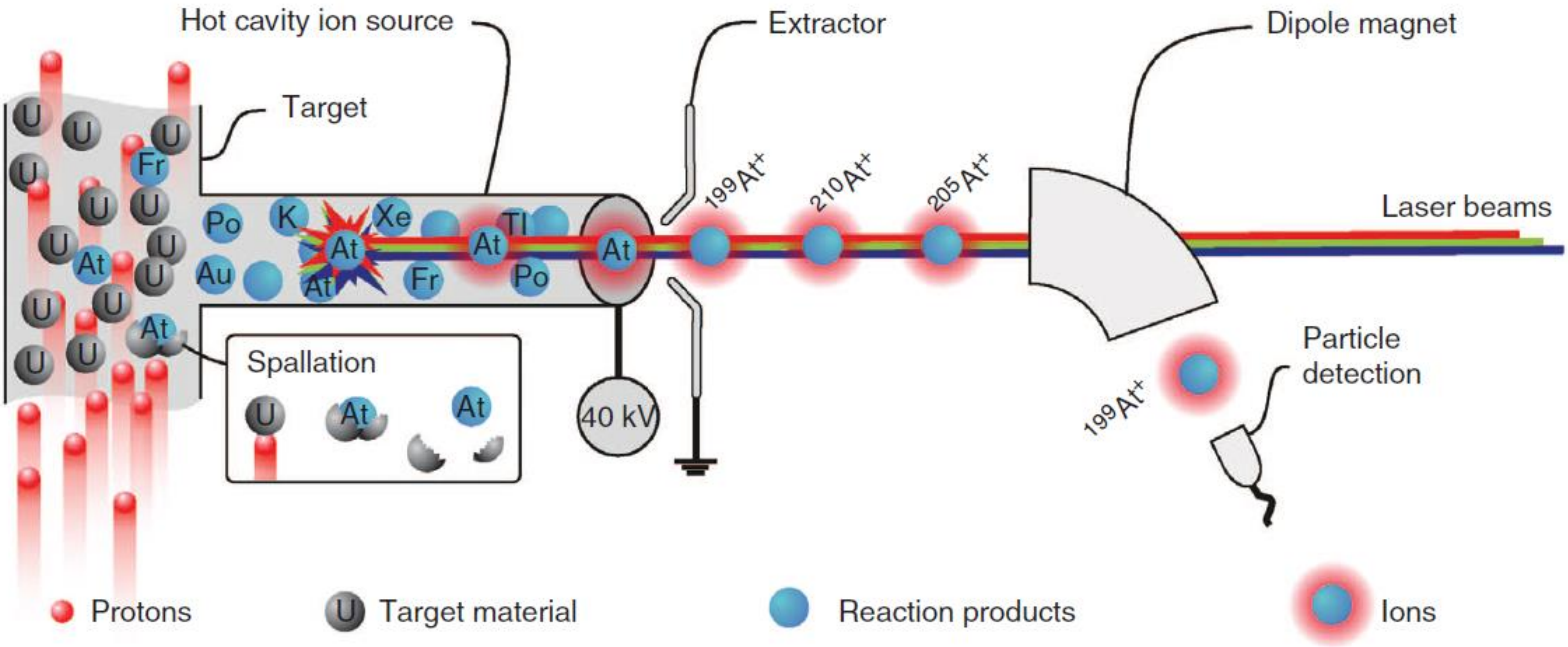


Production of ^{149}Tb, ^{152}Tb and ^{155}Tb at ISOLDE



radioactive ion beams

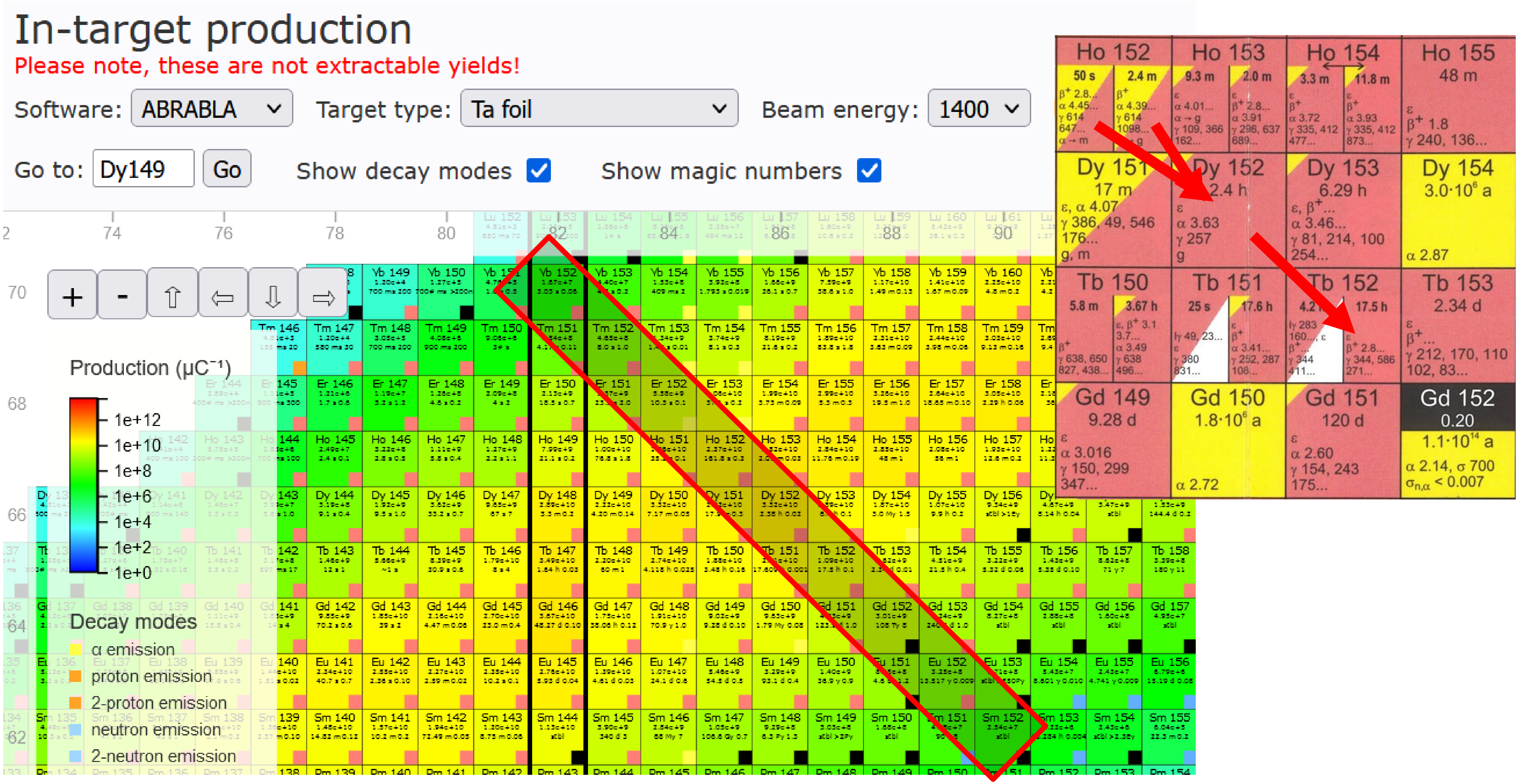
Resonant laser ionization combined with mass separation



Poster by Maryam Mostamand et al.

S. Rothe et al., Nature Comm 2013;4:1835.

Predicted production rates



In-target production

Please note, these are not extractable yields!

Software: ABRABLA

Target type: Ta foil

Beam energy: 1400

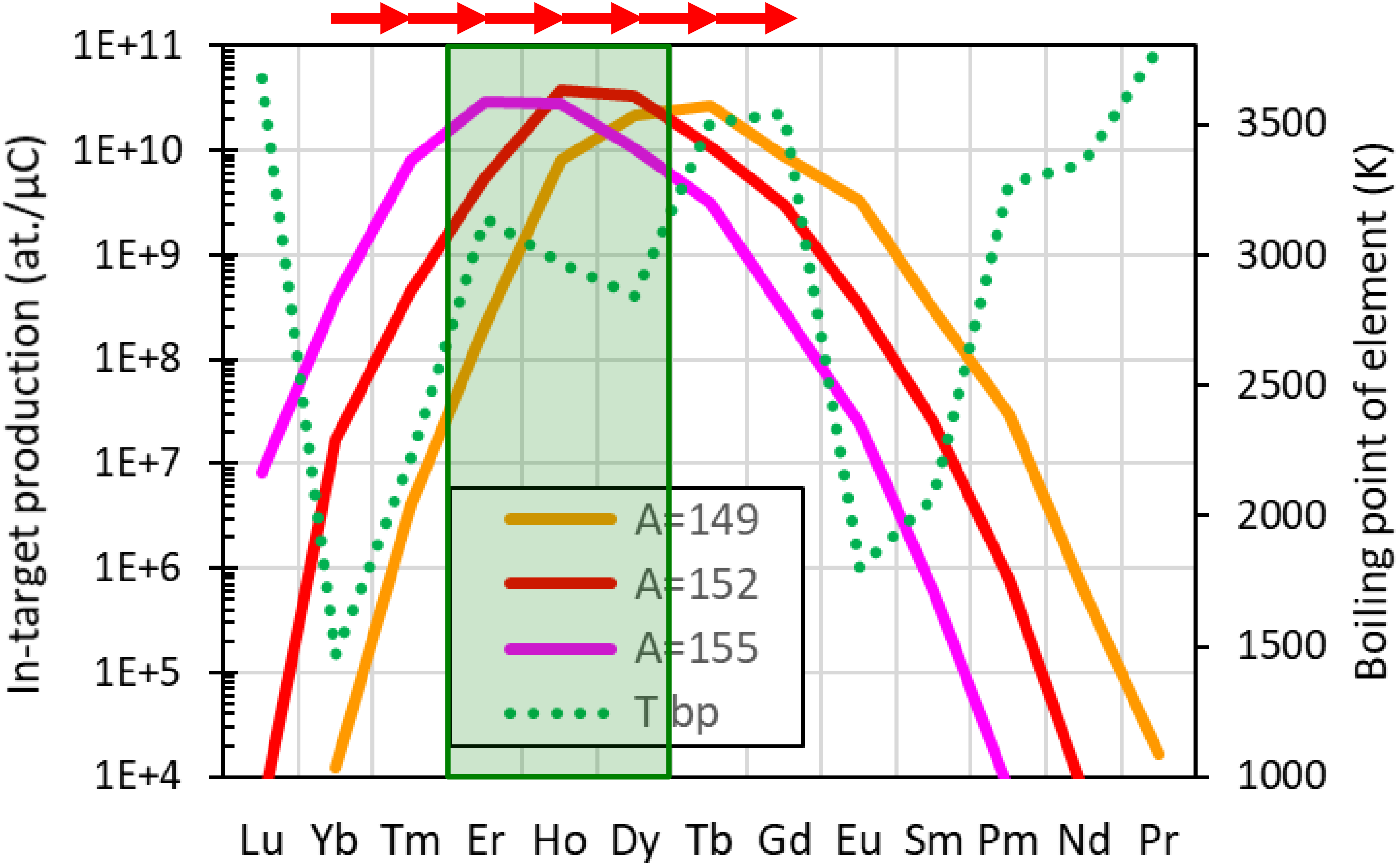
Go to: Dy149

Show decay modes ☑

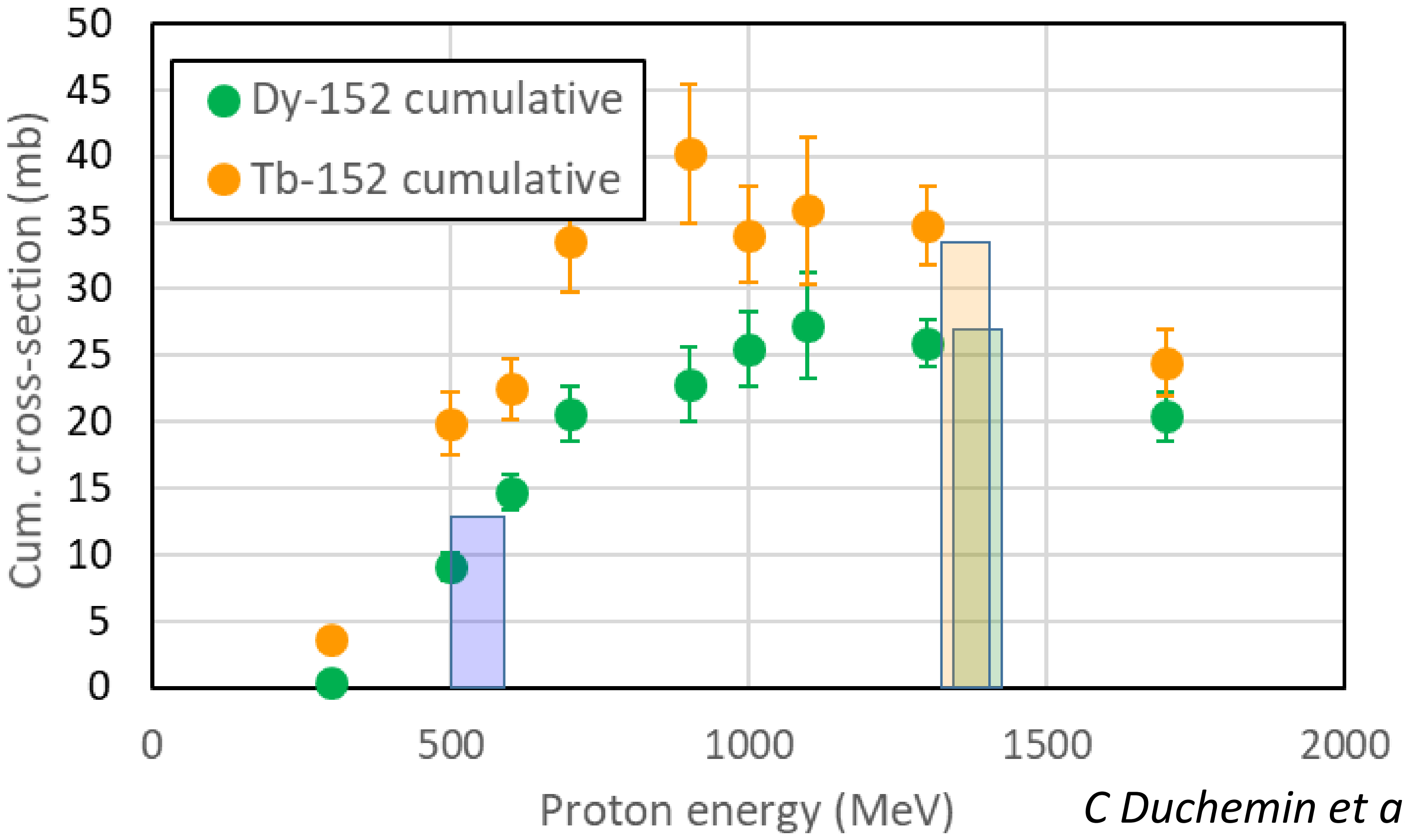
Show magic numbers ☑

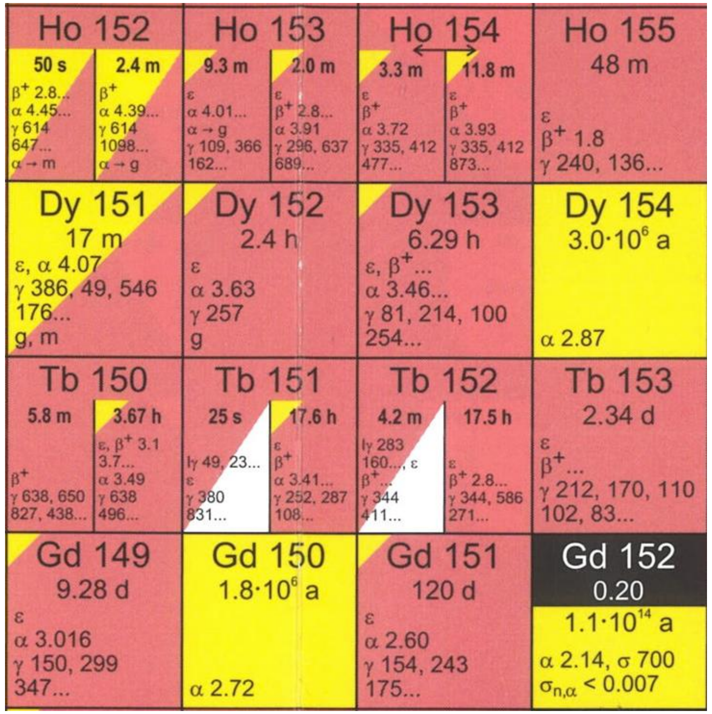
https://isoyields2.web.cern.ch/InTargetProductionChart.aspx

Predicted production rates



^{152}Tb production from Ta targets





C Duchemin et al., Front Med 2021;8:625561.

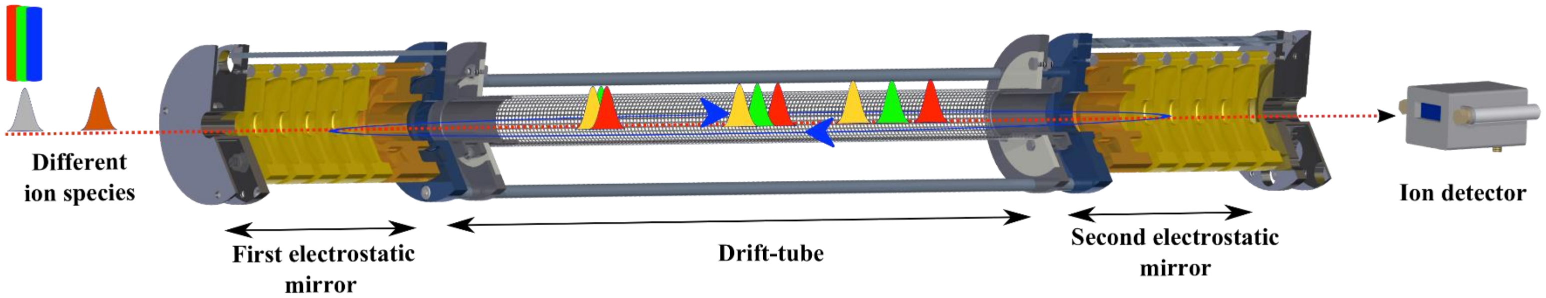
ISOLDE 1.4 GeV protons on 50 g/cm^2 ^{nat}Ta target □25 GBq/µA Dy-152 in target!

MEDICIS 1.4 GeV protons on 50 g/cm^2 ^{nat}Ta target □35 GBq/µA Tb-152 in target!

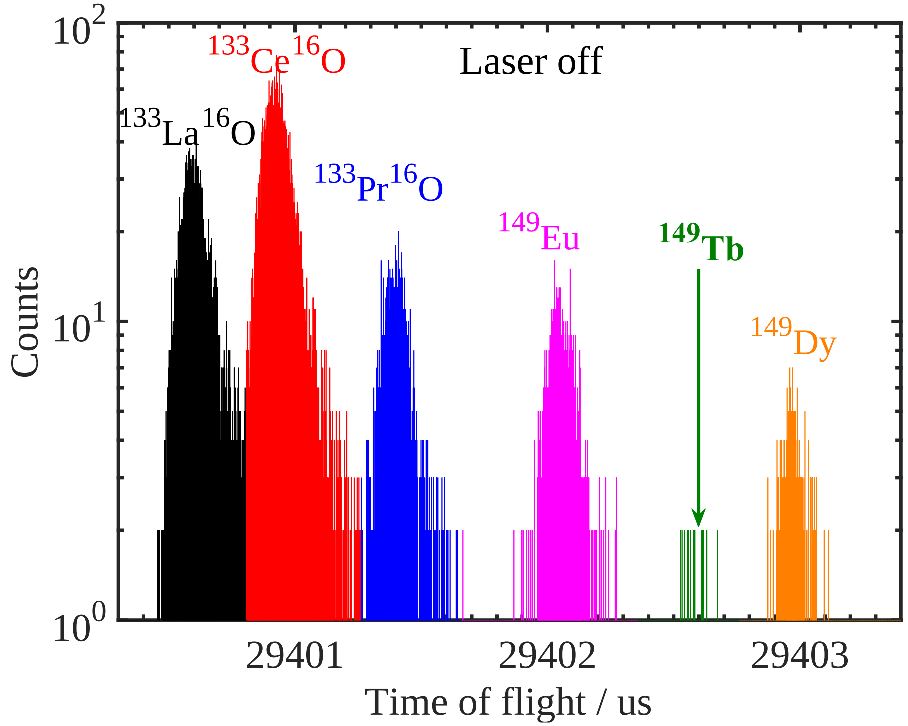
ISOLDE 1.5 µA for 5 h: **□1 GBq at EOI**

TATTOOS 100 µA for 12 h: **□100 GBq at EOI** **□300 patients**

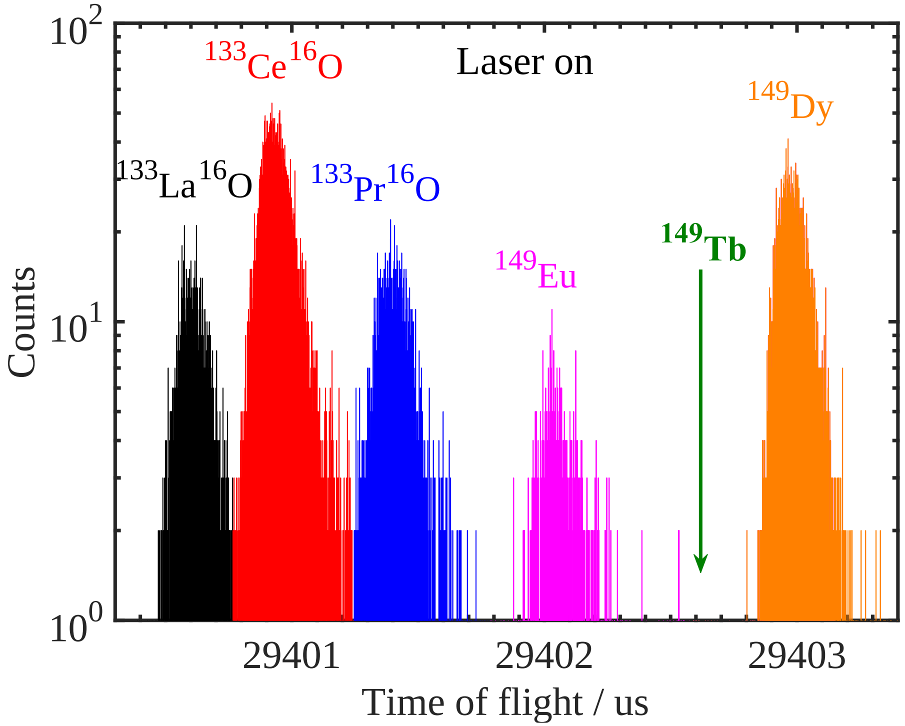
Beam optimization with ISOLTRAP’s MR-ToF-MS



1000 revolutions □ m/□m □ 115000



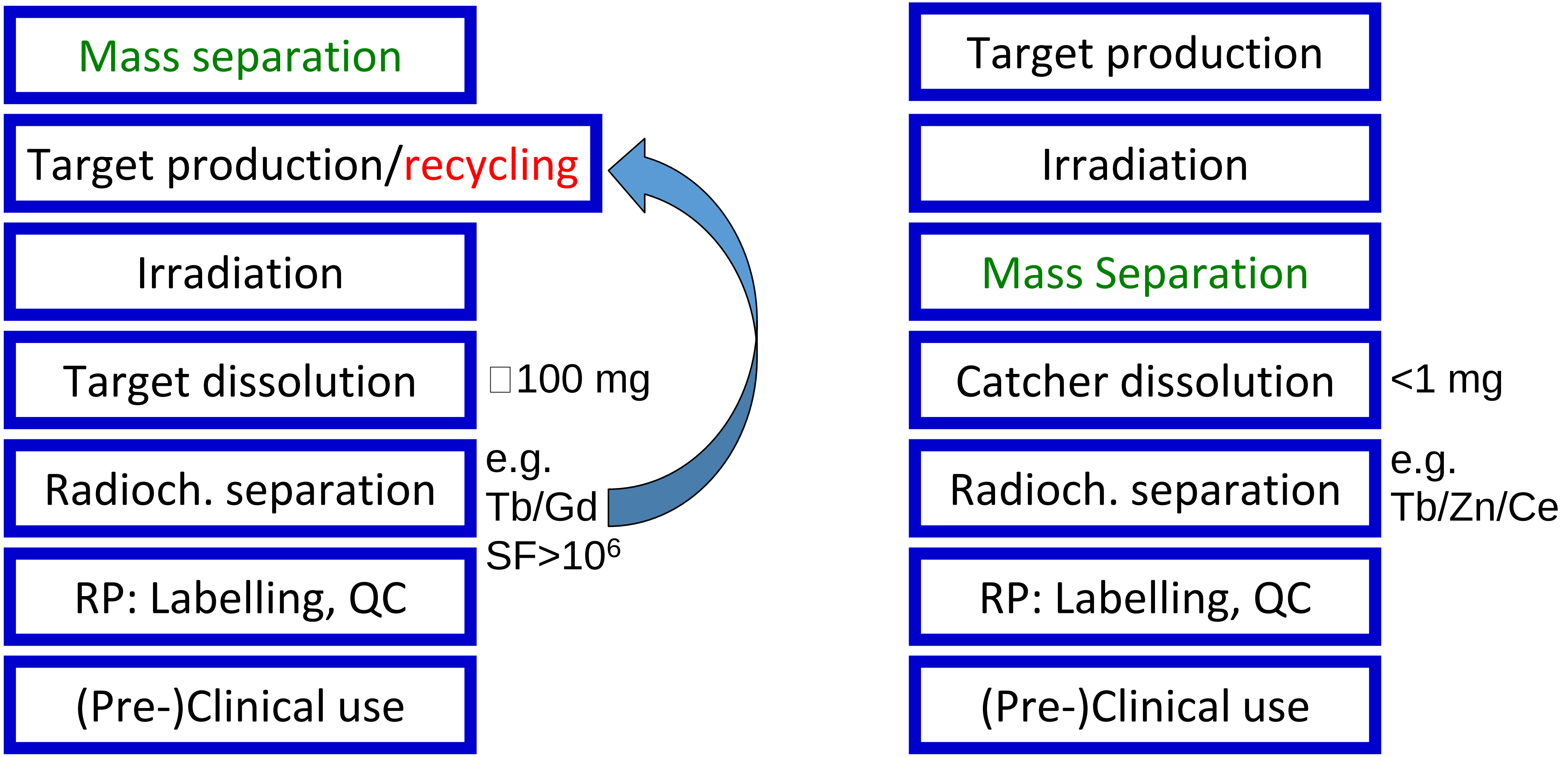
Much more Dy than Tb



Laser on/off ratio □ 10

Poster by Wiktoria Wojtaczka et al.

Production schemes



“Classical” (p,n) reaction

- Mass separation
- Target production/recycling
- Irradiation
- Target dissolution
- Radioch. separation
- RP: Labelling, QC
- (Pre-)Clinical use

□100 mg

e.g.
Tb/Gd
SF>10^6

Spallation + off-line mass separation

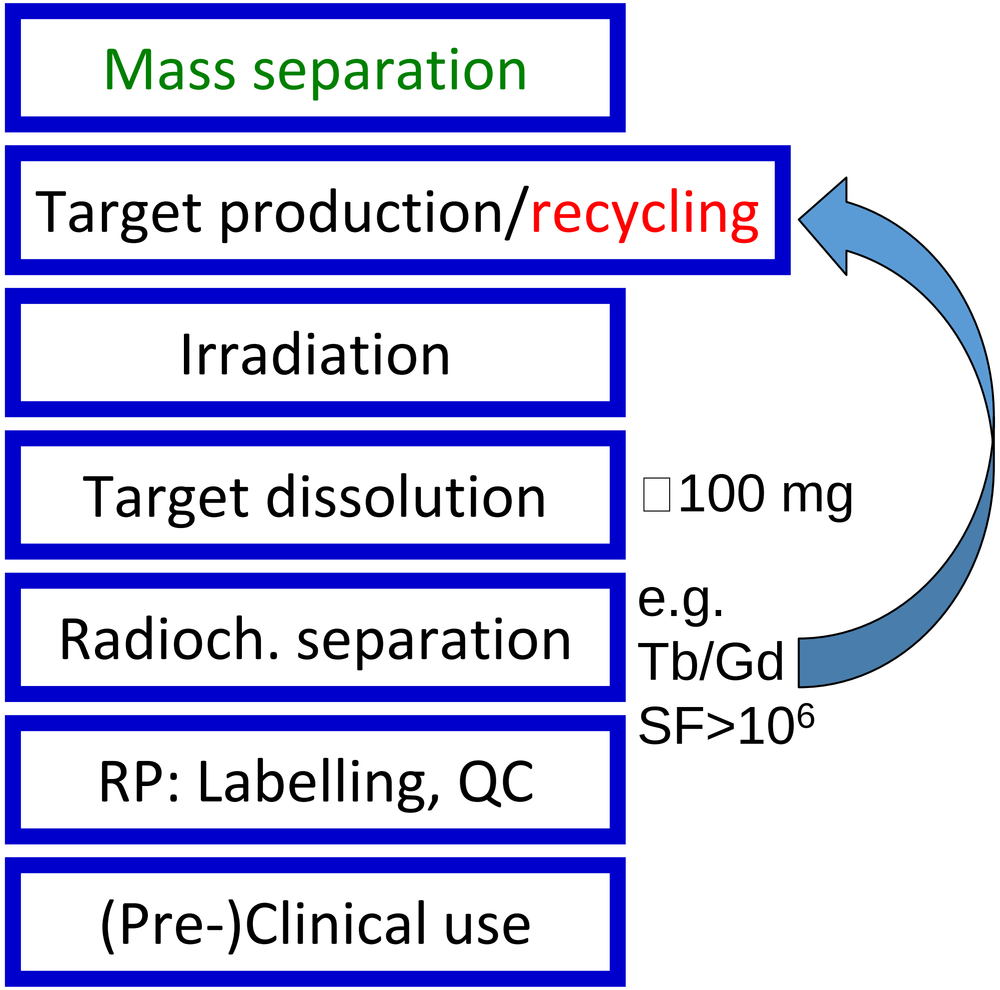
- Target production
- Irradiation
- Mass Separation
- Catcher dissolution
- Radioch. separation
- RP: Labelling, QC
- (Pre-)Clinical use

<1 mg

e.g.
Tb/Zn/Ce

Production schemes

“Classical” (p,n) reaction



- Mass separation
- Target production/recycling
- Irradiation
- Target dissolution □100 mg
- Radioch. separation e.g. Tb/Gd SF$>10^6$
- RP: Labelling, QC
- (Pre-)Clinical use

Spallation + ISOL

isotope separation on-line

- Target production
- Irradiation + Mass Separation
- Catcher dissolution <1 mg
- Radioch. separation e.g. Tb/Zn/Ce
- RP: Labelling, QC
- (Pre-)Clinical use

References

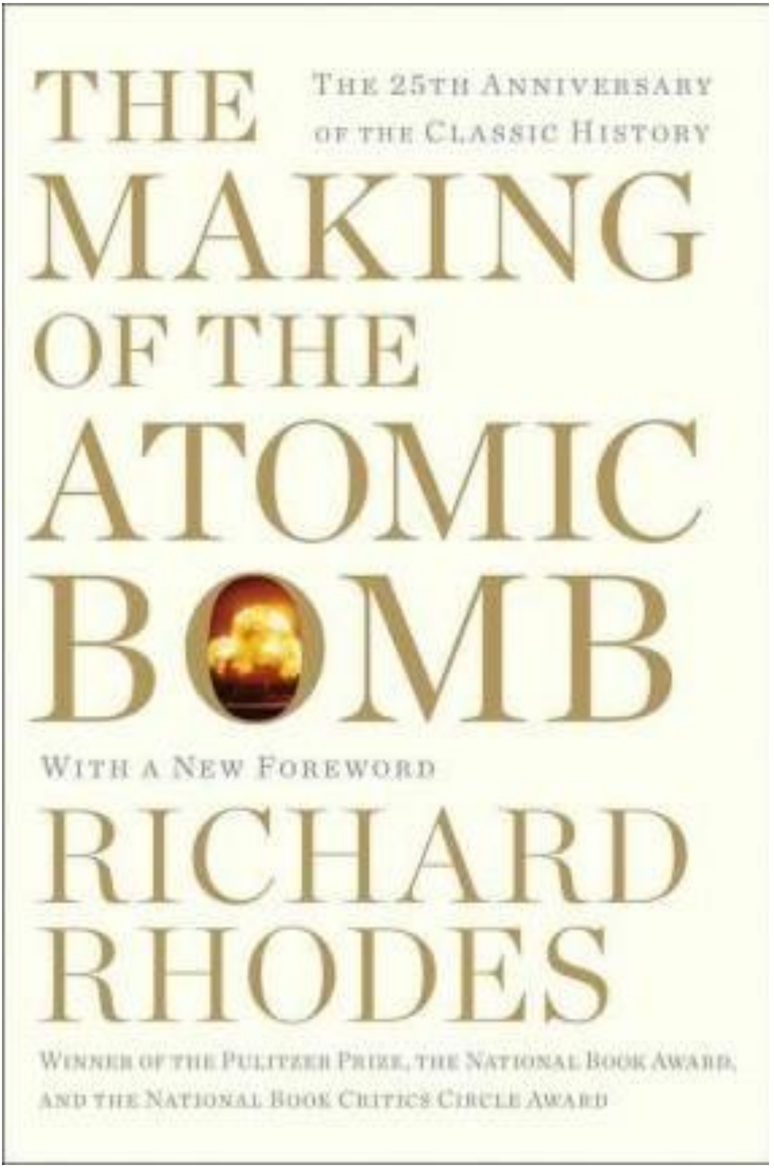
The Making of the Atomic Bomb, Richard Rhodes, Penguin Books, 1986.

Uranium Enrichment and Nuclear Weapon Proliferation, A.S. Krass et al., Taylor & Francis, 1983.

Heavy Water and the Wartime Race for Nuclear Energy, Per F. Dahl, IOP, 1999.

More information on isotopes, enrichment, etc. at:

http://www.wise-uranium.org



THE 25TH ANNIVERSARY OF THE CLASSIC HISTORY

THE MAKING OF THE ATOMIC BOMB

WITH A NEW FOREWORD

RICHARD RHODES

WINNER OF THE PULITZER PRIZE, THE NATIONAL BOOK AWARD, AND THE NATIONAL BOOK CRITICS CIRCLE AWARD

Isotope Separation by gaseous diffusion

Net effusive flow through a hole (ideal gas law):

$J \cdot A = v \cdot A / (4\, R\, T) \cdot (p_f - p_b)$

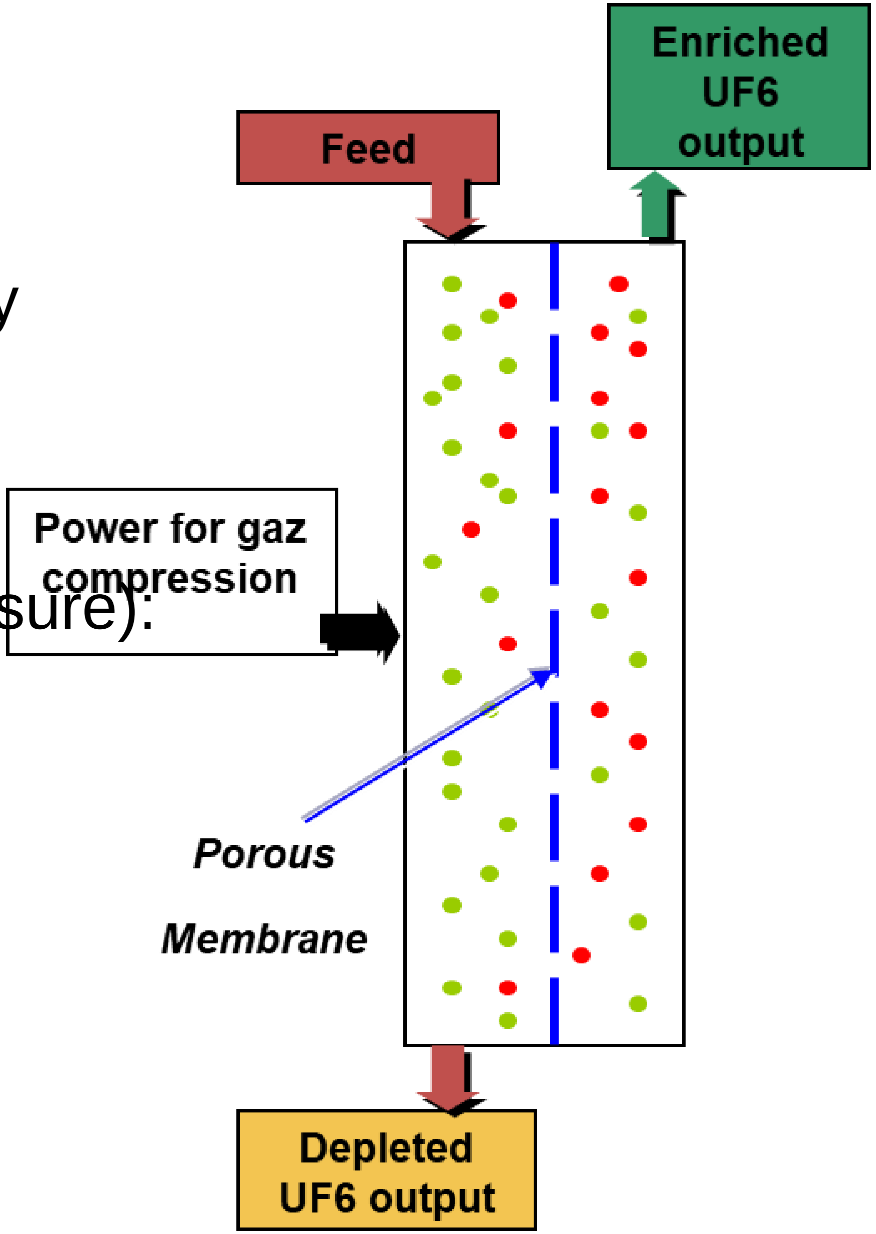
$v = (8\, R\, T / \pi\, \mathbf{M})^{1/2}$ Maxwell mean velocity

J: net flow (mol/s) through surface A

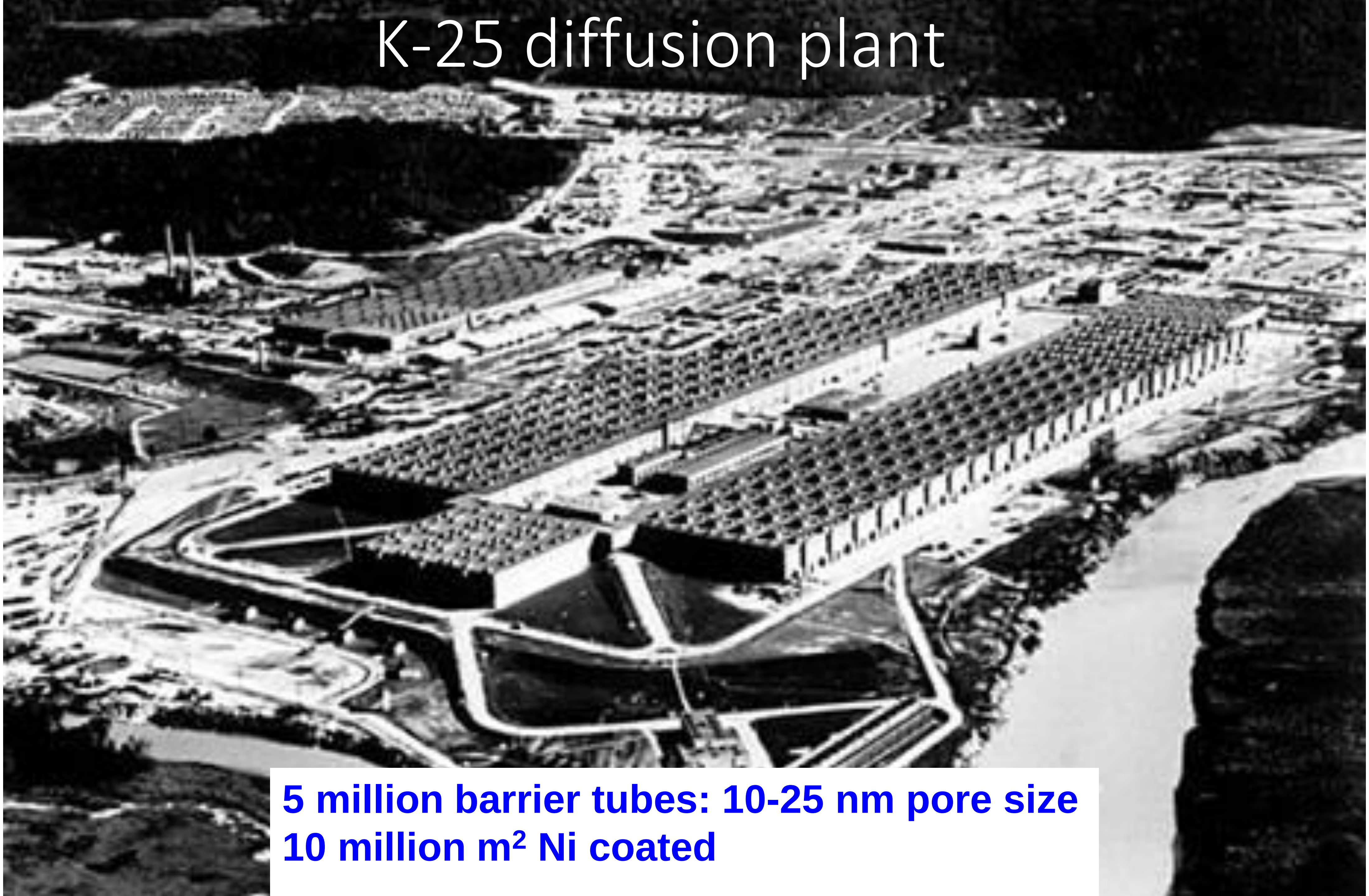
p_f: fore-pressure, p_b back-pressure

ideal elementary separation factor (negligible back-pressure):

$\boldsymbol{\alpha = (M_2 / M_1)^{1/2}}$ for ${}^{235,238}UF_6$: $\alpha = 1.00429$

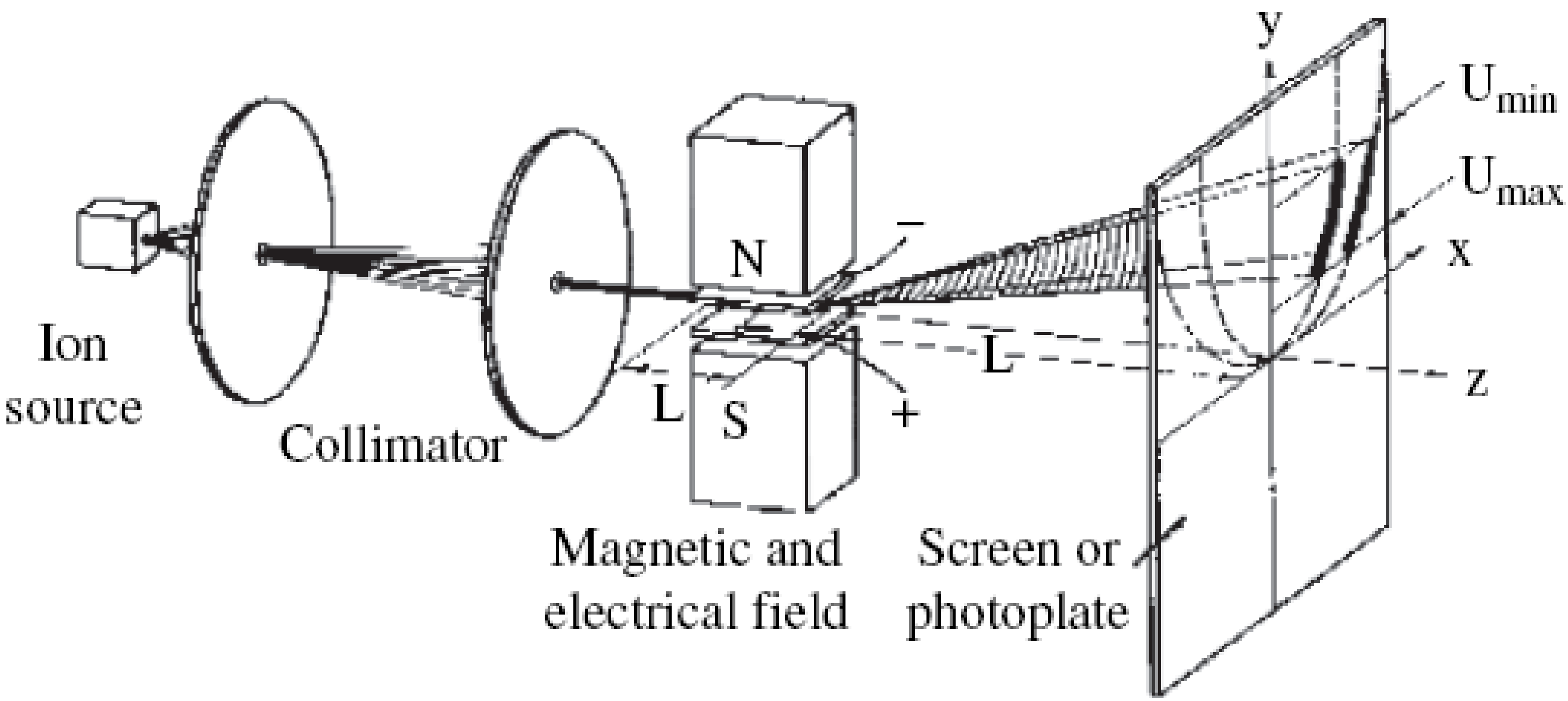


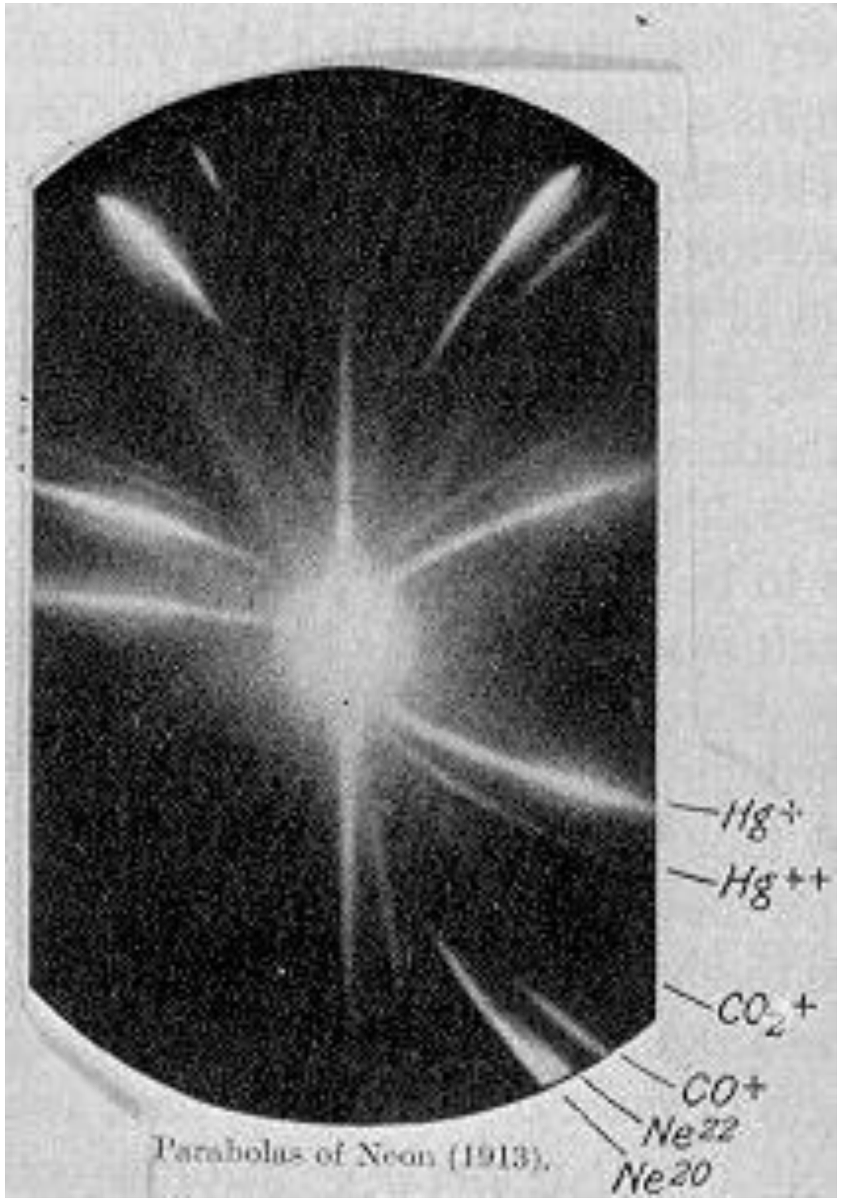
K-25 diffusion plant



5 million barrier tubes: 10-25 nm pore size
10 million m^2 Ni coated

1910: Thomson parabola mass spectrograph





$y = k\,\mathbf{m}/q\,x^2$; $k = 2\,U/(d\,B^2\,L^2)$

1913 discovery of isotopes

The LOHENGRIN fission fragment spectrometer

NEUTRONS
FOR SOCIETY

Efficient parallel operation

ISOLTRAP

^{152}Tb

^{149}Tb

^{155}Tb

HRS setup

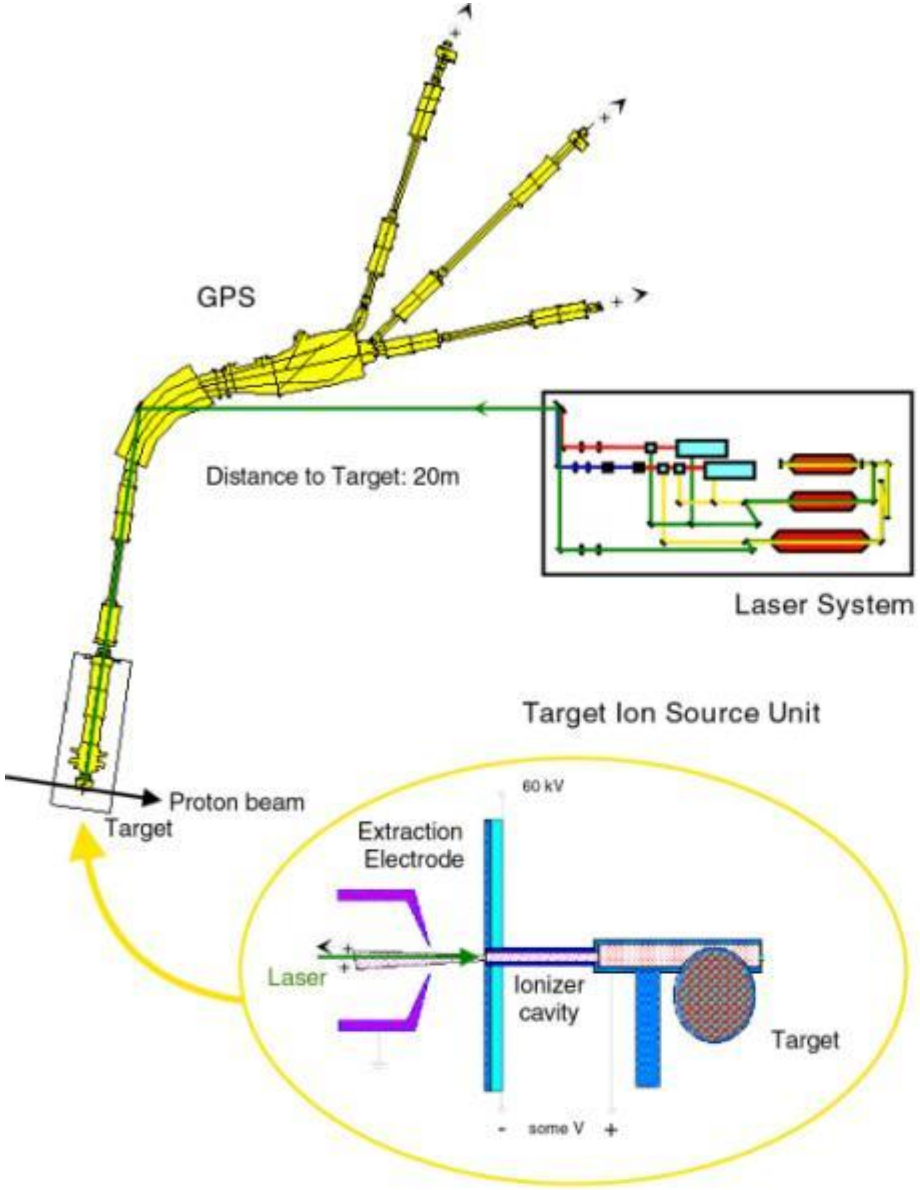
Costs

Compared to n.c.a. ^{177}Lu

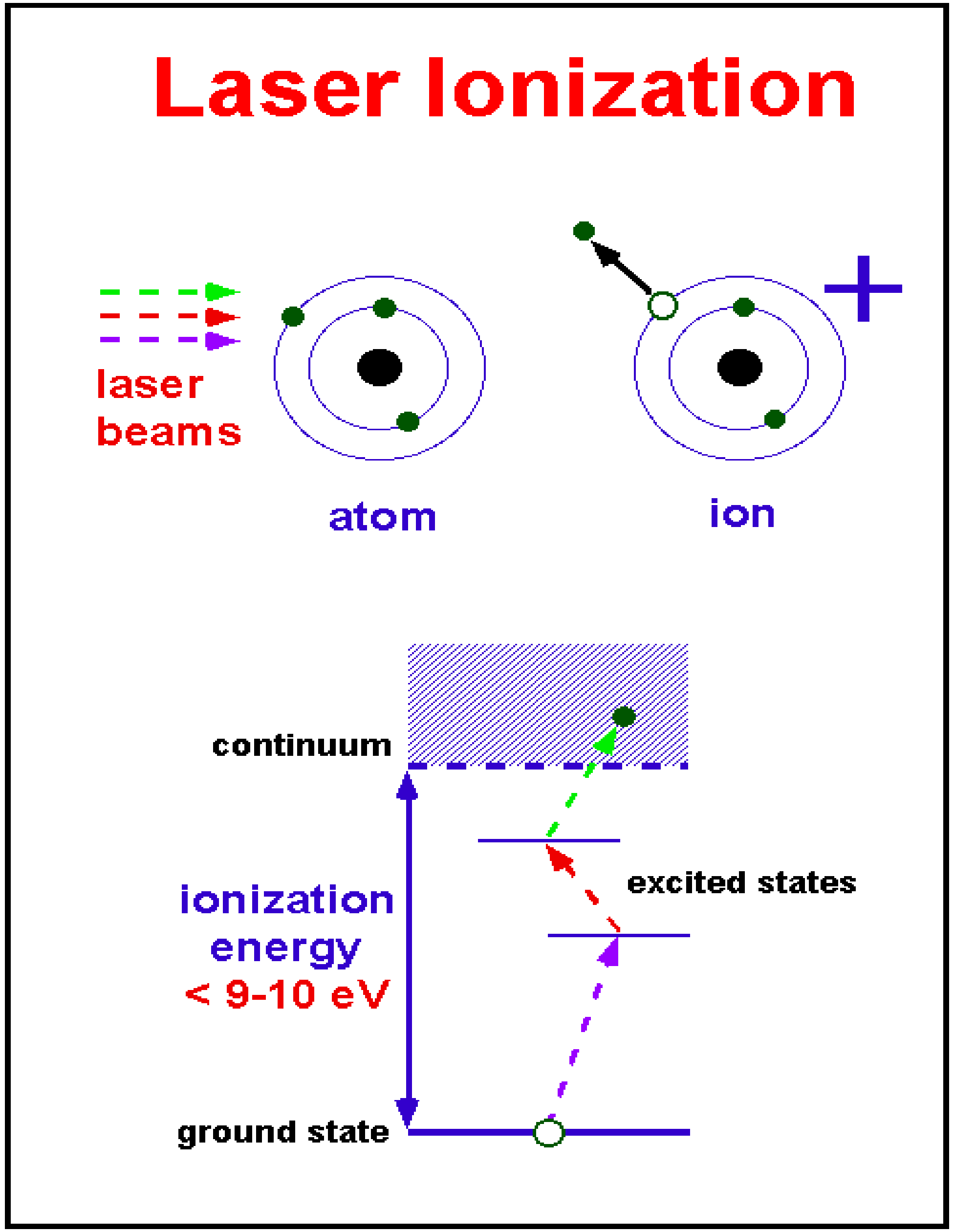
	^{177}Lu	^{161}Tb	Advantage ^{161}Tb
Activity per injection (GBq)	7	⍰5	⍰1.4
Cross-section ^{176}Yb or ^{160}Gd (b)	2.85	1.5	0.55
Historic calutron throughput (g/tank d)	0.16	0.2	1.25
Natural abundance (% ^{176}Yb or ^{160}Gd)	13	21.9	1.7
Co-production of other useful isotopes	^{168}Yb	^{152}Gd ^{155}Gd ^{157}Gd	++
Enriched isotope costs per injection			**⍰ 1 – 1.3**
Chemical separation (Lu/Yb vs. Tb/Gd)	1.54	2.4	++

Industrially produced n.c.a. ^{161}Tb should not be more expensive than n.c.a. ^{177}Lu !

Resonance Ionization Laser Ion Source



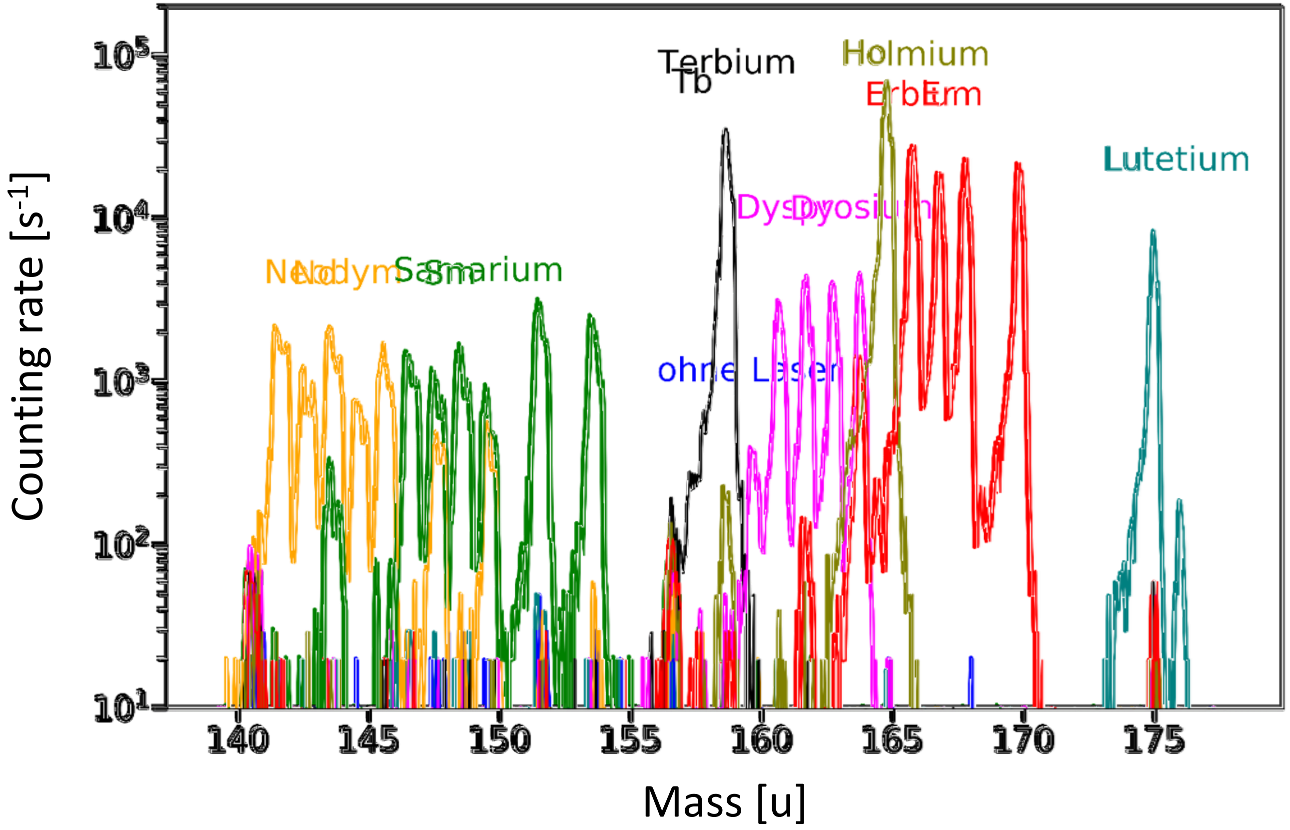
Spectrochim. Acta 2003;B58:1047.

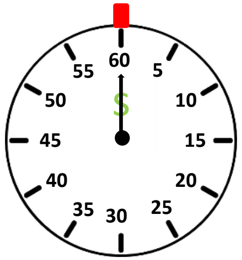


Automated Switching between Ionization Schemes

Nd → 2 min → **Sm** → 2 min → **Tb** → 2 min → **Dy** → 2 min → **Ho** → 2 min → **Er** → 2 min → **Lu**

Ion Source at 1560 °C

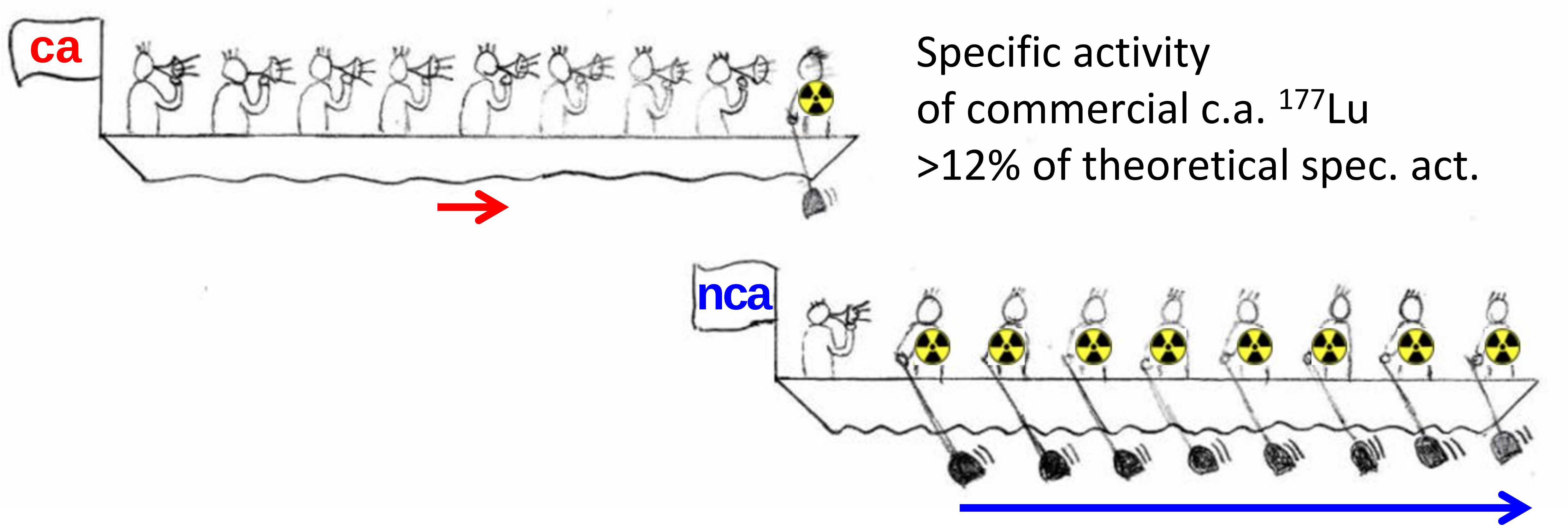




Courtesy of Felix Weber & Vadim Gadelshin, JGU Mainz

JOHANNES GUTENBERG UNIVERSITÄT MAINZ JG|U

Carrier added vs. non-carrier added



Specific activity
of commercial c.a. ^{177}Lu
>12% of theoretical spec. act.

Irradiation of 100% enriched precursor
for 1x $T_{1/2}$ in very high neutron flux ($\varphi = 10^{15}$ cm^{-2}s^{-1}) and 1 day decay:
^{177}Lu ≈70% of theoretical specific activity

^{89}Sr 1.7%, ^{90}Y 0.02%, ^{153}Sm 1.7%, ^{166}Ho 0.2%, ^{169}Er 0.2%

Reactor produced radionuclides

Direct production

Carrier-added (c.a.)
Limited specific activity
Limited radionuclidic purity
“Easy & cheap”
c.a. ^{177}Lu, ^{89}Sr, ^{90}Y, ^{153}Sm, ^{166}Ho, ^{169}Er, ^{186}Re, etc.

Indirect production

No-carrier added (n.c.a.)
Close to theoretical spec. act.
Optimum radionuclidic purity
Needs radiochem. separation
n.c.a. ^{177}Lu, ^{111}Ag, ^{149}Pm, ^{161}Tb
Generator: ^{47}Sc, ^{90}Y, ^{99m}Tc, ^{166}Ho, ^{188}Re, etc.
mainly odd Z radionuclides !

Direct + mass-separation

No-carrier added (n.c.a.)
Close to theoretical spec. act.
Optimum radionuclidic purity
Still under R&D
n.c.a. ^{153}Sm, ^{169}Er, ^{175}Yb, etc.

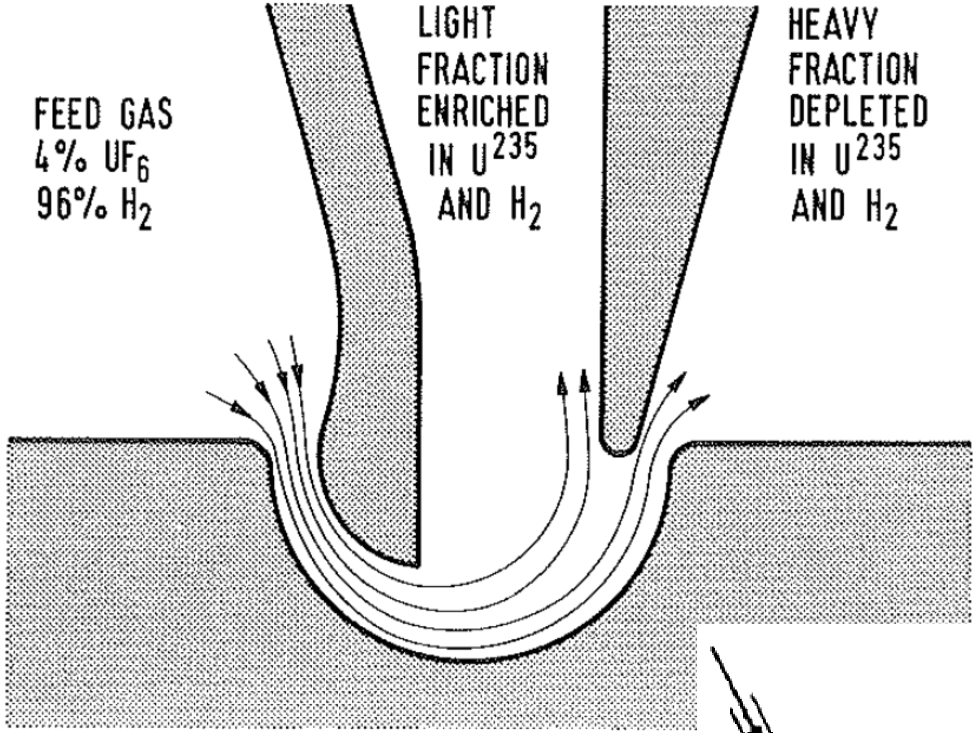
^{161}Tb costs compared to n.c.a. ^{177}Lu

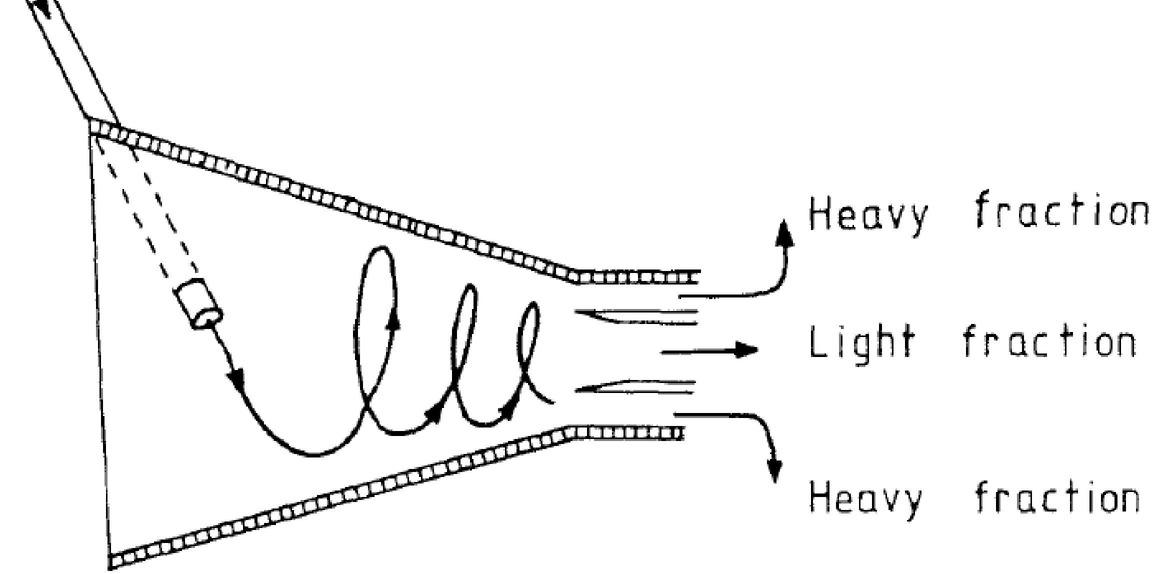
	^{177}Lu	**^{161}Tb**	**Advantage ^{161}Tb**
Equitoxic activity per injection (GBq)	7.4	5.4	1.4
Cross-section ^{176}Yb or ^{160}Gd (b)	2.8	1.4	0.5
Historic calutron throughput (g/tank d)	0.16	0.2	1.25
Natural abundance (% ^{176}Yb or ^{160}Gd)	13	21.9	1.7
Co-production of other useful isotopes	^{168}Yb	^{152}Gd ^{155}Gd ^{157}Gd	++
Enriched isotope costs per injection			** 0.9 – 1.2**
Chemical separation (Lu/Yb vs. Tb/Gd)	1.54	2.4	++

Industrially produced n.c.a. ^{161}Tb should have similar cost to n.c.a. ^{177}Lu.

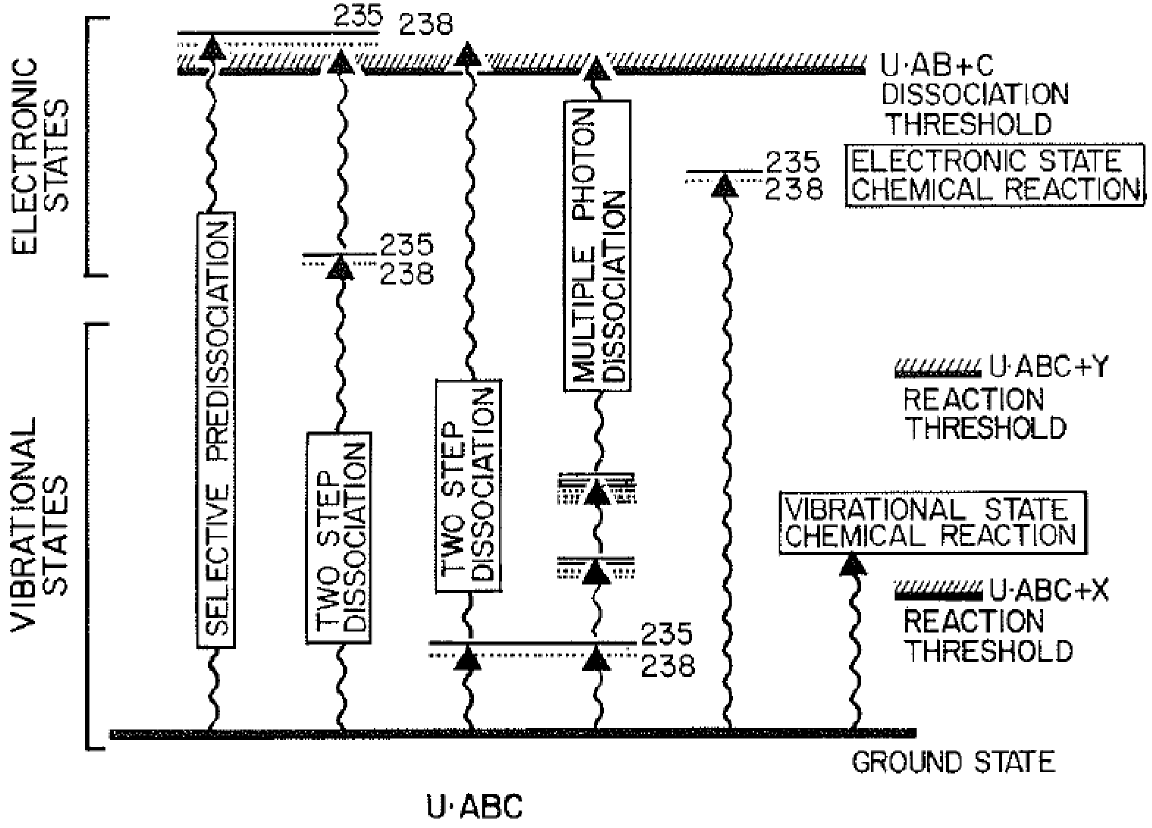
Highly correlated production chain ^{161}Tb is **not** an independent backup !

Isotope separation by nozzle or vortex

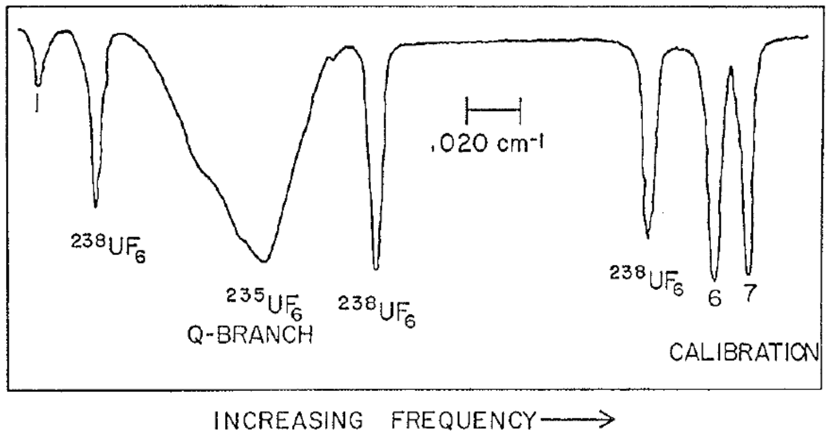




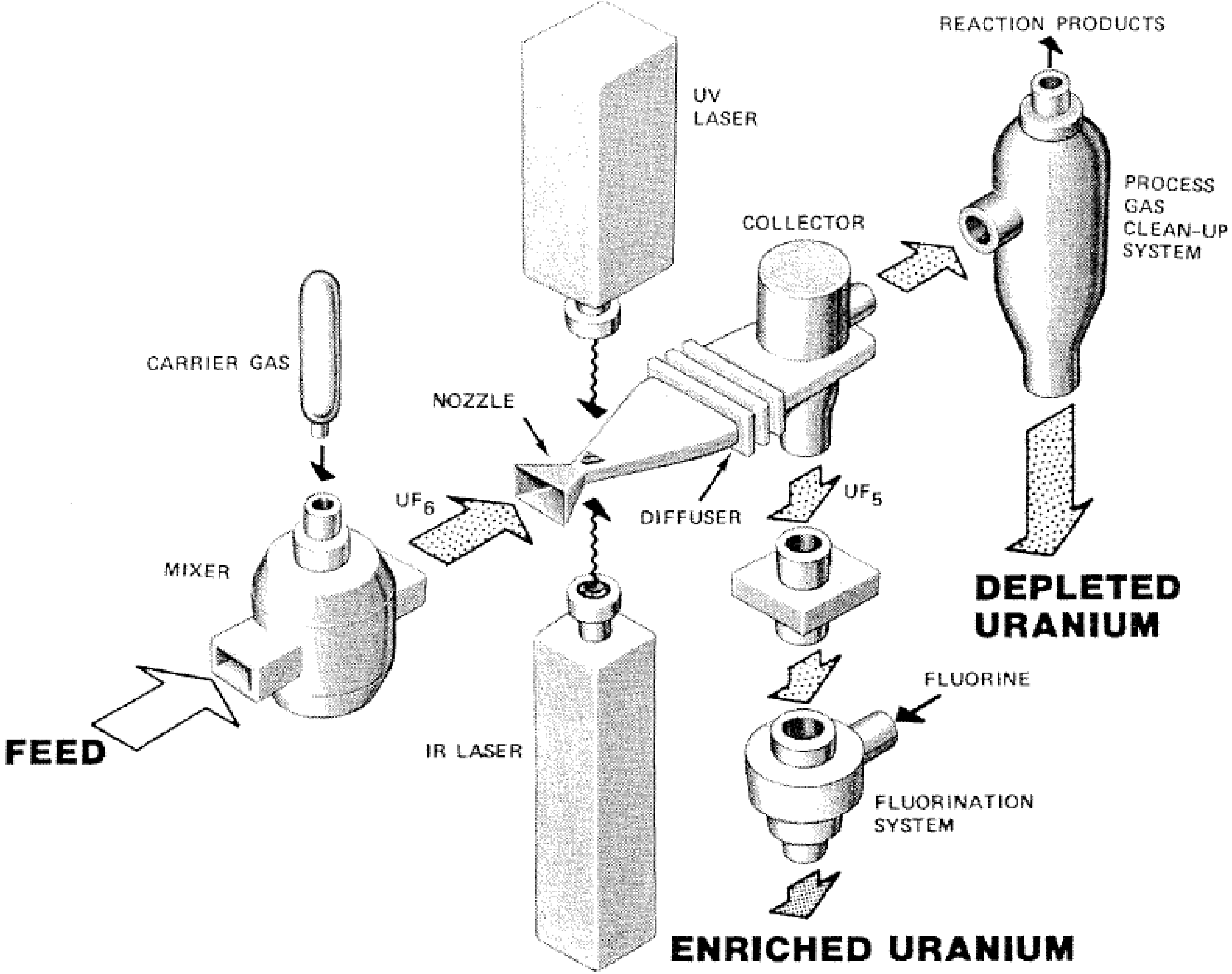
Molecular Laser Isotope Separation



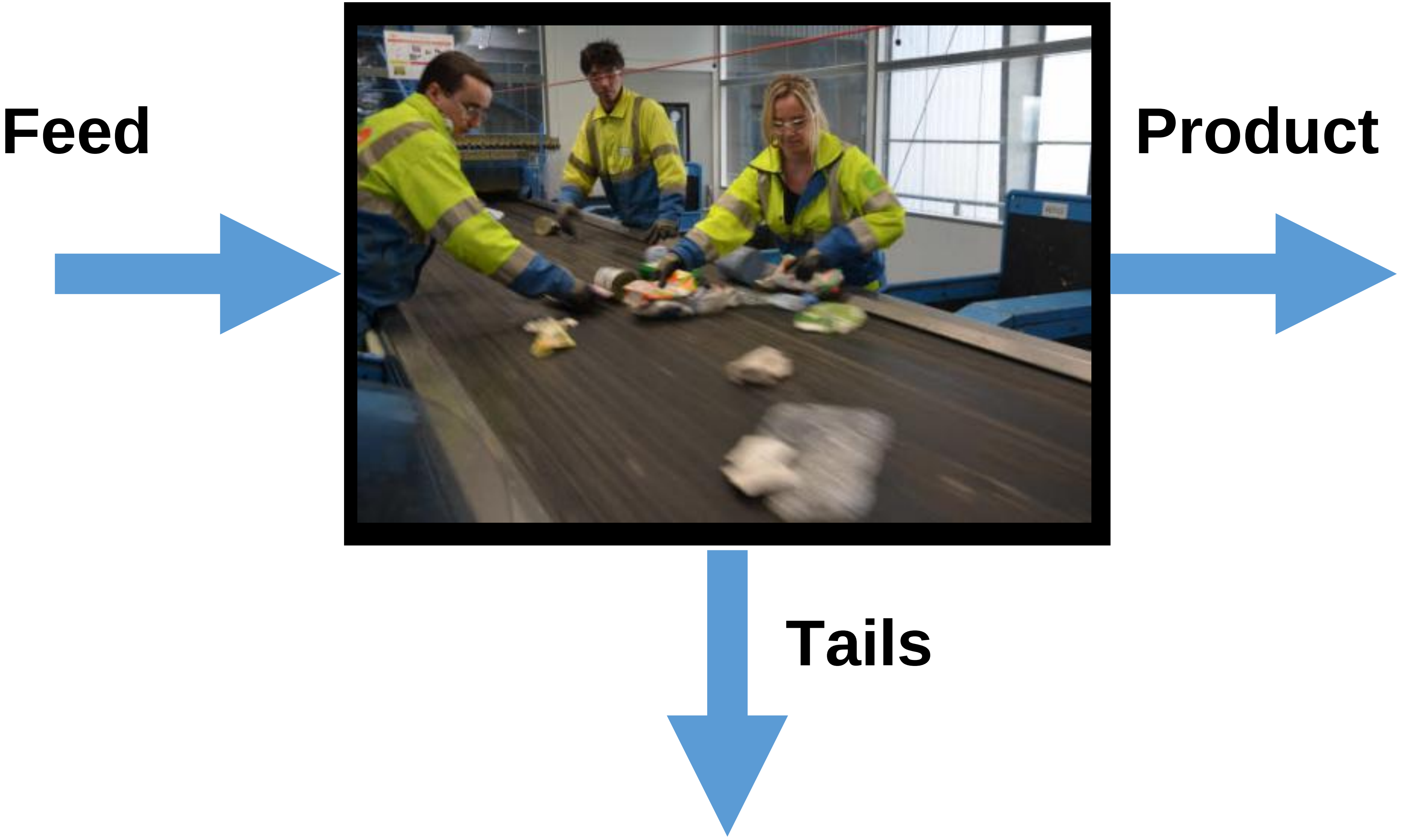
LASER DIODE SCAN OVER THE Q-BRANCH OF THE ν_3 MODE OF $^{235}UF_6$



Molecular Laser Isotope Separation: SILEX process



The Separative Work Unit

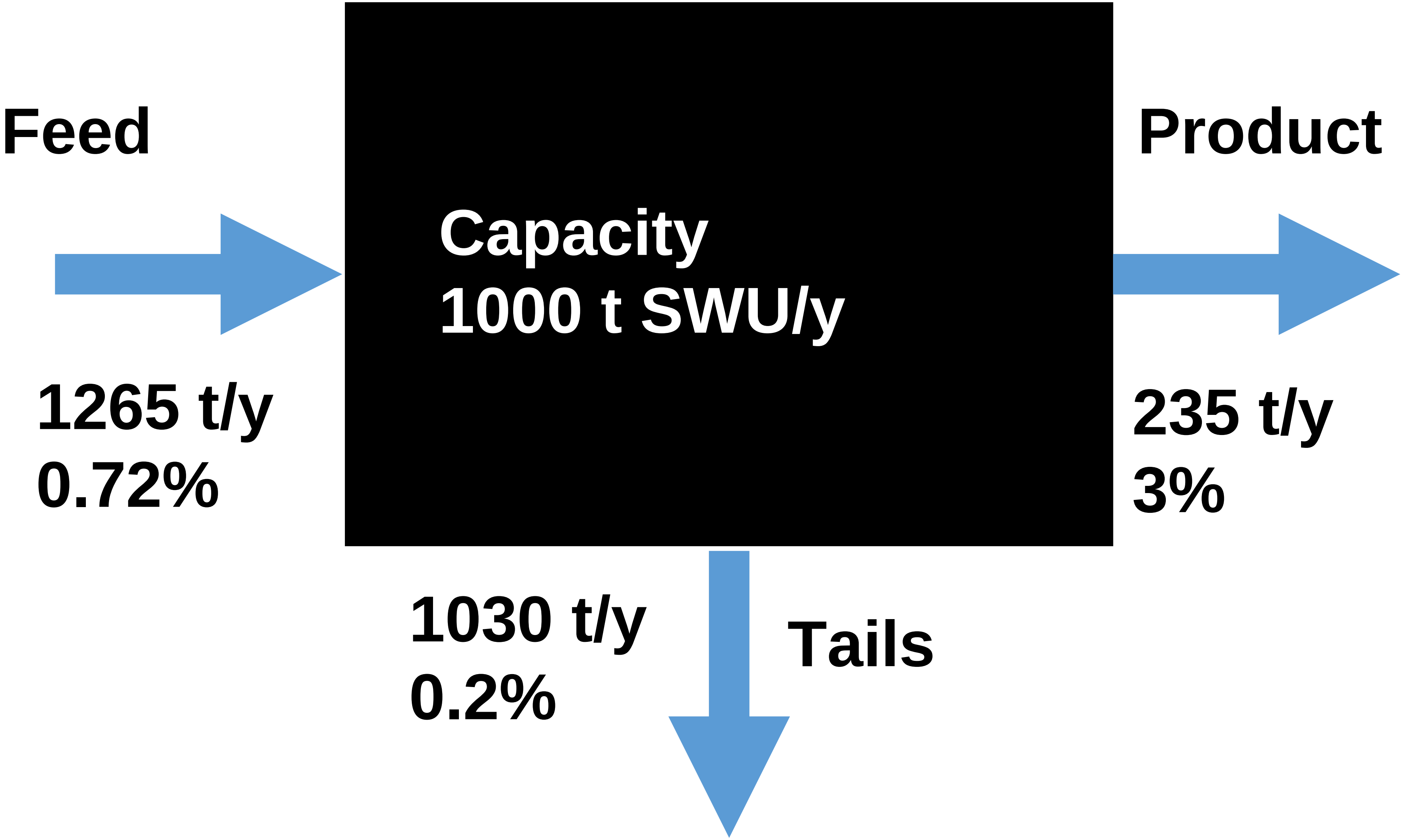


Feed

Product

Tails

The Separative Work Unit



Feed

1265 t/y
0.72%

Capacity
1000 t SWU/y

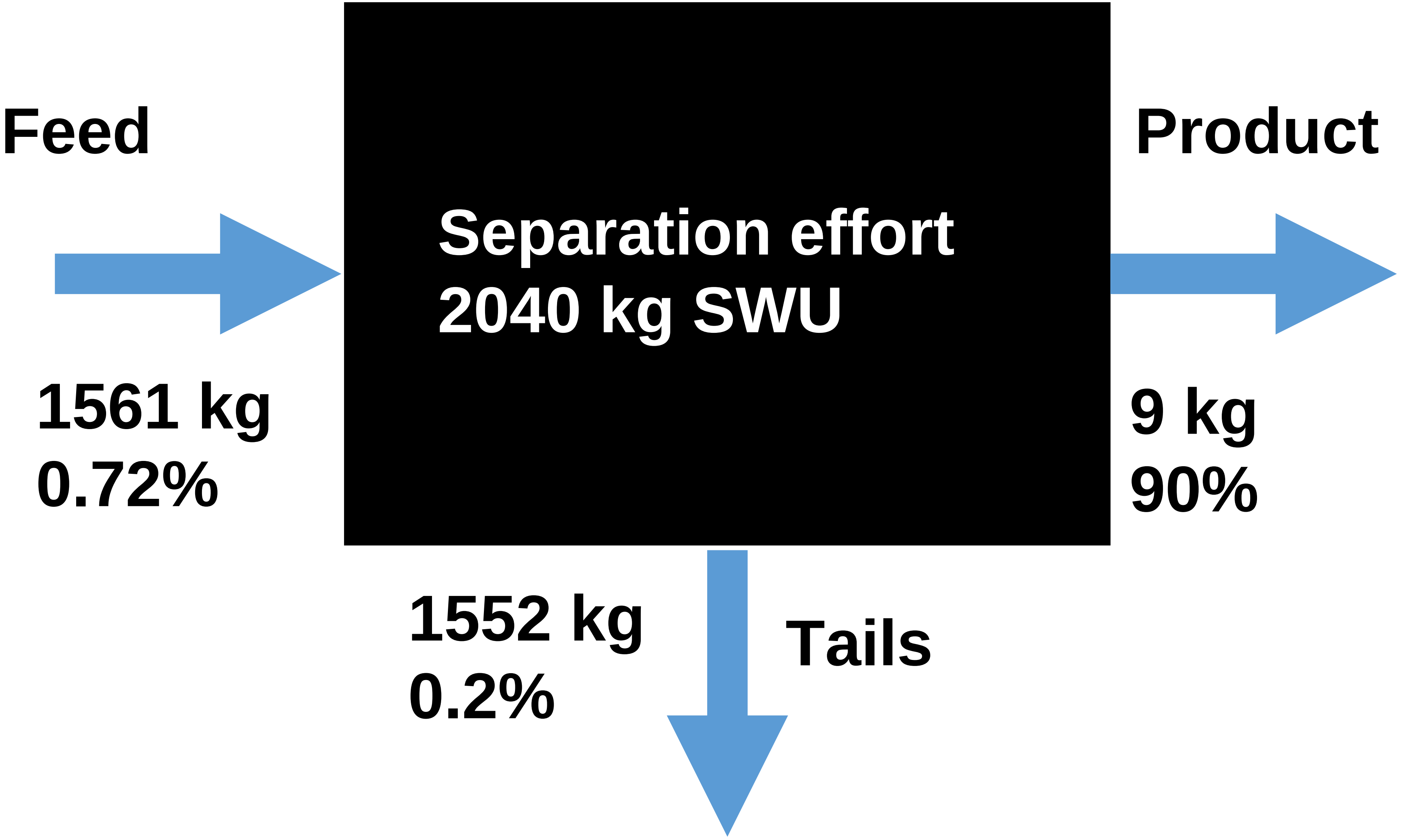
Product

235 t/y
3%

1030 t/y
0.2%

Tails

The Separative Work Unit



Feed

1561 kg
0.72%

Separation effort
2040 kg SWU

Product

9 kg
90%

1552 kg
0.2%

Tails

Spinoffs of uranium enrichment