Oak Ridge National Laboratory

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Production and Processing of Heavy Actinides at ORNL

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U.S. DEPARTMENT of ENERGY

ORNL IS MANAGED BY UT-BATTELLE LLC
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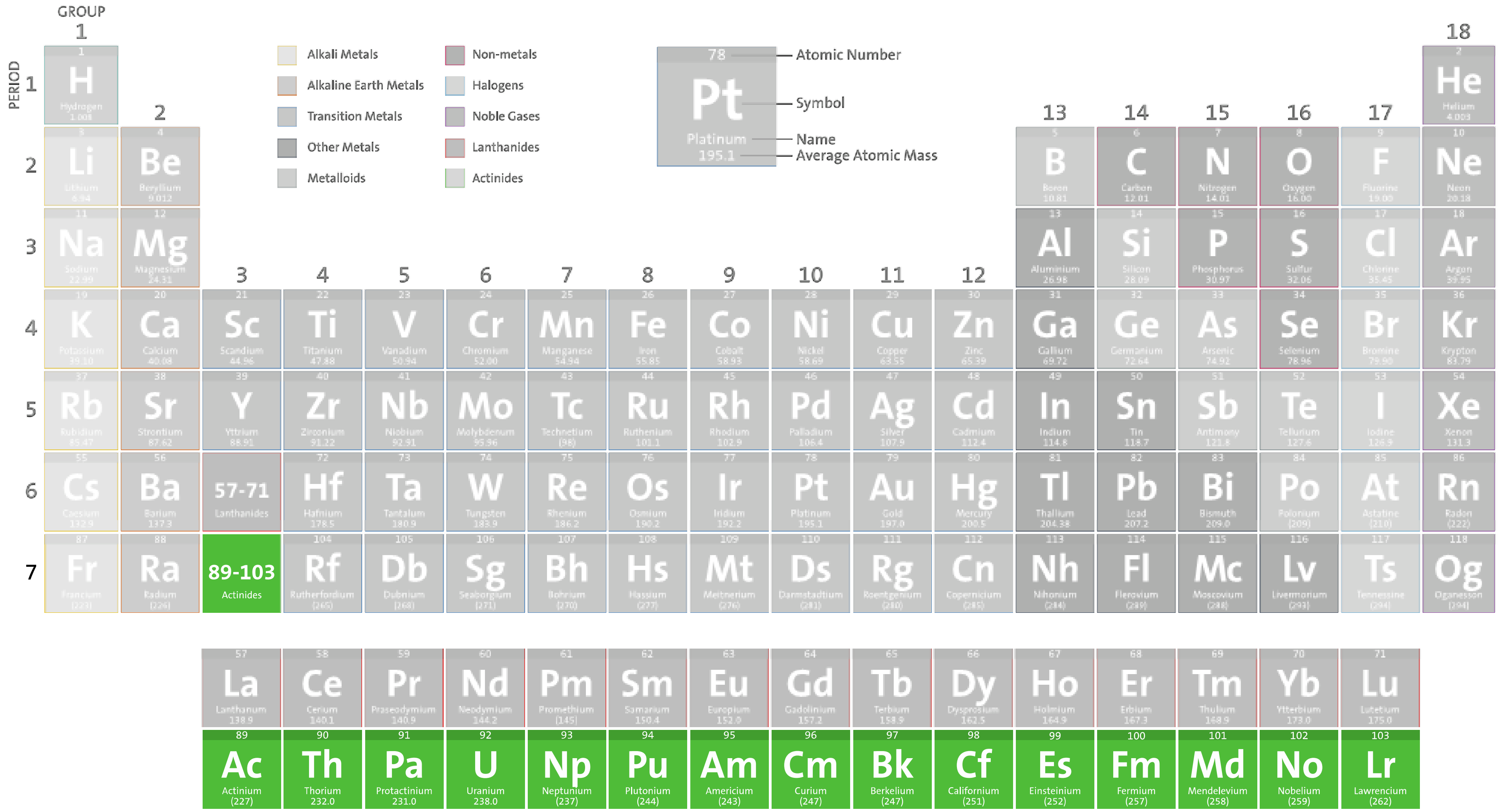
THE ACTINIDES? WHERE DO THEY COME FROM?



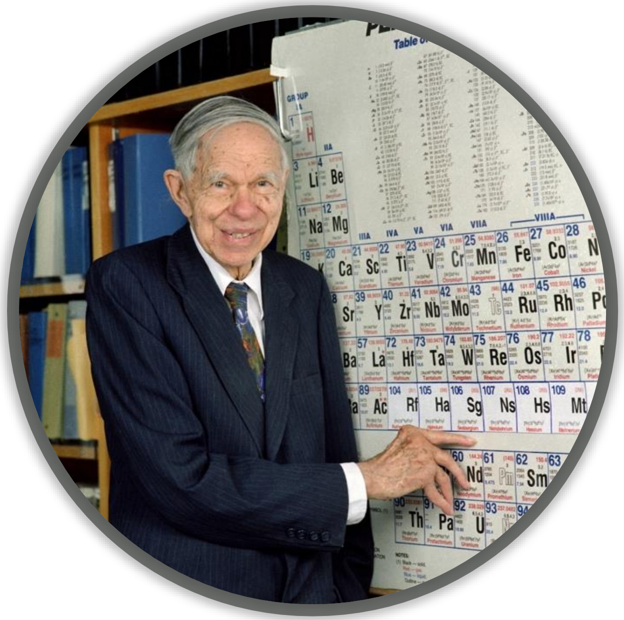
American Chemical Society

www.acs.org/outreach

ACTINIDE CONCEPT (1941)

- Seaborg proposed that the series started with thorium and that all the elements heavier than actinium constituted an “actinide” series

- Because the 5*f* shell began filling in the same relative position as the 4*f* shell, the electronic configuration of elements in the two series would be similar



	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
Year of discovery/ creation	1899	1829	1913	1789	1940	1940	1944	1944	1949	1950	1952	1952–54	1955	1958–66	1961–65
Electron configuration after [Rn] core	$6d^1 7s^2$	$6d^2 7s^2$	$5f^2 6d^1 7s^2$	$5f^3 6d^1 7s^2$	$5f^4 6d^1 7s^2$	$5f^6 7s^2$	$5f^7 7s^2$	$5f^7 6d^1 7s^2$	$5f^9 7s^2$	$5f^{10} 7s^2$	$5f^{11} 7s^2$	$5f^{12} 7s^2$	$5f^{13} 7s^2$	$5f^{14} 7s^2$	$5f^{14} 7s^2 7p^1$
Oxidation states (bold = most common)[a]	+2, **+3**	+1, +2, +3, **+4**	+2, +3, +4, **+5**	+1, +2, +3, +4, +5, **+6**	+2, +3, +4, **+5,** +6, +7	+1, +2, +3, **+4,** +5, +6, +7, +8	+2, **+3, +4,** +5, +6, +7, +8	+2, **+3,** +4, +5, +6	+2, **+3, +4**	+2, **+3,** +4, +5	+2, **+3,** +4	+2, **+3**	+2, **+3**	**+2,** +3	**+3**

Peplow. Chemists explore the periodic table’s actinide frontier. C&EN (2019) Vol. 97(9)

EXPLORING TRANSPLUTONIUM ELEMENTS

Element	Isotope	Year Discovered	Method
Pu	238	1940	U-238 + d
Am	241	1944	Pu-239 + n
Cm	242	1944	Pu-239 + ^{4}He
Bk	243	1949	Am-241 + ^{4}He
Cf	245	1950	Cm-242 + ^{4}He

Ernest Orlando Lawrence, works on the 60-inch cyclotron at the University of California Berkeley in 1937

https://elements.lbl.gov/news/berkeley-lab-history-the-60-inch-cyclotron/

AND ALONG CAME MIKE

Ivy-Mike was the first ever thermal nuclear test conducted at Eniwetok in the Marshall Islands (Nov. 1, 1952). Its denotation was equivalent to 10 million tons of TNT.

This yielded elements 94-100 (Pu-Fm).

Resulted in a multineutron capture in U-238 that was part of the device yielding Cf-253 that beta decayed to form Es-253.

Fm-255 was also part of the new isotopes discovered.

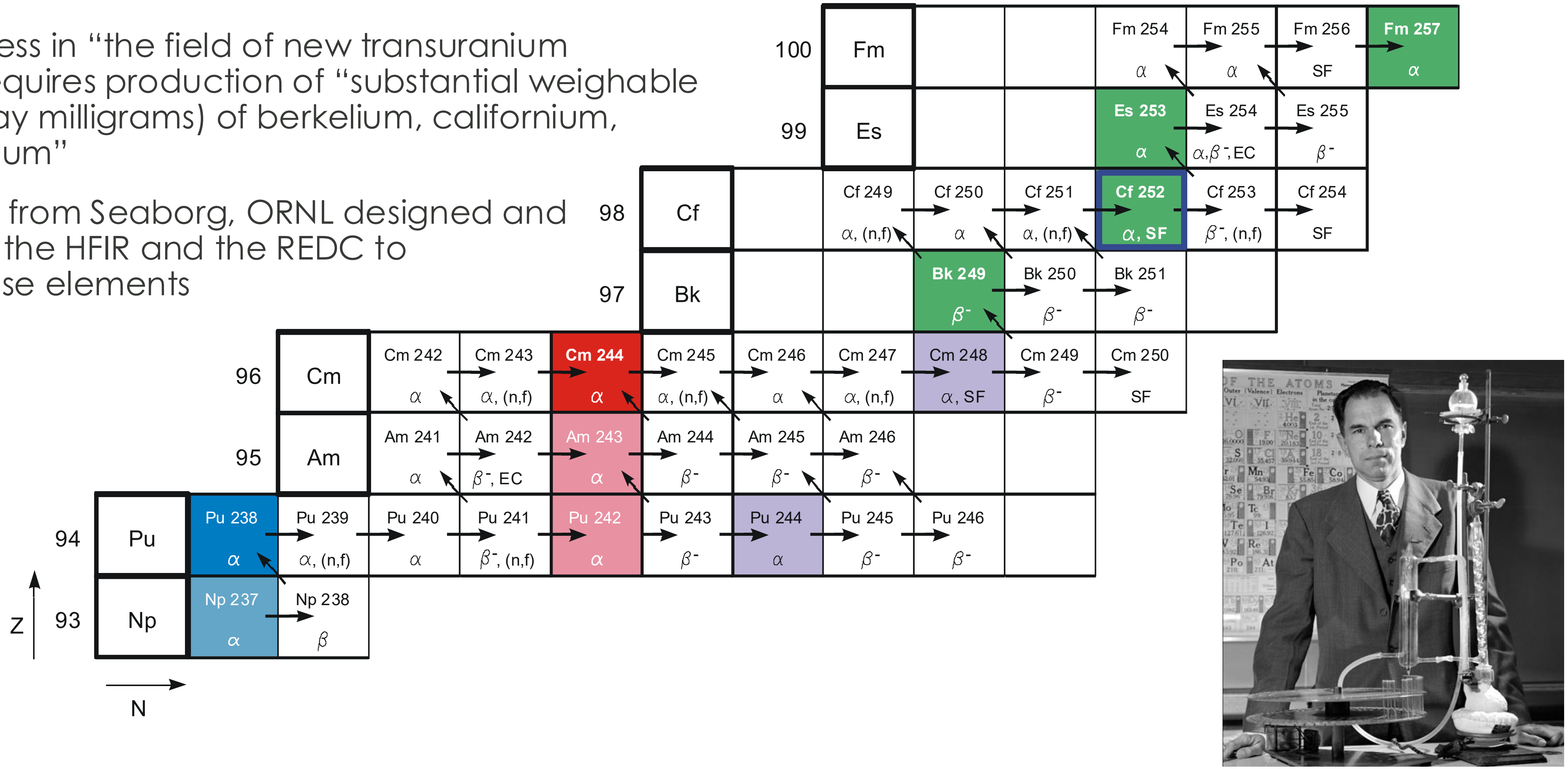
This triggered the potential for high-flux neutron reactors.

https://www.osti.gov/opennet/manhattan-project-history/Science/BombDesign/hydrogen-bomb.html

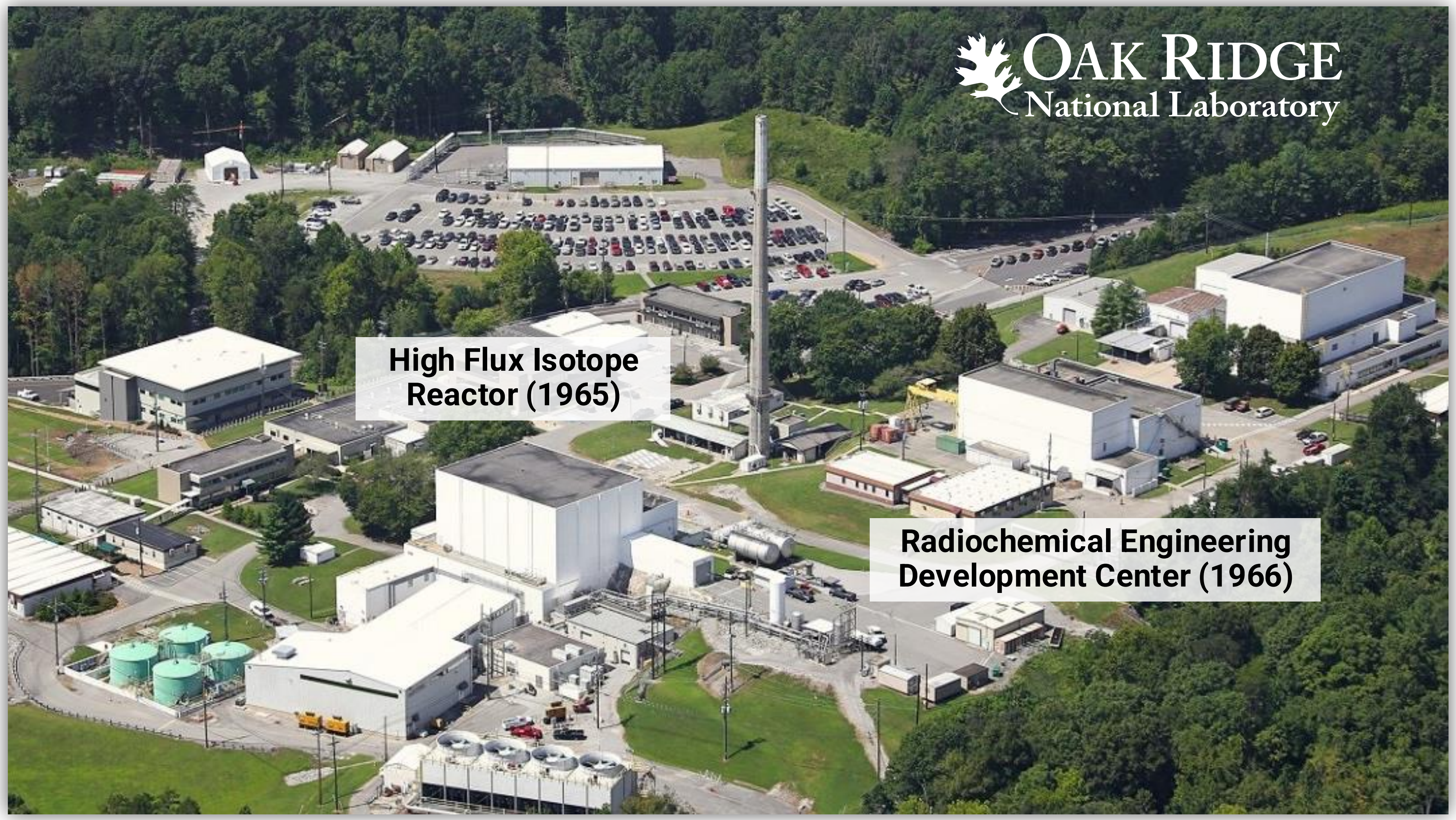
REALIZING SEABORG'S VISION: A 'VERY HIGH FLUX REACTOR' FOR HEAVY ELEMENT PRODUCTION PROGRAM AT ORNL

Letter to AEC Chairman Lewis Strauss, October 24, 1957

- Future progress in "the field of new transuranium elements" requires production of "substantial weighable quantities (say milligrams) of berkelium, californium, and einsteinium"
- With support from Seaborg, ORNL designed and constructed the HFIR and the REDC to produce these elements

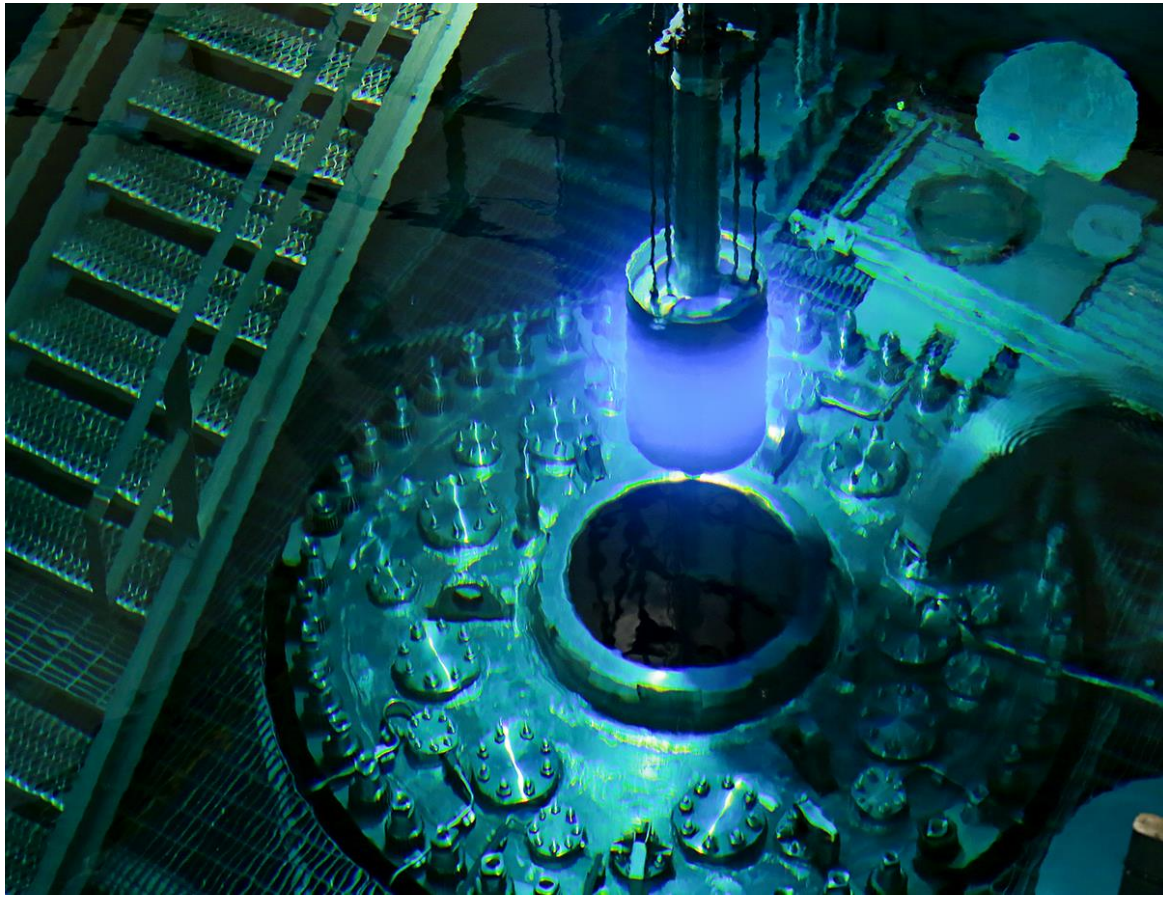


HFIR AND REDC



HFIR: Unique Capabilities

- Highest steady state thermal flux
  - 2.1×10^{15} n/cm^2-s thermal
  - 1×10^{14} n/cm^2-s epithermal (< 1 MeV)
  - 4.7×10^{14} n/cm^2-s fast (> 1 MeV)
- Brightest cold neutron source in world
- Delivers 6–7 irradiation cycles per year
- Missions:
  - Neutron scattering research
  - **Isotope production**
  - Materials irradiation
  - Neutron activation analyses



HFIR operations are supported by the Office of Basic Energy Sciences in DOE's Office of Science

RADIOISOTOPES PRODUCED/PROCESSED AT ORNL

Actinides

- ^{237}Np
- ^{257}Fm
- $^{242, 244,245, 246,248}$Cm
- ^{244,242,240,}Pu
- $^{252,251, 250, 249}$Cf
- ^{254,253}Es
- ^{227,229}Th
- ^{249}Bk
- ^{234,233}U
- ^{243,241}Am
- ^{225,227}Ac

ORNL isotope research and production is part of DOE's Isotope R&D and Production Program

Non-Actinides

- ^{63}Ni
- ^{93}Zr
- ^{209}Po
- ^{225,223}Ra
- ^{75}Se
- ^{188}W
- ^{147}Pm
- ^{133}Ba
- ^{99}Tc
- ^{212}Pb
- ^{89}Sr
- ^{166m}Ho
- ^{117m}Sn

CF-252 IS THE SECOND LARGEST RADIOISOTOPE PRODUCED AT ORNL

98

Cf

Californium-252

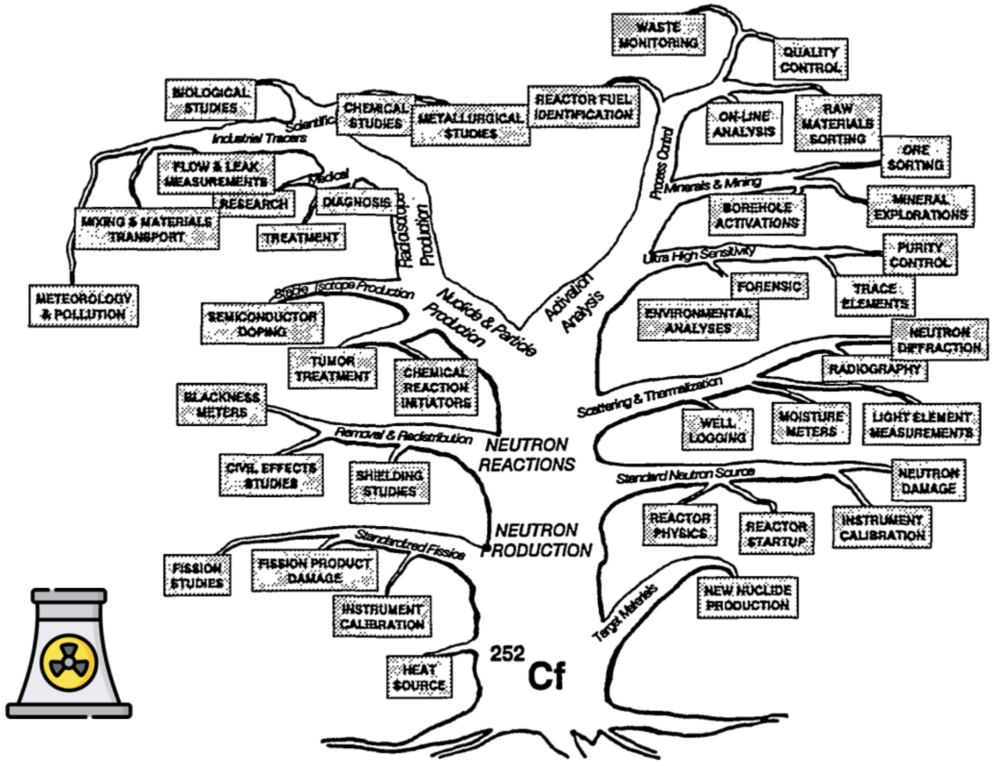
$t_{1/2}$ = 2.65 years

Neutron source

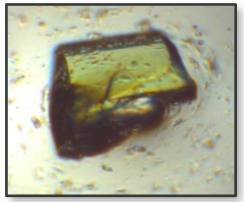
2.3 × 10^6 neutron/s per 1 µg

APPLICATIONS

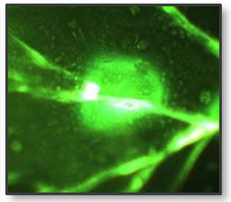
- Oil exploration
- Nuclear Reactors
- Coal analysis
- Detection Devices
- Medical Research



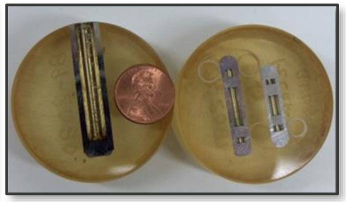
ACTINIDE PRODUCTION PROGRAMS



Crystal of californium dipicolinate



Emission under 420 nm light



Californium source

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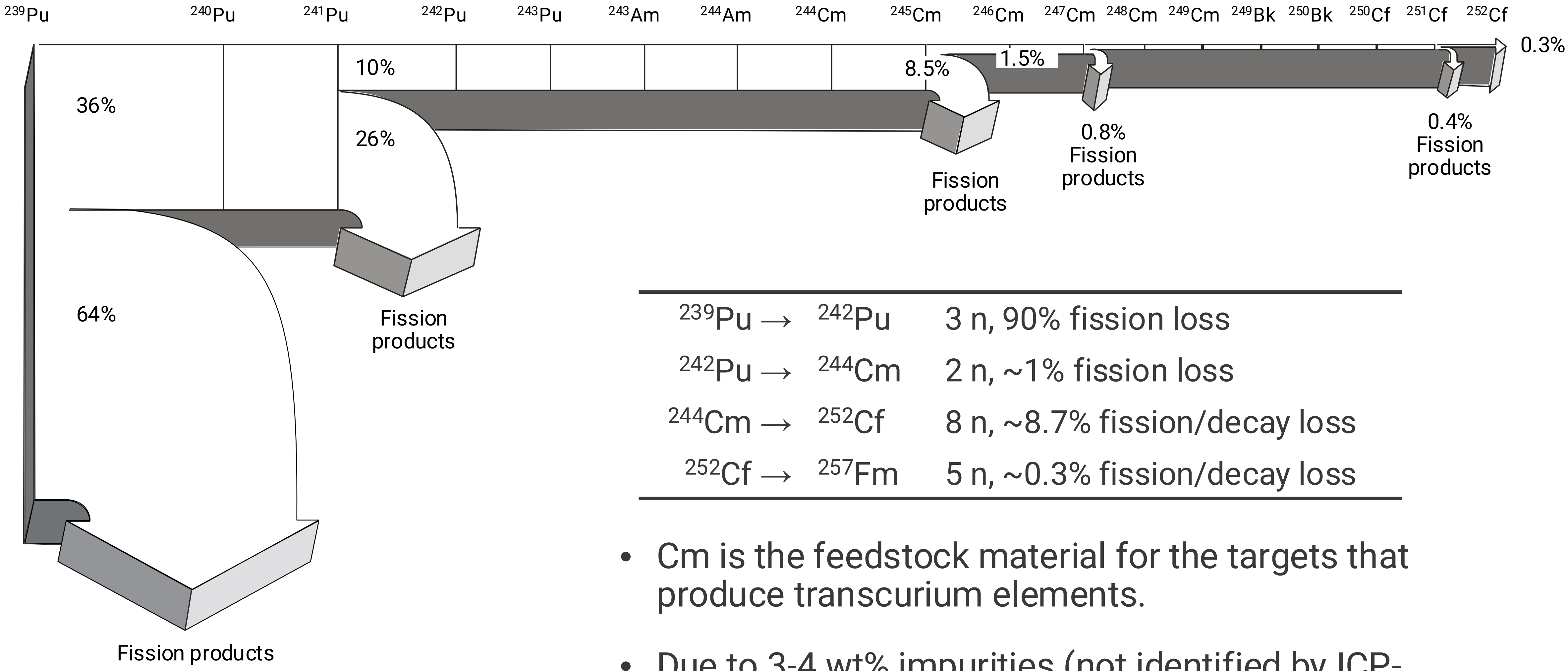
THE IMPORTANCE OF FEEDSTOCK



$^{239}Pu \rightarrow$	^{242}Pu	3 n, 90% fission loss
$^{242}Pu \rightarrow$	^{244}Cm	2 n, ~1% fission loss
$^{244}Cm \rightarrow$	^{252}Cf	8 n, ~8.7% fission/decay loss
$^{252}Cf \rightarrow$	^{257}Fm	5 n, ~0.3% fission/decay loss

- Cm is the feedstock material for the targets that produce transcurium elements.
- Due to 3-4 wt% impurities (not identified by ICP-MS) large conservatism is built in for safety.

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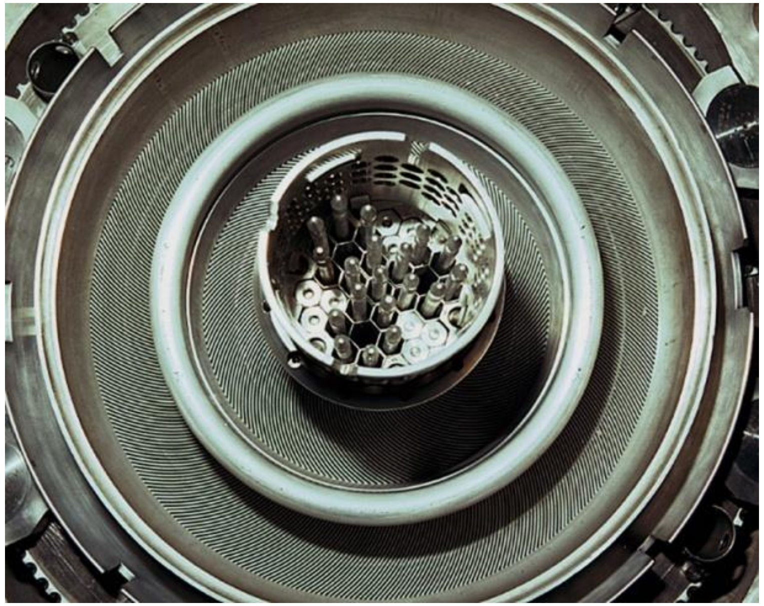
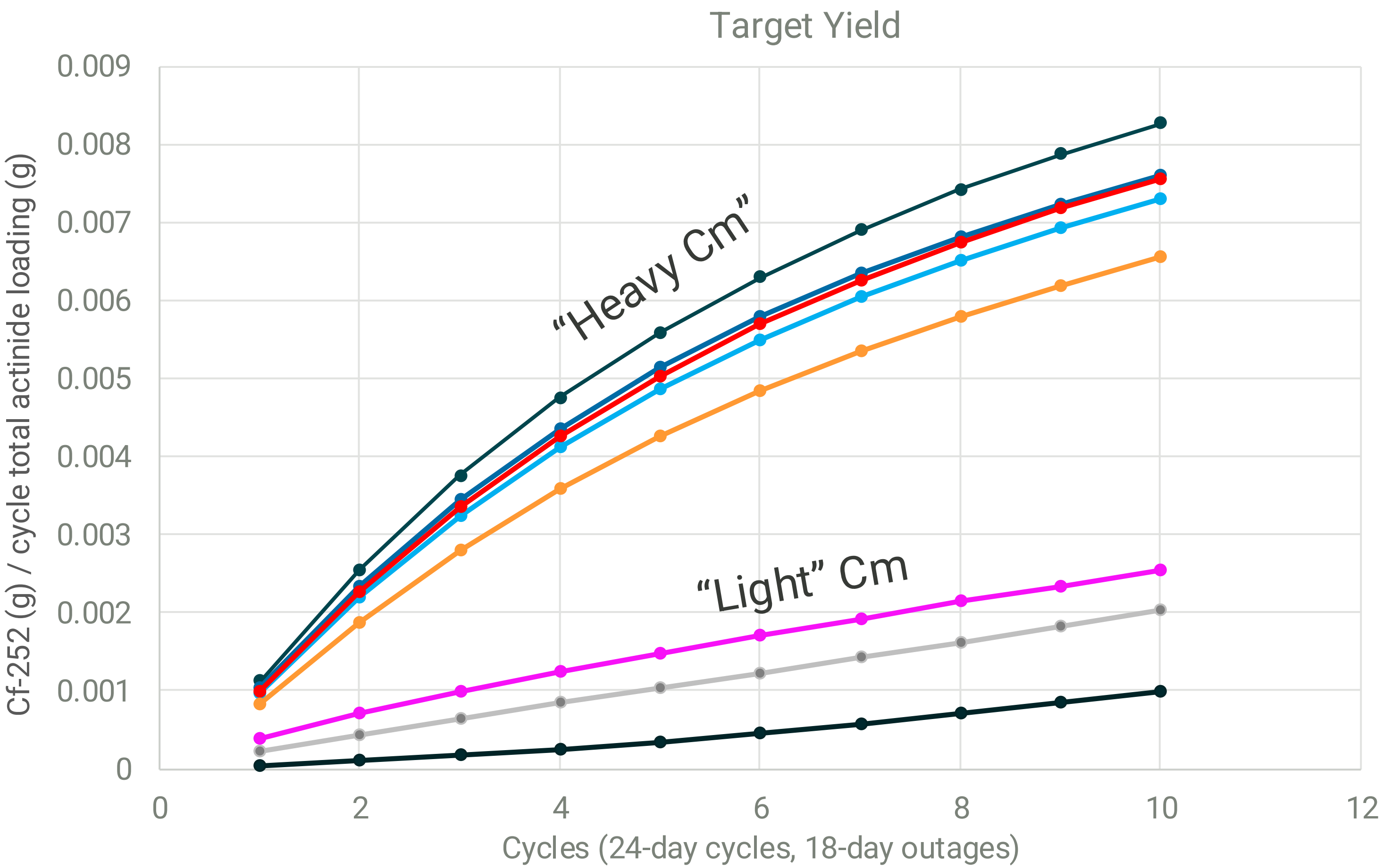
High Flux Isotope Reactor irradiation

- Typically, targets are irradiated for 4-6 cycles.
- 90 days for target cooling to allow decay of ^{131}I





https://neutrons.ornl.gov/hfir/

Heavy Curium Targets are Irradiated

96
Cm
Curium
(247)

98
Cf
Californium-252
$t_{1/2}$ = 2.65 years
Neutron source

HEAVY ACTINIDES

^{249}Bk

97
Bk
Berkelium
(247)

$^{253|254}$Es

99
Es
Einsteinium
(252)

^{257}Fm

100
Fm
Fermium
(257)

milligrams **micrograms** **picograms**

Some Major Scientific Impacts of the Heavy Element Products

Discovery of new heavier elements and isotopes using heavy element radionuclides as accelerator/ cyclotron targets

Basic research on the physics of heavy elements

- Electron behavior in orbitals
- Nuclear properties
- Nuclear reactions

Discovery of bimodal fission in some nuclides with $Z \geq 100$

Basic research on the chemistry of heavy actinides

- Chemically stable compounds
- Structure and bonding
- Speciation

^{253}Es (0.17 mg, self-illuminated)

The heavy element program was endorsed twice by the National Research Council

100+ publications resulted each year from research using heavy element products

E.K. Hulet, "Bimodal Symmetric Fission Observed in the Heaviest Elements," *Phys. Rev. Lett.* 56, 313–316 (1986)

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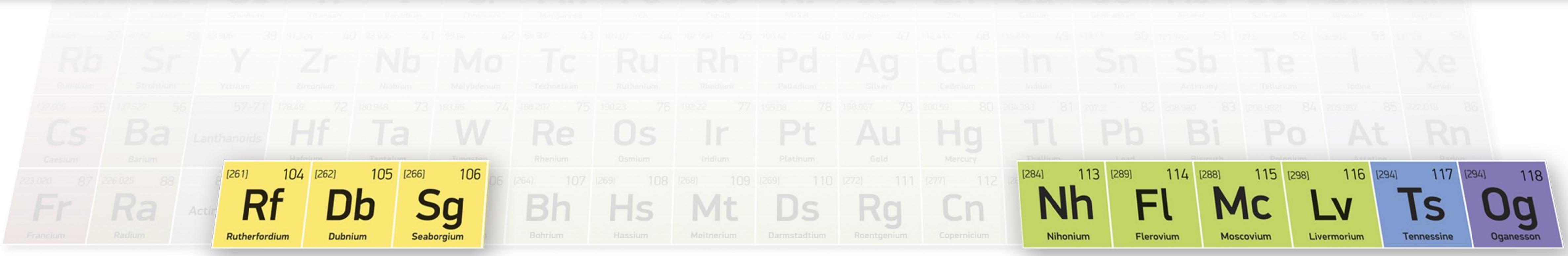
SUPER-HEAVY ELEMENT DISCOVERIES ENABLED BY ORNL-PRODUCED MATERIALS AND ISOTOPES

Element	104 Rutherfordium	105 Dubnium	106 Seaborgium	113 Nihonium	114 Flerovium	115 Moscovium	116 Livermorium	117 Tennessine	118 Oganesson
Year produced	1964	1970	1974	2004	2000	2004	2005	2010	2006
Target	^{242}Pu, ^{249}Cf	^{249}Bk, $^{249,250}Cf$	^{249}Cf	^{243}Am (decay from 115)	^{244}Pu	^{243}Am	$^{245,248}Cm$	^{249}Bk	^{249}Cf



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CHEMICAL PROCESSING AFTER IRRADIATION

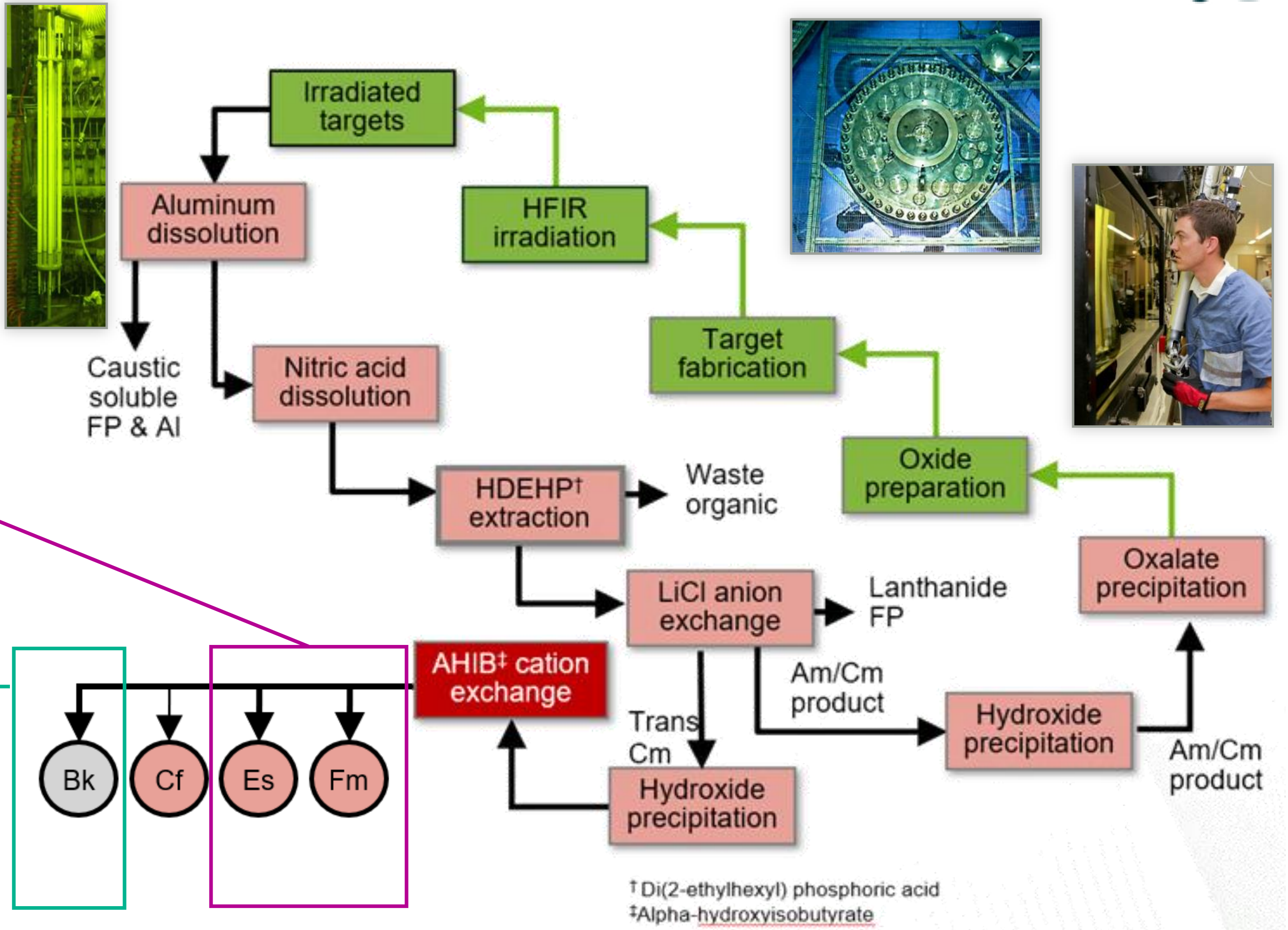
GLOVE BOXES

^{249}Bk | $^{253/254}$Es | ^{257}Fm

FINISHING in the glove boxes

BERKEX

HOT CELLS – 7920



Robinson, et al. Radiochimica Acta 108.9 (2020): 737-746.

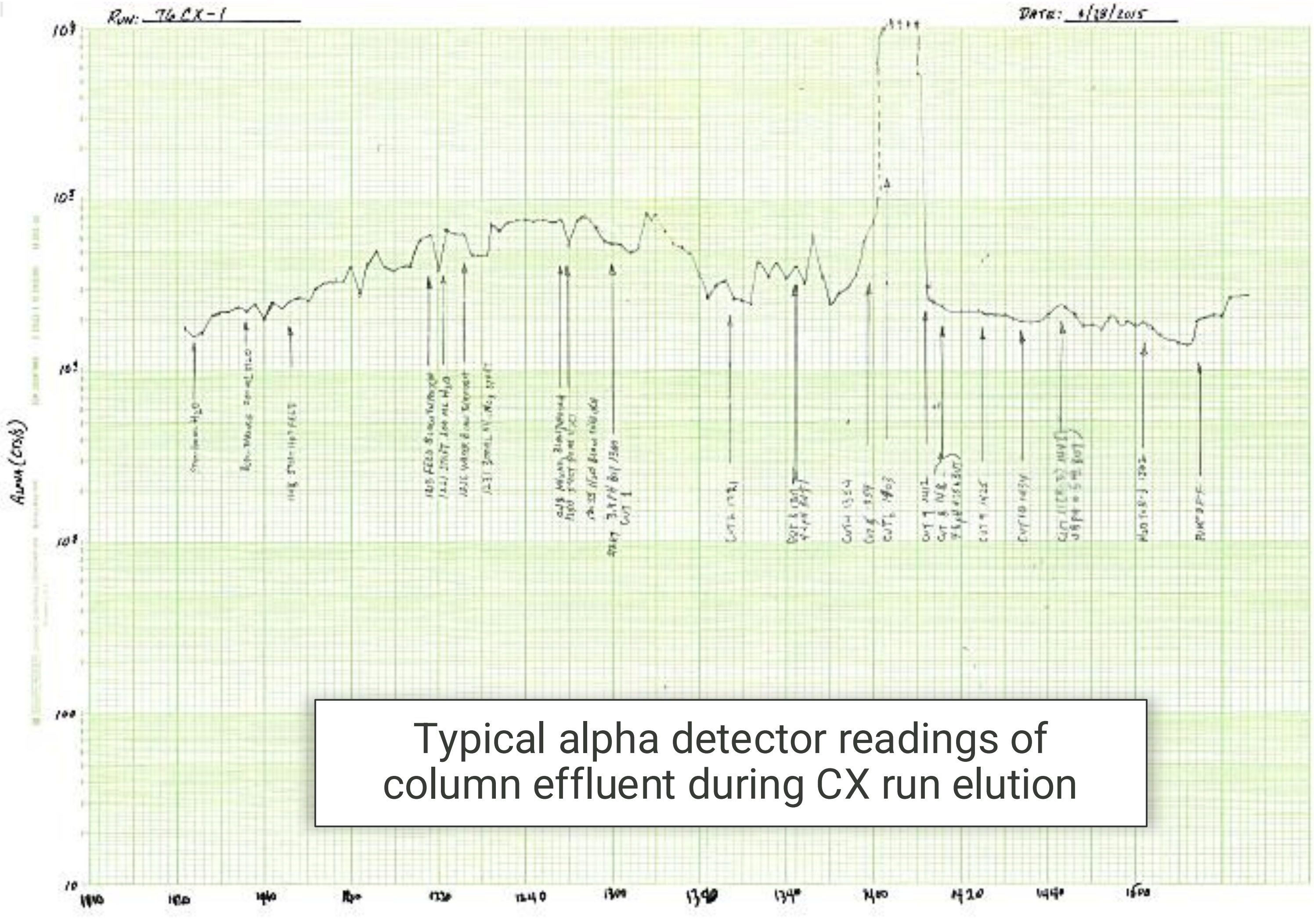
TRANSCURIUM ELEMENT SEPARATIONS VIA CATION EXCHANGE ELUTION USING AHIB

- Conversion from chloride form to nitrate by hydroxide precipitation, filtration, and re-dissolution in HNO3

- Elemental separation by ammonium alpha-hydroxyisobutyrate (AHIB)at varying values of pH

- Column is heated ~70°C

- Element (band) separation followed with neutron and alpha probes



Typical alpha detector readings of column effluent during CX run elution

C81 CAMPAIGN

97
Bk
Berkelium-249 Processing
$t_{1/2}$ = 327 days
β decay → ^{249}Cf

IMPURITIES

Ce-141	Cf-252
Ce-144	Cf-249

ADVANTAGES

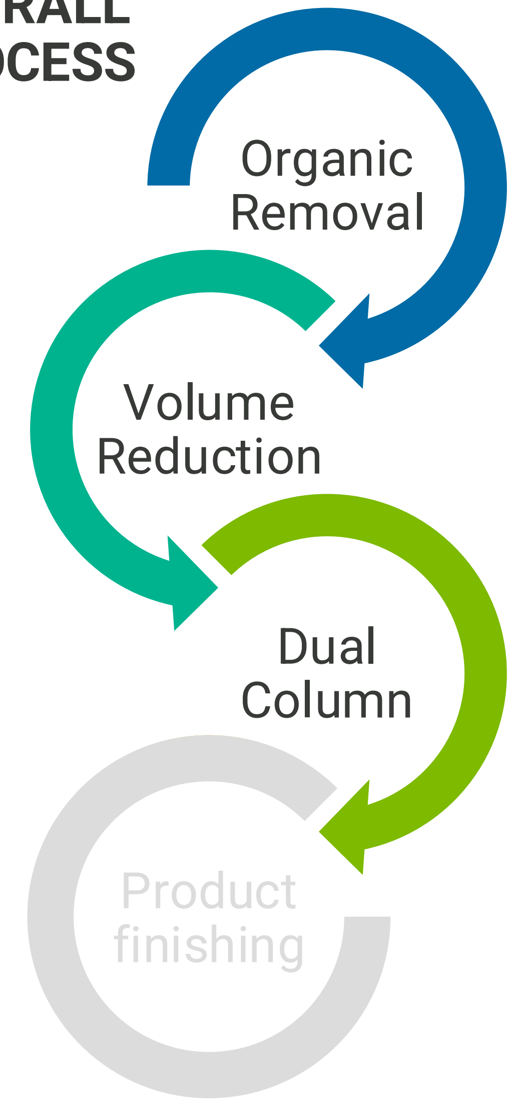
- **Bk^{4+} is accessible** compared to heavier An
- **Cleaner crude product** from Hot Cell bank
- Ce^{3+} and Ce^{4+} **ionic sizes differ** from Bk

Nathan Sims
Technician
Isotope Processing and Manufacturing Division

BERKERLIUM-249 PURIFICATION

97
Bk
Berkelium
(247)

OVERALL PROCESS



Dual column: Anion exchanger + LN column

Species after oxidation

$[Ce^{IV}(NO_3)_n]^{(n-4)-}$	Anionic complex
$[Bk^{IV}(NO_3)_3]^{+}$	Cationic complex
$[Cf^{III}(NO_3)_3]$	Neutral complex

Main impurities
- Ce-141/144
- Cf-252
- Cf-249

Oxidation

Loading

Washing

Fully loaded

LN Anion exchange

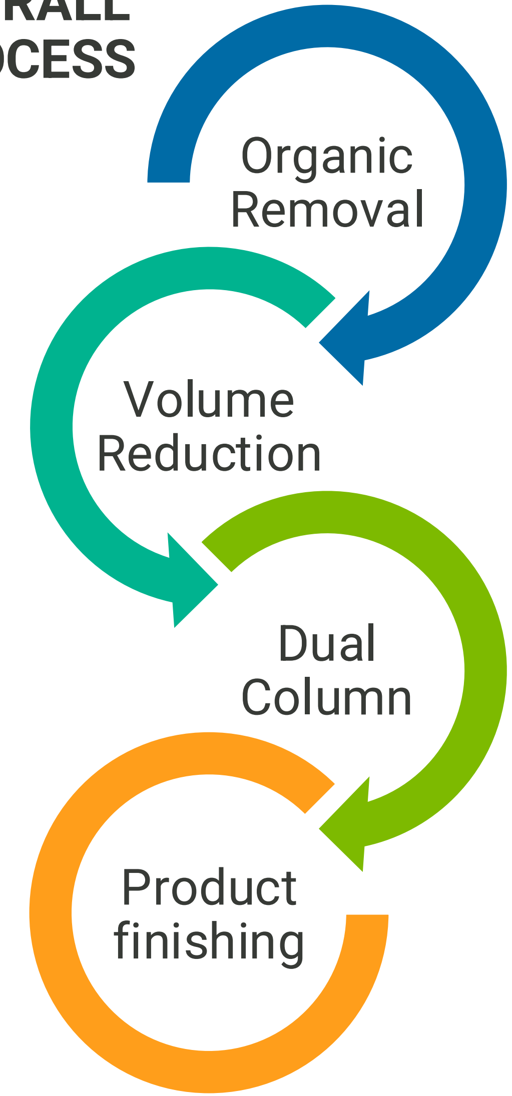
STRIPPING

LN

MP-1

BERKERLIUM-249 PURIFICATION

OVERALL PROCESS



Product finishing

The Bk-249 product is supposed to be ready but…

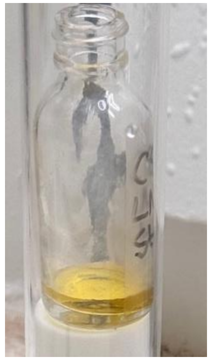
HDEHP residue?

Instead…
Peroxide + HNO_3 treatment

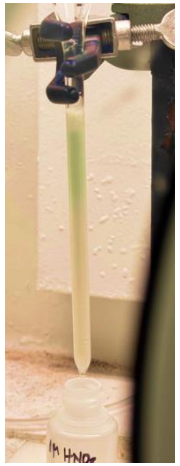
wait a minute...

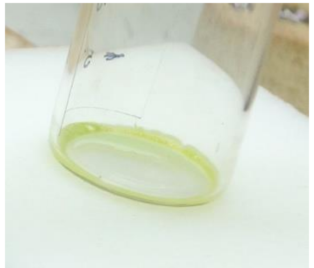
Bk is supposed to be GREEN?

- ✅ Radiochemical purity
- ❌ Mass spec revealed traces of **Fe** and **Cr**



DGA column for transition metal rejection





- ✅ **Radiochemical purity**
- ✅ **Chemical purity**

C81 CAMPAIGN

99
Es

Einsteinium-253/254 Processing

$t_{1/2}$: 21 days (^{253}Es); 276 days ^{254}Es
α decay → ^{249}Bk ; ^{250}Bk

CHALLENGES

- Cf-252 main impurity – **high dose!!!**
- **Poor Cf/Es** separation
- **Underdeveloped chemistry**

Ashley Harvey
Technician
Radioisotope Science and Technology Division

IMPURITIES

Cf-252	Pu-238
Ce-144	Eu-154

100
Fm

Fermium-257 Processing

$t_{1/2}$ = 100 days
α decay → ^{253}Cf

CHALLENGES

- **Picogram scale**
- **Short-lived surrogate,** Fm-255
- **Little is known** about Fm chemistry

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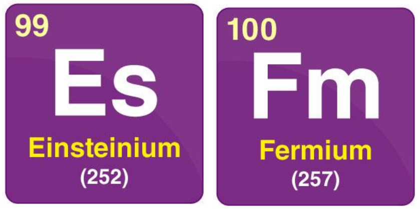
EINSTEINIUM/FERMIUM PURIFICATION



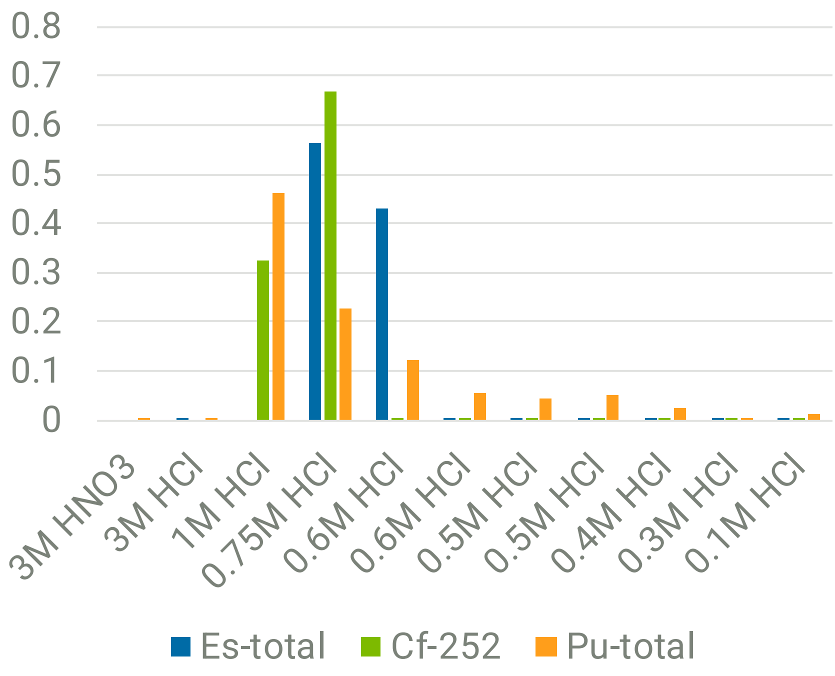
Previous campaigns relied on **AHIB cation exchange** runs as a continuation of the run in the hot cells:

Challenges: slight changes in temperature and pH affect tremendously performance. Product not chemically clean enough.

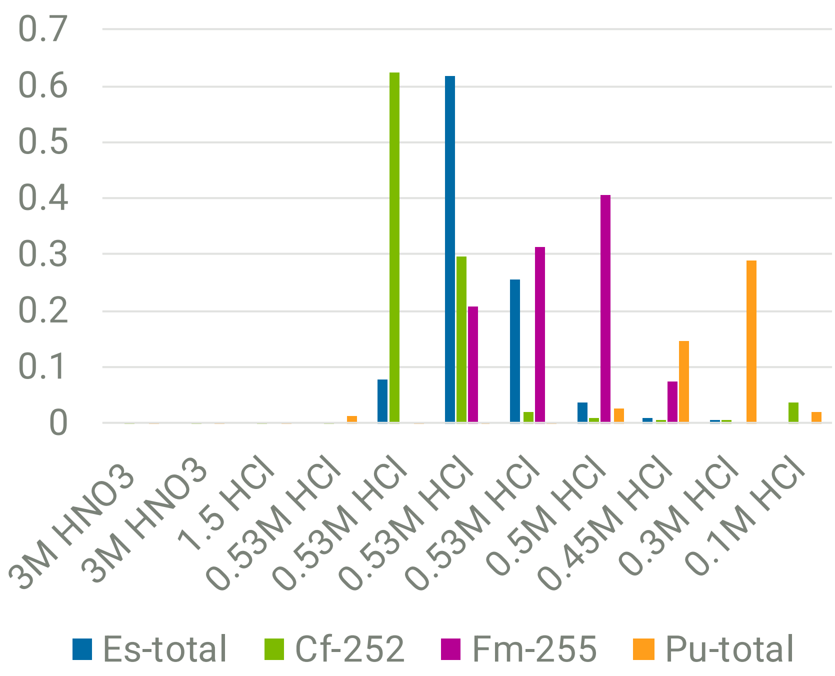
C81 – Turning to DGA and LN resins

Cf-252 debulk

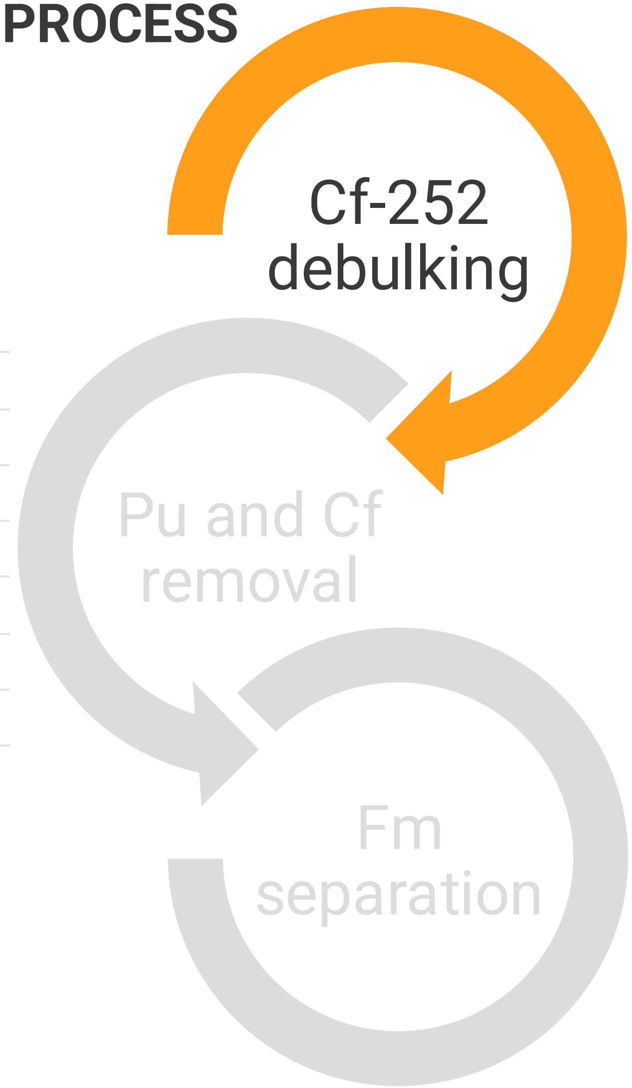
DGA – Hot cells



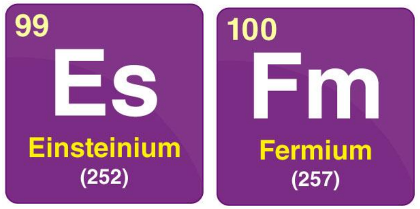
DGA – Glove box



OVERALL PROCESS



EINSTEINIUM/FERMIUM PURIFICATION

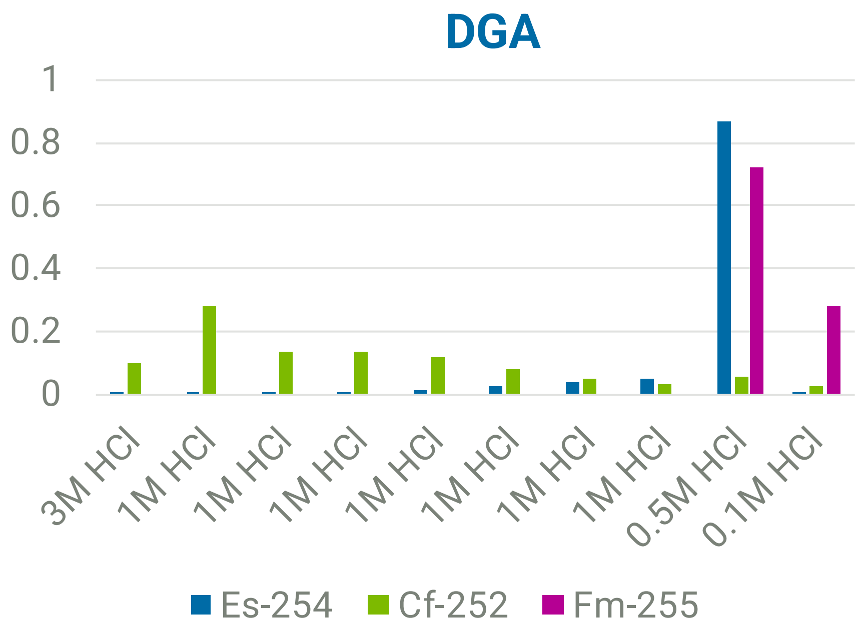


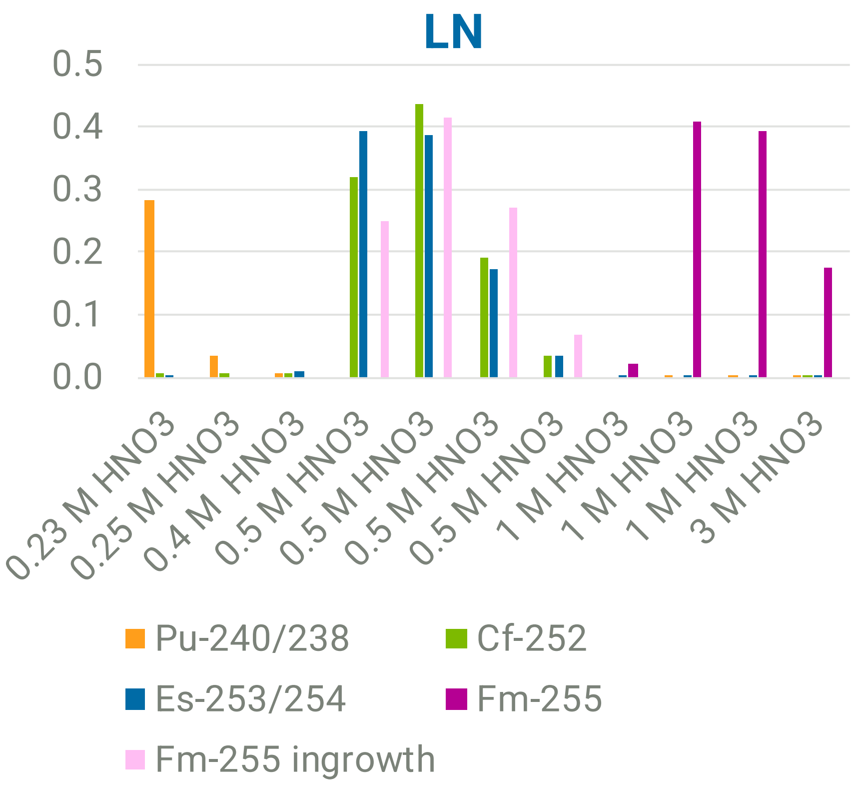
Pu and Cf removal

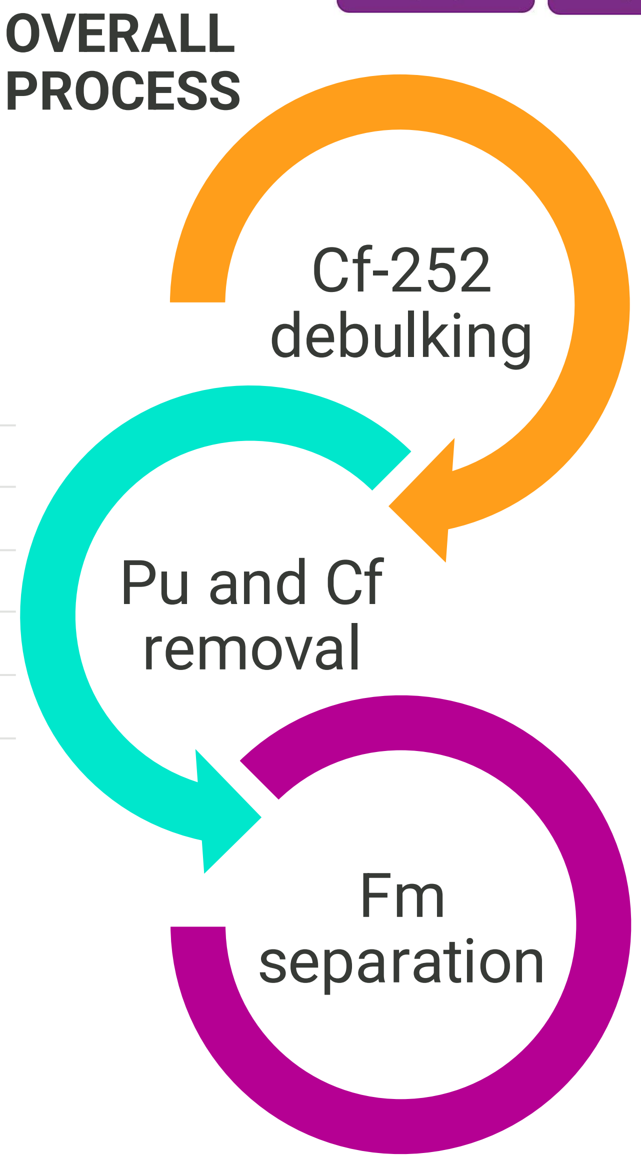
After **optimizing DGA conditions**, Cf/Es separation is improved under 1M HCl.

As expected, **LN** does not separate Cf/Es but does a good job removing Pu and Fm out.

Only 30% Pu is recovered suggesting the elution of PuO_2^{2+} only.







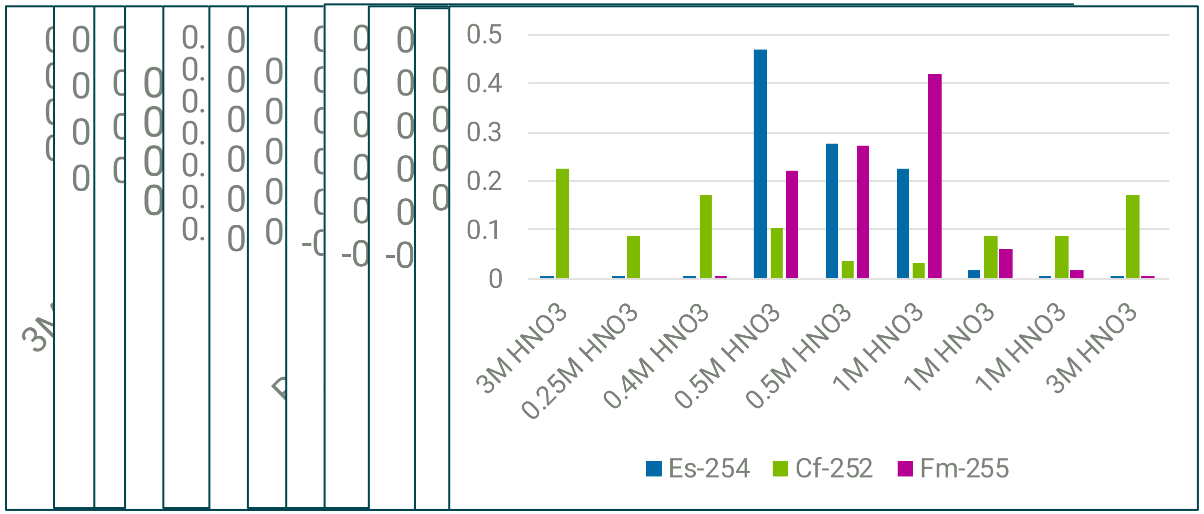
EINSTEINIUM/FERMIUM PURIFICATION

99 Es Einsteinium (252)

100 Fm Fermium (257)

Product

Significant improvements were made along the way… and a great effort made.



C79 product

C80 product

Final product C81

99.93% chemical purity

Seeing **NOTHING is good** in the Es campaign!

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Outlook

Significant progress has been made to improve the processing of transcurium elements

Prepping analytical methods for the quantification of Fm-257 (for the first time decades at ORNL)

R&D on heavy actinide continues in preparation for C82 for optimized removal of Cf-252

Potential increase of material for C82 due to heavier Cm targets

Acknowledgements

The heavy element team

Radiochemical lab technicians

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Sam Schrell
Cf-252 program manager

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Program management:

- Roy Copping (Radioisotope portfolio manager)
- Susan Hogle (Division Director)

Processing support:

- Sara Gilson (Staff Scientist)
- Karlee Eardley (Technical Professional)

Radiological Control Technicians (RCTs)

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