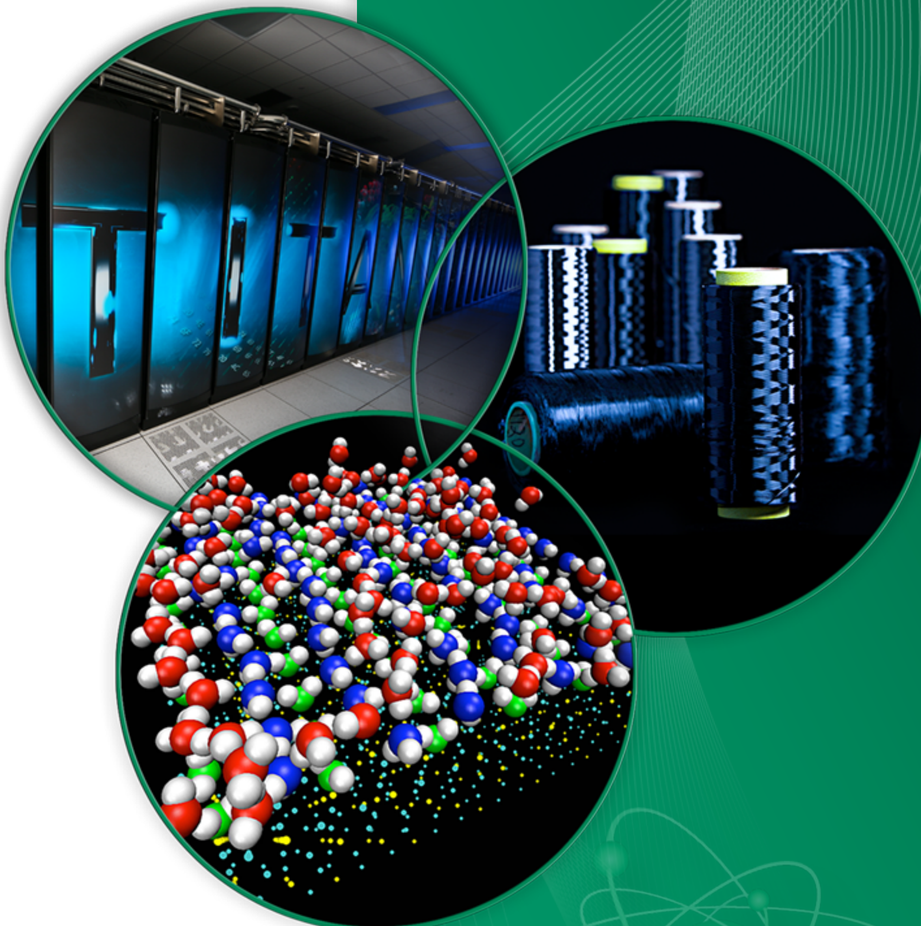
Actinide Targets for Super-Heavy Element Research

Presented at

International Workshop on Super-Heavy Element Research SHE-15



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Outline

- Actinides and heavy element discovery
- Production and availability of actinides
  - Reactor production
  - Chemical processing
- Actinide materials for SHE research
  - Current inventories
  - Berkelium
  - Californium
  - Einsteinium
  - Novel materials
- Actinide targets
  - Fabrication
  - Projected target needs

Actinides

- Radioactive elements with Z=89-103
  - Identified as a new row in the Periodic Table in the 1940s (Seaborg)
  - Only two actinides occur naturally in substantial quantities (Th and U)
- All actinides heavier than uranium discovered using actinide targets
  - Np, Pu, Am, Cm, Bk, and Cf in the 1940s (Berkeley)
  - Es and Fm from neutron irradiation of uranium in a thermonuclear explosion (1952)
  - Md, No, and Lr in the 50s and 60s (Berkeley and Dubna)
- Production in high-flux reactors required for more than trace amounts of actinides heavier than uranium
  - Only Np, Pu, Am, Cm, Bk, and Cf available in quantities needed for SHE targets
- 9 of 15 super-heavy elements (Z=104-106, 113-118) synthesized using actinide targets
  - Most target materials from ORNL and RIAR

"Hot fusion" using ^{48}Ca beams on actinide targets has significantly expanded SHE research

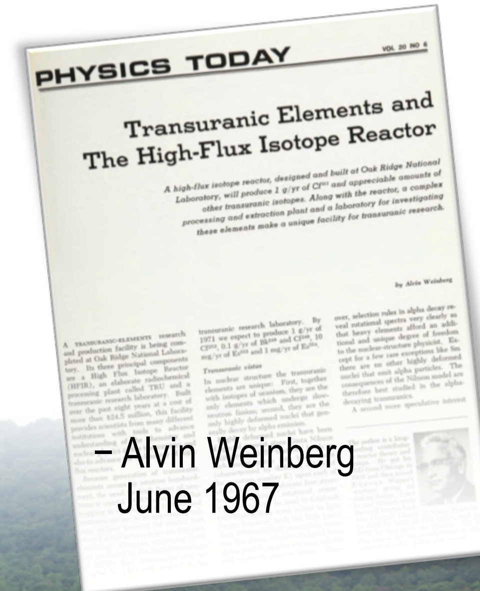
Six new elements (Z=113-118) synthesized using ^{48}Ca beams on actinide targets since 2000

Element (Z)	Year first produced	Target	Beam	Nuclei produced to date (directly/ total including decay chains)
113	2004	^{243}Am (decay from 115)	^{48}Ca	2/89 (directly in 2007 from ^{237}Np)
Flerovium (114)	2000	^{244}Pu	^{48}Ca	64/99
115	2004	^{243}Am	^{48}Ca	63/87
Livermorium (116)	2005	$^{245,248}Cm$	^{48}Ca	31/35
117	2010	^{249}Bk	^{48}Ca	22
118	2006	^{249}Cf	^{48}Ca	4

- More than 50 new isotopes
- A total of more than 300 nuclei (Z=113-118)
- "Hot fusion" has increased SHE production rates for Z ≥ 113 by one or more orders of magnitude

HFIR/REDC: ORNL's actinide production and research complex

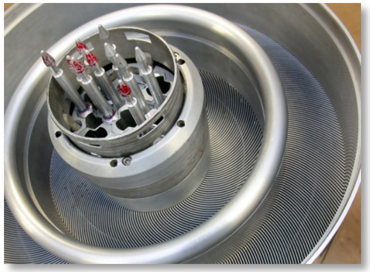
High Flux Isotope Reactor (HFIR)	Radiochemical Engineering Development Center (REDC)
• High-power (85-MW) compact core provides intense thermal neutron fluxes in a central flux trap • Isotope production (gram quantities of Cf-252), neutron scattering, and materials irradiation	• Heavily shielded hot cells for target preparation and processing irradiated materials from HFIR


PHYSICS TODAY
VOL 20 NO 6
Transuranic Elements and The High-Flux Isotope Reactor
A high-flux isotope reactor, designed and built at Oak Ridge National Laboratory, will produce 1 g/yr of Cf^{252} and appreciable amounts of other transuranic isotopes. Along with the reactor, a complex processing and extraction plant and a laboratory for investigating these elements make a unique facility for transuranic research.
by Alvin Weinberg

– Alvin Weinberg
June 1967

Actinide production by irradiation of Am/Cm targets in HFIR

Targets specially designed for reactor conditions	Irradiation in the HFIR flux trap
• Composition controls fission and gamma heating • Targets typically remain in the reactor for 3-6 cycles (up to a year)	• Intense thermal neutron flux (2.5×10^{15} neutrons/$cm^2 \cdot s$) • 31 target positions (6-8 targets typical for Cf production) • Typical campaign: Up to 200mg ^{252}Cf, 20mg ^{249}Bk, ~2μg ^{254}Es, picograms ^{257}Fm



Target positions in the flux trap of HFIR fuel assembly

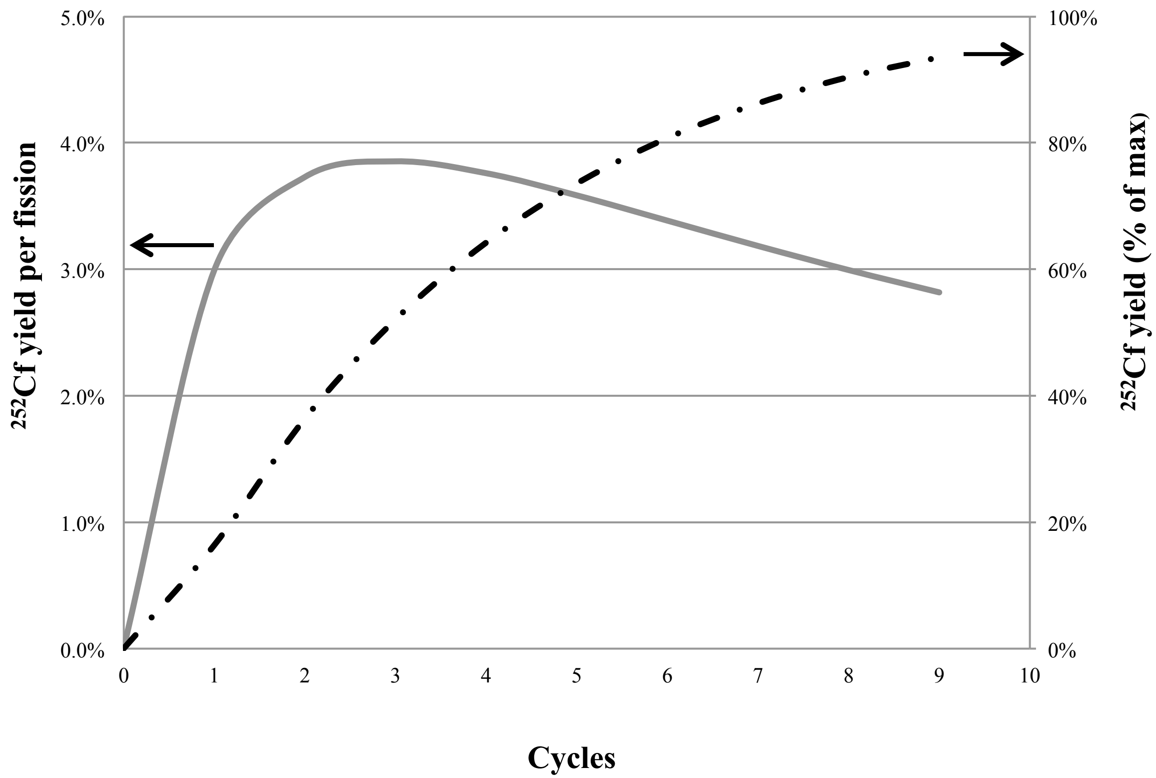
Upcoming paper on actinide production in PIK by Onegin

Inventories of selected actinides at ORNL

Isotope	Approximate amount (mg)	Isotopic %	Notes
Np-237	1000	>99%	
Pu-242	5500	>99%	
Am-241	3500	>99%	
Am-243	1000	>99%	
Cm-244	1000	>90%	
Cm-248	1700	80-95%	80 mg at >95%, remainder requires recovery processing
Bk-249	—	>99%	Requires HFIR production and chemical processing (up to 20 mg per campaign when combined with Cf-252 production)
Cf-249	170	>99%	
Cf-251	40-150	~35%	Requires recovery and processing from old Cf sources (isotopic separation required for higher enrichment)

- HFIR/REDC is a unique resource for production and chemical processing of actinides
- ORNL is the repository for actinide isotopes for DOE's Isotope Development and Production for Research and Application Program in the Office of Nuclear Physics

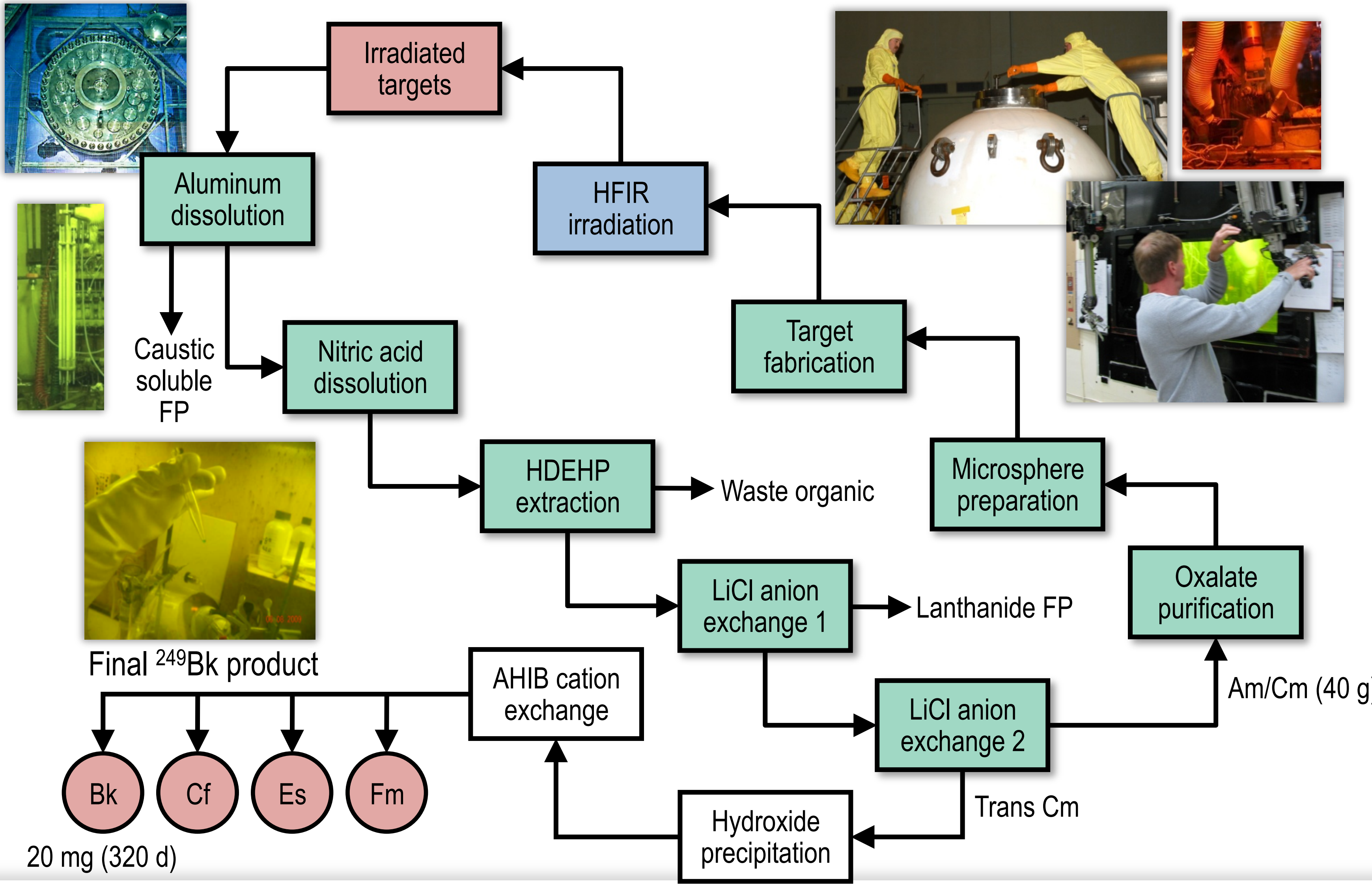
^{252}Cf yield as a function of the number of reactor cycles in HFIR



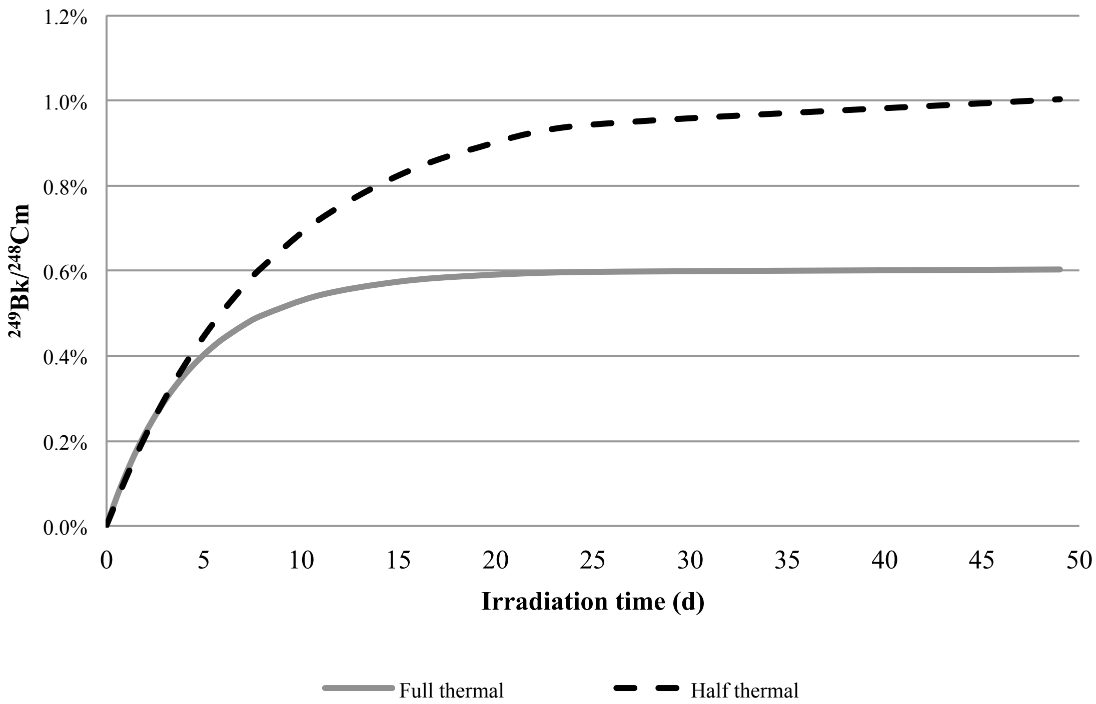
- Production efficiency highest in first 2 cycles
- To maximize overall production, a typical campaign is 3-5 reaction cycles (24 days per cycle)

Bk production/separation cycle at ORNL

24 months Cf/Bk, 8 months Bk only



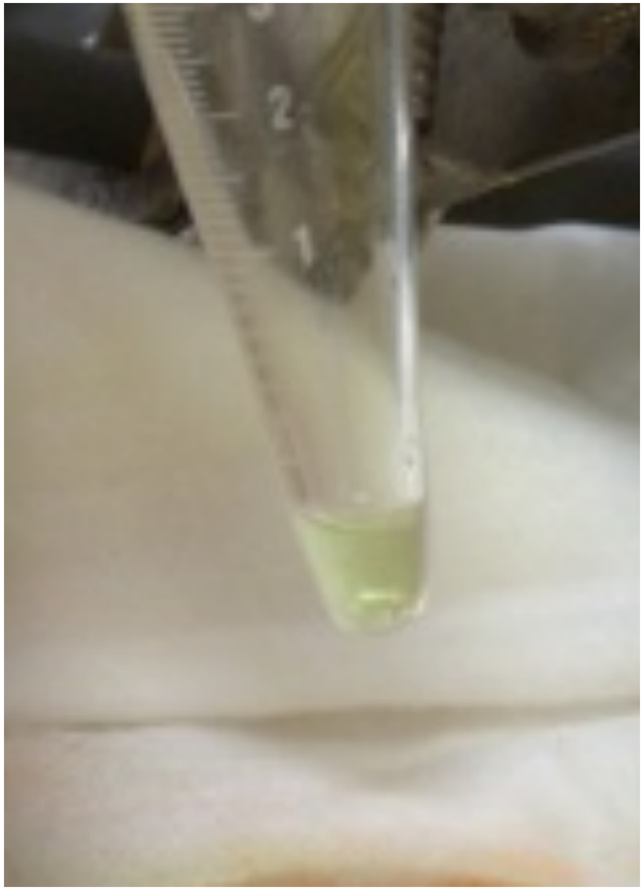
Bk production peaks after one reactor cycle (24 days)



- ^{249}Bk yield is typically 0.6% of the ^{248}Cm content of the target
- Suppressing thermal neutron flux can further increase yield

Bk-249 outlook

- Cf-252 production currently under contract is ~80 mg per campaign
  - Reduces Bk-249 yield to ~9 mg per campaign
- Increased Bk production
  - Use additional Cm-248 feedstock for one-cycle HFIR irradiations coordinated with Cf-252 production (allows leveraging of Cf processing)
  - Use thermal neutron filtering to further increase production by factors of two or more (requires development)
  - >100 mg possible, but unlikely due to resource requirements
  - 30 mg a reasonable goal assuming availability of Cm-248 feedstock



Additional details in upcoming paper by Boll

Cf isotopic distribution following production and after several decades decay

Isotope	Atomic % (as produced)	Half-life (y)
^{249}Cf	3.41	351
^{250}Cf	8.7	13.08
^{251}Cf	2.60	898
^{252}Cf	85.27	2.645
^{253}Cf	0.004	0.049
^{254}Cf	0.010	0.166

- After several decades, ^{252}Cf substantially decays
- Remaining Cf a mixture of $^{249,\ 250,\ \text{and}\ 251}Cf$, with ~35% ^{251}Cf (heaviest SHE target material)
- Mixed target must be handled in a shielded glove box due primarily to ^{250}Cf content
- Possibility of producing two new heaviest isotopes of element 118

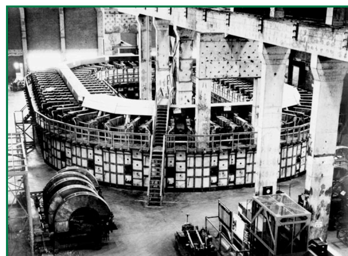
Cf recovered from decayed sources				
Cf-249 mg	Cf-250 mg	Cf-251 mg	Cf-252 mg	Total Cf mg
7.6	2.5	5.7	0.007	15.8
48.1%	15.6%	36.3%	0.04%	

Reestablishing isotope enrichment capability at ORNL

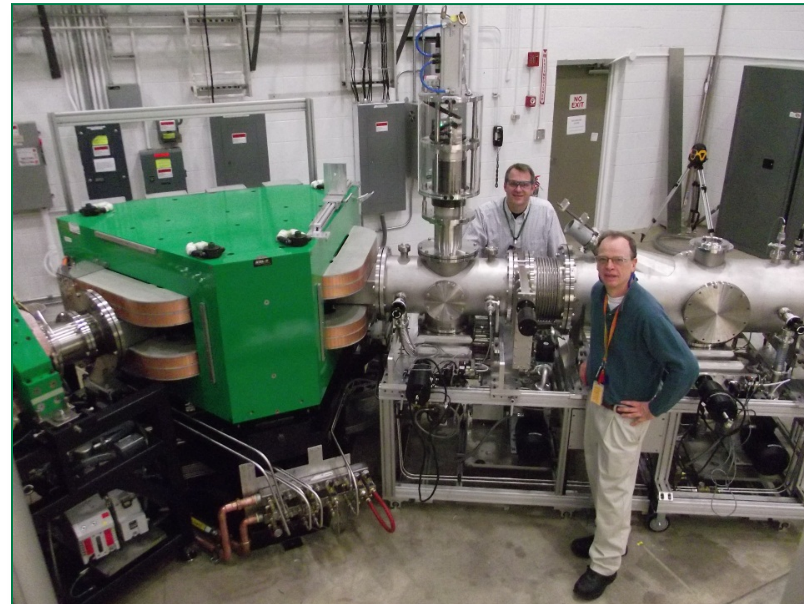
Electromagnetic Isotope Separator (EMIS)

- Designed and built by ORNL
- Sponsored by DOE Office of Nuclear Physics

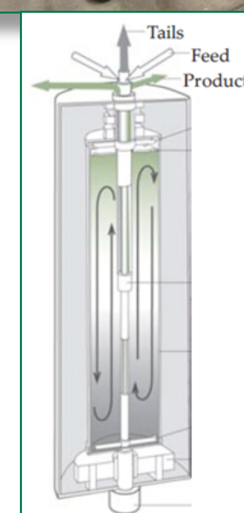
Separated beams of molybdenum isotopes by new EMIS



1943-1995

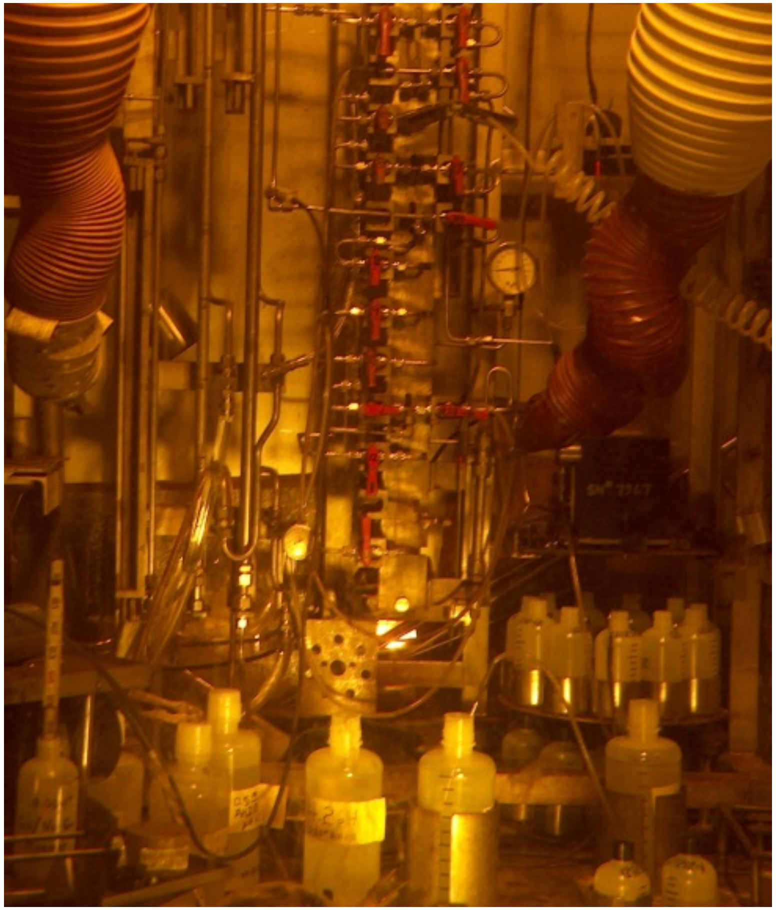


- Currently capable of producing mg to g quantities of >98% enriched stable isotopes
- Combined with gaseous centrifuge to increase throughput
- Proposed radioactive EMIS for actinides



Revisiting the Large Einsteinium Activation Program (LEAP)

- A special HFIR campaign proposed in the 1980s to maximize production of ^{254}Es
- Beginning with ~1g of ^{252}Cf, LEAP estimated to yield 40 µg of ^{254}Es
  - 300 times smaller than the typical amount of target material for SHE experiments
- Can neutron spectrum filtering increase this yield?
- ^{254}Es attractive (if available) as path to element 119 (using Ca beams) and possibly 121 (Ti beams)
  - SHE Factory-scale approach essential for consideration of using ^{254}Es targets
  - Significant target heating and damage considerations



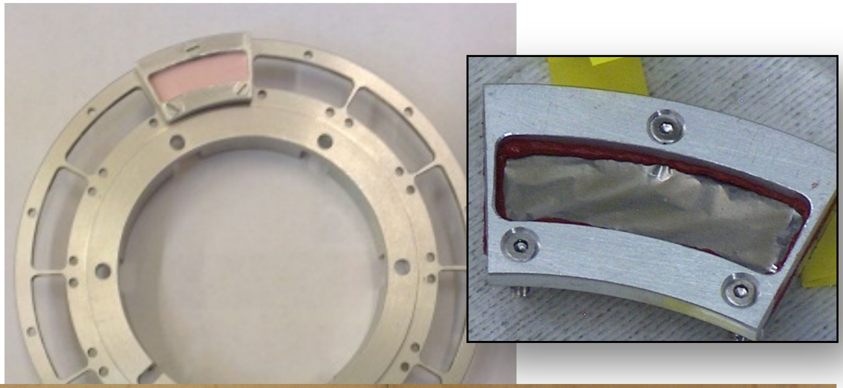
Novel materials for research on extreme nuclei

- ^{60}Fe
  - Neutron-rich long-lived element
  - Can be produced in HFIR (~1mg per 2.5g of 65% ^{58}Fe in 4 reactor cycles)
  - Requires isotopic separation and low-level radioactive handling
  - Interesting for supernova research and fusion reactions near shell closures, could in principle get us one neutron closer to the island

- ^{10}Be
  - Be reflector material in HFIR enriched in ^{10}Be from neutron irradiation (~1 mg per 10g reflector Be after 20 cycles)
  - Requires isotopic separation and Be handling
  - Interesting for neutron capture cross sections of astrophysical importance, studies of neutron-rich light elements, and producing new heavy actinide isotopes

Actinide target fabrication

- Ammonium diurante was the first actinide target for nuclear studies
  - Used by Meitner, Hahn, and Frisch to investigate irradiation of uranium with neutrons
- Target fabrication techniques
  - Molecular plating
  - Polymer-assisted deposition
  - Other techniques include sputtering, painting, electrophoresis, electrospray, and intermetallics
- Fabrication requirements
  - Deposition yields near 100%
  - Non-interfering substrates (low Z, typically titanium)
  - Reproducibility for multi-sector targets
  - Durability
  - Remote handling capability for radioactive materials



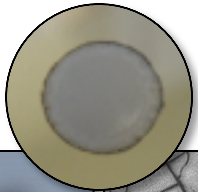
Thin film target fabrication at ORNL

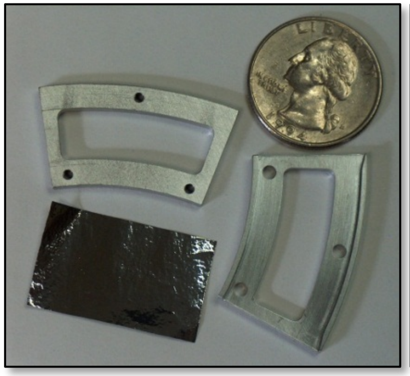
Actinide film deposition: Cf, Am, Cm, and Bk

- Metal substrates (Pt, stainless steel, Cu, Fe, Au, Gd, and Ti foils)
- Electrochemical deposition methods: isopropanol, isobutanol, and ammonium acetate
- Thin film analysis and characterization using gamma imaging and scanning electron microscopy (SEM)

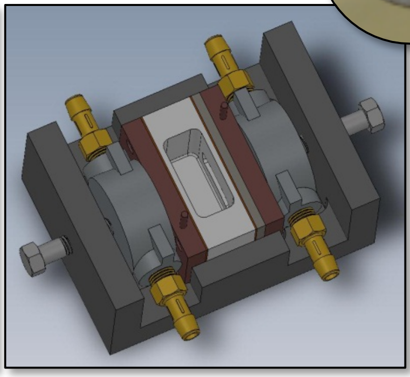
Upcoming papers on actinide targets by Eberhardt and Boll

Deposition on stainless steel

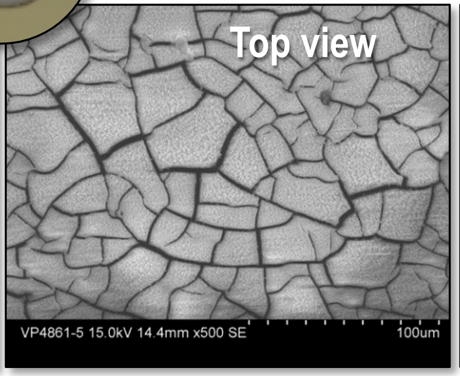


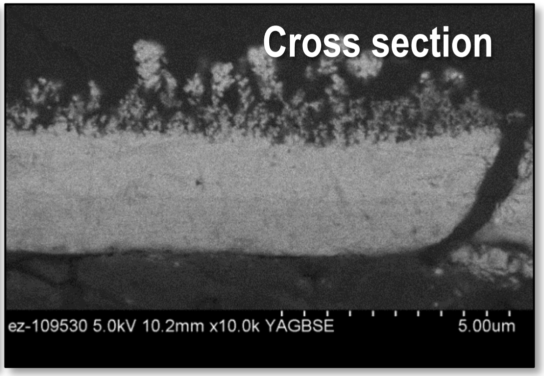


Ti foil and mounting frame



Electrodeposition unit





SEM of lanthanide deposition

Projected target needs for SHE experiments (2015-2020)

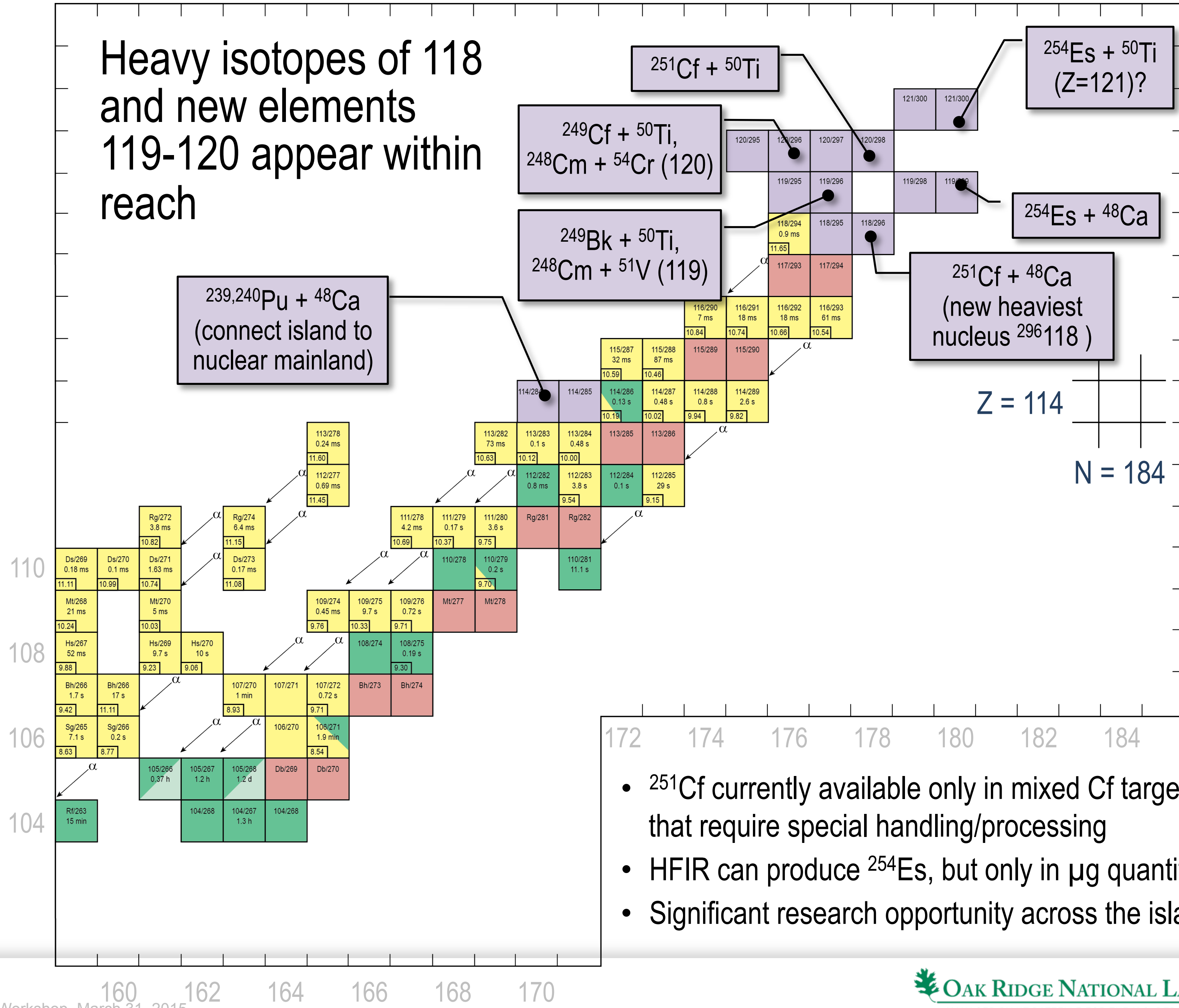
Date	Facility	Isotope	Experiment	Comments
2015	JINR	Mixed Cf	Heavy isotopes of Z=118, 296(118) would be the heaviest nucleus to date	Targets produced at ORNL, shipped to JINR in January 2015
2015	JINR	Pu-239/240	Connect SHE island to nuclear mainland	
2015	RIKEN	Cm-248	Decay properties of Z=116, new Z=118 isotopes, search for Z=120	Multi-year campaign
2015	LBNL	Am-243 Np-237	Alpha, gamma, x-ray spectroscopy, mass measurements	
2017	GANIL	Am-243 Np-237	Extremes of nuclear mass and charge, new isotopes	S3 spectrometer
2017	JINR/ Factory	Cm-248 Am-243	Reaction mechanisms for beams heavier than Ca, search for 296(118)	Development and commissioning of high current beams at SHE Factory
2018	ANL	U-238 Pu-242/244 Cm-248	Radioactive beam commissioning of AGFA spectrometer, reaction mechanisms	New gas-filled spectrometer and upgraded accelerator capabilities
2018	LBNL	Cm-248 Pu-244	Alpha, gamma, x-ray spectroscopy, mass measurements	Excited states of SHE island nuclei
2019	GANIL	Cm-248 Pu-244	New neutron-rich isotopes, search for Z=120	High intensity LINAC, S3 spectrometer
2019	RIKEN	Cm-248	Multi-year campaign to search for Z=119	
2019	JINR/ Factory	Bk-249	Spectroscopy of odd-Z nuclei, search for Z=119	Requires Bk production in HIFR (ORNL)
2020	JINR/ Factory	Mixed Cf	Search for heaviest nuclei 297(118) and 298(120)	Requires processing of old Cf sources at REDC (ORNL)

Requested targets will enable a wide-ranging international SHE research program

Facility	2015	2016	2017	2018	2019	2020
JINR and Factory	Heavy isotopes of 118, connect island to nuclear mainland (^{239,240}Pu, mixed Cf targets)		Reaction mechanisms (new beams), search for 296(118) using Ti beams, Factory commissioning (^{248}Cm/^{243}Am)		Spectroscopy of odd-Z nuclei, search for 119 (^{249}Bk)	Search for 120 and 297(118) (mixed Cf)
RIKEN	Decay properties of 116, new 118 isotopes, search for 120 (^{248}Cm target)				Search for 119 (^{248}Cm target)	
LBNL	Alpha, gamma, x-ray, mass measurements (Z=113-114) (^{243}Am/^{237}Np targets)			Alpha, gamma, x-ray, mass measurements (Z=115-116), excited states (^{248}Cm, ^{244}Pu)		
ANL				AGFA commissioning, reaction mechanisms (^{238}U, ^{242,244}Pu, ^{248}Cm targets)		
GANIL (S3)			New isotopes with extreme nuclear mass and charge (^{243}Am/^{237}Np targets)		New neutron-rich isotopes, search for 120 (^{244}Pu, ^{248}Cm targets)	

- Heaviest nuclei ever produced
- New elements: Searches for 119 and 120 (multiple independent approaches)
- Significant advances in SHE reaction mechanisms and spectroscopy

Heavy isotopes of 118 and new elements 119-120 appear within reach



- ^{251}Cf currently available only in mixed Cf targets that require special handling/processing
- HFIR can produce ^{254}Es, but only in µg quantities
- Significant research opportunity across the island

Discussion