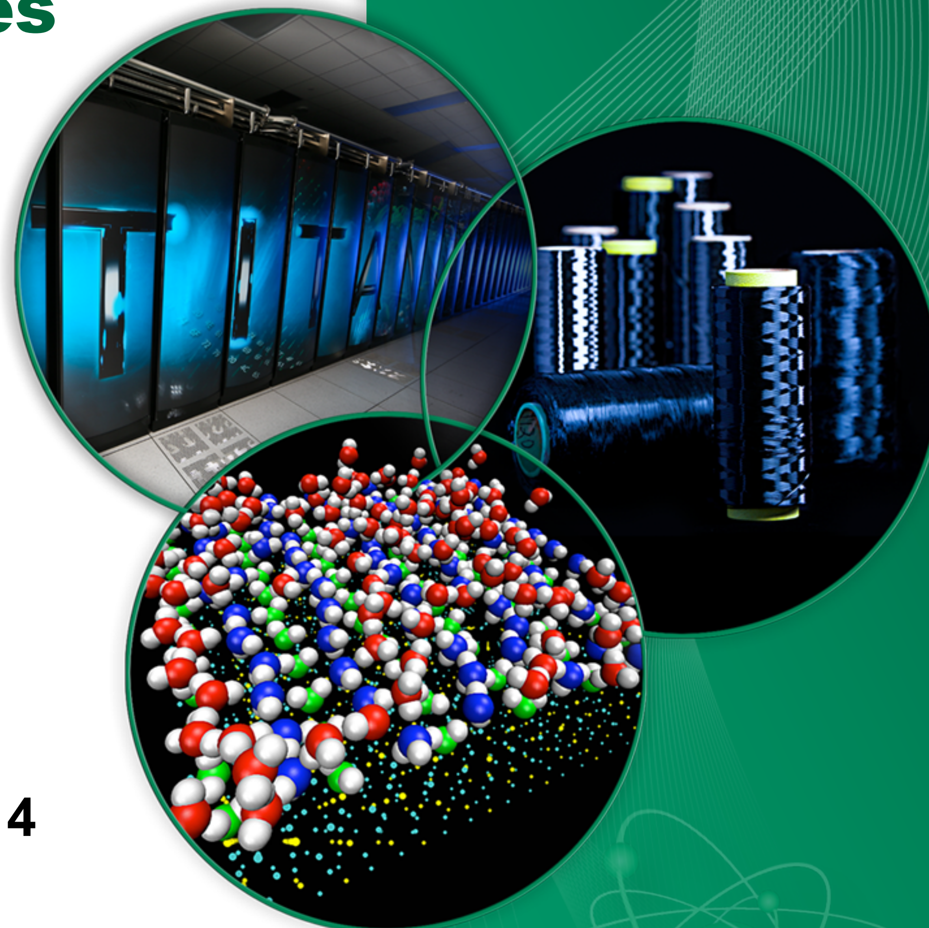
Production of Actinides at Oak Ridge National Laboratory and Their Applications



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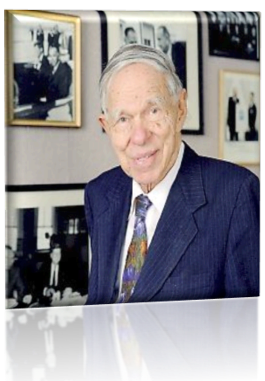
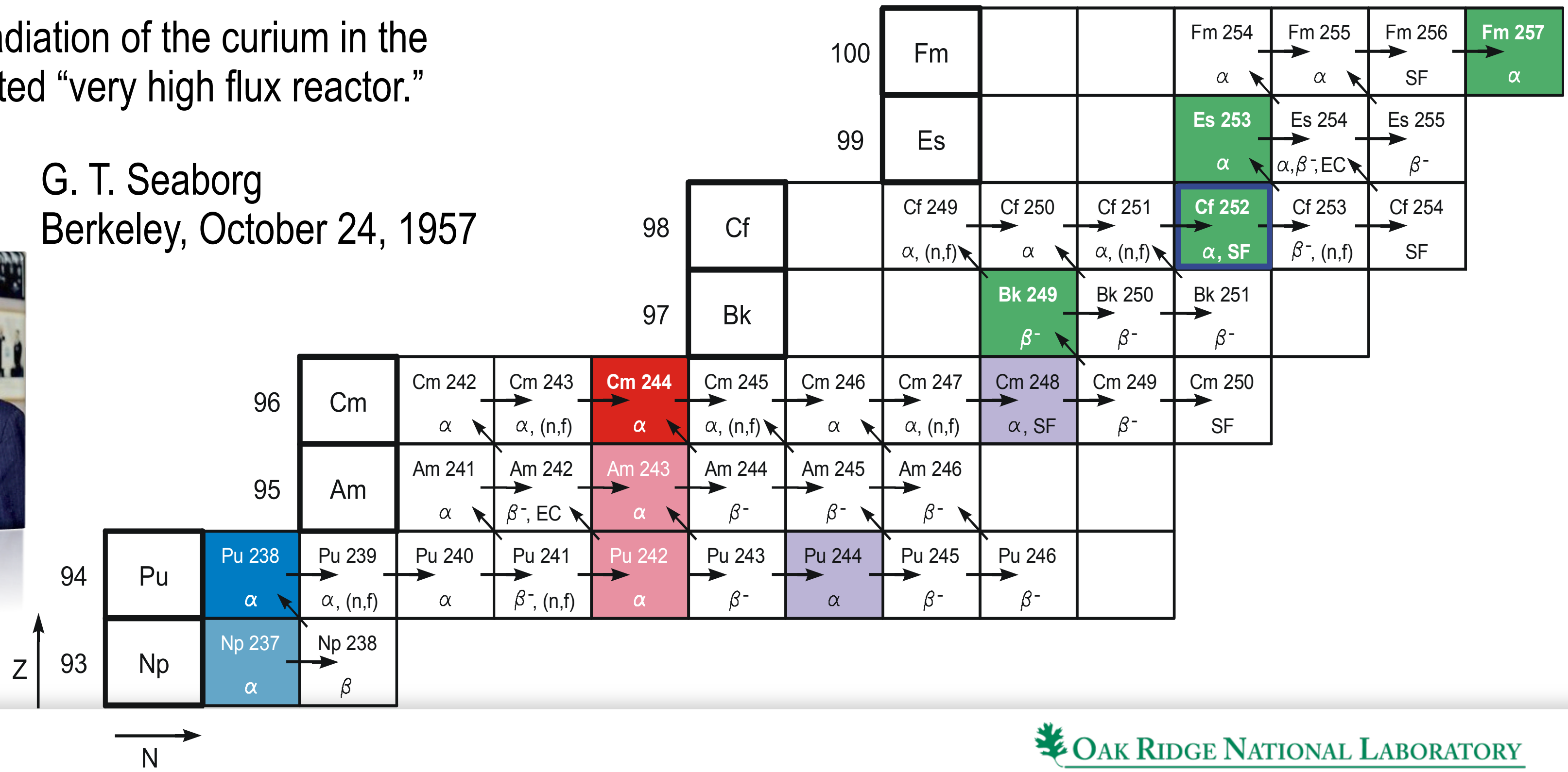
Vision of a Unified Heavy Element Production Program

- The field of new transuranium elements is entering an era where the participating scientists in this country cannot go much further without some unified national effort which can only be authorized and coordinated by the Atomic Energy Commission itself.
- The future progress in this area depends on substantial weighable quantities (say milligrams) of berkelium, californium, and einsteinium. The acquiring of this depends upon our country's entrance into a two-fold program
- The irradiation of substantial quantities of ^{239}Pu as reactor fuel element, and the reirradiation of the products…to form hundred gram amounts of ^{244}Cm and higher curium isotopes…
- The irradiation of the curium in the suggested "very high flux reactor."

G. T. Seaborg
Berkeley, October 24, 1957





ORNL Heavy Element Production Capabilities

- Oak Ridge National Laboratory undertook the design and construction of both the High Flux Isotope Reactor (HFIR) and the Radiochemical Engineering Development Center (REDC) for The irradiation of curium in the suggested **"*very high flux reactor.*"** and the recovery of ***substantial weighable quantities (say milligrams) of berkelium, californium, and einsteinium*.**
- HFIR and REDC began operation in 1965 and 1966
- Operation expected to continue until at least 2040.

The HFIR and REDC Facilities in 1970

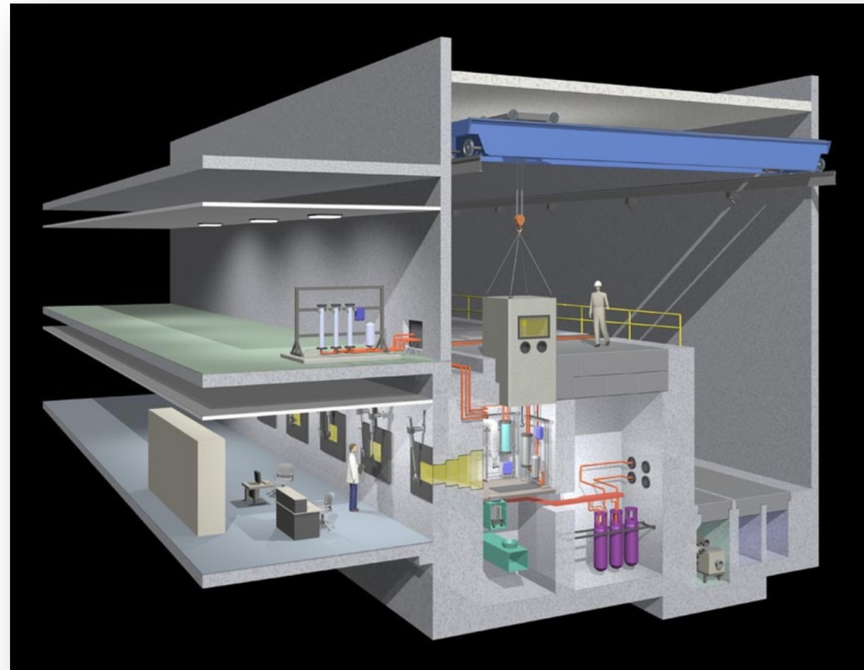
Melton Valley Complex

Radiochemical Engineering Development Center

- Category II multipurpose radiochemical processing facility based around the use of heavily shielded hot cells
- Process irradiated fuel elements and targets for various DOE Programs including Office of Science, National Nuclear Security Administration (NNSA), National Aeronautical and Space Administration (NASA) and Nuclear Energy
- Primary thrust areas include:
  - Heavy element production and R&D
  - Medical and industrial isotope production and R&D
  - Fuel cycle R&D
  - National Nuclear Security R&D
- Past major programs included: Transuranium Element Production, Mark 42 Processing, Cf Industrial Sales/University Lease, Consolidated Fuel Reprocessing, and Advanced Fuel Cycle Initiative (AFCI)
- Facility design allows for flexibility in adding new processes and equipment to incorporate new tasks and process improvements within the limits of the documented safety analysis
- A number of process development activities are ongoing especially in programs for Nuclear Physics – Isotope Production, NNSA, NASA – Pu-238 Supply Program, and Nuclear Energy – Fuel Cycle Research & Development

REDC Building 7920 Features for Radiochemical Processing and R&D

- Nine heavily shielded hot cells for radiation control and alpha containment
  - Three hot cells dedicated to pellet production and target fabrication
  - Four hot cells for chemical processing
  - One hot cell for sample analysis
  - One hot cell for waste handling
- One lab with two shielded caves for final product purification and process R&D applications
- Four glove box labs for final product purification and other R&D

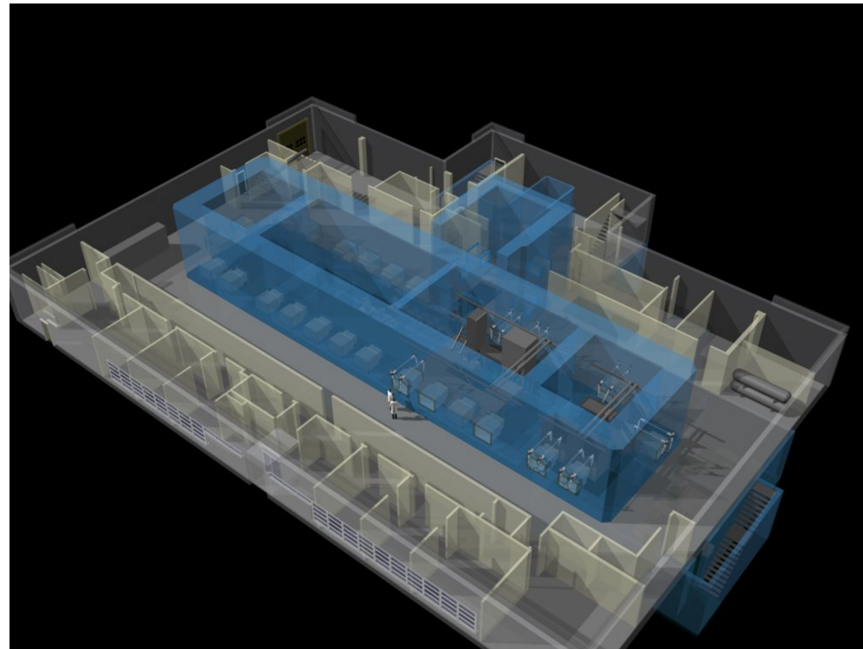


- Radiochemical labs for sample preparation and analysis
- Four cold labs for chemical make up, cold testing, and target fabrication support

Transuranium (Heavy) Element Production

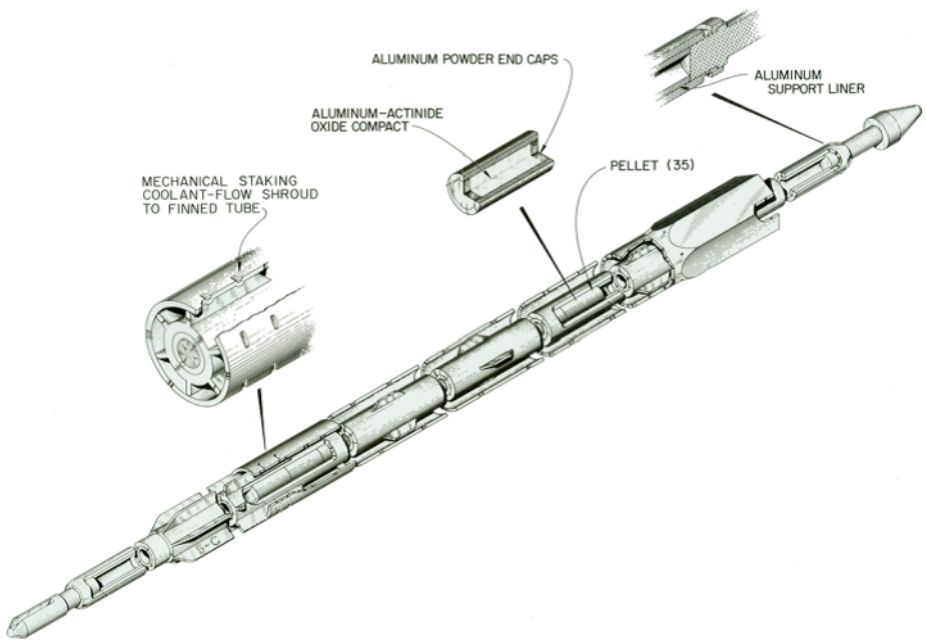
- A historical mission of the REDC has been to provide Cf, Bk, Es, and Fm for research purposes and industrial uses
- Americium/curium cermet targets are fabricated at the REDC
- Targets are irradiated at the HFIR to produce transcurium isotopes up to ^{257}Fm
- Irradiated targets are processed at the REDC to chemically purify and separate the transplutonium actinides
- Actinide products are packaged and shipped to the research community and other users
- Heavy element processing campaigns are conducted approximately every 24 months with a primary focus on ^{252}Cf production

REDC Cf Facility (Building 7930)



- Contains six heavily shielded and one unshielded hot cells for radiation control
- One cell is used for purification of Cf for fabrication into neutron sources, one cell is used for final encapsulation, decontamination, leak checking, and loading/unloading of shipping casks, and one cell is used for loading/unloading of small shipping casks
- One glove box lab, a counting lab, and additional chemistry lab areas available to support facility operations
- A water-filled pool is used as a storage basin for fabricated neutron sources to provide radiation shielding
- Over 300 ^{252}Cf sources currently in pool storage and many available for recovery of ^{248}Cm and mixed Cf
- Over 200 ^{252}Cf sources on loan to university and DOE facilities

Target Assembly

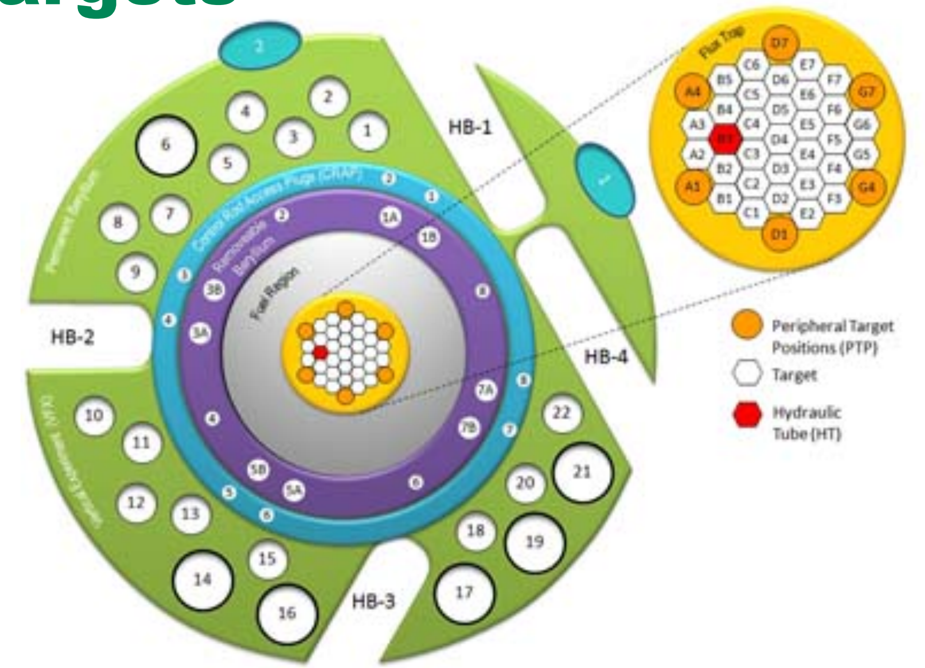


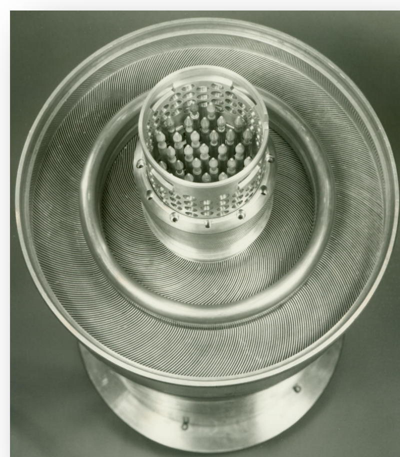
- 35 pellets per target
- Maximum 10 g actinides per target
- Maximum allowable heat flux 1.24×10^6 Btu-h^{-1}-ft^{-2}

- Remote welding, hydrostatic compression, and shroud installation
- Remote QA inspection
  - Radiography (pre- and post- compression)
  - He leak tested to <1.0×10^{-8} std-cc/s

Reactor Irradiation of Targets

- Targets are designed to meet reactor conditions:
  - Compositions designed to control fission and gamma heating
  - Number of cycles and projected operating schedule for irradiations
- Irradiation in the HFIR Flux Trap
  - Working thermal-neutron flux of 2.0×10^{15} neutrons/(cm²·s)
  - 31 target positions
  - 6-8 targets typically irradiated
    - Produces ~35 mg ^{252}Cf per target
    - Smaller quantities of Bk, Es, Fm, others
  - Additional irradiation positions available in reflector region



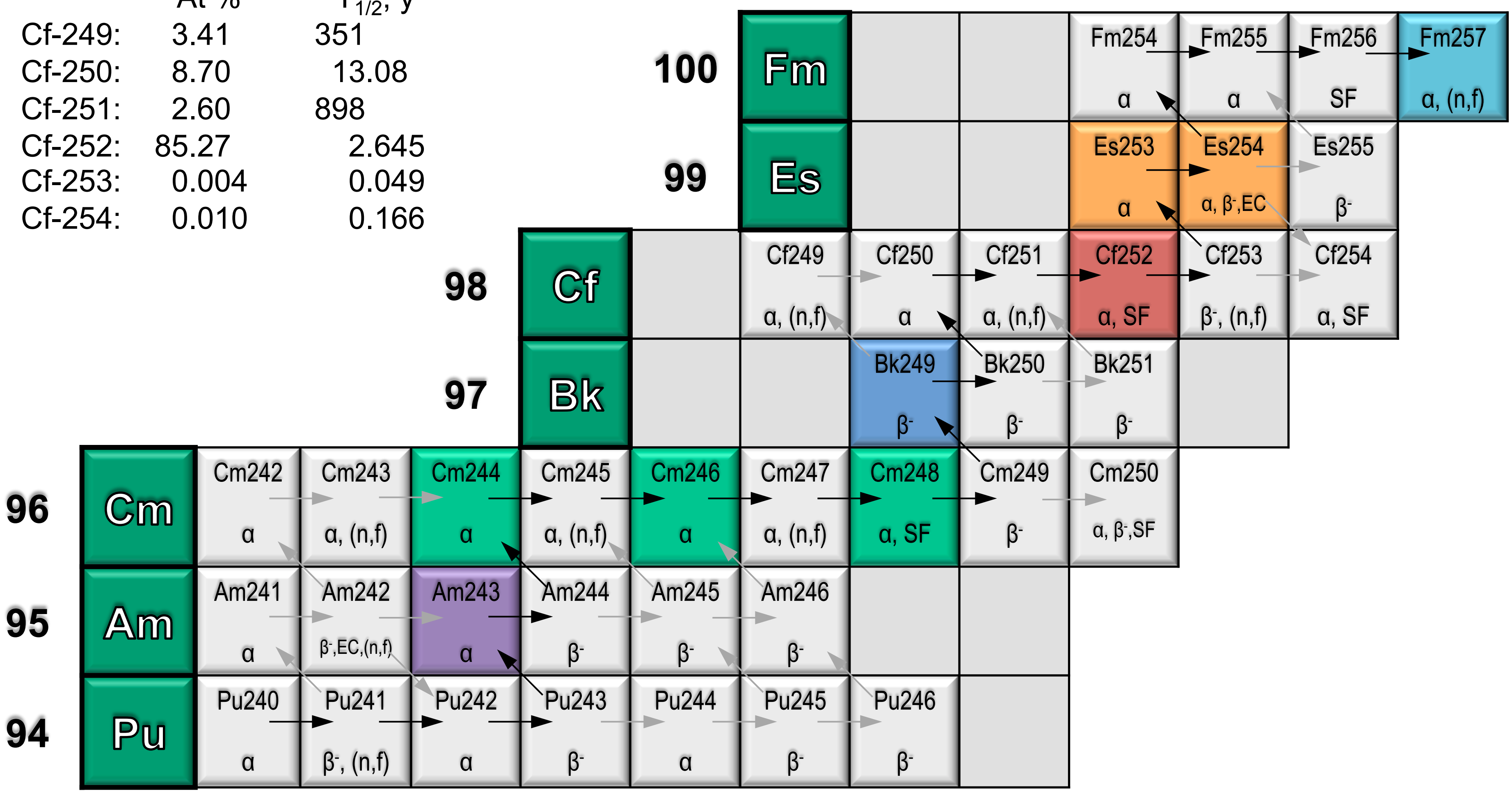


Target positions shown occupied in this HFIR reactor core mock-up

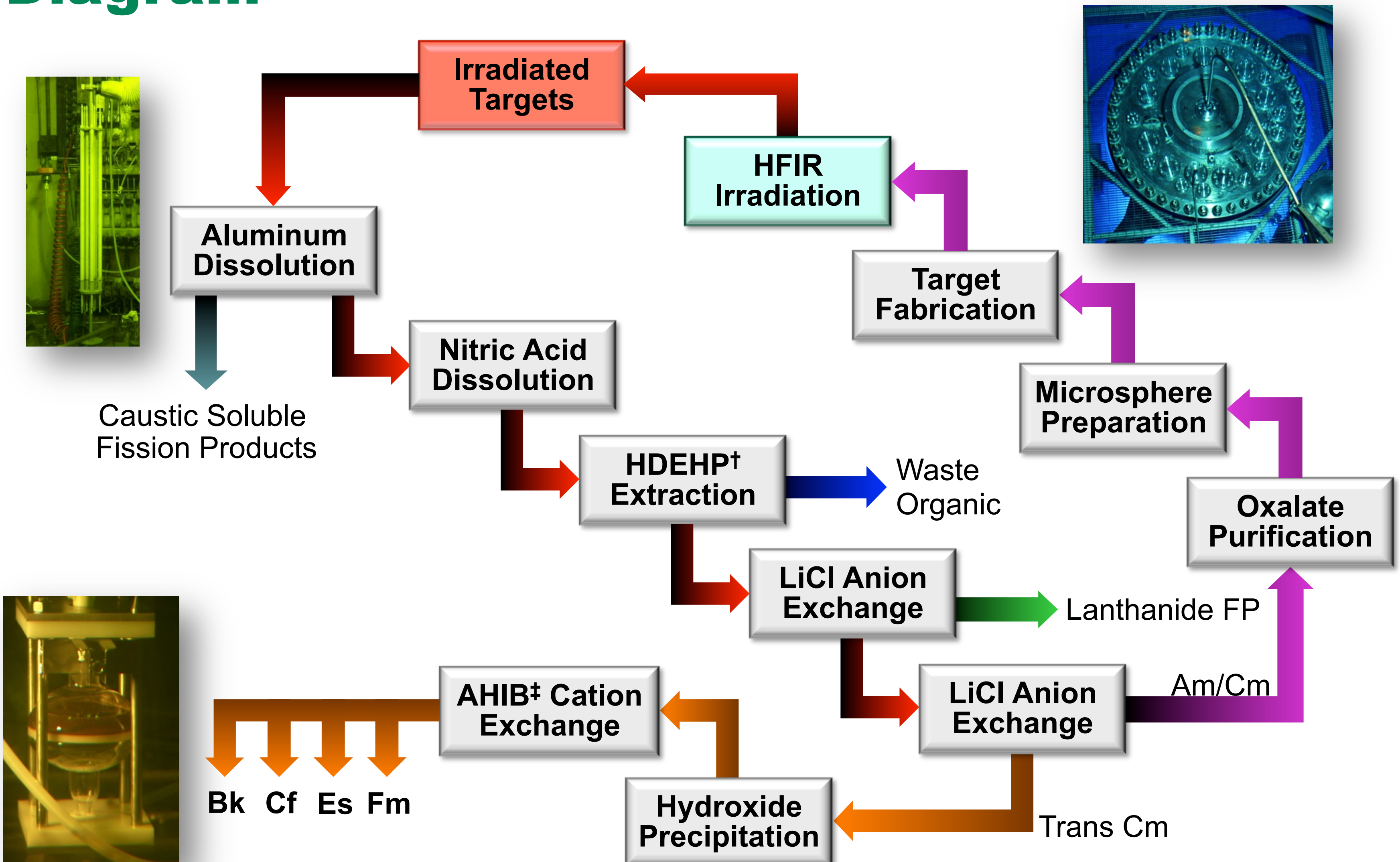
Transuranium Nuclide Production Paths

Typical Cf product isotopic distribution :

	At %	$T_{1/2}$, y
Cf-249:	3.41	351
Cf-250:	8.70	13.08
Cf-251:	2.60	898
Cf-252:	85.27	2.645
Cf-253:	0.004	0.049
Cf-254:	0.010	0.166



Irradiated HFIR Target Processing Flow Diagram

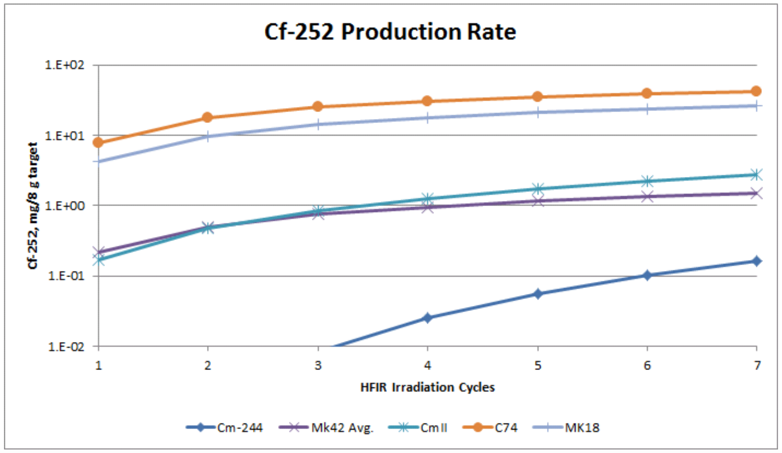


† Di (2-ethylhexyl) phosphoric acid
‡Alpha-hydroxyisobutyrate

Californium Program

Mission

- Purify and fabricate ^{252}Cf neutron source materials for industrial, research, and medical applications
- Recover the ^{248}Cm alpha-decay daughter of ^{252}Cf for heavy-element research
- Successfully completed the initial 4-year production contract
- In FY 2013, DOE signed a new 6-year production contract
- Recently completed Campaign 75 processing which yielded ~160 mg ^{252}Cf



Various ^{252}Cf neutron source designs

Applications of ^{252}Cf

Homeland Security

- Handheld Contraband Detector
- Monitor down-blending of Former Soviet Union Highly Enriched Uranium

National Defense

- Identification of Unexploded Ordinance and Land Mine Detection
- Quality Assurance of weapons stockpile
- Inspection of fighter aircraft wings

Industrial Applications

- Reactor startup sources
- Fuel-rod Scanners, Neutron radiography
- Calibration standards
- Neutron Activation Analysis (NAA)
- Prompt Gamma Neutron Activation Analysis (PGNAA) for on-line monitoring of coal, cement, etc.
- Waste analysis (fissile and transuranic materials)
- Federal Highway Administration measurements for corrosion (bridges, highway infrastructure)

DOE-Complex

- 28 loans of ^{252}Cf sources (109 sources) within DOE and other Agencies
- Determination of Na and actinides in glass encapsulated High Level Waste
- Calibration of personnel dosimeters and radiation instrumentation
- Neutron activation/calibration systems
- Geophysical well-logging
- Neutron shufflers for actinides in waste
- Criticality measurements for spent fuel
- Measurement of Spontaneous Fission Products

Medical and University Uses

- Currently 103 ^{252}Cf sources loaned to 30 Universities and 3 Medical Centers
- Cancer research – inoperable brain tumors
- Laboratory Instructional/Educational Purposes
- Instrument Calibrations
- NAA and PGNAA
- Research in Plasma Desorption Mass Spectrometry

Transcurium Isotopes

^{249}Bk

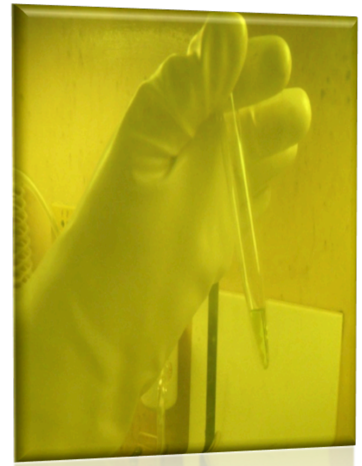
- Utilized in the discovery and confirmation of super-heavy element 117

^{253}Es/^{254}Es and ^{257}Fm

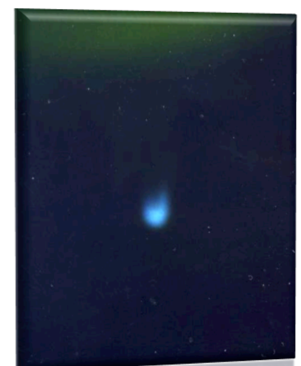
- Predominantly used to determine nuclear physics characteristics

^{251}Cf

- Planned for use in super-heavy element discovery experiments for elements beyond 117



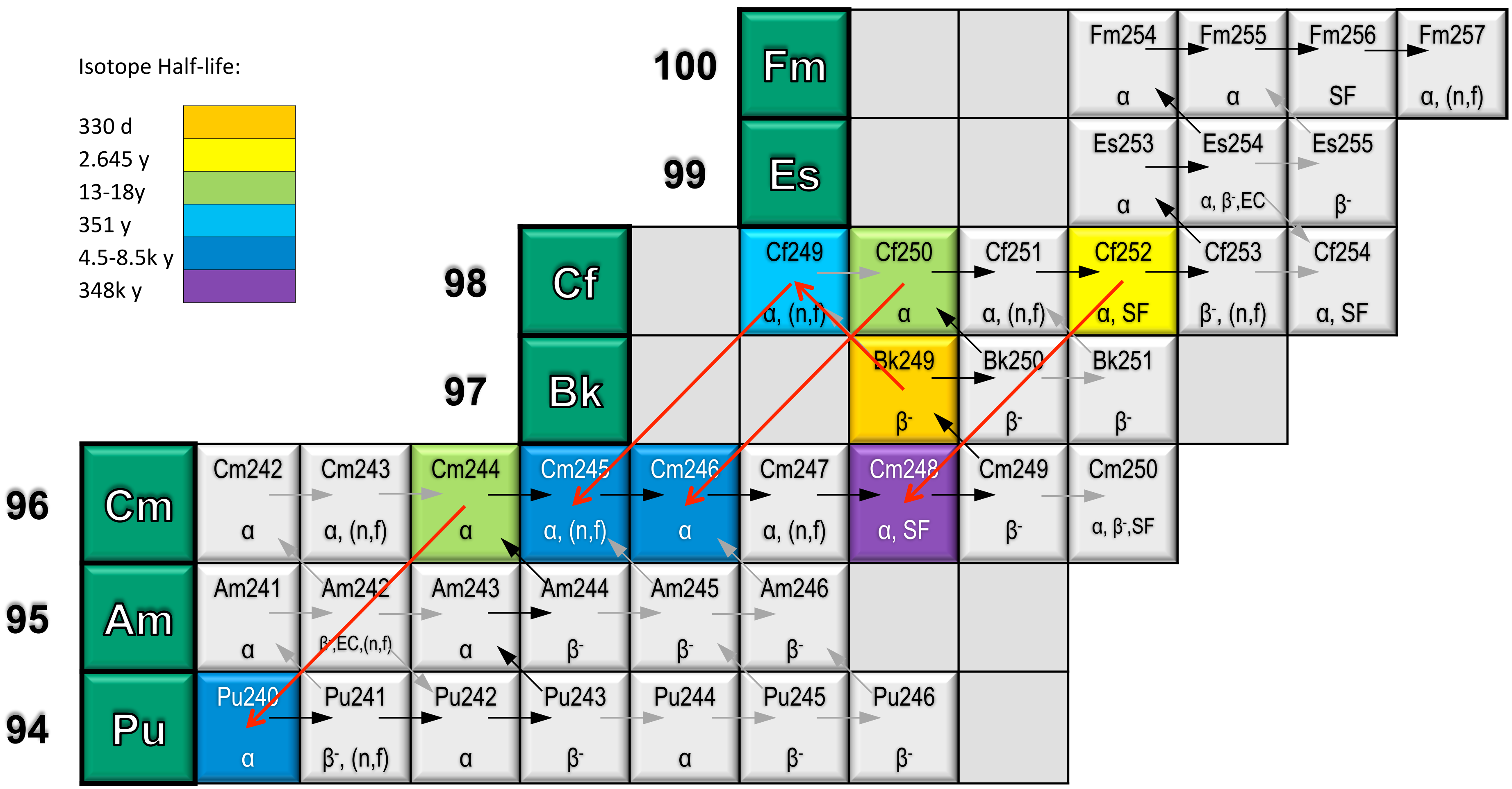
^{249}Bk (22mg)



^{253}Es (0.30mg)

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

Historic ORNL Production and Decay Can Provide Isotopically Pure Materials



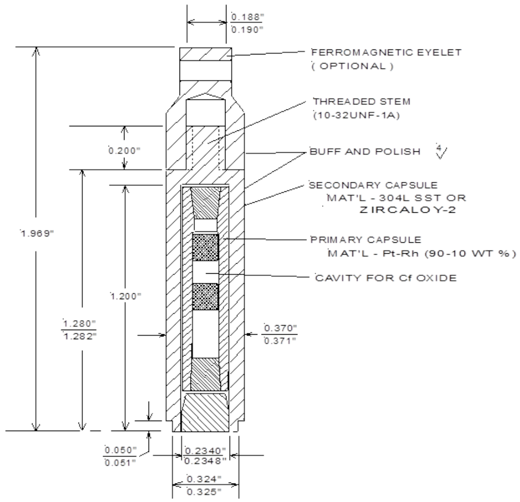
Recovery of mixed Cf for Accelerator Target

- Reviewed Cf content of ~160 candidate sources
- Selected 17 SR-Cf-100 series sources to provide desired material for target, ~ 15 mg total Cf needed
- Sources fabricated in 1971 and 1972; therefore, 40 years of decay

Prediction of Cf content from historic data

Cf Fraction				Total	Cm Fraction				Total
Cf-249	Cf-250	Cf-251	Cf-252	mg	Cm-245	Cm-246	Cm-247	Cm-248	mg
10.5	3.2	7.6	0.004	21.3	0.58	10.7	0.16	19.8	31.2
49.2%	14.9%	35.9%	0.02%		1.9%	34.3%	0.52%	63.3%	

Source Disassembly for Cf Leaching

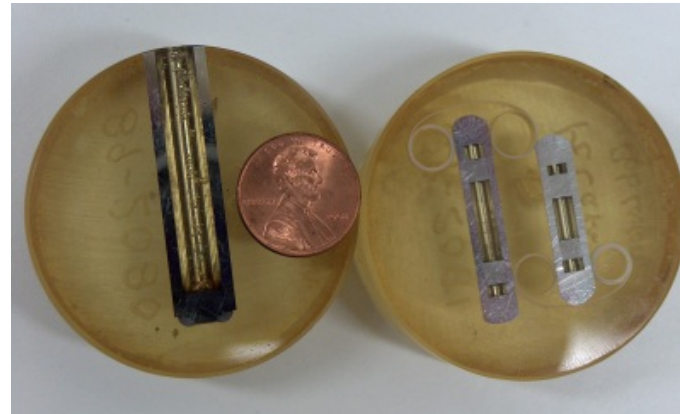


SR-Cf-100 SERIES NEUTRON SOURCE
(PRIMARY AND SECONDARY CAPSULES ASSEMBLED)

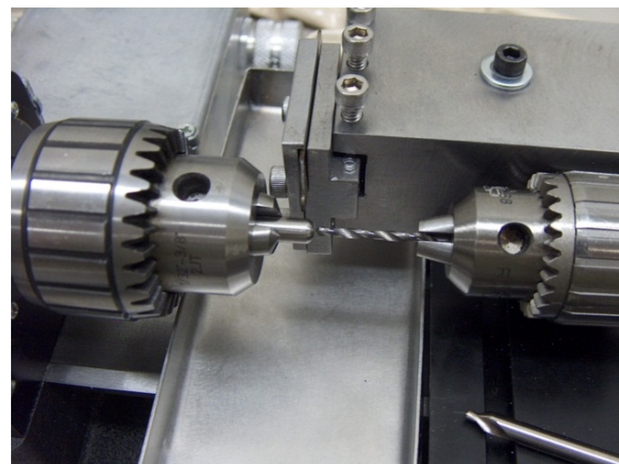
SR-Cf-100 series:

← assembly

section →



- Modified lathe used to remotely remove outer capsule and drill inner capsule for acid leaching
- Inner capsules were cut in half for final leaching



Drilling of inner Pt capsule in preparation for acid leaching

Purification of Mixed Cf Product

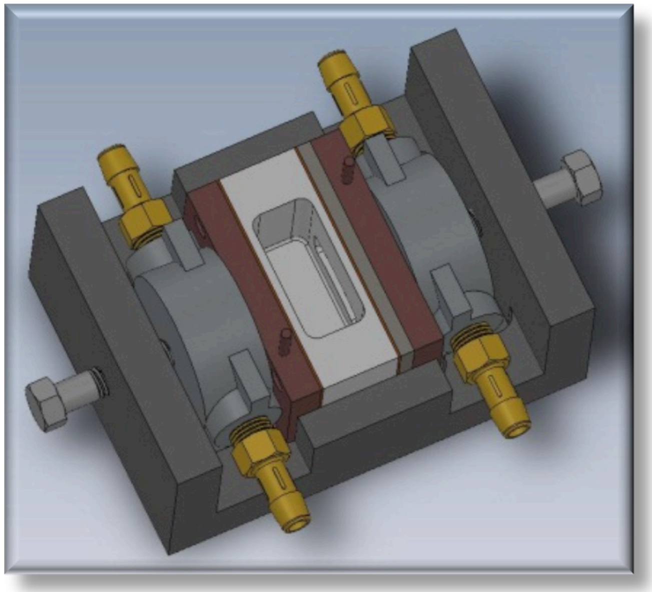
- Capsules leached with 0.5 *M* HNO_3 followed by 4.0 *M* HNO_3

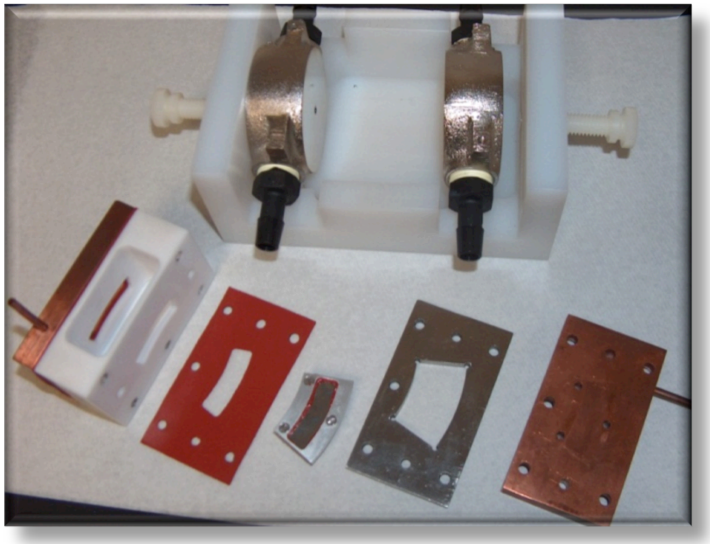
Cf Recovery by Acid Leaching				
Cf-249	Cf-250	Cf-251	Cf-252	Total Cf, mg
7.6	2.5	5.7	0.007	15.8
48.1%	15.6%	36.3%	0.04%	

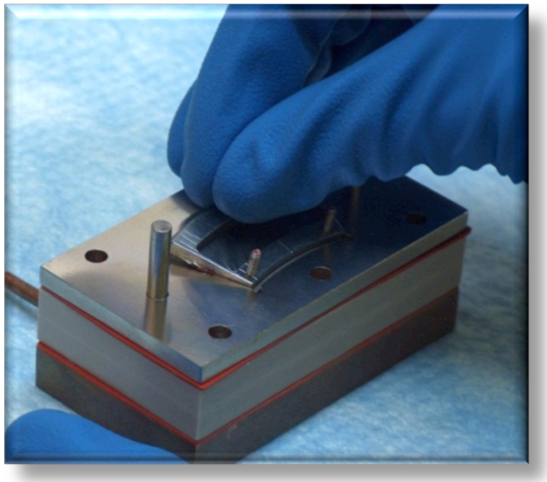
- Cf and Cm separated by cation exchange with 0.25 *M* AHIB at pH 4.2 and 4.6 with excellent recovery
- Cf fraction concentrated by cation exchange
- Additional purification required before product can be used for electrodeposition

New Electrodeposition Equipment

- Fabricated new equipment for electrodeposition on target segments
  - Target segment mounts on side of deposition well; unit is water cooled
- Goal is to electrodeposit ~ 1 mg Cf per target segment

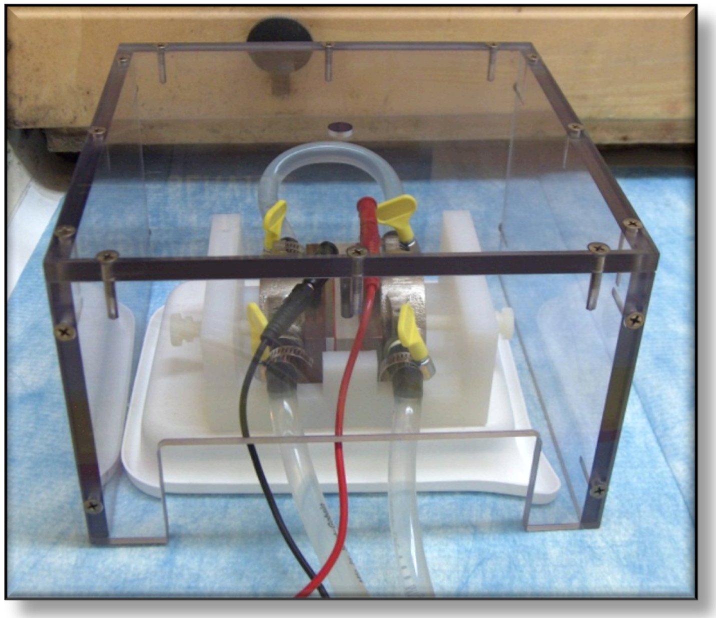






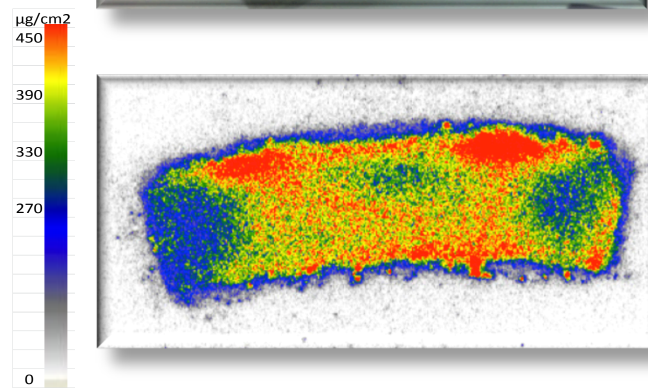
Electrodeposition Development with Surrogate Material

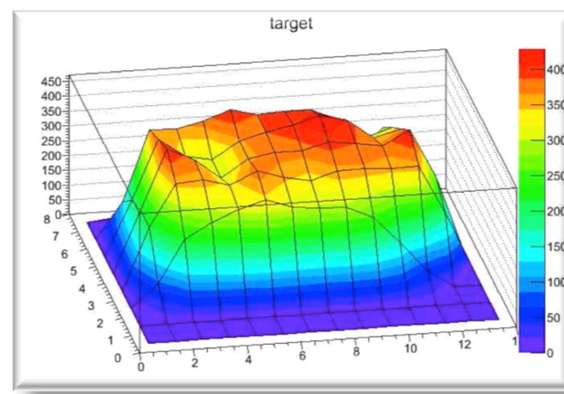
- Developing parameters for electrodeposition with isobutanol using Sm surrogate
- Performed nine deposition trials
  - Feed solution contained 1200 µg Sm with ^{153}Sm tracer
  - Deposition voltage limits in the range, 250-599 volts
  - Deposition current of 0.3 mA/cm^2
  - Tested bare, anodized, and Teflon-coated Al frames
  - Examined heated and non-heated targets for flaking
  - Magnetic stir bar for solution mixing provided more uniform deposition



Trial Electrodeposition Results

- Successful depositions obtained in trials
  - 8 of 9 showed greater than 94% deposition
  - Average deposition was 390 μg/ cm^2
  - Flaking reduced to ~1% with heating (30 min under IR lamp)
- Radiographic image of target shows relatively uniform deposition
  - Gray, no deposition; Blue to Red indicate Sm deposition intensity
  - 99% deposition, 382 μg/cm^2 Sm





Additional Development Activities

- Tested electrodeposition method with ^{249}Cf
- One target segment electrodeposited with a Cf mixture, rich in ^{251}Cf
- Investigated possible use of 2 micron thick Ti foil
  - Minimize wrinkles when mounted on frame
  - May increase uniformity of the depositions
  - Targets would be less fragile during handling and shipping
- Remaining segment electrodeposition in progress
  - Current plan is to fabricate 15 segments with 12 required for target wheel
  - Desire is to minimize time between deposition and start of experiment to ensure target durability

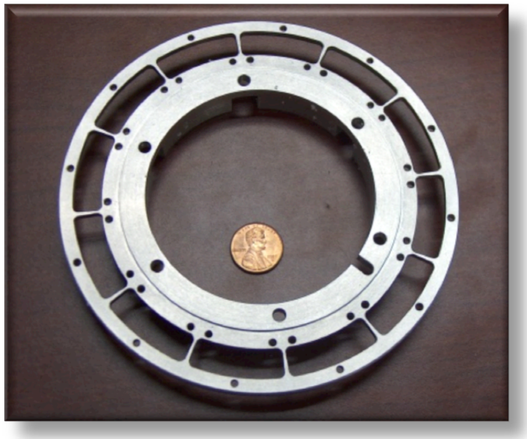
Transportation of Target Segments

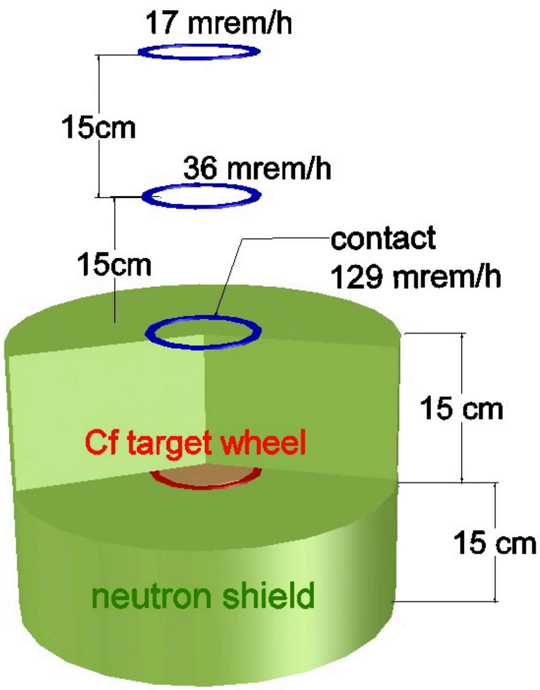
- Analysis of transportation of target segments was favorable
  - 1/12 of feed would be 0.549 of A_2 limit (normal form)
  - 1/15 of feed would be 0.439 of A_2 limit (normal form)
- Two segments could be packaged as normal form in single US Department of Transportation Type A container
- Shipment will be 7-8 individual drums, 5 or 10 gal size

- Avoids the need for Type B international shipment

Dose Calculations on Target Assembly

- Calculation of expected doses from unshielded assembled target wheel completed
  - Max = 180, 1.09, 0.303 rem/h at distances of 0.05, 15.24, 30.48 cm, respectively
- Borated polyethylene shielding (21.7 cm dia. x 15.24 cm high) provides reduced dose in target assembly
  - Max = 0.129, 0.036, 0.017 rem/h at contact, 15.24, and 30.48 cm above the shielding for the assembled target wheel, respectively
  - Max dose at side of assembled wheel approximately ½ of above values





Final Outcomes

- Successful preparation of 15 segments
  - Sufficient material recovered for preparation of all segments
  - Electrodeposition efficiency sufficient to obtain desired Cf density
- Protection of segments to prevent damage during shipment
  - Designed holder for packaging and shielding of segments
- Shipment and receipt of materials in Dubna
- Final approval for assembly/use of Cf target in Dubna facility
- Handling of target segments and assembly of target wheel with minimal worker dose
- Successful experiment!!

Thank You

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Questions??