Californium-252 Production in the High Flux Isotope Reactor

Test Research and Training Reactors 2013 Annual Conference

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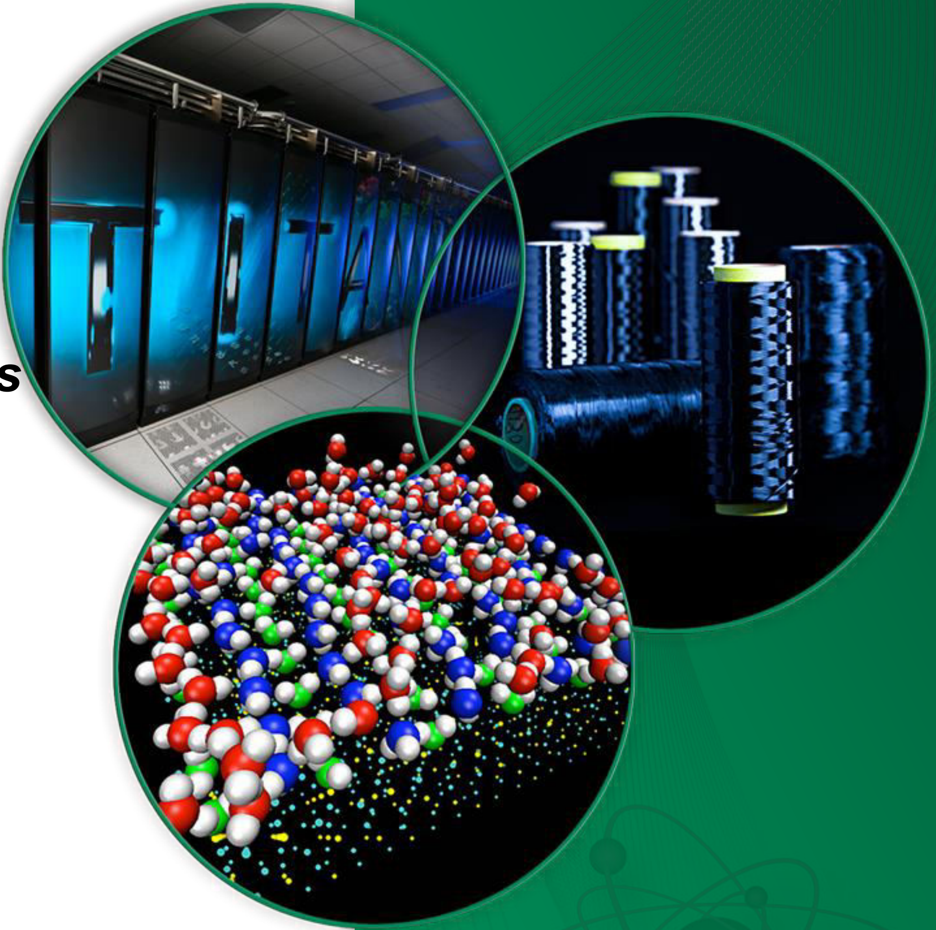
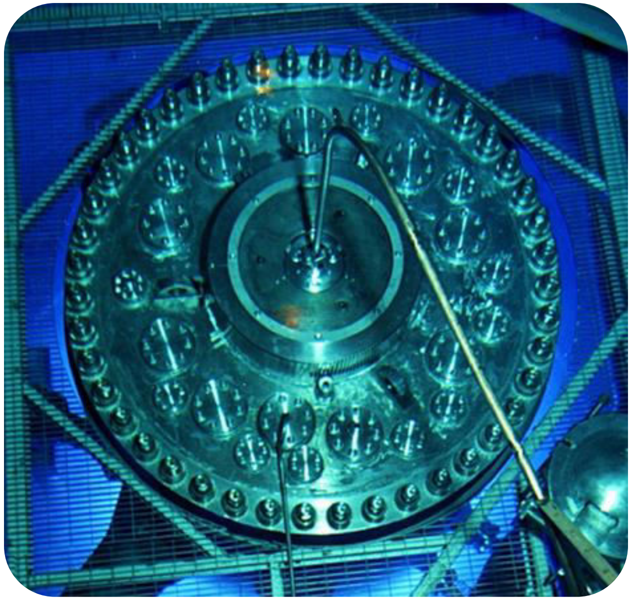


St. Louis, Mo
2013, September 23-26

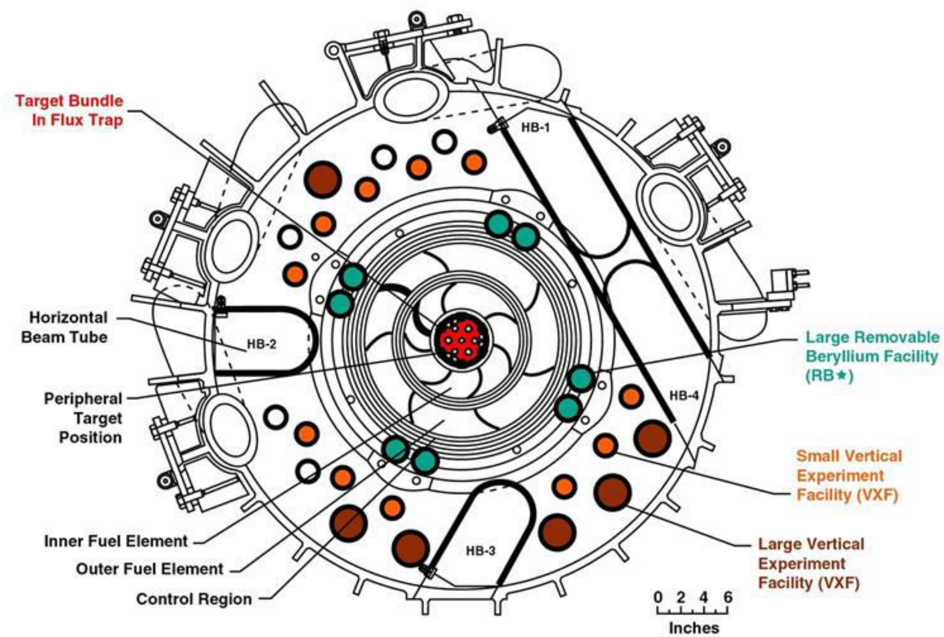
Overview of the HFIR

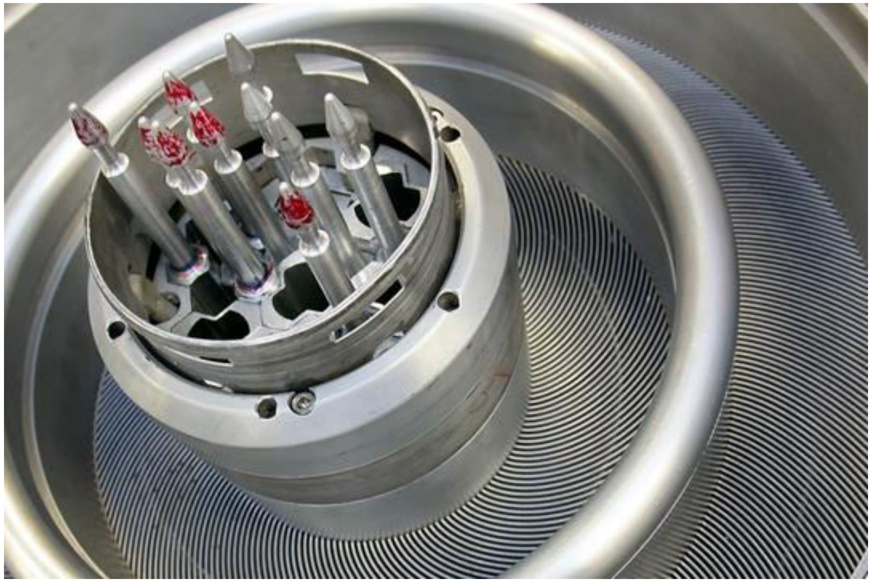
- Light water cooled, Be reflected, pressurized flux-trap style research reactor
- Operates at 85 MW, rated for 100 MW
- 93% HEU involute shaped fuel plates
- Two concentric poison bearing control elements
- Currently operating for 6-7 cycles per year of 24-25 days per cycle
- Mid-plane centerline thermal flux ~2.2 x 10^{15} n/cm^2·s



HFIR Flux Trap Target Island

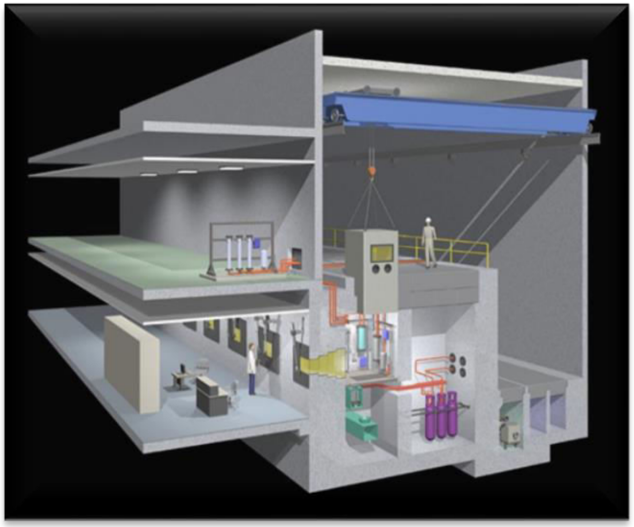
- Flux Trap target island of ~12.7 cm diameter containing 37 vertical target positions
- Each target rod ~1.6 cm diameter, 90 cm height
- One hydraulic tube for experiments <1 cycle in length





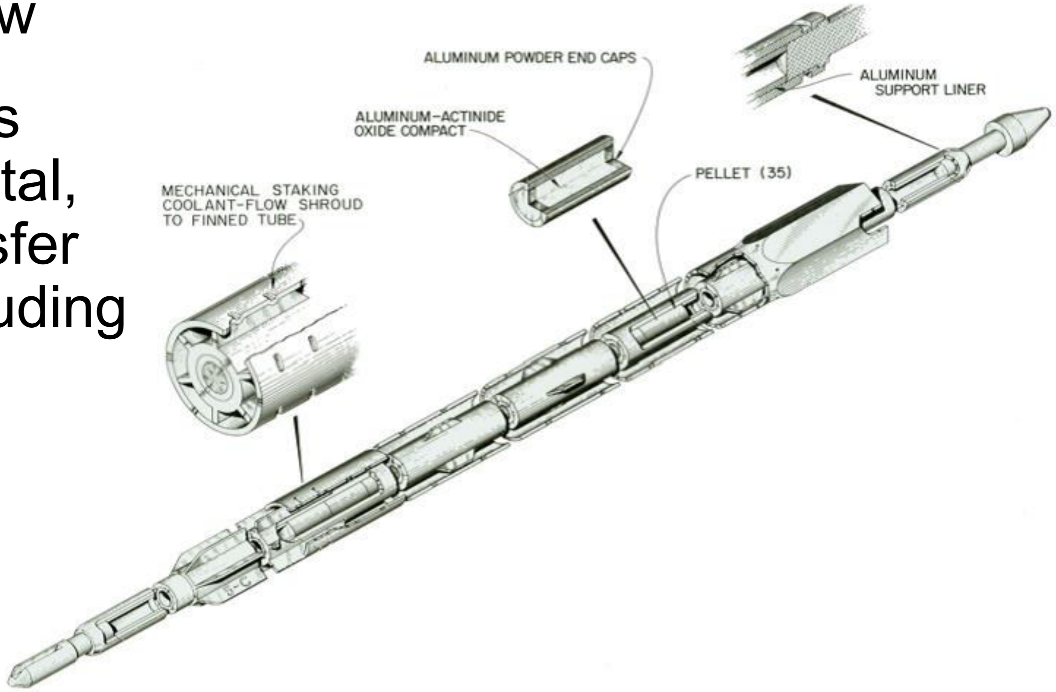
Radiochemical Engineering Development Center

- Flexible hot cell and glove box facility for advanced material handling and recycling
- Highly suited for handling alpha-emitting isotopes
- Located next to the HFIR for easy material transfer
- Provides ~70% of the world's supply of neutron emitting Cf-252 along with other research-oriented heavy isotopes



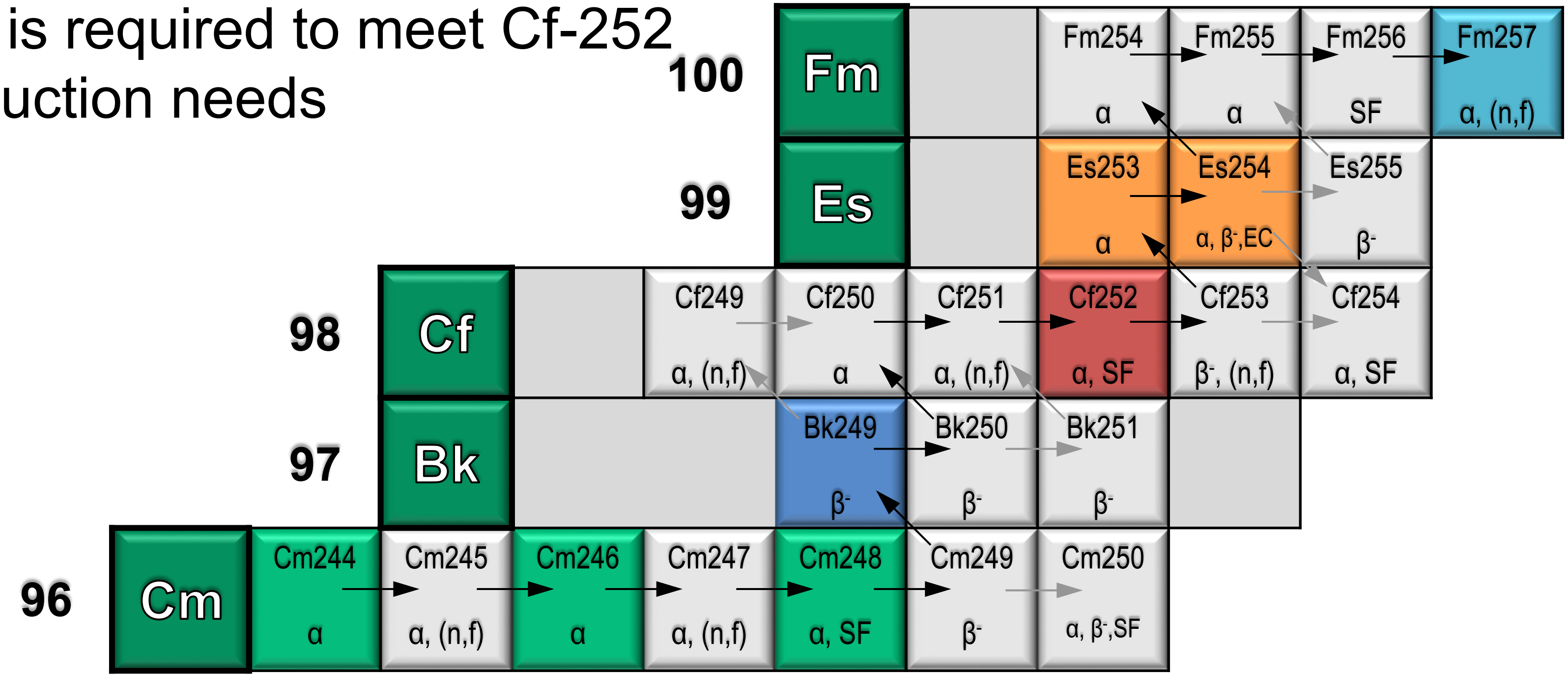
Transcurium Production Targets

- Typical targets composed of mixed curium and americium
- Actinides in oxide formed pressed into pellets in an aluminum matrix
- Pellets are held within a shrouded aluminum target allowing for cooling water flow
- Each target contains 5-8 g of actinide metal, limited by heat transfer considerations, including void for fission gas production.



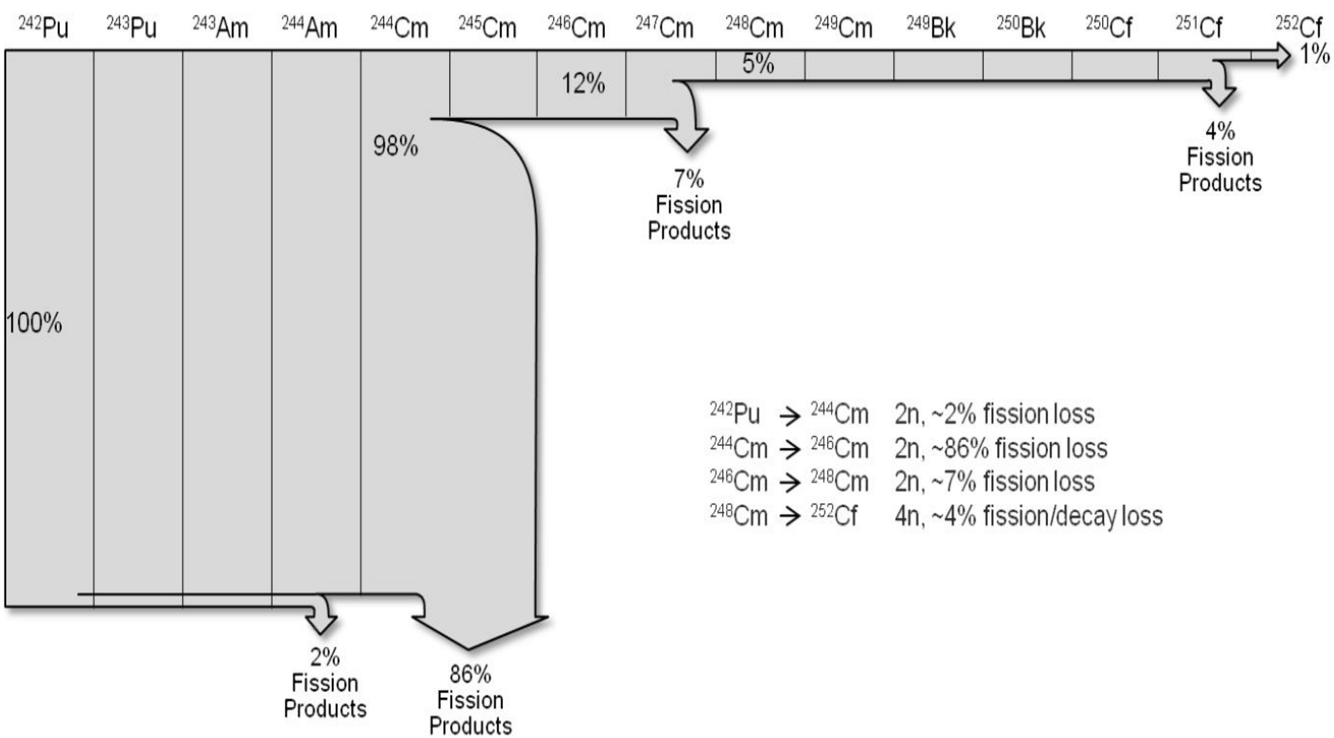
Transcurium Production Chain

- Transmutation from Cm-244 requires 8 neutron captures, while only 4 if starting from Cm-248
- A unique feedstock rich in heavier isotopes of Cm, in addition to a high flux, is required to meet Cf-252 production needs



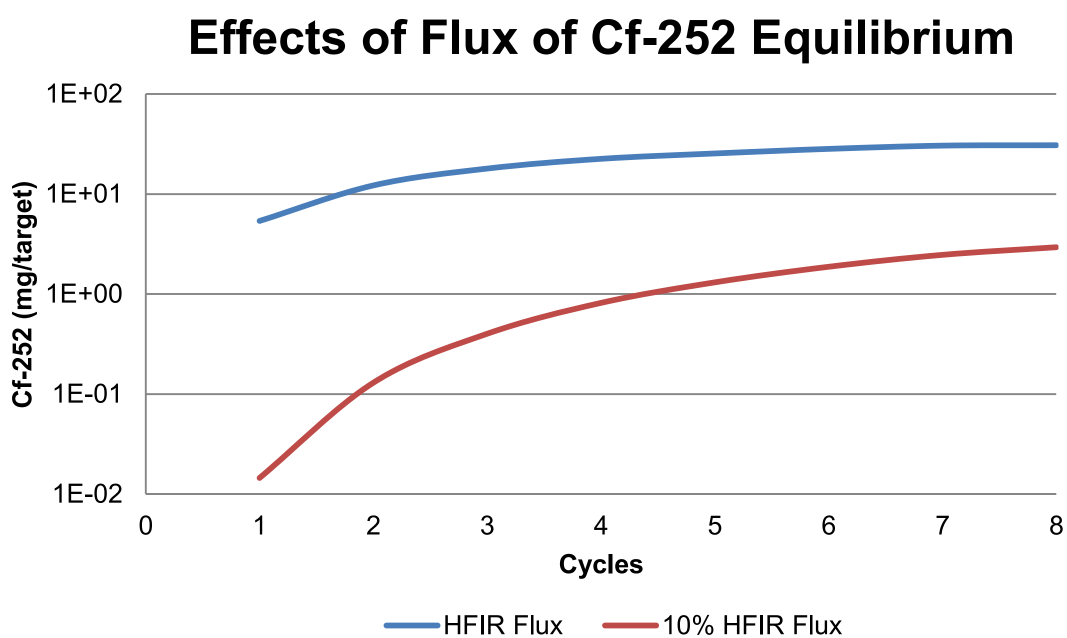
Transcurium Fission Losses

- Largest loss of feedstock occurs during capture in Cm-245
- Starting with Cm-246 instead of Cm-244 results in nearly 10 times more Cf-252 if fully transmuted, and nearly100 times more from the same amount of irradiation



Determining Irradiation time

- Cf-252 reaches equilibrium within ~7-8 cycles in HFIR when decay and transmutation rates of Cf-252 equal production rate
- At this point decay accounts for ~30% of destruction in reactor
- Production efficiency drops severely after 3-4 cycles
- At lower flux this point is reduced and takes longer to reach

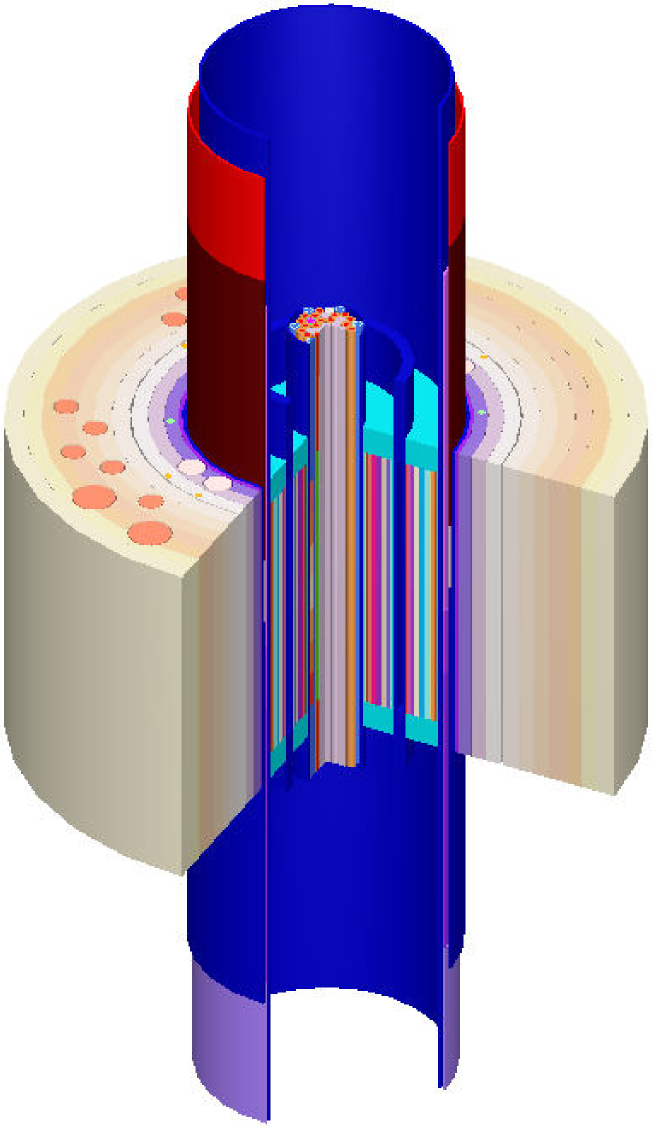


Determining Target Composition

- Supply of heavy curium is of vital interest to the Cf Program
- Looking to the future, it is beneficial to "replace" any heavy curium that is consumed, for sustainable production
- Lighter curium targets irradiated to make new feedstock.
- Irradiated light feed is blended into the heavy feed, with ratios constantly being examined to balance the Cf production needs with the feedstock replacement needs
- Increasing the "weight" of feedstock not only means better feedstock is available for Cf production, but reduces losses due to decay of Cm-244
- Target composition optimization is an ongoing effort

Heavy Use of Modeling and Simulation

- As part of the heavy elements production program, REDC staff perform detailed modeling and simulation of the HFIR
- Models in use includes simple Bateman-chain solvers, deterministic 1-D and 2-D and full 3-D Monte-Carlo using MCNP and SCALE
- REDC researchers frequently collaborate with Research Reactors, Reactor Physics, and other ORNL divisions
- Developing advanced optimization models using genetic algorithms and evolutionary learning



Advanced Heavy Isotope Production

- REDC produces additional heavy isotopes for use in international research and development
- Bk-249, Cf-251, Es-253/254
- Production of rare isotopes can be optimized with more advanced control of the reaction rates
- Dr. Glenn Seaborg initiated the Large Einsteinium Activation Program (LEAP) to produce large quantities of Es-253 and Es-254
- Project was never funded, and prolonged shutdown of HFIR depleted isotope reserves
- Prolonged effort will be needed to get back to these levels

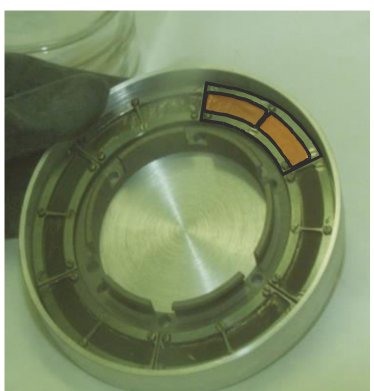
The future progress in this area depends on substantial weighable quantities (say milligrams) of berkelium, californium, and einsteinium. The acquiring of this depends upon our country's entrance into a two-fold program. The irradiation of substantial quantities of ^{239}Pu as reactor fuel element, and the re-irradiation 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."

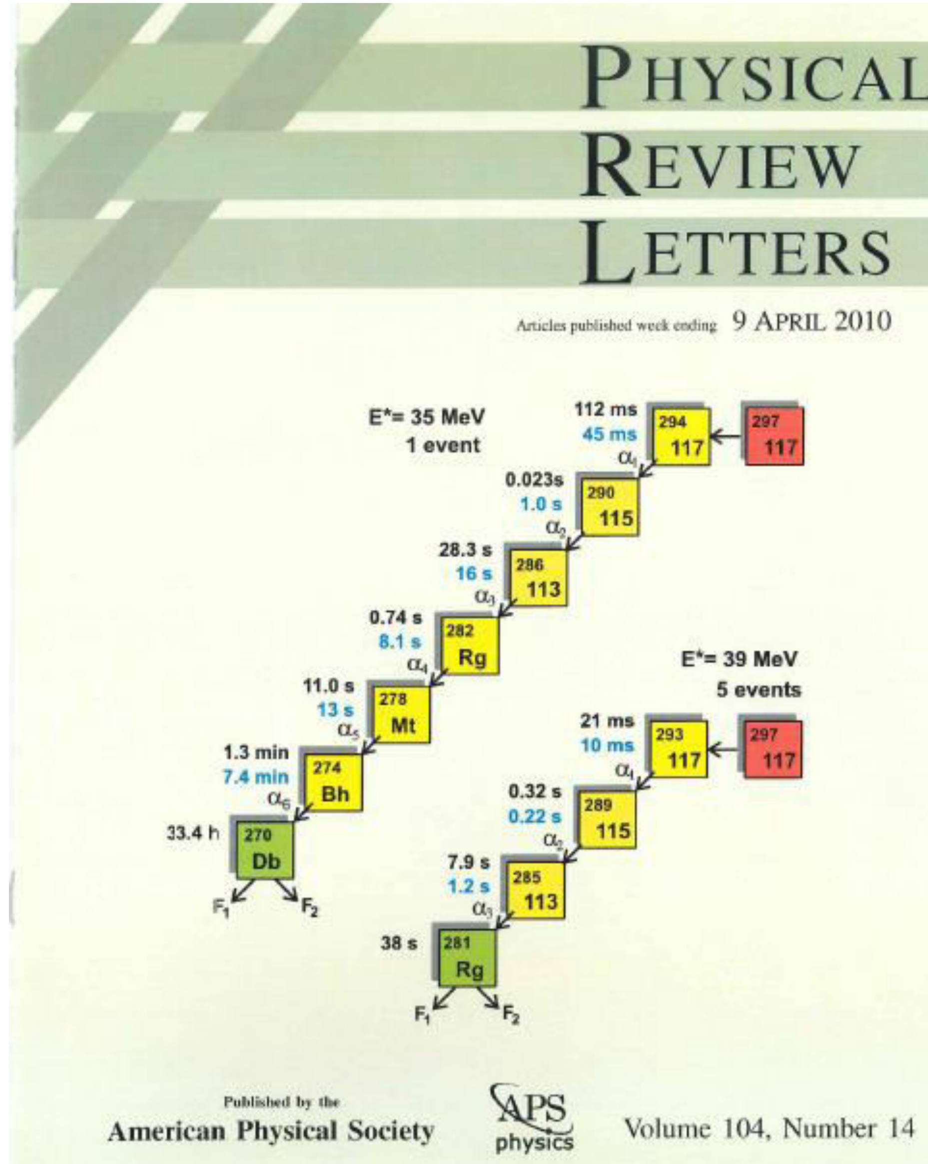
G. T. Seaborg, Berkeley

October 24, 1957

Super Heavy Element 117 Discovery

- Production of 22mg (36 Ci) of pure Bk-249 in HFIR
- 3 mo chemistry at REDC
- 36 cm^2 target wheel produced at RIAR Dimitrovgrad
- Bombarded in ^{48}Ca beam at JINR Dubna



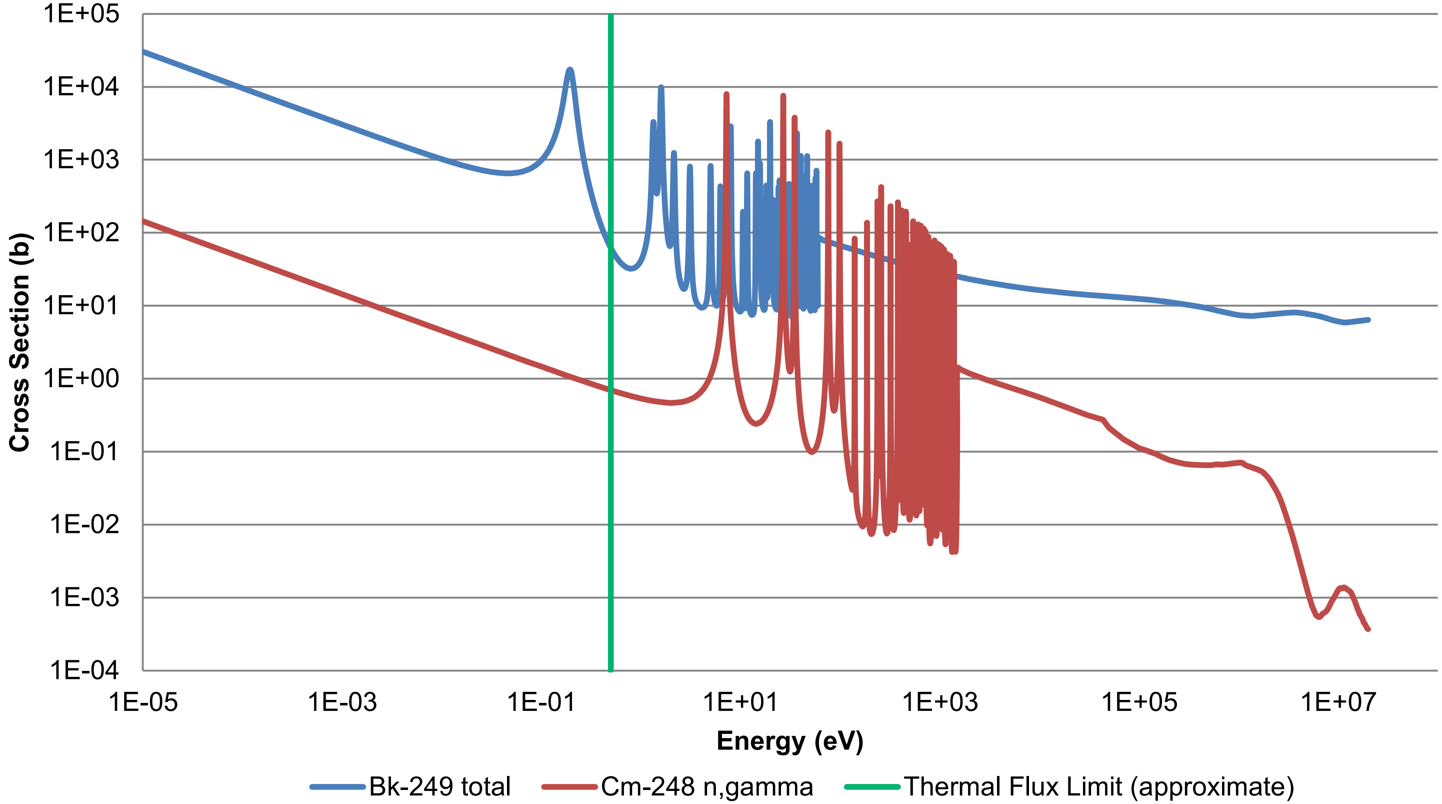


Advanced Bk-249 Production

- Bk-249 is in high demand for target material in the production of heavy and superheavy elements
- Bombarded by ion beams for the discovery/confirmation of element 117 as well exploratory for element 119
- Routinely generated during production of Cf-252, however, this production method is highly inefficient for Bk-249
- Bk-249 reaches equilibrium in ~14 days due to a large burn-up cross section relative to capture in Cm-248
- Bk-249 burnup can be reduced without significantly reducing production rate by filtering out thermal neutrons
- Transmutation Cm-248 cross section is primarily driven by the resonance integral

Bk-249 Production

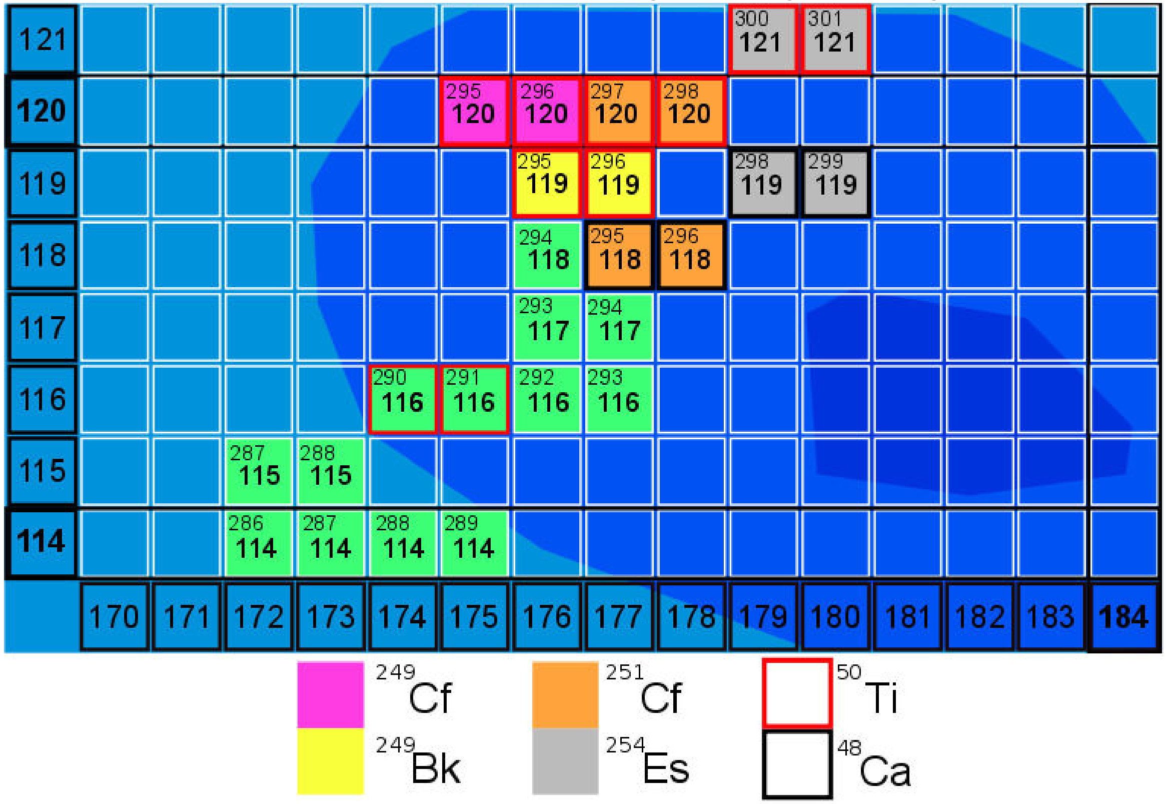
- Absorption past the thermal flux limit (~0.5 eV) results in less burnup of Bk-249 and larger equilibrium values



Advanced Cf-251 Production

- Current Cf-251 sources are made from ~30 year old decayed Cf-252 sources
- As demand increases Cf-251 production will be necessary with shorter decay times
- Similar to Bk-249 production process,
- Thermal neutron filtered Cf-250 targets reducing Cf-251 absorption
- Requires unfiltered pre-irradiation of Bk-249 to produce Bk-250, decaying to Cf-250 in 3.2 hours.

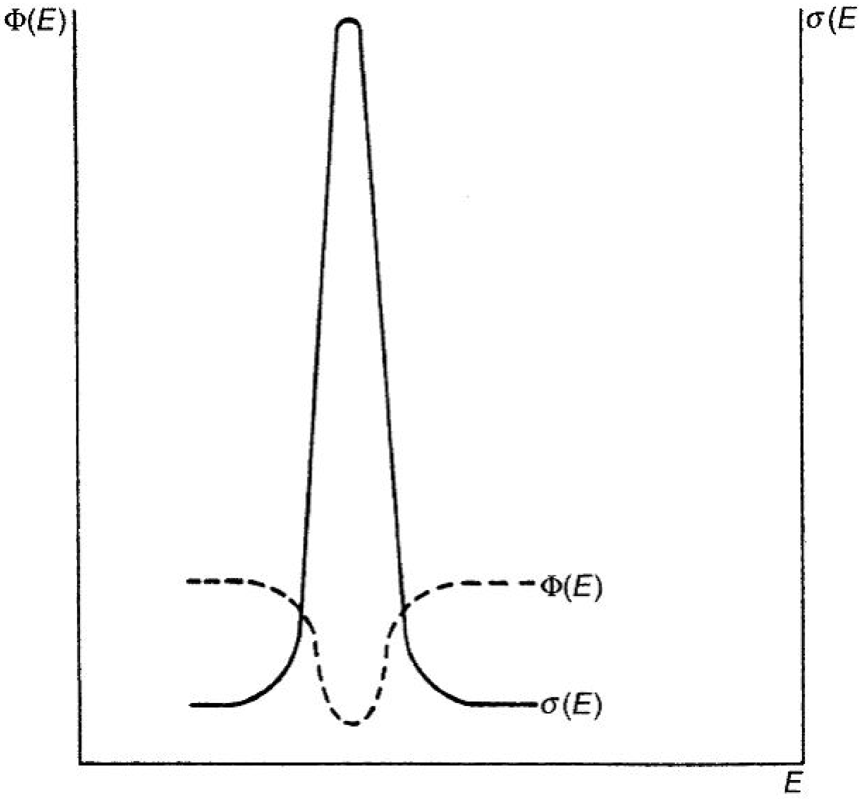
Courtesy of Krzysztof Rykaczewski



Resonance Filtering

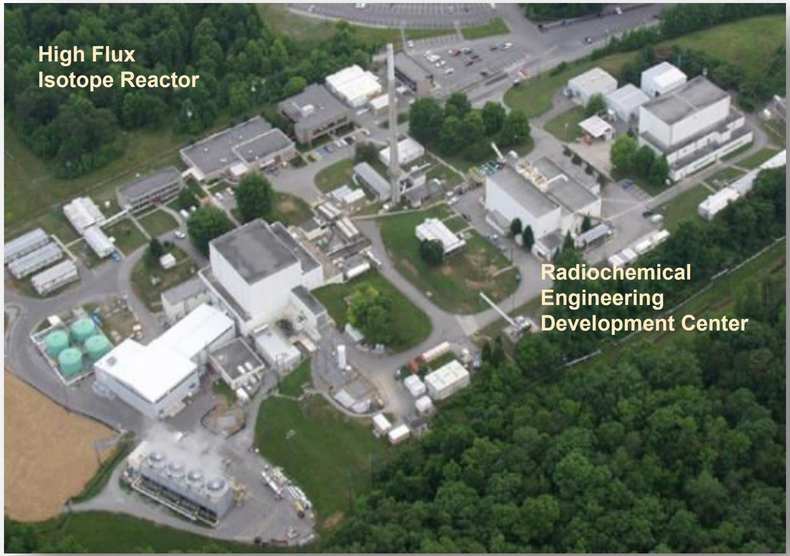
- Preliminary experimental work is currently in development looking at directed resonance filtering
- This technique looks at reducing the flux in specific energy bands where undesirable absorptions are most prevalent
- Potential to increase heavy isotope production per unit of feedstock consumed
- Office of Science funded project to irradiate filtered rabbits in 2014 to examine burn-up rates

Local flux reduction around large resonance



Summary

- HFIR and REDC are uniquely suited for production of Cf-252 and other heavy isotopes due to the very high flux produced in the HFIR flux trap, and REDC's capability to handle and process highly active alpha and neutron emitting isotopes
- For sustainable californium production, optimization of target design and irradiation schemes is necessary in order to maintain the current heavy curium feedstock
- Advanced production of heavy isotopes is becoming more important as demand increases - significant effort is necessary to develop these capabilities
- Experimental work is upcoming to examine advanced techniques for resonance filtering aimed at reducing fission losses



Acknowledgements

Research supported by the Isotope Program, Office of Nuclear Physics of the U.S. Department of Energy.

ORNL is managed by UT-Battelle, LLC, for the U. S. Department of Energy under contract DE-AC05-00OR22725.