Optimizing HFIR Isotope Production through the Development of a Sensitivity-Informed Target Design Process

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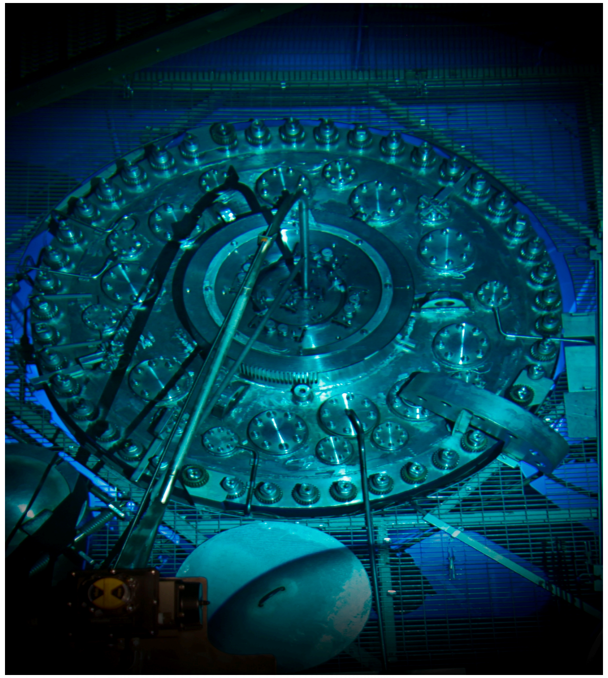
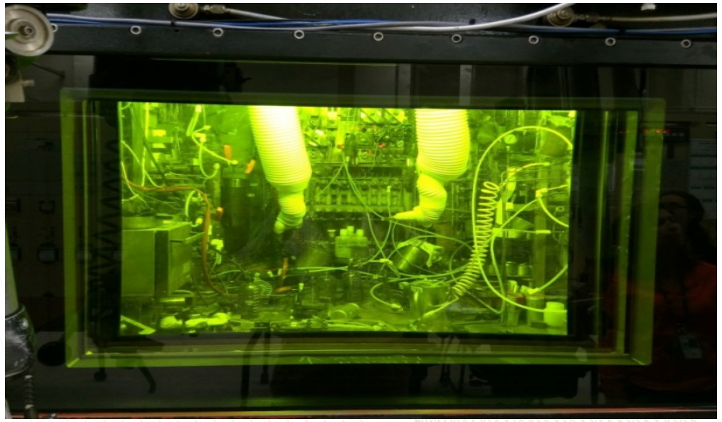
Isotope Production Opportunities

- The High Flux Isotope Reactor (HFIR) at the Oak Ridge National Laboratory (ORNL) provides one of the highest intensity neutron fluxes
- HFIR can provide unique isotopes, some of which have no alternative US production source, including:
  - ^{14}C: medical applications such as studying diabetes, gout, anemia, and acromegaly
  - ^{63}Ni: detection of explosives and airport security
  - ^{229}Th: ^{225}Ac for α-particle cancer therapy
  - ^{238}Pu: radioisotope power for space exploration
  - ^{254}Es: production of super-heavy elements
  - ^{252}Cf: source of neutrons for nuclear reactor startup, study of materials with neutron diffraction, oil well logging, and neutron spectroscopy





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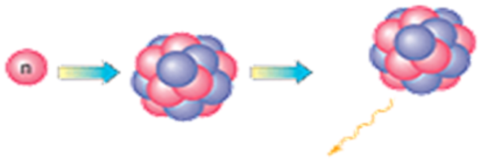
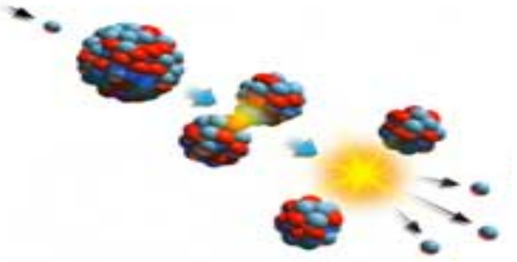
Opportunity to Improve Isotope Production

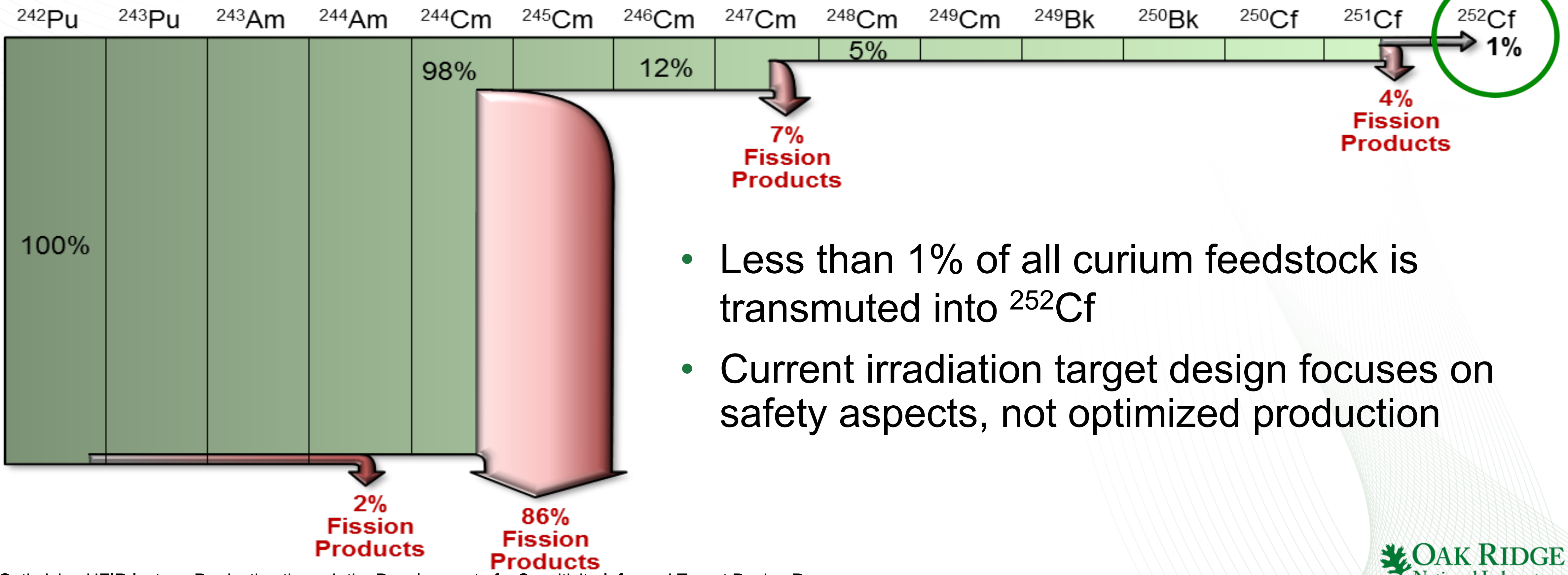
Neutron capture: produces heavier isotopes



Nuclear fission: destroys heavy isotopes



The Long Road to ^{252}Cf



- Less than 1% of all curium feedstock is transmuted into ^{252}Cf
- Current irradiation target design focuses on safety aspects, not optimized production

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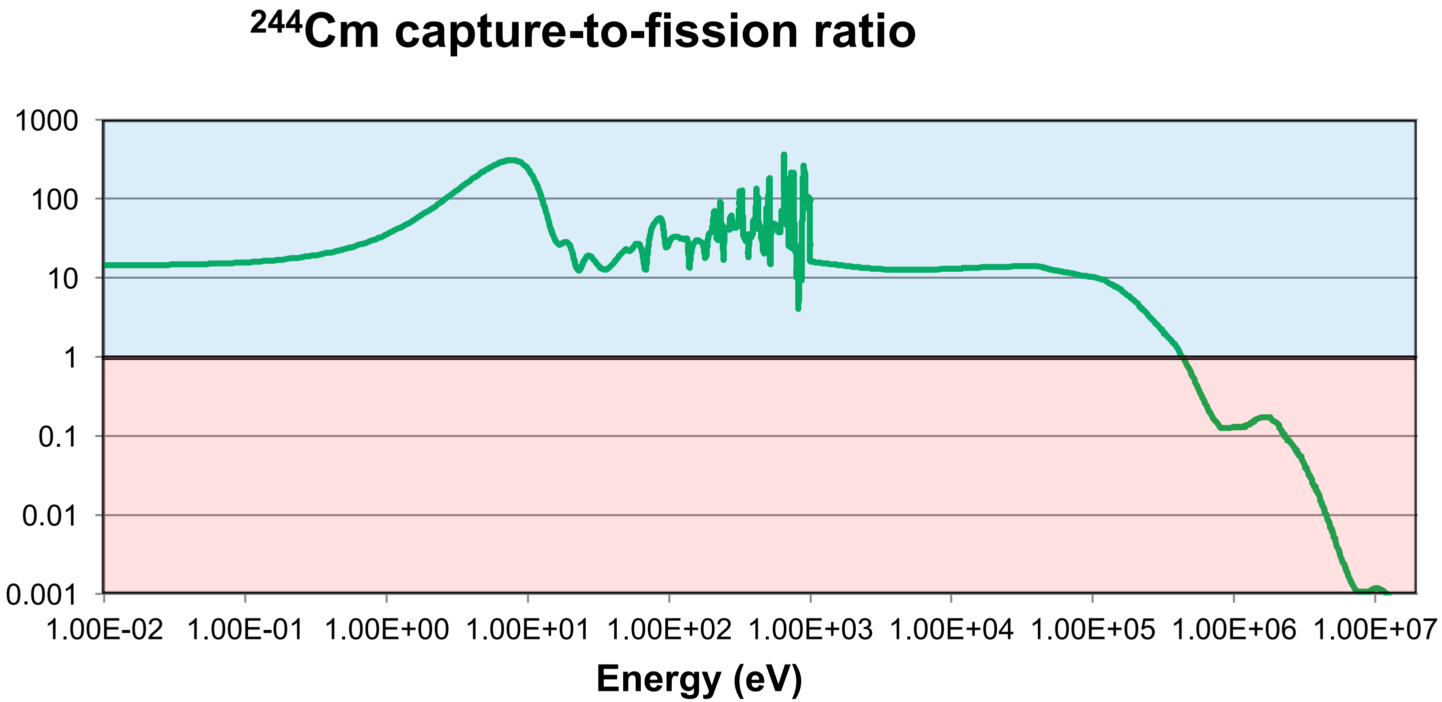
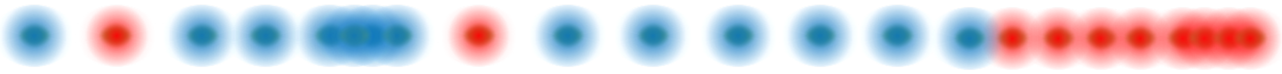
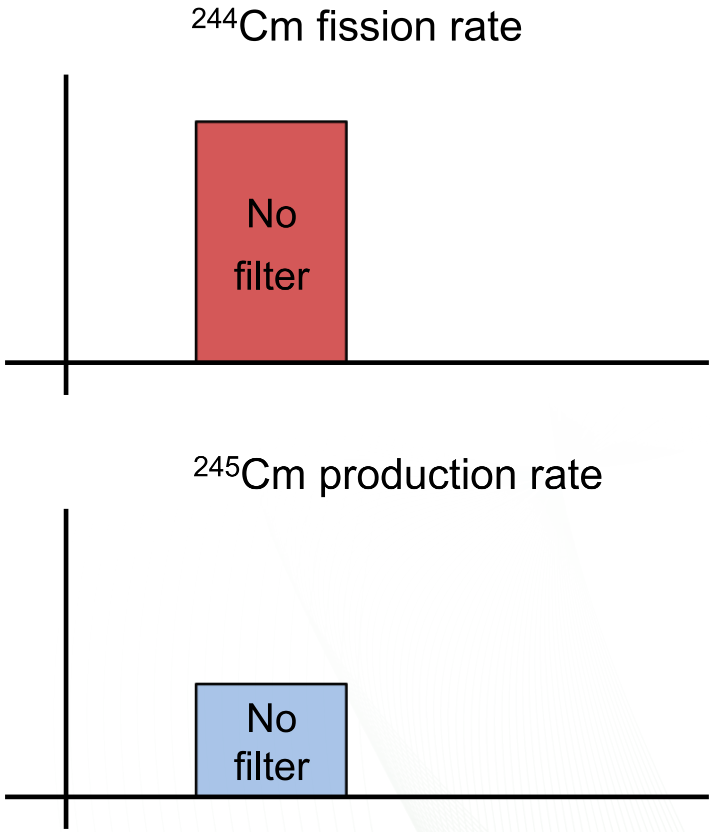
Neutron Filter Design







Fission reaction

Capture reaction

Neutron flux

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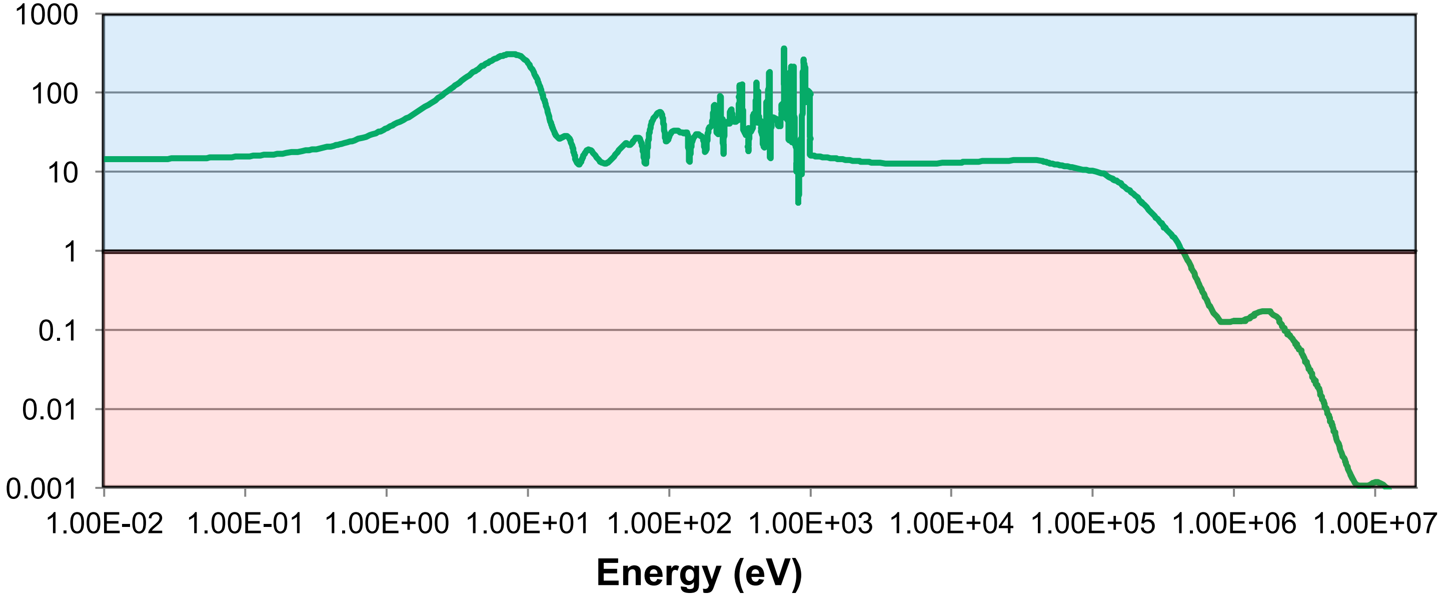
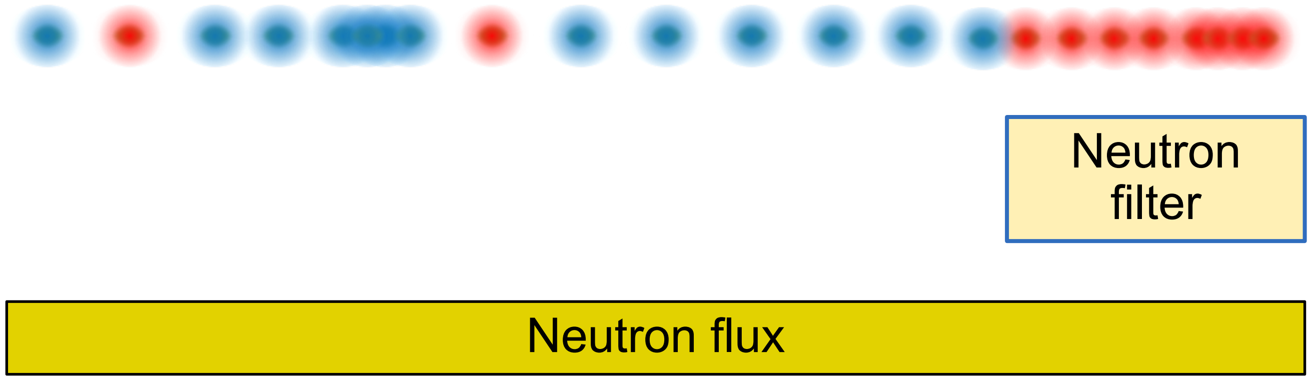
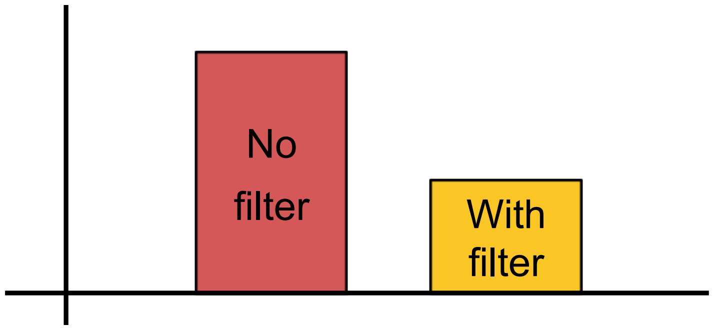
Neutron Filter Design

^{244}Cm capture-to-fission ratio

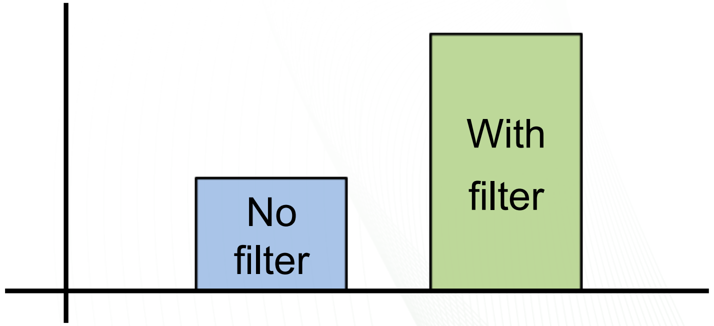


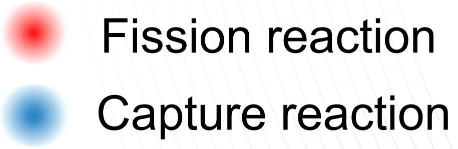


^{244}Cm fission rate



^{245}Cm production rate

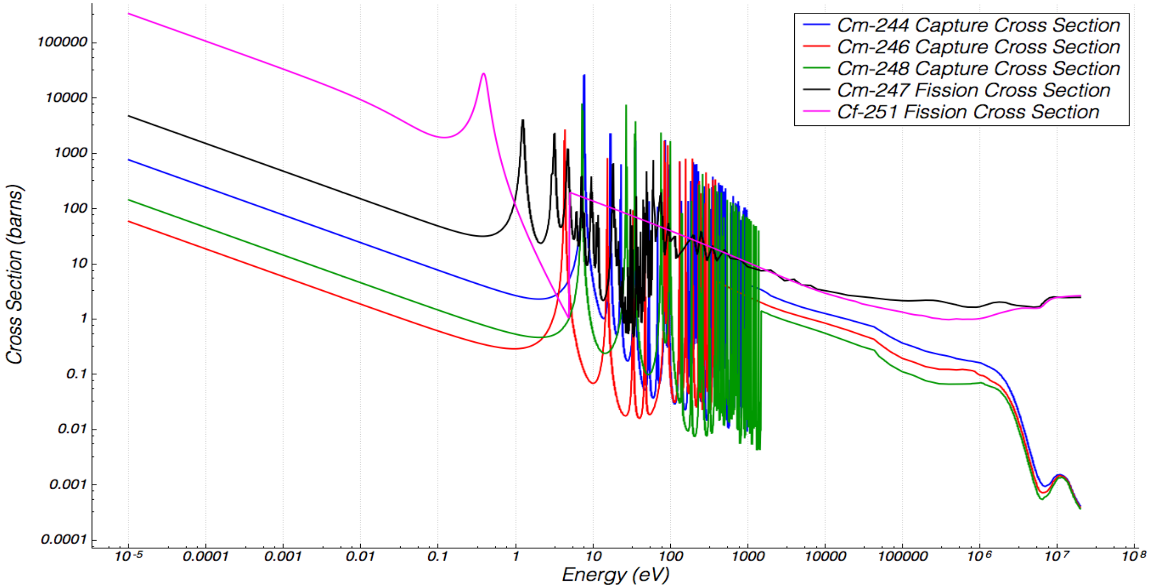




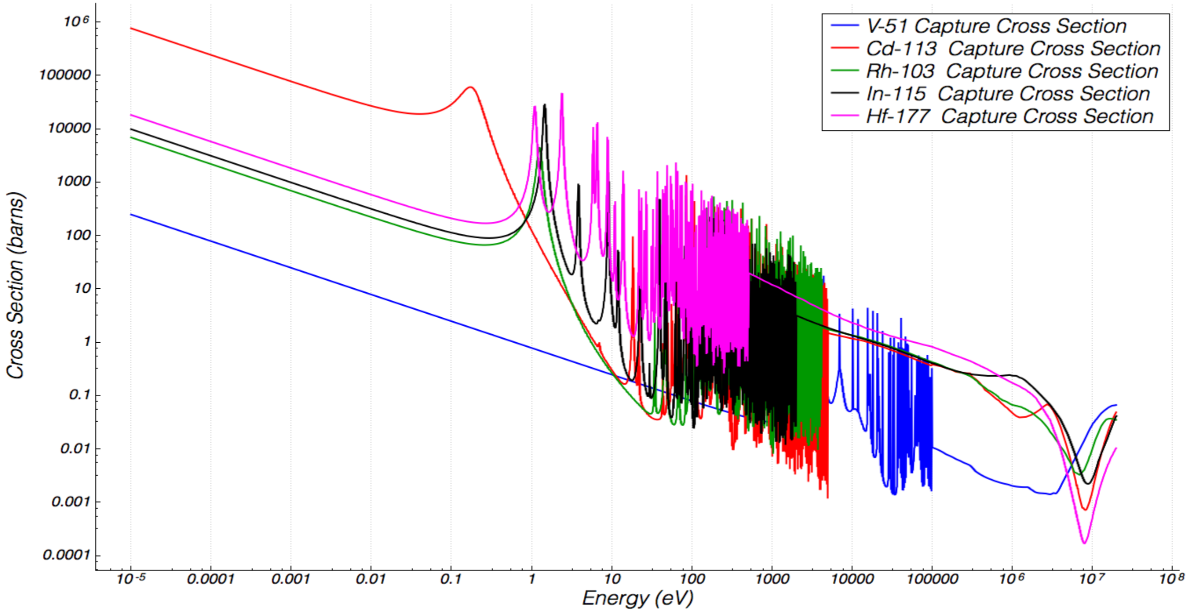
Neutron Filter Design

- Actinide fission and capture cross sections are not well-behaved…
- …and neither are filter material cross sections

Actinide fission and capture cross sections



Cross sections for candidate filter materials



- Selecting an optimal neutron filter is not simple or intuitive
- Current filter design relies on expert judgment or approximate methods

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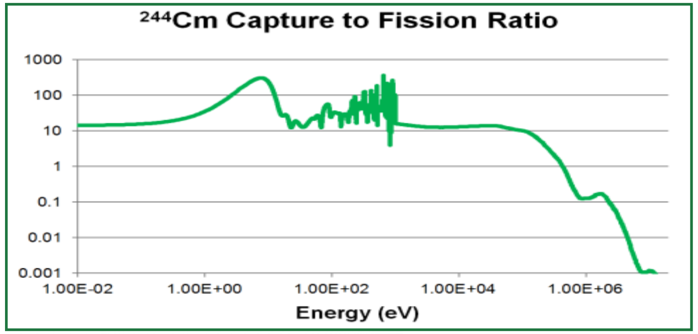
Emerging Sensitivity Methods

- Sensitivity coefficients (i.e., relative derivatives) predict how changing a design parameter will affect system responses

$$S_{R,\Sigma_x} = \frac{\delta R/R}{\delta \Sigma_x/\Sigma_x}$$



- Generalized Perturbation Theory (GPT) sensitivity analysis capabilities[1] in TSUNAMI-3D (in the SCALE code system) allow analysts to quantify how modifying the system parameters will affect reaction rate ratios (i.e., capture-to-fission ratios)
- This information enables designers to tweak system parameters that improve isotope target performance while mitigating undesired effects

[1] C. M. Perfetti, B. T. Rearden, "Development of a Generalized Perturbation Theory Method for Uncertainty and Sensitivity Analysis using Continuous-Energy Monte Carlo Methods," *Nucl. Sci. Eng.*, **182**, *3*, 354–368 (2016).

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Project Scope

- This project used sensitivity analysis to inform and optimize production of ^{252}Cf in HFIR isotope production targets
- Optimization parameters included:
  - the geometry of isotope production targets, and
  - the addition of a neutron filter foil around targets to block neutrons that are likely to induce fission in heavy actinides
- Before any optimization could be performed, the TSUNAMI-3D GPT sensitivity algorithms required adaption for use in high-performance computing (HPC) environments

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Presentation Outline

1. Introduction to ^{252}Cf Isotope Production
2. Adapting TSUNAMI-3D Sensitivity Algorithms to HPC
3. HFIR Irradiation Target Design Optimization
   a. Optimizing the Target Geometry
   b. Consideration of a Neutron Filter Foil
4. Results of Irradiation Target Design Optimization

Adapting GPT Algorithms for High-Performance Computing

- The TSUNAMI-3D GPT sensitivity algorithms must track tally data for a particle's history for several generations until it can determine the "importance" of the data
- These algorithms generate:
  1. a large amount of tally data for each particle history, and
  2. a small amount of data that describe the importance of the particle's fission chain
- Previously, this information was communicated with each particle history

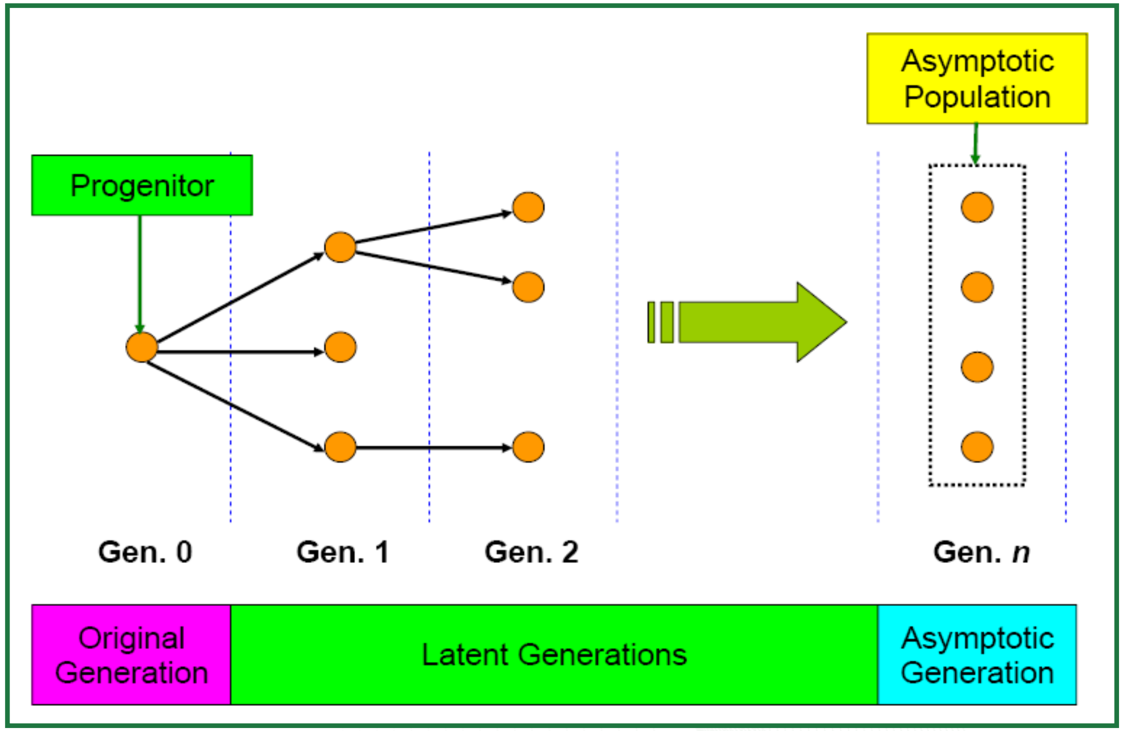
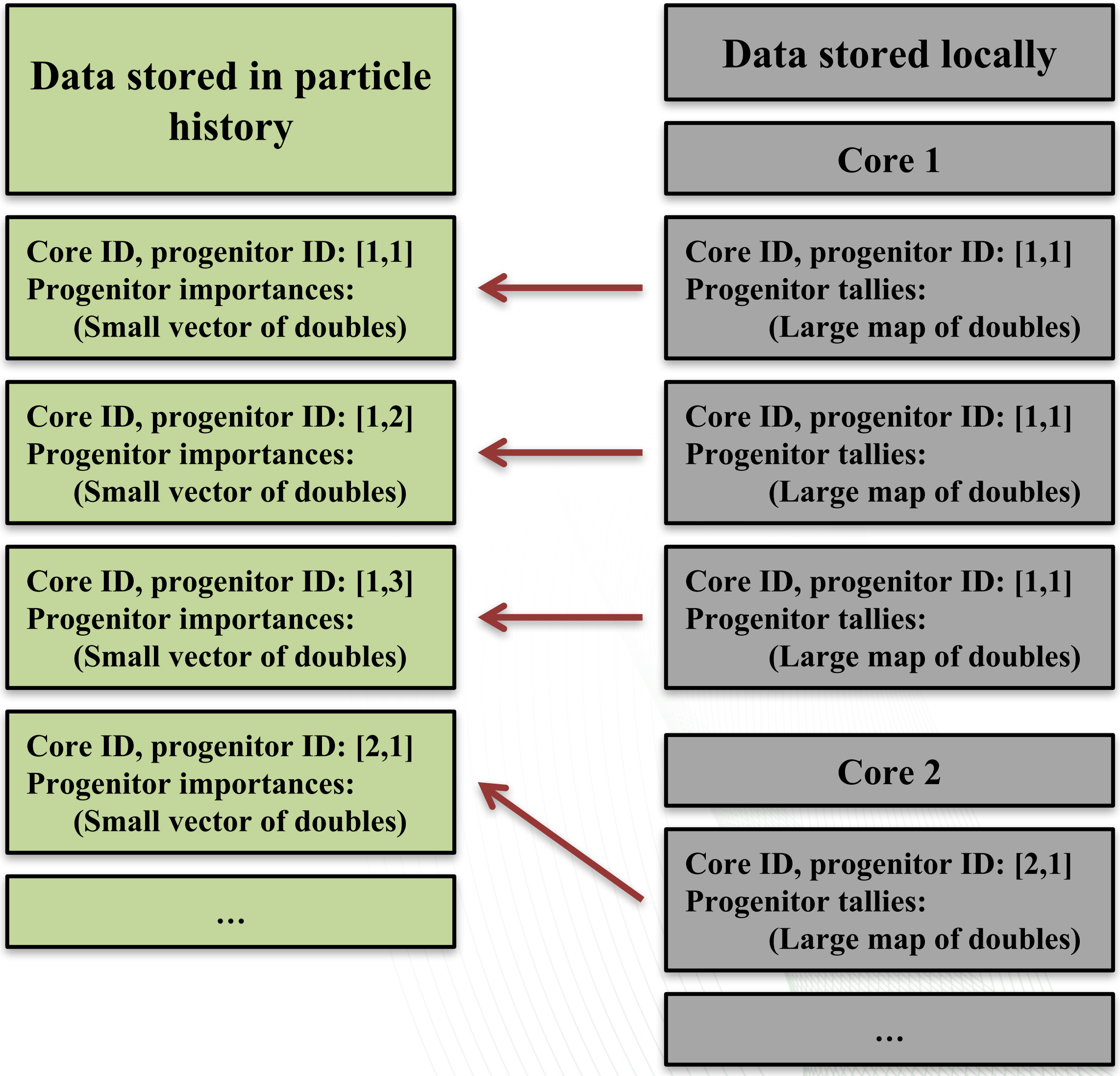


Illustration of the iterated fission probability process (image courtesy of Brian Kiedrowski)

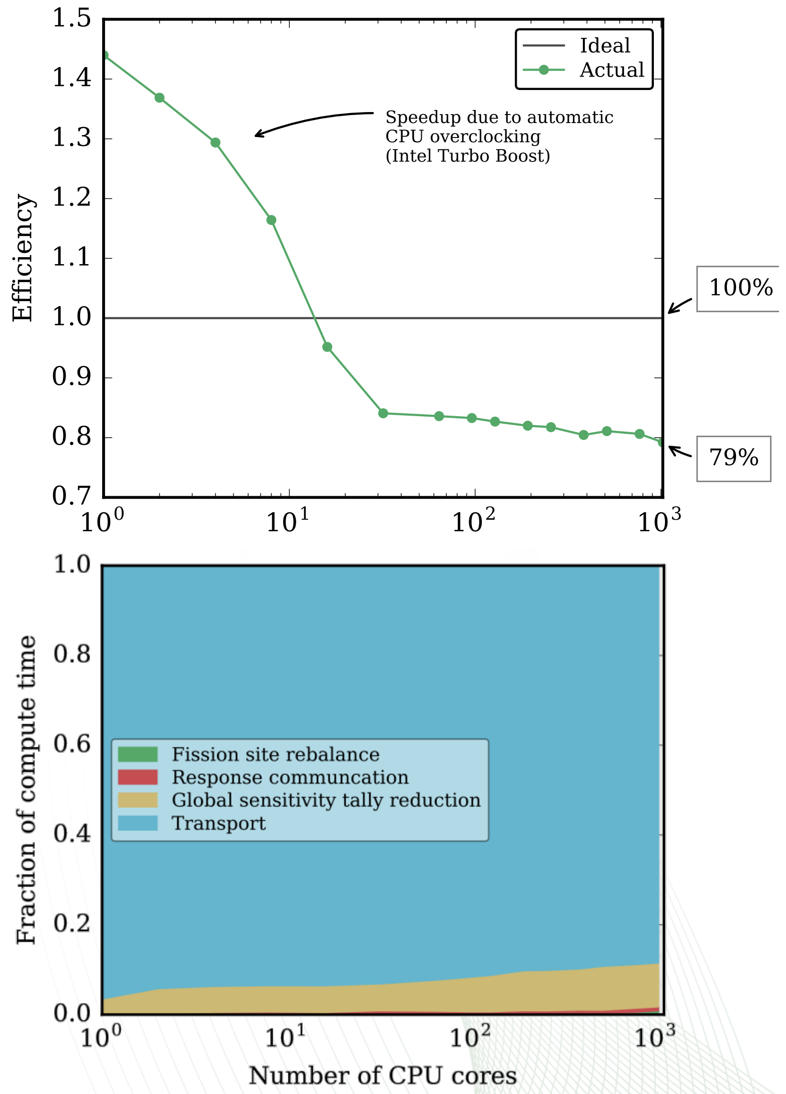
Adapting GPT Algorithms for High-Performance Computing

- The TSUNAMI-3D GPT algorithms have been modified so that:
  1. the large tally data are stored locally on the node where the particle was simulated, and
  2. only the small importance information is communicated with the particle history
- When the fission chain terminates, the importance information is broadcast to the node where the original particle's tally data were stored, where it can then bank sensitivity tallies for the particle.



Adapting GPT Algorithms for High-Performance Computing

- The changes to the GPT sensitivity algorithms allowed them to be used in high-performance computing environments with reasonable parallel efficiency (79% efficiency on 1,000 CPU cores)
- The sensitivity tally global sum operations (required for batch statistics) were responsible for the majority of the loss in parallel efficiency
- Work continues to further improve the parallel efficiency of the GPT sensitivity algorithms



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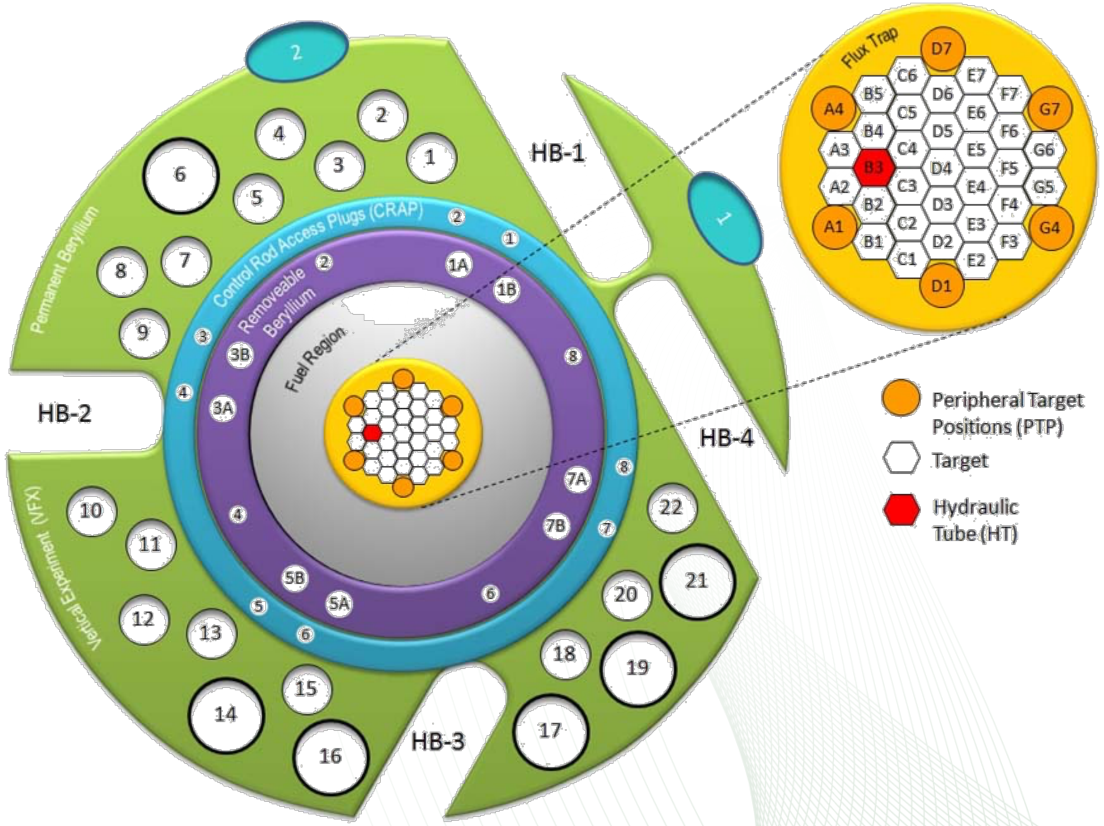
Isotope Production Target Geometry Optimization

- Only a limited number of changes to the ^{252}Cf isotope production target geometry are possible because the targets are located in HFIR's central flux trap
- Possible changes include:
  1. using an annular target,
  2. using a thin target, and
  3. adjusting the isotope density in the target



Isotope Production Target Geometry Optimization

- To determine the optimal geometry of the isotope production targets, sensitivity coefficients were computed for key actinide reaction rate ratios with respect to the density of the inner, middle, and outer regions of each isotope production target. For example, low or negative sensitivities in the inner target zones suggest moving to an annular design
- Relative importances were used to weight each reaction rate ratio based on its importance for the production or ^{252}Cf

Reaction rate ratio	Relative importance	Sensitivity coefficient	Net sensitivity effect
^{251}Cf capture / ^{252}Cf capture	14.84%	-2.44%	-0.36%

- These importances were determined empirically based on data from historic HFIR operations

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Isotope Production Target Geometric Sensitivities

Reaction rate ratio	Relative importance	Inner layer density sensitivity	Middle layer density sensitivity	Outer layer density sensitivity
^{244}Cm capture / fission	0.00%	-3.97%	-10.37%	-12.10%
^{245}Cm capture / fission	0.03%	-0.06%	-0.07%	-0.06%
^{246}Cm capture / fission	0.49%	-7.10%	-12.10%	-10.39%
^{247}Cm capture / fission	6.76%	-0.17%	-0.22%	-0.21%
^{248}Cm capture / fission	1.33%	-7.51%	-12.21%	-10.53%
^{249}Bk capture / fission	0.00%	-0.63%	-0.72%	-0.66%
^{250}Cf capture / fission	0.00%	-12.37%	-11.87%	-10.80%
^{251}Cf capture / fission	9.12%	-0.12%	-0.14%	-0.13%
^{244}Cm capture / ^{252}Cf capture	1.70%	-1.69%	-8.03%	-10.41%
^{245}Cm capture / ^{252}Cf capture	24.49%	-2.63%	-2.76%	-2.83%
^{246}Cm capture / ^{252}Cf capture	11.27%	-1.91%	-7.07%	-6.16%
^{248}Cm capture / ^{252}Cf capture	29.29%	-3.23%	-8.83%	-7.53%
^{251}Cf capture / ^{252}Cf capture	14.84%	-2.44%	-2.52%	-2.61%
Net sensitivity		**-2.35%**	**-4.82%**	**-4.37%**

Isotope Production Target Geometry Optimization

- Each heavy actinide reaction rate ratio was negative, suggesting that all design changes would improve the efficiency of ^{252}Cf production. These results suggest that targets are over-self-shielding neutron flux at energies where capture is likely
- Targets using annular, thin, or low density will therefore be considered in this study
  - The low density target uses 50% of the nominal heavy actinide density in an isotope production target
  - The annular and thin targets are each 50% of the volume of a nominal target
- Additional irradiation locations are available in the HFIR flux trap for the ORNL ^{252}Cf Isotope Production Program, so these design changes are feasible

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Incorporating Neutron Filter Foils

- Thin neutron-absorbing foils can be wrapped around isotope production targets to improve the efficiency of ^{252}Cf production by filtering neutrons likely to induce fission in heavy actinides
- Ideally, these foils will not filter neutrons at energies likely to be captured
- Candidate filter materials[2] included Rh, In, ^{176}Lu, and ^{149}Sm
- To determine which filter material would be optimal, isotope production targets were modeled with low density (20% of natural density) filter materials, and sensitivity coefficients were computed for key actinide reaction rate ratios with respect to the density of the filter material. Filter materials that produce large, positive sensitivity coefficients are ideal

[2] S. Hogle, "Optimization of Transcurium Isotope Production in the High Flux Isotope Reactor," Doctoral Dissertation, University of Tennessee, Knoxville (2012).

Neutron Filter Foils Sensitivity Coefficients

Reaction rate ratio	Relative importance	Sensitivity to Rh	Sensitivity to In	Sensitivity to ^{176}Lu	Sensitivity to ^{149}Sm
^{247}Cm C/F	6.76%	-1.06%	-1.16%	-0.11%	-0.23%
^{248}Cm C/F	1.33%	-0.99%	-1.83%	-0.76%	-1.46%
^{251}Cf C/F	9.12%	-0.62%	-1.08%	-0.75%	-5.31%
^{244}Cm capture / ^{252}Cf capture	1.70%	3.10%	3.45%	2.18%	2.59%
^{246}Cm capture / ^{252}Cf capture	24.49%	5.91%	7.51%	3.74%	5.19%
^{247}Cm capture / ^{252}Cf capture	11.27%	-6.60%	-7.93%	0.35%	0.36%
^{248}Cm capture / ^{252}Cf capture	29.29%	6.00%	7.89%	3.99%	5.20%
^{251}Cf capture / ^{252}Cf capture	14.84%	-11.25%	-16.51%	-6.19%	-26.92%
Net sensitivity		**0.61%**	**0.47%**	**1.10%**	**-1.88%**

Incorporating Neutron Filter Foils

- The calculated sensitivity coefficients suggest that Rh, In, and ^{176}Lu will improve the efficiency of ^{252}Cf production
- ^{149}Sm is predicted to decrease the efficiency of ^{252}Cf production
- Unlike geometric sensitivities, foil sensitivities are both positive and negative, suggesting that one must account for multiple competing effects when selecting an ideal neutron filter material

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Results of Irradiation Target Design Optimization

- HFIR models were simulated using the SCALE Triton-3D depletion tool to evaluate the efficacy of the proposed ^{252}Cf isotope production target design changes
- The Rh, In, and ^{176}Lu filtered designs were anticipated to improve the efficiency of ^{252}Cf production, as were each of the modified geometry designs
- Several metrics were used to evaluate the efficacy of the design changes:
  1. the overall yield of ^{252}Cf,
  2. the amount of "potential" ^{252}Cf produced in the target,
  3. the amount of "potential" ^{252}Cf destroyed in the target, and
  4. the "heavy actinide efficiency" of the irradiation, or the overall yield of ^{252}Cf (1) divided by the potential ^{252}Cf that was destroyed (3)

Determining the Potential ^{252}Cf in a Target

- The amount of potential ^{252}Cf present in an isotope production target was determined using conversion factors based on data from historic HFIR operations

Isotope	Potential californium factor
^{244}Cm	0.0010
^{245}Cm	0.0033
^{246}Cm	0.0141
^{247}Cm	0.0850
^{248}Cm	0.1800
^{249}Bk	0.3500
^{250}Cf	0.3500
^{251}Cf	0.3500
^{252}Cf	1.0000

1) Overall Yield of ^{252}Cf

- All target geometry changes increased the yield of ^{252}Cf
- All neutron filter foils significantly decreased the yield of ^{252}Cf

Filter material	Standard geometry	Annular target	Thin target	Low (50%) density target
Unfiltered target	Baseline	+3.82%	+2.29%	+12.98%
Lu filter	–25.19%	–23.66%	–24.43%	–15.27%
Rh filter	–39.69%	–37.40%	–56.49%	–30.53%
In filter	–45.80%	–43.51%	–44.27%	–37.40%
Sm filter	–58.02%	–56.49%	–58.02%	–51.15%

2) Potential ^{252}Cf Produced

- All design changes had a small but generally positive effect on the amount of potential ^{252}Cf produced
- The foils appear to slow ^{252}Cf transmutation

Filter material	Standard geometry	Annular target	Thin target	Low (50%) density target
Unfiltered target	Baseline	–0.04%	–0.86%	–0.04%
Lu filter	+0.69%	+0.64%	–0.17%	+0.64%
Rh filter	+1.55%	+1.50%	+1.72%	+1.67%
In Filter	+1.76%	+1.72%	+0.90%	+1.89%
Sm filter	+1.72%	+1.72%	+0.90%	+1.85%

3) Potential ^{252}Cf Destroyed

- The neutron filter foils significantly decreased loss of heavy actinides to fission

Filter material	Standard geometry	Annular target	Thin target	Low (50%) density target
Unfiltered target	Baseline	+2.13%	+0.00%	+0.00%
Lu filter	–34.04%	–31.91%	–34.04%	–31.91%
Rh filter	–76.60%	–74.47%	–85.11%	–82.98%
In filter	–89.36%	–85.11%	–87.23%	–95.74%
Sm filter	–87.23%	–85.11%	–87.23%	–93.62%

4) Potential ^{252}Cf Isotope Production Efficiency

- The overall effect is substantial increases to the efficiency of ^{252}Cf production

Filter material	Standard geometry	Annular target	Thin target	Low (50%) density target
Unfiltered target	Baseline	+0.9%	+1.3%	+10.5%
Lu filter	+11.3%	+10.5%	+11.5%	+24.5%
Rh filter	+157.2%	+147.4%	+181.1%	+328.8%
In filter	+375.9%	+273.3%	+319.4%	+1,312.5%
Sm filter	+208.0%	+181.1%	+229.1%	+570.1%

Results and Conclusions

- All proposed geometry changes improved the yield and efficiency of ^{252}Cf production
- The low (50%) density design was especially effective, resulting in a 12.98% increase in the ^{252}Cf yield
- The filtered designs all produced less overall ^{252}Cf yield, but they significantly decreased the amount of potential ^{252}Cf destroyed
  - Overall effect of filters was a significant increase in the efficiency of ^{252}Cf production
  - The maximum efficiency increase observed was 1,312%, occurring in the low density Indium-filtered target

Results and Conclusions

- The optimal design depends on the priorities of the ^{252}Cf isotope production program
  - ➢ Lower density targets should be used if there is space in the HFIR flux trap for additional isotope production targets
  - ➢ Filtered targets should be used if conserving the consumption of heavy curium feedstock is a higher priority than maximizing the production of ^{252}Cf

Results and Conclusions

- The sensitivity coefficients were effective at predicting the effect of the geometry changes, but they were not effective at predicting the effect of incorporating a neutron filter foil material
  - ^{176}Lu was predicted to be the most effective foil material, but Indium was in fact more effective
  - ^{149}Sm was predicted to lower the efficiency of ^{252}Cf production, but it was the second most effective filter material
- Reasons for these discrepancies include (1) imperfect ^{252}Cf relative importance values and (2) the possibility that calculated sensitivities change during the transmutation of ^{252}Cf; developing a depletion perturbation theory capability would mitigate both concerns

Thank you for your attention!

Please forward any questions to Chris Perfetti at perfetticm@ornl.gov