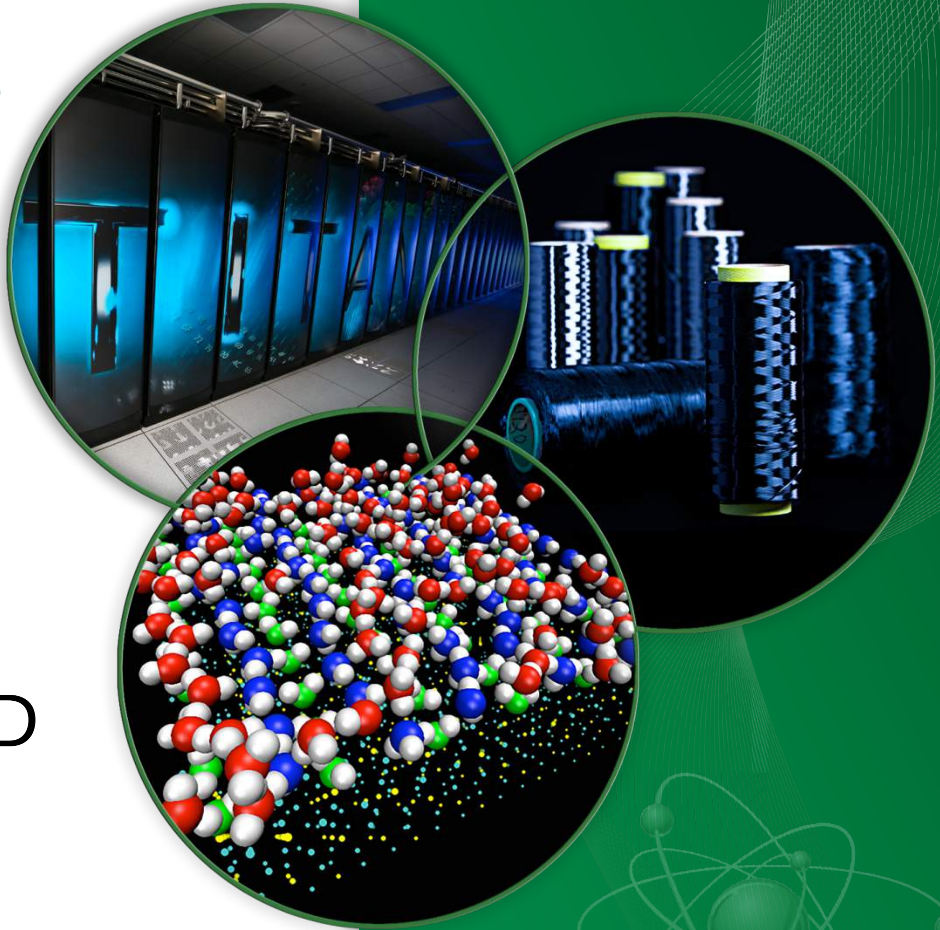
Using High-Fidelity Sensitivity and Uncertainty Analysis to Optimize Isotope Production and Improve Modeling & Simulation Capabilities in HFIR



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Presentation Outline

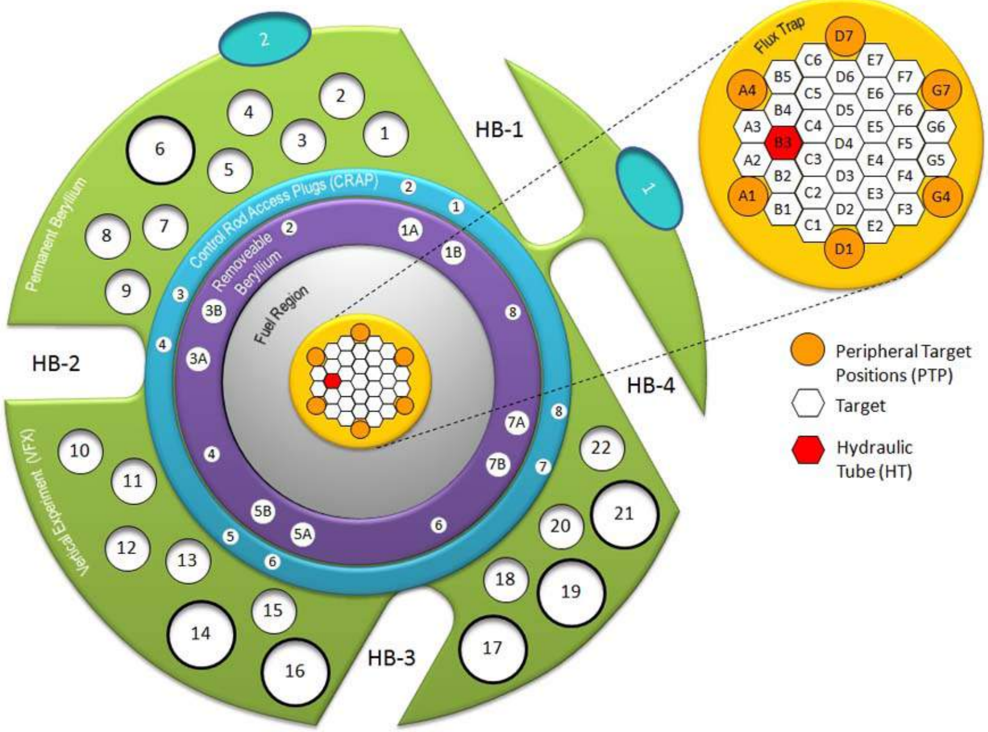
- **Part 1 - Introducing the Problem:**
  Optimizing ^{252}Cf Production in HFIR

- **Part 2 - Current Work:**
  Reaction Rate Sensitivity and Uncertainty Analysis

- **Part 3: Future Opportunities:**
  Data Assimilation for Irradiation Experiments

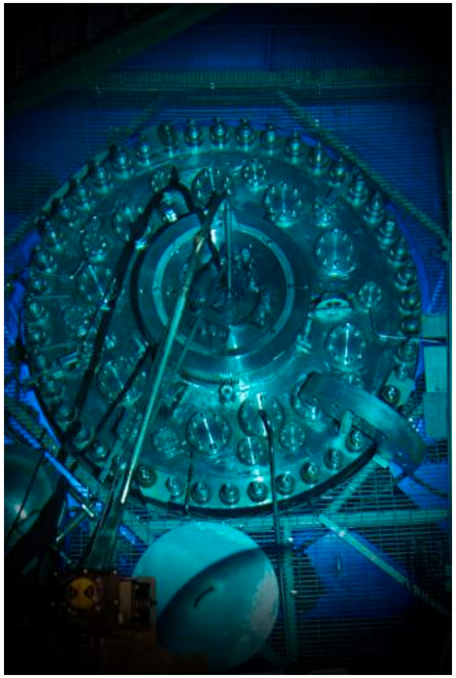
Part 1 – Introducing the Problem: Optimizing ^{252}Cf Production in HFIR

HFIR provides one of the highest neutron fluxes of any research reactor in the world. Thirty-seven (37) target positions in the flux trap and 42 positions in the beryllium reflectors provide locations for material irradiation experiments (MIE) and isotope production (IP) targets.



Isotope Production Opportunities

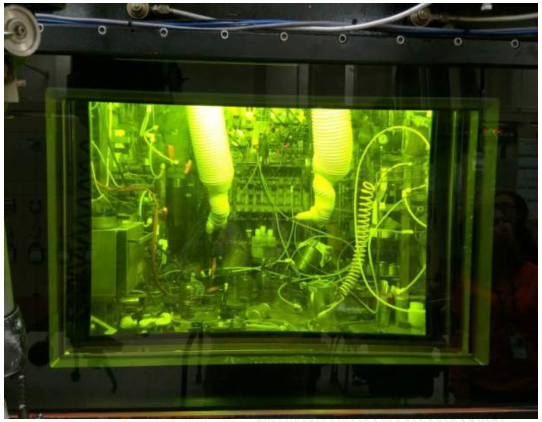
- HFIR can provide unique isotopes, some of which have no alternative U.S. production source, including:
  - ^{14}C useful in medical applications such as studying diabetes, gout, anemia, and acromegaly
  - ^{63}Ni explosives detection, airport security
  - ^{229}Th provides ^{225}Ac for α-particle cancer therapy
  - ^{238}Pu radioisotope power systems for space exploration
  - ^{254}Es production of super-heavy elements
  - ^{252}Cf source of neutrons for nuclear reactor startup and study of materials with neutron diffraction and neutron spectroscopy



Problem:

Our ability to produce these and other isotopes is threatened by many factors, including:

- Limits in the supply of heavy actinide feedstock,
- Availability of irradiation locations in HFIR,
- Heat generation in irradiation targets, and
- Disruption of neutron scattering beamline operations.



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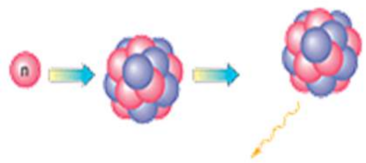
The Challenge

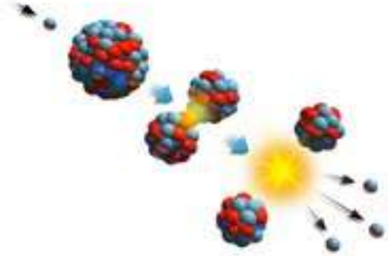
- The potential exists to improve isotope production at HFIR by using recent breakthroughs in sensitivity analysis to better understand:
  - The physical phenomena that lead to the production and destruction of specific isotopes,
  - The sources of uncertainty in design-limiting target heat generation safety calculations, and
  - The design and selection of irradiation experiments to improve the predictive capability of isotope transmutation and optimization calculations.

Opportunities to Improve Isotope Production

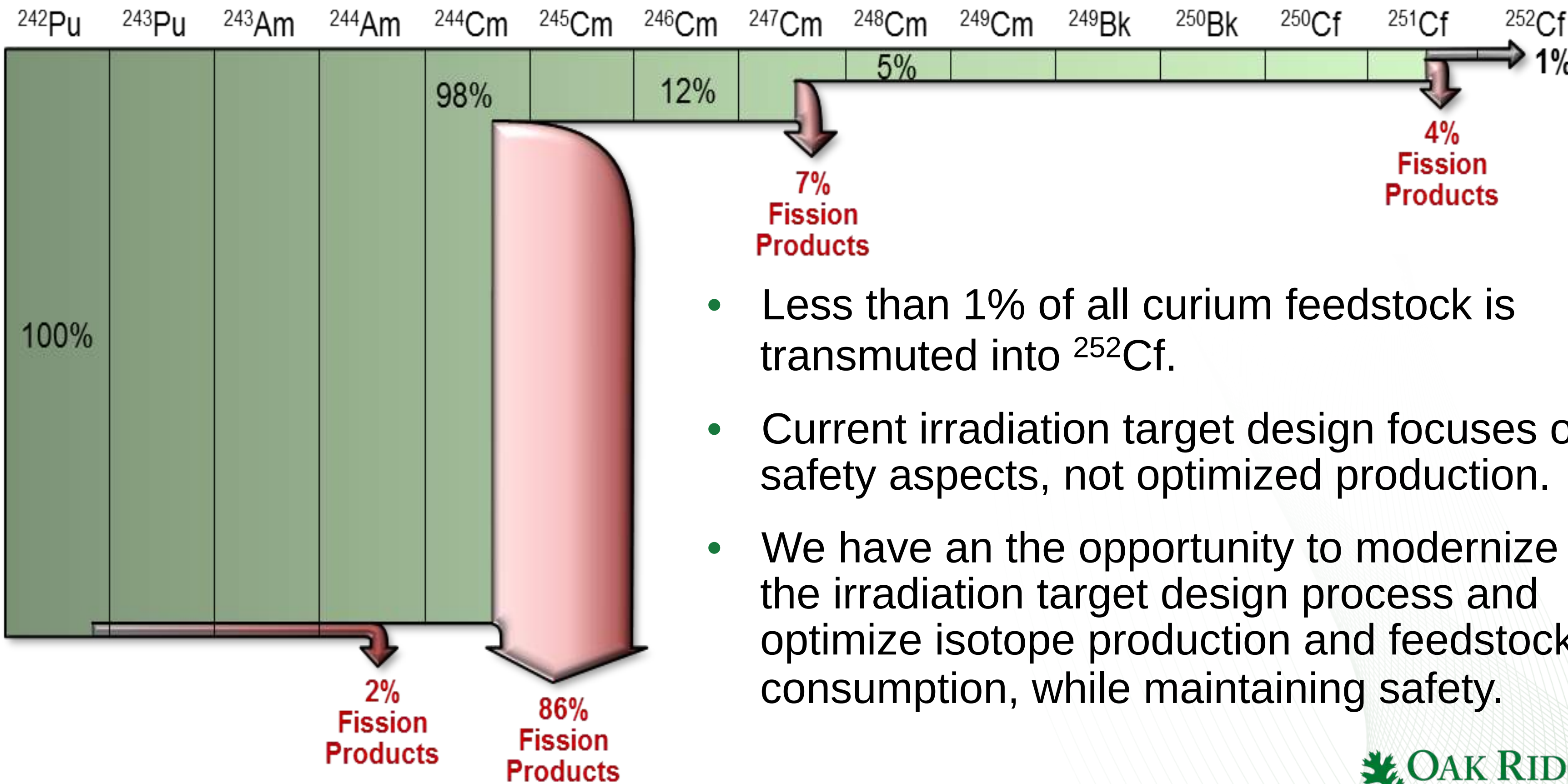
Neutron capture:
Produces heavier isotopes



Nuclear fission:
Destroys heavy isotopes

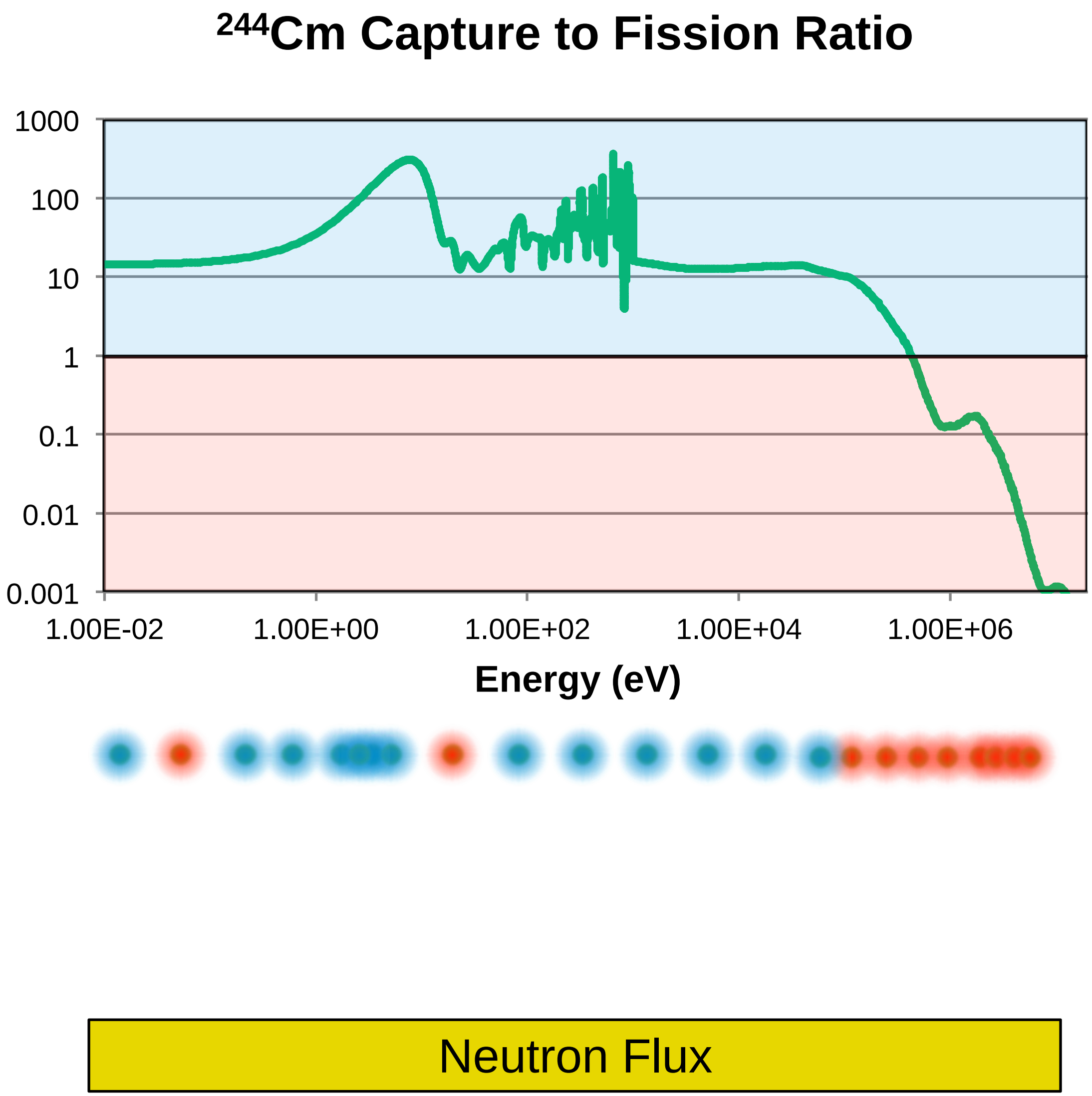


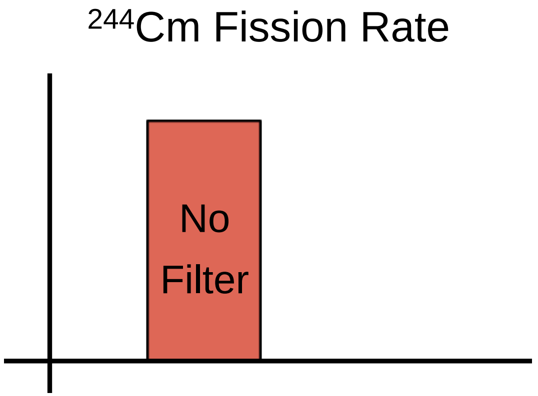
The Long Road to ^{252}Cf

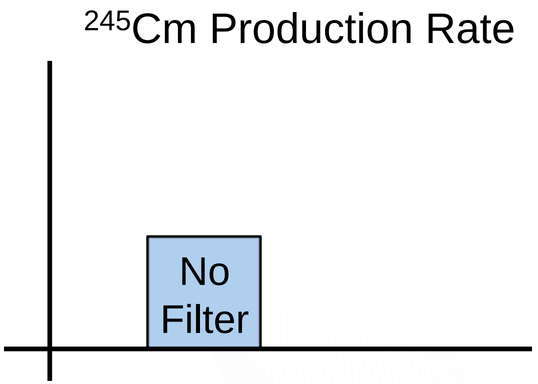


- Less than 1% of all curium feedstock is transmuted into ^{252}Cf.
- Current irradiation target design focuses on safety aspects, not optimized production.
- We have an the opportunity to modernize the irradiation target design process and optimize isotope production and feedstock consumption, while maintaining safety.

Neutron Filter Design





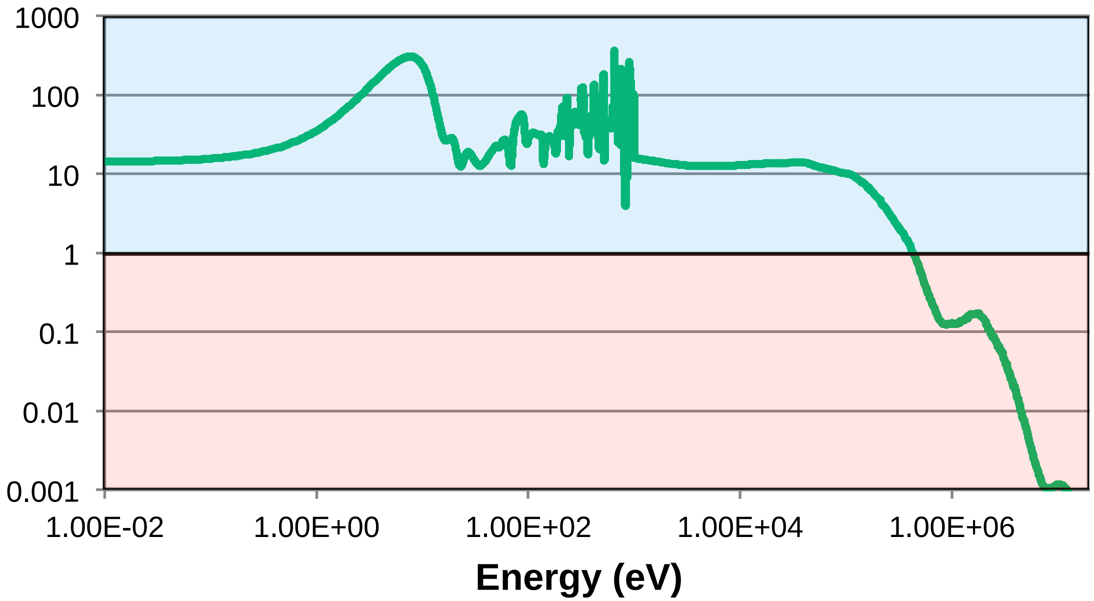


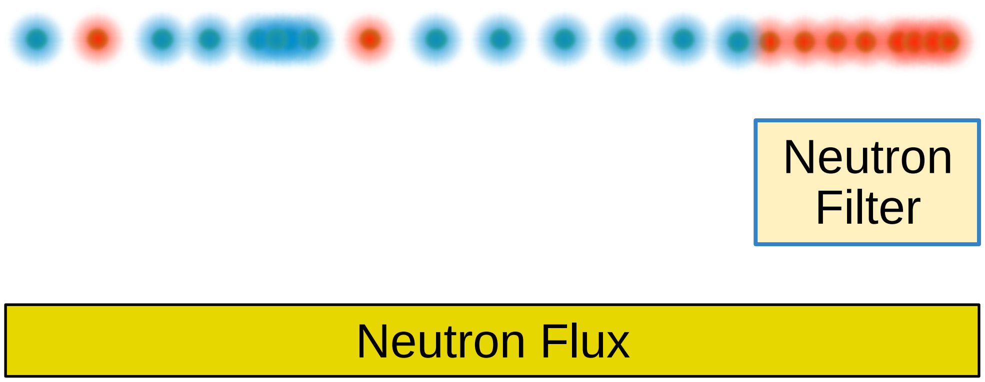
Fission Reaction

Capture Reaction

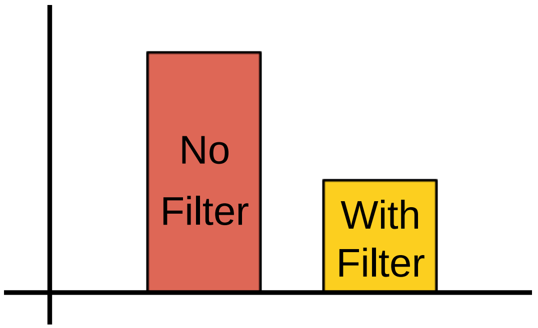
Neutron Filter Design

^{244}Cm Capture to Fission Ratio

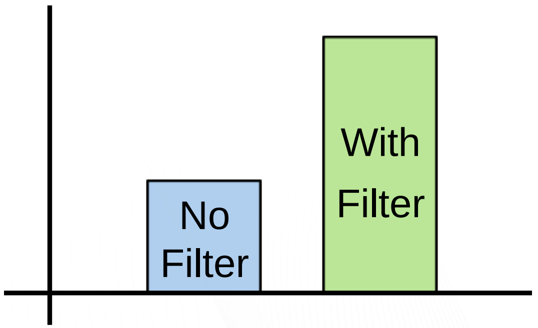




^{244}Cm Fission Rate



^{245}Cm Production Rate



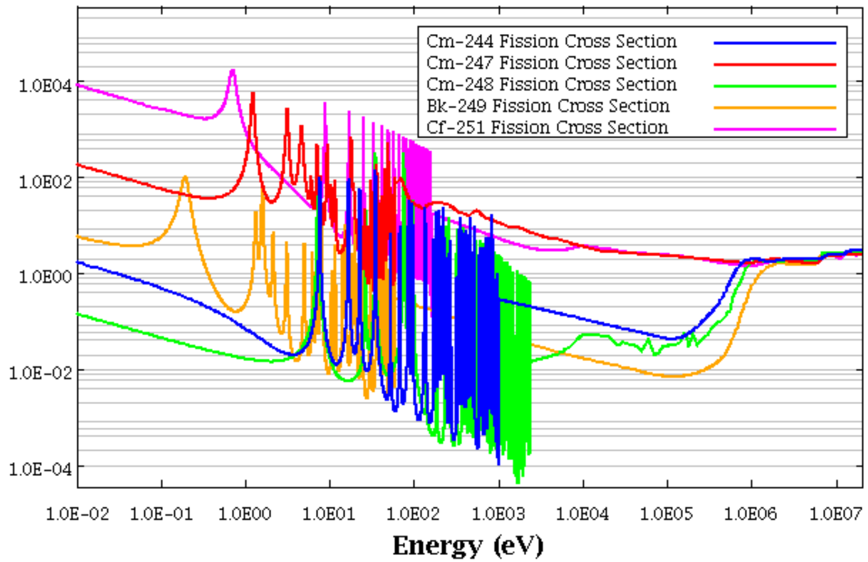
Fission Reaction

Capture Reaction

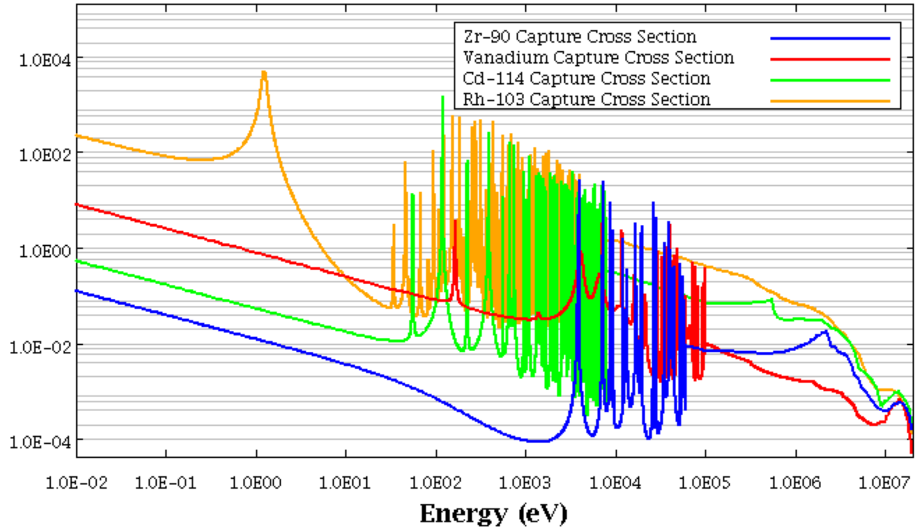
Neutron Filter Design

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

Actinide Fission Cross Sections



Nuclear Cross Sections for Candidate Filter Materials



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

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Part 2 – Current Work:
Reaction Rate Sensitivity Calculations

- Sensitivity coefficients describe the fractional change in a response that is due to perturbations, or uncertainties, in system parameters.

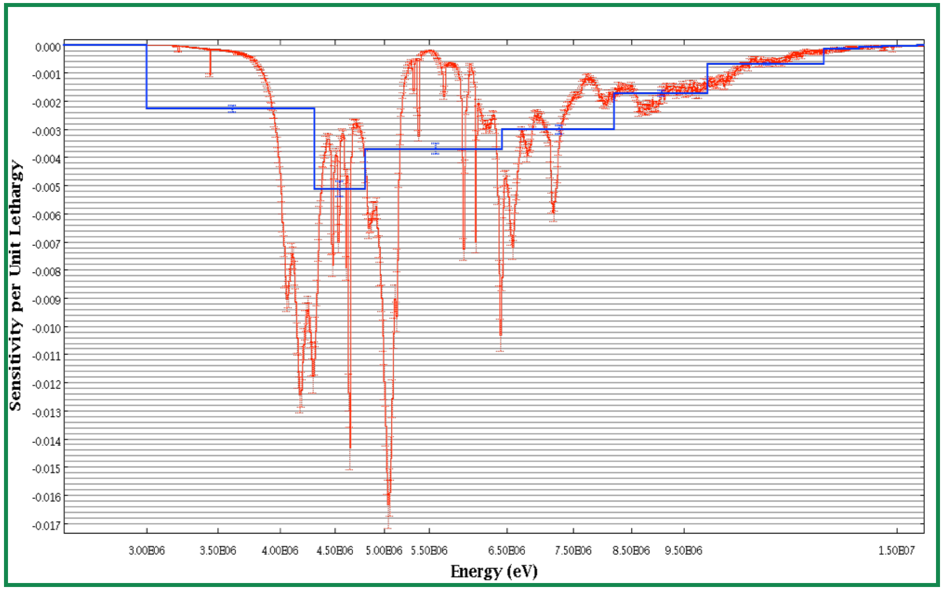
$$S_{k,\Sigma} = \frac{\partial k / k}{\partial \Sigma / \Sigma}$$

- The SCALE code contains a suite of eigenvalue (k_{eff}) sensitivity and uncertainty analysis tools using the TSUNAMI code, which has proven indispensable for numerous application and design studies for nuclear criticality safety and reactor physics.

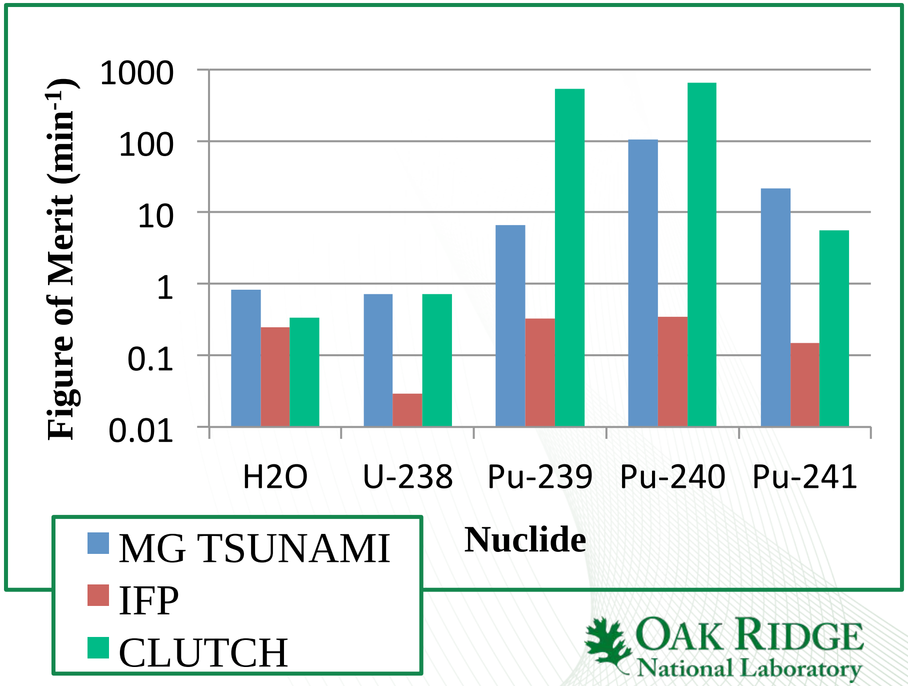
Part 2 – Current Work: Reaction Rate Sensitivity Calculations

- In SCALE6.2 the multigroup TSUNAMI-3D code has been extended to perform continuous-energy (CE) sensitivity coefficient calculations.
    - ➢ This work involved the development of the CLUTCH sensitivity method, a new and efficient approach for calculating eigenvalue sensitivity coefficients.

O-16 Capture Sensitivity
238-group VS
Microgroup CLUTCH



MIX-COMP-THERM-004-001
FoM Comparison



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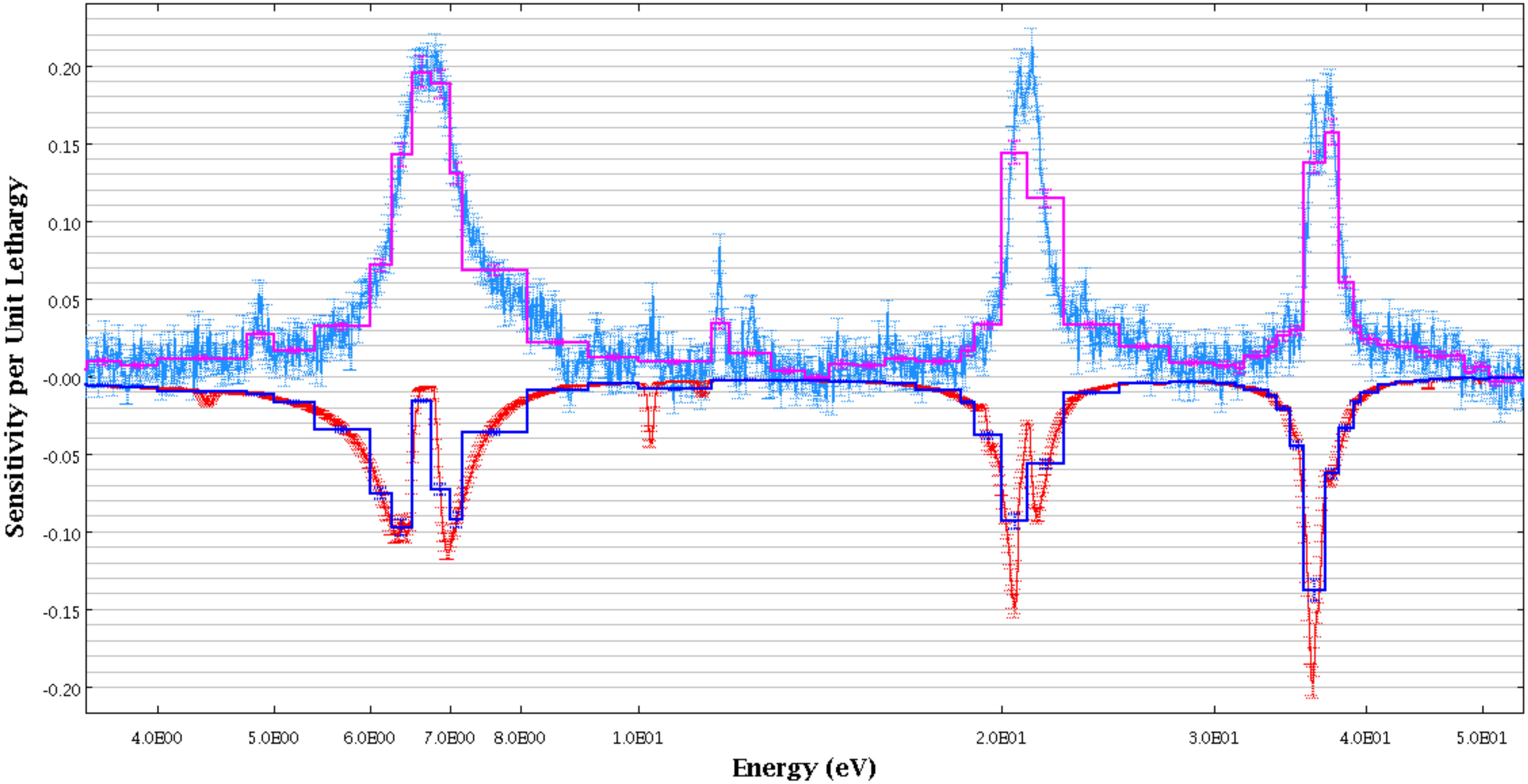
H-1 Elastic Scatter Sensitivity
238-group CLUTCH VS
Microgroup CLUTCH

U-238 Capture Sensitivity
238-group CLUTCH VS
Microgroup CLUTCH



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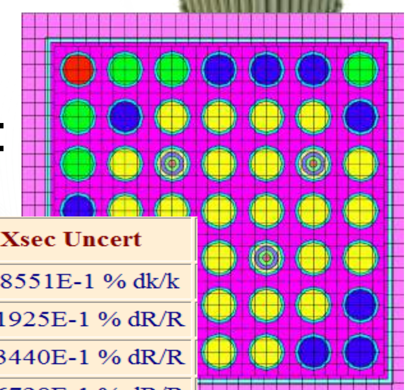
Generalized Perturbation Theory

- Recent developments by Perfetti and Rearden have enabled the calculation of generalized response sensitivity coefficients using high-fidelity, continuous-energy Monte Carlo methods.

- Generalized Perturbation Theory (GPT) calculates sensitivity coefficients for any system response that can be expressed as the ratio of reaction rates.

$$R = \frac{\langle \Sigma_1 \phi \rangle}{\langle \Sigma_2 \phi \rangle}$$



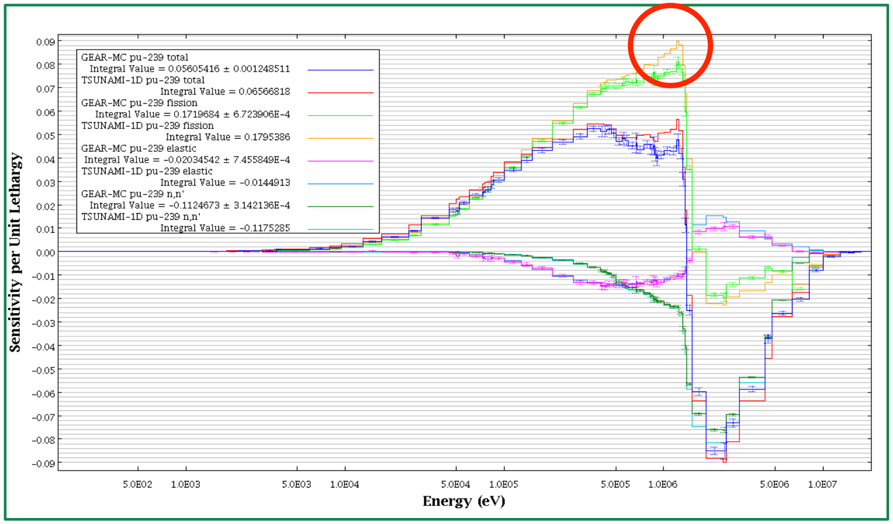
- Applications for GPT sensitivity/uncertainty analysis include:
  - Relative powers
  - Isotope Conversion Ratios
  - Multigroup Cross Sections
  - Experimental Parameters

NUMBER	EXPERIMENT	Type	Format	Value	Xsec Uncert
1	k_infinity	keff	Relative	1.1083E+0	4.98551E-1 % dk/k
2	fission_grp_1	gpt	Relative	1.9155E-3	6.91925E-1 % dR/R
3	fission_grp_2	gpt	Relative	2.7748E-2	3.23440E-1 % dR/R
4	absorpt_grp_1	gpt	Relative	7.1637E-3	8.36728E-1 % dR/R
5	absorpt_grp_2	gpt	Relative	5.3702E-2	2.38082E-1 % dR/R
6	cornerrod_fpf	gpt	Relative	1.1458E+0	1.67147E-1 % dR/R

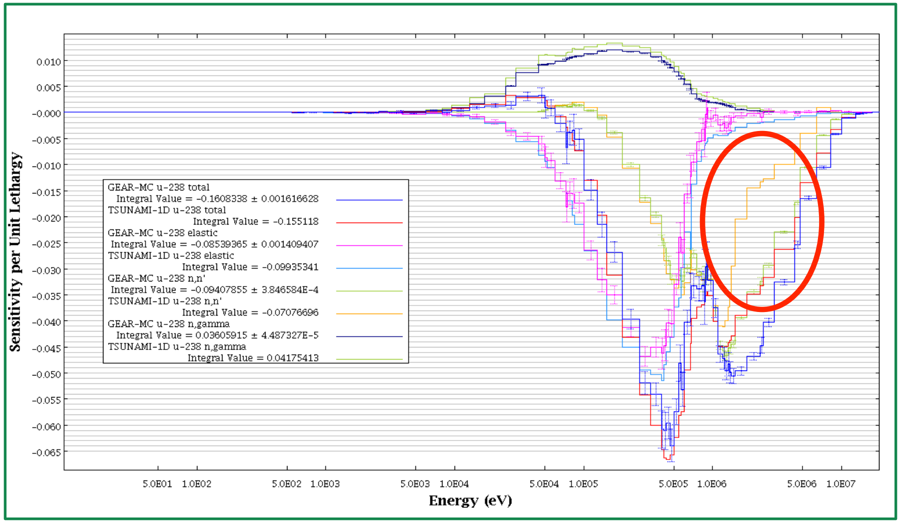
OECD UAM GPT Benchmark Phase 1-2 Results

GPT Flattop Foil Response Sensitivity Coefficients

F28/F25 Pu-239 Sensitivity Coefficients



F37/F25 U-238 Sensitivity Coefficients



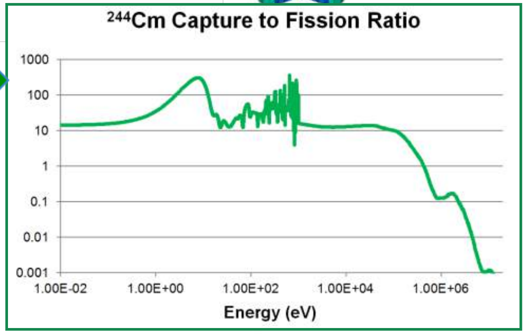
Flattop Total Nuclide Foil Response Sensitivities

Experiment	Response	Isotope	Reference	TSUNAMI-1D	GEAR-MC
Flattop	F28 / F25	U-238	0.8006 ± 0.0533	0.8024 **(0.03 σ)**	0.7954 ± 0.0018 **(-0.10 σ)**
		Pu-239	0.0528 ± 0.0043	0.0657 **(2.99 σ)**	0.0561 ± 0.0012 **(0.73 σ)**
	F37 / F25	U-238	-0.1540 ± 0.0102	-0.1551 **(-0.11 σ)**	-0.1608 ± 0.0016 **(-0.66 σ)**
		Pu-239	0.0543 ± 0.0048	0.0736 **(3.99 σ)**	0.0489 ± 0.0010 **(-1.10 σ)**

Emerging Sensitivity Methods

- Sensitivity coefficients (i.e. derivatives) describe how changing system parameters affects system responses.

$$\text{Sensitivity} = \frac{\delta(\text{System Response})}{\delta(\text{System Parameter})}$$

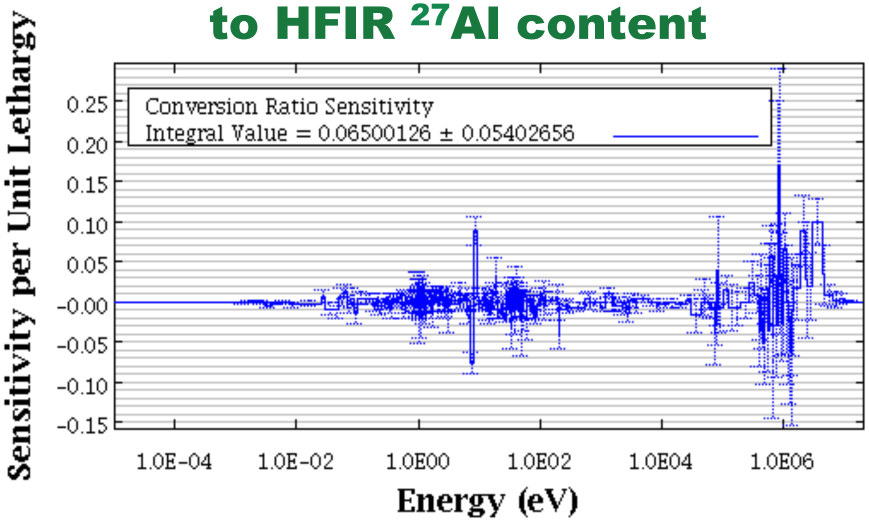


- These recent breakthroughs in sensitivity analysis allow us to quantify how modifying system parameters will affect capture-to-fission ratios and heat generation rates in irradiation targets.
- This information enables system designers to tweak the parameters that improve target performance while reducing the effect of undesired features.

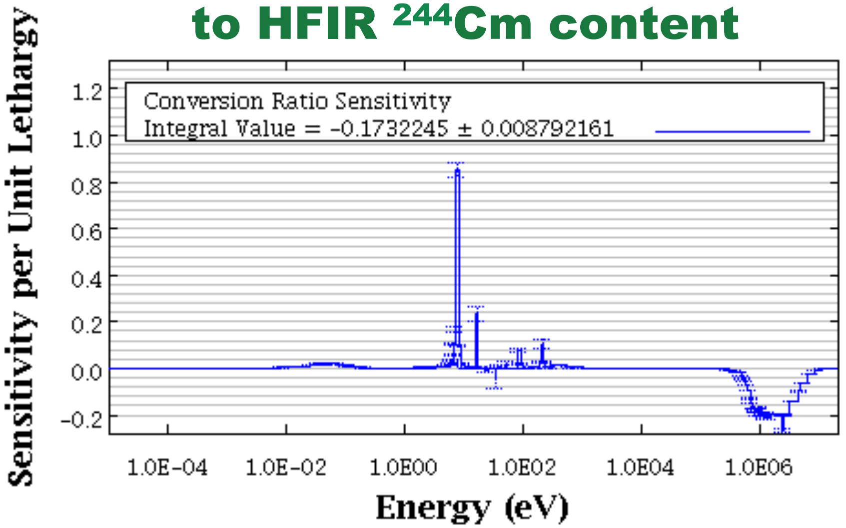
Emerging Sensitivity Methods

- These breakthroughs enable the calculation of the sensitivity of these responses to every nuclear reaction in the system in ONE calculation.

Sensitivity of the ^{244}Cm Capture-to-Fission Ratio to HFIR ^{27}Al content



Sensitivity of the ^{244}Cm Capture-to-Fission Ratio to HFIR ^{244}Cm content



- For the first time ever, these methods allow for sensitivity calculations using high-fidelity Monte Carlo M&S tools.

- These breakthroughs enable a sensitivity analysis that scales much better than previous sensitivity methods, allowing for analysis of complex systems, such as HFIR.

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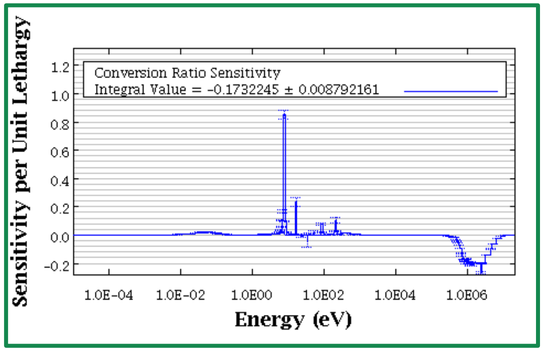
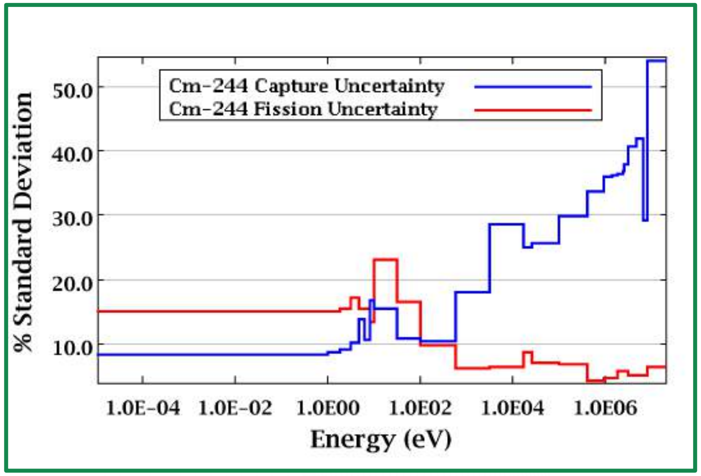
Propagation of Cross-Section Uncertainties

- Cross-section uncertainties can be propagated to quantify the uncertainty in k_{eff} and reaction rate tallies using the "Sandwich Equation."

Sensitivity Coefficients



Cross-Section Covariance Data



$$\sigma = \sqrt{S C_{\alpha\alpha} S^{T}}$$

…where:

- S is a vector of all energy-dependent sensitivity data for all nuclides and reactions; and
- $C_{\alpha\alpha}$ is a matrix containing energy-dependent cross section covariance data evaluated for all nuclides and reactions, α.

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Using Uncertainty Quantification to Identify Nuclear Data Needs

- Uncertainty quantification is useful for identifying areas where nuclear data improvements could improve simulation accuracy.

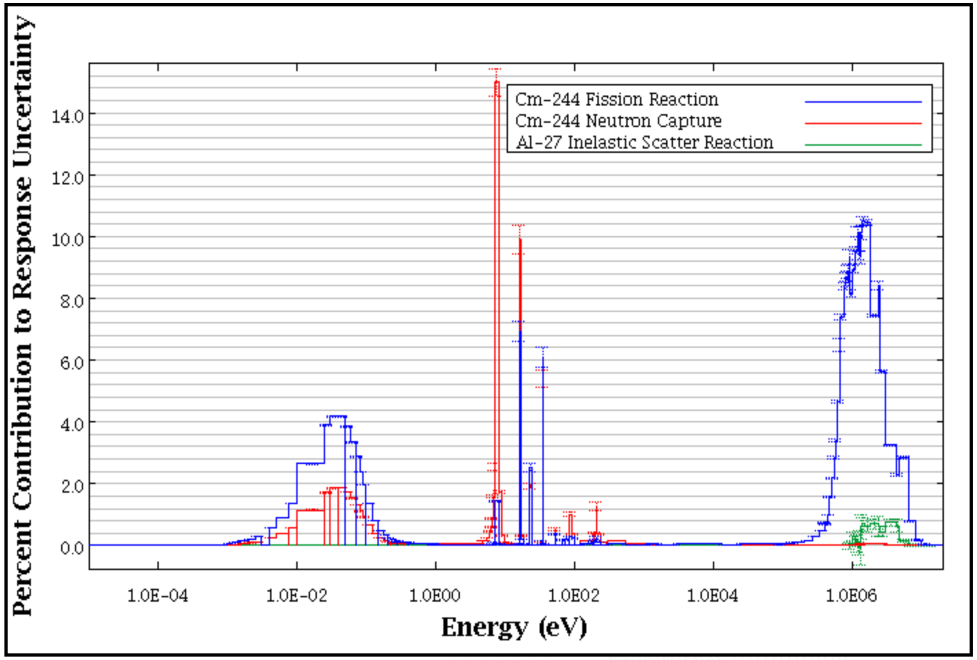
- Optimization efforts are meaningless if computational models cannot accurately predict isotope production due to low-fidelity nuclear data evaluations.

Data-Induced Uncertainty in Curium Capture-to-Fission Ratios in HFIR ^{252}Cf Production Targets	
Curium Isotope	**Uncertainty**
^{244}Cm	**18.33%** ± 0.02%
^{245}Cm	**5.38%** ± 0.02%
^{246}Cm	**14.80%** ± 0.24%
^{247}Cm	**21.08%** ± 0.08%
^{248}Cm	**20.40%** ± 0.57%

Using Uncertainty Quantification to Identify Nuclear Data Needs

- Sensitivity-based uncertainty analyses offer insight on which reactions and neutron energies contribute the most uncertainty to responses of interest.

Reaction Contributions to the Uncertainty in the ^{244}Cm Conversion Ratio	
^{244}Cm Fission Reaction	17.62%
^{244}Cm Neutron Capture	4.96%
^{27}Al Inelastic Scatter Reaction	0.72%
^{244}Cm Elastic Scatter Reaction	0.59%
^{1}H Elastic Scatter Reaction	0.56%
Total Data-Induced Uncertainty	**18.33%**



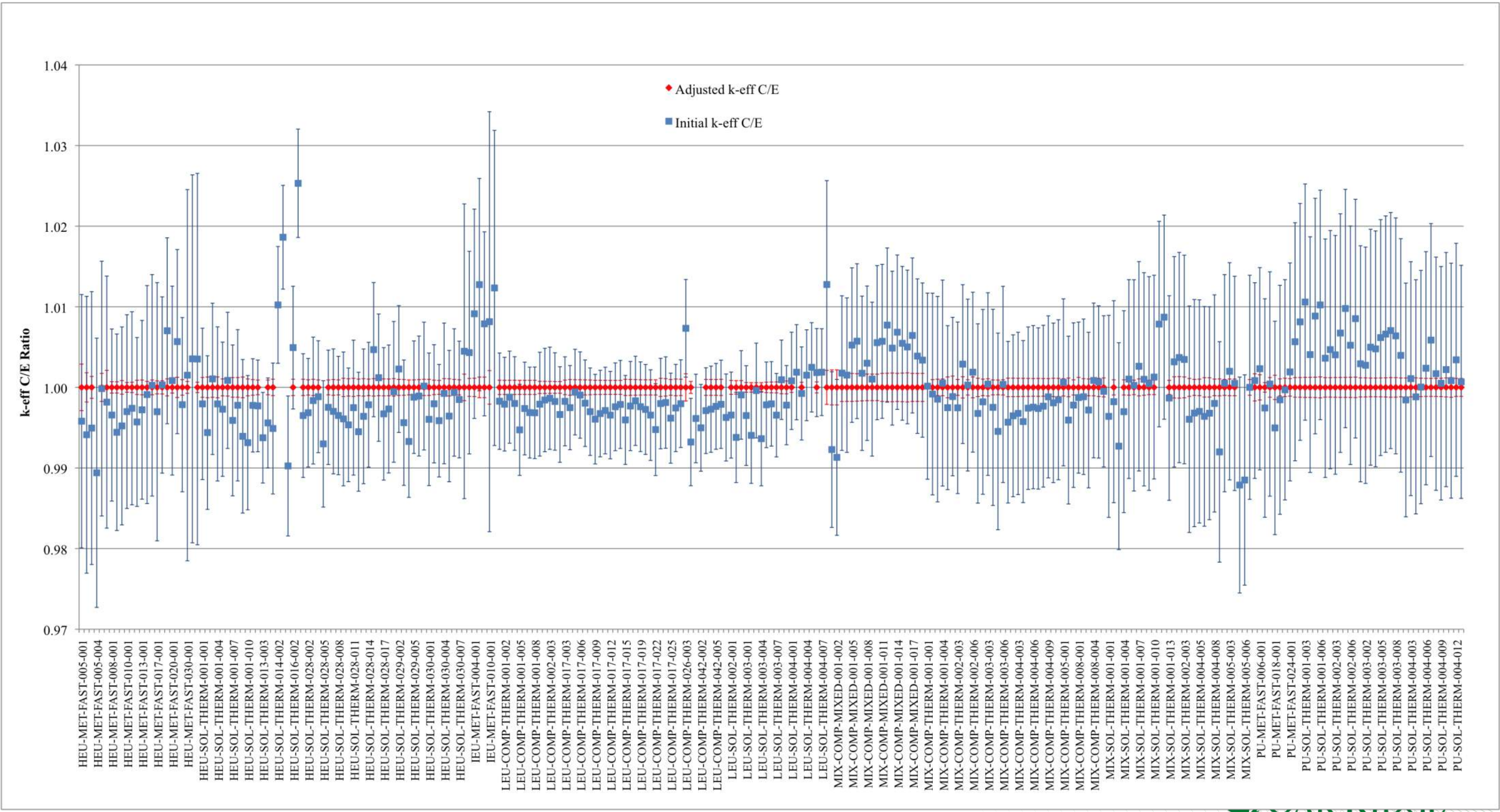
Energy-dependent contributions to the uncertainty in the ^{244}Cm conversion ratio.

Part 3 – Future Opportunities: TSURFER Tools for Data Adjustment and Experimental Data Assimilation

- ***TSURFER:*** ***T***ool for ***S***/***U*** analysis of ***R***esponse ***F***unctionals using ***E***xperimental ***R***esults
  - Biases are observed as differences between benchmark and computed quantities (k_{eff}, reaction rates, etc.)
  - TSURFER adjusts the nuclear data to reconcile biases between integral experiment results and computational predictions.
  - By taking into account the uncertainties and correlations between nuclear data and integral experiments, a consistent set of data can be formed that eliminates biases for the benchmarks, within a known uncertainty.
  - Where the cross sections and covariance data are modified, the modifications can be used to project biases from the benchmarks to a bias and bias uncertainty for targeted application systems.

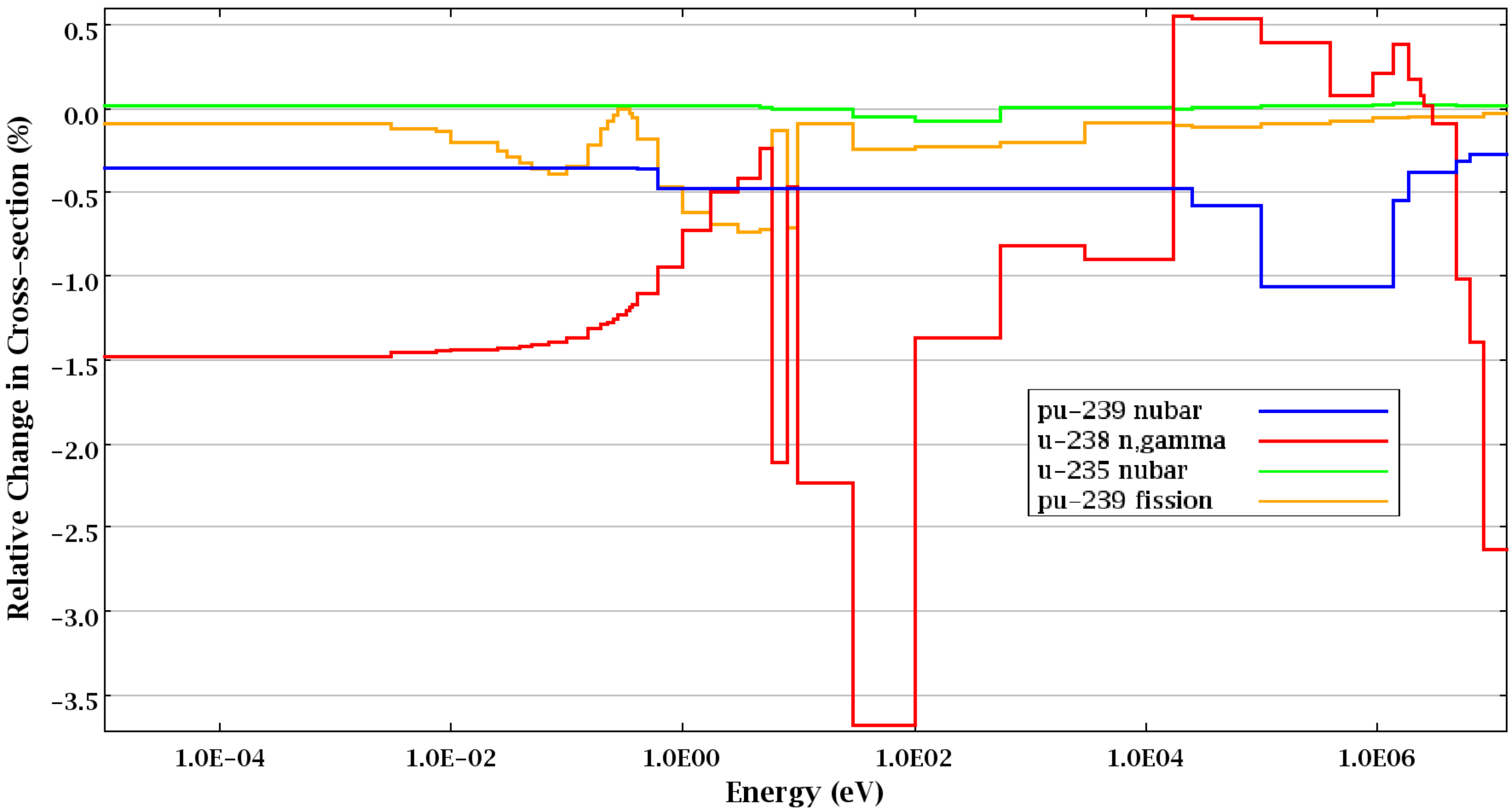
Data Adjustment Techniques:

Experimental benchmark data (E) is used to improve the accuracy of the initial computed responses (C).



Cross Section Adjustments to Minimize Bias:

A consistent set of data adjustments is produced to minimize biases in all integral experiments. Reduced cross section uncertainties are also produced.

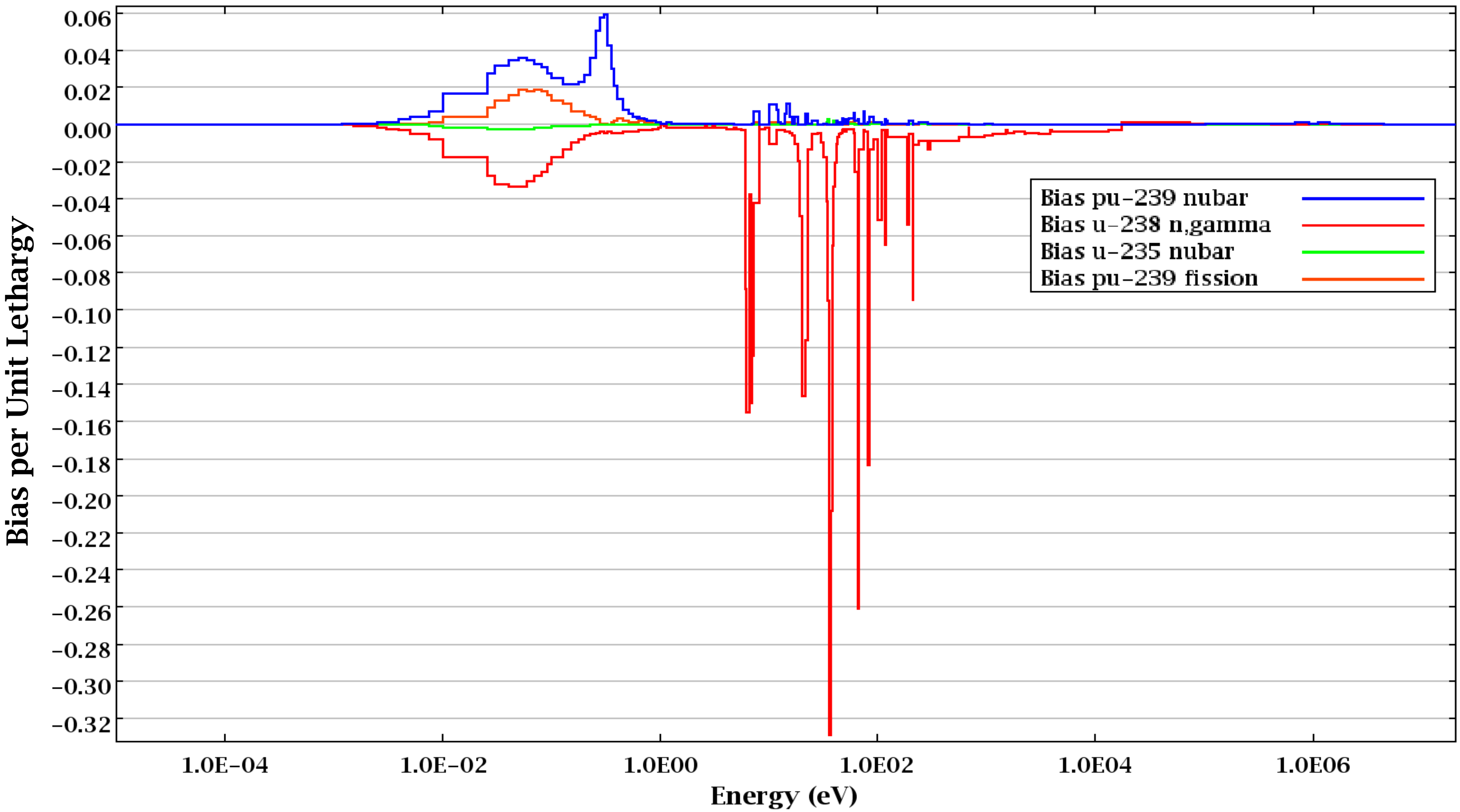


Adjusted Cross Sections Reduce Data-Induced Biases

- Original Uncertainty is:
  0.520% Δk/k
- Adjusted Uncertainty is:
  0.119% Δk/k
- Interpretation: ~80% of uncertainty is quantified through validation with experiments.
- Remaining uncertainty highlights gaps in available validation data.

NUCLIDE	REACTION	CONTRIBUTION TO BIAS % dk/k
u-238	n,gamma	-2.1084E-01
pu-239	nubar	1.2761E-01
pu-239	fission	3.9872E-02
o-16	elastic	3.2243E-02
pu-239	n,gamma	-2.5810E-02
pu-239	chi	1.0248E-02
u-235	chi	2.9940E-04
fe-56	n,gamma	1.7158E-02
u-235	fission	-1.2351E-02
pu-240	n,gamma	-1.3162E-02
u-238	elastic	2.7715E-03
u-235	n,gamma	1.0599E-03
h-1	elastic	2.7348E-03
u-238	n,n'	-6.8963E-03
u-235	nubar	-4.1298E-03
fe-56	elastic	-6.0079E-03
h-1	n,gamma	4.1893E-03
u-238	nubar	3.1408E-03

Energy-dependent Bias is Produced for each Nuclide and Reaction



Future Work: Extending Data Assimilation to Isotope Production Applications

- Valuable experimental data exists for transcurium irradiation experiments, but current methods CANNOT assimilate this data and gain knowledge on nuclear data biases, knowledge gaps, etc.
- A tool performing **sensitivity analysis for the isotopics in depletion and transmutation calculations** is needed to enable this kind of uncertainty analysis.

$$\frac{\delta(Reaction\ Rates)}{\delta\Sigma} \times \frac{\delta(Isotopics)}{\delta(Reaction\ Rates)} = \frac{\delta(Isotopics)}{\delta\Sigma}$$

- These new CE TSUNAMI-3D capabilities for performing **generalized reaction rate sensitivity calculations** have the potential to be combined with ongoing work by Mark Williams for **developing an adjoint version of ORIGEN** to produce such a **transmutation sensitivity tool**.

One Slide to Rule Them All: Presentation Summary

- Actinide production applications are sensitive to low-fidelity nuclear data because they involve extremely rare isotopes and difficult-to-measure reactions.
  Need: Accurate heavy actinide data
- Recent developments allow us to calculate the sensitivity/ uncertainty of isotope production rates to nuclear data parameters using high-fidelity Monte Carlo methods.
  Need: Accurate covariance estimates
- Improved capabilities for sensitivity analysis and integral data assimilation would allow data scientists to better understand the sources of bias/disagreement in isotope production applications, and allocate our efforts to most effectively improve the fidelity of nuclear data.
  Need: Isotope number density sensitivity capability

Questions???

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