Integral Measurements of Independent and Cumulative Fission Product Yields Supporting Nuclear Forensics and Other Applications

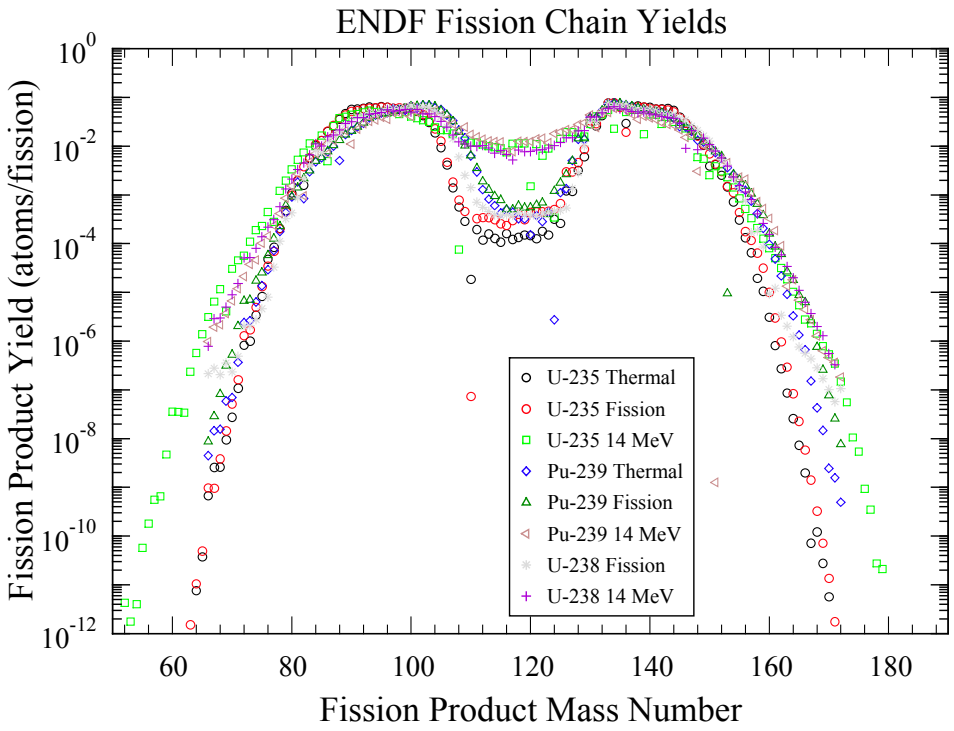
Todd Bredeweg
Los Alamos National Laboratory

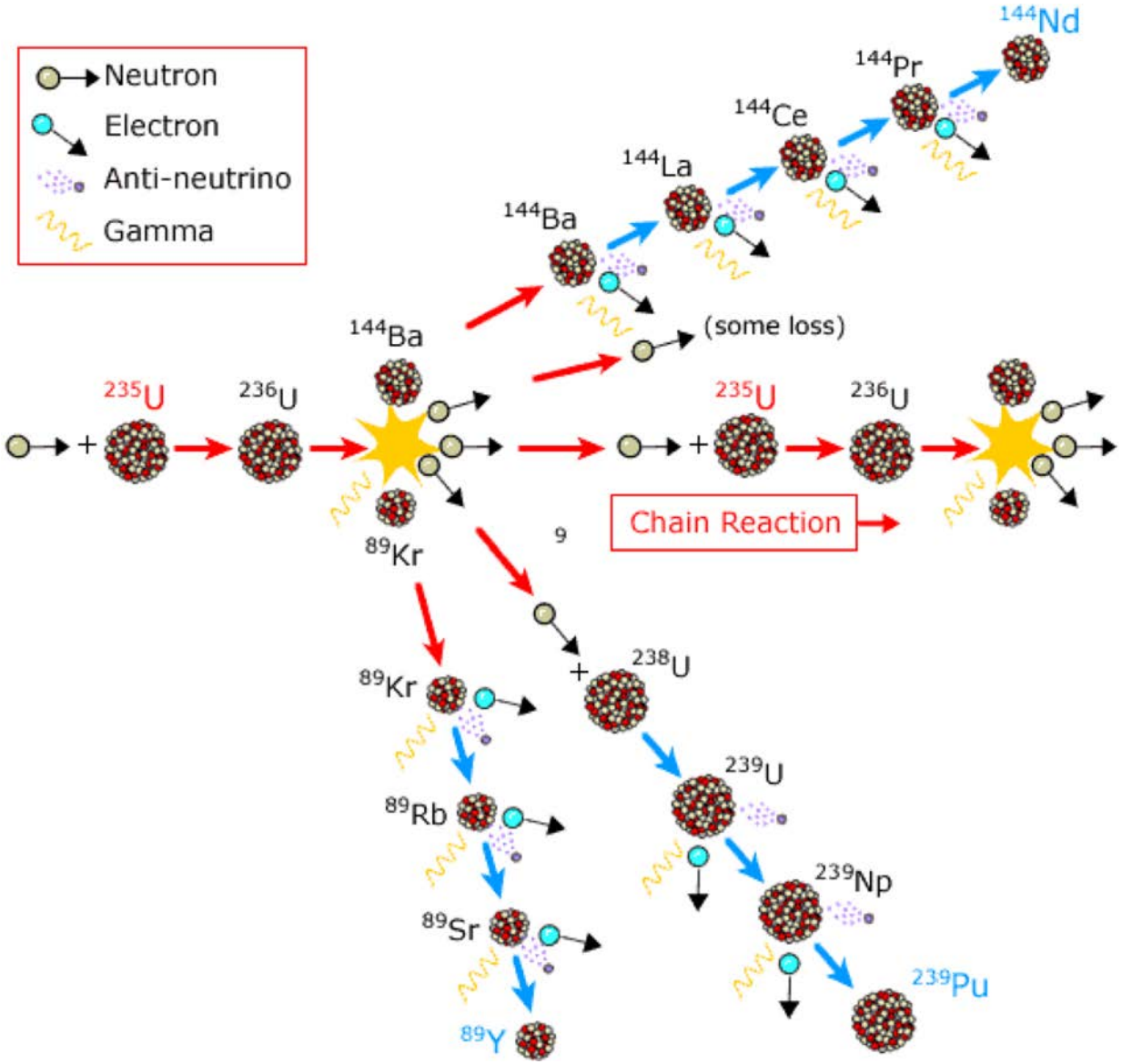
5 March 2020

Why is nuclear physics / nuclear data important to the mission

The defining signature of a nuclear detonation is the presence of nuclear reaction products in the debris. These can be from

- Fission products (FP) from the fuel
- Activation products (AP) from the fuel or nearby materials
- Production/Destruction depends on fuel type and neutron spectrum





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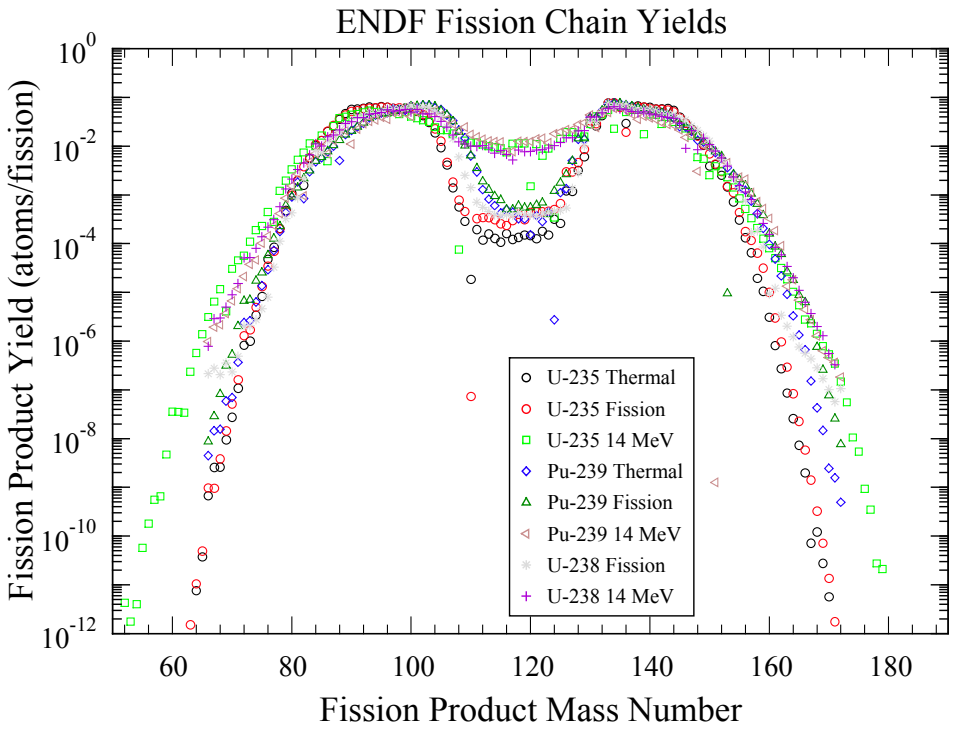

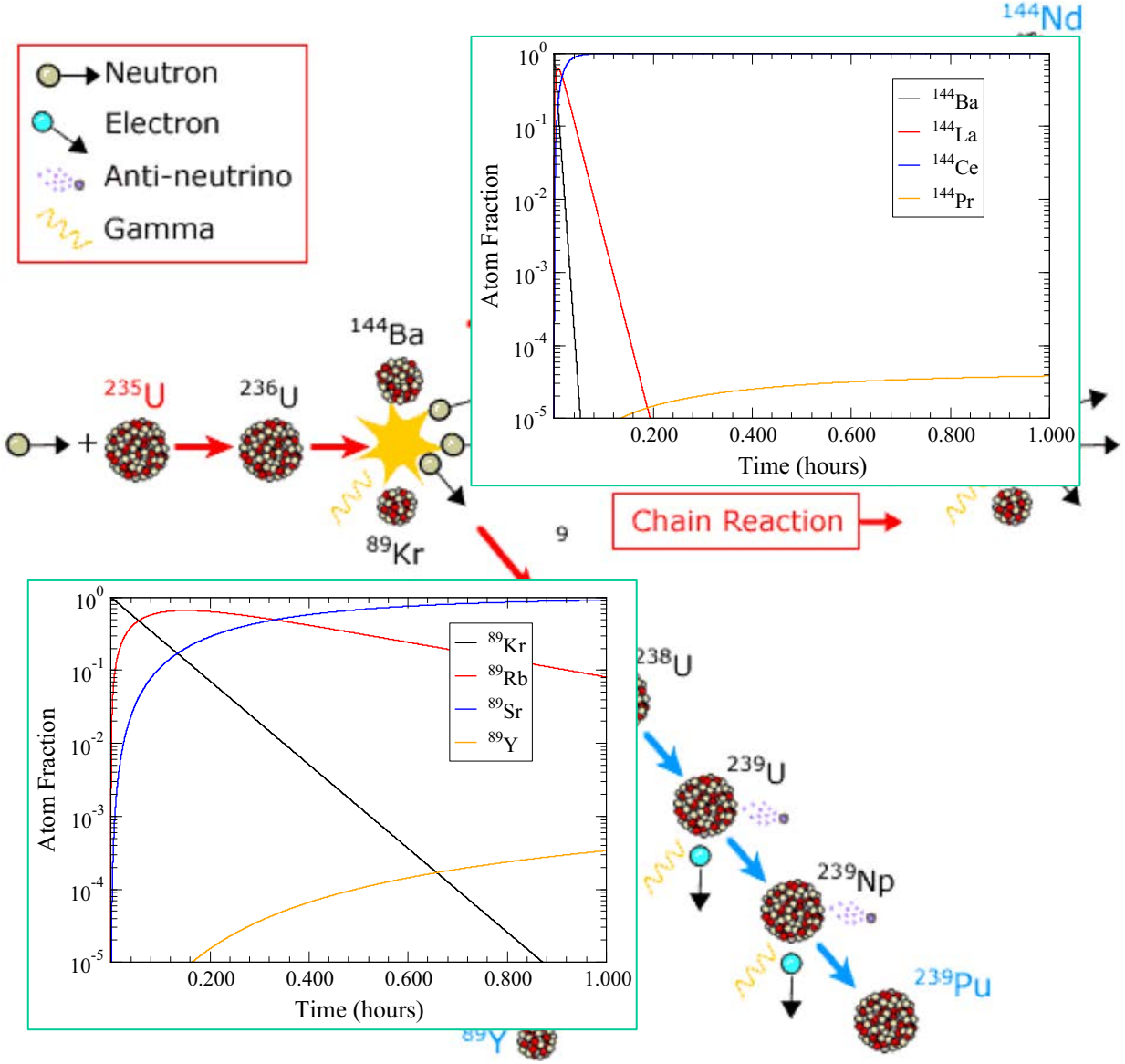

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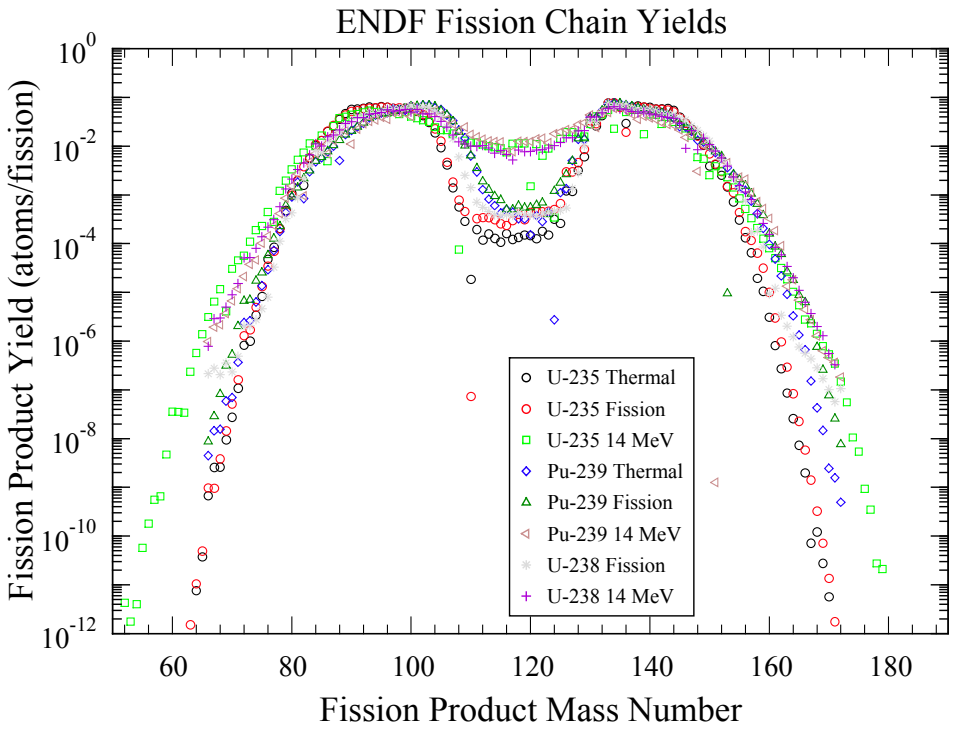

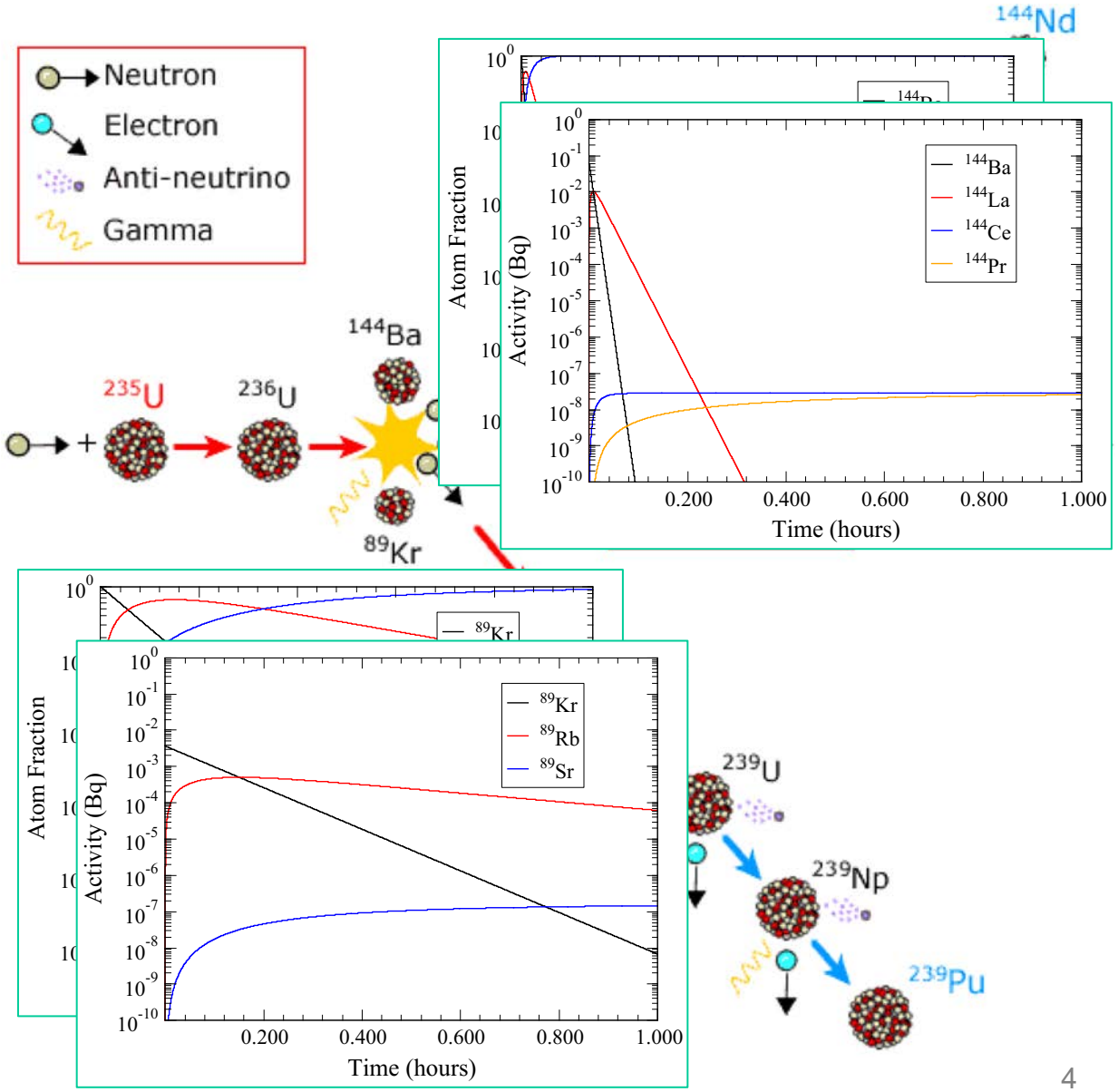

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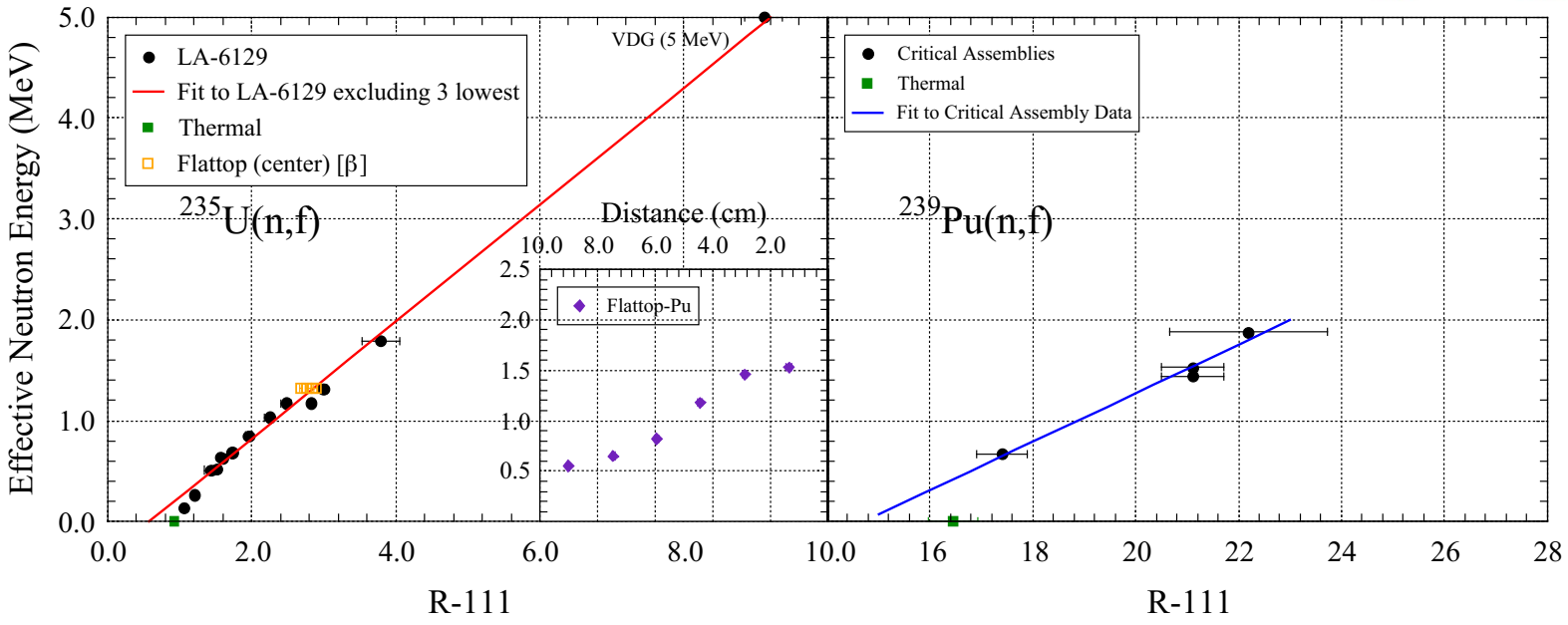

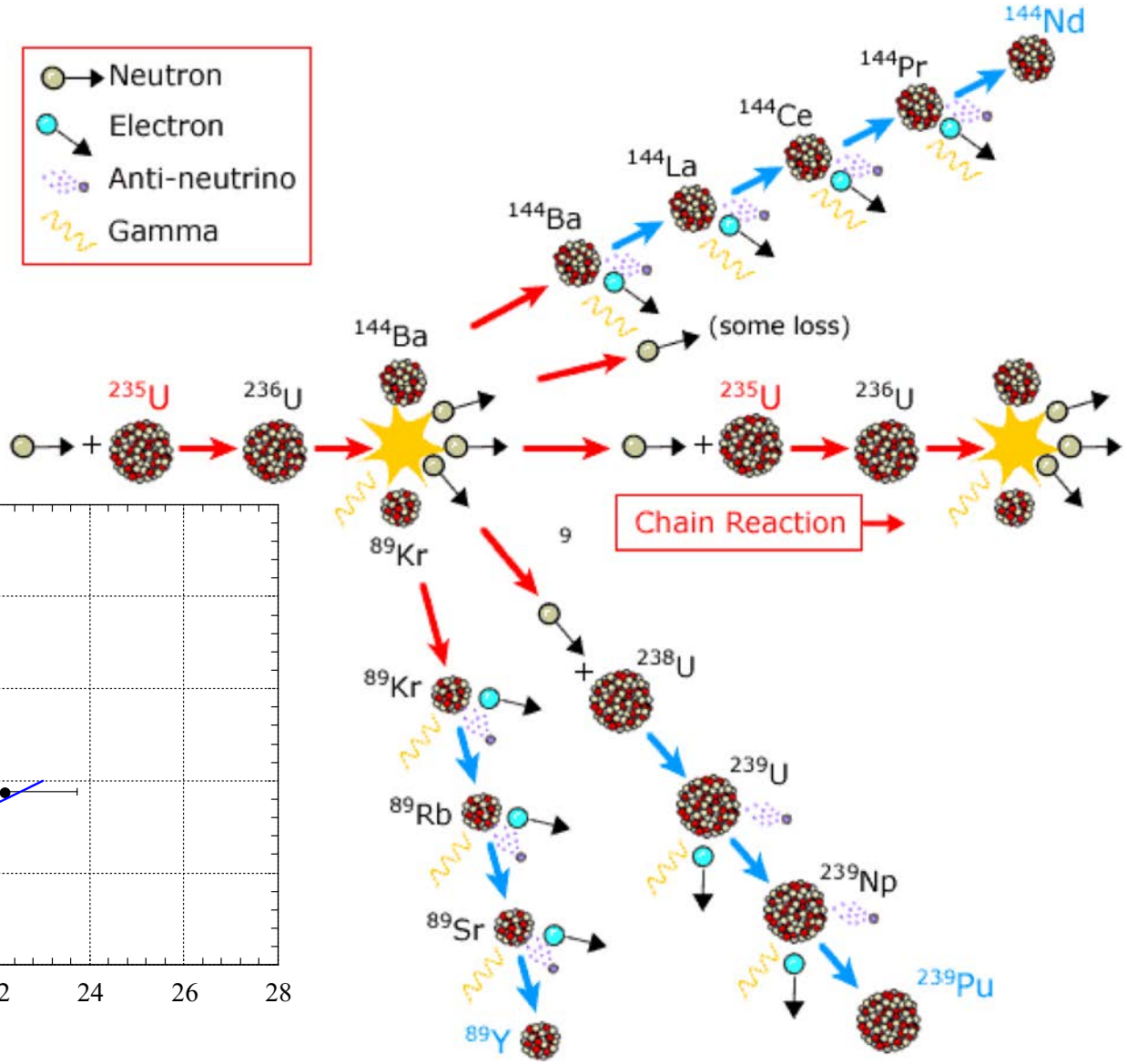

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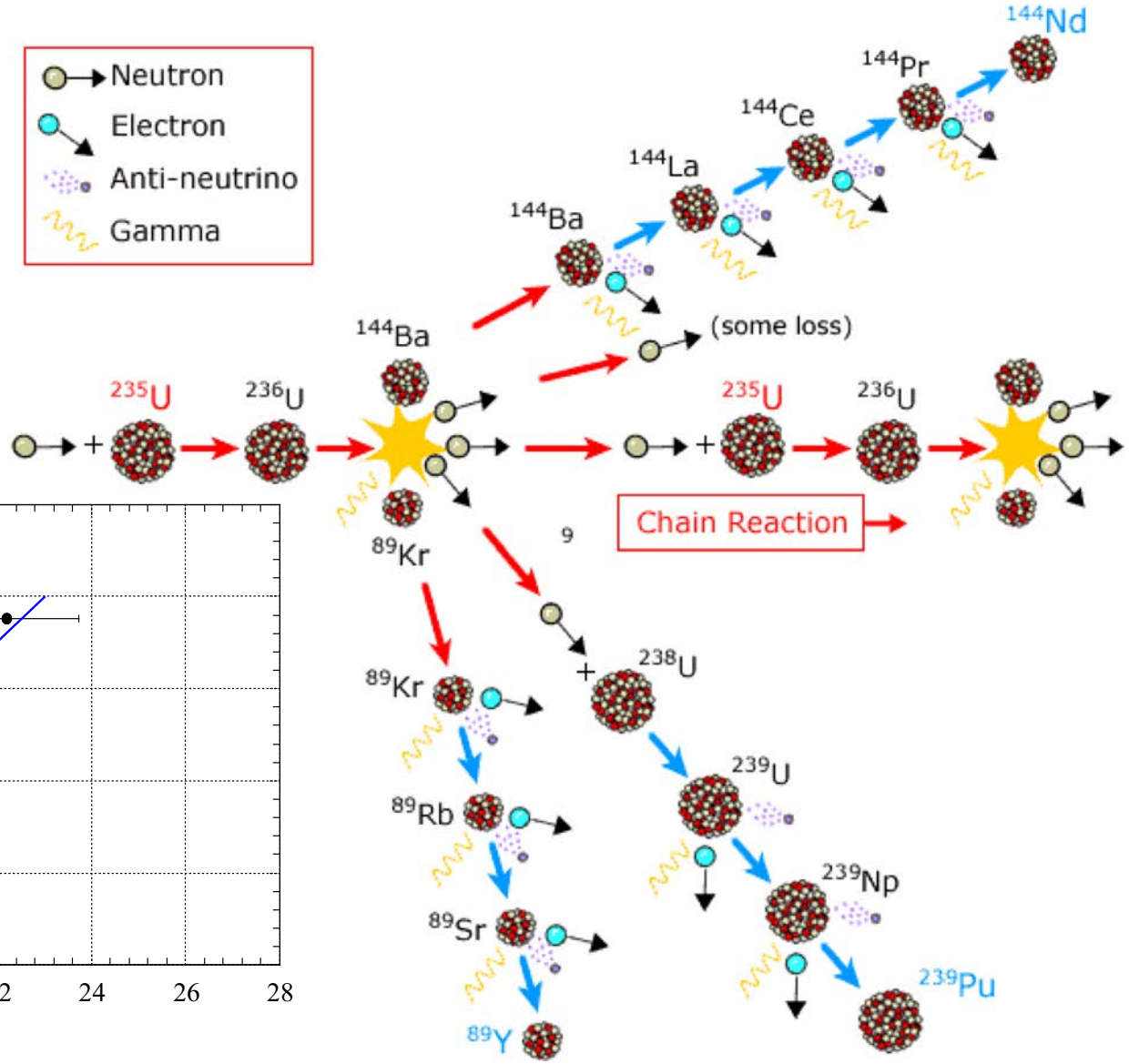
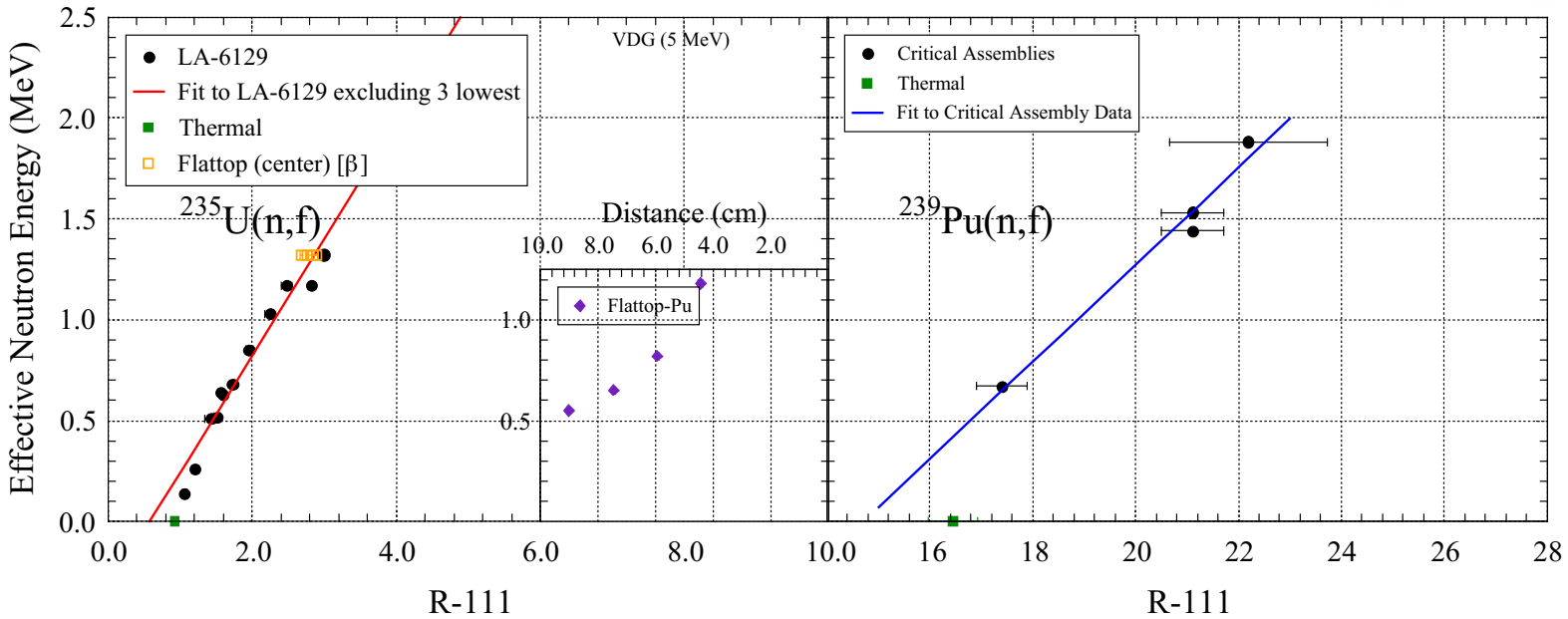
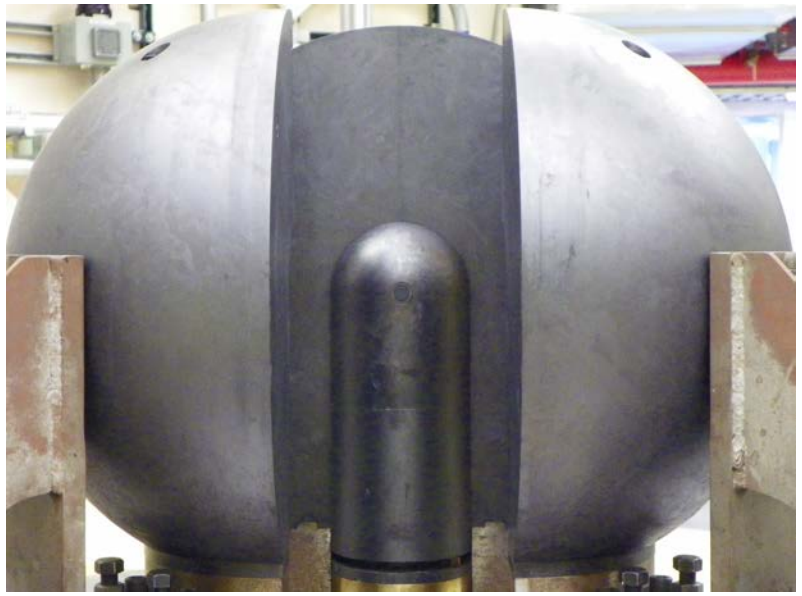




Neutron Sources

Flattop (NCERC)

- Fast/fission Spectrum
- U(93) (17.7 Kg) & WG Pu (6 kg) cores / ^{Nat}U Reflector (~1000 kg)
- Horizontal ("traverse") glory hole
- 10^{13} fissions/g on samples



Godiva IV (NCERC)

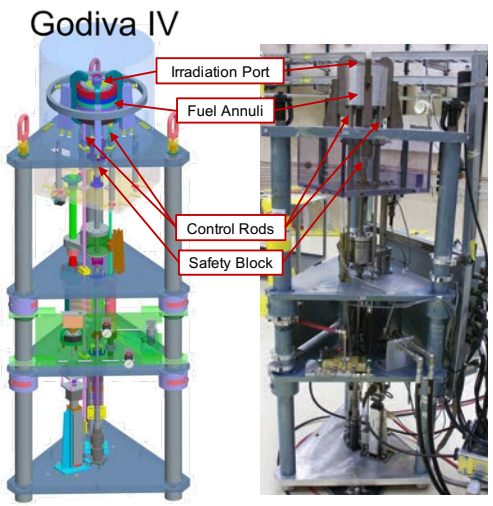
- Fast/fission neutron spectrum
- U(93) (65.5 kg, 1.5% Mo by weight)
- Super-Prompt Critical Operations
- Vertical glory hole for samples
- 1-4 x 10^{16} Total Fissions / burst

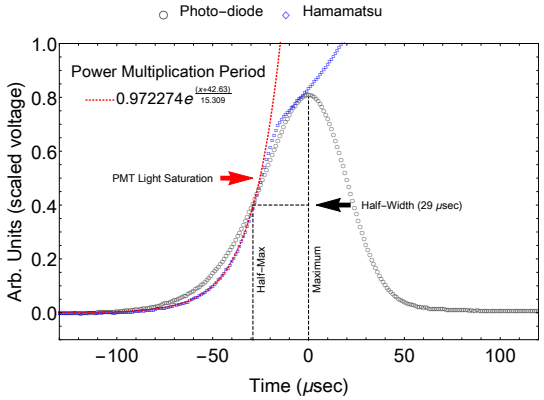
D-T Generator (PNNL)

- Thermo D711 neutron generator
- Low scatter facility at PNNL
- Max neutron flux of 1×10^{9} n/cm^2/s

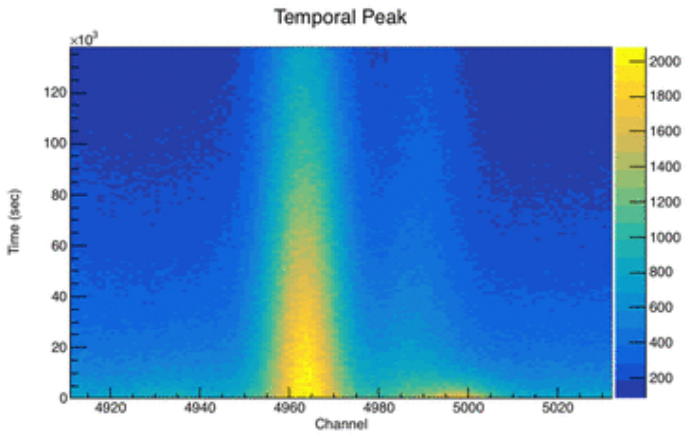
Short-Lived Fission Product Yields (FASTER)

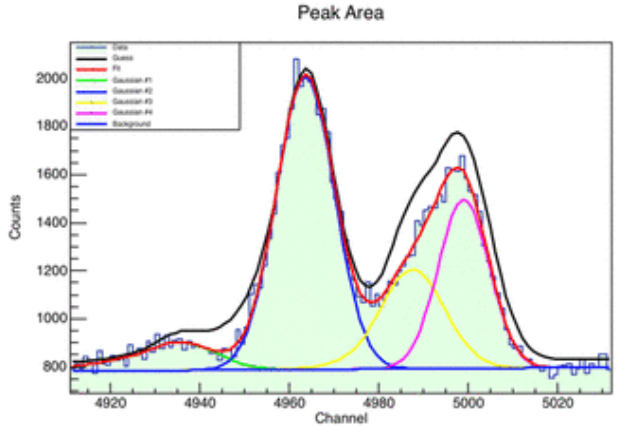
Fabricate High Quality Targets → Pulse Irradiate at NCERC → Acquire γ spectral data → Symmetric Temporal Spectral Analysis (LLNL) → Asymmetric Temporal Spectral Analysis (PNNL) → Evaluate Neutron Fluence & Fissions → Publish Fission Yields in Nuclear Data Sheets for NNDC



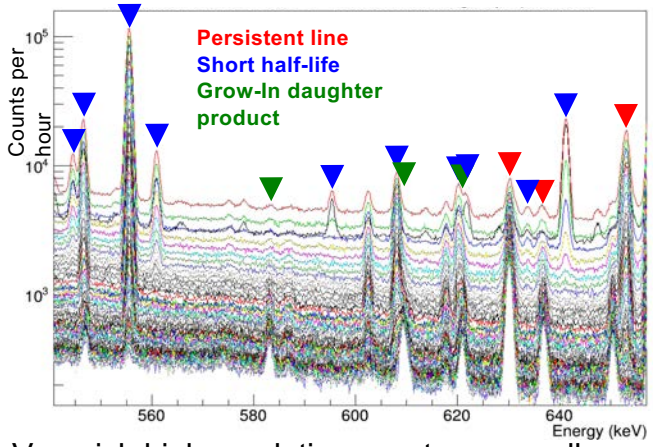


Godiva Irradiation Pulse Width

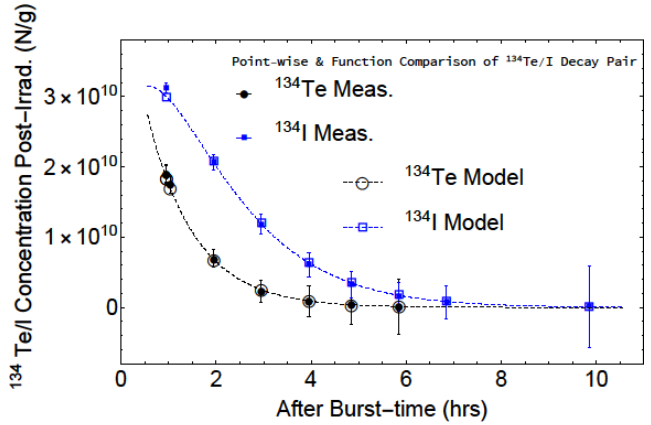




Symmetric Temporal Fit of ^{112}Ag & ^{134}I



Very rich high-resolution spectroscopy allows us to unfold the time dependent behavior of FPs



Asymmetric Temporal Assay of ^{134}I

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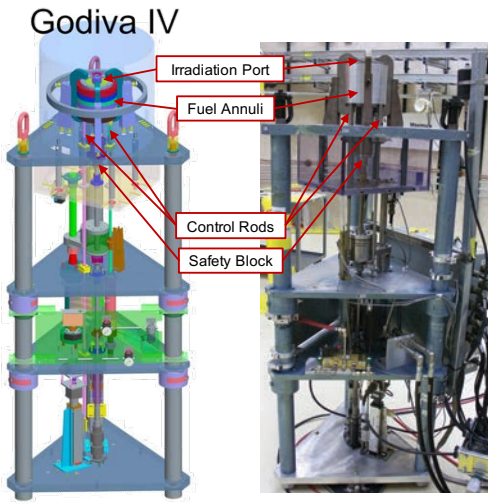
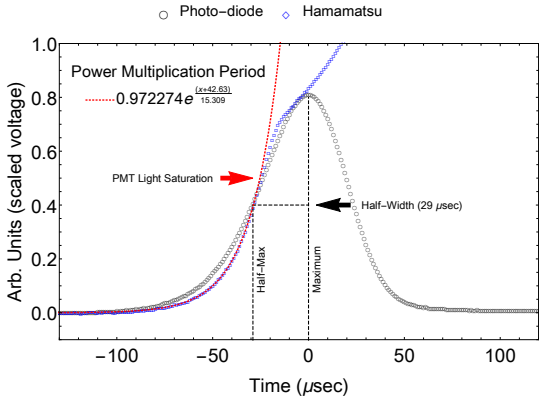




Godiva Irradiation Pulse Width

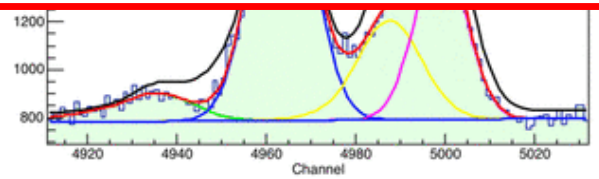
MEASUREMENTS TO DATE

^{235,238}U: Completed

^{239}Pu: Finalizing Analysis

^{237}Np: Data Collected Jan 2020

We are seeing good improvement in FPYs and issues with branching ratios effecting FPY results. This has implications for user community. Results are published in NDS.



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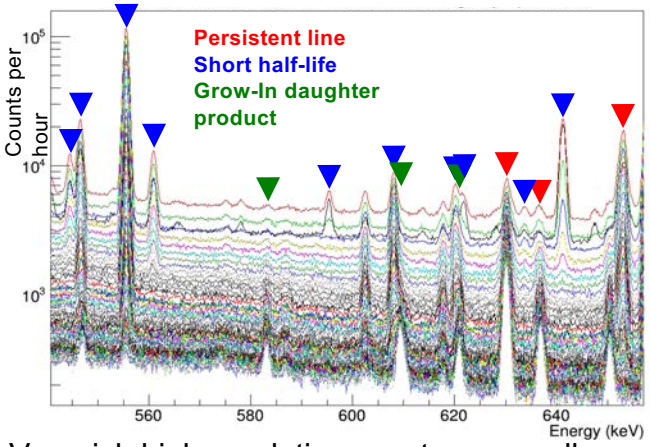

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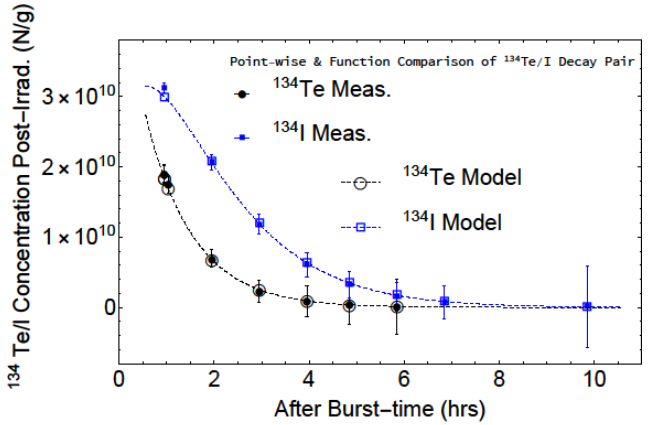
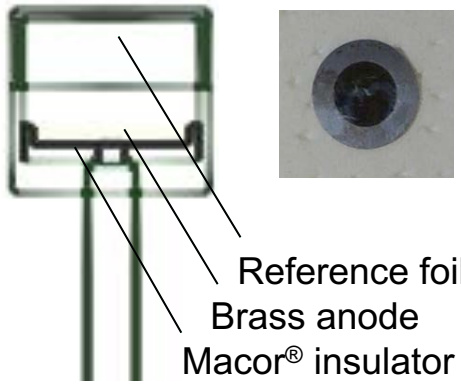
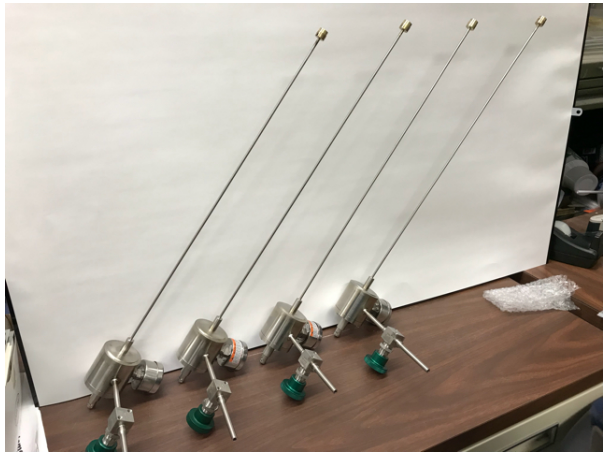


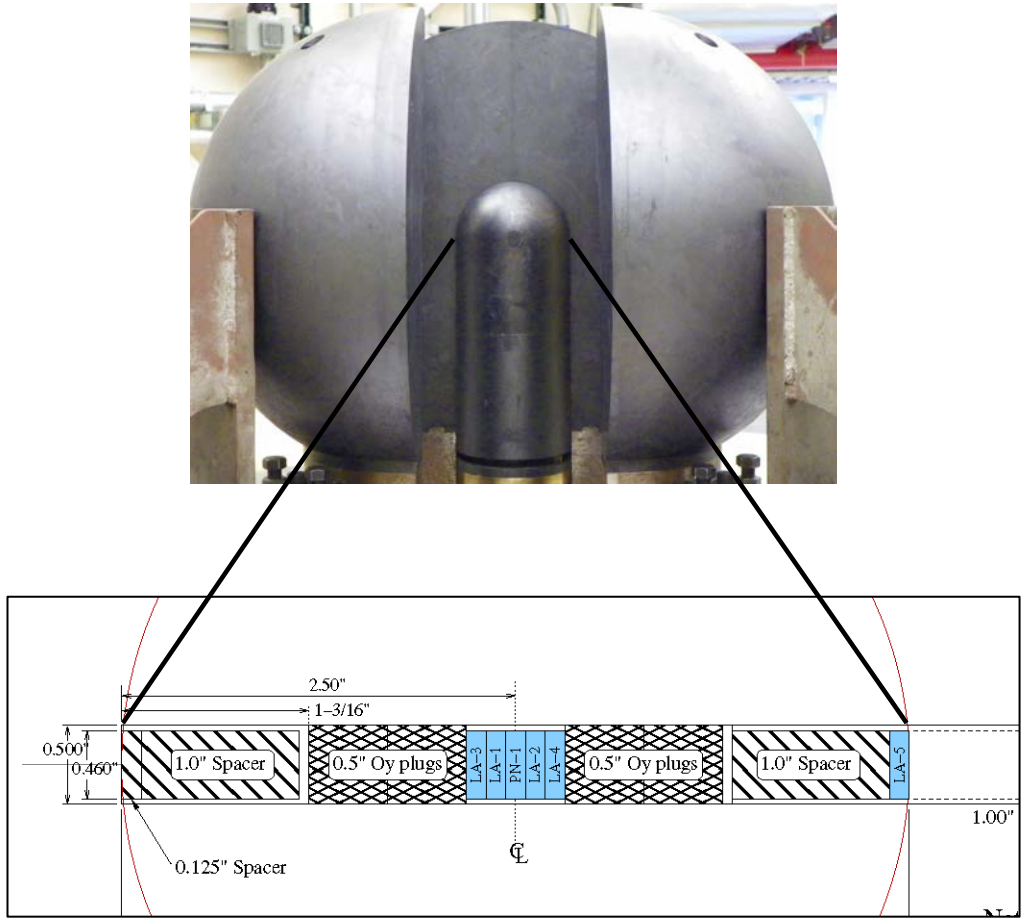
Asymmetric Temporal Assay of ^{134}I

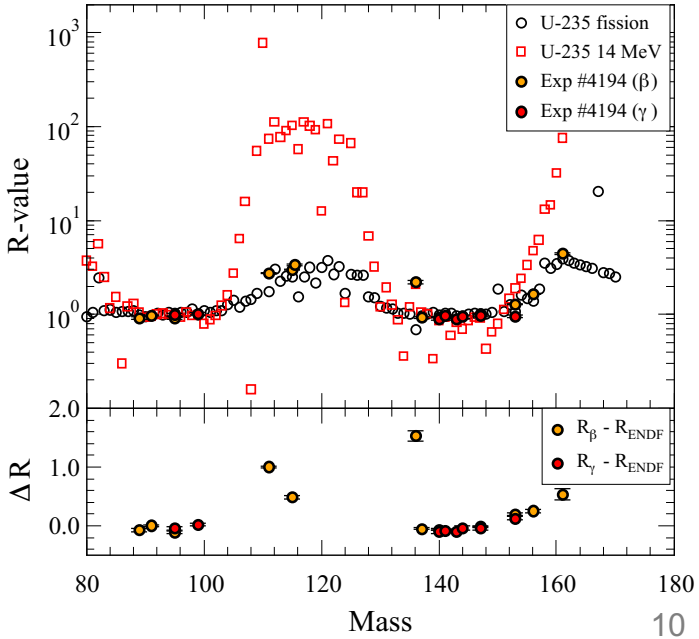
Cumulative Fission Product Yields (BETTER)

Fabricate Fission Chambers to Determine Absolute FPYs → Prepare High Quality Reference and Macro-foils → Irradiate Samples at NCERC and PNNL → Post-Irradiation Whole Foil γ–Counting → Radiochemical Dissolution and Analysis → R-values ($R_i^{j,k}$) and Absolute FP Yields ($Y_i^{j,k}$) → Publish Results and Post in XFOR

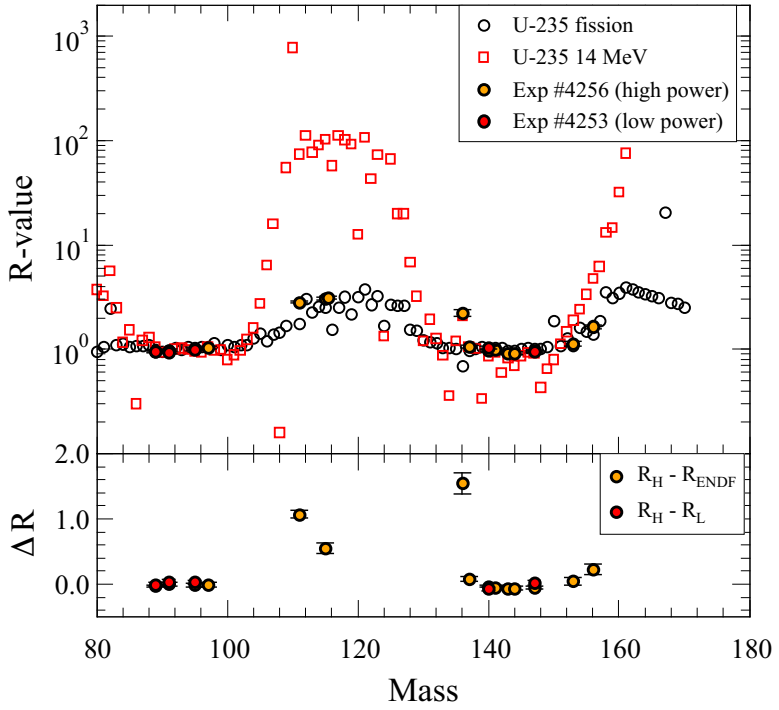


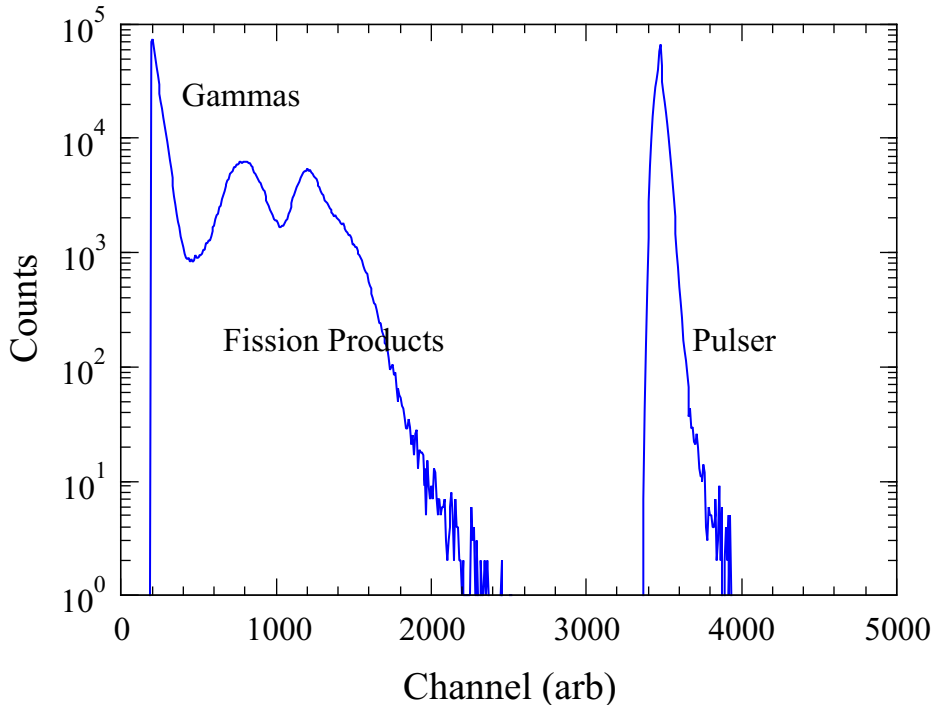






Relative to Absolute Fission Yields





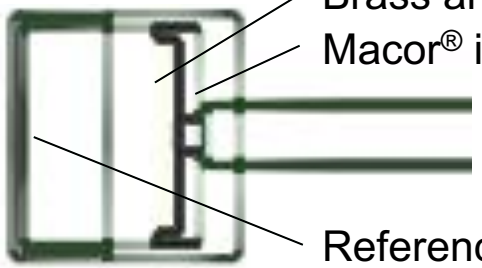
$$R_i^{j,k} = \left(\frac{A_i^{j,k} / A_{99}^{j,k}}{A_i^{25,th} / A_{99}^{25,th}} \right)$$

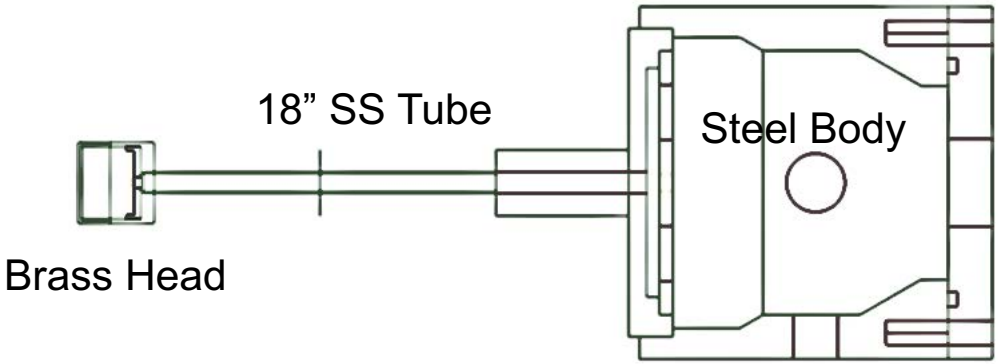
$$Y_i^{j,k} = \frac{(1+\alpha_i) A_i^{j,k}}{\lambda_i \epsilon_i j_i N_f^*}$$

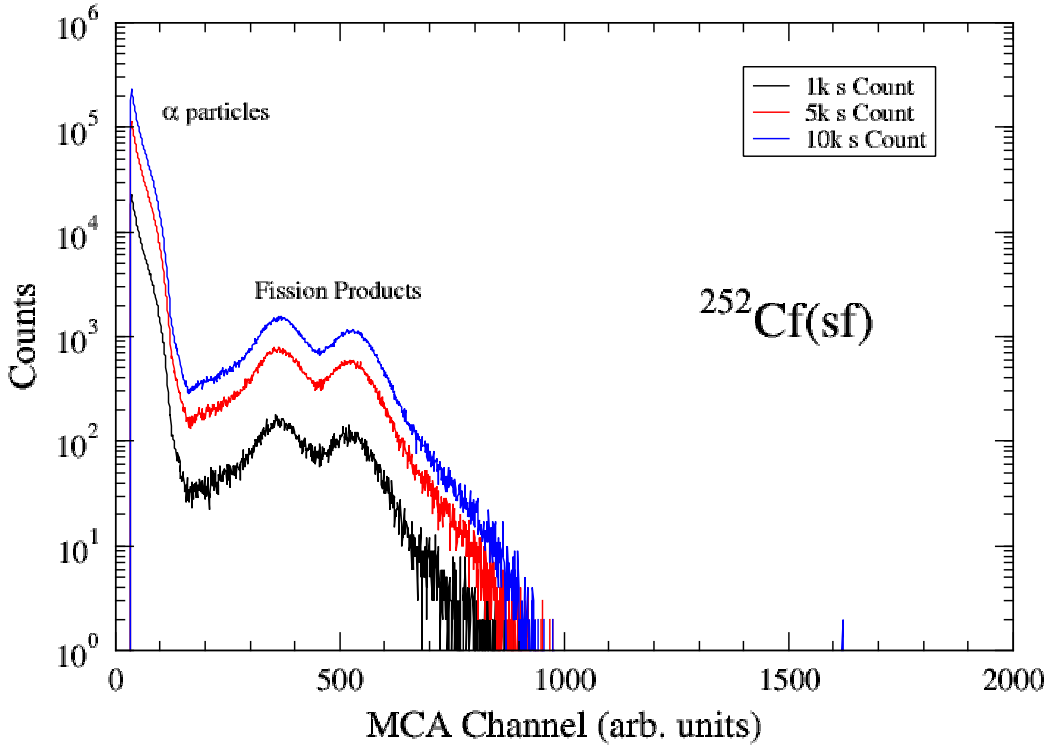
Fission Chamber

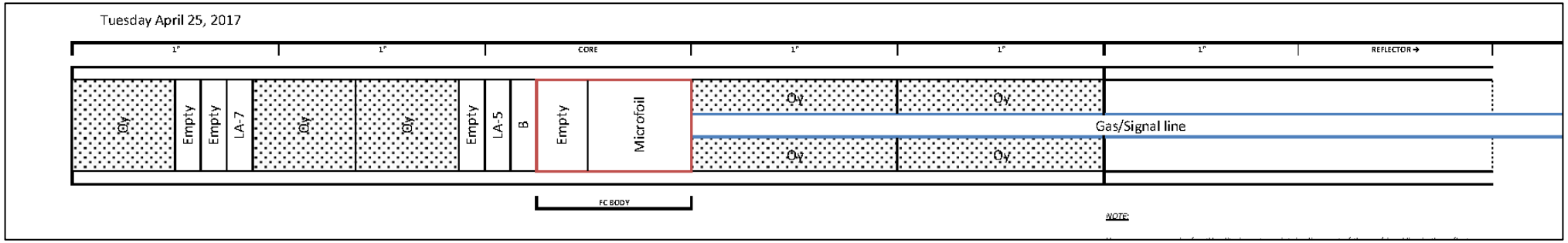
Radiochemistry

Mark II Fission Chamber





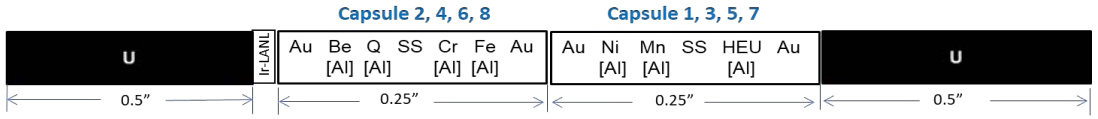




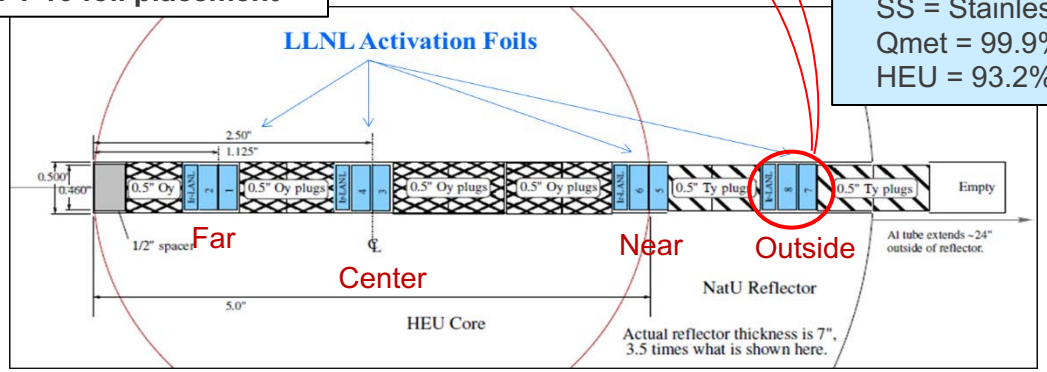
Continuing R&D Activities

- Develop and improve radiochemical and atom-counting techniques for high-fidelity assay of key APs of interest to DD
- Irradiate structural alloys and elemental APs at various neutron energies/spectra:
  - Other facilities for thermal, fission, and 14 MeV neutrons
  - At NCERC facilities with varying SNM fuel types and chemical compositions
  - Extract capture cross sections at different neutron energies
- Modeling of different NCERC configurations in support of signature evaluations

Flattop traverse mode irradiation of SS and activation foils

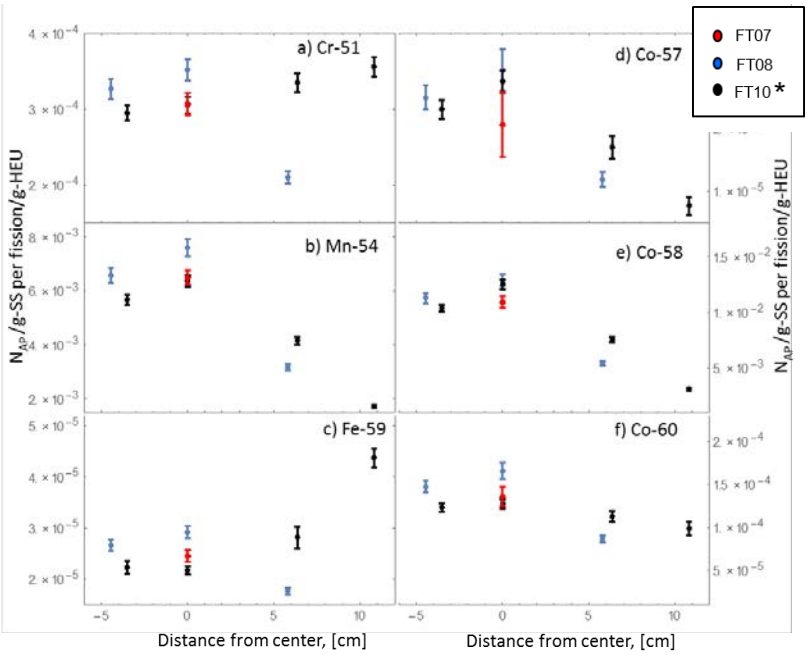


FT 10 foil placement

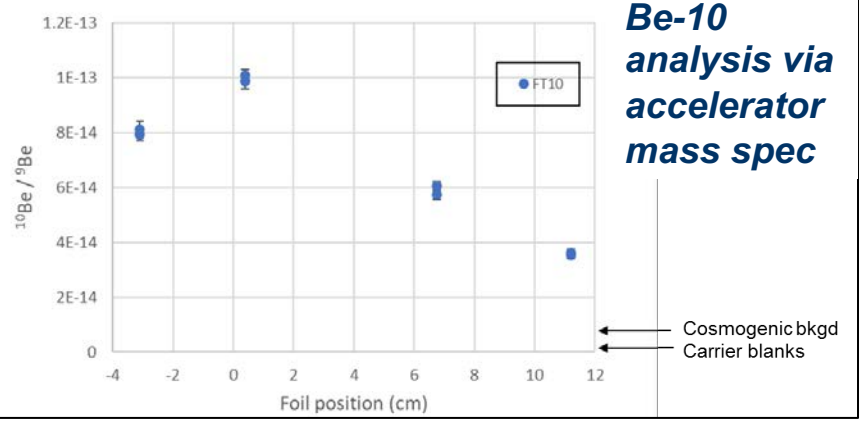


Foil compositions:
SS = Stainless Steel 304
Qmet = 99.9% U-238
HEU = 93.2% U-235

Stainless steel activation results



Be-10 analysis via accelerator mass spec



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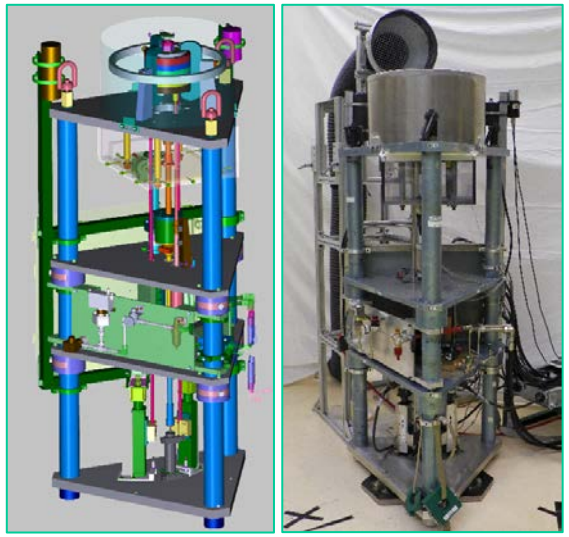
EXTRA SLIDES

Critical Assemblies at NCERC

Planet

- Light Capacity Vertical Lift Assembly
- 10^{13} fissions/g on samples, Variable Spectrum

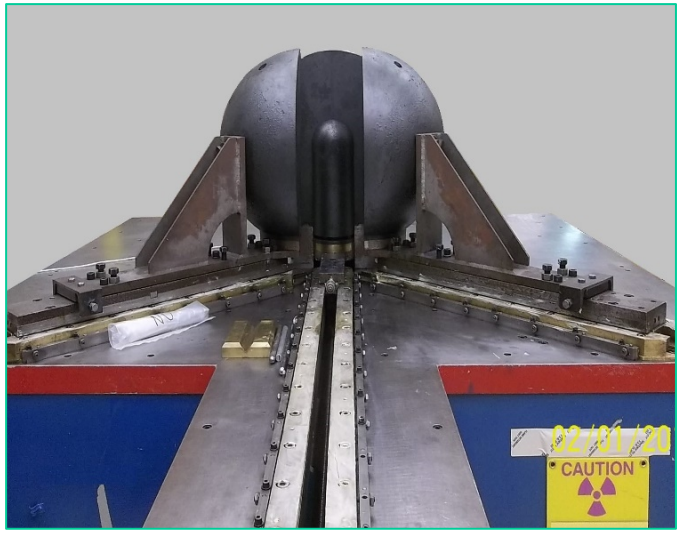


Godiva-IV

- 65.5 kg HEU (1.5% Mo by weight)
- Super-Prompt Critical Operations
- Short-Lived Fission Products
- 1-4 x 10^{16} Total Fissions, Hard Spectrum

Comet

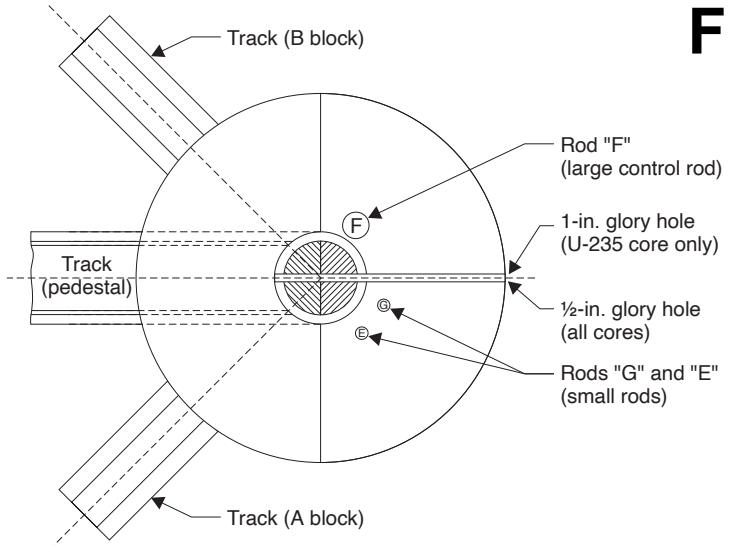
- High Capacity Vertical Lift Assembly
- Supports Large Experiments
- 10^{13} fissions/g on samples, Variable Spectrum



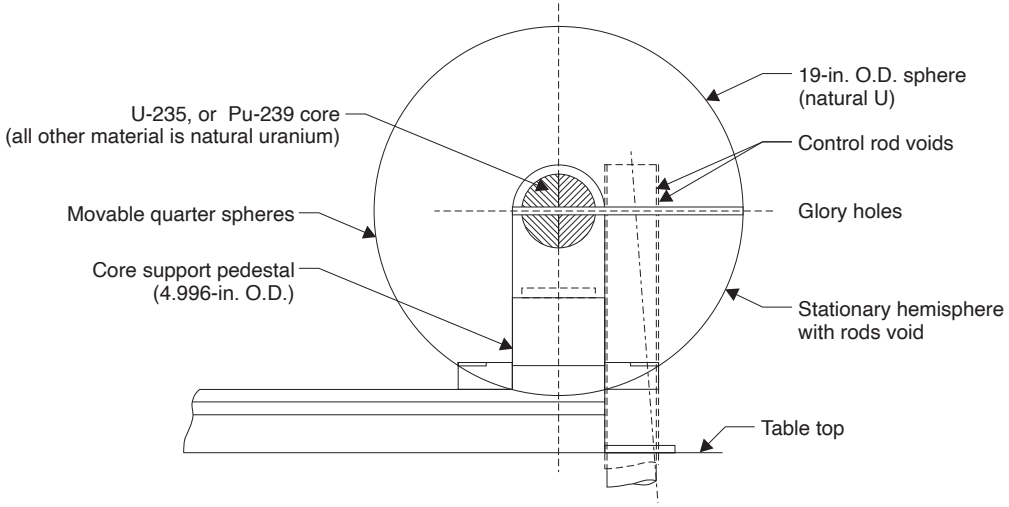
Flat-Top

- Assembly with either a 17.7 kg HEU core or a 6 kg WG Pu core. Reflected by ~1000 kg NU.
- 10^{13} fissions/g on samples, Hard Spectrum

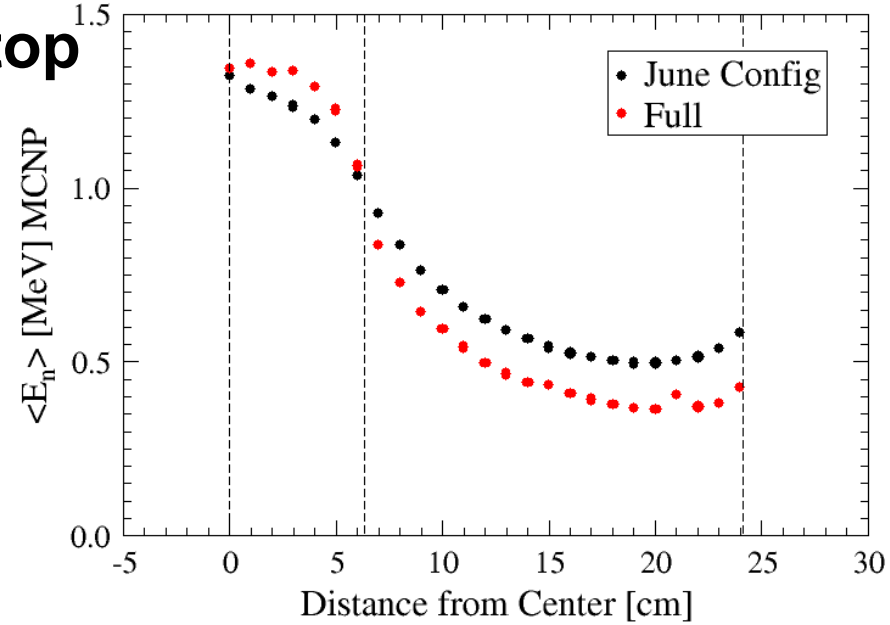
Flattop

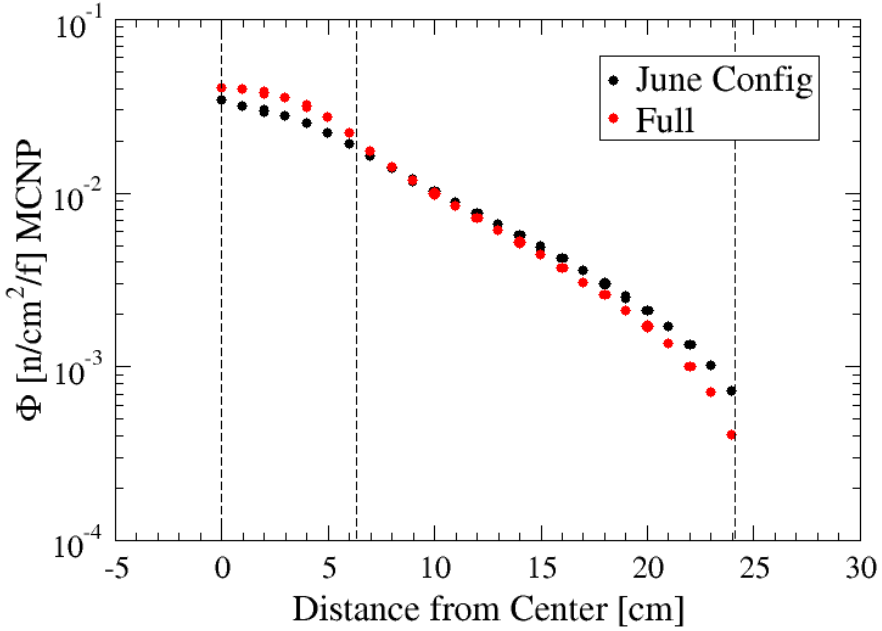


Plan View



Elevation View





Background

Fission Product Yields (FPYs) were historically determined by radiochemical analysis of the irradiated material, and reported as ratios of activities known as R-values*

$$R_i^{j,k} = \left(\frac{A_i^{j,k} / A_{99}^{j,k}}{A_i^{25,th} / A_{99}^{25,th}} \right) = \left(\frac{Y_i^{j,k} / Y_{99}^{j,k}}{Y_i^{25,th} / Y_{99}^{25,th}} \right)$$

Fission chamber experiments were used to calibrate radiation detectors for a small set of FPs, e.g. ^{99}Mo, to quickly determine # of fissions that occurred in a sample

$$K_i^{j,k} = \frac{N_f^*}{A_i^{j,k}} \quad \Rightarrow \quad N_f^* = K_i^{j,k} A_i^{j,k}$$

The K-factor was then used to determine the number of fissions that occurred in other samples directly from the measured activity of the chosen fission product.

NOTE: THE K-FACTOR IS DETECTOR SPECIFIC!

* G.P. Ford and A.E. Norris, LA-6129 (1976)

- The fission chamber (FC) is used to determine the number of fissions, N_f, in a thin deposit of fissile material, called the reference foil.
- The number of fissions, N_f^*, occurring in a "macro-foil" is scaled from the reference foil by mass.

$$N_f^* = \frac{m_M}{m_R} N_f$$

- Radiochemical (RC) analysis of the macro-foil provides $A_i^{j,k}$. The atom number and FPY is then determined by the standard equations

$$N_i^{j,k} = \frac{A_i^{j,k}}{\lambda_i \epsilon_i} = \frac{(1+\alpha_i) A_i^{j,k}}{\lambda_i \epsilon_i f_i}$$

$$Y_i^{j,k} = \frac{N_i^{j,k}}{N_f^*} = \frac{(1+\alpha_i) A_i^{j,k}}{\lambda_i \epsilon_i f_i N_f^*}$$

- Use MCNP and other tools to model as-run configuration and compare results with measured values to test nuclear data.

Fission Yield Data Applications

Pacific Northwest NATIONAL LABORATORY

Lawrence Livermore National Laboratory

NCERC NATIONAL CRITICALITY EXPERIMENTS RESEARCH CENTER

Modelling & Validation

- Criticality Safety — ICSBEP
- Nuclear Forensics — LLNL/LANL
- Shielding & Dose — SINBAD
- Detector Response — ATLAS

Nuclear Data Cycle

New Libraries Used in Modeling and Simulation Codes

Mission Impacting Nuclear Data Gaps Identified

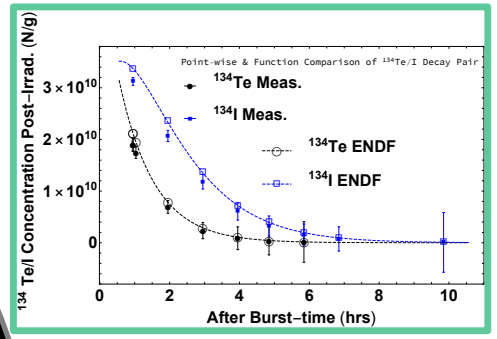
Experimental Campaign is Conducted to Collect New Data

Experimental Results are Vetted and Published

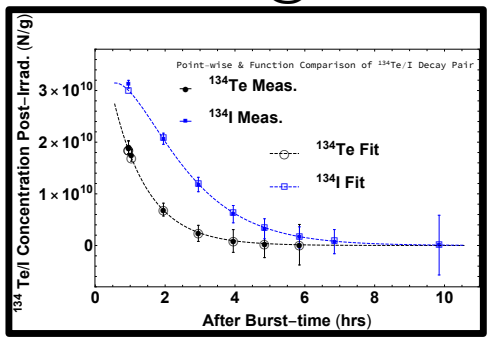
Published Results are Evaluated Against Model Constraints

Evaluation used to Update Nuclear Data Libraries

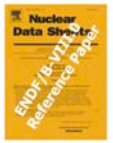
ENDF



Measured @ NCERC



Evaluated Nuclear Data File (ENDF)



ENDF ENDF/B-VIII.0 released February 2, 2018

ENDF/B-VIII.0 fully incorporates the new Neutron Data Standards, includes improved thermal neutron scattering data and uses new evaluated data from the Coordinated International Evaluation Library Organization (CIELO) pilot project for neutron reactions on ^{1}H, ^{16}O, ^{56}Fe, ^{235}U, ^{238}U and ^{239}Pu. Notable advances include updated evaluated data for light nuclei, structural materials, actinides, fission energy release, prompt fission neutron and gamma-ray spectra, thermal neutron scattering data, and charged-particle reactions.

Experimental Nuclear Reaction Data
NNDC
Database Version of 2019-12-18
Software Version of 2019-12-10

Nuclear Data Sheets **155**, 86 (2019).
Nuclear Data Sheets **163**, 249 (2020).