Integrated Experiment Diagnostics

WANDA 2025

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Samantha A. Labb, on behalf of Kelly N. Kmak | NACS

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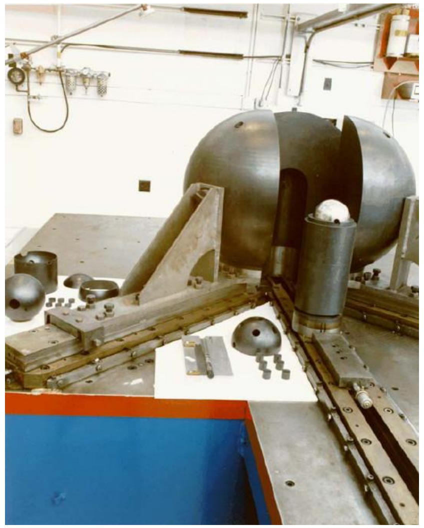
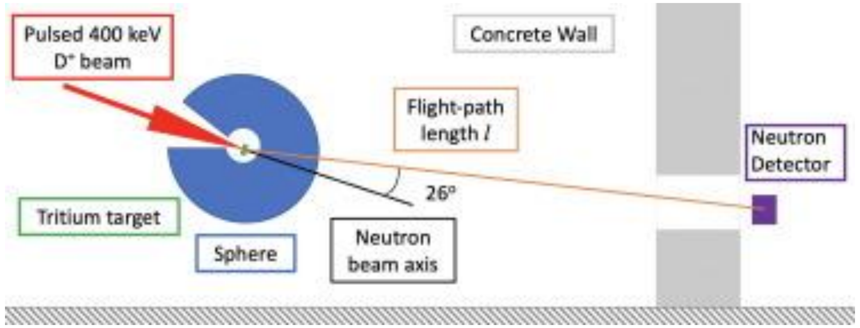
Diagnostics for Integral Data

- Examples of integral experiments
  - Critical assemblies (prompt fission spectrum)
    - Flattop, JEZEBEL, Godiva, BIG-10
    - Steady state, µs-ms bursts
  - Accelerators and neutron generators (fast neutron spectrum)
    - Steady state, pulsed
  - Bethe Sphere (Combined Spectrum)
    - Steady state



Flattop critical assembly, Hayes, LA-UR-12-22906



LLNL Pulsed Spheres, Neudecker et al., Ann. of Nuc. En. 159 (2021) 108345

Integral data provides validation or constraints to differential data

Radiochemistry to diagnose NIF capsule performance

- Diagnosing capsule mix via Tc activation products from Mo capsules
  - Ratios used to inform nuclear physics models
- Radiochemistry required to remove gamma-ray interferences from fission products to reliably identify Tc isotopes
  - Separation Mo and Tc from each other and other FPs
  - Remove interferences in gamma spectroscopy: Nb, Au activation, V activation
- Currently measuring both ^{96g}Tc (4.3 days) and ^{95g}Tc (20 h)

^{92}Tc	^{93}Tc	^{94}Tc	^{95}Tc	^{96}Tc	^{97}Tc	^{98}Tc	^{99}Tc	^{100}Tc	^{101}Tc
^{91}Mo	^{92}Mo	^{93}Mo	^{94}Mo	^{95}Mo	^{96}Mo	^{97}Mo	^{98}Mo	^{99}Mo	^{100}Mo

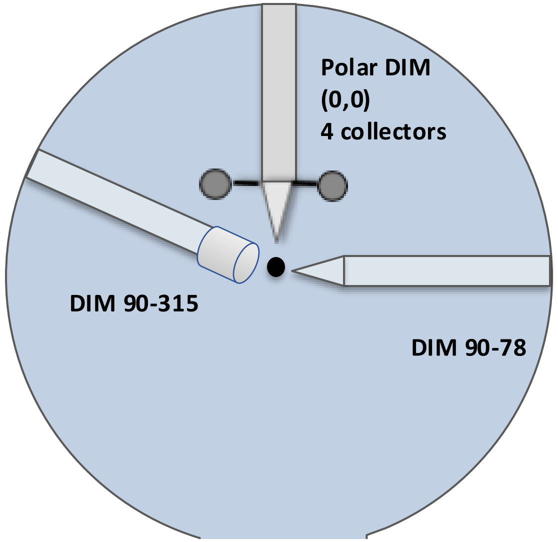
Radiochemical Diagnostics

Chemistry Development

- Separations optimized over 14 trials
- First tested on N231001
- Routine Measurements started in Nov ‘24 with ^{96g}Tc/^{99}Mo ratios reported for N231126 and shots since then
- Refined over the next several PSS shots
- In May 2023, expanded measurements to both ^{95g}Tc and ^{96g}Tc

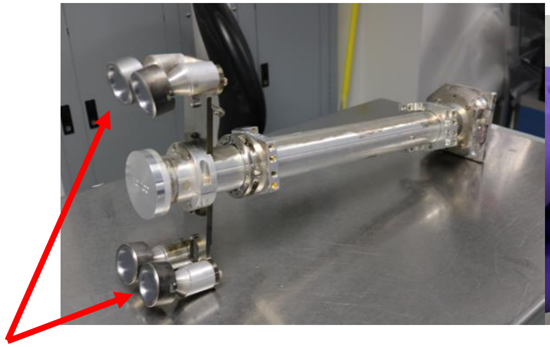
Future Goals

- Expand radchem capabilities to allow for processing more SRCs
- Working with University of Wisconsin (Jonathan Engle) to produce ^{97m}Tc
    - Allow for simultaneously tracing the chemistry and measuring both ^{95g}Tc and ^{96g}Tc for all samples

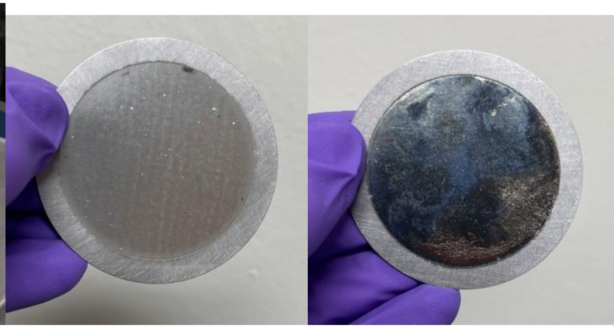


SRCs geometric collection efficiency (based on solid angle) is ~0.1%

Solid debris collection



SRCs in bracket
(4 per DIM)



Polar DIM vs 90-78 for N240616 (PSS)

SRCs retrieved from NIF
- SRCs are “super-rushed”; retrieved from NIF (~2 hrs post shot)

SRC preparation
- Brackets from 90-315 DIM are brought to B154
- SRCs mounted for counting

Initial Counting
- NCF counts the SRCs for initial amount of ^{99}Mo (8-12 hours)

Chemistry
- Separations performed and samples prepared for counting
- ~7 hours for 4 SRCs

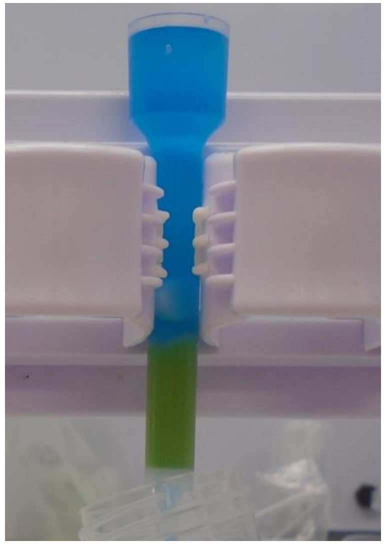
Final Counting at NCF
- Samples counted for 2 hours to 10 days
- ^{99}Mo, ^{95g}Tc and ^{96g}Tc are measured

Data Analysis
- Spectral analysis
- ratio calculation

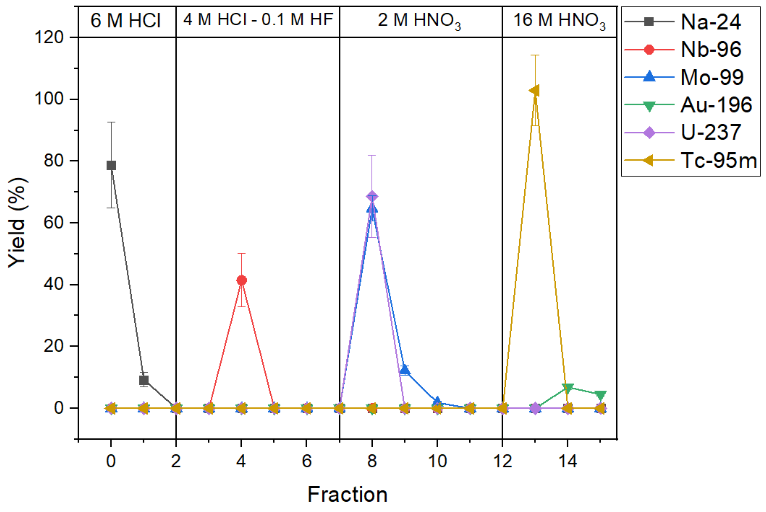
Leaching

Before and after

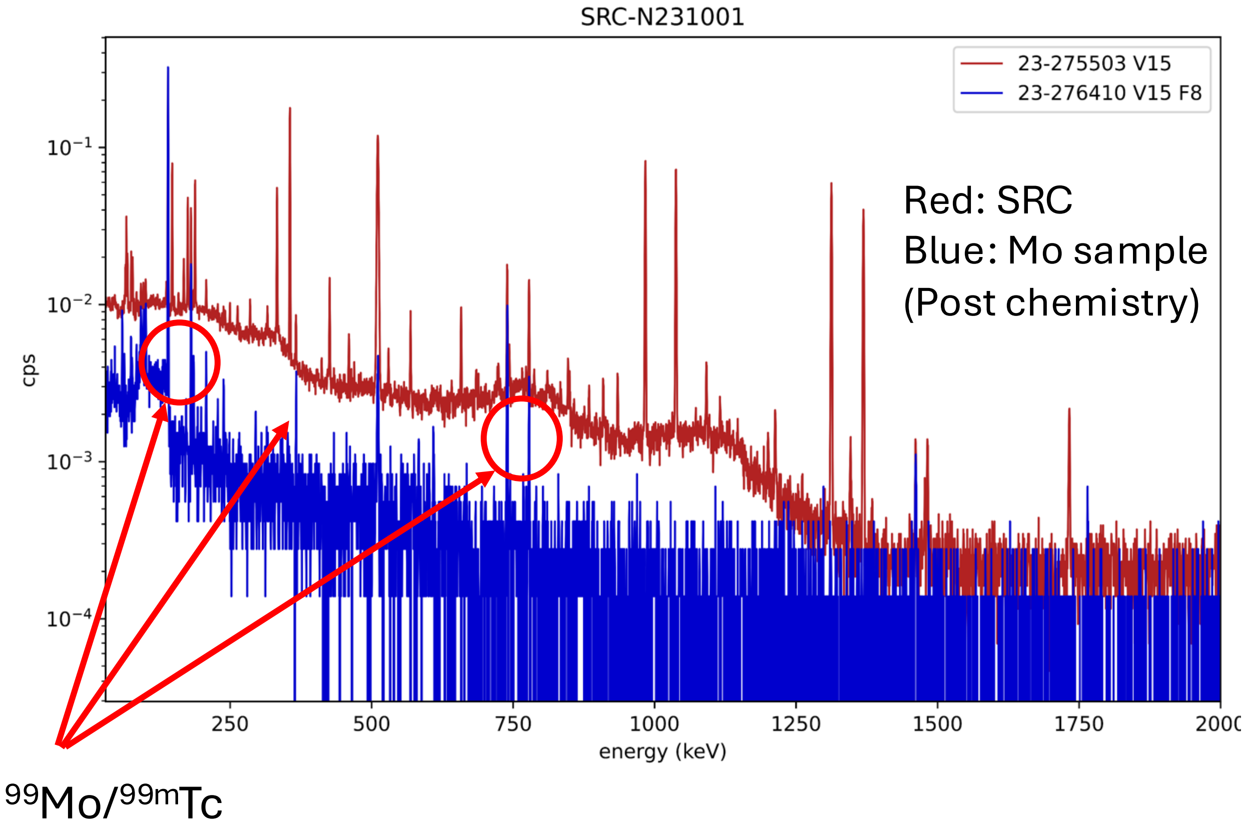
Column Elution

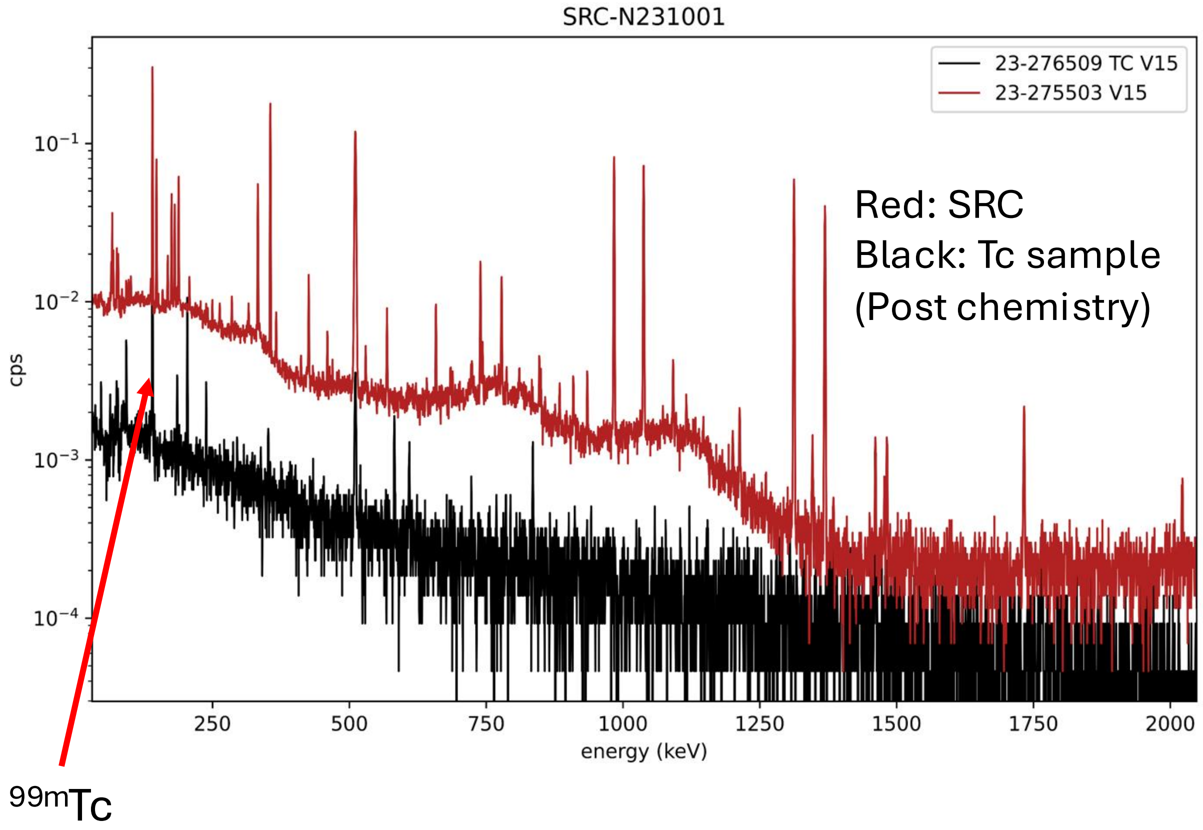


Separation Results

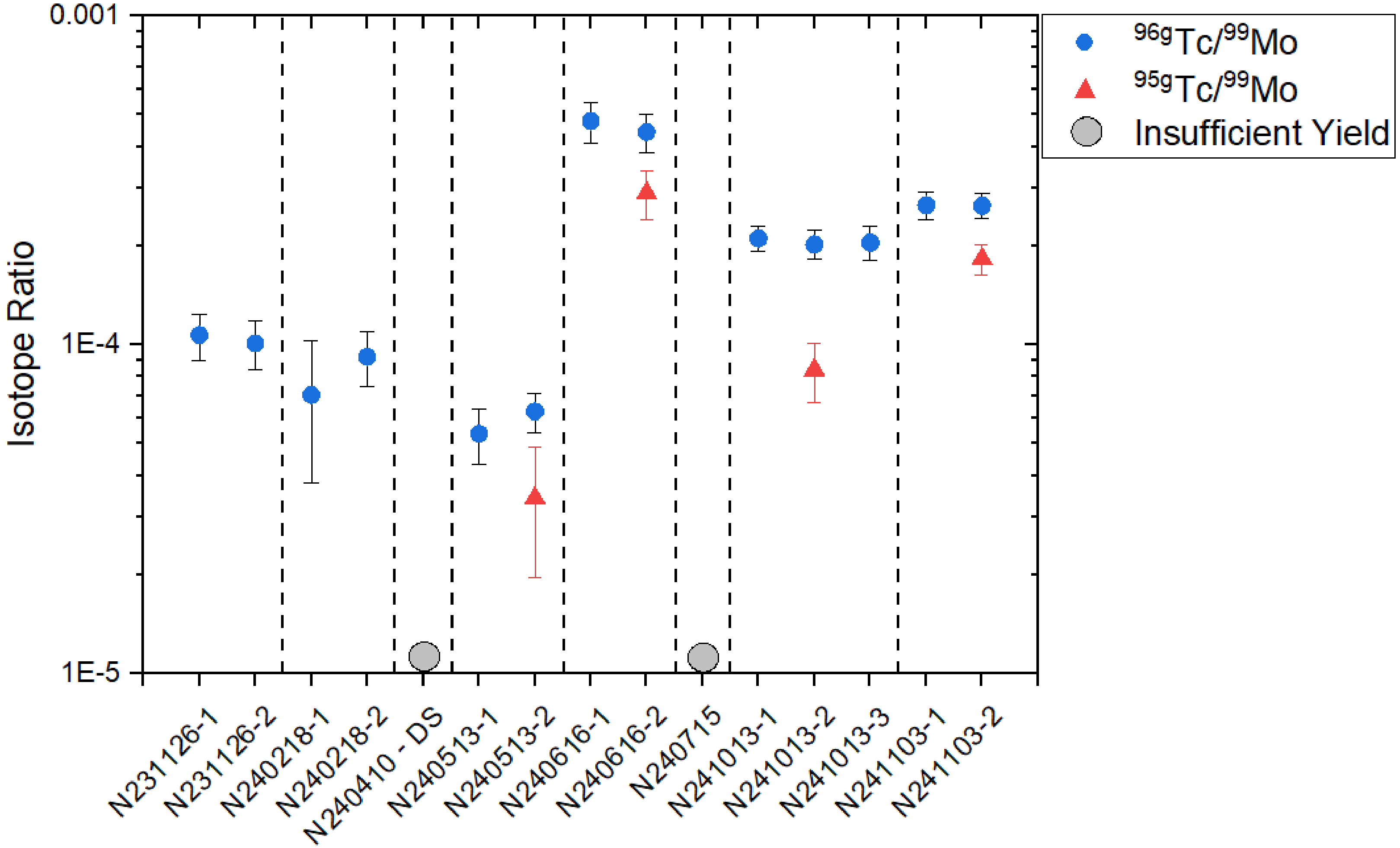


Before and After Chemical Separations (N231001)





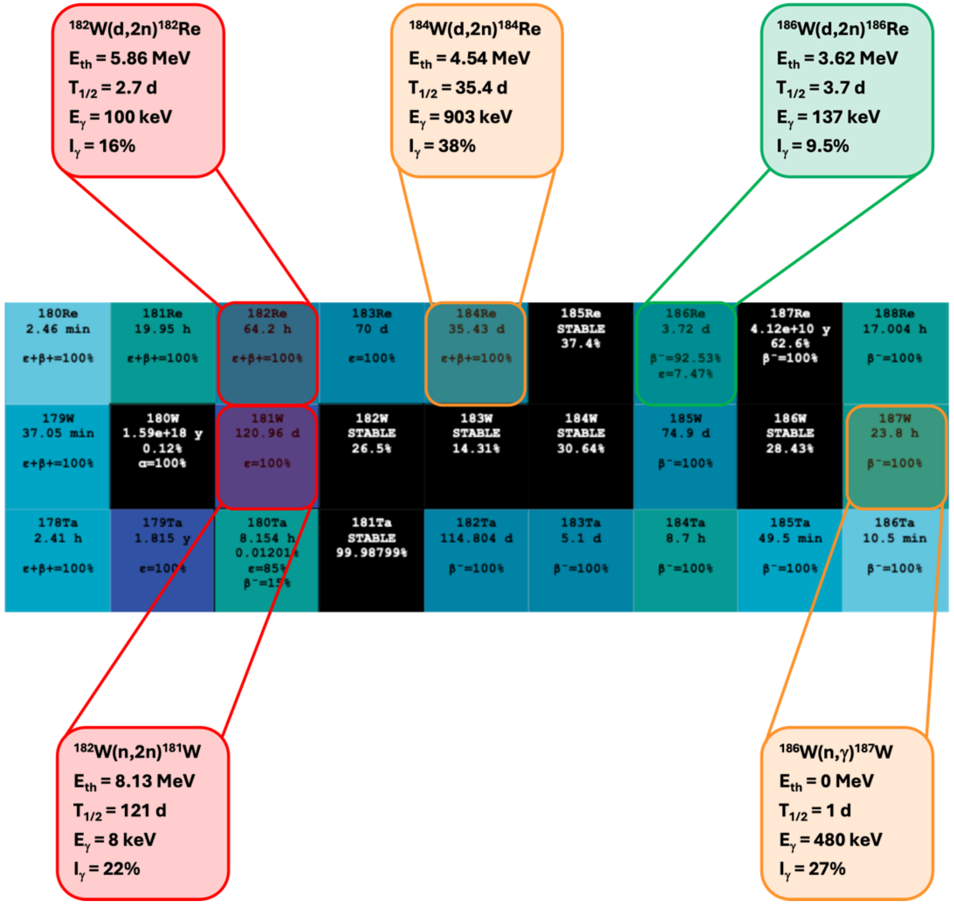
Results in FY24 – First Measurements of $^{96}Tc/^{99}Mo$ Ratio



We have developed rapid, high-yield chemical procedures and are able to provide routine results for the PSS campaign and are working to expand capabilities

Exploring tungsten radiochemistry as a possible mix diagnostic for various burn platforms (PSS, ICF)

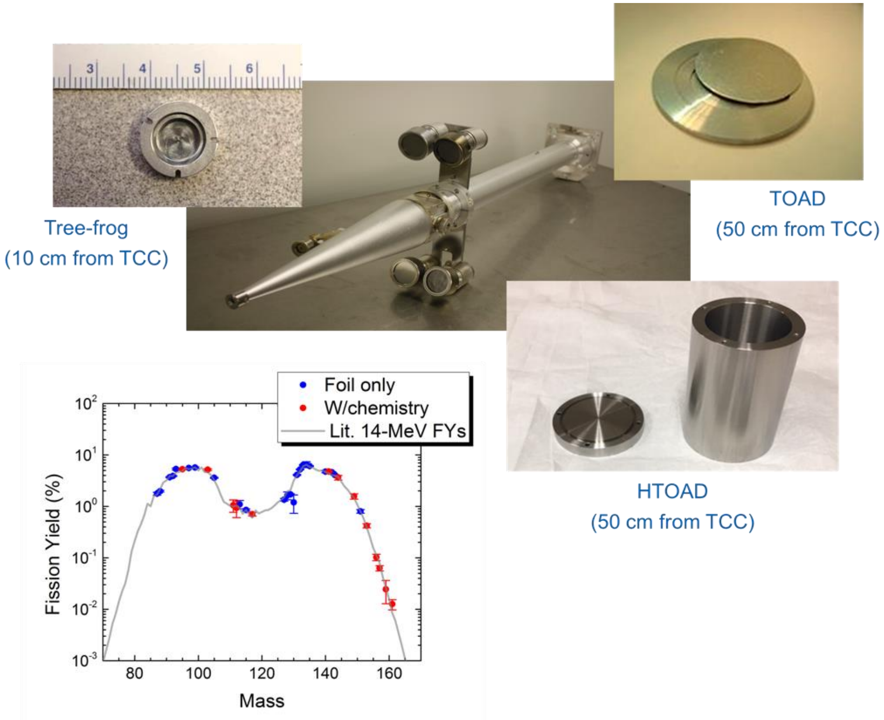
- Several programs interested in using tungsten as a mix diagnostic
- W and Re are difficult to detect due to nuclear decay properties
- Very challenging separation chemistry
  - W/Re have multiple oxidation states
  - Require stabilization with HF
  - Cannot use any existing chemical procedures
- Identifying isotopes of interest in FY25 with R&D in FY26 & FY27 (depending on program needs)



Possible reactions of interest in the W, Re system (PSS, ICF)

Fission product measurements on exotic materials possible at NIF using TOADS

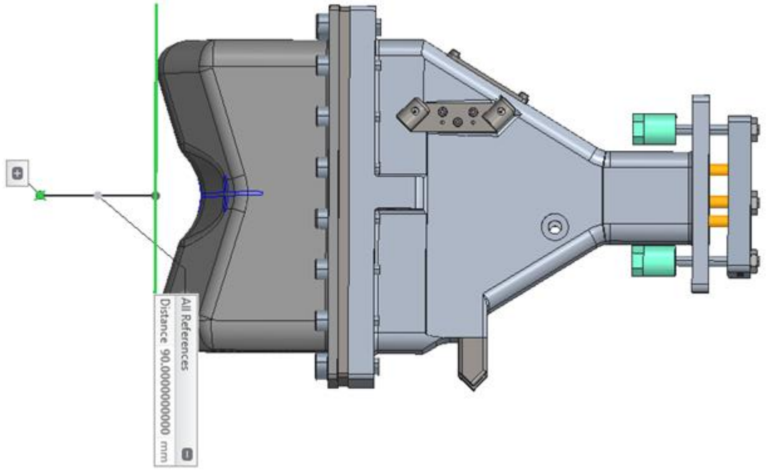
- NIF provides a unique platform for measuring 14.1 MeV fission yields (FPYs) of actinide isotopes
- Target Option Activation Device (TOAD)
- Materials of interested will be fielded in TOADs with flux monitors
  - Isolated from shot debris and see a high neutron flux
  - A combination of nuclear decay counting, and radiochemistry will be used to make measurements on fission products of interest
- TOADs can be used to field other actinide materials
  - $^{244-248}$Cm, $^{238-244}$Pu, ^{237}Np, $^{241-243}$Am, etc.
- Ignition and EYC yields enable even more FPYs to be measured at lower uncertainty



Various types of toads, fielded behind SRC at 35-50 cm, with initial TOAD based DU fission product measurement

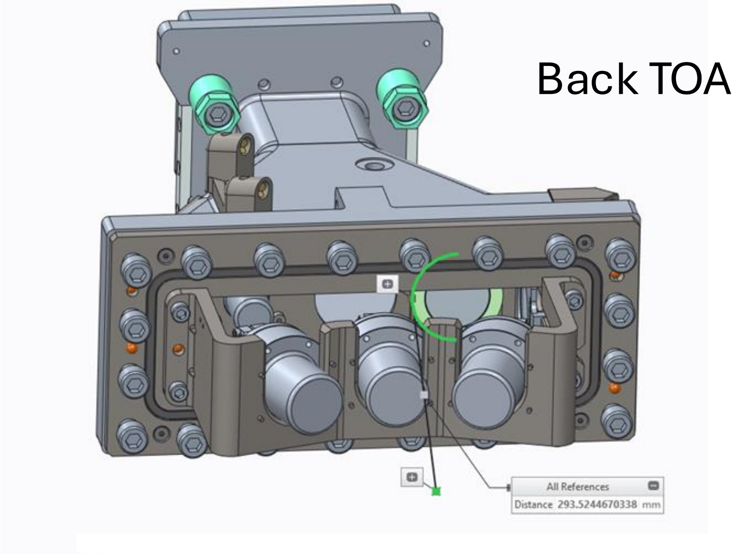
TOAD Experimental Set-up

- Measurements from two DU TOAD samples deployed on N240907-001 in CryoXNBS Config 6
  - Foils delivered to chemistry lab ~17 days post shot Neutron yield: 2.9E+17
- Multi-programmatic, cross-cutting data obtained
- Fission yield measurements contribute to DPI project
- Provided fission product data to benchmark simulated fission product predictions

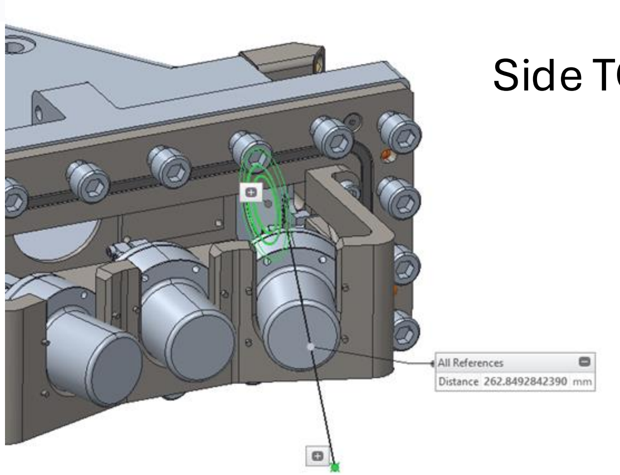


CryoXNBS
Snout Tip Standoff = 9 cm

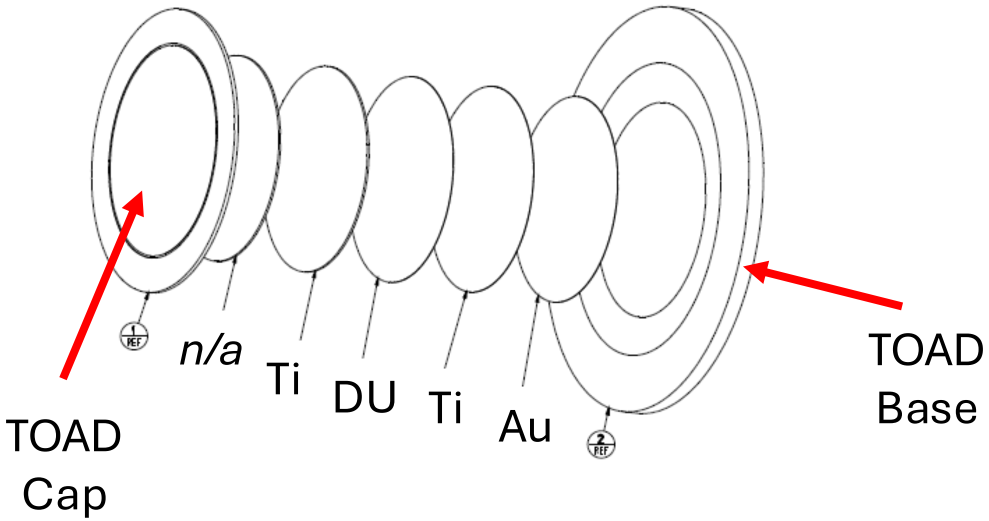
CryoXNBS Configuration



Back TOAD: 1.2103g DU



Side TOAD: 1.225g DU



Au foils are used as a flux monitor

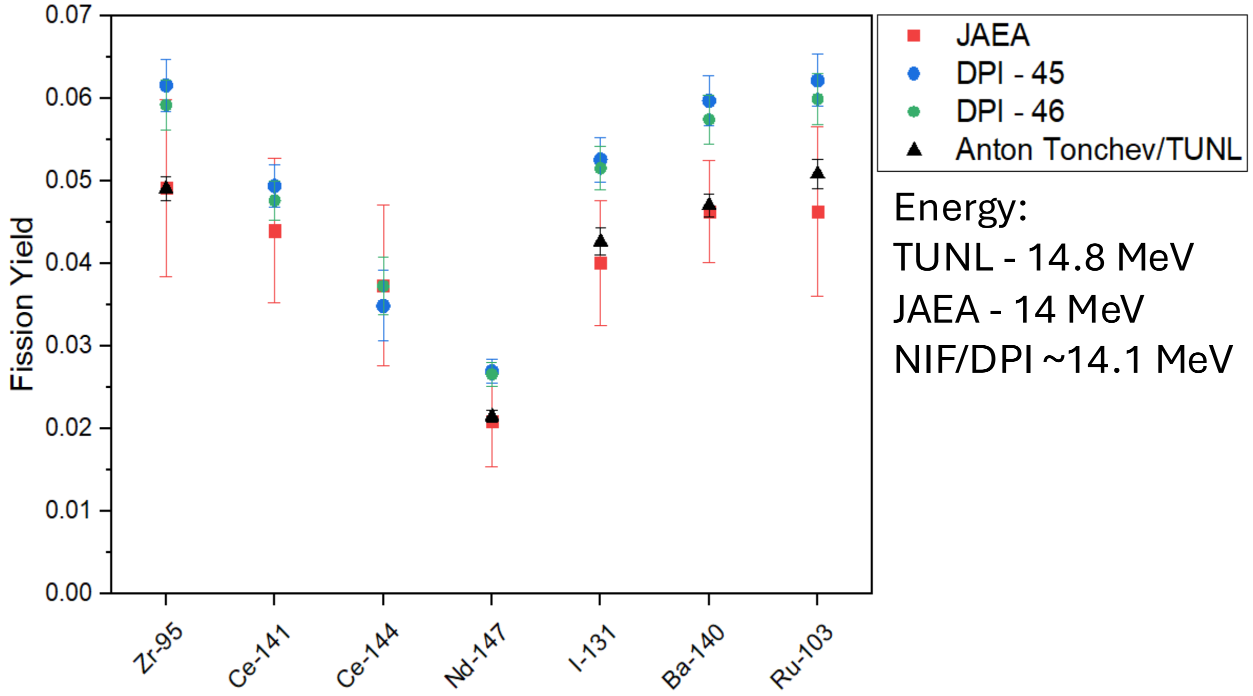
Ti catcher foils ensure no FPs are lost to Au

TOAD	Flux (n/cm²/s)	Error	% Error
Back	1.40E+23	4.81E+21	3.45
Side	1.80E+23	6.31E+21	3.50

Images from John Gundelfinger & Charles Yeamens

Fission Yield Results

- TOADS are fully sealed – not exposed to NIF debris wind
- TOAD samples delivered to chemistry lab ~17 days post shot
  - Counted on days 18, 19, 21 post shot
- DPI samples give consistently higher FPYs than measurements using traditional methods (JAEA and TUNL)
  - Work is ongoing to confirm results and improve error

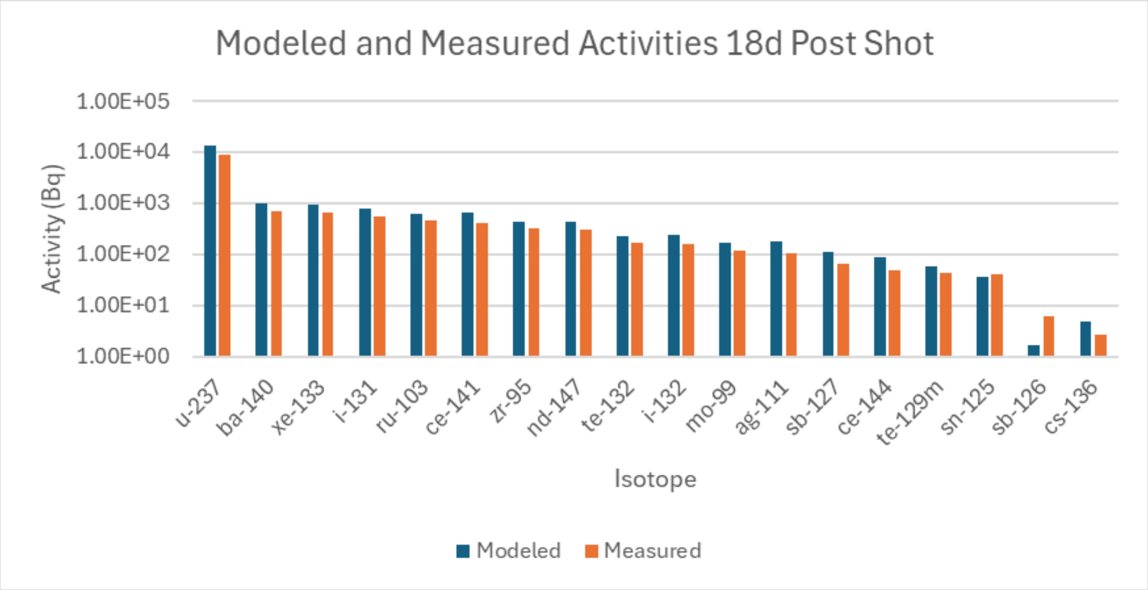


Energy:
TUNL - 14.8 MeV
JAEA - 14 MeV
NIF/DPI ~14.1 MeV

JAEA/TUNL agree very well; DPI yields are higher. Errors are due to counting statistics.

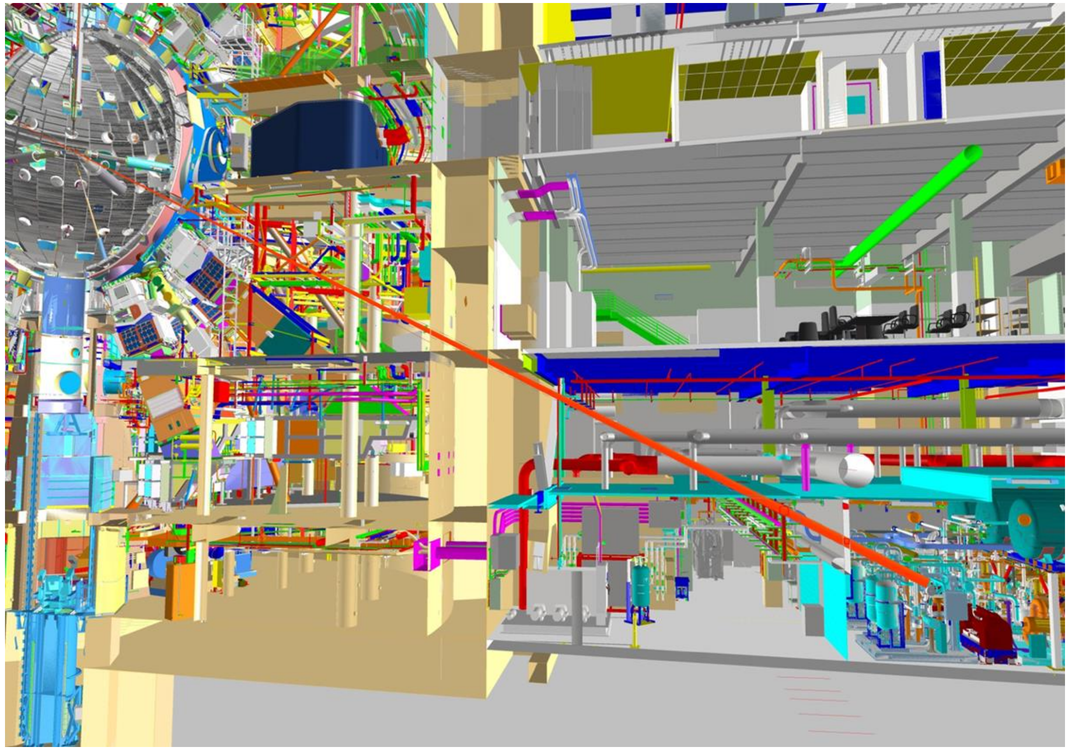
Comparison with Models
collaboration with J. Gundelfinger

- Benchmark simulated fission/activation product predictions against real life measurements
- Simulations conducted with snout/TOAD configuration matching using MCNP -> ALARA simulation workflow
- The model consistently overpredicted isotope activities compared with measurements
- Average Overprediction factors:
  - Side Sample: 18 days post shot: 1.38
  - Back Sample: 18 days post shot: 1.25



RACER: Rapid Collection for Experimental Radiochemistry

- Concept: Drop an SRC collection foil post-shot into a tube that travels down to the HMMA in the NIF basement
- Sample would be controlled via vacuum and pneumatic air
- Sample could be retrieved and measured ~1 minute post-shot regardless of shot yield
- Essential for shorter half-life reaction products requested by programs

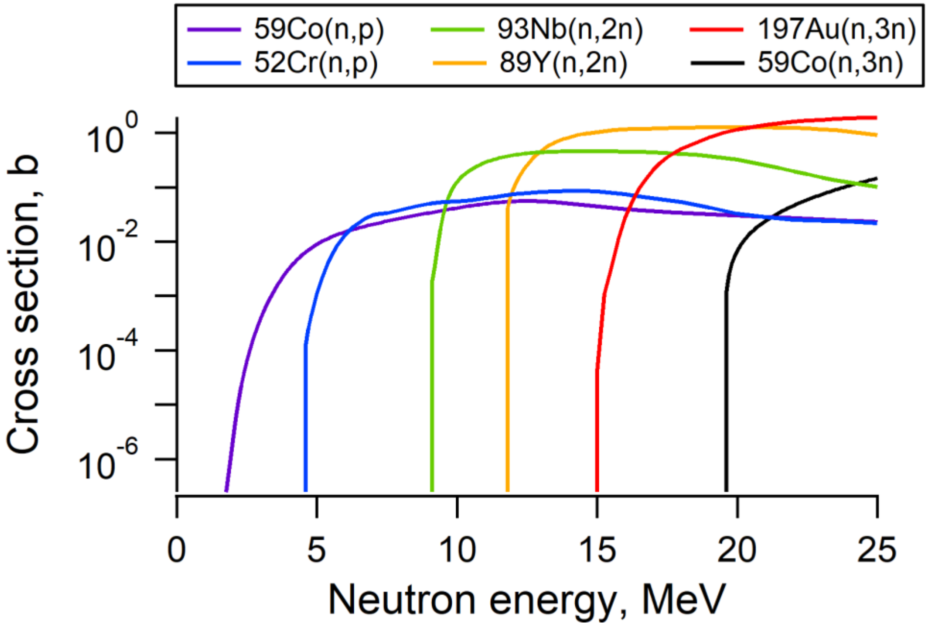


Candidate port with direct line (red) to HMMA (lower right corner).

Rapid sample retrieval at ignition yields would open up many potential nuclear data measurements relevant to programs
Examples include independent fission yields, multiple order processes, excited state reactions, and activation cross sections

Activation foils provide “spectra” for integrated experiments

- Foils comprised of isotopes with neutron cross section thresholds
- Using reaction sensitivity to neutron energy, “unfold” spectrum



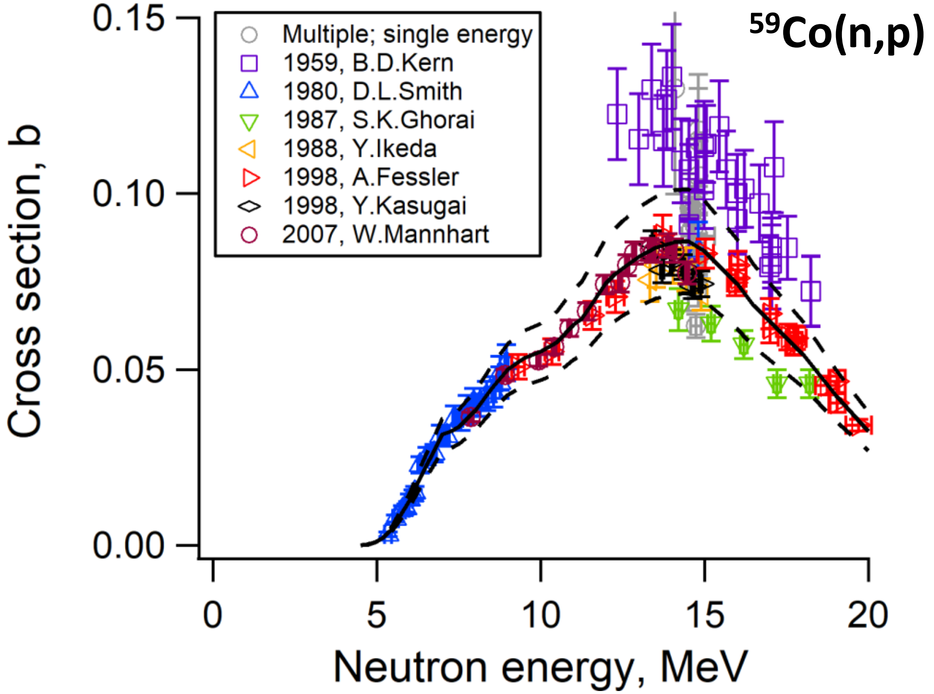
Cu, Au, Co, Fe, and Ti

a) Godiva critical assembly; b) J. Goda loading a sample holder; c) samples and witness pack

Activation foils are an important diagnostic for integrated systems; low cost and simple to field

Options for unfolding algorithms exist, but there is room for improvement

- Different methods suggest different neutron spectra when unfolded
  - Need common benchmark to test and validate options
- Few methods include uncertainties
- Do any include reaction data uncertainties?
  - Uncertainties missing for some candidate reactions, e.g., ^{197}Au(n,3n)
  - Uncertainties may need to be revisited even if they exist



Uncertainty for ^{59}Co(n,p) reflects scatter in the data rather than uncertainty in the cross section?

Future Investment Recommendations

- Radiochemistry
  - Tracers used in extreme environments – need well known nuclear data and defined radiochemical procedures
  - Tracer production, expand nuclear counting capabilities, R&D
- Facility upgrades to enable rapid sample retrieval
  - Necessary for short-lived reaction products related to independent fission yields, multiple order processes, and excited state reactions
- Improvement of reaction cross section measurements to improve uncertainties
  - Radioactive species for tracers, stable targets used as activation foils

Lawrence Livermore National Laboratory

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