Cross Section Measurements Of $^{17,18}O(\alpha,n)^{20,21}Ne$ And $^{27}Al(\alpha,n)^{30}P$ For Nuclear Safeguards

Rebecca Toomey

LLNL-PRES-2015740
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LLNL (α,n) Team

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- And collaborators:

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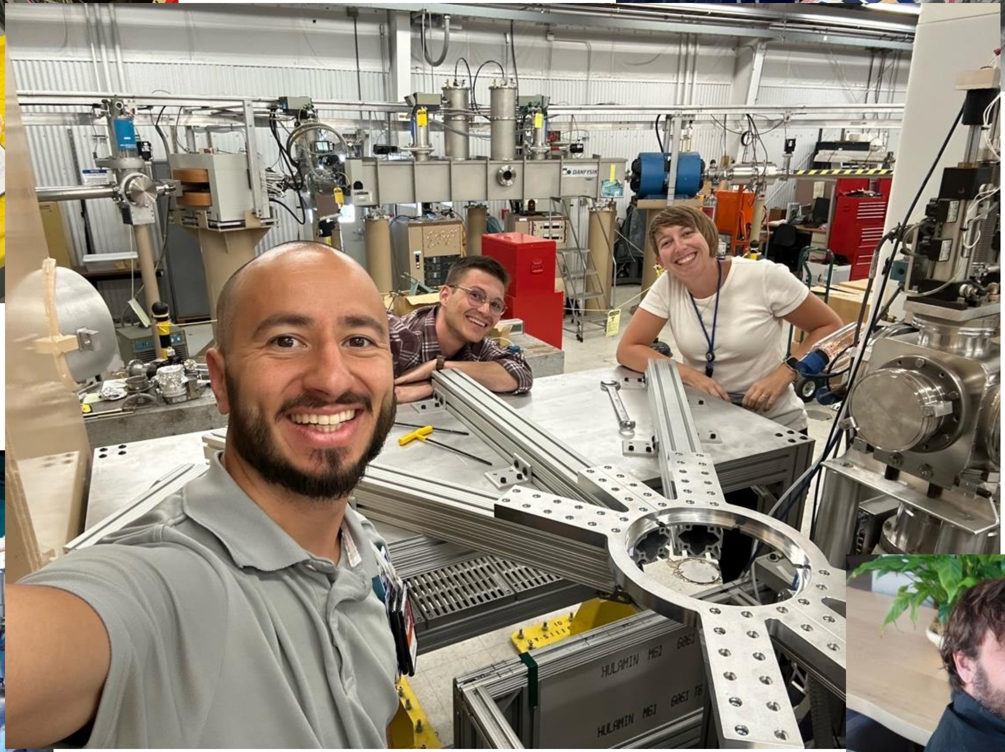
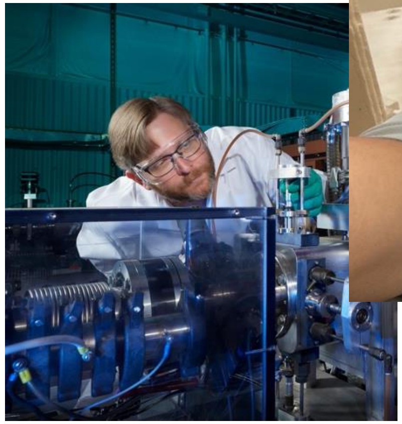
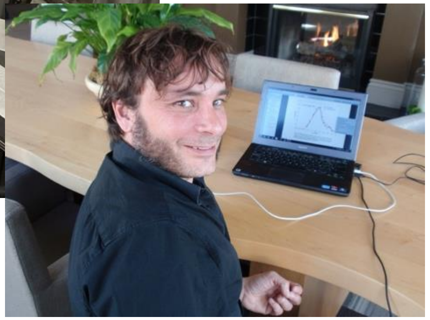
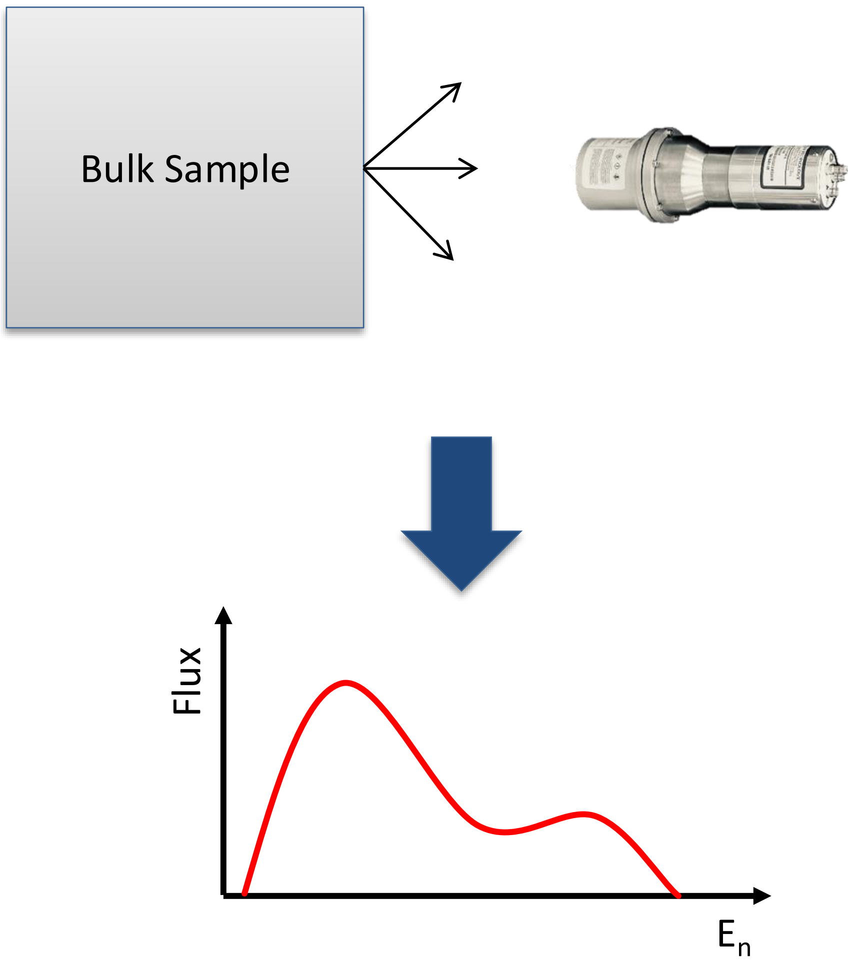






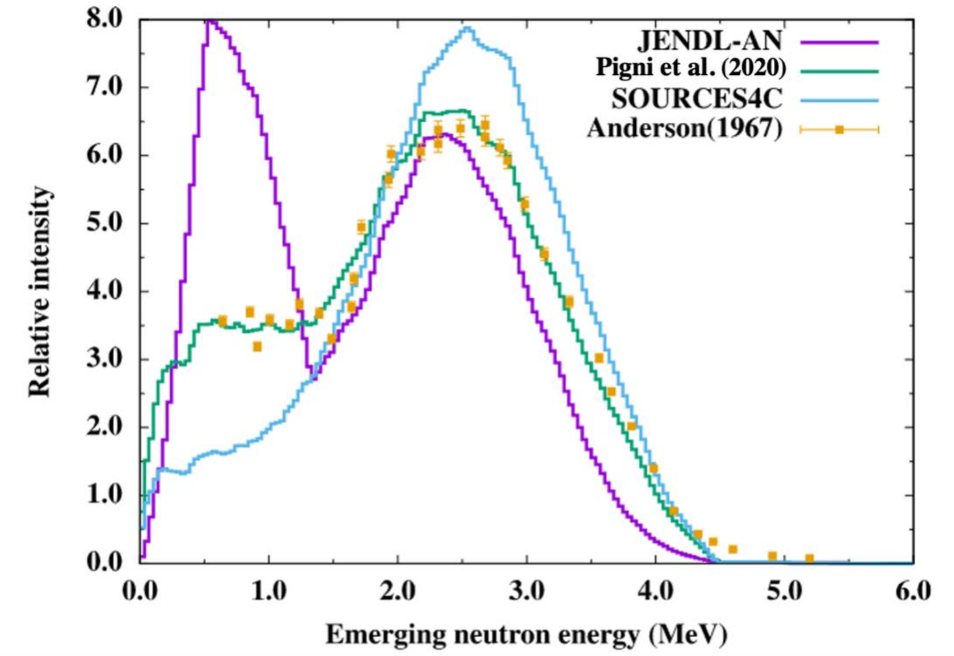
Why are (α,n) reactions important for nonproliferation?

- Nondestructive Analysis (NDA) provides a quick and inexpensive assessment of Special Nuclear Material (SNM) in a bulk sample
- Understanding the neutron emission spectra from SNM improves NDA measurements
  - Neutron source calculations to predict NDA spectra rely heavily on nuclear data
- Neutron energy spectra of SNM will be heavily determined by contributions from excited states
  - Partial cross sections crucial for confidence identifying SNM



Why are (α,n) reactions important for nonproliferation?

- Nuclear fuel oxides matrices e.g. UO_2 or PuO_2
  - Low burnup fuels for weapons-grade materials, $^{17,18}O(\alpha,n)^{20,21}$ reactions identified as dominant components[1]
  - Actinide alpha decay on oxygen in the matrix
- For aluminum matrices containing transuranic wastes, the $^{27}Al(\alpha,n)^{30}P$ reaction is important
  - Also, structural components in molten salt reactors
- Nuclear data evaluations are based on experimental data with models to fill in the gaps
  - Different models use different assumptions

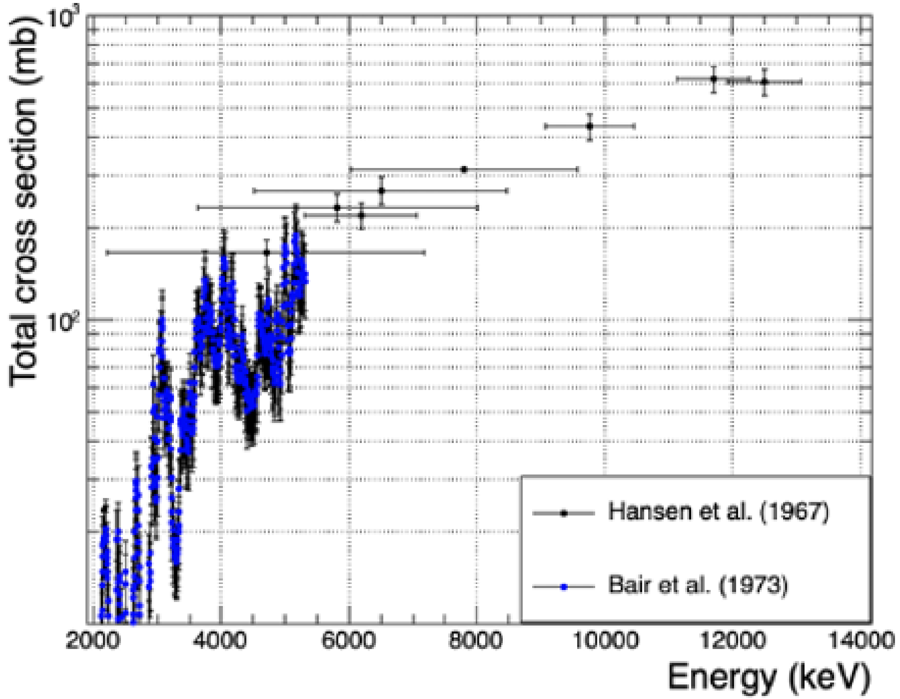


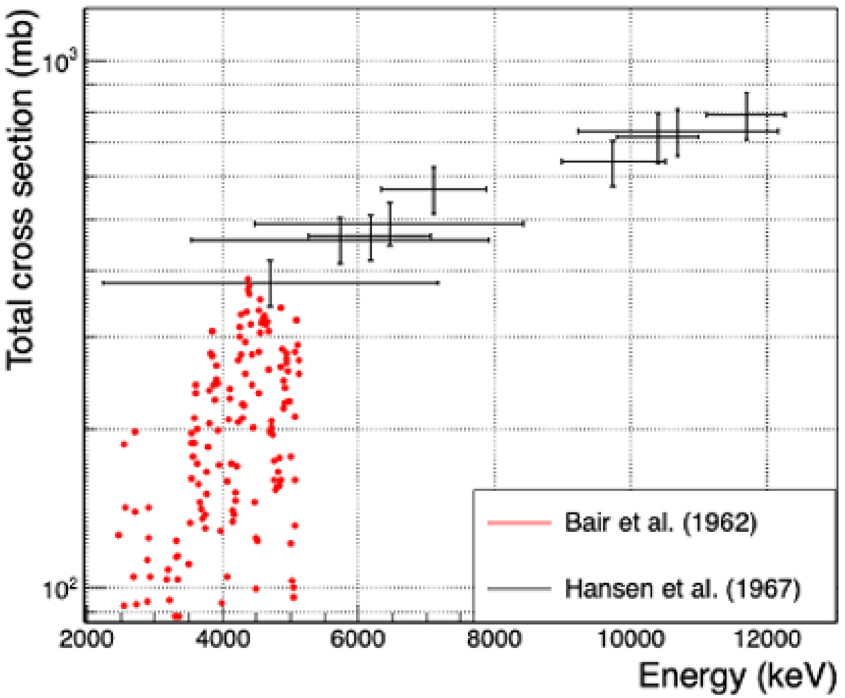
[1] M.T. Pigni et al., Prog. Nucl. Energy 118 (2020)

Note: $^{16}O(\alpha,n)^{19}Ne$ threshold is E_α ~ 15 MeV

Current state of existing data… $^{17,18}O(\alpha,n)^{20,21}Ne$

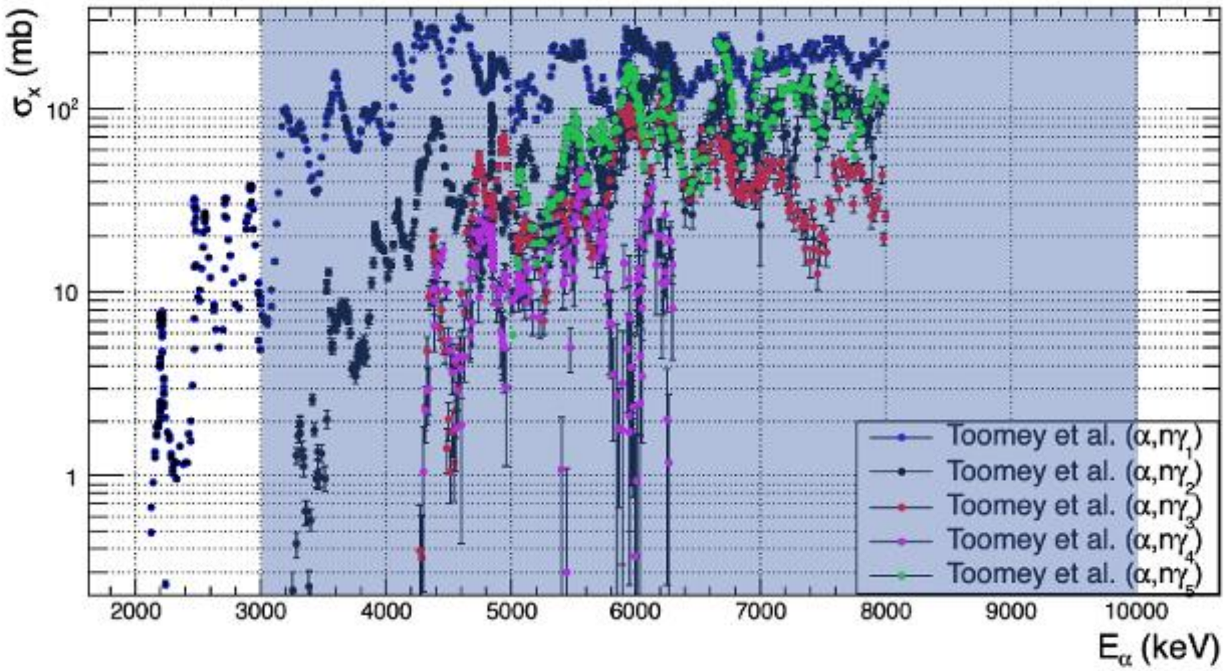
- Mostly total cross sections that are >50 years old
  - No excited state partial cross sections or angular distributions
- Large uncertainties in region of interest for actinide alpha decay ($3<E_\alpha<10$ MeV)
  - ~10% in cross section, but large energy uncertainty with washed out structure

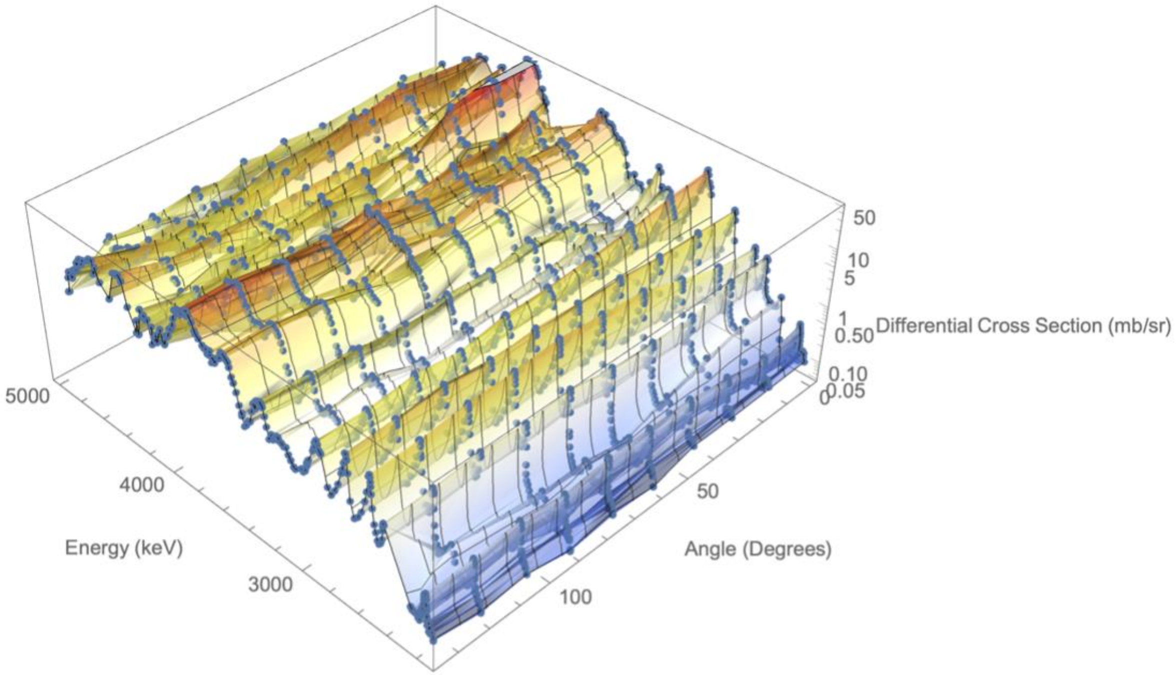




Current state of existing data… $^{17,18}O(\alpha,n)^{20,21}Ne$

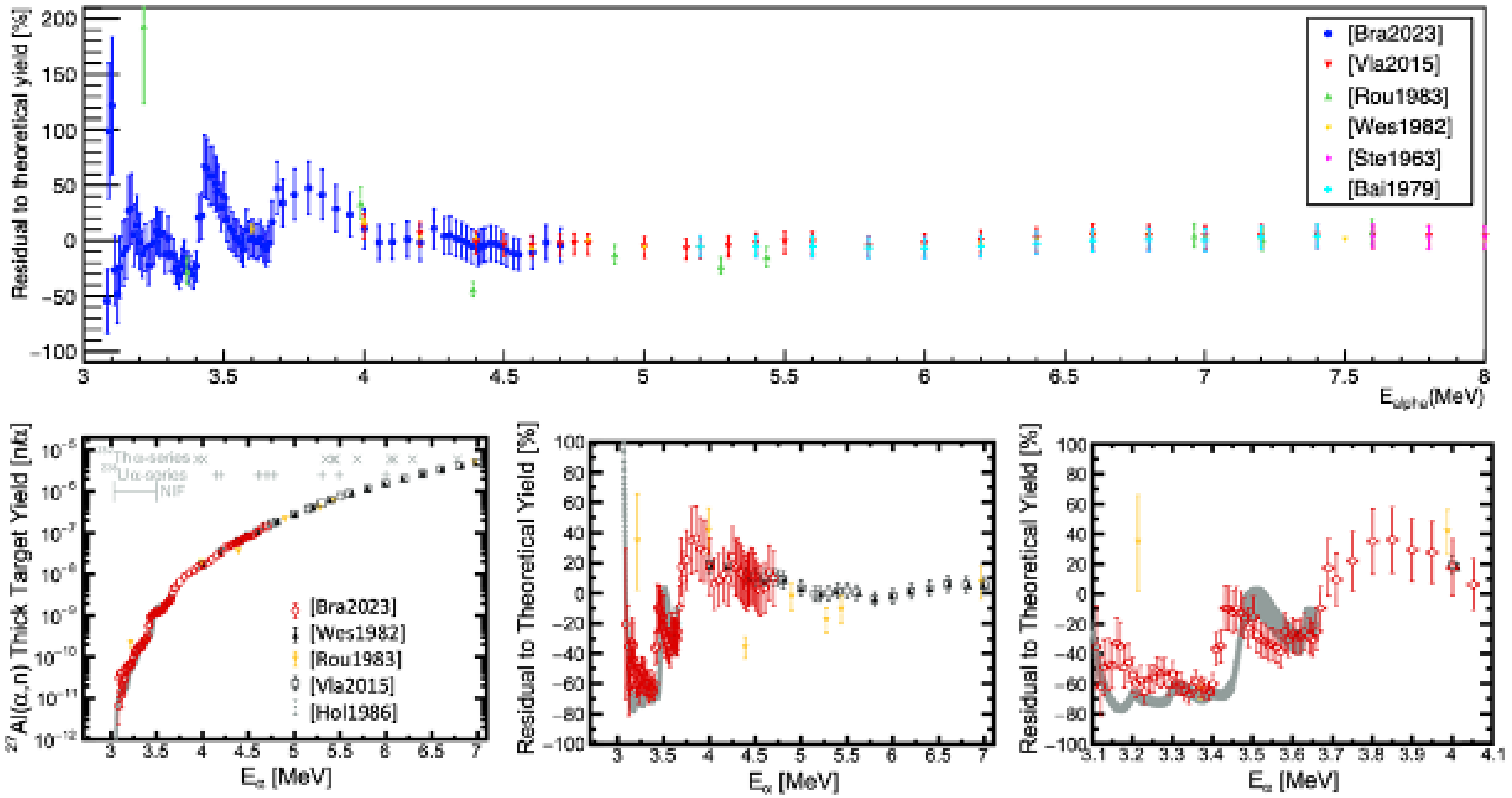
- Recent measurement of $^{18}O(\alpha,n)^{21}Ne$ in 2019 at Notre Dame
  - Primary neutron and secondary gamma ray spectroscopy
- Didn't cover full energy range of interest and angular distribution analysis ongoing
  - Can be used to benchmark our measurements





Current state of existing data… $^{27}Al(\alpha,n)^{30}P$

- Six thick-target yield datasets
- Only 2 datasets E_α~3 MeV and they have large disagreement
- Most recent measurement in 2023 highlights discrepancy of up to 40% for E_α< 5 MeV



We will measure the $^{17,18}O(\alpha,n)^{20,21}Ne$ and $^{27}Al(\alpha,n)^{30}P$ reactions

- Objective 1: Establish high-resolution (α,n) capability at LLNL
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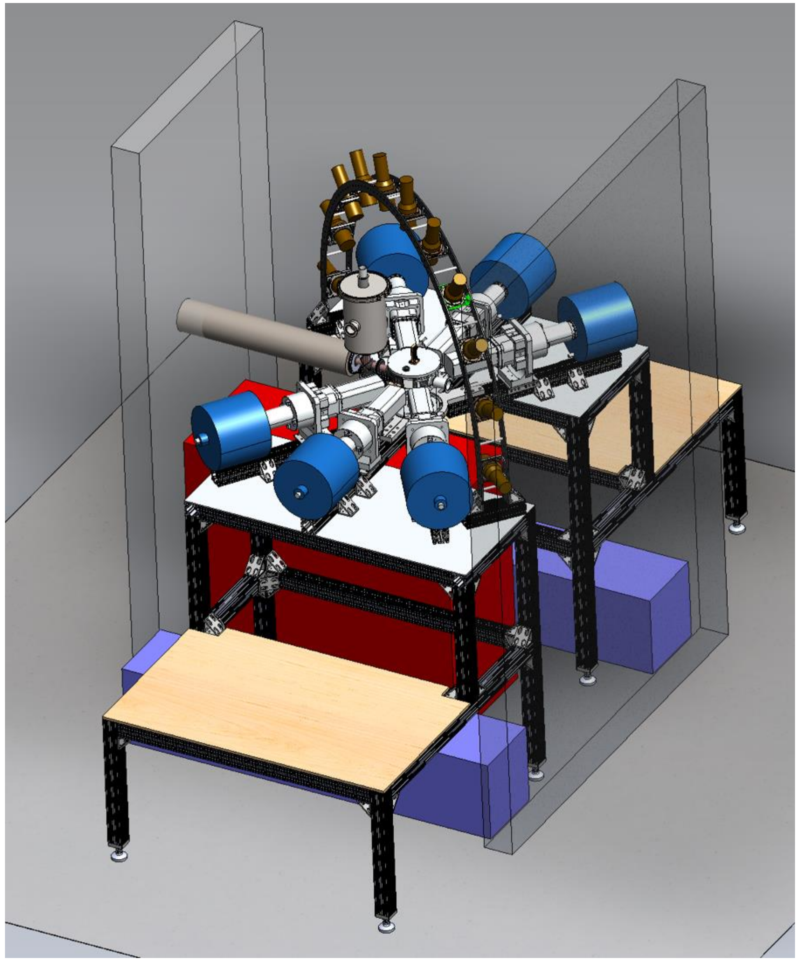

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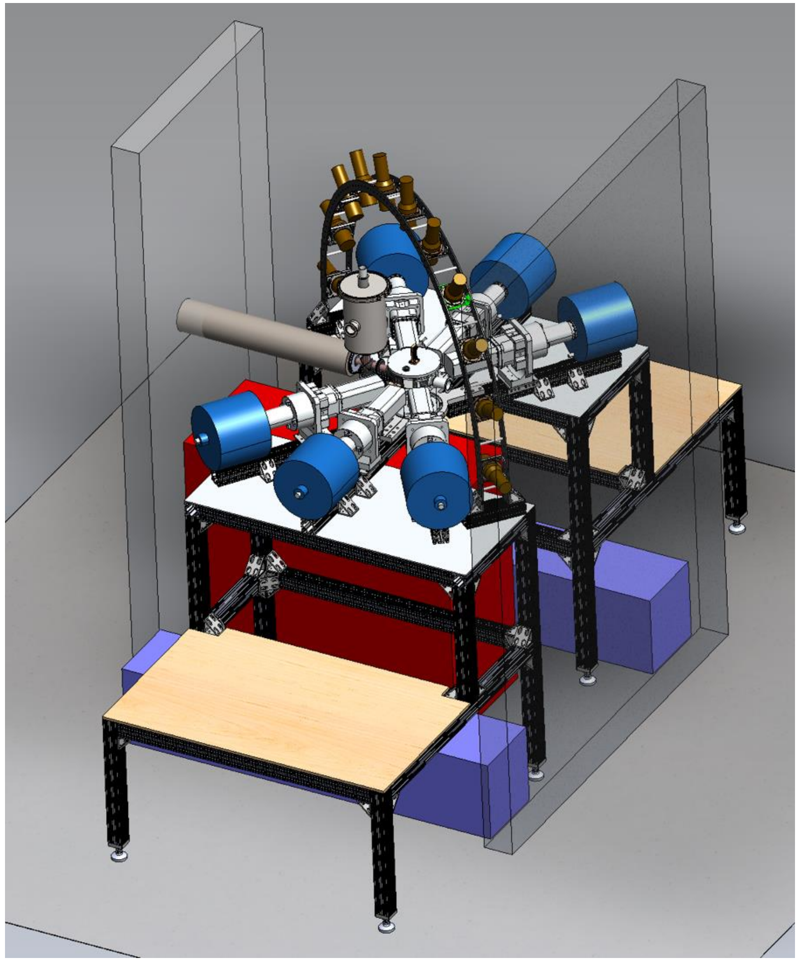
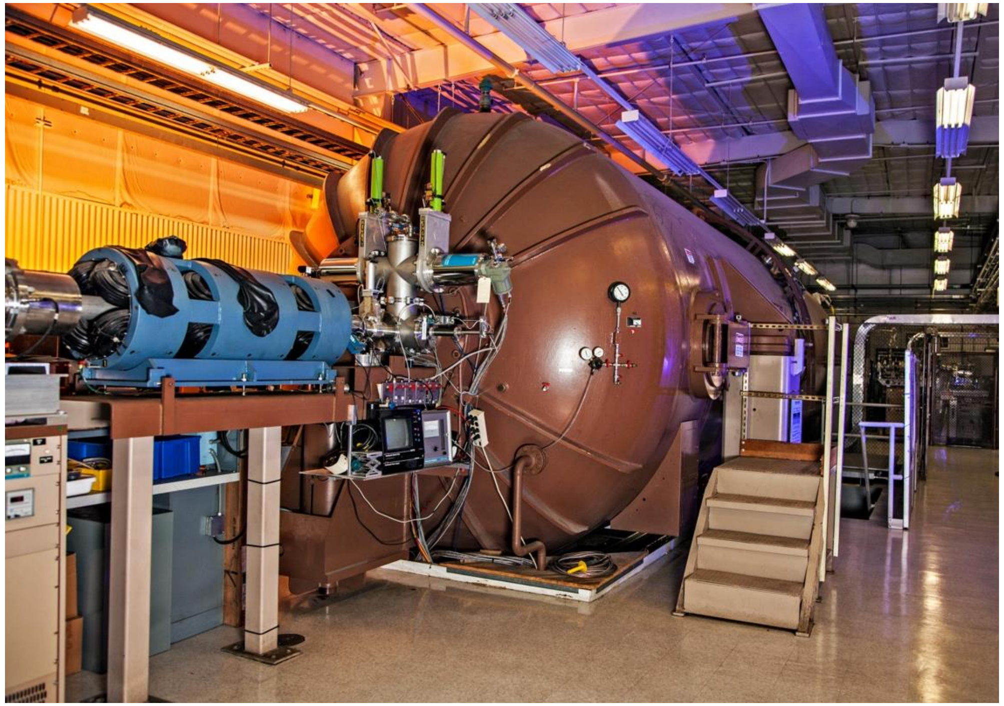


The Center for Accelerator Mass Spectrometry

- 10 MV FN model tandem accelerator
  - Electrostatic accelerator (~1 kV noise)
  - 4 ion sources, cesium sputter and plasma
  - 4 beamlines, 1 multi-use end station
  - 2-18 MeV p/d ; 3-27 MeV alphas
  - AMS analysis
    - $^{7/10}Be$, ^{14}C, ^{26}Al, ^{36}Cl, ^{60}Fe, ^{90}Sr, ^{129}I, ^{236}U, ^{240}Pu
- Online- and offline nuclear science
  - Excitation functions
  - Production yields
  - Light-ion induced fission distributions



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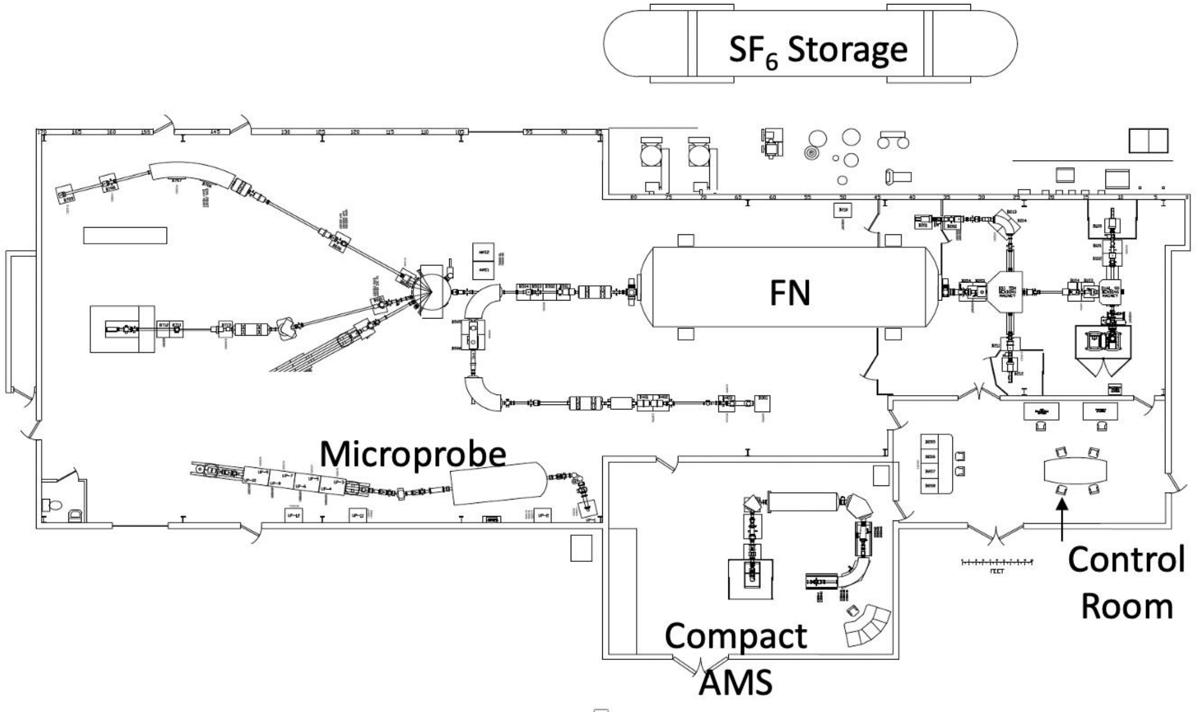
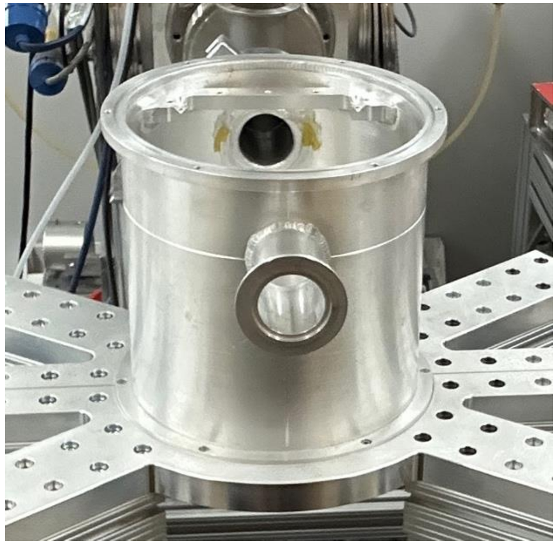


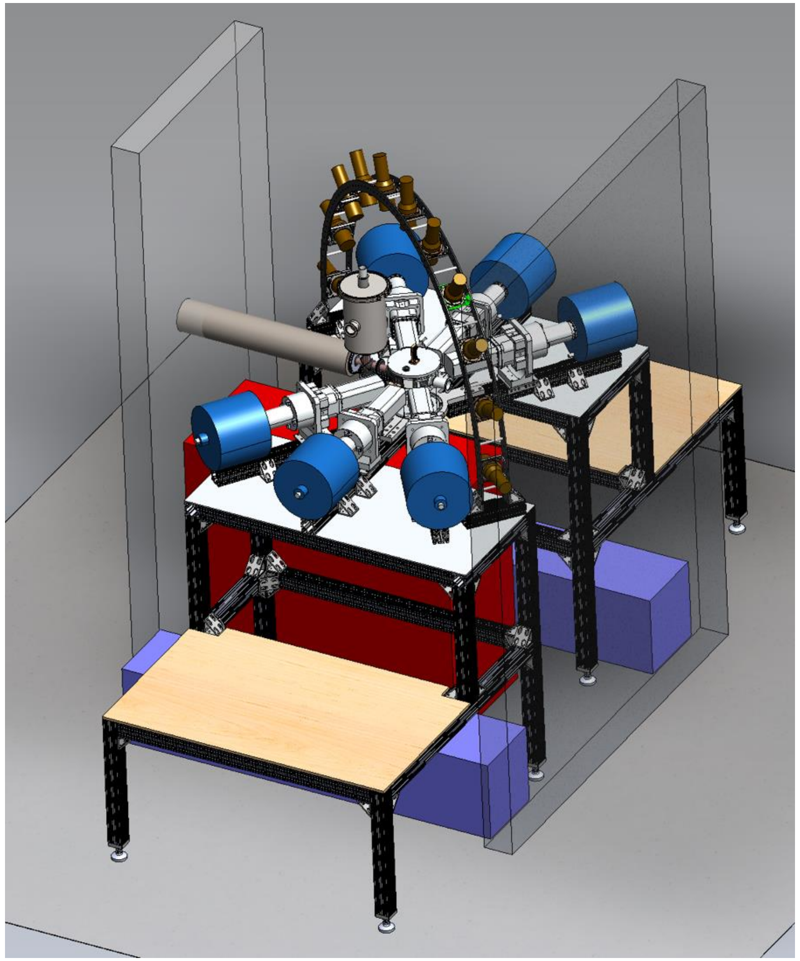
Utilizing Existing Arrays

- 11 deuterated scintillators for neutron spectroscopy
  - 5 EJ301D and 6 EJ315M – 3”x3”
  - Neutron spectroscopy via spectrum unfolding
- 6 Hyperion HPGe clovers for gamma ray spectroscopy
  - 4 leafs – 5 cmx5 cmx8 cm
  - 150% relative efficiency
- STARS target chamber
  - Thin-walled aluminum



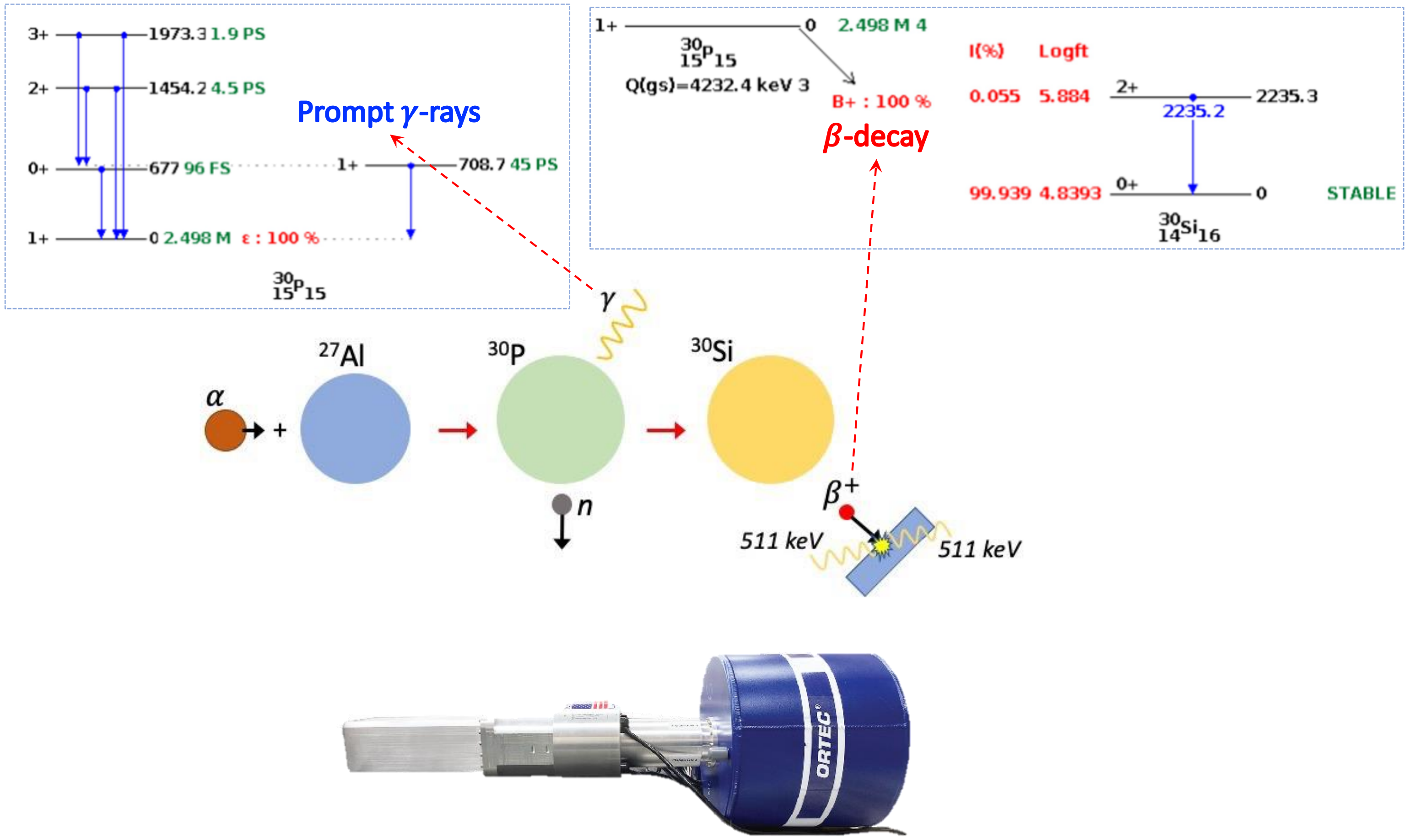
Experimental Setup

- 11 neutron detectors
  - 38-142° angular coverage at 32” distance
- 6 HPGe clovers
  - Fixed angle 55°, 90°, 125°
  - Adjustable distance
- Array of 4 beta detectors
- Stepper motor for target ladder
- Cold finger to mitigate carbon buildup
- Thermal camera to monitor target



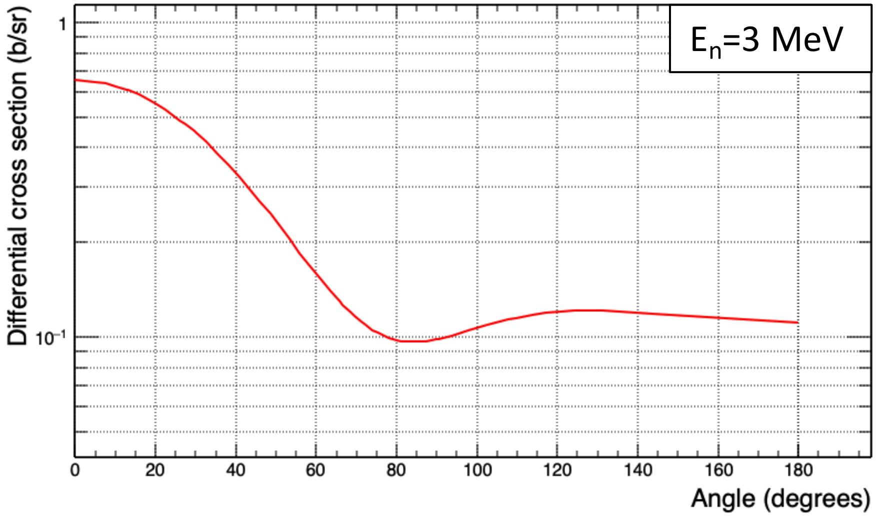
$^{27}Al(\alpha,n)^{30}P$ Experimental Design

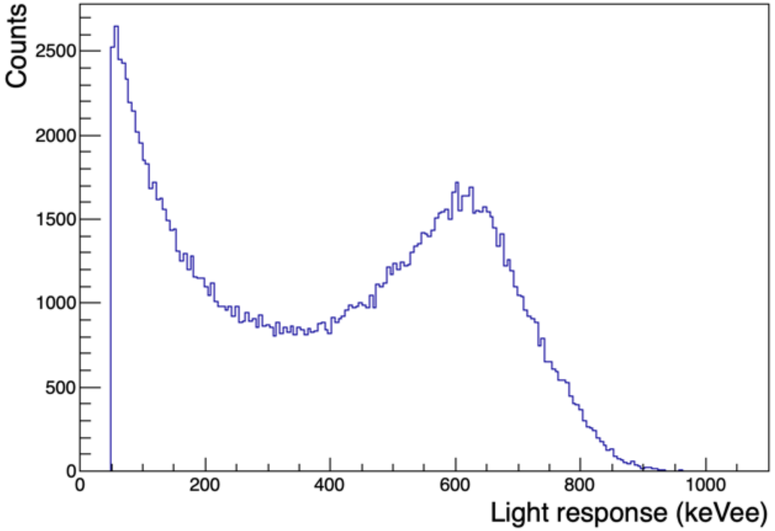
- Direct method – beam on
  - Thick target yield/cross section directly extracted from prompt reaction neutrons/gamma rays
- Activation method – beam off
  - Number of reaction events determined by measuring the 511 keV decay curve from the reaction product, ^{30}P



Spectrum unfolding – how does it work?

- Deuterated organic liquid scintillators
  - Non-isotropic elastic scattering cross section
- Forward-going recoil produces characteristic feature
  - Directly related to incident neutron energy ($E_{d,max} = {}^{8}/_{9}E_n$)
- Can deconvolve incident neutron energy spectrum using known detector response using a maximum-likelihood expectation maximization (MLEM) algorithm

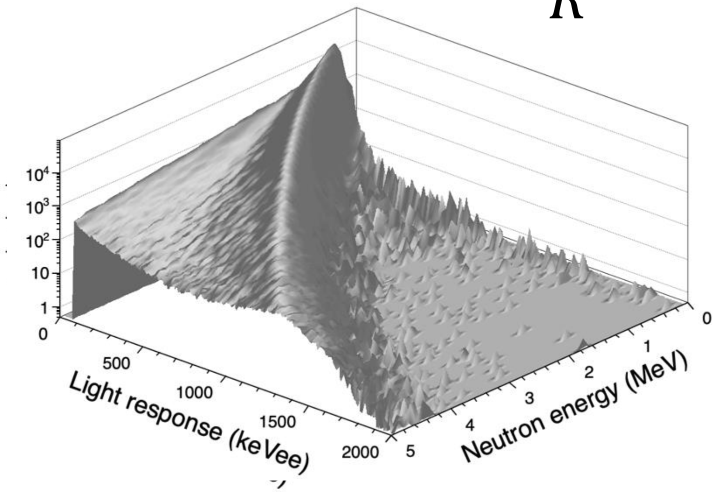




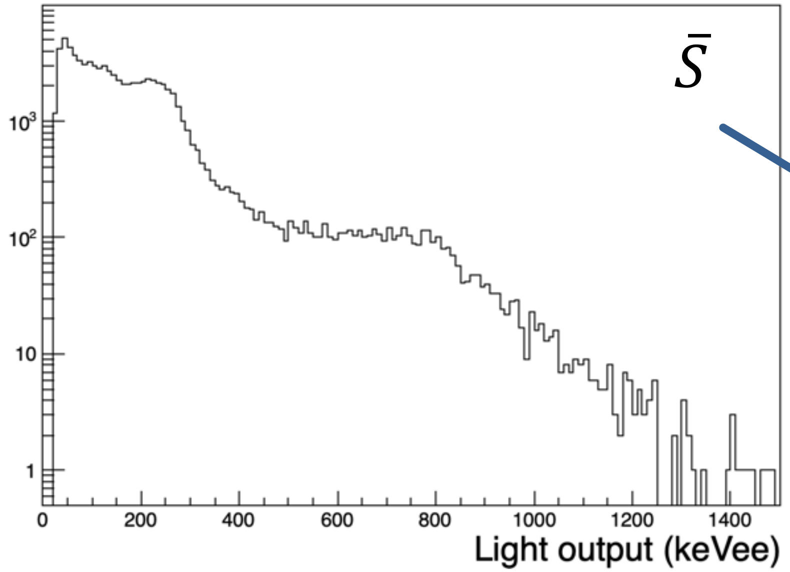
Spectrum unfolding – how does it work?

- Using MLEM and the detector response matrix, $\bar{\bar{R}}$, a neutron energy spectra can be extracted
  - Able to extract spectroscopic information independent of ToF

$\bar{\bar{R}}$

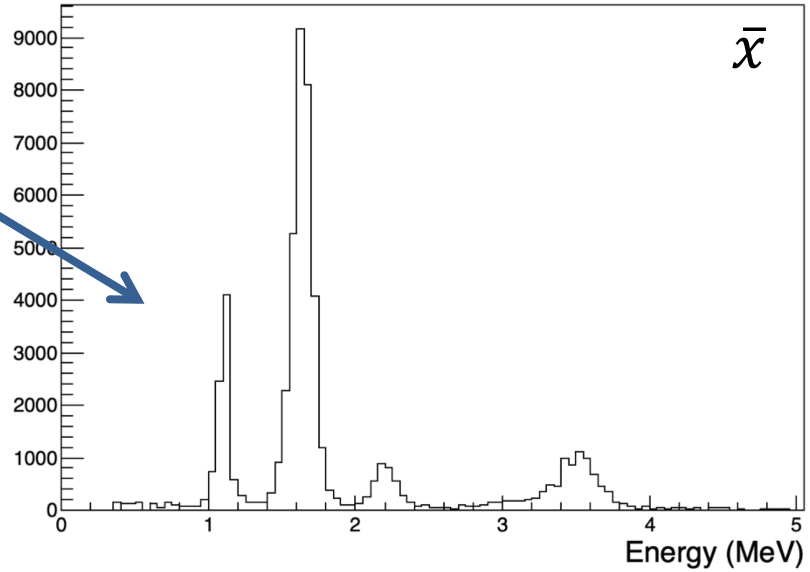


$\bar{S}$



$\bar{\bar{R}}^{-1}$

$\bar{x}$



$$\bar{S} = \bar{\bar{R}}\bar{x}$$

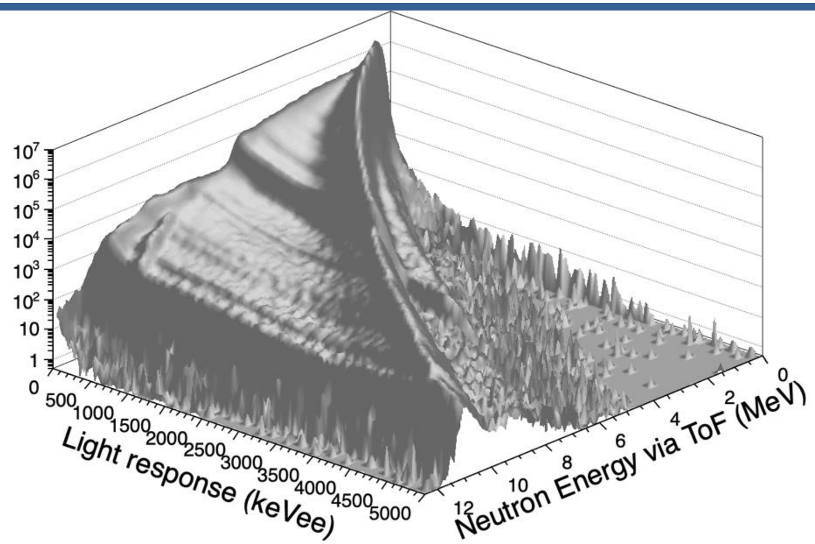
$\bar{S}$ = Measured light spectrum

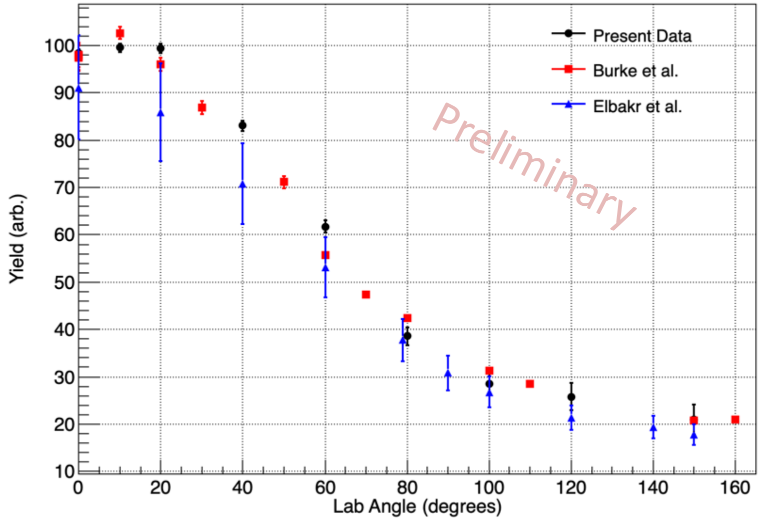
$\bar{\bar{R}}$ = Response matrix

$\bar{x}$ = Neutron energy spectrum

Deuterated Detector Characterization

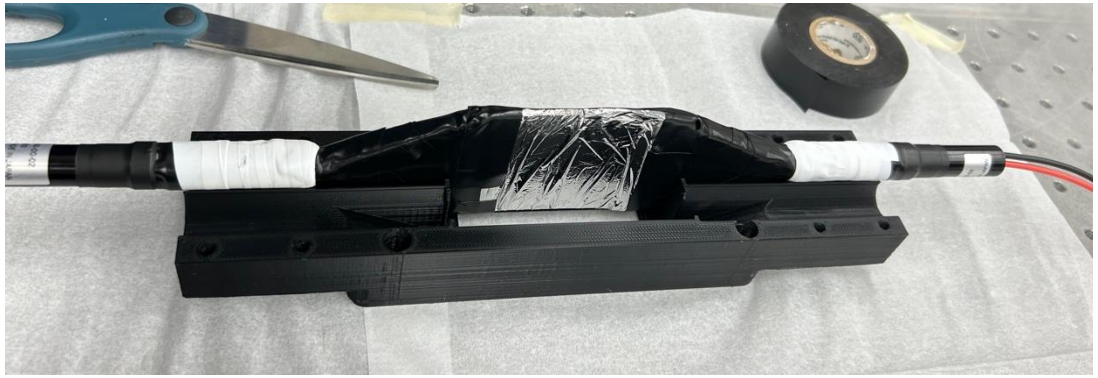
- Detectors fully characterized at Edward's Accelerator Laboratory
- Detector response matrix up to E_n ~12 MeV
  - Light response, resolution, intrinsic efficiency etc.
- Also measured ^{7}Li(p,n) angular distributions
  - Commonly used for neutron scattering
  - E_p = 3.6 MeV and E_p = 4.0 MeV

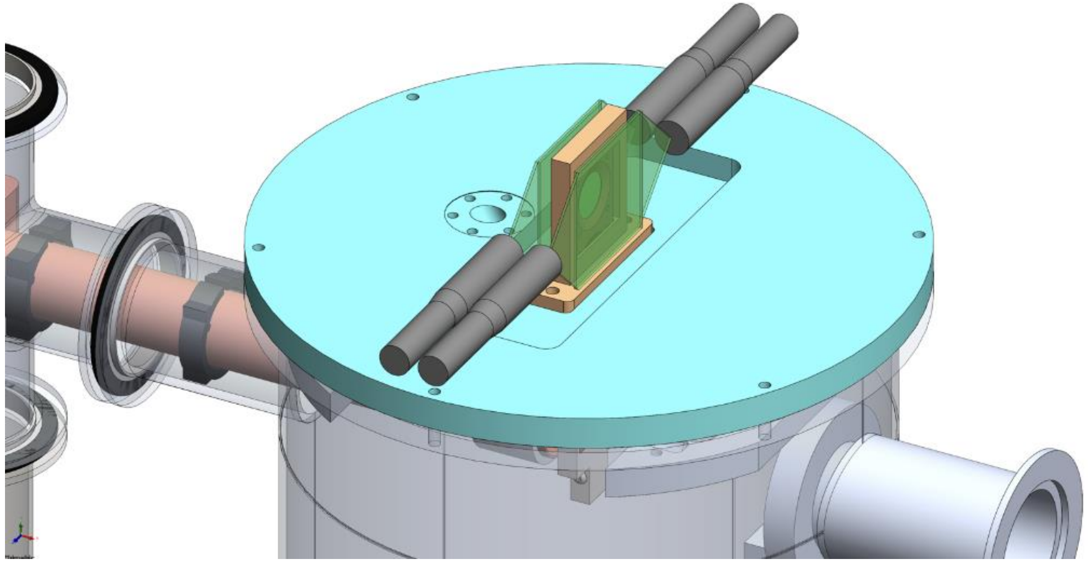


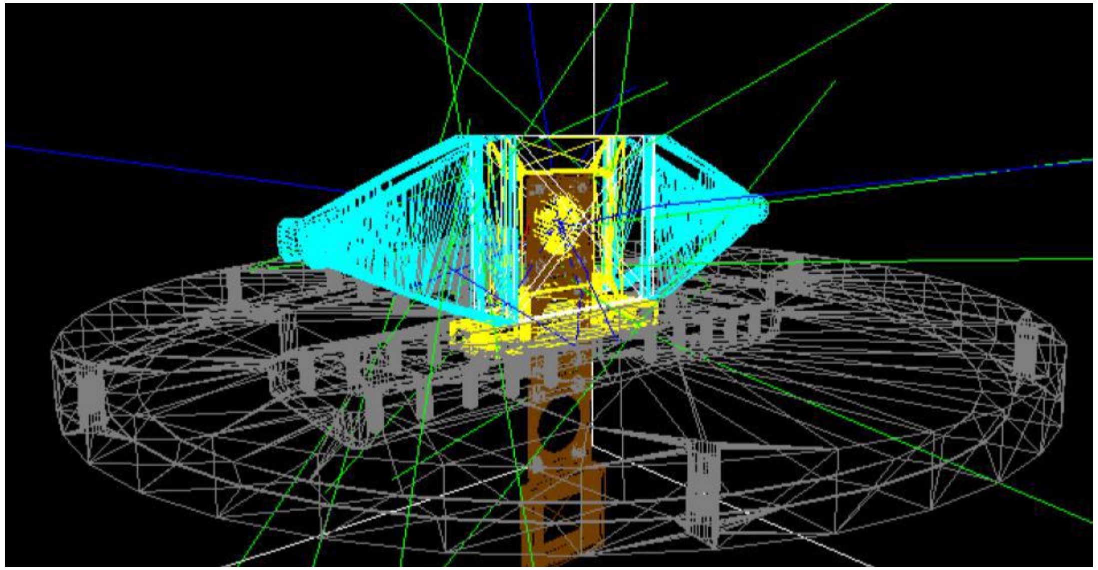


Beta Detection

- Beta detection consists of 4 EJ-212 paddles with fishtail light guides
  - 50 x 53 x 1 mm
  - 2 paddles mounted each side of thin target window







Enriched Oxygen Target Fabrication

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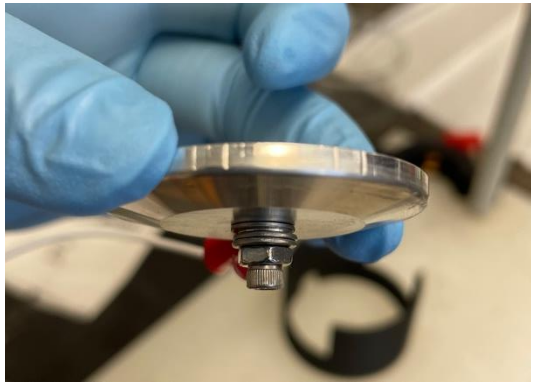
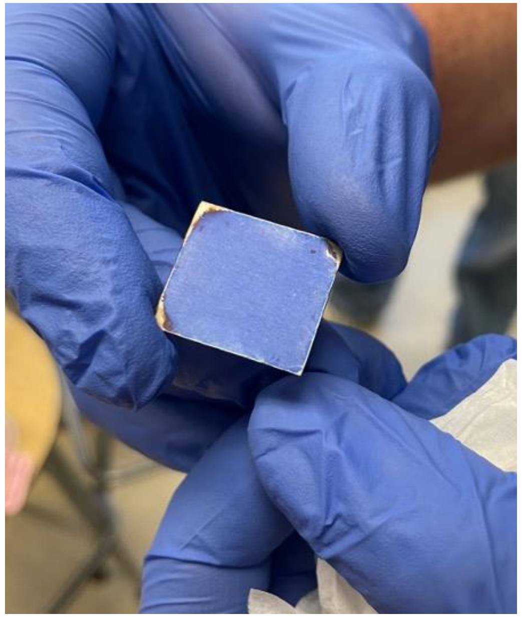
- Isotopically-enriched $Ta_2{}^{17,18}O_5$ via anodization

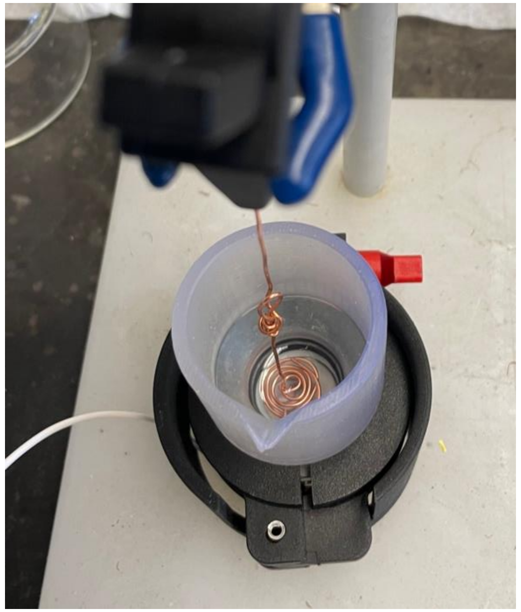
- To minimize fluorine contamination we have developed an etching process for the tantalum backings
  - 1 Molar ascorbic acid @ 90°C for 65 minutes
  - Rinse with DI water

- Etching process verified with Raman spectroscopy
  - See significant reduction in surface contaminants
  - Still identifying all features in spectrum

- Thickness analyzed via RBS at LLNL







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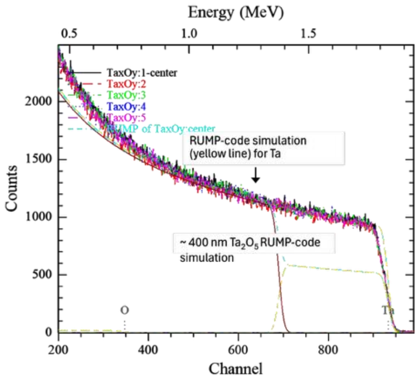
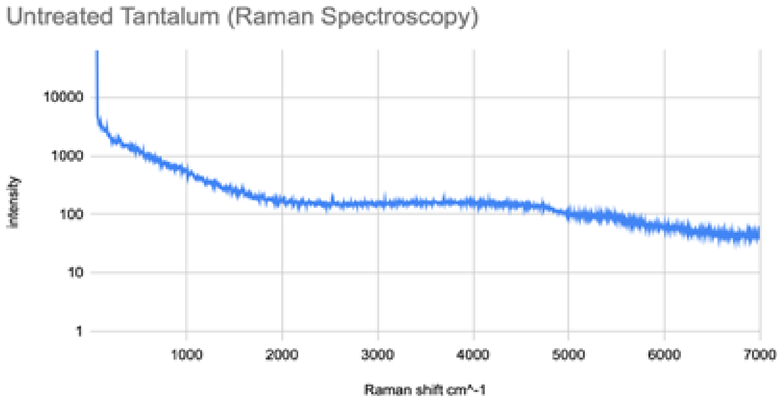
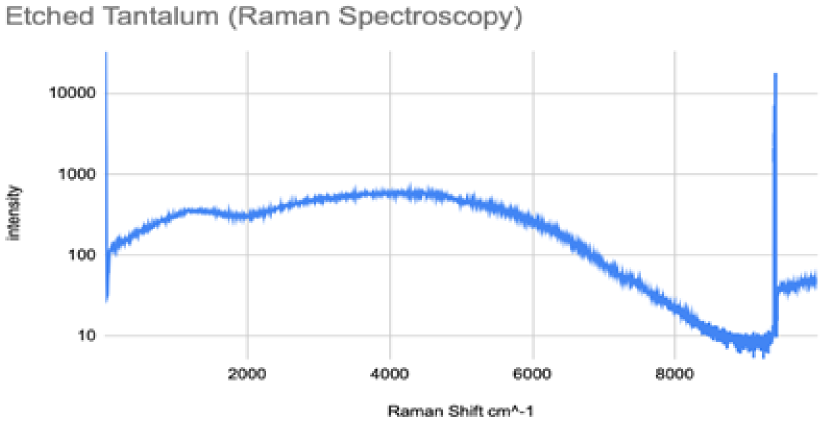
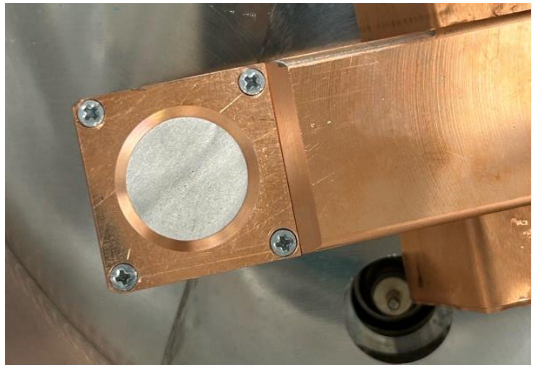


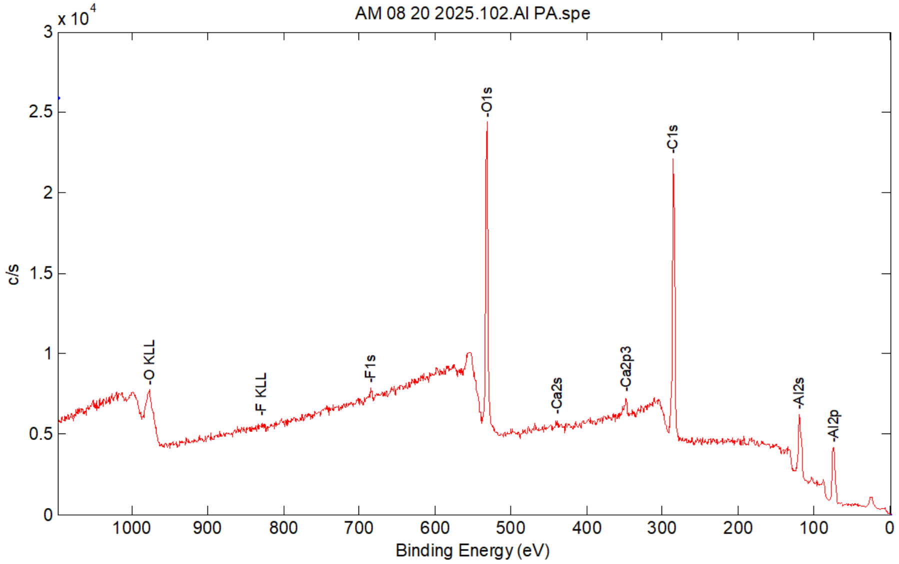




^{27}Al Target Characterization

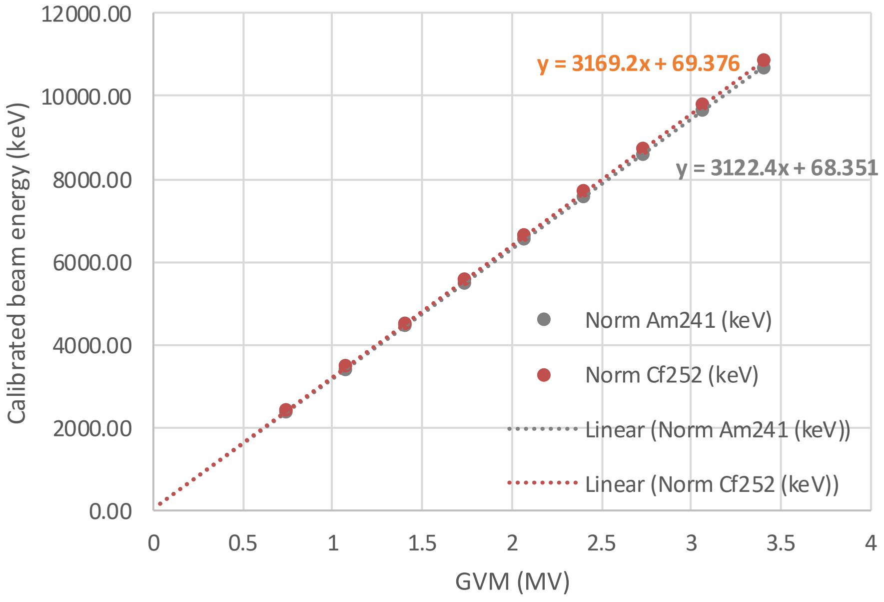
- Carbon buildup has been highlighted as a major issue in previous measurements
- 99.9999% purity
- 2cm dia x 0.5mm thick
- X-ray fluorescence shows expected oxide layer with a small quantity of carbon
- Prior to beam, target surface was argon plasma etched and kept in an inert environment before installation

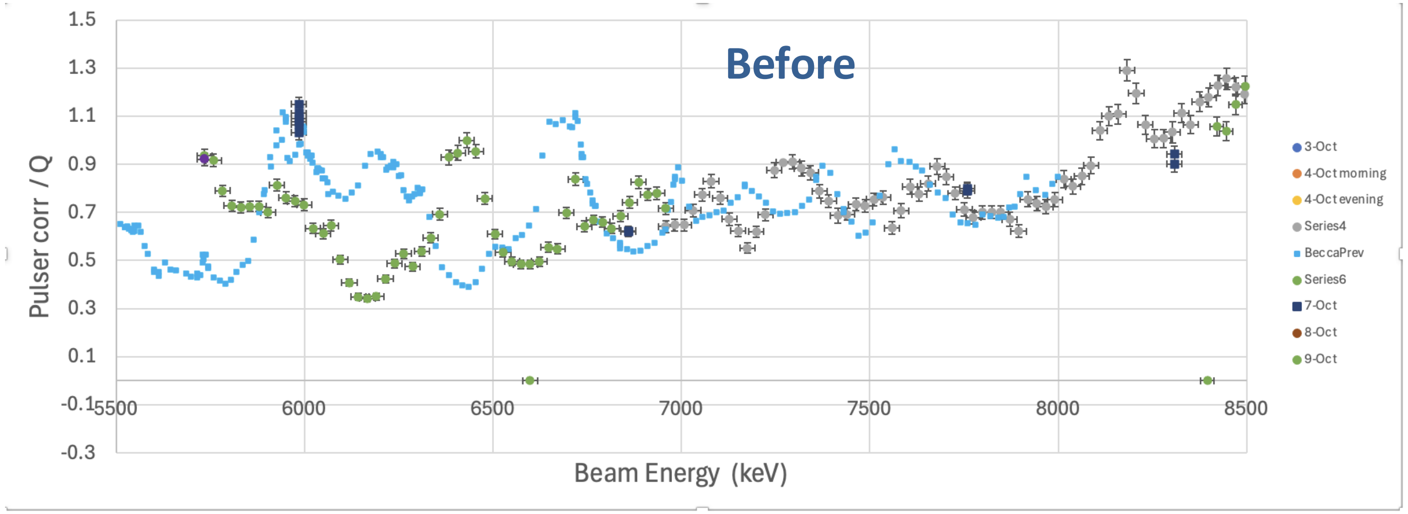


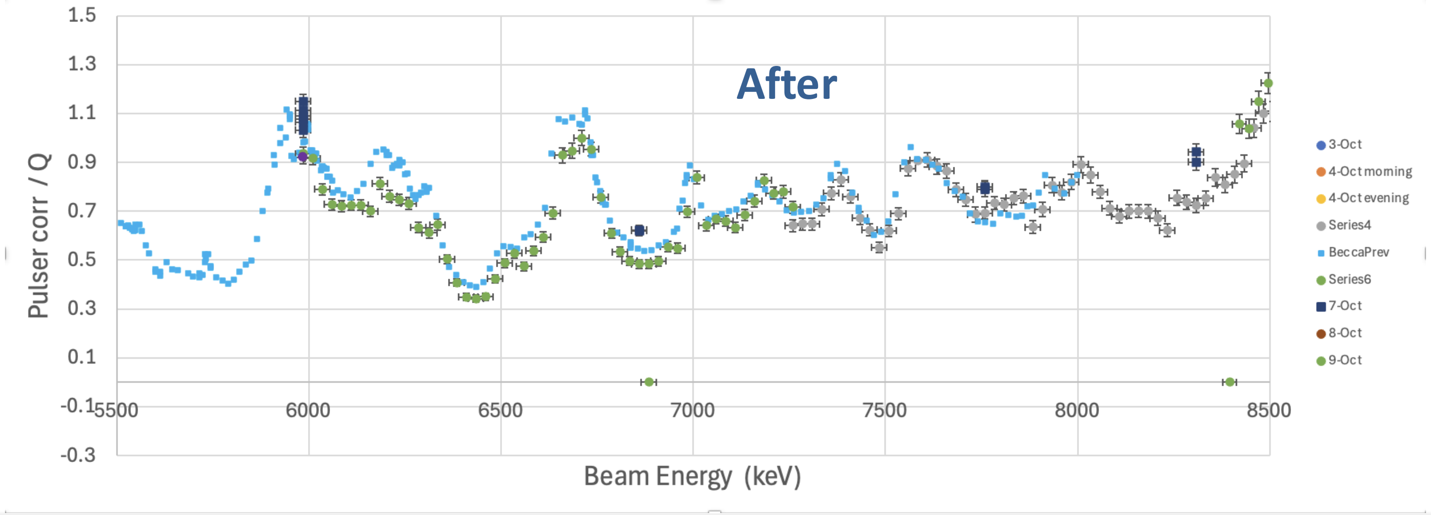


CAMS Beam Energy Calibration

- AMS is relative – precise beam energy (~1-3 keV) not needed
- Mounted silicon detector in beam to precisely calibrate beam energy







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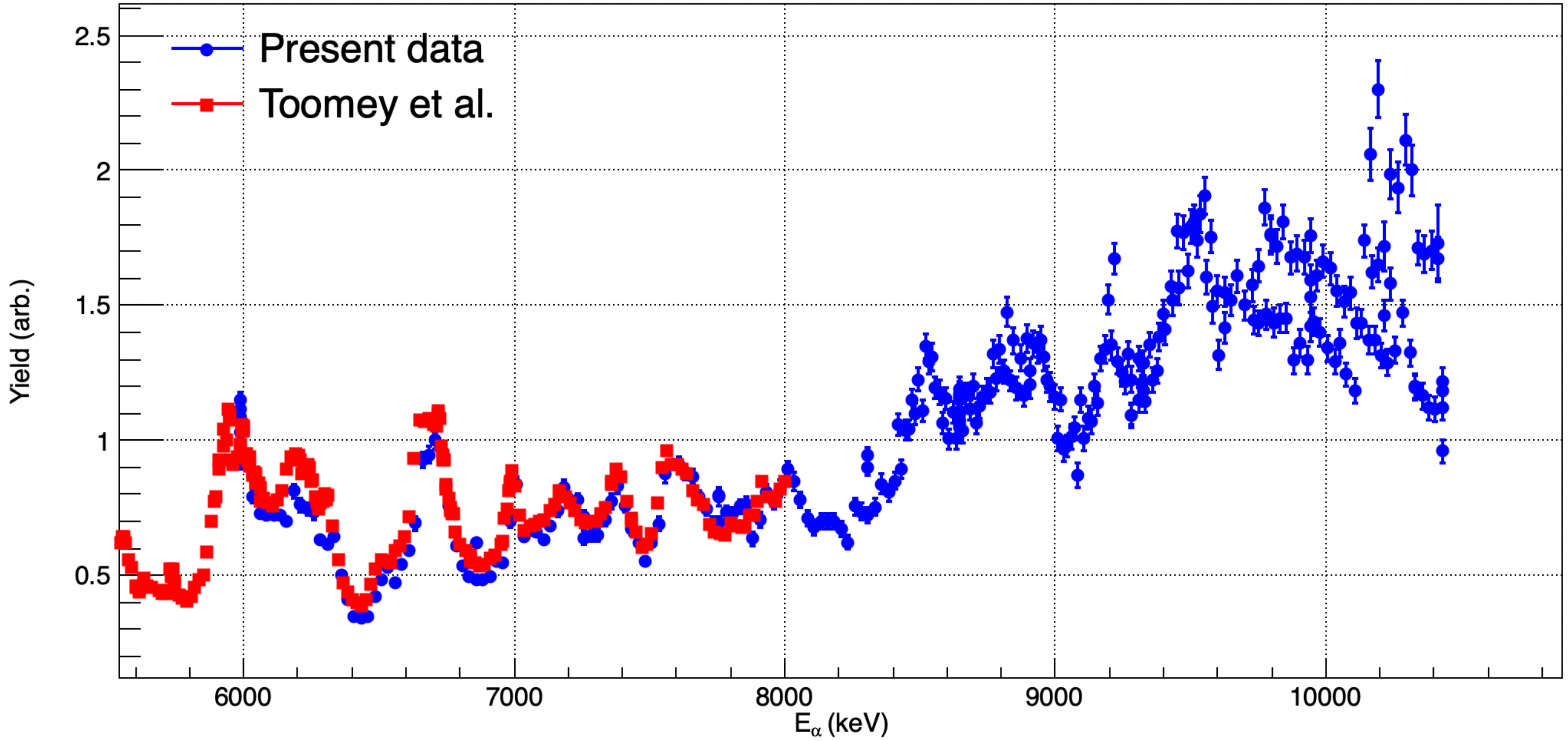
Measurement of the $^{18}O(\alpha,n)^{21}Ne$ Reaction

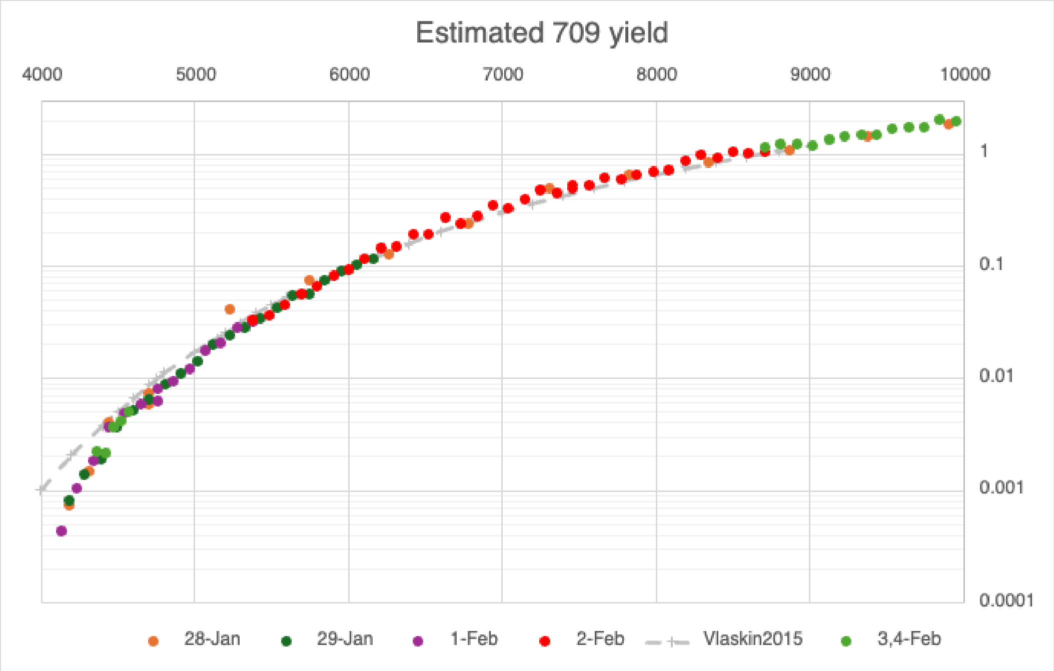
- Experiment to benchmark new high-resolution (α,n) capability
  - Lots of hurdles!

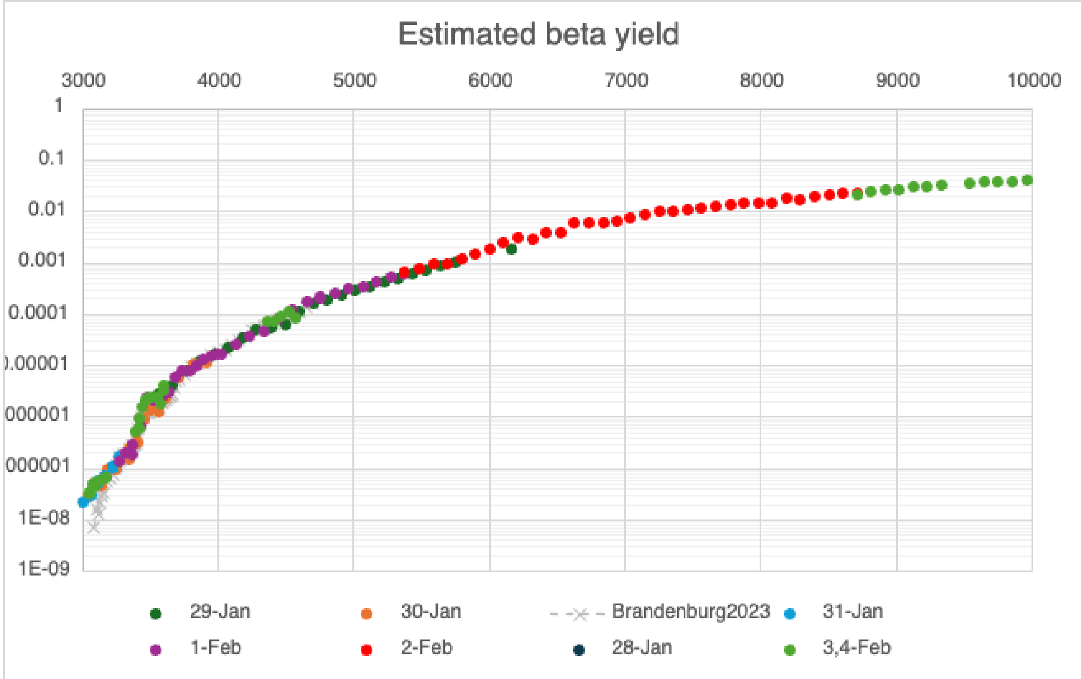
- Beam time: September 8th – 19th and September 29th – October 10th



Measurement of the $^{27}Al(\alpha,n)^{30}P$ Reaction

- Beam time: January 26th – February 4th 2026
- Measured $3 < E_{\alpha} < 10$ MeV
- Also observed (α,p) and (α,d) reaction channels





Summary and Outlook

- High-resolution (α,n) reaction nuclear data is important for NDA in the nuclear safeguards space
  - $^{17,18}O(\alpha,n)^{20,21}Ne$ and $^{27}Al(\alpha,n)^{30}P$ have been highlighted

- LLNL has successfully established a high-resolution (α,n) reaction capability
  - Characterized neutron detectors at Ohio University
  - Calibrated CAMS beam energy

- We have measured the $^{18}O(\alpha,n)^{21}Ne$ and $^{27}Al(\alpha,n)^{30}P$ reactions

- Next steps:
  - Analyze experimental data
  - Perform $^{17}O(\alpha,n)$ experiment – Late 2026
  - Assess data via R matrix analysis
  - Incorporate directly into SCALE for immediate use in safeguarding spaces

This work was supported by NNSA DNN R&D.