

# Nuclear Reaction and Decay Data for Medium Energy Radionuclide Production

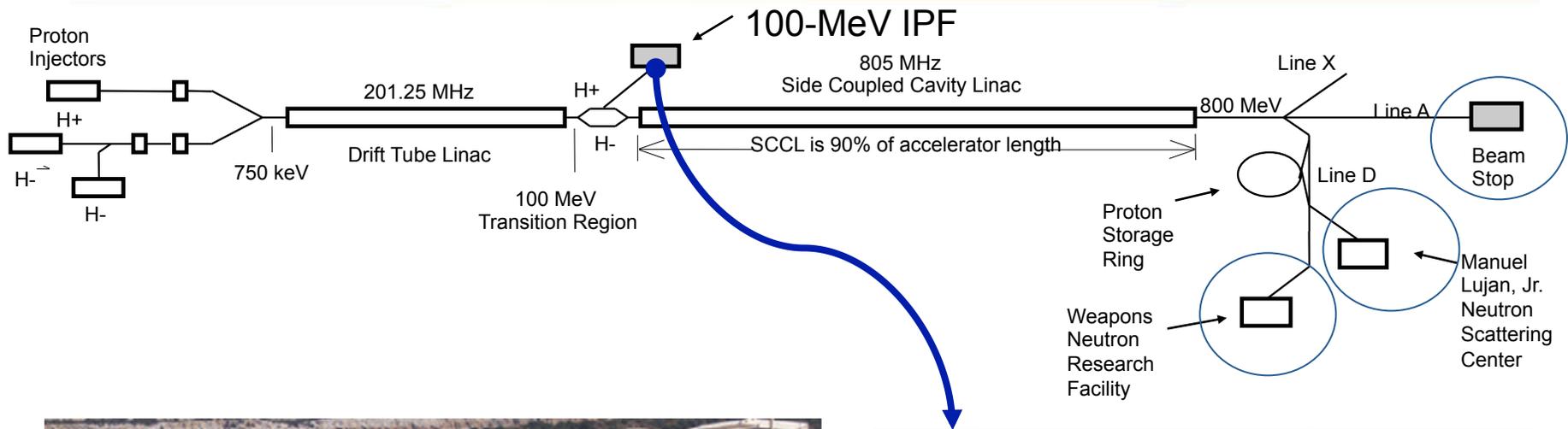
Jonathan W. Engle

Los Alamos National Laboratory, LA-UR 15-23573

Thursday, May 28<sup>th</sup>, 2015

# Medium-Energy Radionuclide Production

## Context at LANL IPF



IPF at LANSCE

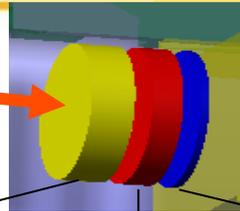


Slide 2

# Medium-Energy Radionuclide Production

Context at LANL IPF

100 MeV H<sup>+</sup>  
(240 μA limit)

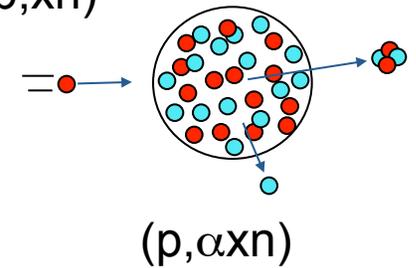
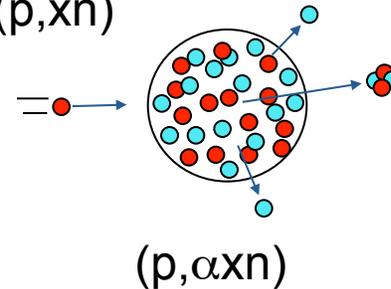
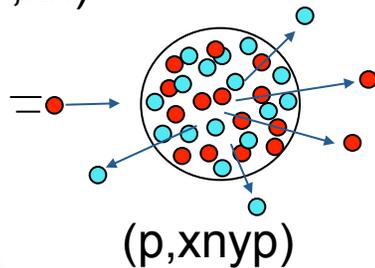
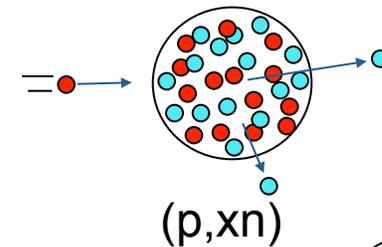
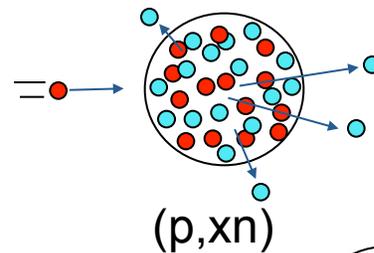
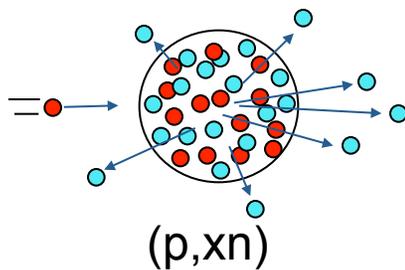


3 Stack Target Irradiation

High Energy  
(93-70 MeV)

Medium Energy  
(65-40 MeV)

Low Energy  
(31-0 MeV)



# Radionuclides of Interest at LANL

Red for those with nuclear data being actively or recently pursued.

Isotope	Half-life	Main Use
$^{82}\text{Sr}$	25.5 d	Parent of $^{82}\text{Rb}$ used in cardiac perfusion studies with PET
$^{68}\text{Ge}$	270 d	Parent of $^{68}\text{Ga}$ being tested for diverse PET applications
$^{22}\text{Na}$	2.6 a	PET isotope used as a tracer and sources material
$^{32}\text{Si}$	153 a	Environmental tracer; produced in partnership w/ TRIUMF
$^{73}\text{As}$	80.3 d	Tracer for toxicology studies
$^{109}\text{Cd}$	462.6 d	Source for X-ray fluorescence
$^{225}\text{Ac}$	10 d	Alpha emitter used in cancer therapy clinical trials
$^{186g}\text{Re}$	90.6 h	Bone pain palliation, cancer therapy
$^{44}\text{Ti}$	58.9 a	Generator for PET isotope $^{44}\text{Sc}$
$^{236}\text{Np}$	$1.5 \cdot 10^5$ a	Standard for Np quantification by IDMS
$^{119}\text{Sb}$	38.5 h	Auger emitter for cancer therapy

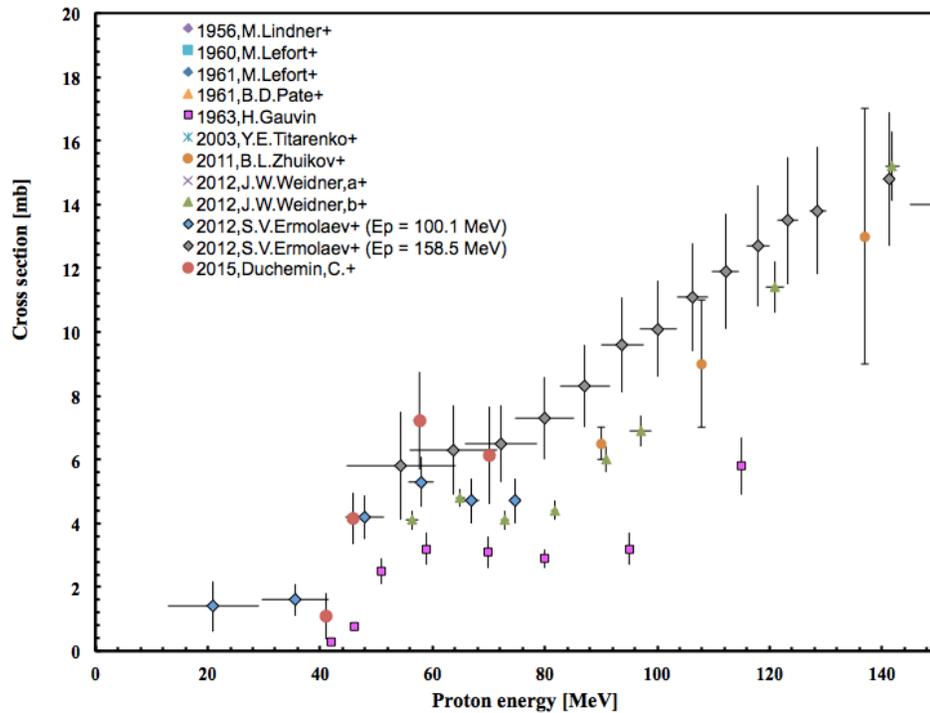
# Summary

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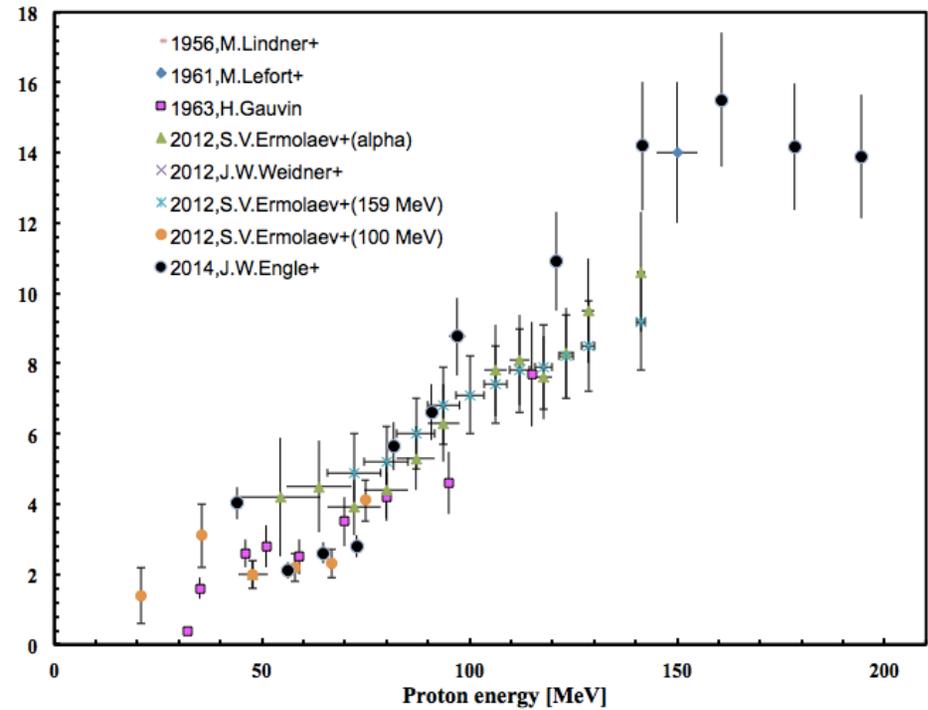
- Nuclear Excitation Functions – Targeted measurements in the context of global isotope/data needs and existing production facilities
  - Needed to predict Yield and Impurities and to guide Target Design
  - Needed to resolve discrepancies
  - Useful for V&V of codes and nuclear models
- The necessary quality and breadth of capabilities
  - Accuracy in Standard Foil Stack Activation experiments
    - Energy and distribution, intensity... Analyzing magnets, Faraday cups
    - Target characterization
      - Target preparation and pre/post-irradiation characterization
      - Signal attribution due to overlapping gammas
  - Charged and neutral particles
- Nuclear Decay Data

# To Predict Yields and Impurities

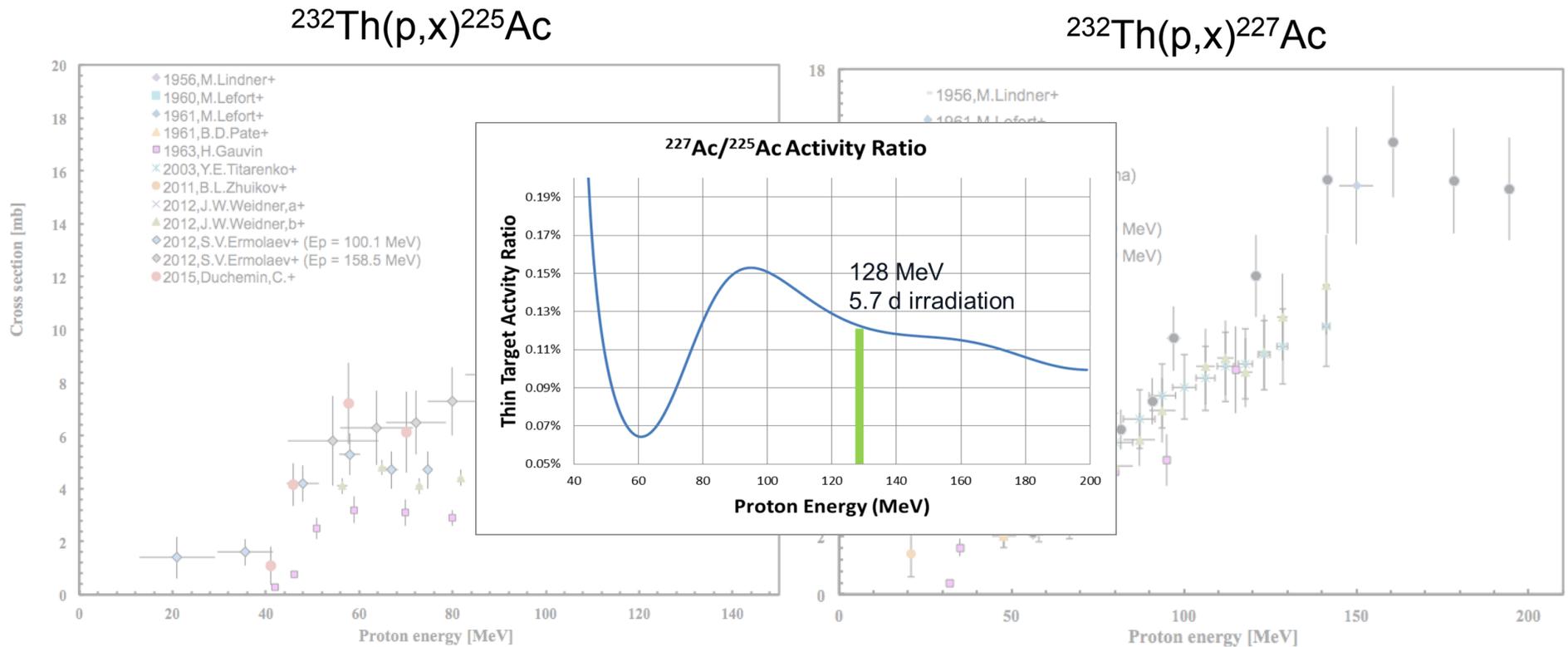
$^{232}\text{Th}(p,x)^{225}\text{Ac}$



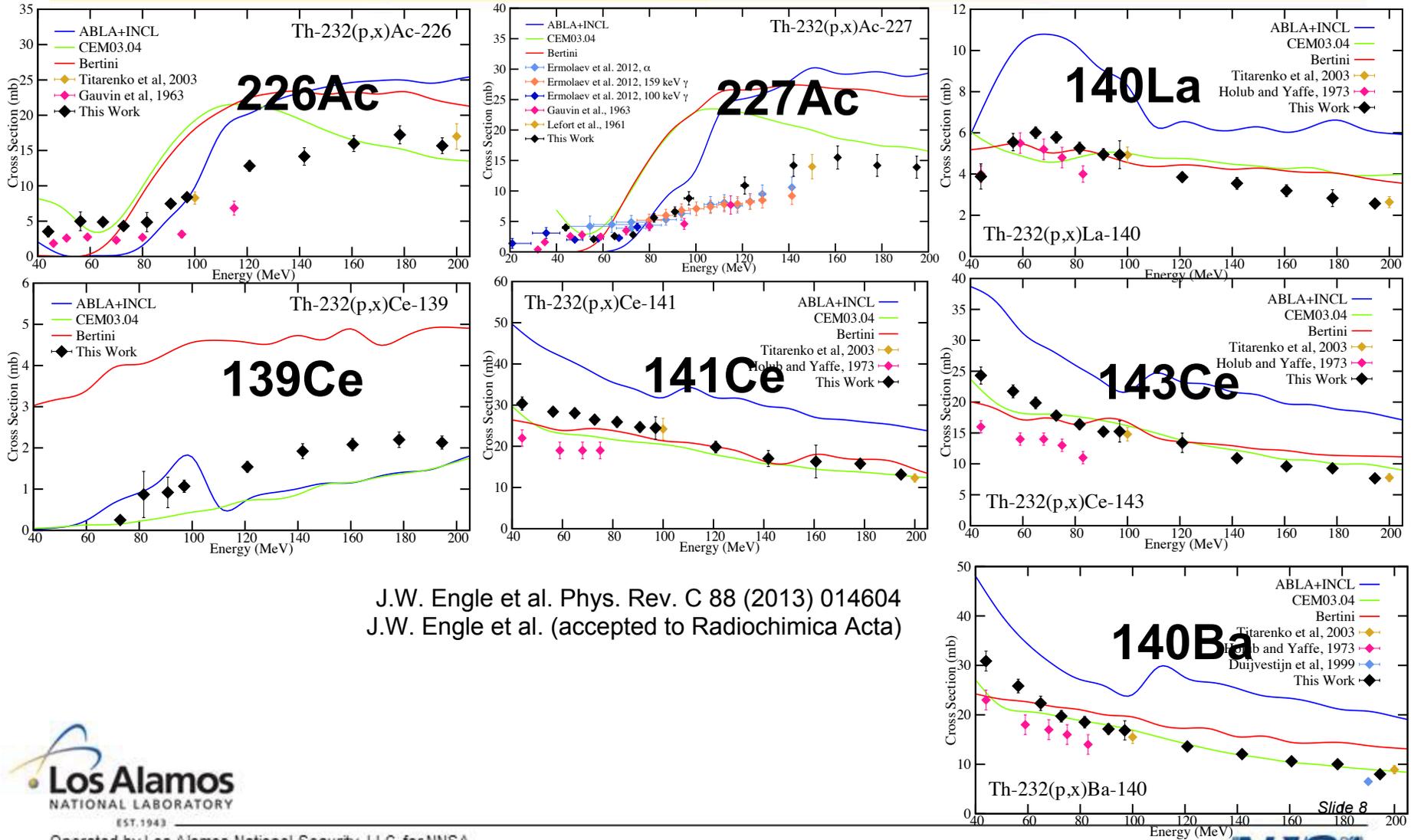
$^{232}\text{Th}(p,x)^{227}\text{Ac}$



# To Predict Yields and Impurities



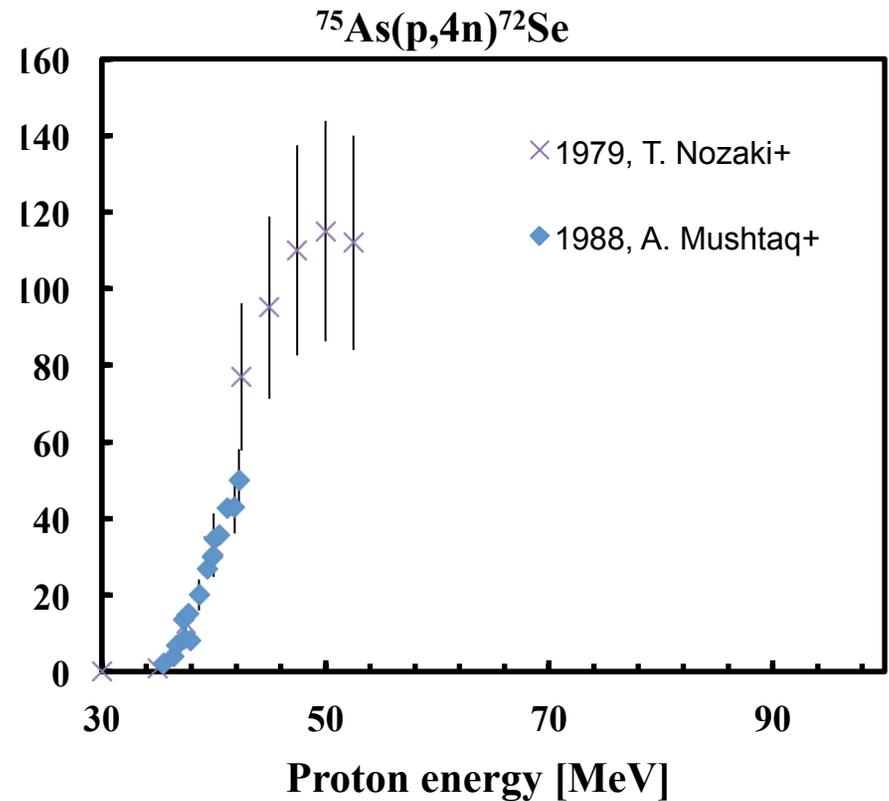
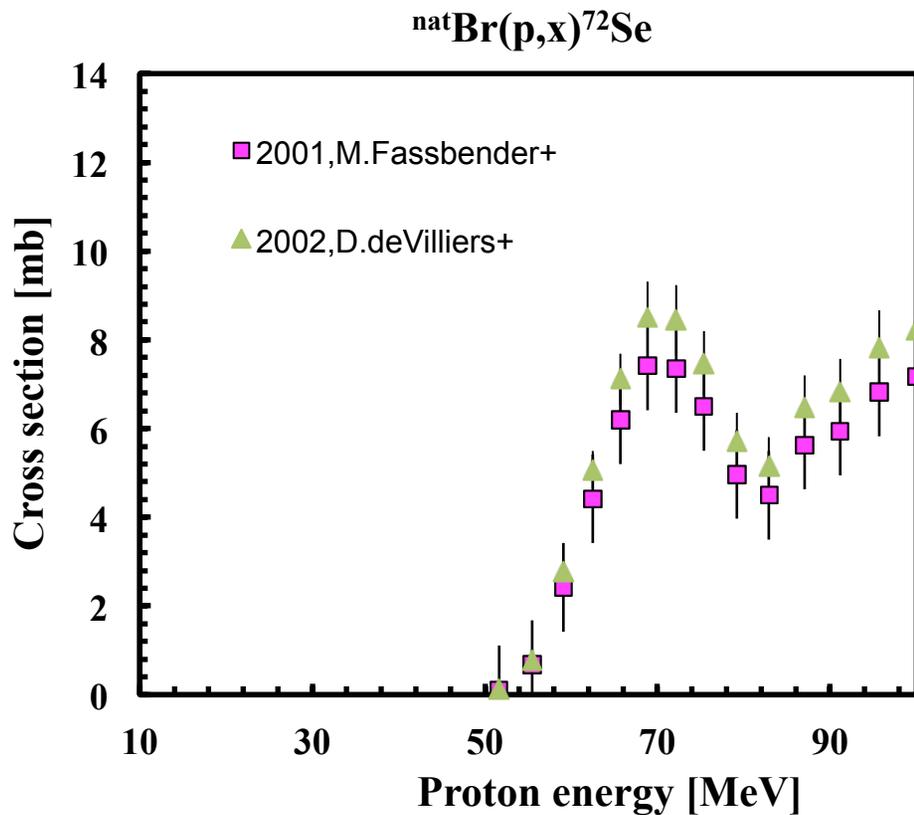
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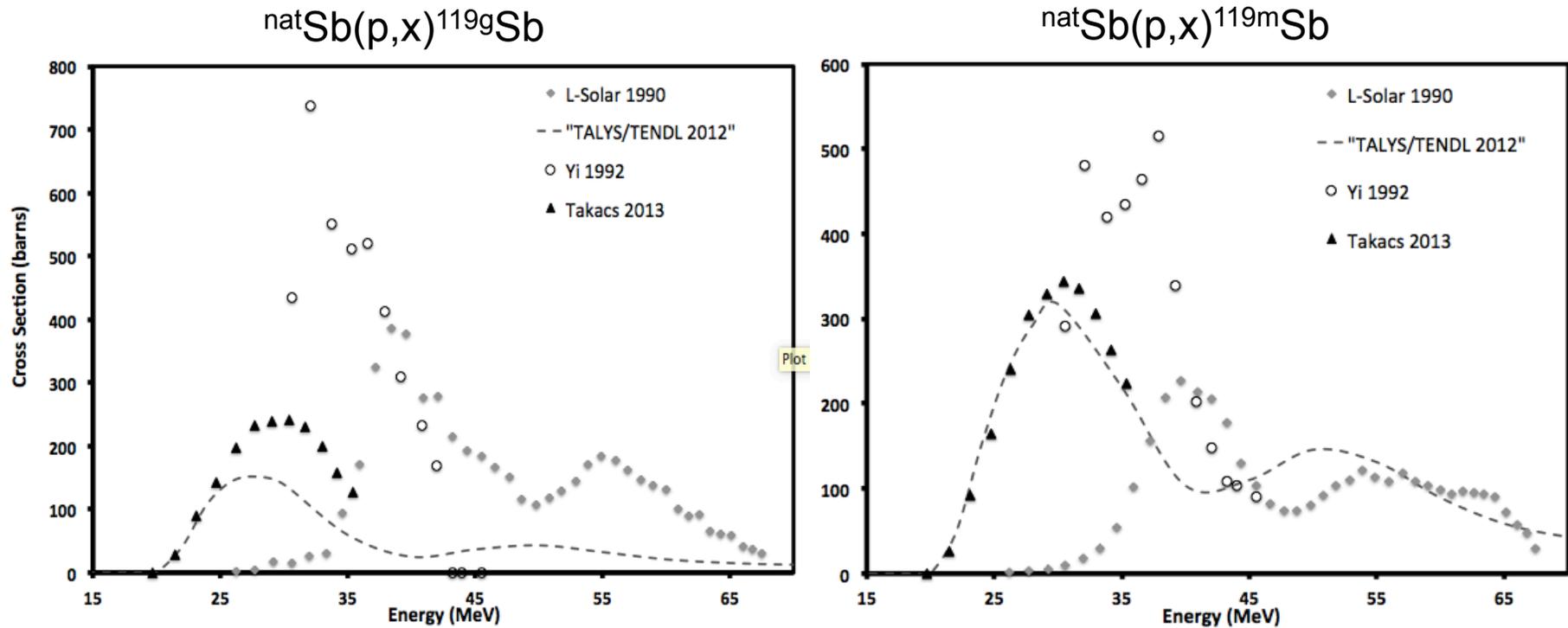
J.W. Engle et al. Phys. Rev. C 88 (2013) 014604  
 J.W. Engle et al. (accepted to Radiochimica Acta)

# To Guide Target Design

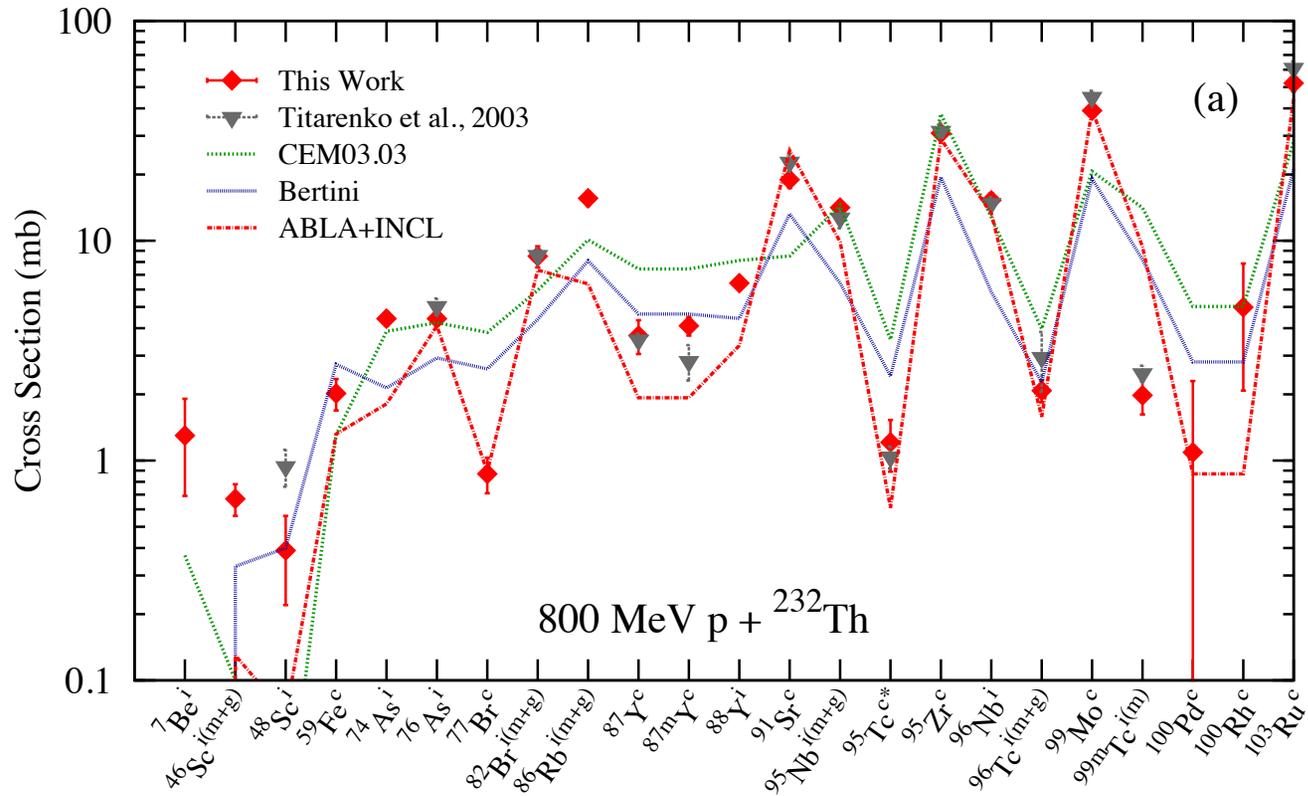
Example:  $^{75}\text{As}(p,4n)^{72}\text{Se}$ ,  $^{\text{nat}}\text{Br}(p,x)^{72}\text{Se}$



# New Measurements are Also Needed to Resolve Discrepancies



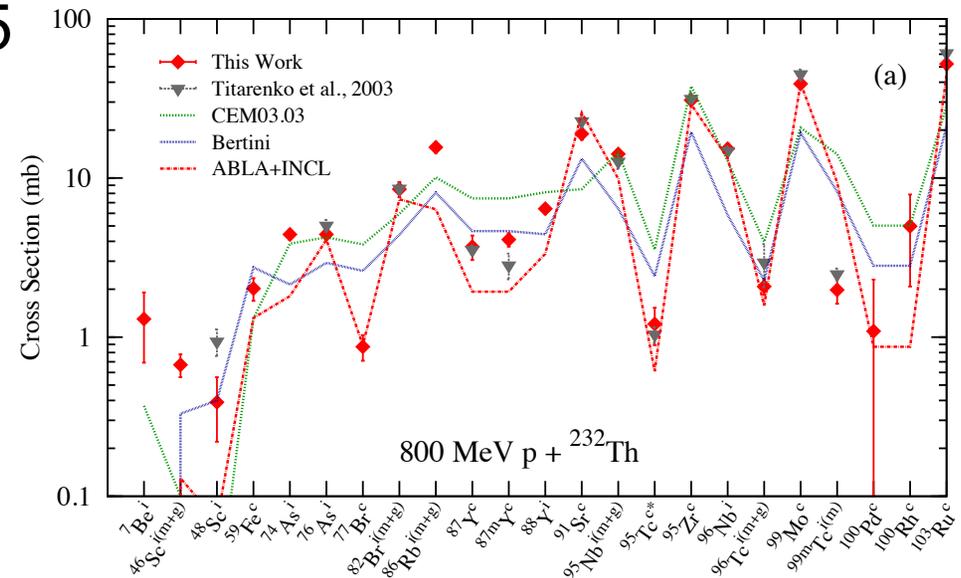
# Data as a Standard for V&V of Nuclear Codes



# Data as a Standard for V&V of Nuclear Codes

Assumed accuracy in code predictions for charged particle-induced reactions:

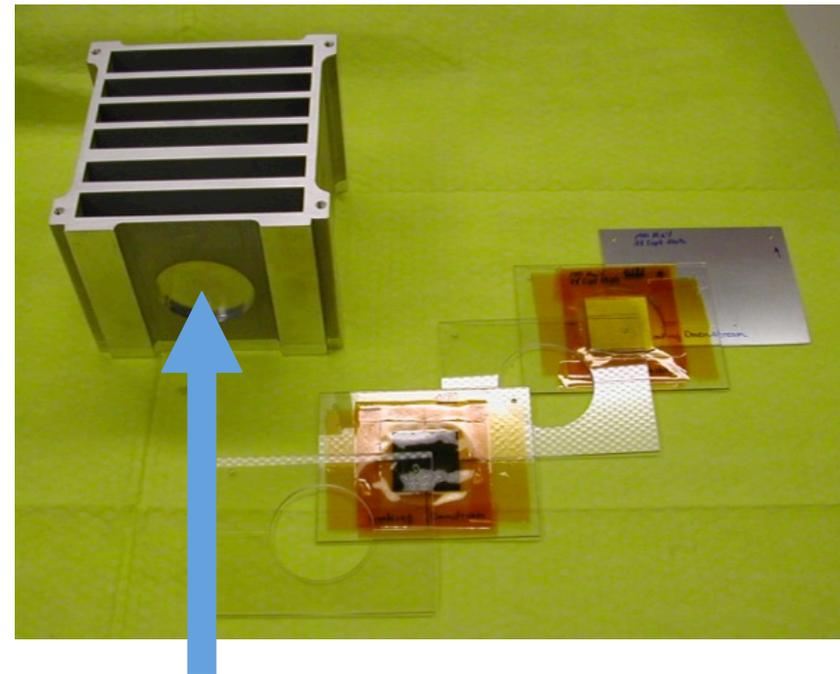
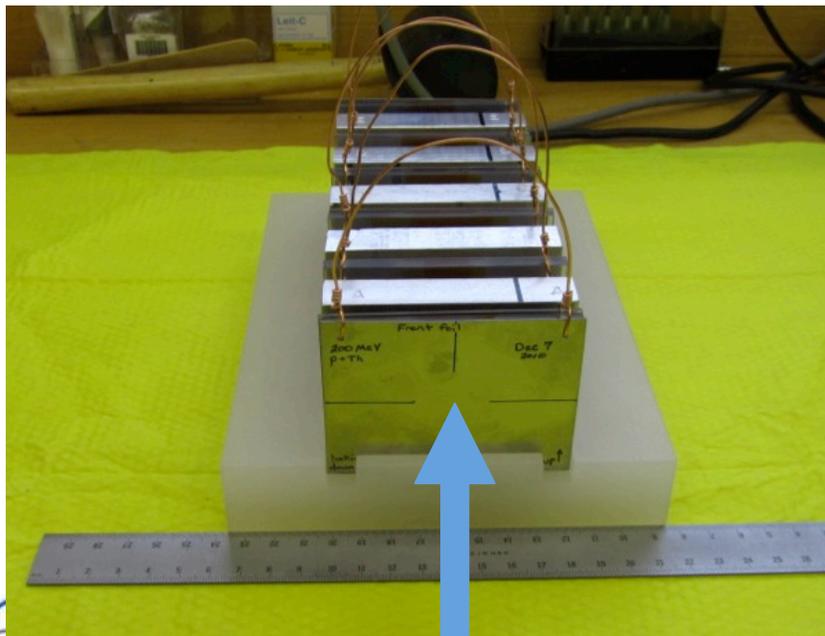
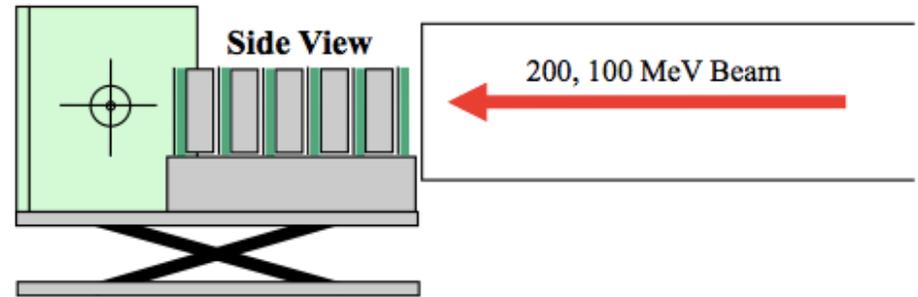
- Magnitude: Factors of 1-5
- Structure: Within 10 MeV



Useful measurements will improve on this.

# The Standard Stacked Foil Activation Method

1. Activation of well-characterized thin samples
2. Quantification of produced radioactive residuals
3. Calculation of reaction probability



# The Standard Stacked Foil Activation Method

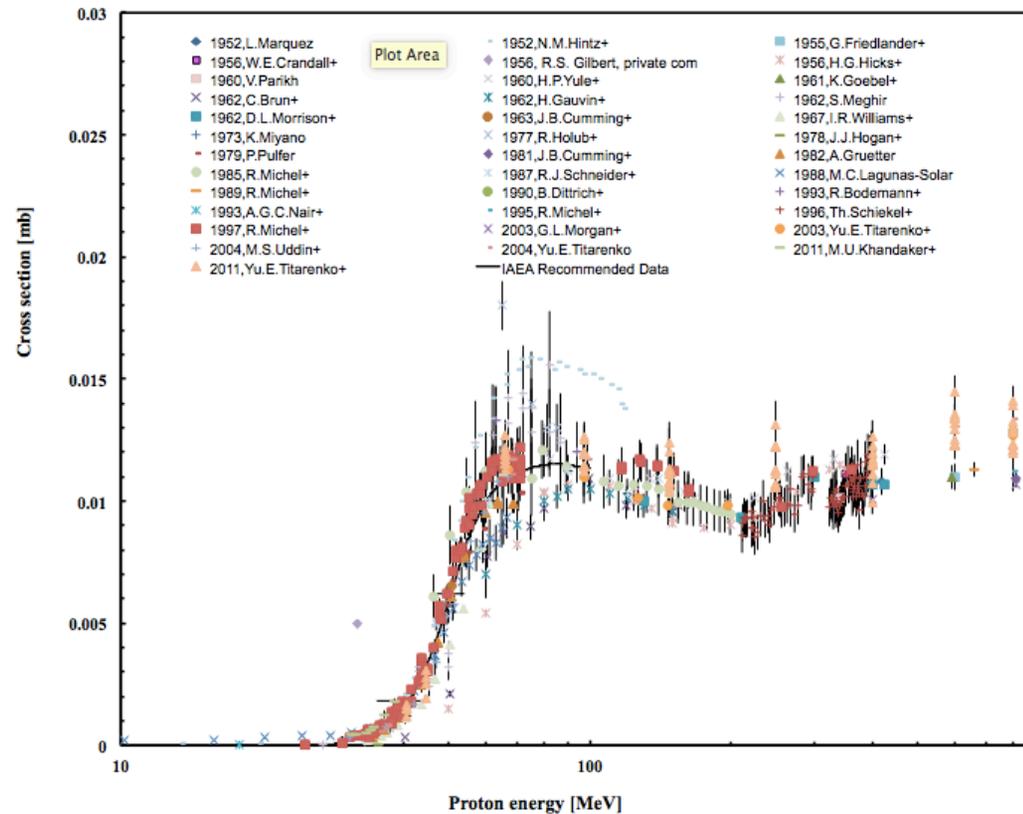
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- Driving, Reported Uncertainties at Present:
  - Incident particle flux during measurement: 4-12%
  - HPGe Assay: 3-5% efficiency calibration. 1-3% peak fitting.
  - Characterization of atomic density of irradiated sample: 1-?%  
(challenging when not using metallic foils)
  - Peak deconvolution / Radionuclide identification: a problematic, systematic source of misattribution error

Combined Uncertainty is generally >6%.

# Particle Intensity

- Commonly tracked with monitor reactions, which lend their own uncertainty



# A solution: Faraday Cups

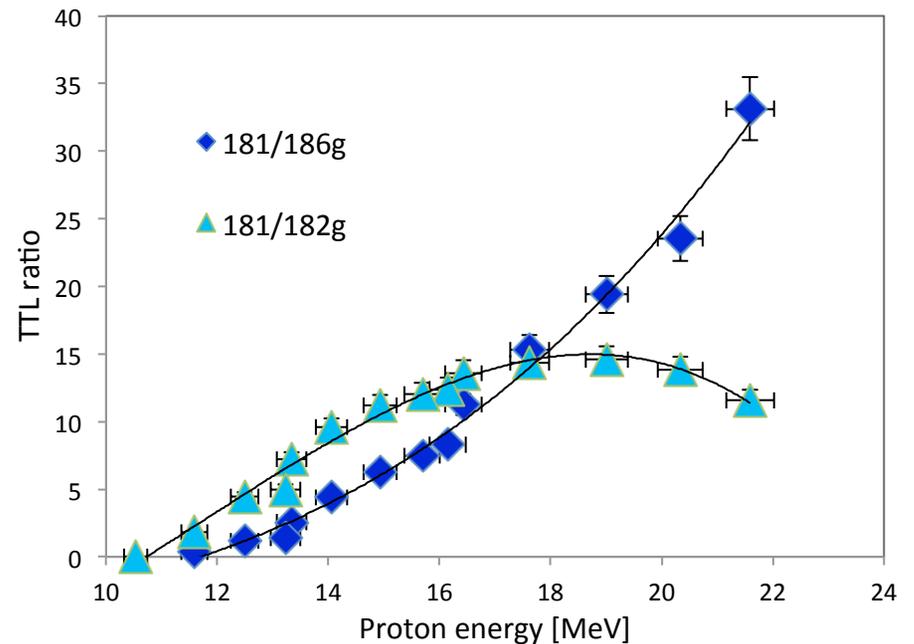
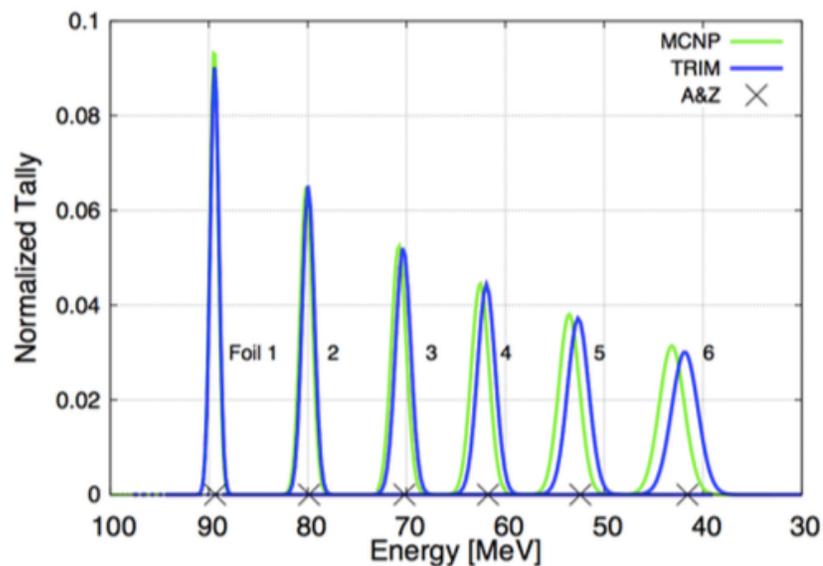
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- Of course they have their own challenges:
  - secondary electron suppression,
  - target electrical insulation,
  - particle and flux dependences are not always simple
- Alternative real-time, verifiable particle flux monitors
  - Offline-Faraday Cup calibrated SEEM or LEM



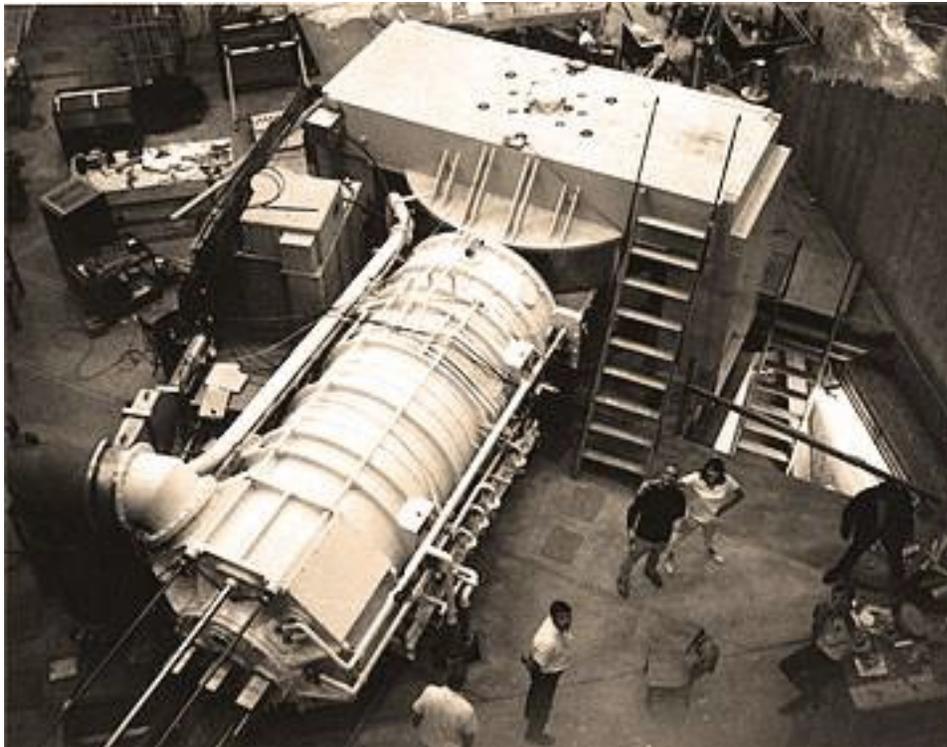
# Primary Beam Energy

- Energy degradation has a limited effective range,
  - Commonly “verified” by monitor foils giving reactions with known thresholds (indirect measurement),
  - or calculated by established codes (computational)



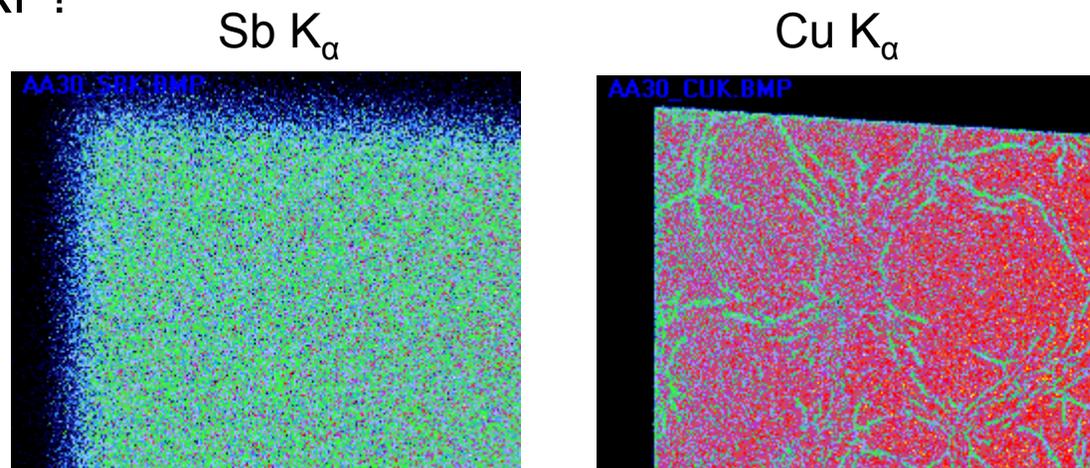
# Tunable Beam Energies, Analyzing Magnets

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# Target Characterization

- Some measurements are avoided because of the challenge of characterizing targets (e.g.,  $^{72}\text{Se}$ )
  - We need better ways to prepare them and measured  $\text{mg}/\text{cm}^2$
  - e.g.: XRF?

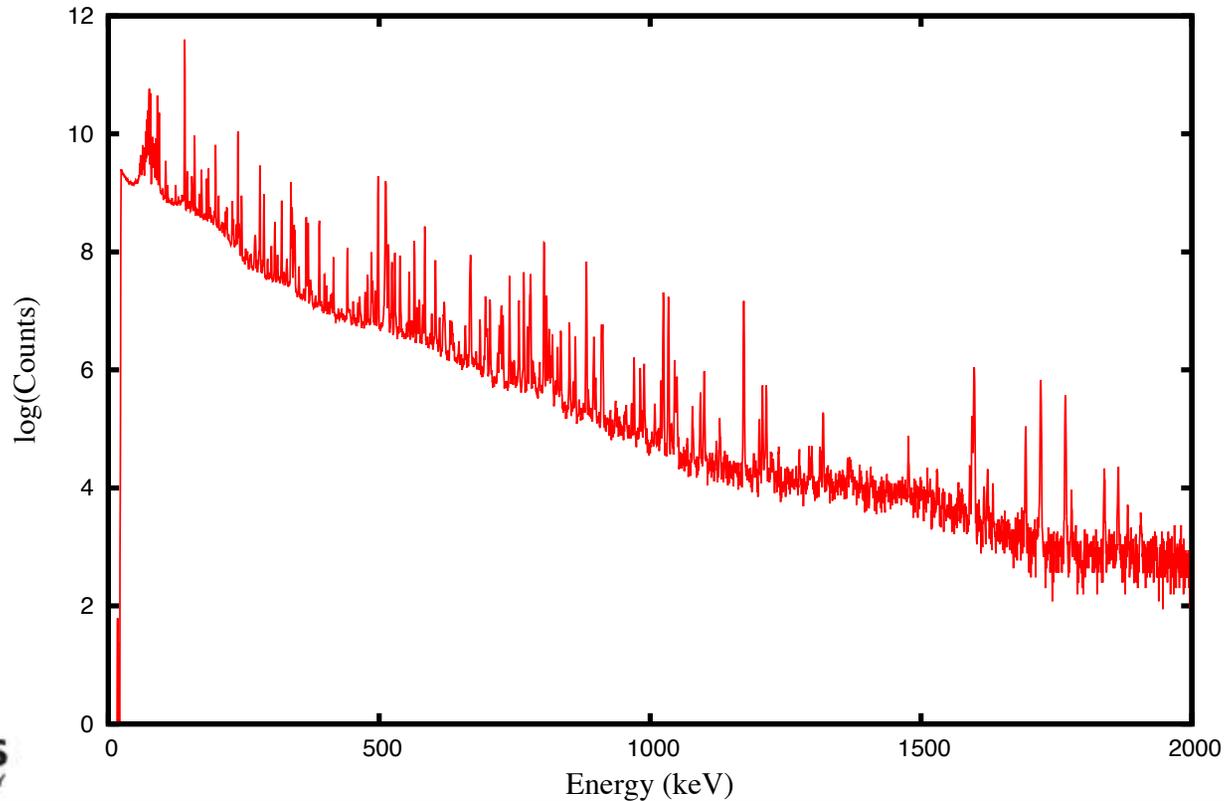


- $K_\alpha$  map shows very low intensity all around the edges of the foil  
Intensity decreases by a factor of  $\sim 2$  in the green region relative to the blue
- The unexpected presence of Cu lines reveals estimated 40-50% contamination from Cu

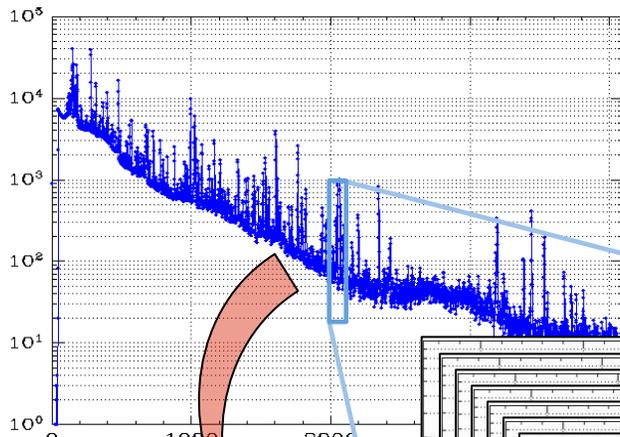
# Residual Quantification

- At higher energies, or for more massive target nuclei, spectral analysis is challenging

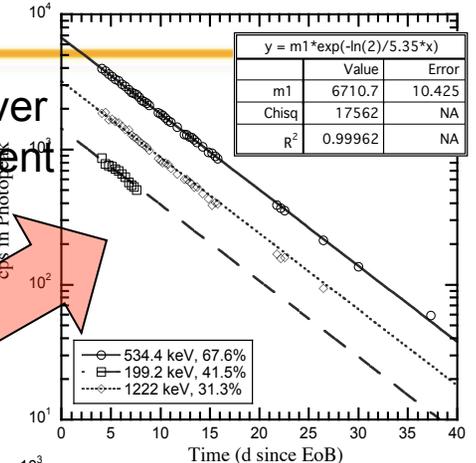
Example Spectrum of 800 MeV p + Th-232



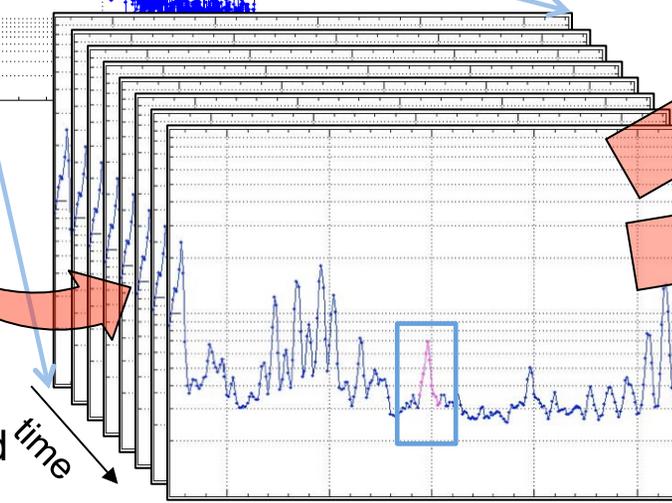
# Identification and Quantification of Residuals by Gamma-Ray Spectrometry



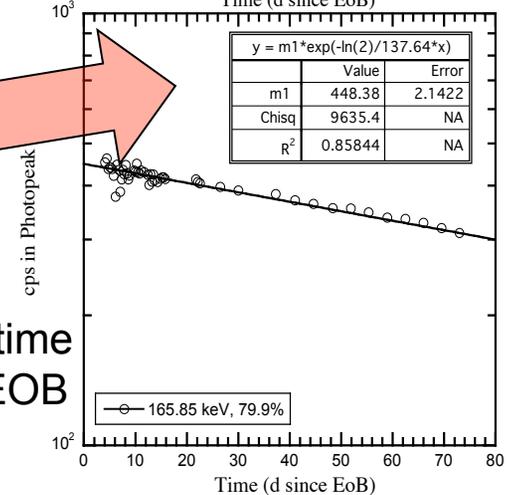
Decay of three gamma lines over time, providing three independent EOB activity measurements



Peak areas extracted from multiple recorded spectra

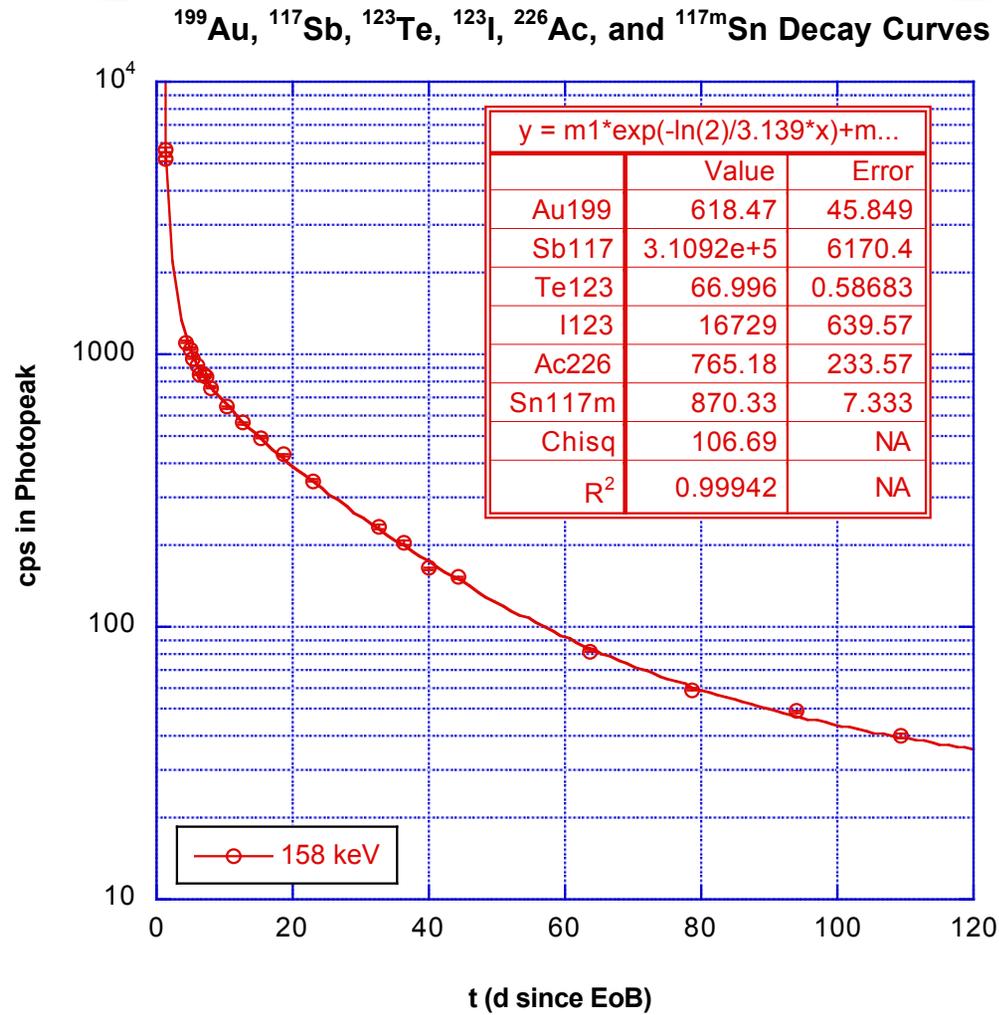


Decay of one gamma line followed over time to determine EOB activity



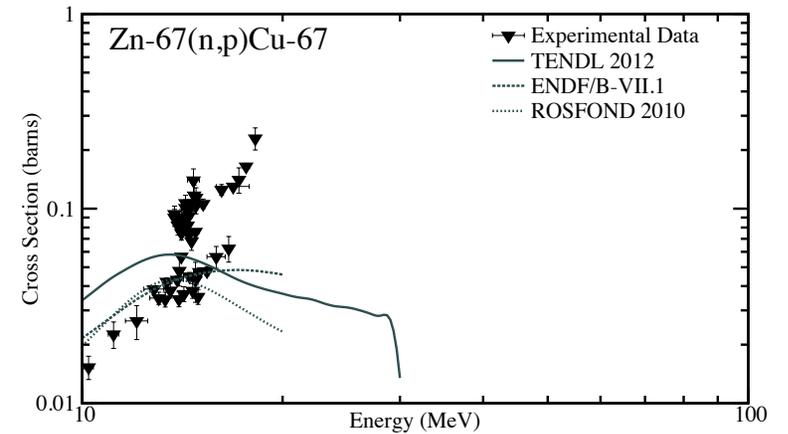
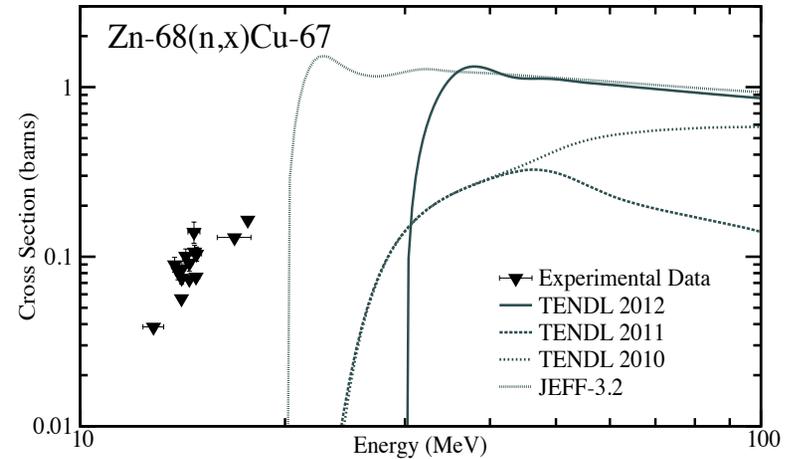
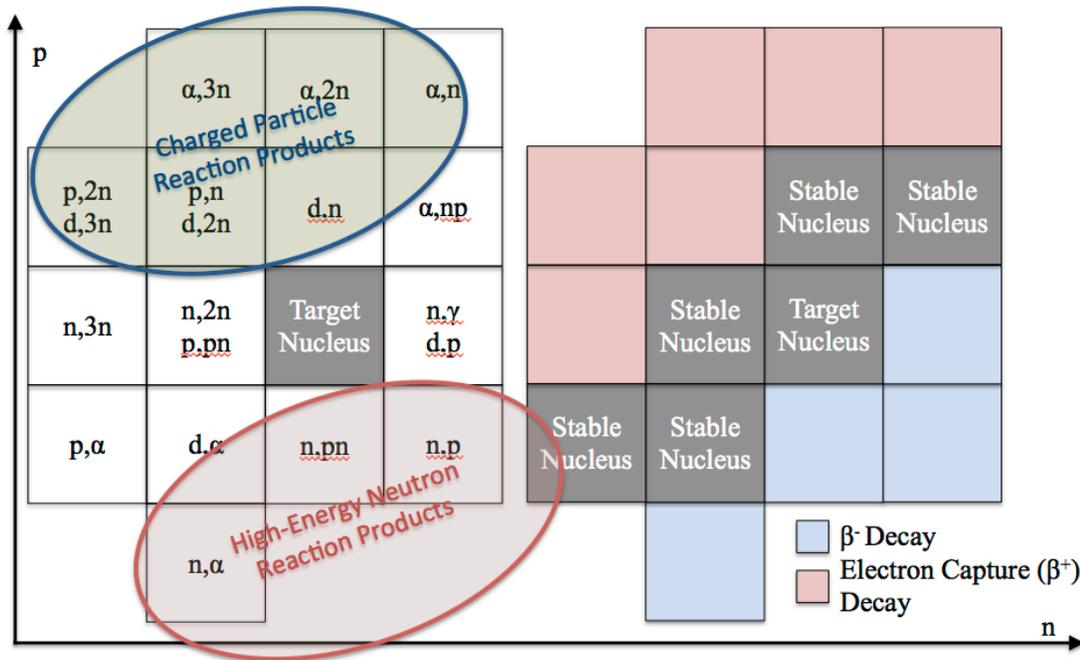
One analysis cycle produces a single cross section data point

# What is possible

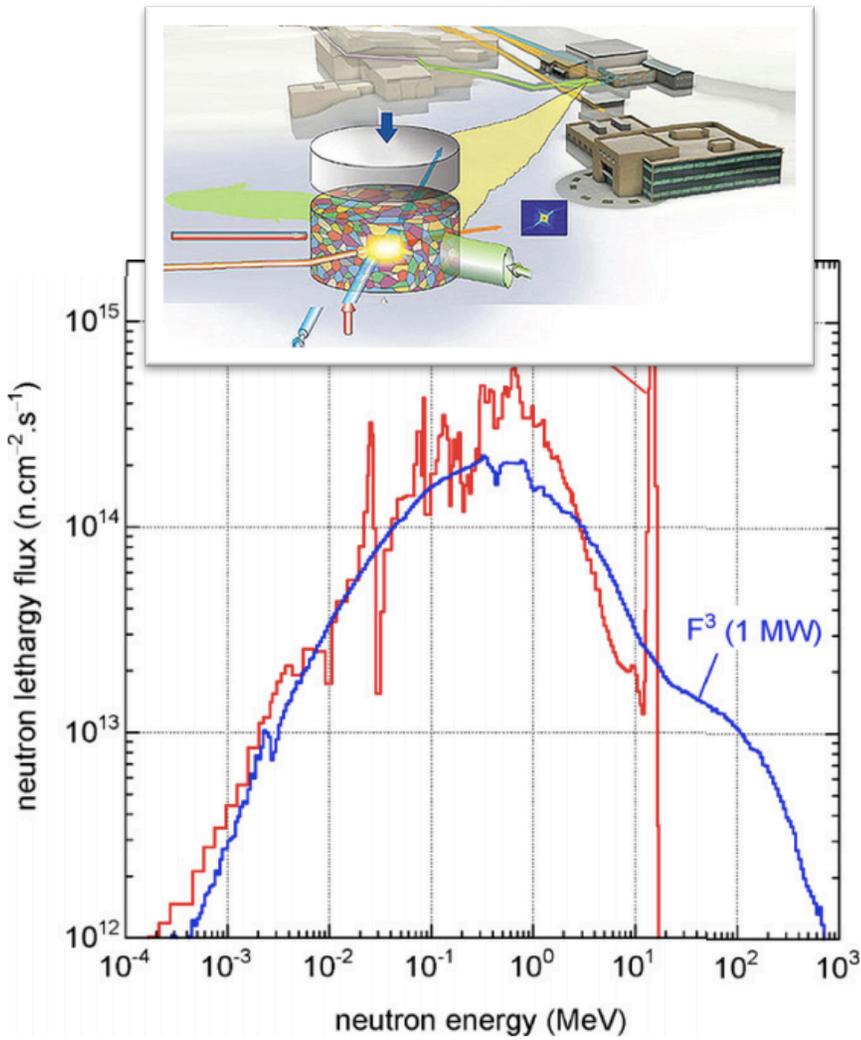


# Neutral Particles

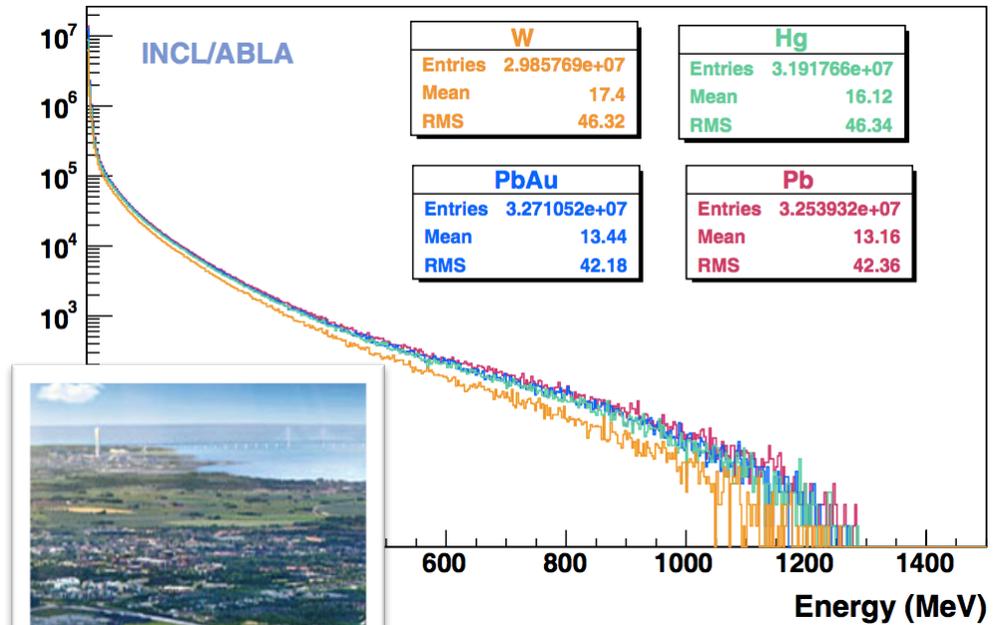
- Neutrons (the very fast ones)
- Also for gammas



# Data to Buttress Capability Development at New Facilities



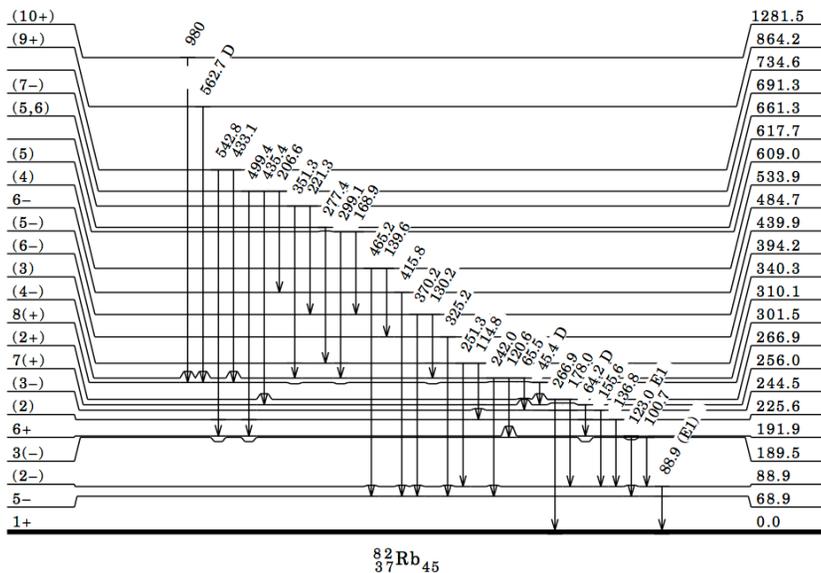
Spallation Neutrons Energy Spectra



Slide 24

# Nuclear Decay Data – An example

	Measured BR	Source
776.50 keV gamma of $^{82}\text{Rb}$	$13.4 \pm 0.5 \%$	<i>H.-W. Muller 1987 (extensively used)</i>
	$15.08 \pm 0.16 \%$	J.K. Tuli 2003 & NNDC
	$14.93 \pm 0.37\%$	C.J. Gross et al., 2012



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Nuclear Data Sheets 98 (2003) 209–334

**Nuclear Data Sheets**

**Nuclear Data Sheets for A = 82\***

**J. K. Tuli**  
National Nuclear Data Center  
Brookhaven National Laboratory, Upton, NY 11973  
(Received July 12, 2002; Revised January 7, 2003)

PHYSICAL REVIEW C **85**, 024319 (2012)

**Measuring the absolute decay probability of  $^{82}\text{Sr}$  by ion implantation**

C. J. Gross,<sup>1,\*</sup> K. P. Rykaczewski,<sup>1</sup> D. W. Stracener,<sup>1</sup> M. Wolinska-Cichocka,<sup>1,2</sup> R. L. Varner,<sup>1</sup> D. Miller,<sup>3</sup> C. U. Jost,<sup>1</sup> M. Karny,<sup>1,4</sup> A. Korgul,<sup>4</sup> S. Liu,<sup>5</sup> and M. Madurga<sup>3</sup>

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<sup>3</sup>Department of Physics and Astronomy, University of Tennessee, Knoxville, Tennessee 37996, USA  
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<sup>5</sup>UNIRIB/Oak Ridge Associated Universities, Oak Ridge, Tennessee 37831, USA

(Received 21 December 2011; published 27 February 2012)

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**Thank you for your attention**

# Related initiatives

## Engagement in IAEA data evaluation efforts

- Cross section measurement effort gained national and international attention
- Invited to participate in an IAEA Consultants' Meeting in 2011
- Follow-up invitation to participate in a 3 year IAEA Coordinated Research Project starting in 2012 (11 countries represented)
- First of three Research Coordination Meetings occurred in Dec 2012
- Summary Report published in February 2013



**IAEA**  
International Atomic Energy Agency

INDC(NDS)-0630  
Distr. G+NM+SD

### INDC International Nuclear Data Committee

#### Summary Report

#### First Research Coordination Meeting on

#### Nuclear Data for Charged-particle Monitor Reactions and Medical Isotope Production

IAEA Headquarters  
Vienna, Austria

3 – 7 December 2012

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