Cross Section measurements at LANL: $^{232}Th + p$

Meiring Nortier

Los Alamos National Laboratory, U.S.A.

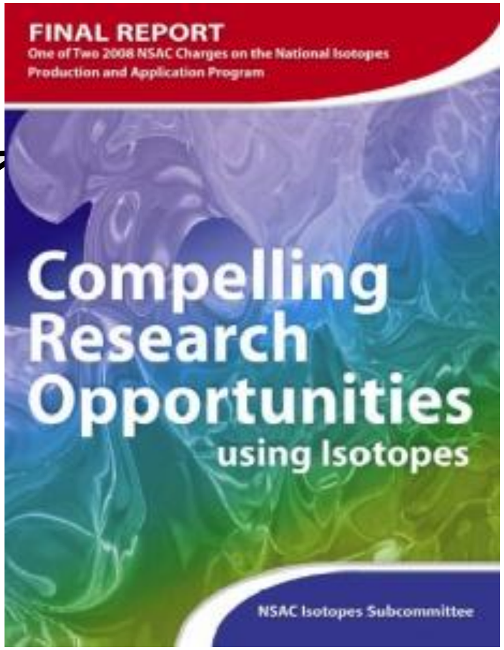
Outline

- **Basic measurement approach**
- **Cross sections for $^{232}Th + p$**
  - Energies: 800 MeV and 40-200 MeV
  - Primary Isotopes of interest: ^{225}Ra, ^{225}Ac and ^{227}Ac, ^{227}Th, ^{223}Ra
- **Comparison with theory**
- **Efforts at ORNL and Fermi Lab**
- **Ongoing efforts at LANL**

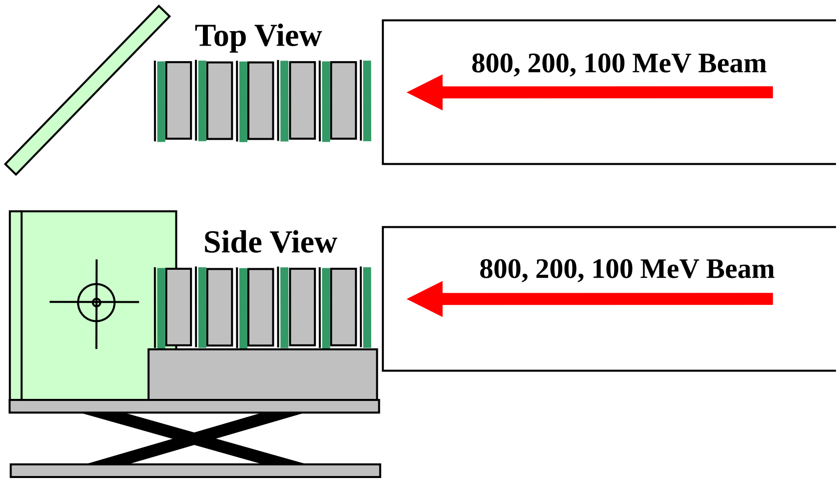
Why Ac-225 and Ra-223?

First of 6 Recommendations to the DOE by the ***Isotopes Subcommittee*** of the ***Nuclear Science Advisory Committee*** (NSAC)

Medicine#1:
Invest in new production approaches of alpha-emitters with highest priority for Ac-225.

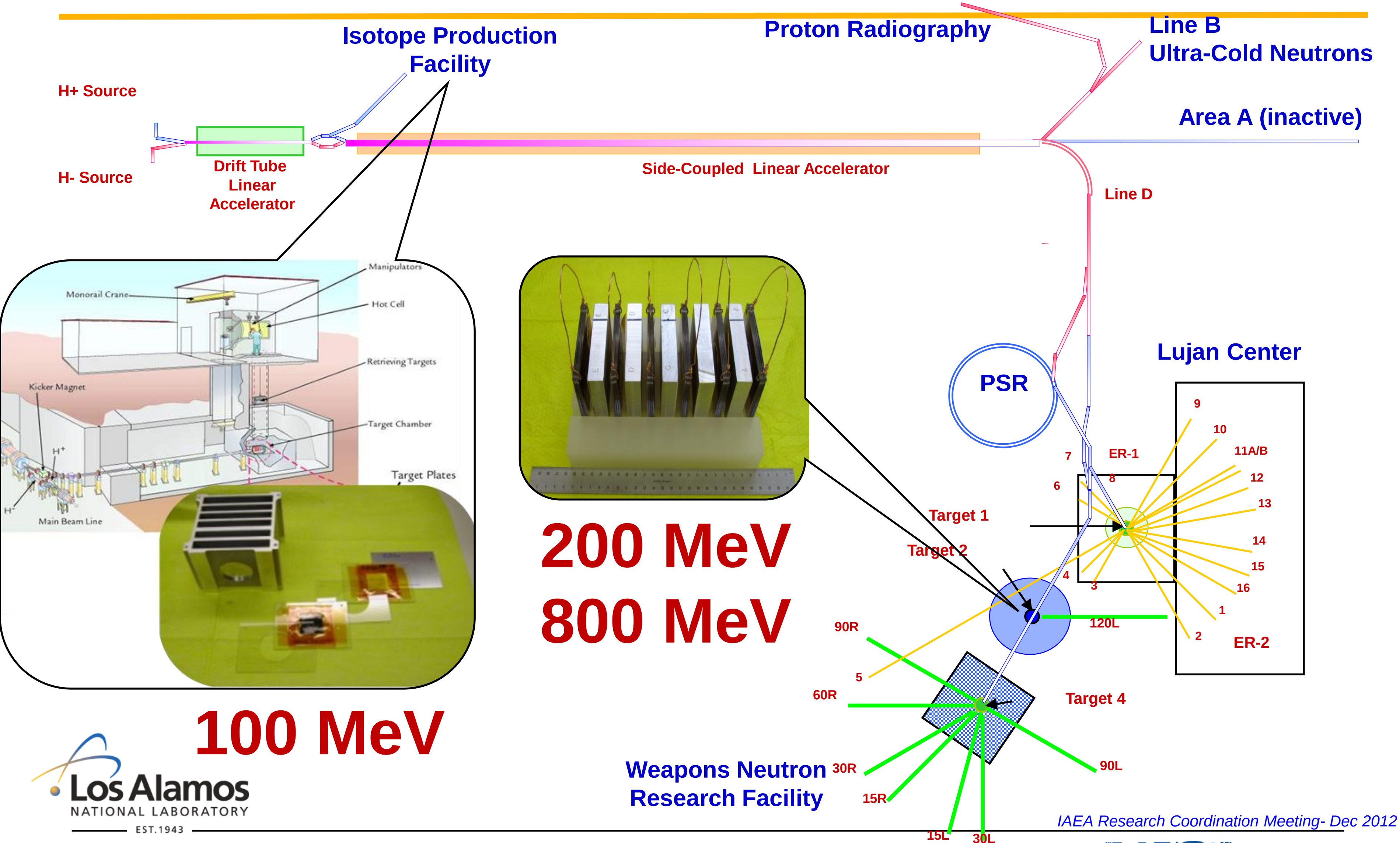


Basic Measurement Approach at LANL



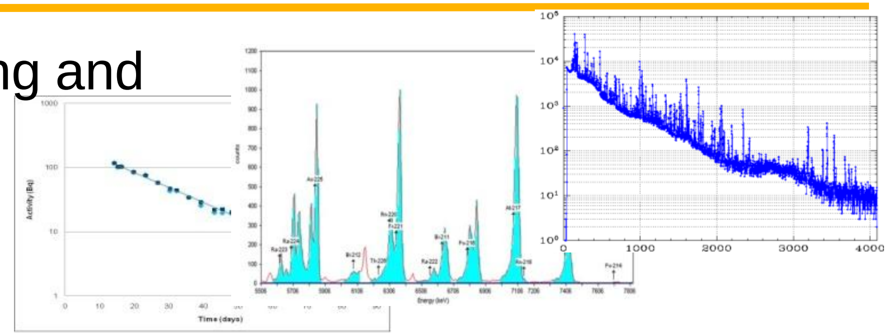
- Apply the well-known stacked foil technique
- Target foils and proton fluence monitor foils are irradiated with proton beams having primary energies of 100, 200 and 800 MeV
- Measurements cover the energy range up to 800 MeV

Irradiations are conducted at WNR and IPF
100 nA, 30-60 min

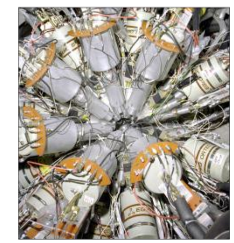


Radio-assay and Chemistry - Samples are assayed via various counting methods

- Use primarily non destructive γ counting and data analysis as well as α counting capabilities of Chemistry Division's world-class Count Room

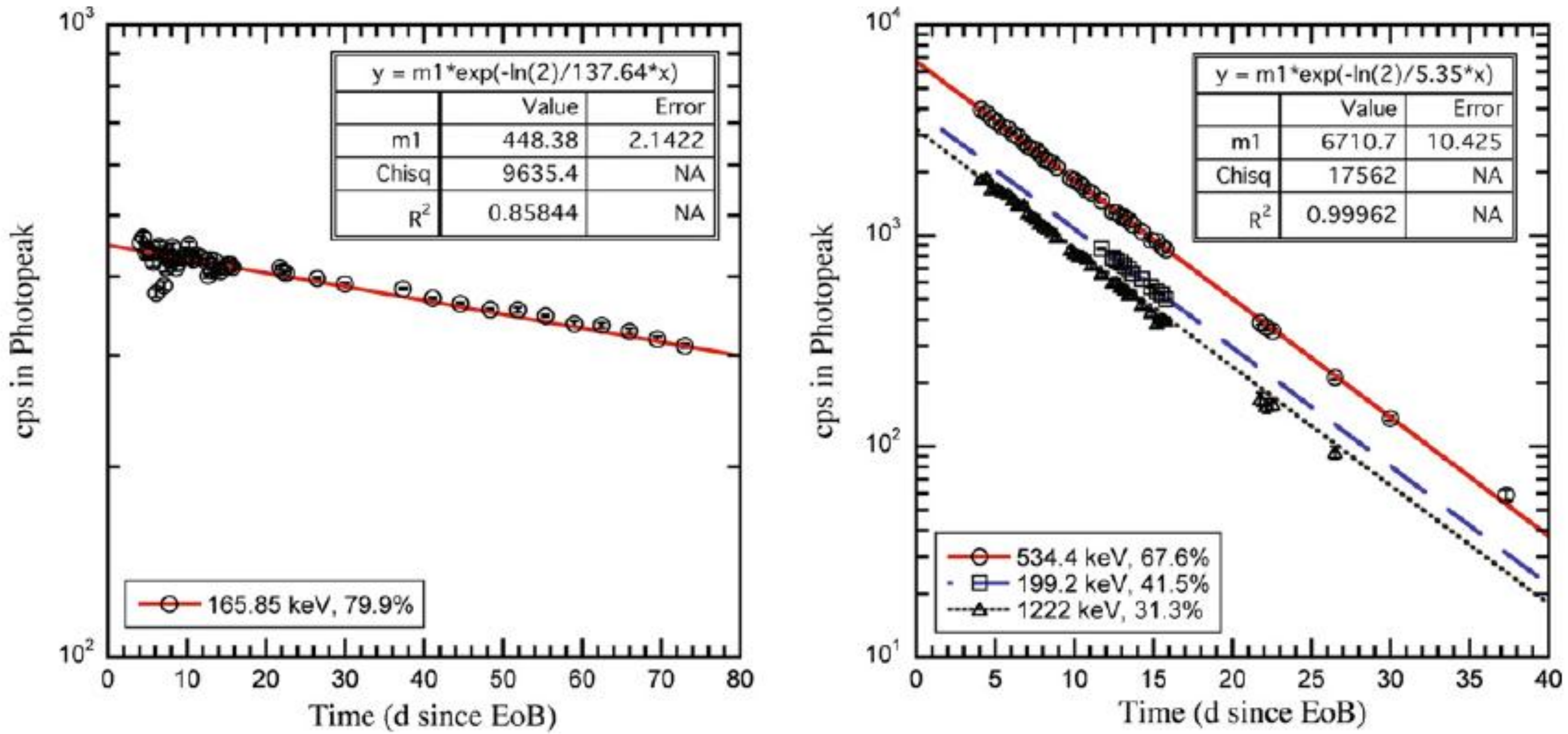


- Sometimes the γ-γ coincidence counting capability of LANCE's GEANIE detector is utilized in parallel with other nondestructive counting



- When required, chemical separations are performed using Chemistry Division's radiochemistry expertise

Decay of isotopes is followed over time to identify residuals and extract accurate production cross sections



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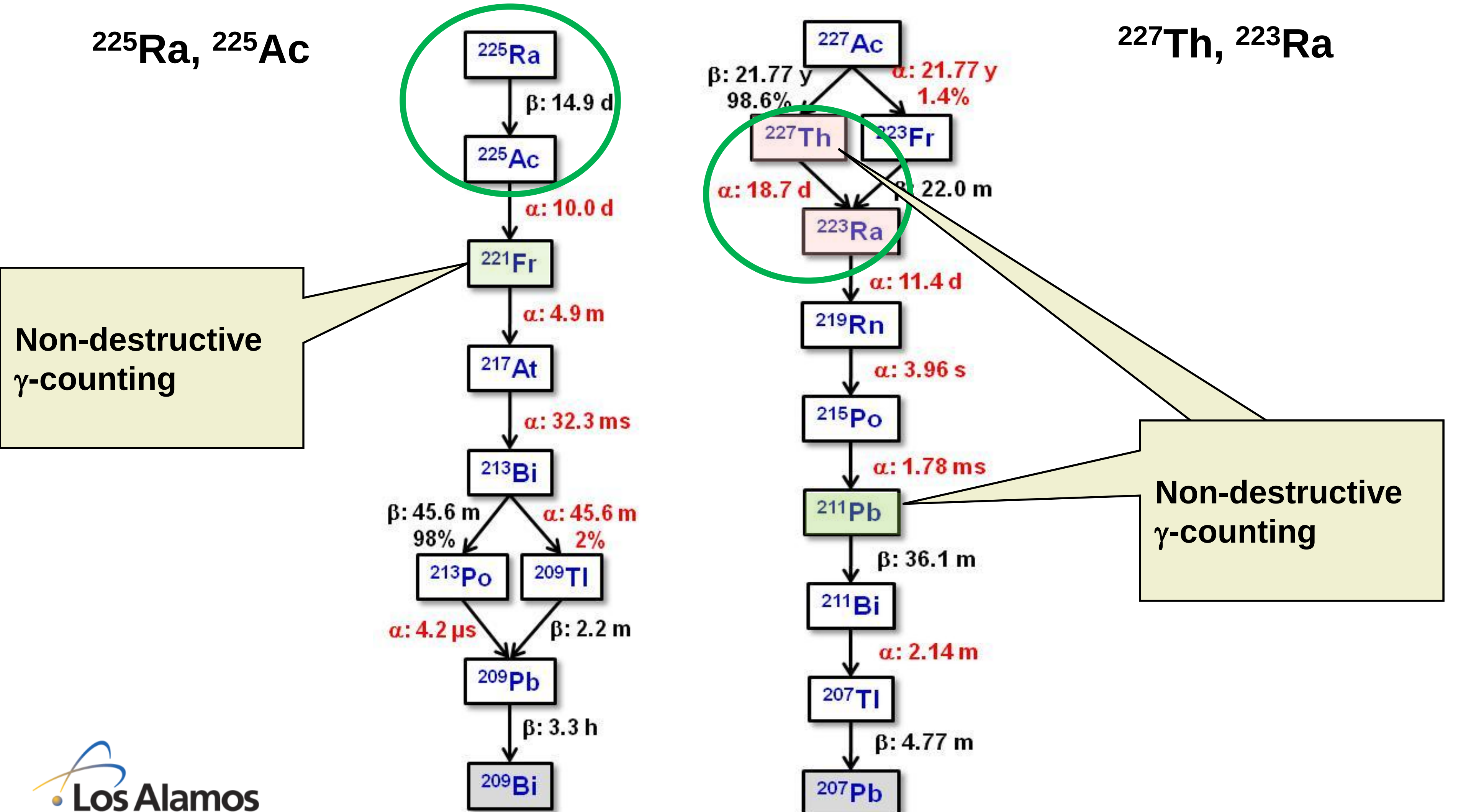
Isotopes of interest in recent measurements



Production Cross Sections at 800 MeV

Reaction	LANL Cross Section (mb)	Counting Method	Literature value (mb)	Theoretical values (CEM) (mb)
$^{232}Th(p,x)^{225}Ac$	14.4 ± 1	γ, α	20.3 ± 5.1*	15.0
$^{232}Th(p,x)^{225}Ra$	3.4 ± 0.4	$\gamma_{indirect}$, $\alpha_{indirect}$	None	1.54
$^{232}Th(p,x)^{227}Ac$	19.5 ± 0.7	α	None	11.0
$^{232}Th(p,x)^{227}Th$ $^{232}Th(p,x)^{223}Ra$	12.8 ± 1.1 5.8 ± 0.6	γ, α γ, α	None None	18.6 3.1

*Titarenko *et al*. (2002), INDC(CCP)-434

New data for $^{223,225}Ra$, ^{227}Ac, and ^{227}Th

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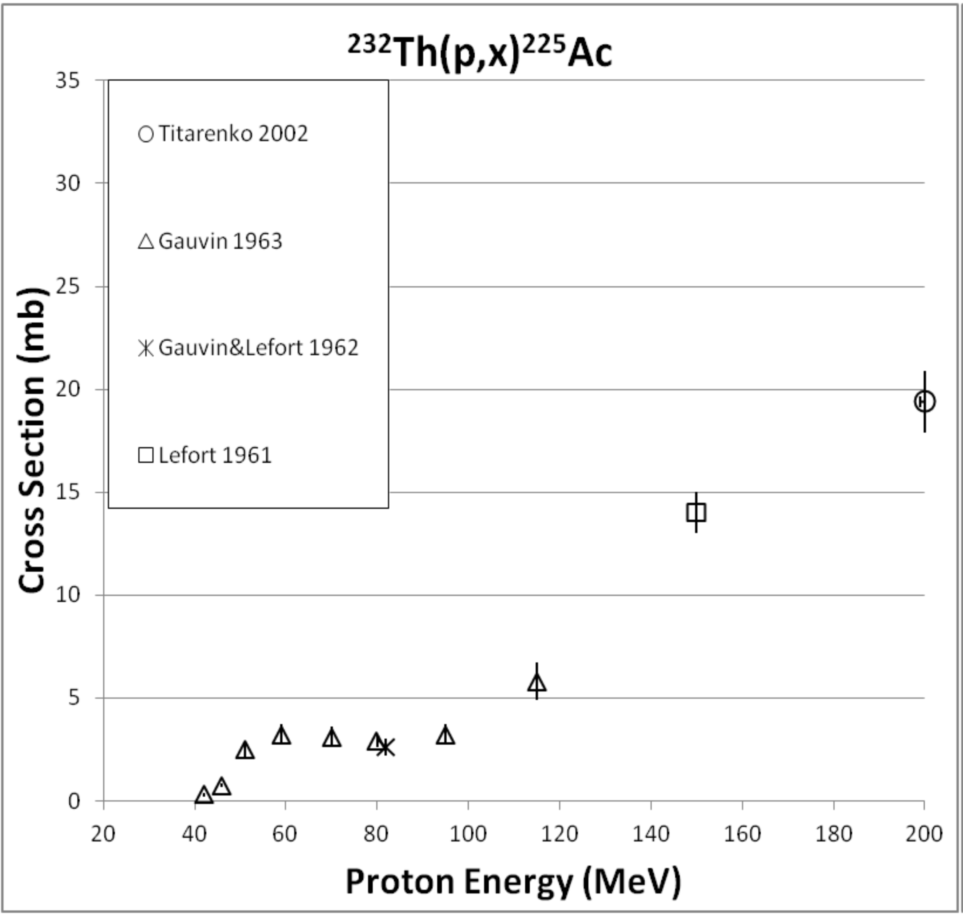
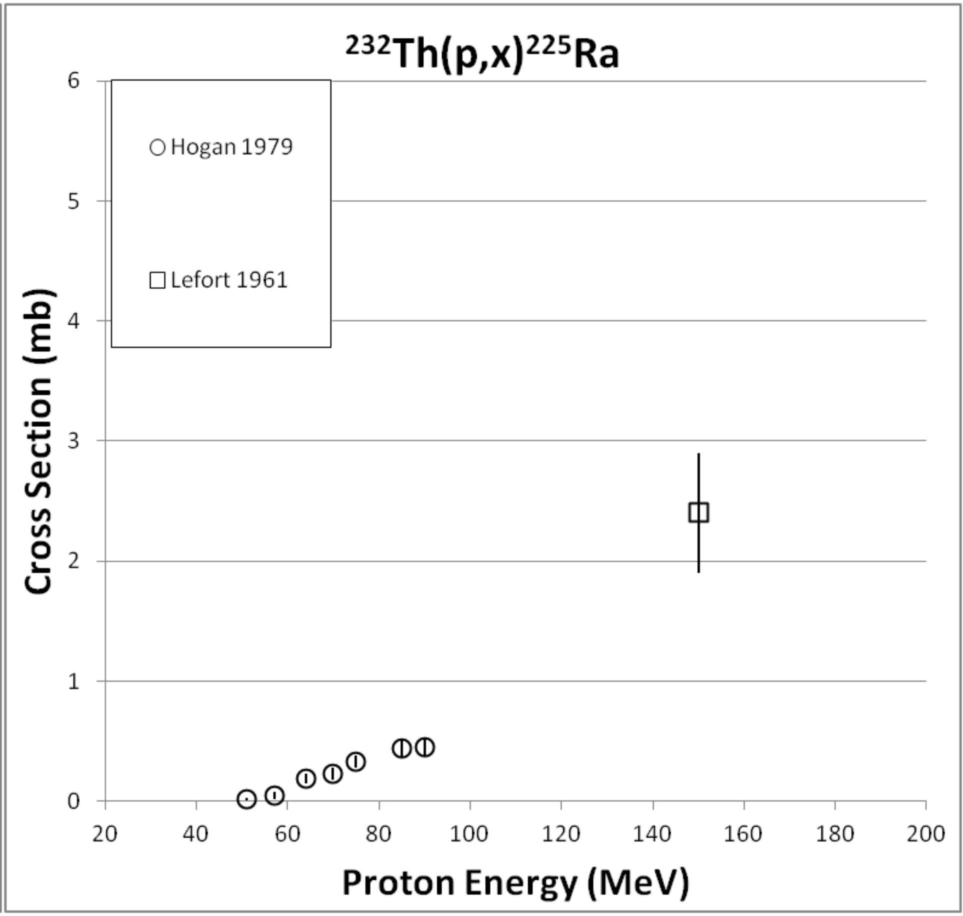
Production Cross Sections below 200 MeV
Existing Data for Ac-225 and Ra-225





Production Cross Sections below 200 MeV
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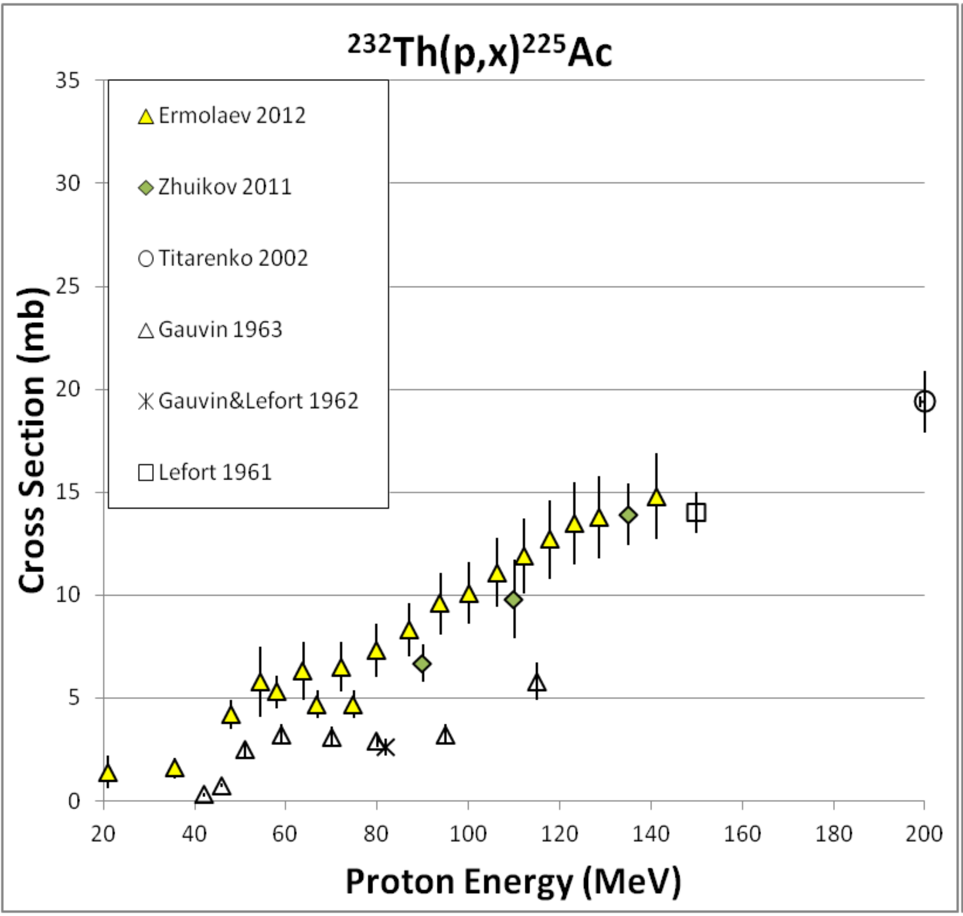
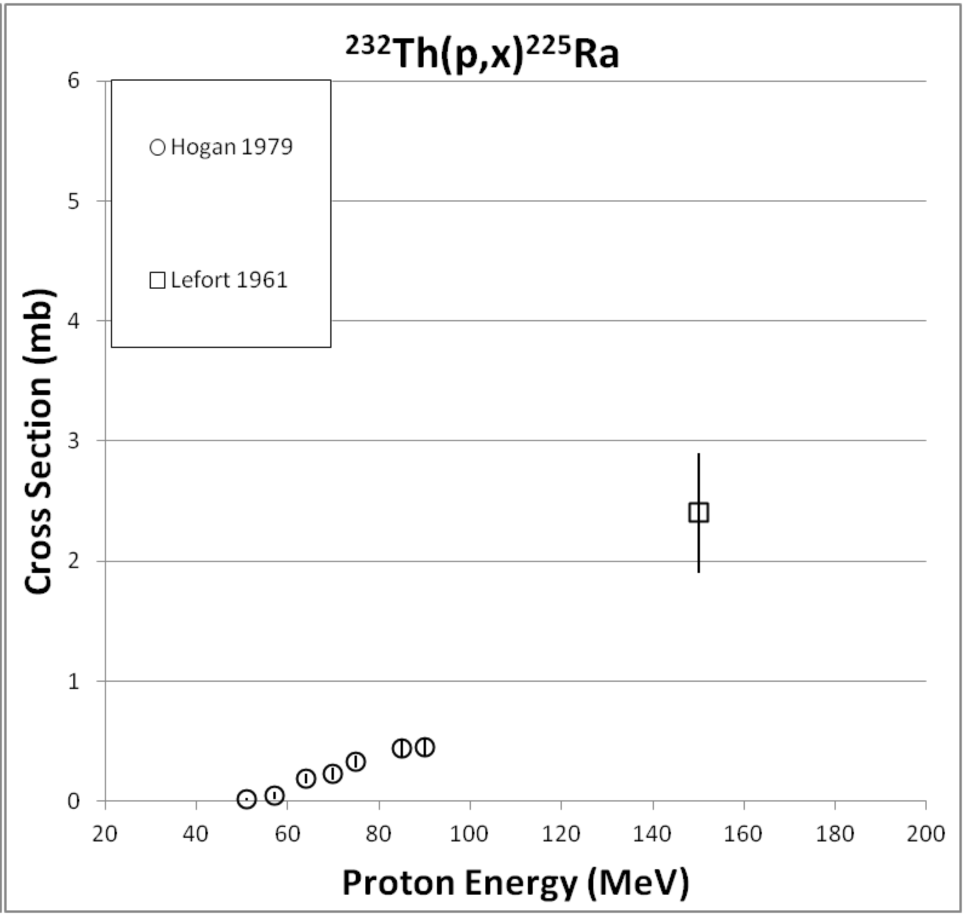
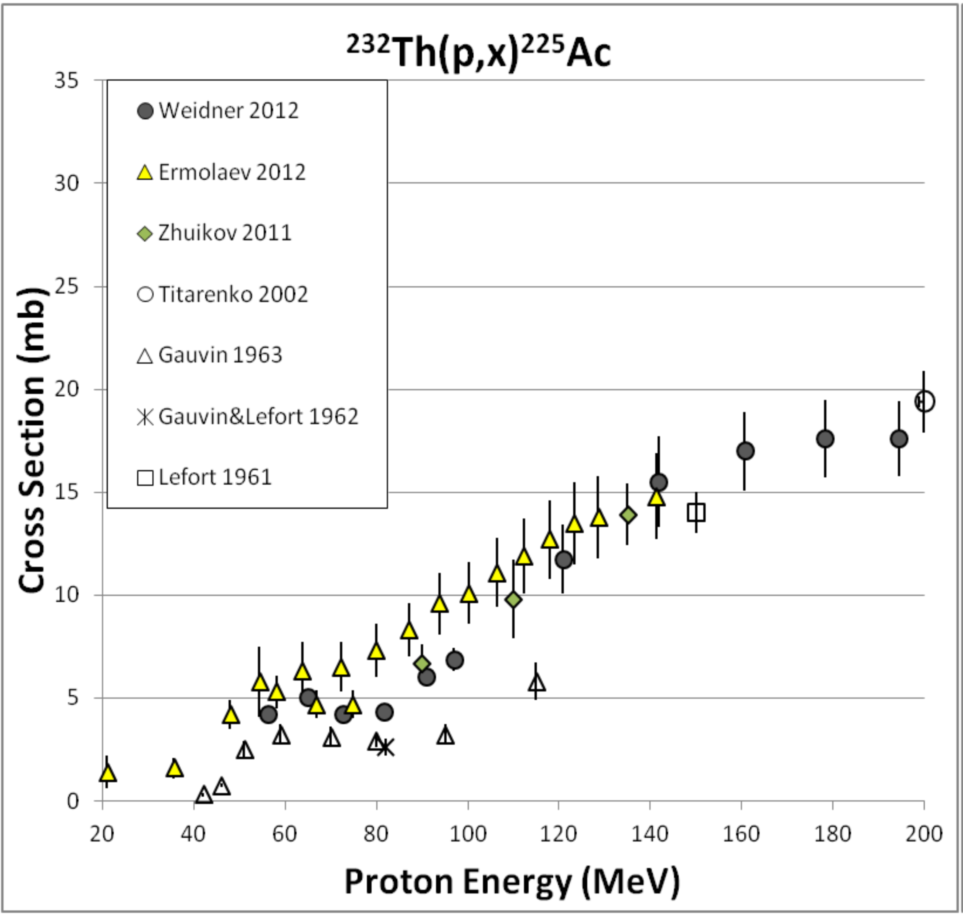
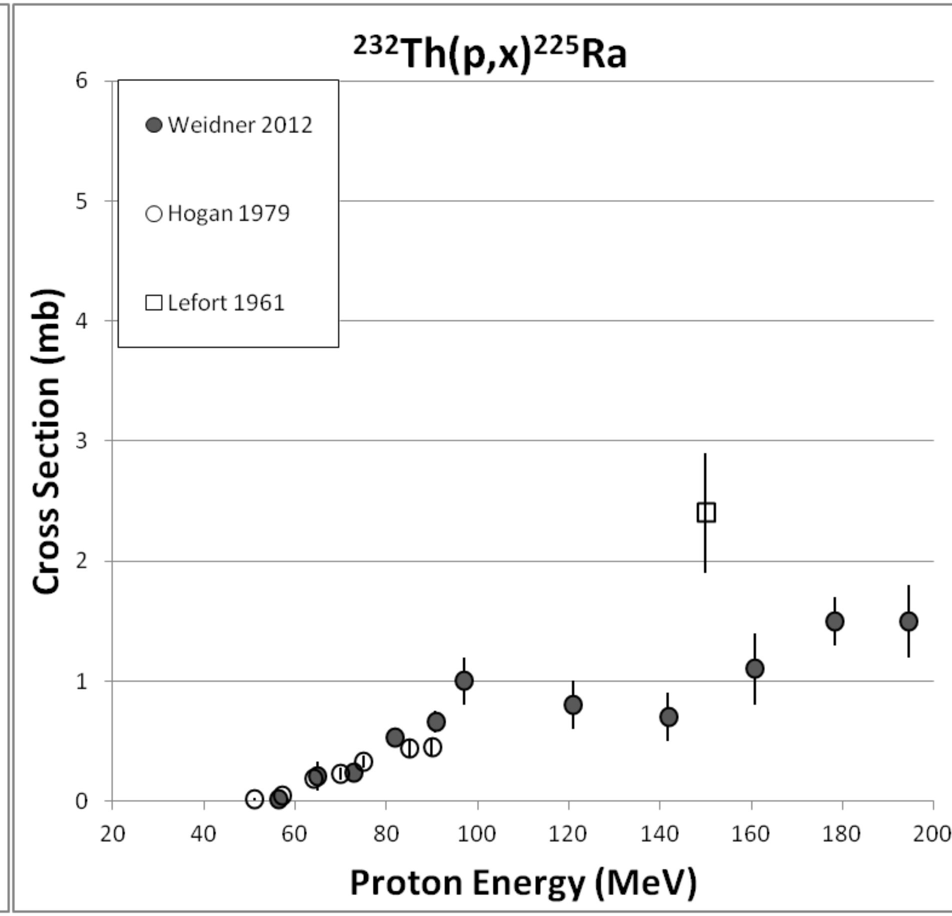




Production Cross Sections below 200 MeV
LANL Data for Ac-225 and Ra-225





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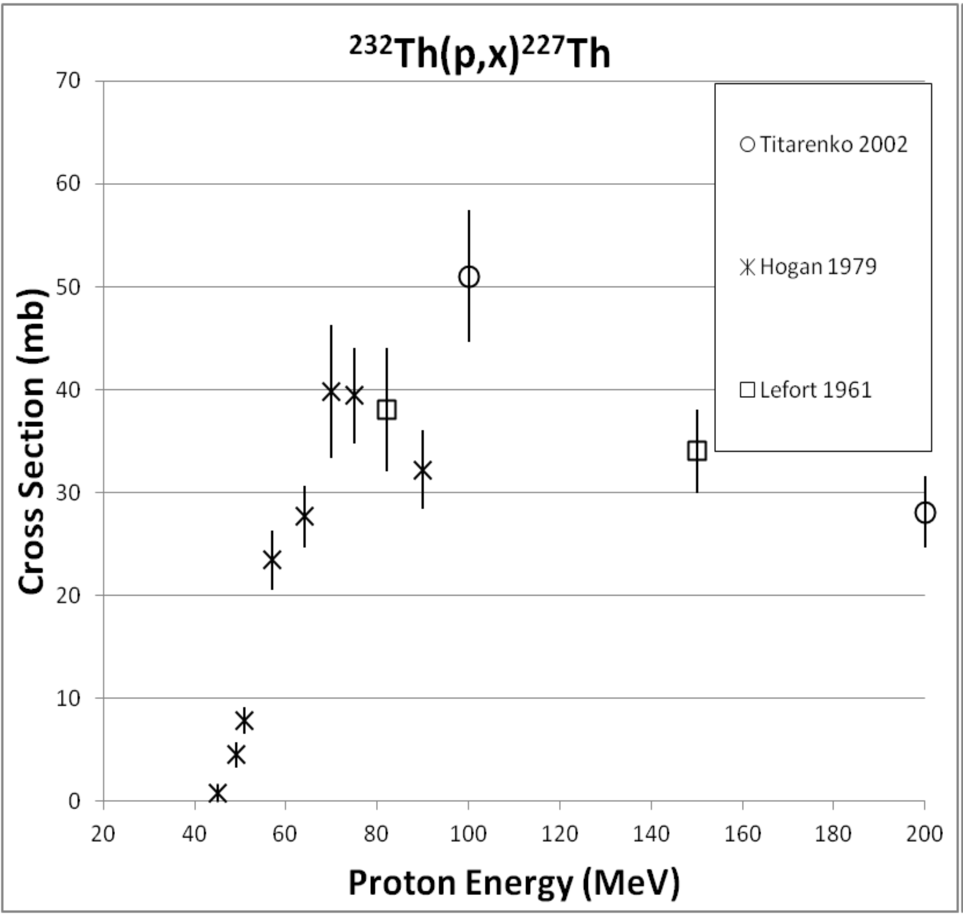
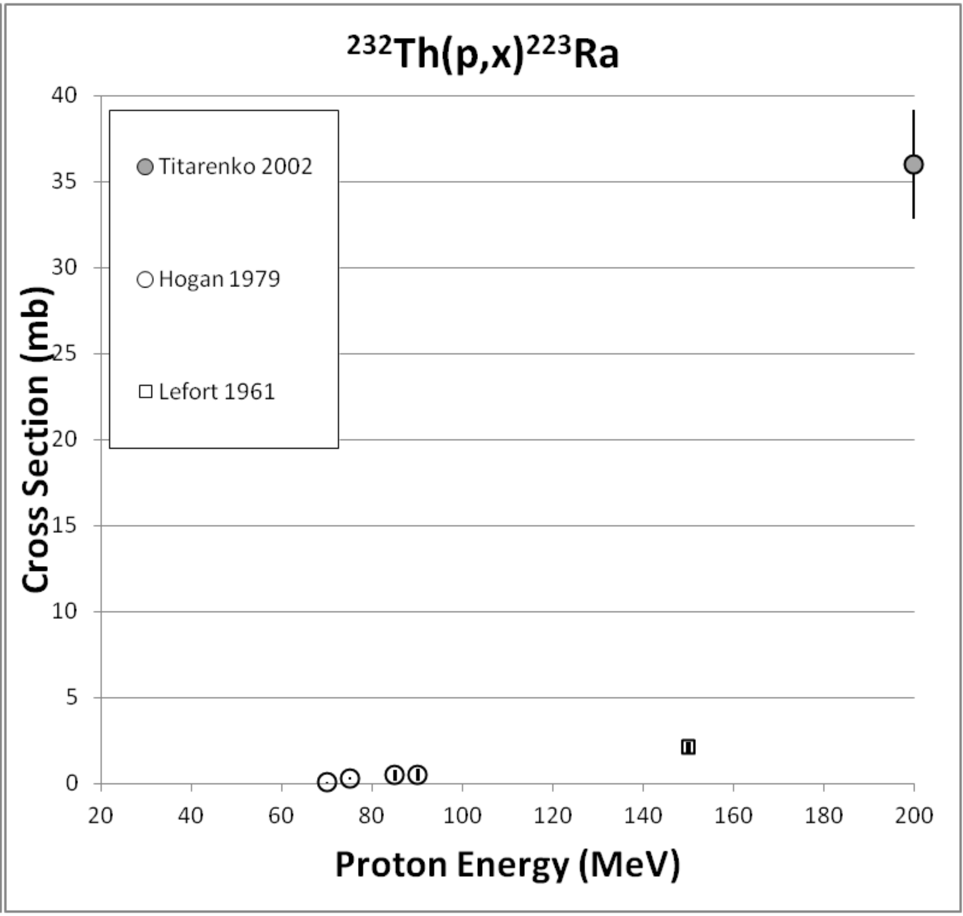
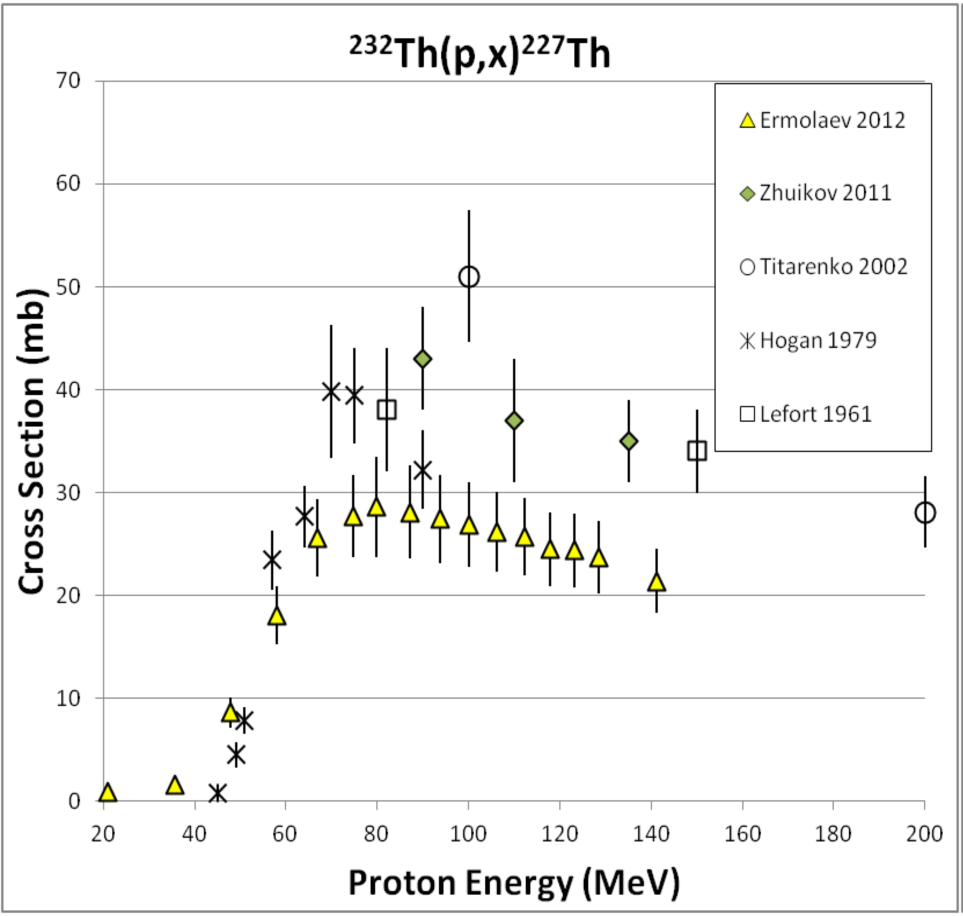
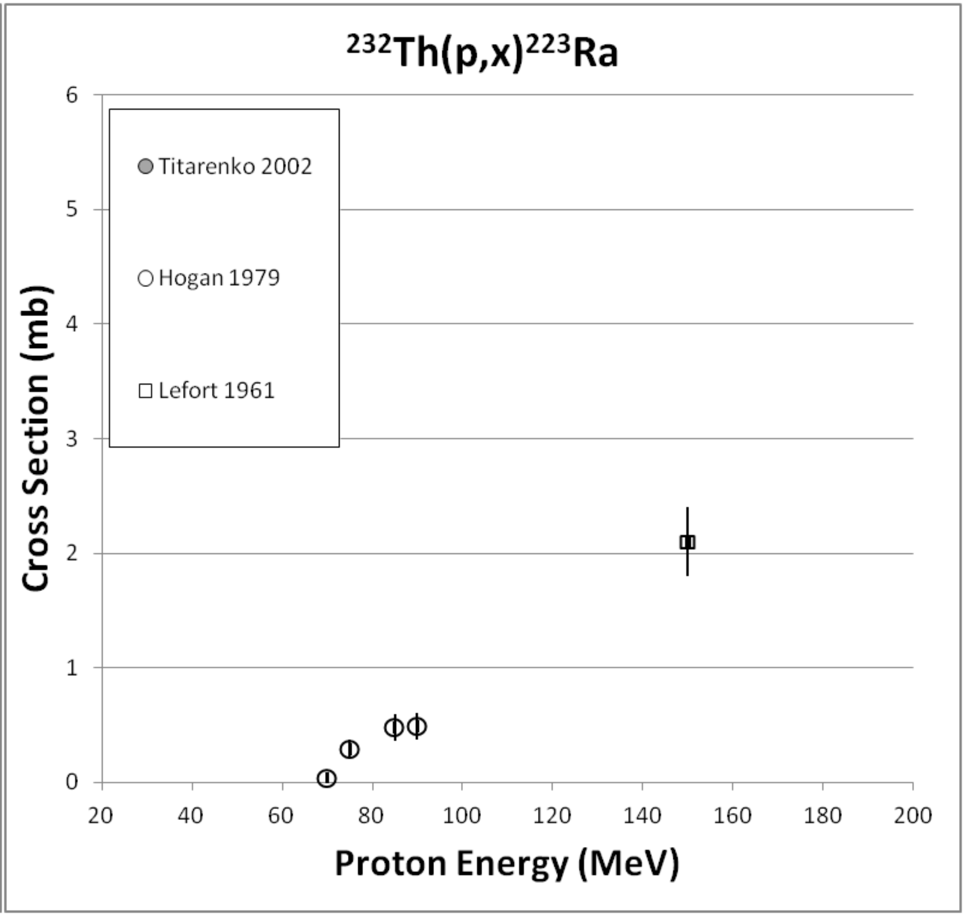
Production Cross Sections below 200 MeV Existing Data for Th-227 and Ra-223



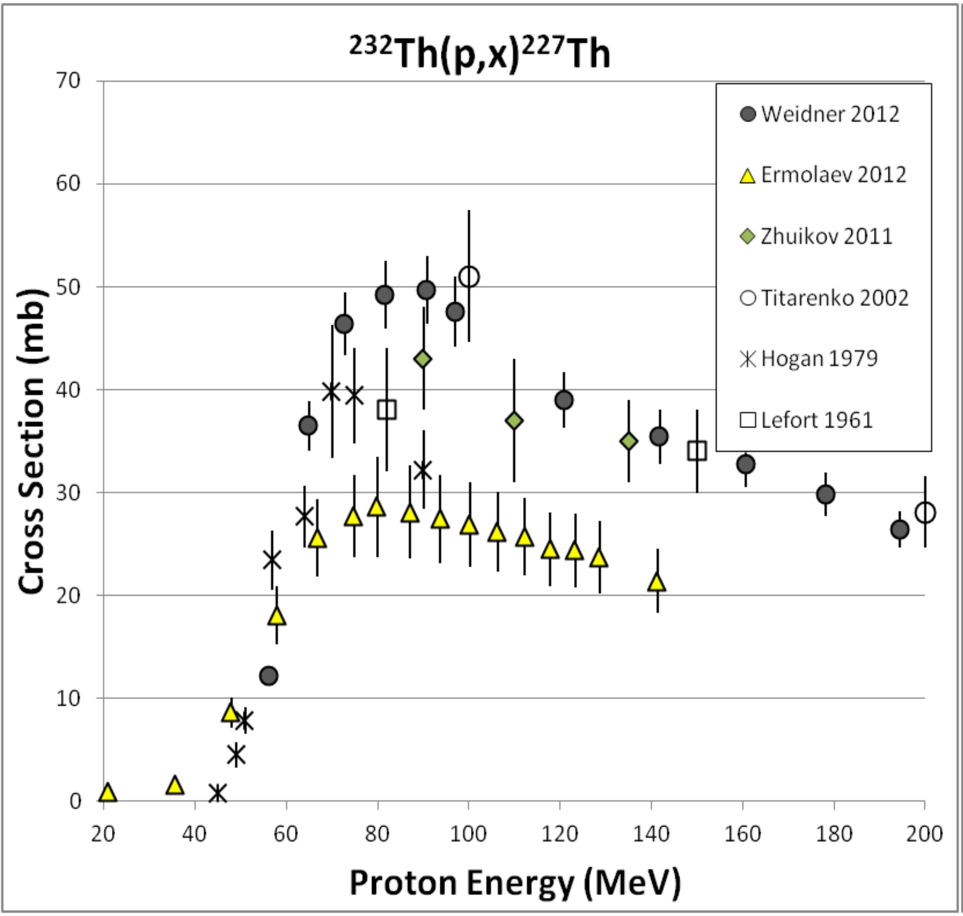


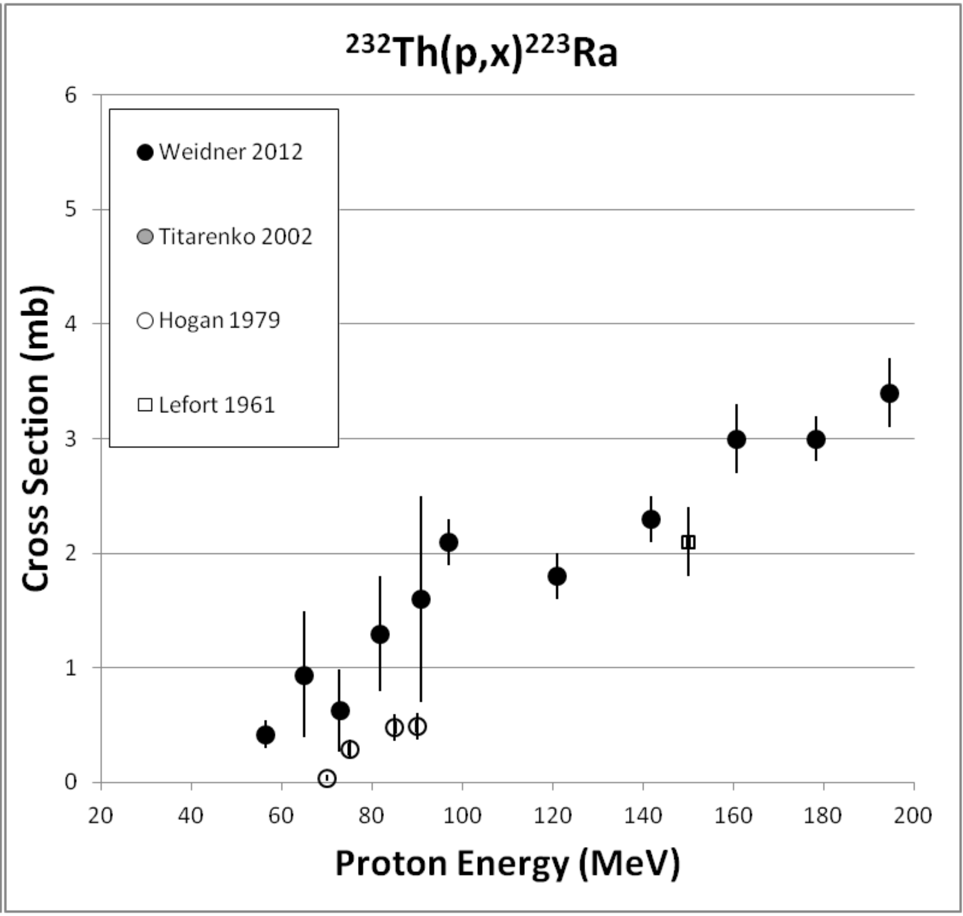
Production Cross Sections below 200 MeV
Existing Data for Th-227 and Ra-223





Production Cross Sections below 200 MeV
LANL Data for Th-227 and Ra-223





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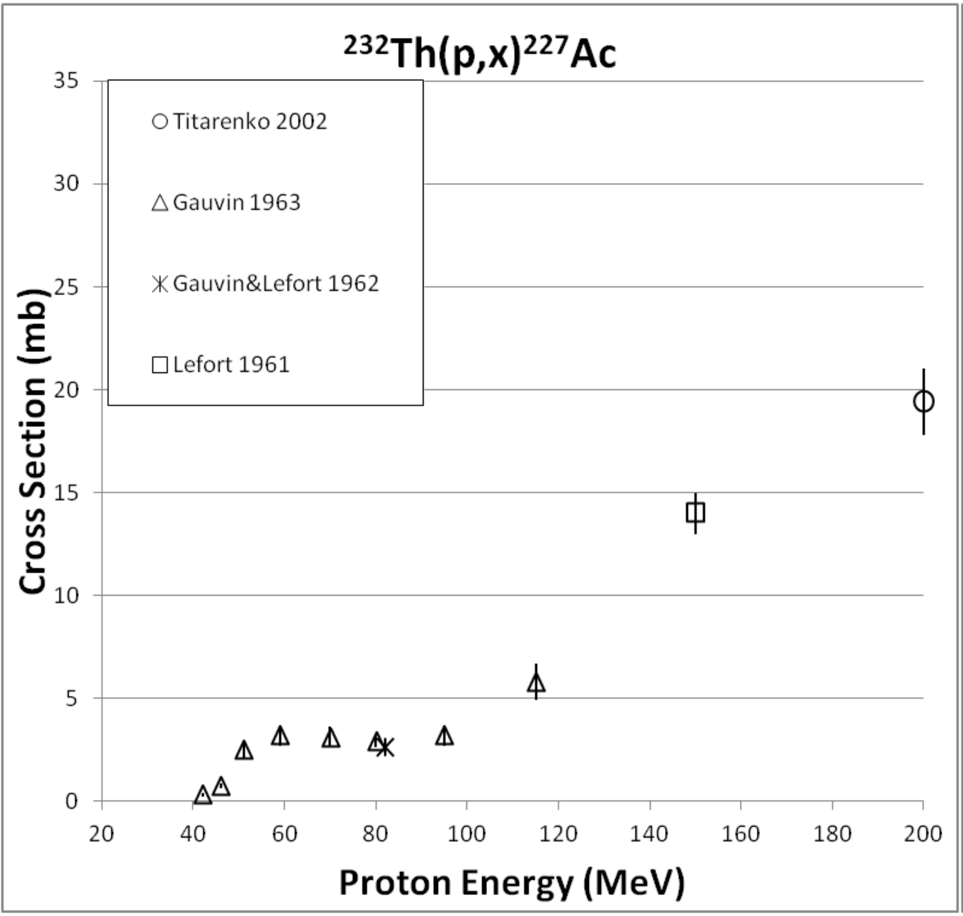
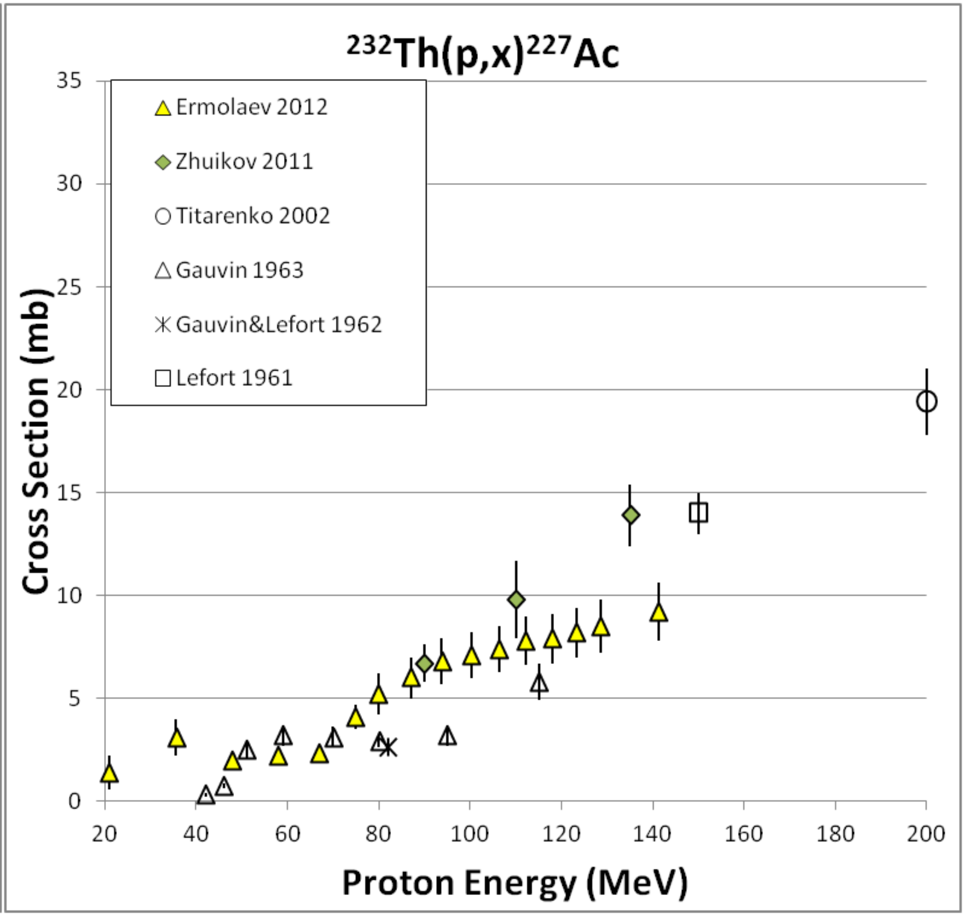
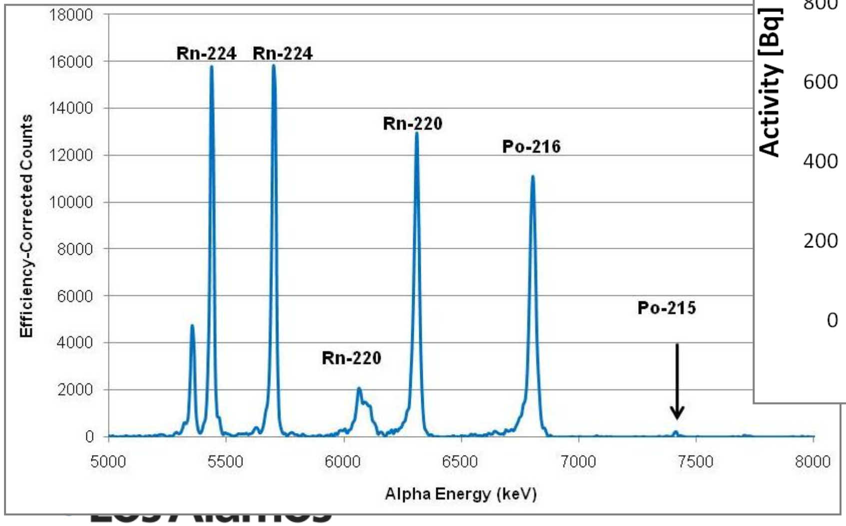
Production Cross Sections below 200 MeV
Existing Data for Ac-227

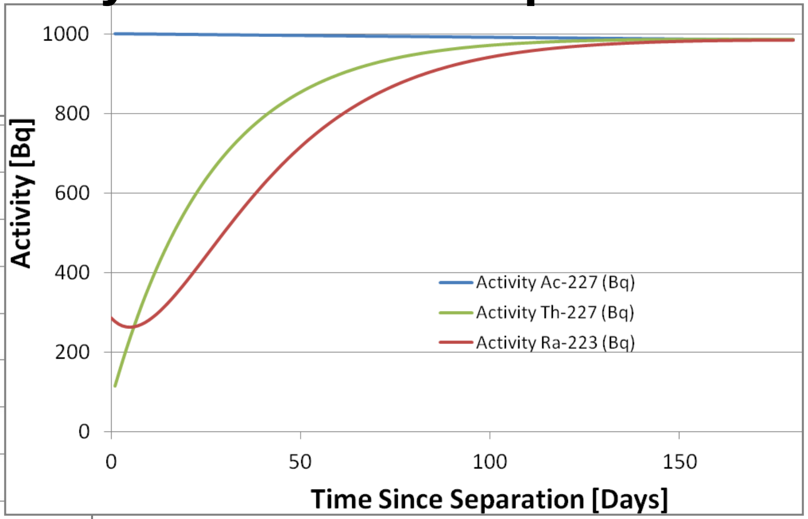


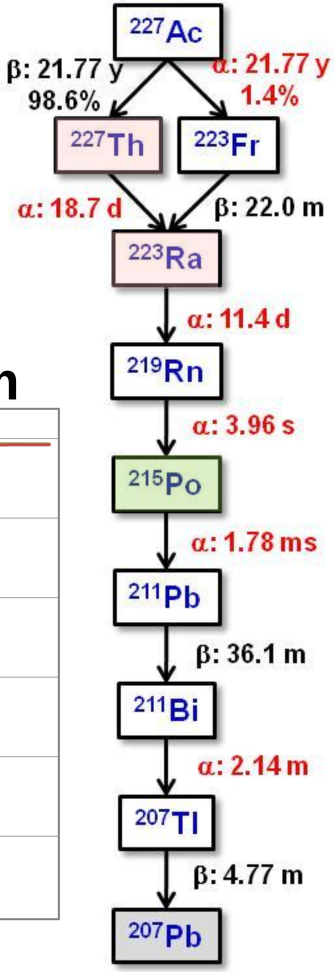


Ac-227 (21.7 y) Cross Sections Below 200 MeV
α-counting and analysis

- **LANL measurements are still in progress**
- **Ac-227 activity at EOB is based upon the measured Po-215 activity obtained from alpha spectroscopy**
- **Requires chemical separation prior to counting**
- **170 days for the Po-215 activity to come into equilibrium with the Ac-227**

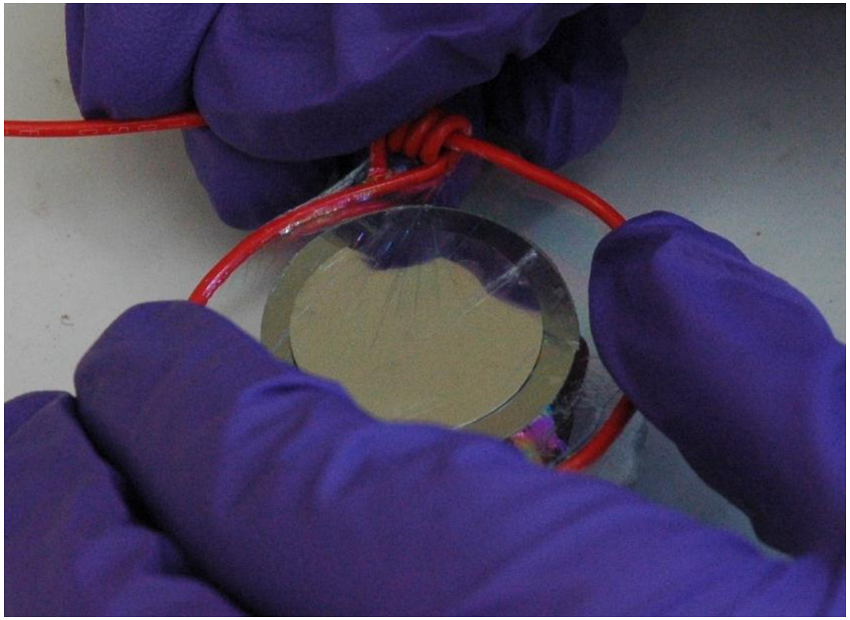


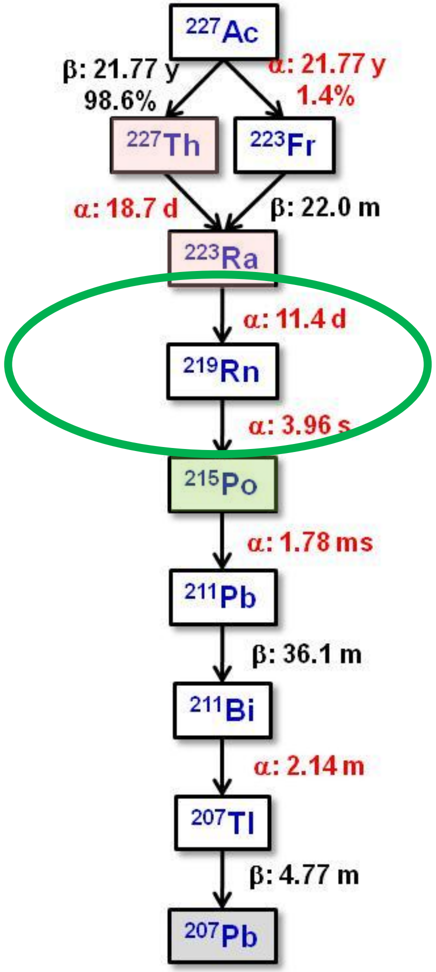




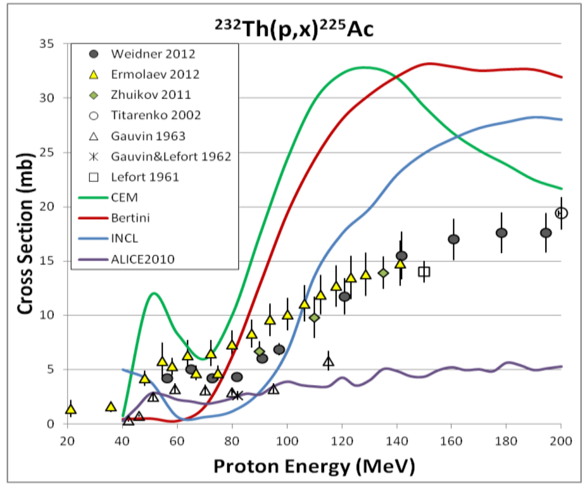
Ac-227 (21.7 y) Cross Sections Below 200 MeV
α-counting and analysis

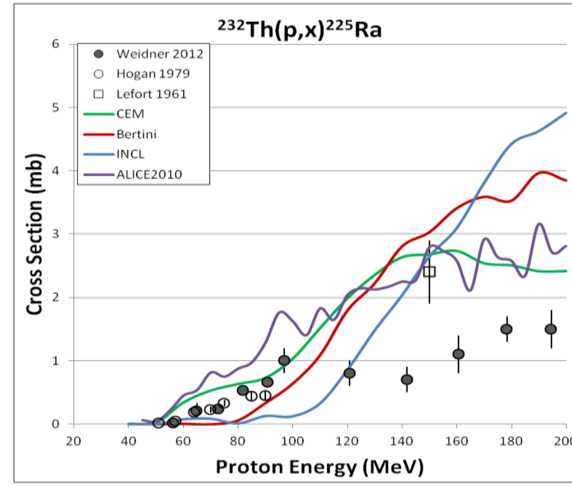
- **Extra care must be taken in quantifying Po-215**
- **Po-215 forms via decay of gaseous Rn-219**
- **To prevent the loss of Rn-219, a very thin polymer membrane is applied to the counting sample**

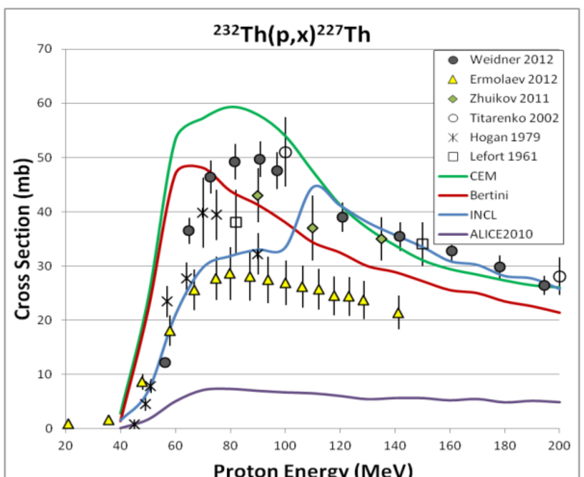


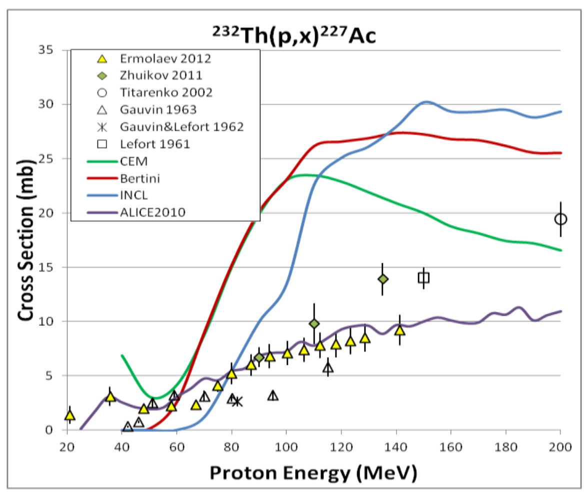


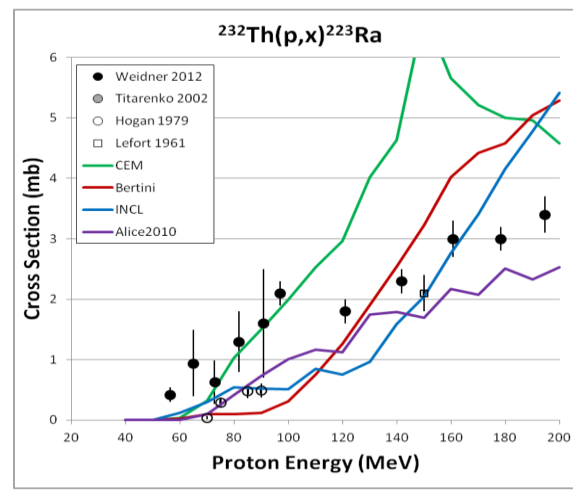
Comparison of Theory with Experiment (<200 MeV)











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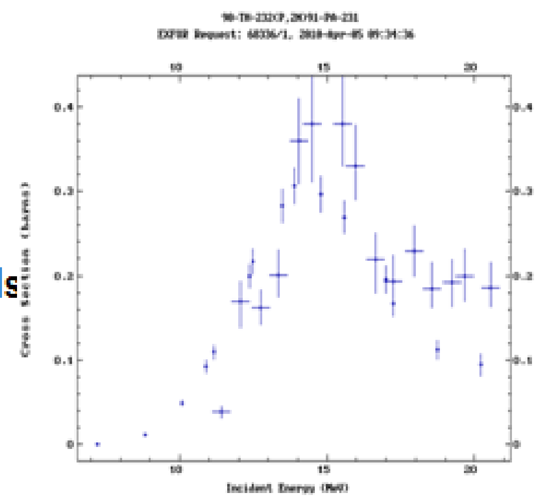
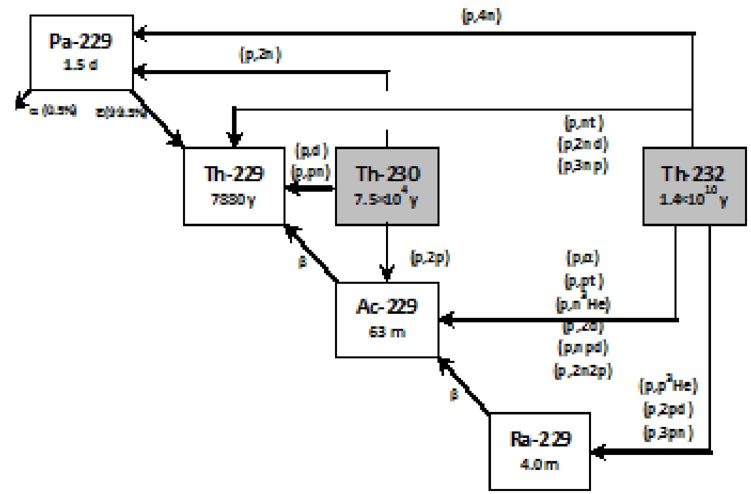
Other Measurement Efforts
^{229}Th production at ORNL (<40 MeV)

^{230,232}Th Proton Bombardment

Research Goals

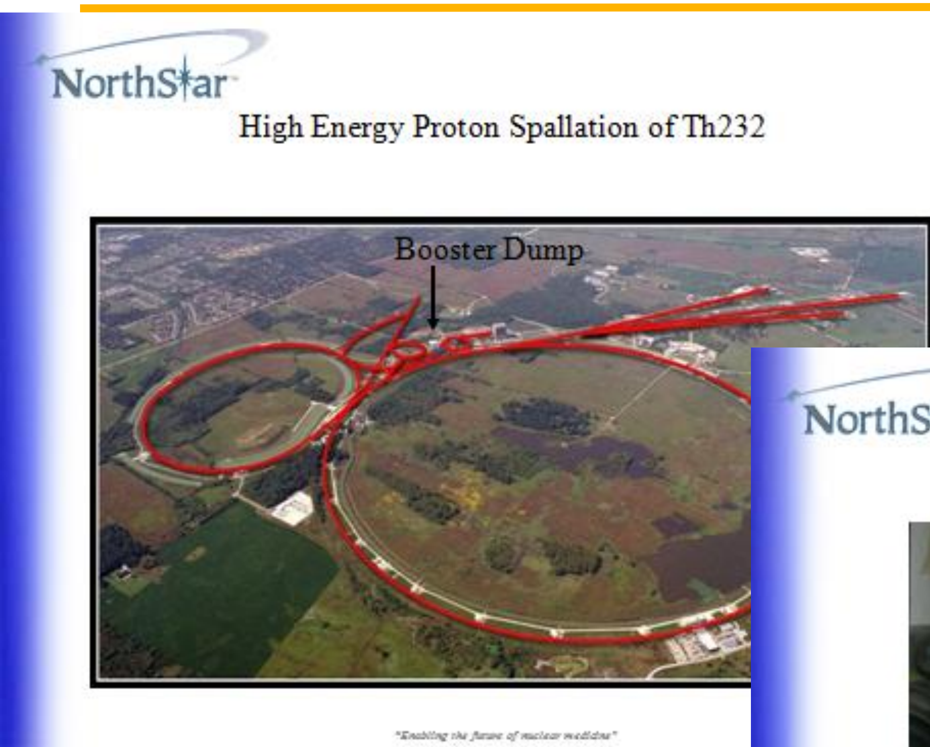
- Measure excitation functions for reactions producing ^{229}Th (and significant impurities) in ^{230}Th and ^{232}Th targets
- Evaluate thick target yields for ^{229}Th production from ^{230}Th and ^{232}Th targets

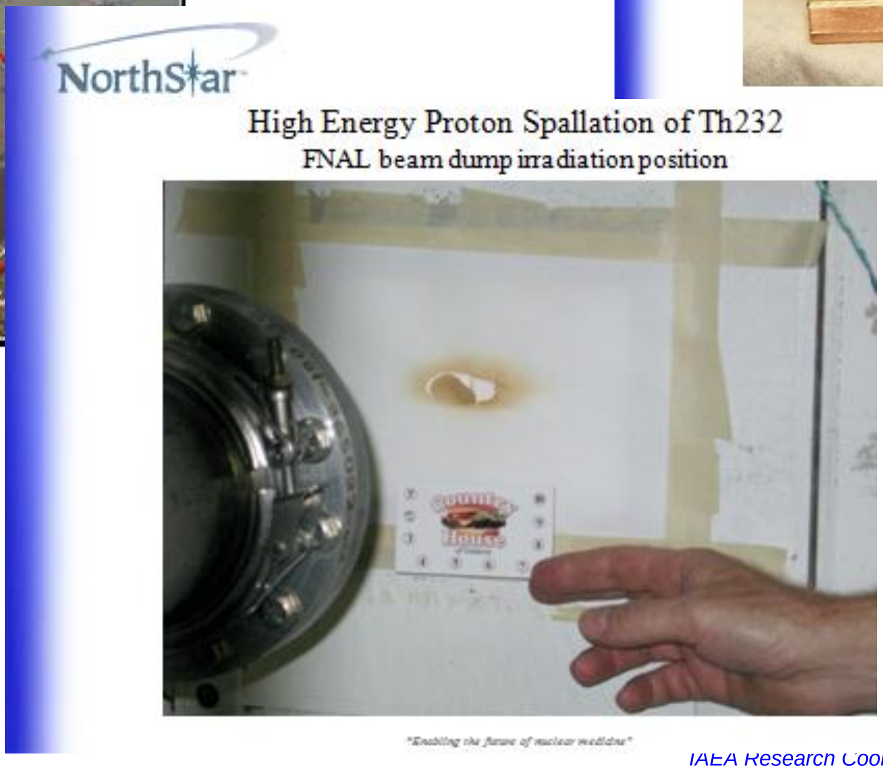




Slides Courtesy of
Saed Mirzadeh

Other Measurement Efforts
^{225}Ac production at Fermi Lab (8 GeV)

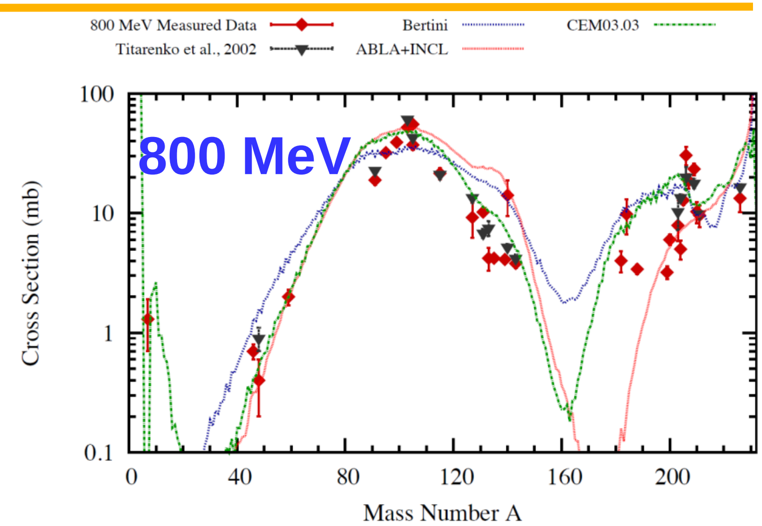




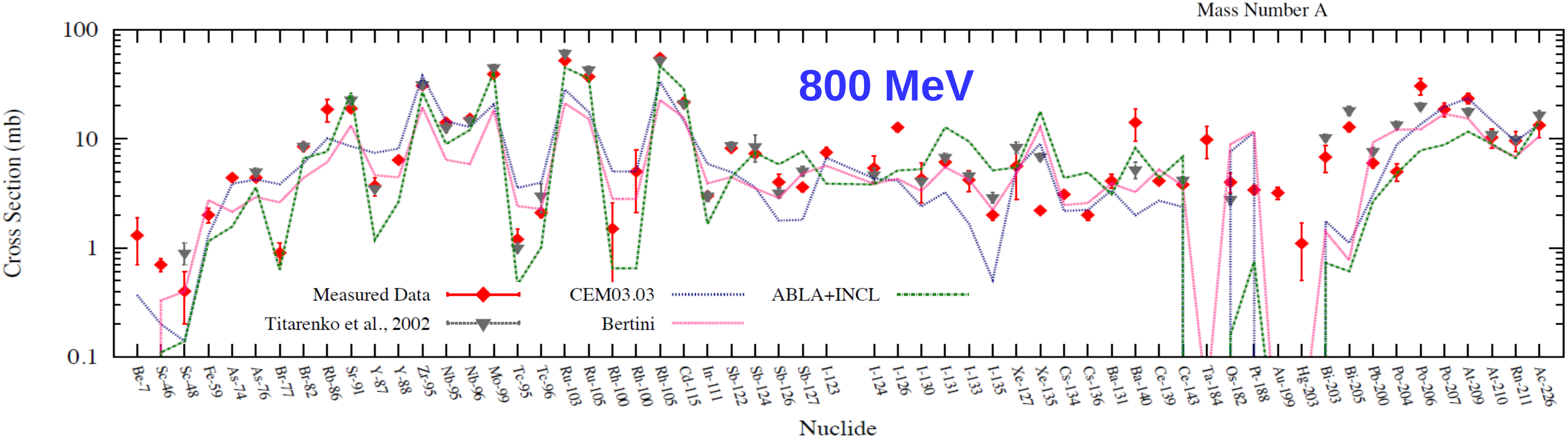
Slides Courtesy of
Jim Harvey, NorthStar

Continuing analysis of ^{232}Th +p (800 MeV and <200 MeV)

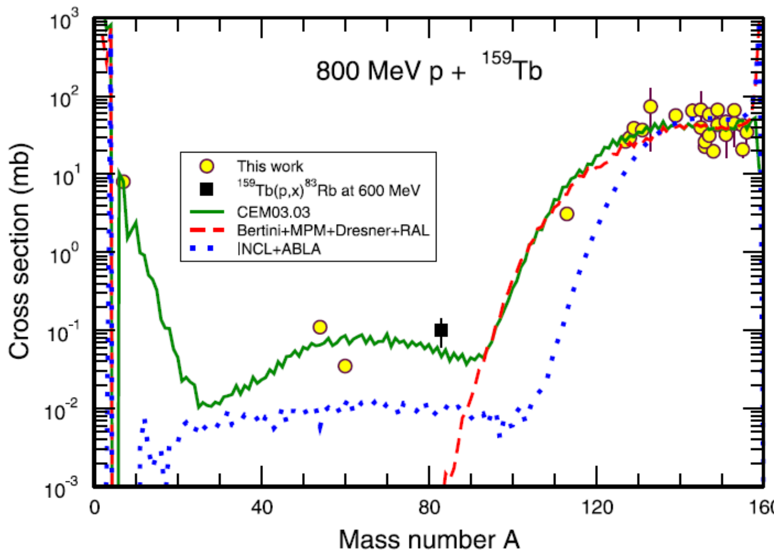
Extraction of fission product cross sections for code verification, validation and development



Analysis work performed by Dr. Jonathan Engle



Recent ^{159}Tb+p measurements at 800 MeV
Analysis of <200 MeV measurements in progress - Engle



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Cross sections from 800 MeV proton irradiation of terbium

J.W. Engle *, S.G. Mashnik, H. Bach, A. Couture, K. Jackman, R. Gritzo, B.D. Ballard, M. Fassbender, D.M. Smith, L.J. Bitteker, J.L. Ullmann, M.S. Gulley, C. Pillai, K.D. John, E.R. Birnbaum, F.M. Nortier [1]

Los Alamos National Laboratory, Los Alamos, NM 87545, USA

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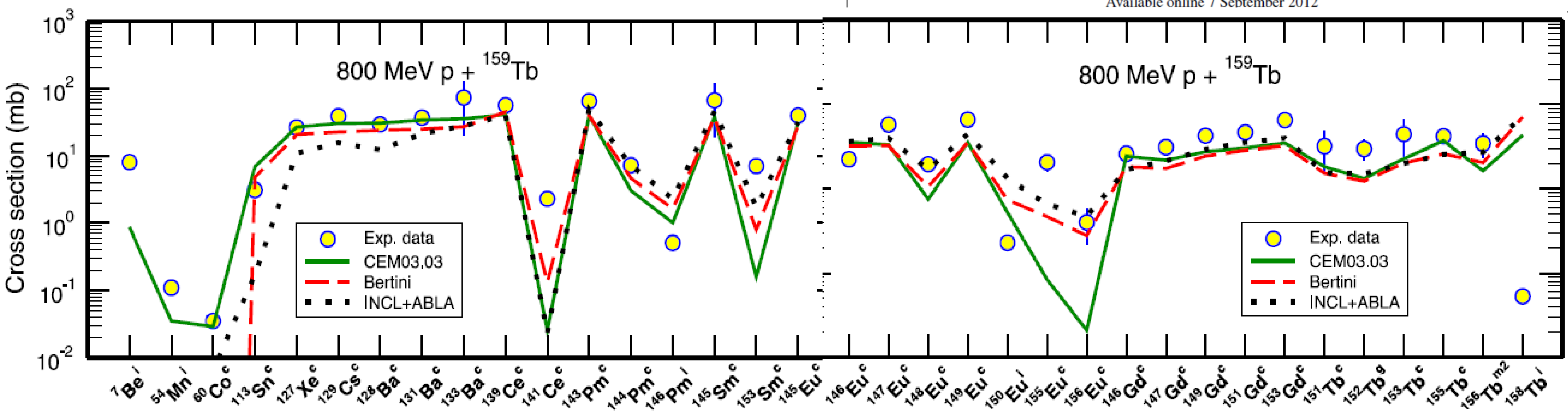


Summary

- LANL has re-established a cross section measurement capability for charged particle induced nuclear reactions
- First LANL measurements were aimed at evaluating production potential of ^{225}Ac and ^{223}Ra in natural Th targets using 100, 200, 800 MeV beams – for IPF, BLIP and spallation production routes
- Results include new cross section data which were published in two separate papers
- Measurement of ^{227}Ac in the energy range <200 MeV is still in progress
- Complimentary measurements at ORNL (<30 MeV) and FermiLab (8 GeV)
- Extraction of many more cross sections continues in support of theoretical model verification and validation

- **Additional slides**

Various $^{225}Ac/^{229}Th$ Production Routes

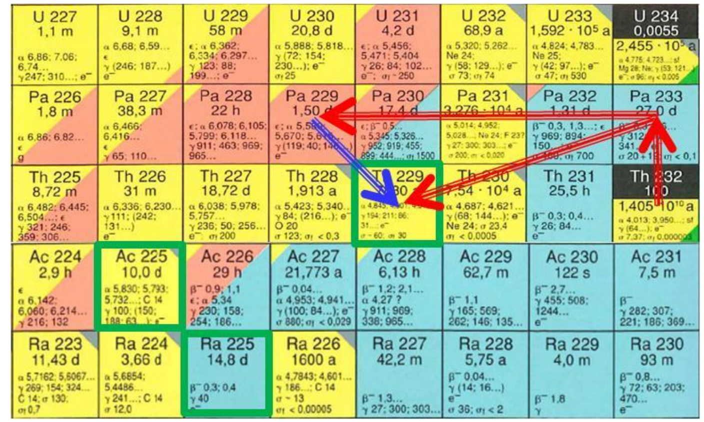
Facility	Nuclear Reaction
Reactor (thermal neutrons)	$^{226}Ra(3n,\gamma)^{229}Ra \rightarrow ^{229}Ac \rightarrow ^{229}Th$
Reactor (fast neutrons)	$^{226}Ra(n,2n)^{225}Ra \rightarrow ^{225}Ac$
Accelerator (low energy particles)	$^{226}Ra(p,2n)^{225}Ac$ $^{226}Ra(\alpha,n)\ ^{229}Th$ $^{232}Th(p,x)\ ^{229}Th$ $^{230}Th(p,x)\ ^{229}Th$
Accelerator (high energy protons)	$^{232}Th(p,x)^{225}Ac$ $^{232}Th(p,x)^{225}Ra \rightarrow ^{225}Ac$ $^{232}Th(p,x)\ ^{229}Th$
Accelerator (electrons)	$^{226}Ra(\gamma,n)^{225}Ra \rightarrow ^{225}Ac$

Evaluate Higher Energy Accelerator Production Routes using thorium targets

Th-229 production

$^{232}Th(p,4n)^{229}Pa \xrightarrow{99.5\%} {}^{229}Th$

$^{232}Th(p,p3n)^{229}Th$



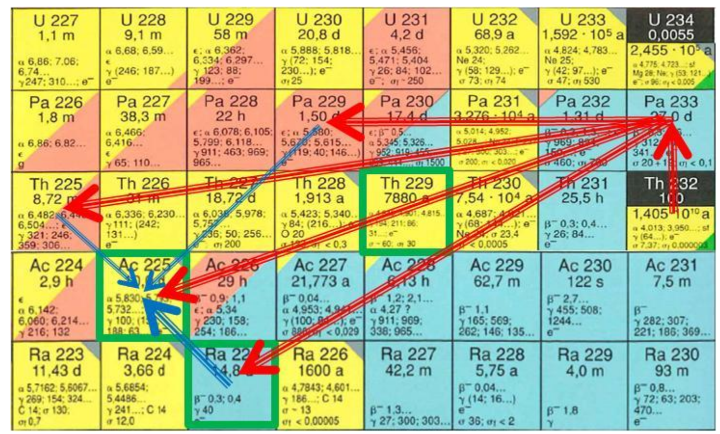
Ra-225/Ac-225 production

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

$^{232}Th(p,x)^{225}Ra \xrightarrow{100\%} {}^{225}Ac$

$^{232}Th(p,x)^{229}Th \xrightarrow{10\%} {}^{225}Ac$

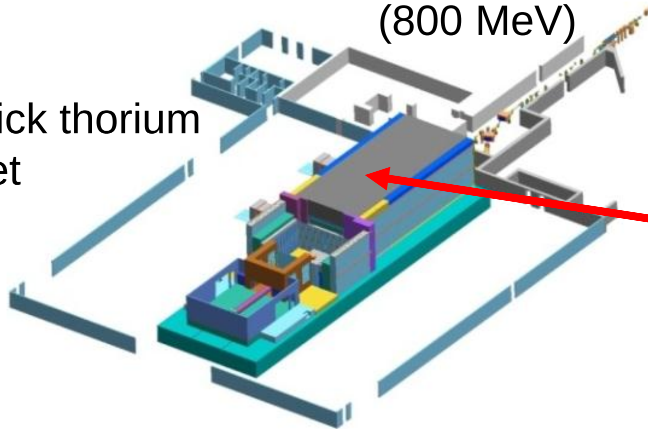
$^{232}Th(p,4n)^{229}Pa \xrightarrow{0.5\%} {}^{225}Ac$

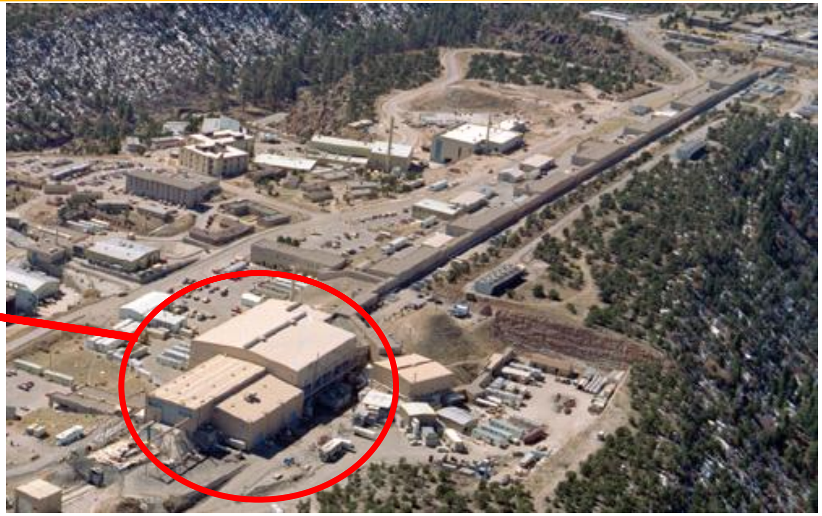


800 MeV production potential at the future MTS

1250 µA proton beam
(800 MeV)

3 g/cm^2 thick thorium
metal target





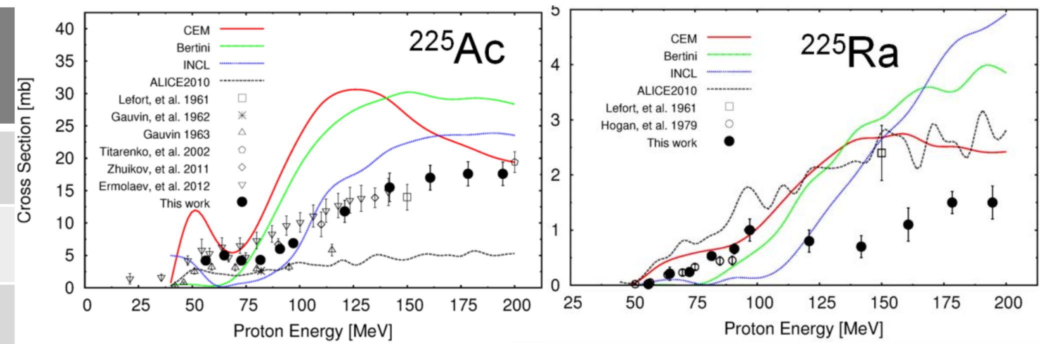
Measured production cross sections translate into a production potential of :

- 1.6 Ci of directly-produced ^{225}Ac per day (0.17% ^{227}Ac impurity level)
- 250 mCi of ^{225}Ra per day, which translates into 140 mCi of pure ^{225}Ac

Isotope	$T_{1/2}$	Expected Yields
^{225}Ra	14.8 d	250 mCi/day (~140 mCi of pure ^{225}Ac)
^{225}Ac	10 d	1.6 Ci per day (0.17% ^{227}Ac)
^{223}Ra	11.4 d	550 mCi per day
^{227}Th	18.7 d	780 mCi per day
^{227}Ac	21.7 y	1.0 Ci per year

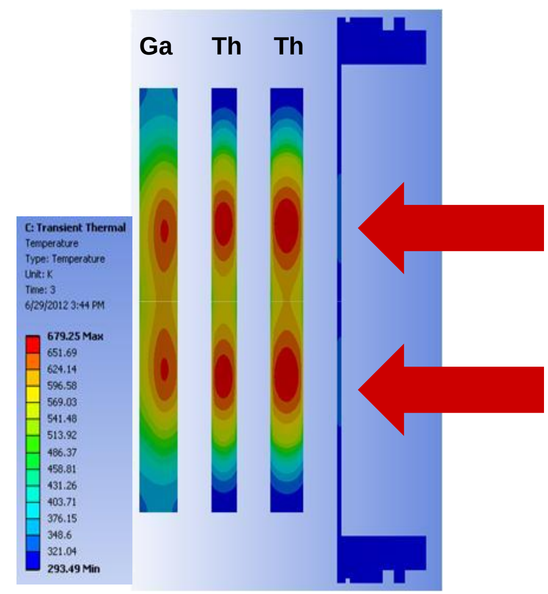
Targetry Advances – Ac-225 production - Cross section measurements

	5 g/cm^2 target yield for a 10 day irradiation	
	Ac-225 (Ci)	**Ra-225** (Ci)
IPF (250 µA)	1.4	0.1
BNL (100 µA)	2.0	0.1



Weidner et al. Appl. Radiat. Isot. 2012

- **Data show that large scale production is feasible at IPF and BLIP**
- **Predicted co-production of ^{227}Ac is non-zero but low (<0.2%)**
- **Small scale proto production foil irradiations proceed at BNL to support ORNL chemical recovery development**
- **High current targetry is being developed by LANL for full-scale production at both facilities**



Production Potential

a Instantaneous production rate, which does not account for decay
b Values calculated from 227Ac cross section measurements by Ermolaev, et al. and ALICE2010 predictions

	IPF (250 µA, 93-72 MeV)		BLIP (100 µA 195-183 MeV)	
	Production Rate[a] [µCi/µA·h]	Yield [Ci]	Production Rate[a] [µCi/µA·h]	Yield [Ci]
^{225}Ac	33.1	1.4	115.6	2.0
^{223}Ra	6.8	0.3	18.8	0.3
^{225}Ra	2.6	0.1	6.7	0.1
^{227}Th	173.1	8.7	95.7	1.9
^{227}Ac[b]	0.04	0.003	0.09	0.002

Production rates and projected yields from a 10-day irradiation of a 5 g/cm2 natural thorium target at the Los Alamos National Laboratory Isotope Production Facility and Brookhaven National Laboratory. The energy range of the protons within the 5 g/cm2 thick thorium target is shown.