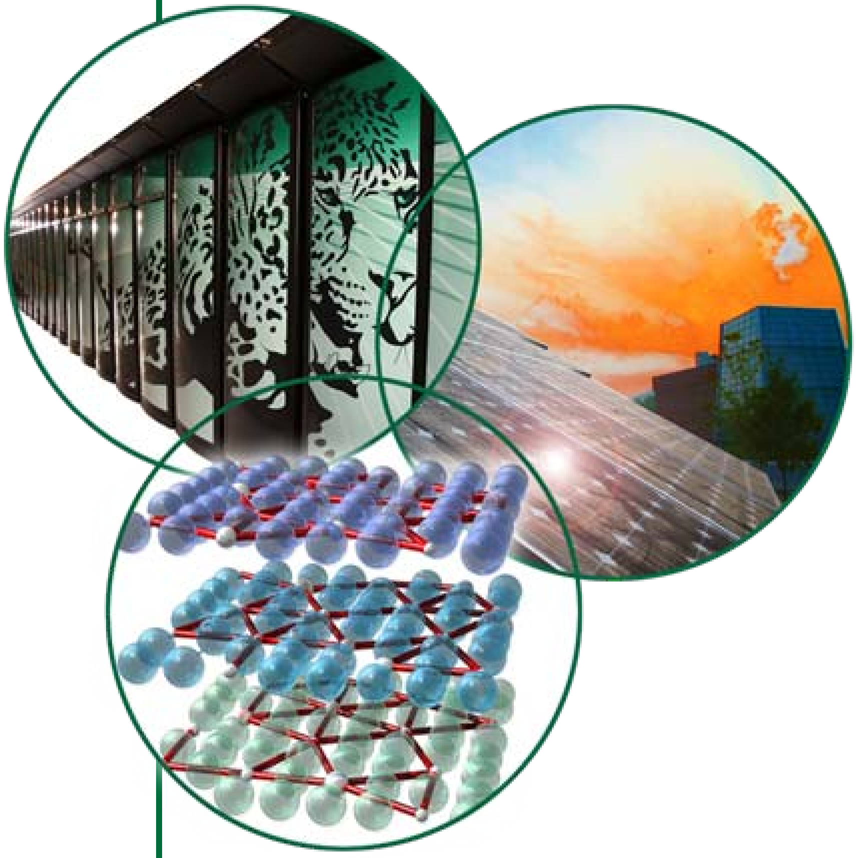
R&D on Accelerator-Based Production of ^{229}Th

Dan Stracener

Office of Science
Nuclear Physics Program
Isotope Program Review

Oak Ridge National Laboratory
Oak Ridge, Tennessee

September 15-16, 2011



U.S. DEPARTMENT OF ENERGY

OAK RIDGE NATIONAL LABORATORY
MANAGED BY UT-BATTELLE FOR THE DEPARTMENT OF ENERGY

Organization of Talk

- Motivation for ^{229}Th project
- Experimental plan and setup
- List of irradiations and results
  - Benchmark experiments
  - Measured cross-sections for nuclei around ^{229}Th
- What still needs to be accomplished

ORNL Isotope Program Goals

The ORNL Isotope Program will be a science and technology leader for ensuring a sustainable supply of specific radioisotopes and enriched stable isotopes for research, medicine, national security, and industry.

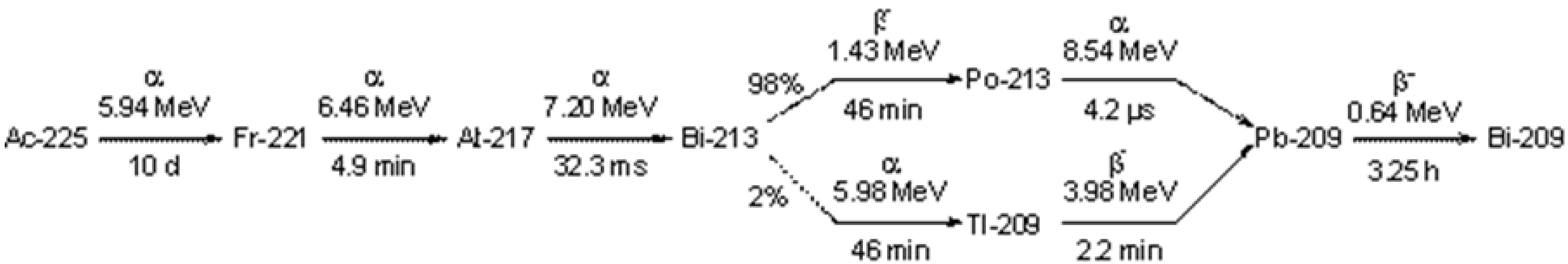
1. Production of specific trans-uranium elements and other radioisotopes that can be uniquely produced by ORNL for research, medicine, and industry. (NSACi 1-3, 2-1)
2. Develop a new capability for the production of enriched stable isotopes. (NSACi 1-5, 2-3)
3. Develop alpha-emitters and other important medical radioisotopes for diagnostics and therapy. (NSACi 1-1, 2-1)
4. Develop a sustained isotope research program. (NSACi 1-4, 2-1)

Overview of ^{229}Th Project

- Main goal:
  - Evaluate an alternative method for production of ^{229}Th (precursor of ^{225}Ac and ^{213}Bi) via proton bombardment of ^{230}Th and ^{232}Th targets
- Specific Aims:
  - Measure excitation functions for the 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
- 2-year project funded by ARRA funds in FY10

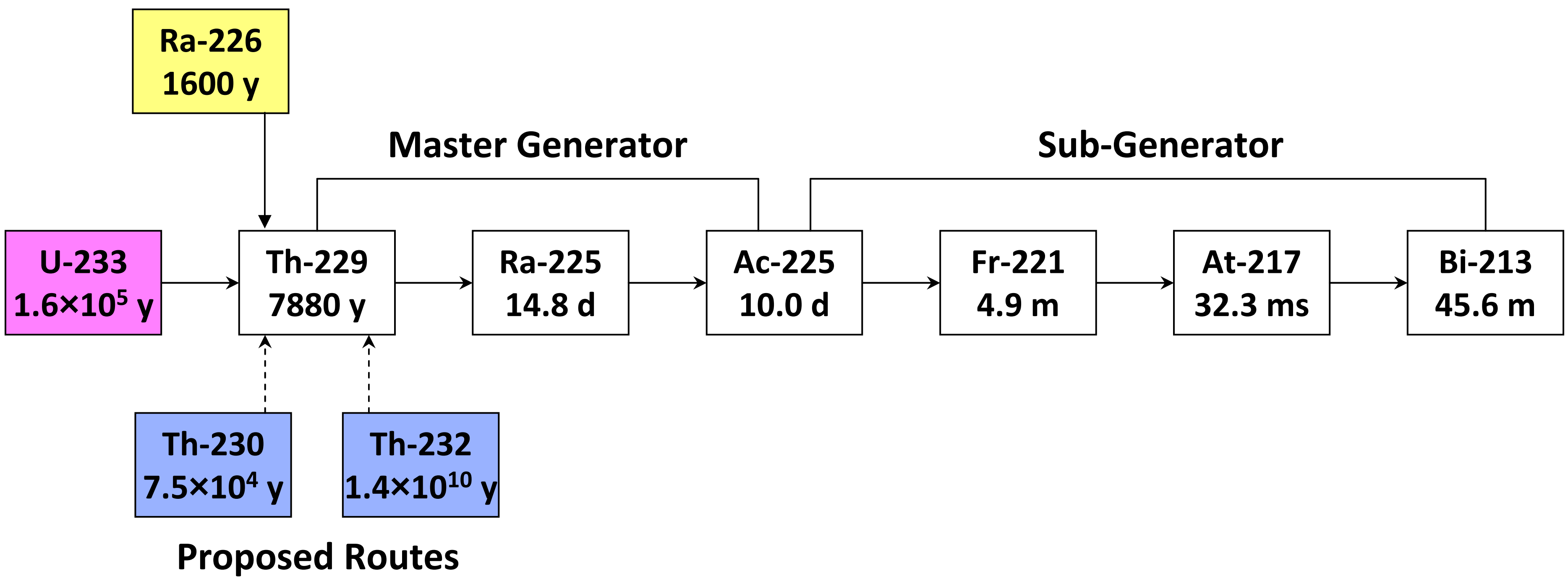
Why use ^{225}Ac / ^{213}Bi for cancer therapy?

- Alpha-emitting radioisotopes are important for cell-directed therapy



- Alpha-emitters with relatively short half-lives are of particular interest
  - ^{225}Ac (10.0 d), ^{213}Bi (45.6 m), and ^{211}At (7.22 h)
  - ^{225}Ac and ^{213}Bi can easily be 'milked' from a long-lived parent, ^{229}Th (7880 y)
  - distribution of ^{229}Th 'cows' could be accomplished with relative easy (perhaps in a way similar to the current use of ^{99}Mo for production of ^{99m}Tc, which is used in about 85% of all radioisotope applications)

^{229}Th decay chain

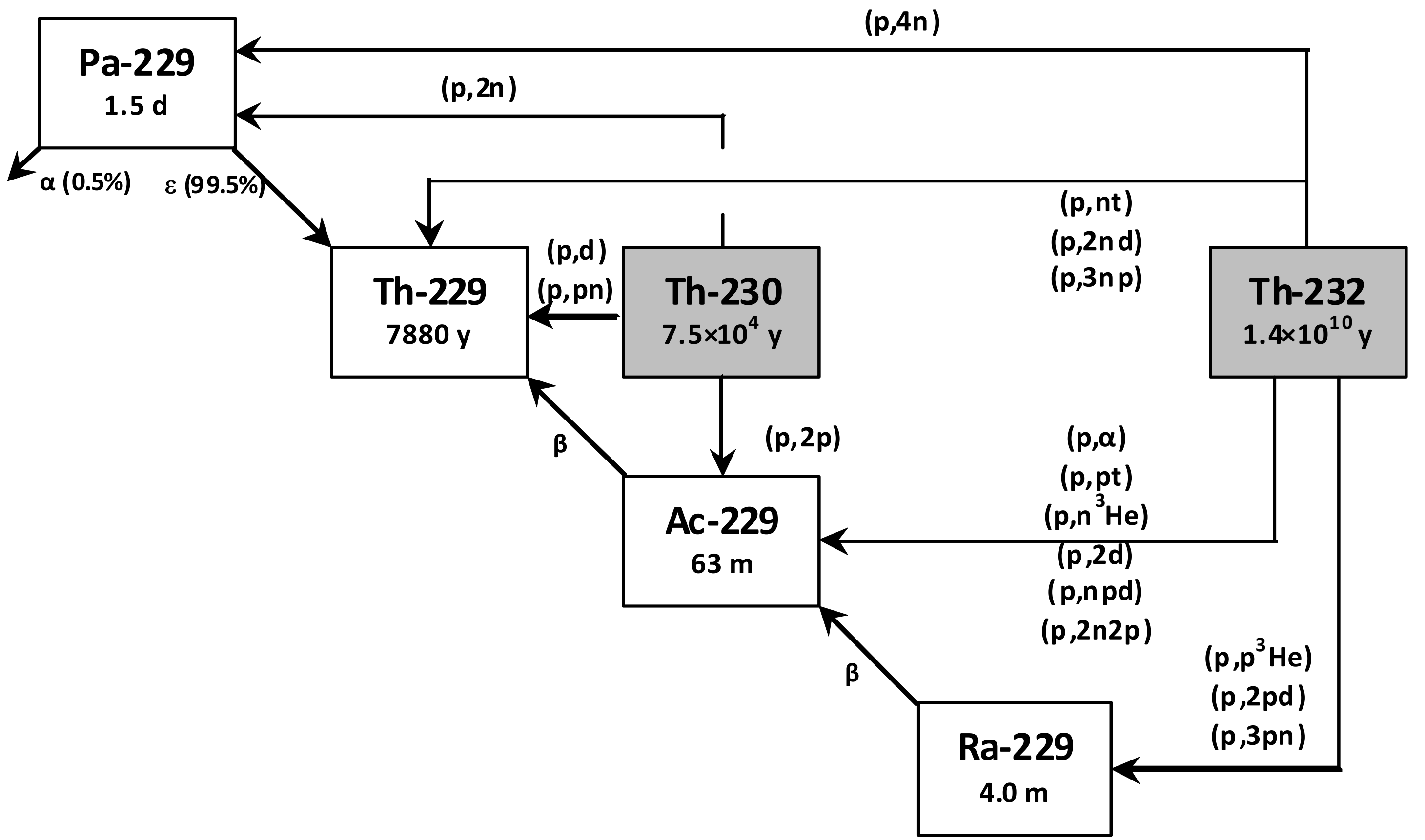


Current Methods used to Produce ^{225}Ac

- Extraction from aged ^{233}U stockpiles
  - Supply of ^{233}U is limited and access is limited
- Irradiation of a ^{226}Ra target (half-life is 1600 years) in a high flux reactor
  - $^{226}Ra(3n,2\beta)^{229}Th$
  - Yield of ^{228}Th is 1000 times greater (half-life is 1.9 years)
  - 2.6 MeV γ-ray in the ^{228}Th decay chain (from ^{212}Po) poses shielding problems for large-scale production
- Direct reactions in a ^{226}Ra target - (p,2n) or (γ,nβ)
  - The relatively short half-lives of ^{225}Ra and ^{225}Ac (14.8 days and 10.0 days) require fast turn-around times for processing
  - Demand would require nearly continuous processing of radium targets, which is far more challenging than 'milking' ^{225}Ra and ^{225}Ac from long-lived ^{229}Th
- Spallation from ^{232}Th using high energy proton beams
  - Requires that ^{229}Th or ^{225}Ac be separated from a large array of reaction products
- **Need to develop a stockpile of ^{229}Th**

Investigation of an alternative method

- Need to develop some additional production capacity since the current availability of ^{225}Ac and ^{213}Bi is insufficient to support existing clinical trials
- Investigate some of the proton-induced reactions shown below to determine the efficacy of this concept to produce ^{229}Th



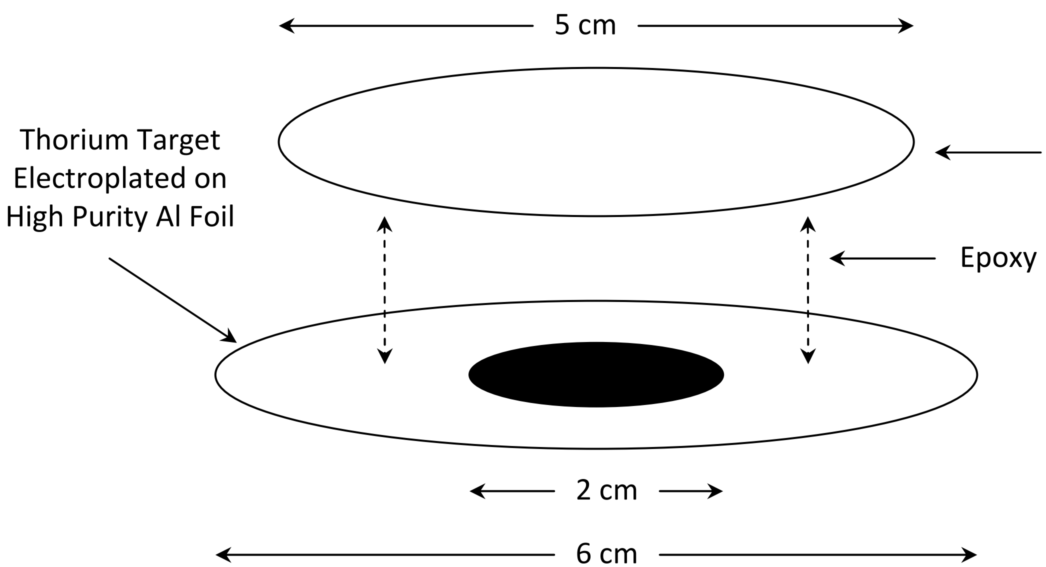
Experimental Approach

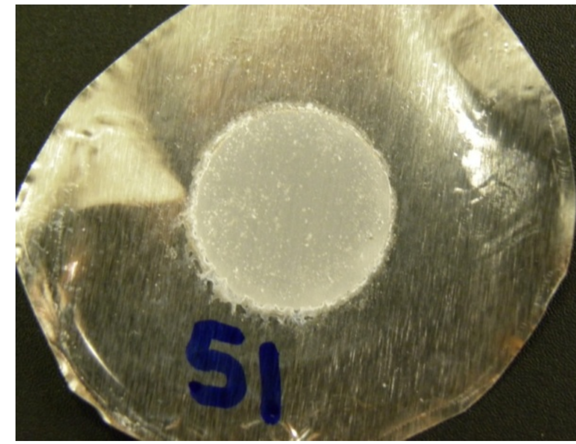
- Produce thin metal foils with known thickness
  - use copper and nickel foils for benchmarking the technique
  - electroplate thin layers of ^{230}Th and ^{232}Th on aluminum support foils
  - make a stack of several foils interspersed with aluminum foils with thickness selected to degrade the beam energy to the distribution desired
- Irradiate these stacks of thin foils using a proton beam with specific and well-defined energy
- Determine the number of radioactive nuclei produced in each foil
  - detect γ-rays from the radioactive decay of each of the reaction products
- Calculate the production cross-section as a function of energy
- Irradiate a thick ^{232}Th target
  - compare the number of ^{229}Th nuclei produced with the number expected from the thin-target measurements described above

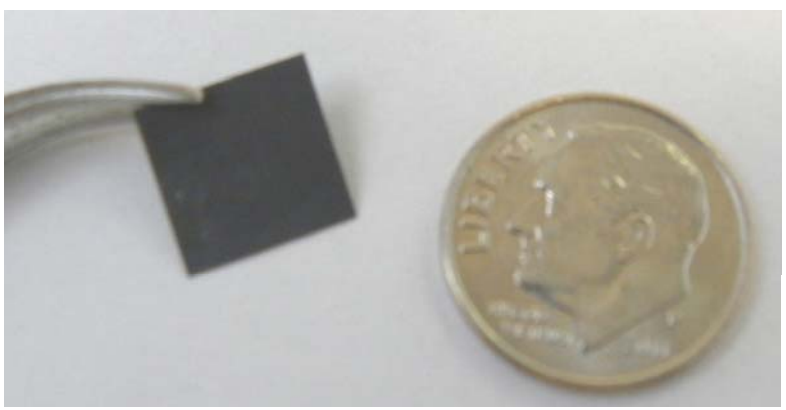
Tandem electrostatic accelerator at HRIBF

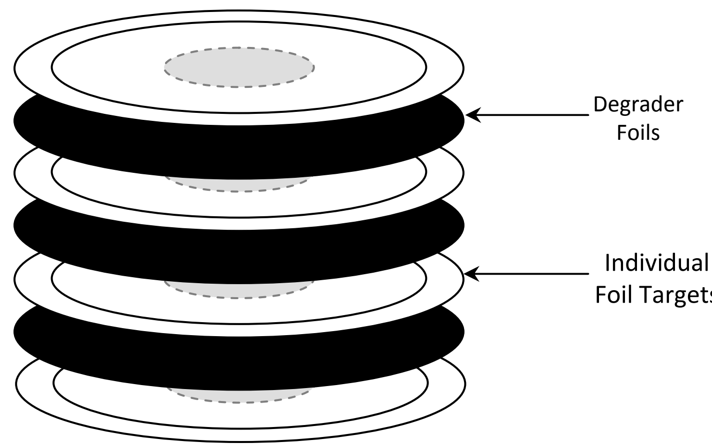
- Wide range of beams and energy
  - Up to 48 MeV protons, deuterons
  - Up to 72 MeV alphas
- Easy energy variation
- Excellent beam quality
  - Small beam spot size (<2 mm)
- Reliable and economical
- Proton beam intensity is presently administratively limited to 50 nA, but can be increased to 1 μA
- Ideal for R&D on isotope production using light ions

Thorium targets for excitation function measurements (see poster by Justin Griswold)





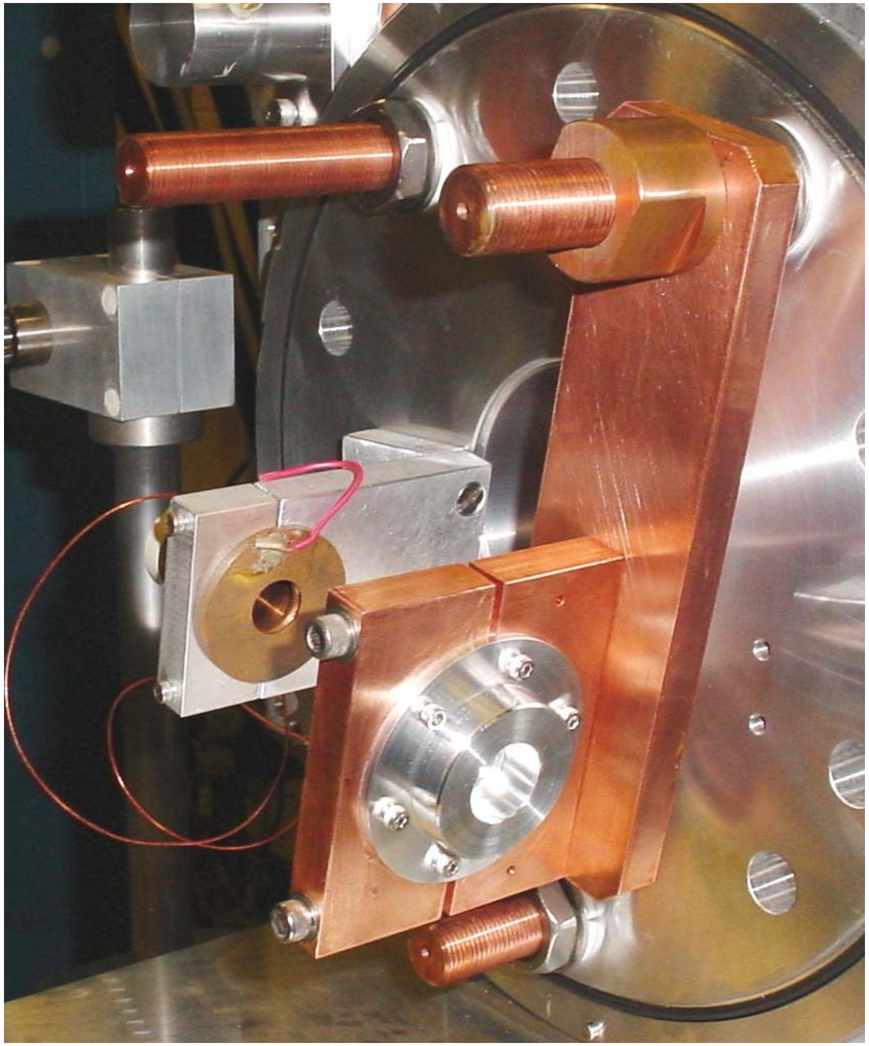




^{232}Th foils (0.125 mm thick) used for 2 target stacks (purchased from Goodfellow and borrowed from LANL)

Target irradiations at the HRIBF

Thin metal foils separated by aluminum foils to degrade the beam energy



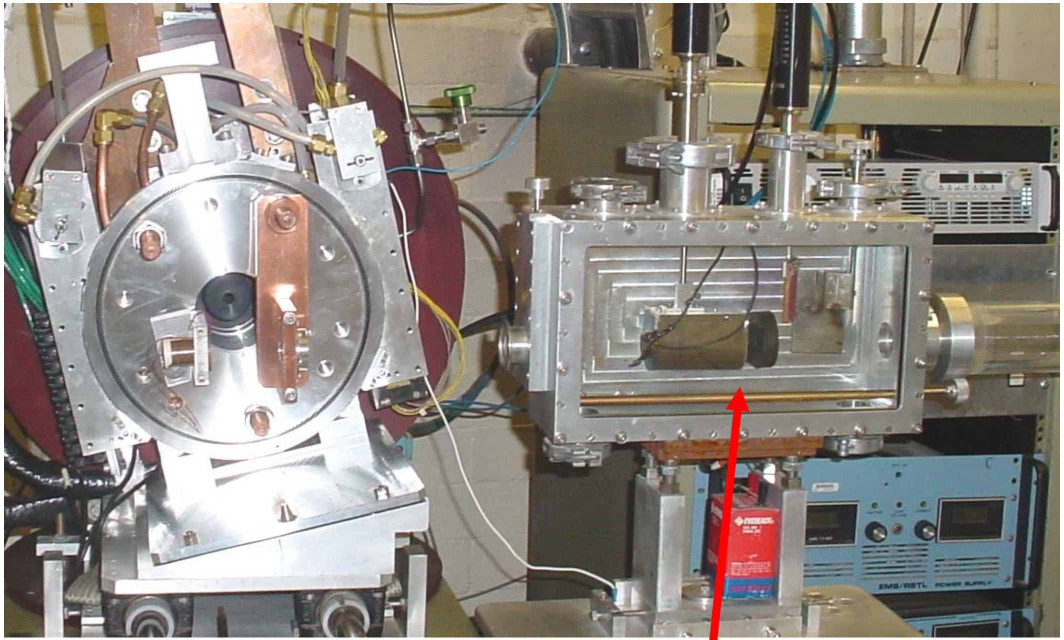
Target holder installed in vacuum chamber with Faraday Cup mounted downstream to measure beam current during experiment

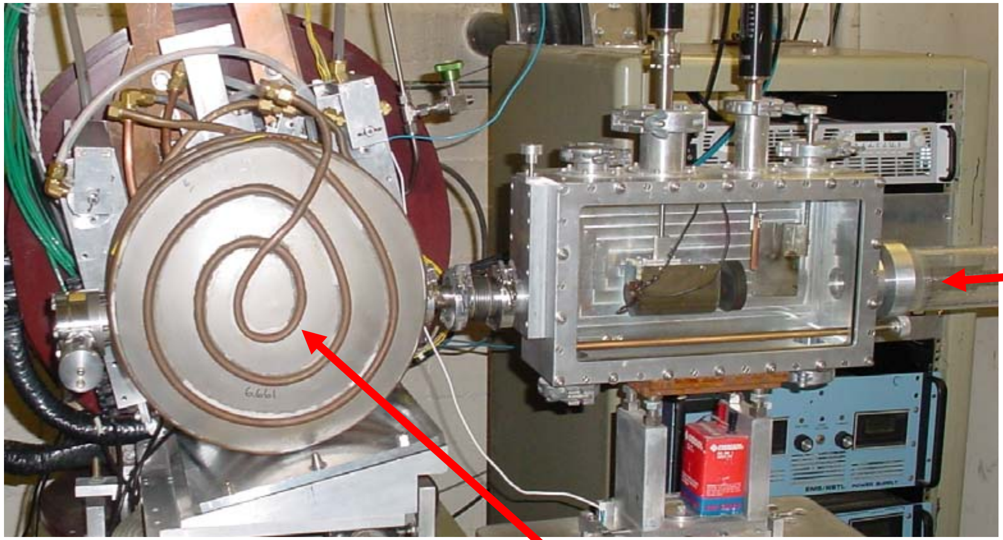
Target holder with ^{232}Th foils

Target irradiations at the HRIBF

Target irradiation setup showing the beam diagnostics and the water-cooled vacuum chamber.

This setup is normally used for testing the release of radioactive ions from a uranium carbide target with an operating temperature of about 2000° C.



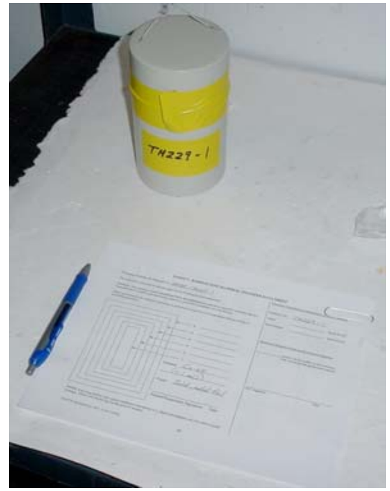


Beam diagnostics
(beam size, position, and current)

Proton beam

Water-cooled vacuum enclosure

Transporting foils from HRIBF to Hot Cells



The radioactive material is contained within the target holder. (So far, no foils have been torn or melted during irradiation.)

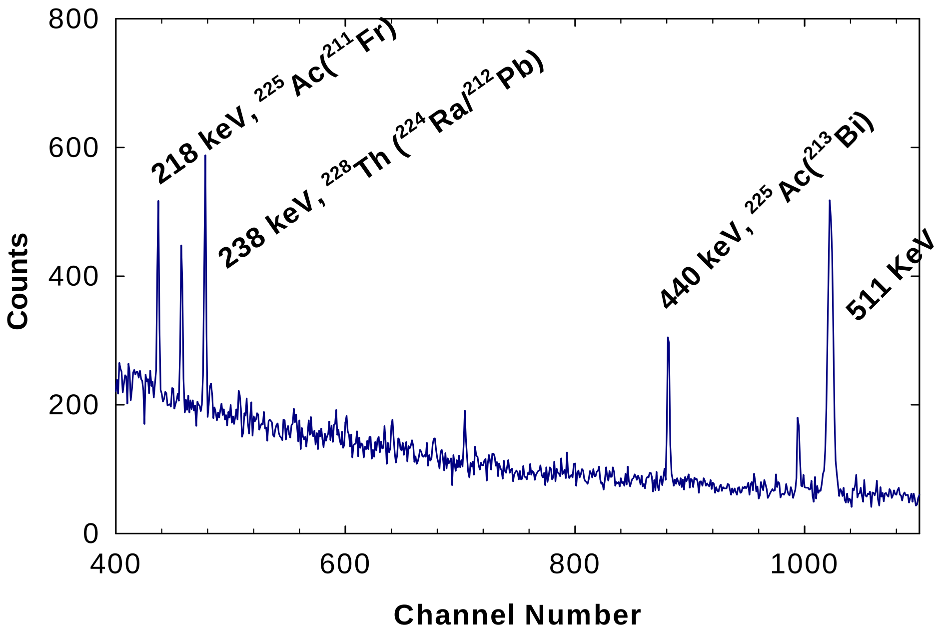
For the thin-target irradiations, the dose rates are low enough for immediate processing (thick targets may need to cool for a few days)

Usually, the target holder is transferred within an hour after the end of irradiation.

Analysis of irradiated foils
(see poster by Justin Griswold)

- Foils are separated and mounted for γ-ray counting
- Radiochemical separations may be needed to reduce the count rate
  - from other nuclei produced in the reaction
  - from long-lived daughter nuclei that increase with time

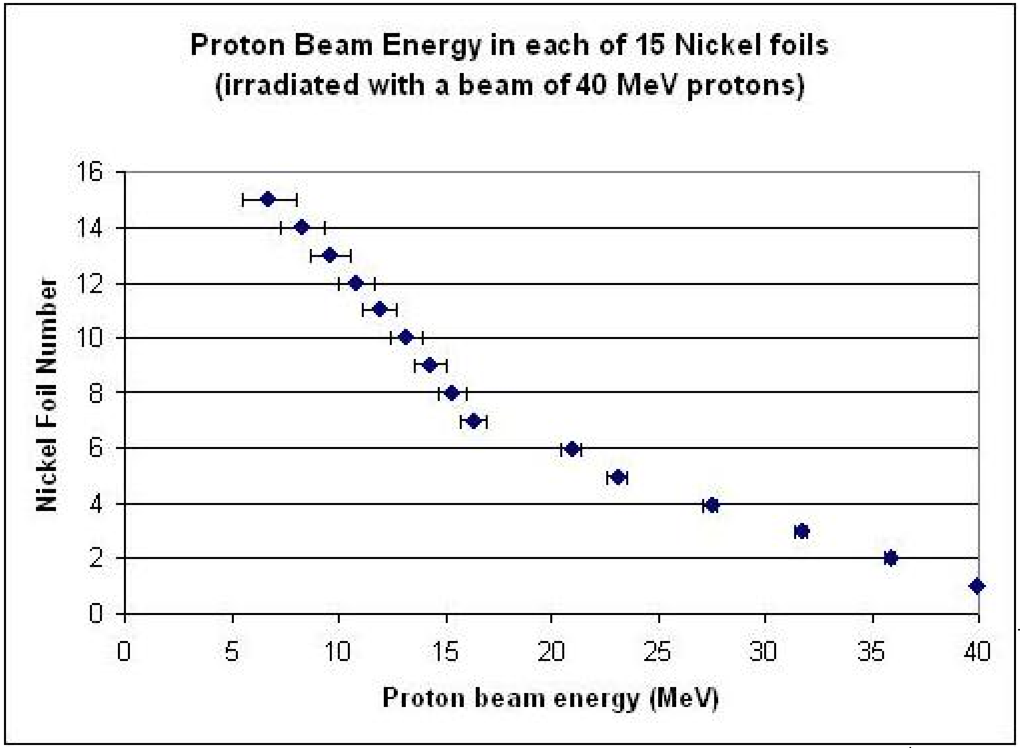
γ-Ray Spectrum of Purified ^{225}Ac from ^{229}Pa α-decay
(Th230-2, PaPPT-T5, MP1 Load, 6/25/2011, T=33.9 h)



List of target irradiations completed

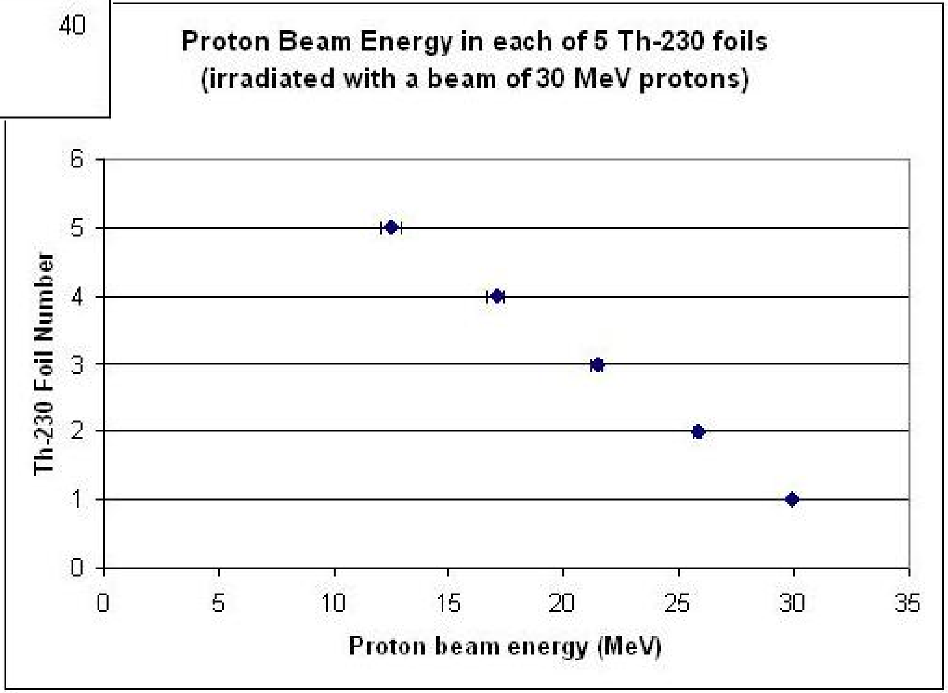
Run ID	Element	# of foils	Total Thickness	Beam Energy	# of hours	# of protons	Nuclei analyzed
			(mg/cm^2)	(MeV)			
Ni-I	nickel	15	33.6	40	11	1.1 x 10^{16}	^{55}Co, ^{56}Ni, ^{57}Ni
Cu-I	copper	15	168	40	13.5	1.3 x 10^{16}	^{61}Cu, ^{62}Zn
Th-232-I	^{232}Th	15	1.602	40	44	4.4 x 10^{16}	^{228}Pa, ^{229}Pa, ^{230}Pa, ^{225}Ac
Cu-II	copper	10	112	35	9	6.8 x 10^{15}	^{61}Cu, ^{62}Zn
Cu-III	Nickel	10	112	26	68	5.4 x 10^{16}	^{61}Cu, ^{62}Zn
Ni-II	nickel	10	22.4	38	11	1.0 x 10^{16}	^{55}Co, ^{57}Ni
Th-230-I	^{230}Th	5	0.361	30	42	4.7 x 10^{16}	^{228}Pa, ^{230}Pa, ^{225}Ac
Th-230-II	^{230}Th	8	0.699	32	37	3.9 x 10^{16}	^{228}Pa, ^{230}Pa, ^{225}Ac
Th-232-II	^{232}Th	8	1096	40			
Th-232-III	^{232}Th	23	3152	40	98	8.5 x 10^{16}	

Energy loss in the Nickel and ^{230}Th foils



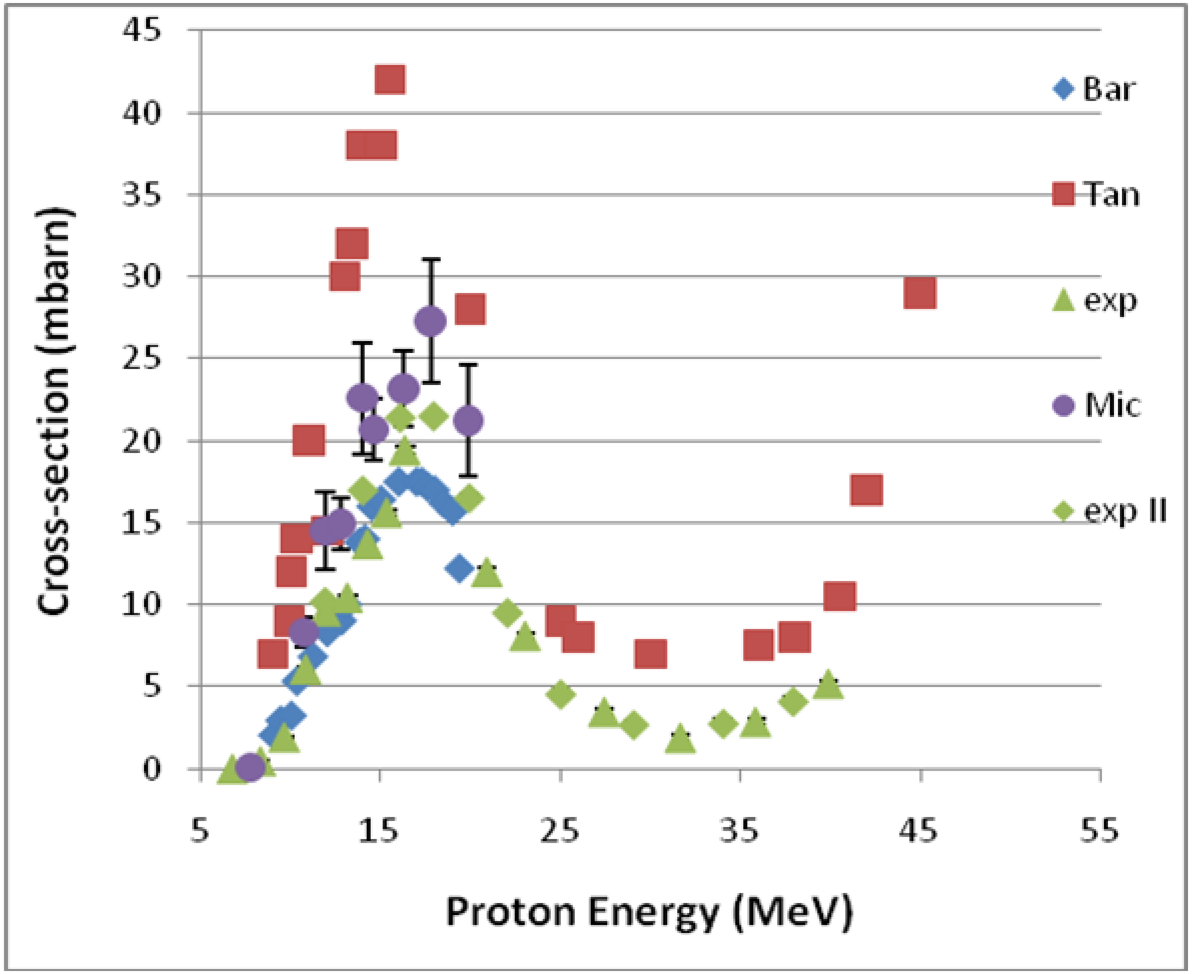
Calculated using SRIM-2008.03
- a program that calculates Stopping and Range of Ions in Matter

The error bars represent the energy spread as the beam passes through the target stack



Calculated cross-sections for ^{55}Co

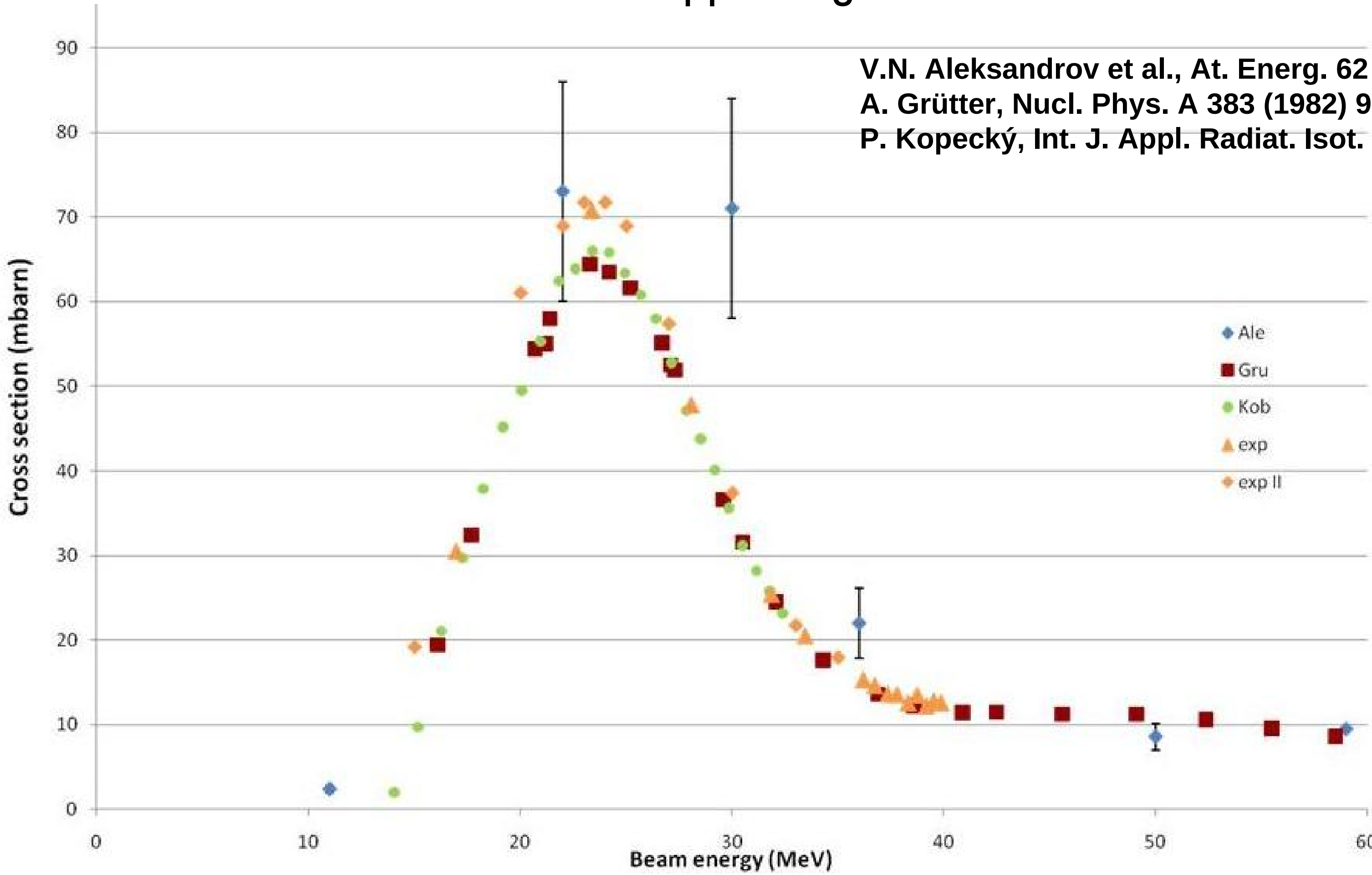
- Produced via the $^{58}Ni(p,\alpha)$ reaction ($T_{1/2}$ = 17.5 hours)
- Includes data from both of the nickel target irradiations
- Good agreement with some of the data sets
- Good reproducibility for data from two irradiations



J.N. Barrandon et al., Nucl. Instr. Meth. 127 (1975) 269.
S. Tanaka et al., J. Inorg. Nucl. Chem. 34 (1972) 2419.
R. Michel et al., Z. Phys. A 286 (1978) 393.

Calculated cross-sections for ^{62}Zn

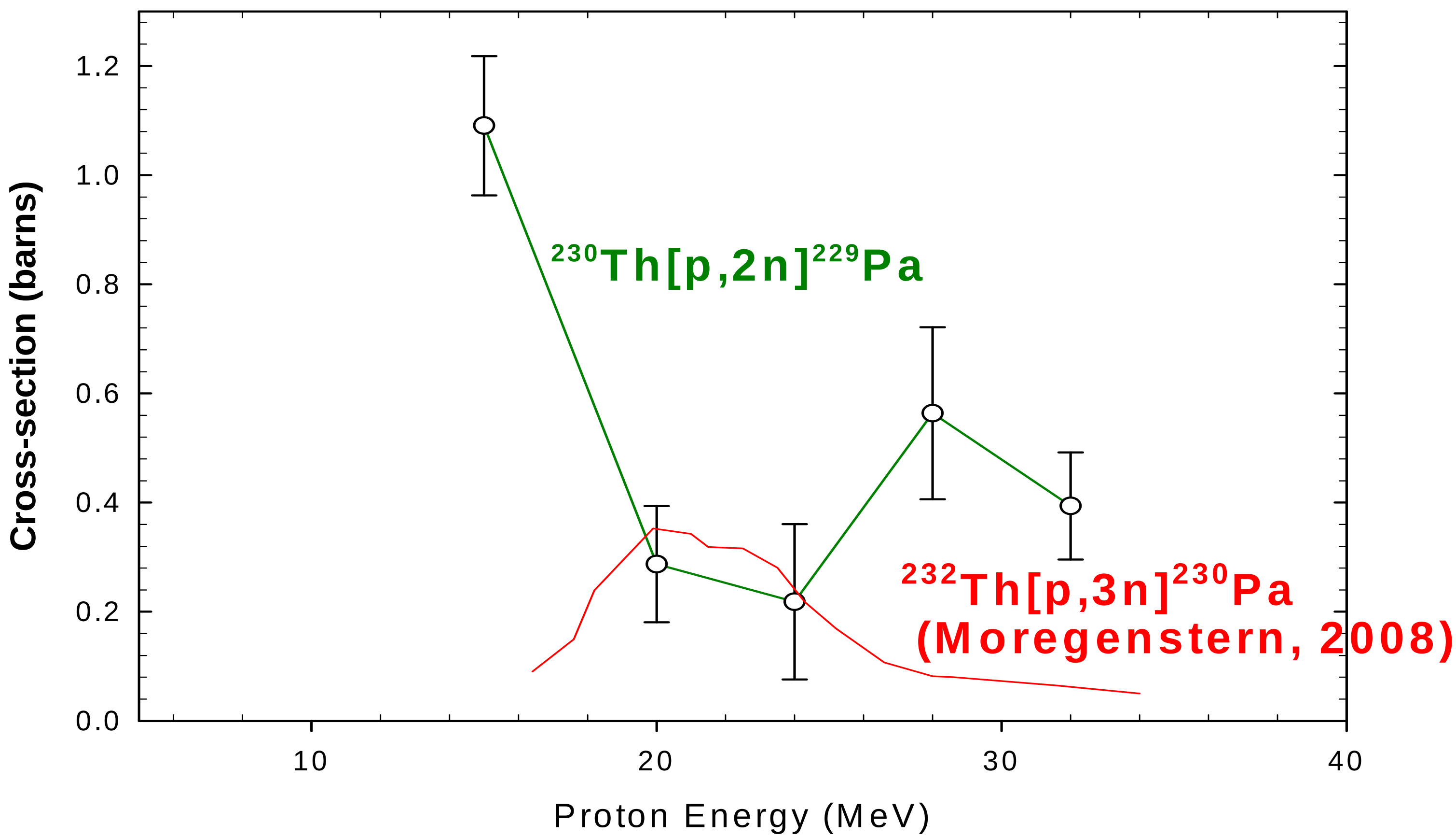
- Produced via the $^{63}Cu(p,2n)$ and $^{65}Cu(p,4n)$ reactions ($T_{½}$ = 9.13 hours)
- Includes data from two of the three copper target irradiations



V.N. Aleksandrov et al., At. Energ. 62 (1987) 411.
A. Grütter, Nucl. Phys. A 383 (1982) 98.
P. Kopecký, Int. J. Appl. Radiat. Isot. 36 (1985) 657.

Preliminary results from thorium irradiations

Excit. Fxn of $^{230}Th[p,2n]^{229}Pa$ ($t_{1/2}$=1.4 d, 0.5% α-decay to ^{225}Ac)



The red curve is data from Karlsruhe by A. Morgenstern, et al., App. Rad. Isotopes **66** (2008) 1275.
The green curve represents preliminary data obtained in this work (to be published).

Tasks still to be completed

- Irradiate the second ^{232}Th thin target (planned for next week)
- Complete γ-ray counting and data analysis of the last two irradiated targets
- Determine feasibility and cost-effectiveness of producing ^{229}Th via proton irradiation of ^{230}Th or ^{232}Th targets

Future R&D plans

- Design a high-power target for the production of ^{229}Th
  - study the effect of high power density by decreasing the beam spot size on target
  - increase beam intensity (up to 1 μA is possible from the Tandem)
  - add cooling, as necessary, to prevent melting of the target foils and high-temperature reactions with the target holder materials
  - measure radiological dose rates as beam power is increased to determine the shielding and remote handling requirements

Summary

- Designed an experiment to investigate an alternative method for producing ^{229}Th (long-lived parent of ^{225}Ac, which is an important radionuclide in the field of radioimmunotherapy)
- Measured cross-sections for isotopes of interest
  - Benchmarked with reactions on copper and nickel nuclei and found good agreement with literature values
  - Measured some reaction cross-sections leading to production of ^{229}Th
- Analysis of recent target irradiations is continuing
  - Complete analysis of data from the thorium targets
  - To determine the thick target yields of ^{228}Th and ^{229}Th
  - To determine the feasibility of this production method

Collaborators

- Radiochemistry
  - Stephanie Bruffey, Research Staff
  - David Denton, Research Staff
  - Justin Griswold, Student, Nuclear Engineering, UT
  - Saed Mirzadeh, Research Staff (co-PI)
- Target Irradiations
  - Cara Jost, post-doc (earned PhD in Nuclear Chemistry in 2010)
  - Dan Stracener, Research Staff (co-PI)