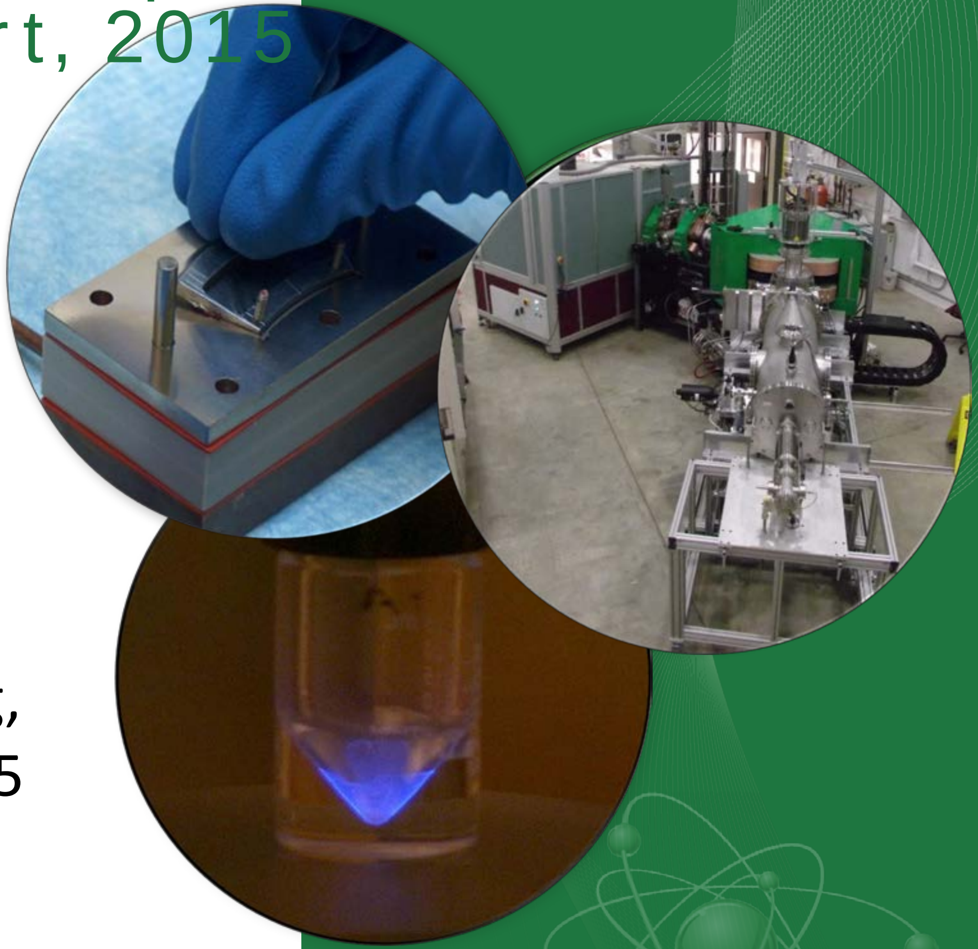
Summary of NSAC-Isotope Subcommittee Report, 2015

Meeting Isotope Needs and Capturing Opportunities for the Future: The 2015 Long Range Plan for the DOE-NP Isotope Program



Saed Mirzadeh

Low Energy Community Meeting, East Lansing, MI, August 21, 2015

ORNL is managed by UT-Battelle for the US Department of Energy

NSAC-I Subcommittee

Kelly Beierschmitt, Idaho National Laboratory	J. David Robertson, University of Missouri
Roy Brown, MBA Mallinckrodt Pharmaceuticals	Thomas Ruth, TRIUMF
Carol Burns, Los Alamos National Laboratory	David Scheinberg, Memorial Sloan Kettering Cancer Center
Lawrence Cardman, (chair) Thomas Jefferson National Accelerator Facility	Sally Schwarz, Washington University, St. Louis
Donald Geesaman, (ex officio) Argonne National Laboratory	Bradley Sherrill, Michigan State University
Suzanne Lapi, Washington University, St. Louis	Mark Stoyer, Lawrence Livermore National Laboratory
Saed Mirzadeh, Oak Ridge National Laboratory	Scott Wilbur, University of Washington
Eugene Peterson, Los Alamos National Laboratory	Frank Yeager, Eckert and Ziegler Isotope Products
Lee Riedinger, University of Tennessee	

The mission of the DOE Isotope Program:

- Produce and/or distribute radioactive and stable isotopes that are in short supply, associated byproducts, surplus materials, and related isotope services.

- Maintain the infrastructure required to produce and supply isotope products and related services.

- Conduct R&D on new and improved isotope production and processing techniques that can make available new isotopes for research and applications.

Background

The Isotope Program is a relatively small federal program:

FY15 federal appropriation: $19.84 M
FY15 anticipated isotope sales of ~$36 M

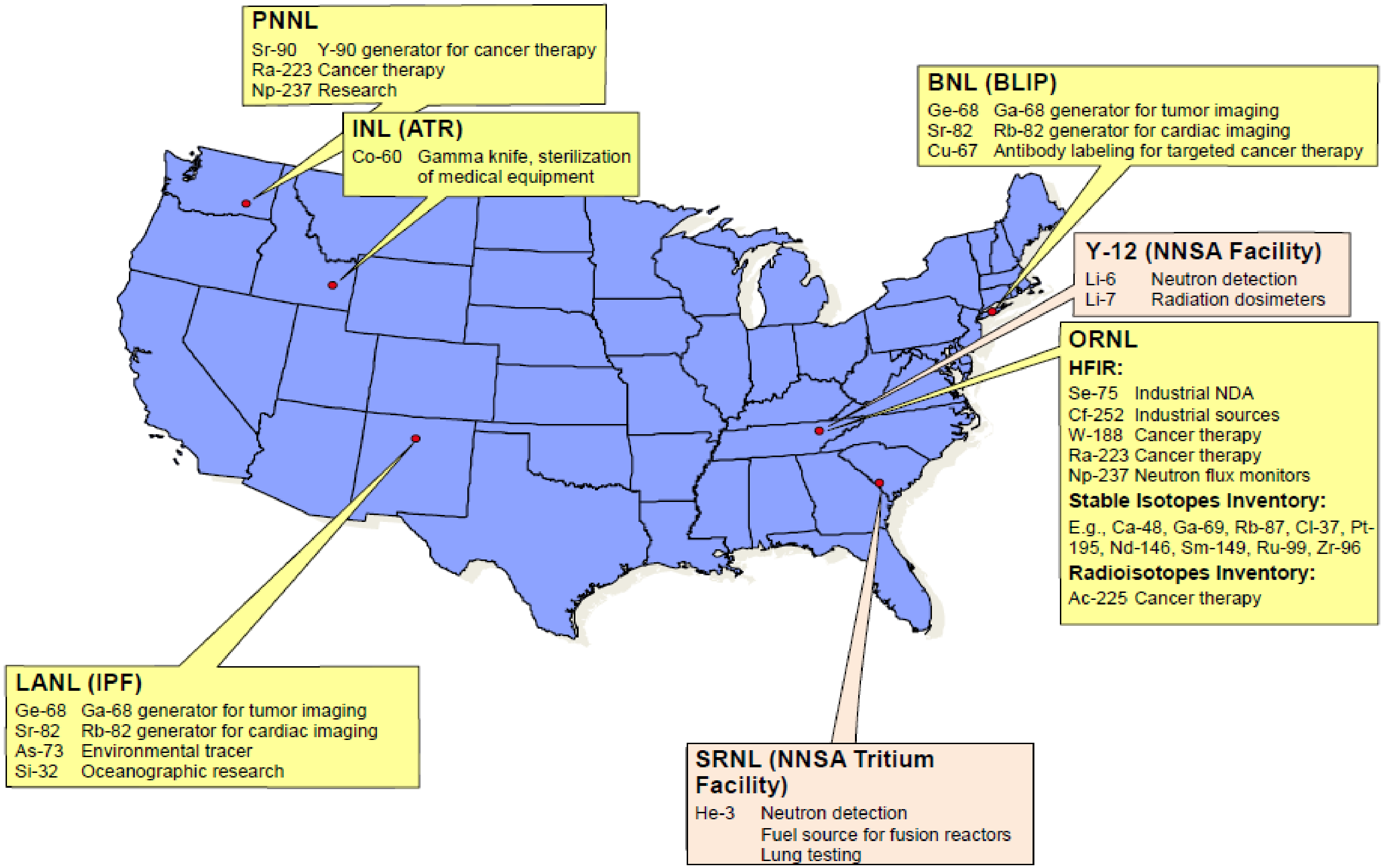
This small programs, however, enables and is immersed in billion-dollar enterprises, including medical diagnosis and treatment, research, national security, and critical industries. These applications touch the lives of almost every citizen.

NSAC-I 2015 Report

Contents

Application	Selected Isotope Examples
Diagnostic imaging of human disease	*Gamma imaging:* ^{201}Tl, ^{99m}Tc, ^{111}In, ^{131}I, ^{133}Xe
	Positron (PET) imaging: ^{18}F, ^{124}I, ^{68}Ga, ^{89}Zr, ^{64}Cu, ^{86}Y, ^{11}C, ^{15}0, ^{82}Sr/^{82}Rb
Therapy of human disease	*Beta-emitters:* ^{131}I, ^{90}Y, ^{89}Sr, ^{177}Lu, $^{186/188}$Re, ^{153}Sm
	Alpha-emitters: ^{223}Ra, ^{225}Ac, ^{211}At, ^{213}Bi, ^{212}Pb/^{212}Bi
Fixed therapeutic sources	*External beams:* ^{60}Co
	Internal brachytherapy: ^{192}Ir, ^{125}I
Biological, biochemical, and chemical research tracers	^{3}H, ^{32}P, ^{33}P, ^{125}I, ^{35}S, ^{51}Cr, ^{75}Se, ^{14}C
Power sources	^{244}Cm, ^{238}Pu, ^{210}Po, ^{90}Sr,
Nuclear and particle physics, chemistry and engineering	**^{237}Np, ^{239}Pu, ^{244}Pu, ^{243}Am, ^{248}Cm, ^{249}Bk, ^{249}Cf, ^{225}Ra, ^{48}Ca**
Fission and reactor function	^{233}U, ^{235}U, ^{238}Pu
Environment, security and safety	*Sensors, tracers and detectors:* ^{241}Am, ^{137}Cs, ^{252}Cf, ^{6}Li, ^{14}C, ^{3}He, ^{75}Se, ^{133}Cs, ^{65}Ni
	Coolants: ^{7}Li, ^{10}B
Stable Isotopes	^{13}C, ^{15}N, ^{18}O, Many

The Network of DOE Isotope Production Sites and examples of isotopes produced or distributed from each site.



Recommendations

- **significant increase of funding for Research and Development**

  a) ***Continue support for R&D on the production of alpha-emitting radioisotopes.***

  b) ***Support R&D into the production of high specific activity theranostic radioisotopes:*** ^{99m}Tc/^{188}Re, ^{111}In/^{90}Y.

  c) ***Support for R&D on the use of electron accelerators for isotope production:***
  ^{68}Zn(γ,p)^{67}Cu,
  ^{232}Th(γ,spall)^{225}Ac
  ^{232}Th(γ,spall)^{211}Rn($t_{1/2}$=14.6 h, *EC*)^{211}At.

  a) ***Support R&D on the development of irradiation materials for targets that will be exposed to extreme environments to take full advantage of the current suite of accelerator and reactor irradiation facilities***

  b) **Completion and the establishment of effective, full intensity operations of the stable isotope separation capability at ORNL**

Recommendations (cont.)

- **Increase in the annual appropriated budget for high-impact infrastructure investments and to maintain a stable funding base for reliably operating and continually improving facilities.**

    a) ***Infrastructure for isotope harvesting at FRIB***

    b) ***Develop a strategy for the re-establishment of a separator for radioactive isotopes to support research***

    c) ***Increase the base infrastructure budget to sustain and expand production capacity at the Isotope Program facilities at BNL-BLIP and LANL-IPF***

- **Continuation and expansion of the effort to integrate the university facilities with the Isotope Program**

Applications and Uses of Isotopes in Nuclear Physics

Investigation of the structure and reactions of atomic nuclei

- The Argonne Tandem Linac Accelerator System (ATLAS) and Californium Rare Ion Breeder Upgrade (CARIBU); requiring **1 Ci ^{252}Cf source 1½ - 5 years**

- National Superconducting Cyclotron Laboratory (NSCL); **^{48}Ca, ^{86}Kr, ^{82}Se**, and others

- Rare Isotope Beams (FRIB); new opportunity to access a broad range of radioactive isotopes through isotope harvesting. Example include:

  a) Harvesting ^{48}V or ^{147}Eu-^{154}Eu, and potential to provide increased quantities of ^{223}Ra and ^{225}Ra, and search for more sensitive candidates, such as ^{229}Pa.

  b) Providing beams of ^{16}C, ^{17}N, ^{20}O for synthesizing very long-lived isotopes of rutherfordium, dubnium, and seaborgium with projected $t_{1/2}$> 1 y

  c) Measurement of cross-section of very short-lived radionuclides via inverse kinematics reactions

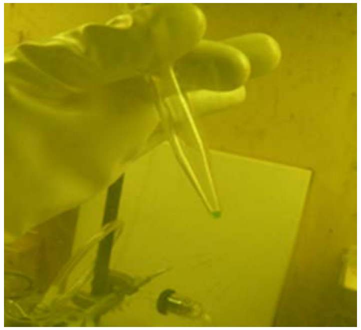
Nuclear Physics (cont.)

Permanent electric dipole moment (EDM) of a quantum system
Requiring multi mCi of ^{225}Ra and ^{223}Ra as potential high-sensitivity deformed nuclei

Neutrinoless Double Beta Decay:

$^{76}Ge \rightarrow ^{76}Se$ $^{136}Xe \rightarrow ^{136}Ba$, $^{130}Te \rightarrow ^{130}Xe$, $^{48}Ca \rightarrow ^{48}Ti$. Requiring a ton of active material

Nuclear Physics (cont.)



22 mg of chemically purified ^{249}Bk prior to shipment to Dmitrovgrad.

Synthesize of Heaviest Elements using Actinide Targets

Six heaviest elements, A=113-118, was discovered using actinide targets of ^{237}Np, ^{239,240,242,244}Pu, ^{243}Am, ^{248}Cm, ^{249}Bk and ^{249}Cf target mainly produced in the U.S.

In 2009, Element 117 was synthesized via ^{48}Ca + ^{249}Bk reaction by a 150-day irradiation of 22 mg of ^{249}Bk in Dubna at the U-400 cyclotron with ^{48}Ca beam

Future targets are envisioned to require > 100 mg.

Planetary science

Use of ^{238}U, ^{234}U, ^{230}Th, ^{232}Th and ^{226}Ra as geochronologic tools for study of magmatic differentiation or deposition of carbonate rocks; requiring very high purity ^{233}U and ^{229}Th (very low contamination from both ^{230}Th and ^{232}Th) as calibration standards of mass spectrometer

Solid-state physics

Avogadro's project – on going project to develop a mass standard based on fundamental units; measuring the uncertainty of Avogadro constant to 0.01 ppm. Project requiring kilograms of isotopically pure ^{28}Si .

Radioisotopes for calibration of neutrino detectors

^{51}Cr, ^{144}Ce, freshly irradiated Cm targets, fresh HFIR spent fuel, etc.

For example, preliminary calculations indicate that six fresh Cm targets arranged in a flux-trap configuration could provide the highest flux of man-made neutrinos.

In the later three examples, the key challenge is that the magnitude and size of required radioisotopes require inter-agency agreements, and substantial funds.

Nuclear Data for Weapons Physics Research

With the cessation of nuclear testing in 1992, it has become necessary to develop sophisticated simulations of weapons performance that incorporate many different simultaneous nuclear reactions taking place in the high neutron flux density environment.

Simulations must be empirically validated by comparing the historic data from U.S. weapons tests against predicted values. This generates need for accurate nuclear data.

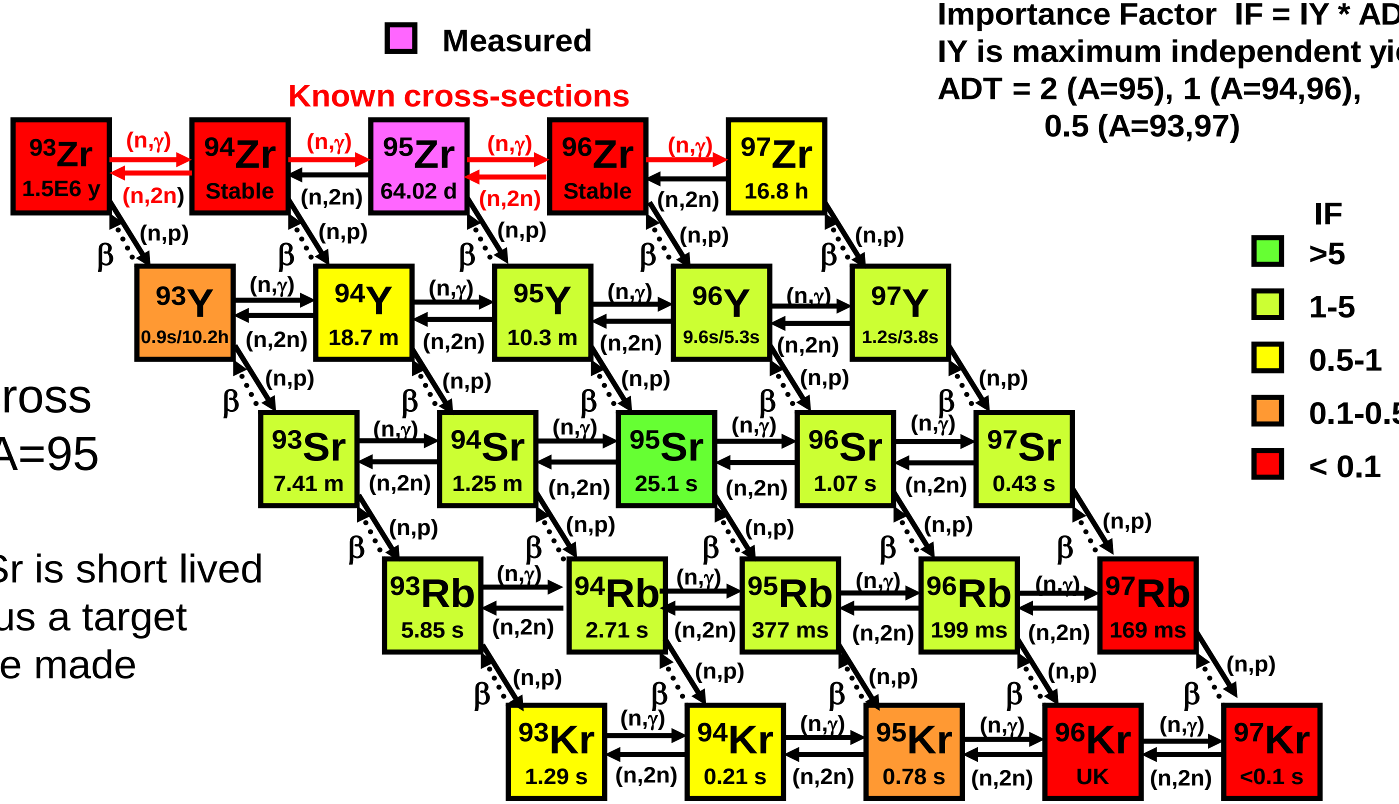
Principal reactions are: (n,γ),(n,f), (n,p), (n,α), $(n,2n)$

While cross-sections are well known for stable isotopes, **there is lack of data for short-lived radionuclides, and for reaction leading to metastable radionuclides**;

$$\sigma({}^{A}Z[n,n']^{Am}Z) >> \sigma({}^{(A-1)}Z[n,\gamma]^{Am}Z)$$

Importance Factor IF = IY * ADT
IY is maximum independent yield
ADT = 2 (A=95), 1 (A=94,96),
0.5 (A=93,97)

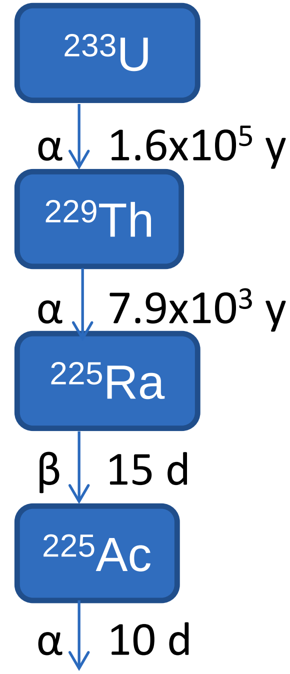
Key reaction cross sections near A=95



But ^{95}Sr is short lived and thus a target can't be made

- National Criticality Experiments Research Center (NCERC) located at the Nevada National Security Site provides access to relevant fission-spectrum irradiation capabilities.
- For very short-lived species, FRIB will provide the opportunity to examine in-beam reactions

Nuclear Data for Production of Actinium-225

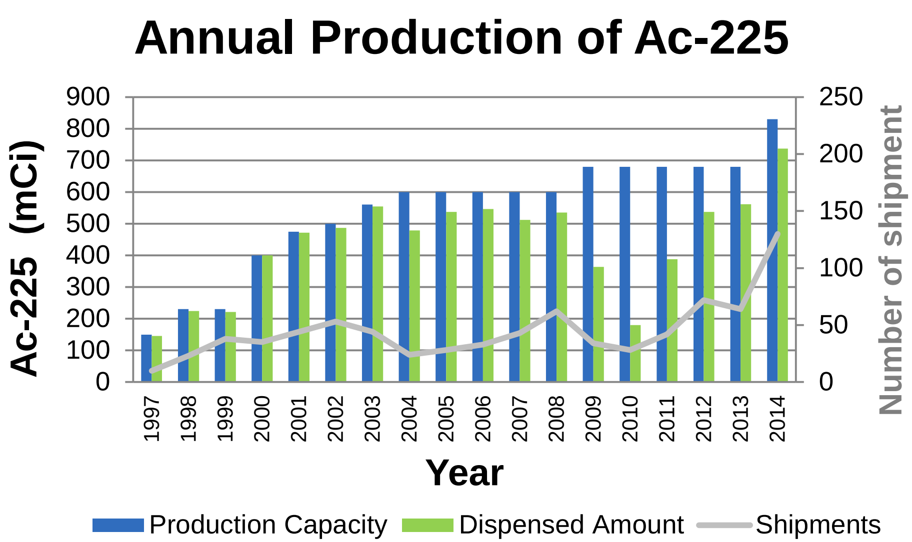


Background of Ac-225 production

- ORNL has been the main supplier of ^{225}Ac (via decay of existing ^{229}Th stock) since 1997,
- 700-900 mCi of ^{225}Ac is harvested annually from 130–mCi ^{229}Th stock at ORNL.
- 6-12 campaigns are performed per year, and campaign **120** is currently underway

Rationale for R&D related to production of Ac-225

- The present supply of ^{225}Ac is insufficient for current medical and research demands of ~6 Ci/year.



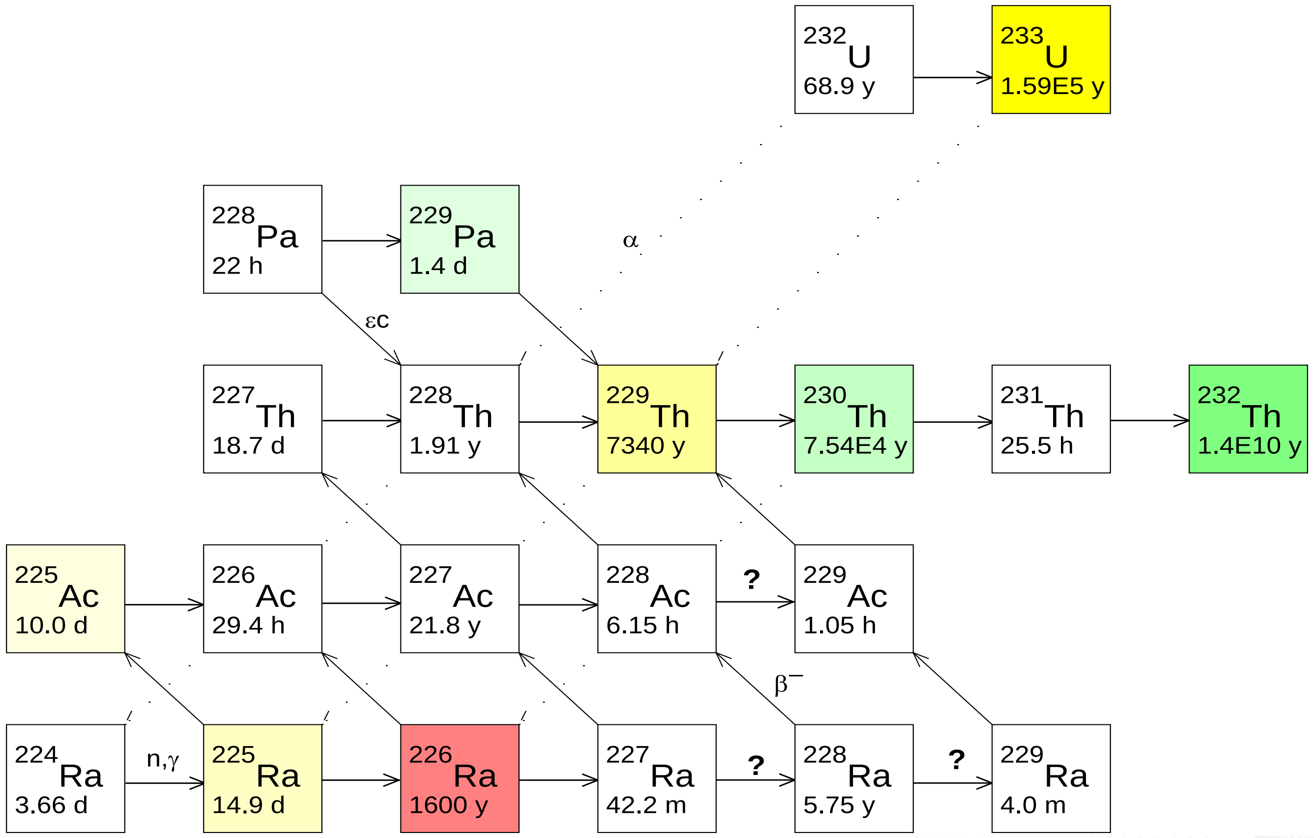
New Initiatives to Enhance Production of Actinium-225 for Cancer Treatment

The initiatives are a direct response to the NSAC-I recent citation on the gap between production and demand of this medically useful isotope

Projects	Time frame	Comments
Direct production of Ac-225 in a proton accelerator	2012-Cont.	Target irradiation at BNL and LANL, processing at ORNL
Nuclear Data for Reactor Production of ^{229}Th and ^{227}Ac at ORNL-HFIR	2012-2013	NP R&D Grant
Nuclear Data for Production of ^{229}Th via low energy protons at ORNL Tandem	2010-2011	NP R&D Grant

Direct production of ^{225}Ac in a proton accelerator

The new collaboration between ORNL, BNL and LANL aims at developing a plan for full-scale production and stable supply of ^{225}Ac by irradiating ^{232}Th targets in the BNL BLIP and LANL IPF

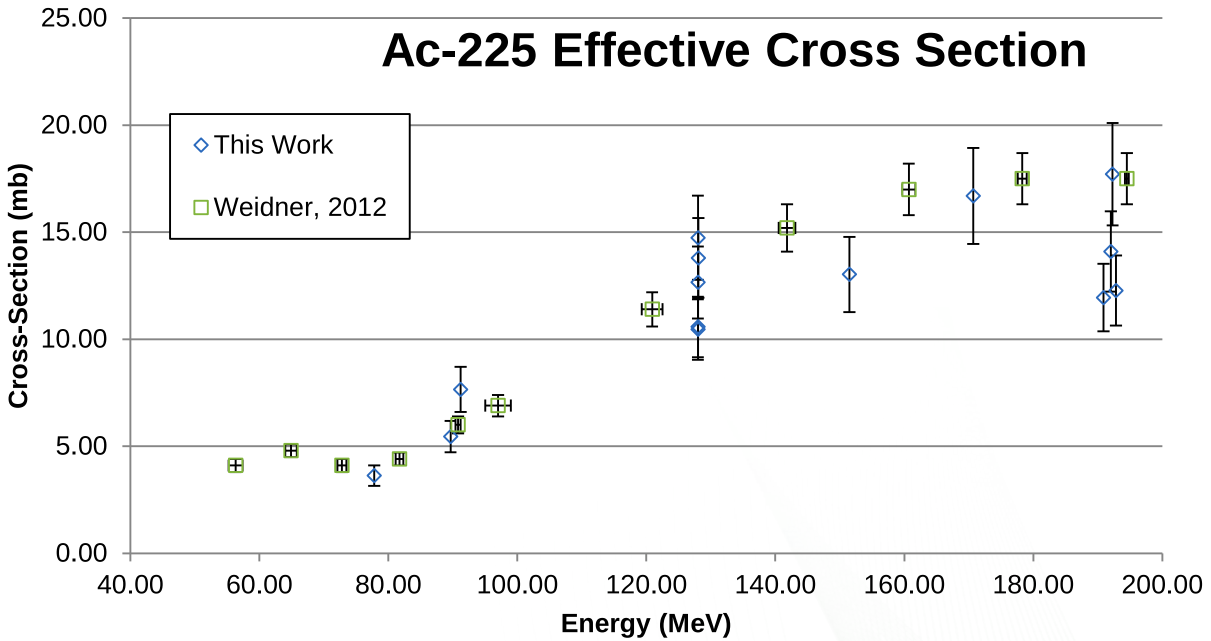


Direct production of ^{225}Ac in a proton accelerator (cont.)

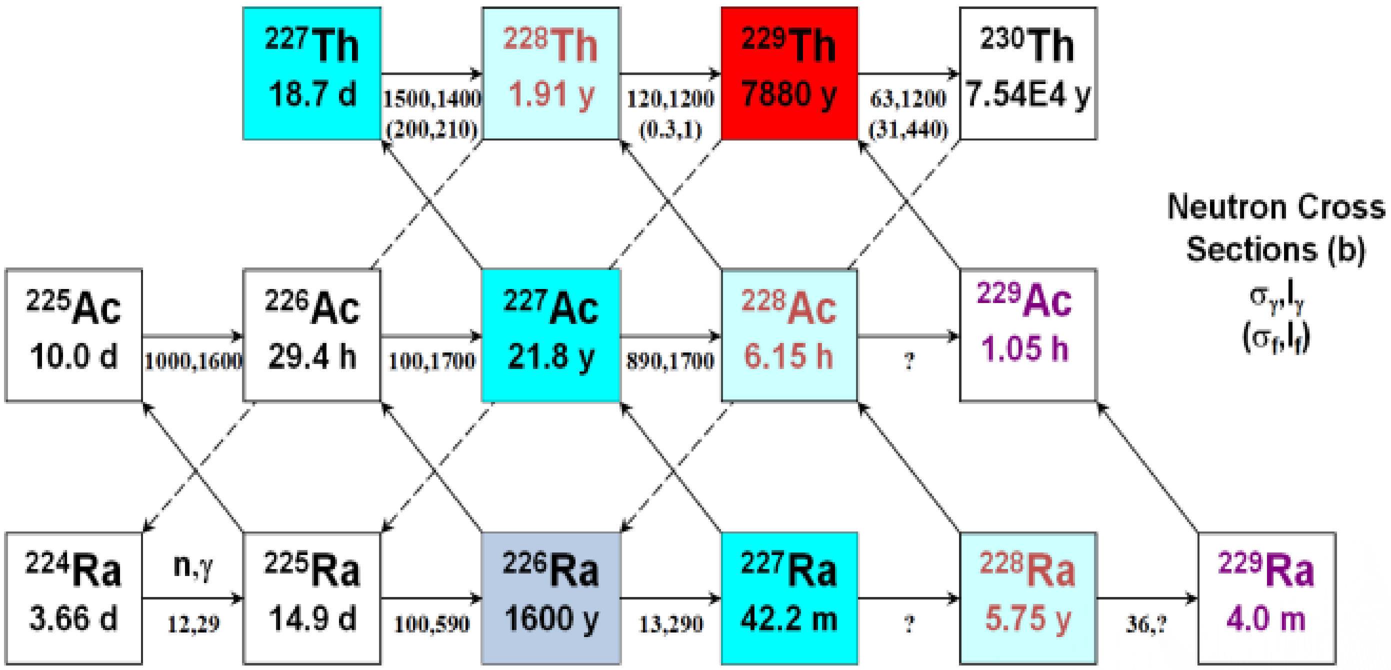
- BNL targets: 1-3 foils ~850 mg each. LANL : one 10-g target
- Irradiations: BLIP: 45-85 µA at 128-193 MeV, IPF: 200 µA of 80-95 MeV

Issues:

- Transportation: Rad Dose & Activity Limits
- Radiological Limits of Processing Facility
- Very Complex Chemistry
- Rather short half life of ^{225}Ac, and ~0.2% contamination with ^{227}Ac at EOB

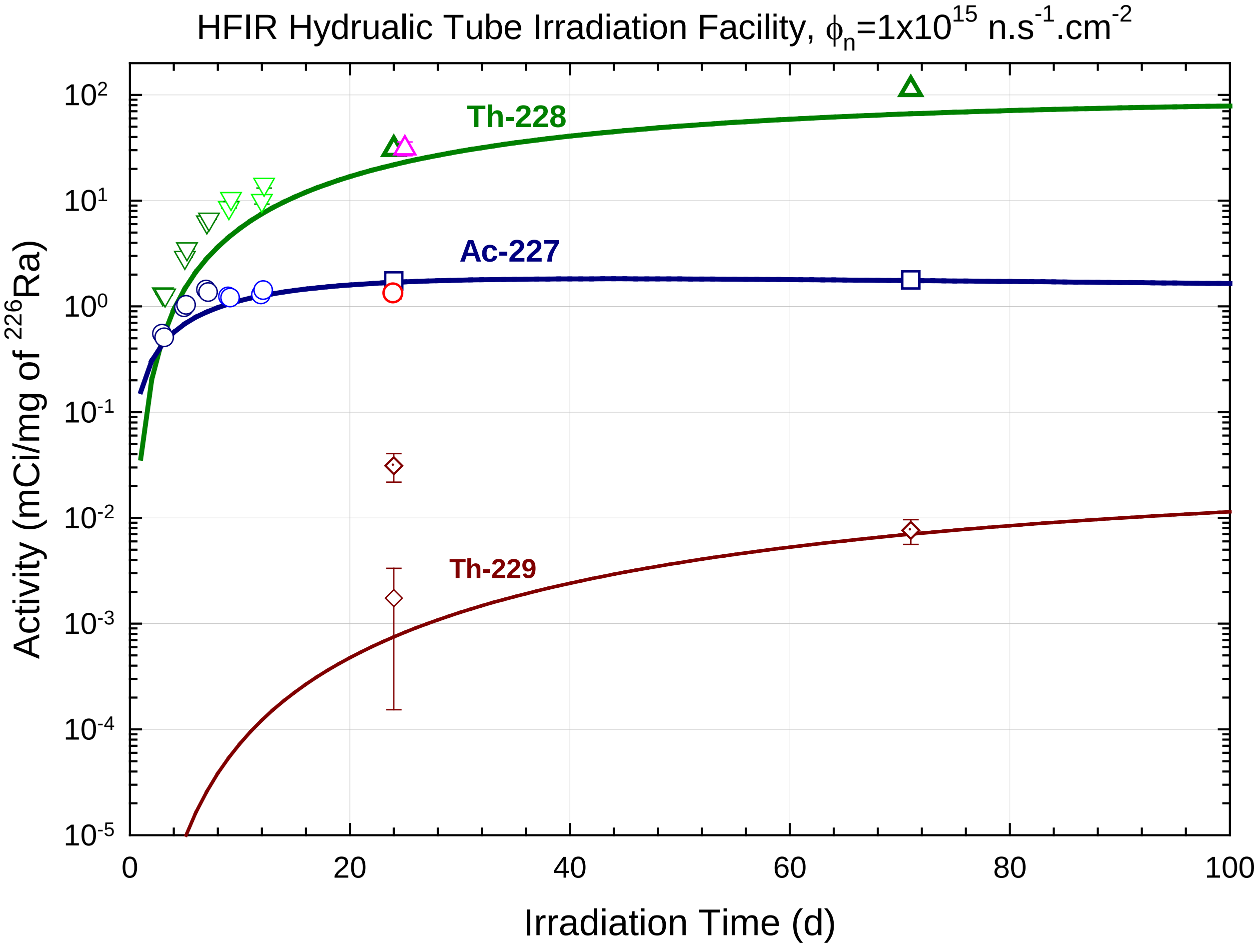


Nuclear Data for Reactor production of Th-229



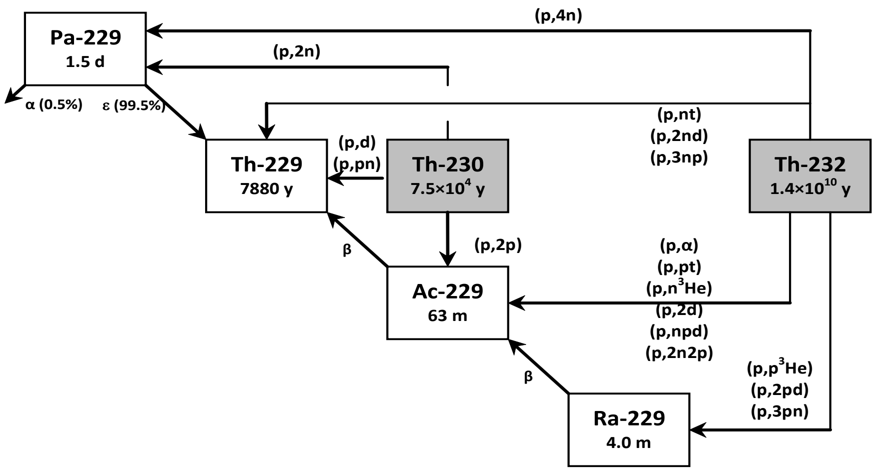
$\sigma = 457 \pm 195$ b

Reactor Production of ^{227}Ac, ^{228}Th and ^{229}Th from ^{226}Ra Target



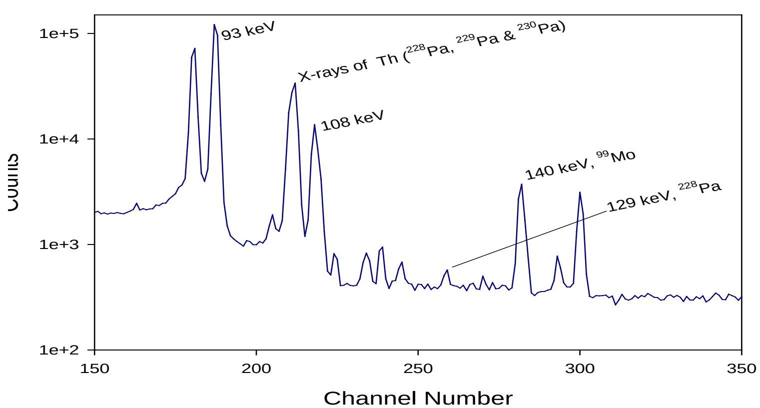
- Projected ^{229}Th yield for 6 cycle irradiations: 18-23 mCi per g of ^{226}Ra, with ^{228}Th and ^{227}Ac contaminations of ~3000 and 50 times larger.
- 20 mCi of ^{229}Th will generate ~140 mCi of ^{225}Ac per year

Nuclear Data for Production of ^{229}Th via Proton-induced Reactions on ^{232}Th

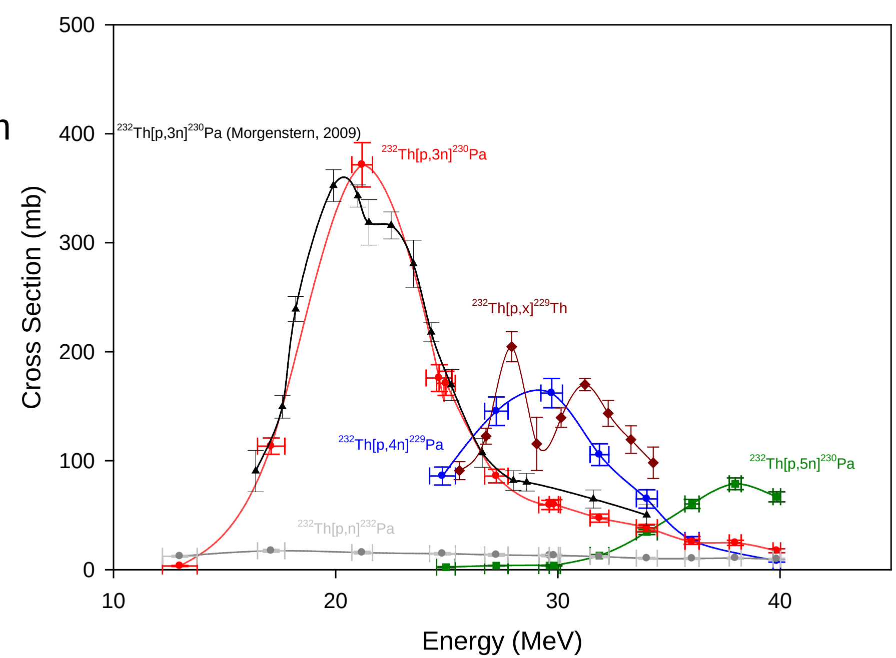


Thorium Proton Bombardment Reaction Block Diagram

γ-Ray Spectrum of Purified Pa Fraction
(Th230-2, PaPPT-T5, 6/15/2011)



Various Excitation Functions for Proton Bombardment of ^{232}Th



ORNL-DWG 95M-6490