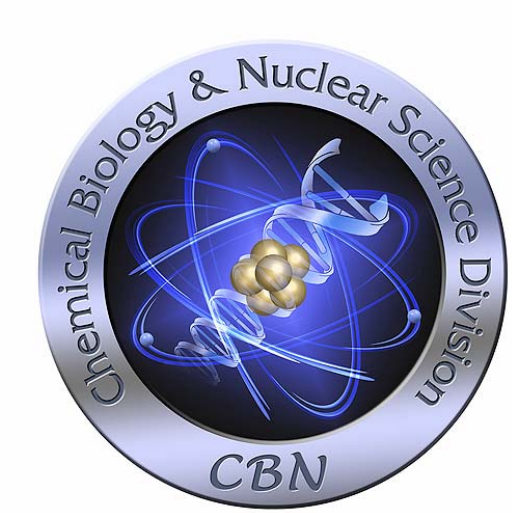


Harvesting of Isotopes at High-Powered Accelerator Facilities

2nd High Power Targetry Workshop
Oak Ridge National Laboratory
October 10-14, 2005

Mark A. Stoyer

For Larry Ahle

Physics & Advanced Technologies
LN Division

Outline

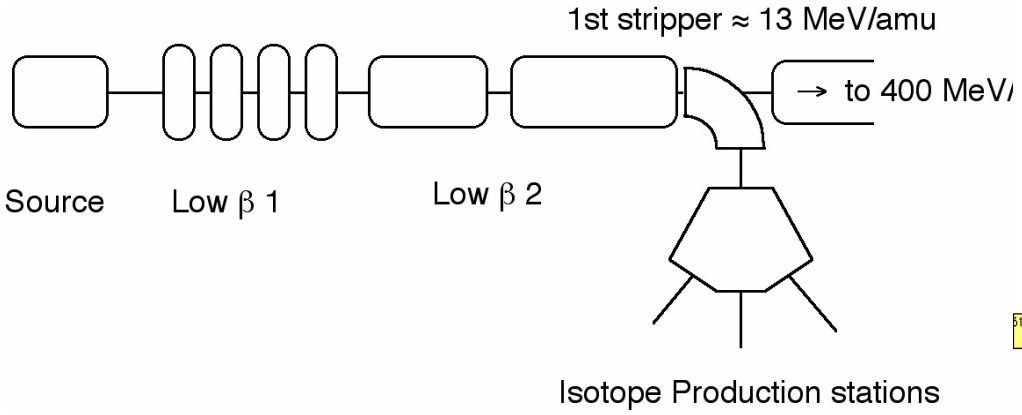
- Why are we interested in Isotope Harvesting?
  - —Medical isotopes
  - —SSP Radiochemistry
- How might it be accomplished?
  - —ISOL
  - —PF
  - —Neutron source
  - —Radiochemistry
- Novel target materials
- Conclusions

Medical Isotope Summary

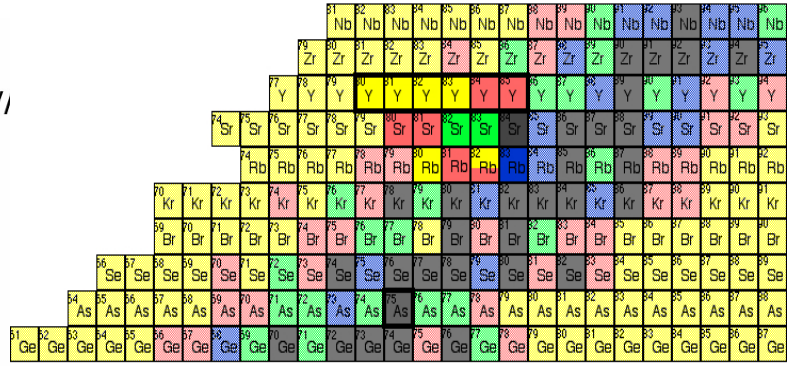
- **RIA will enable exploration into new production methods for research quantities of medical isotopes.**
- **Light ion reactions at ~10 MeV/A offer new approach beyond current medical cyclotrons.**
- **Target flexibility and kinematics effects ease post handling requirements.**

IBA's Cyclone 30

High-current H^- cyclotron for isotope production

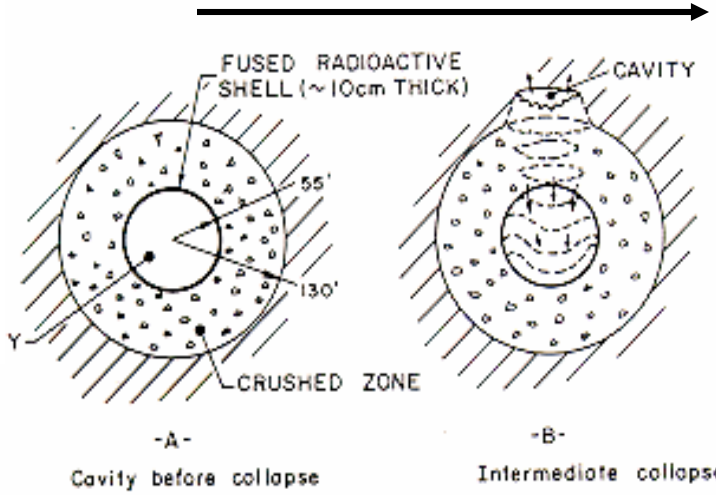


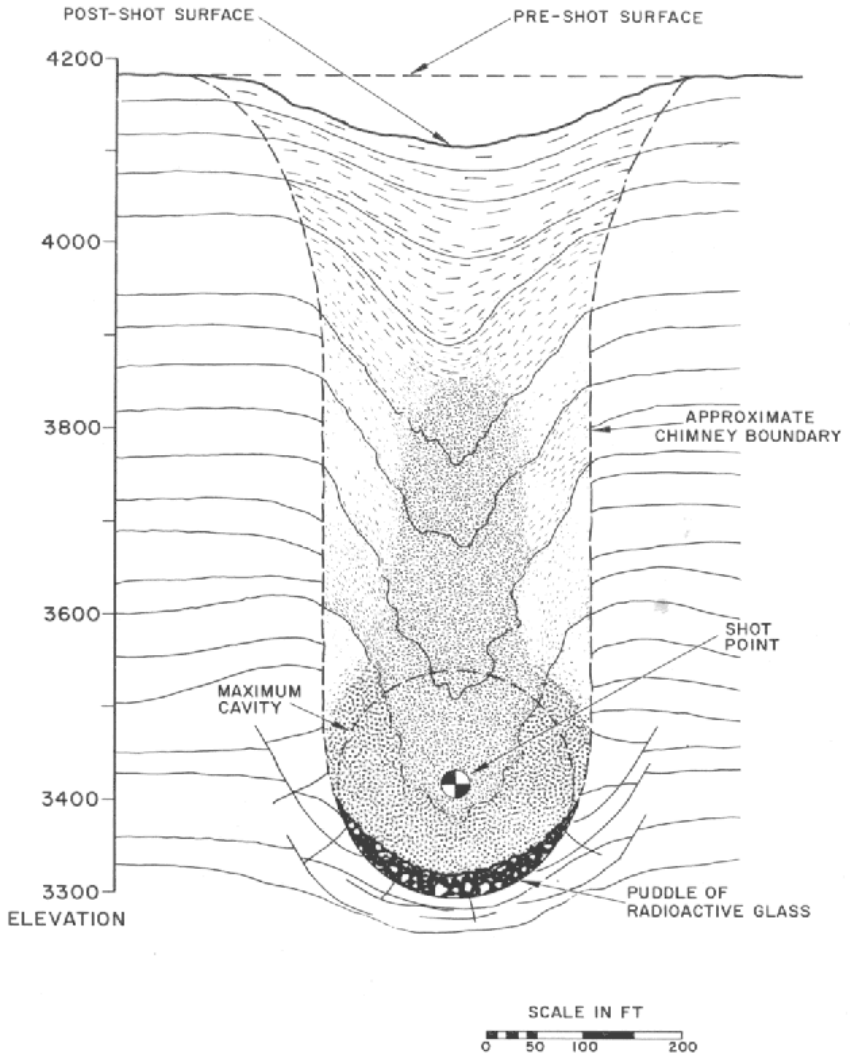
Isotope Production stations



After the Dominic series of tests, ending in 1962, nuclear testing moved underground

Now, we not only had to worry about representative samples, but how to retrieve them from depths of as much as a mile

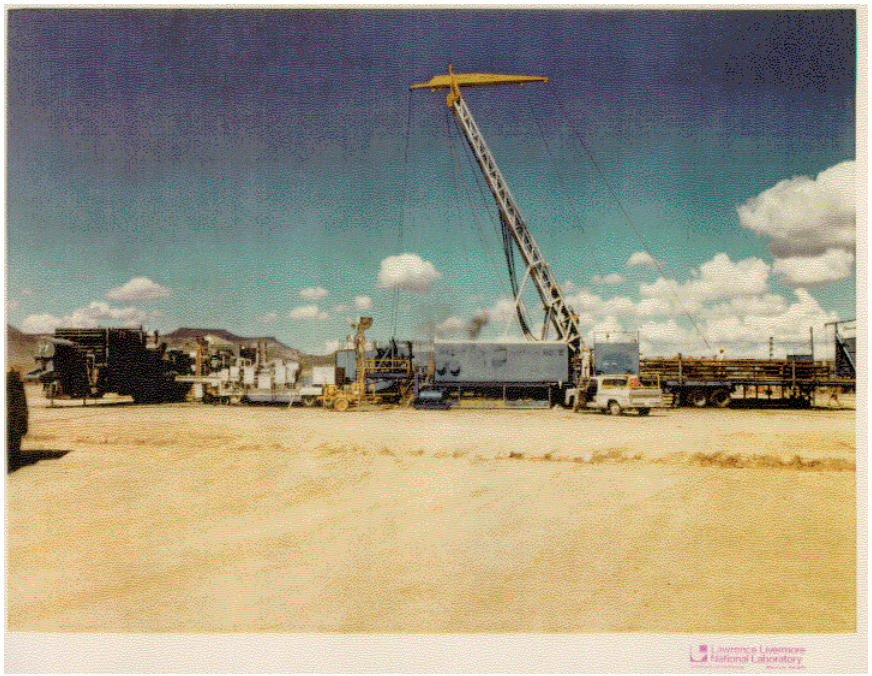




Cores for radiochemical analysis are retrieved by slant drilling

After subsidence is complete, the rig is moved in and set up

Finding the debris puddle is more of an art than a science

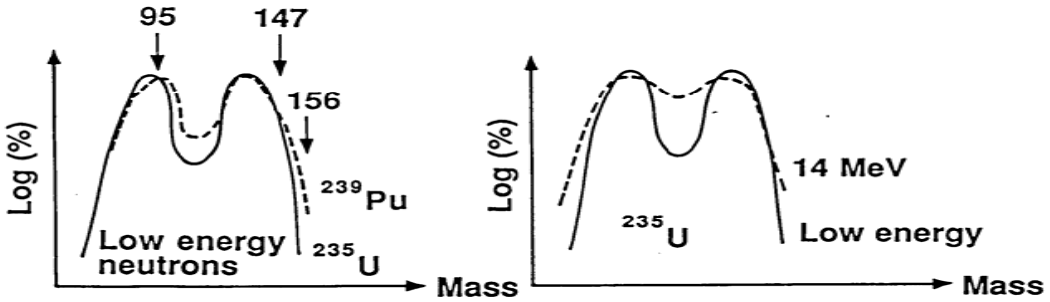


Sample recovery costs more than sample analysis in most cases

In principle, it is possible to obtain the total fission yield of the device from a single cumulative fission yield and a bomb fraction

In practice, this is complicated by the existence of several nuclear fuels in the same device and the resulting "fission split"

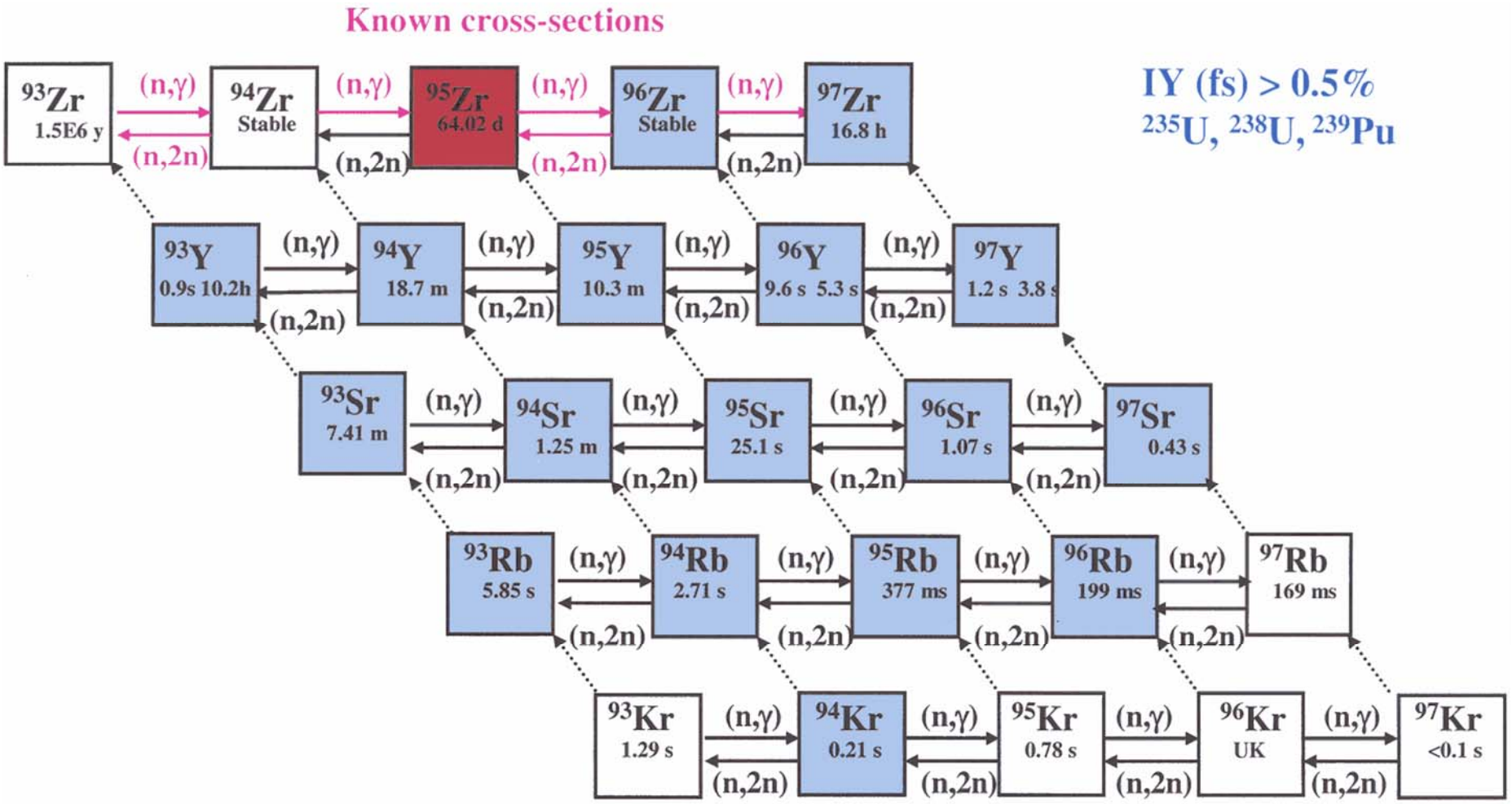
The "double-humped fission distribution" is slightly different for each fuel and is a function of neutron energy



We break the fission yield into those portions resulting from the three main nuclear fuels (^{235}U, ^{238}U, and Pu) and two energy groups (the fission-spectrum low-energy group and the 14-MeV high-energy group)

The six-group split is usually taken from model calculations

The fission product we measure days later is not what was initially present



Just as the fission yield can be determined from the fission products, the fusion yield can be determined from the products of fusion (mainly ^{4}He)

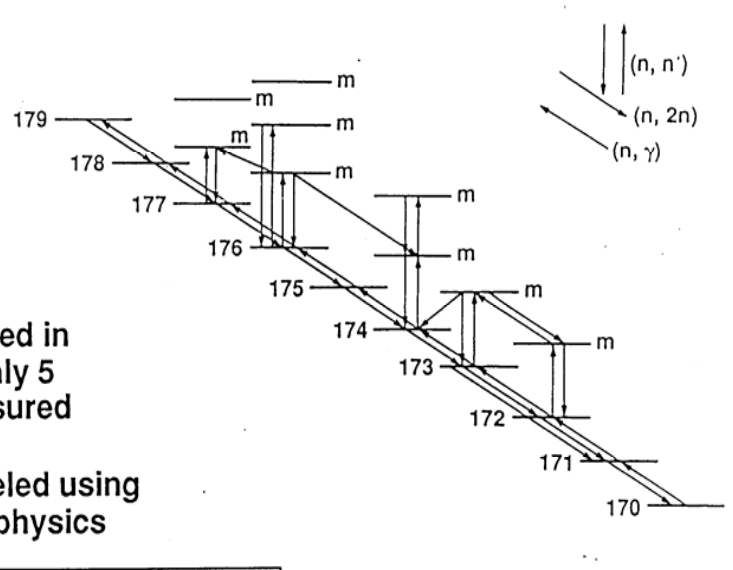
However, it is often advantageous to infer the fusion yield through the activation of radiochemical detectors loaded in thermonuclear fuel

Conversion of detector activations to thermonuclear yield is accomplished through explosion code calculations and a good set of reaction cross sections

Unfortunately, many of the relevant reactions involve transient radioactive nuclides as targets, produced in the high fluences present in the explosion

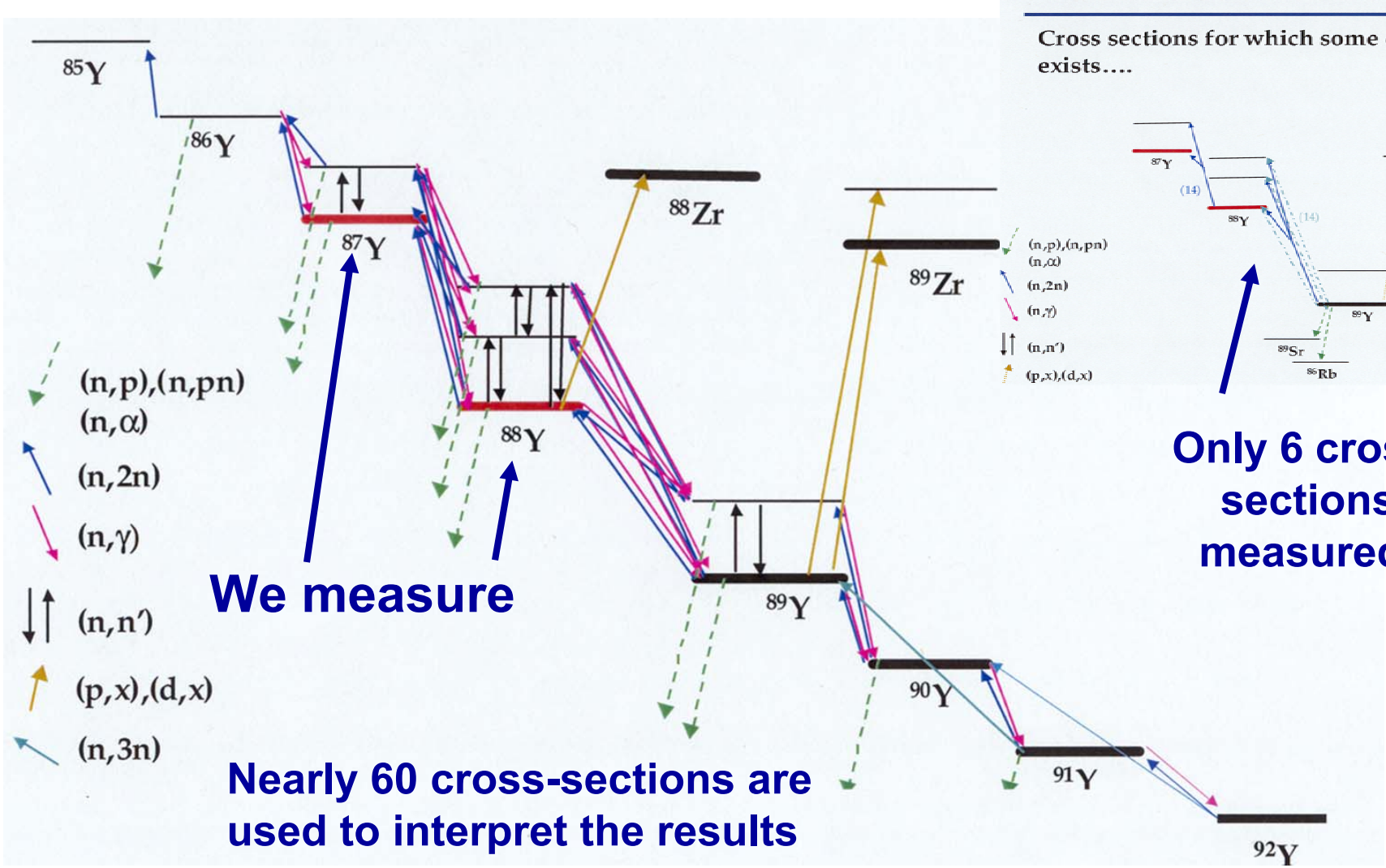
Most of these cross sections are calculated rather than measured

- 25 nuclear states are used for Lu
- (n, 2n), (n, 3n), (n, n′), and (n, γ) reactions connect the states
- 192 reactions are included in analyzing the data — only 5 cross sections are measured
- The remainder are modeled using state-of-the-art nuclear physics



We have a reliable, well-tested set of cross sections for Lu which are used to extract mix information

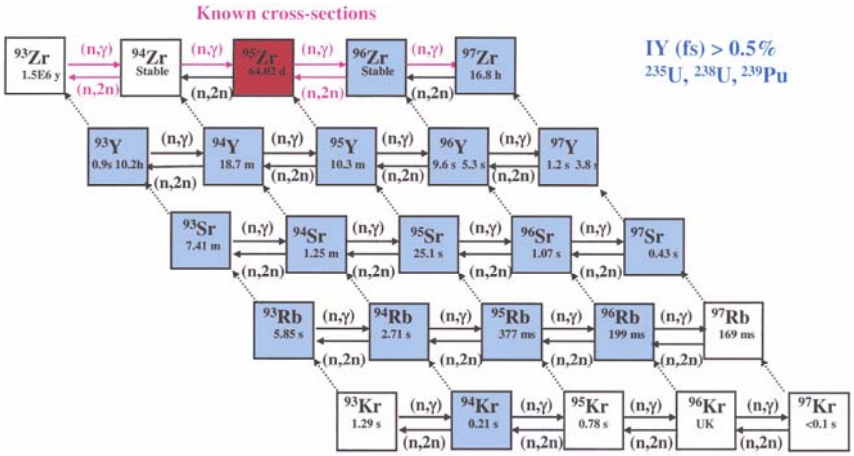
The required nuclear data (cross-section set) is extensive and largely modeled



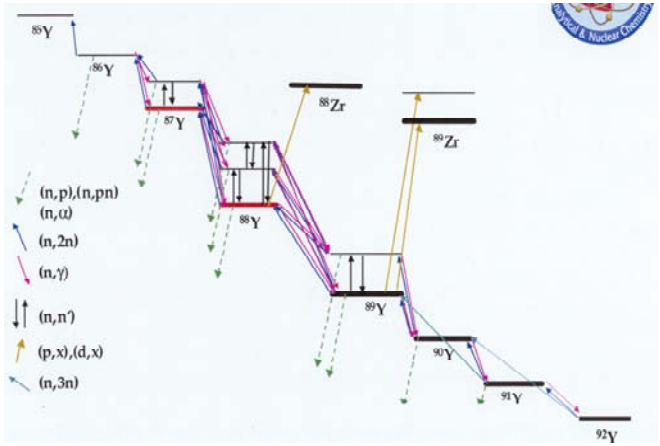
We are required to improve our understanding of the performance of nuclear weapons and quantify the uncertainties

Among other things, this means improving the nuclear data used to calculate past nuclear events

Burn-up of fission products

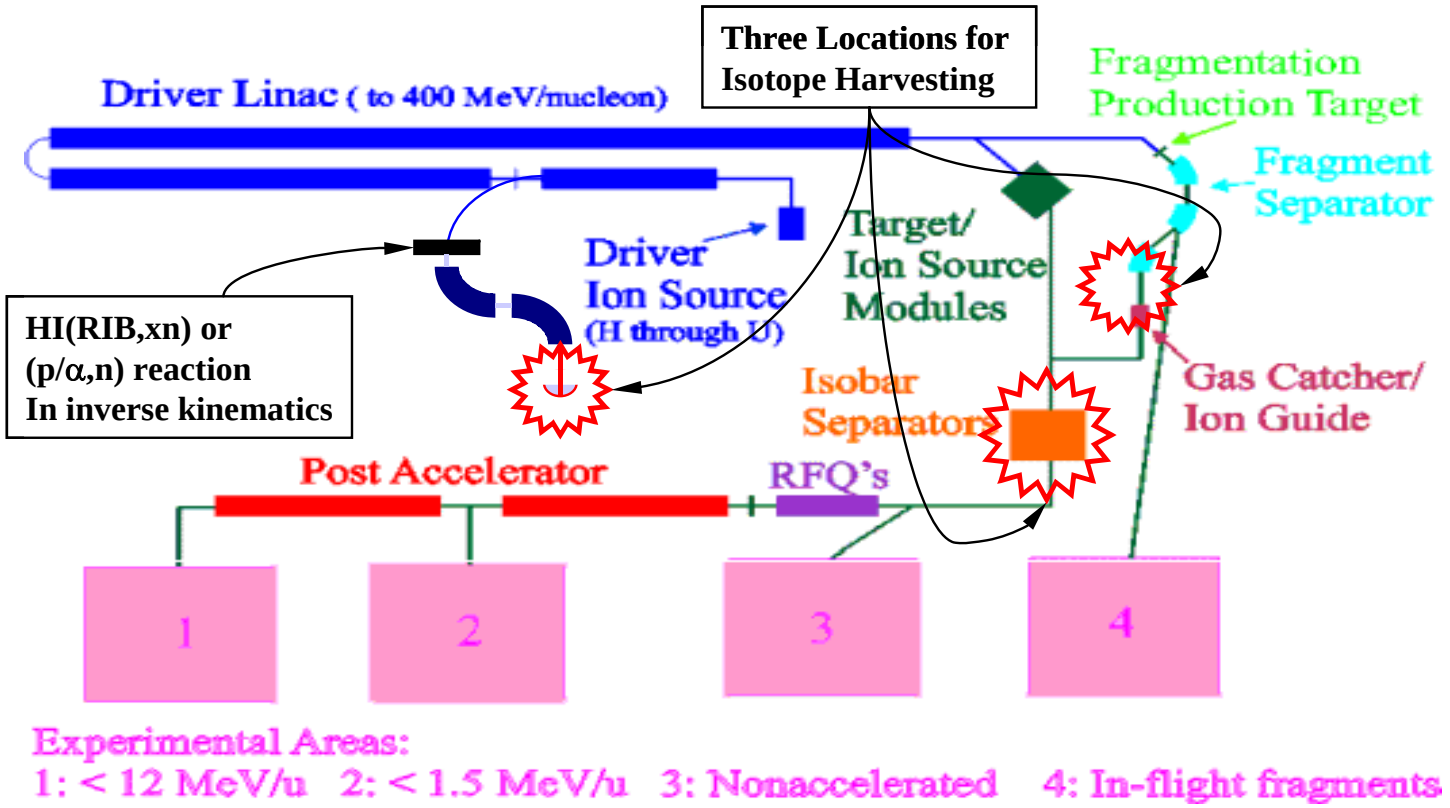


Modeled vs. measured cross sections for thermonuclear reactions



RIA will enable us to measure neutron-induced cross-sections on radioactive isotopes currently un-measurable

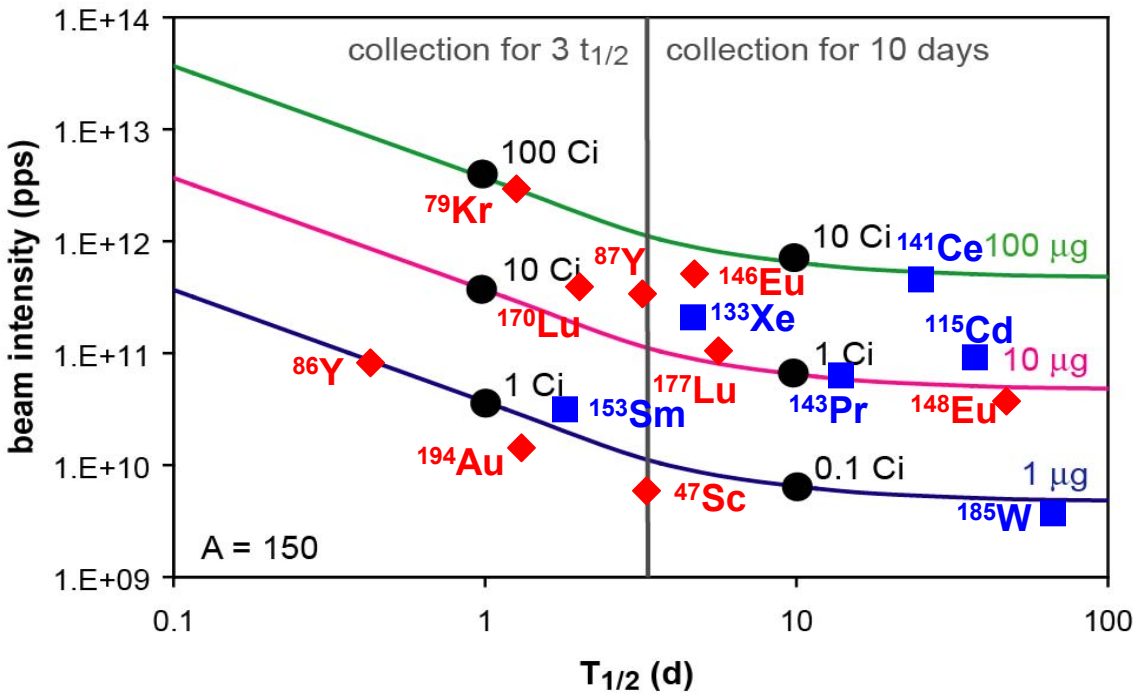
Harvesting Isotopes at RIA



1. Production at first stripper – Direct Reactions
2. ISOL with Mass Separator
3. Fragmentation with IGISOL system

How Much Can Be Harvested at RIA

$$dN(t)/dt = P - \lambda N(t) \rightarrow N(t) = P/\lambda(1 - e^{-\lambda t})$$



Assume collection period of 3 half lives or 10 days, whichever is shorter.

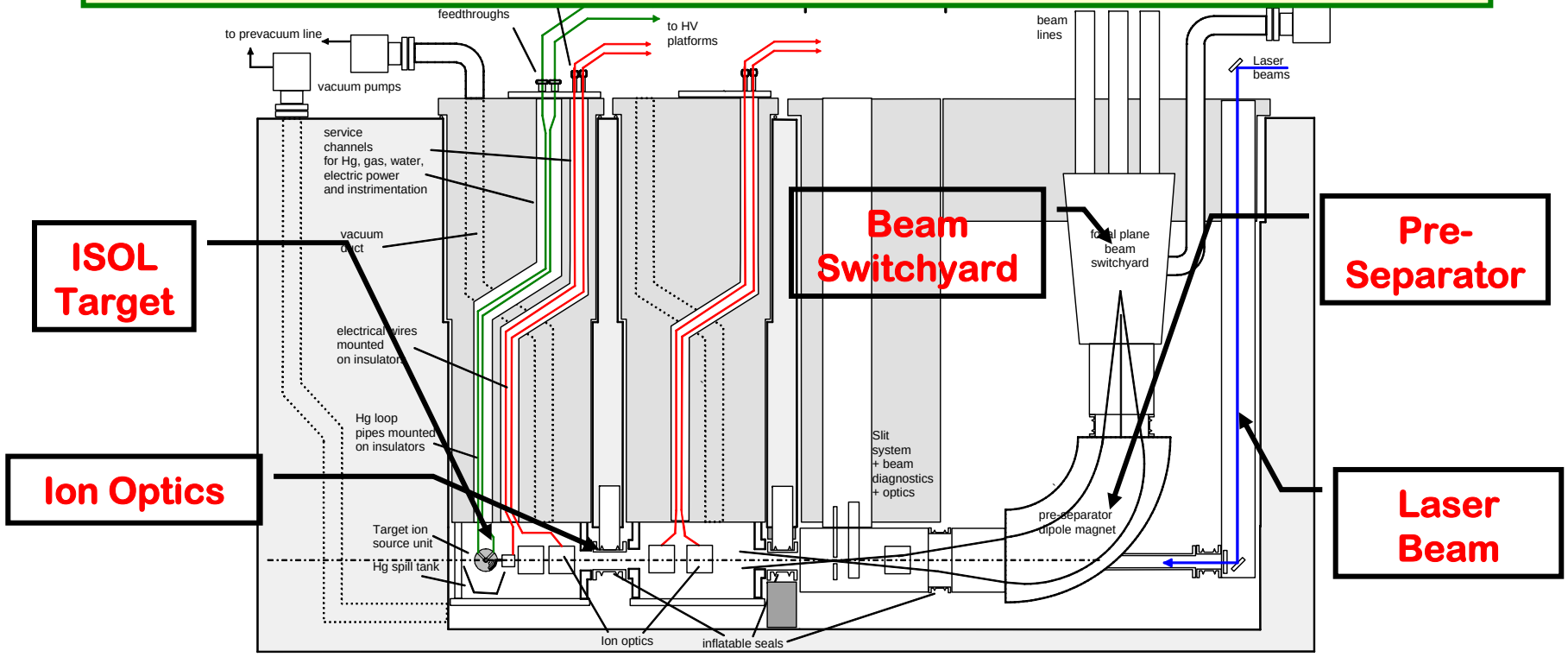
Blue squares are several s-process branch points and red diamonds are several important isotopes for Stockpile Stewardship

For 1 day half-life isotope with production rate of 10^{11} pps, $1x10^{16}$ atoms (~ 2 Curies) can be collected in 3 days.

For 1 year half-life isotopes with production rate of 10^{11} pps, $8x10^{16}$ (~ 50 mCuries) can be collected in 10 days.

Isotope Harvesting - ISOL

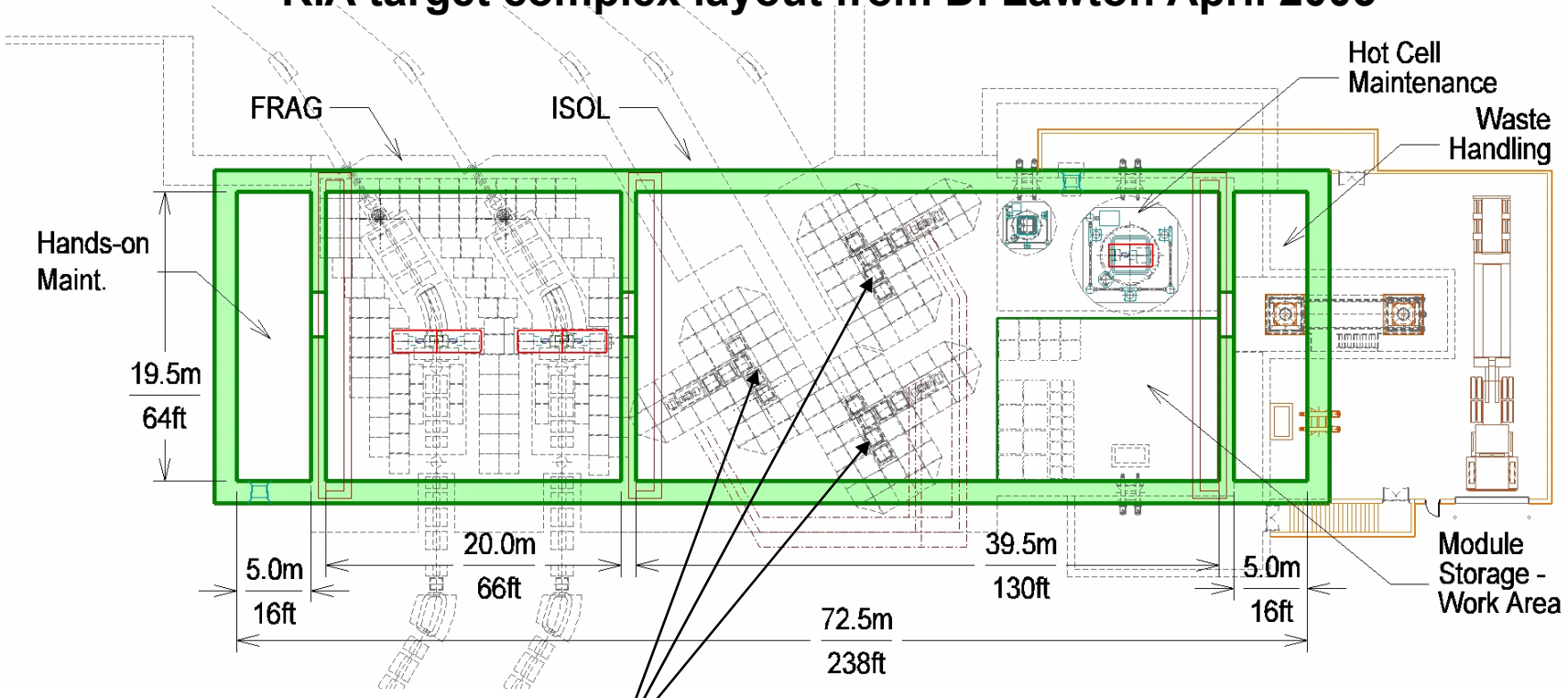
ISOL Produces ~60 keV beams, ideal for implantation into foils.



- Can more than one beam be extracted (Parasitic collection)?
- If so, how far apart in mass can secondary and primary be?

Multiple target stations might be utilized for production of certain isotopes

RIA target complex layout from D. Lawton April 2005

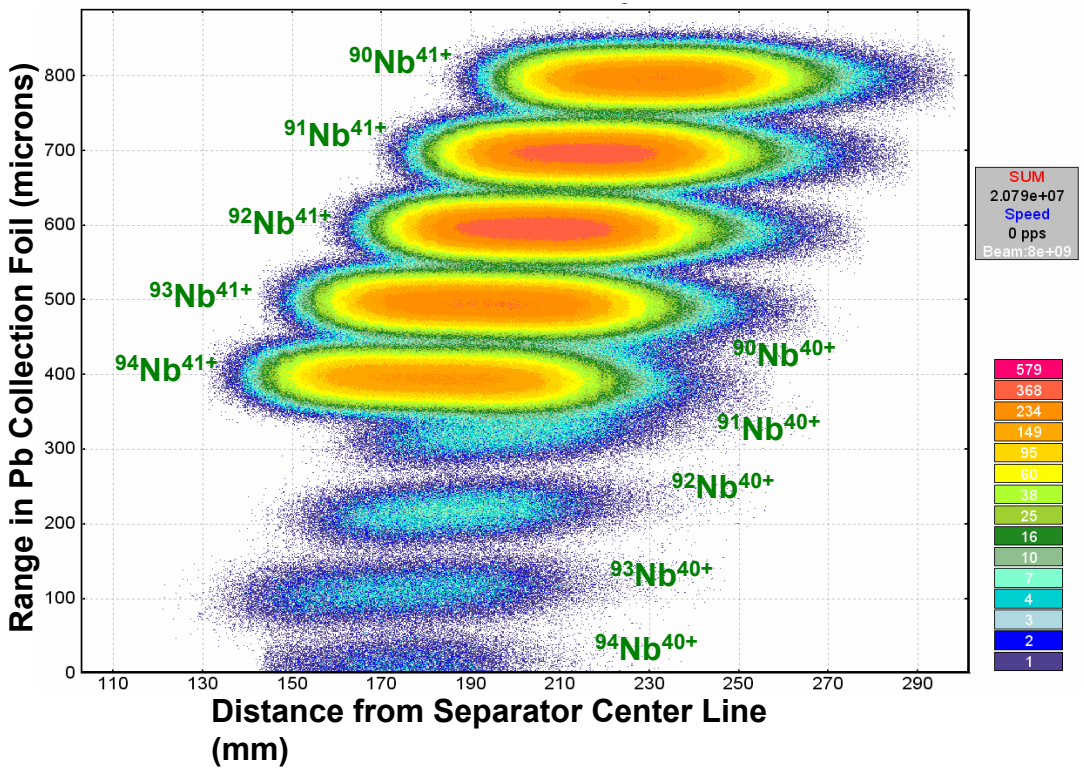


"Secondary" production of isotopes might be accomplished with a different target module located downstream from the primary target to use the leftover proton beam

Isotope Harvesting - Fragmentation

Parasitic Harvesting Difficult for Fragmentation

- **Only isotopes near to primary fragment make it through separator.**



LISE Simulation

Beam: ^{136}Xe

Primary Fragment: ^{130}Cd

Secondary Fragment: ^{92}Nb

Geometry:

1. **Dipole**
2. **Wedge (Pb)**
3. **Collection Foil (Pb)**

Only $^{90\text{-}94}Nb$ tracked

Reactions in collector foils (not simulated) will degrade purity for both parasitic and primary

Many isotopes are planned for experiments benefiting both SSP and Astrophysics communities

Summary of 1997 Workshop at Livermore

			Rad-Chem		s-process		
Isotope	Half-life			Koehler-Priority	Kappeler Feasible?	G. Mathews	Wilhelmy
63 Ni	100	y		3	X		
79 Se	1.00E+04	y		1	X	X	X
85 Kr	10.7	y		1	X	X	X
86 Rb	18.8	d		1			
88 Y	106.6	d	R	4			
89 Sr	50.5	d		2			
90 Sr	28.8	y		2	X		
93 Zr	1.00E+06	y	R			X	X
95 Zr	64	d	R	1			X
94 Nb	2.00E+04	y		2	X		
95 Nb	3.50E+01	d		2			
99 Tc	2.00E+05	y				X	X
106 Ru	367	d		2	X		
107 Pd	1.00E+06	y				X	X
119 Sn	Stable					X	X
134 Cs	2	y		1			X
135 Cs	3.00E+06	y		2	X		X
137 Cs	30.17	y		2	(X)		
141 Ce	32	d				X	X
147 Nd	11	d		1			
147 Pm	2.6	y		1	X		X
151 Sm	90	y				X	X
152 Eu	13	y	R	1			X
154 Eu	8.5	y	R	1			
155 Eu	5	y	R	1	X		X
153 Gd	241	d		1	X	X	X
160 Tb	72.1	d		1			
161 Tb	6.9	d		2			
163 Ho	33	y		1	X (x-ray)		X
166 Ho	1200	y		2			
169 Er	9.4	d		1	X	X	
170 Tm	128	d	R	1	X (?)	X	X
171 Tm	1.92	y	R	1	X		
175 Yb	4.19	d		2	(X)		
176 Lu	3.60E+10	y					X
181 Hf	42.4	d		2			
182 Hf	9.00E+06	y		2	X		
179 Ta	1.70E+00	y		1	X		
185 W	75	d		1	X		X
186 Re	1.00E+05	y		1		X	X
191 Os	15.4	d		1	X		
192 Ir	74	d	R	1			X
193 Pt	50	y		1	X (x-ray)	X	X
198 Au	2.69	d		2			
203 Hg	46.8	d		1			
204 Tl	3.77	y		1	X	X	
205 Pb	1.00E+07	y					X
210m Bi	3.00E+06	y		2	X		
210g Bi	5.01	d		2	X		

Isotope production at the ILL, 2002

Target	Product	Minimum	Half Life
Nd-146	Pm-147	3.8 mg	2.62 yr
Sm-154	Eu-155	8.9 mg	4.76 yr
Er-170	Tm-171	9.4 mg	1.92 yr

Stockpile Stewardship Target Plan

FY 2002	FY 2003	FY 2004	FY 2005
^{171}Tm	^{238}Pu	^{192}Ir	^{232}U
^{173,174,176}Lu	^{170}Tm	2 tbd	2 tbd
^{155}Eu	1 tbd		

s-Process Branch Point Nuclei

(Half-lives greater than 1 yr)

^{151}Sm, ^{147}Pm (FY2002)

^{63}Ni, ^{85}kr, ^{93}Zr, ^{99}Tc, ^{134}Cs, ^{152}Eu

^{154}Eu, ^{155}Eu, ^{163}Ho, ^{171}Tm, ^{176}Lu, ^{179}Ta

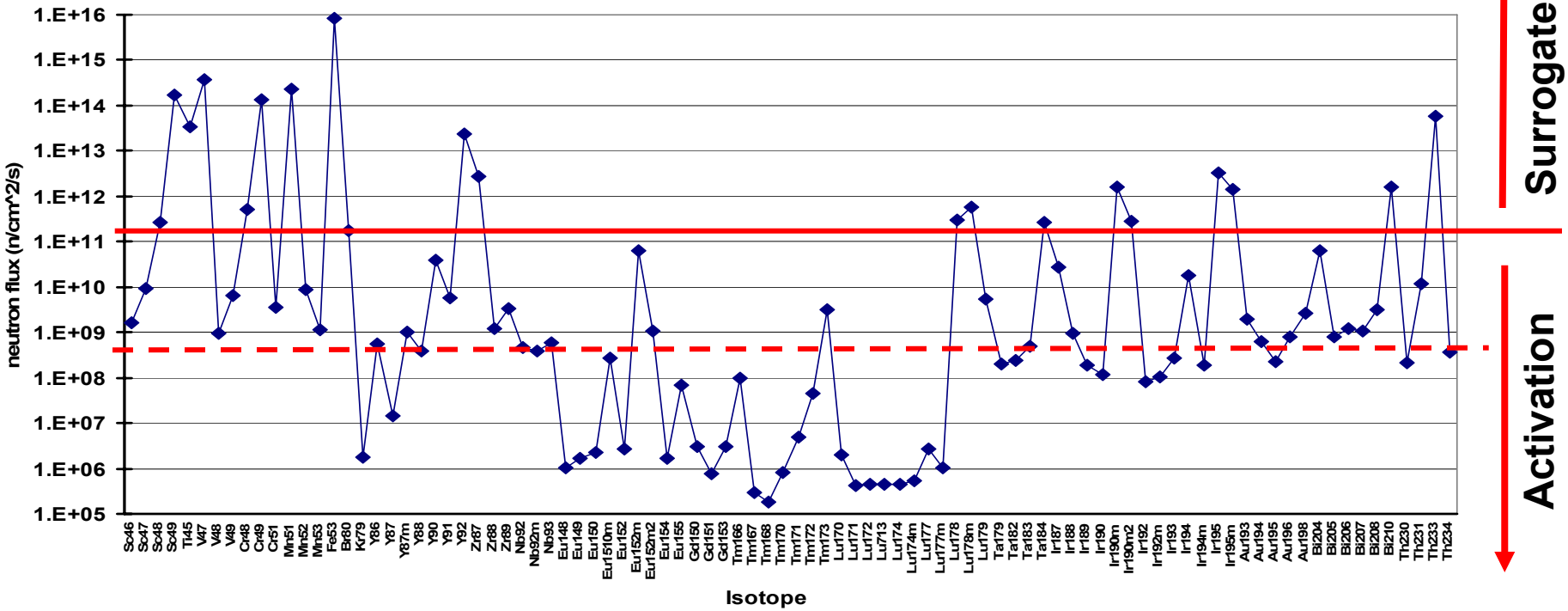
^{186}Re, ^{193}Pt, ^{204}Tl, ^{205}Pb

Required Flux For Activation Measurements at RIA

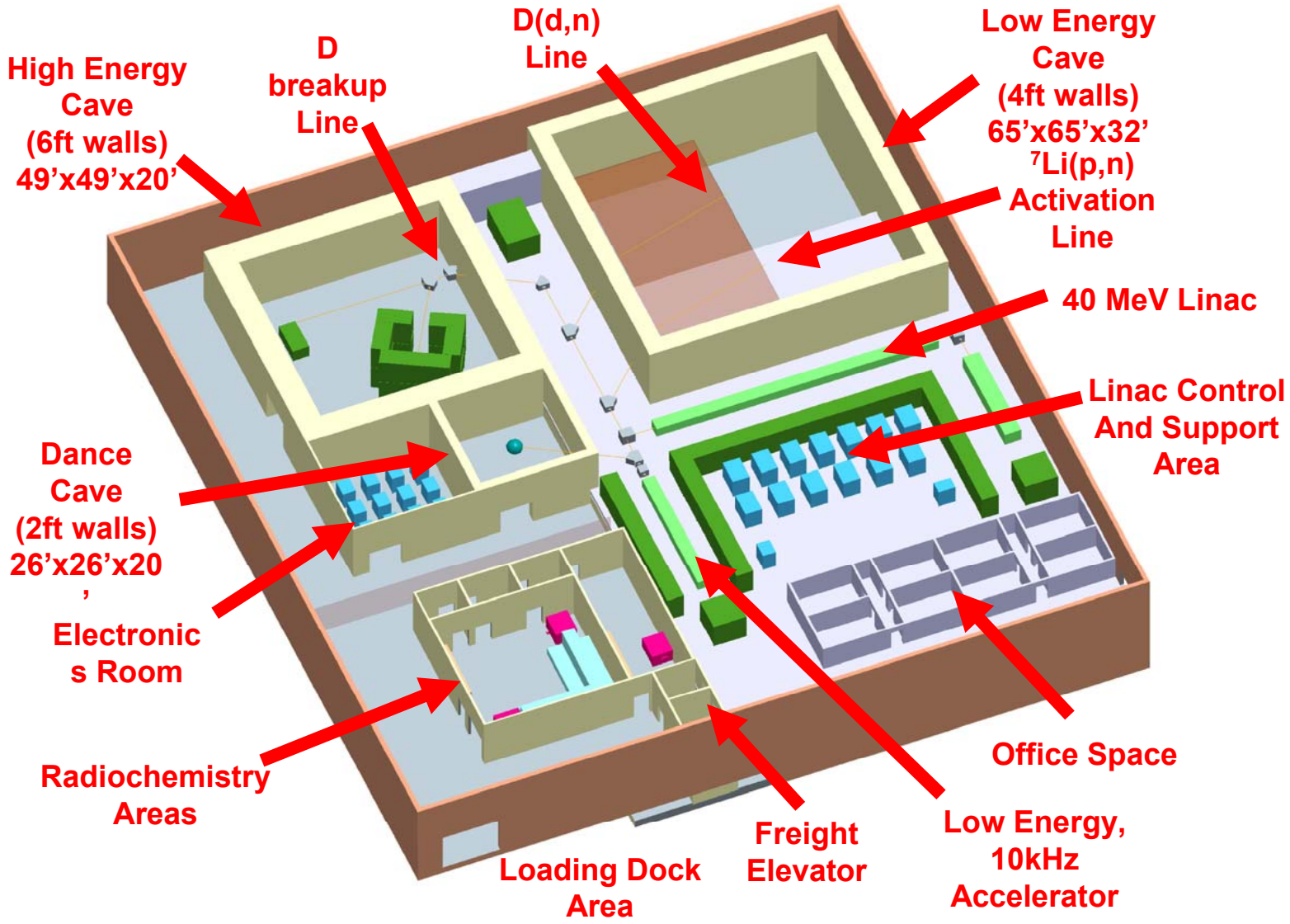
Assumptions

- RIA production rates from ANL website
- 10 day collection period, 10 day neutron irradiation
- (n,γ) cross section at 40 keV
- 1% detection efficiency (activation method)
- 1000 detected events needed (~3% statistical error)

Shorter-lived isotopes require surrogate methods also



On Site Neutron Source for RIA



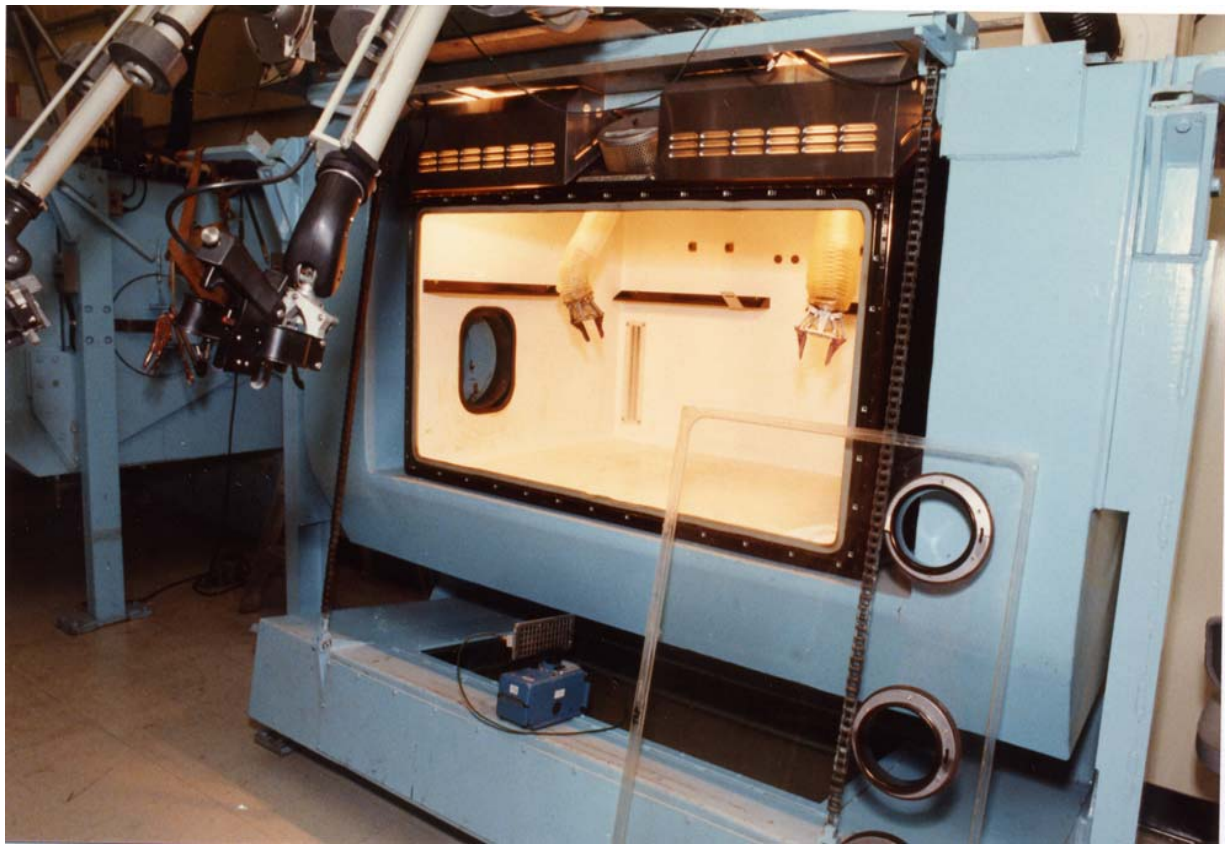
The basic elements of a manipulator cell for chemical operations

The shielding is separate from the contamination enclosure

Chemical operations favor an effectively closed rather than a truly closed system

- Containment using air flow prevents corrosion and build up of gases

There is an intake filter as well as an outlet (HEPA) filter



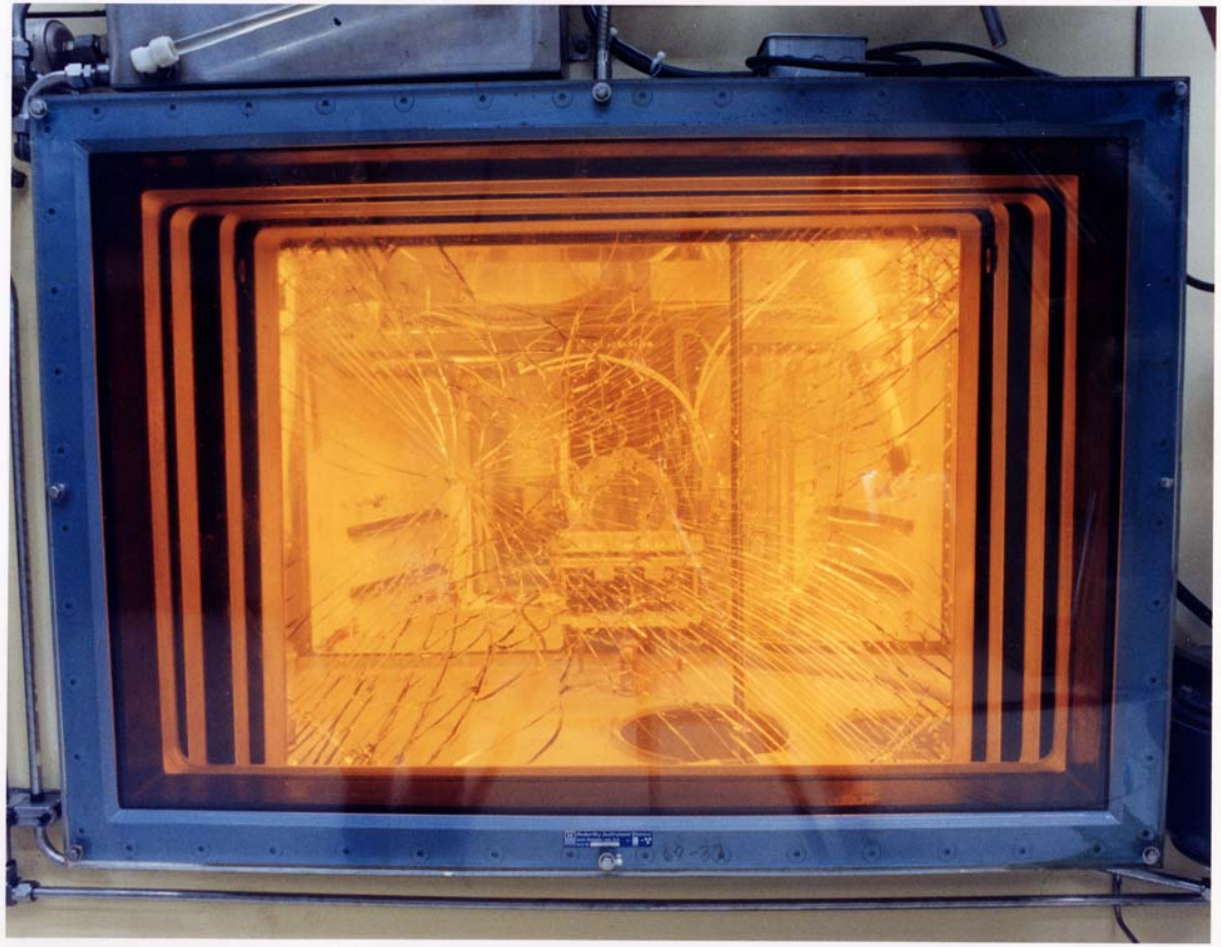
The 1-ft water window can be slung below the cell, leaving the glove box intact

Some engineering issues may arise after the cell goes into operation

For instance, it should not be possible to apply force to the inside of the window with a manipulator hand

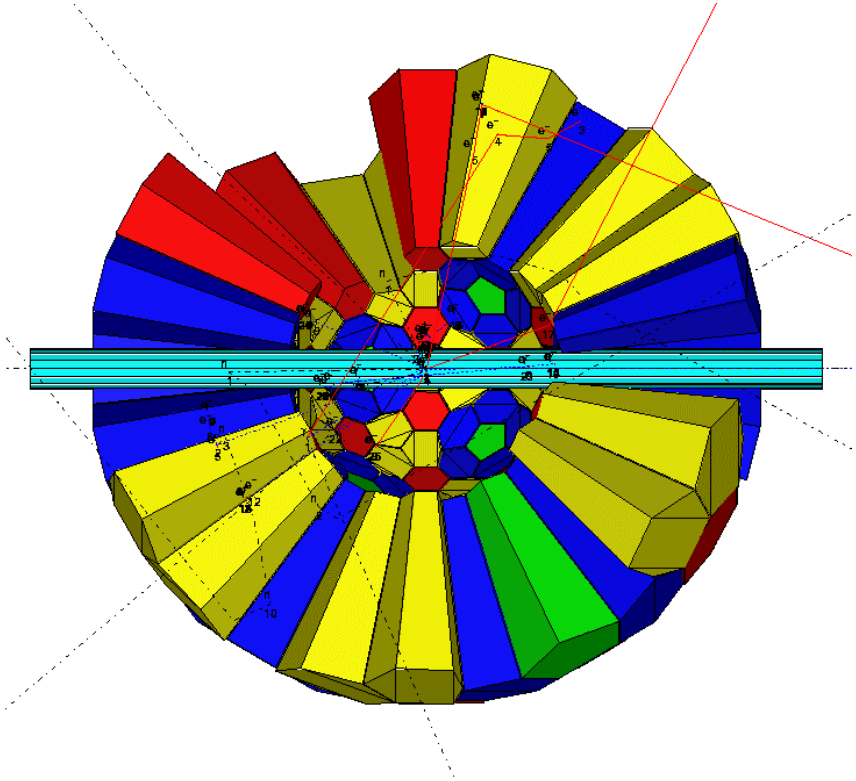
This is **bad**

Correction of this "little problem" took almost a year



A containment breach is the third-from-worst thing that can happen in-cell

The DANCE barium fluoride array



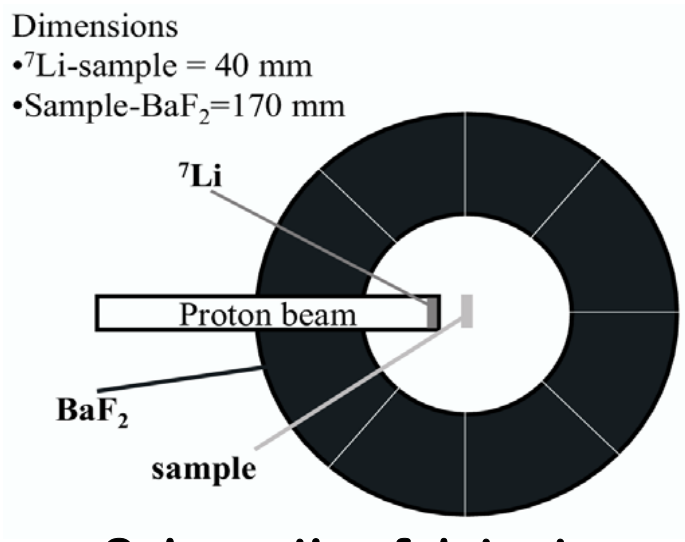
- 162 segments with 4 different shape crystals (159 segments with crystals)
- High efficiency will allow measurements on milligram samples
- Highly segmented to allow detection of radioactive targets
- Hit pattern analysis and reaction calorimetry to minimize backgrounds
- Inner radius = 17 cm
- Crystal depth = 15 cm
- Extensive Monte Carlo simulations to design detector
- All crystals will be delivered in FY2002
- State-of-the-art fast digitizers for data acquisition
- Array will be completed in 2002, but some data may be obtained with partial array.
- M. Heil, et al., Nucl. Instr. Meth. A459, 229-246 (2001)

Monte Carlo (GEANT) Simulations
- M. Heil
- R. Reifarth
- F. Kaeppeler

Forschungzentrum Karlsruhe

Measure (n,γ) cross-sections on radioactive targets

DANCE at RIA - Overview

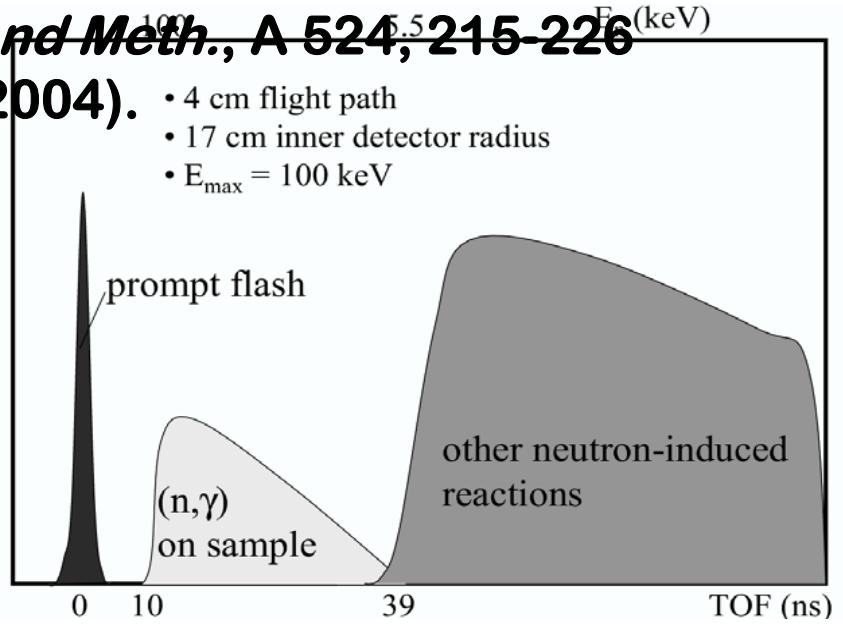


Schematic of detector setup proposed for RIA

Use BaF_2 array similar to DANCE

^{7}Li(p,n) instead of spallation neutron source

R. Reifarth, et al., *Nucl. Inst. And Meth.*, A 524, 215-226 (2004).



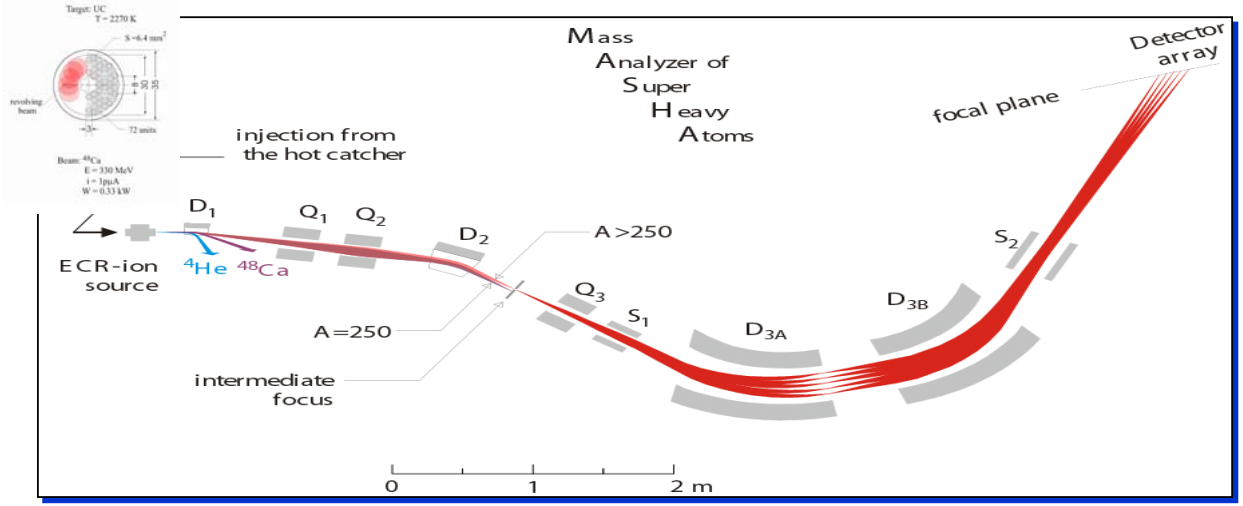
Time dependence of gamma spectrum seen by BaF_2 detector

Required Neutron Flux Reduced by factor of 10.

- **Efficiency near 100%**
- **10 times more detected events needed for background subtraction**

Novel target materials are being explored to increase robustness and release rates

- **High temperature ceramic targets for MASHA are being developed and tested for the harvesting of very rare isotopes– namely SHE produced ~1 atom/month**



Ca + Pu → 114 (eka-Pb)

Zr stabilizes the matrix over a large temperature range so there is no phase transition

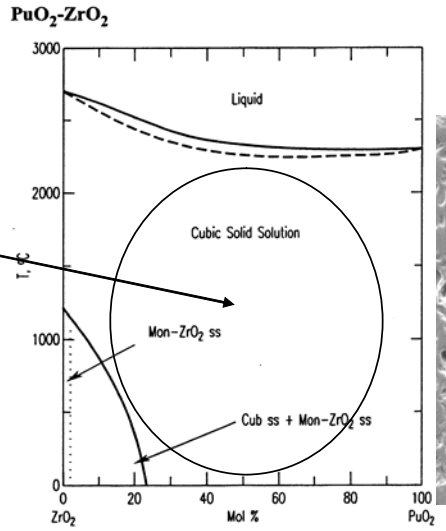
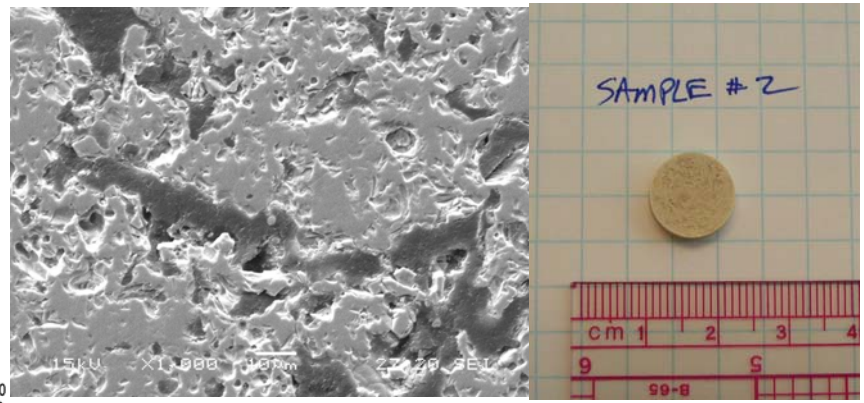


Fig. Zr-187—System ZrO_2-PuO_2. Mon = monoclinic; Cub = cubic.
D. F. Carroll, *J. Am. Ceram. Soc.*, **46** [4] 194-195 (1963).

In preparation for making Pu ceramics, we made a surrogate (Ca + Sm → Pb)



65% Sm_2O_3 35% ZrO_2

Conclusions

- Isotope harvesting will benefit a variety of projects
- The feasibility of parasitically harvesting isotopes at RIA is being investigated
- Investigation of using the leftover primary proton beam in a second target to produce isotopes has started
- Novel target materials are being manufactured and tested
- A combination of cross-section measurement techniques will be used to measure interesting cross-sections
  - —Activation experiments
  - —Surrogate reactions