Production of interesting isotopes with plutonium-242 targets

Paul A. Ellison

University of California, Berkeley, Department of Chemistry

2011 DOE NNSA SSGF Annual Conference, July 21st, 2011

Acknowledgements

- UC Berkeley / LBNL Heavy Element Nuclear and Radiochemistry Group
  - J.P. McLaughlin, C. Fineman-Sotomayor, J. Champion, Z. Dvorakova, L. Stavsetra, K.E. Gregorich, **H. Nitsche**
- LBNL 88-Inch Cyclotron
  - Scientists / Postdocs / Ion source and cyclotron staff
- Funding
  - US DOE Offices of High Energy and Nuclear Physics and Basic Energy Sciences
  - US DOE NNSA Stewardship Science Academic Alliances Grant
  - **US DOE NNSA Stewardship Science Graduate Fellowship**
    - Krell Institute for outstanding administration

Overview

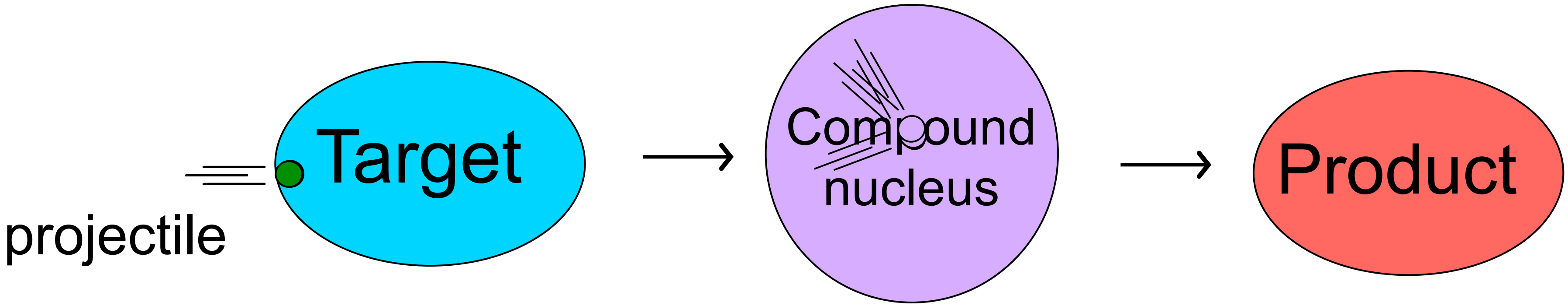
1 H																	2 He
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
55 Cs	56 Ba	57 La	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
87 Fr	88 Ra	89 Ac	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Cn	(113)	114	(115)	(116)	(117)	(118)

Lanthanides	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
Actinides	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr

Overview

- Low energy nuclear reaction studies
- ^{242}Pu target production
- **Part I:** Production of ^{240}Am
  - Motivation / Background
  - Results
- **Part II:** Production of ^{285}114
  - Motivation / Background
  - Results

Low energy nuclear reaction studies



Number of target atoms being bombarded

Intensity of bombarding ions

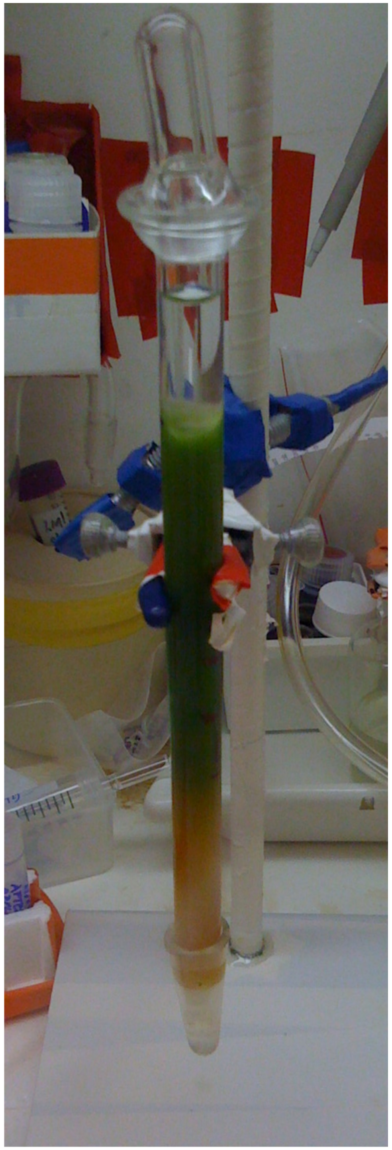
Net production rate $= N \cdot I \cdot \sigma$

Cross section

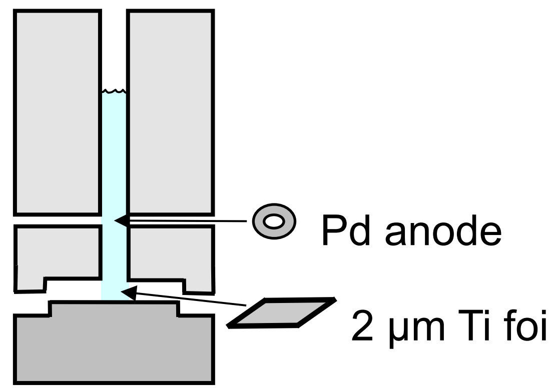
- dependent on projectile, energy of projectile, and target
- essentially the probability that product will be formed
- measured in units of area (1 barn = 10^{-24} cm^2)

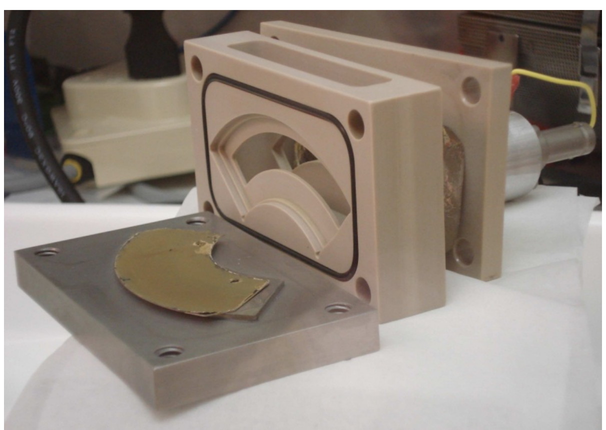
^{242}Pu target production

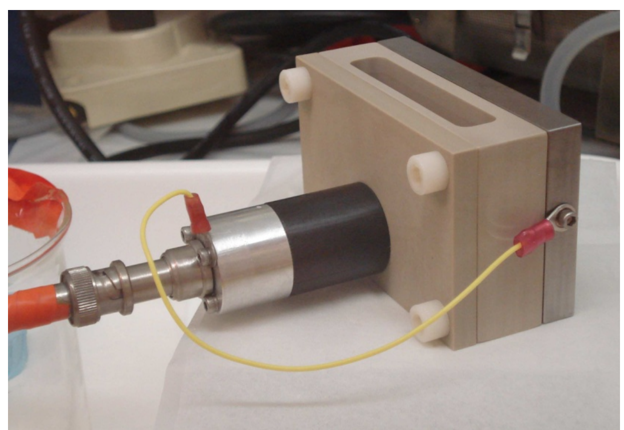
- From ORNL to LBNL in 1987 as $^{242}PuO_2$ powder
  - ^{238}Pu: 0.004%
  - ^{239}Pu: 0.005%
  - ^{240}Pu: 0.022%
  - ^{241}Pu: 0.035%
  - ^{242}Pu: 99.932%
  - ^{244}Pu: 0.002%
- Dissolved ~150 mg and purified using anion exchange chromatography
  - Removed significant ^{241}Am contamination

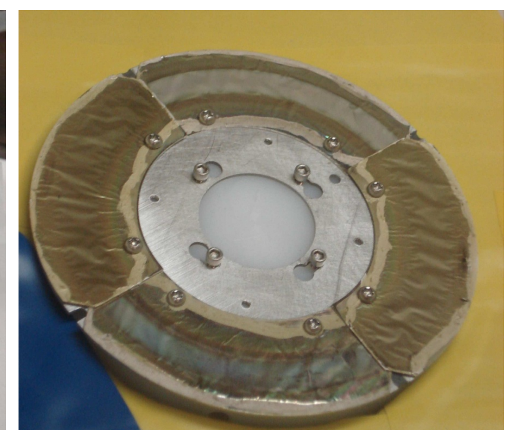


^{242}Pu target production





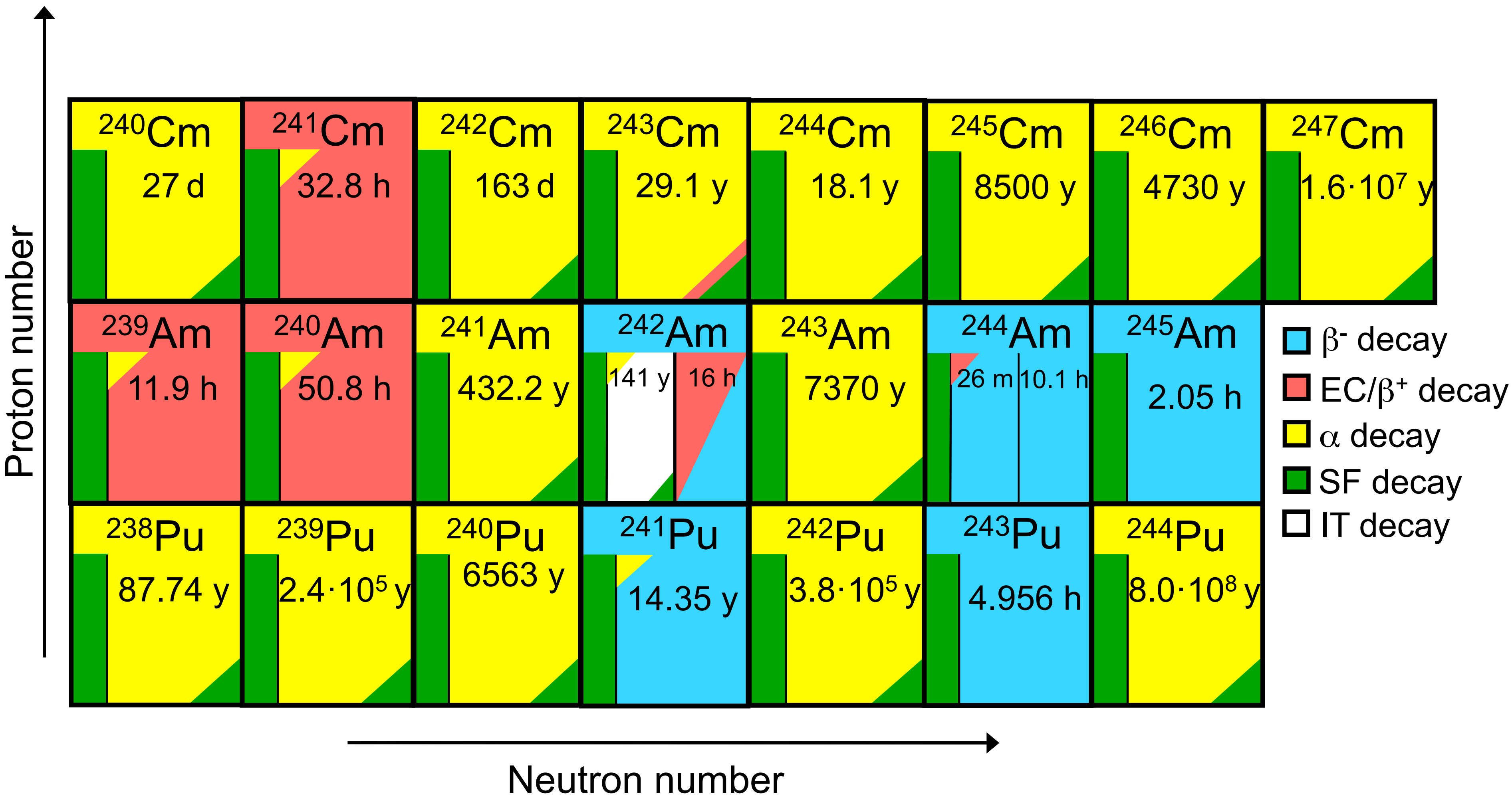




Overview

- Low energy nuclear reaction studies
- ^{242}Pu target production
- **Part I:** Production of ^{240}Am
  - Motivation / Background
  - Results
- **Part II:** Production of ^{285}114
  - Motivation / Background
  - Results

Chart of nuclides around americium-240



Why make ^{240}Am?

- Want to measure ^{240}Am(*n*,*f*) cross section
  - Post detonation nuclear forensics
  - Science-based stockpile stewardship
  - Predictions of nuclear waste isotopics of minor actinides

- Possible to measure at LANL LANSCE lead slowing down spectrometer
  - Need 10 – 100 nanograms of ^{240}Am

Producing ^{240}Am

- Cannot be produced in nuclear fission reactor
- Previously studied nuclear reactions have only low cross section (2-3 millibarn)
- ^{242}Pu(p,3n)^{240}Am has high predicted cross section of 100 – 300 millibarn, but never experimentally measured
  - Mashnik, S.G., et al., LANL Report LA-UR-05-7321, Los Alamos (2005).
  - Mashnik, S.G., et al., Research Note X-3-RN(U)-07-03, LANL Report LA-UR-06-8652, Los Alamos (2006).

Measuring the ^{242}Pu(p,3n)^{240}Am cross section

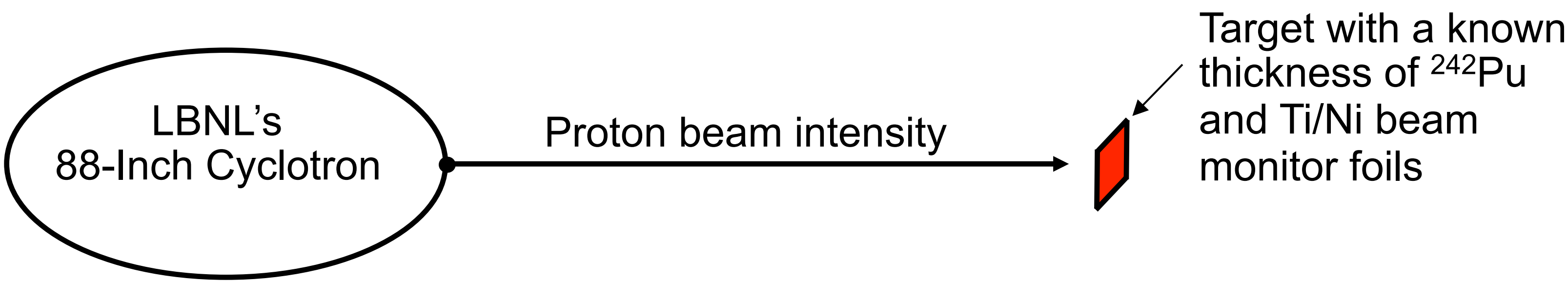
Intensity of proton beam

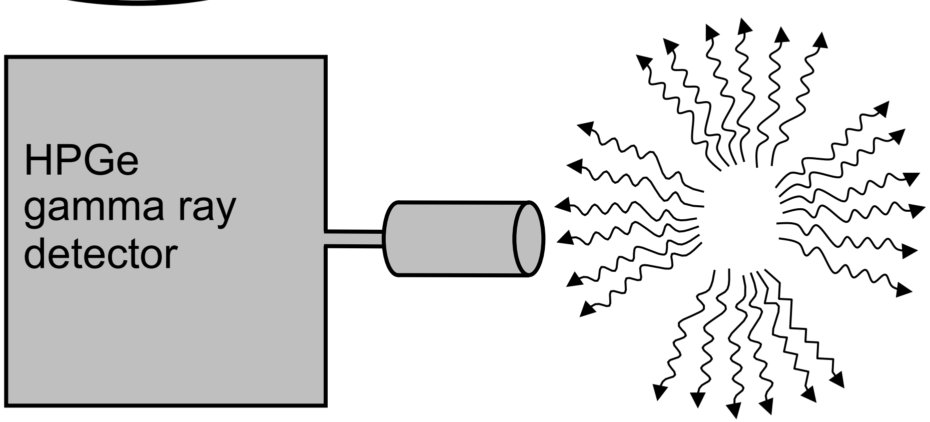
Decay rate of ^{240}Am

Thickness of ^{242}Pu target

$$\sigma = \frac{N \cdot I \cdot (1 - e^{-\lambda \cdot t})}{A_t}$$

Produced Am activity after irradiation time t

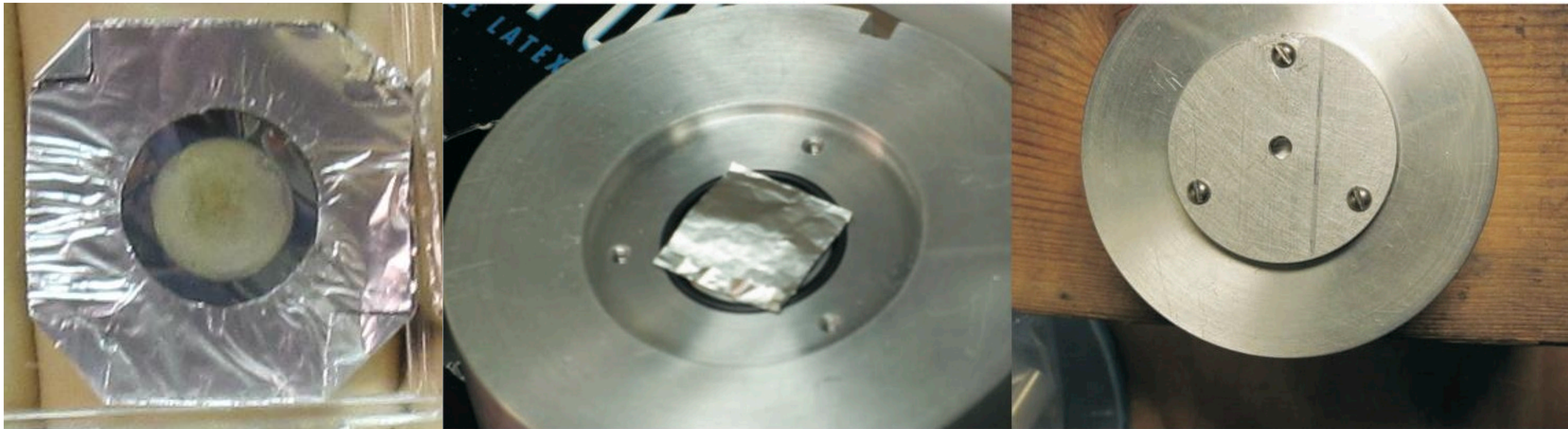


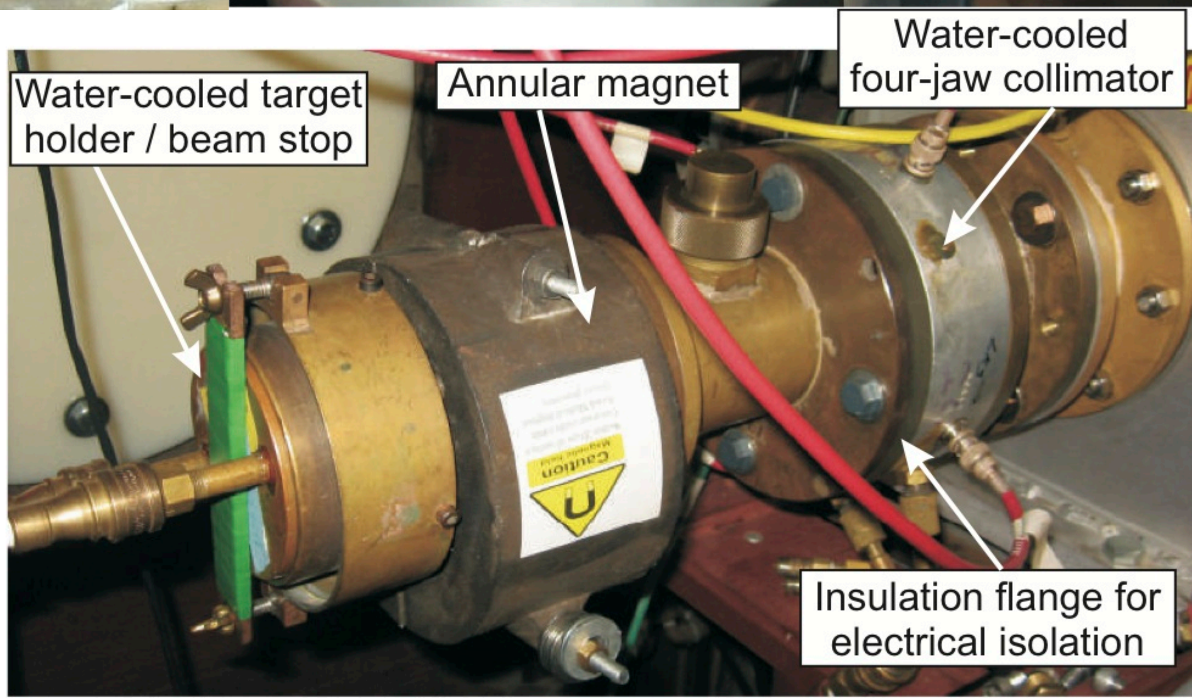


987.76 keV (73.2%)

888.80 keV (25.1%)

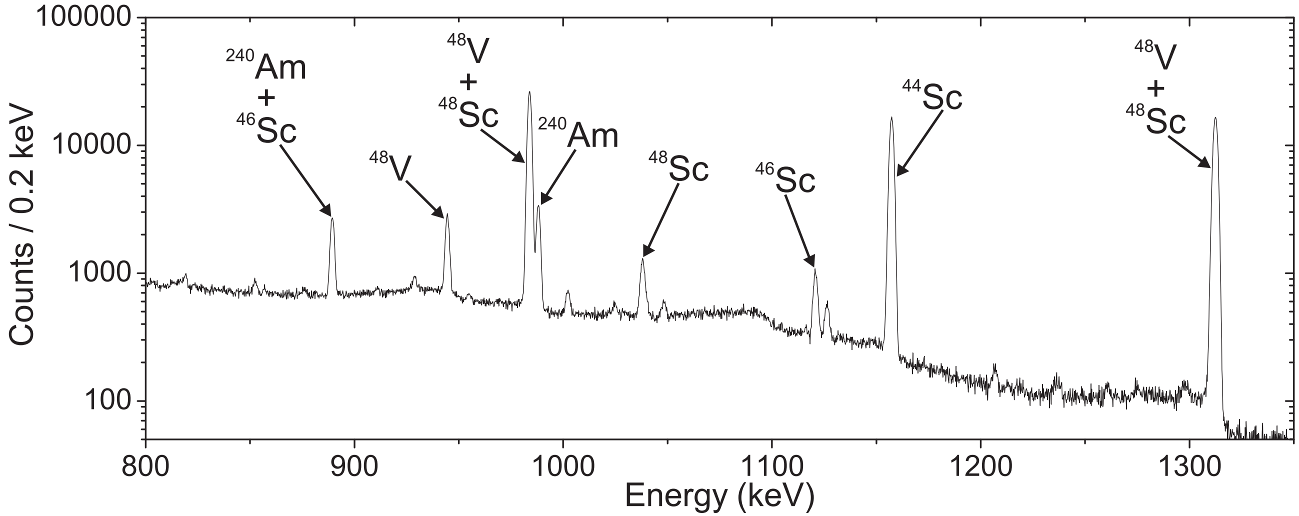
^{242}Pu(p,$3n$)^{240}Am targetry





$^{242}Pu(p,3n)^{240}Am$ post-irradiation characterization

- Irradiations of ^{242}Pu targets on Ti backings were performed during 32 hours of beam time at LBNL's 88-Inch Cyclotron in five separate experiments
  - — Involved data analysis techniques for gamma spectra of irradiated foils



Four hour gamma spectrum taken of ^{242}Pu deposited on Ti foil 34 hours after irradiation with 25 MeV protons.

$^{242}Pu(p,3n)^{240}Am$

$^{nat}Ti(p,xn)^{48}V$

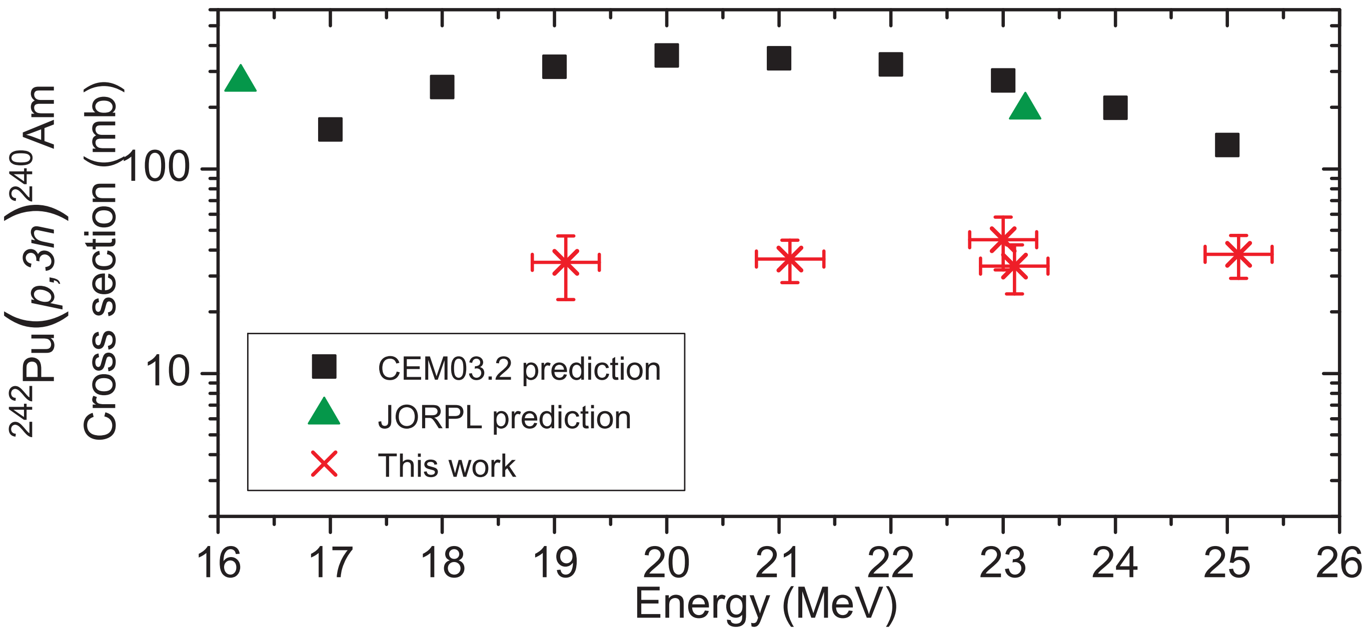
(used as proton monitor reaction)

$^{nat}Ti(p,\alpha xn)^{46}Sc$

$^{50}Ti(p,p'p''n)^{48}Sc$

$^{242}Pu(p,3n)^{240}Am$ cross section

- Cross sections measured to be **~40 millibarn**
  - Predicted to be 100 - 300 millibarn



- While less than predicted, remains the most promising heavy ion reaction for producing ^{240}Am for a LSDS irradiation

Overview

- Low energy nuclear reaction studies
- ^{242}Pu target production
- **Part I:** Production of ^{240}Am
  - Motivation / Background
  - Results
- **Part II:** Production of ^{285}114
  - Motivation / Background
  - Results

Transactinide elements

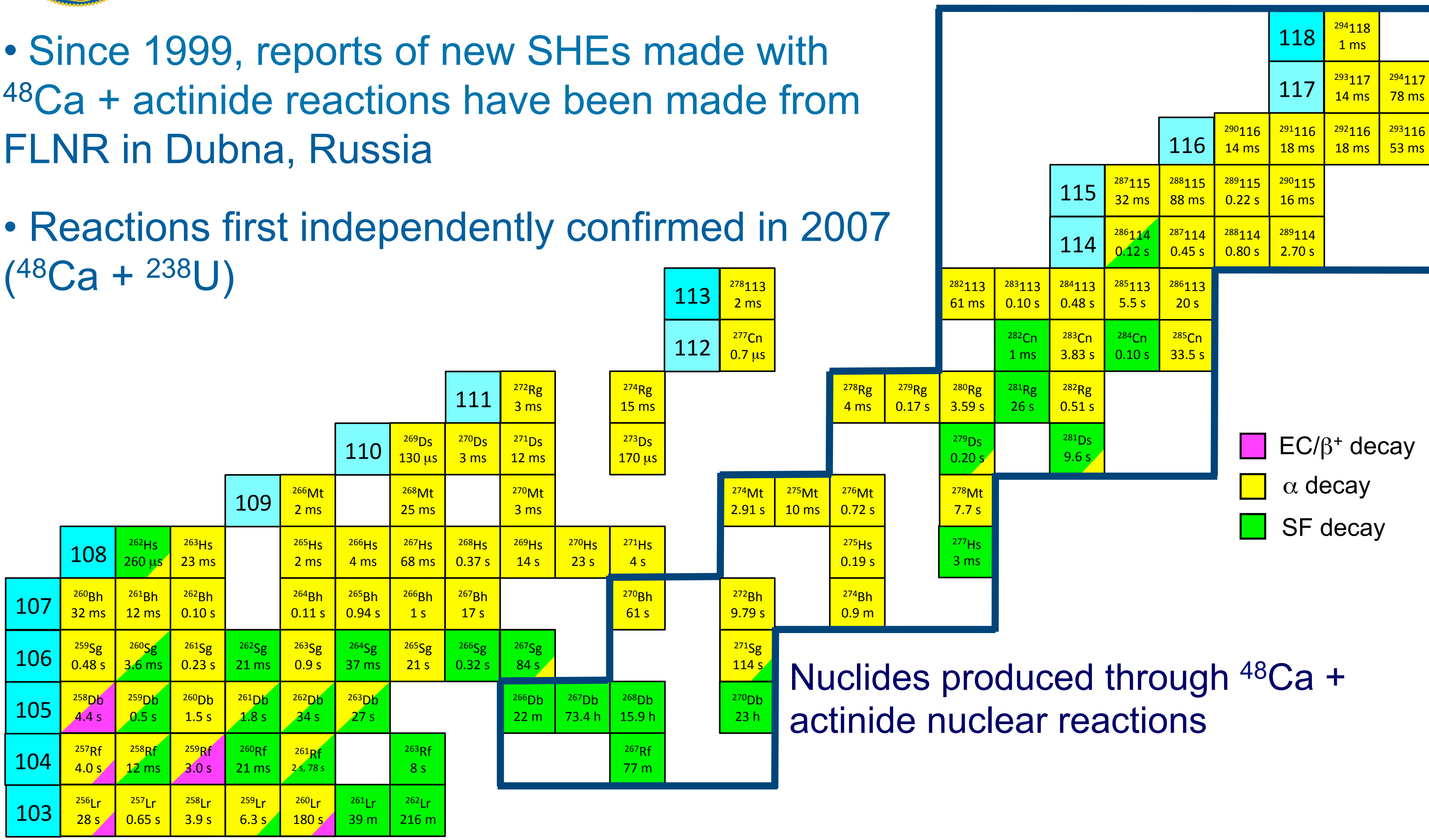
1 H																	2 He
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
55 Cs	56 Ba	57 La	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
87 Fr	88 Ra	89 Ac	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Cn	(113)	114	(115)	(116)	(117)	(118)

Lanthanides	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
Actinides	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr

Upper end of the nuclide chart

- Since 1999, reports of new SHEs made with ^{48}Ca + actinide reactions have been made from FLNR in Dubna, Russia

- Reactions first independently confirmed in 2007 (^{48}Ca + ^{238}U)



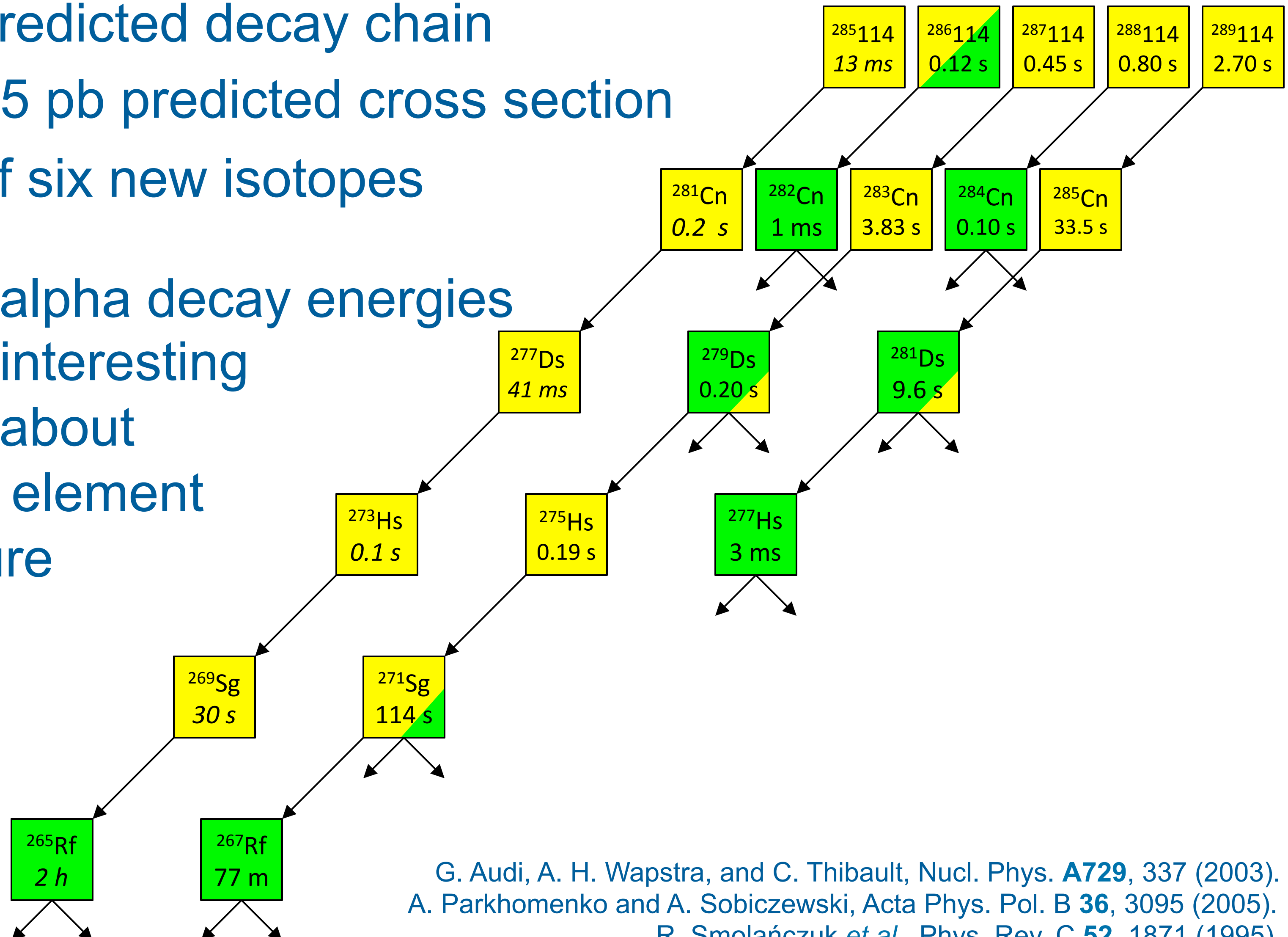
Yu. Ts. Oganessian, J. Phys. G **34**, R165 (2007).

S. Hofmann *et al.*, Eur. Phys. J. A **32**, 251 (2007).

Motivation for ^{242}Pu(^{48}Ca,5n)^{285}114

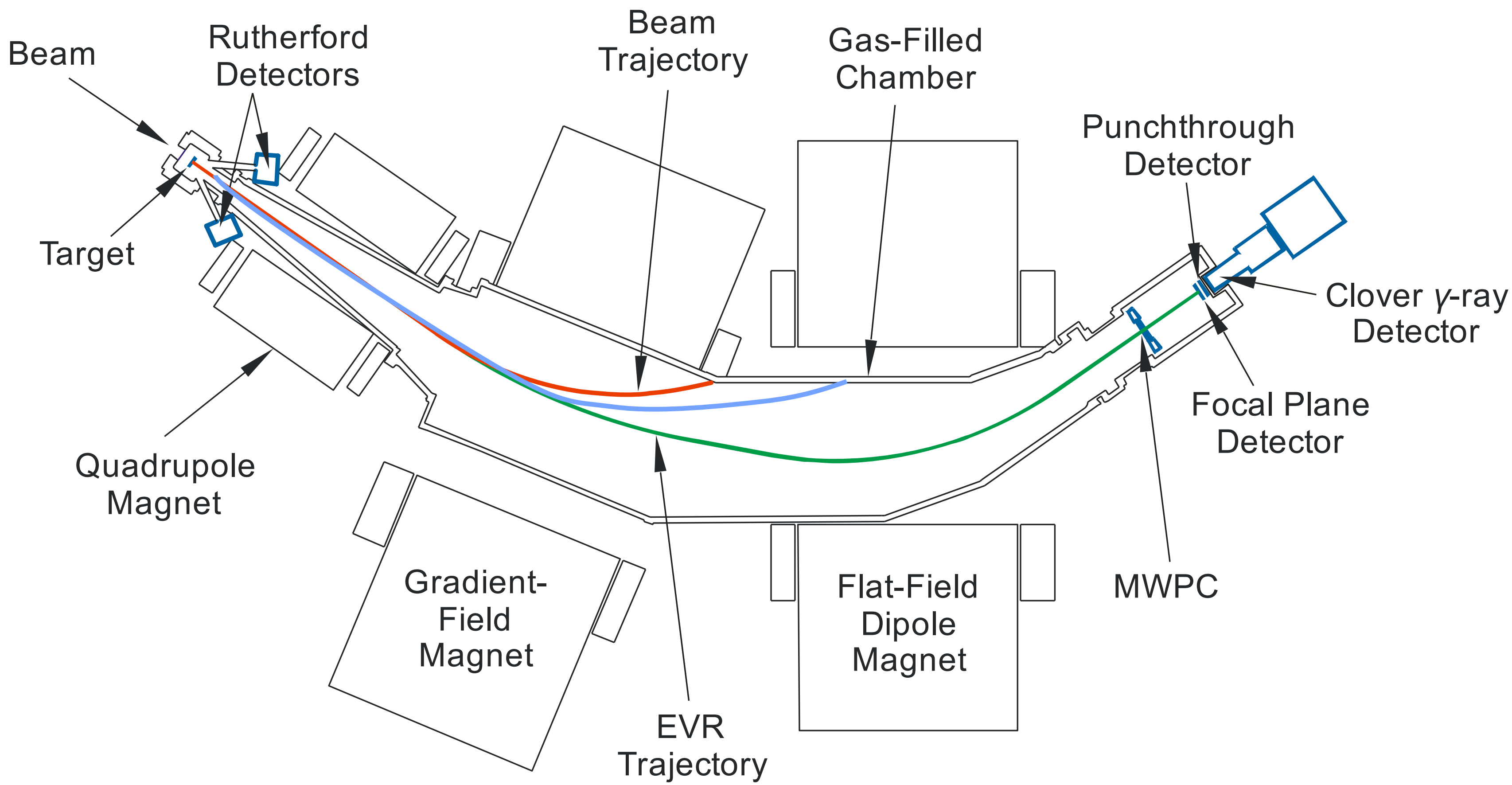
- Long predicted decay chain
- 0.5 - 1.5 pb predicted cross section
- Total of six new isotopes

5 predicted alpha decay energies would yield interesting information about superheavy element shell structure

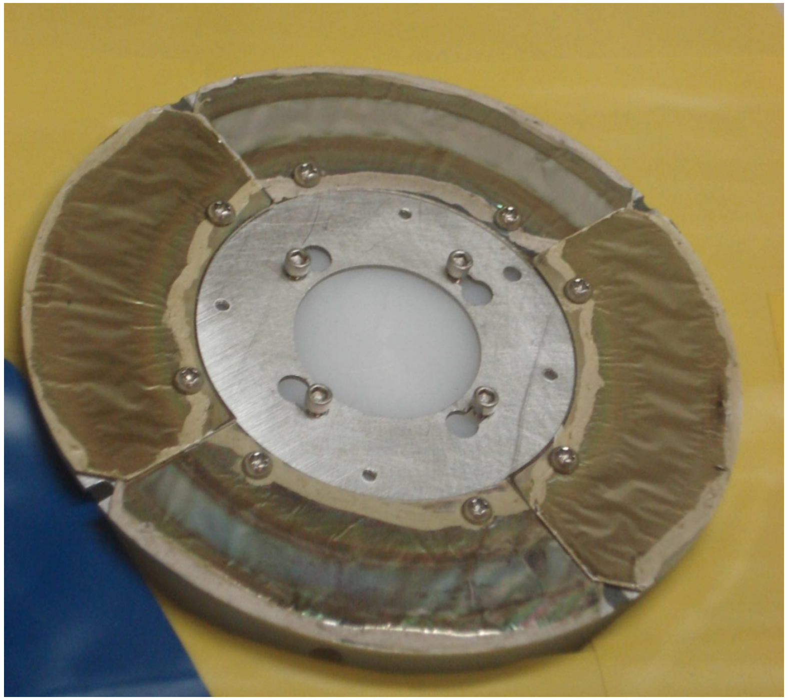


G. Audi, A. H. Wapstra, and C. Thibault, Nucl. Phys. **A729**, 337 (2003).
A. Parkhomenko and A. Sobiczewski, Acta Phys. Pol. B **36**, 3095 (2005).
R. Smolańczuk *et al.*, Phys. Rev. C **52**, 1871 (1995).

Berkeley Gas-filled Separator (BGS)

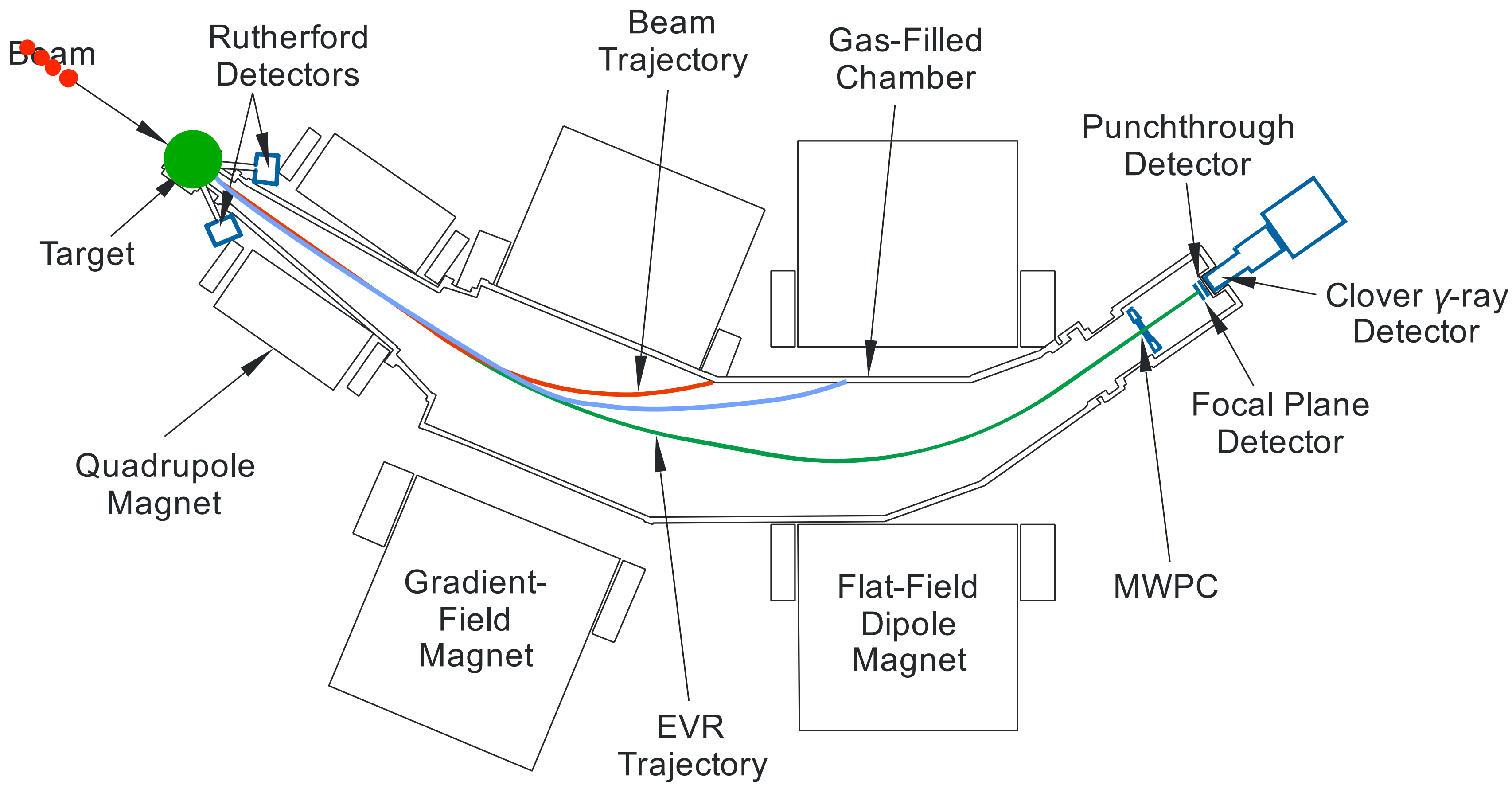


Plutonium target apparatus

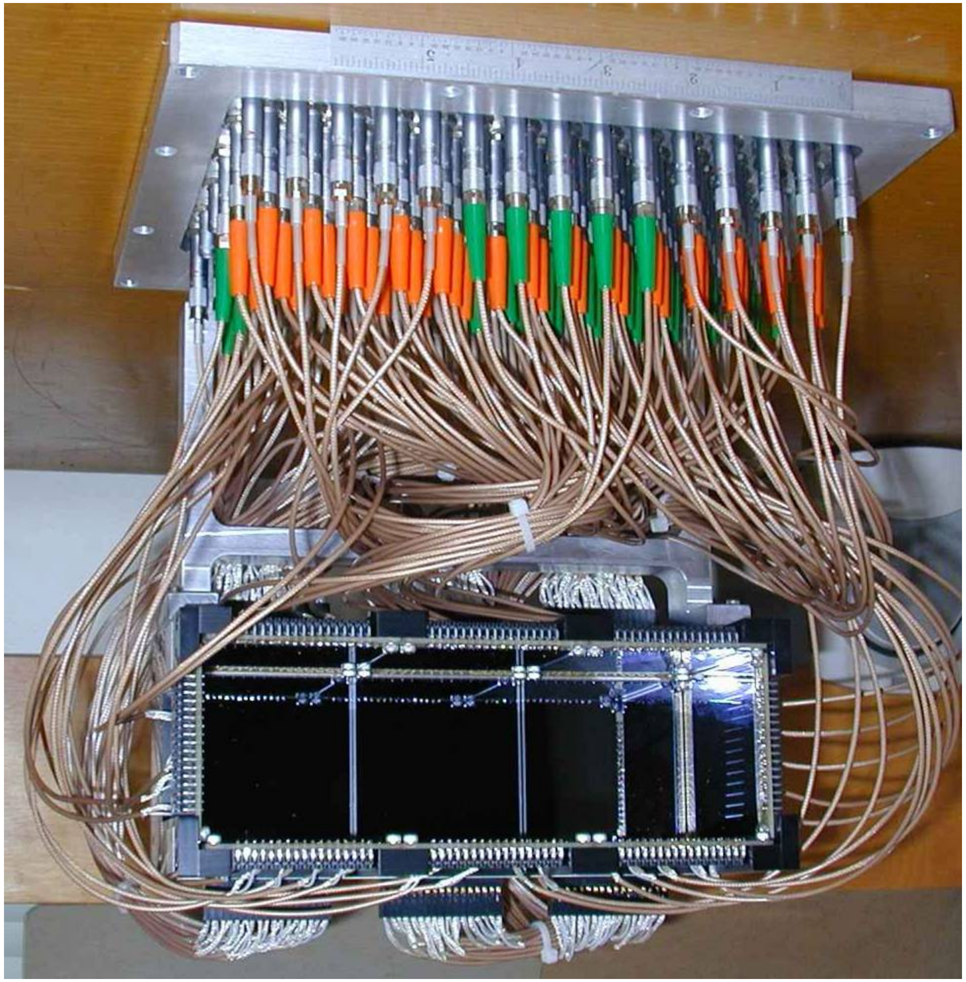


- 270 – 440 μg / cm^2 of ^{242}Pu on 2.4-μm Ti foils
- 9.5 cm diameter wheel

Berkeley Gas-filled Separator (BGS)

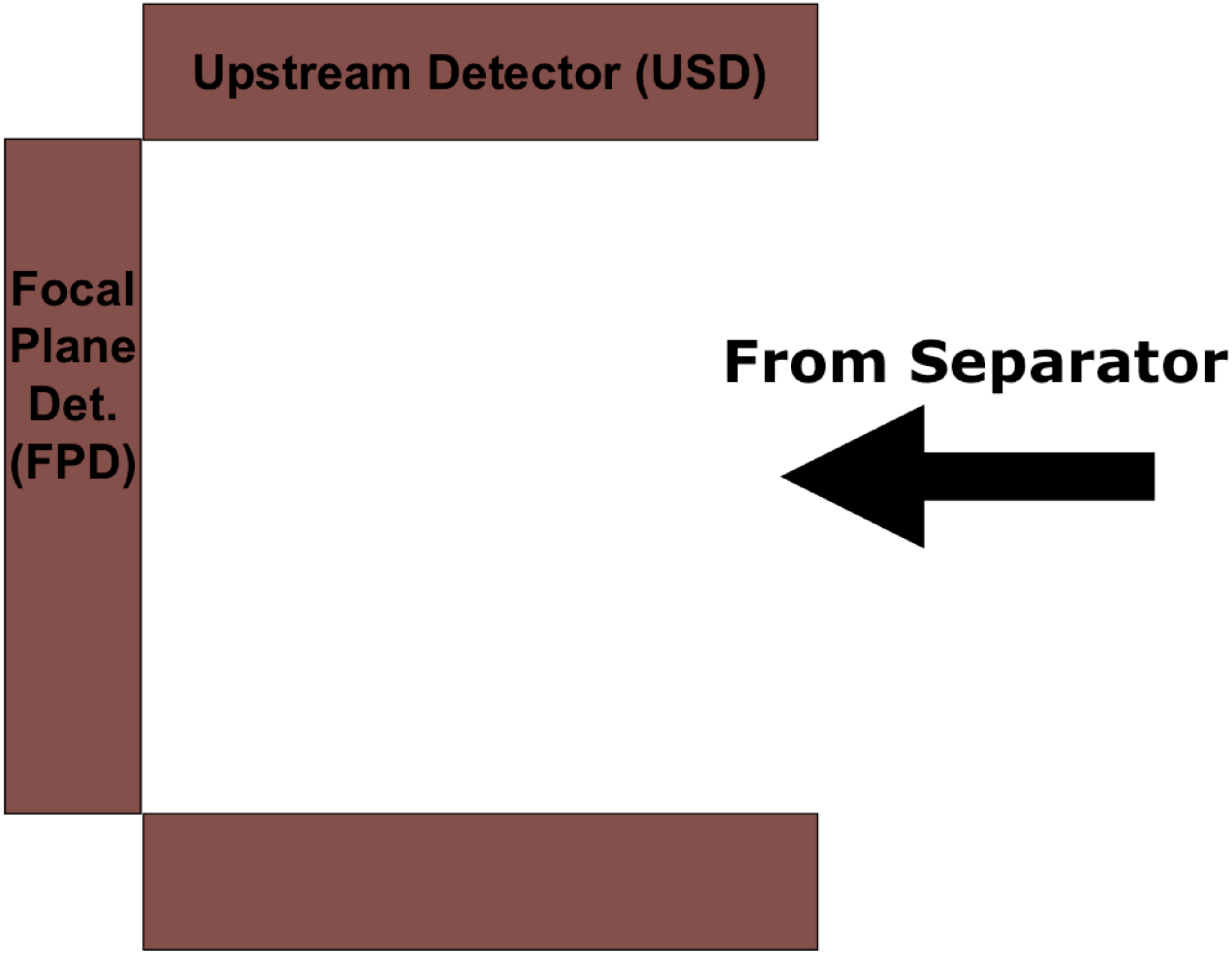


Focal-plane detector box

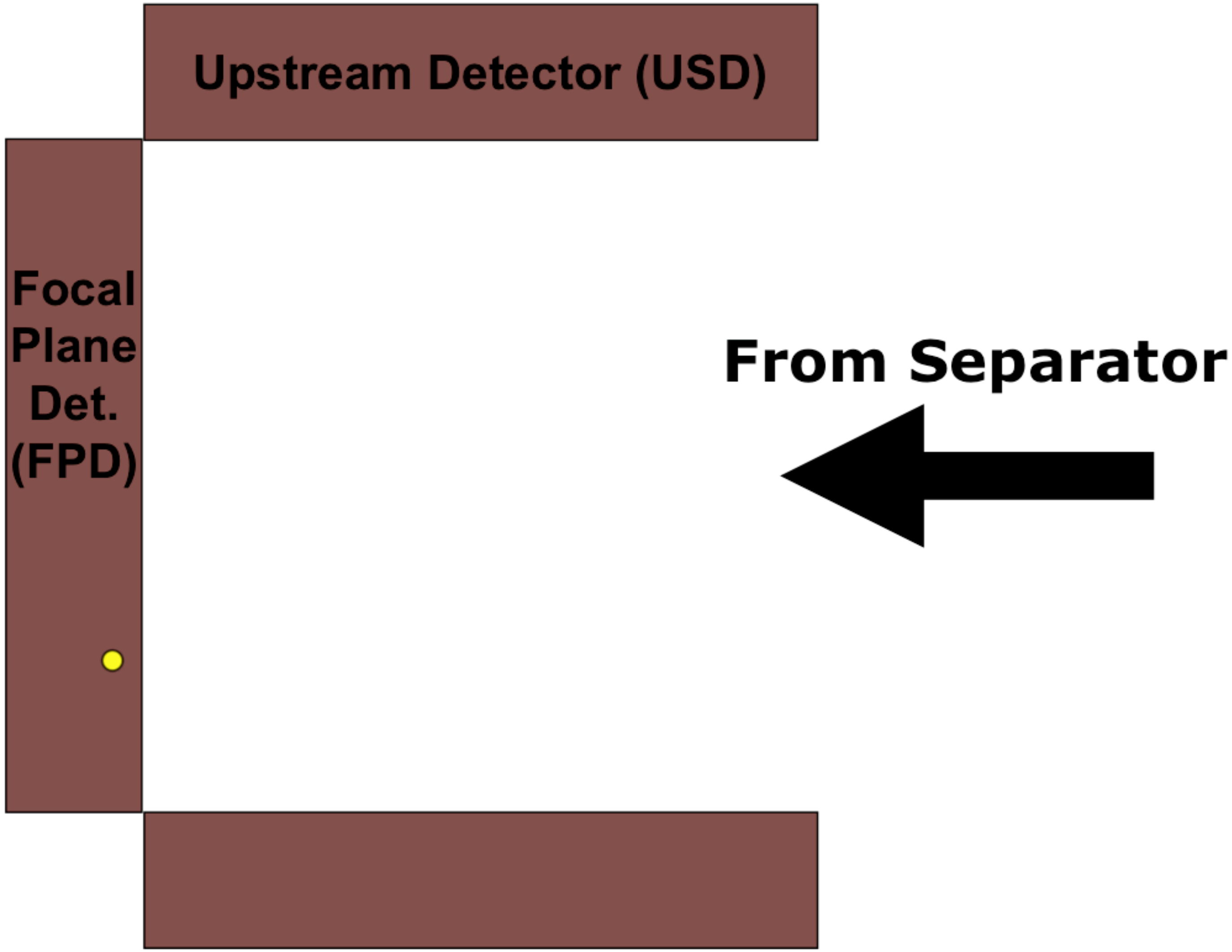


Vertical position determined by resistive charge division

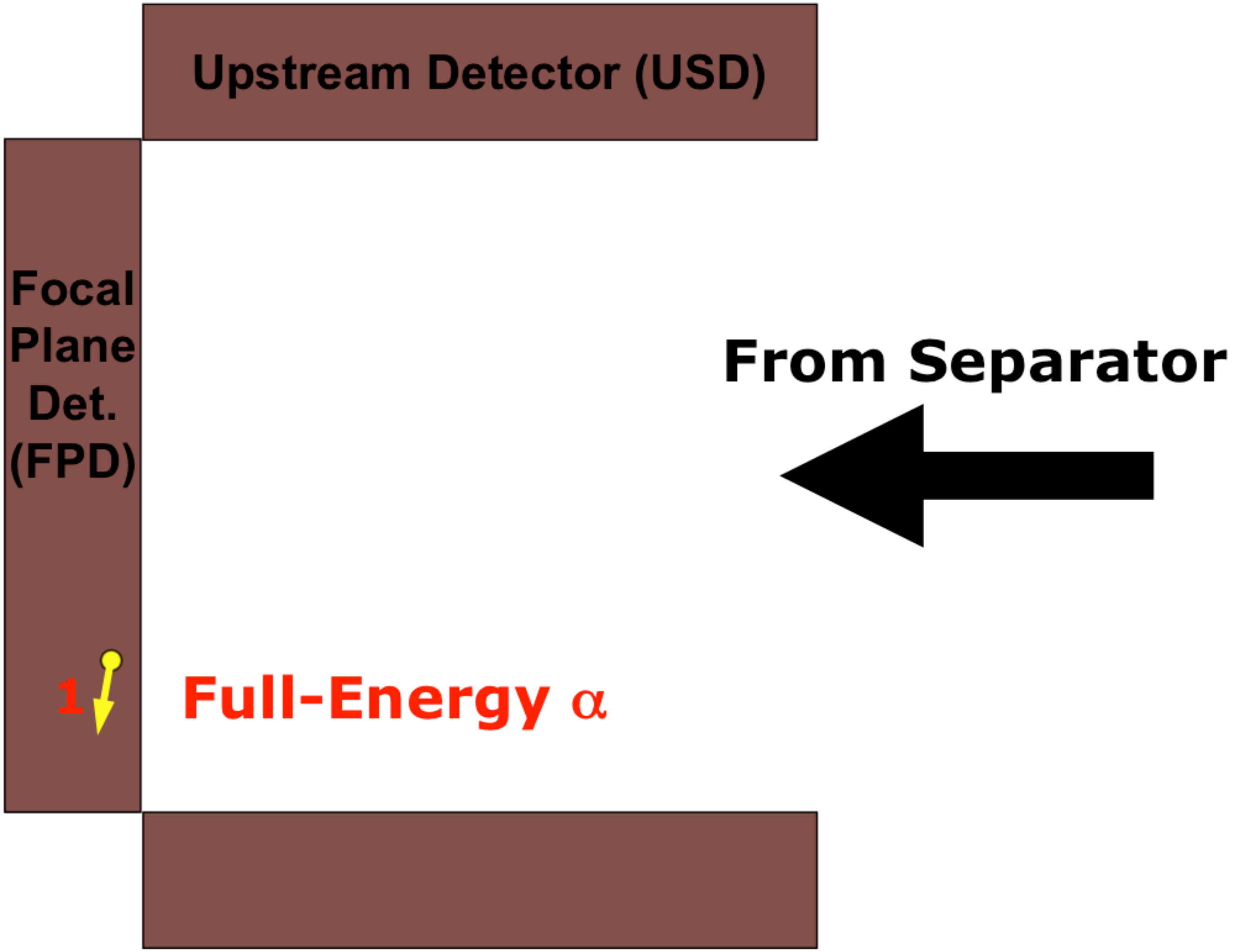
Particle Detection



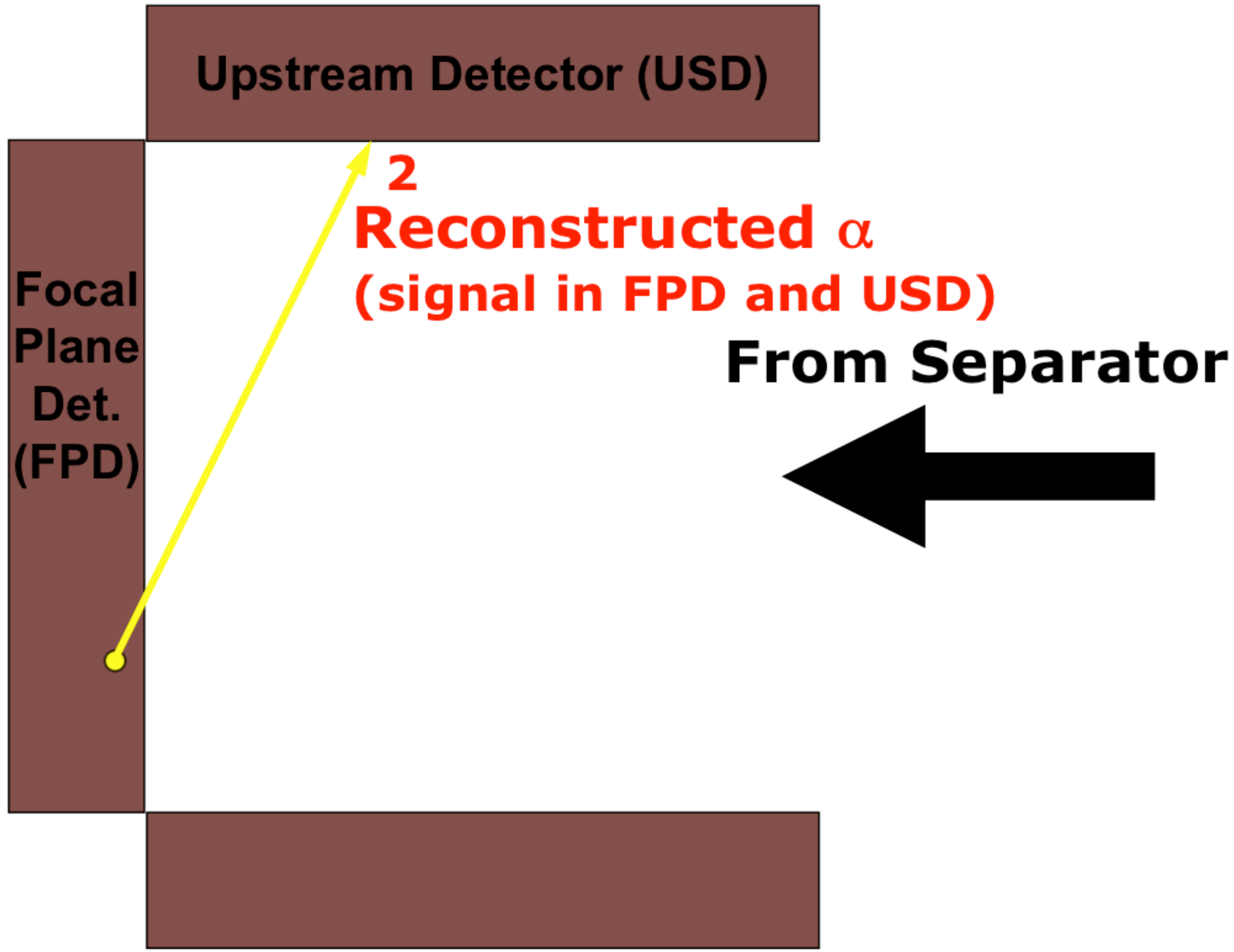
Particle Detection



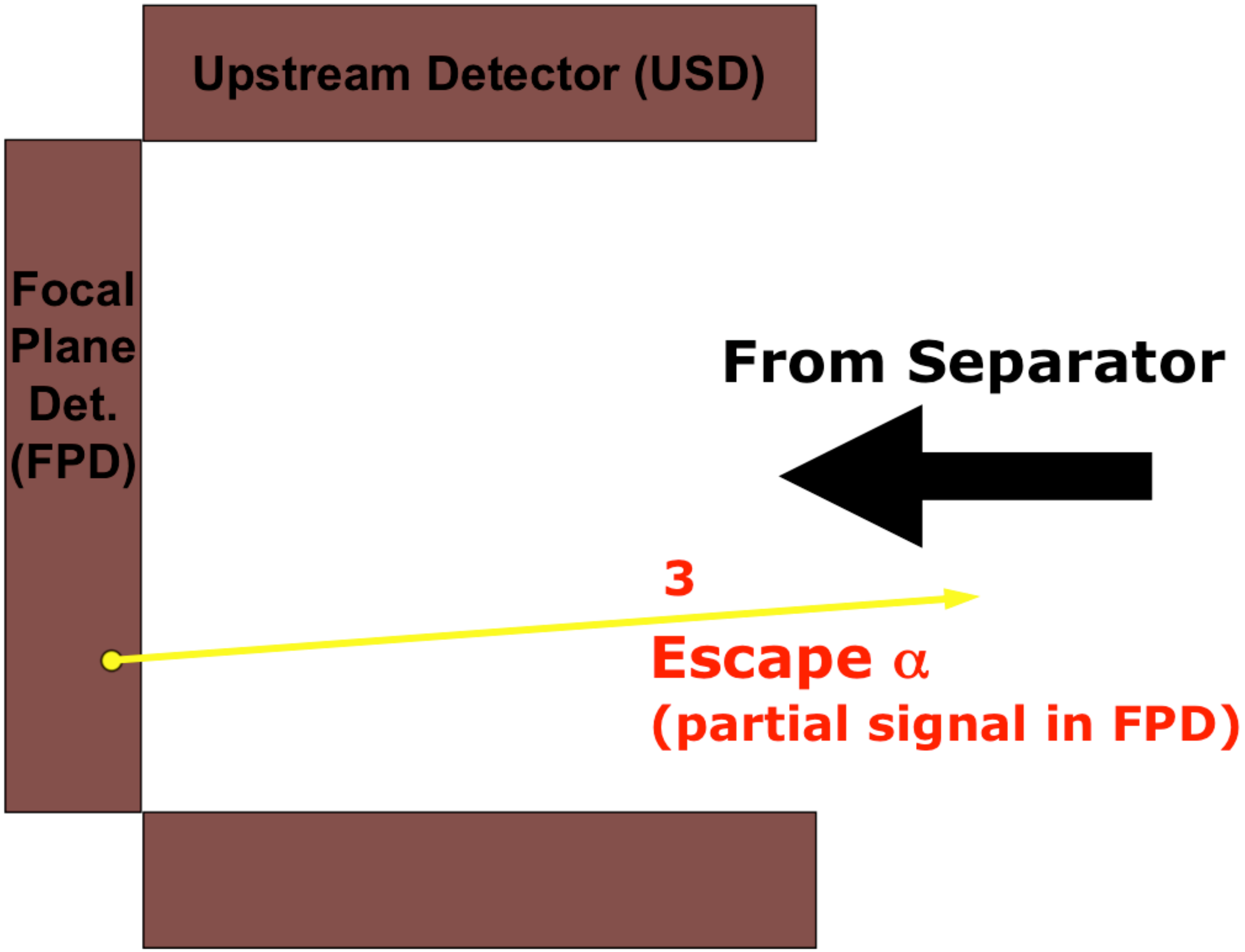
Particle Detection



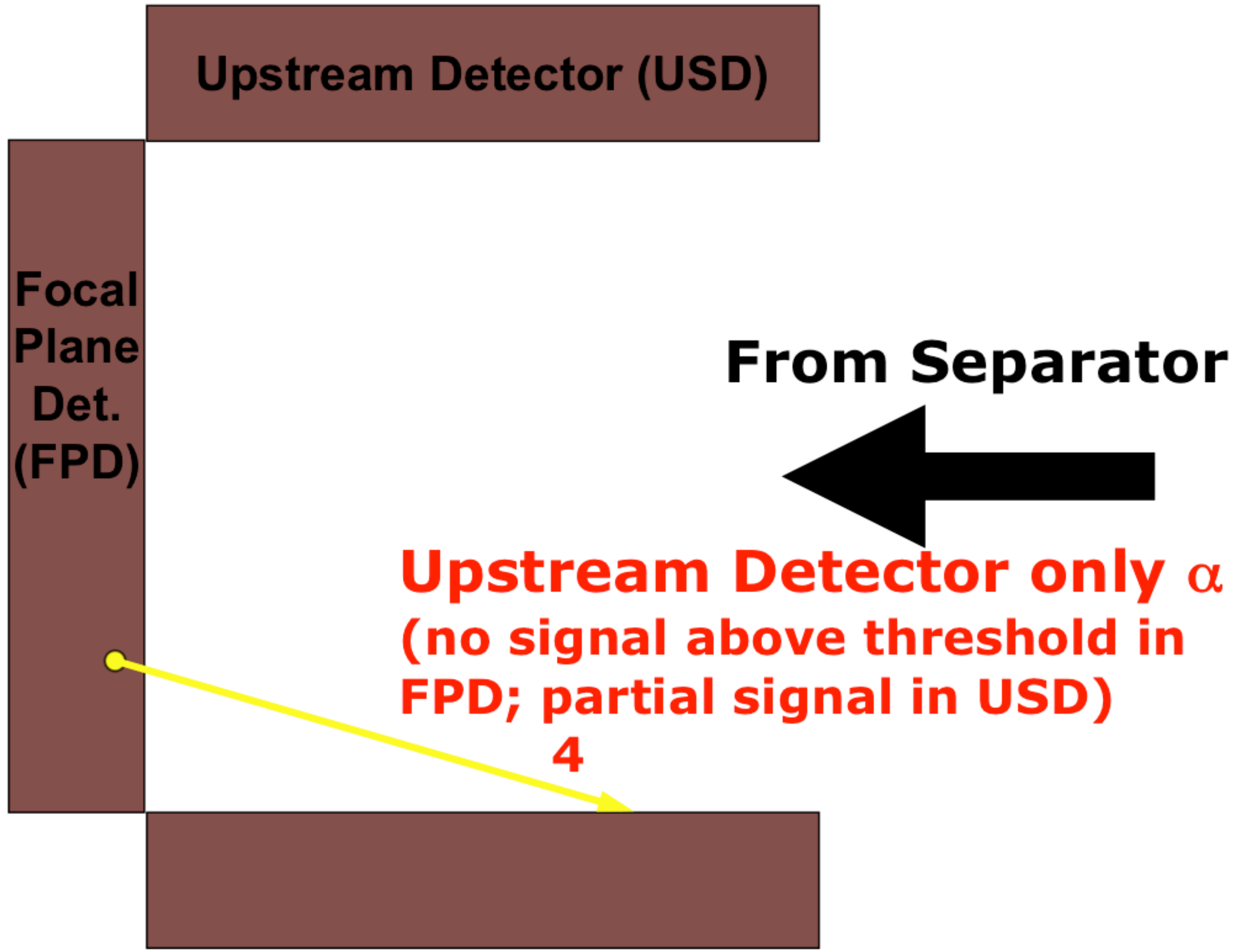
Particle Detection



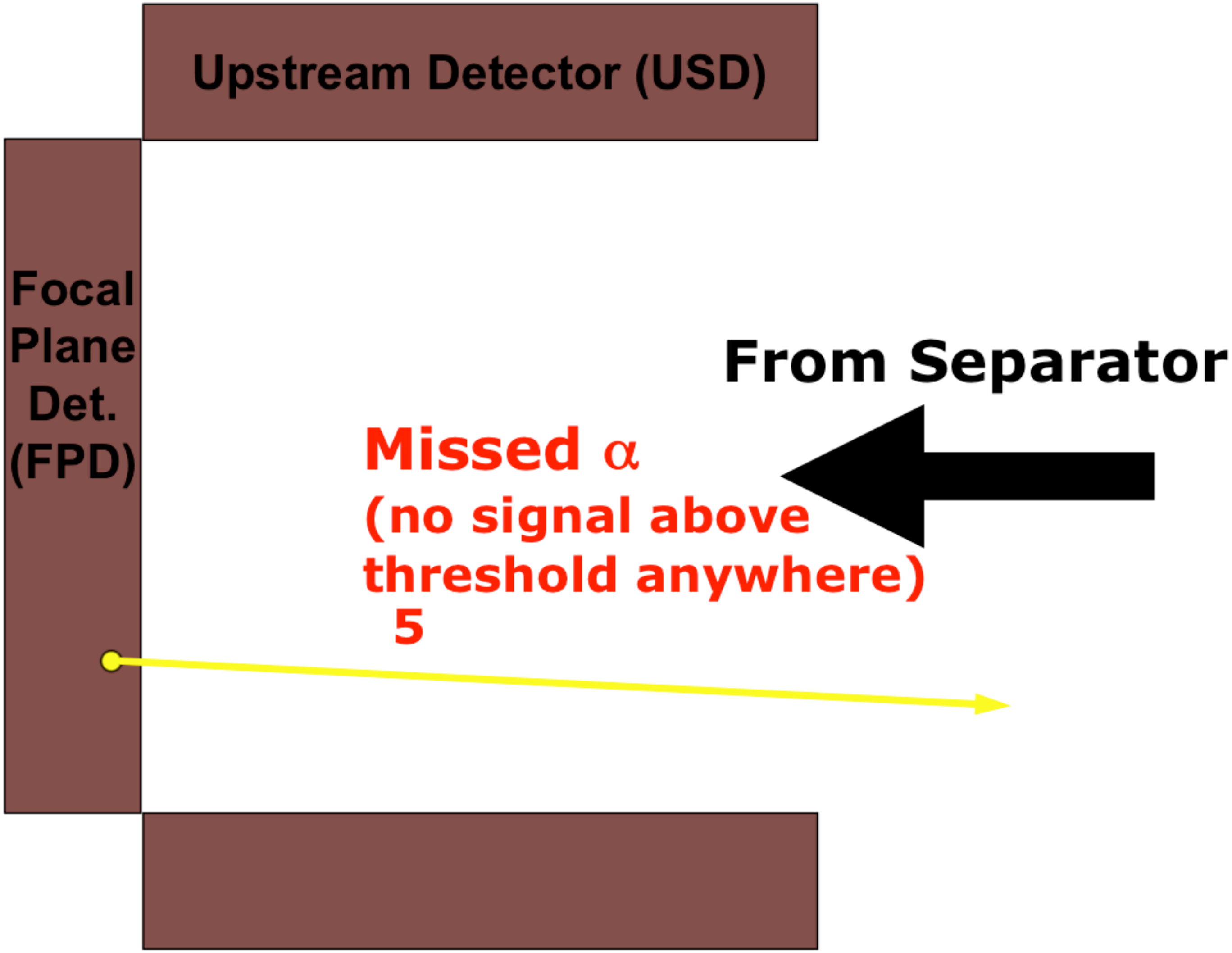
Particle Detection



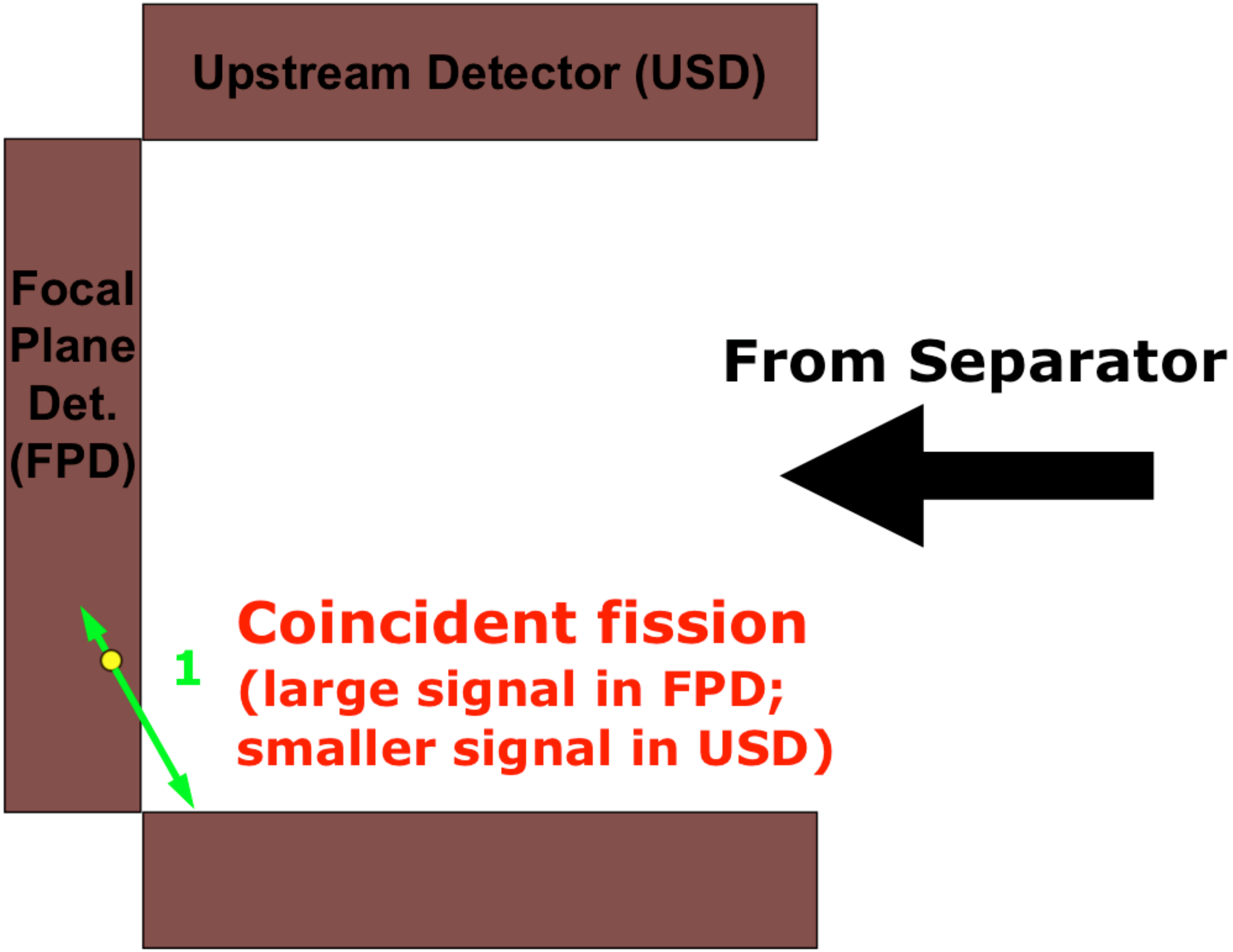
Particle Detection



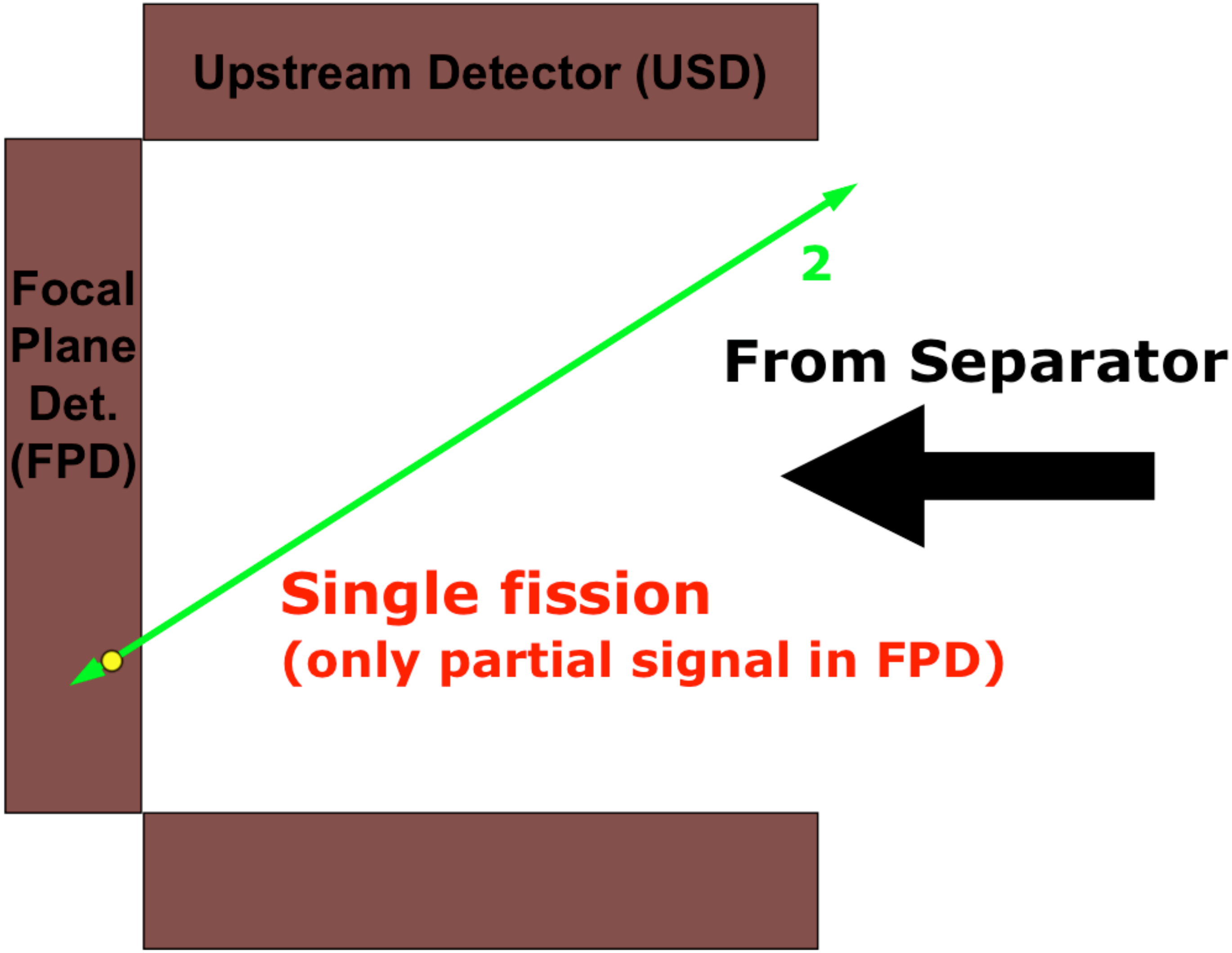
Particle Detection



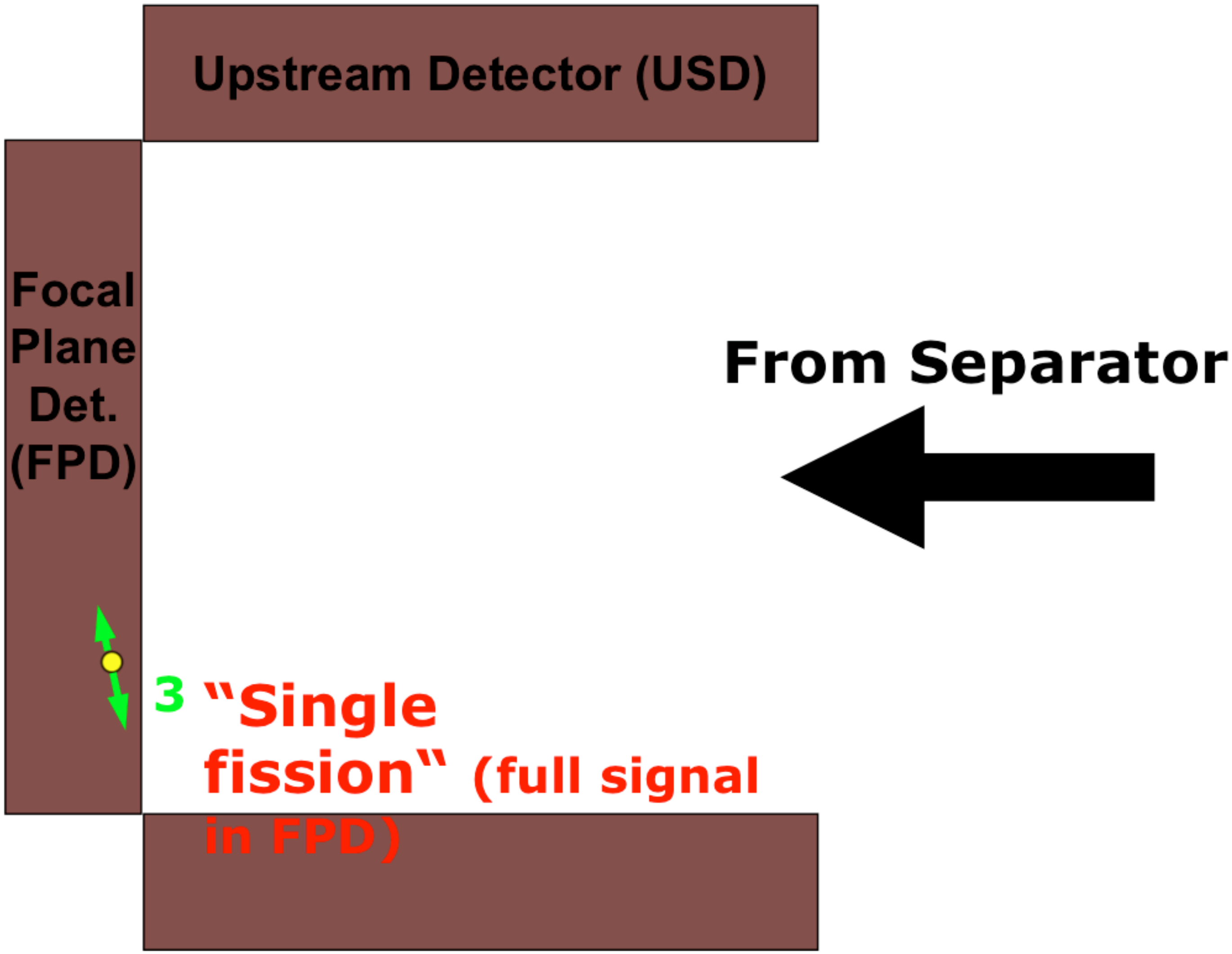
Particle Detection



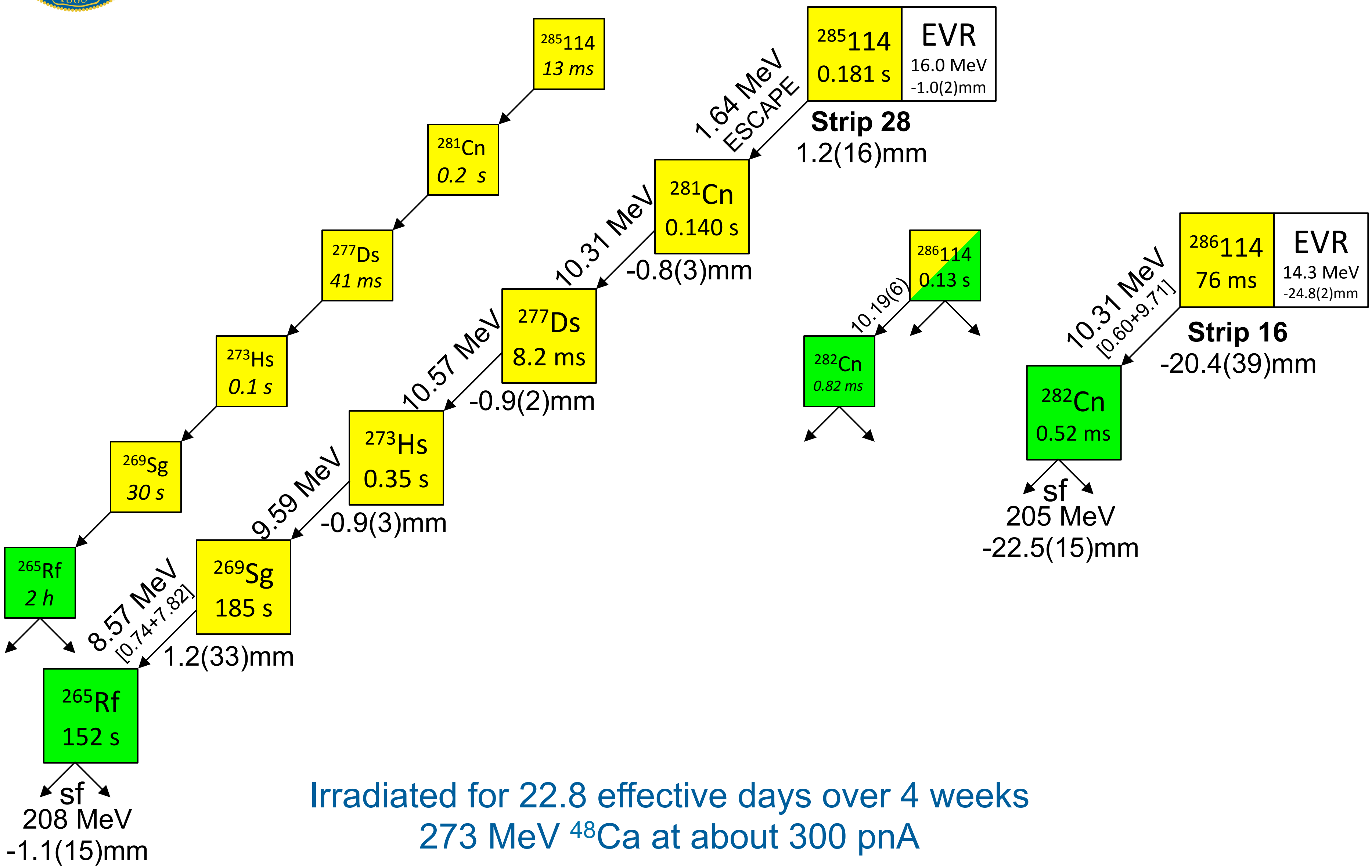
Particle Detection



Particle Detection

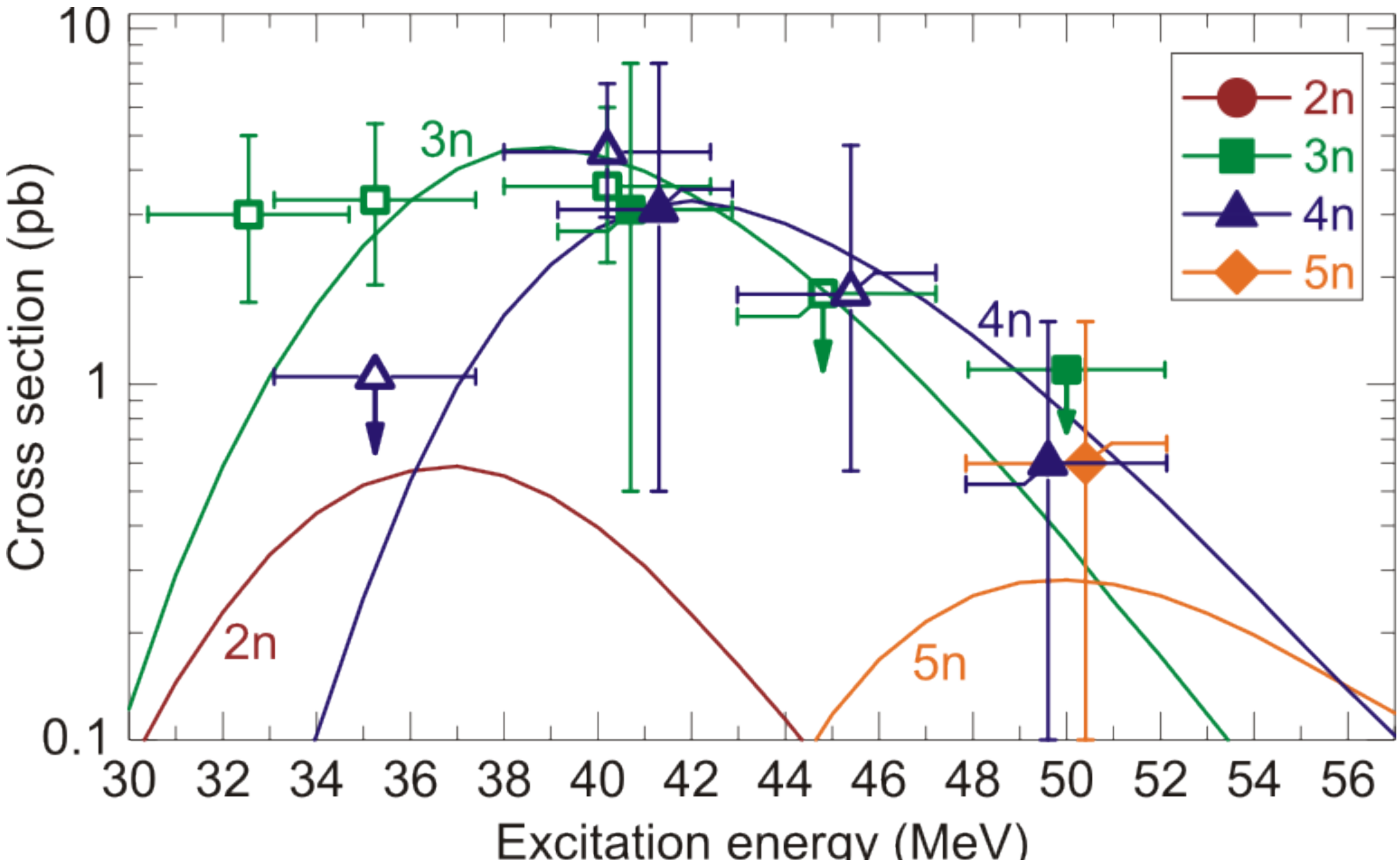


^{286}114 and predicted ^{285}114 decay properties



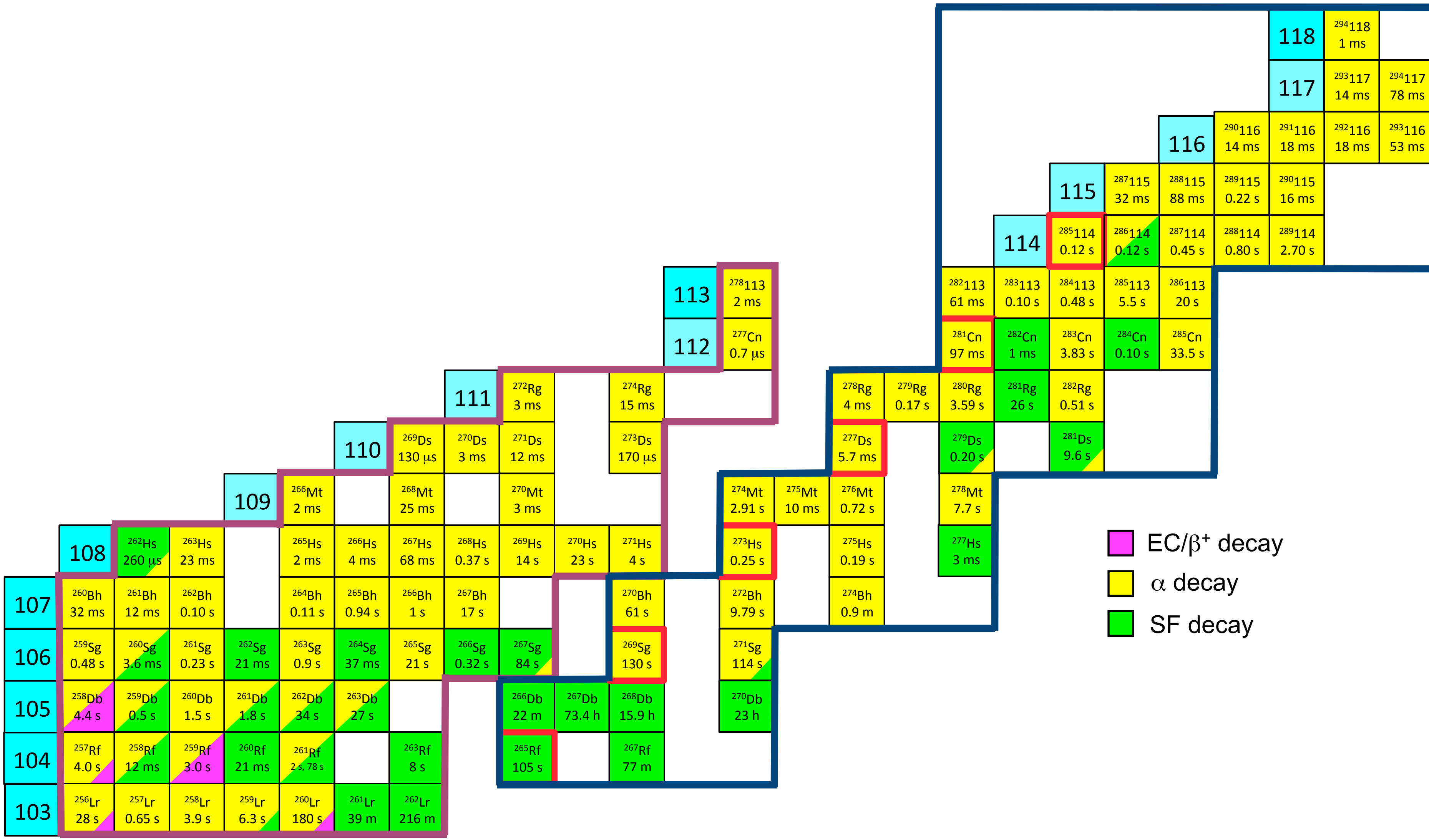
Irradiated for 22.8 effective days over 4 weeks
273 MeV ^{48}Ca at about 300 pnA

3n, 4n, and 5n cross sections



V.I. Zagrebaev, Nucl. Phys. **A734**, 164 (2004).
V.I. Zagrebaev (private communication).

Upper end of the nuclide chart



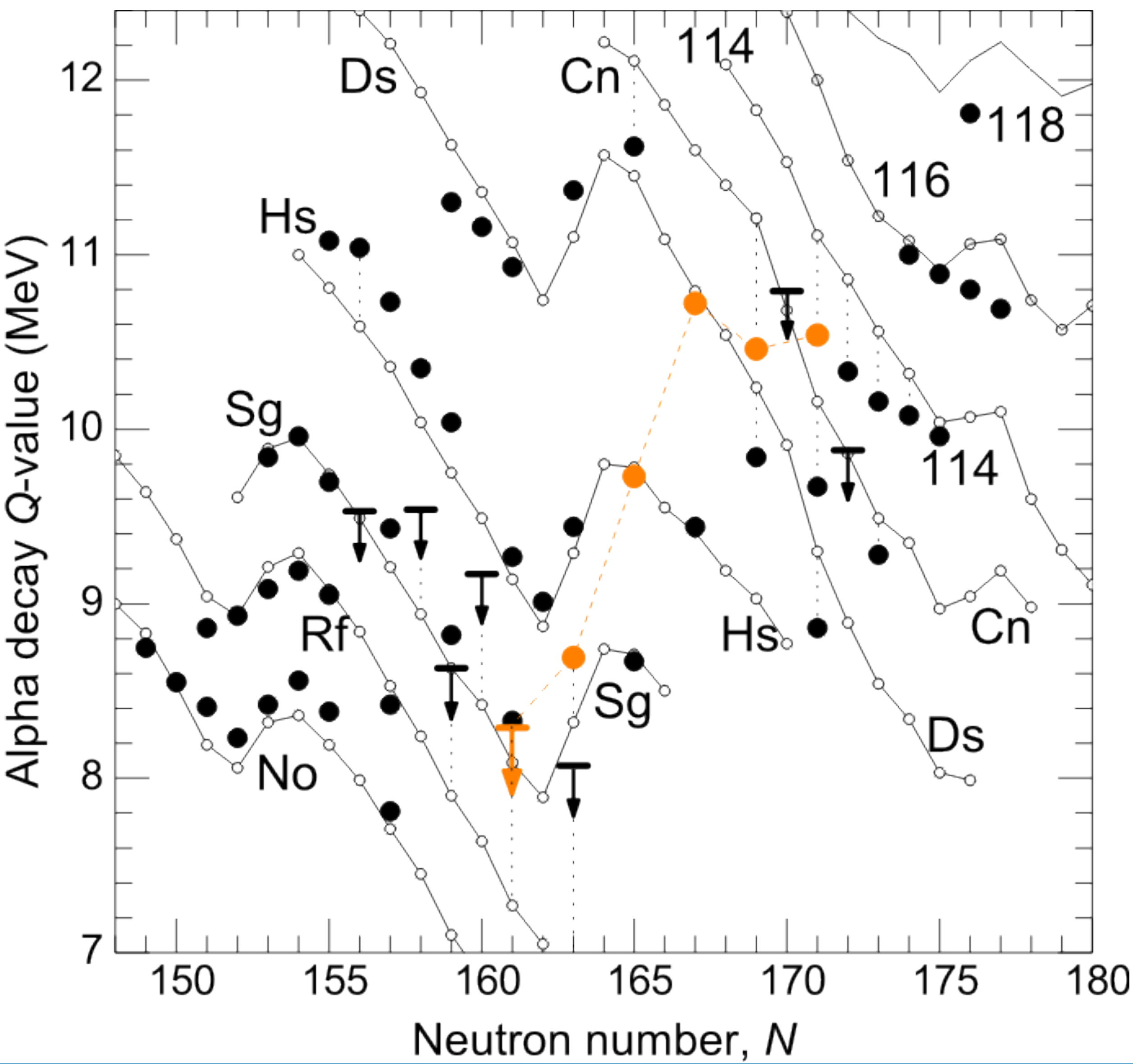
Summary

- Low energy nuclear reaction studies
- ^{242}Pu target production
- **Part I:** Production of ^{240}Am
  - — Measured cross section for ^{242}Pu($p,3n$)^{240}Am reaction
- **Part II:** Production of ^{285}114
  - — Discovery of six new superheavy element isotopes

Thank you for your attention

BERKELEY, CALIFORNIA

Even-Z experimental / theoretical Q_{alpha} values

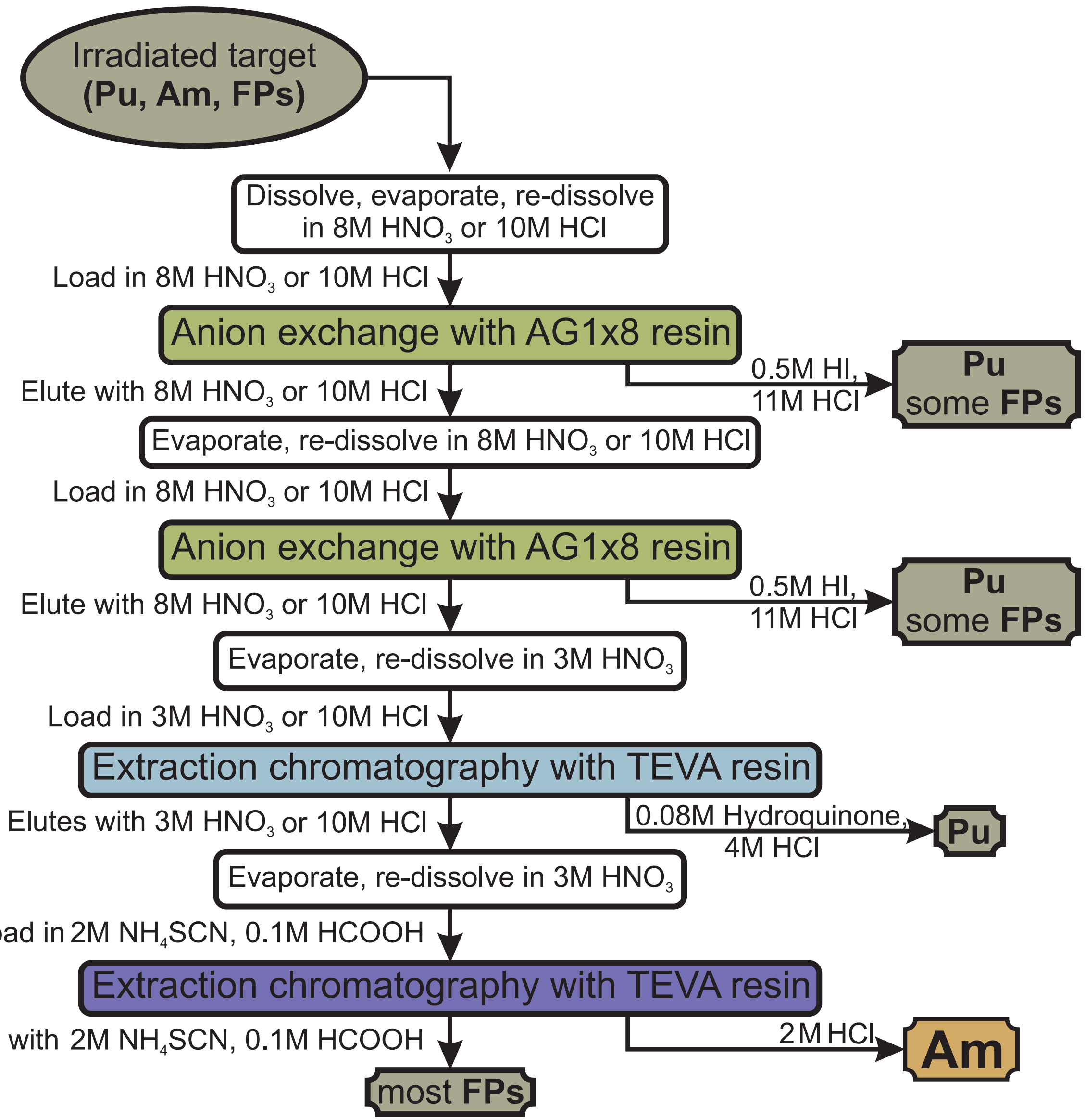


Purification of ^{240}Am

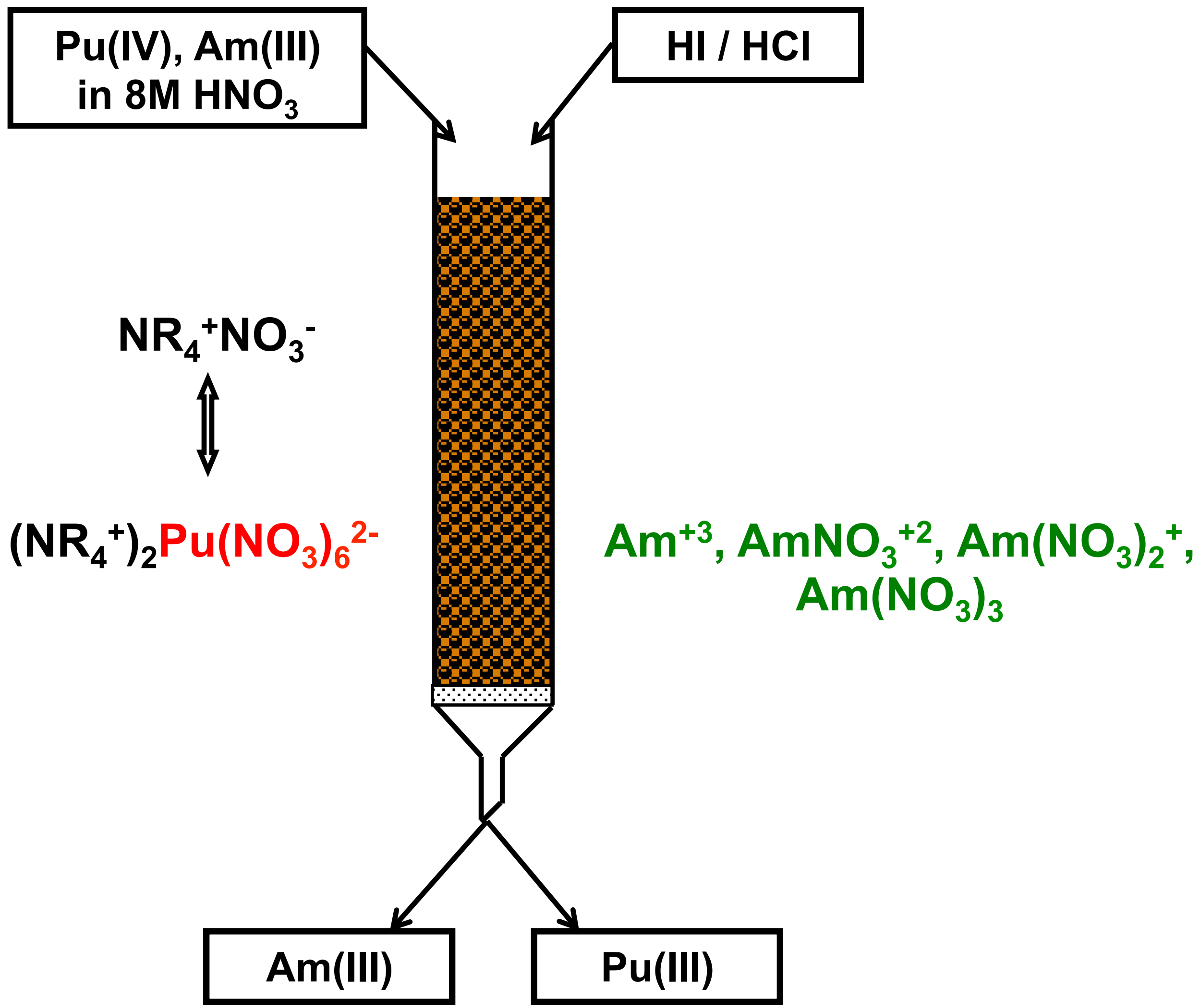
- Irradiated plutonium target will consist of
  - — ~1 g ^{242}Pu + ~ 50-100 ng ^{240}Am
  - — Products of ^{242}Pu(p,f) reaction: Z=38 (Sr) to Z=62 (Sm)
    - ^{97}Zr-^{97}Nb, ^{99}Mo-^{99m}Tc, ^{126}Sb, ^{132,133}I, ^{140}La
      - T. Ohtsuki *et al.*, *Phys. Rev. C* **40**, 2144 (1989).

- challenges of separation
  - — High Pu/Am mass ratio in the target (separation factor ~ 10^8)
  - — Similar chemistry of Am and some FP (lanthanides)
  - — High radioactivity (~2 R/hr=20 mSv/hr at 30 cm 1 day after irr.)
  - — Time restriction ($T_{1/2}$ of ^{240}Am is 50.8 h)

Am/Pu/FP separation summary



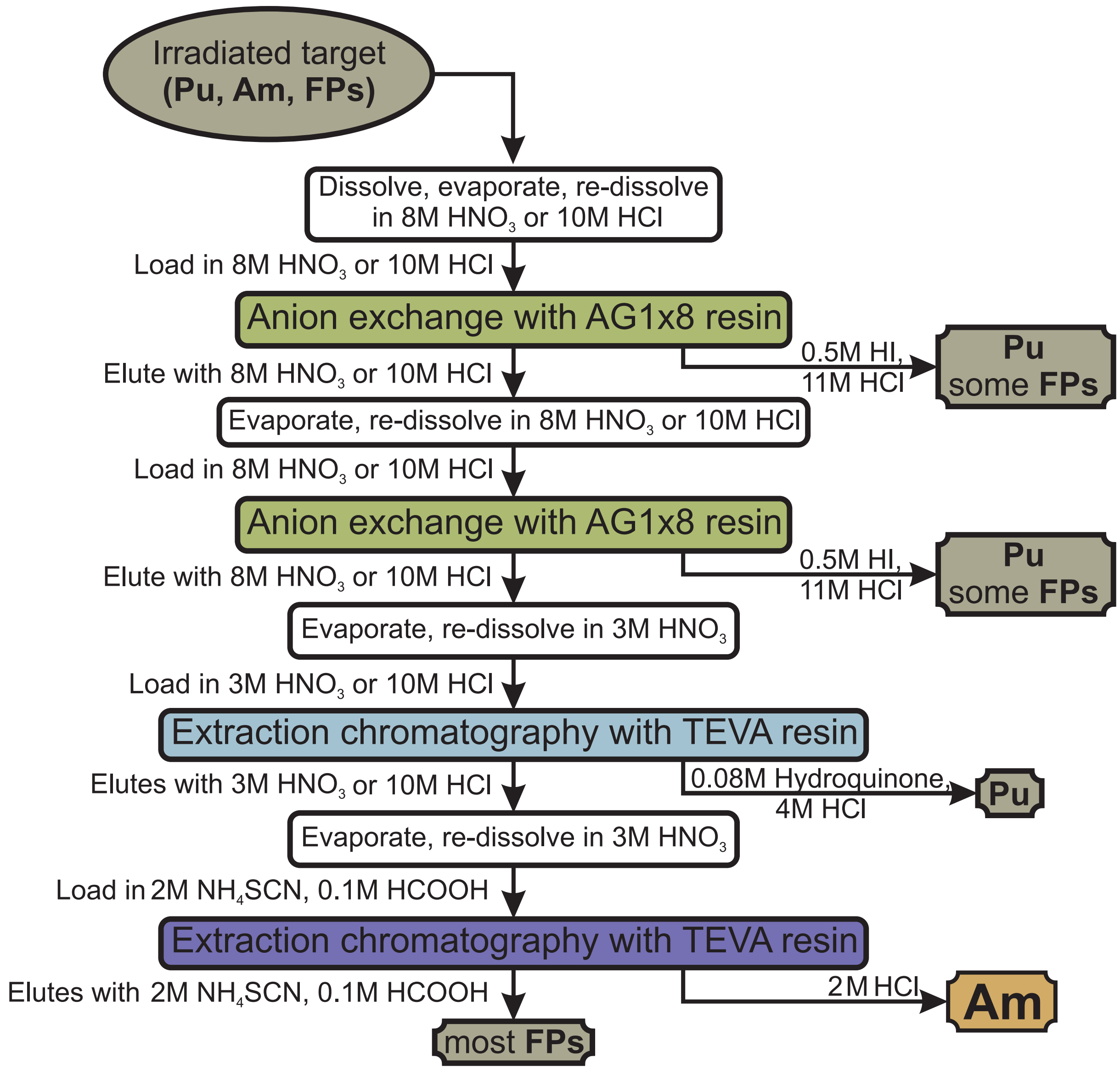
Anion Exchange Chromatography



Anion Exchange Chromatography

Pu complexed to column, Am being eluted

Am/Pu/FP separation summary

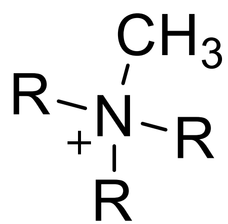


TEVA Liquid Extraction Chromatography

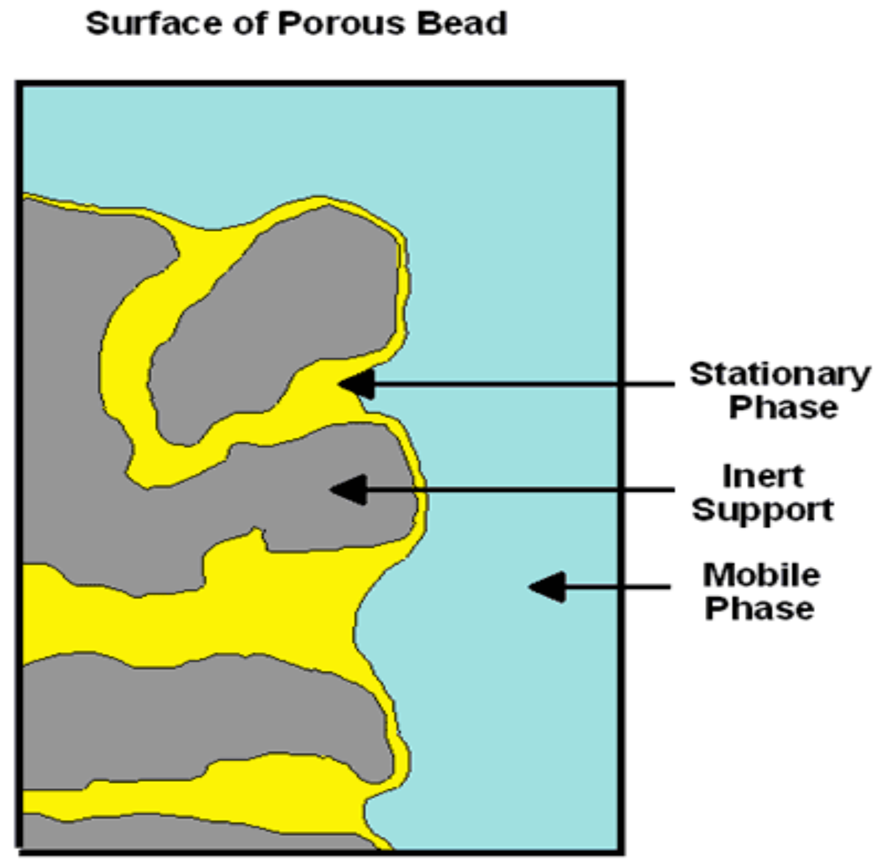
TEVA extractant:

Quaternary ammonium salt

(liquid anion exchanger)



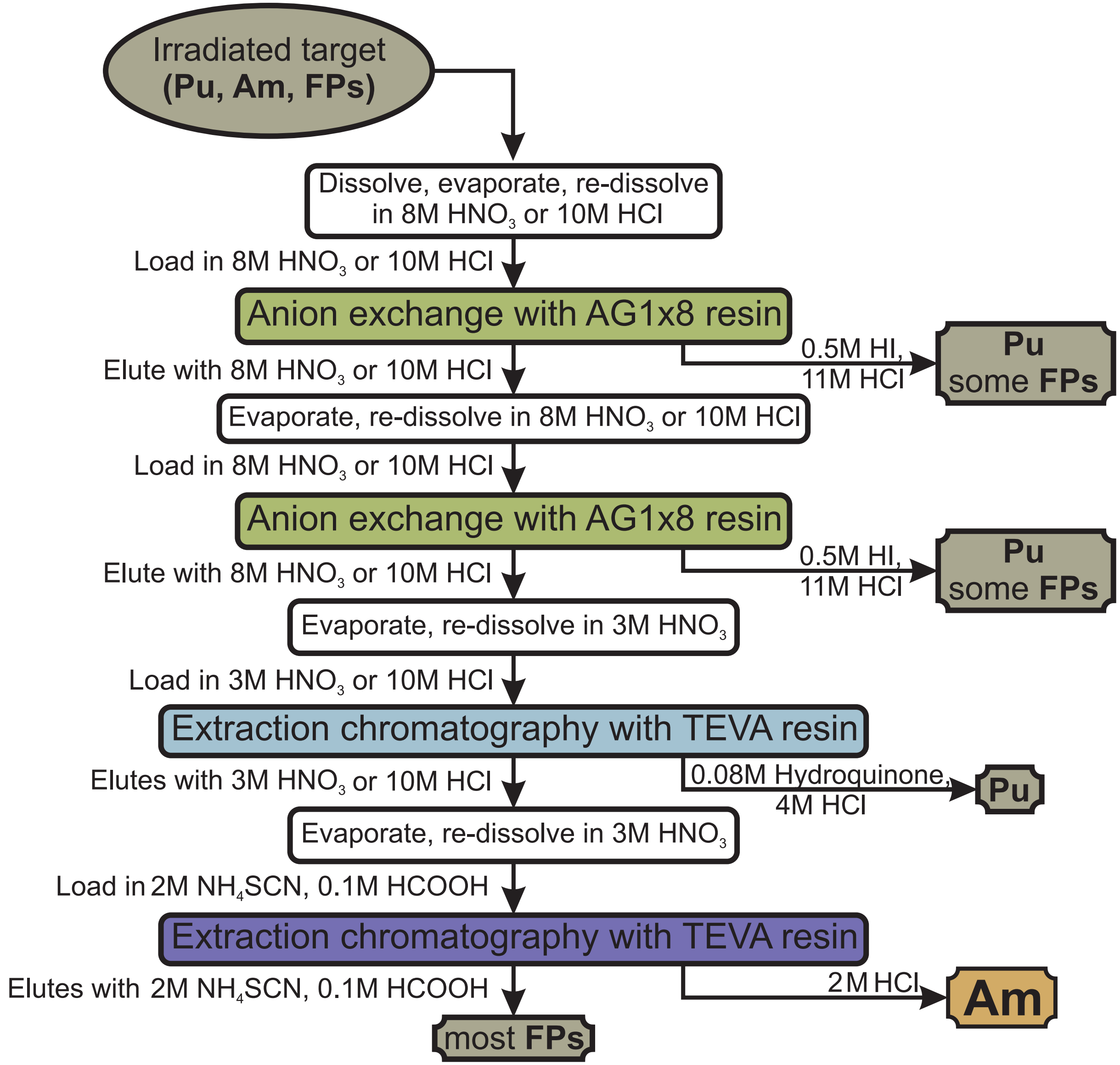
R=C_8H_{17} and $C_{10}H_{21}$



$$\mathbf{Pu^{4+} + 4NO^{3-} + 2\ E{\bullet}NO_3 \Longleftrightarrow E_2^{+}{\bullet}Pu(NO_3)_6^{2-}}$$

(E=Extractant)

Am/Pu/FP separation summary

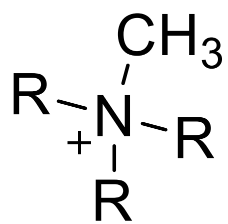


TEVA Liquid Extraction Chromatography

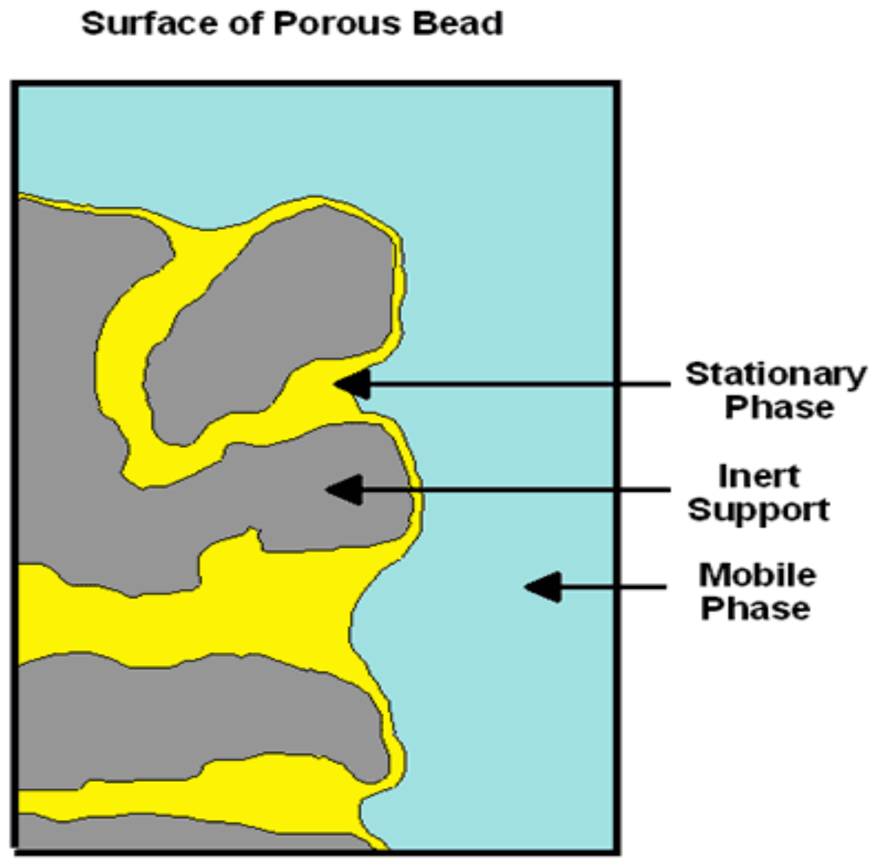
TEVA extractant:

Quaternary ammonium salt

(liquid anion exchanger)



$R = C_8H_{17}$ and $C_{10}H_{21}$

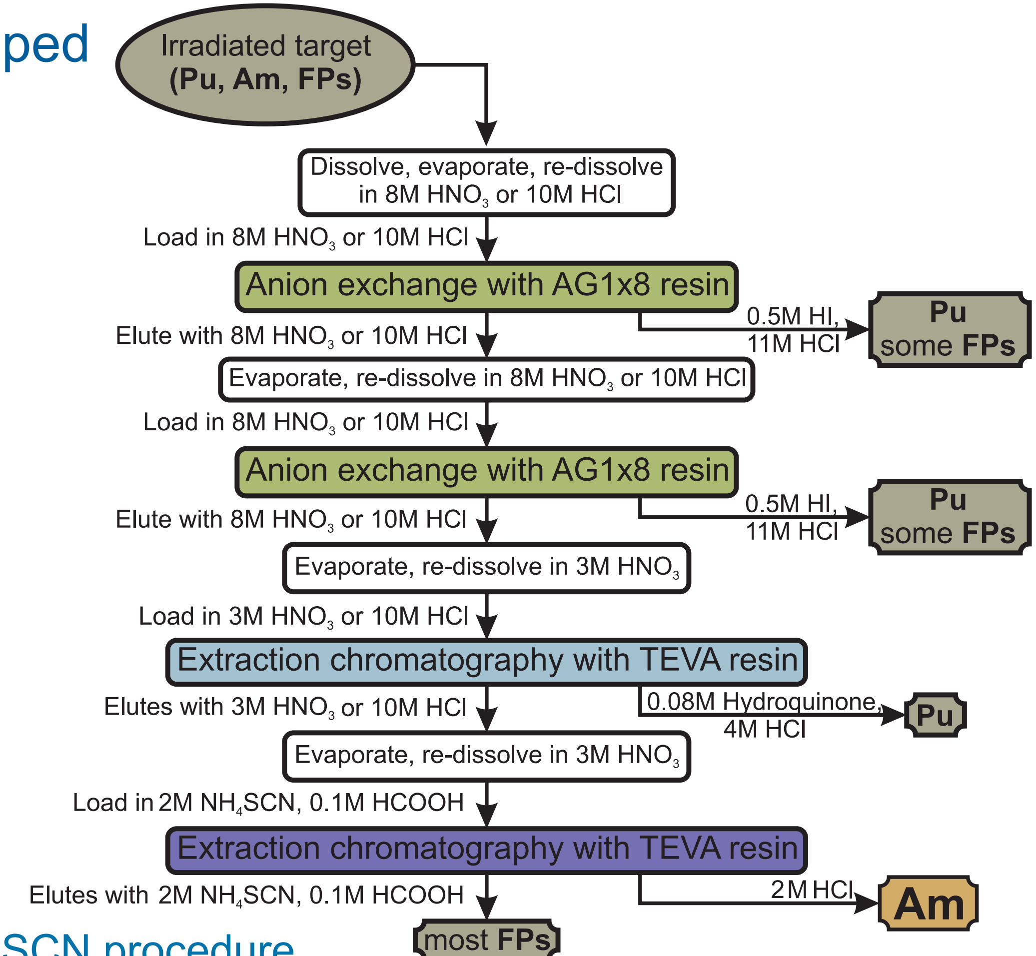


$$Am^{3+} + 4SCN^{-} + 2\ E{\bullet}SCN \Longleftrightarrow E_2^{+}{\bullet}Am(SCN)_4^{-}$$

(E=Extractant)

Am/Pu/FP separation summary

- Separation procedure developed and tested on tracer scale
  - — ^{241}Am, ^{239}Pu, ^{95}Zr, ^{95}Nb, ^{152}Eu



- Procedure takes ~1 day

  Am yield: ~70%

  Am/Pu

  separation factor: >2 x 10^7

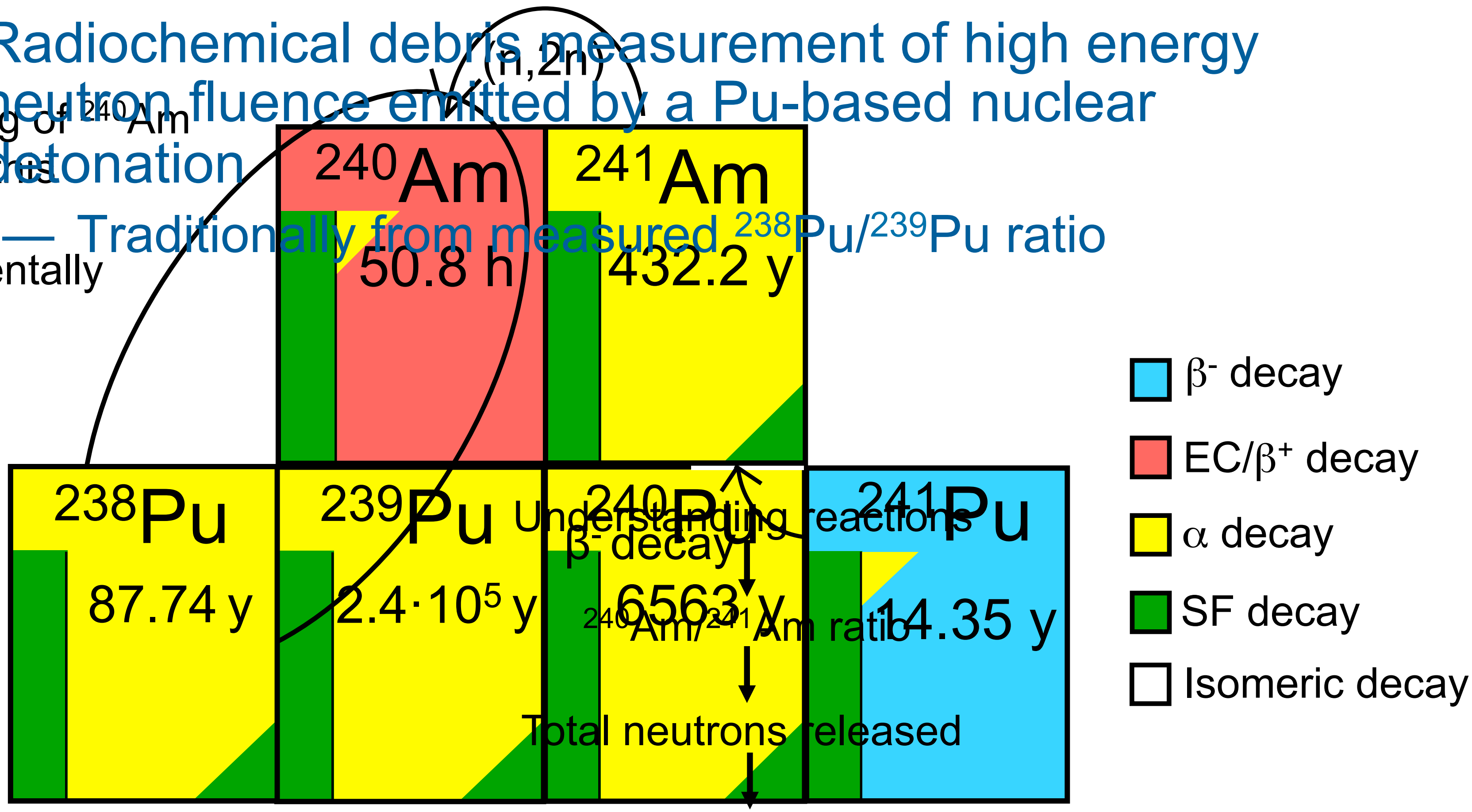
- **Eu** separated from Am in TEVA/NH_4SCN procedure
- **Nb** separated from Am in anion exchange and TEVA/NH_4SCN procedure
- **Zr** separated partially from in part in anion exchange and TEVA/NH_4SCN

  Difficulties due to formation of polynuclear hydrolysis products

Why ^{240}Am?

- Radiochemical debris measurement of high energy neutron fluence emitted by a Pu-based nuclear detonation
  - Traditionally from measured $^{238}Pu/^{239}Pu$ ratio

10-100 ng of ^{240}Am to study this reaction experimentally



Understanding reactions

$^{240}Am/^{241}Am$ ratio

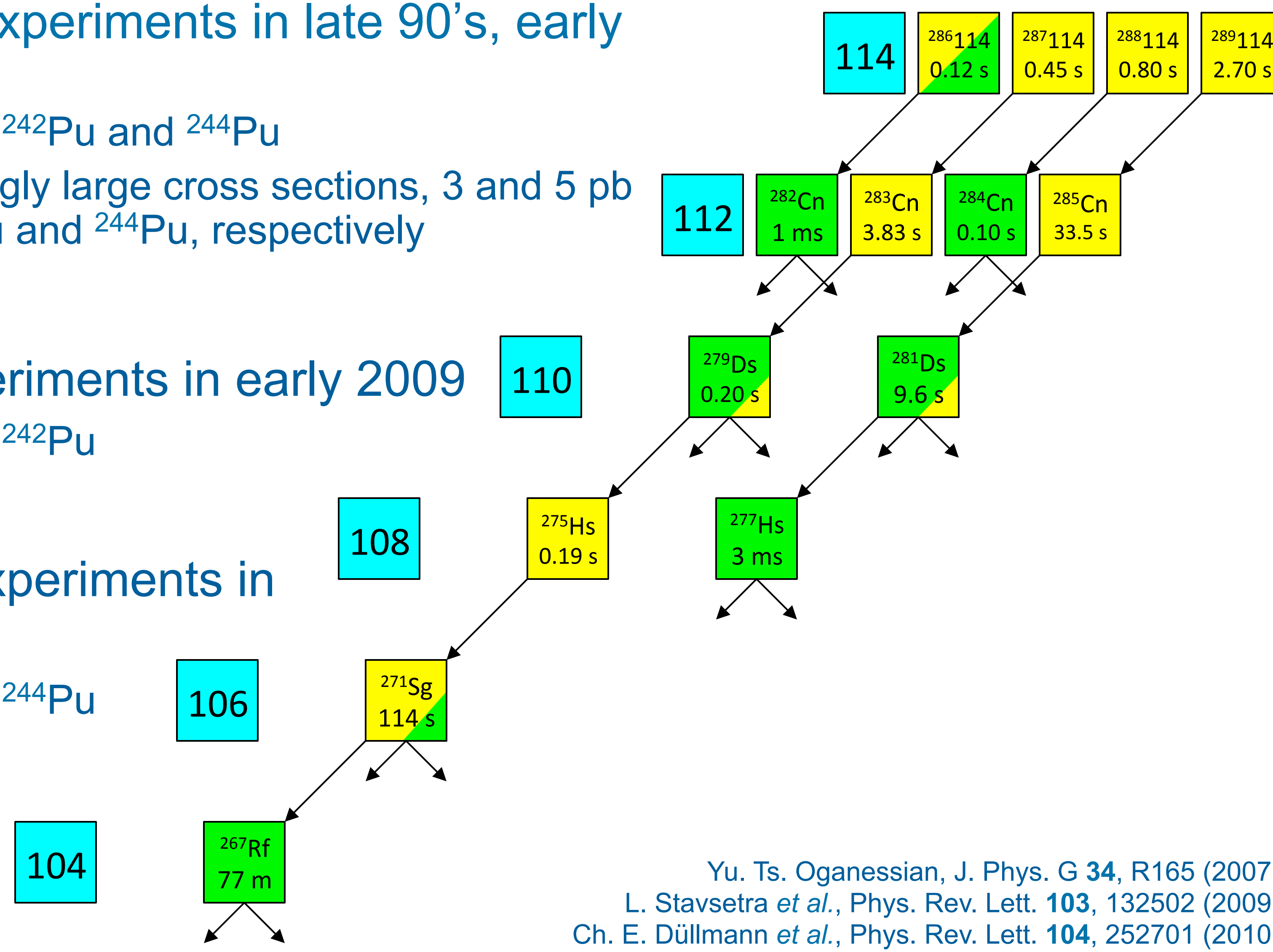
Total neutrons released

Isotopic composition and age of fuel

Information about the fuel's production location and date

Element 114 background

- DGFRS experiments in late 90’s, early 00’s
  - ^{48}Ca on ^{242}Pu and ^{244}Pu
  - Suprisingly large cross sections, 3 and 5 pb for ^{242}Pu and ^{244}Pu, respectively
- BGS experiments in early 2009
  - ^{48}Ca on ^{242}Pu
- TASCA experiments in mid 2009
  - ^{48}Ca on ^{244}Pu



Yu. Ts. Oganessian, J. Phys. G **34**, R165 (2007).
L. Stavsetra *et al.*, Phys. Rev. Lett. **103**, 132502 (2009).
Ch. E. Düllmann *et al.*, Phys. Rev. Lett. **104**, 252701 (2010).

Random rates

- Background rates (for whole array):
  - — EVR-like rate (5 < E (MeV) < 18): 0.38 Hz
  - — α-like rate (8 < E (MeV) < 12): 0.011 Hz
  - — SF-like (E (MeV) > 80): 9 events (only 3 with E(MeV) > 101)

- Defined "^{285}114-like" decay chain as:
  - — EVR-like event, 3 α-like events, and an SF-like event
  - — Correlated within 20 hours
  - — One α-lifetime less than 3 seconds
  - — **9.2 x 10^{-4} such expected correlations**

- ^{286}114-like decay chain as:
  - — Within decay 10 half-lives of observed properties (~1.5 seconds)
  - — **1.7 x 10^{-9} such expected correlations**