Results and Analysis of ^{232}Th Spallation Effort to Produce ^{225}Ac

D. R. McAlister[1], E. P. Horwtiz[1], and J.T. Harvey[2]

[1]PG Research Foundation/Eichrom Technologies, Inc. Lisle, IL
[2]Northstar Medical Radioisotopes, Madison, WI

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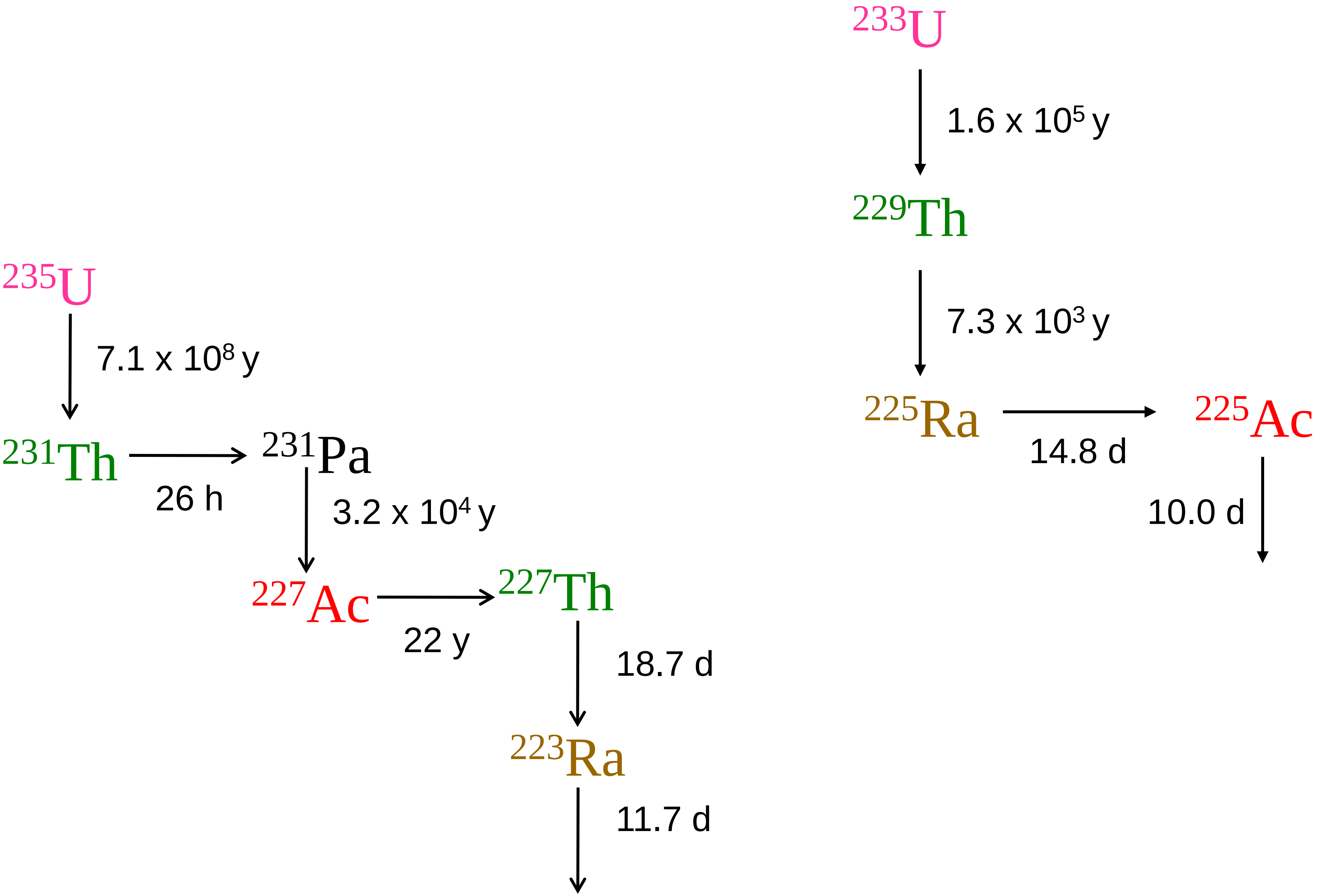
Sources of Actinium Isotopes



Ac-225 Sources

ORNL-150mCi Th-229 (on-going; ~600mCi Ac-225 annually)

INL-27MT LWBR fuel; ^{232}Th/^{233}U(~5000mCi/month Ac-225)

Chemical Separation of Th-229 from existing U-233 stocks (~6000mCi/month Ac-225)

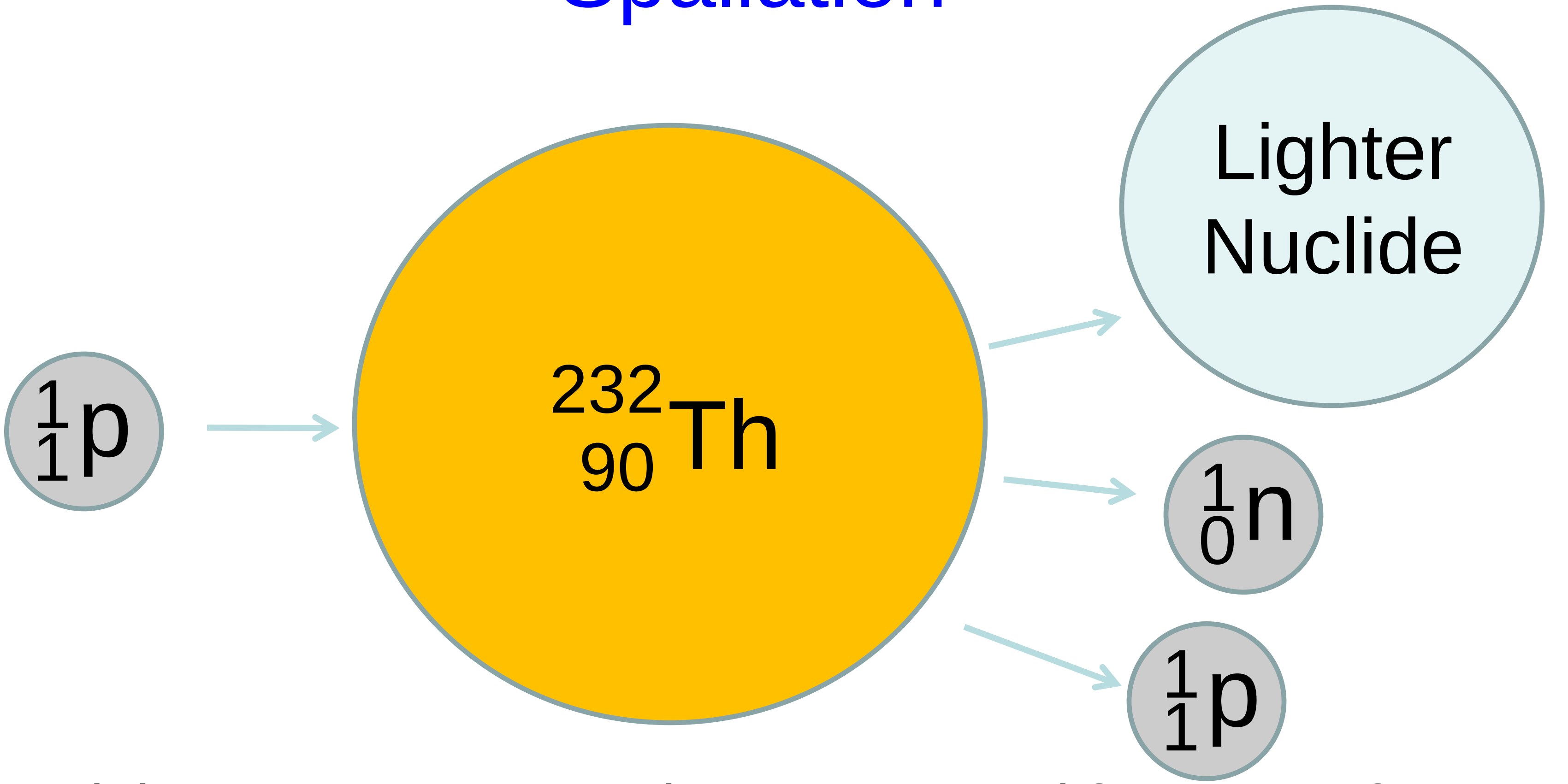
Cyclotron Production via Ra-226(p,2n)Ac-225 (~200mCi/month/cyclotron)

Photonuclear transmutation via Ra-226(γ,n)Ra-225→Ac-225 (~400mCi/month/LINAC)

Reactor production of Th-229; Ra-226→Th-229 or Th-228(n,γ)Th-229

High Energy Proton Spallation of Th-232 (~10,000mCi/month)

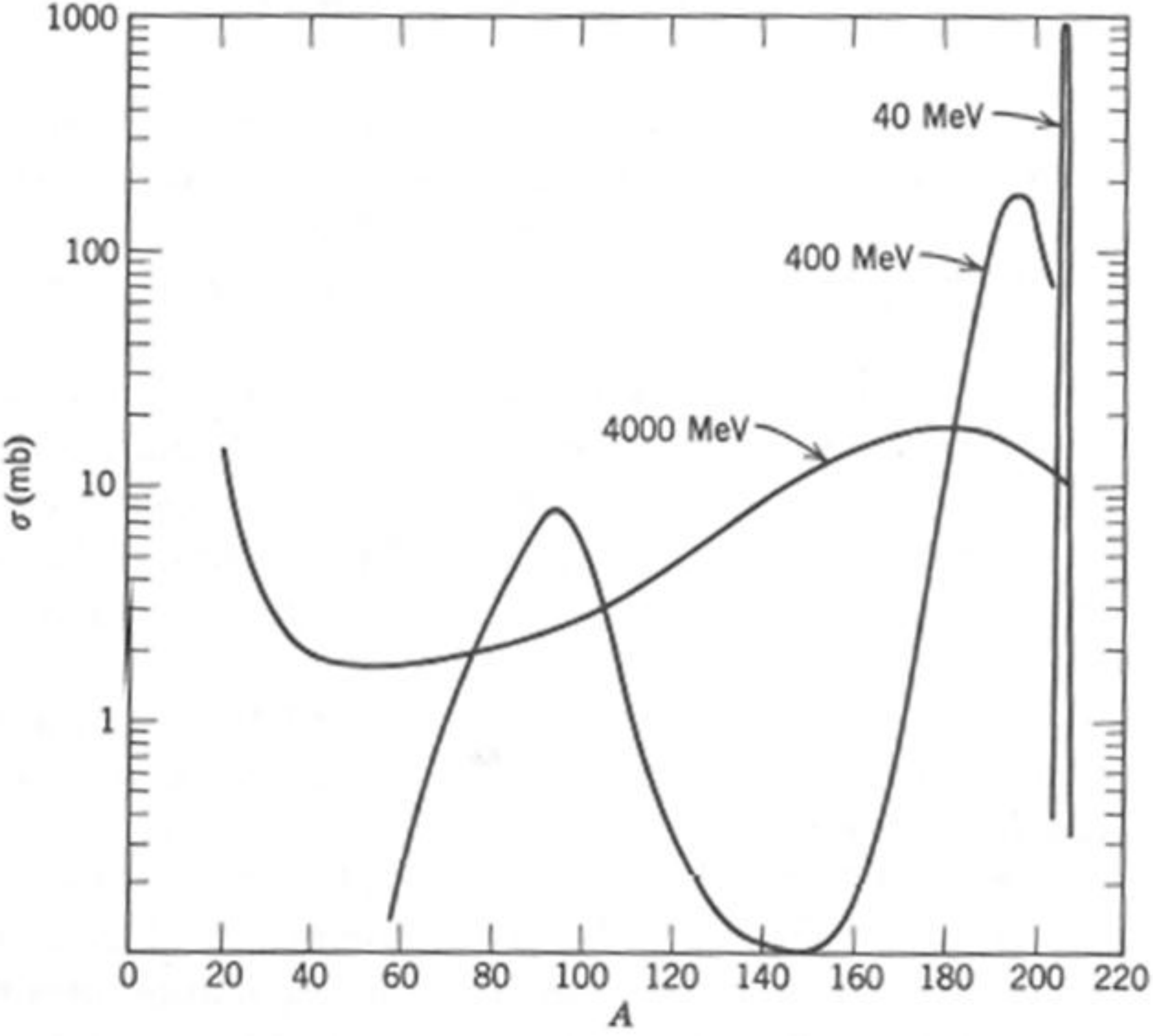
Spallation



High energy protons strip neutrons and fragments from thorium forming lighter nuclides.

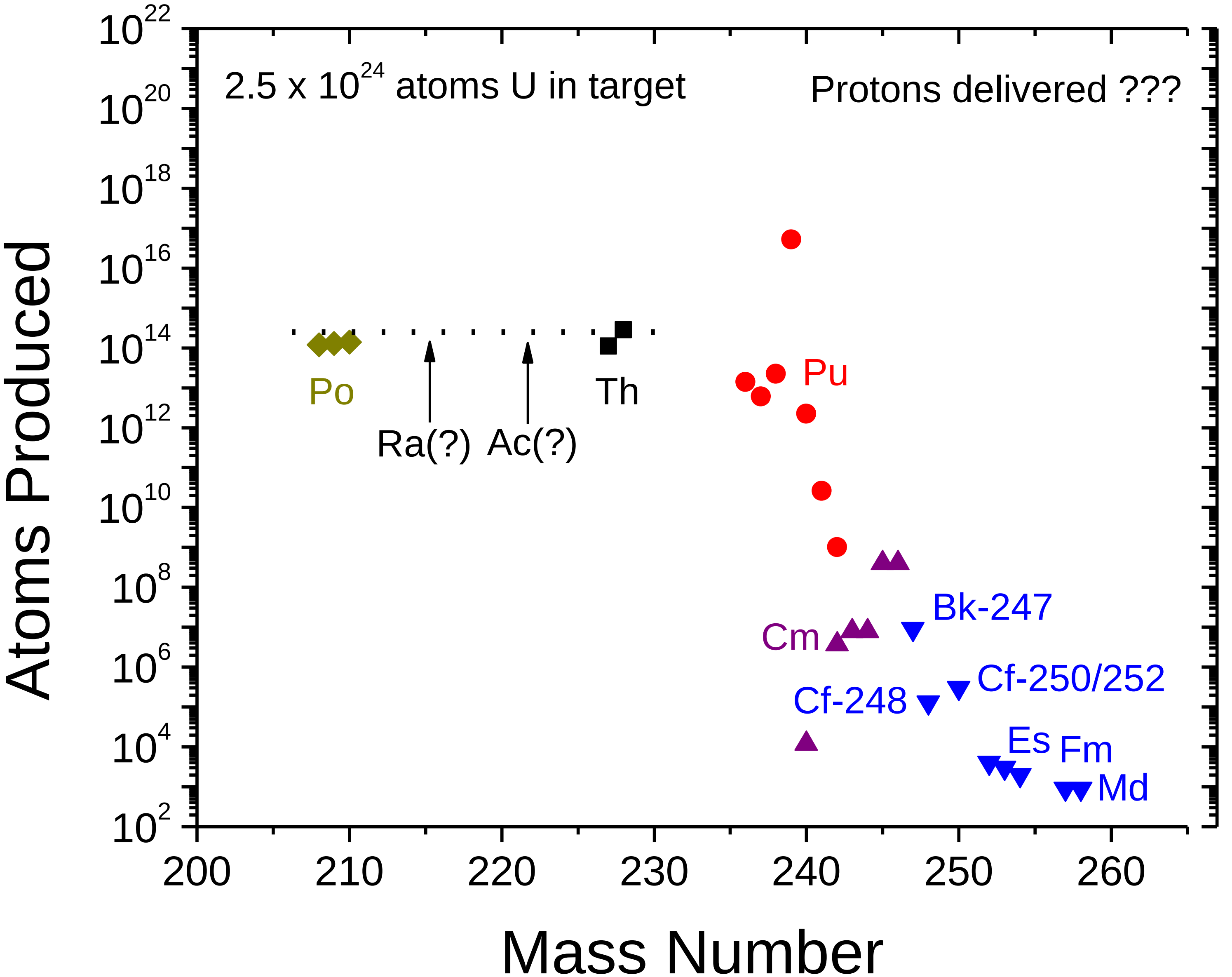
Fragments can also combine with thorium to form heavier nuclides.

Mass Distribution for Reaction of Protons with ^{209}Bi



Friedlander, G.; et al. Nuclear and Radiochemistry. 3rd Ed. John Wiley and Sons, New York, 1981, p 172.

U beam-stop from ZG-Synchrotron (ANL), 12 GeV



Unik, J.P.; Horwitz, E.P.; et al.; Production of Actinides and the search for super-heavy elements using secondary reactions induced by GeV protons, Nucl. Phys. A191, 233-244 (1972)

Th-232 Spallation

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Pate, B. D.; Poskanzer, A. M. Spallation of Uranium and Thorium Nuclei with BeV-Energy Protons, *Physical Review*, **123(2)**, 647-654 (1961).

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Hahn, R.L.; Bertini, H.W.; Calculations of Spallation-Fission Competition of Protons with Heavy Elements at Energies <3 GeV *Phys. Rev. C.* **6(2)**, 660-669 (1972).

Th-232 Spallation

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Harvey, J.; et al.; Production of ^{225}Ac via high energy proton induced spallation of ^{232}Th, Proceedings of Application of high energy proton accelerators, Fermilab, Chicago, IL, October 19-21, 2009, eds. Rajendran Raja and Shekhar Mishra, pp. 321-326.

"Production of ^{225}Ac via high energy proton induced spallation of ^{232}Th," DOE/SC0003602-1, (2011). http://www.osti.gov/scitech/biblio/1032445.

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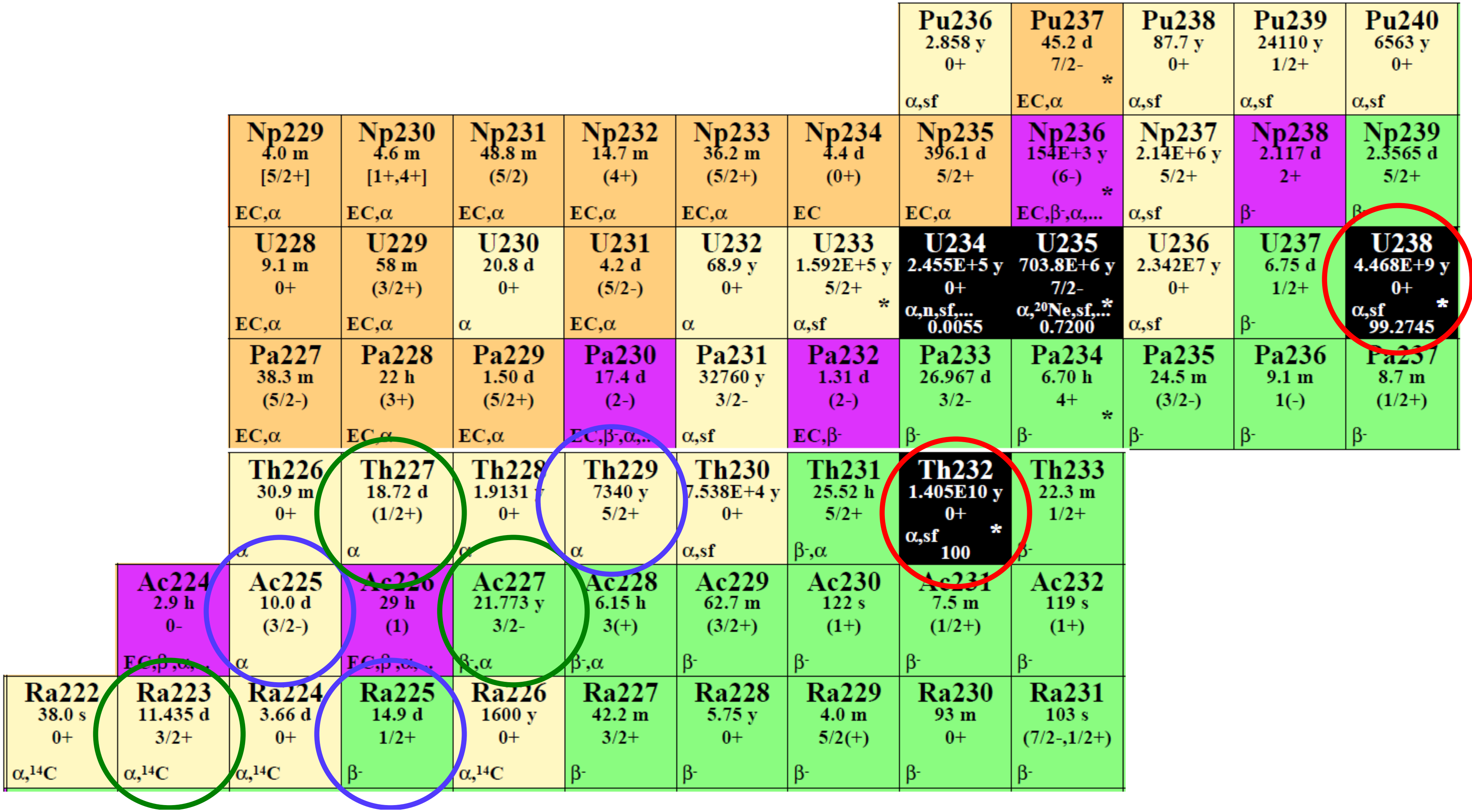
Ermolaev, S.V.; et al.; Production of actinium, thorium and radium isotopes from natural thorium with protons up to 141 MeV *Radiochimica Acta* **100**, 223-229 (2012).



$$^{232}\mathrm{Th}(p,2p\,6n)^{225}\mathrm{Ac} \text{ and/or } ^{232}\mathrm{Th}(p,\alpha\,4n)^{225}\mathrm{Ac} \quad (1)$$

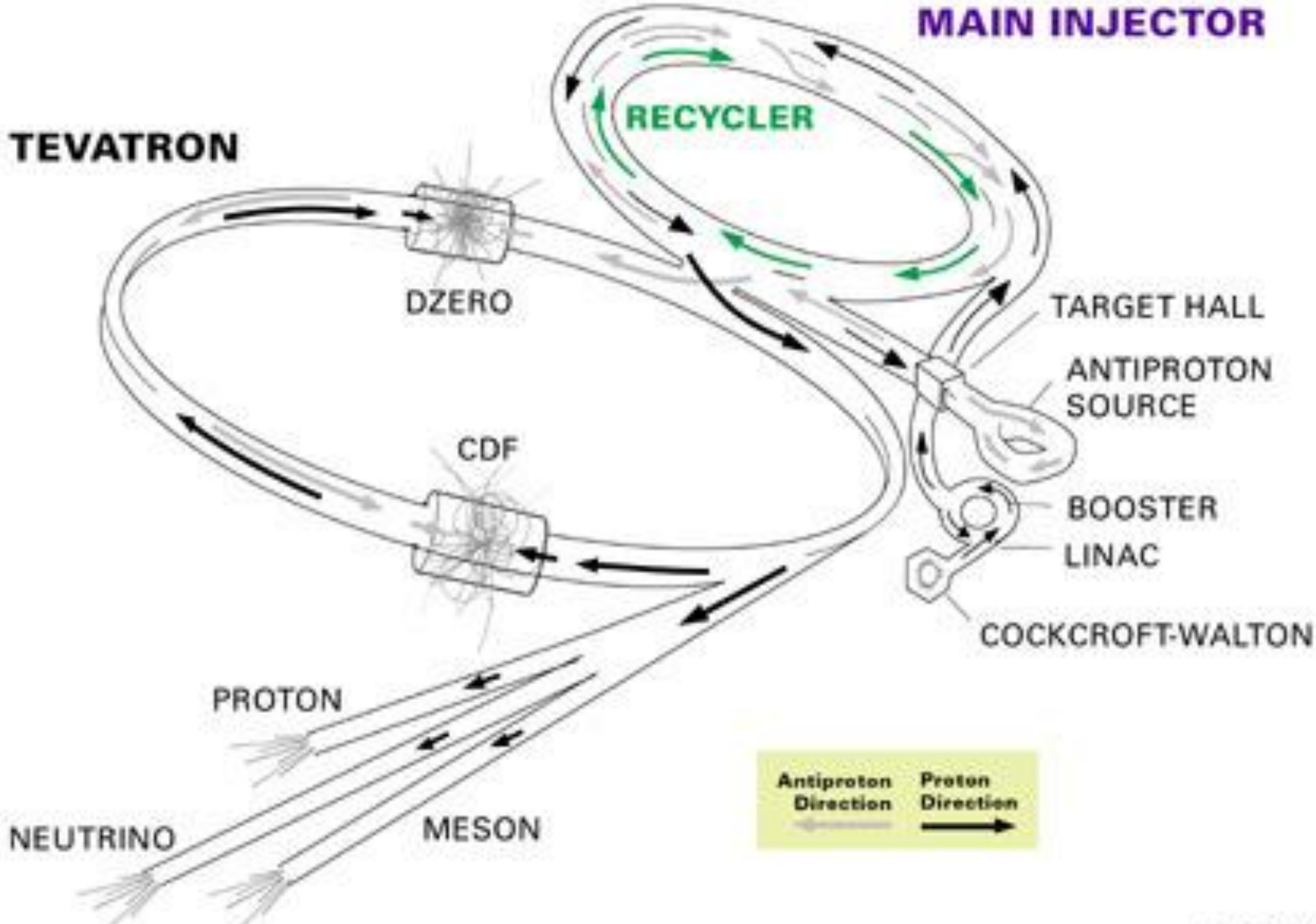
$$^{232}\mathrm{Th}(p,p\,7n)^{225}\mathrm{Th}\ (T_{1/2}=8\text{ min, EC, }10\%) \to {}^{225}\mathrm{Ac} \quad (2)$$

$$^{232}\mathrm{Th}(p,4n)^{229}\mathrm{Pa}\ (T_{1/2}=1.4\text{ d}, \alpha, 0.48\%) \to {}^{225}\mathrm{Ac} \quad (3)$$

$$^{232}\mathrm{Th}(p,3p\,5n)^{225}\mathrm{Ra}\ (T_{1/2}=14.8\text{ d}, \beta^-, 100\%) \to {}^{225}\mathrm{Ac} \text{ and/or } ^{232}\mathrm{Th}(p,\alpha\,p\,3n)^{225}\mathrm{Ra} \to {}^{225}\mathrm{Ac} \quad (4)$$

Zhuikov, B.L.; et al.; Production of 225Ac and 223Ra by Irradiation of Th with Accelerated Protons *Radiochemistry*, *53(1)*, 73-80 (2011).

FERMILAB'S ACCELERATOR CHAIN

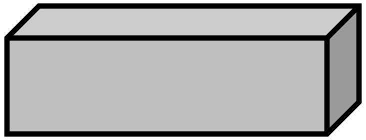


une 1-7 2011, 5.8x10^{17} 8GeV protons

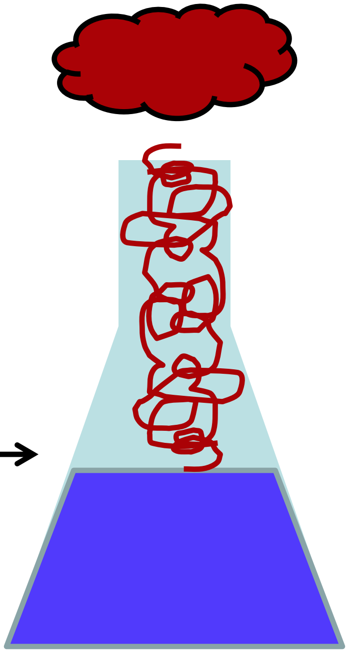
Target Dissolution (July 2011)

Cu clad target
(0.127 cm Cu)

Physical
De-cladding



35g Th Metal
(2.3 x 1.0 x 1.3 cm)



Dissolve Th
8M HNO_3 +
0.01M HF

Complex
Residual F^-
with Boric acid

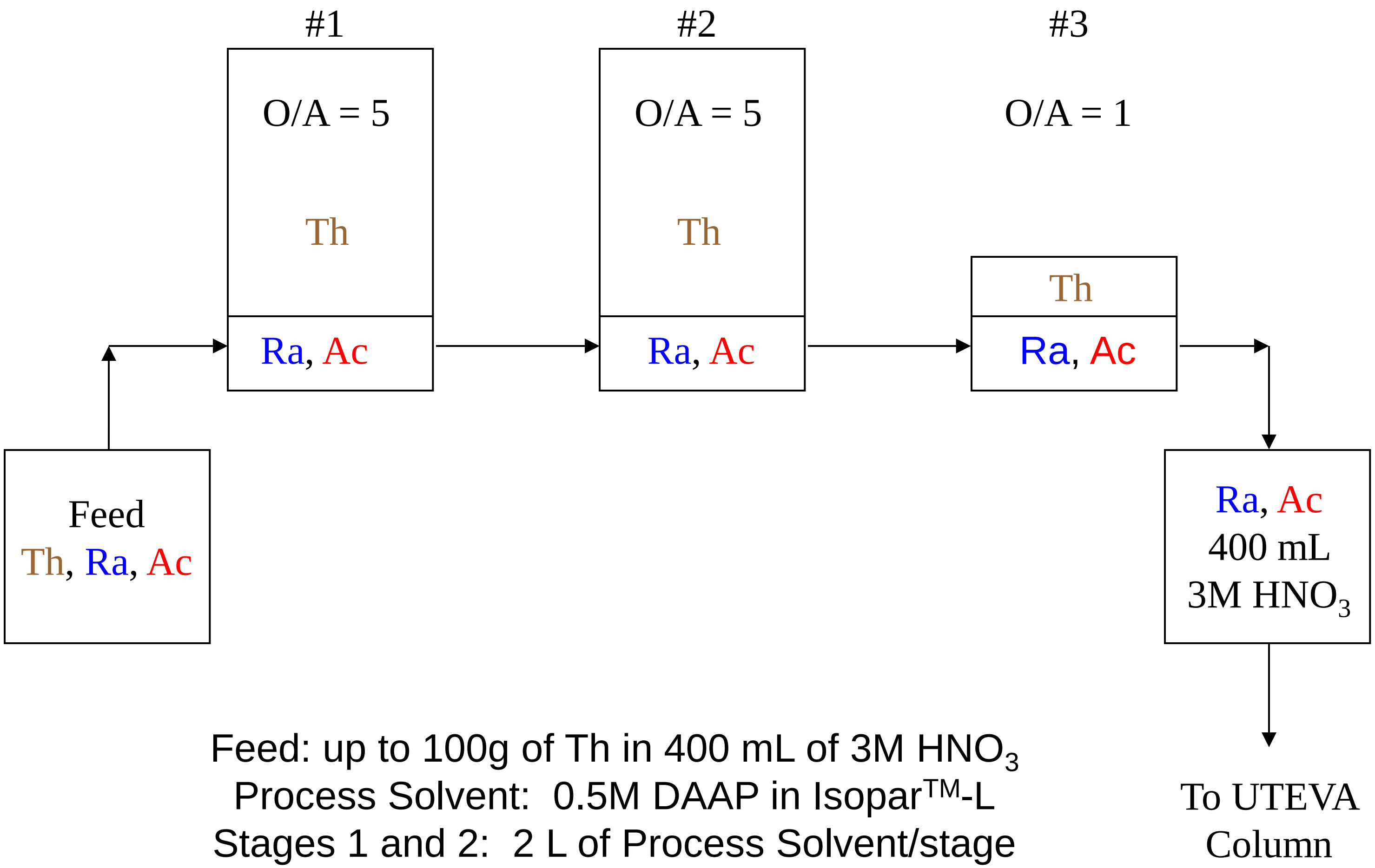
Physical
De-cladding

Cu Cladding
analyzed for beam
characterization

Primary Separation for Actinium Recovery and Purification

Separation of Ra and Ac from Th Stock

(Batch Extraction Process)



Feed: up to 100g of Th in 400 mL of 3M HNO_3

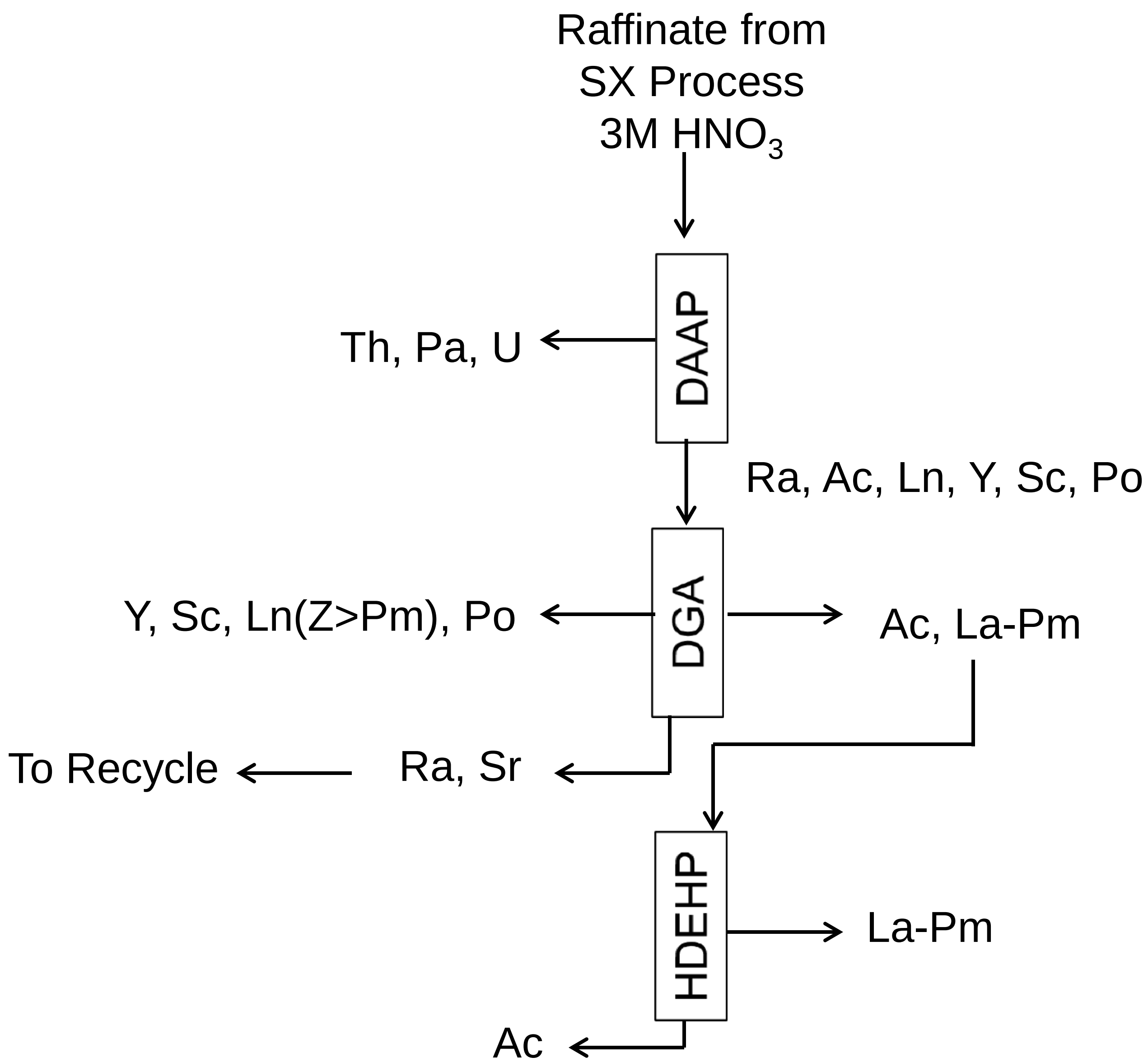
Process Solvent: 0.5M DAAP in Isopar™-L

Stages 1 and 2: 2 L of Process Solvent/stage

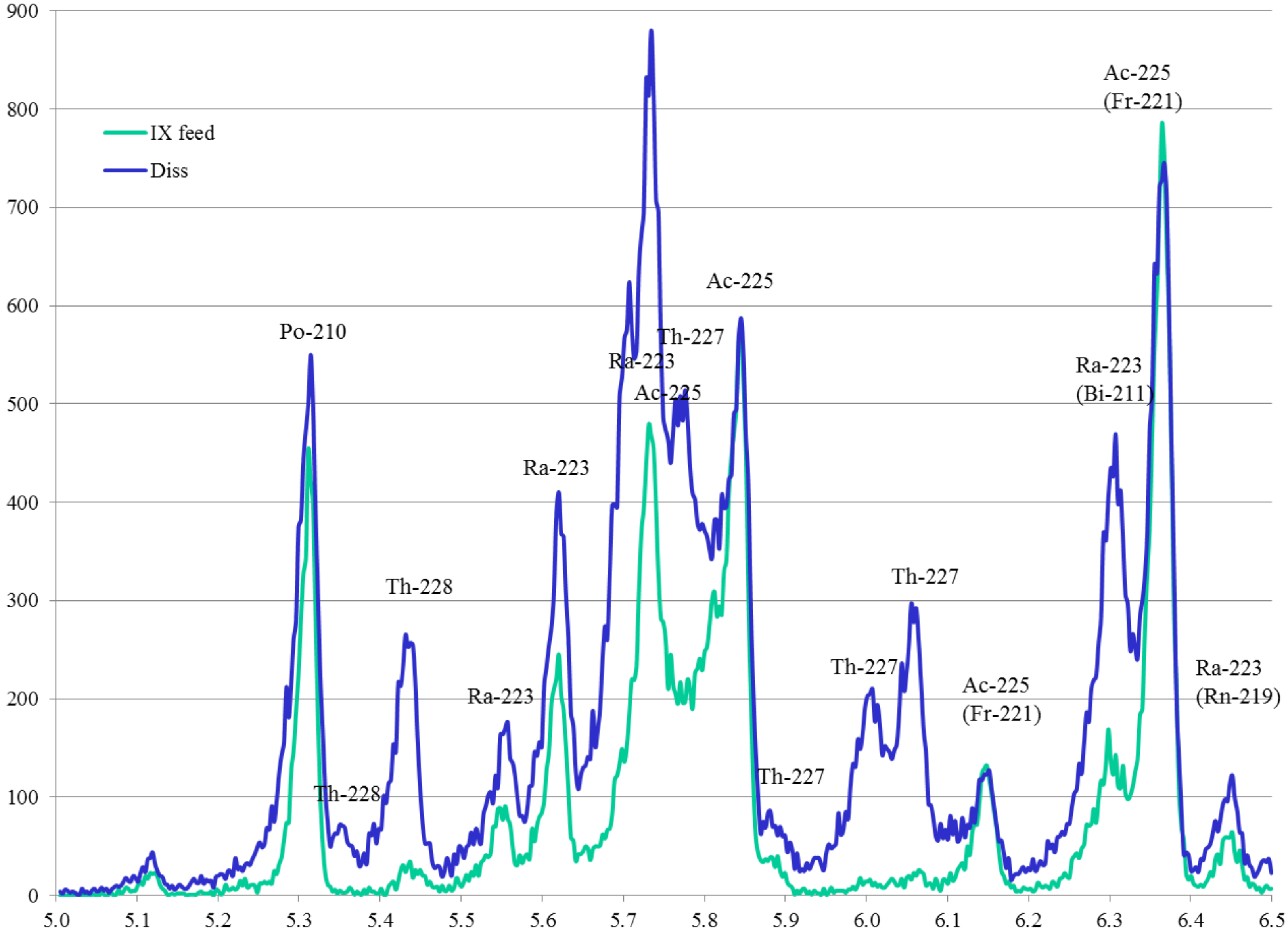
Stage 3: 400 mL of Process Solvent

Mixing Time: 2 minutes/stage

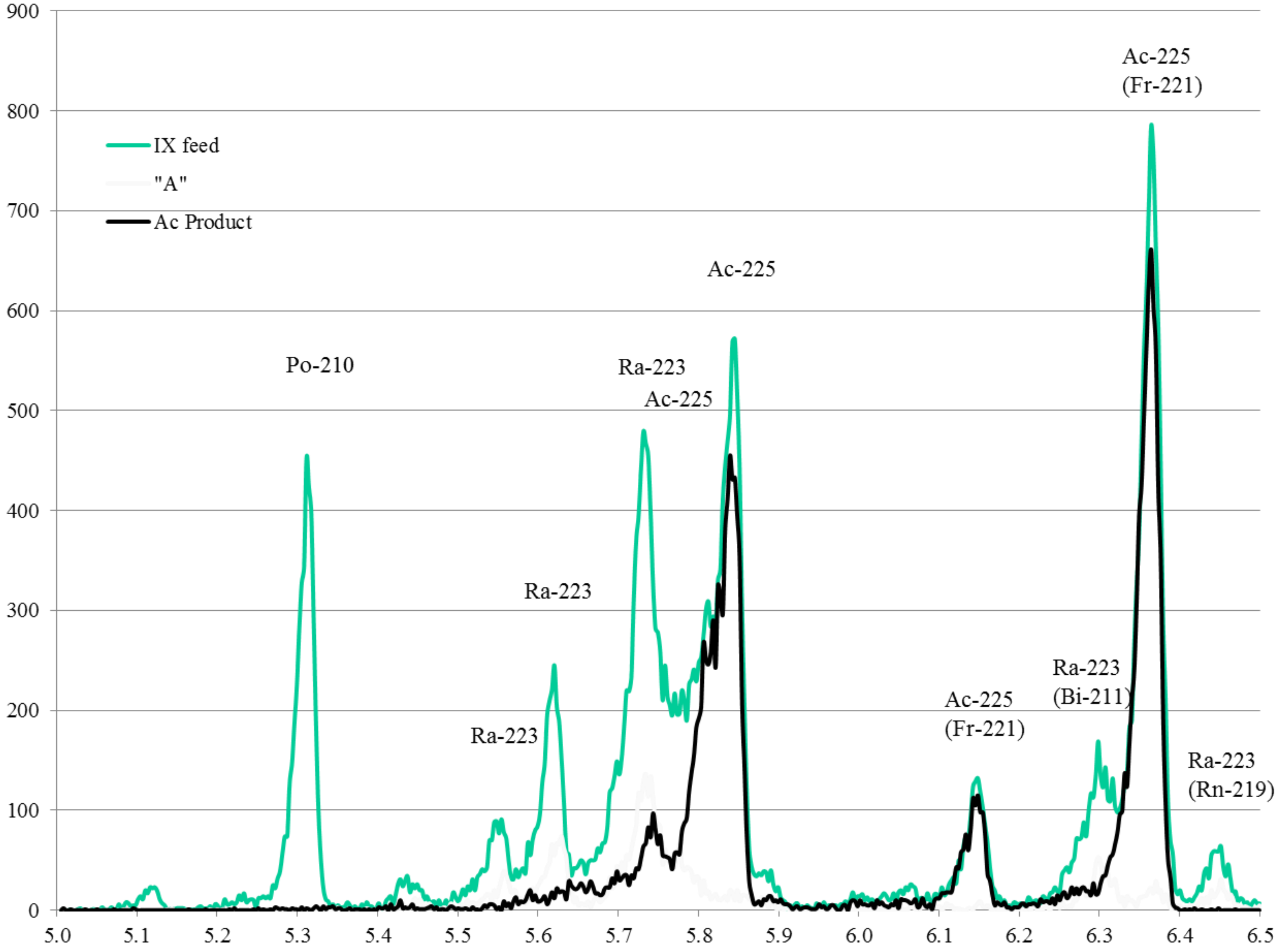
Tandem Column System for the rapid Extraction and Purification of Ac-225



Initial Dissolution and Ion Exchange Feed



UTEVA/DGA Separation

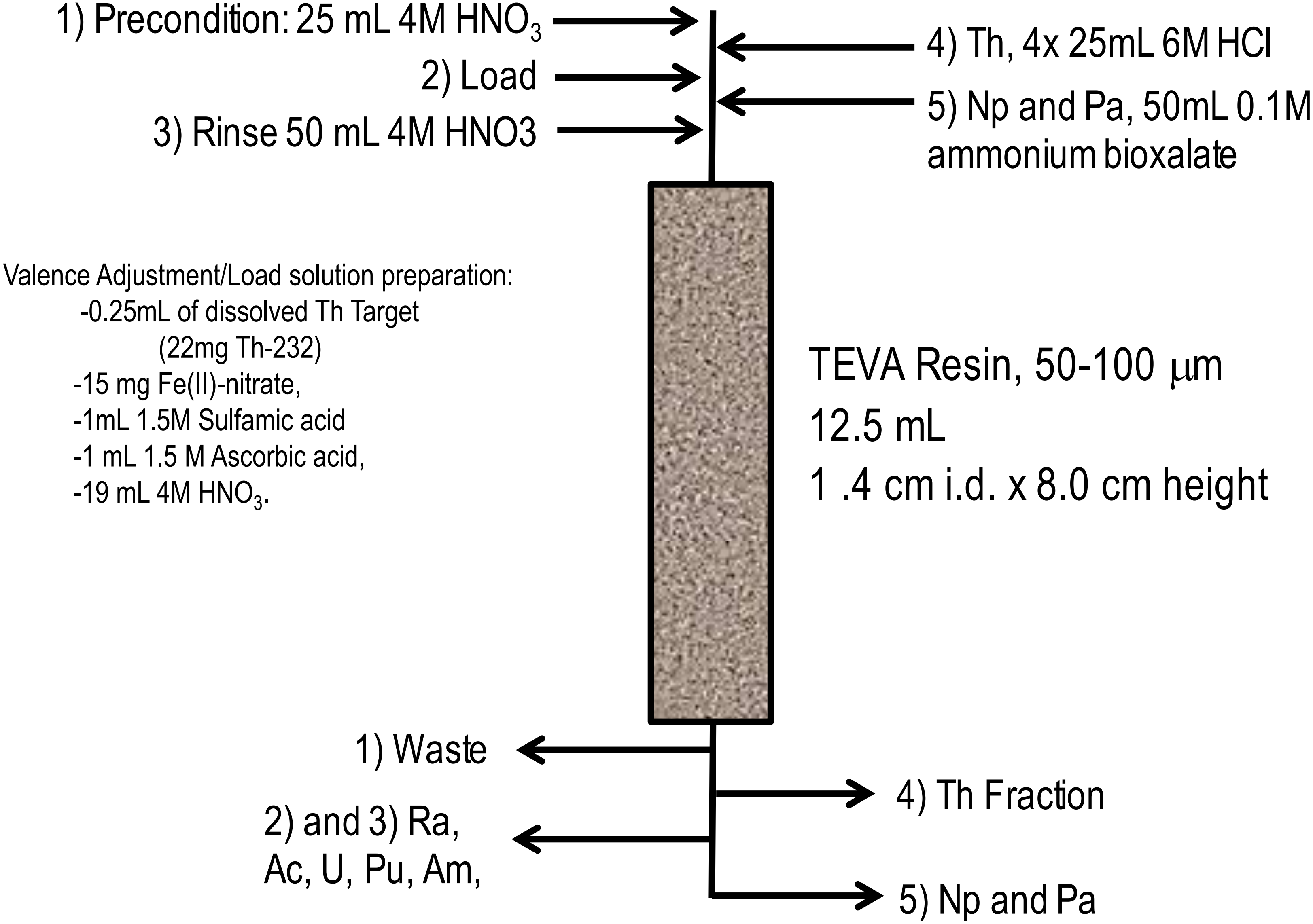


Yields of Key Isotopes
(5.9×10^{16} protons on 30g Th-232)

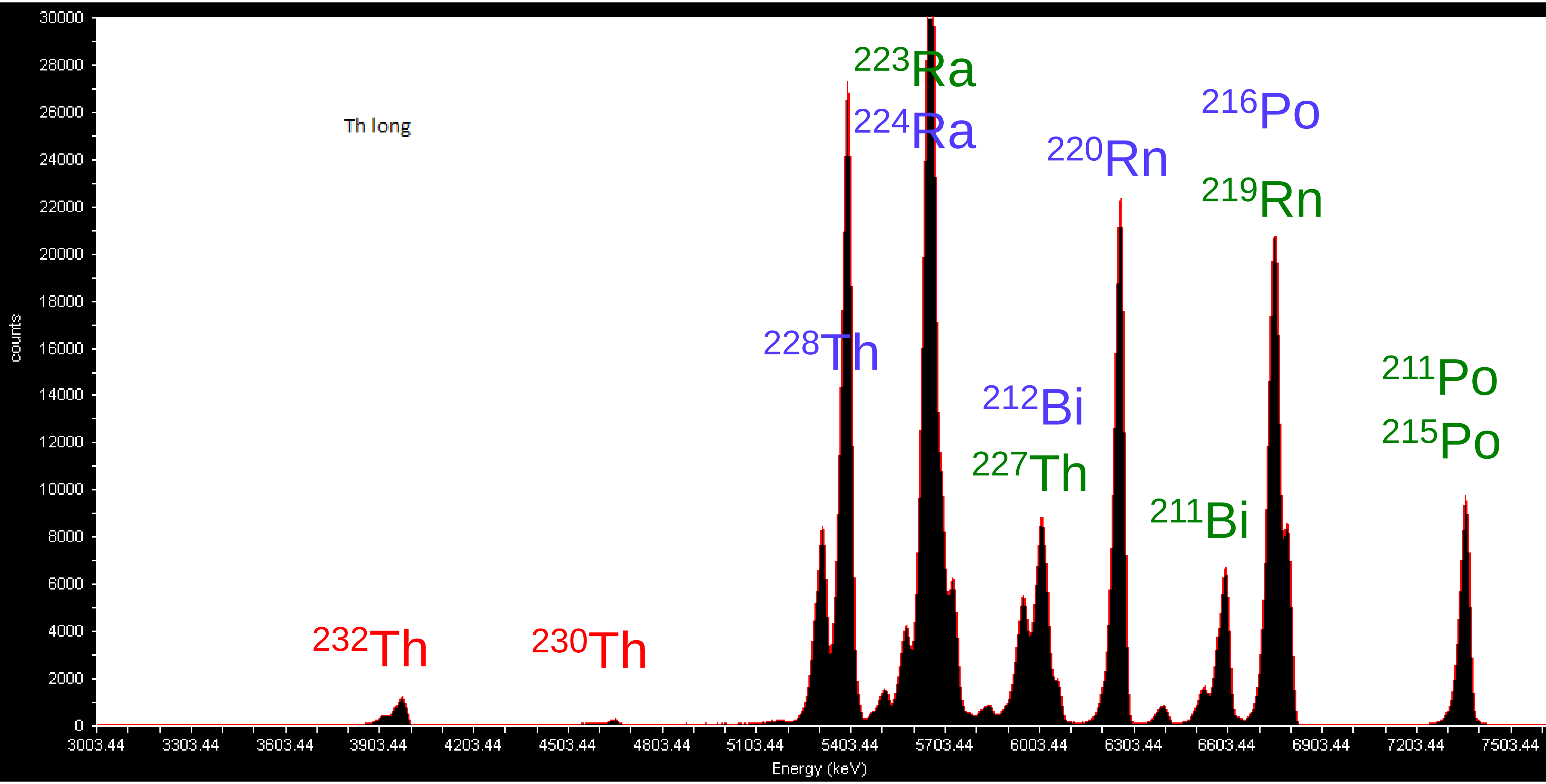
Isotope	Half-life	Atoms	uCi
$_{89}$Ac-225	10 d	7.7×10^{13}	1700
$_{89}$Ac-227	22 y	7.3×10^{13}	2.0
$_{88}$Ra-225	14 d	2.0×10^{13}	290
$_{90}$Th-227	18.72 d	8.1×10^{13}	940

Analytical Separations for Byproduct Determination

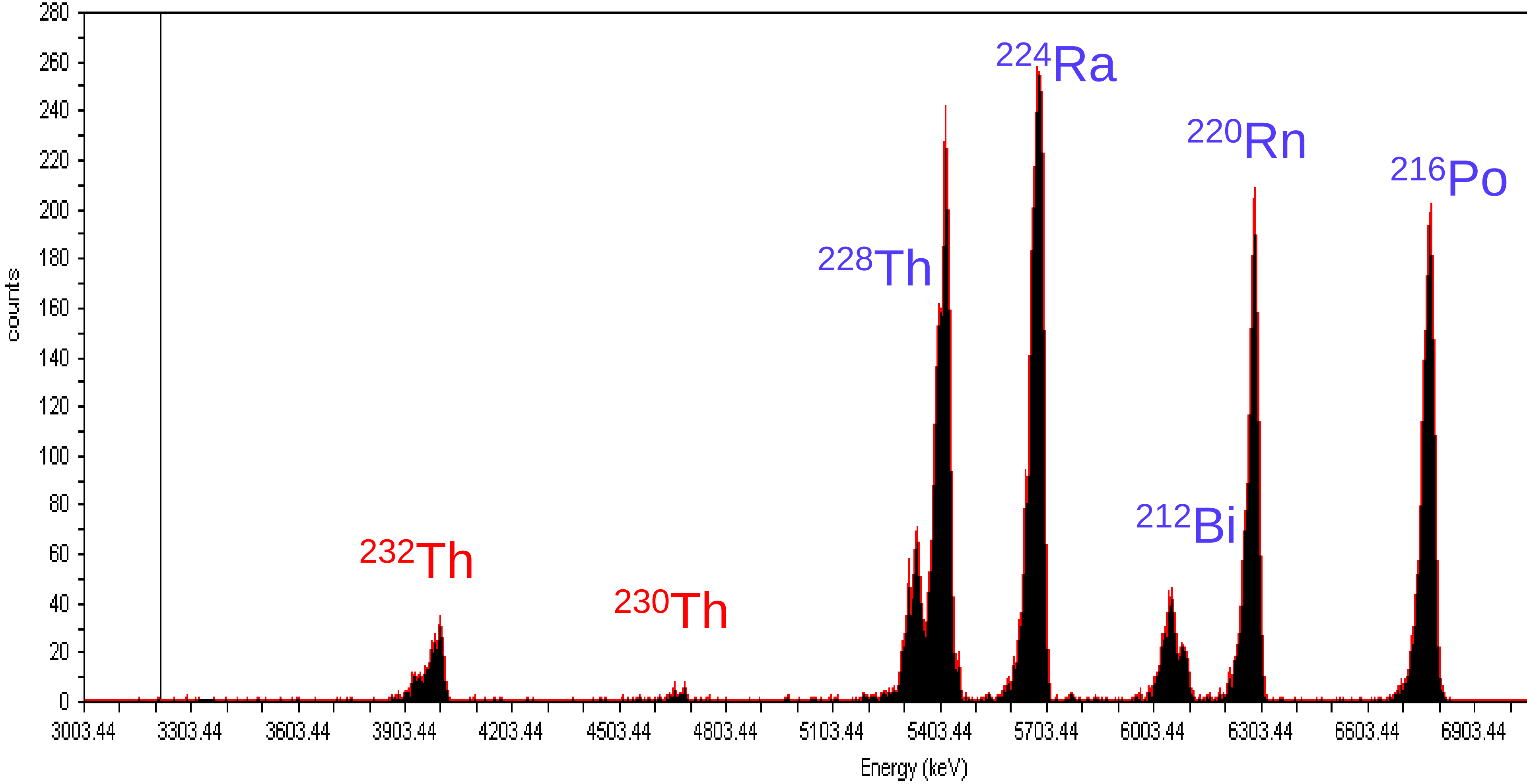
Bulk Th Removal



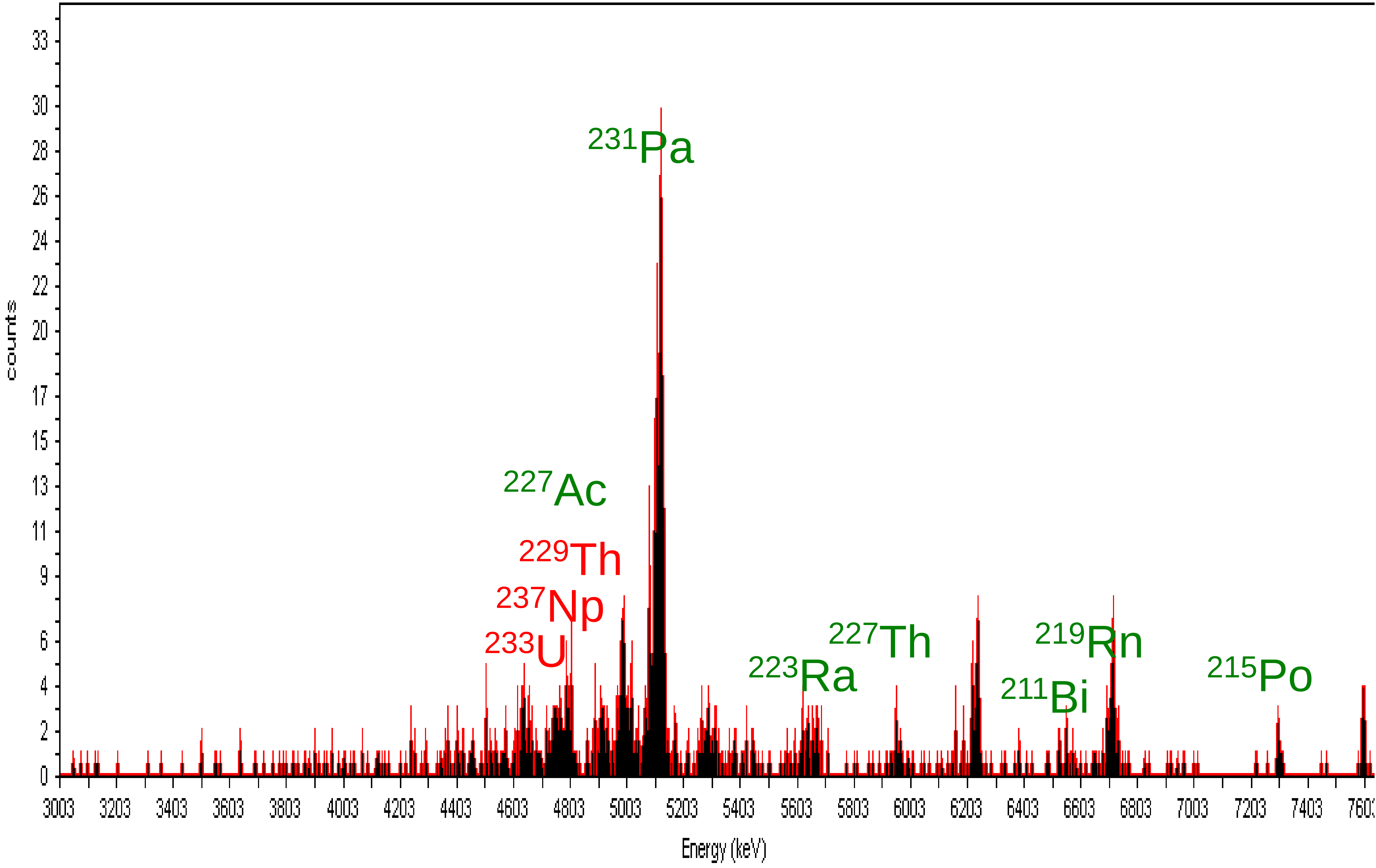
Th Alpha Spectrum (CeF_3 ppt, immediate)



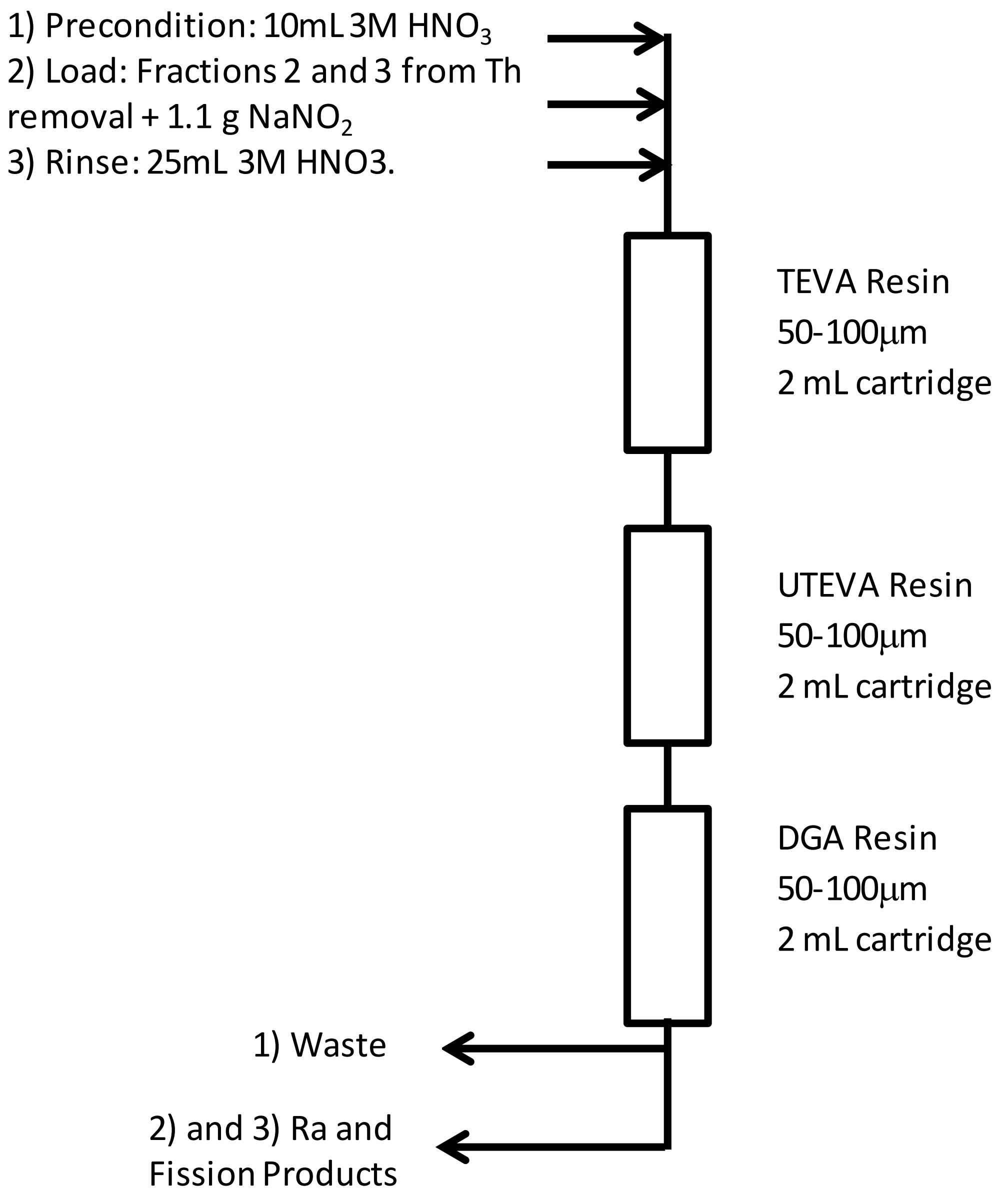
Th Alpha Spectrum (CeF_3 ppt, 3 years)



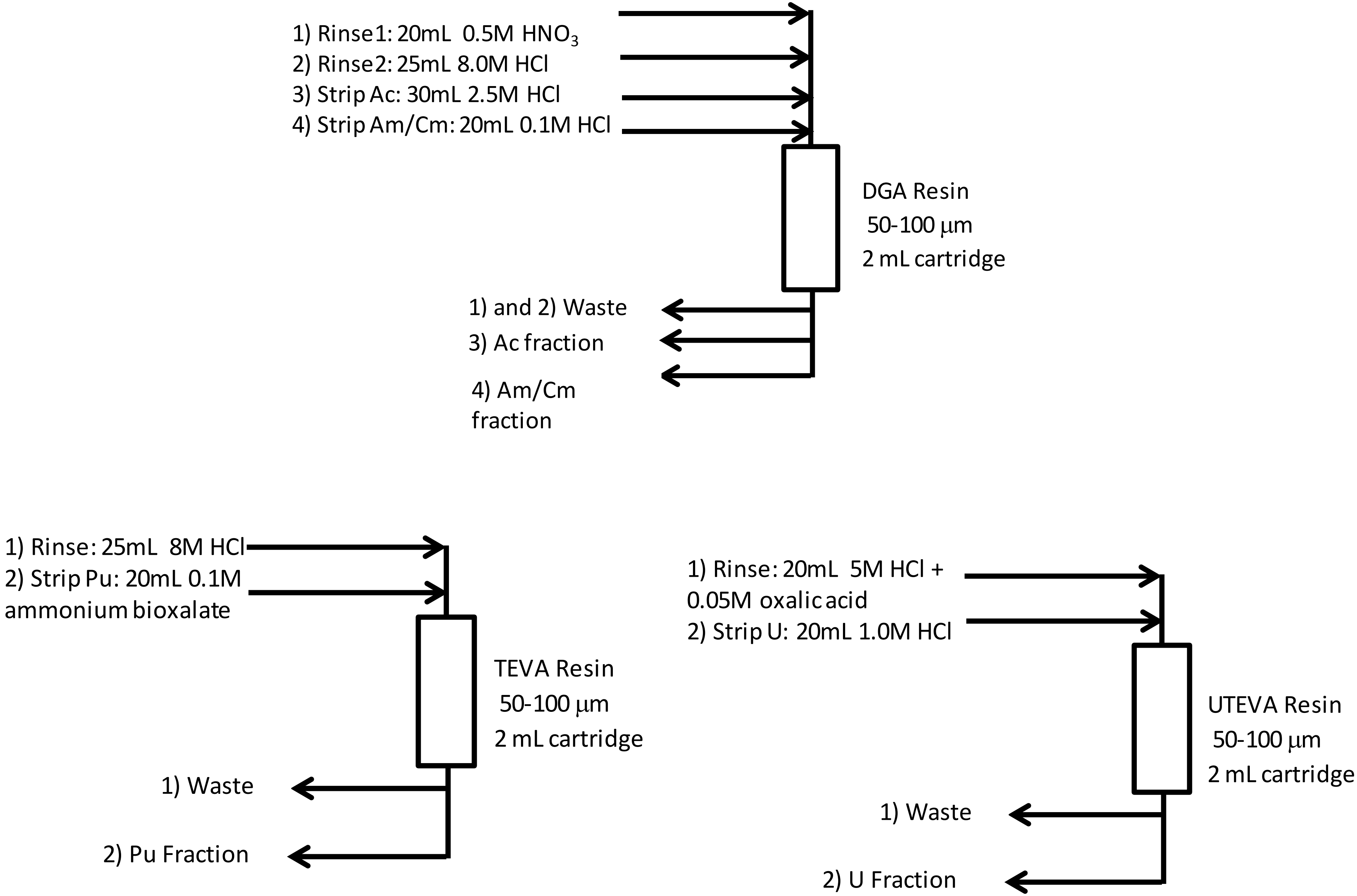
Np-Pa Alpha Spectrum (CeF_3 ppt, 3 years)



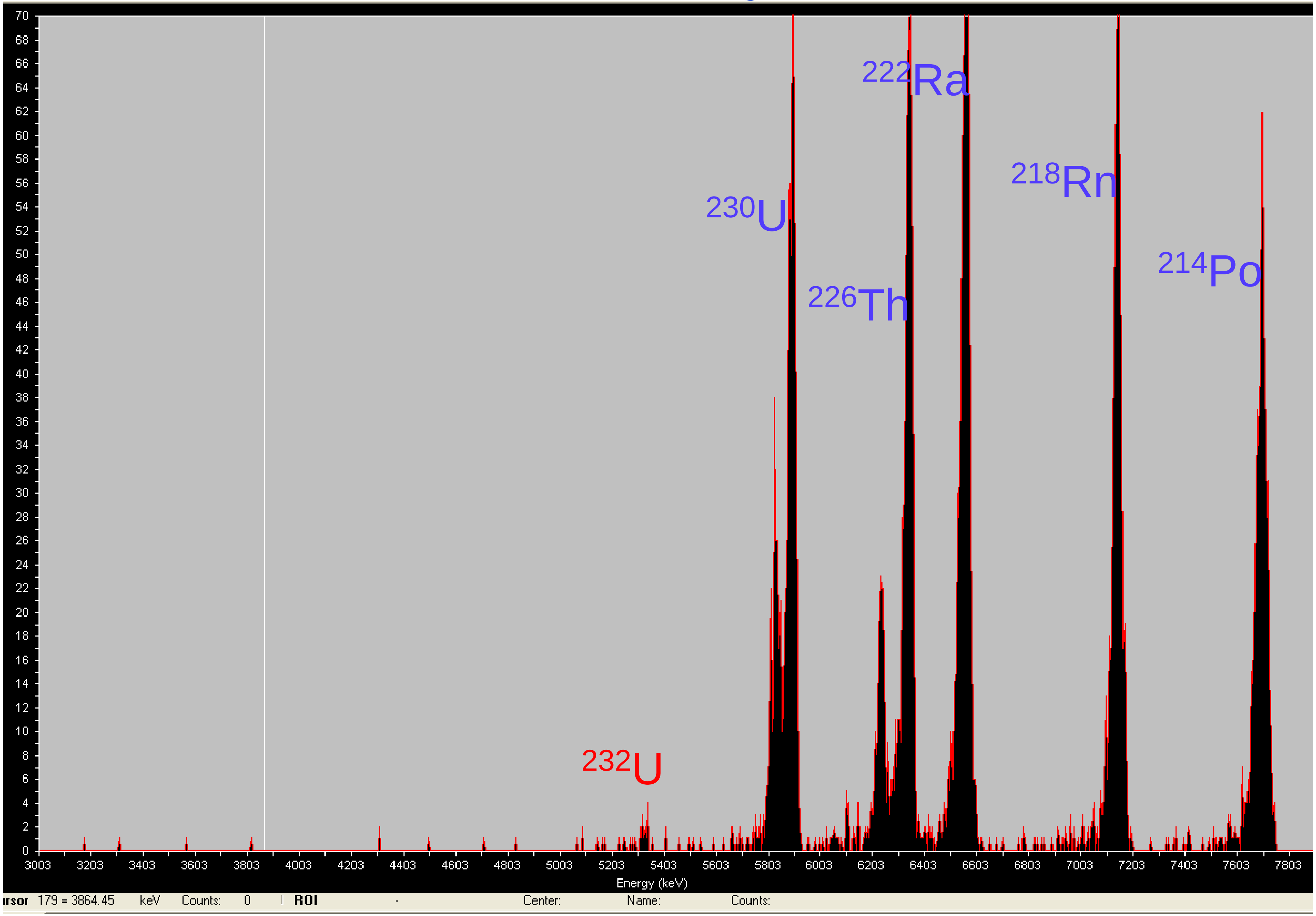
Actinide Separations



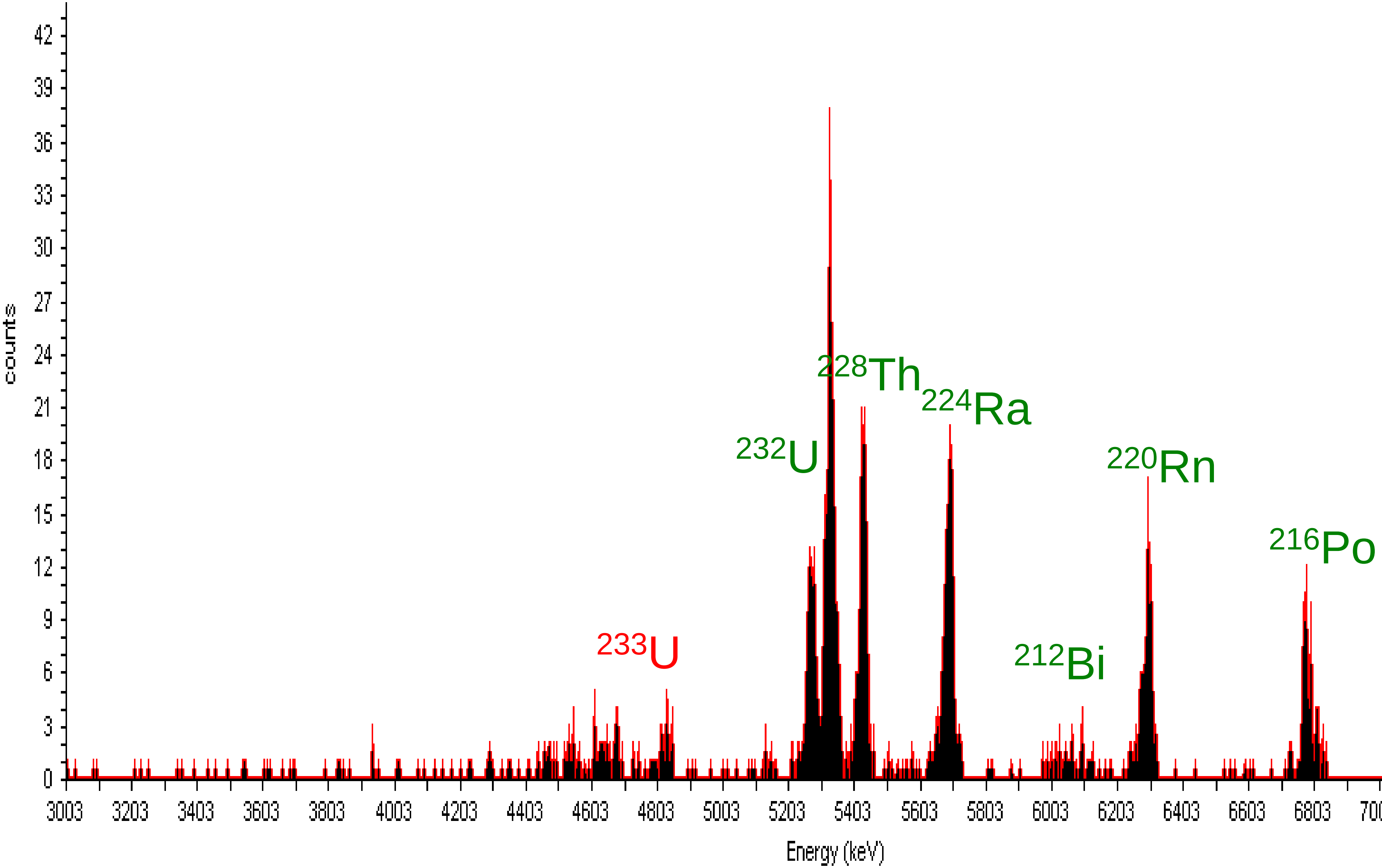
Actinide Separations



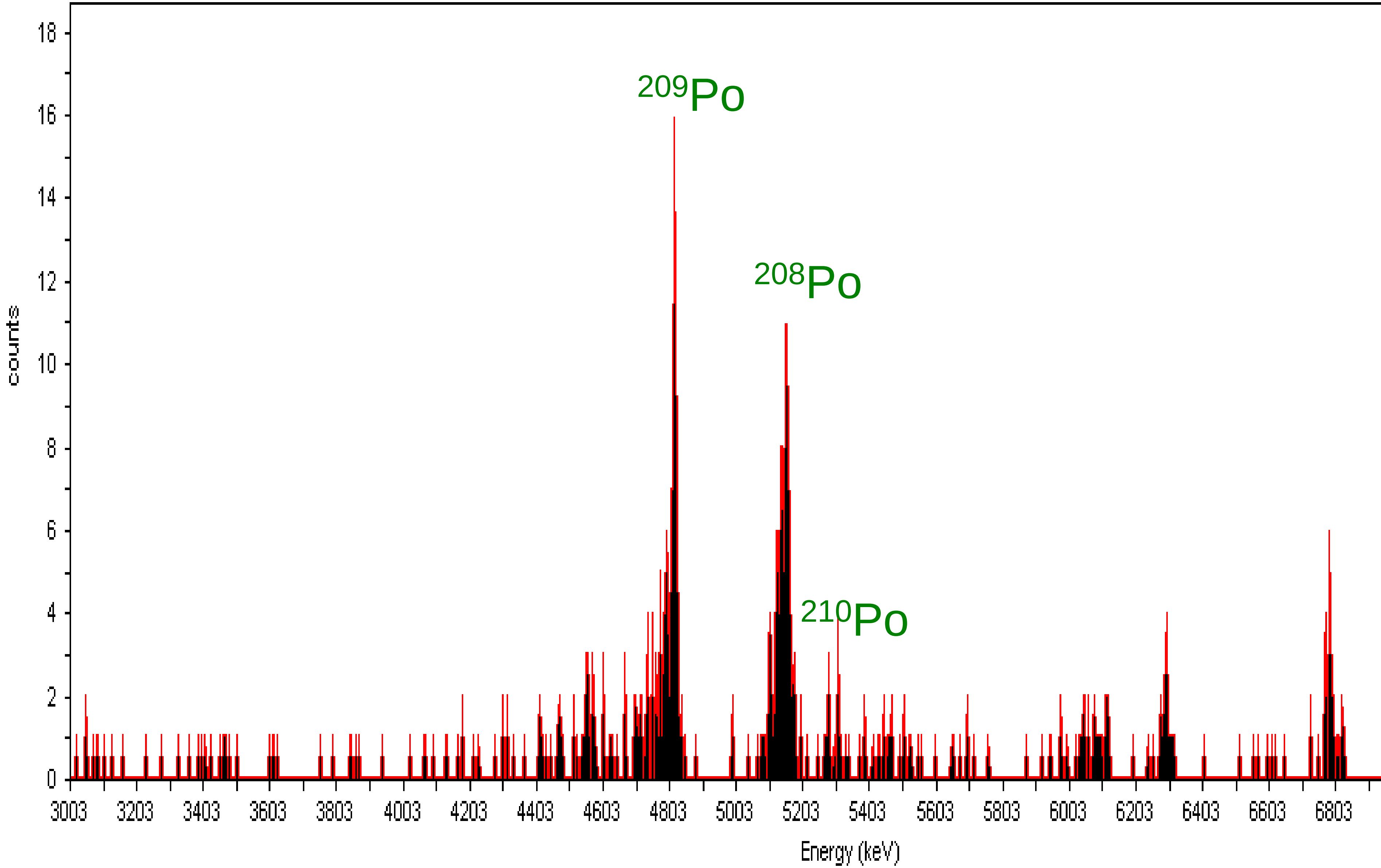
U Alpha Spectrum (CeF_3 ppt, immediate)



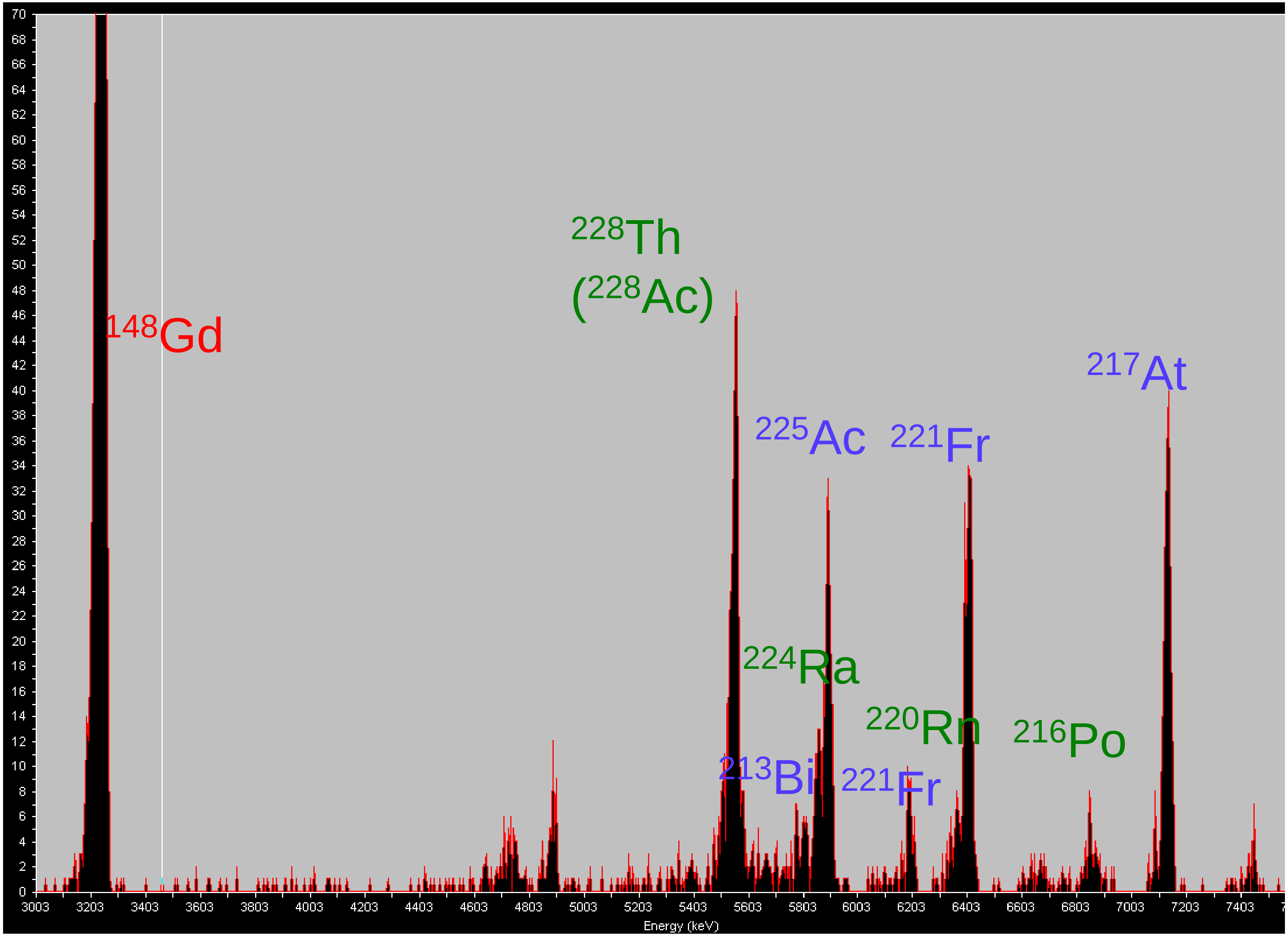
U Alpha Spectrum (CeF_3 ppt, 3 years)



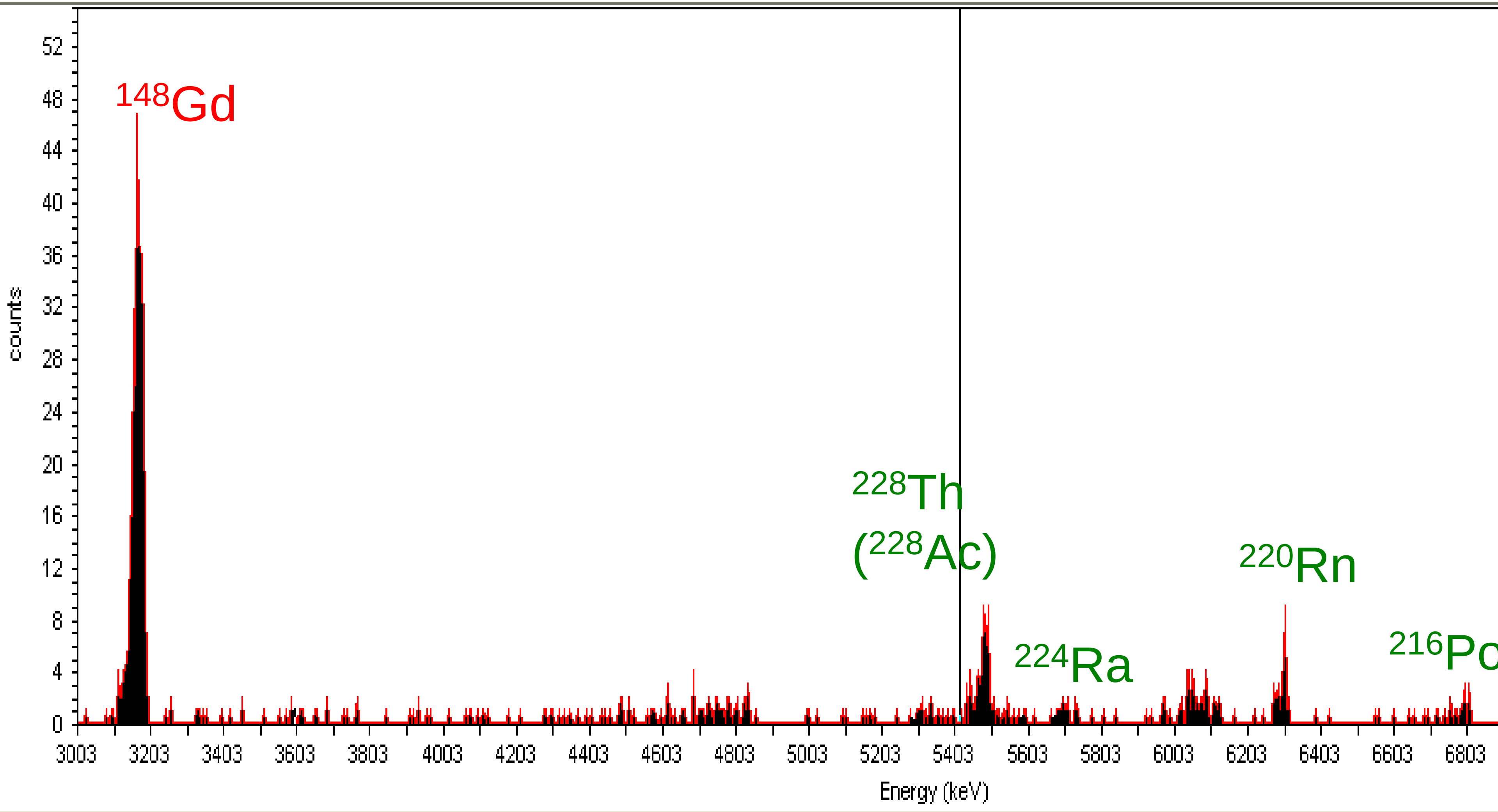
Pu Alpha Spectrum (CeF_3 ppt, 3 years)



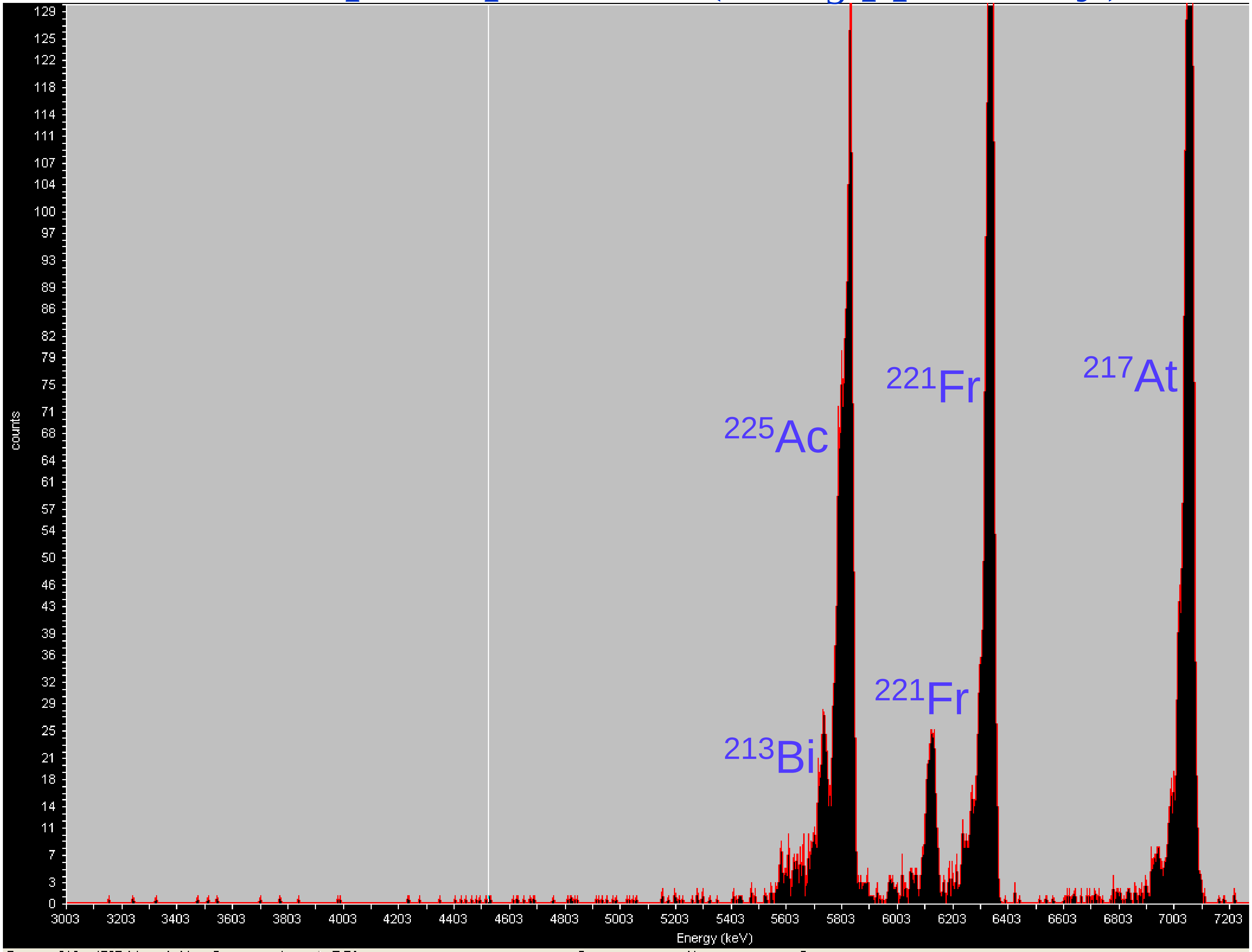
Am-Cm Alpha Spectrum (CeF_3 ppt, immediate)



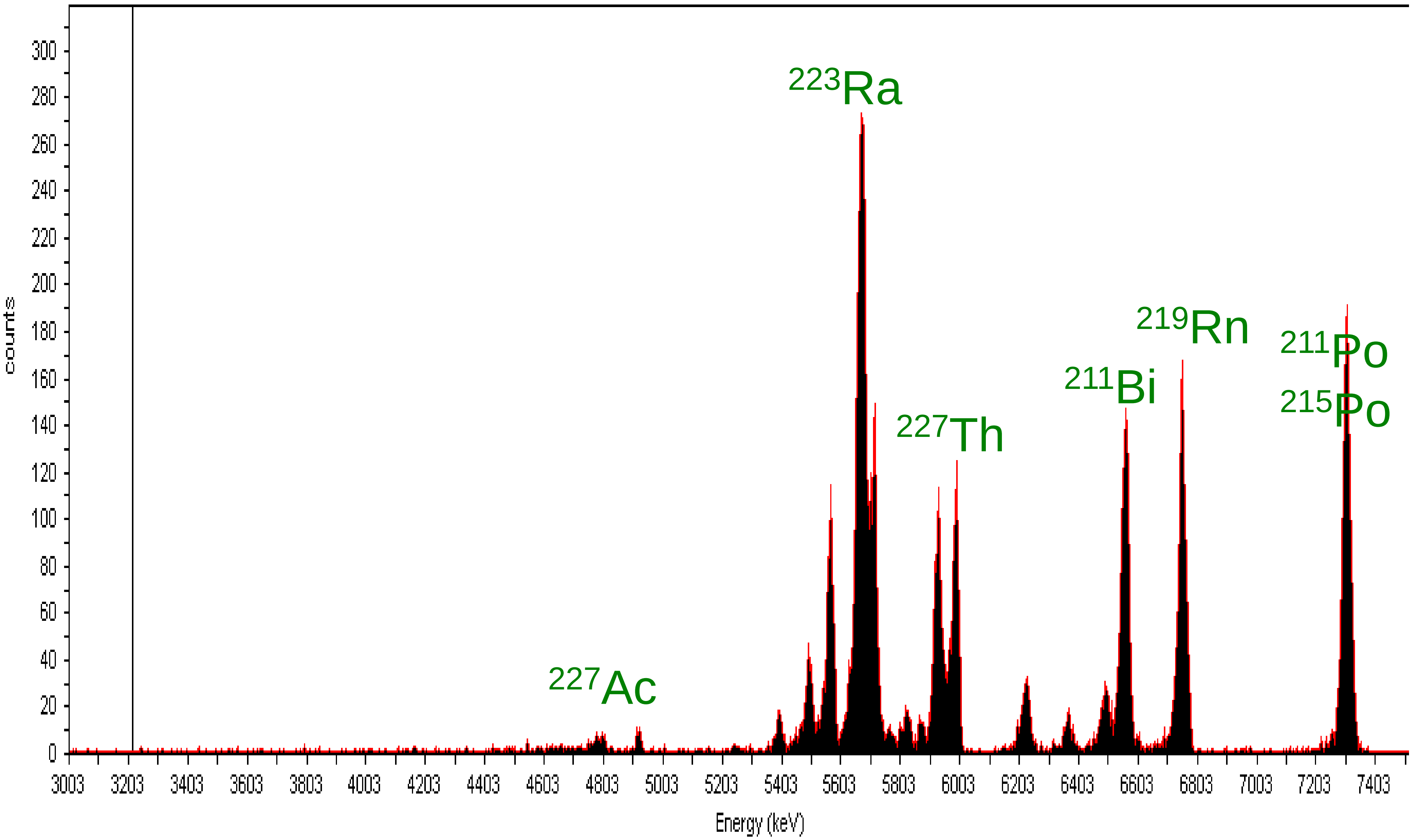
Am-Cm Alpha Spectrum (CeF_3 ppt, 3 years)



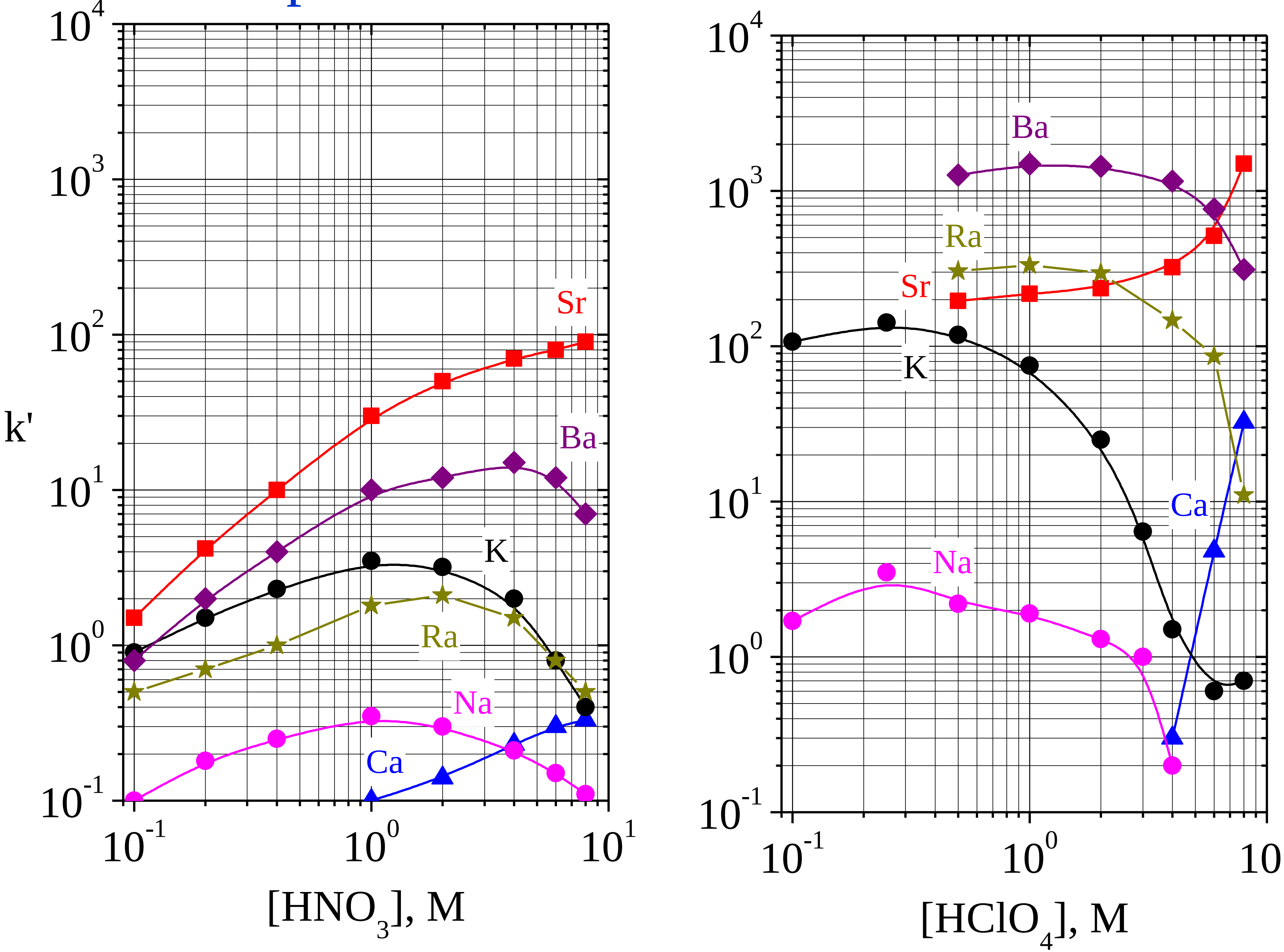
Ac Alpha Spectrum (CeF_3 ppt, 1 day)

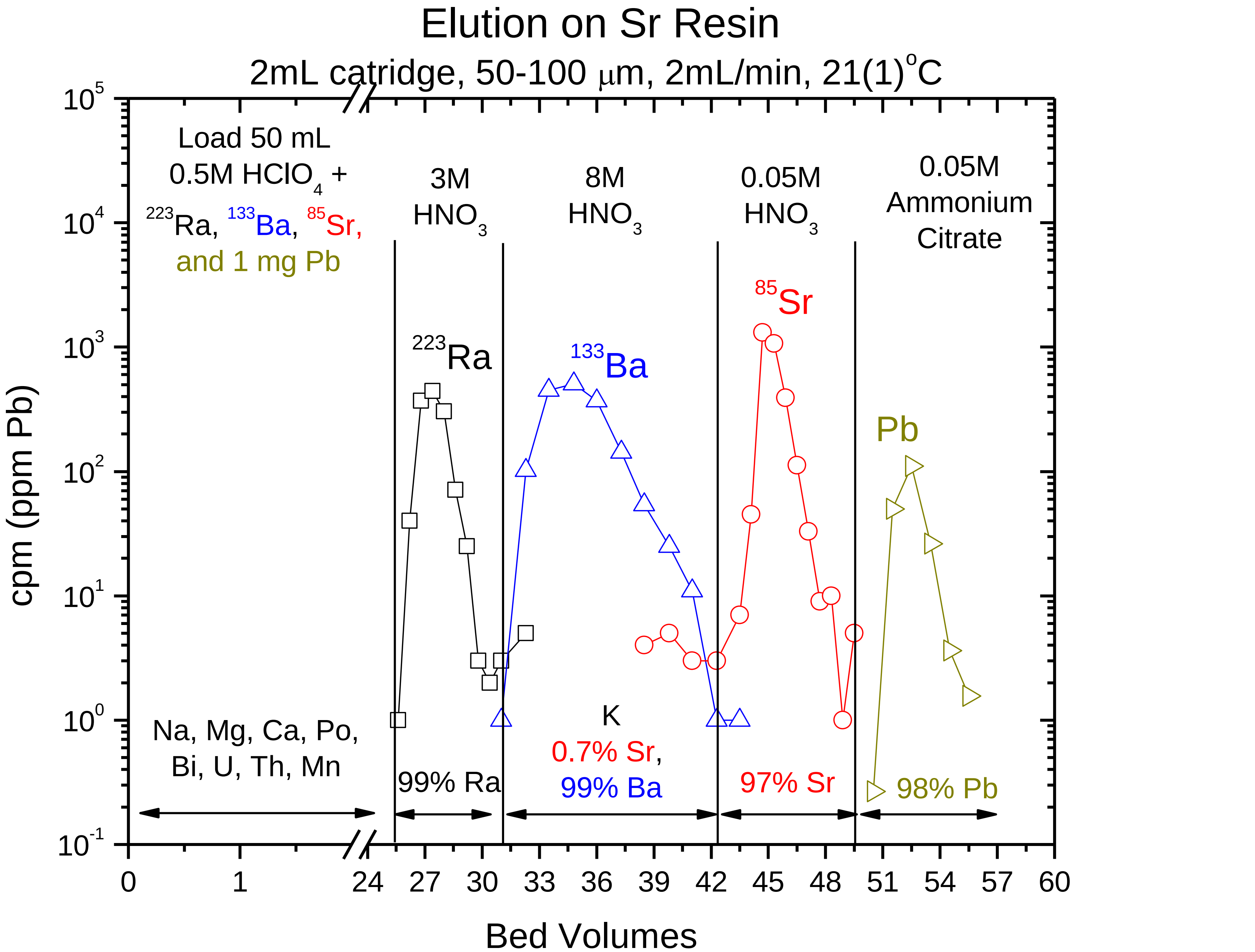


Ac Alpha Spectrum (CeF_3 ppt, 3 years)



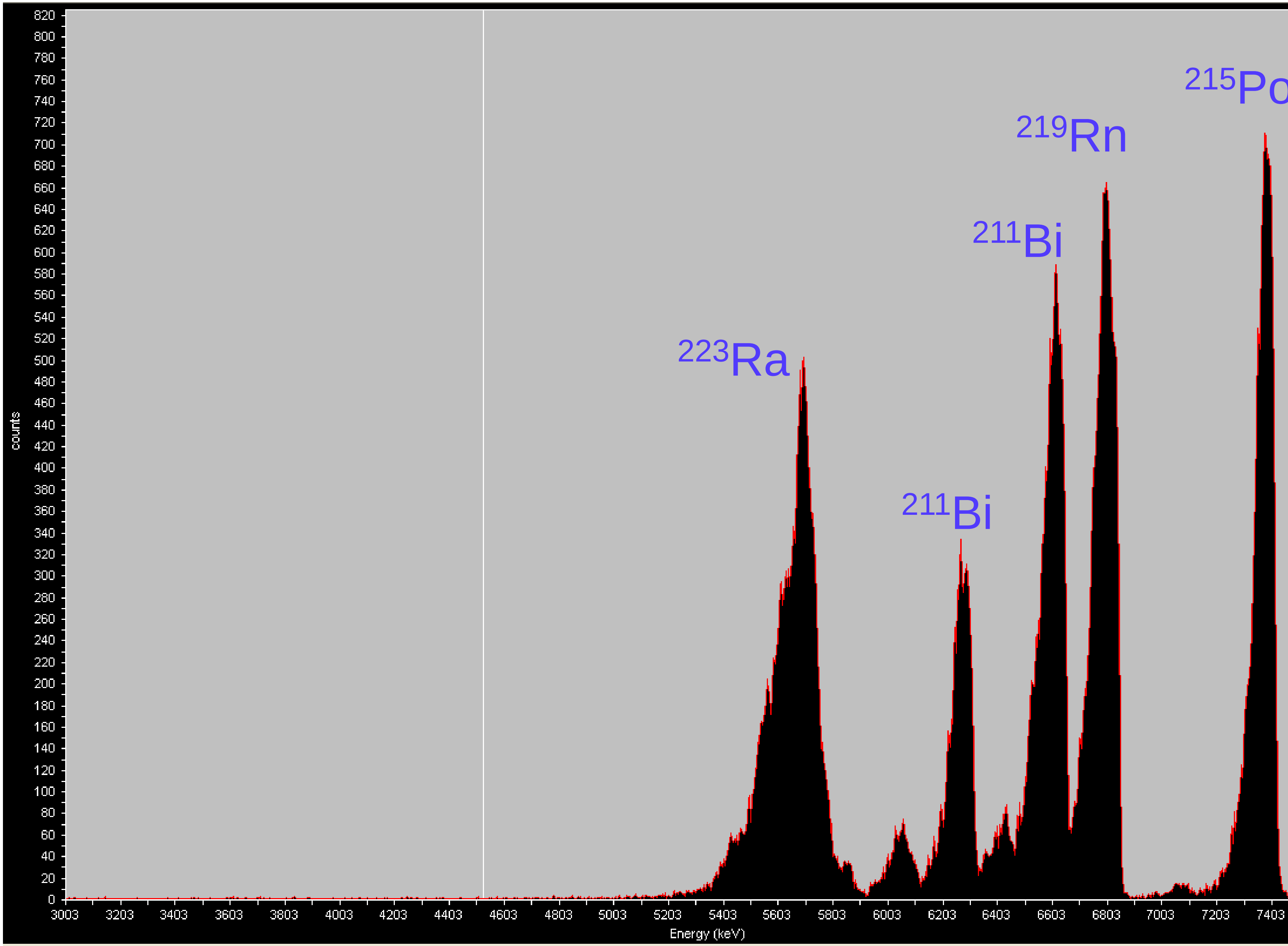
Uptake of Metal Ions on Sr Resin



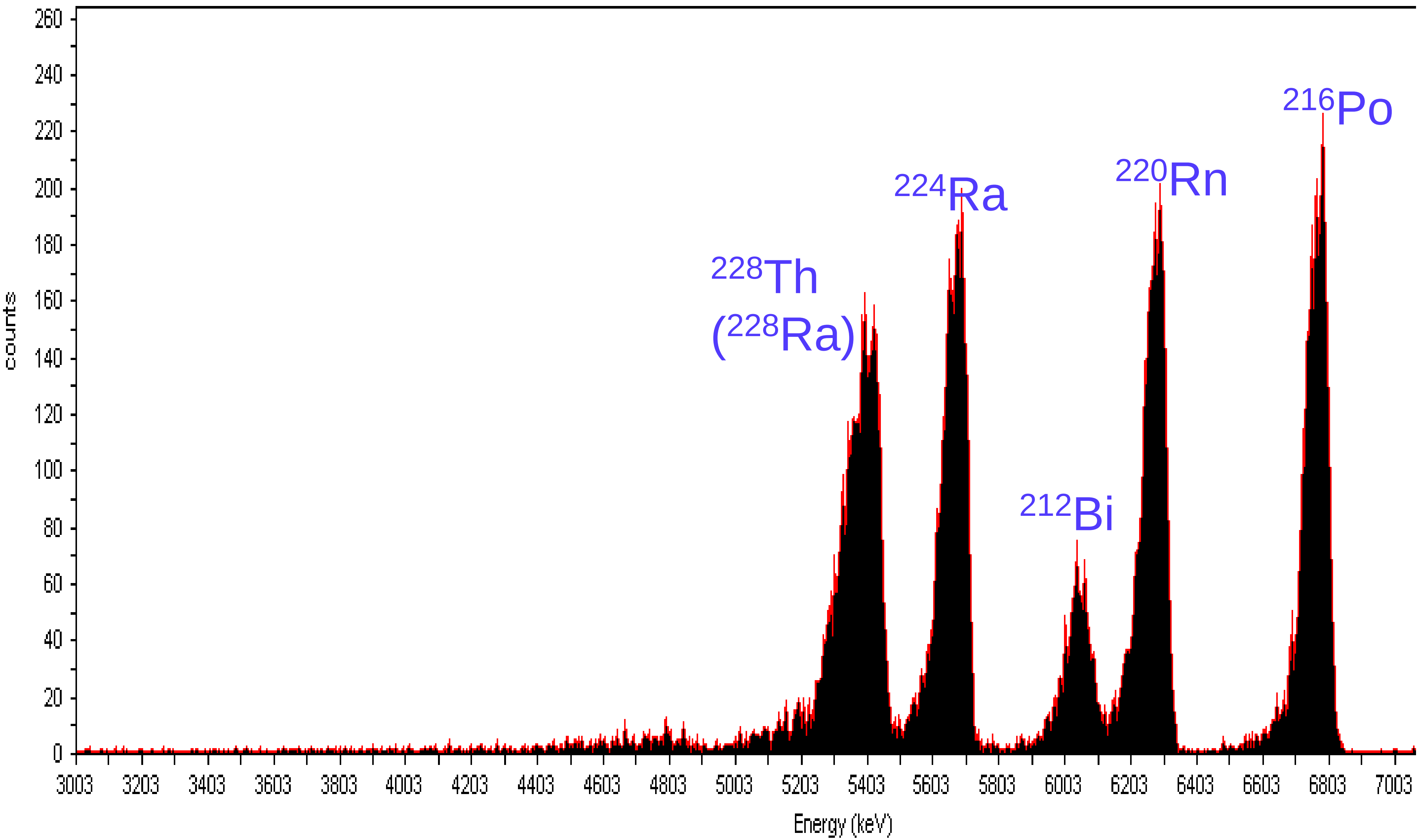


"Radium Determination Method Based on Extraction by Crown Ether from Dilute Perchloric Acid," D. R. McAlister, E. P. Horwitz, 57th Annual Radiobioassay and Radiochemical Measurement Conference, San Destin, FL, October 31- November 4, 2011.

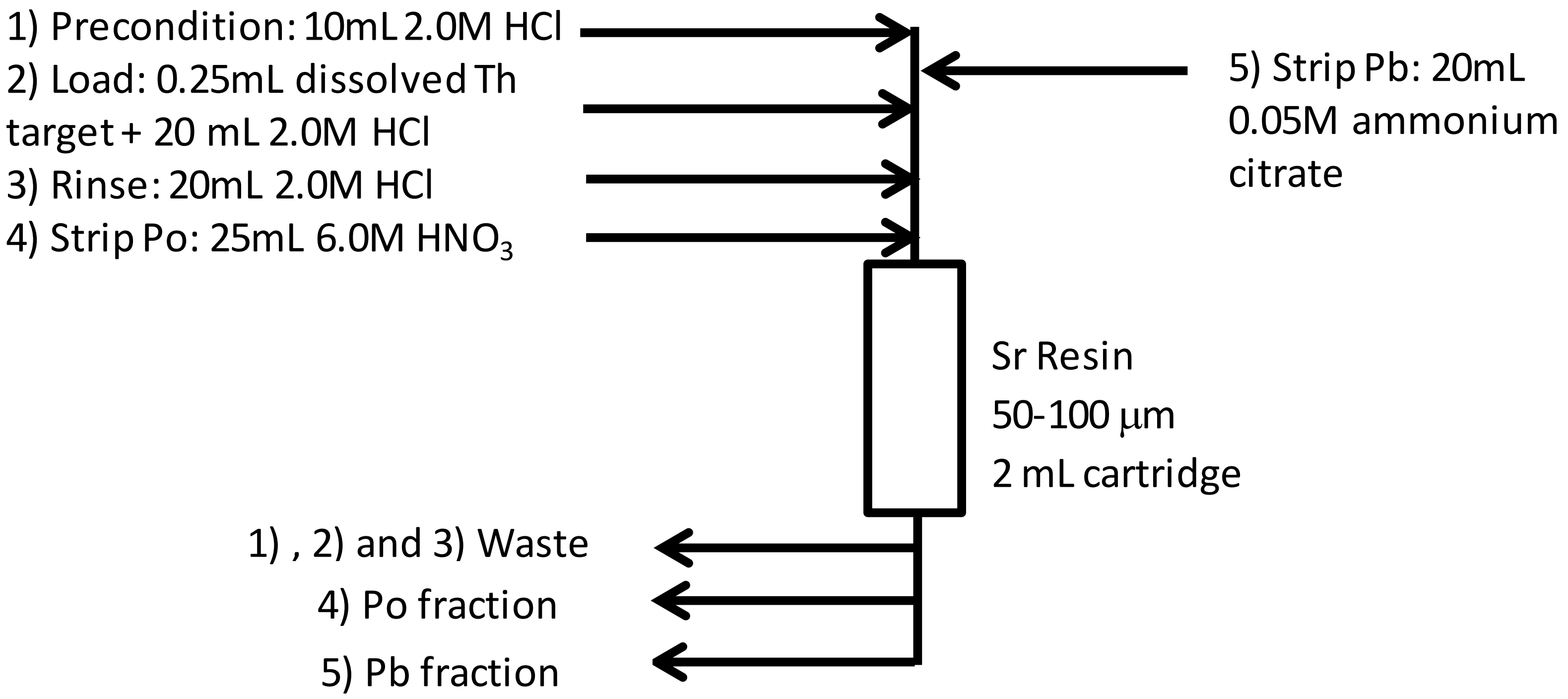
Ra Alpha Spectrum ($BaSO_4$, immediate)



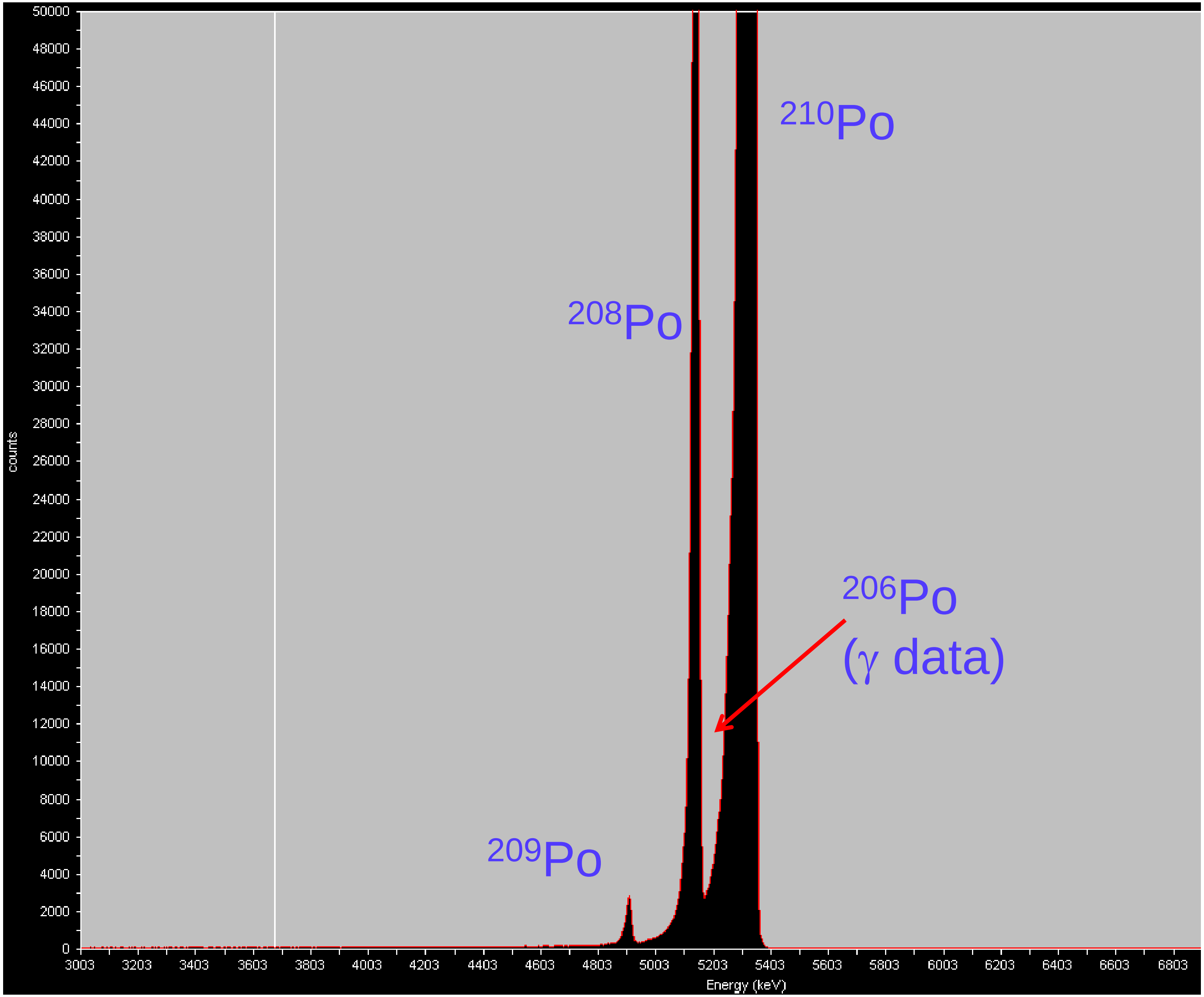
Ra Alpha Spectrum ($BaSO_4$, 3 years)



Pb/Po Separation



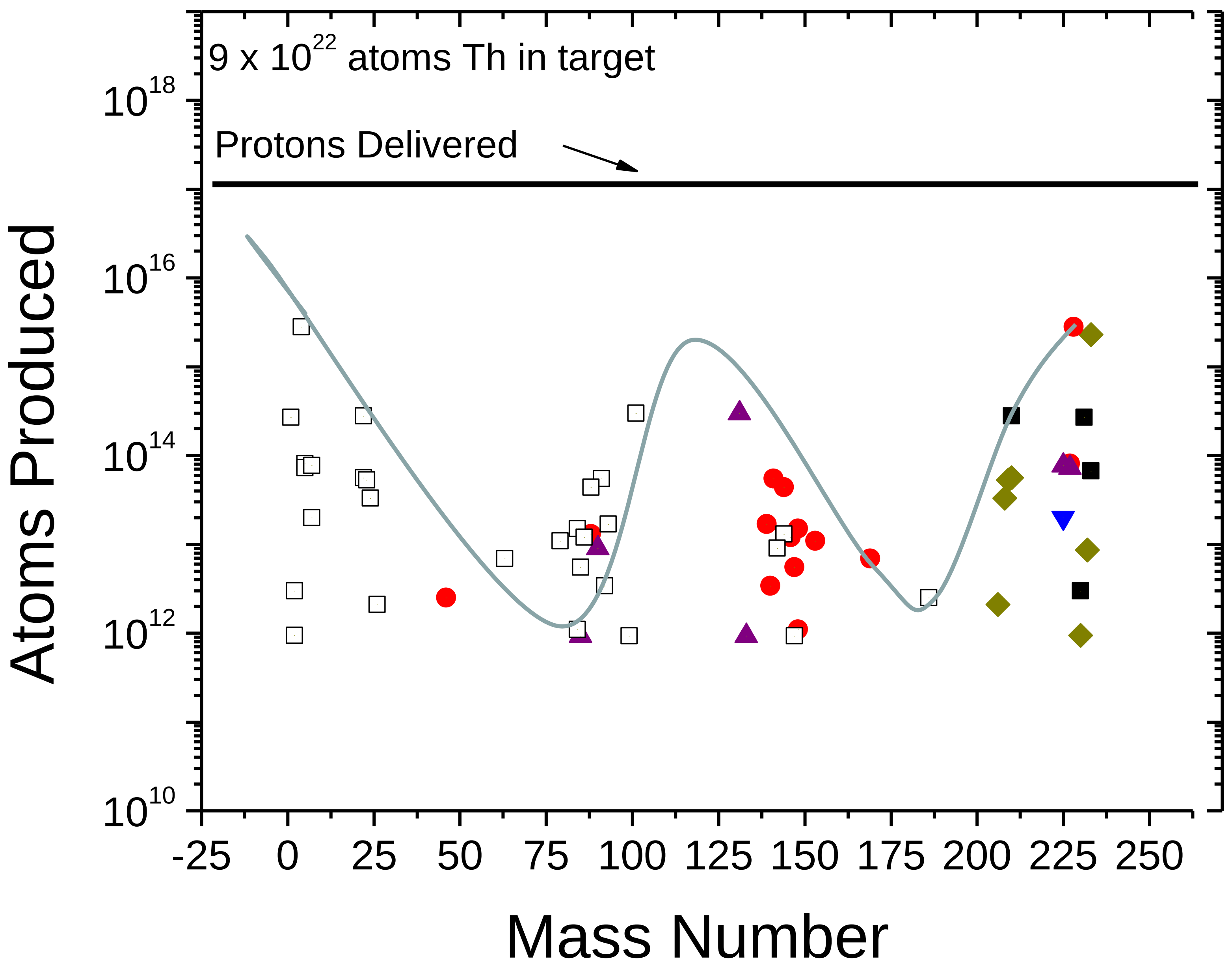
Po Alpha Spectrum (Nickel Disk)

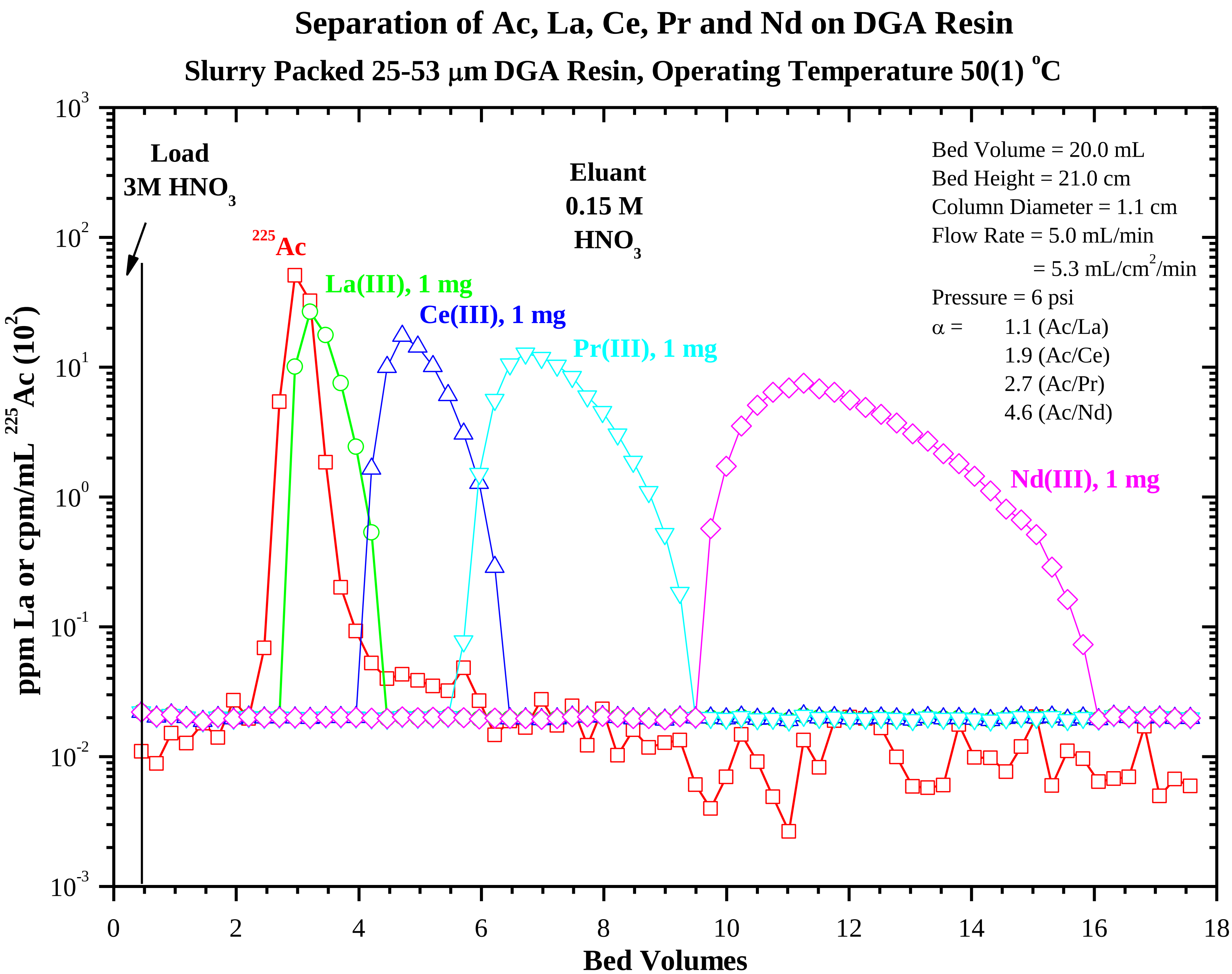


Light Nuclides Formed by Spallation of Thorium Target with Protons

$_{90}Th$	230, 228, 227, 226	$_{63}Eu$	147, 146
$_{89}Ac$	227, 225	$_{61}Pm$	148m
$_{88}Ra$	225, 223	$_{58}Ce$	144, 141, 139
$_{84}Po$	210, 209, 208, 206	$_{56}Ba$	140, 133, 131
$_{82}Pb$	210	$_{39}Y$	88
$_{70}Yb$	169	$_{38}Sr$	90, 85
$_{64}Gd$	153, 148, 146	$_{21}Sc$	46

Spallation Yield for Thorium-232 with 8 GeV Protons

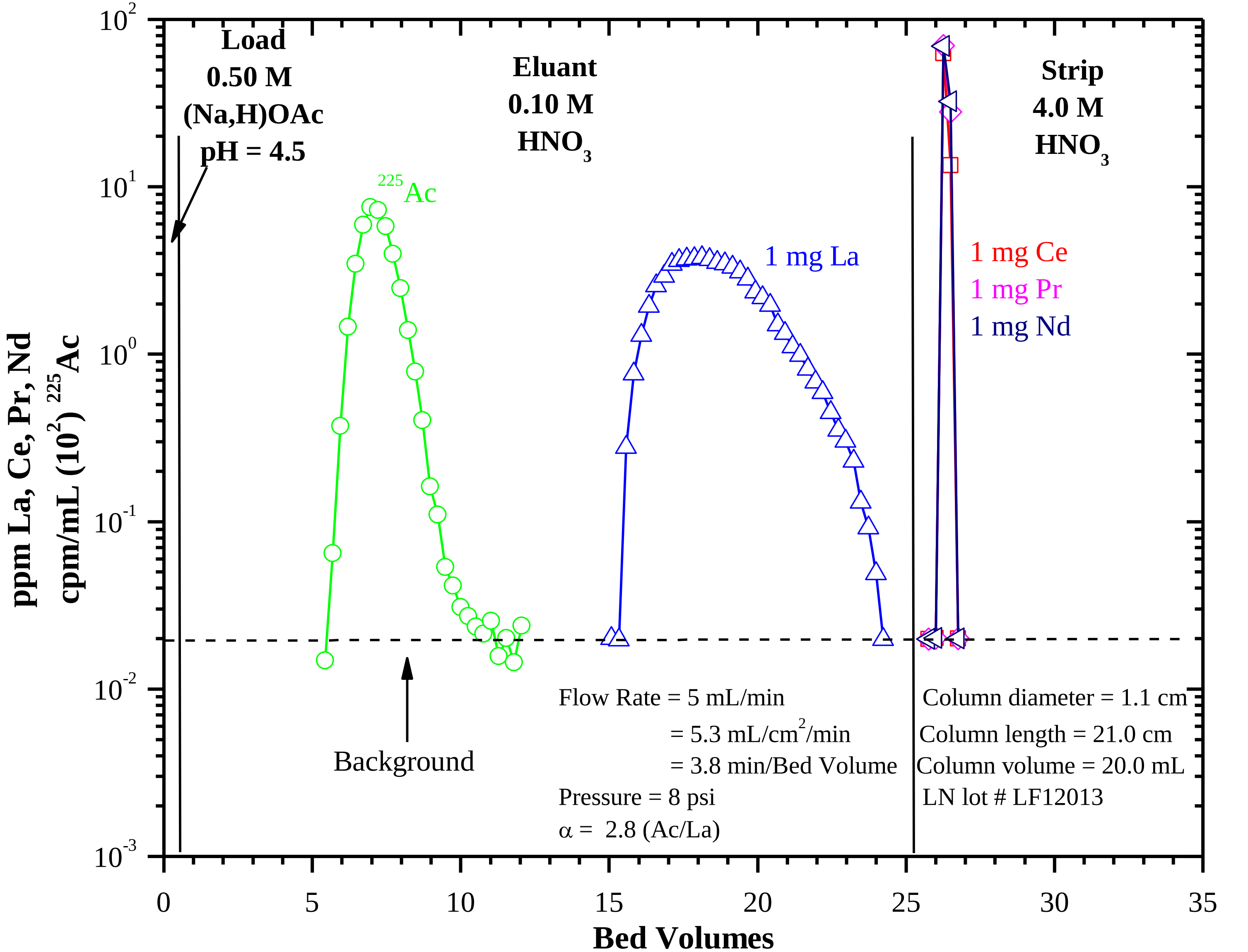




Separation of Ac, La, Ce, Pr and Nd on DGA Resin
Slurry Packed 25-53 μm DGA Resin, Operating Temperature 50(1) °C
Load
3M HNO3
Eluant
0.15 M
HNO3
225Ac
La(III), 1 mg
Ce(III), 1 mg
Pr(III), 1 mg
Nd(III), 1 mg
Bed Volume = 20.0 mL
Bed Height = 21.0 cm
Column Diameter = 1.1 cm
Flow Rate = 5.0 mL/min
= 5.3 mL/cm2/min
Pressure = 6 psi
α = 1.1 (Ac/La)
1.9 (Ac/Ce)
2.7 (Ac/Pr)
4.6 (Ac/Nd)
ppm La or cpm/mL 225Ac (10²)
Bed Volumes
10³
10²
10¹
10⁰
10⁻¹
10⁻²
10⁻³
0
2
4
6
8
10
12
14
16
18

ppm/mL vs. Bed Volumes of Eluate

Slurry Packed 25-53 μm LN Resin, Preconditioned with 0.50 M (Na,H)OAc, 50(1) °C



Conclusion/Future Work

Demonstrated the feasibility of producing Ac-225, Ra-225, Ac-227 using high energy proton (8 GeV) bombardment of Th-232 target (30g).

Repeat using lower energy protons (200-400 meV) and smaller targets ~3g.

- Higher yield of key nuclides
- Fewer bi-products
- Easier processing of targets

Production of Actinium-225 via High Energy Proton Induced Spallation of Thorium-232. **Final Technical Report DE-SC0003602.** http://www.osti.gov/bridge/servlets/purl/1032445/1032445.pdf