Recent Advances in the Recovery and Purification of Actinium Isotopes

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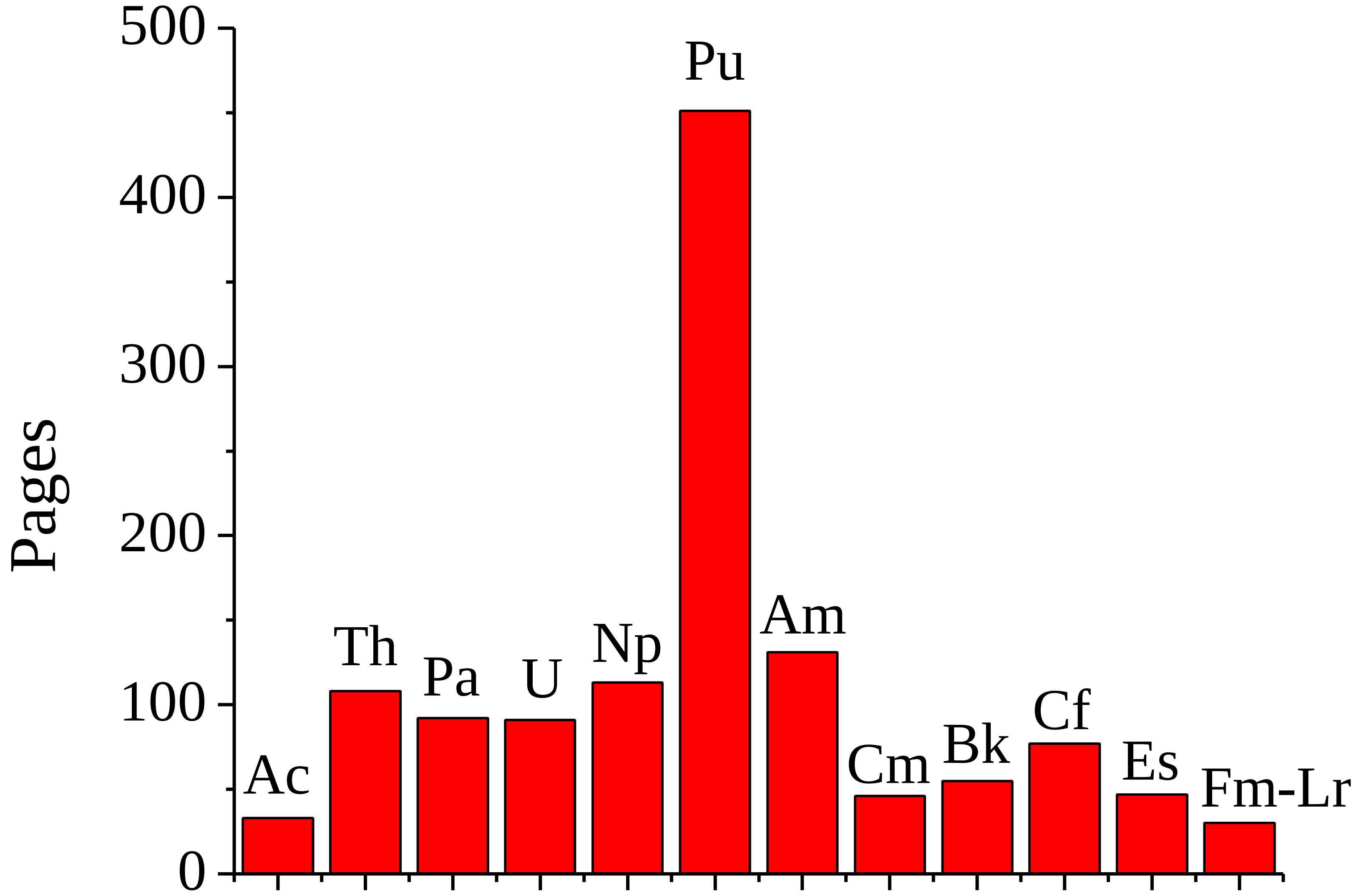
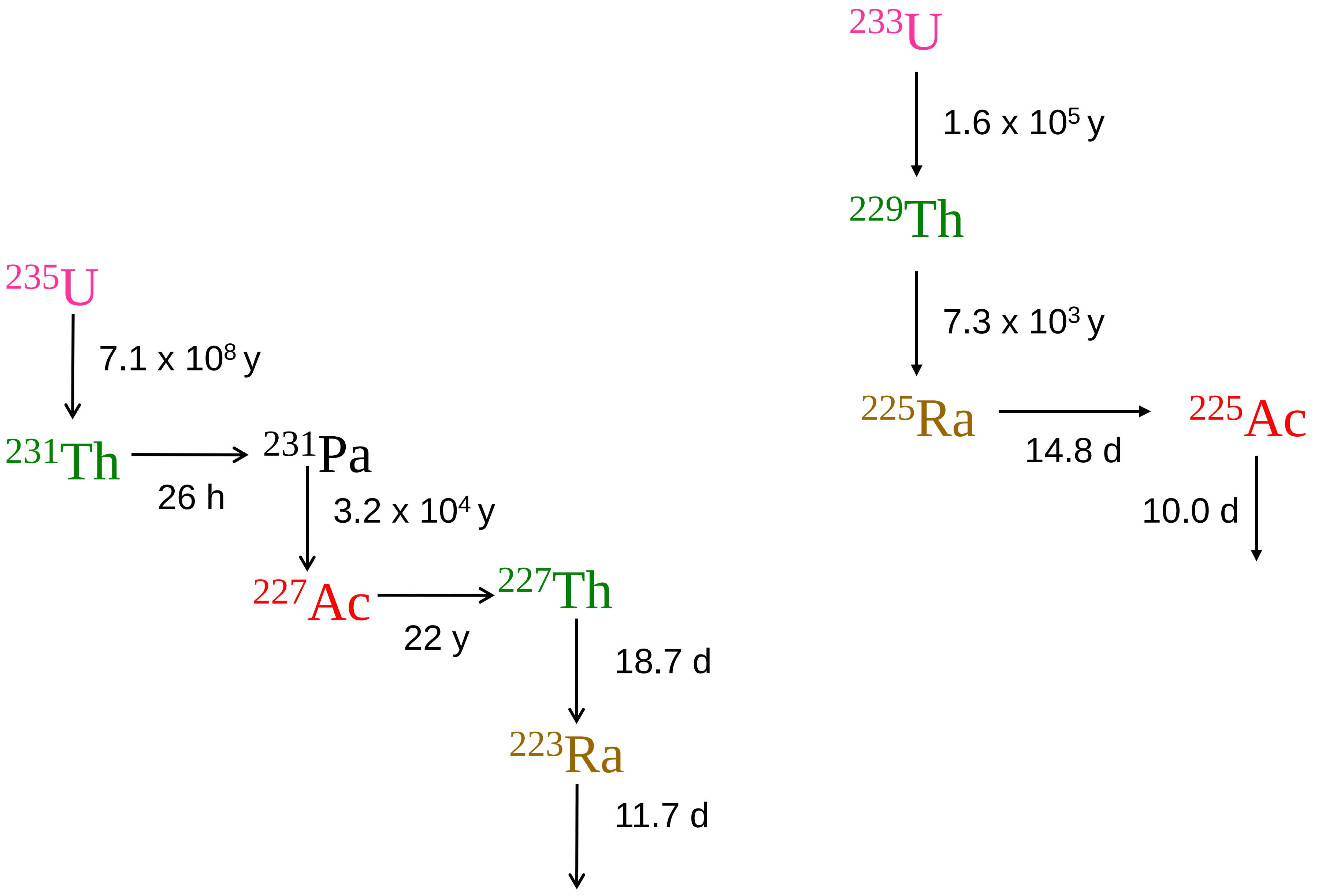
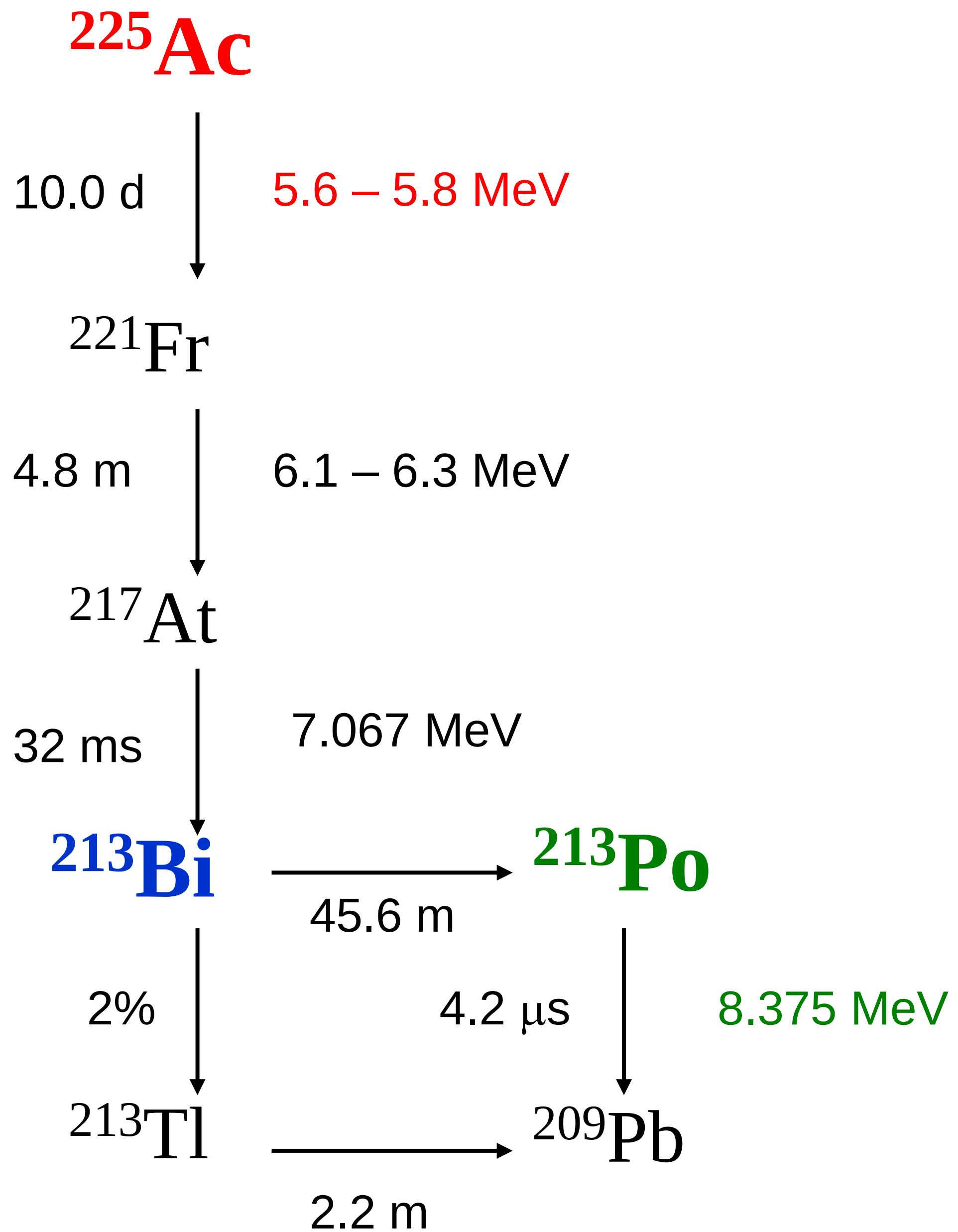
The Chemistry of the Actinide and Transactinide Elements



Sources of Actinium Isotopes



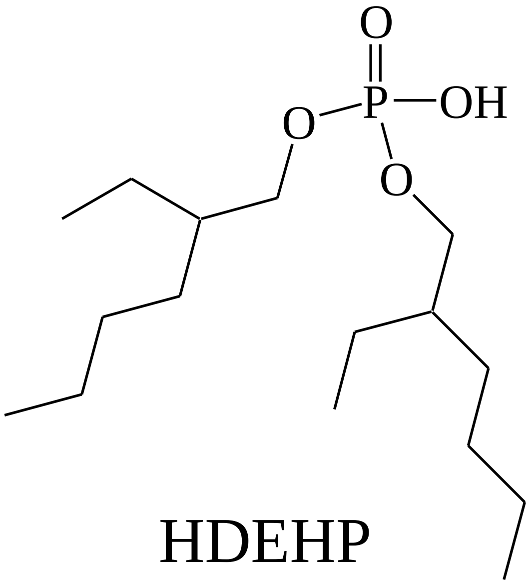
Decay of Actinium-225



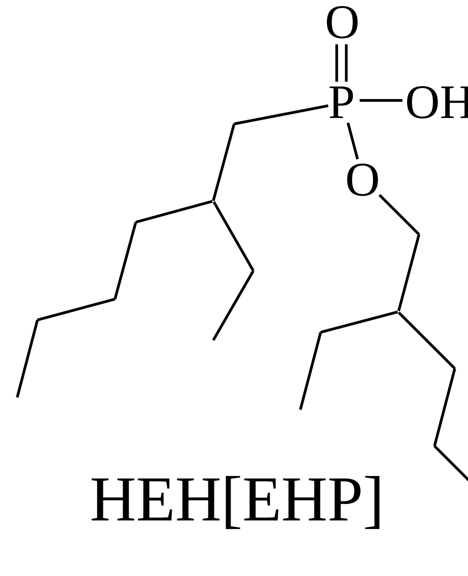
Charge to Radius Ratio for Selected Trivalent Cations

Element	Effective Ionic Radius (CN = 6)	Charge to Radius Ratio
Ac^{3+}	1.12Å	2.68
La^{3+}	1.03Å	2.91
Ce^{3+}	1.01Å	2.97
Pu^{3+}	1.00Å	3.00
Am^{3+}	0.975Å	3.08

Extractants

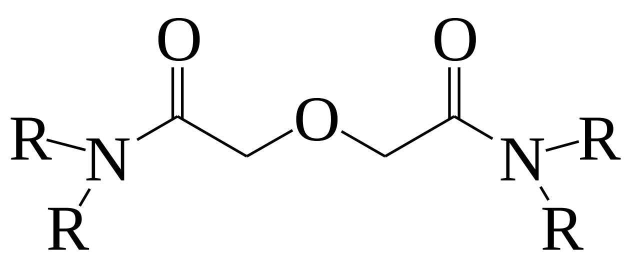


HDEHP



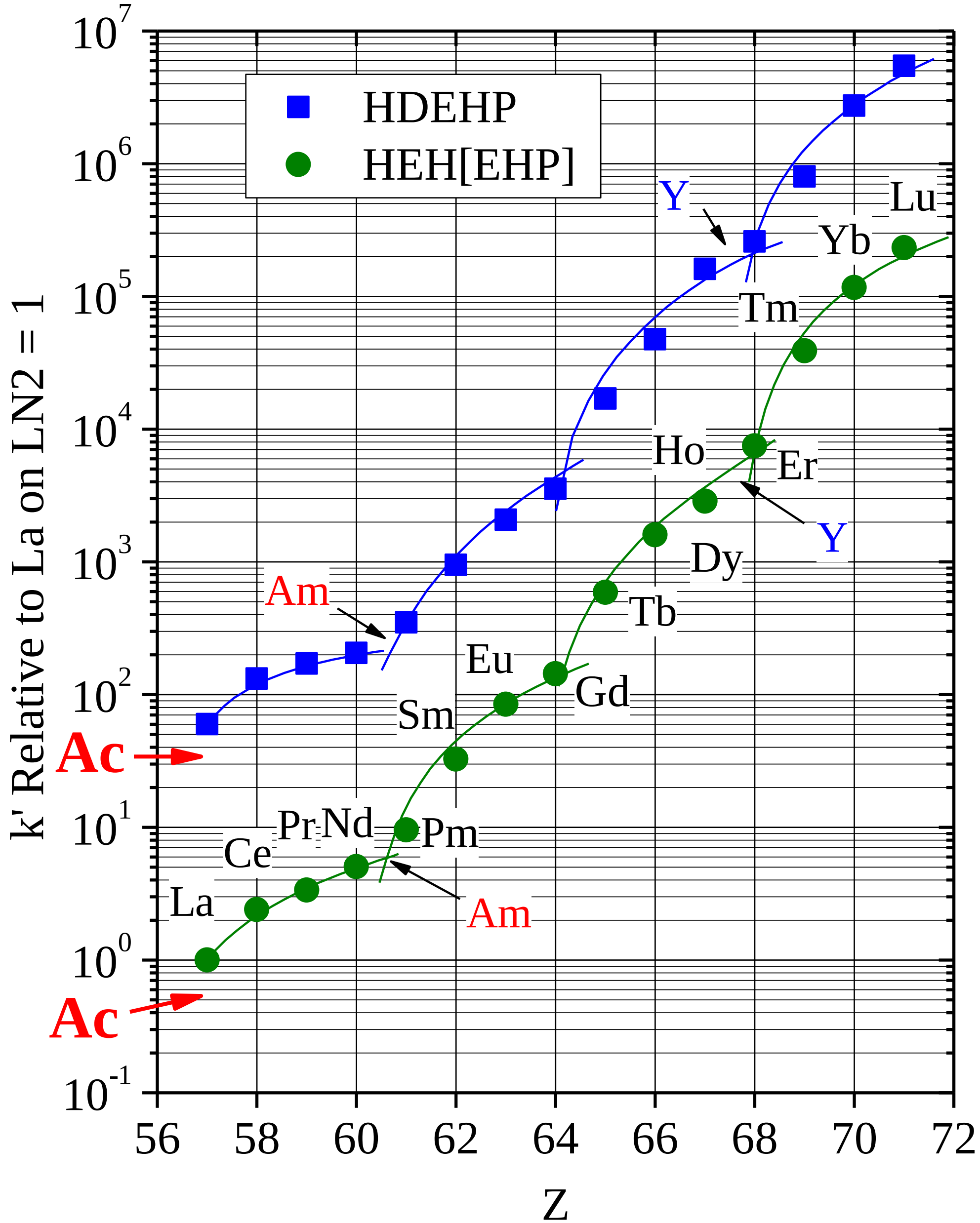
HEH[EHP]

$$\overline{3(\mathrm{HY})_2} + \mathrm{M}^{3+} \leftrightarrow \overline{\mathrm{M}(\mathrm{HY}_2)_3} + 3\mathrm{H}^+$$

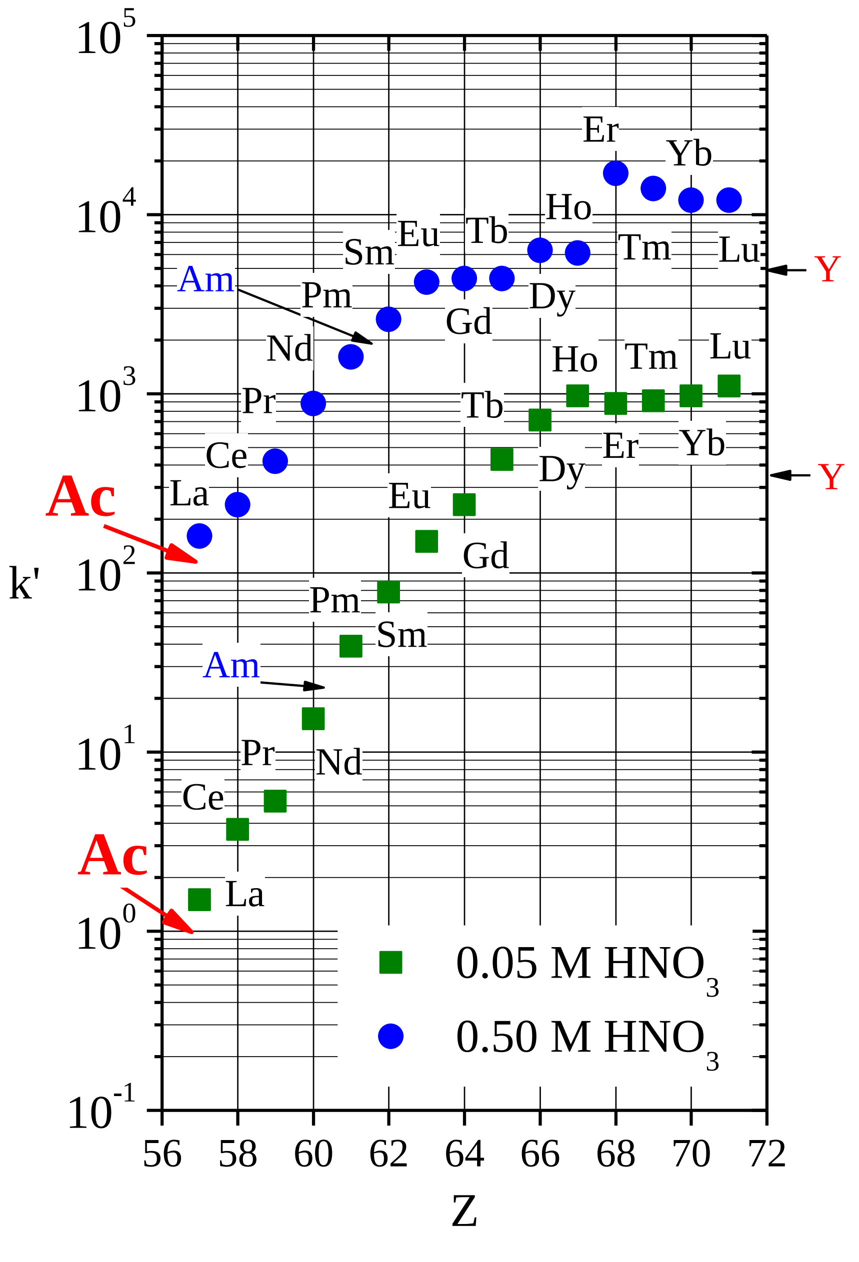


$$\overline{3\mathrm{TODGA}} + \mathrm{M}^{3+} + 3\mathrm{NO}_3^- \leftrightarrow \overline{\mathrm{M}(\mathrm{TODGA})_3(\mathrm{NO}_3)_3}$$

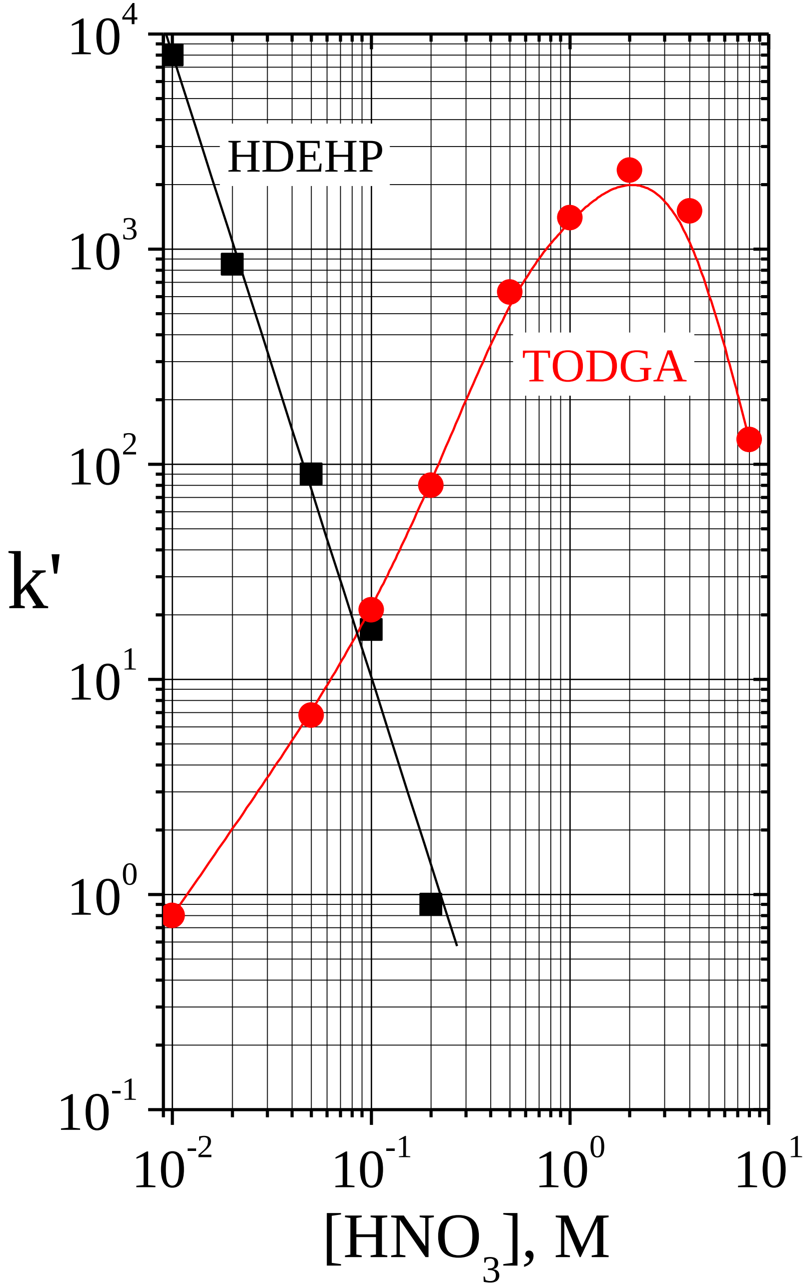
Selectivity of HDEHP and HEH[EHP]

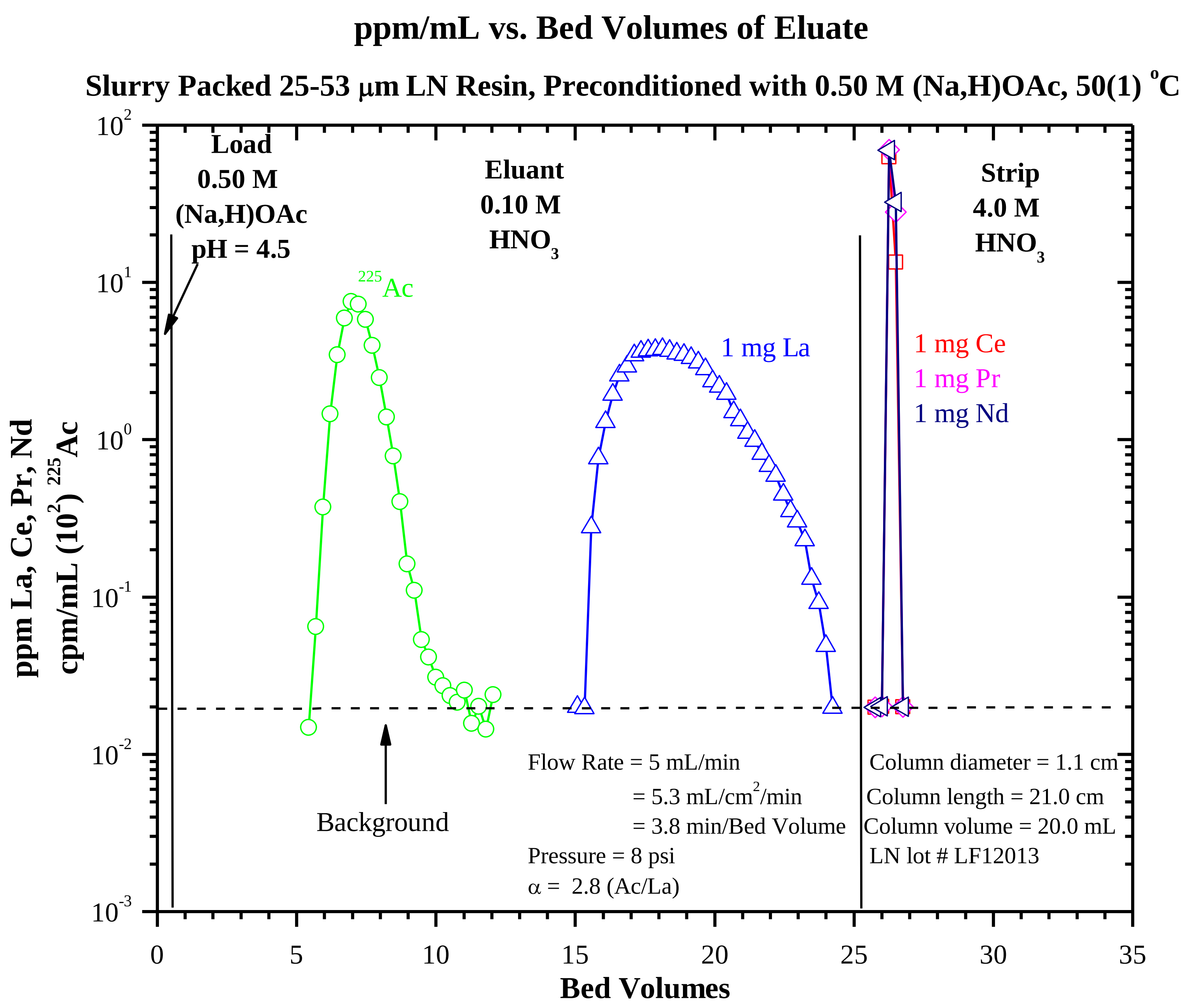


Selectivity of TODGA



k’ Ac from HNO_3 on TODGA and HDEHP Resins




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
Load
0.50 M
(Na,H)OAc
pH = 4.5
Eluant
0.10 M
HNO3
Strip
4.0 M
HNO3
225Ac
1 mg La
1 mg Ce
1 mg Pr
1 mg Nd
Background
Flow Rate = 5 mL/min
= 5.3 mL/cm2/min
= 3.8 min/Bed Volume
Pressure = 8 psi
α = 2.8 (Ac/La)
Column diameter = 1.1 cm
Column length = 21.0 cm
Column volume = 20.0 mL
LN lot # LF12013
ppm La, Ce, Pr, Nd
cpm/mL (10²) 225Ac
Bed Volumes
0
5
10
15
20
25
30
35
10²
10¹
10⁰
10⁻¹
10⁻²
10⁻³

Ac-225 Sources

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

INL-27MT LWBR fuel; ~14MT unirradiated + ~13MT “lightly irradiated” (~5000mCi/month Ac-225)

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

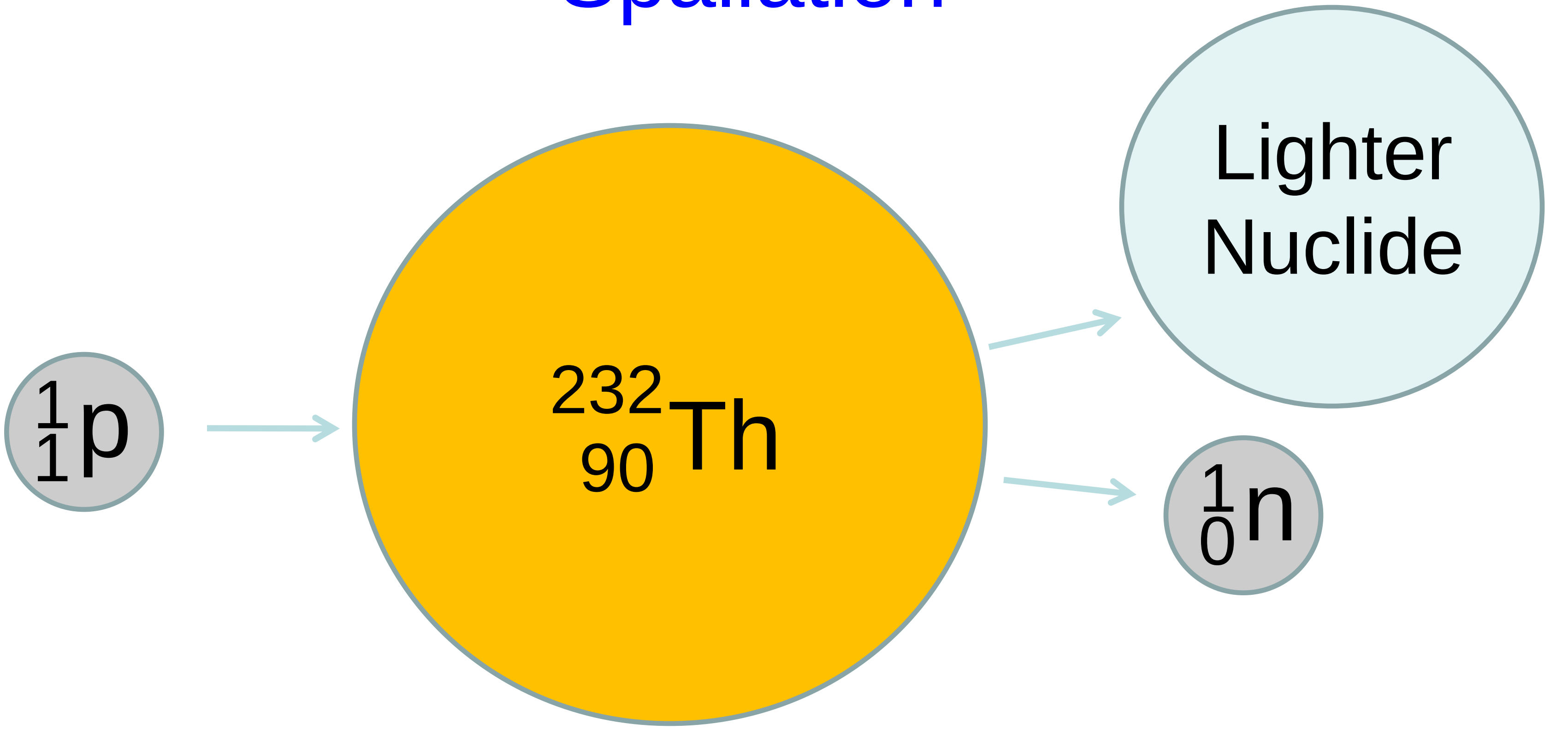
Cyclotron Production via Ra-226(p,2n)Ac-225 (~200mCi/month/cyclotron)

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.

Light Nuclides Formed by Spallation of Thorium Target with Protons

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

Heavy Nuclides Formed by Spallation of Thorium Target with Protons

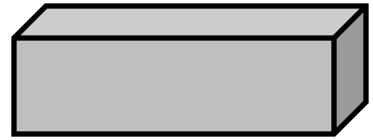
$_{91}Pa$ 233, 231, 230

$_{92}U$ 233, 232, 230

Target Dissolution

Cu clad target
(0.127 cm Cu)

Physical De-cladding →



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

↓

Dissolve in minimum volume of 7M HNO_3 + 0.1M HF (~100mL)

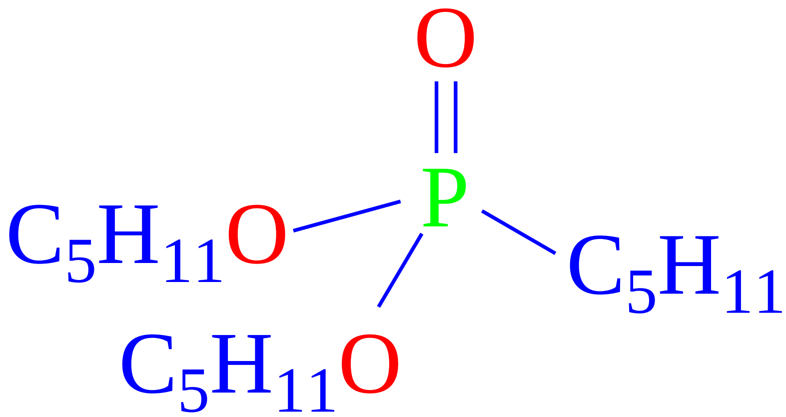
↓

Adjust to 4M HNO_3,
Add Boric acid

↓

SX Process
DA[AP]

U(VI) and Tetravalent Actinide Selective Extractant



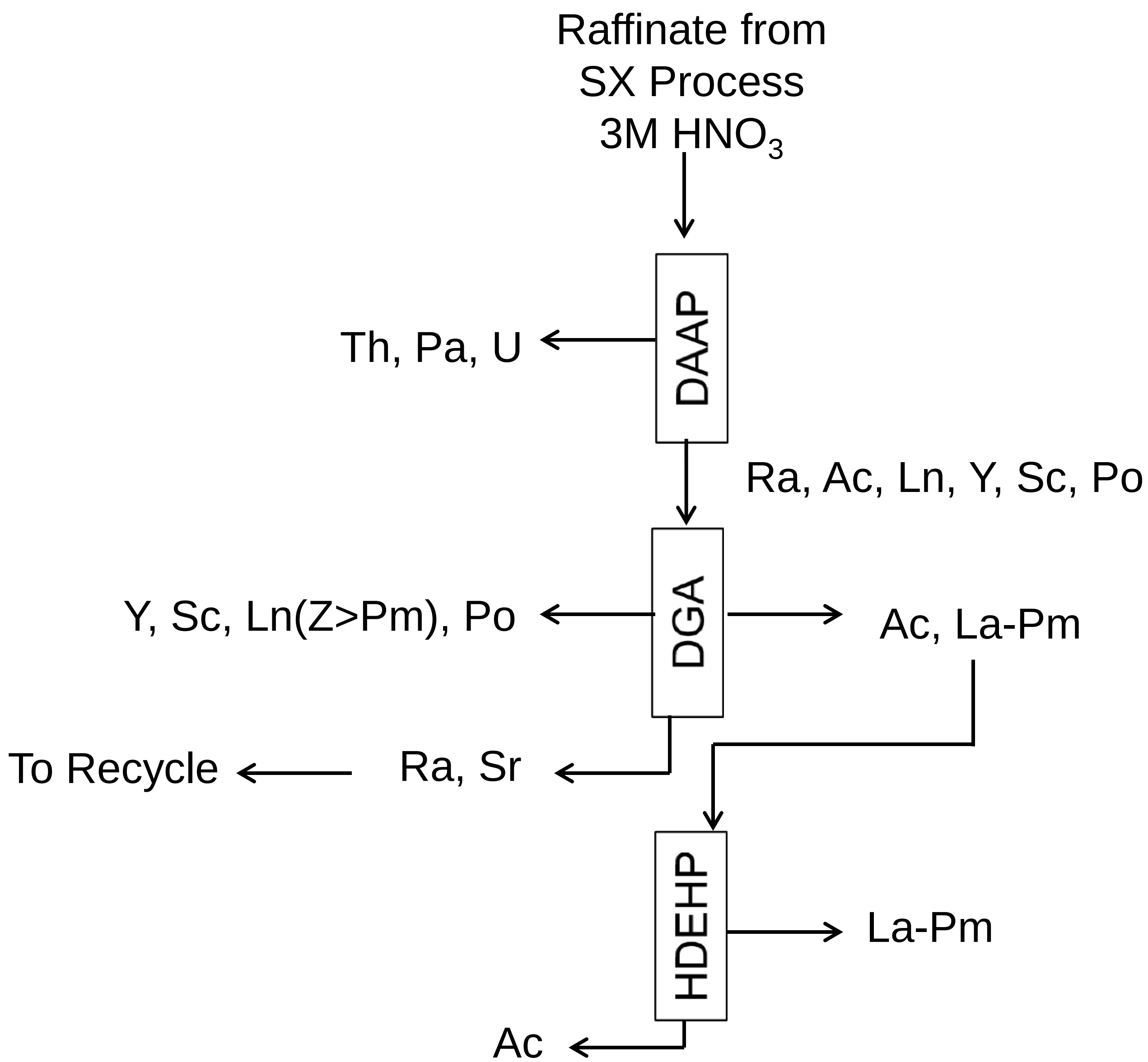
Diamyl amylphosphonate (DA[AP])

$$Th^{4+} + 4NO_3^- + \overline{2E} \leftrightarrow \overline{Th(NO_3)_4 \cdot 2E}$$

$$UO_2^{2+} + 2NO_3^- + \overline{2E} \leftrightarrow \overline{UO_2(NO_3)_2 \cdot 2E}$$

Ac, Ra no significant extraction

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



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

Isotope	Half-life	Atoms	uCi
$_{89}$Ac-225	10 d	7.7×10^{13}	1.7×10^{3}
$_{89}$Ac-226	29 h	N/A	N/A
$_{89}$Ac-227	22 y	7.3×10^{13}	2.0

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