Removing a Needle from a Haystack with a Small Fork: Or can a series of small columns handle a kilogram of matrix?

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Recovery of ^{225}Ac from Aged Light Water Breeder Reactor Fuel

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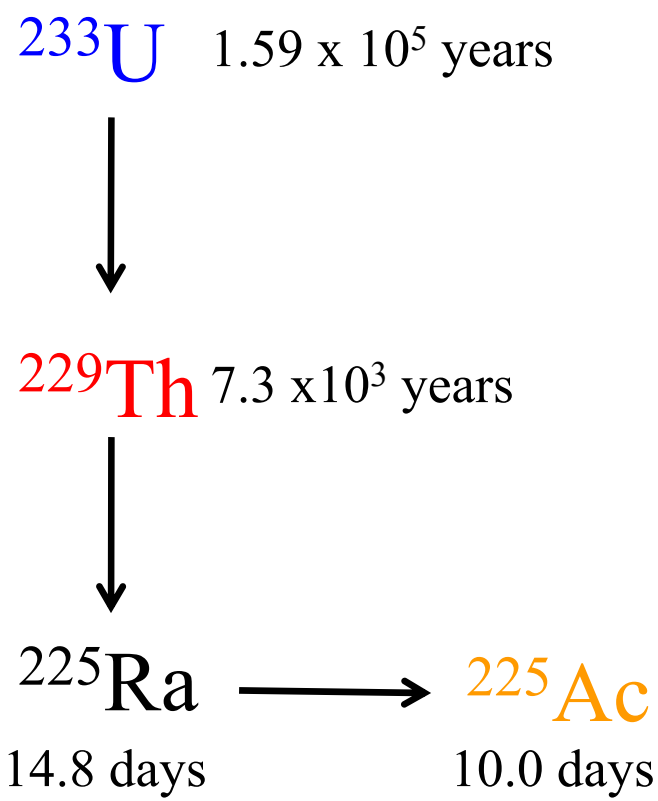
-A chemical process has been developed and patented, no. 7,157,061, to separate ^{225}Ac from dissolved LWBR fuel (97% ^{232}Th and 3% ^{233}U, as oxides)

-Laboratory scale studies funded by CRADA with Northstar Nuclear Medicine completed on actual aged LWBR fuel pellets

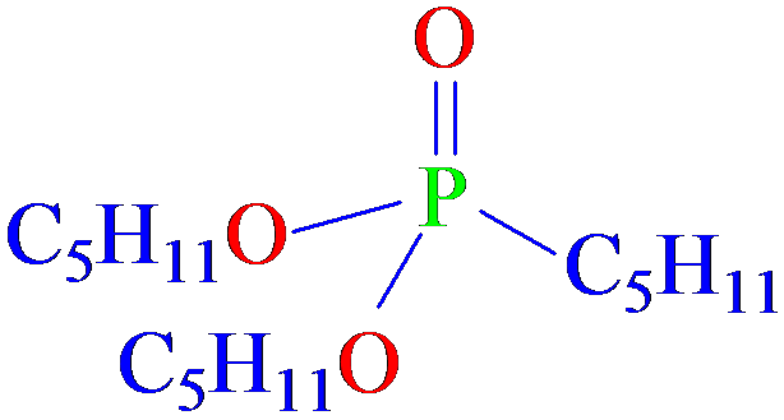
-Aged ^{233}U, via in-growth of ^{229}Th, is the best source of ^{225}Ac

-LWBR fuel, 97% ^{232}Th, thought unusable for ^{225}Ac recovery because the ^{229}Th is not isolatable.

Decay of ^{233}U



Diamyl amylphosphonate (DA[AP])

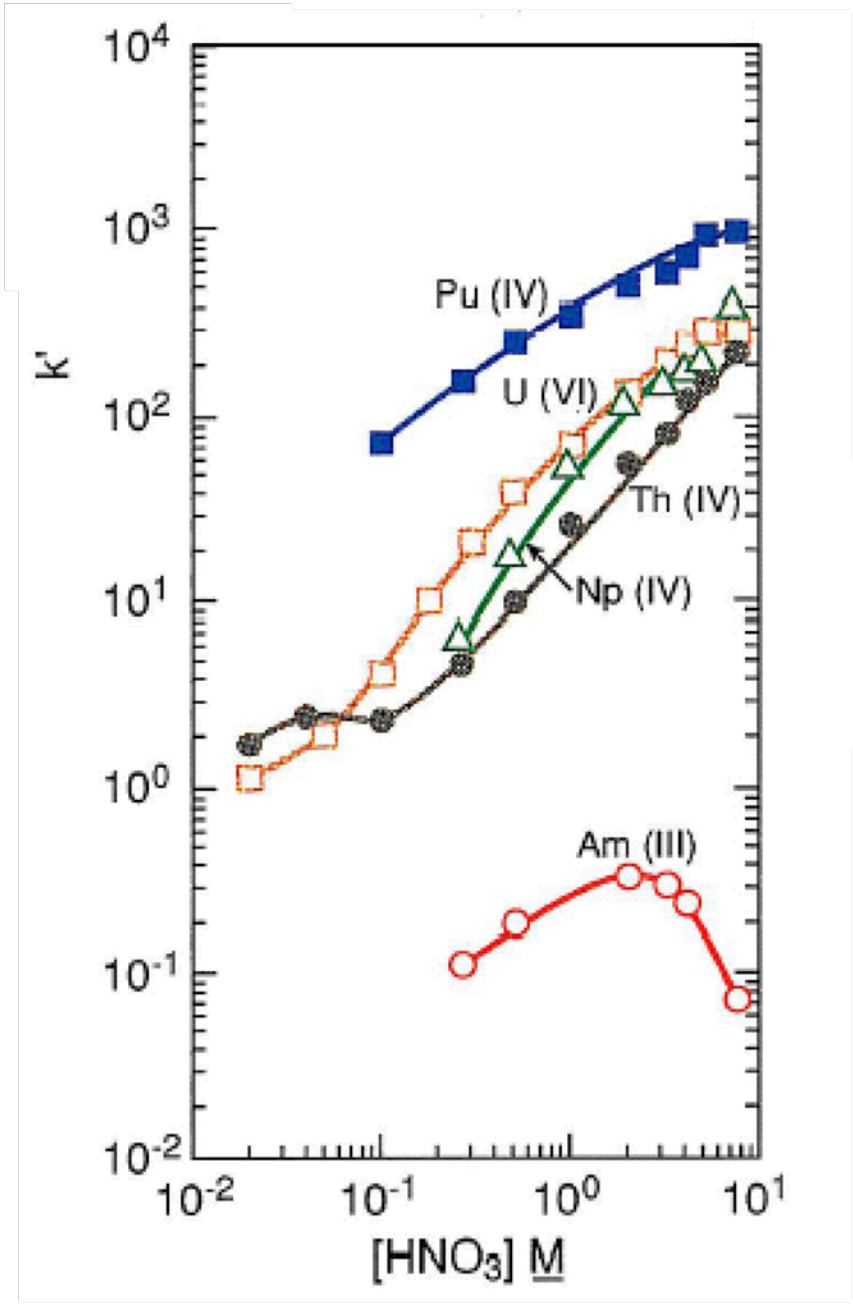


- Very selective for Th and U from acidic nitrate media

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

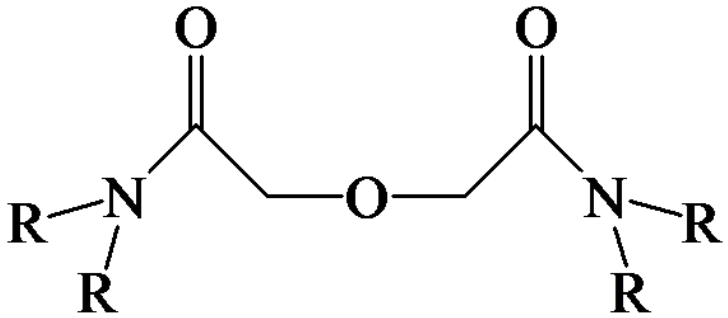
E = DA[AP]

k' for Actinides on UTEVA Resin vs HNO_3



UTEVA Resin is 40% (w:w) DA[AP] on an inert support

Diglycolamide Extractant



R = *n*-octyl

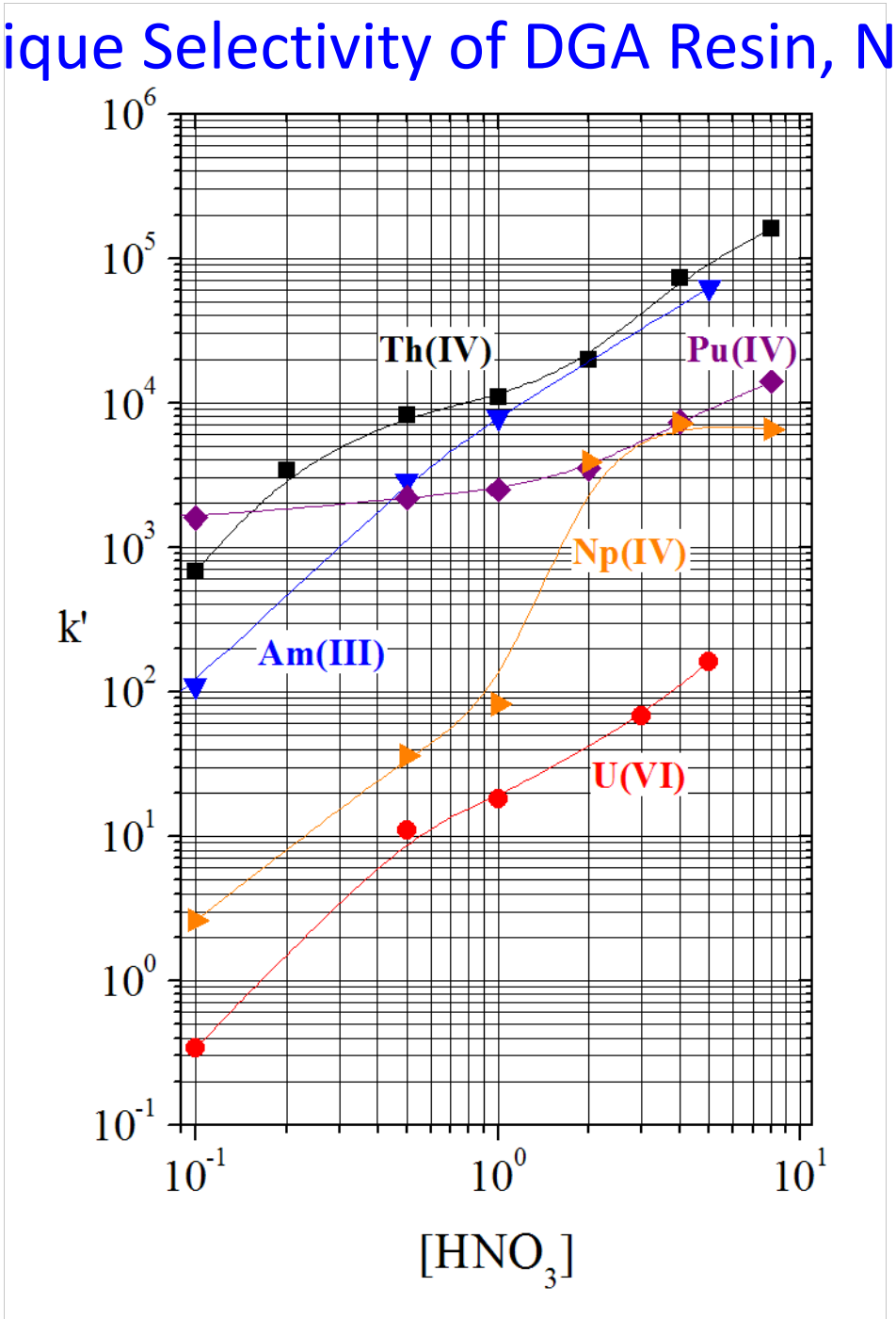
DGA or DGA, Normal

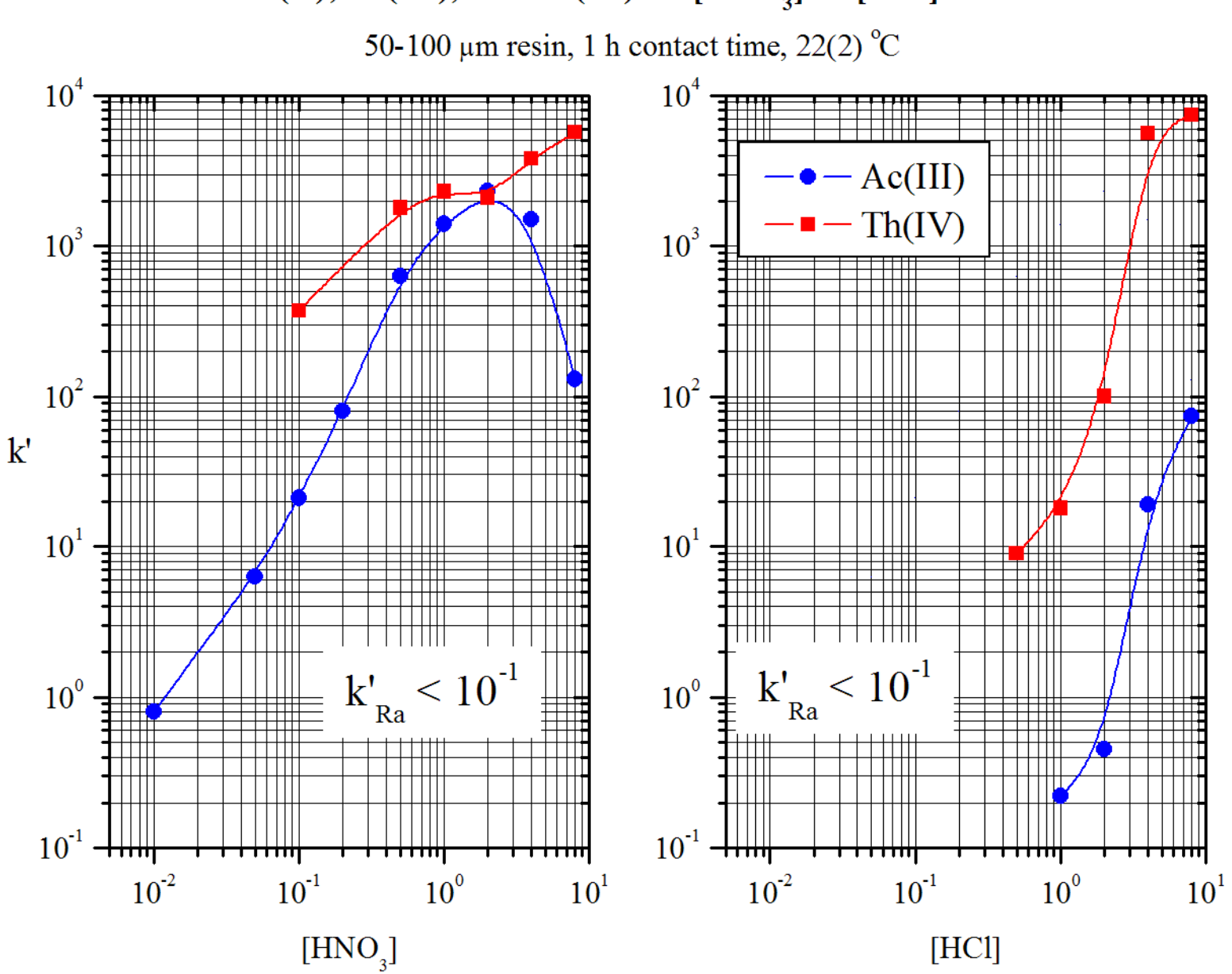
$$\overline{3DGA} + Am^{3+} + 3X^{-} + aHX \leftrightarrow \overline{Am(DGA)_3(X)_3.(HX)_a}$$

X = NO_3^- or Cl^-

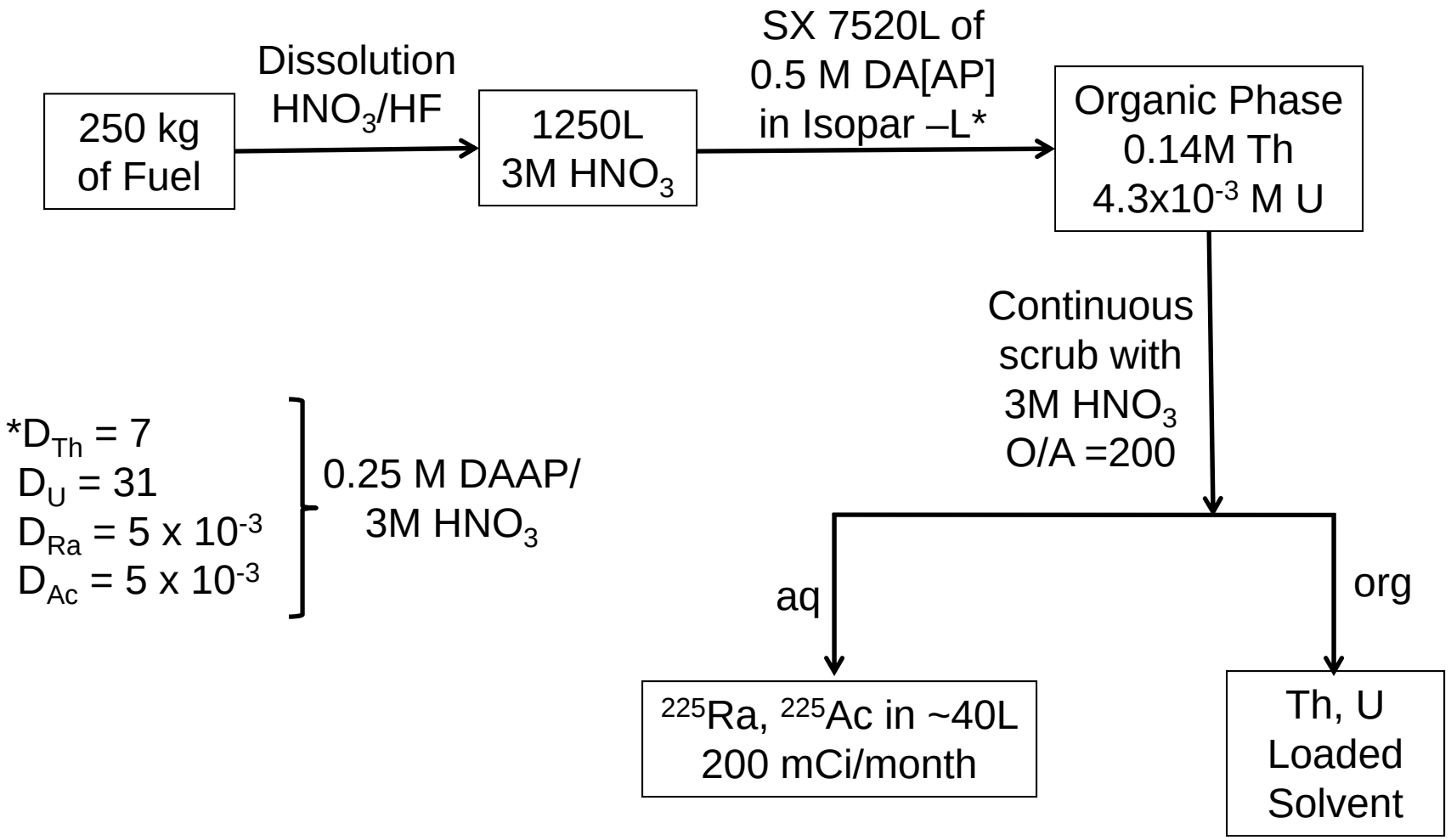
Unique Selectivity: An(III) ~ An(IV) > U(VI)

Unique Selectivity of DGA Resin, Normal

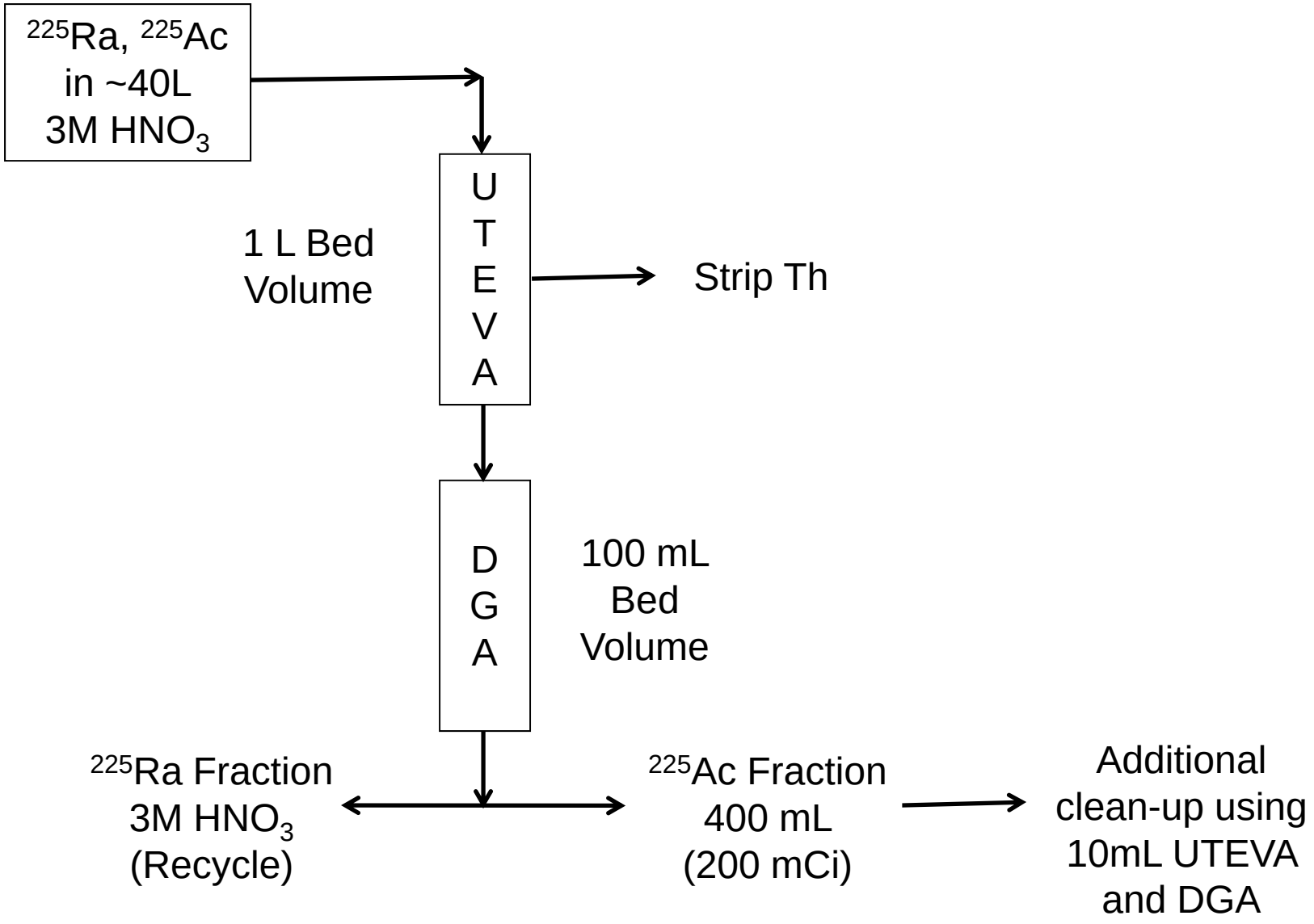



k' Ra(II), Ac(III), and Th(IV) vs. [HNO3] or [HCl] on DGA Resin
50-100 µm resin, 1 h contact time, 22(2) °C
k'
[HNO3]
[HCl]
Ac(III)
Th(IV)
k'Ra < 10-1
k'Ra < 10-1

Conceptual Flowsheet for the Recovery of ^{225}Ra and ^{225}Ac from INL Breeder Reactor Fuel



^{225}Ra and ^{225}Ac Flowsheet (continued)



Flowsheet Tested on 75g of LWBR Fuel

-75 g of fuel was dissolved with 150 mL of 13M HNO3/0.03M HF in a PARR Reactor at 107^{o}C for 24 hours

-Overall recovery of ^{225}Ac (80 μCi) 80% yield

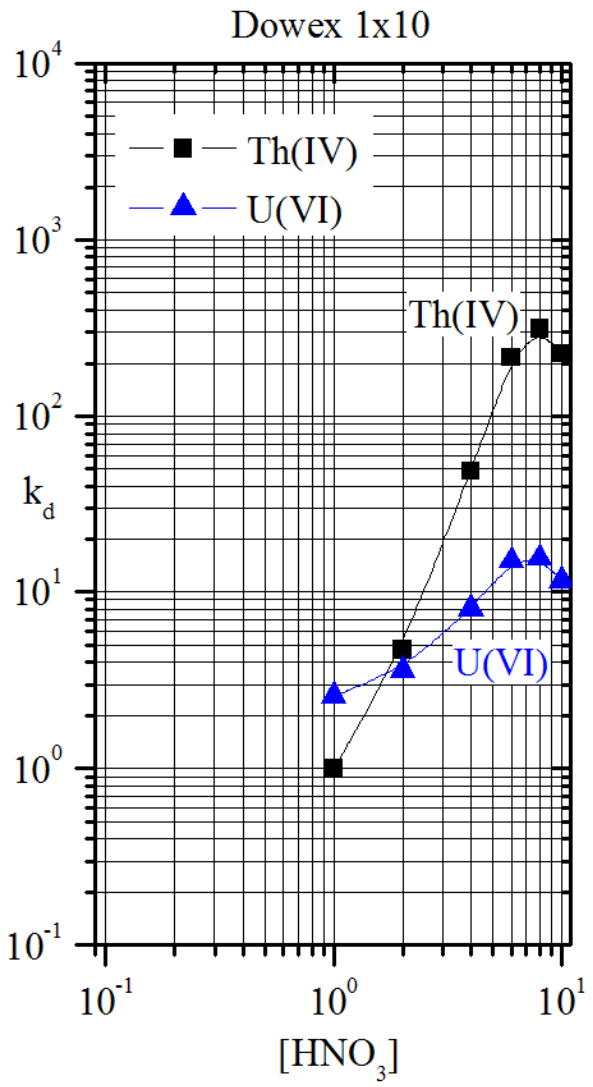
- -D.F. from ^{229,232}Th and ^{233}U > 10^{11}
- -D.F. from Ra > 10^{8}
- -Impurities <<ug/mCi of ^{225}Ac

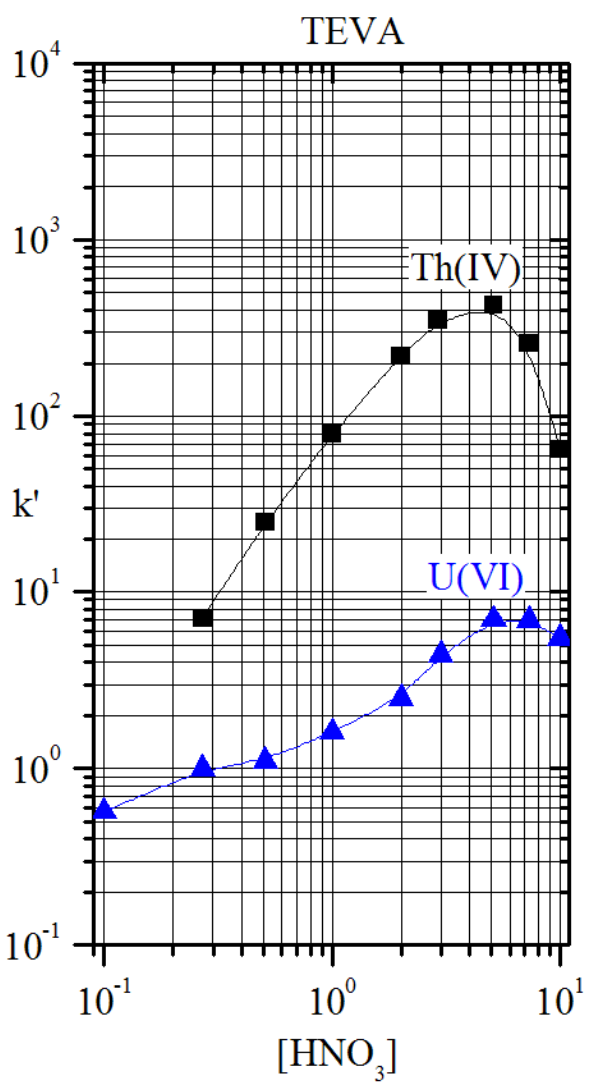
Separation of Trace Th from Macro Uranium

- ^{234}Th from ^{238}U (useful laboratory tracer)

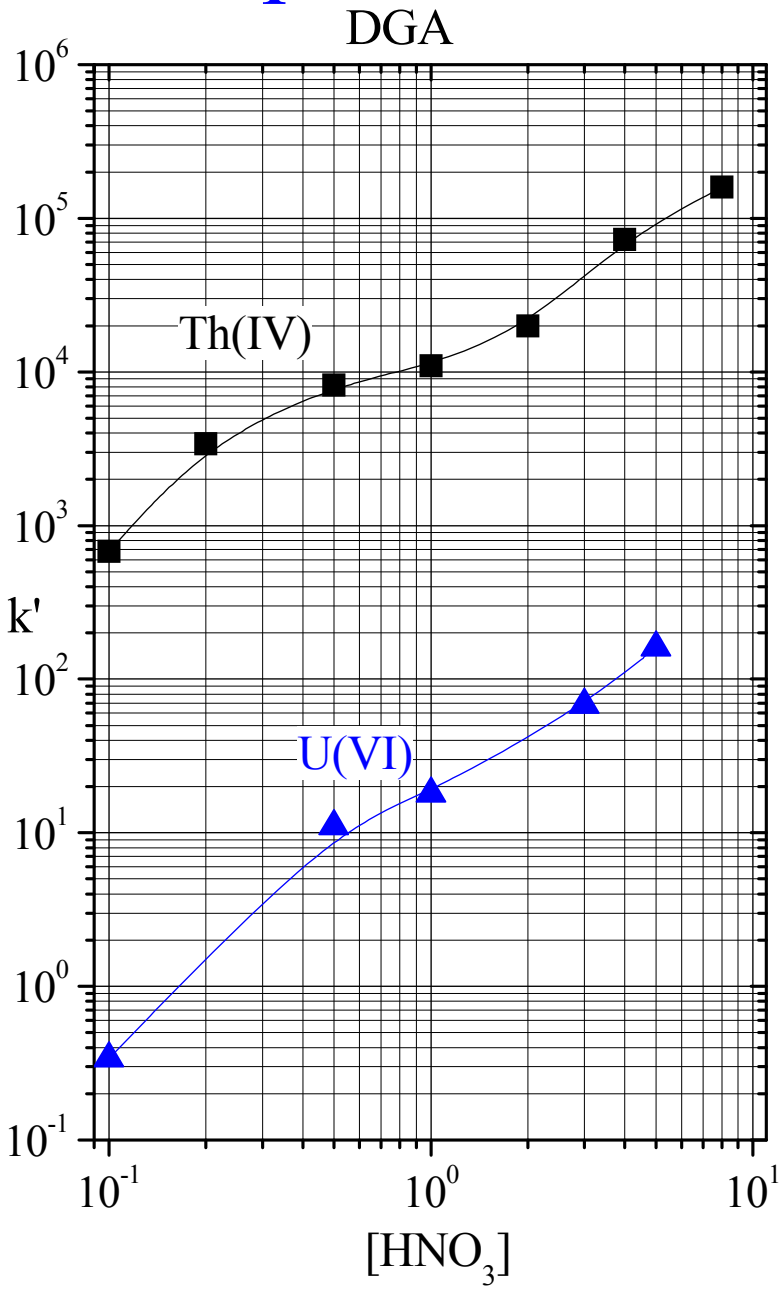
- ^{229}Th from ^{233}U (parent of ^{225}Ac, medical radioisotope)

Th/U Separation Factor

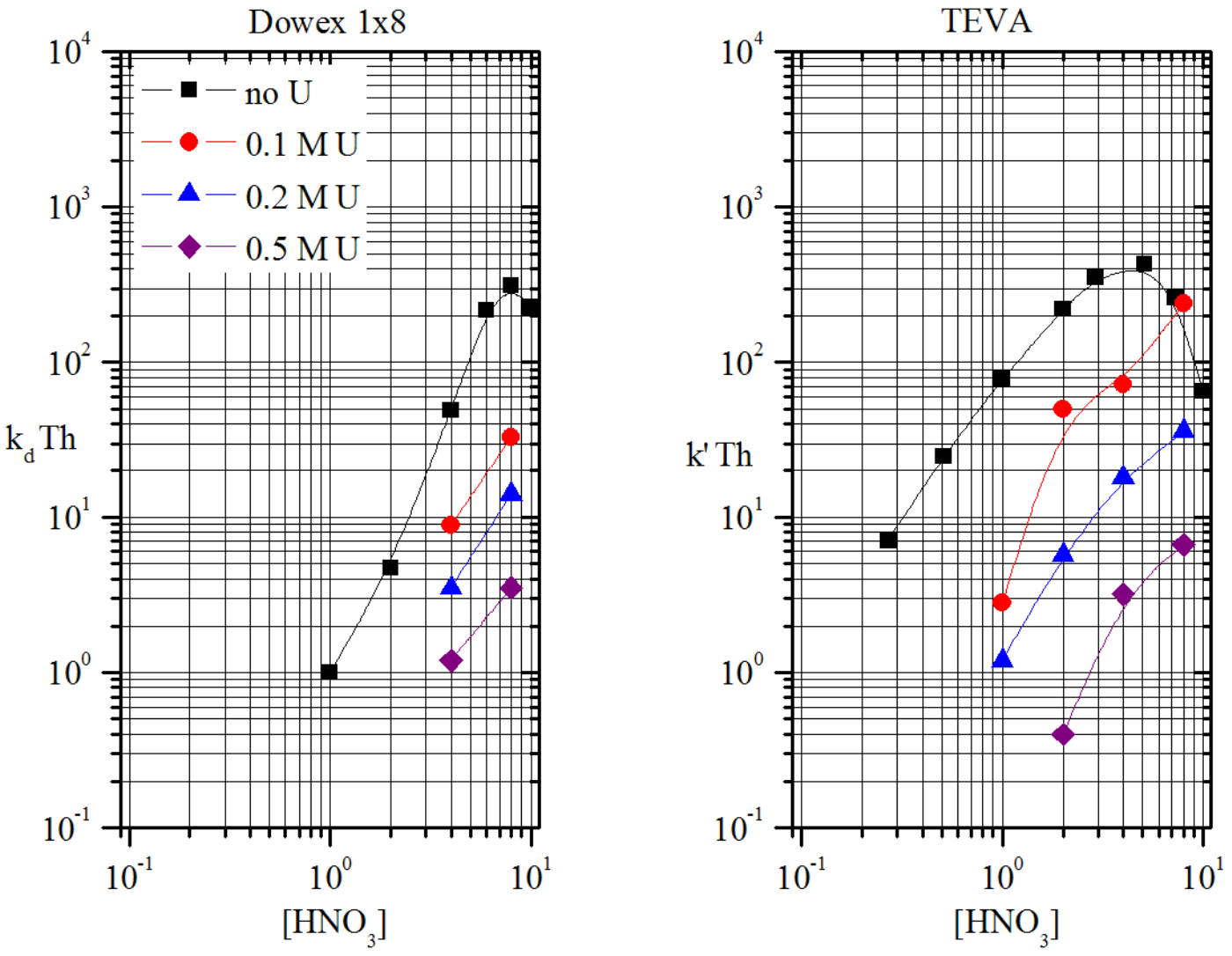




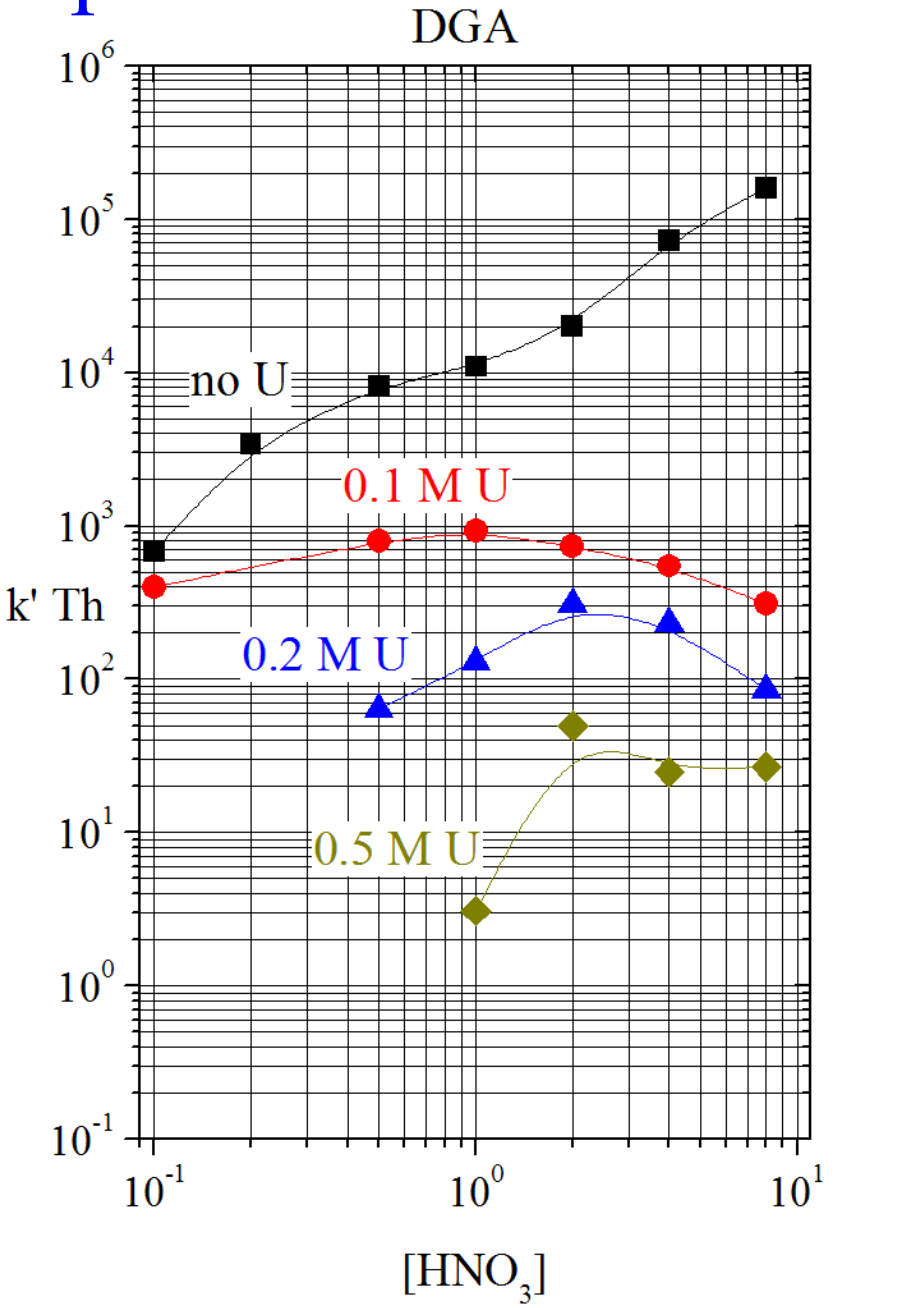
Th/U Separation Factor



Th uptake vs U concentration



Th uptake vs U concentration



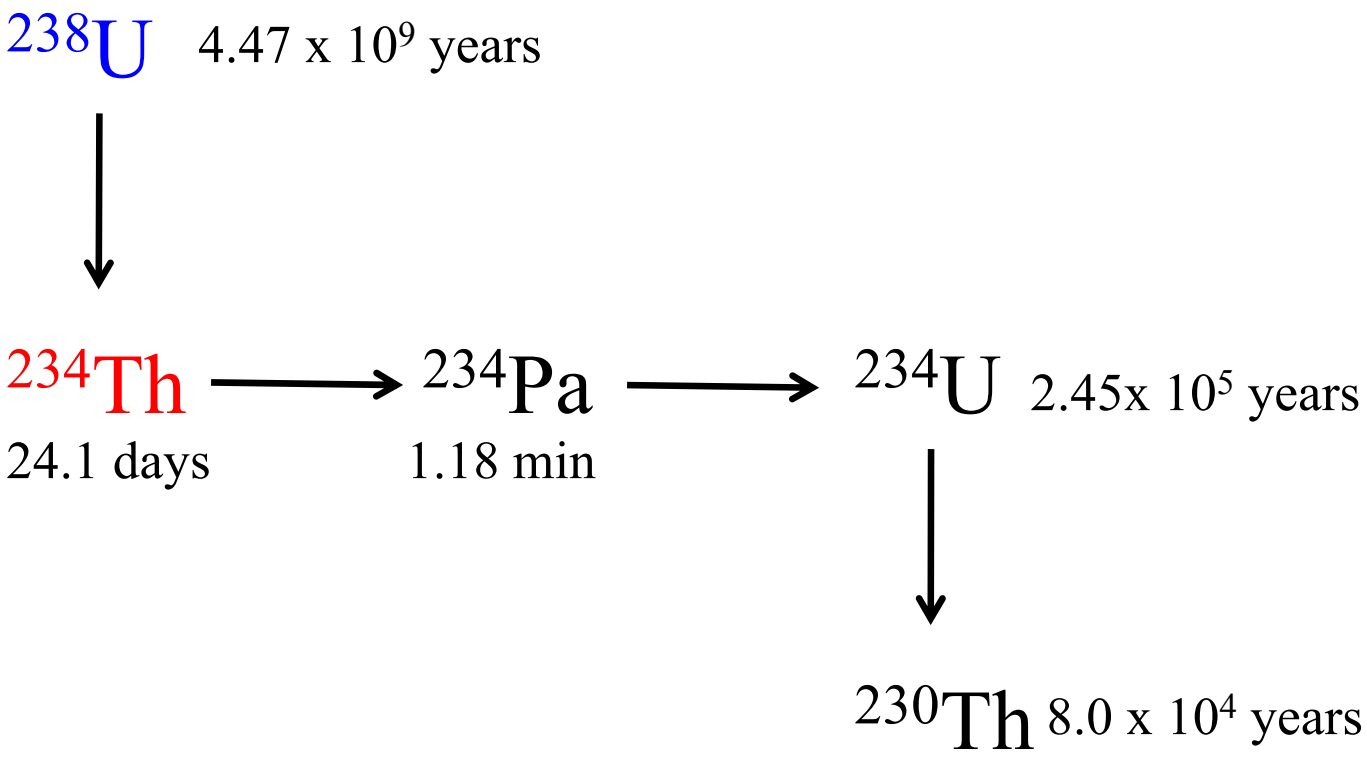
Optimization for k' for Th

Resin	k'	Acid
Dowex 1x8	300	8M HNO_3
TEVA	400	4M HNO_3
DGA	$>10^5$	8M HNO_3
	$>10^4$	4M HNO_3

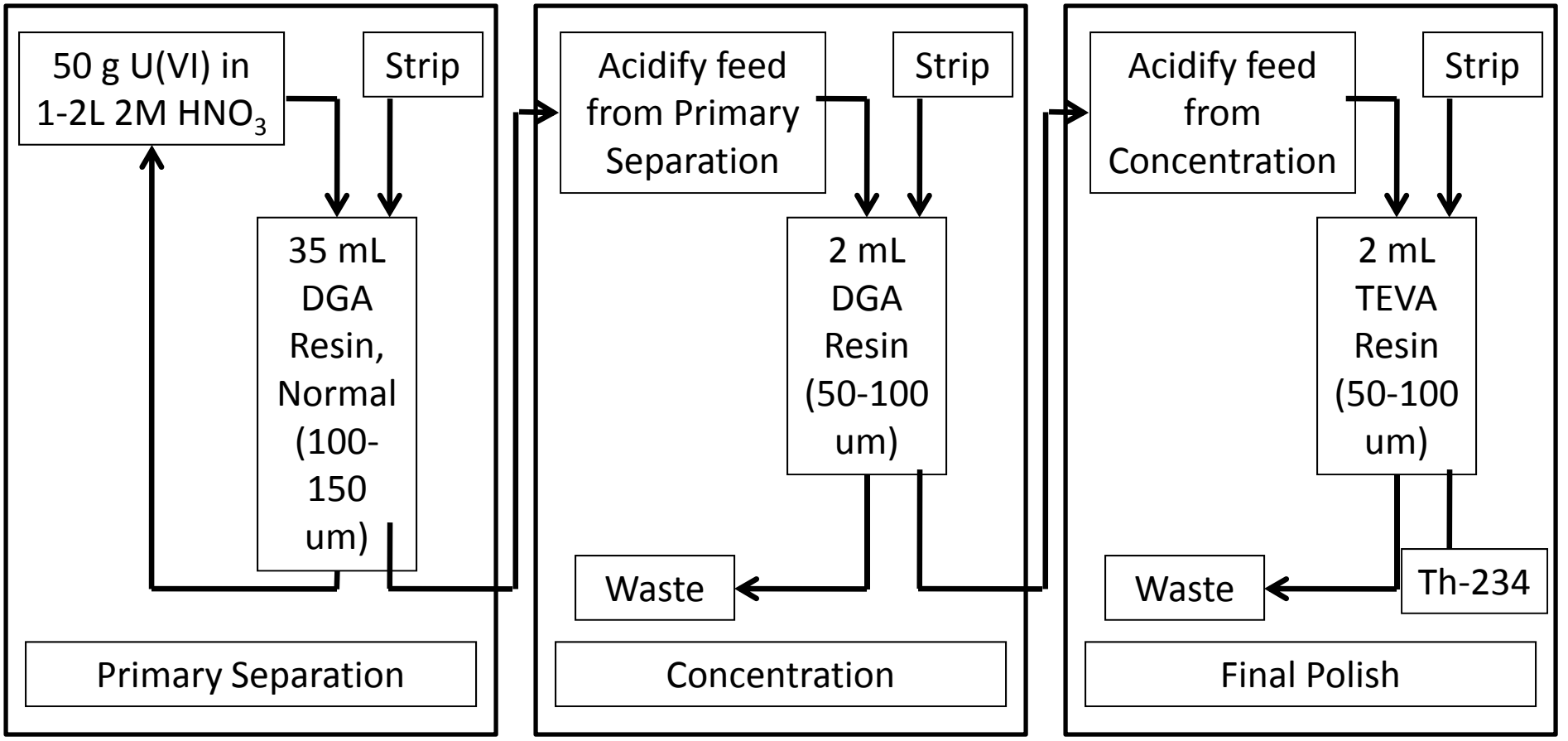
Optimization for k' for Th from 0.2M U

Resin	k'	Acid
Dowex 1x8	14	8M HNO_3
TEVA	20	4M HNO_3
DGA	300	2M HNO_3

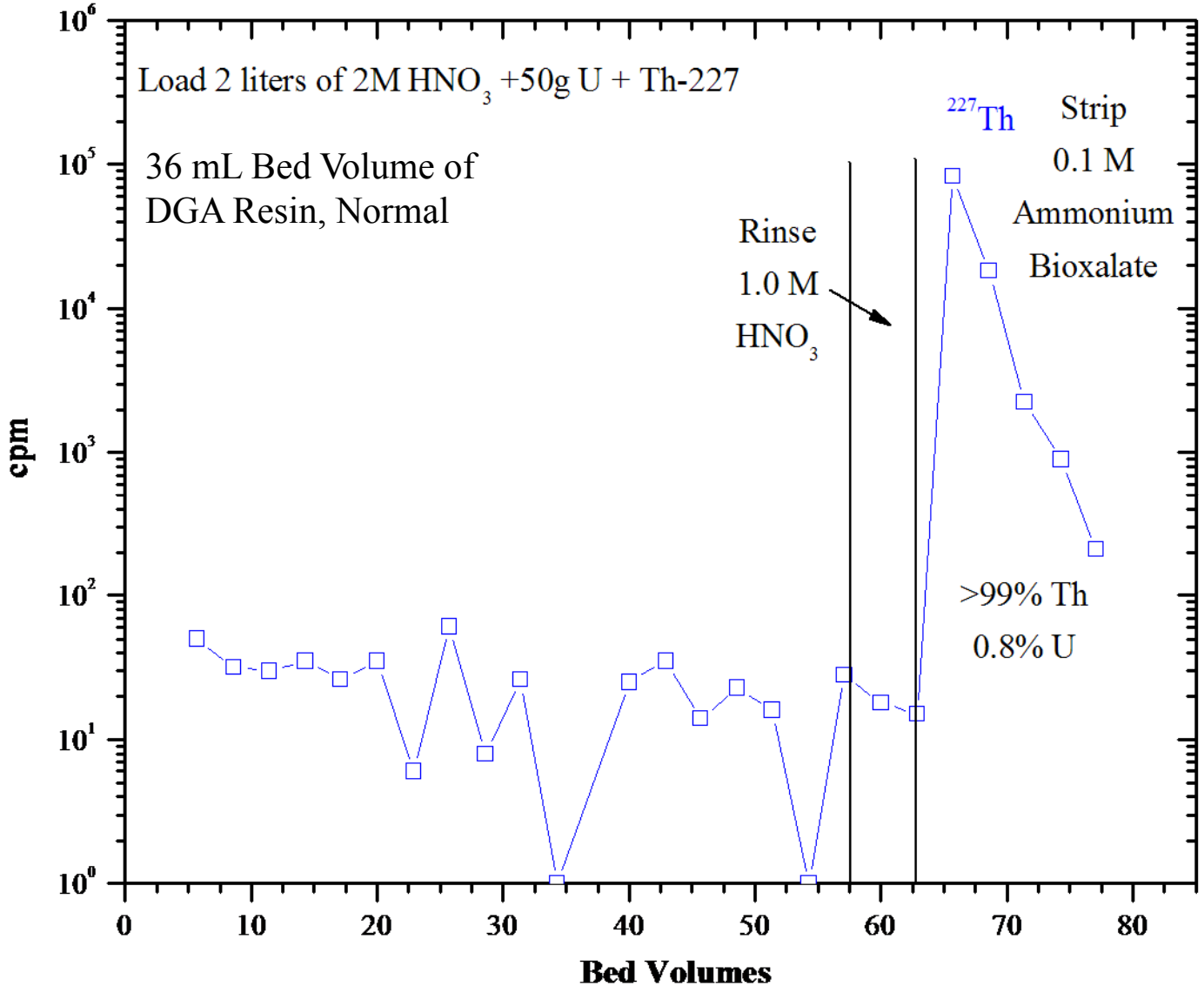
Decay of ^{238}U



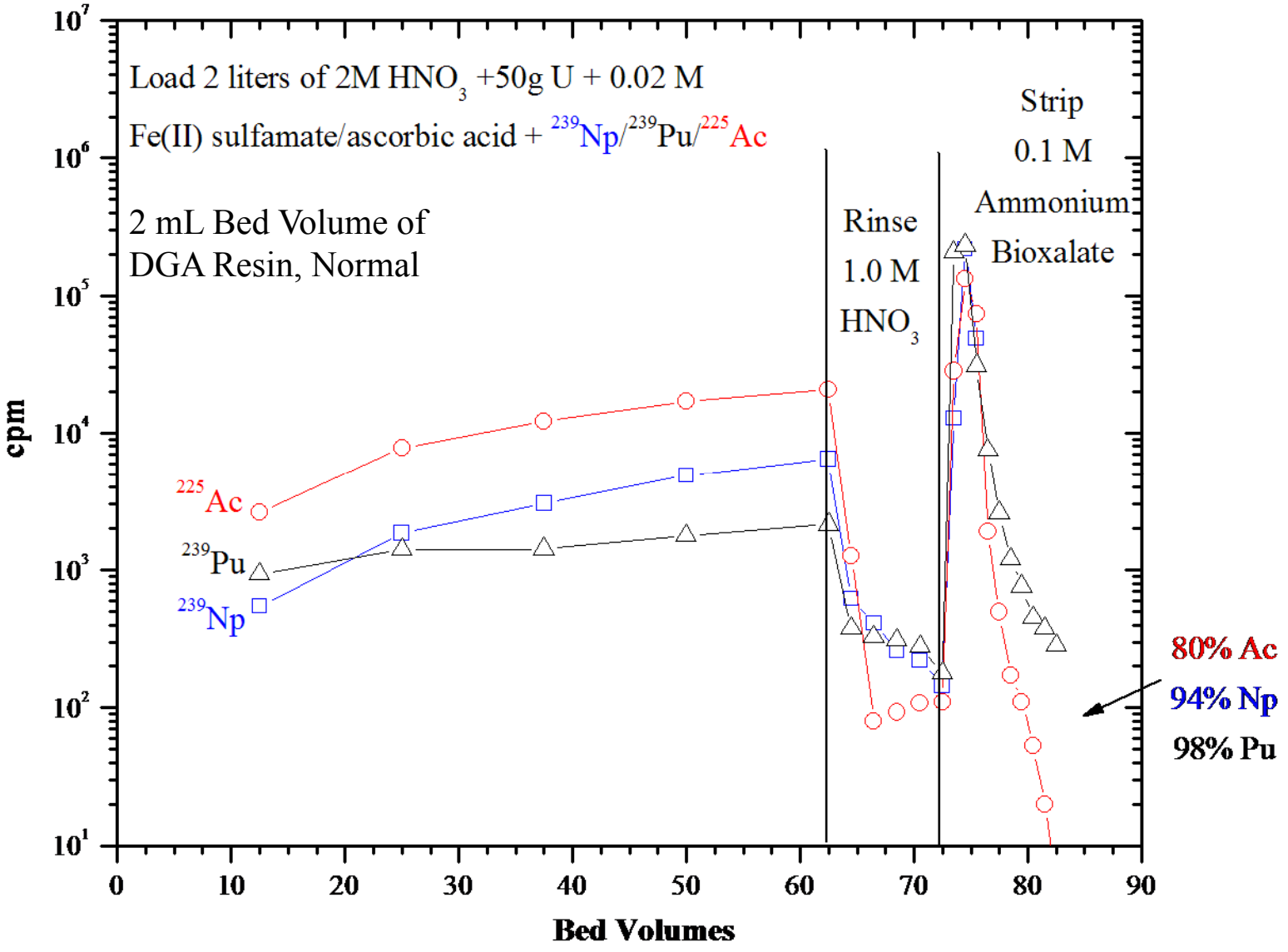
^{234}Th Separation from 50 g ^{238}U



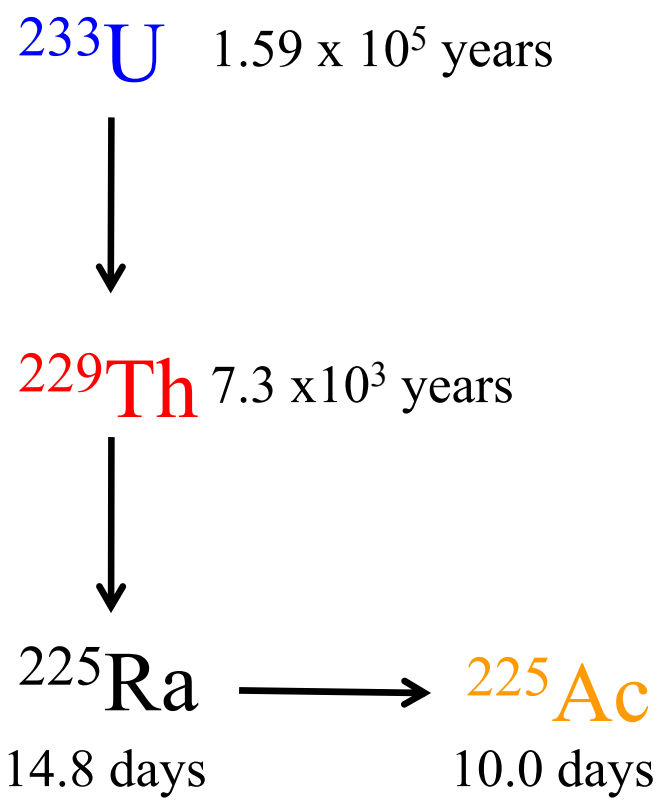
Primary Separation



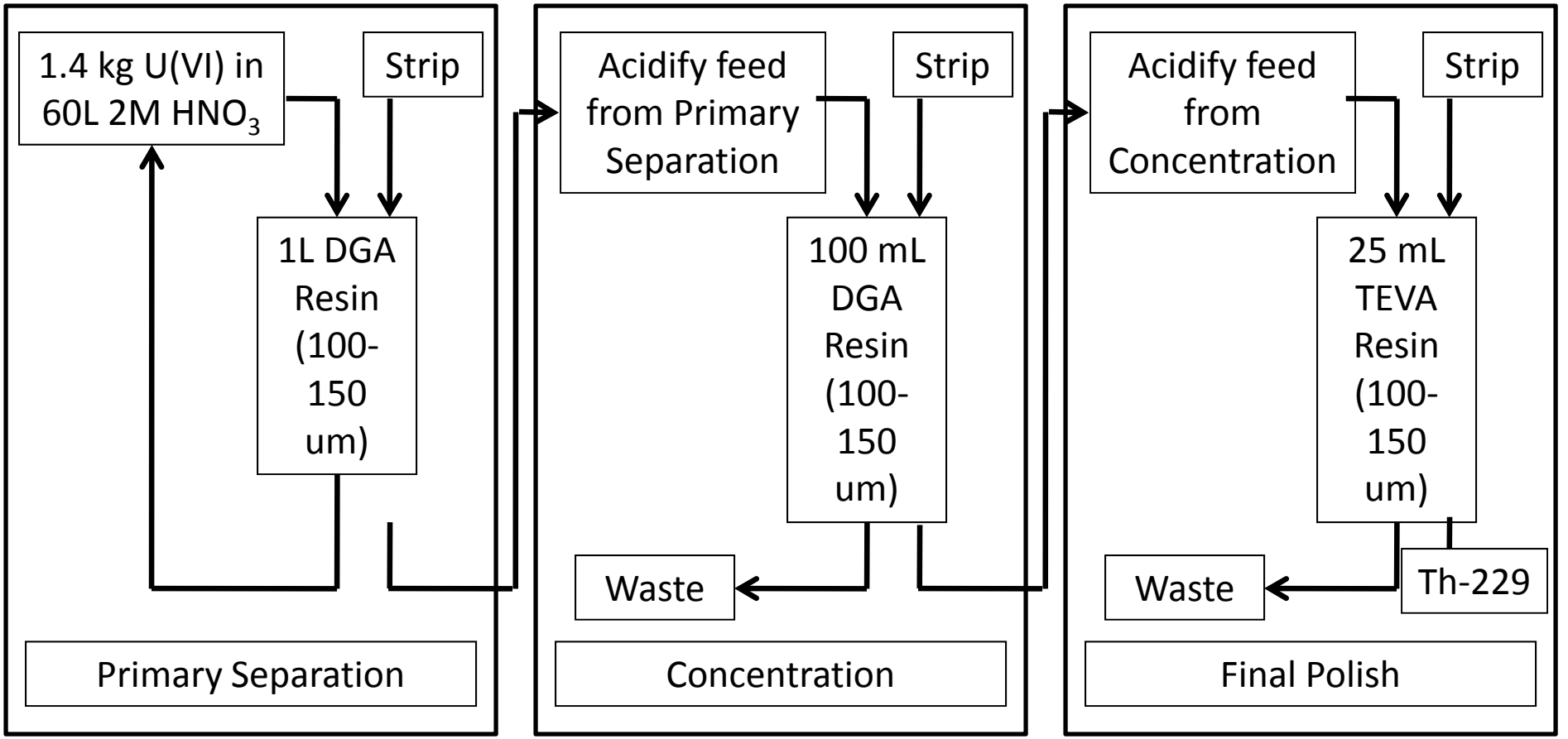
Separation of Np, Ac, Pu from 0.1 M U



Decay of ^{233}U



^{229}Th Separation from 1.4 kg ^{233}U



Separation of ^{176}Hf from kg amounts of Lu

^{176}Lu (2.59%)

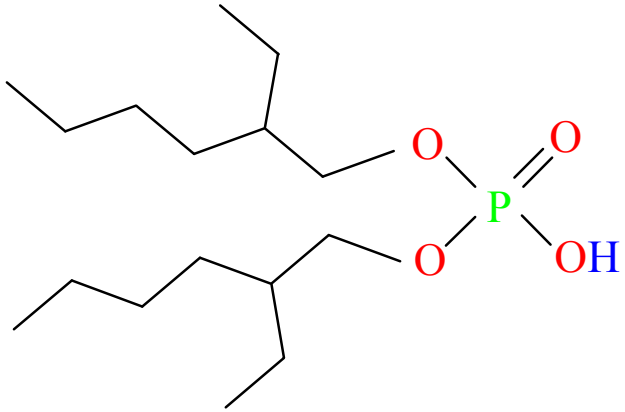
↓ $2.2 – 4.6 \times 10^{10}$ y

^{176}Hf Stable

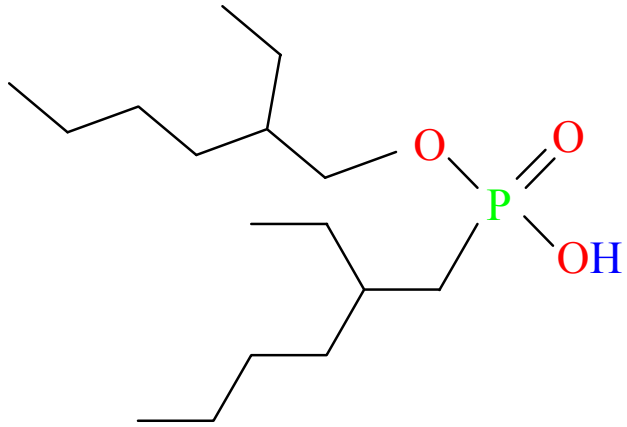
^{176}Lu $t_{1/2}$ = 3.8×10^{10} y (CRC - 2003)

3.59×10^{10} y (Browne/Firestone - 1986)

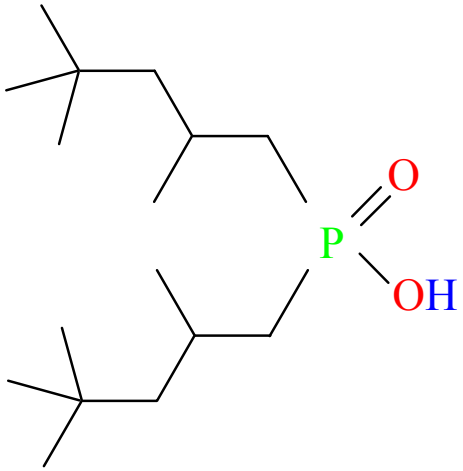
The LN Series of Resins



HDEHP (LN)

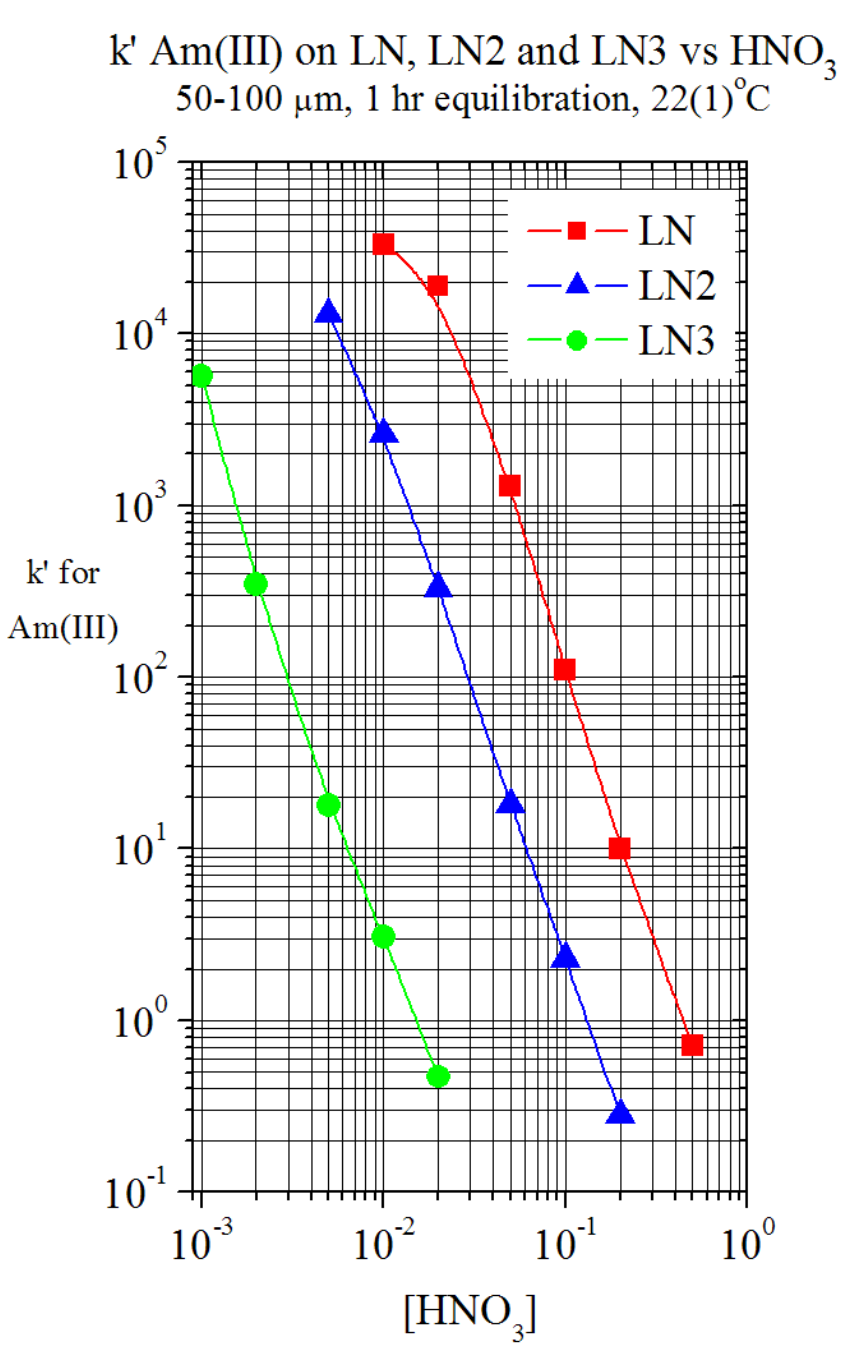


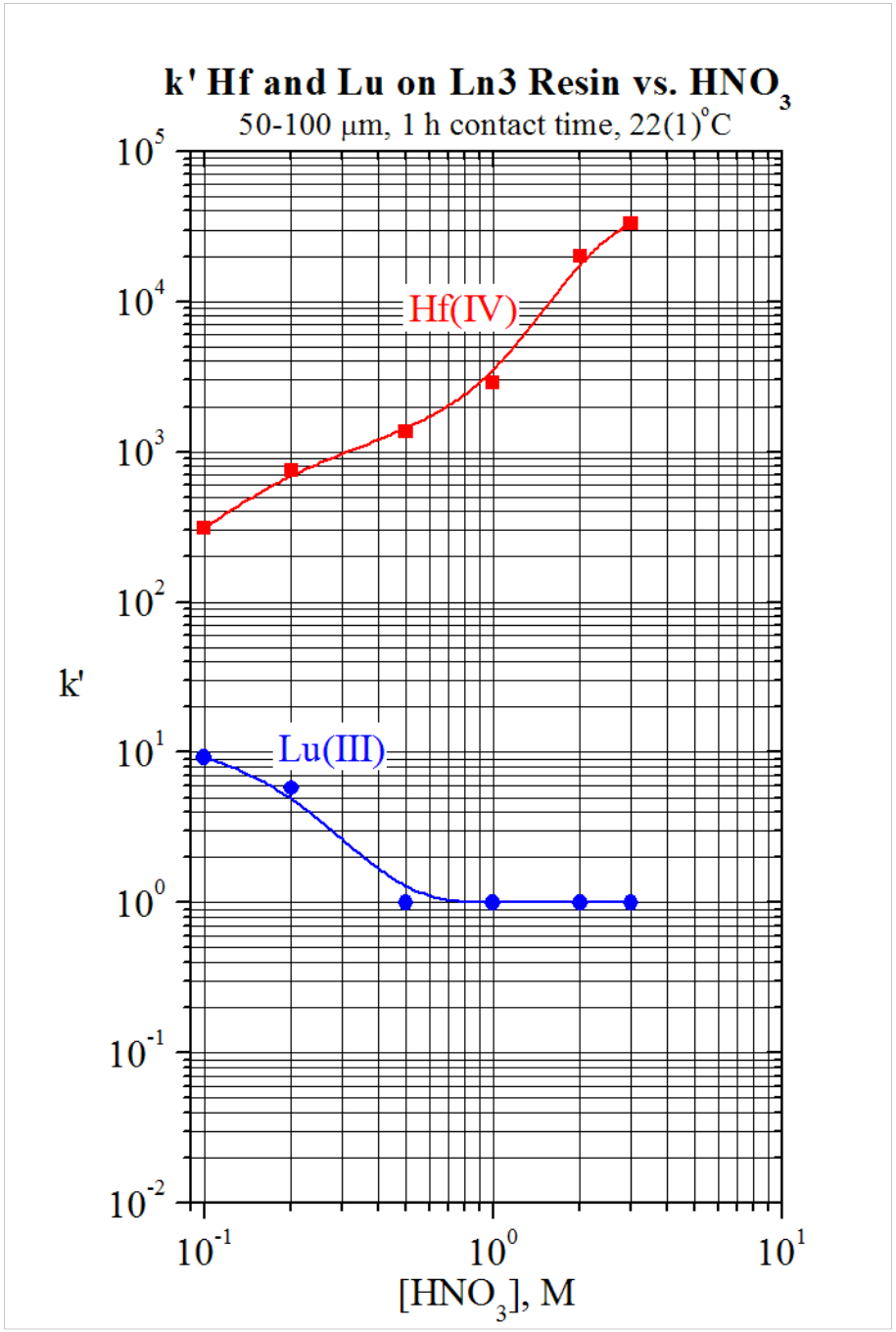
HEH[EHP] (LN2)



H[DTMPeP] (LN3)

$$M^{3+} + 3\overline{(HY)}_2 \leftrightarrow \overline{M(HY_2)_3} + 3H^+$$

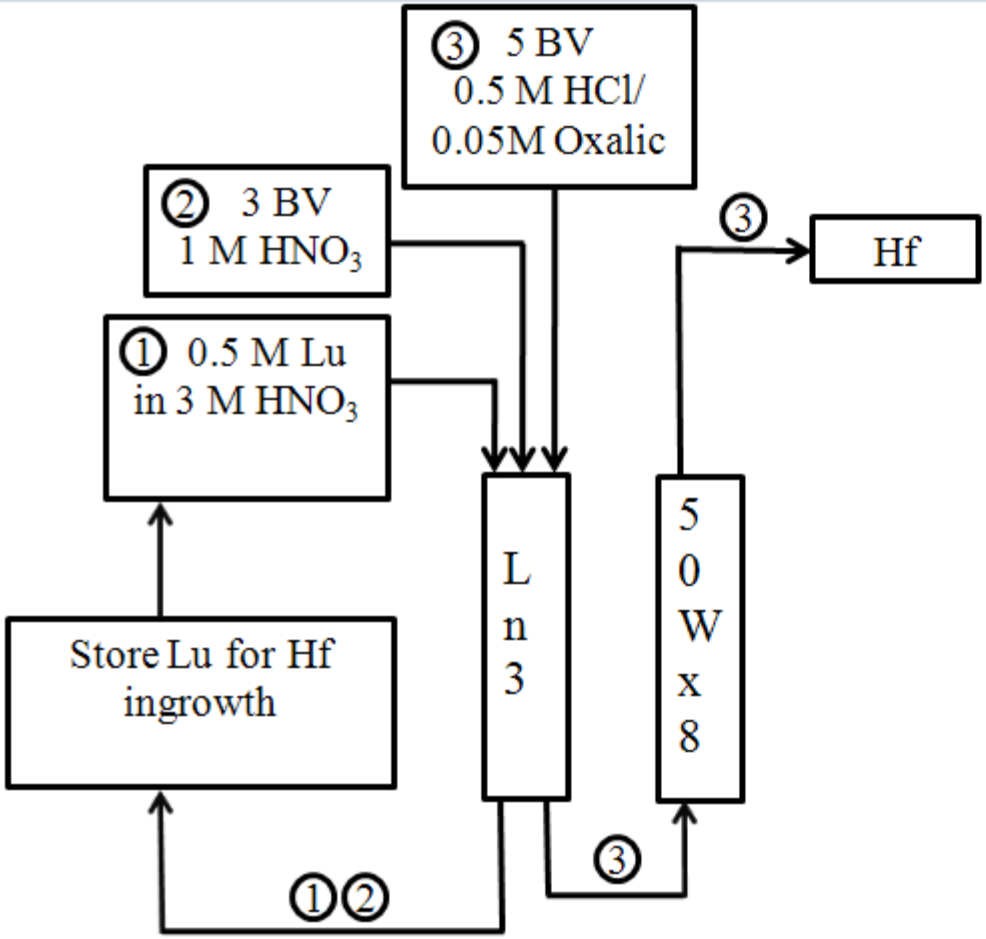

k' Am(III) on LN, LN2 and LN3 vs HNO3
50-100 µm, 1 hr equilibration, 22(1)°C
LN
LN2
LN3
k' for Am(III)
10^5
10^4
10^3
10^2
10^1
10^0
10^-1
10^-3
10^-2
10^-1
10^0
[HNO3]



αHf/Lu on Ln-3 Resin

HNO3	k'-Hf	k'-Lu	αHf/Lu
0.1	312	9.3	34
0.2	753	5.8	130
0.5	1365	<1	>1300
1	2891	<1	>2900
2	20164	<1	>20,000
3	33245	<1	>33,000

Separation of ^{176}Hf from kg amounts of Lu

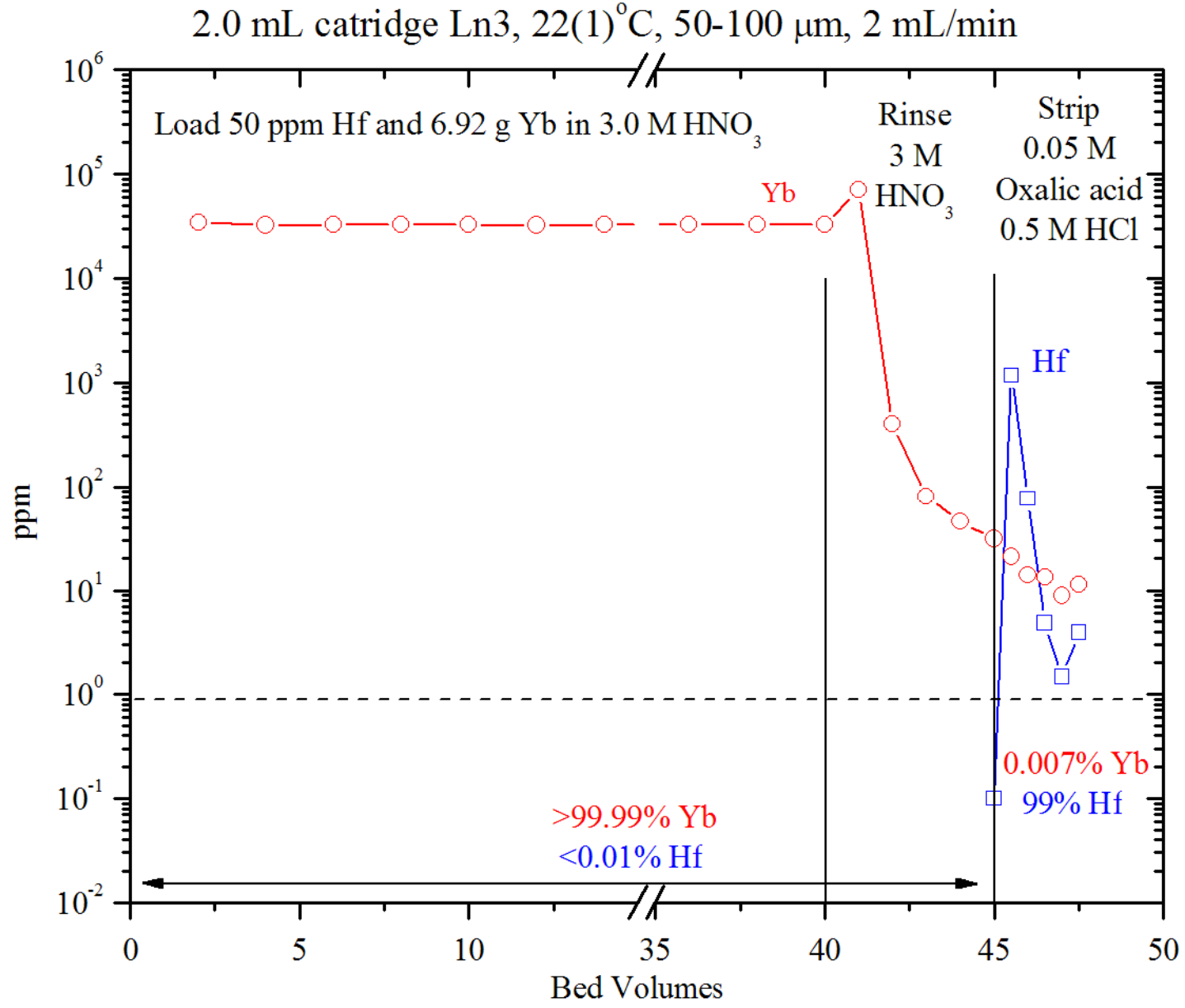


Ln 3 = ~25 mL/L 0.5M Lu in 1M HNO_3

50wx8 = 1 mL/10 mL Ln3

Precondition Ln3 with 1 Bed Volume of 1M HNO3

Precondition 50Wx8 with 2 Bed Volumes of 0.5M HCl/0.05M Oxalic acid


2.0 mL catridge Ln3, 22(1)°C, 50-100 µm, 2 mL/min
Load 50 ppm Hf and 6.92 g Yb in 3.0 M HNO3
Yb
Rinse
3 M
HNO3
Strip
0.05 M
Oxalic acid
0.5 M HCl
Hf
0.007% Yb
99% Hf
>99.99% Yb
<0.01% Hf
ppm
Bed Volumes
10^6
10^5
10^4
10^3
10^2
10^1
10^0
10^-1
10^-2
0
5
10
35
40
45
50