TANDEM SEPARATIONS OF RADIONUCLIDES USING AN AUTOMATED SYSTEM

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Outline

-ARSIIe Automated System

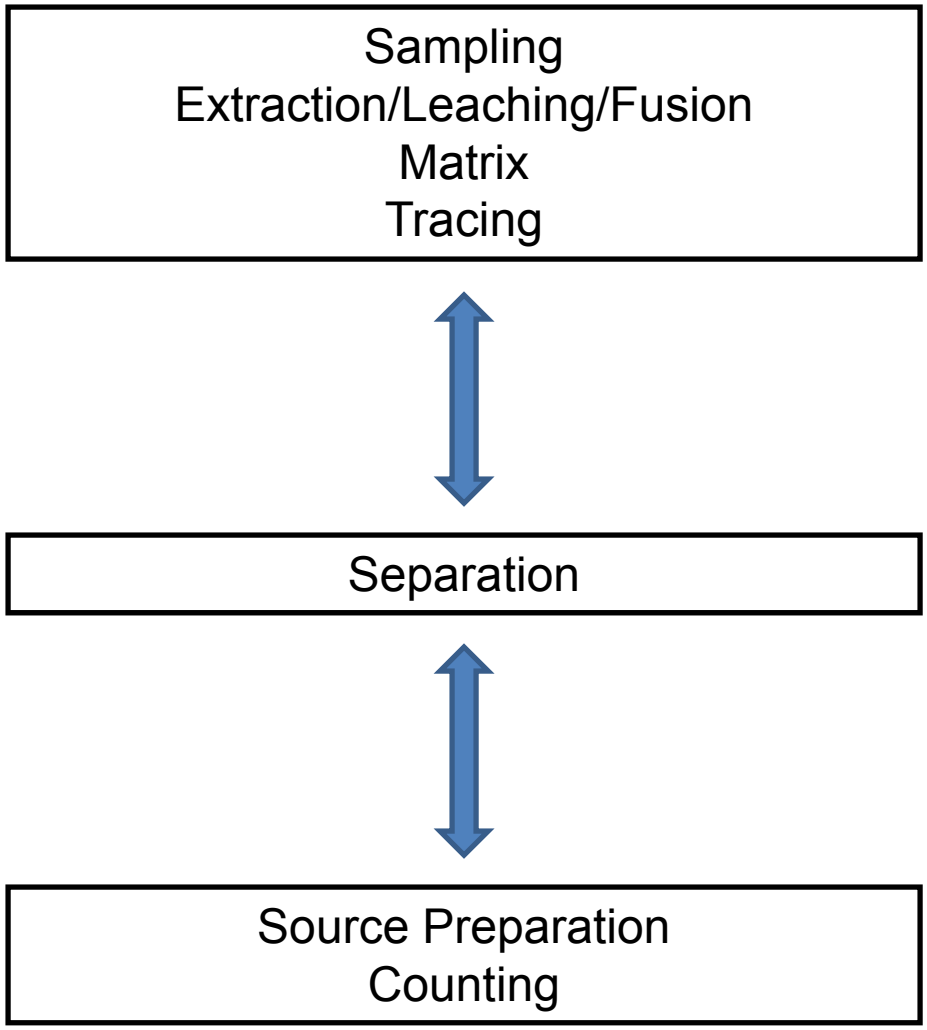
-Separation Challenges

-Examples of 2, 3, and 4 column systems

ARSIIe


ARS IIe
NorthStar
ENGINEERED TECHNOLOGIES, LLC
STOP
START/CONTINUE
RADIOACTIVE MATERIAL

Separation Challenges/Goals



Separation Challenges/Goals

- Obtain desired analytes:
  - in the necessary purity
  - in a matrix which is compatible with the available measurement technique(s).

- Integrate system with sample extraction/fusion method

- Use as few different reagents as possible

- Use reagents that have long shelf-lives

- Use reagents that compatible with each other

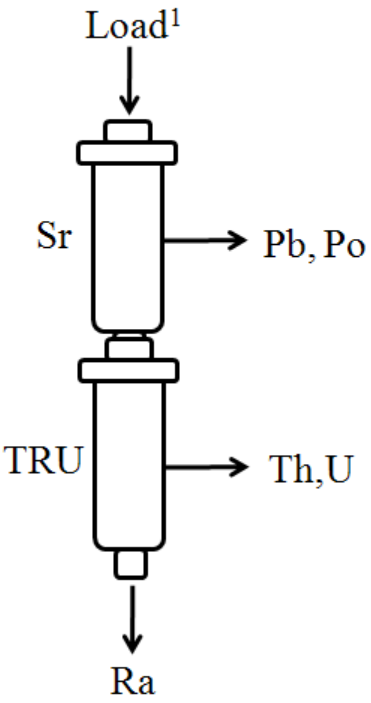
- Minimize elution volumes

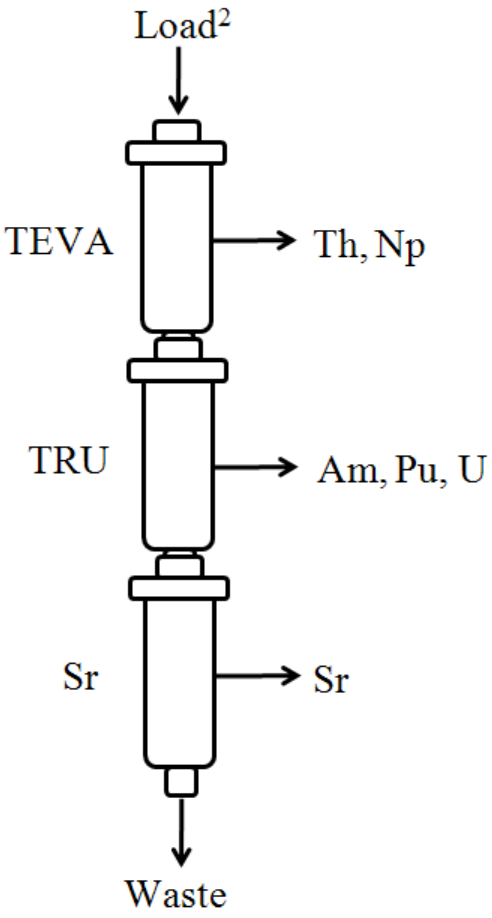
Available Tools

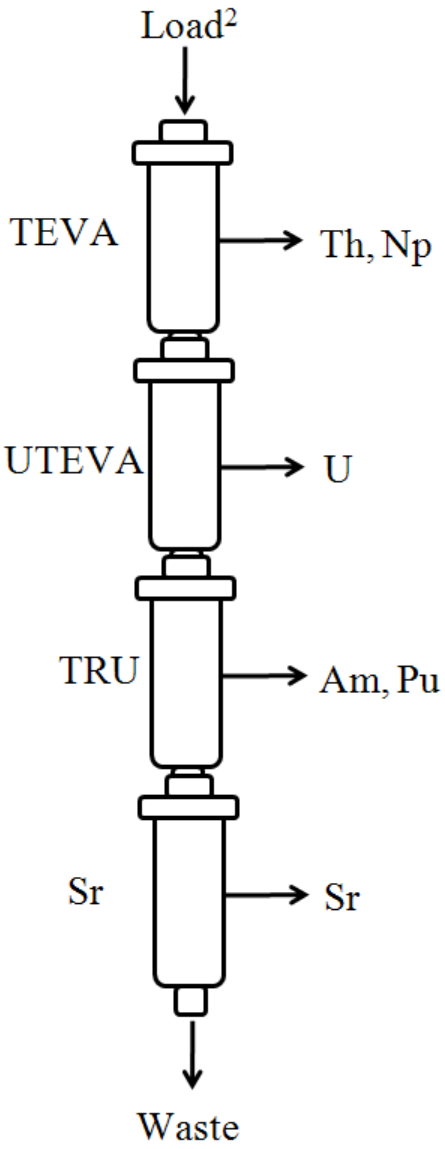
- TEVA
  - -Quaternary Amine: Th, Tc, Pu(IV), Np(IV), U(VI)
- UTEVA
  - -Phosphonate: Th, U(VI), Pu(IV), Np(IV), Po
- TRU
  - -Carbmoylphosphine oxide: Th, U(VI), Np(IV), Pu, Am
- DGA
  - -Diglycolamide: Th, U(VI), Np(IV), Pu, Am, ~Sr, ~Pb
- Sr Resin
  - -Crown Ether: Sr, Po, Pb, Pu(IV)

HNO_3 HCl Both

Examples







[1]Load = 4M HCl + 0.1 M Ascorbic acid
[2]Load = 3M HNO_3 + 0.1M Ascorbic acid + 0.1 M Ferrous Sulfamate

1st Example: Pb, Po, U, Th, Ra

- NATO Project, Uranium Mining/Processing Sites in former Soviet Republics

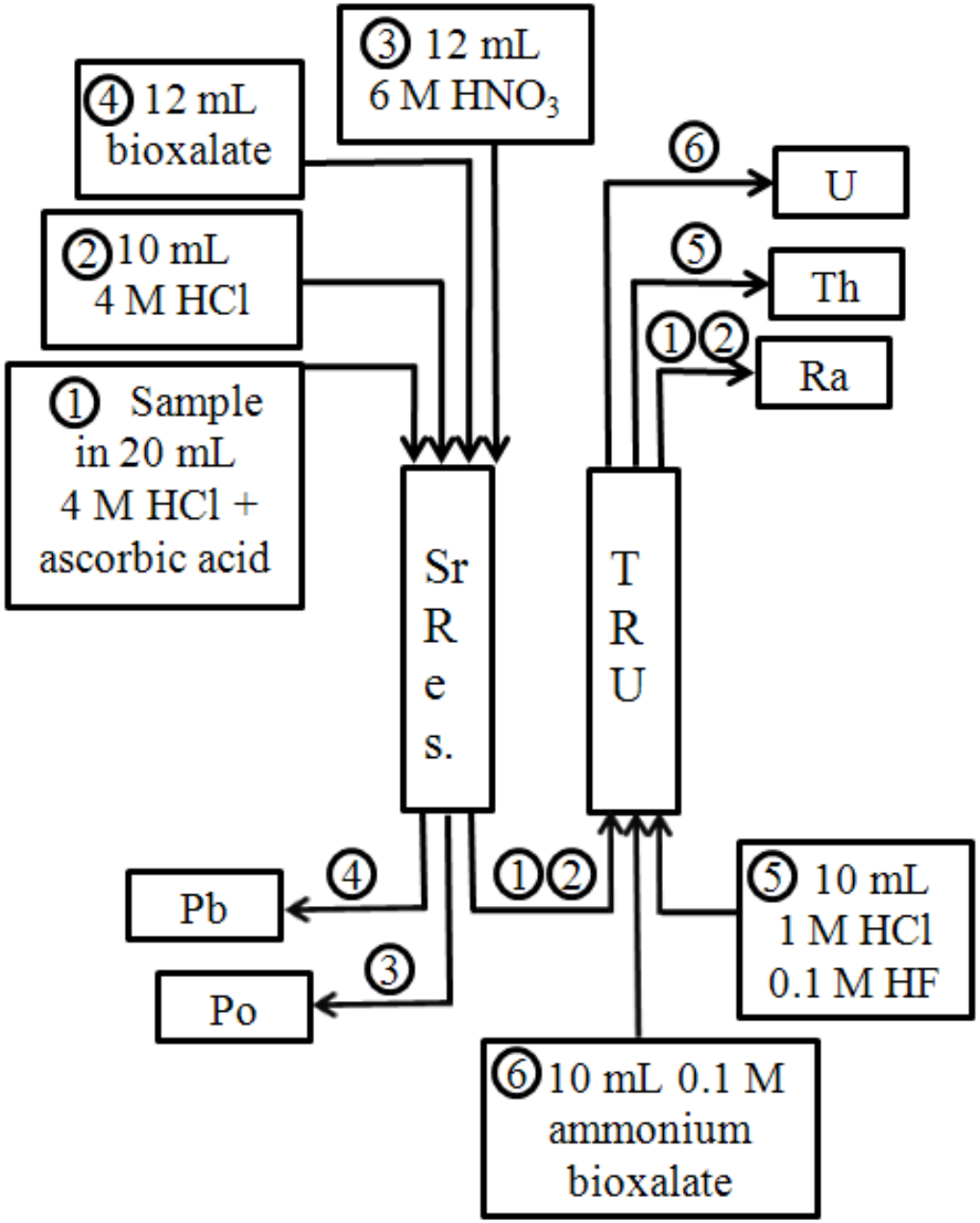
- ^{238}U, ^{234}U, ^{230}Th, ^{226}Ra, ^{210}Pb and ^{210}Po from ^{238}U decay chain

-^{232}Th, ^{228}Th, ^{224}Ra from ^{232}Th decay chain

-Soil samples leached with HCl

-Ascorbic acid added to limit interference from Fe(III)

Pb, Po, U, Th, Ra



Direct precipitation or deposition of column eluate:

$BaSO_4$ precip → Ra

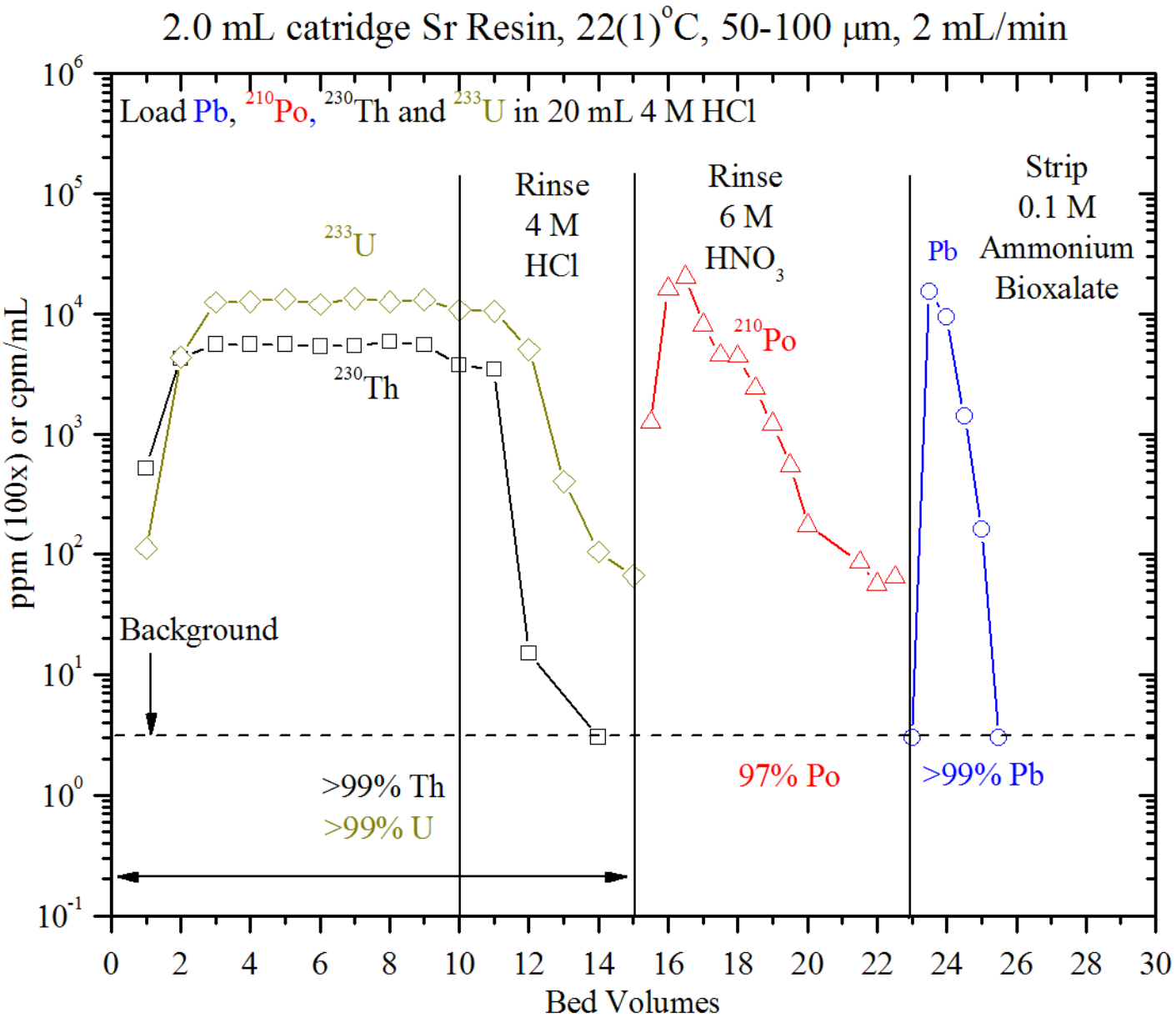
$PbSO_4$ precip → Pb

Autodeposition → Po

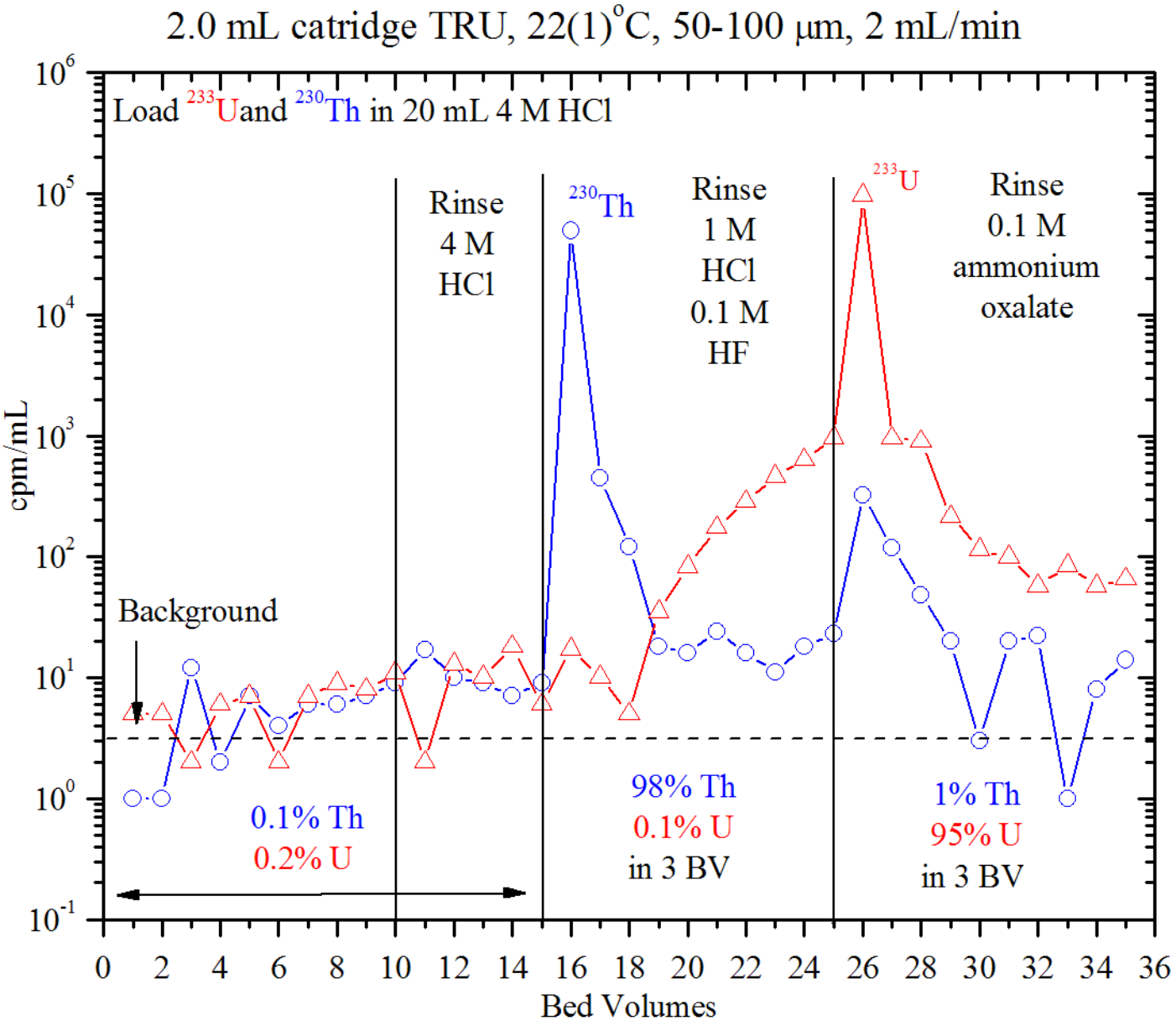
CeF_3 precip → U/Th

Followed by alpha spectroscopy

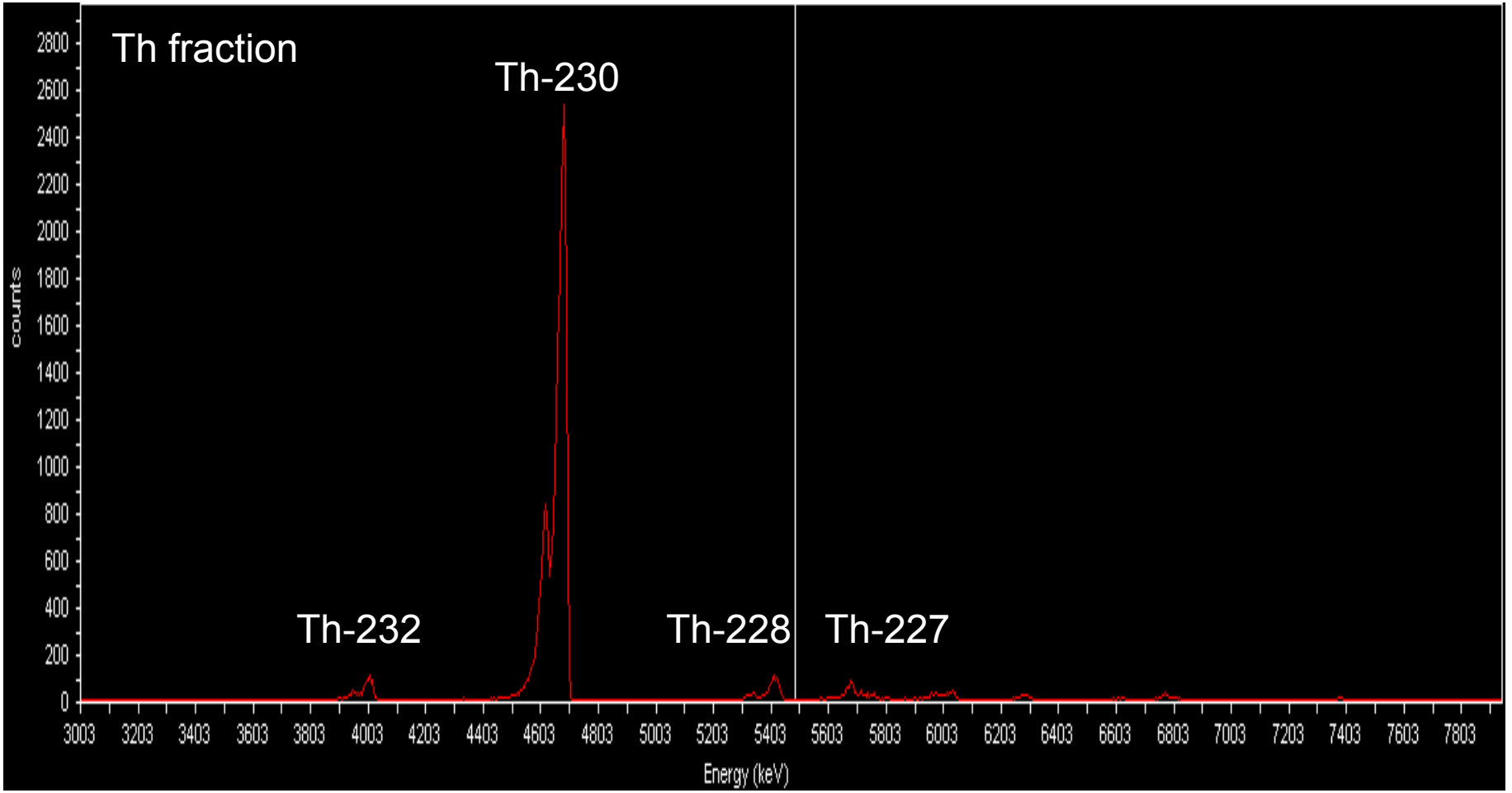
Pb, Po, U, Th, Ra


2.0 mL catridge Sr Resin, 22(1)°C, 50-100 µm, 2 mL/min
Load Pb, 210Po, 230Th and 233U in 20 mL 4 M HCl
Rinse 4 M HCl
Rinse 6 M HNO3
Strip 0.1 M Ammonium Bioxalate
233U
230Th
210Po
Pb
Background
>99% Th
>99% U
97% Po
>99% Pb
ppm (100x) or cpm/mL
Bed Volumes

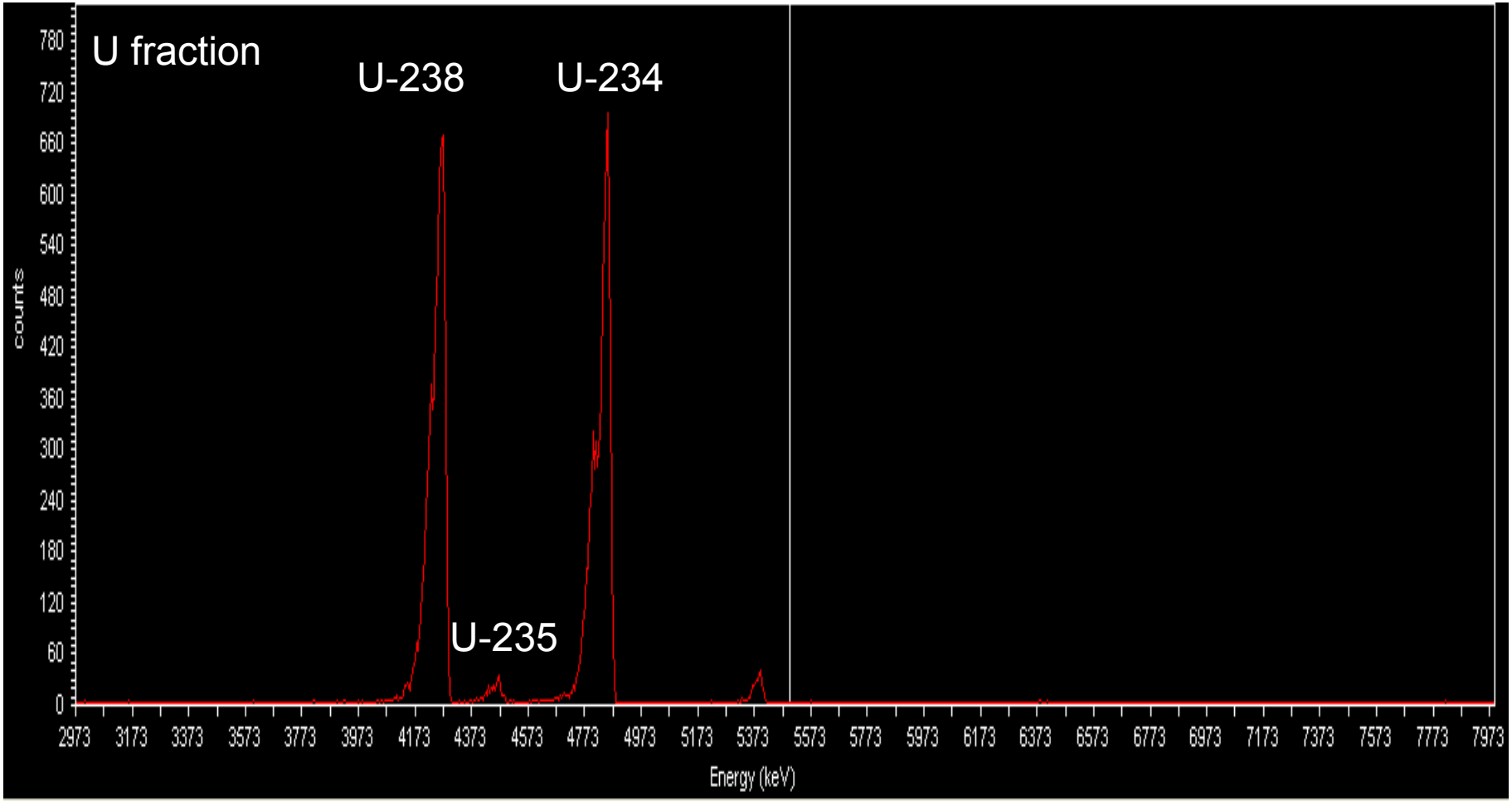
Pb, Po, U, Th, Ra


2.0 mL catridge TRU, 22(1)°C, 50-100 µm, 2 mL/min
Load 233Uand 230Th in 20 mL 4 M HCl
Rinse 4 M HCl
230Th
Rinse 1 M HCl 0.1 M HF
233U
Rinse 0.1 M ammonium oxalate
Background
0.1% Th
0.2% U
98% Th
0.1% U
in 3 BV
1% Th
95% U
in 3 BV
cpm/mL
Bed Volumes

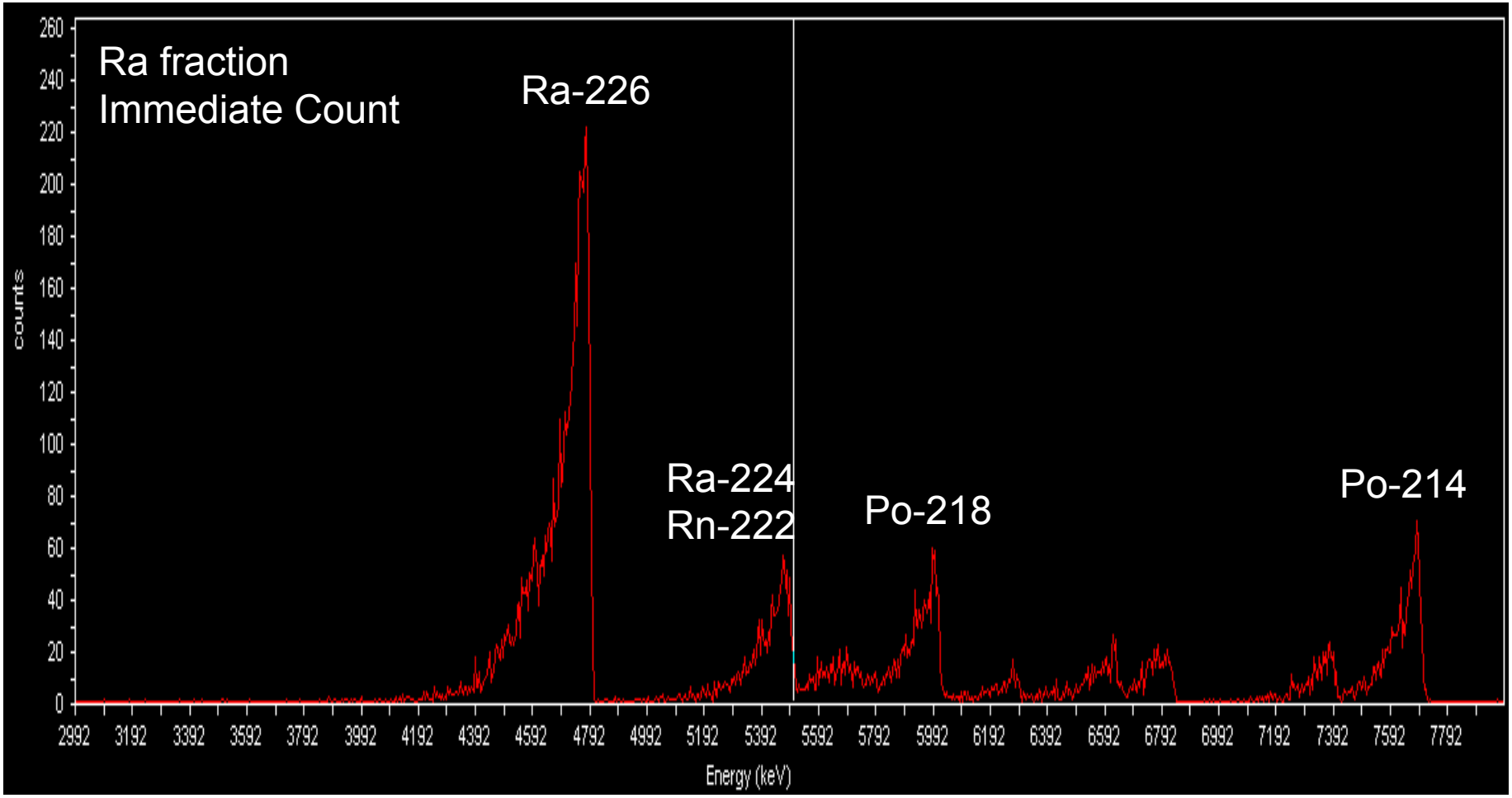
Pb, Po, U, Th, Ra



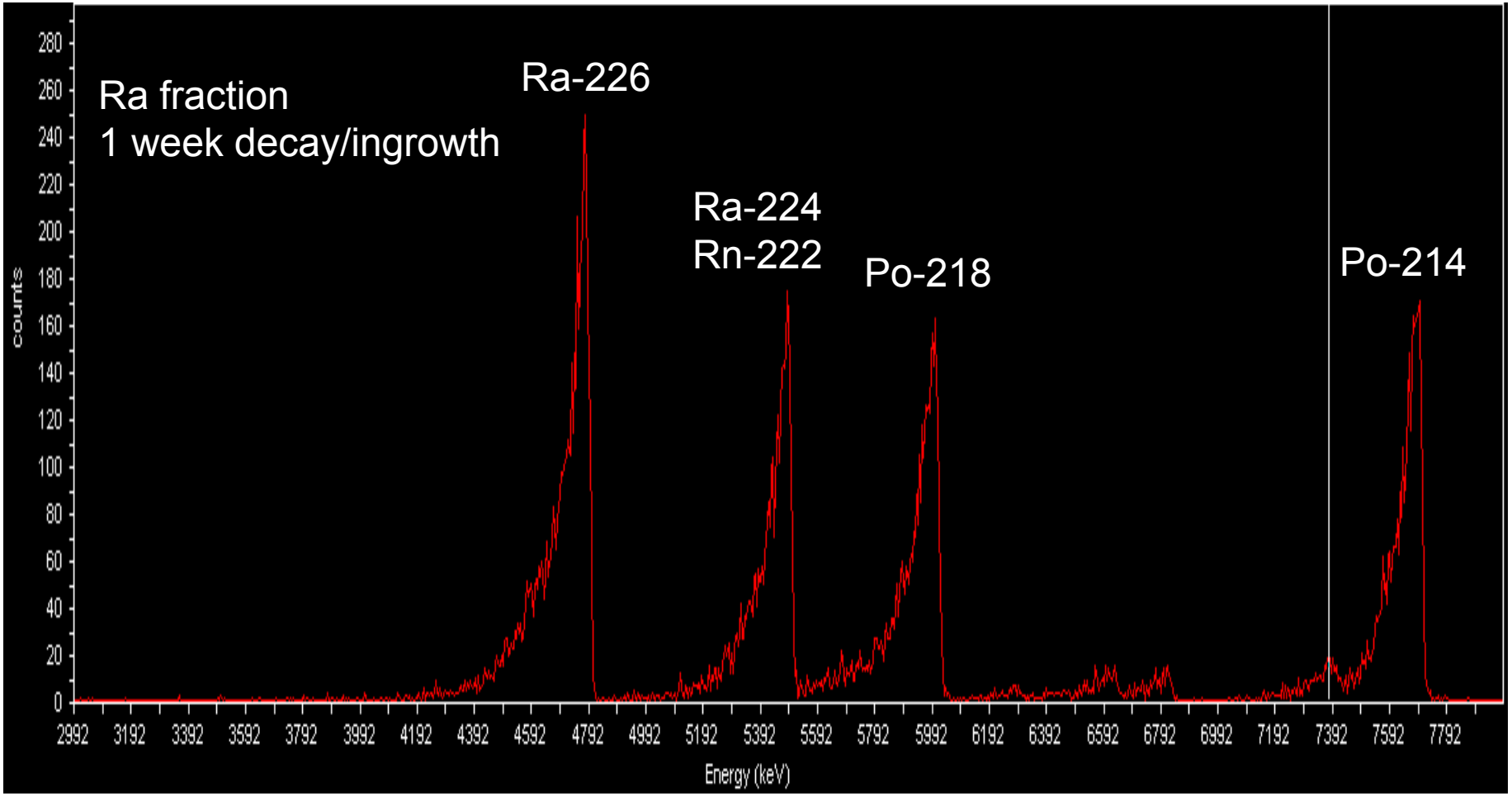
Pb, Po, U, Th, Ra


U fraction
U-238
U-234
U-235
counts
780
720
660
600
540
480
420
360
300
240
180
120
60
0
2973
3173
3373
3573
3773
3973
4173
4373
4573
4773
4973
5173
5373
5573
5773
5973
6173
6373
6573
6773
6973
7173
7373
7573
7773
7973
Energy (keV)

Pb, Po, U, Th, Ra


Ra fraction
Immediate Count
Ra-226
Ra-224
Rn-222
Po-218
Po-214
counts
Energy (keV)

Pb, Po, U, Th, Ra

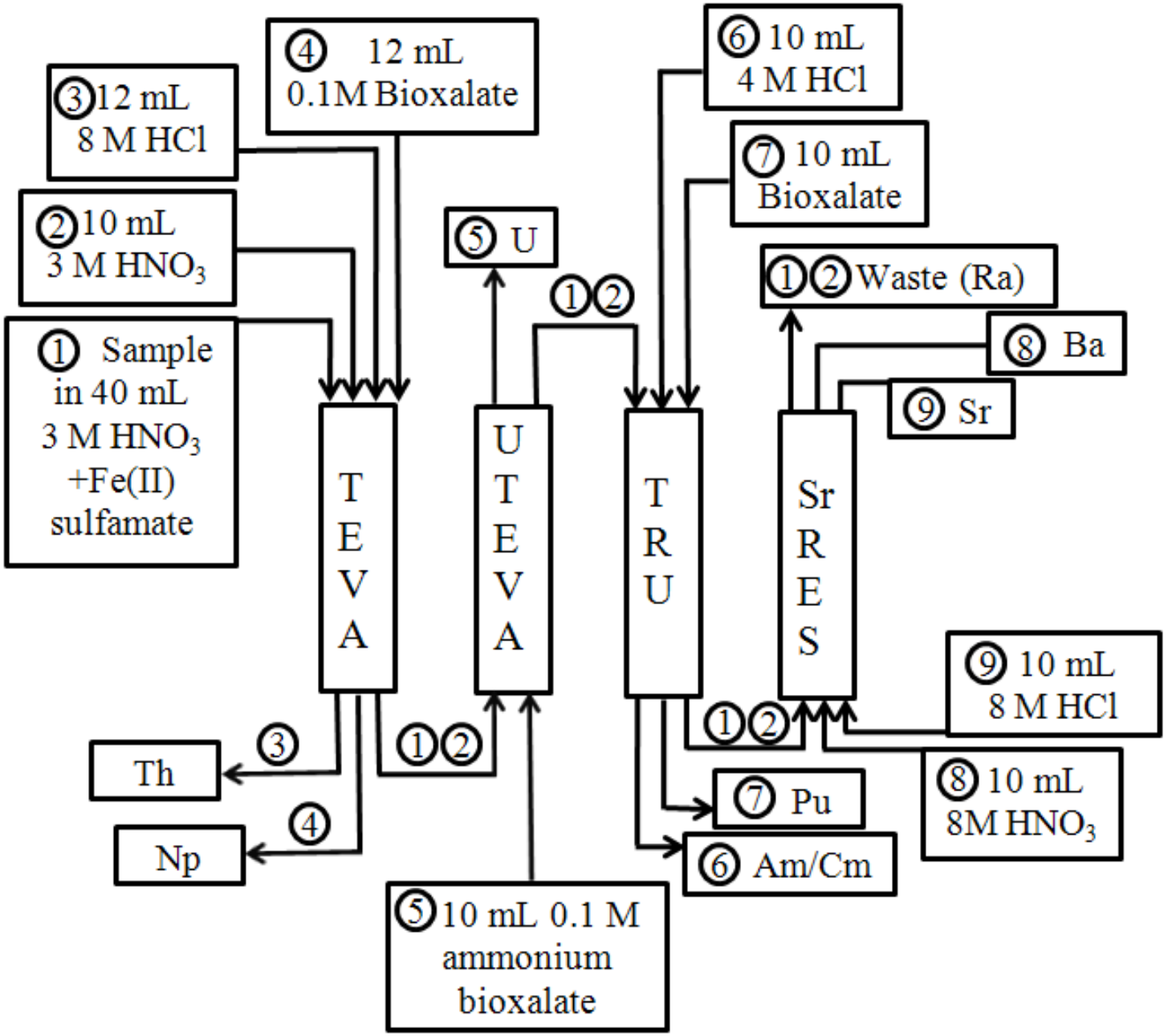

Ra fraction
1 week decay/ingrowth
Ra-226
Ra-224
Rn-222
Po-218
Po-214
counts
Energy (keV)

2nd Example: Th, Np, U, Am/Cm, Pu, Sr

-USAF Project (Marty Johnson Friday at 10:10)
-Soil samples, lithium borate fusion
-Dissolve Fusion with HNO_3
-Filter to remove silicates
-Adjust oxidation states with Ferrous Sulfamate/Ascorbic acid:
    -Np(IV)
    -Pu(III)

-Th, U, Am/Cm, Pu by alpha spectroscopy (CeF_3 precip.)
-Np by alpha, gas flow proportional and/or LSC (CeF_3 precip.)
-Sr by LSC or gas flow proportional

Th, Np, U, Am/Cm, Pu, Sr



Th, Np, U, Am/Cm, Pu, Sr

Soil	Fraction	Nuclide	Tracer Recovery (%)[d]	% Yield[e]
S2	TEVA-Th Strip	Th-228		
	12 mL	Th-229	219.7[f]	
	8M HCl	Th-230		33.7[f]
		Th-232		
S2	TEVA-Np	Np-237	N/A	
	12 mL 0.1 M Biox			
S2	UTEVA- U Strip	U-232	58.4	
	10 mL	U-233		
	0.1 M Bioxalate	U-234		98.6
		U-235		103.6
		U-238		109.8
S2	TRU-Am/Cm Strip	Am-241		123.5
	10 mL	Am-243	28.3	
	4M HCl	Cm-242		
		Cm-243		301.3
S2	TRU-Pu Strip	Pu-236		
	10 mL	Pu-238		127.8
	0.1 M Bioxalate	Pu-240		115.0
		Pu-242	62.7	
S2	Sr Resin Strip	Sr-90	86.8	
	10 mL 8 M HCl			

[a]Columns: 2mL TEVA, UTEVA, TRU, Sr Resin (50-100 µm)

[b]Load: 45 mL 3M HNO_3 + Fe(II) sulfamate/ascorbic acid

[c]Rinse: 10 mL 3M HNO_3 + 0.1 M $NaNO_2$

[d]Absolute recovery of tracer through column chemistry and dissolution of melt

[e]Yeild corrected for tracer recovery

[f]Poor alpha spectrum resolution, overlap of Th-230 and Th-229 peaks

Th, Np, U, Am/Cm, Pu, Sr

Soil	Fraction	Nuclide	Tracer Recovery (%)[d]	% Yield[e]
S3	TEVA-Th Strip	Th-228		
	12 mL	Th-229	86.2	
	8M HCl	Th-230		113.0
		Th-232		
S3	TEVA-Np	Np-237	N/A	
	12 mL 0.1 M Biox			
S3	UTEVA- U Strip	U-232	66.6	
	10 mL	U-233		
	0.1 M Bioxalate	U-234		102.7
		U-235		113.1
		U-238		102.2
S3	RE-2-Am/Cm Strip	Am-241		114.9
	10 mL	Am-243	55.9	
	4M HCl	Cm-242		
		Cm-243		94.0
S3	RE-2-Pu Strip	Pu-236		
	10 mL	Pu-238		87.2
	0.1 M Bioxalate	Pu-240		89.7
		Pu-242	97.6	
S3	Sr Resin Strip	Sr-90	107.0	
	10 mL 8 M HCl			

[a]Columns: 2mL TEVA, UTEVA, RE-2, Sr Resin (50-100 µm)

[b]Load: 45 mL 3M HNO_3 + Fe(II) sulfamate/ascorbic acid

[c]Rinse: 10 mL 3M HNO_3 + 0.1 M $NaNO_2$

[d]Absolute recovery of tracer through column chemistry and dissolution of melt

[e]Yeild corrected for tracer recovery

Conclusion

- Demonstrated two separation methods on the ARSIIe
    - -2 column (Po, Pb, U, Th, Ra)
    - -4 column (Th, Np, U, Am/Cm, Pu, Sr)

- Develop additional separations on ARSIIe
    - -Single column methods (4 simultaneous samples)
    - -Additional multiple column methods

Acknowledgment

Glenn Isensee, Mike Krummey, Lionel Johnson and Jim Harvey of Northstar Engineered Technologies for providing the ARSIIe instrument and training.

Marty Johnson for helpful discussions while developing the separation method.