Determination of Am, Pu, U and Sr from large amount of waste samples originated from NPP

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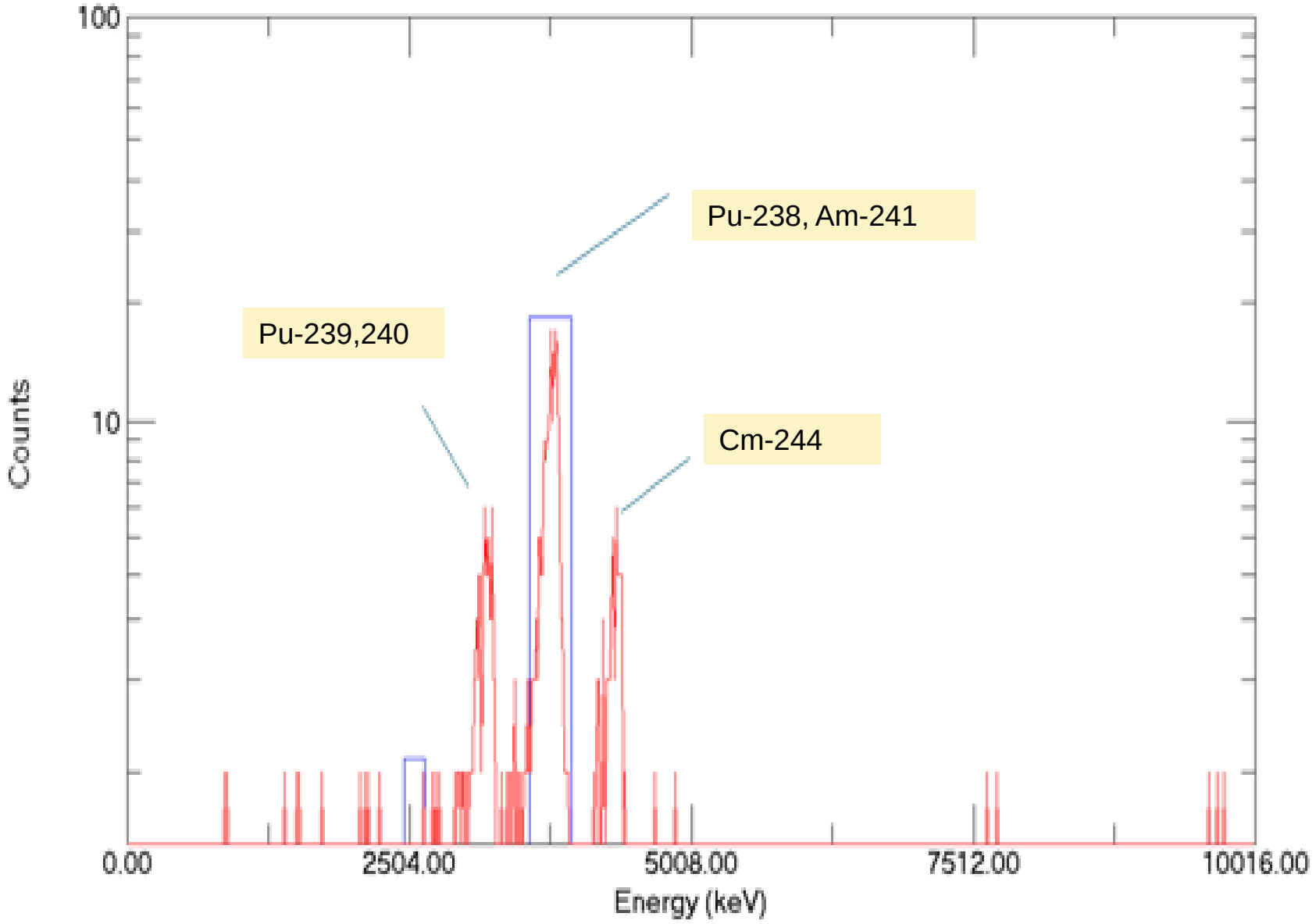
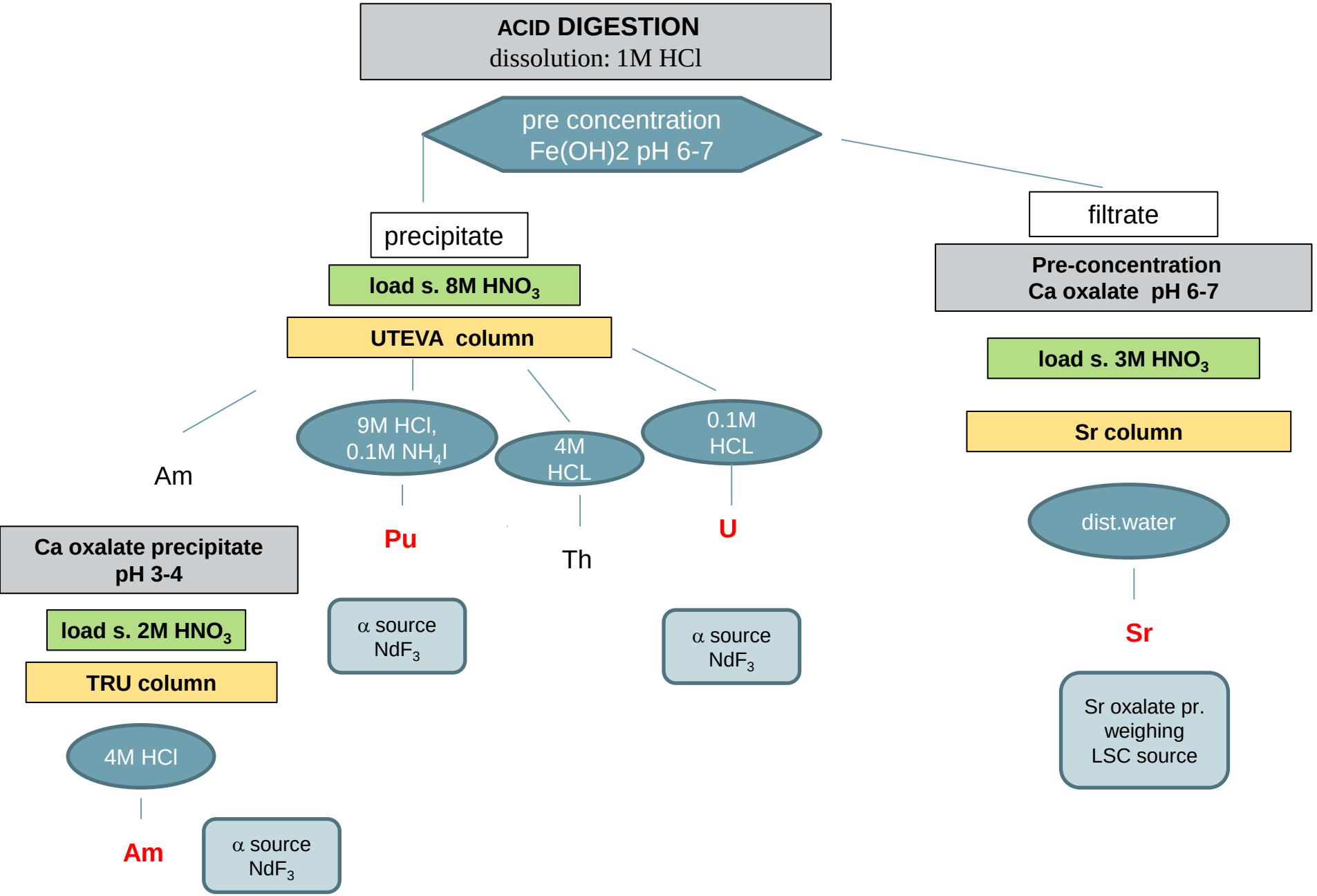
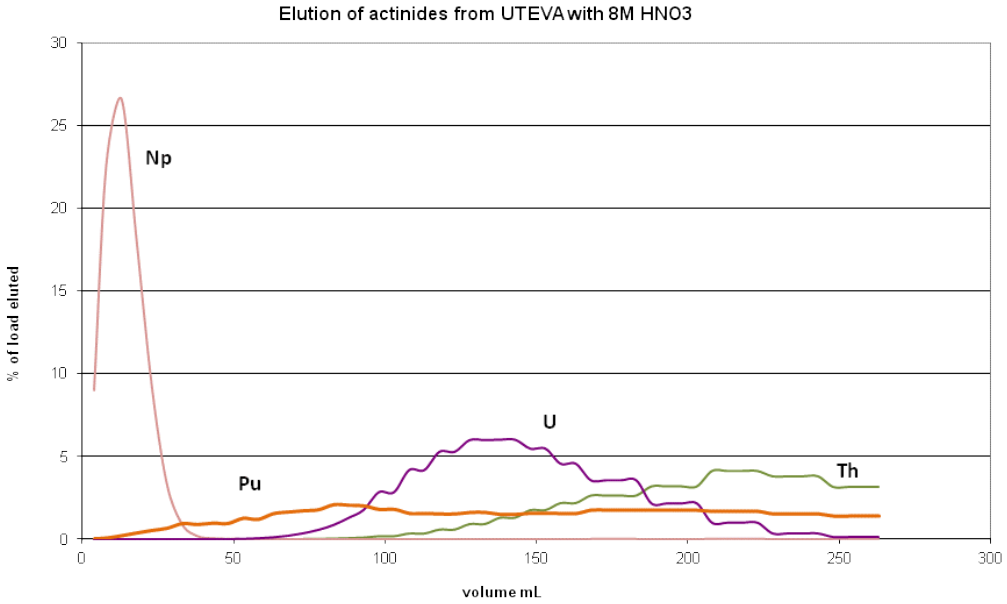

100
10
Counts
Pu-238, Am-241
Pu-239,240
Cm-244
0.00
2504.00
5008.00
7512.00
10016.00
Energy (keV)


ACID DIGESTION
dissolution: 1M HCl
pre concentration
Fe(OH)2 pH 6-7
precipitate
filtrate
load s. 8M HNO3
UTEVA column
Pre-concentration
Ca oxalate pH 6-7
load s. 3M HNO3
Sr column
9M HCl,
0.1M NH4I
4M
HCL
0.1M
HCL
Am
Pu
Th
U
dist.water
Ca oxalate precipitate
pH 3-4
load s. 2M HNO3
TRU column
α source
NdF3
α source
NdF3
Sr
4M HCl
Sr oxalate pr.
weighing
LSC source
Am
α source
NdF3

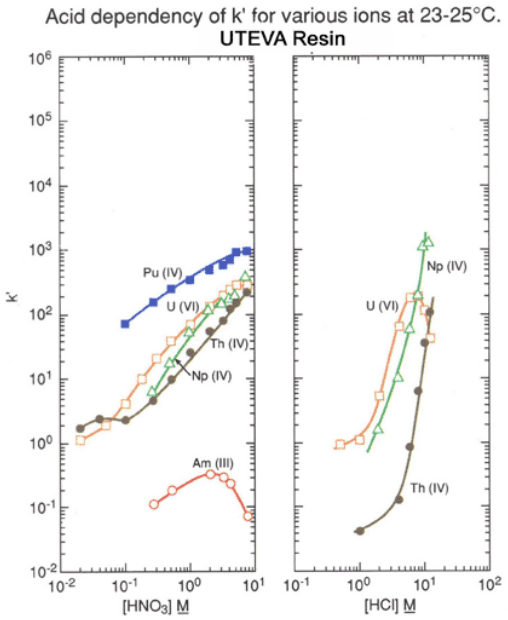
Separation of actinides on UTEVA column from 8M HNO_3

U(VI) and Pu(IV) are well retained on the column
Am goes through the column
Oxidation state of Pu?



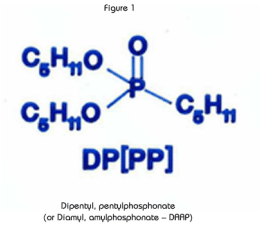
Am, Np are in the effluent,
Pu is in different oxidation states

Figures 2 and 3

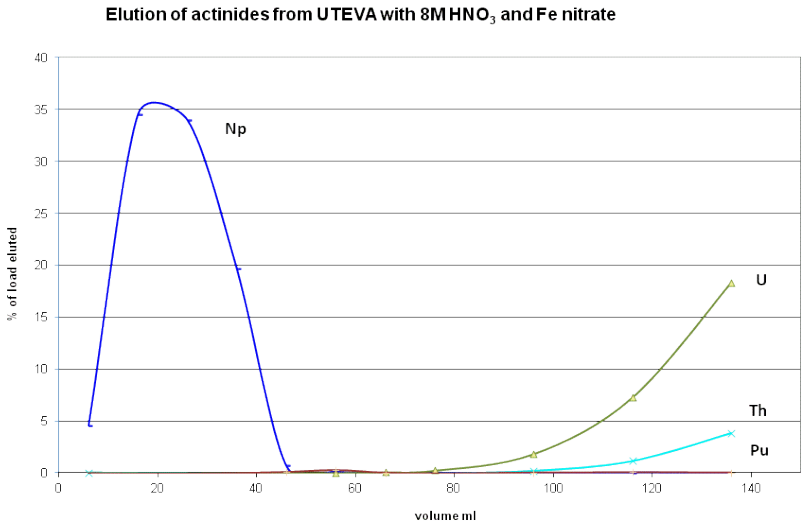


Horwitz, et al. (HP392)

Figure 1

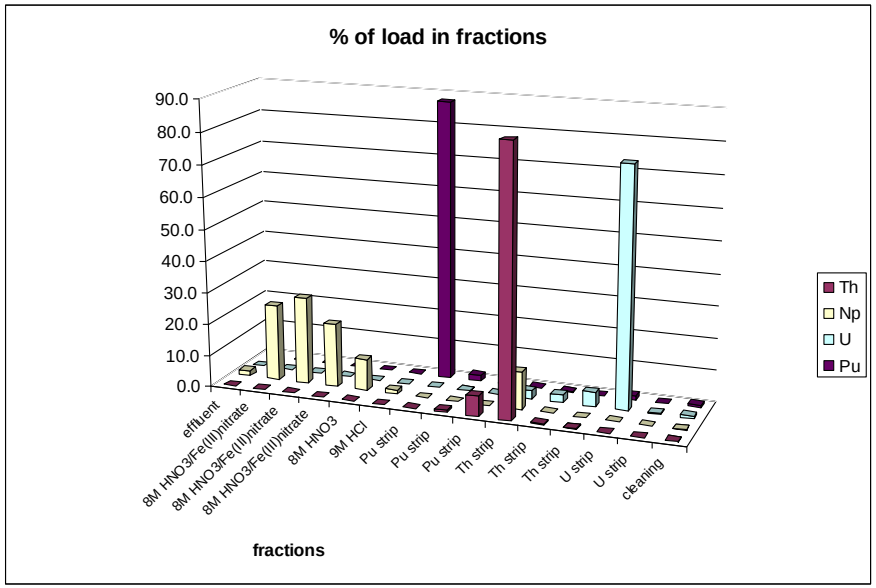


Separation of actinides on UTEVA column from 8M HNO_3/Fe(III)



Load: 8M HNO_3/Fe(III)
UTEVA (34mm)
Pu strip: 9M HCl/0.1M NH_4I
Th strip: 4M HCl
U strip: 0.1M HCl

U, Th and Pu are well retained
volume of loading solution could be more than 50 ml
Am and Np are in the load solution



Separation Am, Cm on TRU column

Load solution after UTEVA column
(Triskem method)

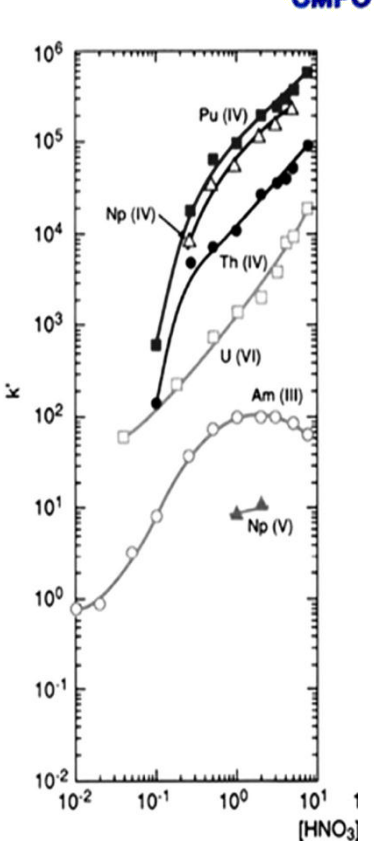
evaporation

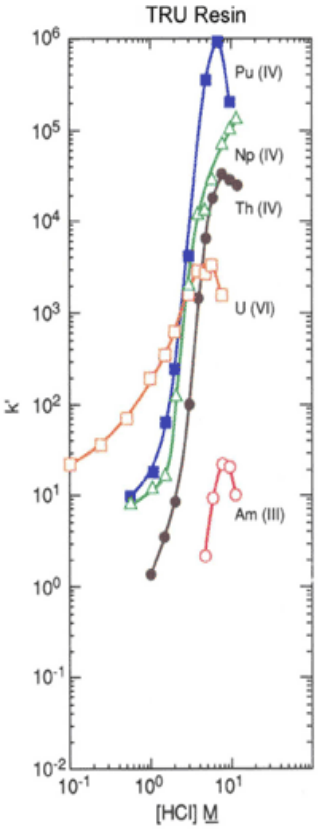
Ca oxalate precipitation pH 3-4

evaporation
oxalate destruction

Load: 2M HNO_3 (ascorbic acid)
TRU column (34mm)
Am strip: 4M HCl

TRU resin for Am





Separation of Sr on Sr resin

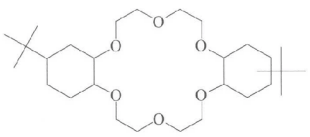
Filtrate after $Fe(OH)_2$ precipitation

Ca-oxalate precipitation pH 5-6
0.5 g $CaCl_2$
filtrate
oxalate destruction

Load: 3M HNO_3
Sr resin column (3g)
Sr strip: dist.water
determination chemical recovery by weighing Sr oxalate

Figure 1

4,4'(5')-di-t-butylcyclohexano 18-crown-6

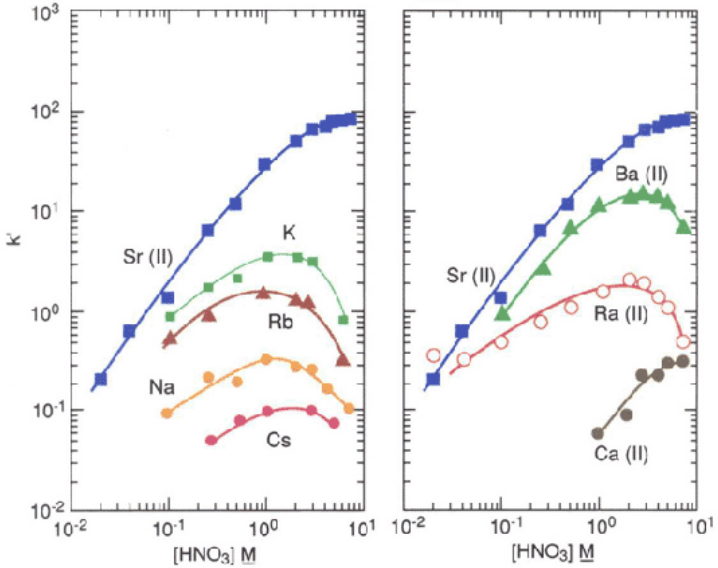


Diluent: 1-octanol

Figures 2 and 3

Acid dependency of k' for various ions at 23-25°C.

Sr Resin



Horwitz, et al., (HP292)

Source preparation:
α source: co-precipitation by NdF_3
β source: Sr-oxalate is disolved in 2 ml of 1M HNO_3, add 10 ml of Prosafe HC

Method validation

2013. Measurement of SRM

MAPEP -06-MaS15

Isotope	Measured A Bq/kg		σ Bq/kg	Reference value A Bq/kg		σ Bq/kg	z-score	prec. index %
U-238	35.3	+-	2.2	37.0	+-	3.8	0.4	11.9
U-234	36.0		2.2	38.9	+-	4.5	0.6	13.2
Pu-239,240	44.6	+-	2.7	45.9	+-	6.4	0.2	15.1
Pu-238	53.0		2.9	57.9	+-	4.2	1.0	9.1
Am-241	55.0		2.8	57.1	+-	5.5	0.3	10.8
Criteria							≤ 3	≤ 25%

Isotope	Sr-90 Bq/kg	σ	z-score	prec. index %
MAP1	298.95	14.22	0.33	14.5
MAP2	318.12	8.89	0.09	14.0
MAP3	287.66	9.56	0.60	14.1
MAP1 (2)	269.84	16.97	0.96	15.1
MAP2 (2)	293.42	7.00	0.47	13.9
MAP3 (2)	276.87	11.33	0.83	14.3
			≤ 3	≤ 25%

Where do we use the procedure?

- Measurement of nuclear wastes and processed wastes

- Determination of Sr and Pu isotopes in ground water samples
  (we analyse 70-80 l of water)
  procedure: Water goes through on-site cation exchanger

    - strip cations with HCl

    - coprecipitation of Pu with Fe(II)-hydroxide → UTEVA

    - coprecipitation of Sr with Ca oxalate → Sr resin

- Measurement of environmental samples as soils, aerosols etc.

Some results

Nuclear waste:

Sample code	H13-3			H13-4		
Isotope	A (Bq/l)		σ (Bq/l)	A (Bq/l)		σ (Bq/l)
U-234	2.55E+02	±	3.48E+01	2.46E-01	±	1.38E-02
U-235		≤	3.67E+01	3.51E-02	±	4.90E-03
U-238	7.55E+01	±	1.79E+01	1.06E-01	±	8.15E-03
Pu-239,240	9.45E+03	±	4.23E+02	2.71E-01	±	2.75E-02
Pu-238	9.23E+03	±	4.12E+02	2.98E-01	±	2.93E-02
Am-241	1.16E+04	±	5.25E+02	2.44E-01	±	1.60E-02
Cm-244	1.27E+03	±	1.32E+02	4.02E-02	±	5.88E-03
Cm-242		≤	8.62E+01		≤	3.97E-03
Sr-90	1.73E+07	±	1.22+06	1.03+02	±	5.26+00

Two samples from 2013
the concentrations are very different
in the 1st case we analyzed „20 μl" of the sample without pre-concentration

Activity concentration of Sr-90 in ground water:

Sample	Volume (L)	Chem. recovery	^{90}Sr Bq/l		σ Bq/l		DL Bq/l
T2	76	62 %	8.47E-03	±	2.64E-04	≤	1.31E-04
T4	76	33 %	4.26E-04	±	1.03E-04	≤	2.38E-04
T14	78	57 %	2.45E-04	±	5.96E-05	≤	1.37E-04
T18	53.5	45 %				≤	2.57E-04
T19	78.6	55 %	2.24E-03	±	9.24E-05	≤	1.39E-04
T39	70.8	41 %	4.10E-03	±	3.64E-04	≤	7.84E-04

Activity concentration of Pu isotopes in ground water:

Sample	Chem. recovery	Volume (L)	Pu-239/240 Bq/l		Pu-238 Bq/l	
T2	82%	119.2	≤	3.07E-06	≤	3.07E-06
T4	29%	160.6	≤	9.99E-06	≤	9.99E-06
T14	60%	163.4	≤	4.46E-06	≤	3.83E-06
T18	79%	114	≤	2.44E-05	≤	2.44E-05
T19	89%	163.4	≤	2.00E-06	≤	2.00E-06
T39	82%	117	≤	4.63E-06	≤	3.98E-06

Thank you for your attention