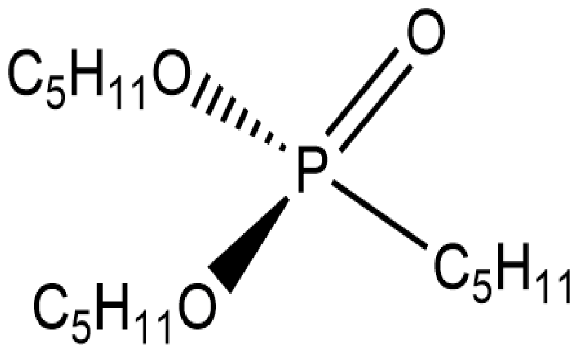
Determination of distribution coefficients of radionuclides set for UTEVA resin

A.P. Marinova, G.M. Marinov, J.A. Dadakhanov, S. Happel, V.I. Radchenko, D.V. Filosofov

Joint Institute for Nuclear Research, DLNP, Dubna, Russian Federation
University of Sofia, Faculty of Chemistry and Pharmacy, Sofia, Bulgaria
Triskem International, Rue Maryse Bastié, Campus de Ker Lann, Bruz, France
Johannes-Gutenberg University Mainz, Mainz Germany

UTEVA Resin

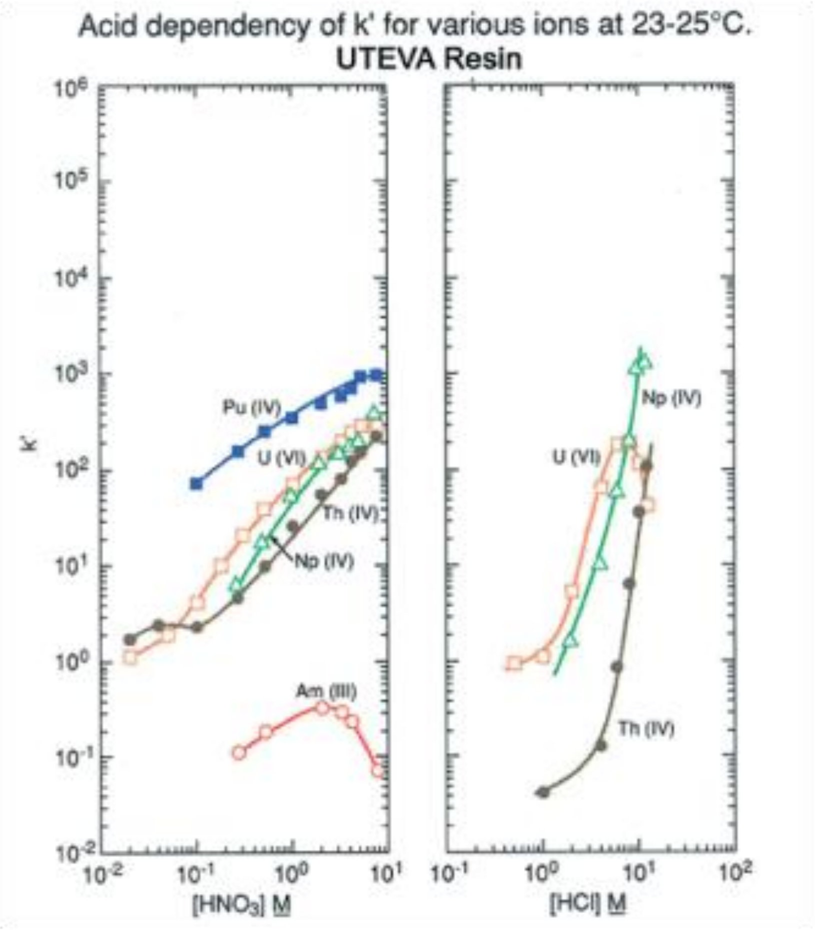
- UTEVA Resin - Uranium and Tetravalent Actinides is:
    - extraction chromatographic resin;
    - primarily used for the separation of actinides;
    - Provided by «TRISKEM INTERNATIONAL SAS».



Dipentyl pentylphosphate (DP[PP]) also called Diamyl amylphosphate (DAAP).

Dependence of distribution coefficients for several elements on UTEVA resin

- Graph of the retention factors for UTEVA resin for the actinides Th, U, Np in HNO_3 and HCl **(Triskem International)**



Purpose of the research

- Determining the distribution coefficient of the following elements: In, Sn, Sb, Te, Bi, Co, Fe, Nb, Sr, Ba, Ag, Cd, Hf, Zr, Ti for UTEVA for:
  - HCl;
  - HNO_3;
  - H_2SO_4.

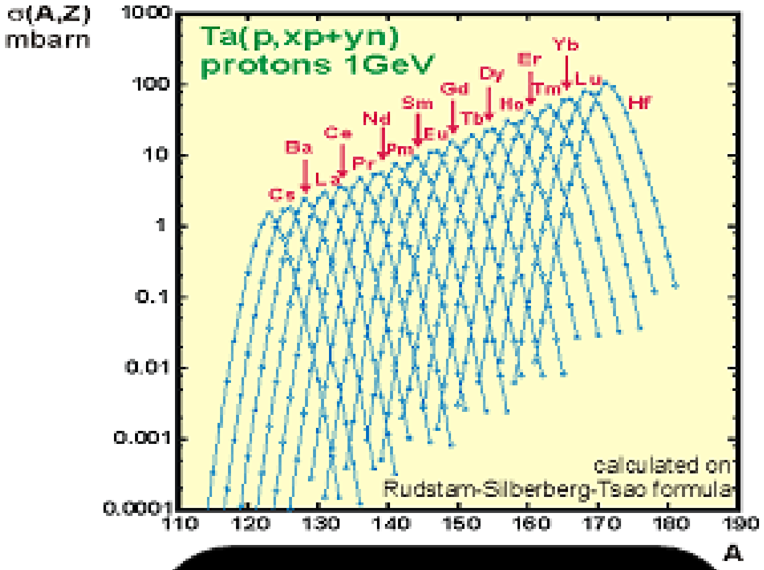
Available irradiation facilities

- Phasotron LNP (E_p=65-660 MeV, I=6 μA)

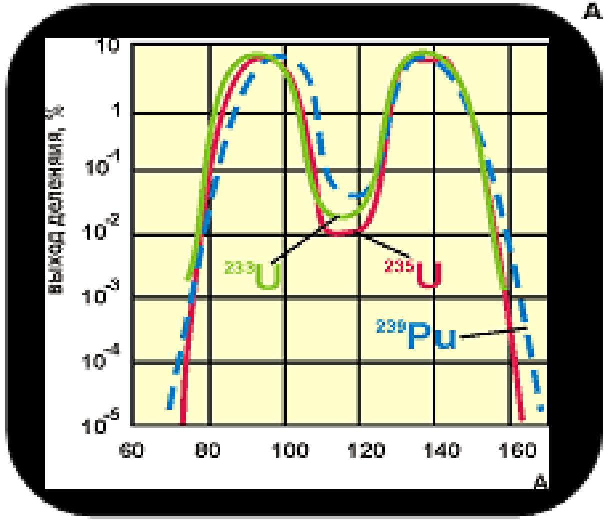
- U-200 of Laboratory of Nuclear Reaction (LNR) (E_α=36 MeV, I=70μA)
- Microtron LNR (E_β=25 MeV, I=30 μA)
- Ractor IBR-2 (2×10^{12} n/s $\times cm^2$)

Production of radionuclides

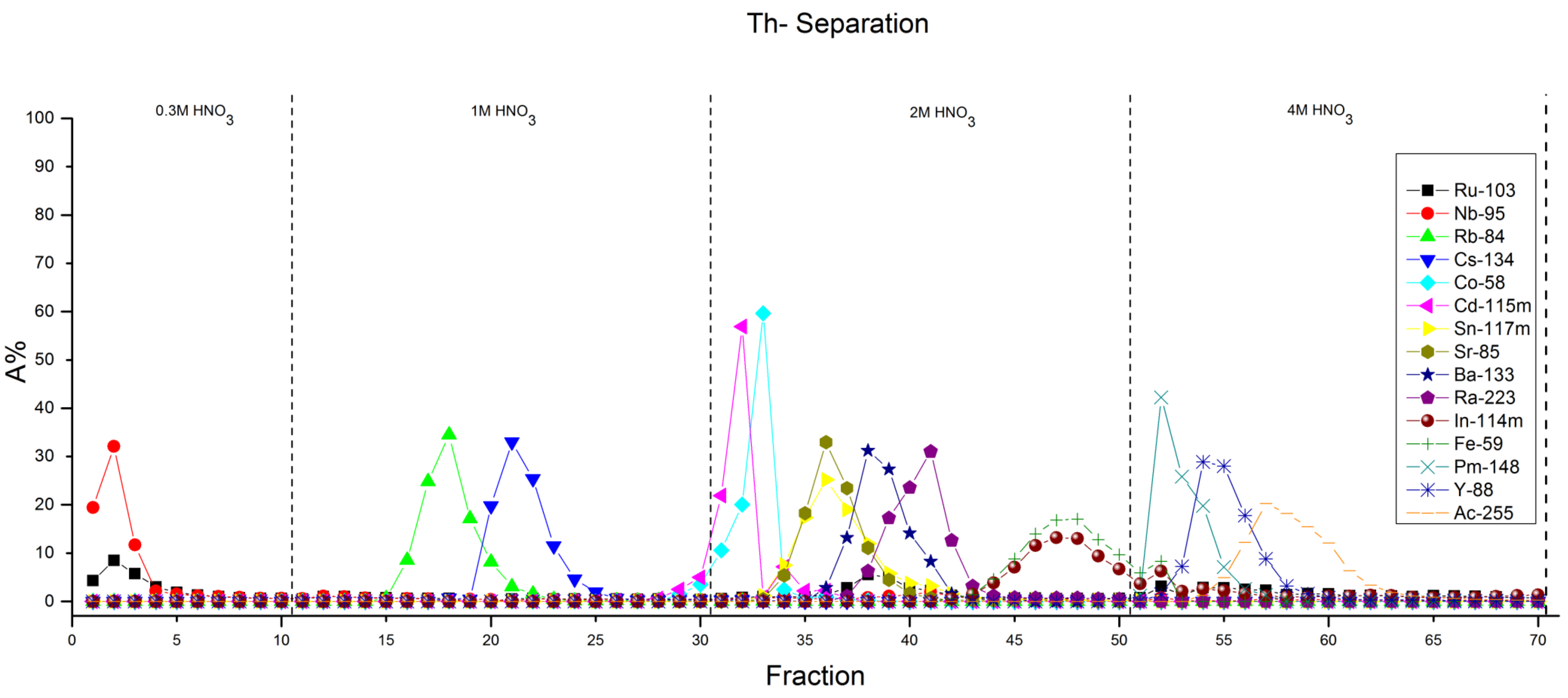
Spallation



Fission



Separation of products of nuclear reaction in thorium target



γ – lines of the used radionuclides

Isotopes	$T_{1\backslash 2}$	Energy [KeV]	Yield %
^{114m}In	49.51 d	190	15.56
^{121m}Te	154 d	212	81
^{133}Ba	10.51 y	356	62.05
^{113}Sn	115.09 d	392	64
^{85}Sr	64.84 d	514	96
^{124}Sb	60,20 d	603	98.26
^{95}Nb	34.975 d	766	100
^{207}Bi	31.55 y	570	97.74
^{59}Fe	44.503 d	1099	56.5
^{60}Co	5.2714 y	1333	99.99
^{109}Cd	462.6 d	88	3.61
^{175}Hf	70 d	343	84
^{88}Zr	83.4 d	393	97.29
^{44}Ti	63 y	68	94.4
^{110m}Ag	249.79 d	658	94.04

- Isotopes used for determination of the distribution coefficient.
- Half lifes of the isotopes, Energies and Yield of the gamma –lines are rendered in the present table.

Stock cocktail solutions

- Stock cocktail solutions are prepared using 1M HNO_3, 1M HCl and 0.1M H_2SO_4. For each stock solution, a set of radionuclides is chosen as follows:

Stock	Elements
G1	^{121m}Te, ^{124}Sb, ^{95}Nb, ^{113}Sn, ^{85}Sr, ^{133}Ba, ^{114m}In, ^{59}Fe, ^{207}Bi, ^{60}Co
G4	^{113}Sn, ^{85}Sr, ^{133}Ba, ^{114m}In
G11	^{124}Sb, ^{95}Nb, ^{109}Cd, ^{60}Co
G14	^{207}Bi, ^{110m}Ag
G21	^{88}Zr, ^{175}Hf, ^{44}Ti
G24	^{105}Ag
G34	^{207}Bi
G25	^{175}Hf, ^{88}Zr

Stock solutions with HNO_3,

containing the targeted radionuclides

Stock	Elements
G2	^{121m}Te, ^{124}Sb, ^{95}Nb, ^{113}Sn, ^{85}Sr, ^{133}Ba, ^{114m}In, ^{59}Fe, ^{207}Bi, ^{60}Co
G12	^{121m}Te, ^{124}Sb, ^{109}Cd, ^{60}Co, ^{59}Fe, ^{113}Sn, ^{114m}In, ^{95}Nb, ^{133}Ba
G22	^{88}Zr, ^{175}Hf, ^{44}Ti
G26	^{88}Zr, ^{175}Hf

Stock solutions with HCl, containing the targeted radionuclides

Stock	Elements
G3	^{121m}Te, ^{124}Sb, ^{95}Nb, ^{113}Sn, ^{85}Sr, ^{133}Ba, ^{114m}In, ^{59}Fe, ^{207}Bi, ^{60}Co
G13	^{121m}Te, ^{124}Sb, ^{109}Cd, ^{60}Co, ^{59}Fe, ^{113}Sn, ^{114m}In, ^{95}Nb, ^{133}Ba
G23	^{88}Zr, ^{175}Hf, ^{44}Ti

Stock solutions with H_2SO_4, containing the targeted radionuclides

Method

1 - initial radionuclide cocktail.

2 - filtered radionuclide cocktail.

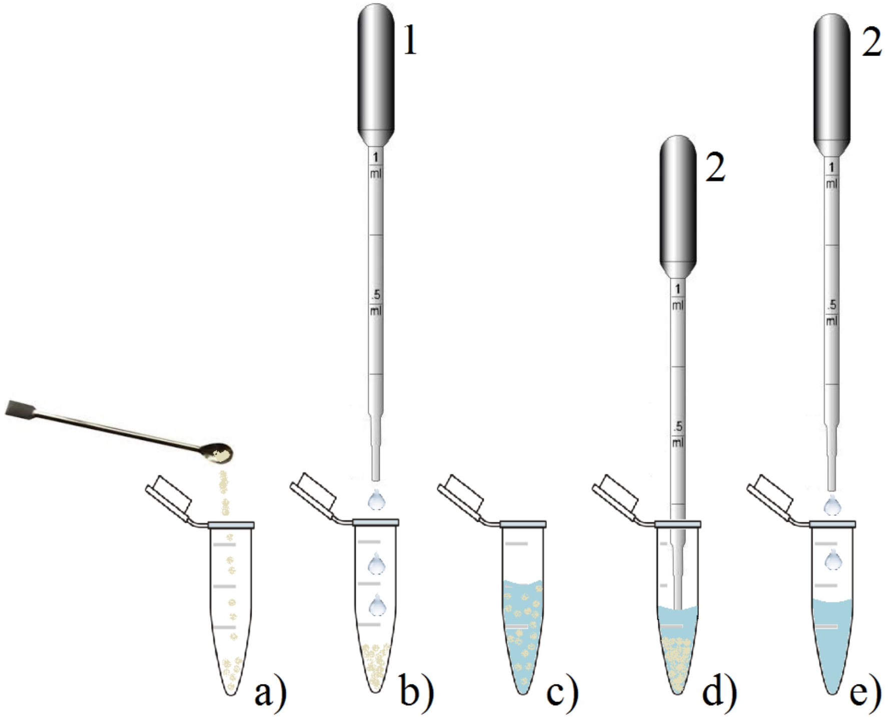
a) Loading of resin UTEVA - B.

b) Addition of a cocktail solution of radionuclides.

c) Mixing of the radionuclide cocktail and the resin for best sorption then measuring the test tubes on a γ - spectrometer.

d) Pipetting the solution without the resin.

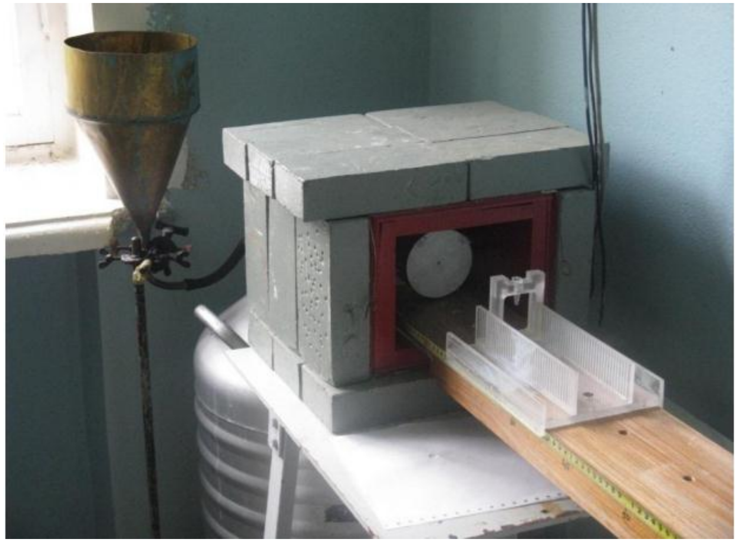
e) The pipetted solution is put in a clean test tube and measured on a γ- spectrometer.

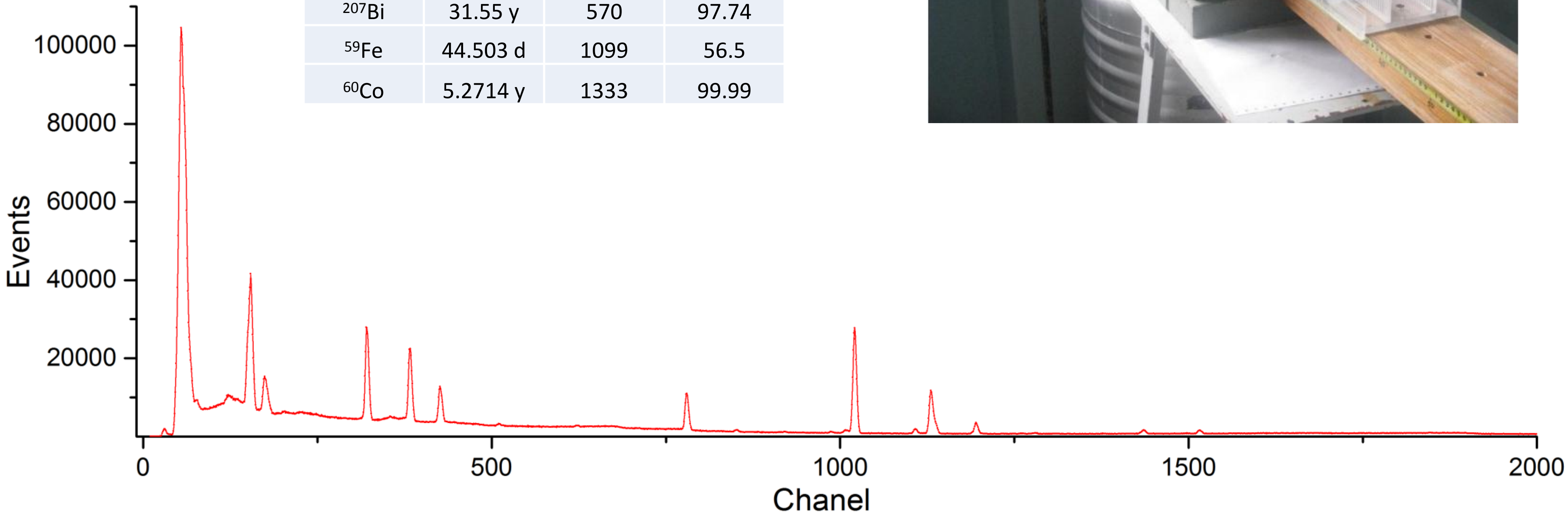


The resin particle size is 100 – 150 μm

Example of obtained spectra

Isotopes	$T_{1\backslash 2}$	Energy [KeV]	Yield %
^{114m}In	49.51 d	190	15.56
^{121m}Te	154 d	212	81
^{113}Sn	115.09 d	392	64
^{85}Sr	64.84 d	514	96
^{124}Sb	60,20 d	603	98.26
^{95}Nb	34.975 d	766	100
^{207}Bi	31.55 y	570	97.74
^{59}Fe	44.503 d	1099	56.5
^{60}Co	5.2714 y	1333	99.99





Distribution coefficient

- The distribution coefficient shows the retention property of each individual element on the resin.
- The formula for K_d consists of a relation of concentrations. However, in this case the formula that is used is a relation of radioactivity instead of concentrations as follows

$$K_d = \frac{C_{eq1}}{C_{eq2}} = \frac{A_0 - A_{eq}}{A_{eq}} * \frac{V}{m}$$

Where C_{eq1} -phase 1,
C_{eq2} -phase 2,
A_0 -activity of the initial solution,
A_{eq} - activity of the equilibrium solution,
V -volume of the equilibrium solution,
m - the resin's mass.

Distribution coefficients of radionuclides set for UTEVA resin with HNO_3 media

K_d															
HNO_3 [M]	^{114m}In	^{121m}Te	^{133}Ba	^{113}Sn	^{85}Sr	^{124}Sb	^{95}Nb	^{207}Bi	^{59}Fe	^{60}Co	^{109}Cd	^{175}Hf	^{88}Zr	^{44}Ti	^{110m}Ag
0.01	< 1	< 1	< 1	25	< 1	31	73	< 1	< 1	< 1	< 1	31	12	< 1	21
0.03	< 1	< 1	< 1	5	< 1	18	10	< 1	< 1	< 1	< 1	19	11	~ 1	13
0.06	< 1	< 1	< 1	4	< 1	11	11	< 1	< 1	< 1	< 1	15	6.7	~ 1	20
0.1	< 1	< 1	< 1	< 1	< 1	16	15	< 1	< 1	< 1	< 1	8.3	4.3	< 1	23
0.3	< 1	< 1	< 1	< 1	< 1	61	< 1	3.7	< 1	< 1	< 1	4.1	3.3	< 1	4.3
0.6	< 1	< 1	< 1	< 1	< 1	21	3	10	< 1	< 1	< 1	3.1	4.1	< 1	5.8
1	< 1	< 1	< 1	< 1	< 1	4.5	11	7.1	< 1	< 1	< 1	2.3	7.5	< 1	~ 1
3	< 1	< 1	< 1	< 1	< 1	3.8	< 1	1.5	< 1	< 1	< 1	2.2	31	< 1	3.5
3.6	-	-	-	-	-	~ 1	2.9	< 1	-	< 1	-	4.5	43	< 1	2.2
6	< 1	< 1	~ 1	< 1	< 1	~ 1	24	< 1	< 1	< 1	< 1	64	280	< 1	~ 1
8	-	-	-	-	-	~ 1	17	< 1	-	< 1	-	615	790	< 1	< 1
10	< 1	< 1	< 1	~ 1	< 1	5.3	47	< 1	< 1	< 1	< 1	< 1000	< 1000	< 1	< 1

Distribution coefficients of radionuclides set for UTEVA resin with HCl media

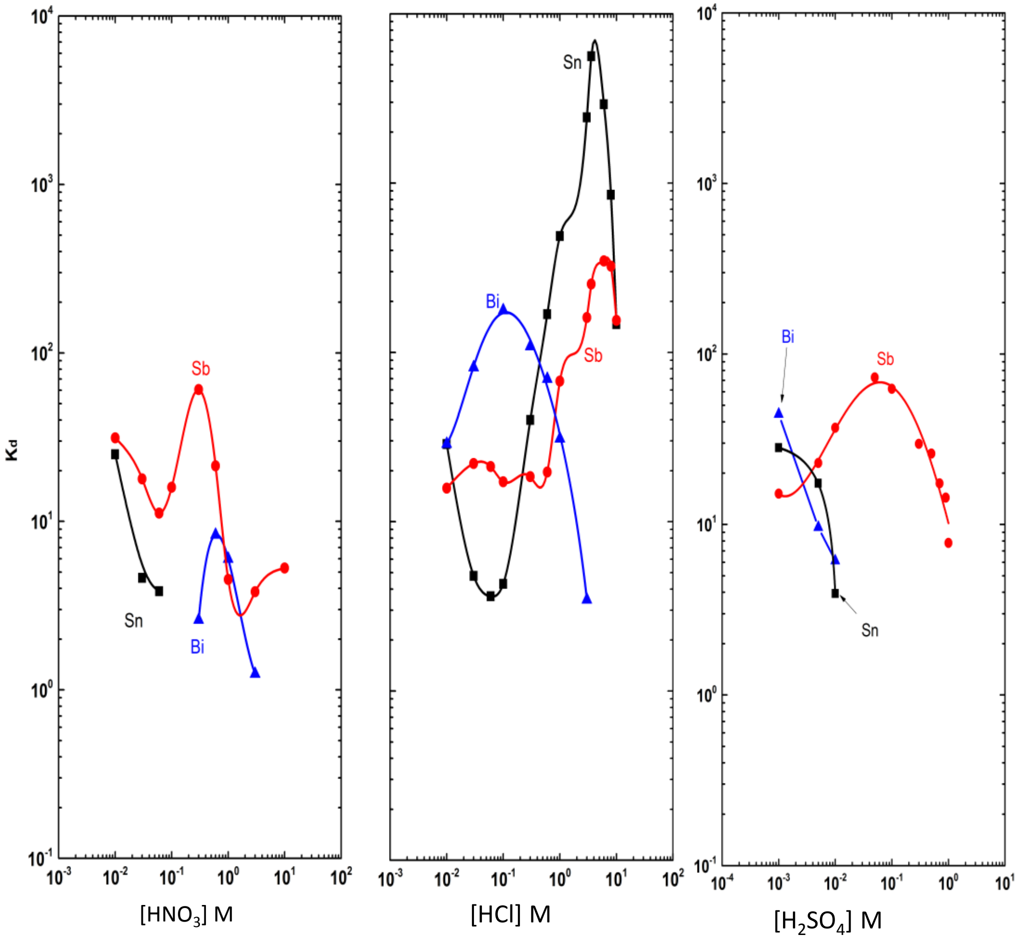
K_d														
HCl [M]	^{114m}In	^{121m}Te	^{133}Ba	^{113}Sn	^{85}Sr	^{124}Sb	^{95}Nb	^{207}Bi	^{59}Fe	^{60}Co	^{109}Cd	^{175}Hf	^{88}Zr	^{44}Ti
0.01	< 1	~ 1	< 1	29	< 1	16	34	29	~ 1	< 1	< 1	210	77	< 1
0.03	< 1	~ 1	< 1	4.8	< 1	22	28	82	< 1	< 1	< 1	67	23	< 1
0.06	< 1	~ 1	< 1	3.6	< 1	21	14	58	< 1	< 1	< 1	25	12	< 1
0.1	< 1	~ 1	< 1	4.3	< 1	17	12	180	< 1	< 1	< 1	8.8	2.4	< 1
0.3	< 1	~ 1	< 1	40	< 1	18	7.6	110	< 1	< 1	< 1	4.4	2	< 1
0.6	< 1	~ 1	< 1	170	< 1	20	5.3	70	2.5	< 1	< 1	~ 1	< 1	< 1
1	3.2	5.1	< 1	490	< 1	68	5.4	31	7.5	< 1	< 1	< 1	< 1	< 1
3	25	150	< 1	< 1000	< 1	160	4.6	3.5	120	< 1	15	< 1	< 1	< 1
3.6	62	220	< 1	< 1000	< 1	250	36	< 1	630	< 1	38	< 1	< 1	< 1
6	110	290	< 1	< 1000	< 1	350	700	< 1	610	< 1	16	12	31	1.9
8	39	320	< 1	850	< 1	320	460	-	270	< 1	< 1	590	400	2.8
10	12	280	< 1	150	< 1	160	400	< 1	240	< 1	< 1	< 1000	< 1000	94

Distribution coefficients of radionuclides set for UTEVA resin with H_2SO_4 media

K_d														
H_2SO_4 [M]	^{114m}In	^{121m}Te	^{133}Ba	^{113}Sn	^{85}Sr	^{124}Sb	^{95}Nb	^{207}Bi	^{59}Fe	^{60}Co	^{109}Cd	^{175}Hf	^{88}Zr	^{44}Ti
0.001	5.7	< 1	< 1	28	< 1	15	50	44	4.7	< 1	< 1	140	75	4.8
0.005	2	< 1	< 1	17	< 1	23	38	9.6	3.1	< 1	< 1	220	71	< 1
0.01	< 1	< 1	< 1	3.9	< 1	37	13	6.1	< 1	< 1	< 1	110	30	< 1
0.05	< 1	< 1	< 1	< 1	< 1	73	1.9	< 1	< 1	< 1	< 1	3.9	1.3	< 1
0.1	< 1	< 1	< 1	~ 1	< 1	62	9	~ 1	< 1	< 1	< 1	1.3	< 1	< 1
0.3	< 1	< 1	< 1	< 1	< 1	30	3.2	< 1	< 1	< 1	< 1	< 1	< 1	< 1
0.5	< 1	< 1	< 1	< 1	< 1	26	12	< 1	< 1	< 1	< 1	< 1	< 1	< 1
0.7	< 1	< 1	< 1	< 1	< 1	17	1	< 1	< 1	< 1	< 1	< 1	< 1	< 1
0.9	< 1	< 1	< 1	< 1	< 1	14	3.3	< 1	< 1	< 1	< 1	< 1	< 1	< 1
1	< 1	< 1	< 1	< 1	< 1	7.8	5.9	~ 1	~ 1	< 1	< 1	< 1	< 1	< 1

Distribution coefficients of Sn, Sb, Bi for UTEVA with HNO_3, HCl and H_2SO_4

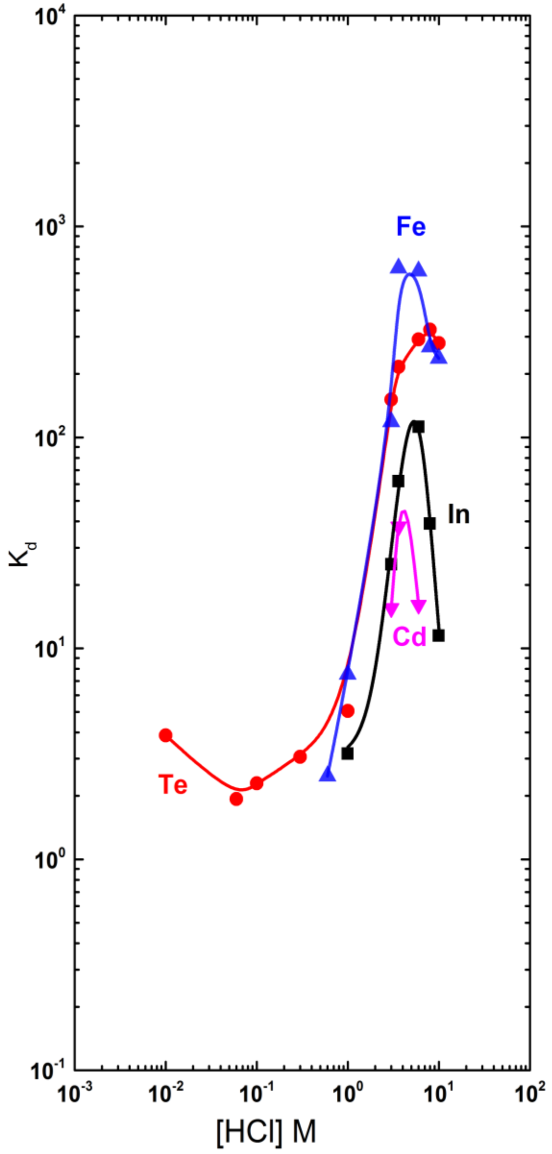
- *Distribution coefficients for the elements Bi, Sn and Sb with respect to the concentration of nitric, hydrochloric and sulfuric acid.*



Distribution coefficients of Te, Fe, In, Cd for UTEVA Resin

- *Distribution coefficients of Te, Fe, In, Cd with respect to the concentration of hydrochloric acid.*

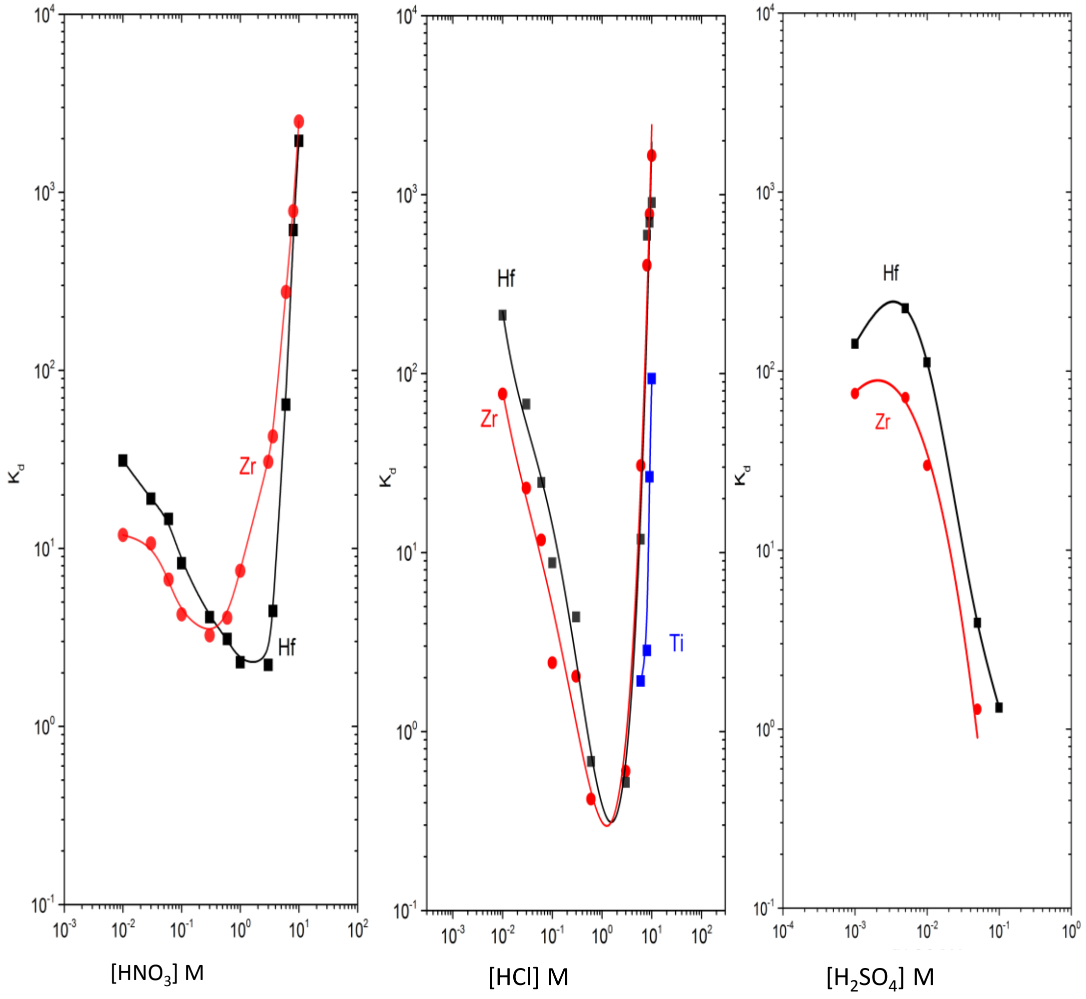
- *Te, Fe, Cd and In do not retain on UTEVA resin when using nitric or sulfuric solution.*



Distribution coefficients of the elements Hf, Zr and Ti for UTEVA Resin with HNO_3, HCl and H_2SO_4

- *Distribution coefficients for the elements Hf, Zr and Ti for UTEVA resin with respect to the concentration of nitric, hydrochloric and sulfuric acid.*

- *Titanium does not retain on UTEVA resin with HNO_3 or H_2SO_4 it retains only in high concentrations of HCl.*



Conclusions

- Data for the distribution coefficients of the elements: In, Sn, Sb, Te, Bi, Co, Fe, Nb, Sr, Ba, Ag, Cd, Hf, Zr, Ti is obtained for UTEVA in HCl, HNO_3 and H_2SO_4.
- The resulting information allows the development of methods for the separation of a number of elements from each other, for example - Hf, Zr, Ti.
- The obtained distribution coefficients with low values allow analysis of samples containing actinides (U, Th and Pu).
- The data received from the research shows that UTEVA can be used for effective purification of low-background samples from actinides.