DTU Nutech
Center for Nuclear Technologies

Extraction chromatography in determination of radionuclides in environment, waste and radiopharmaceutics

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Importan radionuclides in environment and waste

- Important natural radionuclides in environment

  ^{3}H, ^{7}Be, ^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl, ^{41}Ca, ^{129}I, $^{228, 226}Ra$, ^{210}Po, ^{210}Po, Isotopes of U and Th

- Important artifical radionuclides in environment

  ^{3}H, ^{14}C, ^{90}Sr, ^{137}Cs, ^{134}Cs, ^{99}Tc, ^{129}I, $^{238,239,240,241}Pu$, ^{237}Np, ^{241}Am, etc.

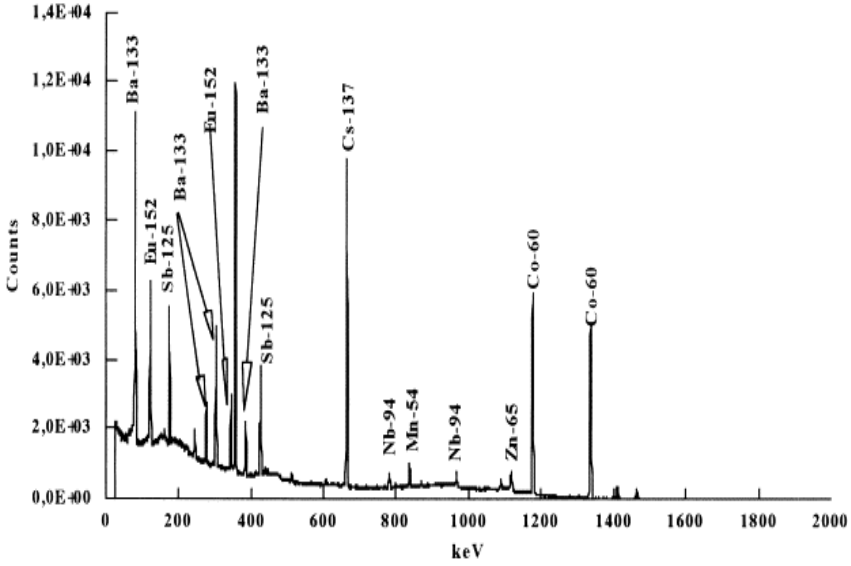
- Important radionuclides in nuclear waste

  ^{3}H, ^{14}C, ^{90}Sr, ^{137}Cs, ^{134}Cs,, ^{99}Tc, ^{129}I, $^{238,239,240,241}Pu$, ^{237}Np, ^{241}Am, ^{242}Cm, ^{36}Cl, ^{41}Ca, ^{55}Fe, ^{59}Ni, ^{63}Ni, ^{60}Co, ^{133}Ba, ^{135}Cs ^{152}Eu, ^{79}Se, ^{93}Zr, ^{93}Mo, ^{94}Nb, etc.

Major Radionuclides in the environment and waste

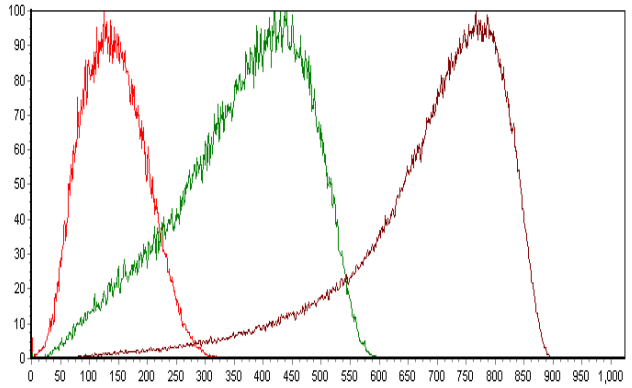
- γ- radionuclides

  ^{60}Co, ^{133}Ba, ^{137}Cs, ^{134}Cs, ^{106}Ru, $^{152,154,155}Eu$, ^{58}Co, ^{54}Mn, ^{59}Fe, ^{110m}Ag, ^{94}Nb.

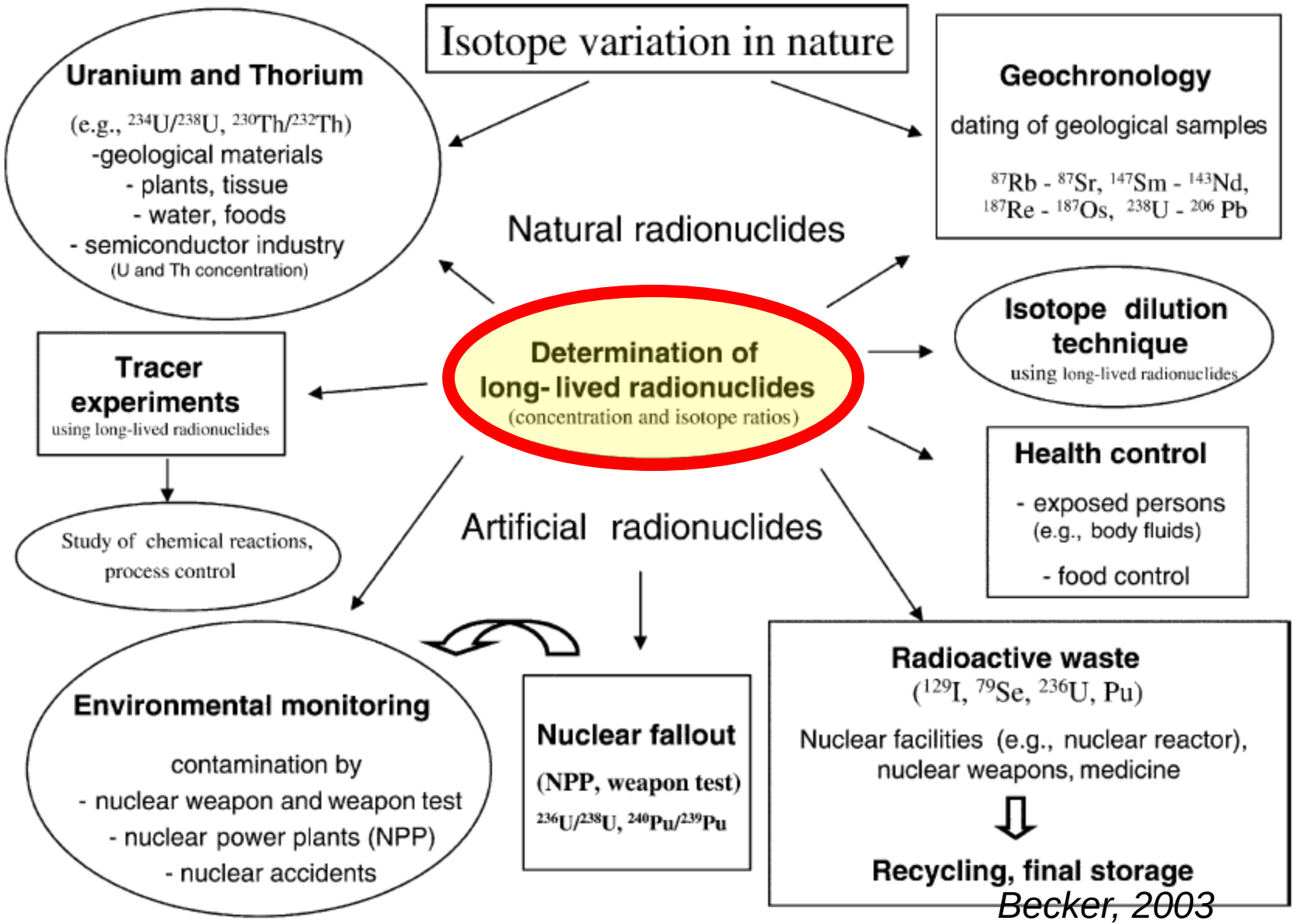


Difficult-to-measure radionuclides

- β- Emitter
  - ^{3}H, ^{14}C, ^{36}Cl, ^{41}Ca, ^{55}Fe, $^{63,59}Ni$, ^{93}Zr, ^{93}Mo, ^{90}Sr, ^{99}Tc, ^{129}I, ^{241}Pu.
- α- emitter (actinides)
  - $^{238\text{-}240}Pu$, ^{241}Am, $^{243,244}Cm$, ^{237}Np

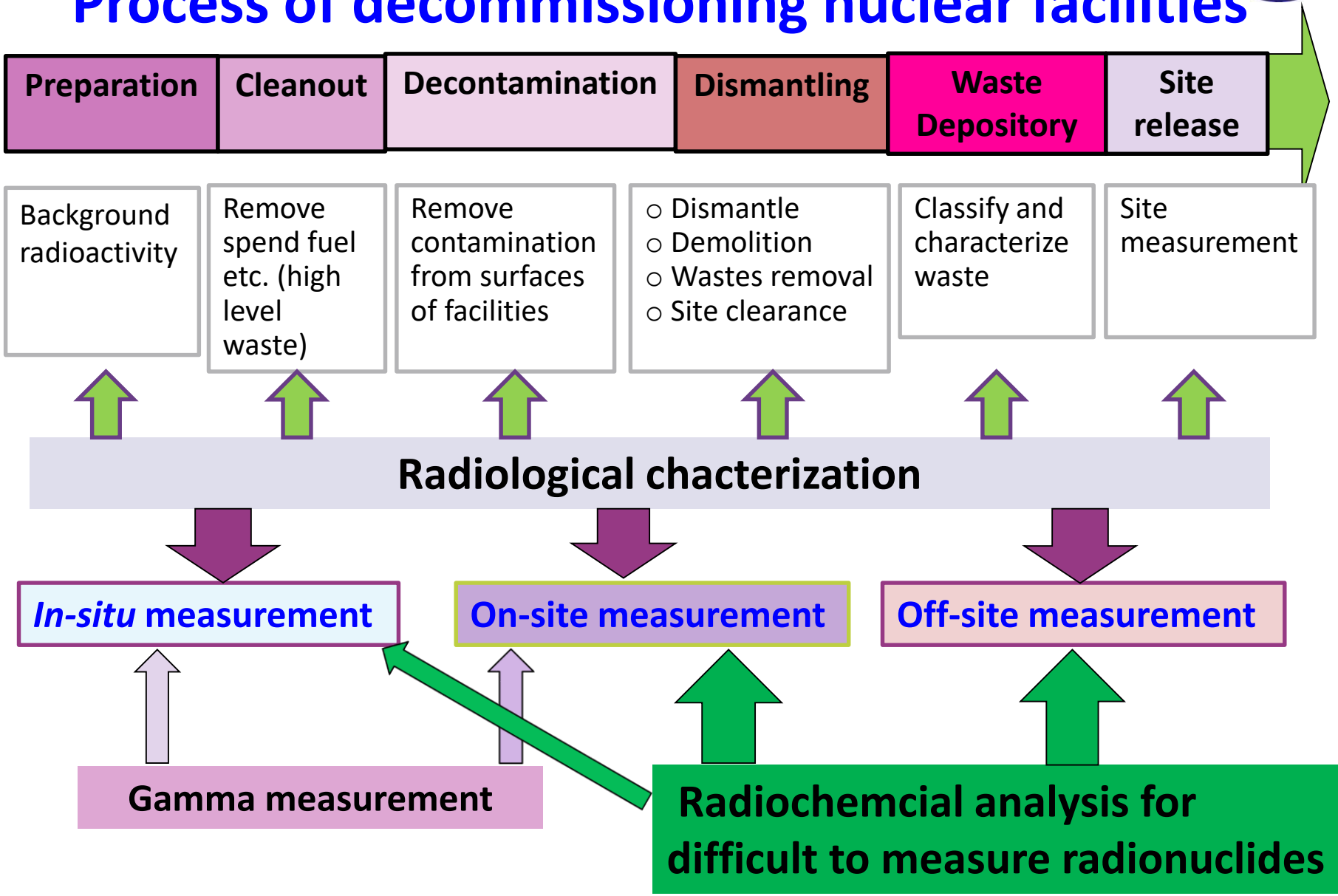


Application of radionuclides



Becker, 2003

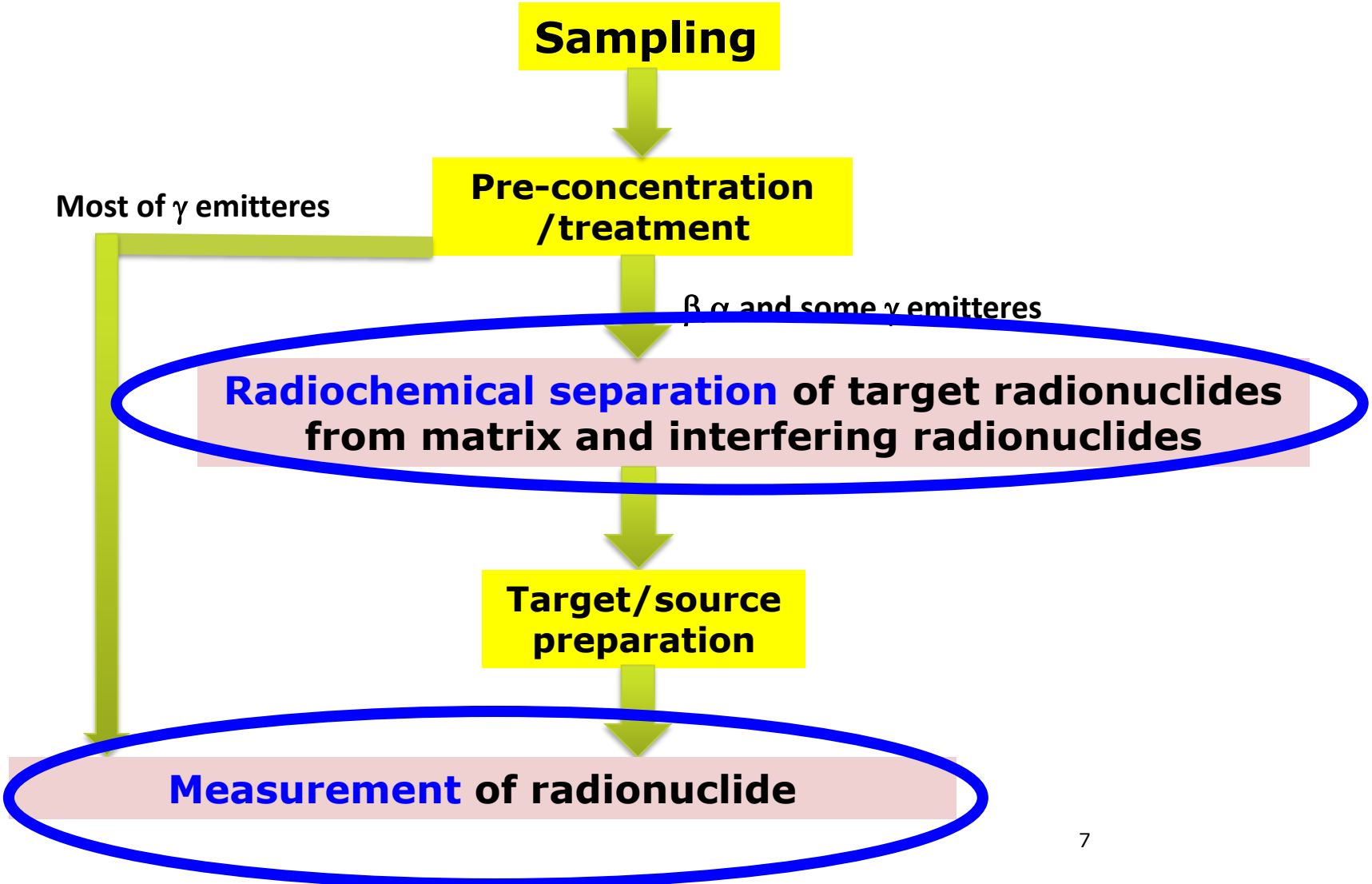
Process of decommissioning nuclear facilities



Production of medical radioisotopes

- **Preparation of irradiation targets**
- Irradiation: production of radioisotope by nuclear reaction
- **Separation of radioisotope:**
  - Remove from the matrix
  - Remove the impurities
  - Recovery of the enrich isotope
- **Quality control of the radioisotope product**
  - Radionuclidic purity
  - Radiochemcial purity

Procedure for Radiochemical Analysis of Radionuclides



Methods for chemcial separation of radionuclides

- Precipitation-co-precipitation (^{90}Sr、^{63}Ni 、^{41}Ca、 ^{226}Ra、 ^{36}Cl、Pu、U、Np)
- Selective adsorption (^{137}Cs、^{210}Po)
- Combustion/heating (^{131}I、^{129}I、^{103}Ru、^{99}Tc)
- Solvent extraction (U、Pu、Np、 Am、 ^{99}Tc、^{131,129}I)
- Ion exchange chromatography (Pu、 Np、Am、^{129}I, ^{99}Tc, ^{64}Cu, ^{10}Be)
- **Extraction chromatography** (^{63}Ni、^{55}Fe, ^{99}Tc、Pu、Np、U、Am、^{90}Sr、^{210}Pb、^{10}Be、etc.)

Application of extraction chromatograhy in determination of radionuclides

- **Environmental radioactivity and tracer studies**
  - ✓ Determination of U, Pu, Np and Am isotopes in environmental samples
  - ✓ Determination of ^{99}Tc in environmental samples.
- **Characteristation of decommissioning waste**
  - ✓ Determination ^{63}Ni and ^{59}Ni in decommissioning waste
- **Application in radioisotope production**
  - ✓ Quality control of ^{99}Mo-^{99m}Tc generator
  - ✓^{67}Ga produced by cyclotron

Determination of U, Pu, Np and Am isotopes in environmental samples

Qiao, Hou, Roos & Miro,
Anal. Chem, 2009; 2011; 2015
Anal Chim Acta, 2010
Talanta, 2011
JAAS 2011
J. Environ. Radioact. 2010, 2012
Environ. Sci. Technol. 2016

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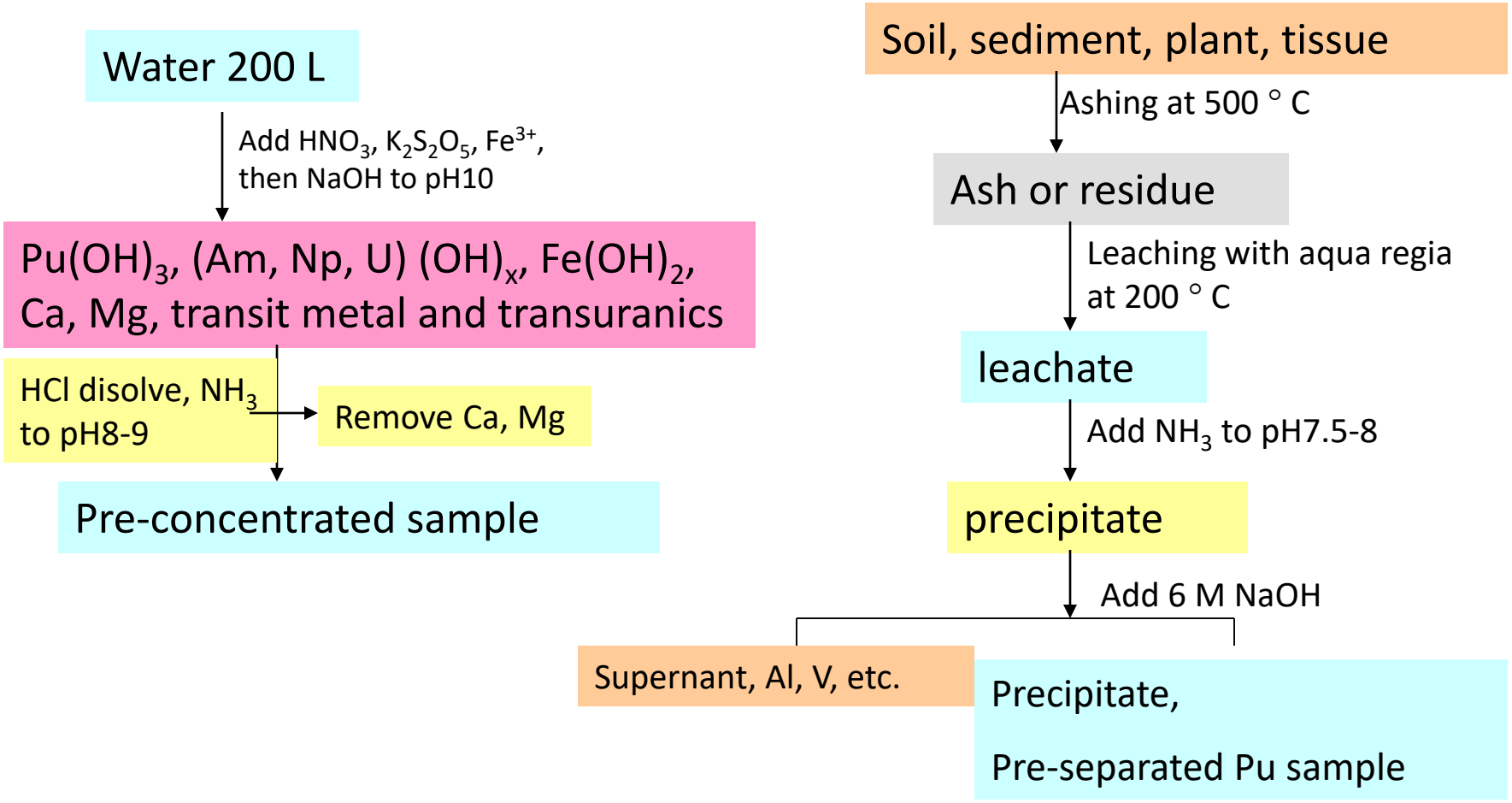
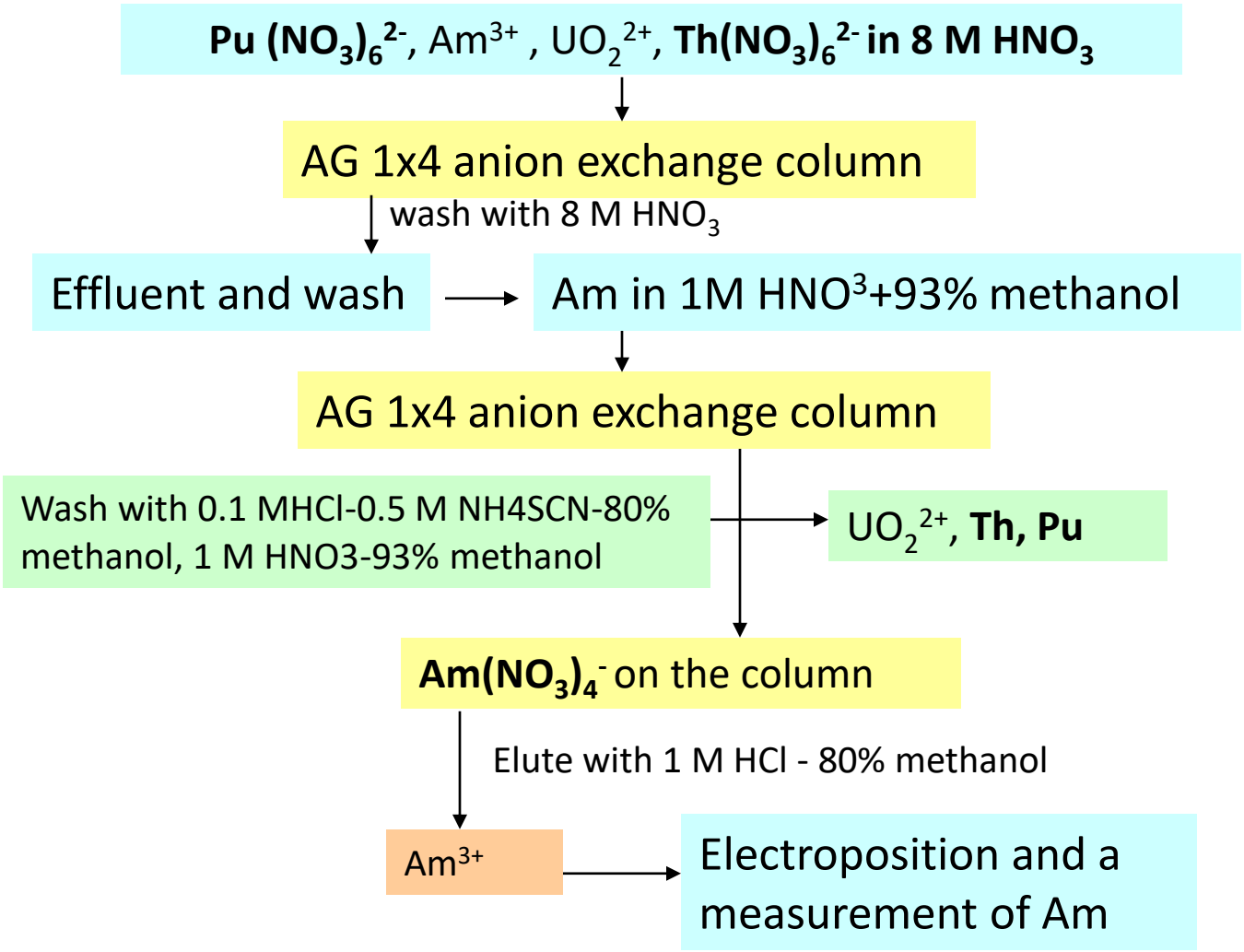
Preconcentration of U, Pu, Np, Am



Formation of complex of Pu, Np, U and Am in HNO_3 and HCl media

Ion	HCl <6M	HCl >6M	HNO_3 <7 M	HNO_3 >7 M
Pu^{3+}	$PuCl^{2+}$, $PuCl_2^{+}$	$PuCl^{2+}$, $PuCl_2^{+}$	$Pu(NO_3)^{2+}$, $Pu(NO_3)_2^{+}$	$Pu(NO_3)^{2+}$, $Pu(NO_3)_2^{+}$
Pu^{4+}	$PuClx^{y+}$	$PuCl_6^{2-}$	$Pu(NO_3)_x^{y+}$	$Pu(NO_3)_6^{2-}$
PuO_2^{2+}	$PuO_2Cl_3^{-}$, $PuO_2Cl_2^{2-}$	$PuO_2Cl_3^{-}$, $PuO_2Cl_2^{2-}$	No complex	$PuO_2(NO_3)_2$
UO_2^{2+}	No anion complex	$UO_2Cl_4^{2-}$	No anion complex	No anion complex
Th^{4+}	No anion compex	No anion complex	No anion complex	$Th(NO_3)_6^{2-}$
Am^{3+}	No anion complex	No anion complex	No anion complex	No anion complex
Po^{2+}		$PoCl_6^{2-}$		$Po(NO_3)_6^{2-}$
Np^{4+}	No	$NpCl_6^{2-}$	No	$Np(NO_3)_6^{2-}$
NpO_2^{+}	No	No	No	$NpO_2(NO_3)_6^{-}$

Separation of Am from Pu, Th and U by anion exchange



Separation of U, Pu, Np, Am based on extraction chromatography

HNO_3 medium

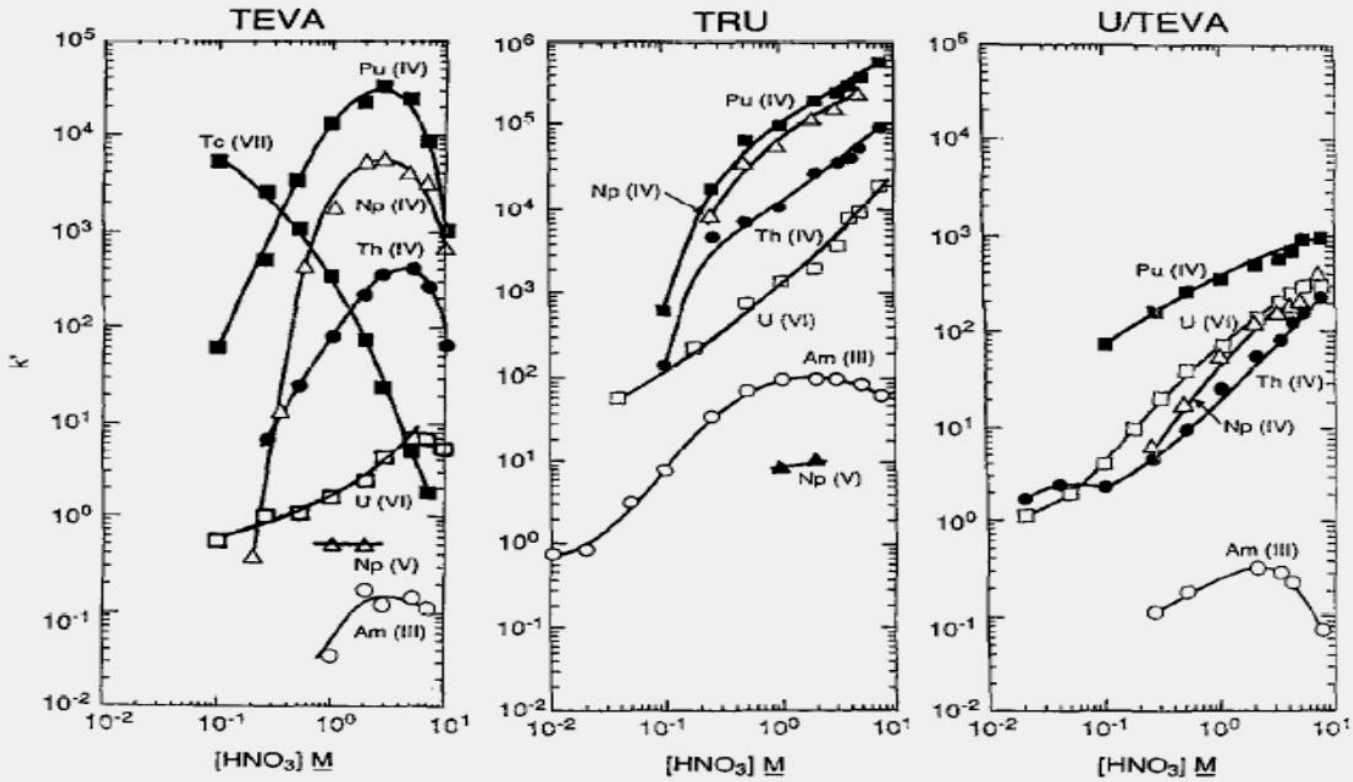


Fig. 1. Nitric acid dependencies of k' for selected elements with the TEVA, TRU, and U/TEVA extraction chromatographic resins ($T = 23–25°$ C; 50–100 μm particle size resins).

E. P. Horwitz, et.al., Anal. Chim. Acta, 1995, 310, 63-7

Separation of U, Pu, Np, Am based on extraction chromatography

HCl medium

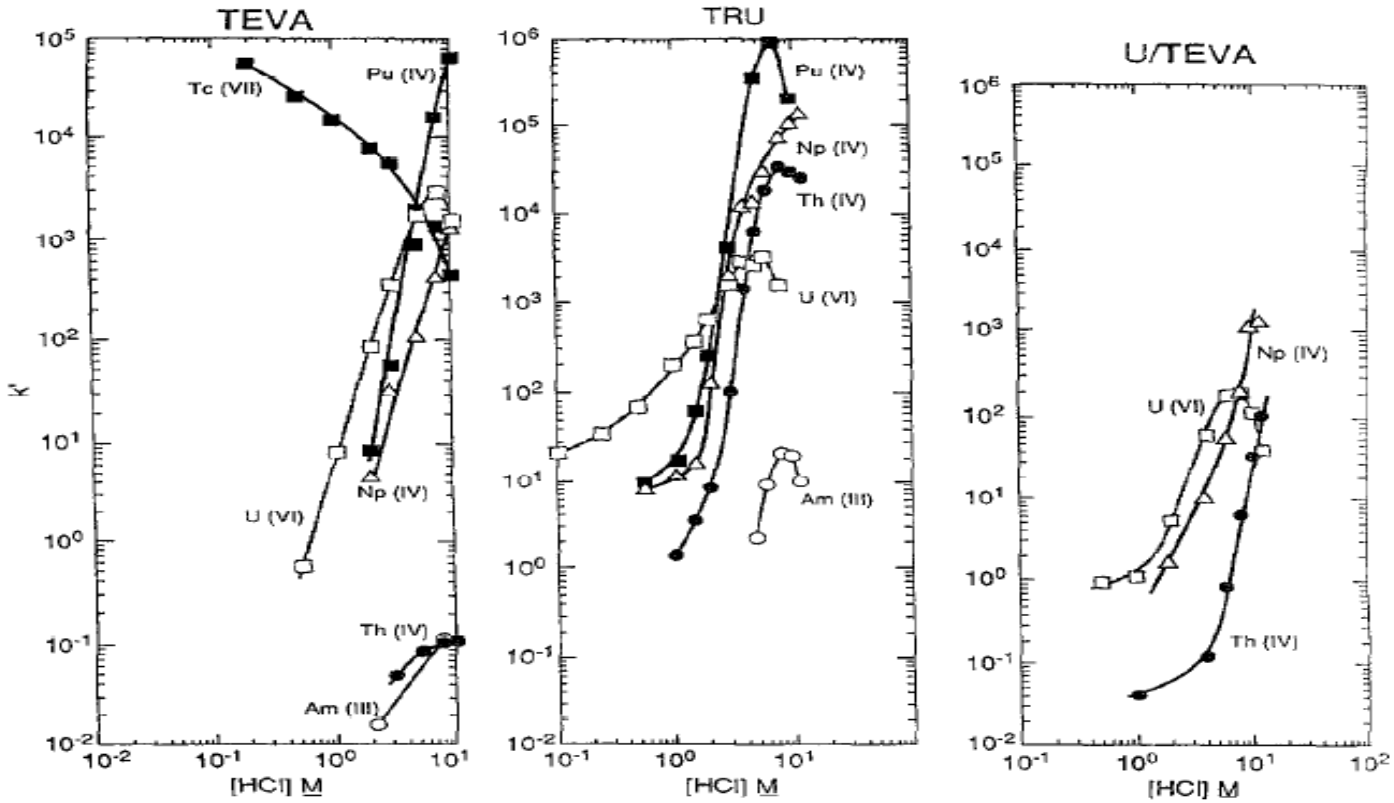


Fig. 2. Hydrochloric acid dependencies of k' for selected elements with the TEVA, TRU, and U/TEVA extraction chromatographic resins ($T = 23–25°$ C; 50–100 μm particle size resins).

E. P. Horwitz, et.al., Anal. Chim. Acta, 1995, 310, 63-7

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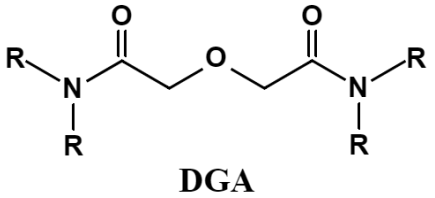
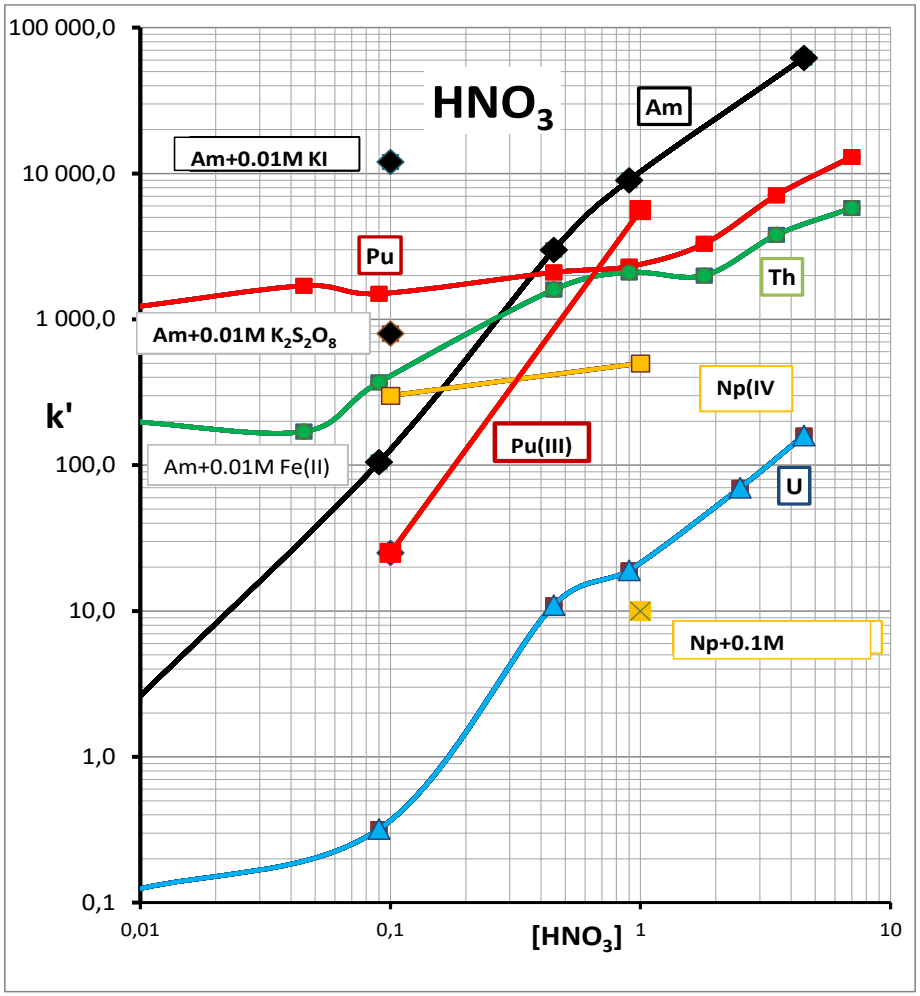
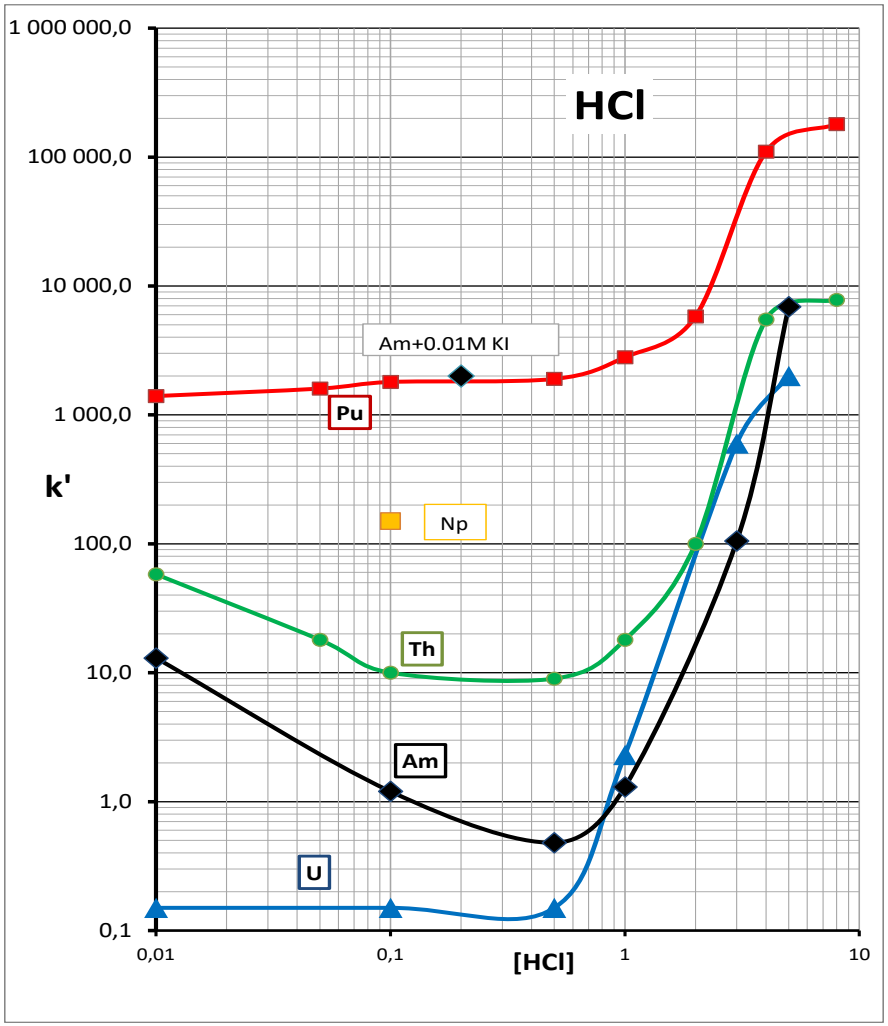
Separation of U, Pu, Np, Am based on extraction chromatography



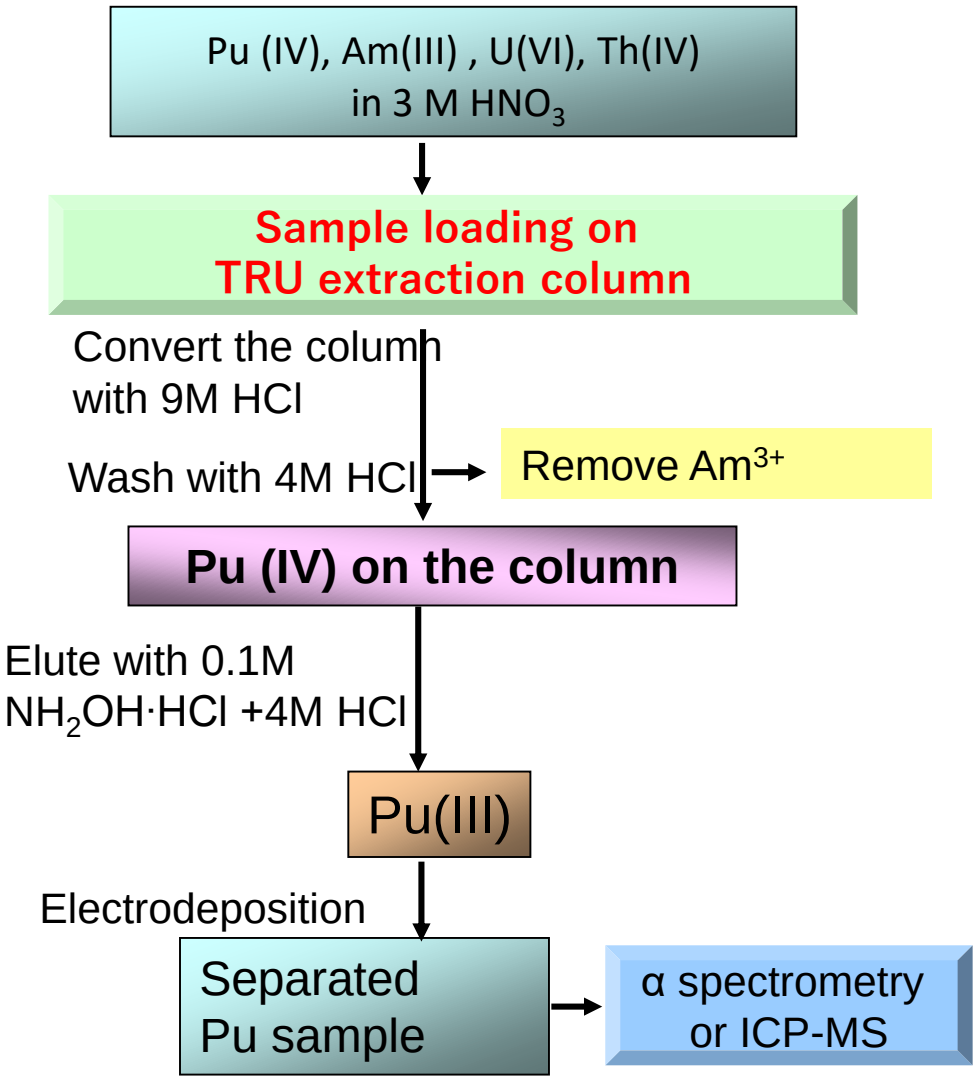
DGA





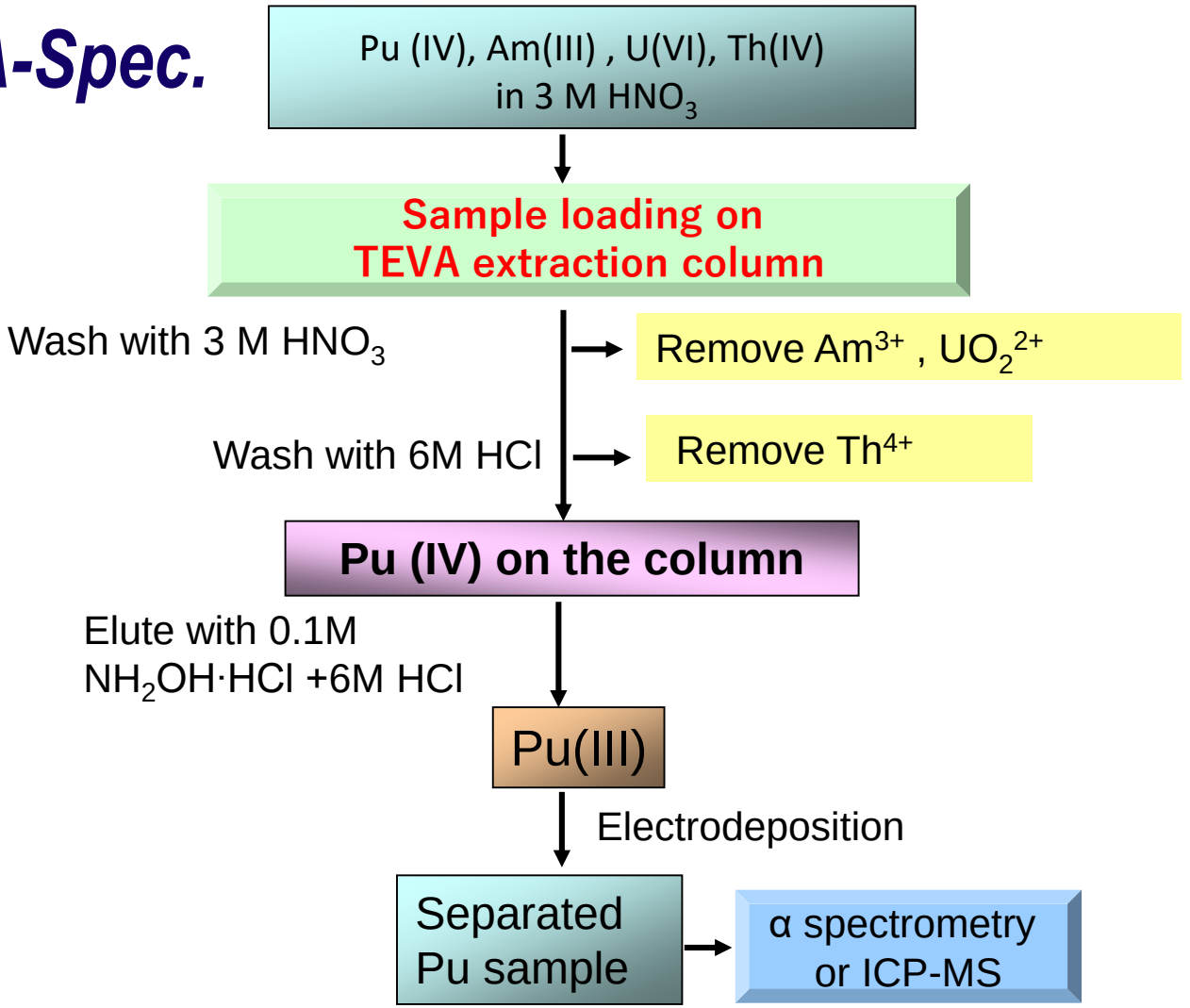
Separation of Pu based on anoin exchange chromatography

using TRU-Spec.

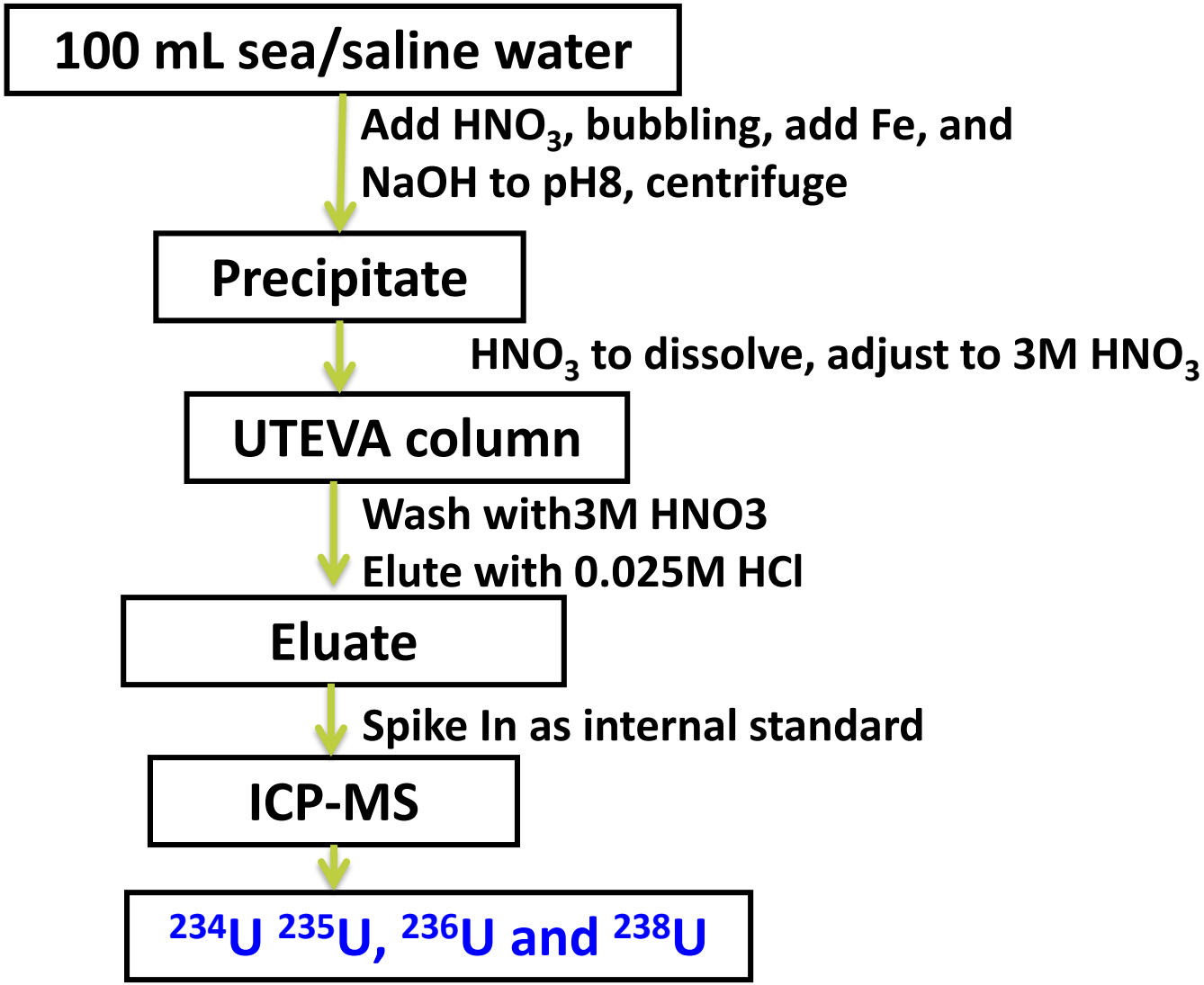


Separation of Pu based on extraction chromatography

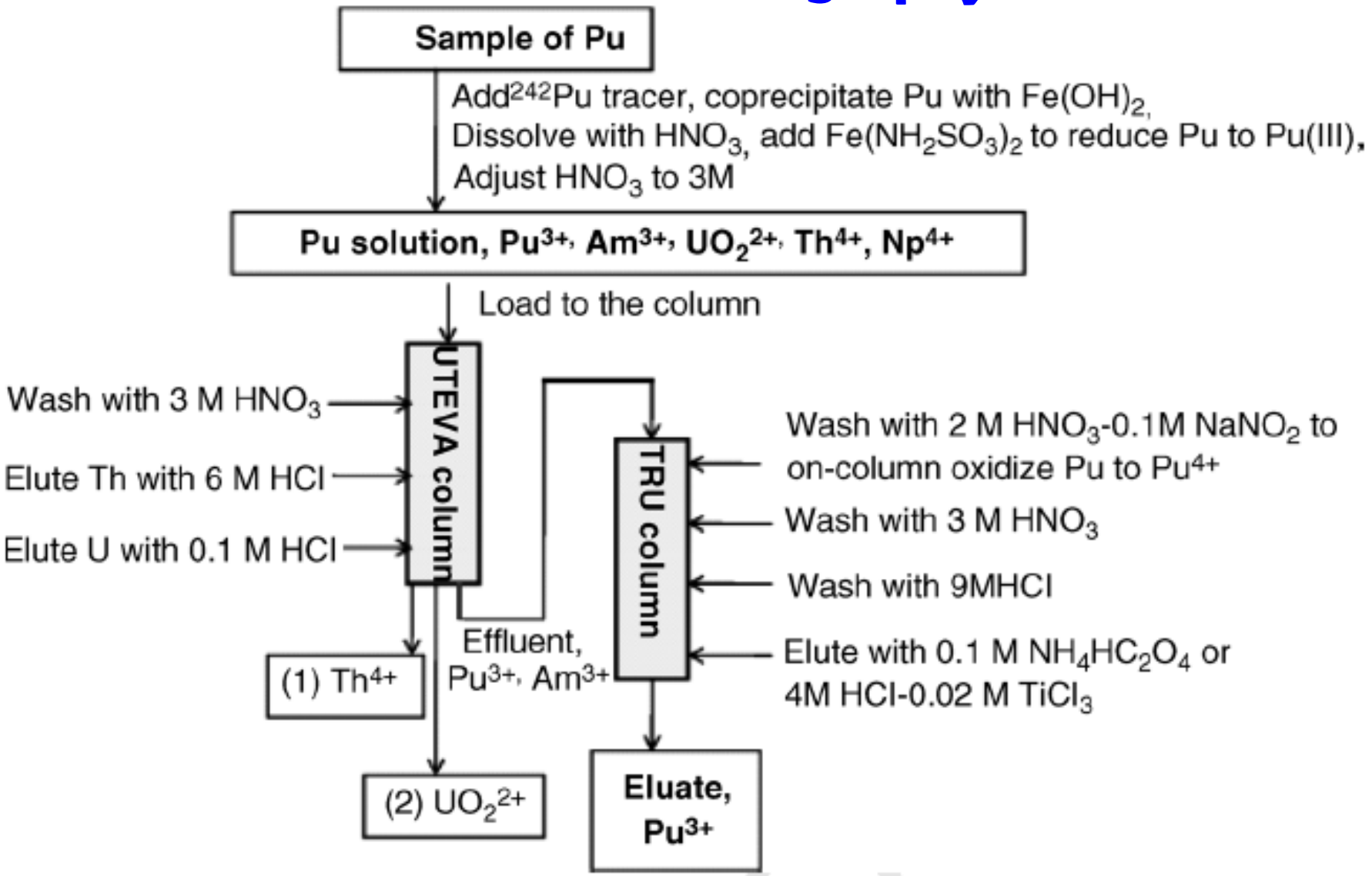
using TEVA-Spec.



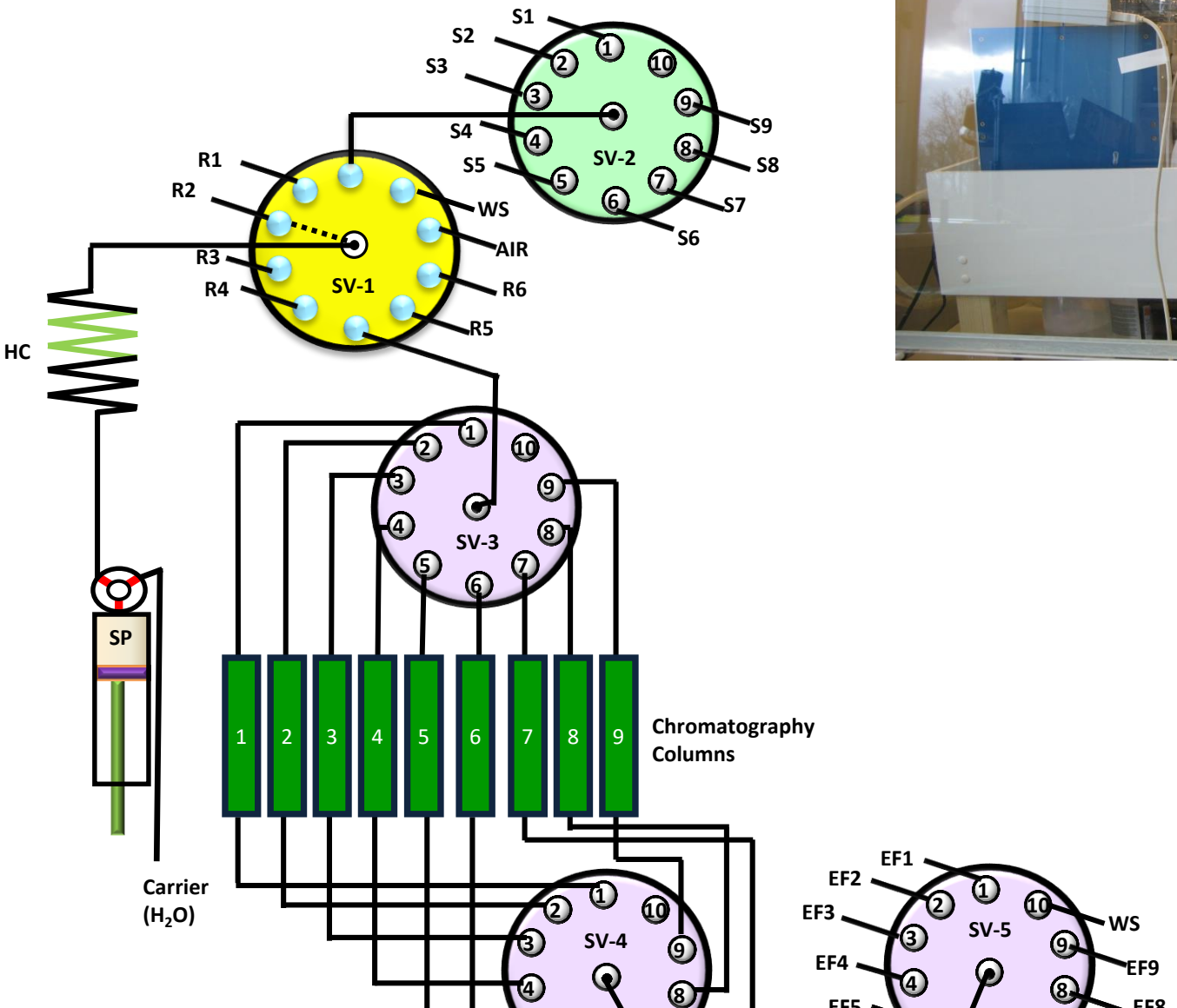
Determination of ^{234}U ^{235}U, ^{236}U and ^{238}U in sea water samples



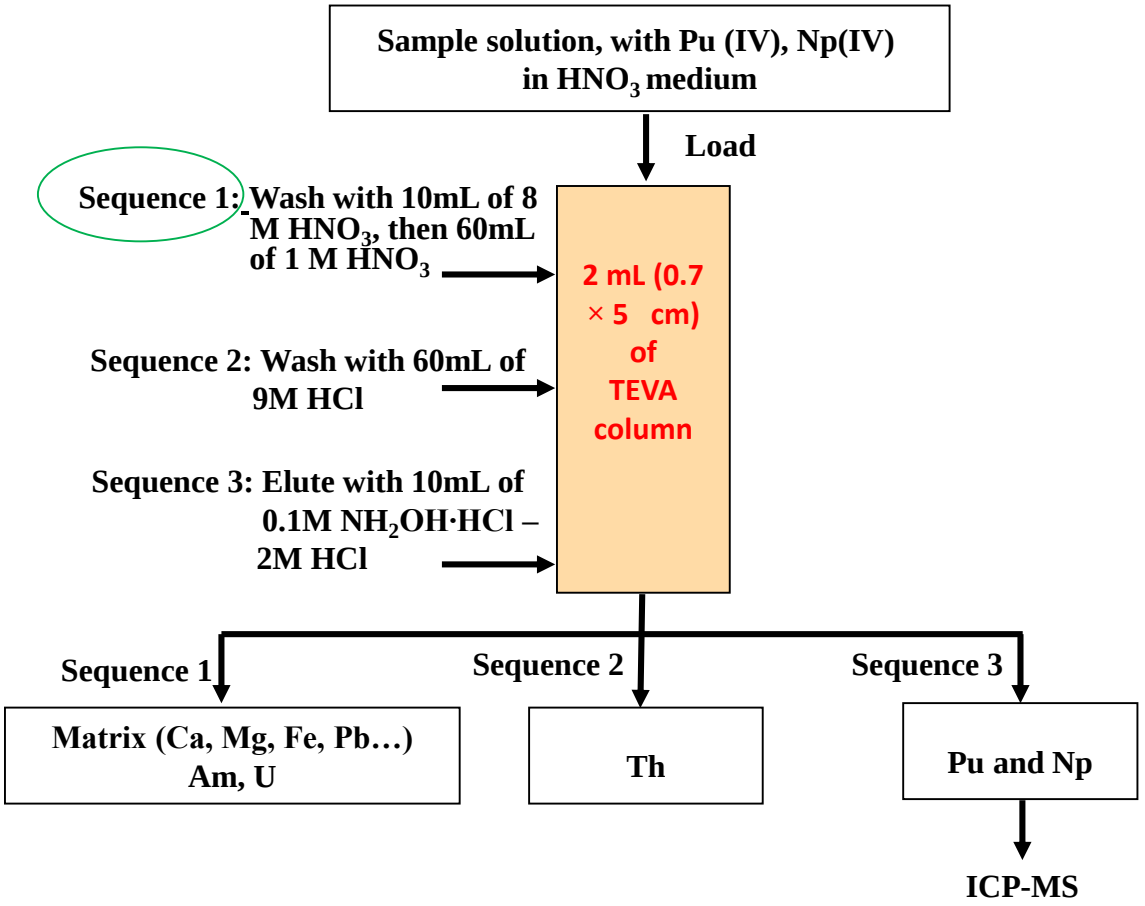
Separation of Pu and U using sequential extraction chromatography



Sepup for automated separation using sequential injection approach



Separation of Np and Np using TEVA column and automated separation system



Loading: 0.2hr

Rinsing: 1hr

Pu & Np elution: 0.3hr

Measurement : 0.5 hr (including sample preparation)

Total: 1.5 + 0.5 = 2hr

Analytical Results of some SRM and samples

Sample name	Recovery†, %	^{239}Pu measured†, Bq/kg	^{240}Pu measured†, Bq/kg	^{237}Np measured†, Bq/kg	Expected concentration, Bq/kg		
					^{239}Pu	^{240}Pu	^{237}Np
IAEA-135 sediment	59.7 ± 6.9	125.7 ± 5.4	93.6 ± 2.6	0.78 ± 0.04	123.0 ± 3.8 [0.87] ‡	93.8 ± 2.3 [0.13]	0.846 ± 0.045 [2.86]
Irish Sea sediment	64.7 ± 15.9	223.7 ± 7.3	191.3 ± 5.9	1.02 ± 0.04	237.0 ± 5.0 [3.16]	181.0 ± 4.9 [3.02]	0.987 ± 0.048 [1.43]
NIST-4359 Seaweed	56.4 ± 2.0	0.079 ± 0.003	0.052 ± 0.005	0.000134 ± 0.000020	0.084 ± 0.003 [1.73]	0.057 (0.049-0.066) [1.73]	0.000173 (0.000152-0.000198) [3.38]
Danish soil*	88.1 ± 3.4	0.14 ± 0.01	0.09 ± 0.02	0.05 ± 0.01	0.140 ± 0.008 [0.69]	0.098 ± 0.006 [0.69]	0.05 ± 0.01 [0.00]
Thule soil&	70.0 ± 9.5	1.45 ± 0.07	0.37 ± 0.07	1.14 ± 0.16	-		-
Danish seaweed#	77.8 ± 11.4	0.40 ± 0.02	0.01 ± 0.01	0.08± 0.01	-	-	-

†All values are the average of three replicates (± standard deviation). ‡Numbers in brackets are $|t|_{exp}$-values, at 95% confidence interval, the critical t-value (t_{crit}) is 4.30 for n=3. * 0.05±0.03 mBq of ^{237}Np was always spiked into each 10 g of Danish soil. & 1.02 ± 0.12 mBq of ^{237}Np was spiked into each 1 g of Thule soil. # 8.13 ± 0.10 mBq of ^{239}Pu and 1.67 ± 0.03 mBq of ^{237}Np were spiked into each 20 g of Danish seaweed.

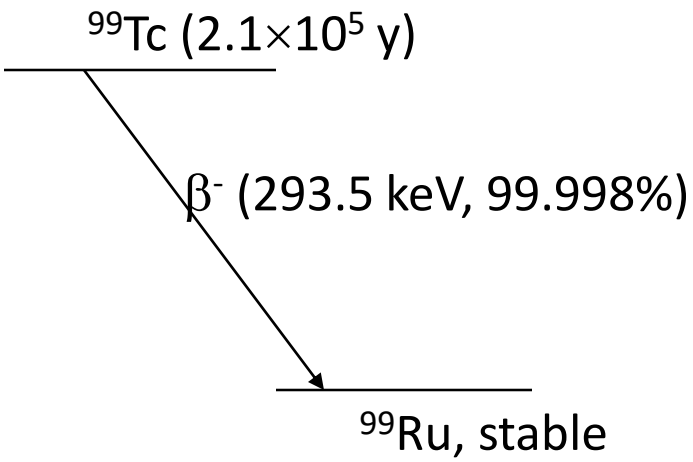
Qiao & Hou, Anal. Chem, 2009; and 2011.

Determination of ^{99}Tc in environmental samples

Shi, Hou, Roos & Wu
Anal Chem., 2012,
Anal. Chim Acta, 2012
Water Research 2012

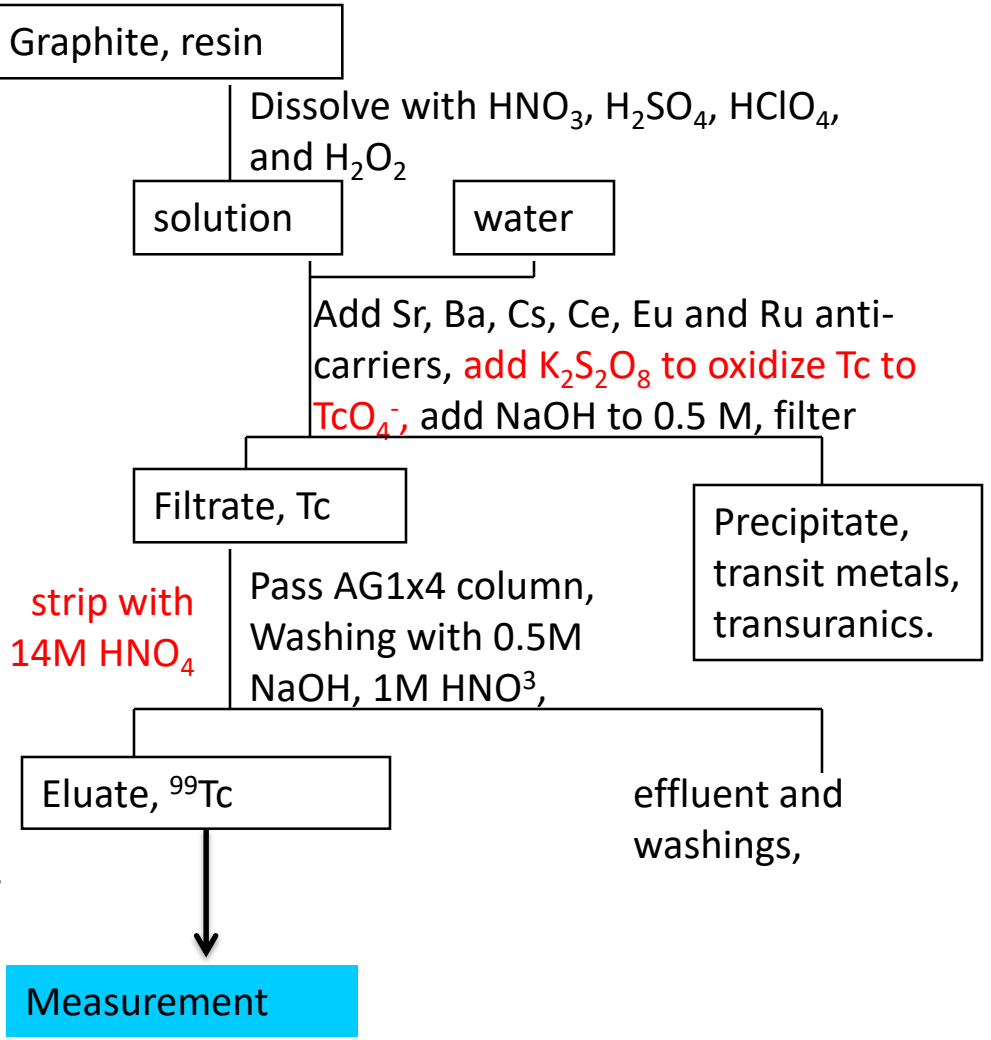
^{99}Tc

- Long half-life (2.1×10^5 y)
- High mobility (TcO_4^-)
- High fission yield (about 6 %)
- Tc is a volatile element, and easily to be loss at high temperature.
- A pure beta emitter
- Measured by GM counter, LSC, and ICP-MS.
- It has to be separated from other radionuclides before measurement by beta counting
- Completely Removal of Ru (^{99}Ru) and Mo ($^1H^{98}Mo$) for ICP-MS.

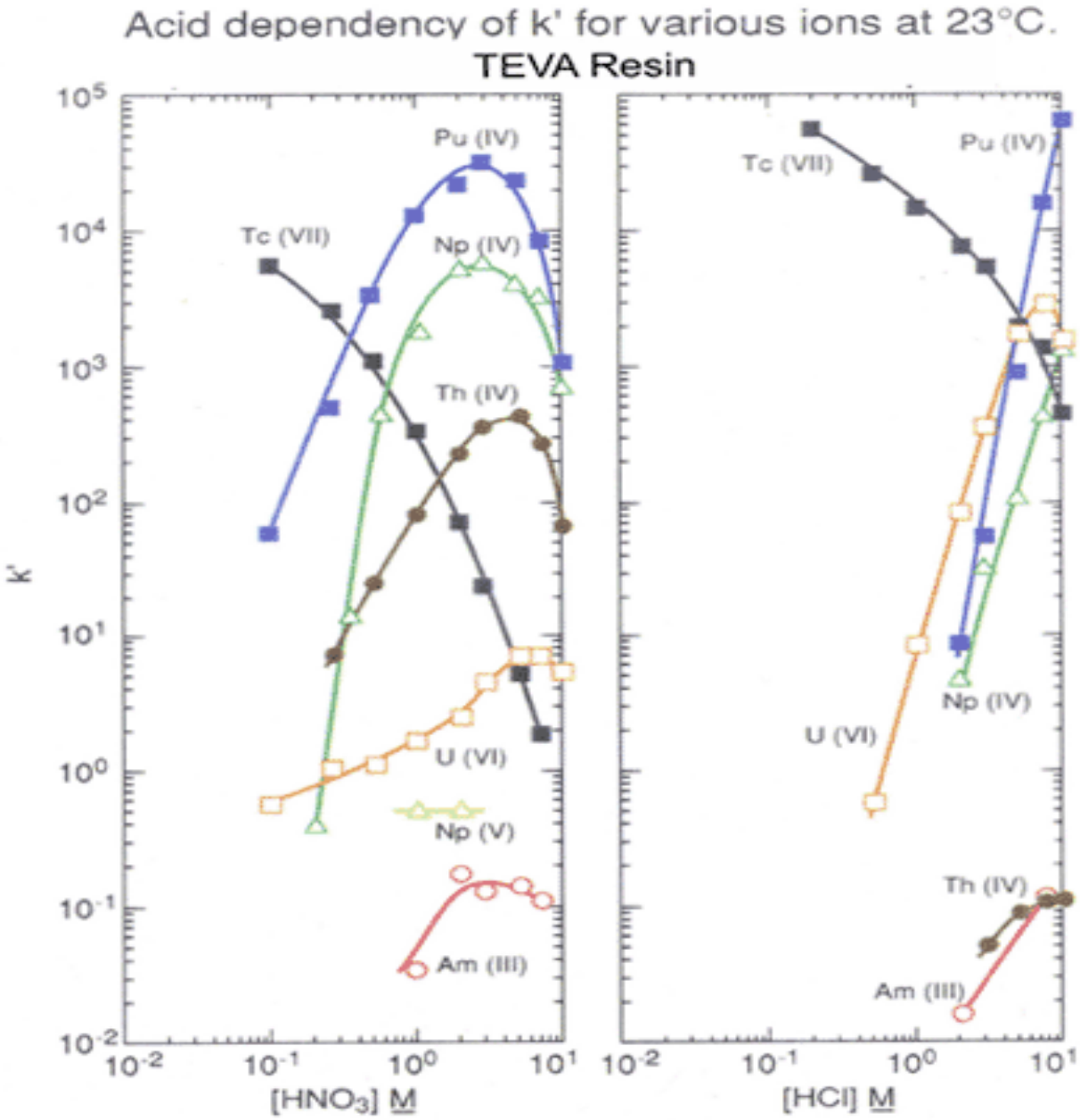


Chemical separation of technetium

- Adjust Tc to TcO_4^- by $K_2S_2O_8$
- Separate Tc from transition metals, transuranics, Po etc by hydroxides, because TcO_4^- can not be precipitate at high pH.
- TcO_4^- can be tightly absorbed by anion exchange column,washing with NaOH and HNO_3 can remove most of interferring nuclides.
- Not satisfactory for removal Ru, and Mo



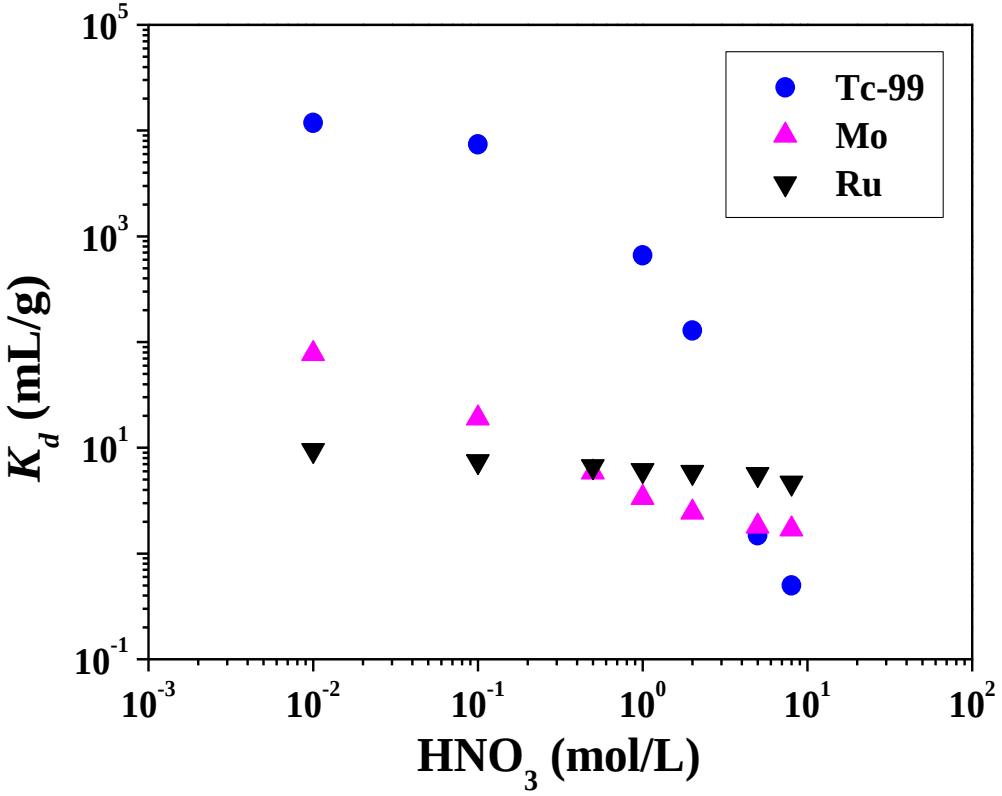
Figures 2 & 3



Horwitz, et al. (HP195)

Behaviors of TcO_4^-, Mo and Ru on TEVA resin

1) Batch experiment; 2) m (resin)/V(solution)=0.01g/mL; 3) Contacted time (3h)

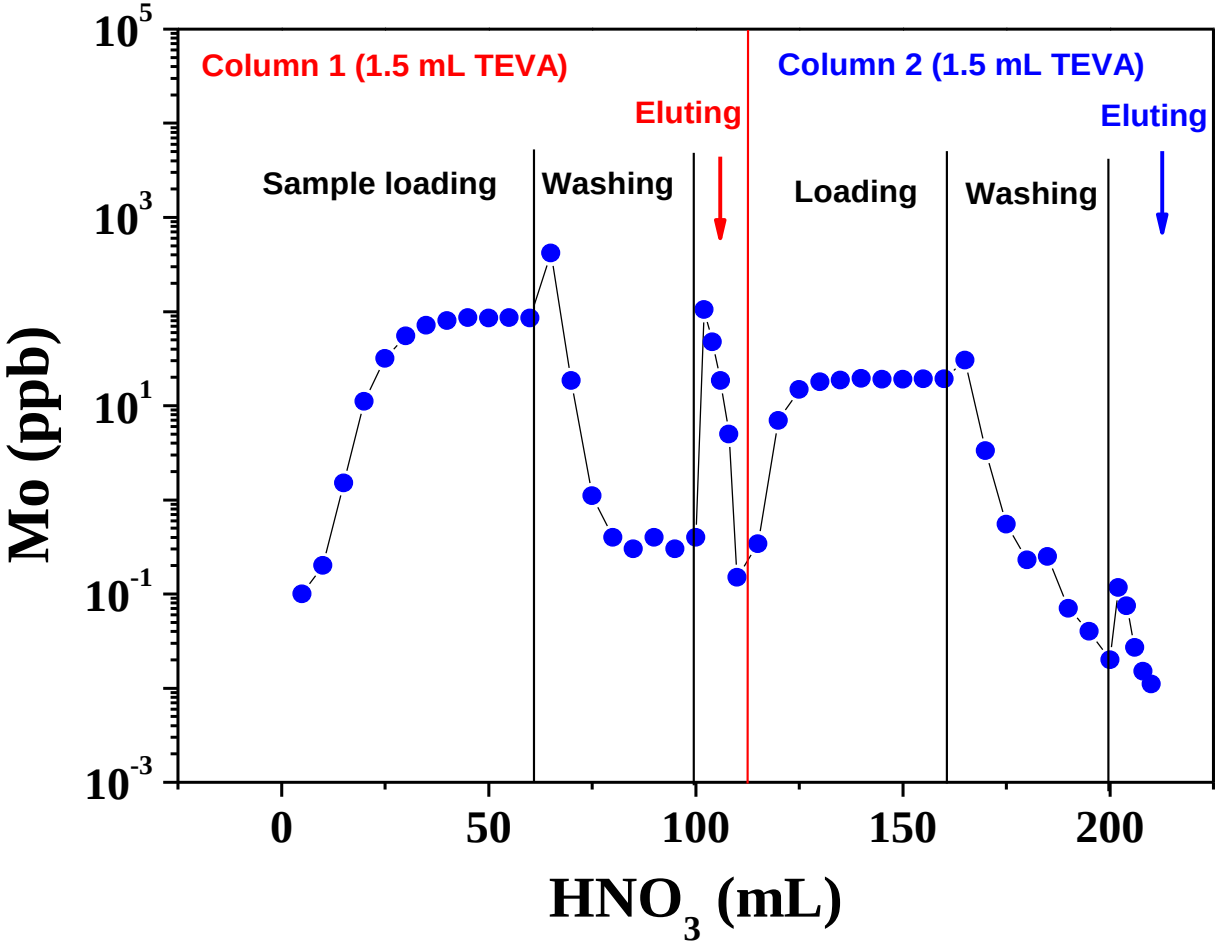


1) TEVA resins express a good sorption ability for TcO_4^-: (K_d > 1000, [HNO_3]< 1mol/L);

2) One small TEVA column is not enough to remove Mo;

3) Ru is insensitive with the change of [HNO_3] when it sorbed on the TEVA resin.

Removal of Mo from TEVA column



Conditions:

1) Loading (0.1M HNO_3 medium)

2) Wanshing (40 mL, 1M HNO_3)

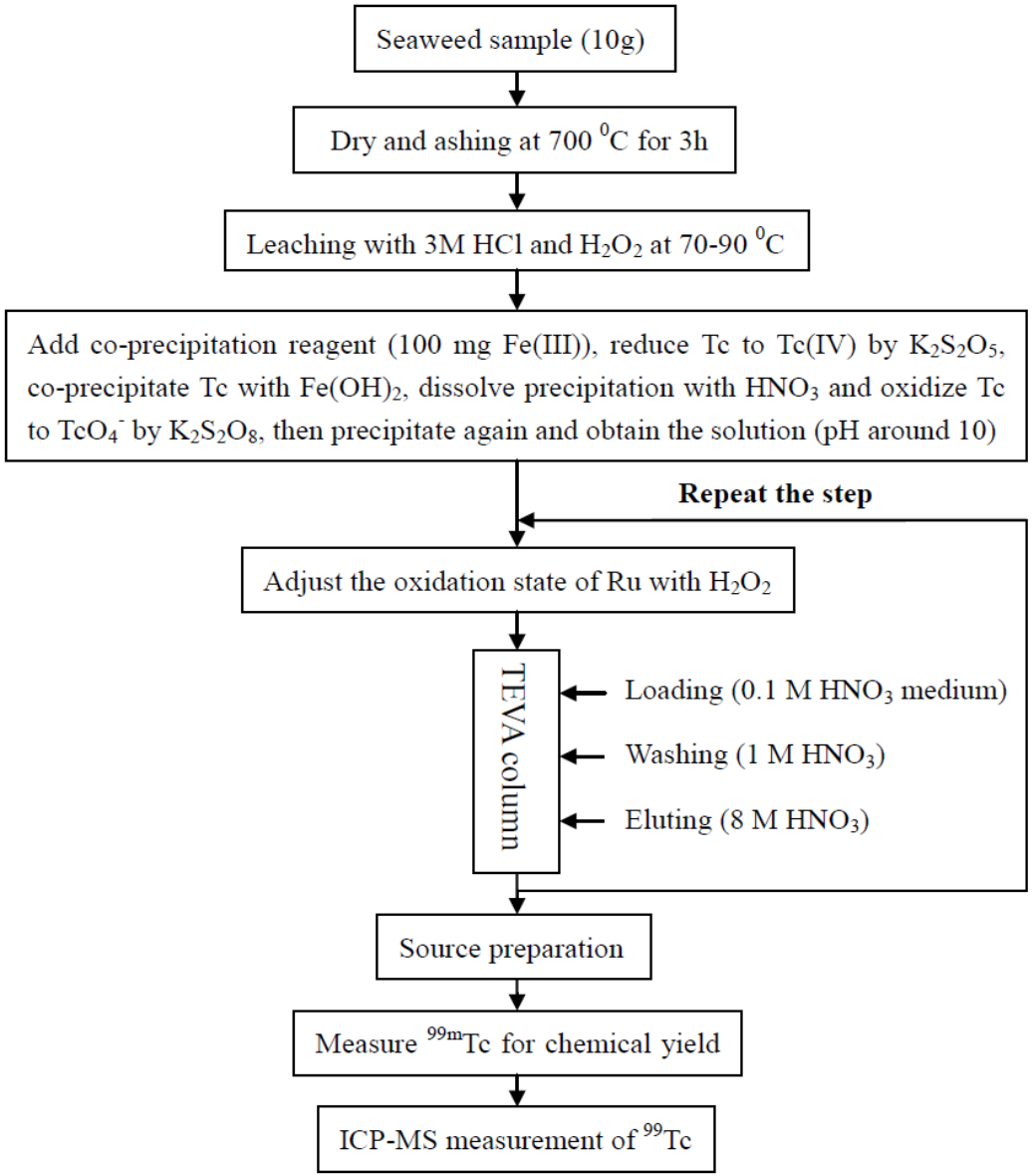
3) Eluting (10 mL, 8M HNO_3)

Effect of H_2O_2 for adsorption of Ru on TEAV column

	Volume (mL)	Pre-treatment with H_2O_2 1M NaOH	H_2O	No H_2O_2 0.1M HNO_3
		Ru (ppb)		
Sample loading	4	0.131	0.102	0.07
	8	0.197	0.150	0.091
	12	0.197	0.159	0.089
	16	0.196	0.154	0.092
	20	0.195	0.154	0.089
Washing	4	0.067	0.054	0.045
	8	< 0.001	0.004	0.003
	12	< 0.001	0.004	0.002
	16	< 0.001	0.005	0.002
	20	< 0.001	0.005	0.003
Removal (%)		**98**	**79**	**49**

* Washing with 0.1 M HNO_3

Separation procedure for determination of ^{99}Tc in soild samples



Sample pre-treatment

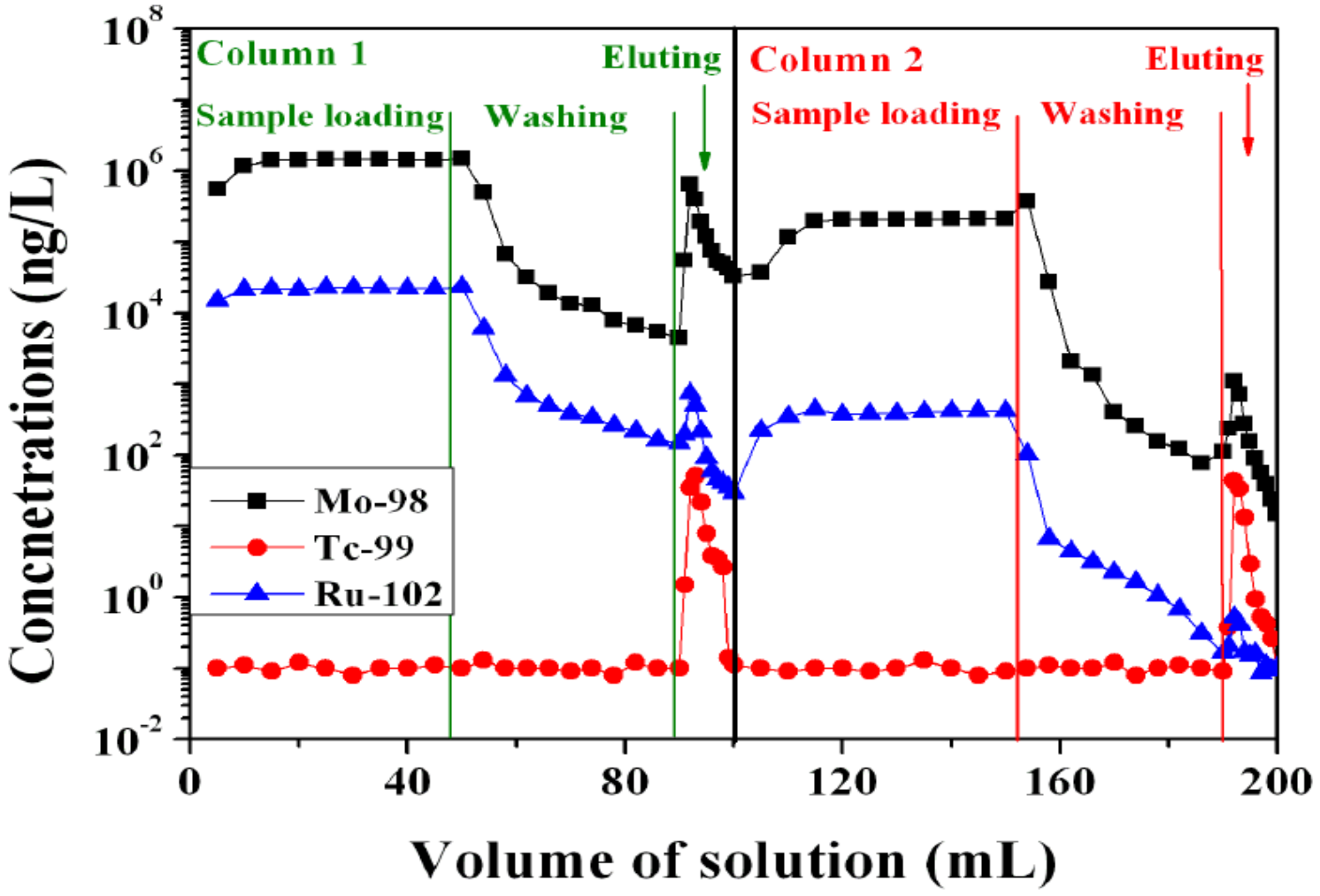
Tc concentration

Separation and purification

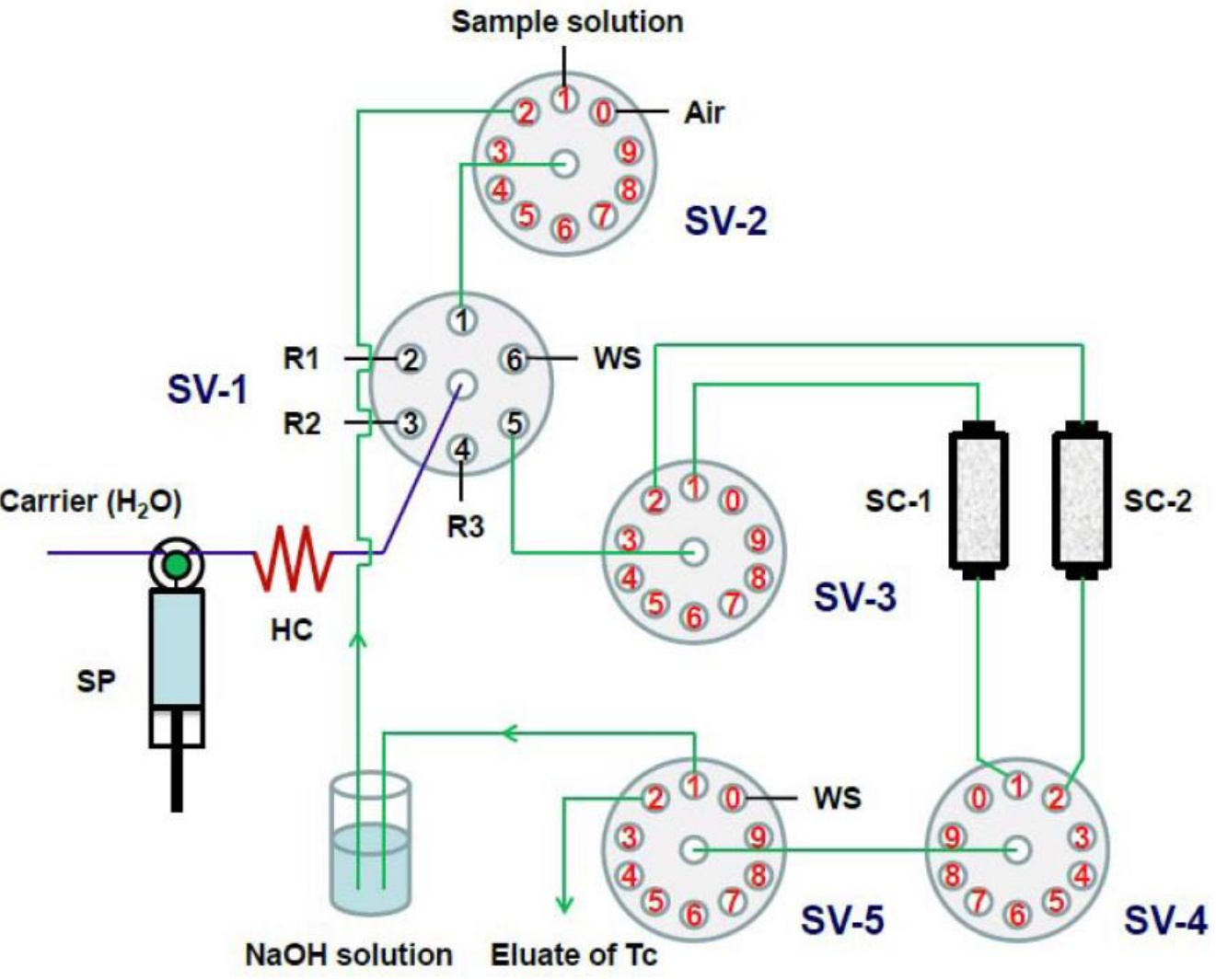
Source preparation

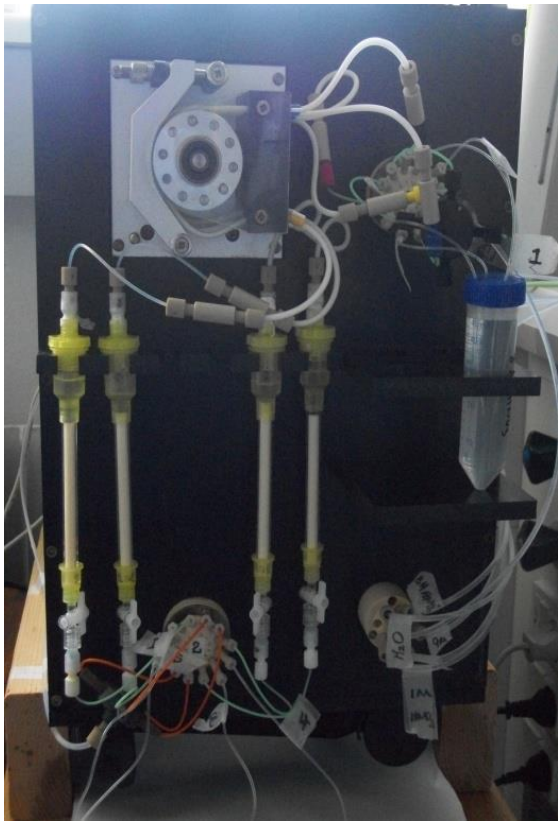
Measurement

Behaviors of Tc, Mo and Tc on two sequential TEVA columns



Separation of ^{99}Tc using an automated system with sequential injection approach





Validation of the method

Sample	Size (g)	Recovary (%)	Concentration of ^{99}Tc (mBq/g)	Reference value (mBq/g)
Danish seaweed	**10**	**85 ~ 95**	**73 ± 2**	**70 ~ 75***
IAEA 446 seaweed	**5**	**70 ~ 75**	**14 ± 2**	**16 ± 2***
NIST 4359 seaweed	**5**	**65 ~ 70**	**23 ± 2**	**17 ~ 48**

*** The value was obtained by a radiomatric method in Risø National Lab, Denmark**

seawater sample (L)	chemical yield (%)	^{99}Tc measured (mBq/L)	reference value of ^{99}Tc (mBq/L)	decontamination factors	
				Mo	Ru
50	68 ~ 75	0.270 ± 0.018	–	$(5.6 \pm 0.8) \times 10^5$	$(1.4 \pm 0.6) \times 10^6$
200	60 ~ 70	0.265 ± 0.021	0.267 ± 0.016 [b]	$(7.0 \pm 1.2) \times 10^5$	$(7.7 \pm 1.5) \times 10^6$

[a] Results are given as the average and 2 SD of three replicates. [b] Values were obtained using the method reported by Chen et al. (Chen et al., 2001).

Determination of ^{63}Ni, ^{59}Ni in decommissioning waste

Hou, et al. Anal. Chim. Acta, 2005

Production of ^{63}Ni and ^{59}Ni in nuclear reactor

- ^{63}Ni:
  - ^{62}Ni(n, γ)^{63}Ni (σ=14.5 b; η_{62Ni}=3.63%)
  - ^{63}Cu(n, p)^{63}Ni, (η_{63Cu}=69.17%)

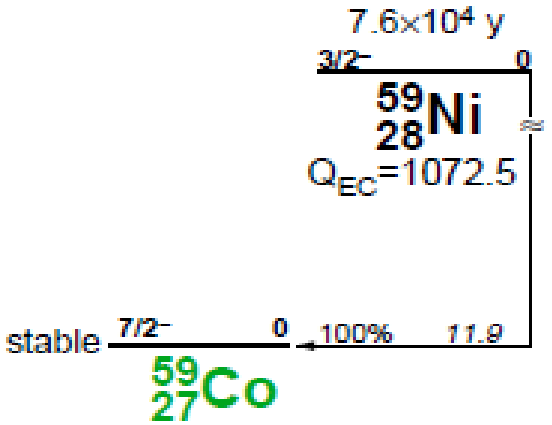
^{59}Ni:

- ^{58}Ni(n, γ)^{59}Ni (σ=4.6 b; η_{58Ni}=68.1%)

Atomic ratio: ^{59}Ni/^{63}Ni=6.5:1

Activity ratio: ^{59}Ni/^{63}Ni=1:133

Radioactive decay of ^{63}Ni 和 ^{59}Ni

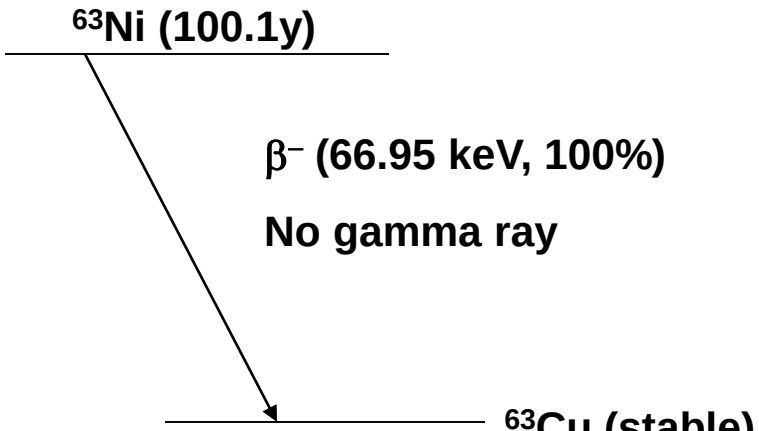


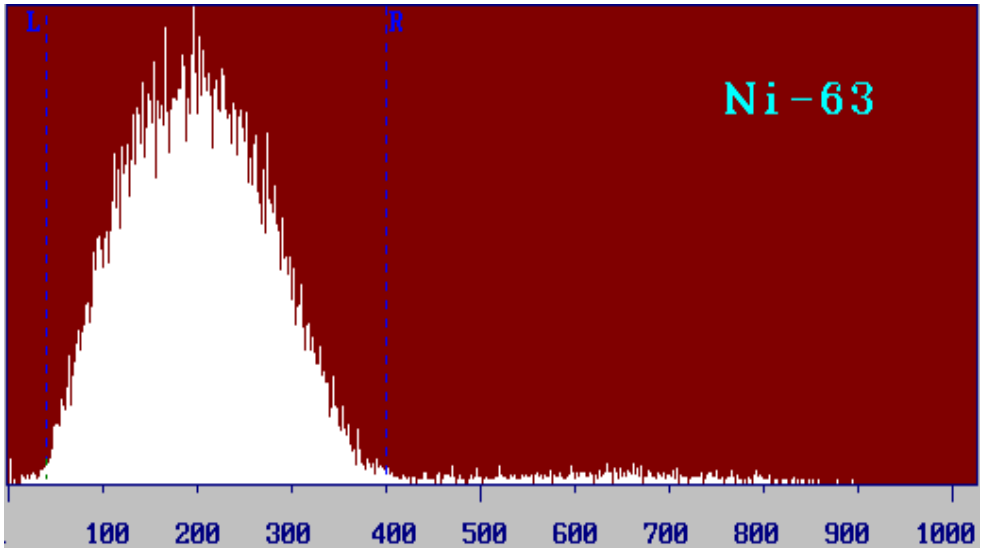
^{59}Ni X-Rays:

6.915 keV (10.4%)

6.930 keV (20.4%)

7.65 keV (3.70%)





Measurement methods for ^{63}Ni, ^{59}Ni

- Due to their low energy of beta particle and measurable electrons, LSC is the most suitable method for their measurement.
- Due to their pure beta and EC decay, they have to be

Mearement methods

^{63}Ni: gass flow counting(anti-coincidence, <10-50%)

Ion implanted silicon detector (1-6%)

LSC (60-80%)

^{59}Ni: X-Ray spectrometry (<1%)

Accelerator mass spectrometry

- Analytical procedure:
  - Decomposition of sample
  - Separation of Ni from matrix elements and all other radionuclides
  - Preparation of a suitable solution for LSC measurement of ^{63}Ni
  - Electroplate Ni on disk for X-ray spectrometry measurement, or prepared as Ni metal for AMS measurement of ^{59}Ni.

Interferences for measurement of ^{63}Ni, ^{59}Ni

Nuclide	Half-life	Decay	Nuclide	Half-life	Decay
^{60}Co	5.27 y	β^-,γ	^{3}H	12.33 y	β^-
^{58}Co	70.86 d	β^+, γ	^{14}C	5730 y	β^-
^{152}Eu	13.54 y	$\varepsilon, \beta^-,\gamma$	^{133}Ba	10.51 y	ε
^{154}Eu	8.59 y	β^-,γ	^{41}Ca	1.03E5 y	ε
^{51}Cr	27.7 d	ε,γ	^{36}Cl	3.01E5 y	β^-, ε
^{65}Zn	244.3 d	$\varepsilon, \beta^+, \gamma$	^{137}Cs	30.7 y	β^-
^{54}Mn	312.3 d	$\varepsilon, \beta^+, \gamma$	^{134}Cs	2.06 y	β^-,ε
^{151}Sm	90 y	β^-	^{90}Y	64 h	β^-
^{90}Sr	28.79 y	β^-			

Conventional metods for separation of Ni

- Precipitation as $Ni(OH)_2$, separation from Sr, Cs, ^{3}H, ^{14}C, Ba, Ca, Cl.
- Precipition by ammonium, separate Ni from Fe, Mn, Eu, Pb, Al, Cr.
  - Low recovery of Ni in this method (Ni can be also partly precipitate in ammonium solution)
  - Cannot separate Cu, Co, etc.
- Ion exchange to separate Ni from Co, Cu, Zn, Fe, and transuranics.
- Precipitation or extraction of complex of Ni with dimethylglyoxime (DMG).
- Co and Cu can also form a complex with DMG and extracted
- Evaporation of $Ni(CO)_6$

Separation of Ni by hydroxides precipitation

Element	Precipitation, %		Solution, %
	NaOH (pH9)	NH_4OH	NH_4OH
Ni^{2+}	>99.8	>20	< 80
Co^{2+}	>99.5	<20	< 80
Ba^{2+}	<30.5	<30.0	>70
Eu^{3+}	>99.8	>99.8	<0.2
Cs^{+}	<0.2	<0.2	>99.8
Sr^{2+}	<37.5	<35.0	>60

- Most of matrix in concrete and environmental samples, such as C, S, Ca, Si, Na will be separated.
- The recovery of Ni is not satisfied using ammonium to separate Ni from other matals by hydroxides precipitation
- Other metals such as Mn, Cr, V, Al, Pb, and transuranics will also be precipitated by NaOH, and cannot be separated from Ni.

Behaviors of Ni and other metals on anion exchange column

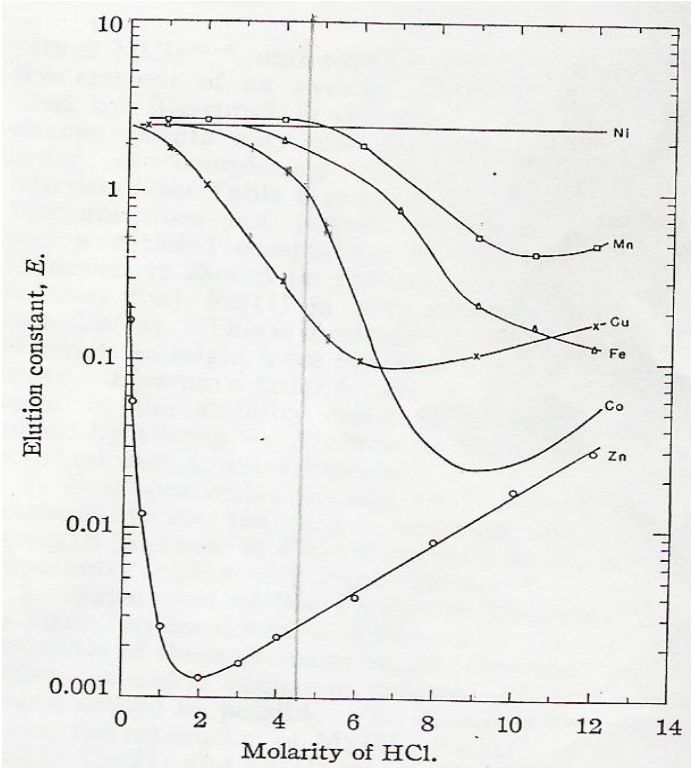


Fig. 1.—Elution constants of some divalent transition elements in hydrochloric acid (data for Zn(II) from unpublished results of F. Nelson).

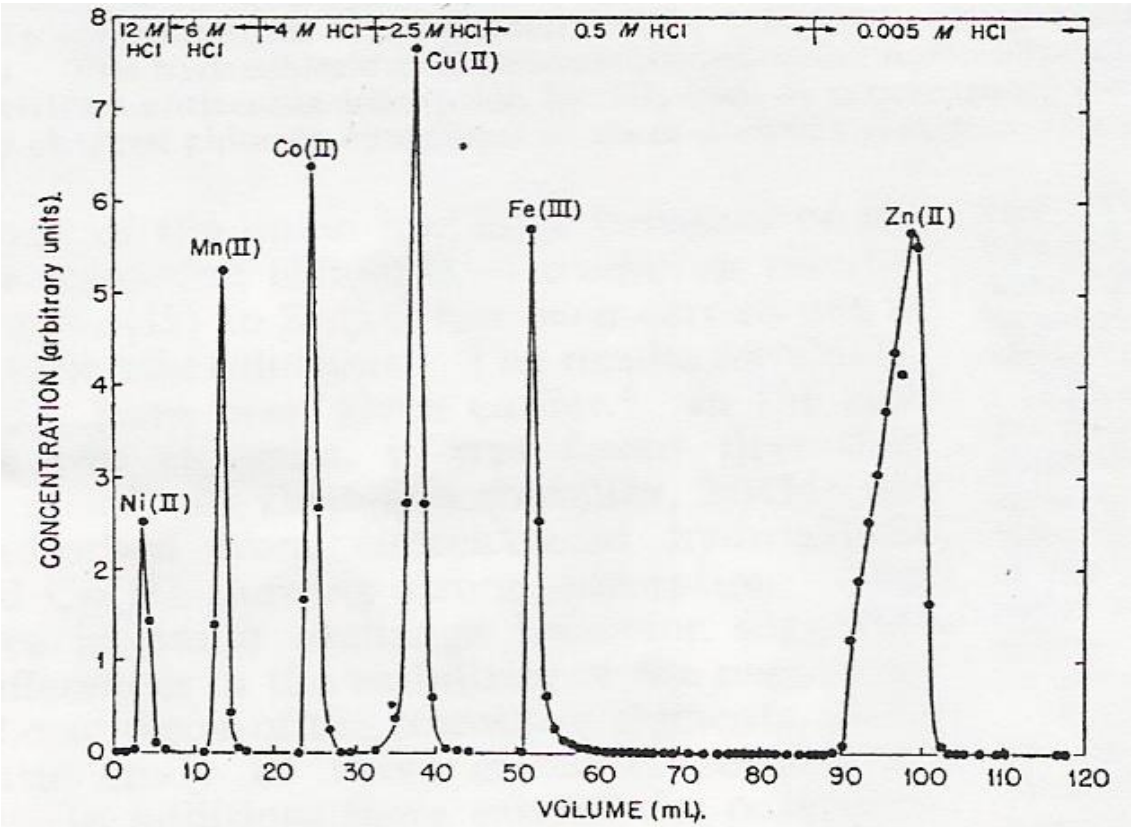


Fig. 2.—Separation of transition elements Mn to Zn (Dowex-1 column; 26 cm. × 0.29 cm.; flowrate = 0.5 cm./min.).

Many metals can form a anion complex with Cl^- in HCl solution (MCl_x^-), so can adsorbed on anion exchange column

Separation of Ni, Co, Eu, Ba by anion exchange chromatography

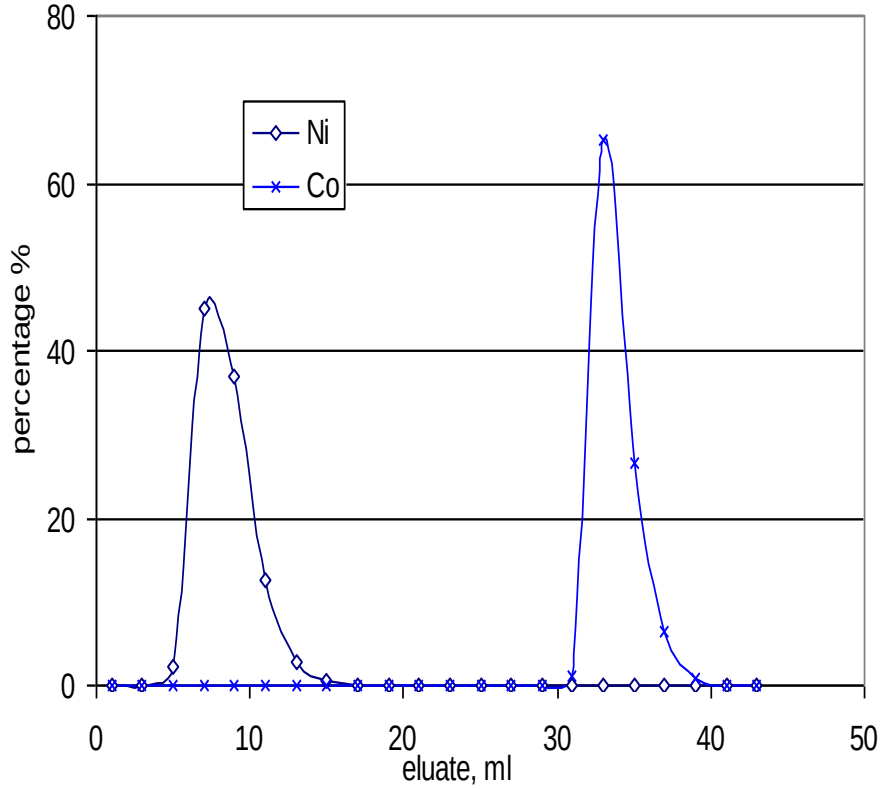
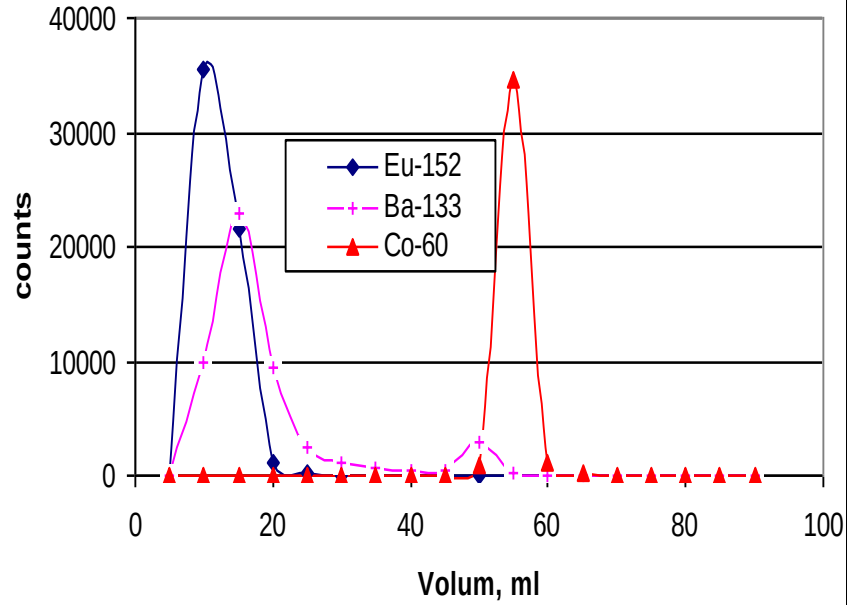


Fig. Separation of Ni from Co by ion exchange, eluted by 9 N HCl (3-20ml) for Ni and 2 N HCl (21-44 ml) for Co. Bio-Rad AG1x4. 1x15 cm column



Separation of Eu, Ba, Co by anion exchange chromatography, Bio-Rad AG1x4, 1x15 cm, 0-40ml:9M HCl, 40-70ml:4M HCl, 70-90ml, 0.05M HCl

Separation of Ni by anion exchange chromatography

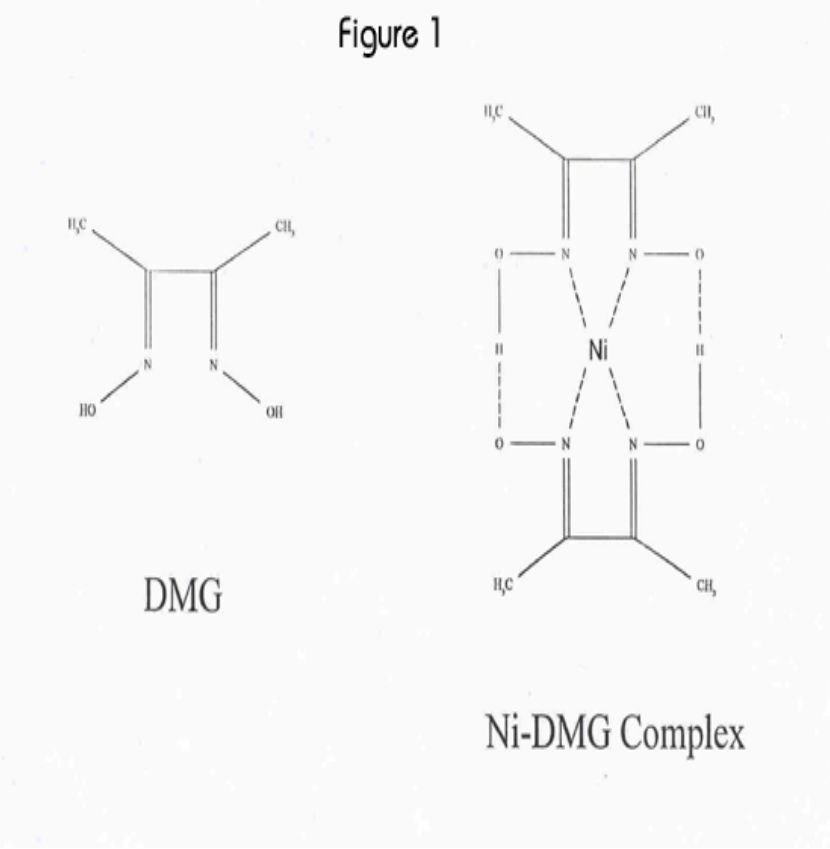
Element	Content, %
Fe^{3+}	<0.001
Ni^{2+}	>99.5
Co^{2+}	<0.01
Ba^{2+}	<7.5
Eu^{3+}	>99.8
Cs^{+}	>99.5
Sr^{2+}	>99.5

- Ni can be completely separated from Fe, Co, Cu, Zn, U, Pu, etc.

- Ni cannot be efficiently separated from Cr, Eu, Sm, Mn, V, Sc, Ti, Zr, Ba, Th, Am. Of them, the radioisotopes of Eu, Sm, Ba, Zr, Mn, Cr and matrix elements of Cr, Mn V in metal and alloy seriously interfer the determination of Ni-63.

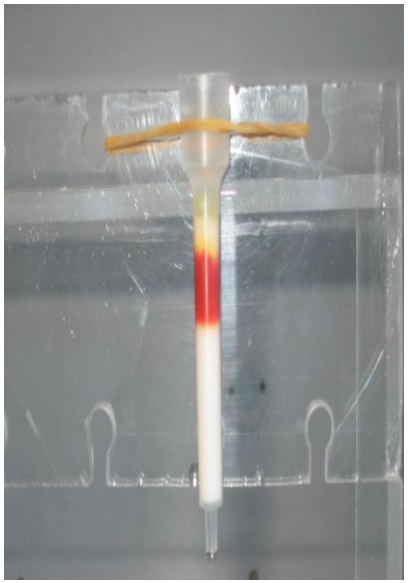
Thus: a further purification for both Ni and Fe is needed.

Application of Ni-DMG complex for the separation of Ni



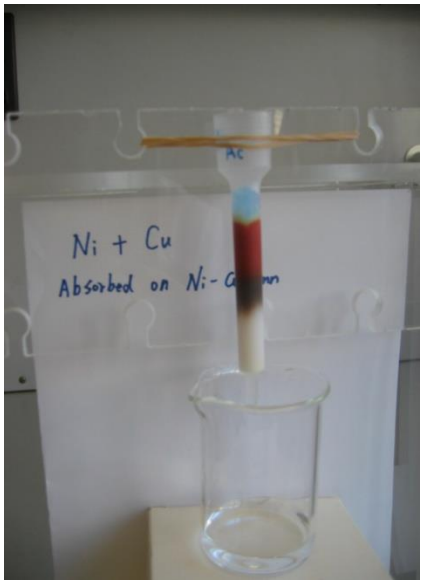
- Ni can form a stable specific complex with dimethylglyoxime. By Ni-DMG precipitation or organic solvent extraction of Ni-DMG complex at low concentration, Ni can be separated from many other elements.
- While, some other metals, such as Co, Cu can also form a complex with DMG and interferring the separation of Ni.

Behaviors of Ni、Co、Cu、Fe on Ni resin column



2mg Ni^{2+}

2mg Ni^{2+} +
2mg Co^{2+}



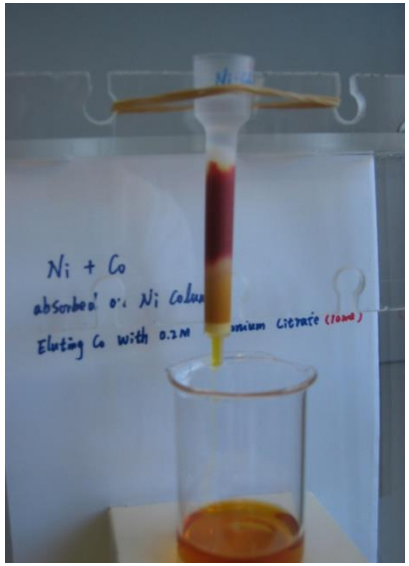
2mg Ni^{2+} +
2mg Cu^{2+}

2mg Ni^{2+} +
8mg Fe^{3+}

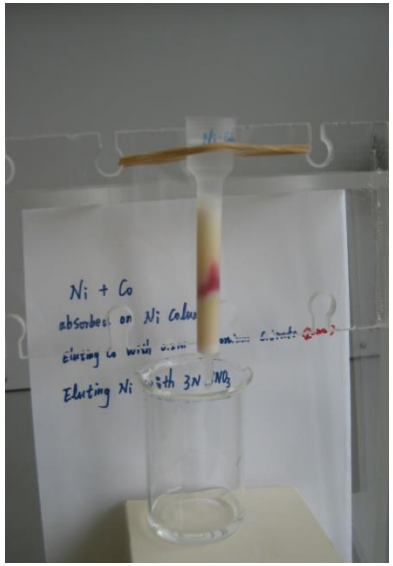
Separation and purification of Ni with Ni resin

The Nickel Resin contains the DMG inside the pores of a polymethacrylate resin. The nickel-DMG precipitate occurs on the resin , where it is held and readily separated from other elements in the supernatant.

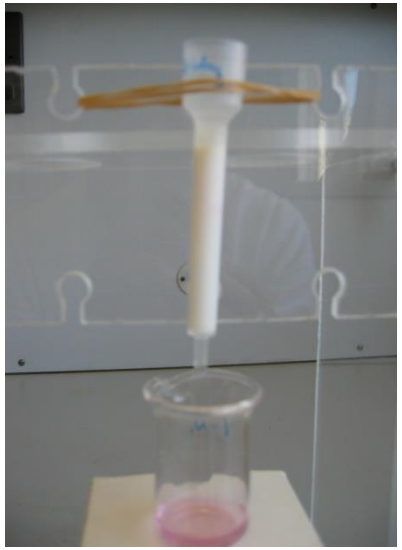
1. Loading of solution



2. Washing with 0.2 M ammonium citrate to remove other elements



3. Eluting Ni using HNO3



4. Evaporte eluted Ni-DMG solution to 0.1-0.2 ml for LSC

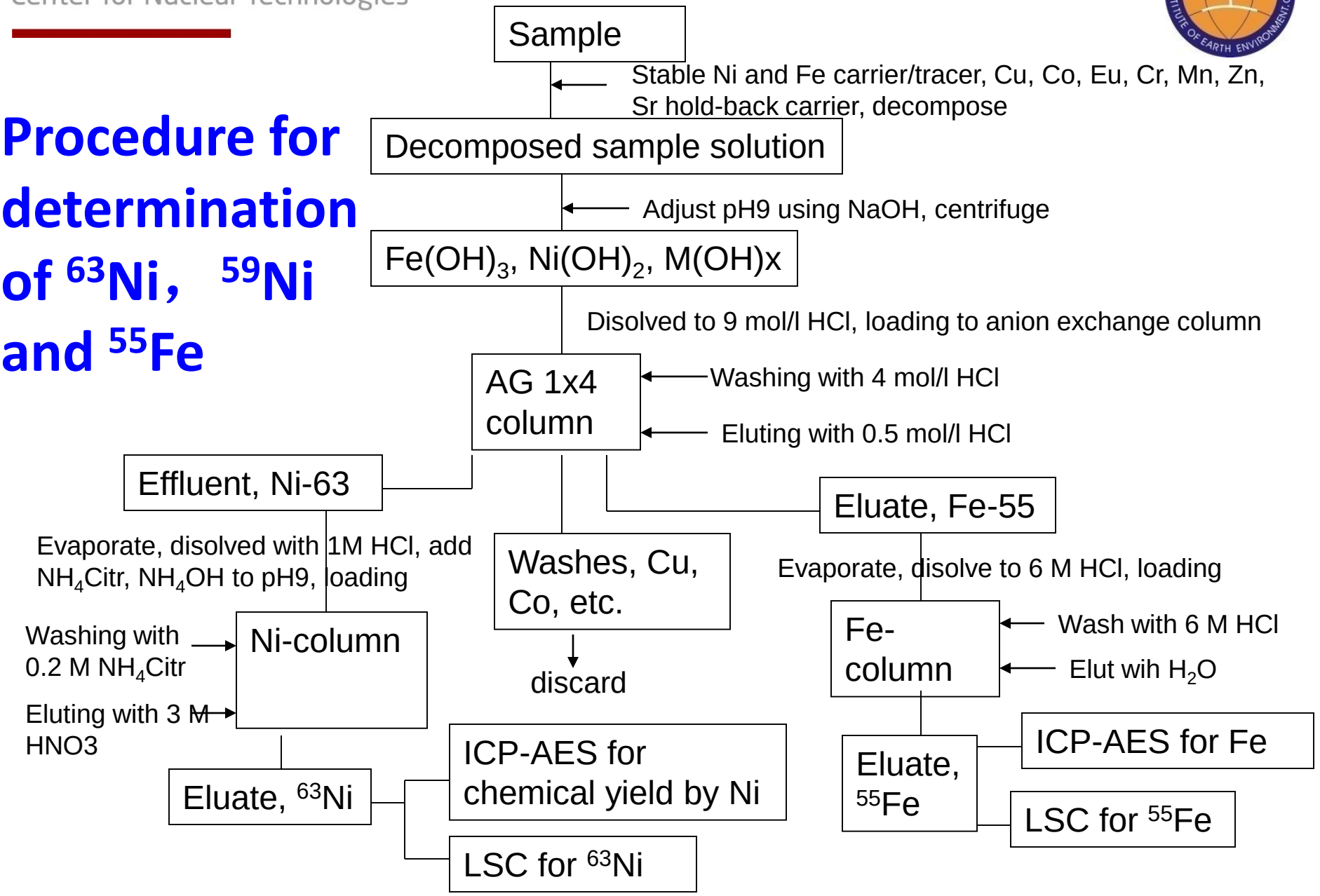
Performance of Ni resin in the separation of Ni

Element	Recovery or decontamination factor
Ni^{2+}	> 98.5%
Fe^{3+}	10^4
Co^{2+}	10^3
Ba^{2+}	10^4
Eu^{3+}	10^4
Cs^{+}	10^4
Sr^{2+}	10^4

Ni specific extraction chromatography has a higher decontamination to most of elements, such as Fe, Co, Cu, Cr. Mn, Ba, Eu, transuranics, etc.

- A higher recovery of Ni can be obtained in the procedure.

Procedure for determination of ^{63}Ni, ^{59}Ni and ^{55}Fe

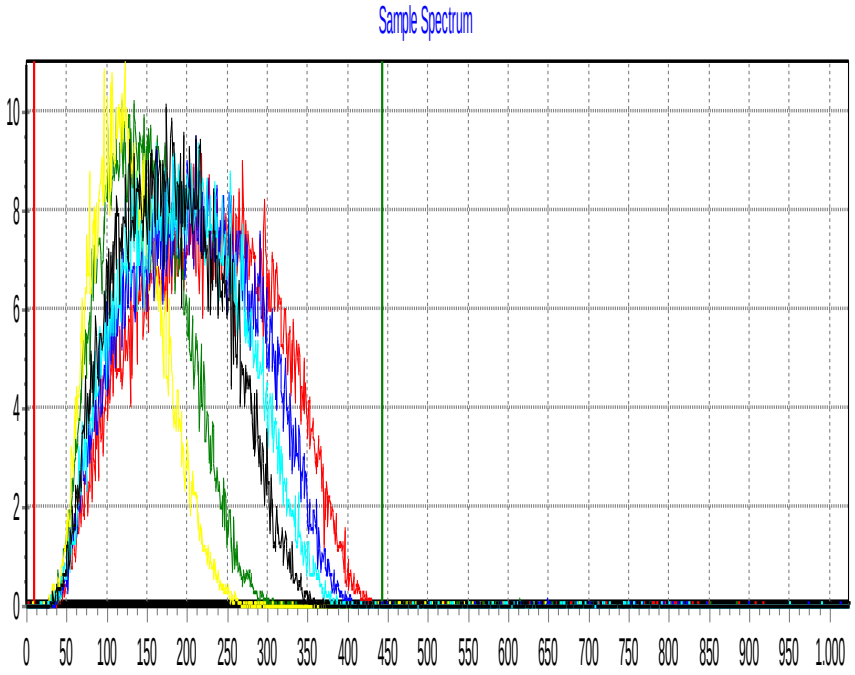


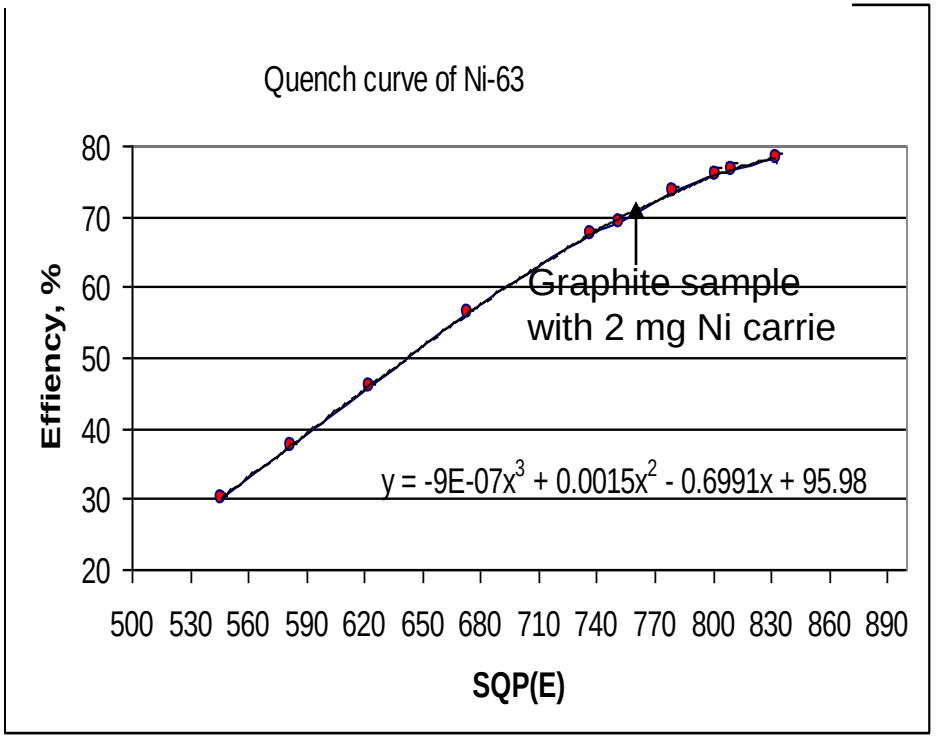
Performance on the separation of Fe and Ni

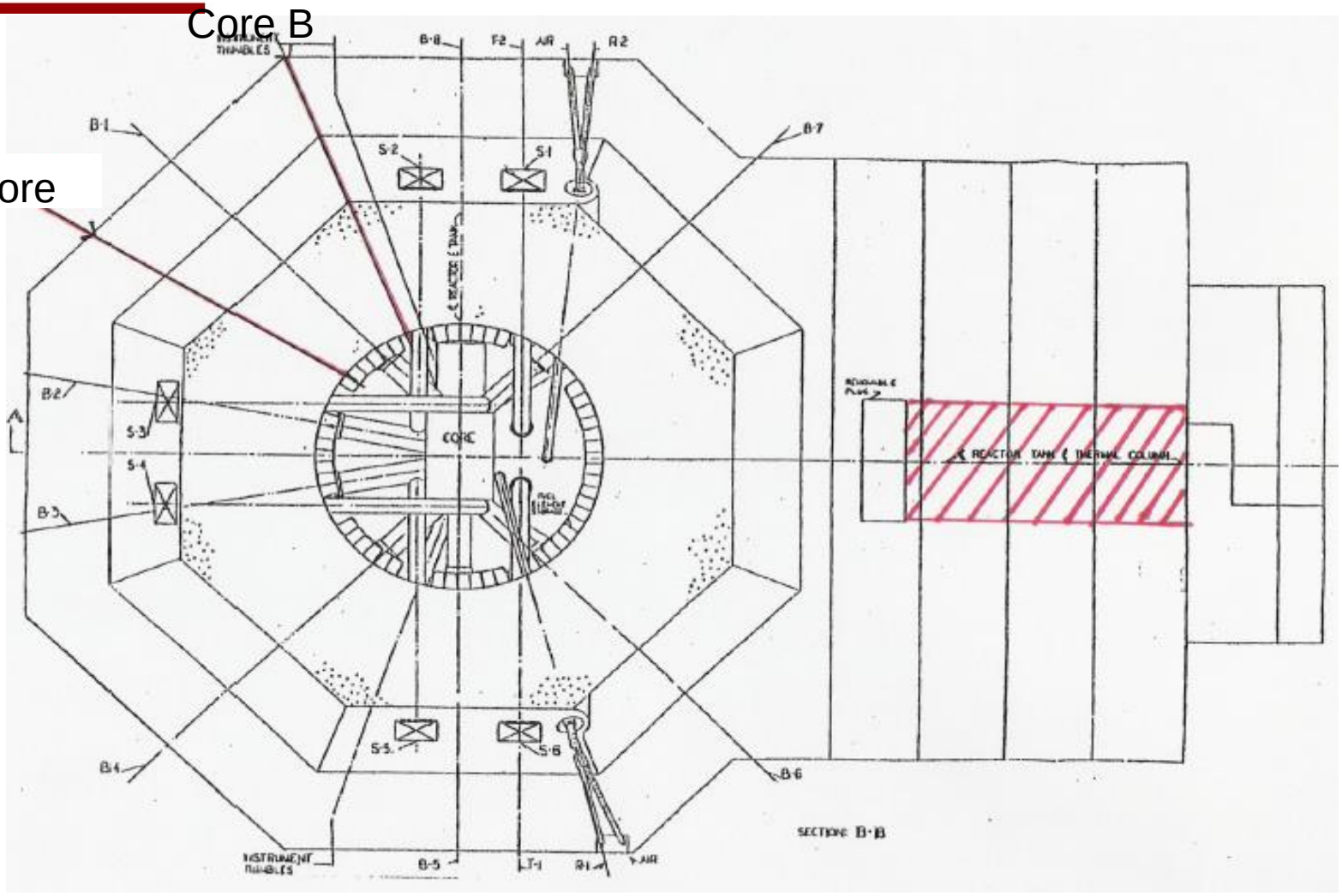
Interference	Recovery/decontamination factor		Interference	Recovery/decontamination factor	
	Fe fraction	Ni fraction		Fe fraction	Ni fraction
^{55}Fe	85-95%	>10^5	^{133}Ba	>10^6	>10^5
^{63}Ni	>10^5	80-95%	^{134,137}Cs	>10^6	>10^6
^{58,60}Co	>10^5	>10^5	^{89,90}Sr	>10^6	>10^6
^{152,154}Eu	>10^6	>10^5	^{41,45}Ca	>10^6	>10^6
^{151}Sm	>10^6	>10^5	^{36}Cl	>10^6	>10^6
^{54}Mn	>10^5	>10^6	^{3}H	>10^6	>10^6
^{51}Cr	>10^6	>10^5	^{14}C	>10^6	>10^6

For all interferring radionuclides, the decontamination factors higher than 10^5.

LSC spectra of ^{63}Ni in samples of decommissioning waste

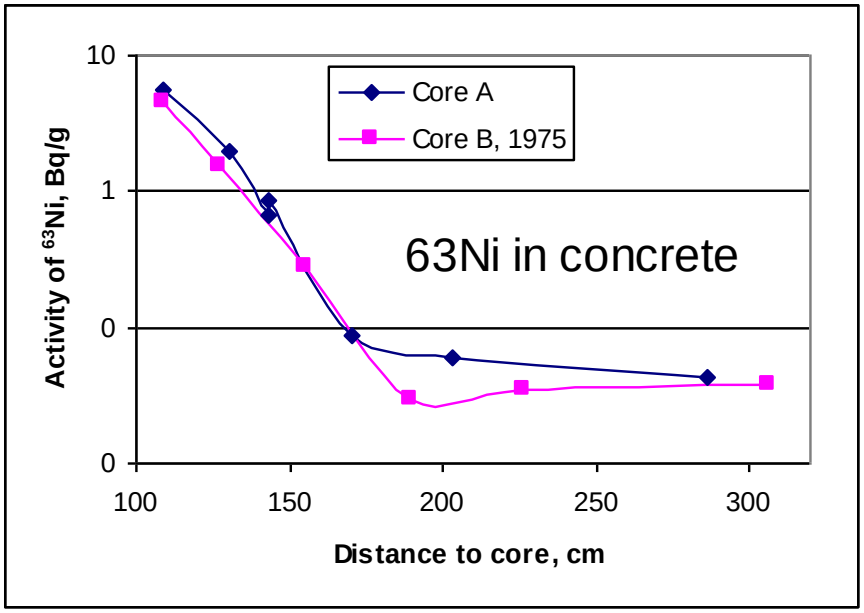






Sampling of concrete and graphite from danish reactor, DR-2

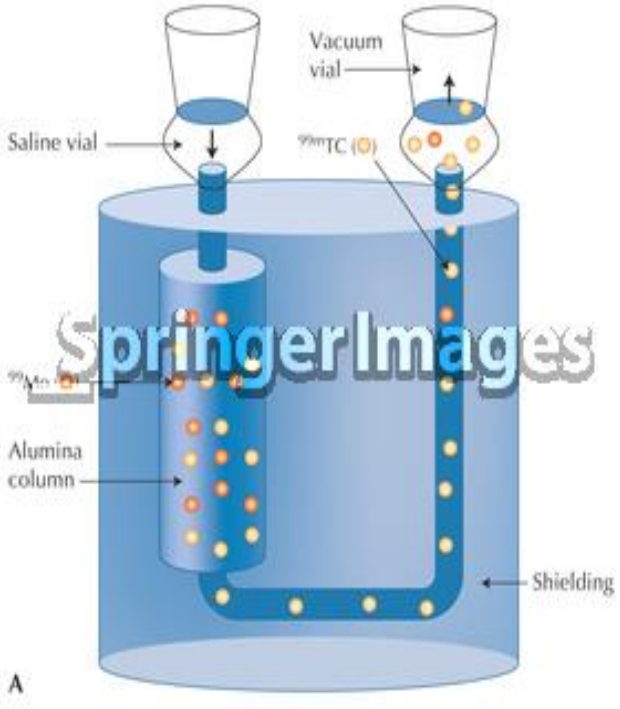
Analytical results of ^{63}Ni and ^{55}Fe in concrete core and graphite from Danish research reactor DR-2



^{55}Fe and ^{63}Ni in graphite of DR-2

Sample No	^{55}Fe		^{63}Ni	
	Recovery,%	Bq/g	Recovery,%	Bq/g
DR-3-T	92.2	545000	94.63	5552
Iy7.5	90.4	0.53	93.89	92.5
Iy5.5	90.6	1.05	93.74	22.3
Yi7.5	92.5	1.92	93.35	7.71
Yi5.5	91.3	9.21	91.56	43.1

Quality control of ^{99}Mo-^{99m}Tc generator



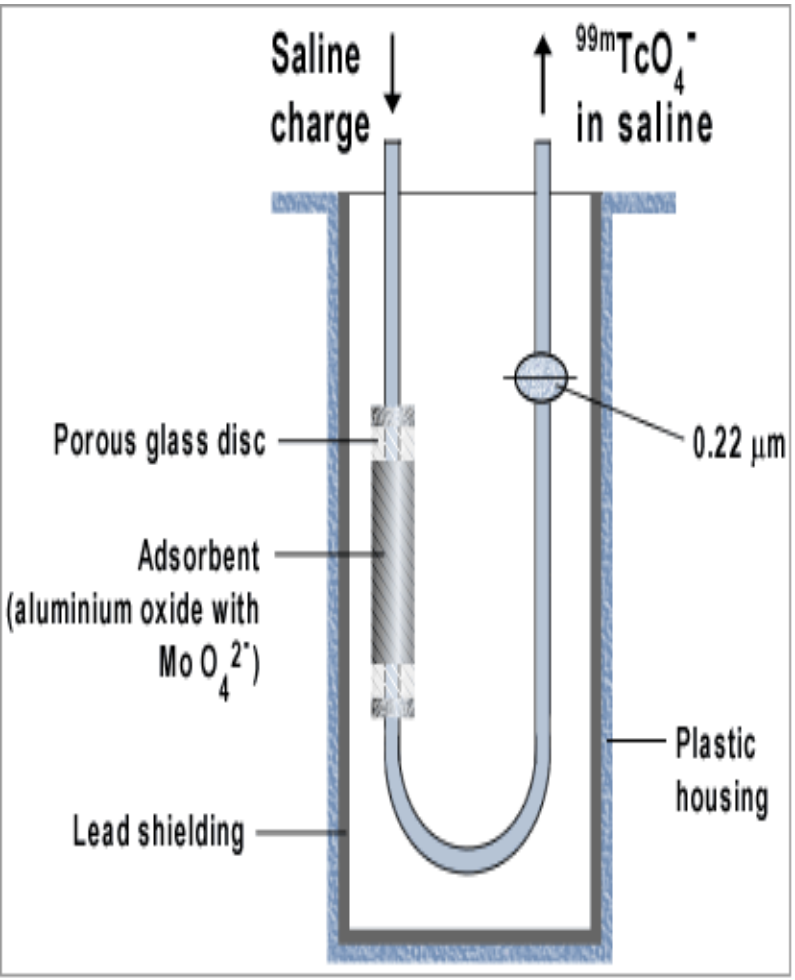
Hou, DTU-Nutech-R, 2012

^{99}Mo production

- **Fission of ^{235}U: ^{235}U(n, f)^{99}Mo**
  - ➢ 6.2% fission yield,
  - ➢ high specific activity (no carrier)
  - ✓ Need separation from uranium and other fission products.
  - ✓ Main Impurities: fission products + activation products including actinides.

- **Neutron activation of ^{98}Mo: ^{98}Mo(n, γ)^{99}Mo (Risø-Generator for analysis of environmental ^{99}Tc)**
  - ➢ Easy production, directly irradiat Mo oxides, and then dissolve irradiated Mo oxide and load it to gernerator column.
  - ✓ Main impurities: activation products

Items for quality control of ^{99}Mo-^{99m}Tc generator



❖ **Chemical Purity:**

- All other elements besides technetium, the most concern is the metals which effect the application of 99mTcO4-, for example Al.

❖ **Radiochemcial Purity**:

➢Definition: For a material, the fraction of the stated isotope present in the stated chemcial form.

➢ The percentage of $^{99m}TcO_4$- in all ^{99m}Tc, mainly $^{99m}Tc^{4+}$/$^{99m}TcO_4$-

❖ **Radionuclidic Purity:**

- Definition: The proportion of the total activity that is present as a specific radionuclide.
- Other radionculides in the eluate of ^{99m}Tc

Radionuclidic purity of ^{99m}Tc eluate from ^{99}Mo-^{99m}Tc generator

Possible radionuclidic impurities:

– fission products for fission ^{99}Mo generator)

Isotope	$t^{1/2}$	γ Energies (keV)	β_{max} Energy (MeV)
^{99}Mo	65.9 h	140.5 (4.5%)	1.350
		739.5 (12.2%)	
^{99}Tc	211,100 yr		0.294
			0.437
^{131}I	8.02 days	364.4 (81.7%)	0.971
^{132}I	2.95 days	522.6 (16.0%)	3.577
^{106}Ru	373.59 days		0.039
^{90}Sr	28.74 yr		0.546
^{90}Y	64.1 h		2.282
^{89}Sr	50.53 days		1.495
^{103}Ru	39.26 days	497.1 (91%)	0.763

Radionuclidic purity of ^{99m}Tc eluate from ^{99}Mo-^{99m}Tc generator

Possible impurities in activation produced ^{99}Mo:

– productions products including actinides

Nuclides	Half-life	Decay model	Energy	Gamma Energy
^{60}Co	5.27 y	beta	318 keV	1173 keV, 1332 keV
^{86}Rb	18.6 d	beta	1774 keV	1076.6 keV
^{124}Sb	60.2 d	beta	1301 keV	602 keV, 1691keV
^{134}Cs	2.06 y	beta	658 keV	604.7 keV, 795.8 keV
^{235}U	703 Ma	alpha	4397 keV	185.7 keV
^{238}U	4468 Ma	alpha	4198 keV	
^{239}Np	2.35 d	beta	436 keV	106.1 keV
^{239}Pu	24110 y	alpha	5156 keV	

Limitation of radionuclic impurities required in 99mTc eluate from a ^{99}Mo-^{99m}Tc generator in European pharmacopoeia 7.0

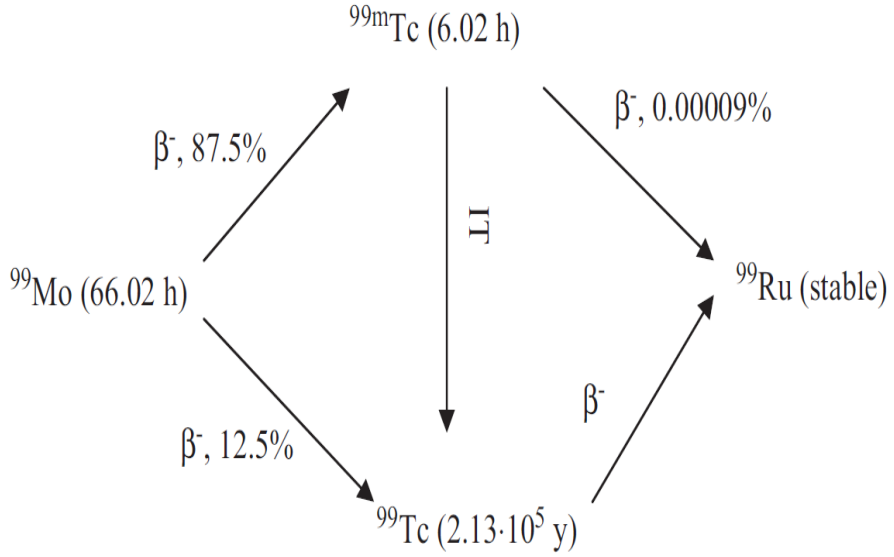
The radioactivity due to radionuclides other than technetium-99m,

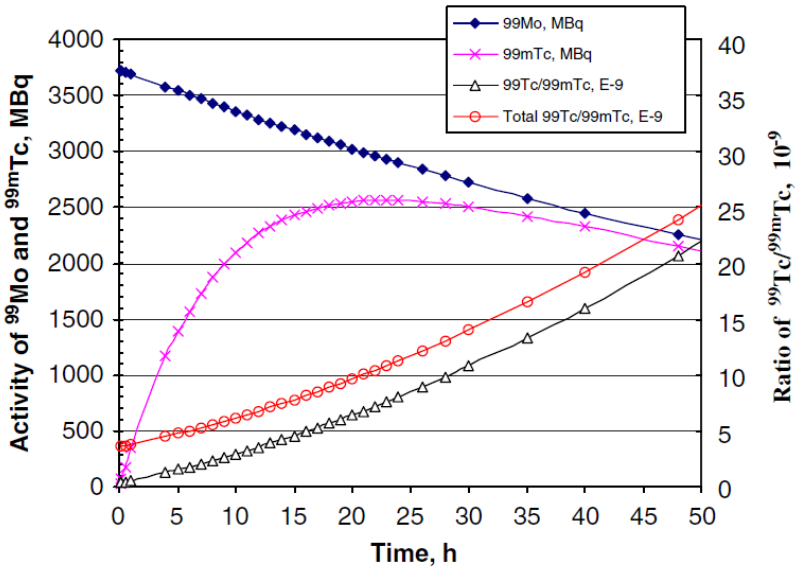
- molybdenum-99: 0.1 %
- iodine-131: 5×10^{-3} %
- ruthenium-103: 5×10^{-3} %
- strontium-89: 6×10^{-5} %
- strontium-90: 6×10^{-6} %
- alpha-emitting impurities: 1×10^{-7} %
- other gamma-emitting impurities: 0.01 %

Analytical methods for determination of impurities in ^{99m}Tc eluate

- **Gamma spectrometry:**
  - ^{99}Mo, ^{131}I, ^{103}Ru and other possible gamma emitters (removal of ^{99m}Tc by 15 days decay)
- **Gross alpha by LSC**
  - ^{99}Mo, ^{99}Tc needs to be removed
- **Measurement of ^{89}Sr和^{90}Sr using LSC by Cerenkov counting**
  - Separation of Sr from all other possible impurities

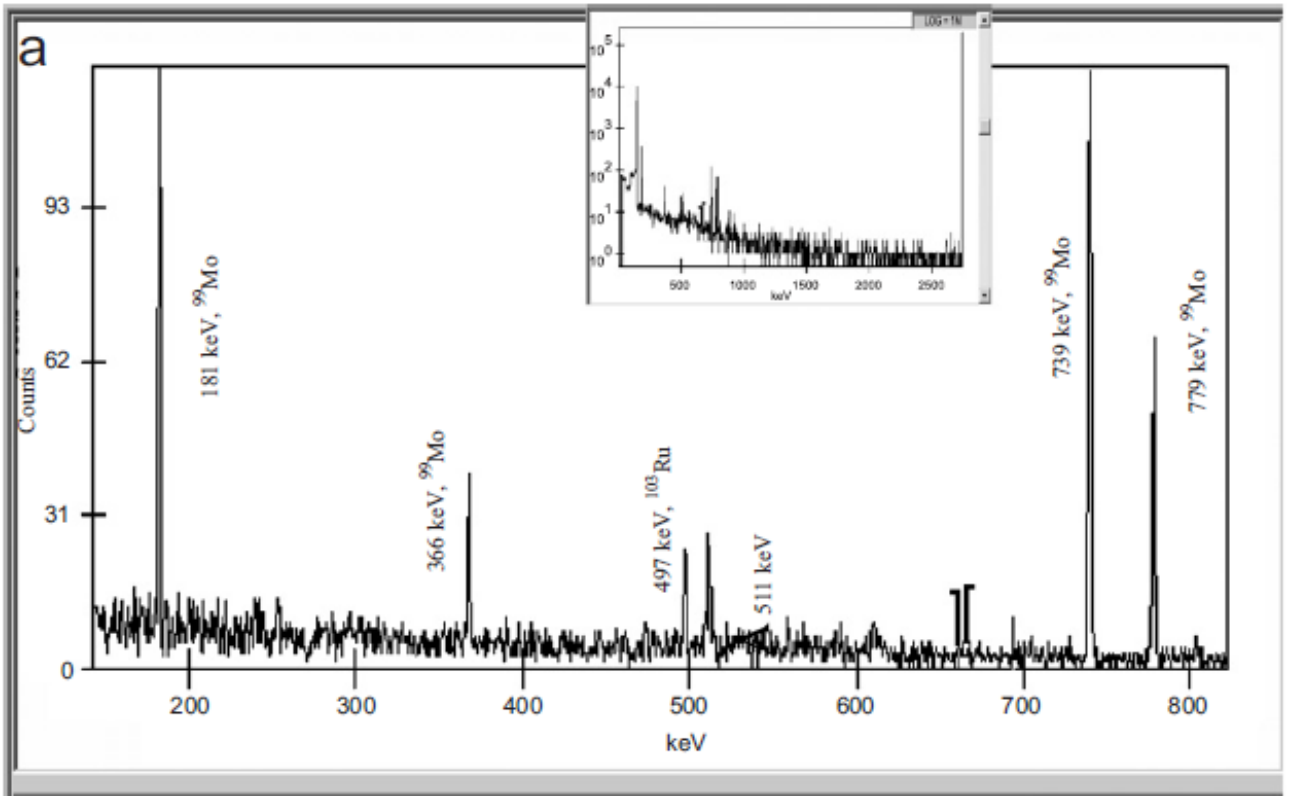
Generation of ^{99m}Tc from the Decay of ^{99}Mo





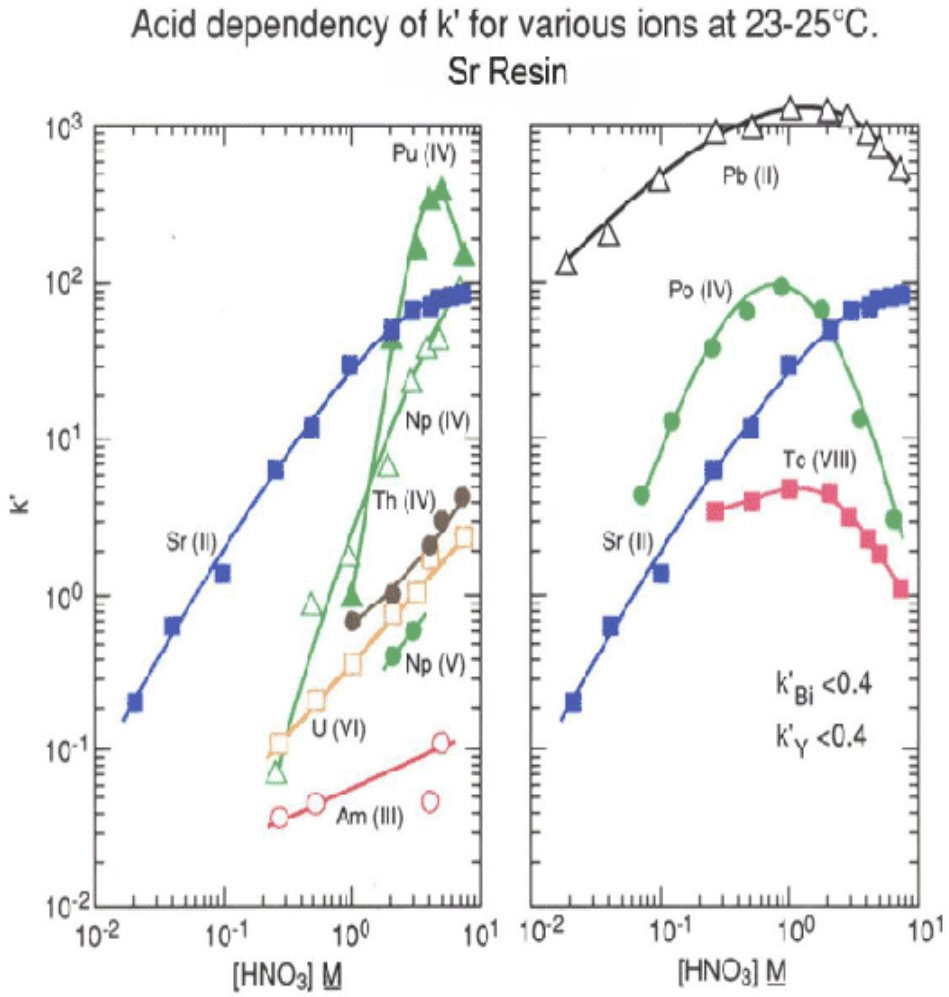
- For 20 h waiting time, the theory activity ratio of ^{99}Tc/^{99m}Tc = 10^{-8} in ^{99m}Tc eluate.
- For a ^{99m}Tc eluate of 20 GBq, ^{99}Tc = 200 Bq.
- A higher ^{99}Tc was observed in the eluate (fresh eluate with short ingrowth time)

^{99}Mo and ^{103}Ru in ^{99m}Tc eluate from ^{99}Mo-^{99m}Tc Generator of GE Healthcare (4 GBq)



Sr resin

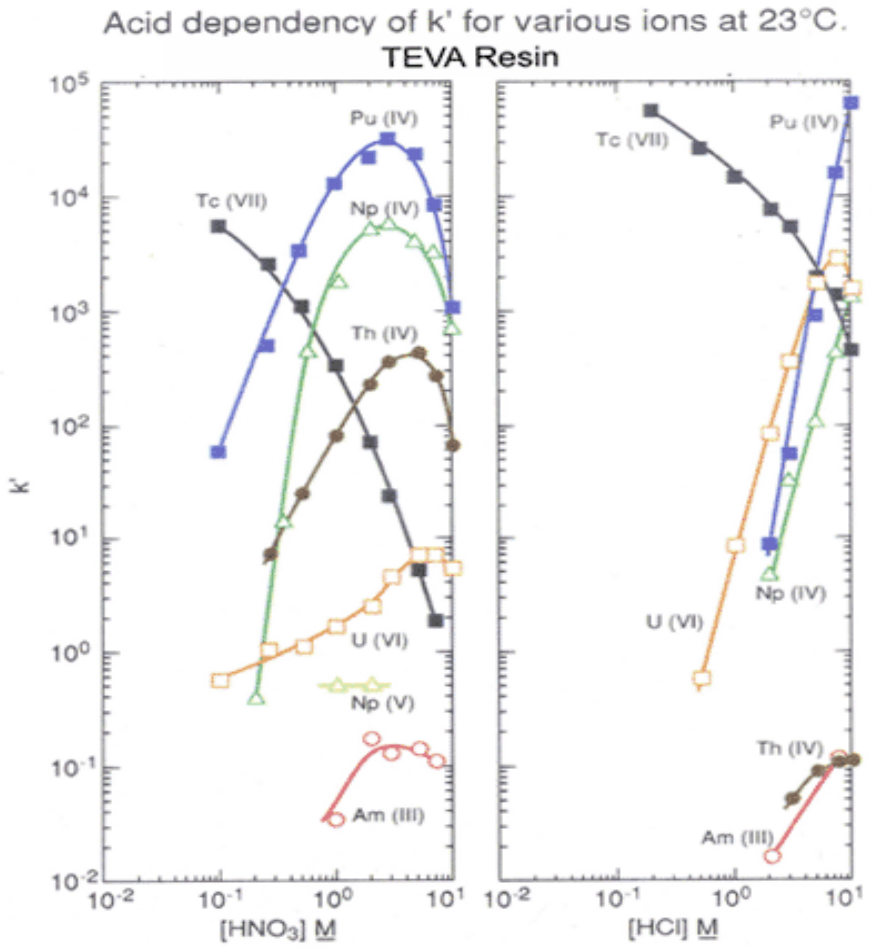
Figures 4 and 5



Horwitz (HP199)

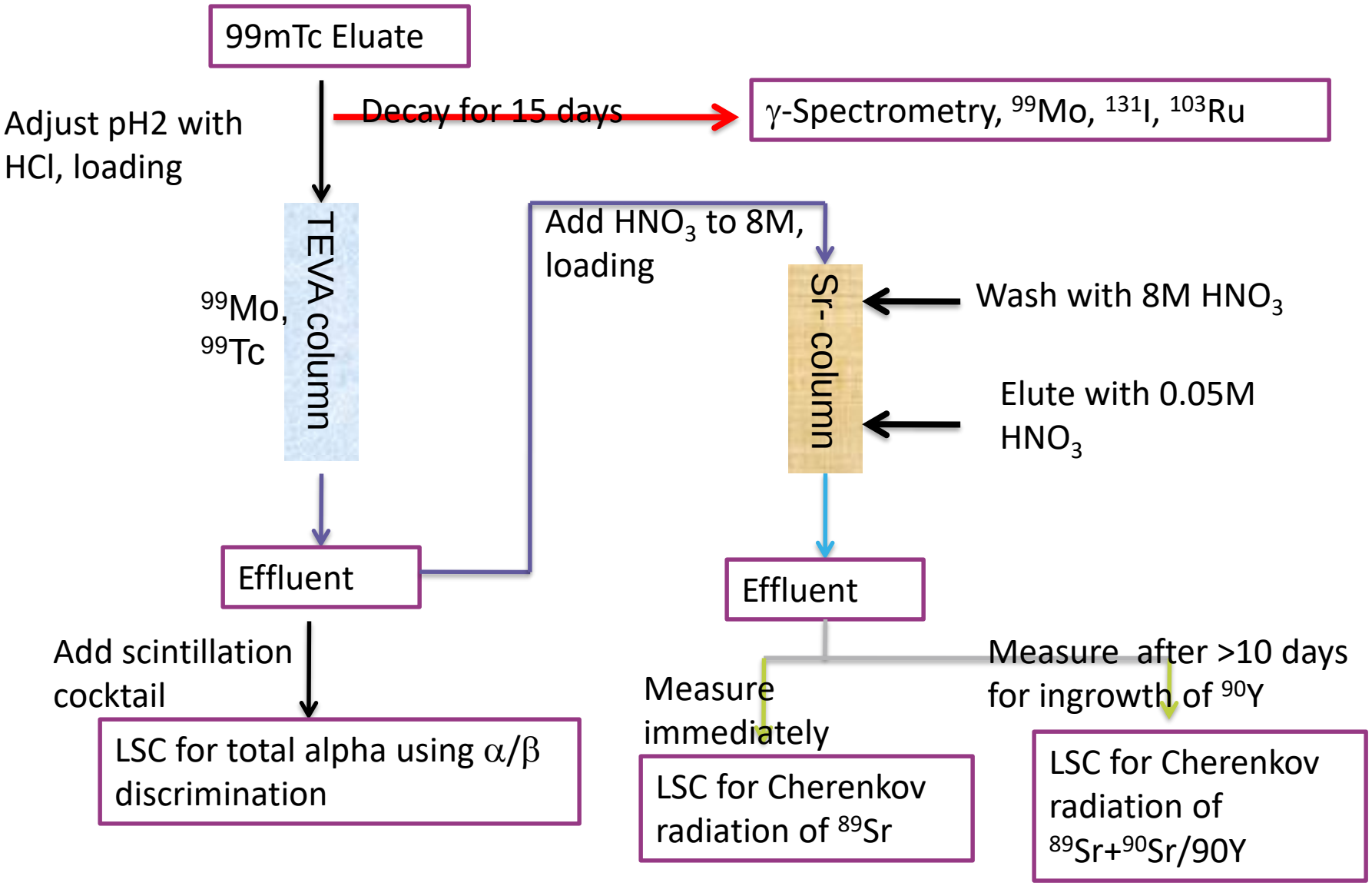
TEVA resin

Figures 2 & 3



Horwitz, et al. (HP195)

Procedure for determination of radionuclidic impurities in ^{99m}Tc eluate



Detection limit to the impurities

Item	Anal. method	Volume of eluate, mL	Detection limit, Bq *	Limitation by Eu Ph. ** Bq
^{99}Mo	γ-spec.	2.0	<250	2×10^6
^{131}I	γ-spec.	2.0	<20	1×10^5
^{103}Ru	γ-spec.	2.0	<6.5	1×10^5
Other gamma #	γ-spec.	2.0	<5	1×10^6
^{89}Sr	LSC	1.0	<0.20	600
^{90}Sr	LSC	1.0	<0.15	60
Total beta #	LSC	1.0	< 0.40	5×10^5
Total alpha	LSC	1.0	< 0.01	1.0

* Considering a decay time of 15 days from the eluting.

Thank you for your attention !