

Determination of Difficult-to-Measure Nuclides in Radioactive Wastes of NPP Paks, Hungary

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Nuclear facilities, radioactive wastes and waste characterization in Hungary

Determination of DTM nuclides at RADANAL Ltd.

Determination of actinides and ^{93}Zr in NPP wastes with combined methods based on the use of

UTEVA,

TRU,

DGA

Determination of DTM fission products:

^{90}Sr ,

^{135}Cs

Determination of DTM activation products:

^{55}Fe - ^{59}Ni - ^{63}Ni

$^{93\text{m}}\text{Nb}$ - ^{94}Nb - ^{125}Sb

Brief overview on the determination of DTM nuclides at INR-ISOTOPTECH Co.

^3H , ^{14}C , ^{36}Cl , ^{129}I , ^{99}Tc , $^{108\text{m}}\text{Ag}$, ^{107}Pd , ^{79}Se

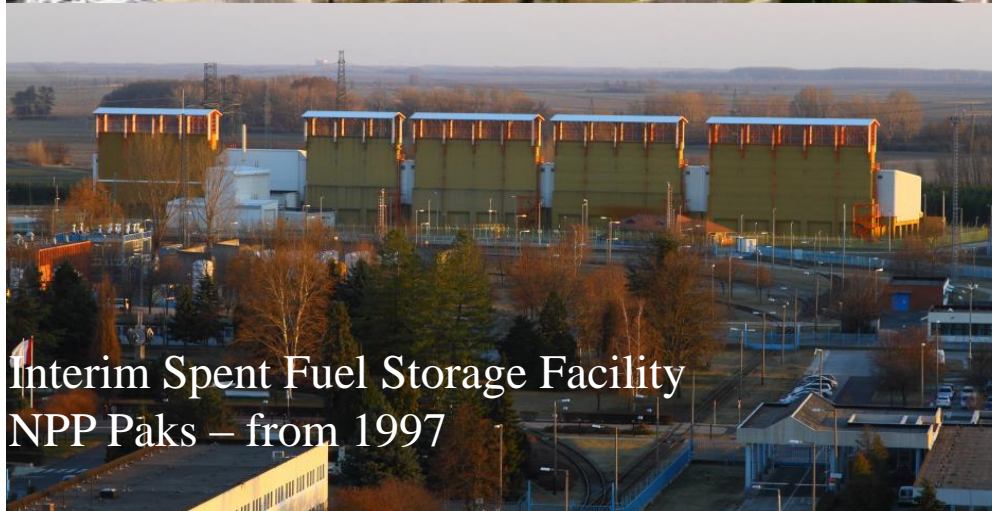


Scaling factors

Nuclear facilities in Hungary

VVER 440/V 213 pressurised water reactors

From
1982

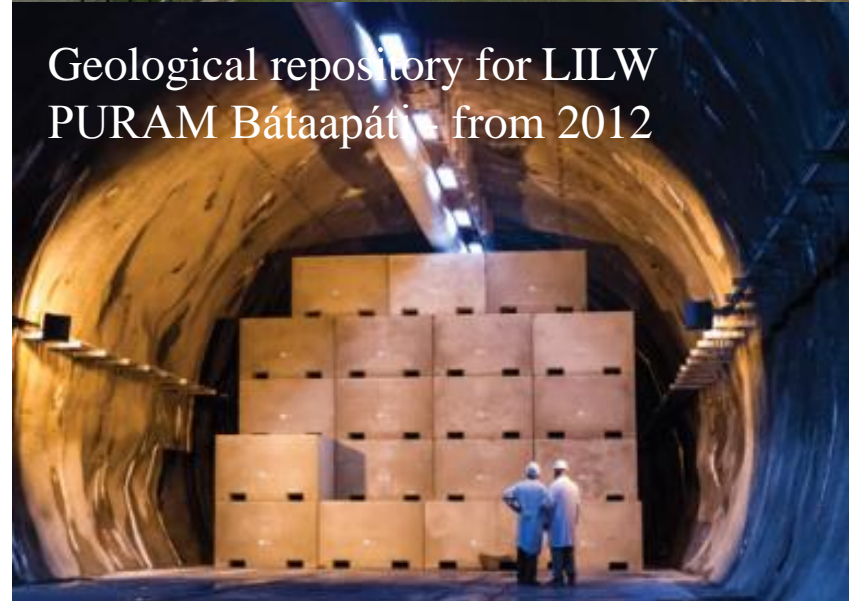


Interim Spent Fuel Storage Facility
NPP Paks – from 1997

Near surface repository for
institutional LILW
PURAM Püspökszilágy



Geological repository for LILW
PURAM Bataapát – from 2012



Waste processing and characterization at NPP Paks

Solid wastes:

collection, compression, storage in drums (210 L), 707 drums in 2014

Characterization by Segmented Gamma Scanning (SGS) of drums and SF method

Liquid radioactive wastes:

radioactivity originates from the PC

collected and processed by evaporation

- **evaporator concentrates:** stored in tanks of 200 m³
 - ~400 g/L salt content (borate), pH ~13 (NaOH),
oxalic acid, citric acid, EDTA
 - LILW
 - ~6500 m³ by 2014

liquid waste treatment technology (LWT):

destruction of organics with KMnO₄, removal of Co by filtration of hydroxide ppt,

ultrafiltration, Cs removal with CsTREAT (FORTUM) ion exchanger, borax removal

solidification by cementization, storage in drums, burial at Waste Depository Bataapáti + clearance

wastes of different composition

- **decontamination solutions:** AP-CITROX technology
- **spent resins**

Characterization by γ scanning, isotope specific analysis → SF calculation



Nuclides to be analyzed

Institute for Nuclear Research, HAS - ISOTOPTECH Co.				
Nuclide	T _{1/2} (year)	MDA [Bq/l]	Type of measurement	Remark
Activation products				
³ H	12,32	1	beta spectrometry	DTM
¹⁴ C	5730	1	beta spectrometry	DTM
³⁶ Cl	301000	10	beta spectrometry	DTM
⁴¹ Ca	102000	10	AMS	DTM
Fission products				
¹²⁹ I	15700000	0,1	beta spectrometry	DTM
⁹⁹ Tc	211000	0,1	beta spectrometry	DTM
¹⁰⁶ Ru	1,01	NR	gamma spectrometry	
¹⁰⁷ Pd	6500000	1	beta spectrometry	DTM
⁷⁹ Se	327000	10	ICP-MS	DTM
¹³⁴ Cs	2,06	NR	gamma spectrometry	
¹³⁷ Cs	30,07	10	gamma spectrometry	KN
¹⁴⁴ Ce	0,78	NR	gamma spectrometry	
¹⁵⁴ Eu	8,8	NR	gamma spectrometry	
Activated corrosion products				
⁵⁴ Mn	0,85	NR	gamma spectrometry	
⁶⁰ Co	5,27	10	gamma spectrometry	KN
^{108m} Ag	418	1	gamma spectrometry	DTM
^{110m} Ag	0,68	NR	gamma spectrometry	
KN	Key Nuclide			
DTM	Difficult-to-Measure Nuclide			

RADANAL Ltd.				
Nuclide	T _{1/2} (year)	MDA [Bq/l]	Type of measurement	Remark
Fission products				
⁹⁰ Sr	29,1	1	beta spectrometry	DTM
¹³⁵ Cs	2300000	1	NAA	DTM
			ICP-MS	
¹²⁵ Sb	2,76	NR	gamma spectrometry	DTM
Activated corrosion products				
⁵⁵ Fe	2,73	1	X-ray spectrometry	DTM
⁵⁹ Ni	75000	1	X-ray spectrometry	DTM
⁶³ Ni	100	10	beta spectrometry	DTM
⁹³ Zr	1500000	1	ICP-MS	DTM
^{93m} Nb	16,1	10	X-ray spectrometry	DTM
⁹⁴ Nb	20300	1	gamma spectrometry	DTM
Actinides				
²³⁴ U	244000	0,05	alpha spectrometry	DTM
²³⁵ U	704000000	0,05	alpha spectrometry	DTM
²³⁷ Np	2100000	0,05	ICP-MS	DTM
²³⁸ U	4470000000	0,05	alpha spectrometry	DTM
²³⁸ Pu	86,4	0,05	alpha spectrometry	DTM
²³⁹ Pu	24400	0,05	alpha spectrometry	DTM
²⁴⁰ Pu	6540	0,05	alpha spectrometry	DTM
²⁴¹ Am	432	0,05	alpha spectrometry	DTM
²⁴³ Am	7380	0,05	alpha spectrometry	DTM
²⁴² Cm	0,45	0,05	alpha spectrometry	DTM
²⁴⁴ Cm	17,9	0,05	alpha spectrometry	DTM

Determination of DTM nuclides at RADANAL Ltd.

Determination of actinides and ^{93}Zr

Goal was to develop a combined procedure for the selective separation of Pu, Am-Cm, Np, U, Th (Zr) using a **single chromatographic column**.

The procedure should be

- adequate for analysis of liquid NPP waste (accurate, sensitive for LLW)
- adequate for measurement by α spectrometry and/or ICP-MS
- simple, cheap, fast for use in emergency situation.

**Resin of high selectivity
for actinides is needed.**

3 methods have been developed:

UTEVA procedure

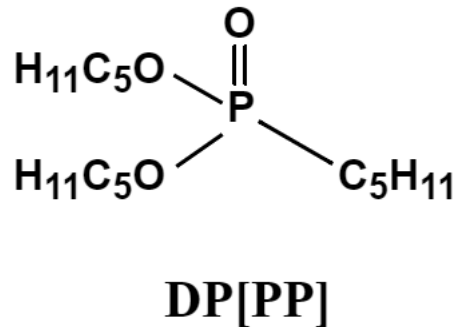
TRU procedure

DGA procedure

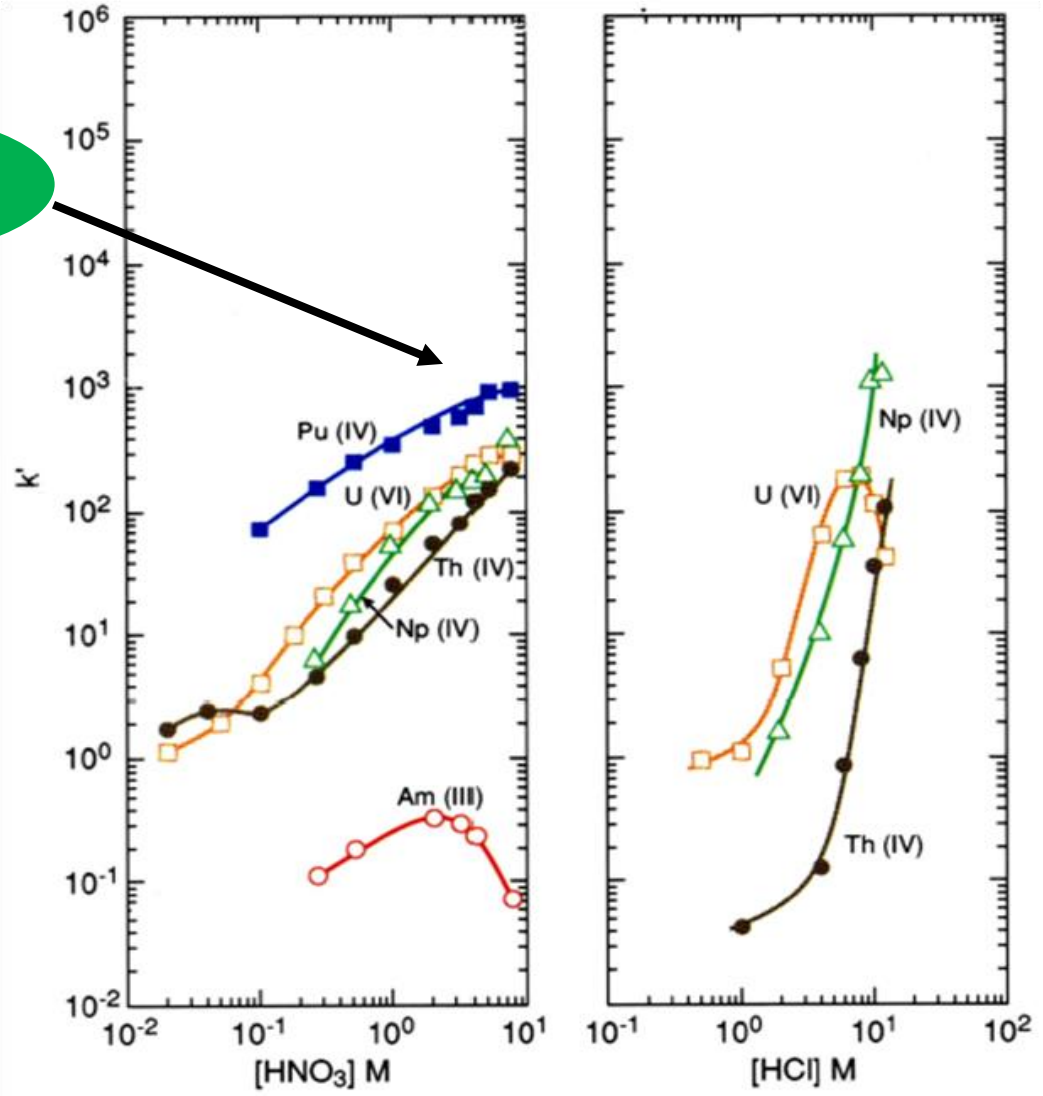
UTEVA® procedure

Chromatographic procedure for actinides (U, Th, Pu, Np)

Pu(IV) or Pu(VI)
 U(VI)
 Th(IV)
 Np(VI)



High k' for
 Ac(IV, VI)
 not for Am



® Triskem International

Horwitz et al.

N. Vajda, Zs. Molnár, E. Kabai, Sz. Osvath: Simultaneous Determination of Long-Lived Radionuclides in Environmental Samples, 9th Int. Symposium on Environmental Radiochemical Analysis, Maidstone, U.K., 18-20 Sept. 2002, (In: Peter Warwick (ed.): Environmental Radiochemical Analysis II; The Royal Society of Chemistry, Cambridge, 2003; pp. 185-196.)

UTEVA procedure: Basic concept of separation

- **Load:**

U(VI), Th(IV), Np(VI), Pu(VI) – $K_2S_2O_8$ or

U(VI), Th(IV), Pu(IV) – $NaNO_2$

retention of actinides from 8M HNO_3 /0.5M $Fe(NO_3)_3$:

**Ac(VI) and Ac(IV)
after oxidation
state adjustment**

**Fe(III) nitrate is
used as salting-out
agent**

- **Elution of Pu(III)** with 9M HCl/ NH_4I

Reduction of Pu to Pu(III)

**On-column
reduction to**

Pu(III), Np(IV)

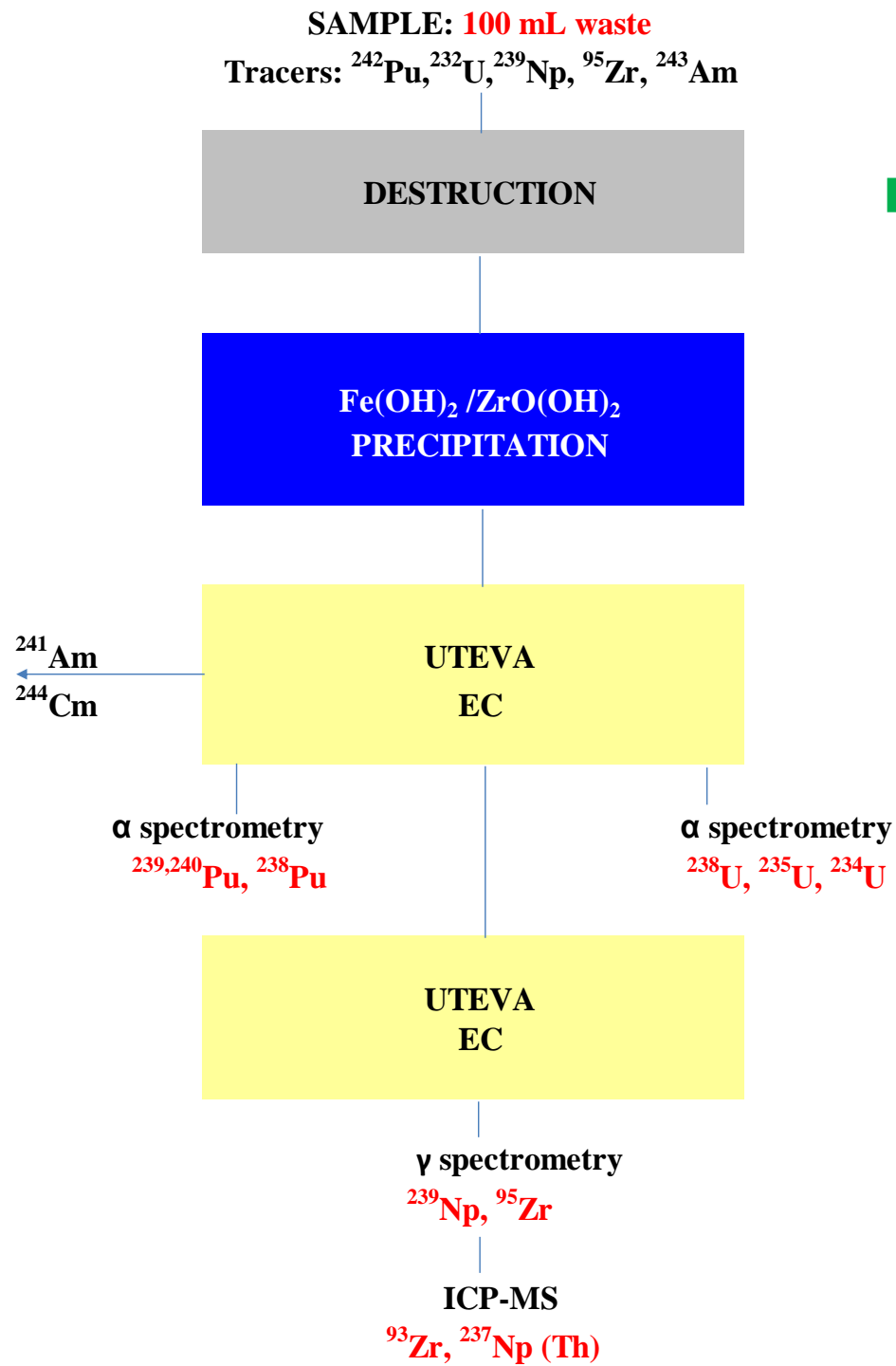
- **Elution of Th, Np** with 4M HCl

- **Elution of U** with 0.1M HCl.

**Ac(III) are not
retained!**

Determination of U, Pu (Np, Zr) in NPP wastes

Flowchart of the UTEVA procedure



Acid destruction

Preconcentration is necessary

Oxidation state adjustment is recommended:
 $\text{K}_2\text{S}_2\text{O}_7$ with Ag^+ catalyst to form Np(VI) , Pu(VI)

Load from 8M HNO_3 with $\text{Fe(NO}_3)_3$ as salting-out agent

Optional: Np, Zr determination

Np-Zr purification with UTEVA from 9M HCl load

Optional: Am determination from the 1st UTEVA effluent:

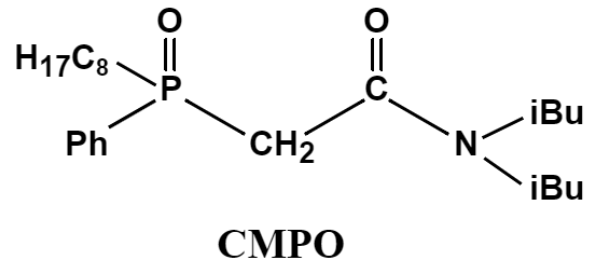
Preconcentration with Ca oxalate

Am separation with TRU resin

TRU® procedure

Chromatographic procedure for all actinides (Ac) in small samples

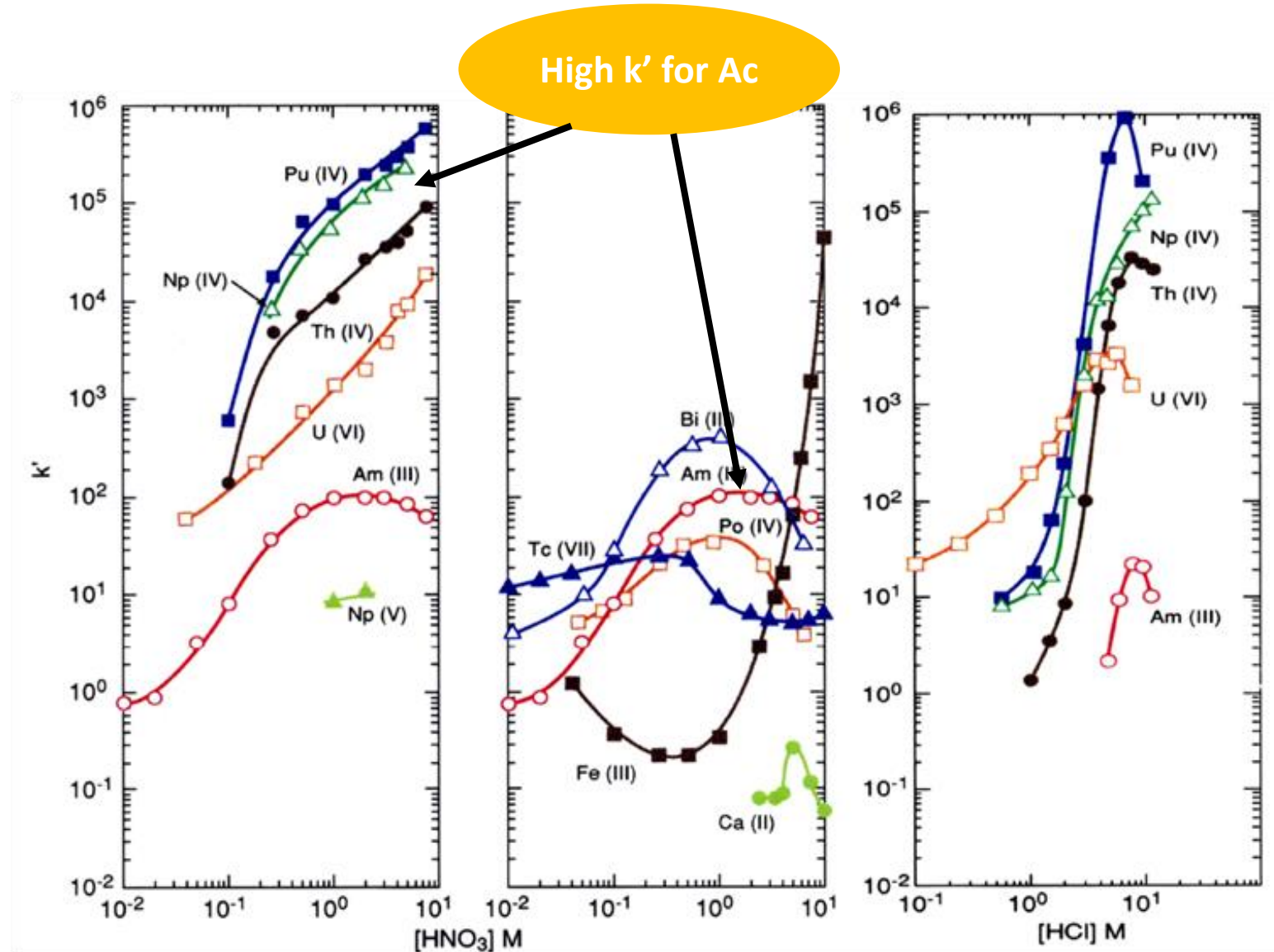
Pu Am
Np
Th
U



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Horwitz et al.

N. Vajda, A. Törvényi, G. Kis-Benedek, C.K. Kim, B. Bene, Zs. Macsik: Rapid method for the determination of actinides in soil and sediment samples by alpha spectrometry. *Radiochimica Acta* **2009**, 97, 395-401



TRU procedure: Basic concept of the separation

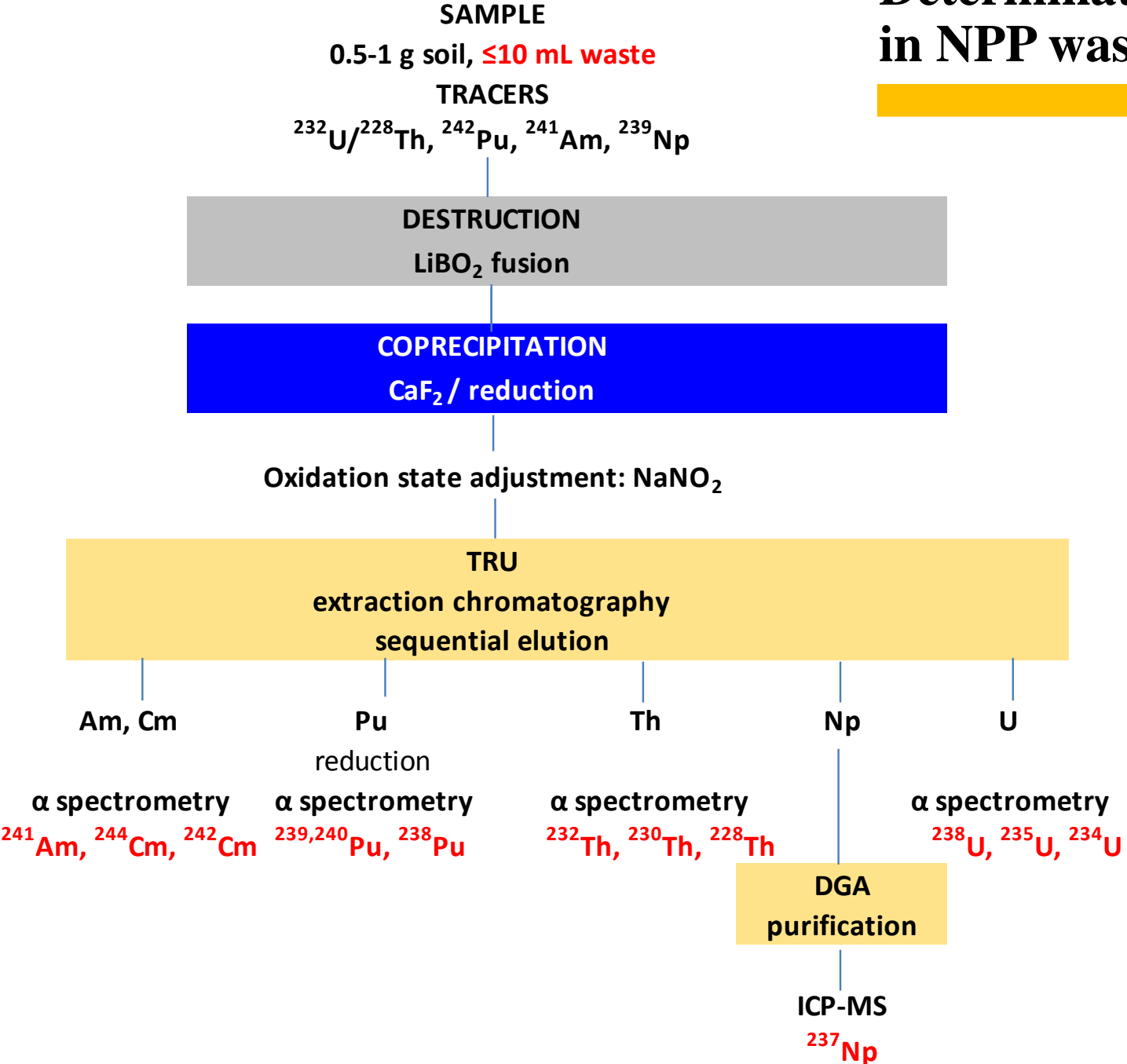
- **Load:** retention of all actinides from 2M HNO₃
Pu(IV), Am(III), U(VI), Th(IV), Np(IV),
- **Elution of Am** with 4M HCl.
- **Elution of Pu(III)** with 4M HCl /Ti³⁺
while
Np is reduced to Np(IV) - retained,
Th and U(IV) are retained
- **Elution of Th** with 2M HCl/Ti⁴⁺
- **Elution of Np** with 2M HCl/Ti³⁺
while U is retained,
- **Elution of U** with 0.1M HCl.

Ac(III), Ac(IV),
Ac(VI) are retained

On-column redox
reactions are
feasible!

Small load
otherwise Am
leakage

Determination of Pu, Am, Cm, U (Np) isotopes in NPP wastes – flowchart of TRU procedure



Rapid method for Am and Pu due to fusion
Optional: destruction with mineral acids

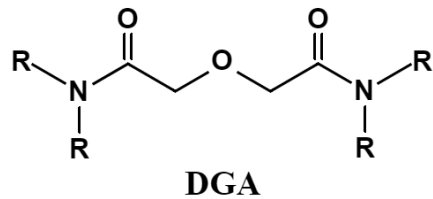
Coprecipitation is necessary
Removal of matrix

Optional analysis of **Pu, Th, Np, U isotopes** after purification with DGA resin columns to remove Ti species using other tracers **by ICP-MS**

DGA® procedure

Chromatographic procedure for all actinides (Ac)

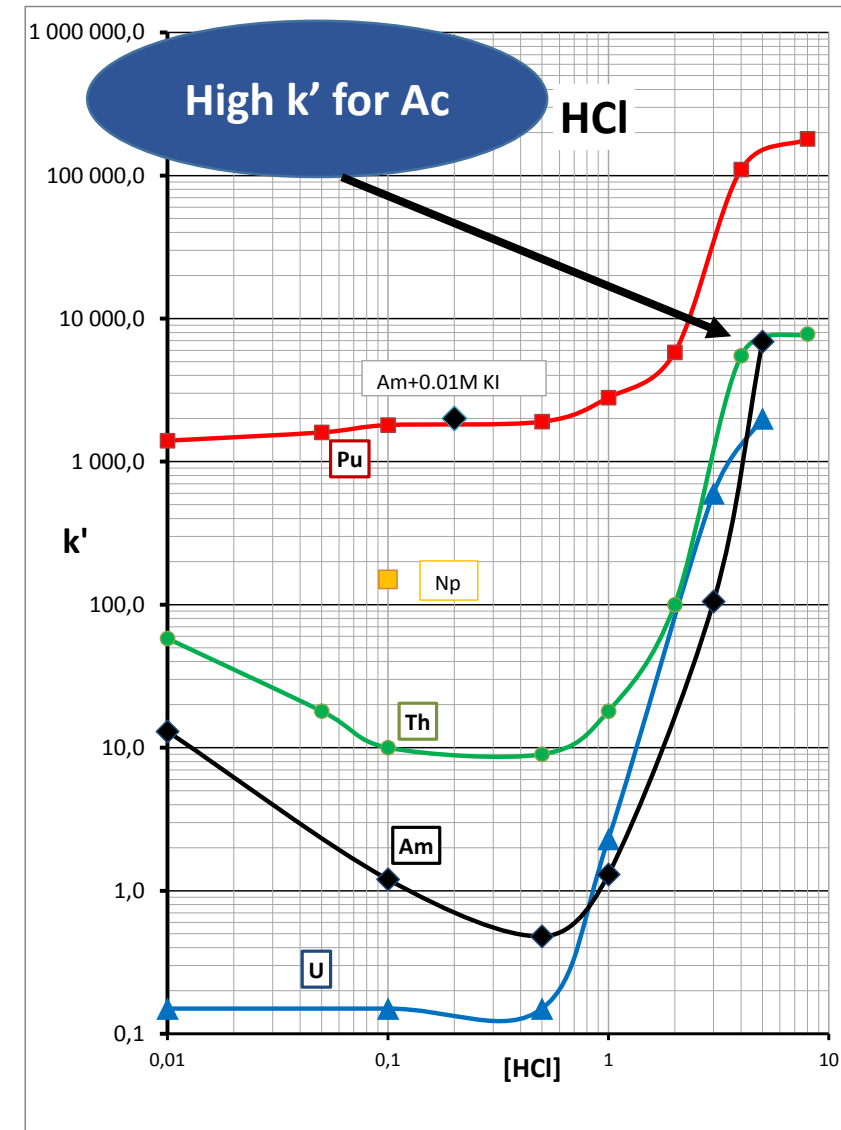
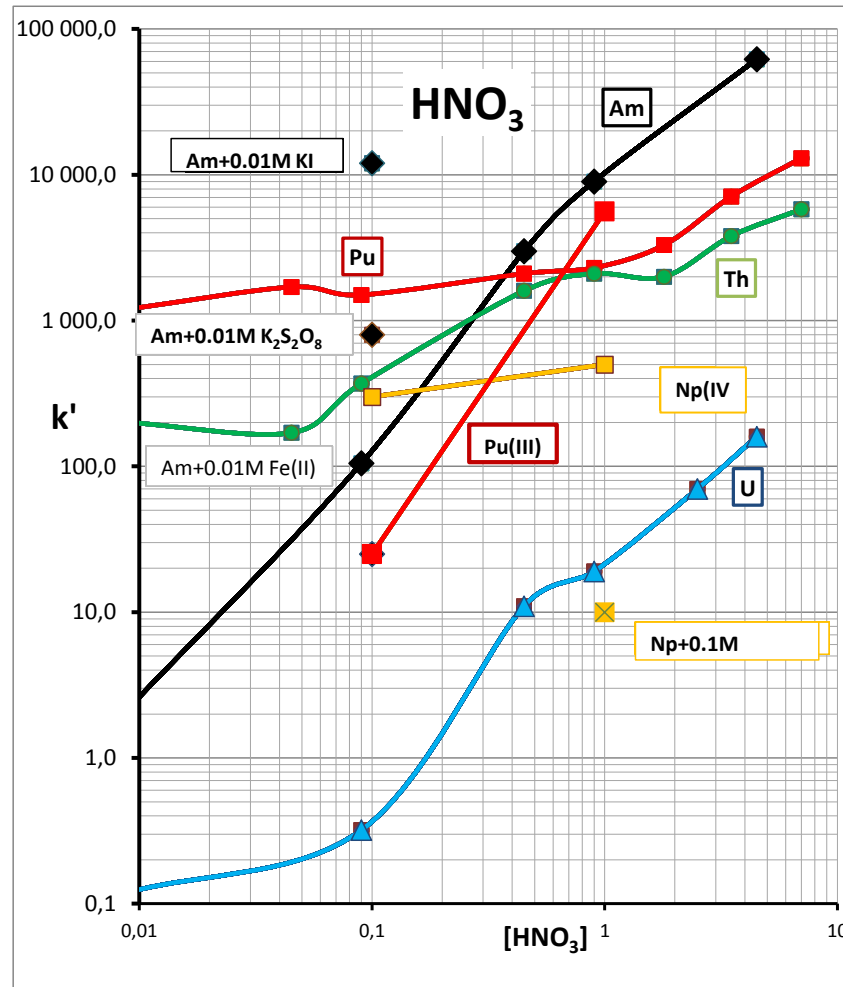
Pu
Am
U
Th
Np



® Triskem International

Horwitz et al.

Groska J, Vajda N, Molnar Z, Bokori E, Szeredy P, Zagyvai M (2016) Determination of actinides in radioactive waste after separation on a single DGA resin column, JOURNAL OF RADIOANALYTICAL AND NUCLEAR CHEMISTRY 309: (3) pp. 1145-1158.



DGA procedure: Basic concept of the separation

- **Load:** retention of all actinides in reduced forms from 4M HCl/ Na₂SO₃
U(IV), Th(IV), Np(IV), Pu(III), Am(III)
- **Elution of U** with dilute HNO₃ after oxidation to U(VI), while
Pu and Np are oxidized to Pu(IV), Np(IV)/Np(VI) - retained,
- Reduction of Pu and Np to Pu(III) and Np(IV),
- **Elution of Th, Np** with complexing agent oxalic acid, while
trivalent actinides (Pu(III), Am) are retained,
- **Elution of Pu** with oxalic acid after oxidation to Pu(IV)
- **Elution of Am** with dilute HCl.

**Ac(III) and Ac(IV)
are completely
retained!**

**On-column redox
reactions are
feasible!**

**Only Ac(IV) form
oxalate complexes !**

Optimization of the DGA procedure

Studied by **model experiments**

with single tracers

^{233}U , ^{230}Th , ^{239}Pu , ^{241}Am

^{239}Np produced from $^{238}\text{U}(n,\gamma)$

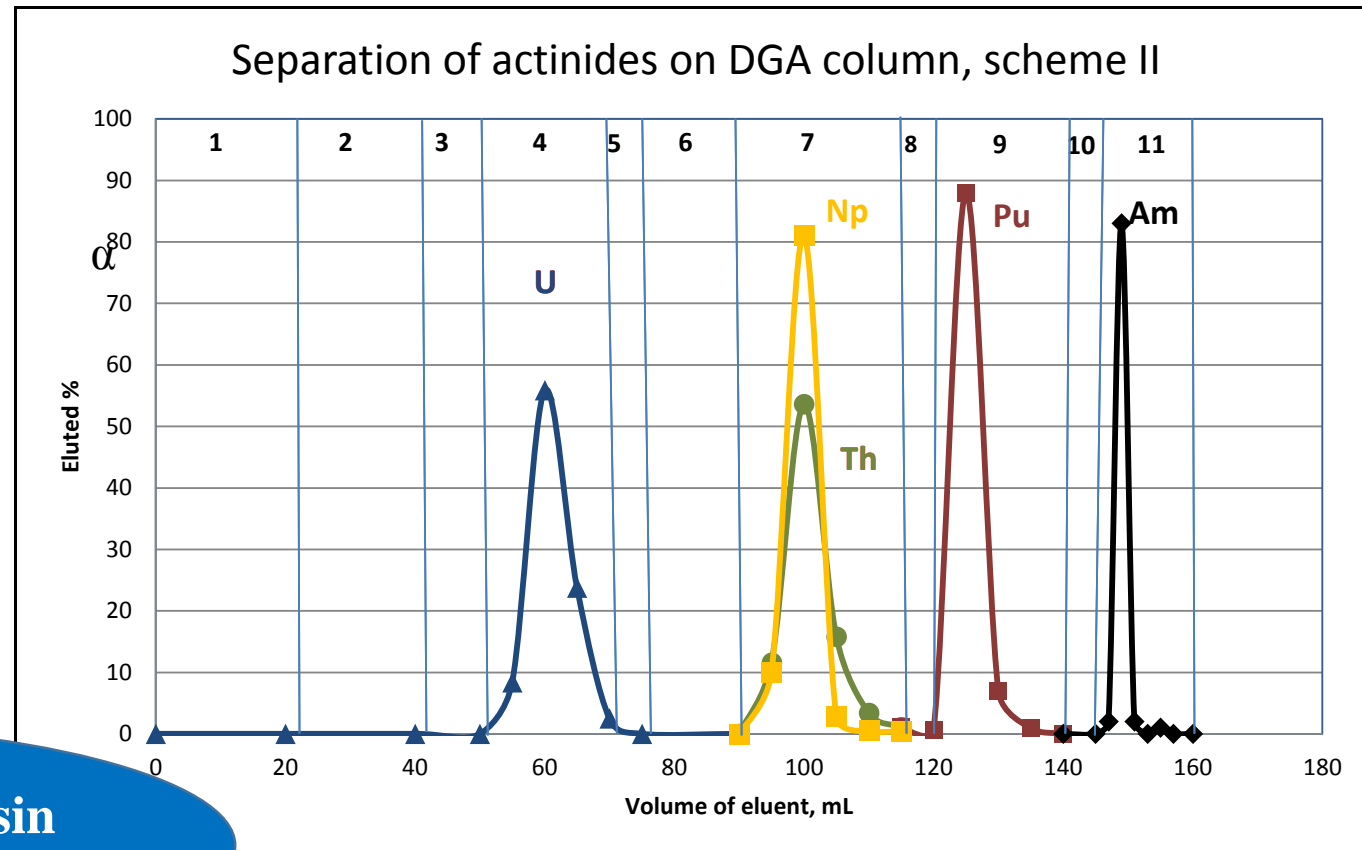
measured by LSC or γ spectrometry

- U, Pu, Am fractions are well separated
- Np and Th are collected together
- Between the strip solutions wash solutions
- Recoveries are high (> 86%)
- Contamination by other actinides is $\leq 1\text{-}2\%$

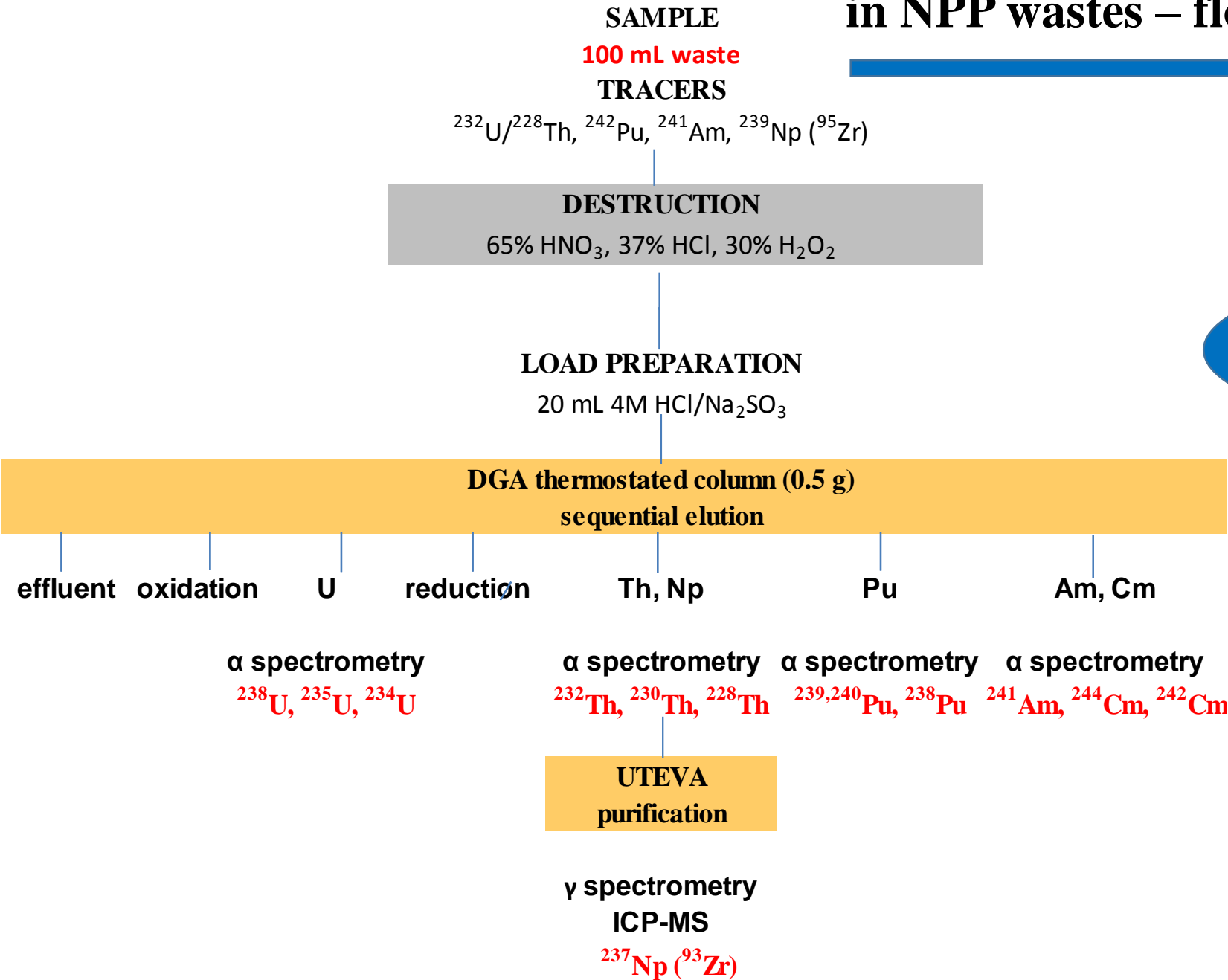
**Major advantage:
Use of a single small resin
column for all Ac in big
samples!**

Recoveries

U	Th	Np	Pu	Am
90%	86%	95%	96%	88%



Determination of U, Pu, Np, Am, Cm, Zr isotopes in NPP wastes – flowchart of DGA procedure



Big volumes are treated.

Conventional destruction.

Preconcentration of Ac and Zr is skipped!

Purification of Np from U is necessary.

Determination of ^{237}Np and ^{93}Zr in NPP wastes

UTEVA procedure:

DESTRUCTION
CO-PRECIIPITATION
 $\text{Fe}(\text{OH})_2/\text{Zr}(\text{OH})_4$

UTEVA: Np, Zr separation
UTEVA: Np, Zr purification

ICP-MS

DGA procedure:

DESTRUCTION

DGA: Np, Zr separation
UTEVA: Np, Zr purification

ICP-MS

Unpredictable behaviour of Zr, because
HF is used to dissolve $\text{ZrO}_2 \leftrightarrow$
 ZrF_4 is not retained on UTEVA

**High and constant
Zr (71%) and Np (84%)
recoveries**

Determination of ^{90}Sr in NPP wastes

Sample: 100 mL
Carrier: 10 mg Sr
AAS

Destruction

Ca(Sr) oxalate precipitation

Sr Resin

time of separation of Y from Sr

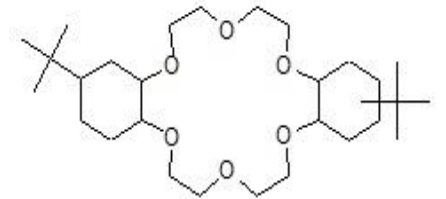
Sr oxalate

Sr yield: gravimetry/ AAS
LSC1
LSC2

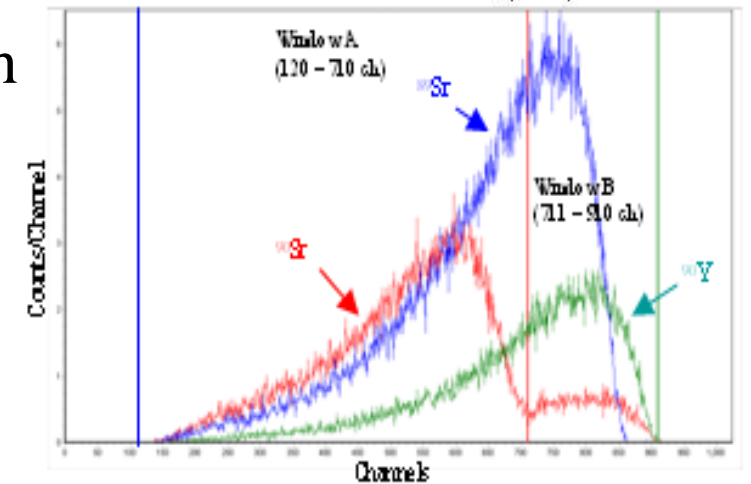
^{90}Sr ^{89}Sr

Sr yield is determined from gravimetry/AAS.

High selectivity for Sr is assured by the use of Sr selective crown ether: Sr Resin



By repeated measurement both ^{90}Sr and ^{89}Sr are determined.



The method is combined with the determination of actinides.

DESTRUCTION

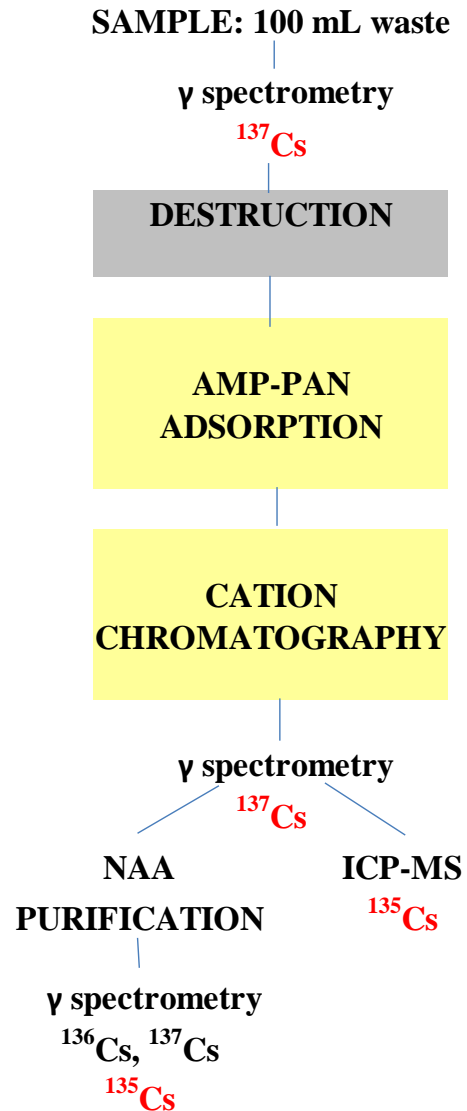
PRECIPITATION, ADSORPTION

EXTRACTION, CHROMATOGRAPHY

Results of U, Pu, Np, Am, Cm, Zr, Sr isotope analyses in NPP wastes

Bq/dm ³	Sample1 (normal evaporator concentrate)		Sample2 (alpha-cont./emergency evaporator concentrate)		Sample3 (decontamination solution)	
	a	unc(a)				
³ H	4,00E+05	2,0E+04	2,12E+05	1,1E+04	7,68E+04	3,8E+03
¹⁴ C	1,01E+04	1,01E+03	2,96E+03	2,96E+02	3,48E+02	3,48E+01
³⁶ Cl	3,65E+01	1,26E+01	9,03E+01	3,46E+01	< 1,1E+01	
⁵⁵ Fe	2,44E+05	2,44E+04	9,37E+06	9,36E+05	1,39E+05	2,08E+04
⁶³ Ni	4,41E+04	2,21E+03	1,75E+06	3,49E+05	2,78E+05	5,56E+04
⁹⁰ Sr	4,92E+02	3,32E+01	1,68E+06	2,35E+05	3,04E+05	2,33E+04
⁹³ Zr	3,52E+02	8,89E+00	8,03E-01	2,43E-02	5,06E-02	1,39E-03
⁹⁴ Nb	7,00E+02	1,40E+02	N/A		8,50E+02	1,00E+02
⁹⁹ Tc	< 1,7E-01		1,77E+01	5,35E+00	1,06E+01	2,07E+00
^{108m} Ag	1,38E+02	8,81E+00	1,34E+04	7,98E+02	1,32E+02	4,71E+00
¹²⁵ Sb	≤ 5,0E+02		N/A		9,00E+03	8,00E+02
¹²⁹ I	1,94E-01	1,75E-02	7,32E-01	8,04E-02	< 2,7E-01	
²³⁴ U	2,20E-01	2,05E-02	≤ 1,67E+03		3,39E+01	6,88E+00
²³⁵ U	≤ 1,13E-02		≤ 1,66E+03		≤ 7,73E+00	
²³⁷ Np	4,55E-03	5,27E-04	1,38E-02	7,95E-04	3,36E-03	1,94E-04
²³⁸ Pu	1,29E+01	4,65E-01	9,37E+04	3,38E+03	1,38E+04	4,48E+02
²³⁸ U	1,49E-01	1,64E-02	≤ 1,14E+03		8,79E+00	3,60E+00
^{239,240} Pu	9,39E+00	3,53E-01	9,84E+04	3,53E+03	1,25E+04	4,08E+02
²⁴¹ Am	7,47E+00	2,60E-01	8,73E+04	3,23E+03	1,33E+04	4,55E+02
²⁴⁴ Cm	3,24E+00	1,24E-01	1,22E+04	7,27E+02	2,77E+03	1,18E+02

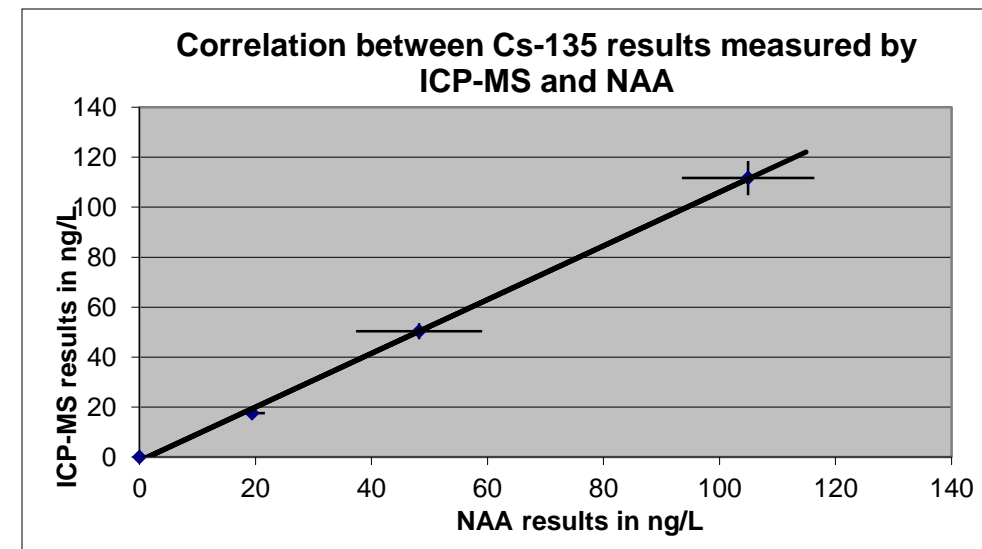
Determination of ^{135}Cs in NPP wastes



AMP is a selective ion exchanger for Cs.

Small cation column is used to separate Ba, the isobar interference for ^{135}Cs in ICP-MS.

NAA and ICP-MS results correlated well:



Measured concentrations in wastes: 20-100 ng/L \sim 1-5 Bq/L

Determination of ^{55}Fe , ^{59}Ni , ^{63}Ni in NPP wastes

SAMPLE
Carriers: 6 mg Fe, 6 mg Ni

DESTRUCTION

MIBK
EC

Fe yield: AAS

LSC/X spectrometry

^{55}Fe

DMG
EC

DMG precipitate

Ni yield: AAS

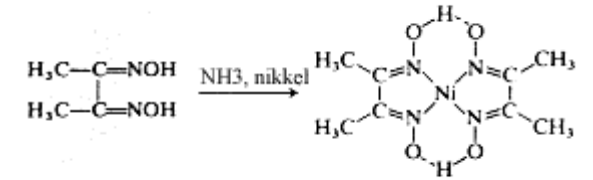
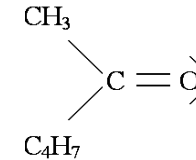
X spectrometry

^{59}Ni

LSC

^{63}Ni

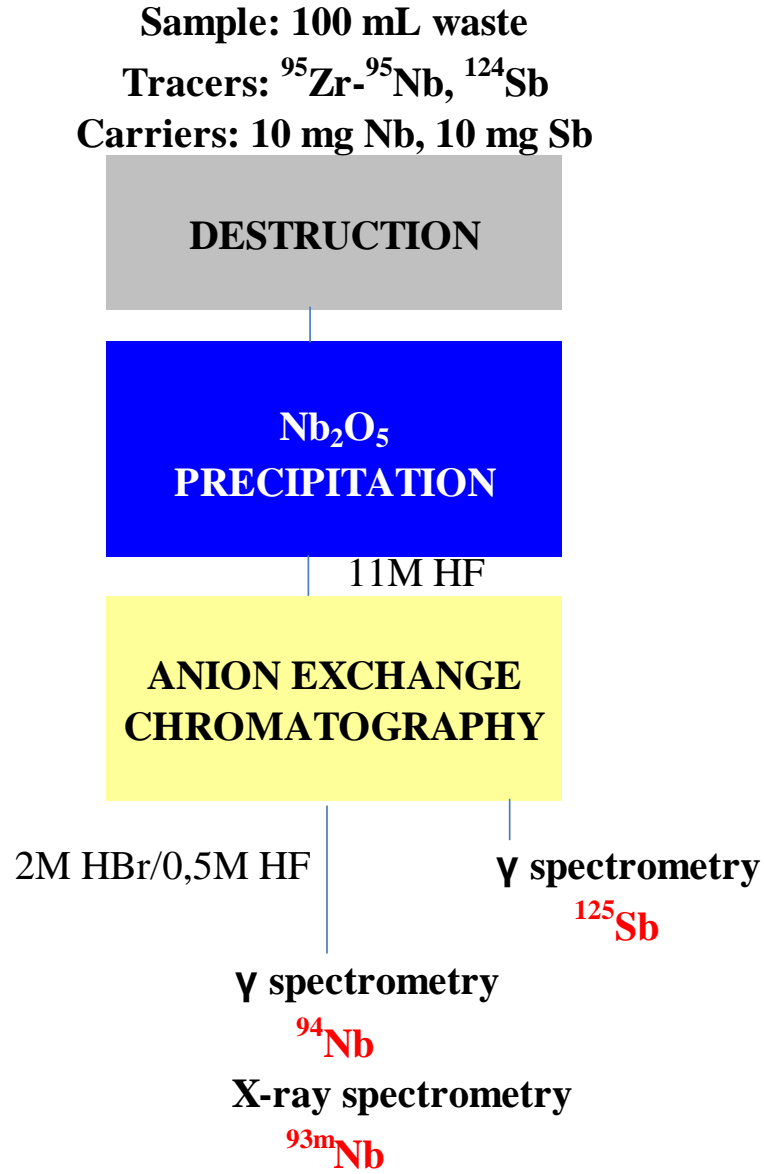
Home made EC
resins are used:



a unc(a)

Bq/dm ³	Sample1 (normal evaporator concentrate)		Sample2 (alpha-cont./emergency evaporator concentrate)		Sample3 (decontamination solution)	
^3H	4,00E+05	2,0E+04	2,12E+05	1,1E+04	7,68E+04	3,8E+03
^{14}C	1,01E+04	1,01E+03	2,96E+03	2,96E+02	3,48E+02	3,48E+01
^{36}Cl	3,65E+01	1,26E+01	9,03E+01	3,46E+01	< 1,07E+01	
^{55}Fe	2,44E+05	2,44E+04	9,37E+06	9,36E+05	1,39E+05	2,08E+04
^{63}Ni	4,41E+04	2,21E+03	1,75E+06	3,49E+05	2,78E+05	5,56E+04
^{90}Sr	4,92E+02	3,32E+01	1,68E+06	2,35E+05	3,04E+05	2,33E+04
^{93}Zr	3,52E+02	8,89E+00	8,03E-01	2,43E-02	5,06E-02	1,39E-03
^{99}Tc	< 1,67E-01		1,77E+01	5,35E+00	1,06E+01	2,07E+00
$^{108\text{m}}\text{Ag}$	1,38E+02	8,81E+00	1,34E+04	7,98E+02	1,32E+02	4,71E+00
^{129}I	1,94E-01	1,75E-02	7,32E-01	8,04E-02	< 2,67E-01	
^{238}Pu	1,29E+01	4,65E-01	9,37E+04	3,38E+03	1,38E+04	4,48E+02
$^{239,240}\text{Pu}$	9,39E+00	3,53E-01	9,84E+04	3,53E+03	1,25E+04	4,08E+02
^{241}Am	7,47E+00	2,60E-01	8,73E+04	3,23E+03	1,33E+04	4,55E+02
^{244}Cm	3,24E+00	1,24E-01	1,22E+04	7,27E+02	2,77E+03	1,18E+02

Determination of ^{93m}Nb , ^{94}Nb , ^{125}Sb in NPP wastes



Results of analysis

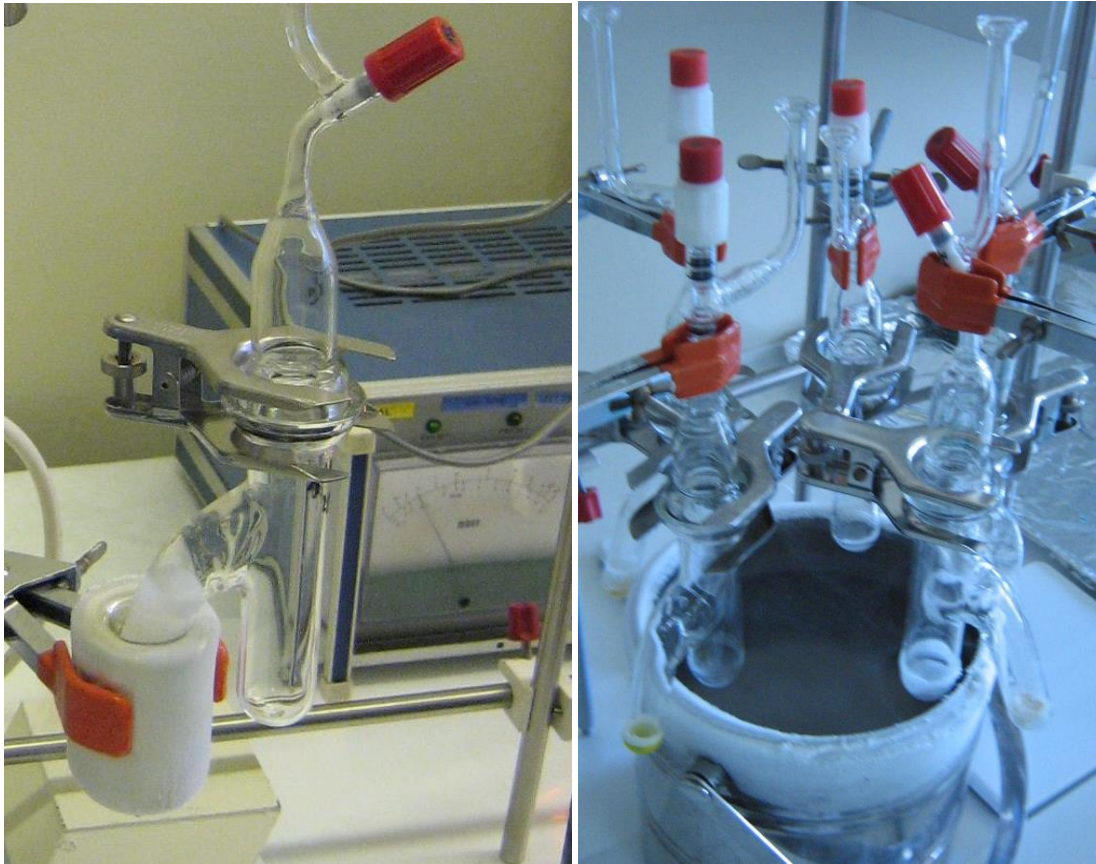
Activity concentration of ^{93m}Nb is usually close to LD.

Bq/dm ³	Sample1 (normal evaporator concentrate)		Sample2 (alpha-cont./emergency evaporator concentrate)		Sample3 (decontamination solution)	
	a	unc(a)				
^3H	4,00E+05	2,0E+04	2,12E+05	1,1E+04	7,68E+04	3,8E+03
^{14}C	1,01E+04	1,01E+03	2,96E+03	2,96E+02	3,48E+02	3,48E+01
^{36}Cl	3,65E+01	1,26E+01	9,03E+01	3,46E+01	< 1,1E+01	
^{55}Fe	2,44E+05	2,44E+04	9,37E+06	9,36E+05	1,39E+05	2,08E+04
^{63}Ni	4,41E+04	2,21E+03	1,75E+06	3,49E+05	2,78E+05	5,56E+04
^{90}Sr	4,92E+02	3,32E+01	1,68E+06	2,35E+05	3,04E+05	2,33E+04
^{93}Zr	3,52E+02	8,89E+00	8,03E-01	2,43E-02	5,06E-02	1,39E-03
^{94}Nb	7,00E+02	1,40E+02	N/A		8,50E+02	1,00E+02
^{99}Tc	< 1,7E-01		1,77E+01	5,35E+00	1,06E+01	2,07E+00
^{108m}Ag	1,38E+02	8,81E+00	1,34E+04	7,98E+02	1,32E+02	4,71E+00
^{125}Sb	$\leq 5,0E+02$		N/A		9,00E+03	8,00E+02
^{129}I	1,94E-01	1,75E-02	7,32E-01	8,04E-02	< 2,7E-01	
^{234}U	2,20E-01	2,05E-02	$\leq 1,67E+03$		3,39E+01	6,88E+00
^{235}U	$\leq 1,13E-02$		$\leq 1,66E+03$		$\leq 7,73E+00$	
^{237}Np	4,55E-03	5,27E-04	1,38E-02	7,95E-04	3,36E-03	1,94E-04
^{238}Pu	1,29E+01	4,65E-01	9,37E+04	3,38E+03	1,38E+04	4,48E+02
^{238}U	1,49E-01	1,64E-02	$\leq 1,14E+03$		8,79E+00	3,60E+00
$^{239,240}\text{Pu}$	9,39E+00	3,53E-01	9,84E+04	3,53E+03	1,25E+04	4,08E+02
^{241}Am	7,47E+00	2,60E-01	8,73E+04	3,23E+03	1,33E+04	4,55E+02
^{244}Cm	3,24E+00	1,24E-01	1,22E+04	7,27E+02	2,77E+03	1,18E+02

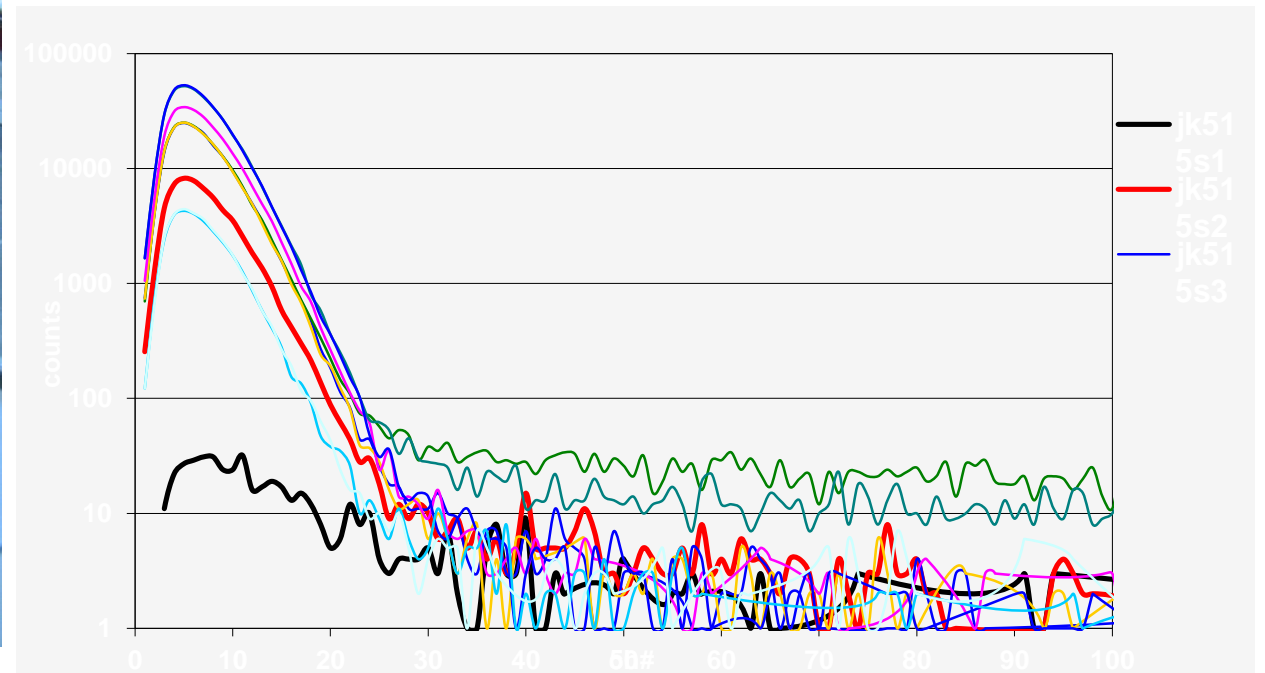
**Determination of DTM nuclides at
INSTITUTE FOR NUCLEAR RESEARCH OF
HUNGARIAN ACADEMY OF SCIENCES (INR HAS) -
ISOTOPTECH Co.**

Determination of ^3H in NPP wastes

Low-pressure cold-distillation



LSC counting



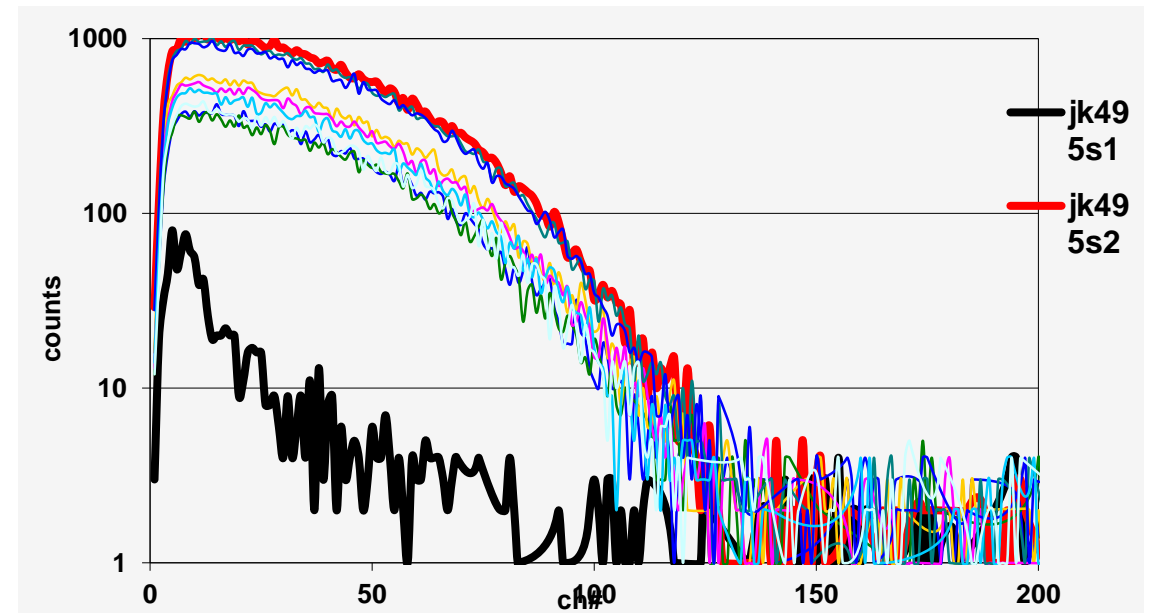
Determination of ^{14}C in NPP wastes

Acidic destruction and 2-step trapping of CO_2

1. step: total oxidation of the sample,
 CO_2 is trapped in $\text{Ba}(\text{OH})_2$ as BaCO_3
2. step: CO_2 is released from BaCO_3 and
trapped in NaOH as NaHCO_3



LSC counting
NaOH solution in HionicFluor



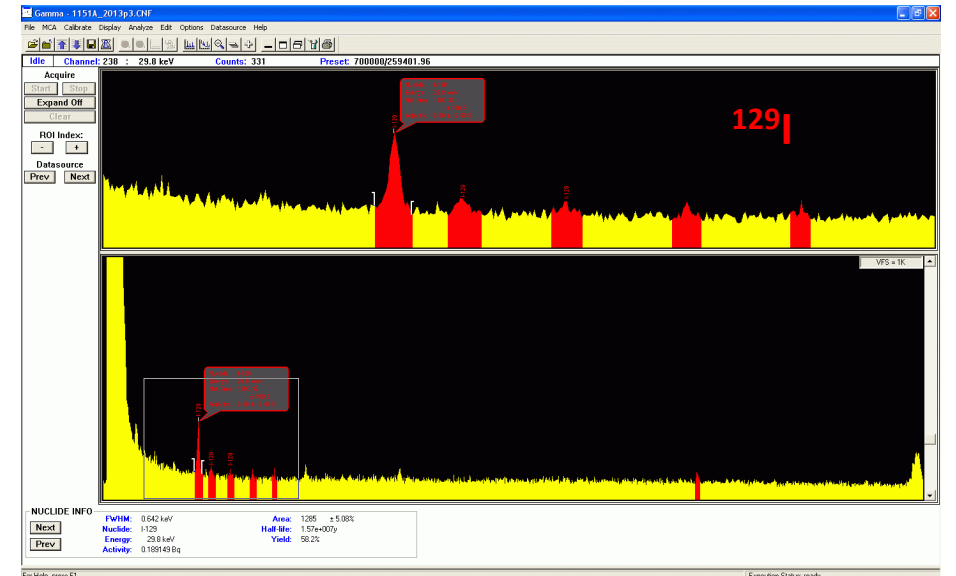
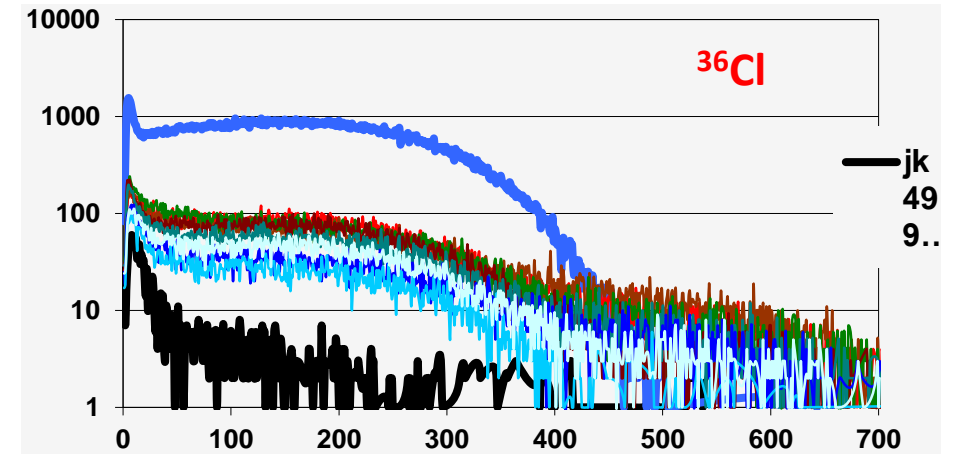
A set of results for 3 sample types

a unc(a)

Bq/dm ³	Sample1 (normal evaporator concentrate)		Sample2 (alpha-cont./emergency evaporator concentrate)		Sample3 (decontamination solution)	
³ H	4,00E+05	2,0E+04	2,12E+05	1,1E+04	7,68E+04	3,8E+03
¹⁴ C	1,01E+04	1,01E+03	2,96E+03	2,96E+02	3,48E+02	3,48E+01
³⁶ Cl	3,65E+01	1,26E+01	9,03E+01	3,46E+01	< 1,07E+01	
⁵⁵ Fe	2,44E+05	2,44E+04	9,37E+06	9,36E+05	1,39E+05	2,08E+04
⁶³ Ni	4,41E+04	2,21E+03	1,75E+06	3,49E+05	2,78E+05	5,56E+04
⁹⁰ Sr	4,92E+02	3,32E+01	1,68E+06	2,35E+05	3,04E+05	2,33E+04
⁹³ Zr	3,52E+02	8,89E+00	8,03E-01	2,43E-02	5,06E-02	1,39E-03
⁹⁹ Tc	< 1,67E-01		1,77E+01	5,35E+00	1,06E+01	2,07E+00
^{108m} Ag	1,38E+02	8,81E+00	1,34E+04	7,98E+02	1,32E+02	4,71E+00
¹²⁹ I	1,94E-01	1,75E-02	7,32E-01	8,04E-02	< 2,67E-01	
²³⁸ Pu	1,29E+01	4,65E-01	9,37E+04	3,38E+03	1,38E+04	4,48E+02
^{239,240} Pu	9,39E+00	3,53E-01	9,84E+04	3,53E+03	1,25E+04	4,08E+02
²⁴¹ Am	7,47E+00	2,60E-01	8,73E+04	3,23E+03	1,33E+04	4,55E+02
²⁴⁴ Cm	3,24E+00	1,24E-01	1,22E+04	7,27E+02	2,77E+03	1,18E+02

Determination of ^{36}Cl and ^{129}I in NPP wastes

Destruction with cH_2SO_4 ,
 I_2 formed is trapped by CCl_4 , backextracted as I^- ,
precipitated as AgI , yield by gravimetry,
 HCl is trapped in H_2O , yield by ion chromatography



A set of results for 3 sample types

a unc(a)

Bq/dm ³	Sample1 (normal evaporator concentrate)		Sample2 (alpha-cont./emergency evaporator concentrate)		Sample3 (decontamination solution)	
	a	unc(a)	a	unc(a)	a	unc(a)
³ H	4,00E+05	2,0E+04	2,12E+05	1,1E+04	7,68E+04	3,8E+03
¹⁴ C	1,01E+04	1,01E+03	2,96E+03	2,96E+02	3,48E+02	3,48E+01
³⁶ Cl	3,65E+01	1,26E+01	9,03E+01	3,46E+01	< 1,07E+01	
⁵⁵ Fe	2,44E+05	2,44E+04	9,37E+06	9,36E+05	1,39E+05	2,08E+04
⁶³ Ni	4,41E+04	2,21E+03	1,75E+06	3,49E+05	2,78E+05	5,56E+04
⁹⁰ Sr	4,92E+02	3,32E+01	1,68E+06	2,35E+05	3,04E+05	2,33E+04
⁹³ Zr	3,52E+02	8,89E+00	8,03E-01	2,43E-02	5,06E-02	1,39E-03
⁹⁹ Tc	< 1,67E-01		1,77E+01	5,35E+00	1,06E+01	2,07E+00
^{108m} Ag	1,38E+02	8,81E+00	1,34E+04	7,98E+02	1,32E+02	4,71E+00
¹²⁹ I	1,94E-01	1,75E-02	7,32E-01	8,04E-02	< 2,67E-01	
²³⁸ Pu	1,29E+01	4,65E-01	9,37E+04	3,38E+03	1,38E+04	4,48E+02
^{239,240} Pu	9,39E+00	3,53E-01	9,84E+04	3,53E+03	1,25E+04	4,08E+02
²⁴¹ Am	7,47E+00	2,60E-01	8,73E+04	3,23E+03	1,33E+04	4,55E+02
²⁴⁴ Cm	3,24E+00	1,24E-01	1,22E+04	7,27E+02	2,77E+03	1,18E+02

Determination of ^{108m}Ag and ^{99}Tc in NPP wastes

Concentration of Ag using ultrafiltration



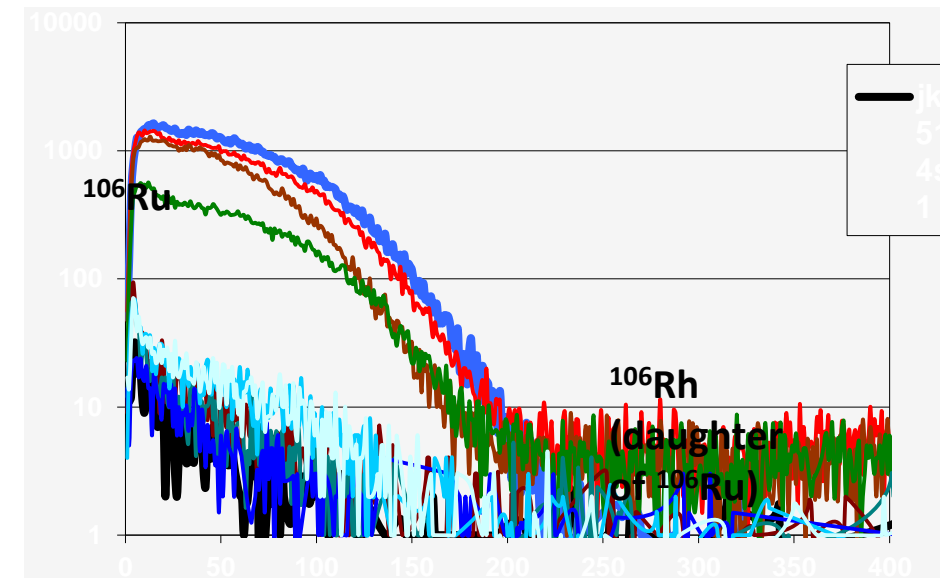
Measurement by gamma spectrometry, e.g.

Isotope	Activity concentration (Bq/dm ³)
^{108m}Ag	$(7,98 \pm 0.21) \text{ E}+02$
^{110m}Ag	$(7,29 \pm 0.06) \text{ E}+03$

Separation of Tc from the Ag permeate: removal of water content, acidic destruction, precipitation with H_2S , purification with cation exchange and extraction chromatography using TEVA resin



Measurement of ^{99}Tc -TEVA using Insta Gel by LSC



A set of results for 3 sample types

a unc(a)

Bq/dm ³	Sample1 (normal evaporator concentrate)		Sample2 (alpha-cont./emergency evaporator concentrate)		Sample3 (decontamination solution)	
³ H	4,00E+05	2,0E+04	2,12E+05	1,1E+04	7,68E+04	3,8E+03
¹⁴ C	1,01E+04	1,01E+03	2,96E+03	2,96E+02	3,48E+02	3,48E+01
³⁶ Cl	3,65E+01	1,26E+01	9,03E+01	3,46E+01	< 1,07E+01	
⁵⁵ Fe	2,44E+05	2,44E+04	9,37E+06	9,36E+05	1,39E+05	2,08E+04
⁶³ Ni	4,41E+04	2,21E+03	1,75E+06	3,49E+05	2,78E+05	5,56E+04
⁹⁰ Sr	4,92E+02	3,32E+01	1,68E+06	2,35E+05	3,04E+05	2,33E+04
⁹³ Zr	3,52E+02	8,89E+00	8,03E-01	2,43E-02	5,06E-02	1,39E-03
⁹⁹ Tc	< 1,67E-01		1,77E+01	5,35E+00	1,06E+01	2,07E+00
^{108m} Ag	1,38E+02	8,81E+00	1,34E+04	7,98E+02	1,32E+02	4,71E+00
¹²⁹ I	1,94E-01	1,75E-02	7,32E-01	8,04E-02	< 2,67E-01	
²³⁸ Pu	1,29E+01	4,65E-01	9,37E+04	3,38E+03	1,38E+04	4,48E+02
^{239,240} Pu	9,39E+00	3,53E-01	9,84E+04	3,53E+03	1,25E+04	4,08E+02
²⁴¹ Am	7,47E+00	2,60E-01	8,73E+04	3,23E+03	1,33E+04	4,55E+02
²⁴⁴ Cm	3,24E+00	1,24E-01	1,22E+04	7,27E+02	2,77E+03	1,18E+02

Methods under development based on ICP-MS/MS measurement



for the sensitive determination of ^{79}Se , ^{107}Pd , ^{129}I , ^{151}Sm , ^{147}Pm

Scaling factors for NPP waste characterization

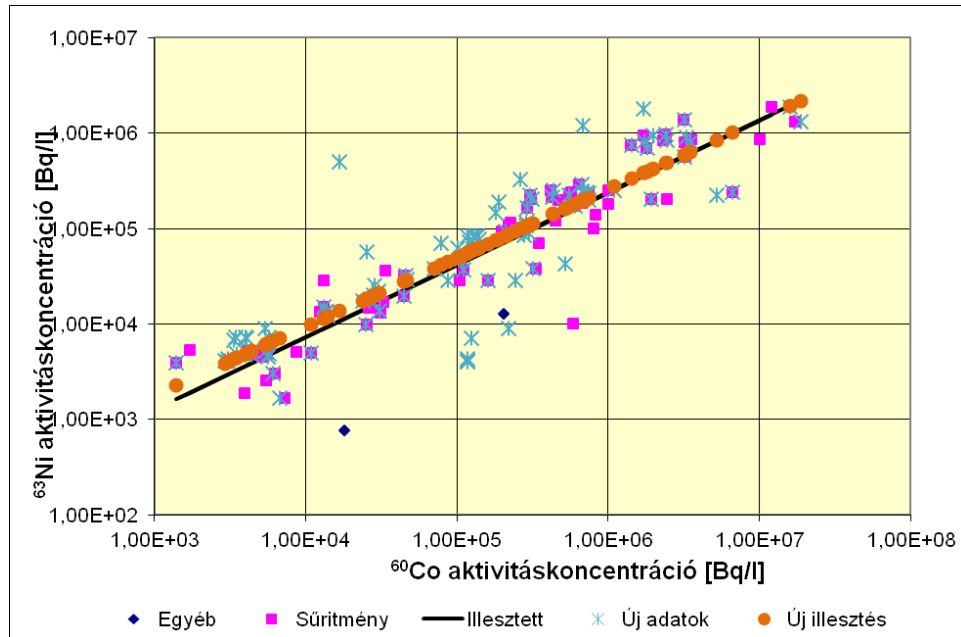
Determination of SF for liquid wastes at NPP Paks

Scaling factors are determined from the analysis of liquid wastes:

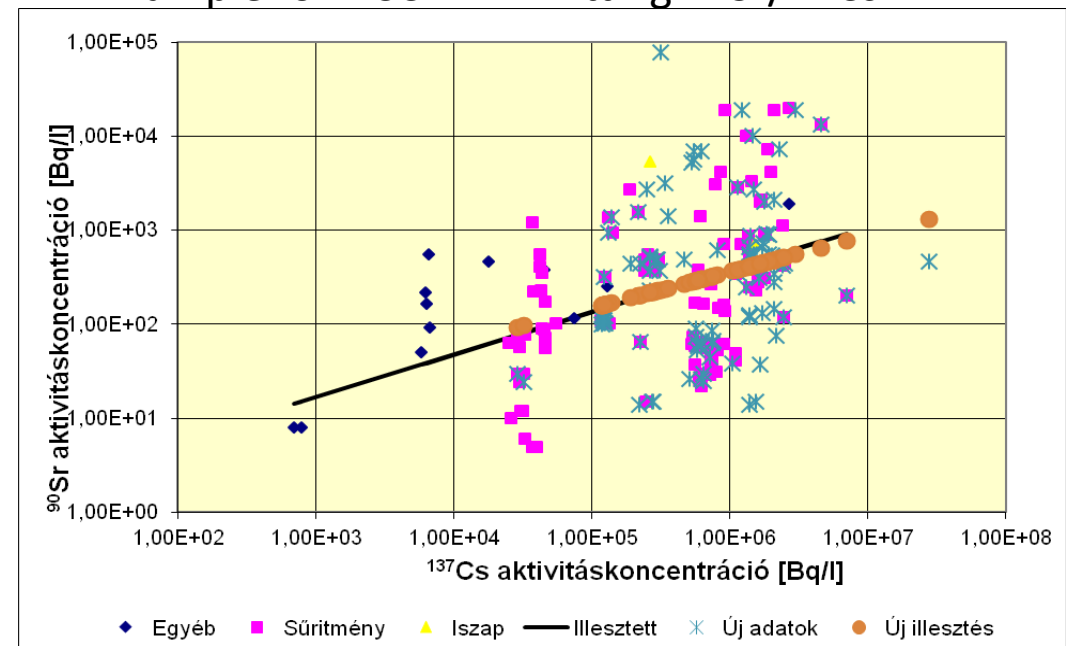
$$C_{DTM} = b * (C_{KN})^m$$

C_{DTM} activity concentration of DTM nuclide
 C_{KN} activity concentration of key nuclide
 m, b parameters

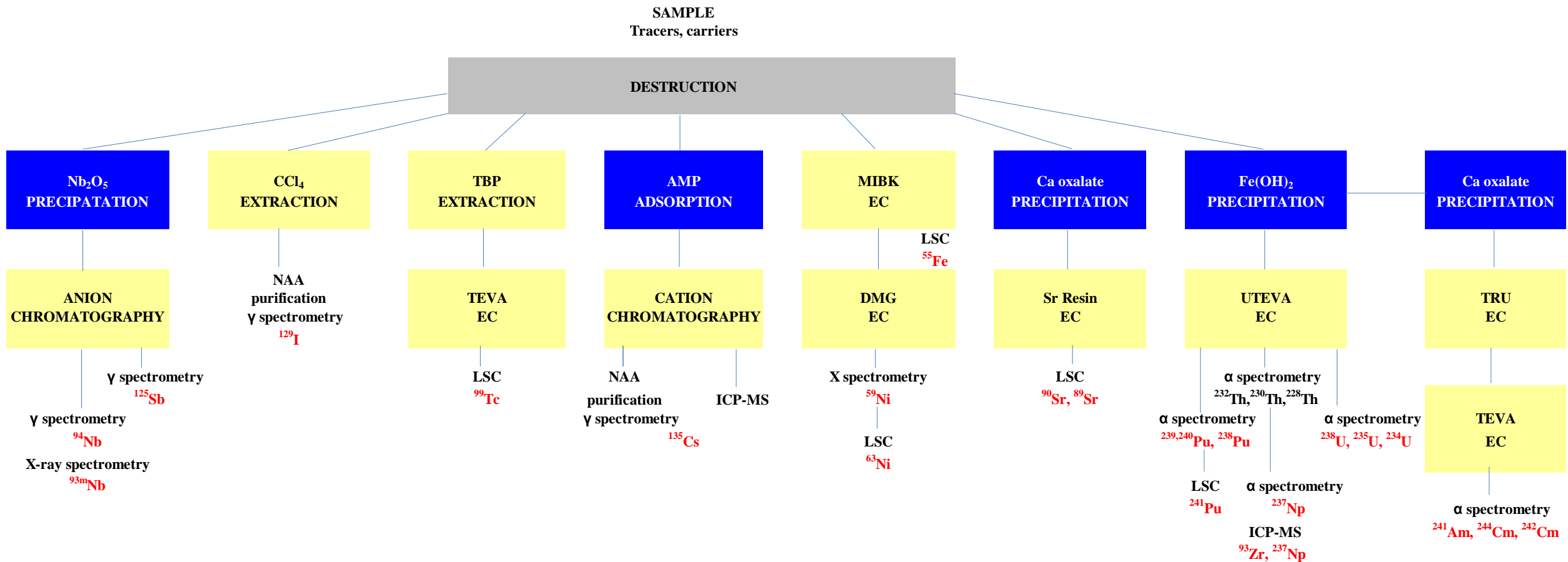
Example for EXCELLENT fitting: $^{63}\text{Ni}/^{60}\text{Co}$

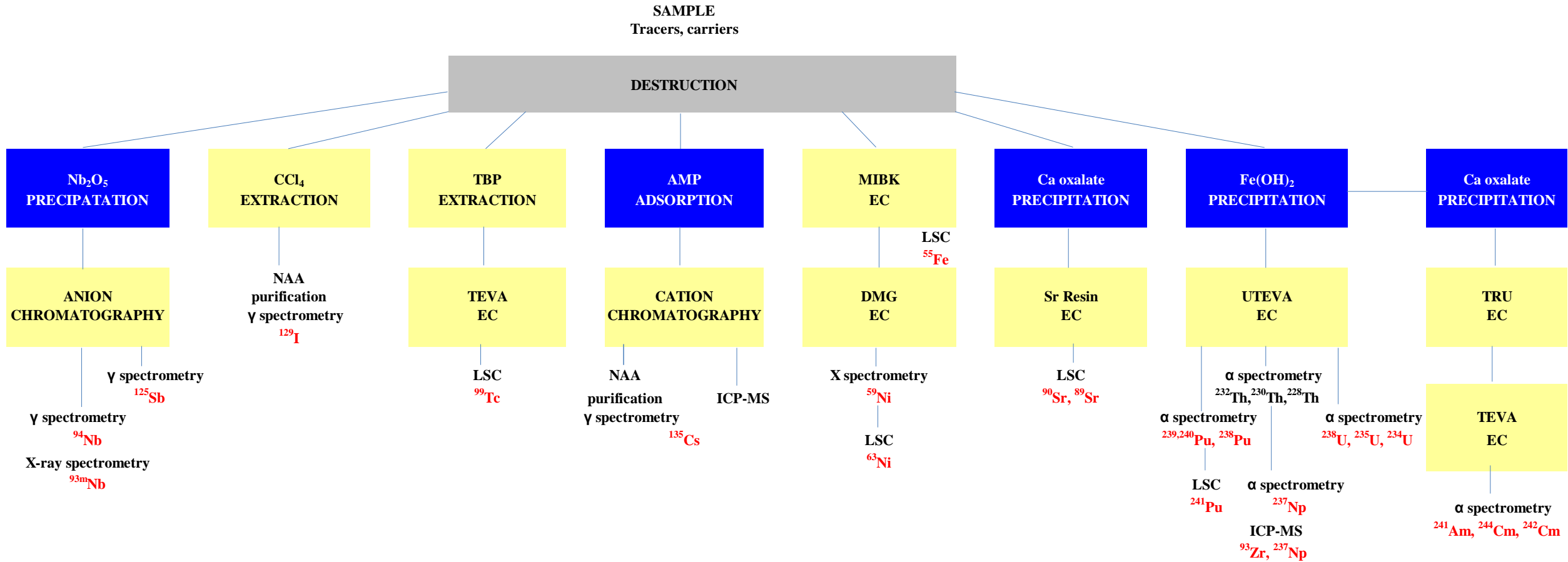


Example for ACCEPTED fitting: $^{90}\text{Sr}/^{137}\text{Cs}$



Instead of a summary...





Thank you for your attention!