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Complex sample preparation using the tritium column

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Aim

The analysis of aqueous samples containing high tritium content whilst enabling subsequent analysis of a range of alpha, beta and gamma fission/activation products.

Tritium

Cs-137 Cs-134 Co-60
Sb-125 Ru-106 Sr90
I-129 Tc-99 Ni-63
Am-241 Pu isotopes Np-237
U isotopes Th isotopes
gross alpha beta

Objectives

- Avoid tritium contamination during radiochemical analysis
- Achieve detection limits across 6 orders of magnitude for more than 7 radioanalytical techniques
- Simplified chemistry to avoid incompatibilities

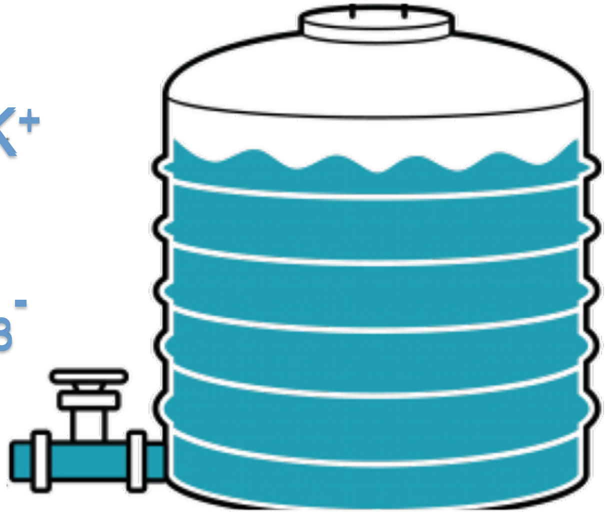
Sample chemistry

Process water sample

- Resulting from chemical treatment
- High cation/anion content
- pH 7-8

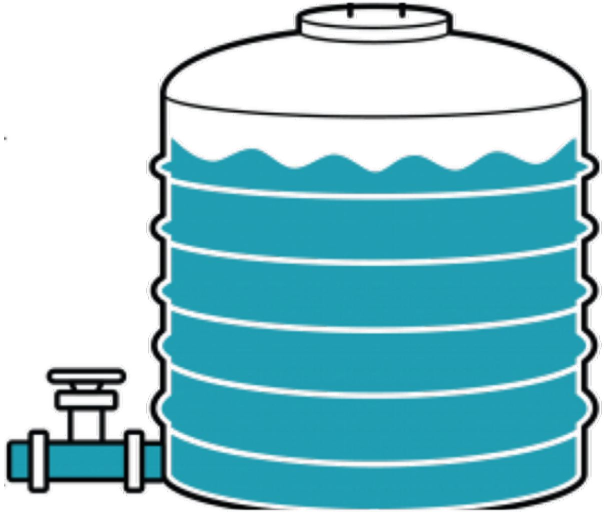
Ca^{+} Mg^{+} Na^{+} K^{+}

HCO_3^{-} Cl^{-} SO_3^{-}



Sample radionuclide content

Tritium H-3	**$1x10^5$ Bq/L**
Gamma emitters (Cs-137, Cs-134, Co-60, Sb-125, Ru-106)	~0.5 Bq/L
Sr-90	~2 Bq/L
I-129	~2 Bq/L
C-14	~10 Bq/L
Ni-63	~2 Bq/L
Tc-99	~1 Bq/L
Gross β	~10 Bq/L
Gross α	~0.05 Bq/L



Measurement Quality Objectives

Radioisotope	MDL Bq/L	Uncertainty %
Tritium H-3	100	<20
Gamma emitters (Cs-137, Cs-134, Co-60, Sb-125, Ru-106)	0.0005	<5
Sr-90	0.02	<5
I-129	0.03	<5
C-14	0.05	<5
Ni-63	0.05	<5
Tc-99	0.02	<5
Gross β	0.1	<25
Gross α	0.06	<25

Analysis procedures

- Liquid scintillation counting
- Gas-flow proportional counter
- Gamma spectrometry
- Alpha spectrometry

Different source preparations
Different interferences

Fit-for-purpose procedure with simple chemistry

Tritium analysis

Proficiency Test sample – Eichrom method

Reported Activity Concentrations

Tritium	29.8 ± 3.5 Bq/L
Sr-90	12.3 ± 2.1 Bq/L
Cs-137	32.6 ± 4.1 Bq/L
Am-241	8.3 ± 1.4 Bq/L

Tritium analysis

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Proficiency test sample – H-3 results

Sample ID	Bq/L	Uncert. k=2	% uncert.		Recovery %
S1	26.0	5.3	20.4		87.3
S2	25.5	5.2	20.6		85.4
S3	32.7	6.2	18.8		110.0
S4	29.3	5.6	19.2		98.3
S5	31.5	5.9	18.6		105.7
AVERAGE	**29.0**	**5.6**	**19.5**		**97.3**
Std Dev	3.2				10.8
Std Dev %	11.1				11.1

Mixed Gamma Experiments

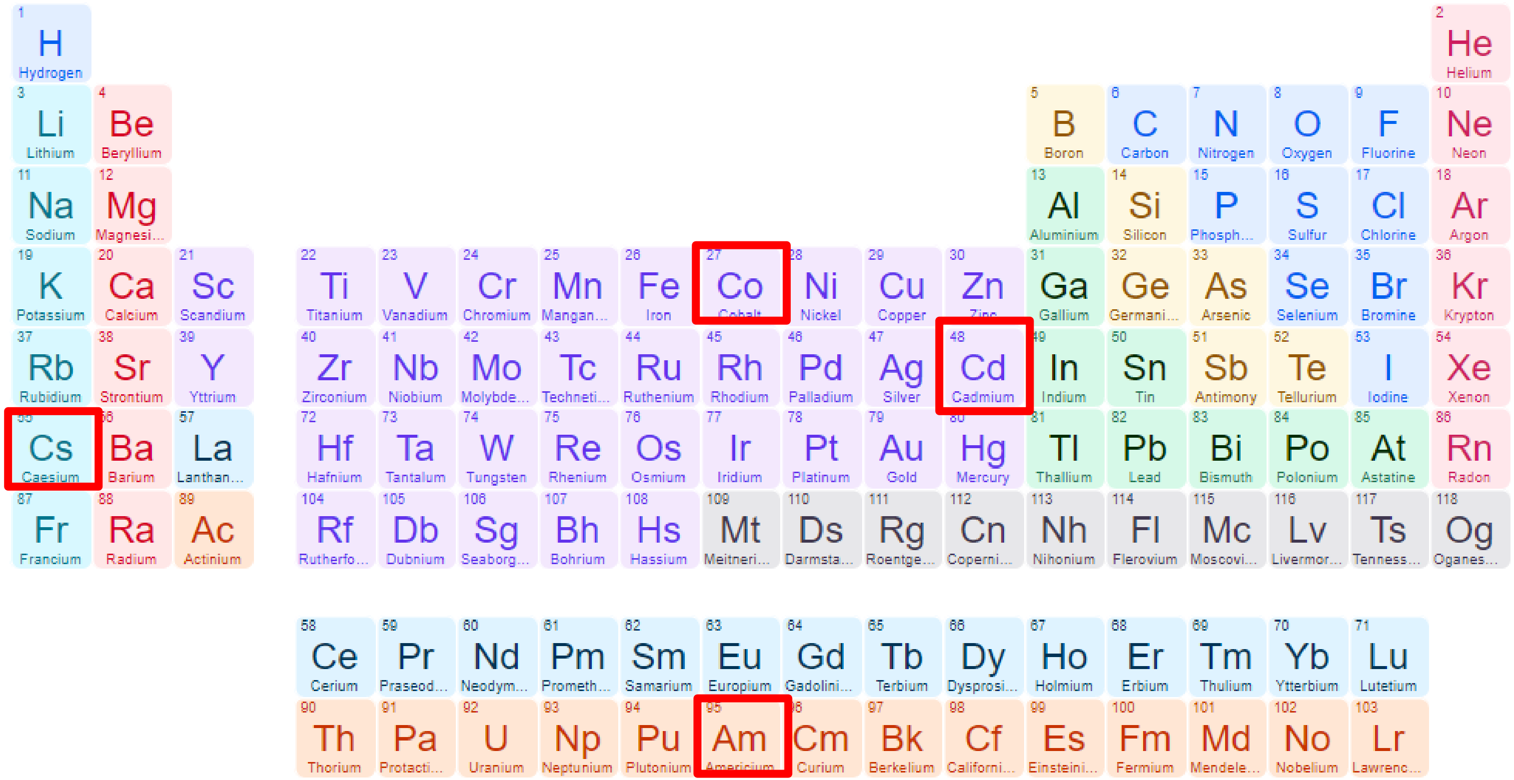
Gamma screening is quick and easy!

Not susceptible to chemical changes like LSC or GFPC!

Mixed gamma sample

Nuclide	Bq/g	Bq/test (25 ml)
Am-241	0.40	10.08
Cd-109	0.22	5.50
Cs-137	0.19	4.68
Co-60	0.15	3.69

Mixed Gamma Experiments



Removal of ‘interferences’

Tritium column resins

- Diphonix® (Monophos) targeting cations
- AG 1x8, Cl^- form targeting anions
- Polymethacrylate resin targeting organic molecules

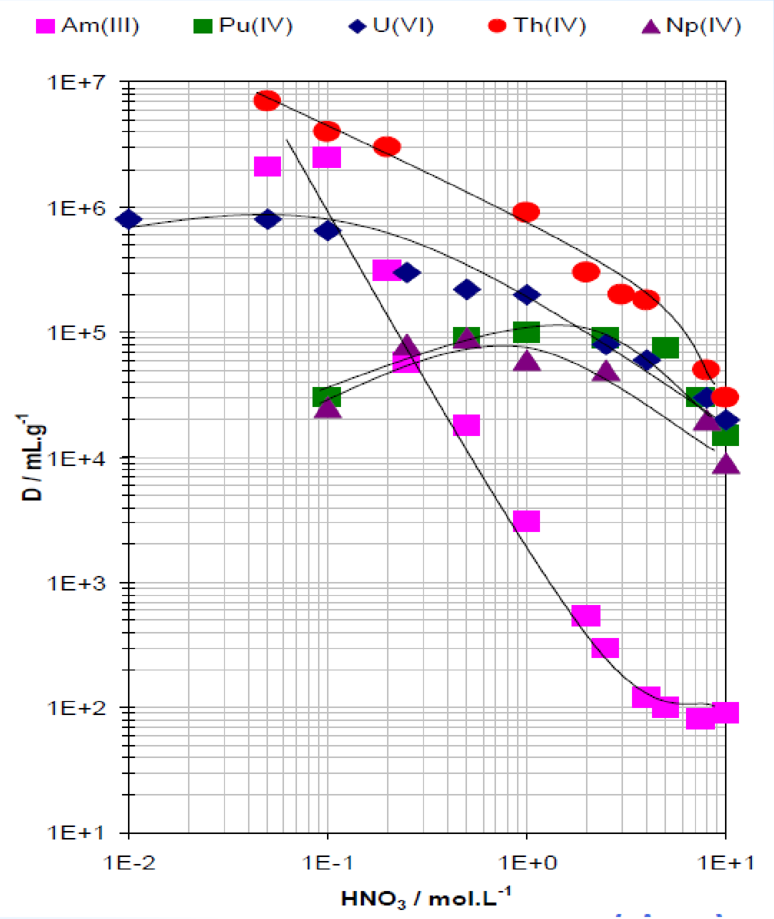
Radioisotope cations adsorbed to Diphonix (Monophos)

Elution from chelating ion exchange resin requires redox reaction or excess reactive reagent.

Potential elutions

Diphonix resin Kd

- Nitric acid
- Phosphoric acid
- Chelating ligand?



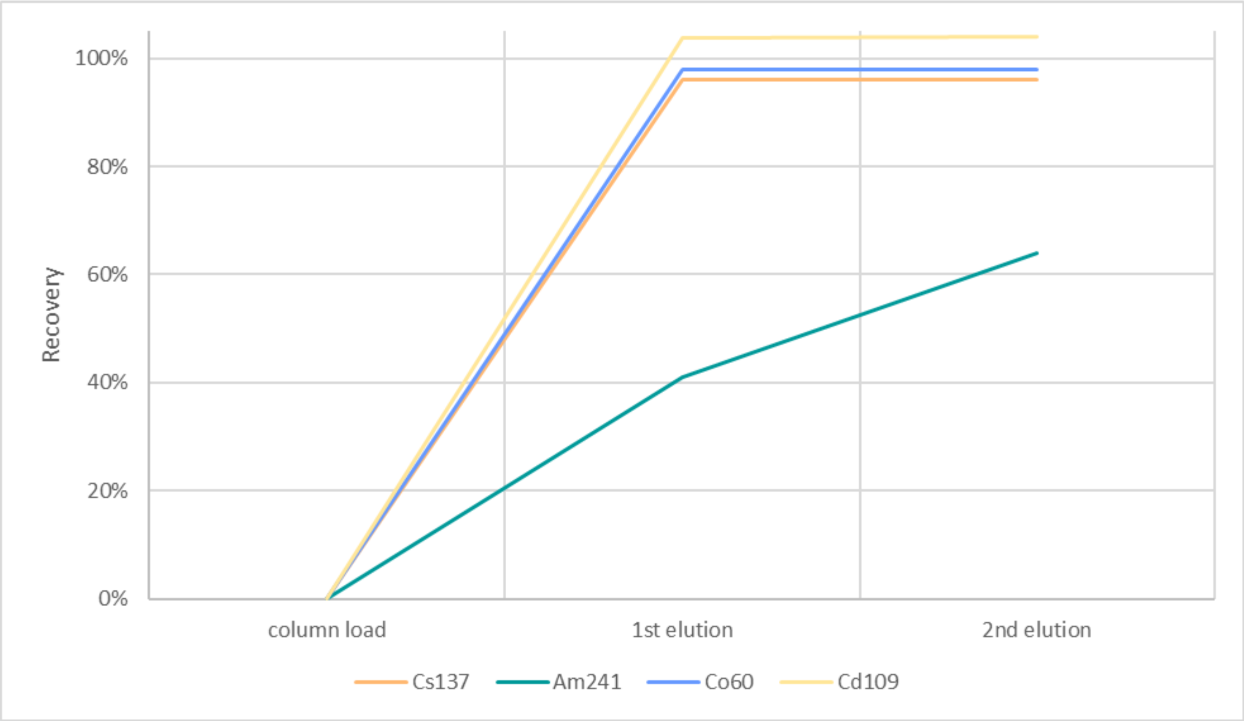
HNO_3 elution

1. 25 mL load solution
2. 15 mL 9M HNO_3

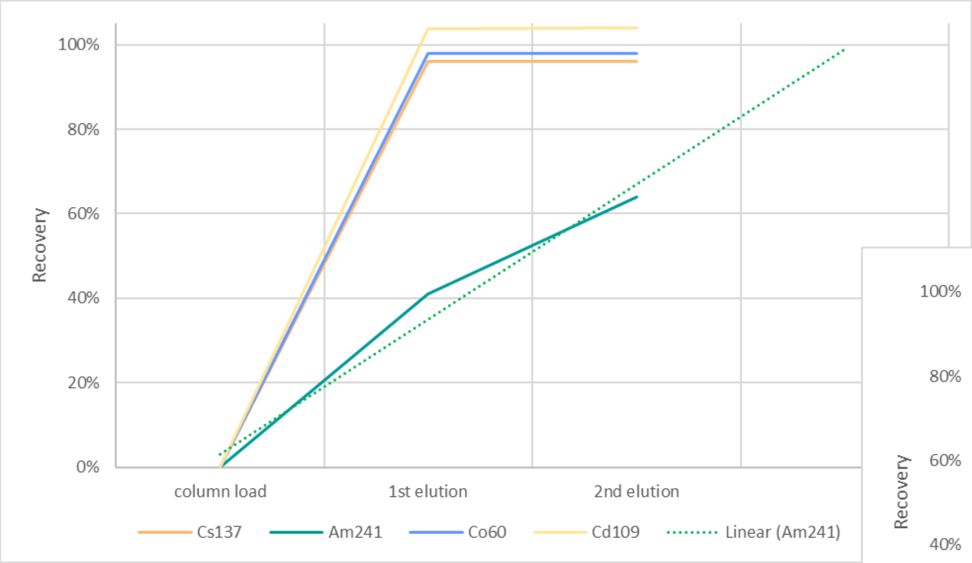
Radioisotope	Column Load CPS	1st elution CPS	Recovery
Cs-137	0.000	0.1678	97%
Am-241	**0.000**	**0.2250**	**41%**
Co-60	0.000	0.0880	97%
Cd-109	0.000	0.0828	104%

2nd HNO_3 elution

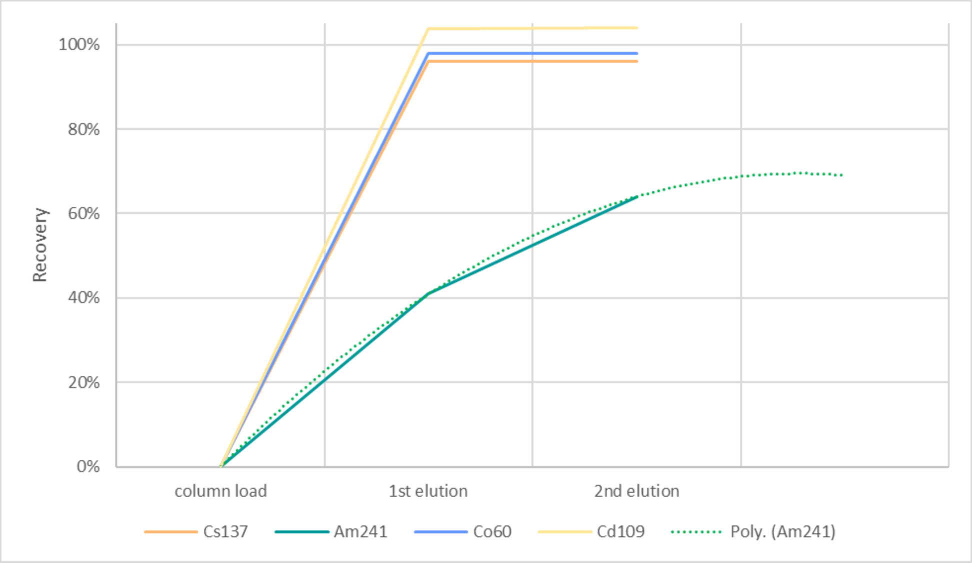
1. 25 mL load solution
2. 15 mL 9M
3. 15 mL 9M



More HNO_3 elutions?



Could it be this simple?



Preliminary conclusions

Main points:

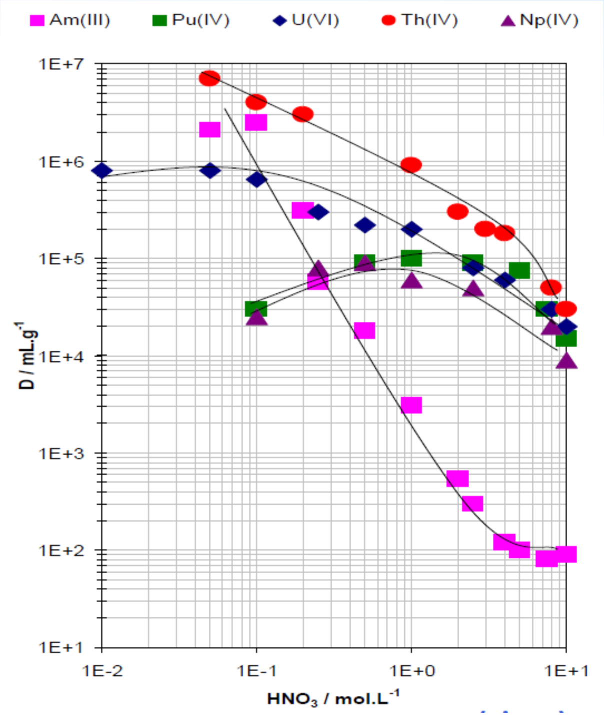
- Cs-137 recovery was acceptable at 97%
- Co-60 recovery was acceptable at 97%
- Cd-109 recovery was acceptable at 104%
- Am-241 was 41-54% with an initial elution of 15ml 9M HNO_3
- An additional 15ml of 9M HNO_3 resulted in an overall recovery of 56-64%

The combination of Diphonix/Monophos and AG 1x8 resins is an effective sample pre-treatment for the separation of tritium from remaining fission/activation products.

Further work is required to determine the acceptable and repeatable recovery for actinides (Am-241).

Further work

- Testing with actinides of more complicated chemistry
- Investigation of oxidizing acids e.g. H_3PO_4
- Investigation of 'light' complexing agents e.g. oxalic acid



Further work

- Development of a QC sample for monitoring the efficiency of elutions
- Testing compatibility with subsequent radiochemical analyses e.g. Sr-90, Ni-63, I-129, Tc-99
- Repeatability and robustness testing for complete validation

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Thank you!