Decontamination of high level activity solutions from cesium and strontium by chromatography

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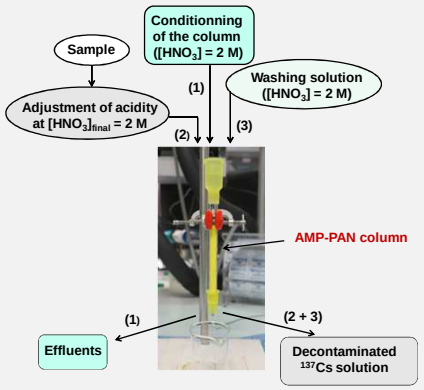
Introduction : High rate of irradiation of certain samples resulting from various programs carried out in ATALANTE facility is essentially due to the presence of ^{137}Cs and ^{90}Sr. The elimination of these radionuclides would make it possible to handle these samples in glove boxes without having to carry out important dilution beforehand. It would allow to analyze these samples by techniques usually implemented in glove boxes (ICP, alpha spectrometry, ...) and to reach lower detection limits. Tests were carried out with real samples by using extraction chromatography as separative technique.

Literature data

AMP-PAN: Inorganic powder of Ammonium Phosphomolybdate embedded in an organic matrix based on PolyAcrylnitrile
Kd(Cs) > 1000 mL/g ($[HNO_3]$ = 1-2M), capacity: 30 mg of Cs/g
density: 0.27 g/mL, Radiation resistance: 10^6 Gray

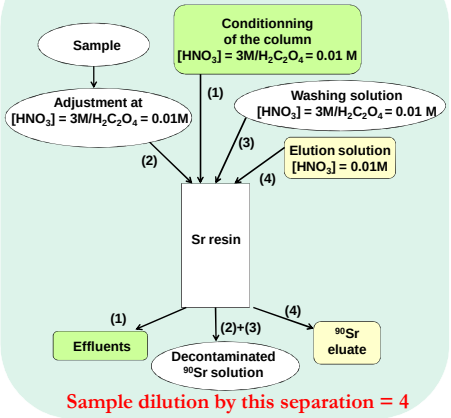
Sr resin : 1.0M 4,4'(5')-di-t-butylcyclohexano 18-crown-6) in 1-octanol 40% (w/w) loaded onto an inert chromatographic support.
Kd(Sr) ~ 100 mL/g ($[HNO_3]$ = 2-4M), capacity: 30 mg of Sr/g
density: 0.35 g/mL,

^{137}Cs separation scheme



Sample dilution by this separation = 4

^{90}Sr separation scheme



Sample dilution by this separation = 4

Conséquences of ^{137}Cs decontamination

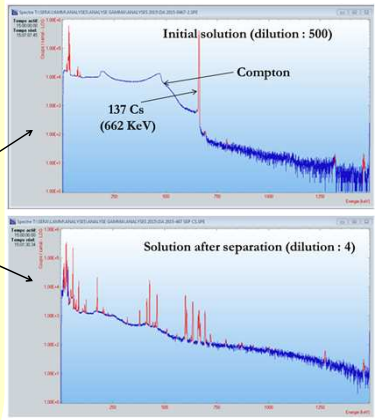
Sample tested: A(^{137}Cs) = 2.10^{10} Bq/L (dissolution solution of residues from a storage tank of fission products)

Alpha analysis: The detection limit (DL) is decreased by a factor > 100 thanks to a smaller dilution of the decontaminated sample (dilution = 4) (Initial solution with Cs: dilution 500). This high DL enhancement is also due to a weak salt concentration (Na^+ < 4 g/L) and a weak total alpha activity (5.10^7 Bq/L) of the decontaminated sample.

Gamma analysis: Elimination of the ^{137}Cs Compton effect, Detection limit improvement : 20-50

ICP-MS: The detection limit (DL) is decreased by a factor > 10 thanks to a smaller dilution of the decontaminated sample (dilution = 4) (Initial solution: 50-fold dilution).

ICP-AES: Same DL between the initial solution measured by ICP-AES in a hot cell and the decontaminated sample measured by ICP-AES in a glove box laboratory.



^{137}Cs separation performances

Initial solution : dissolution solution of residues from a storage tank of fission products
^{137}Cs decontamination factor > 5.10^4
Recovey yields: ^{241}Am 80-100%, Pu : 94 %, Mo : 96%, Zr : 97%
B : 90 %

Validation of recovery yield on a inactive solution
Initial inactive solution : each cation = 50 mg/L, $[HNO_3]$ = 2M

Elements	Recovery yield (%)	Analytical techniques
Al, As B, Ba, Be, Ca, Cd, Ce, Co, Cr, Cu, Fe, Gd, Hg, K, Mg, Mn, Na, Nd, Ni, Pb, Rh, Ru, Sb, Se, Si, Sr, Y, Te, Zn, Zr	> 98	ICP-AES
Ag, Pd	< 5%	
Cs	< 0,04	ICP-MS

High recovery yields are obtained except for Ag and Pd
Mo Recovery yield > 160 % : release of Mo by AMP-PAN ?

Sr separation performances

Sample tested: solution of fuel reprocessing

Sample (V = 2 mL)		Mass balance	
Element X	Activity (Bq/L) or [X] (mg/L)	$X_{(**)}/X_{sample}$ (%)	X_{Eluat}/X_{sample} (%)
^{137}Cs	1.1 10^{12}	95	0.04
^{134}Cs	1.3 10^{11}	94	0.03
^{106}Ru	3.03 10^{10}	87	1.8
^{154}Eu	2.26 10^{10}	119	< 0,5
^{155}Eu	1.04 10^{10}	94	1.06
^{241}Am	2.90 10^{10}	92	< 0.05
Sr	210	*	101.7
U	244000	*	1.9
Fe	190	*	< 3.3
Mo	1139	*	< 0.6
Zr	1080	*	< 1.2
Pd	345	*	< 3.7
Ru	655	*	< 1.0
Tc	256	*	1.3

* not measured **(Effluent feeding + washing)

Initial inactive solution : each cation = 50 mg/L

Elements	$[X]_{(**)}/X_{sample}$ (%)	$[X]_{(eluate)}/X_{sample}$ (%)
Al, As, B, Be, Ca, Cd, Ce, Co, Cs, Cr, Cu, Fe, Gd, Mg, Mn, Mo, Na, Nd, Ni, Rh, Ru, Pd, Pr, Se, Si, Y, Te, Zn, Zr	90-100	DL
Ag, Ba, Hg, K, Pb	DL	DL
Sr	DL	94

DL : detection limit (**) Effluent feeding + washing

- Selective and quantitative Sr recovery in the eluate
- High recovery yield of other elements in the effluent feeding and washing solutions except for Ag, Ba, Hg, K and Pb

Conclusions

Cs separation on AMP-PAN

- Very good selectivity of AMP-PAN
  - . Cs decontamination factor > 5.10^4
    → decrease of the irradiation rate of the sample
  - . High recovery yield for most cations (except Pd and Ag) (behavior of Mo to be confirmed)

- Consequences of the ^{137}Cs separation on sample analyses
  - . Decrease of Detection Limits in gamma spectrometry (because of the minimization of the Cs Compton effect)
  - . Decrease of Detection Limits in ICP-MS and alpha spectrometry (because of a smaller dilution of the decontaminated sample))

- Possible applications
  - . Samples from processes of hydrometallurgical extraction, from dissolution of hulls and spent fuels, radioactive effluent

Sr separation on Sr resin : Very encouraging results to be consolidated with other samples (dissolution of hulls and spent fuels, radioactive effluent, dissolution solution of residues from a storage tank of fission products)
On going studies : Optimization of ^{90}Sr separation by chromatography using Sr resin (breakthrough curve, washing solution volume, ...)
Development of the liquid or solid scintillation analytical technique for ^{90}Sr measurement