Influence of the decontamination of a high radioactive solution from Cesium on analytical performances

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Introduction : In the ATALANTE facility dedicated to the back end nuclear fuel cycle R&D, the high rate of irradiation of certain samples is essentially due to the presence of ^{137}Cs. It results several advantages to decontaminate them from this isotope: 1- a decrease of the irradiation rate, 2- a smaller dilution to make possible the samples manipulation in glove boxes, 3- lower detection limits of analyzed elements. According to the literature, a separation by chromatography using AMP-PAN is a good method to achieve our goals: a high decontamination factor of ^{137}Cs, a high recovery yield for other elements, a small dilution factor of the sample.

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)

AMP-PAN capacity: 30 mg de Cs/g

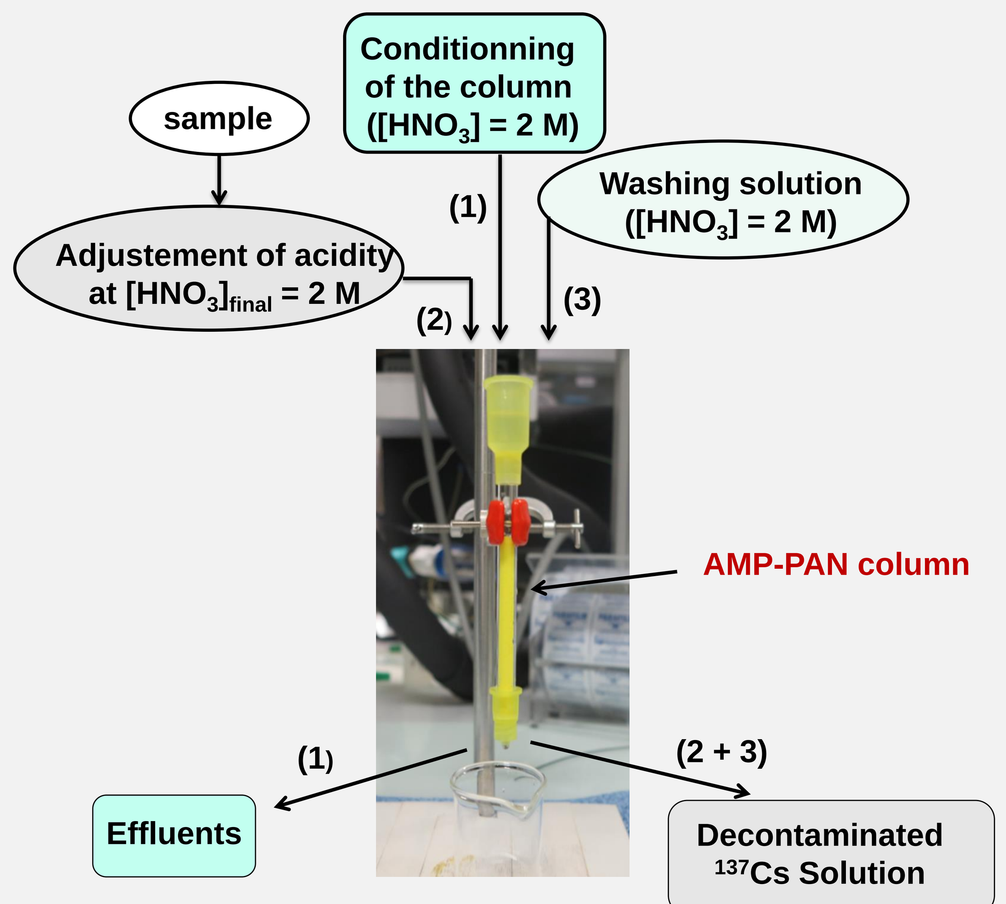
Density: 0.27 g/mL

Radiation resistance: 10^6 Gray

Cationic exchange:

$$(NH_4)_3PMo_{12}O_{40} + 3CsNO_3 \rightarrow Cs_3PMo_{12}O_{40} + 3NH_4NO_3$$

^{137}Cs separation scheme



Sample dilution by this separation = 4

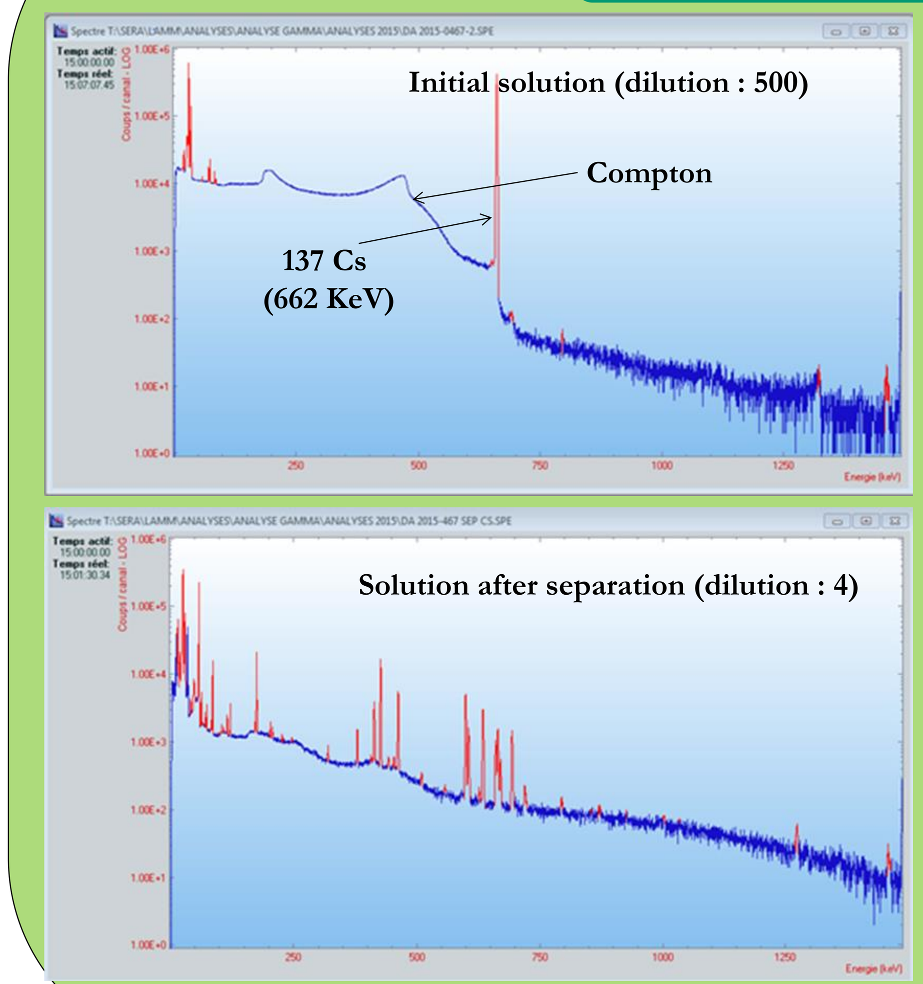
Alpha emitters Analysis

Sample tested: dissolution solution of residues from a storage tank of fission products
Analysis performed in a glove box laboratory

Element	Initial solution	Recovery yield (%)
Pu total	2.5 mg/L	94 +/- 10

- High recovery yield of plutonium
- 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.

Analyses of gamma emitters



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

Consequences of separation:

- Elimination of the Compton effect
- Detection limit improvement (*): 20-50
- ^{137}Cs decontamination factor > 5.10^4
- ^{241}Am recovey yield: 80-100%

(*) measured elements: ^{54}Mn, ^{60}Co, ^{65}Zn, ^{95}Zr, ^{95}Nb, ^{106}(Ru + Rh), ^{110}Ag, ^{144}(Ce + Pr), ^{152}Eu

Multi-elementary analyses by ICP-AES and ICP-MS

Sample tested: dissolution solution of residues from a storage tank of fission products
Analysis performed in a glove box laboratory

Element	Initial solution (mg/L)	Recovery yield (%)
Mo	355	96
Zr	404	97
B	555	90

- High recovery yield of Mo, Zr and B
- 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.

Behavior of some cations with AMP-PAN

- Goal : confirmation of the selectivity of Cs in presence of some cations (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	ICPAES
Ag, Pd	< 5%	
Cs	< 0,04	ICP-MS

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

Influence of Chlorides, Fluorides and Sulfates on Cs behavior

- AMP-PAN feeding inactive solution: Cs = 25 mg/L, $[HNO_3]$ = 2 M, [NaX] = 1 to 1.5 M

[X] feeding solution	Cesium (µg/L) effluents (feeding + washing)	Cesium recovery yield (%)	Analytical technique
[F-] = 1 M	< 10 (DL)	< 0.08	ICP-MS
$[SO_4^{2-}]$ = 1.5 M	< 10 (DL)	< 0.08	
[Cl-] = 1.5 M	< 10 (DL)	< 0.08	
no added anion	< 10 (DL)	< 0.08	

- Conclusion: Cesium is strongly fixed by AMP-PAN.
  Anions like Cl^-, F^-, SO_4^{2-} have no influence on Cesium behavior

Conclusions

- 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 separation using AMP-PAN on sample analyses
  . Decrease of Detection Limits in gamma spectrometry (because of the minimization of the 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