Implementation of americium recycling demonstration from spent nuclear fuel in the highlevel shielded process line CBP in the ATALANTE facility

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DE LA RECHERCHE À L'INDUSTRIE

Implementation of americium recycling demonstration from spent nuclear fuel in the high-level shielded process line CBP in the ATALANTE facility

S. Costenoble, M.-J. Bollesteros, F. Antegnard, J.-N. Calor, V. Vanel, M. Montuir, C. Sorel, D. Espinoux, A. Juvenelle

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10th OCTOBER 2016

www.cea.fr

INTRODUCTION

Study context

- As part of the French Act of June 2006 on sustainable radioactive and waste management:
  - Investigation to recover minor actinides from spent nuclear fuel for heterogeneous recycling in Generation-IV reactors.

- Americium is, after Pu, the main contributor to residual heat of long term radioactive waste:
  - Am recycling in future nuclear reactors would decrease toxicity and residual heat which determines the waste density within geological repository.

- The experimental demonstration of americium recycling is therefore a key aspect.

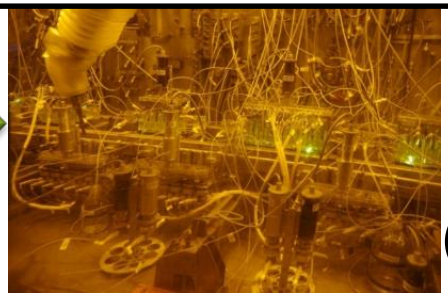
- This demonstration is currently performed in a high-level shielded line called CBP in the ATALANTE facility (CEA Marcoule – France) following different steps.

THE DIFFERENT STEPS OF THE DEMONSTRATION

CBP

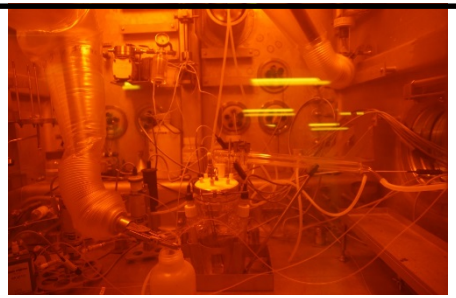
1

Dissolution of spent nuclear fuel and filtration of the solution



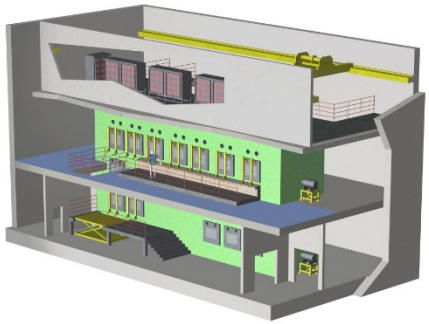
2

PUREX liquid-liquid extraction experiments



3

PUREX raffinate concentration by steam distillation



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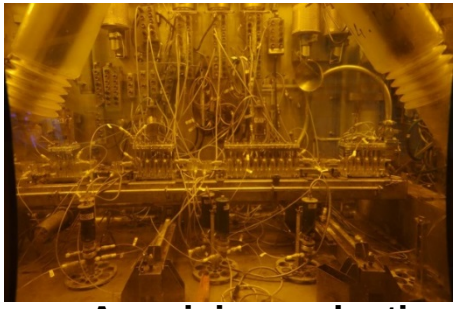
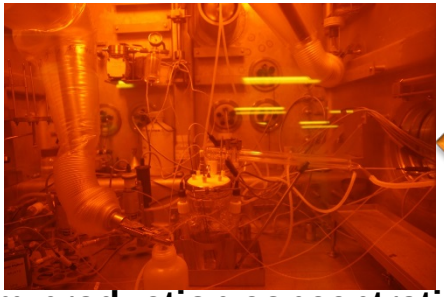


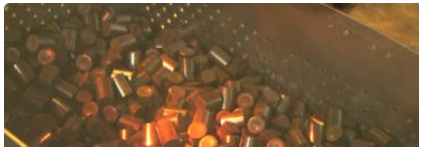
Americium selective recovery



Am production concentration

ATALANTE

U, Am coconversion



$(U,Am)O_2$ pellets production

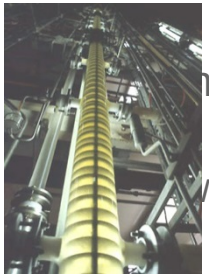
PRESENTATION OF THE PROCESS SHIELDED LINE CBP

Biological shielding of 970 mm concrete and 30 mm of steel.

nts with rem manipulators.

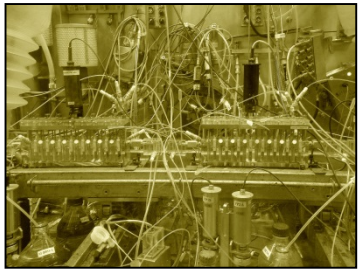
with one me leaded glass.

- Liquid/liquid contactors



Pulsed column - 5,10 m

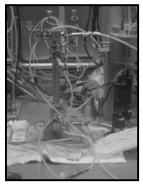
Centrifugal contactors + 0,00 m



Mixer settlers

- Dissolution and filtration

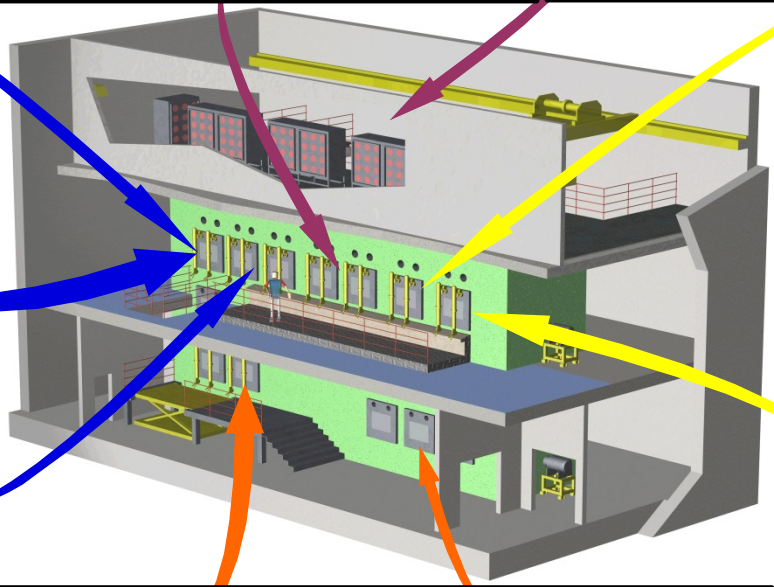
8 or 16 L batch dissolution



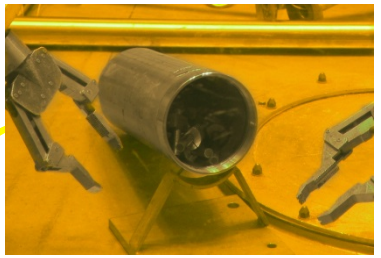
Cross-flow filtration

- Glove boxes for inactive reagents

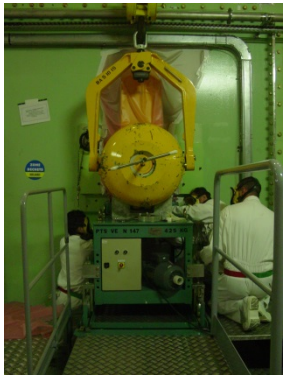
+ 3,80 m



- Spent fuel reception and storage



Fuel reception



PADIRAC connexion

- 5,10 m

- Radwaste packaging
- Reserved for future studies

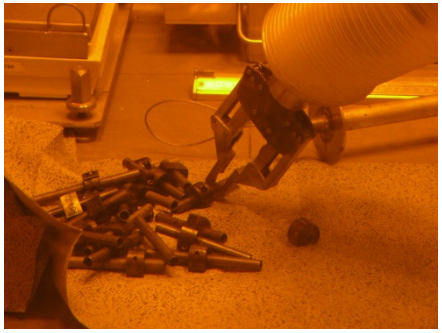
1st STEP: DISSOLUTION AND CLARIFICATION

Nature of the spent nuclear fuel

- 4kg (U+Pu) coming from different reactors of EDF's French plant units.

- UOx fuels (65% mass):
  - Burnup between 30000 and 70000 MWd/t.
  - Enrichment in the range 4-5% ^{235}U.
  - 3-10 years of cooling.

- MOX fuels (35% mass):
  - Burnup between 50000 and 60000 MWd/t.
  - Pu enrichment in the range of 6-7 %.
  - 3-5 years of cooling.



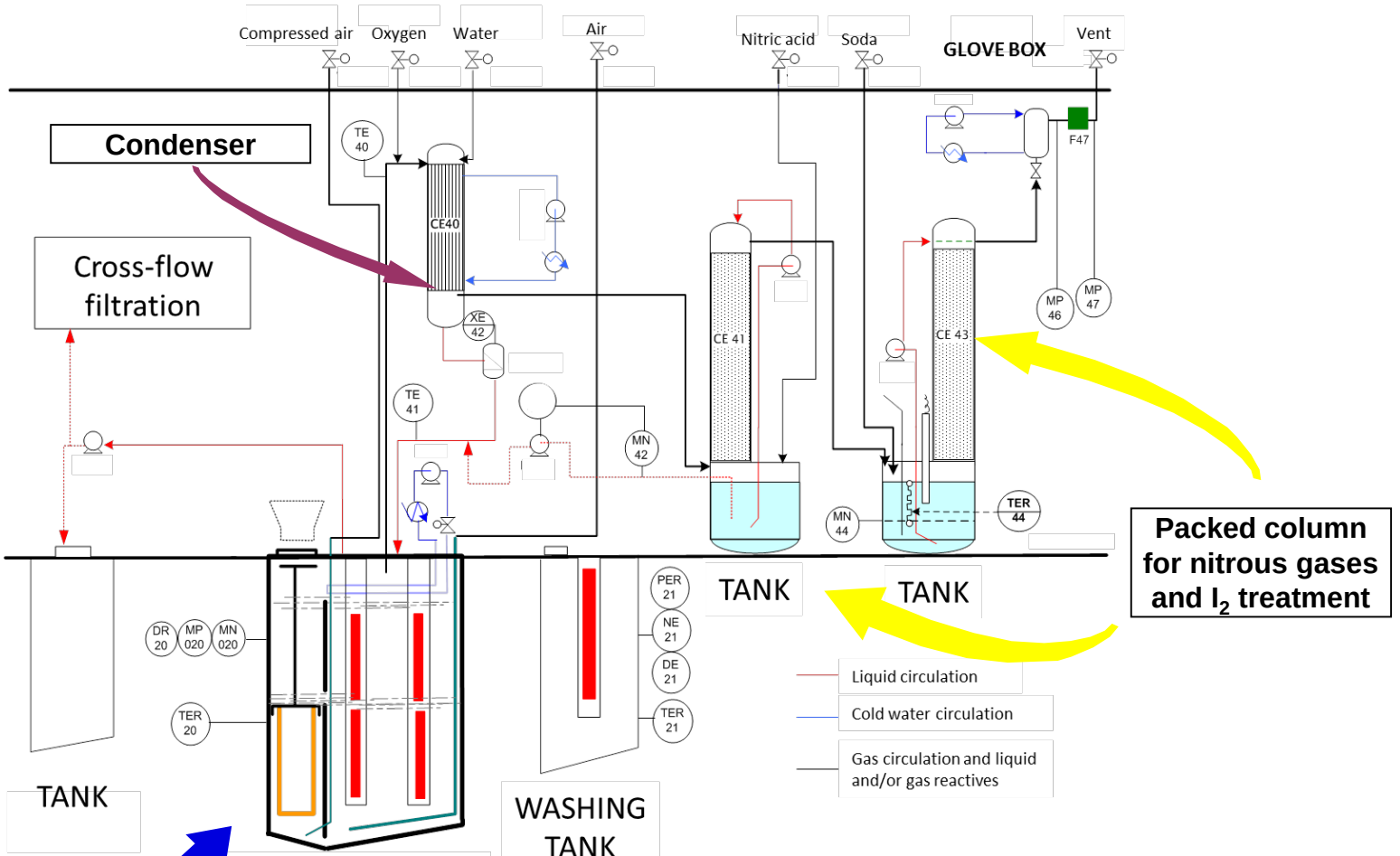
Operating conditions

- Dissolution in boiled nitric acid media with the following reactions.

$$\mathbf{UO_2\ (s)} + 3HNO_3\ (l) \leftrightarrow \mathbf{UO_2(NO_3)_2\ (l)} + 1/2NO\ (g) + 1/2NO_2\ (g) + 3/2H_2O$$

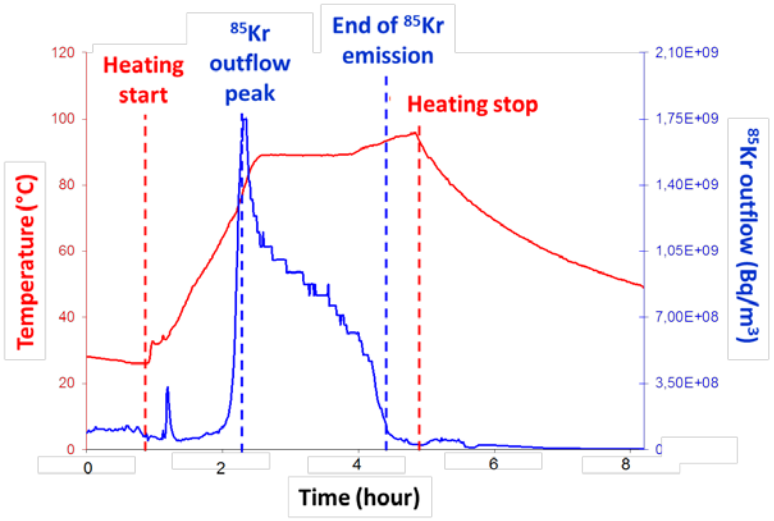
$$\mathbf{PuO_2\ (s)} + 4HNO_3\ (l) \leftrightarrow \mathbf{Pu(NO_3)_4\ (l)} + 2H_2O$$

1st STEP: DISSOLUTION AND CLARIFICATION



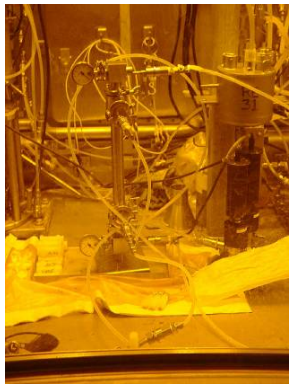
1st STEP: DISSOLUTION AND CLARIFICATION

Monitoring of the process: ^{85}Kr emission



- Kr is trapped into spent nuclear fuel.
- The ^{85}Kr release, measured after gas treatment devices, is controlled during the dissolution.
- The end of emission indicated the end of the reaction.

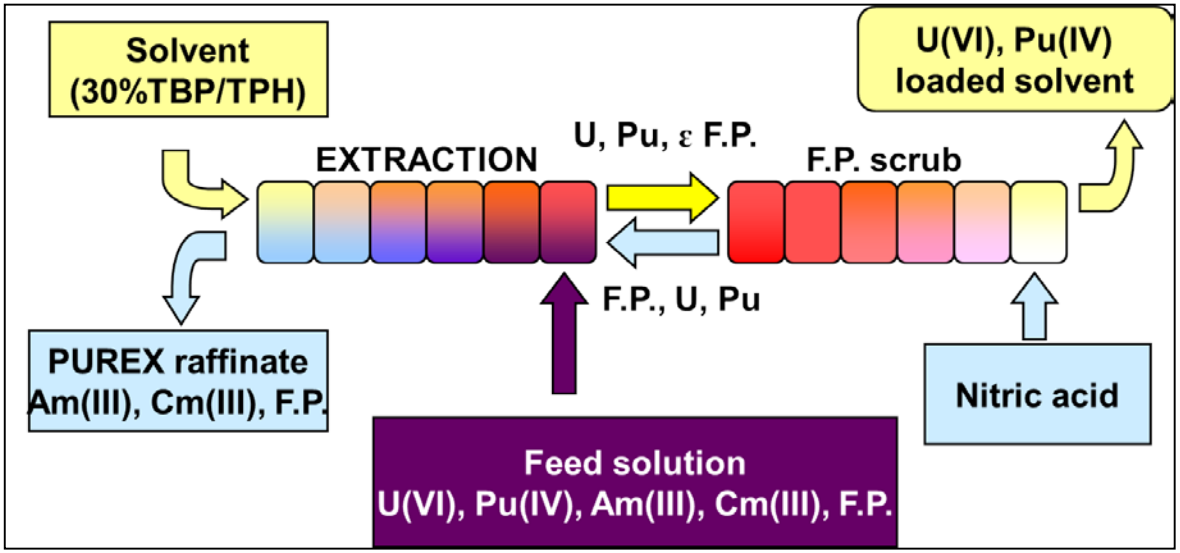
Clarification of the dissolution solutions



- Indispensable step prior to directing them to downstream operations (avoiding any risks of equipment plugging or solvent degradation).
- Performed by tangential filtration through membrane.
- Production of 21L of dissolution solution without fines.
- [U+Pu] = 160 g/L; [H^+] = 4.3 mol/L; [Am] ~ 0.16 g/L; [Cm] ~ 0.05 g/L.

2nd STEP: PUREX LIQUID-LIQUID EXTRACTION EXPERIMENTS

Principle of the process



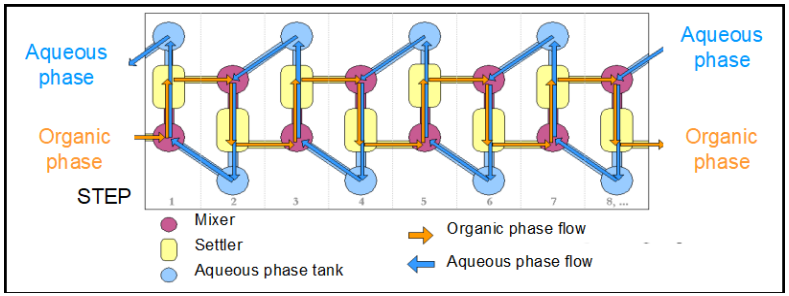
- TBP is very sparingly soluble with an aqueous phase.
- The molecule exhibits much higher affinity for elements in oxidation states +VI (U) and +IV (Pu) and extracts them in the organic phase.
- Extraction of minor actinides and F.P. (+III and +V) is limited.

- The extraction part, performed at 3-4 HNO_3 mol/L, involves two unit operations.
- The scrubbing operation has the purpose of stripping the fission products occuring as impurities in the organic phase.
- The organic phase's preferential affinity for U(VI) and Pu(IV) only operates in the stage in which the aqueous phase is introduced first. The scrubbing operation completes the separation. U and Pu contained in the aqueous phase are sent back to extraction operation.

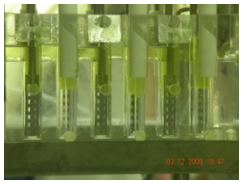
2nd STEP: PUREX LIQUID-LIQUID EXTRACTION EXPERIMENTS

Process devices

- PMMA mixer-settler extractor batteries where the aqueous and the organic phases flow countercurrently.



- Solutions mixed by stainless steel perforated paddle impellers (2000 to 2200 rpm).
- Interphases in settler compartments adjusted with Teflon slides.

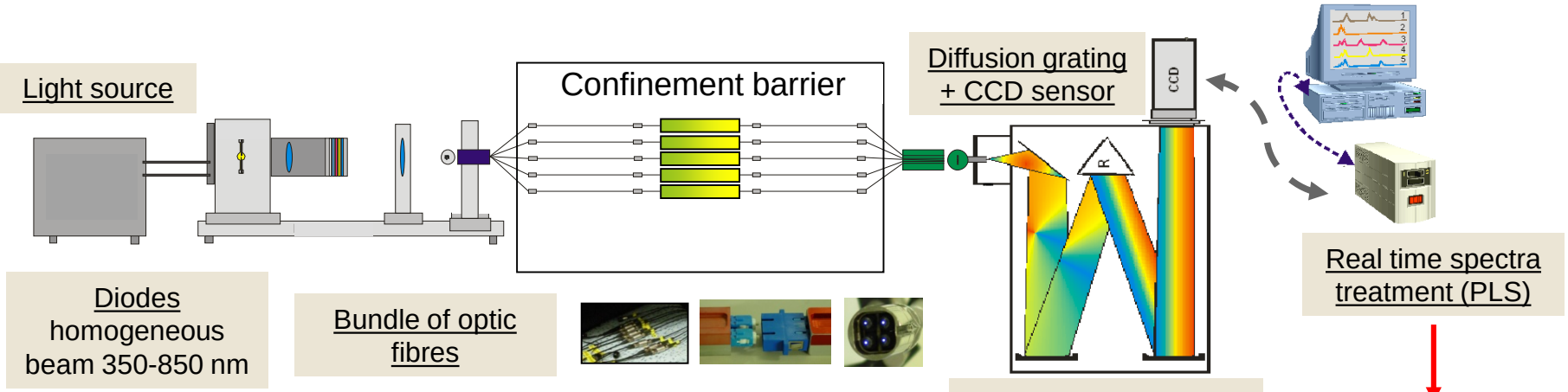


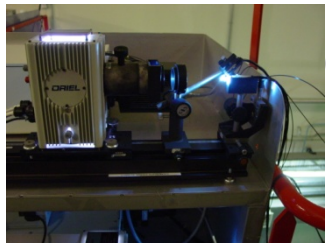
- All reagents are introduced by rotary piston pumps located outside the confinement barrier.
- Flow rate measurements are performed thanks to Coriolis mass flowmeter and controlled by a specific supervision application.

2nd STEP: PUREX LIQUID-LIQUID EXTRACTION EXPERIMENTS

Analytical measurements

- On-line and *in situ* UV-visible spectrophotometric measurements implemented to follow elements (U, Pu, Am…) concentrations in solution.





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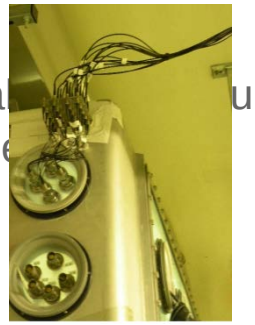
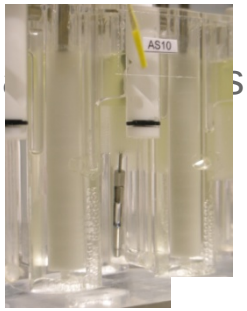
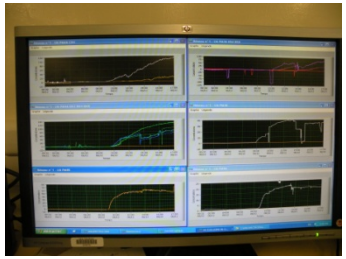
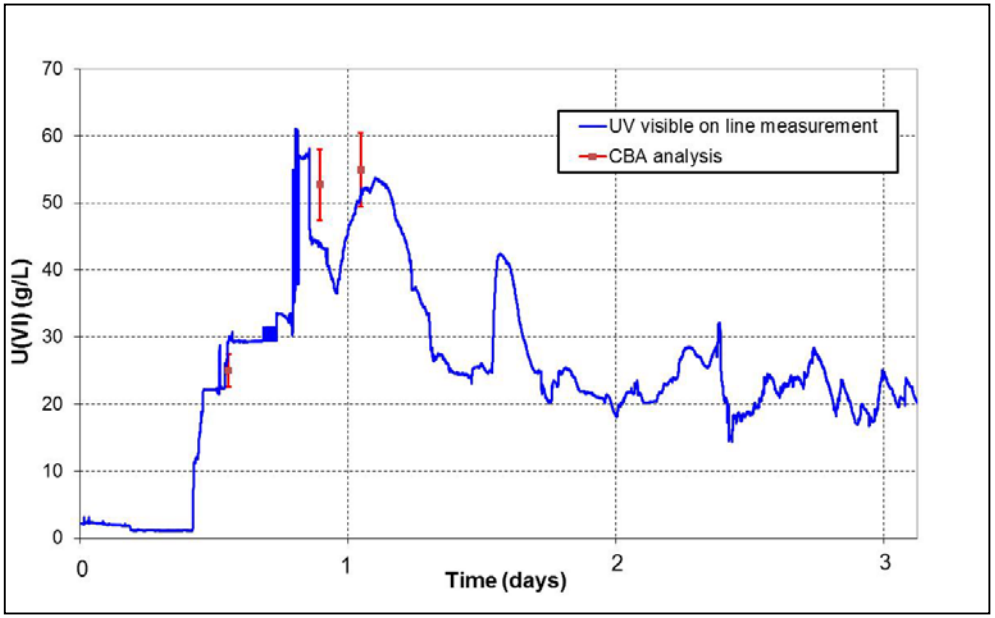






2nd STEP: PUREX LIQUID-LIQUID EXTRACTION EXPERIMENTS

Main results



- Monitoring of the system by U concentration measurement of the aqueous phase coming from F.P. scrubbing mixer settler.
- 24L of PUREX raffinate produced with very low amounts of uranium and plutonium.

[Am] (mg/L)	[Cm] (mg/L)	[H^+] (mol/L)	[Nd] (mg/L)	[Sm] (mg/L)	[Eu] (mg/L)	[Gd] (mg/L)	[Y] (mg/L)	[Mo] (mg/L)	[Zr] (mg/L)
155	18	3,9	740	156	29	38	69	405	290

Main analytical results of the PUREX raffinate

- The solution properties are consistent with the expected results.

3rd STEP: PUREX RAFFINATE CONCENTRATION

Aim of the operation

- The **EXAm process**, which allows the selective and quantitative recovery of americium from a PUREX raffinate by liquid-liquid extraction, was already demonstrated on a genuine PUREX raffinate [1].

- In order to reduce the compacity of the process and future plan associated, the current objective is to adapt this process with a concentrated PUREX raffinate.

Concentration by steam distillation

- This technique has the advantage to increase the concentration of the different elements (Am, Cm, Nd…) while keeping the acidity constant by the release of nitric acid in the distillate.

- The operation is carried out at constant volume; the reactive inflows (PUREX raffinate, water) compensate the distillate outflow (nitric acid).

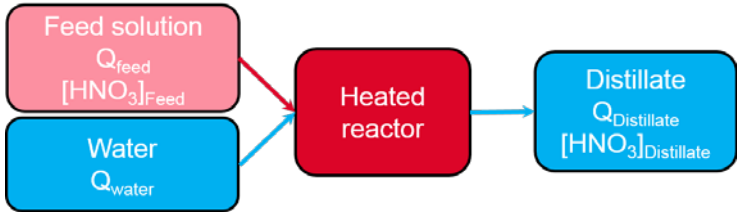
[1] M-J. Bollesteros *et al.* “Implementation of americium separation from a PUREX raffinate”, *Procedia Chemistry*, 7, 2012, 178-183.

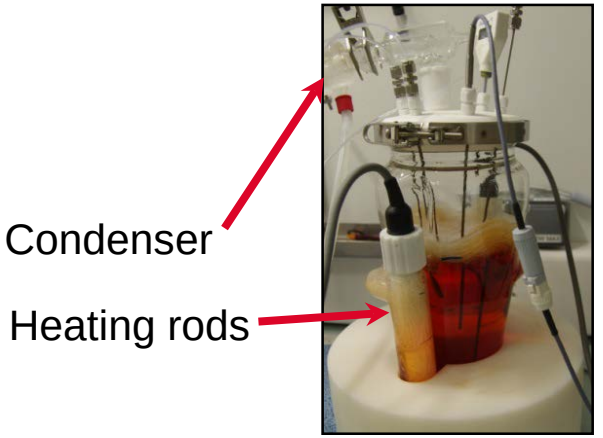
3rd STEP: PUREX RAFFINATE CONCENTRATION

Goals of the steam distillation

- Increase the salt concentrations by a factor of 6.
- Increase the nitric acid concentration around 8 M ($[HNO_3]_{feed\ sol.}$ = 3.9 M).

Principles





- Global mass balance:

$$Q_{Feed} + Q_{Water} = Q_{Distillate}$$

- Nitric acid mass balance:

$$Q_{Feed} = Q_{Distillate} \times \frac{[H^+]_{distillate}}{[H^+]_{Feed}}$$

Measured in given conditions

Initially adjust to 8M

Q_{feed} and Q_{water} determination

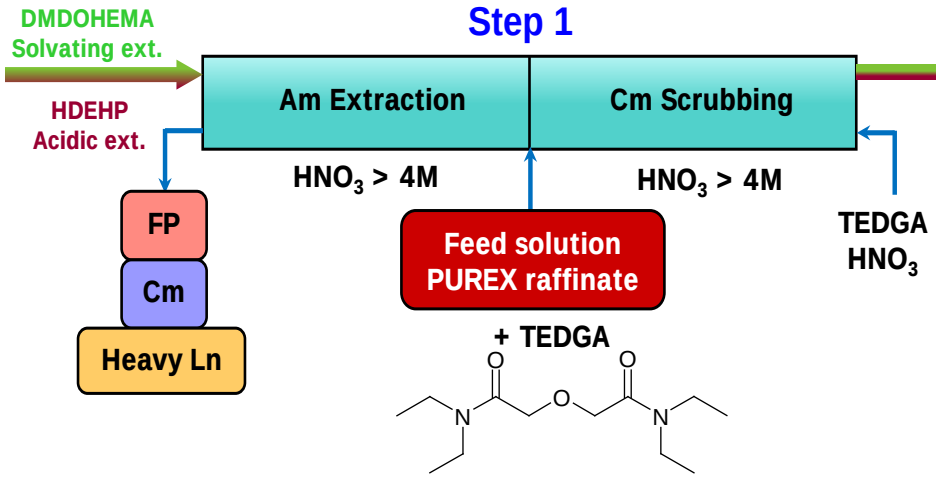
Results

- The final volume of the solution is around 4L. The final nitric acid concentration is measured at 8.2 mol/L.

4th STEP: AMERICIUM SELECTIVE RECOVERY: THE EXAm PROCESS

Flowsheet of the process

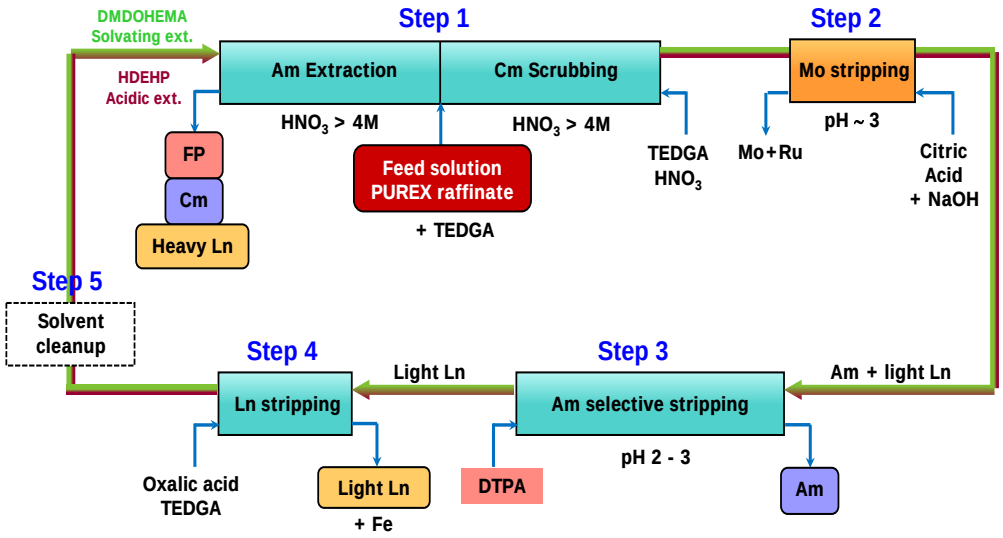
- The separation process used for americium recovery in concentrated conditions was already consolidated with inactive surrogate feed solution and also in alpha conditions with small amounts of americium and curium [2].

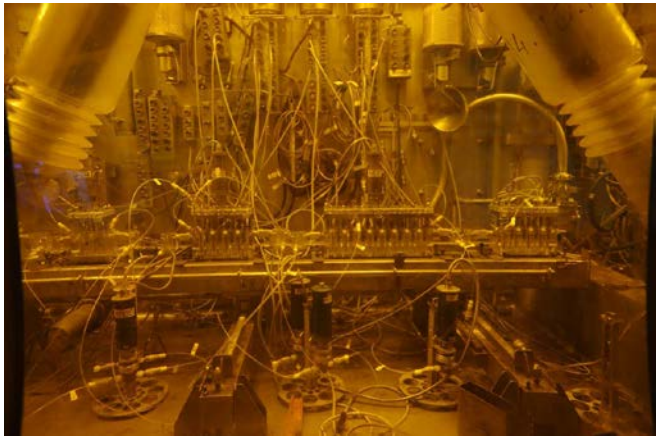


[2] V. Vanel *et al.* "Consolidation of the EXAm process towards the reprocessing of a concentrated PUREX raffinate", *Procedia Chemistry*, in press.

4th STEP: AMERICIUM SELECTIVE RECOVERY: THE EXAm PROCESS

Main results





- **STEP 1**
  - Decontamination factor (Am/Cm) close to 50.
- **STEP 2**
  - Very low americium leak in the Mo+Ru stripping raffinate (less than 0.1%).
- **STEP 3**
  - Decontamination factor (Am/light Ln) equal to 2800 higher than expected (400).
  - **Production of 2.4 g of americium** corresponding to 95.5% of the cations in solution. Impurities are Cm (0.6%), Ln (1.7%) and Fe, Mo, Pd, Ru (2.2%)

CONCLUSION AND PROSPECTS

- Successful demonstration of the americium recycling from spent nuclear fuel in the CBP shielded line.
- After several steps, from spent nuclear fuel dissolution to liquid-liquid extraction experiments (PUREX, EXAm), the aim to produce americium in a selective (without uranium, plutonium, curium, lanthanides, fission products) and quantitative way is attained.
- In order to conclude the demonstration, the americium solution is going to be concentrated before the co-conversion step which will be carried out at the ATALANTE facility.
- The objective of this step is to produce $(U,Am)O_2$ pellets for future irradiation experiments.

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