

Two years of operation of the shielded process line (CBP) in ATALANTE facility

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Abstract

The Atalante shielded process line (CBP) was hot commissioned in December 2003 to reprocess 13 kg of spent UOX fuel with a burnup of $52 \text{ GWd}\cdot\text{t}^{-1}$ after 8 years of cooling. The head-end operations (chopping, dissolution, filtration) were performed in March 2004, producing 48 liters of a solution containing $250 \text{ g}\cdot\text{L}^{-1}$ of U+Pu with a nitric acidity near 4.5 N. The solution was used for a PUREX Np test carried out in April 2005, resulting in quantitative neptunium extraction. The extraction cells were then entirely reconfigured to implement the DIAMEX process for separating the actinides and lanthanides from the other fission products. The PUREX Np extraction raffinate was used to test the DIAMEX flowsheet in November 2005. The PUREX Np and DIAMEX tests contributed toward demonstrating the technical feasibility of enhanced separation in the framework of the French radioactive waste management act of December 30, 1991.

Keywords: *ATALANTE, hot cells, reprocessing studies, partitioning of long-lived radioactive elements*

1. Introduction

Located at Marcoule, in the Rhône Valley, Atalante is one of the CEA's largest nuclear facilities. Mainly devoted to the back-end of the nuclear fuel cycle, this research facility includes several laboratories, one of the most recent of which is the high level shielded process line. Commissioned in 2003, the first program unfolded in the CBP was the demonstration of technical feasibility of minor actinides advanced separation. It is one aspect of the French radioactive waste management act of December 30, 1991 and consists in the implementation and achievement of one PUREX, followed by one DIAMEX and ended by one SANEX processes. Before that, a large amount of spent fuel (about 15 kg) had been received, cut and dissolved in order to make a feed for the PUREX process. All these steps with the used apparatuses are described in this paper.

2. Reception, storage and cutting of spent fuel



Figure 1. TN106 cask connected on the shielded process line

Figure 2. Shear used in the shielded process line

To achieve the technical feasibility of the PUREX process, some 50 liters of a spent fuel dissolution solution was needed, corresponding to the dissolution of about 15 kg of spent fuel. A PWR/UOx fuel had been chosen : its ^{235}U enrichment was 4.5 %, its burn up around 52 GWd/tU and its cooling time comprised between 1 and 8 years depending on its origin.

Some amount of 1400 mm of stumps of spent fuel from CRUAS and GRAVELINES nuclear power plants had been introduced in the shielded process line by connecting a TN 106 transport cask (**Figure 1**).

Then the spent fuel had been cut : the shear is a one by one machine (**Figure 2**). A knife rotates and cuts the spent fuel in pieces about 3.5 cm long.

3. Dissolution of spent fuel

After cutting step, the hulls were introduced in a basket placed into the dissolver tank. The dissolver is designed to make dissolutions of spent fuel in 8 or 16 liters. For this program, 6 dissolutions of 8 liters have been achieved.

The dissolver is shut to be watertight. HNO_3 solution is introduced and the heating system is switched on. During the dissolution, solution density, heat temperature and ^{85}Kr measurements are made. ^{85}Kr measurement is reached after gas treatment : it shows the dissolution progress (**Figure 3**).

The six dissolution batches were assembled for the preparation of the feed solution for the PUREX process. The composition of the resulting solution is shown in **table 1**.

Element	Conc.	Element	Conc.
H^+	4.6 M	Am	245 mg/L
		Cm	24 mg/L
U	228 g/L		
Pu	2.8 g/L	ΣLn	4,000 mg/L
Np	183 mg/L	$\Sigma \text{F.P.}$	6,000 mg/L

Table 1. composition of the feed solution of PUREX process, resulting from the assembly of 6 dissolutions batches

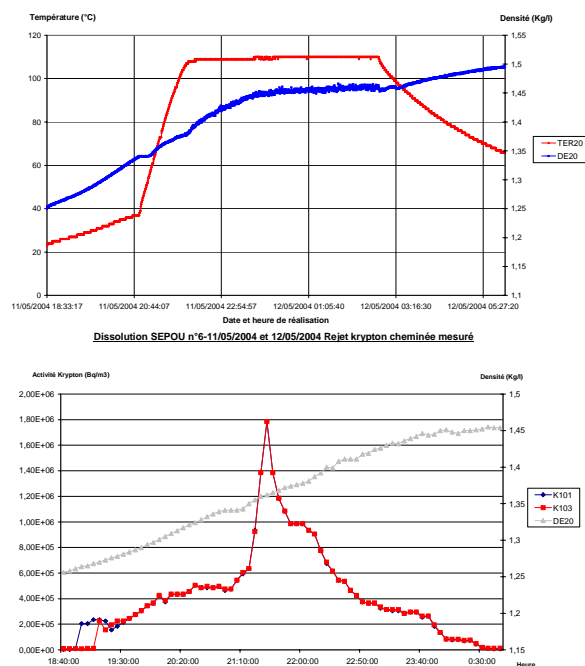


Figure 3. Heat temperature and density of dissolution solution (up)
⁸⁵Kr measurement and density of dissolution solution (down)

4. Liquid/liquid Extraction processes

4.1. Preliminary steps

Before the achievement of the two processes, PUREX-Np and DIAMEX in the liquid section of the shielded process line, one year has been spent to :

- the end of the liquid/liquid contactors implementation, pulsed columns with their instrumented captors (spectrophotometric measurements, retention rate sensors),
- the optimization of gravitating flows and all the parameters of flow rates,
- the characterization of the liquid/liquid contactor performances.

During this one year period, four campaigns of tests have been achieved with nitric acid solutions or solution containing only uranium to optimize all these parameters.

4.2. PUREX-Np process

The demonstration experiment to validate the use of the PUREX-Np flowsheet took place in the CBP line from April 11 to 15, 2005, using 46 liters of dissolution solution.

Target performance and flowsheet

The objective of the modified PUREX process (PUREX-Np) is to recover more than 99% of the neptunium contained in the spent nuclear fuel during the first purification cycle, i.e. together with the plutonium stream. The proposed flowsheet is shown in **Figure 4** :

- two pulsed columns are dedicated to the extraction function (**Figure 5 and 6**),
- one pulsed column is dedicated to the FP scrubbing function,
- the other functions (technetium scrubbing, Pu-Np and U stripping) are performed in benches of mixer-settlers.

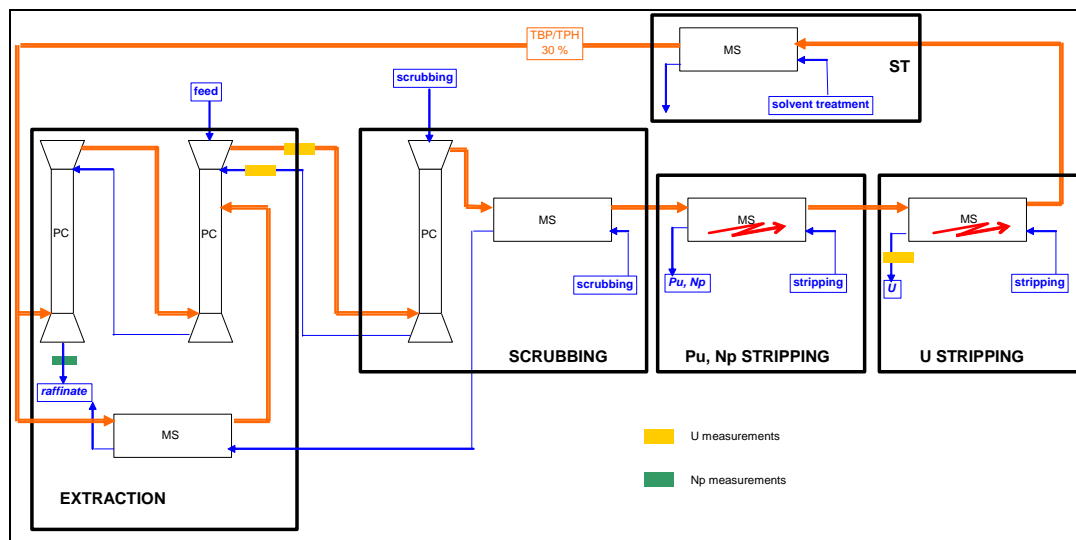


Figure 4. PUREX-Np flowsheet

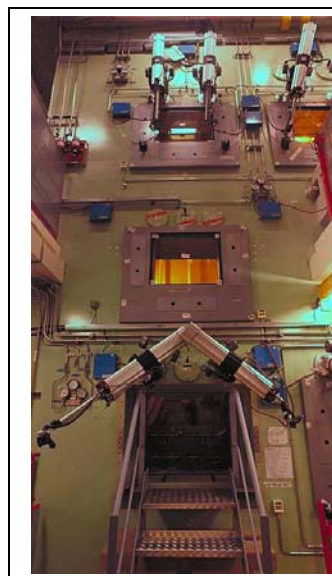


Figure 5. Large high cell containing the pulsed columns



Figure 6. CBP's pulsed column

Test procedure

The PUREX-Np test campaign was preceded by two important steps:

- tests with fictive solutions containing only uranium to validate the efficiency of the pulsed columns;
- optimization of the nitrous acid concentration (in the test solution the HNO_2 concentration is a major parameter that must be monitored to obtain the required selective Np extraction performance).

The test chronology can be summarized as follows:

- Setup (preheating, etc...).
- Startup and chemical equilibrium phase.
- PUREX-Np process on actual spent nuclear fuel batch.
- Scavenging phase.

The process performance was monitored by online spectrophotometric probes, notably in the pulsed columns. These provisions allowed real-time monitoring of the concentration variations of the species (mainly U and Np) in the flows relevant to this test. These results were supplemented during the test by periodic sampling and chemical analysis of solution samples in the Atalante shielded analysis line (CBA).

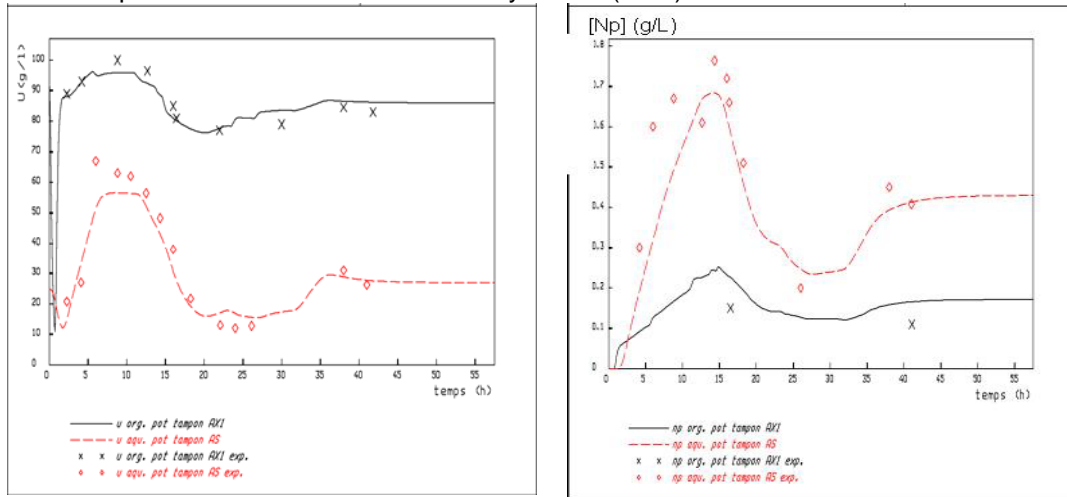


Figure 7. U and Np measurements compared to U and Np predicted values by calculation codes, in the extraction part of PUREX Np process.

Main results

The test performance was satisfactory. For neptunium in particular, the analyses confirmed its quantitative extraction and separation from the “U production” stream (cf. **Table 2**). Additional analyses confirmed the satisfactory fit between the test results and the values predicted by calculation codes (**Figure 9**).

	Extraction raffinate	Pu+Np production	U production
Pu	< 0.06 %	98.7 %	< 0.015 %
Np	< 0.7 %	102 %	< 4 %

Table 2 : U, Pu, Np repartition in the different flows of PUREX-Np process

4.2. DIAMEX process

After the PUREX process, the shielded process line had been prepared for the DIAMEX process, that is needed for the separation of the minor actinides and lanthanides from the fission products.

Target performance and flowsheet

The objectives of this process is to recover more than 99,9 % of americium and curium contained in the extraction raffinate from PUREX process. The second objective is to produce enough solution to achieved the last separation step, that is the SANEX process, dedicated to separate minor actinides from lanthanides. The proposed flowsheet for DIAMEX in shown in **Figure 8** :

- two pulsed columns are devoted for the extraction function (**Figures 5 and 6**),
- one pulsed column is dedicated to the FP scrubbing function,
- 2 banks of mixer-settlers are for the stripping step,
- one bench of 4 ECRAN operating for the solvent treatment.

Test procedure

The DIAMEX campaign consisted in three steps :

- configuration of the shielded process line,

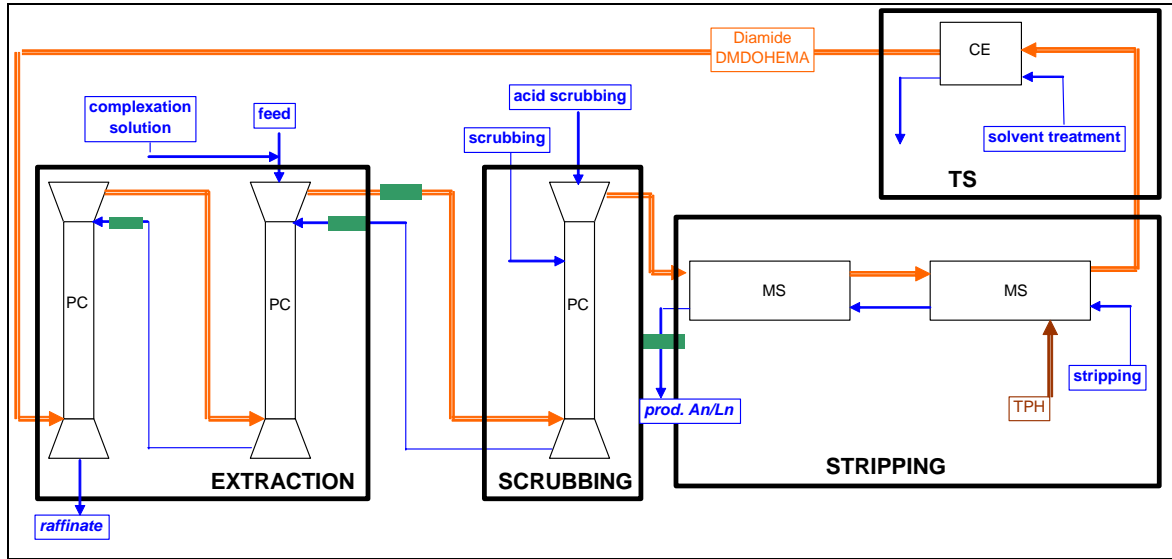


Figure 8. DIAMEX flowsheet

- test with neodymium and some other fission products to validate all the scheme,
- realisation of the DIAMEX test on the genuine solution from the PUREX : the composition of the feed solution of DIAMEX from PUREX Np process is shown in **Table 3**.

Element	Conc.	Element	Conc.
H ⁺	3.8 M	Am	156 mg/L
		Cm	16 mg/L
Pu	< 0.5 mg/L	Σ Ln	2,800 mg/L
Np	< 0.5 mg/L	Σ F.P.	4,800 mg/L

Table 3. composition of the feed solution for DIAMEX process

This campaign took place in the CBP line from September to November, 2005. The DIAMEX test took place between the 22 and 23 November, 2005.

The process performance was monitored by online spectrophotometric probes, notably in the pulsed columns. These provisions allowed real-time monitoring of the concentration variations of the species (mainly Am and Nd) in the flows relevant to this test. These results were supplemented during the test by periodic chemical analysis of solution samples in the Atalante shielded analysis line (CBA) (**Figure 9**).

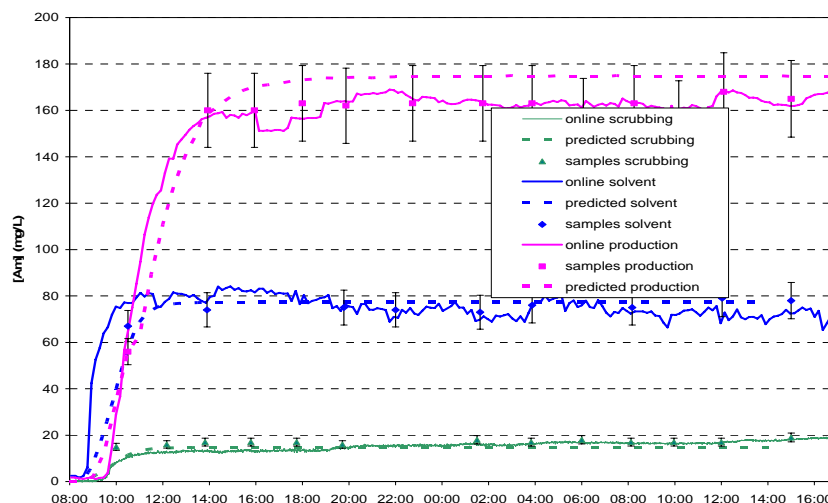


Figure 9. Am measurements compared to Am predicted values by calculation codes, DIAMEX process

Main results

- Actinide (and lanthanide) recovery factor

The americium concentration determined by α or $\beta\gamma$ spectrometry in the extraction raffinate was below $10 \mu\text{g}\cdot\text{L}^{-1}$, or about 0.014% of the quantity in the feed stream, showing very satisfactory extraction performance (the curium concentration was less than $0.4 \mu\text{g}\cdot\text{L}^{-1}$, or about 0.002% of the initial quantity).

Moreover, the quantities of americium and curium in the stripped solvent were below 0.01% and 0.002%, respectively, of the initial quantities.

The overall americium and curium recovery yield reached 99.9%.

- Decontamination with respect to fission products

The purification factors with respect to Zr, Mo and Fe in the first sample profile (T+14 hours) were respectively 409, 64 and 8, respectively. They were increased at the end of the test (profile at T+33 hours) due to the improved efficiency of the scrubbing column (higher pulse pressure). The decontamination factor (DF) with respect to Zr, Mo and Fe then reached >800, 100 and 10, respectively.

Seventeen liters of a solution containing actinides and lanthanides was produced : 4.5 liters was then transfer to C11/C12 facility, in which a SANEX process (actinides separation) has been achieved in December 2005.

5. Conclusion

The first objective of the shielded process line, that was the demonstration of the technical feasibility of the advanced separation, has been achieved. Six dissolution batches, one PUREX process (and its optimization phases) and one DIAMEX, after the entire reconfiguration of the process line has been performed in two years : this shows the high adaptability and flexibility of the facility.

This makes it possible to consider with serenity the new challenges of the future, for example separation and grouped co-conversion of actinides. This will need a new entire reconfiguration of the shielded process line, to integrate new liquid/liquid contactors, as Couette-effect columns, but also new processes as apparatuses for filtration and precipitation steps.