THE REPROCESSING PLANT OF THE FUTURE : A SINGLE EXTRACTION CYCLE

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ABSTRACT

In France, COGEMA has been reprocessing spent nuclear fuel on an industrial scale for over 40 years, and has consistently worked to optimize facility design and operations.

In COGEMA-La Hague's UP3 reprocessing plant, to achieve the necessary decontamination needed to produce purified uranium and plutonium, five extraction cycles were implemented and used at start-up: first cycle for separation of fission products, uranium and plutonium, two uranium purification cycles and two plutonium purification cycle.

By modifying processes at the design stage and making adjustments during operations, we saw that further decontamination of uranium could be achieved with only one cycle. Radiological specification of plutonium was also obtained at the end of the first plutonium purification cycle.

These good performance levels were taken into account for the design of the UP2-800 plant where uranium is purified using a single cycle, and for the recent R4 facility which features only one plutonium purification cycle.

Relevant information on extraction cycles in first-generation French reprocessing plants (UP1 and UP2-400) as well as design characteristics for the extraction cycles of reprocessing facilities currently operating at the COGEMA-La Hague plant is given. Experience shows that we can obtain adequate performance levels using only three cycles. We will also present potential evolutions for extraction cycles, e.g., neptunium decontamination, and demonstrate that one cycle can be sufficient for reprocessing the spent nuclear currently available. The benefits associated with a single extraction cycle will be detailed in the presentation.

INTRODUCTION

Reprocessing activity started in France at the end of the 1950s for the military program and was rapidly adapted to the treatment of civilian reactors fuels. Three plants were constructed, one on the Marcoule site (UP1) and two others on the La Hague site (UP2 and UP3). The PUREX process had to be progressively adapted to the treatment of more and more active fuels: from the single-cycle concept in the original outline of UP1 to the five-cycle design of UP3. Very good results of the new generation plants, UP3 and UP2-800, have already lead to simplifications of

their extraction process without relaxing the product specifications. The aim of this paper is to examine the feasibility of further simplifications to be applied to a future plant, the target being to come back to the single-cycle process.

HISTORICAL BACKGROUND

Reprocessing of spent nuclear fuels began in France in 1958 with the start-up of **UP1** plant in Marcoule. This plant, initially dedicated to the production of plutonium for defense purpose, was using the PUREX process with one extraction cycle only. Given the very low burnup of the fuels, this flowsheet was sufficient to meet the very stringent specifications of Pu product in terms of residual activity and impurities.
UP1 plant has rapidly been adapted to the treatment of spent fuels from the civilian UNGG reactors (gas-cooled, graphite-moderated reactors fuelled with natural uranium metal rods). To comply with the increasing burnup of those fuels (up to 5,000 MWd/t) and with their rather short cooling time (necessarily limited by the corrosion risk during interim storage), two plutonium purification cycles and one uranium purification cycle had to be implemented.

In the meantime, **UP2**, the second French reprocessing plant, was built on the La Hague site and started in 1967. Originally designed for the treatment of UNGG reactor fuels, and based on the experience gained with UP1, this plant was comprised of 3 extraction cycles:

- a first one for U and Pu co-decontamination,
- a second one for further co-decontamination and U/Pu separation,
- a third one to finish plutonium decontamination and to get a sufficiently concentrated solution to feed the plutonium conversion.

All the liquid-liquid extraction operations of the three cycles were performed in mixer-settlers (stage-wise contactors).
In 1976, a new head-end facility, HAO, (French acronym for High Activity Oxide) was commissioned to allow reprocessing of the Light Water Reactors (LWR) fuels in the existing plant. As the capacity of this new facility was 400 tU/year, the plant was renamed UP2-400. To reach the decontamination performances required for those fuels, whose burn-up was close to 30,000 MWd/t, the use of a supplementary uranium purification cycle was necessary. Moreover, it appeared that the decontamination performances of the extraction cycles could be sustained only on condition that the solvent were periodically renewed.

Lessons learned from operation of UP2-400, positive and negative as well, were considered for designing the third generation reprocessing plant, **UP3**. Started in 1989, this plant was dedicated to reprocessing of LWR fuels from foreign utilities. Its nominal capacity was 800 tU/year. Five extraction cycles were implemented and used at start-up (refer to fig. 1.a):

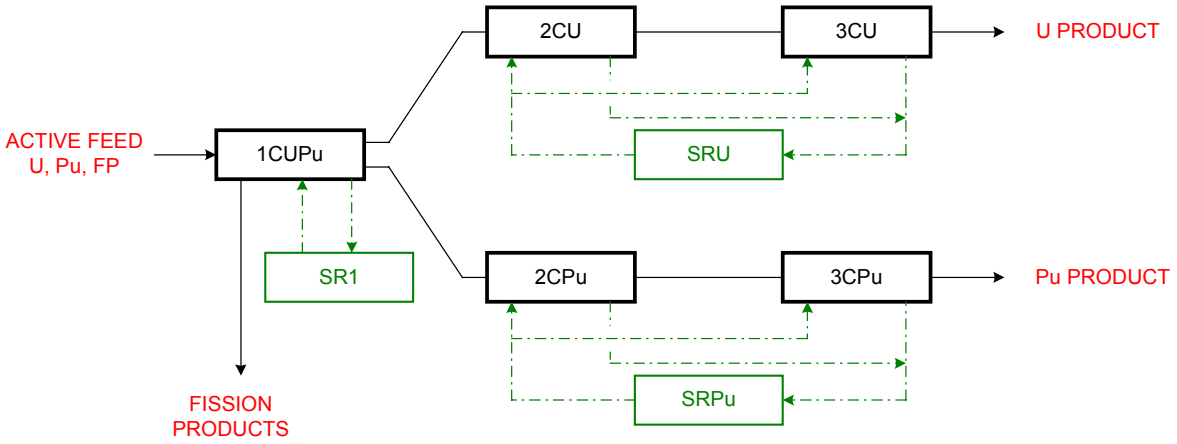
- first cycle for co-decontamination and uranium/plutonium separation (1CUPu),
- two uranium purification cycles (2CU and 3CU),
- two plutonium purification cycles (2CPu and 3CPu).

Particular attention was paid to the solvent management based on three independent solvent loops, each of them fitted with an alkaline Solvent Regeneration unit (SR): SR1 for the first cycle, SRPu for plutonium cycles and SRU for uranium cycles. Like in UP1 and UP2, the extractant is the tributyl phosphate (TBP) diluted in a saturated hydrocarbon diluent. The major

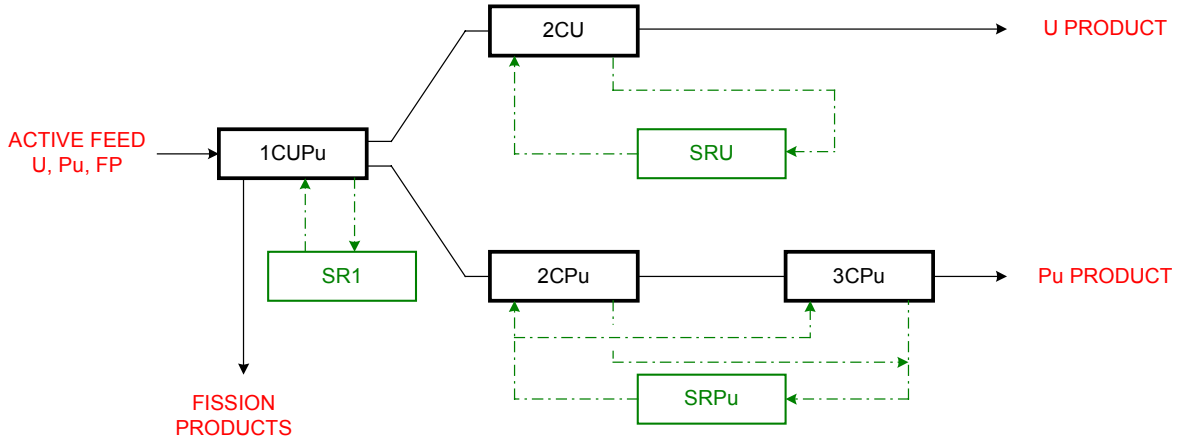
innovation consisted in the Organic Wastes Treatment unit (OWT), based on a vacuum distillation. This unit ensures purification of used solvent and allows recycling of diluent and concentrated TBP into the main process line. This provides a permanent renewal of the solvent stock.
In addition, pulsed columns were selected as contactors in the most active parts of the plant, mainly to comply with the criticality-safety requirement. This technology drastically reduced the solvent degradation rate thanks to the lower residence times and to the better management of interfacial cruds compared to mixer-settlers.

OPERATION OF THE PRESENT REPROCESSING PLANTS

Very soon after the start-up of UP3, in the early nineties, it appeared that the decontamination performances were far better than expected. Thanks to the process improvements described above, the products specifications were almost reached at the outlet of the first extraction cycle.
In actual fact, at the outlet of the first cycle, residual Pu contamination of uranium product was lower than required by product specifications, and with slight flowsheet optimizations, specifications for residual neptunium and $\beta\gamma$ contamination in uranium product could be reached at the outlet of the second uranium cycle (2CU). Operation of the third uranium cycle (3CU) was definitely stopped in 1994 (refer to fig. 1.b).
As for plutonium, although every specifications were met at the outlet of second plutonium cycle (2CPu), the third plutonium purification cycle (3CPu) was kept in operation for the sole purpose of reaching the necessary concentration of plutonium stream to feed the downstream conversion process.



a. Original design



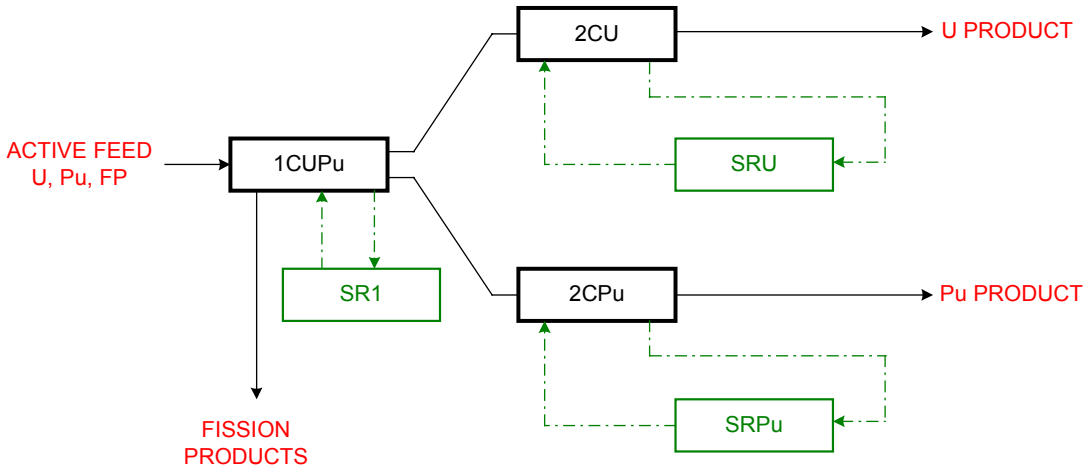
b. Present outline

Fig. 1. UP3 extraction scheme

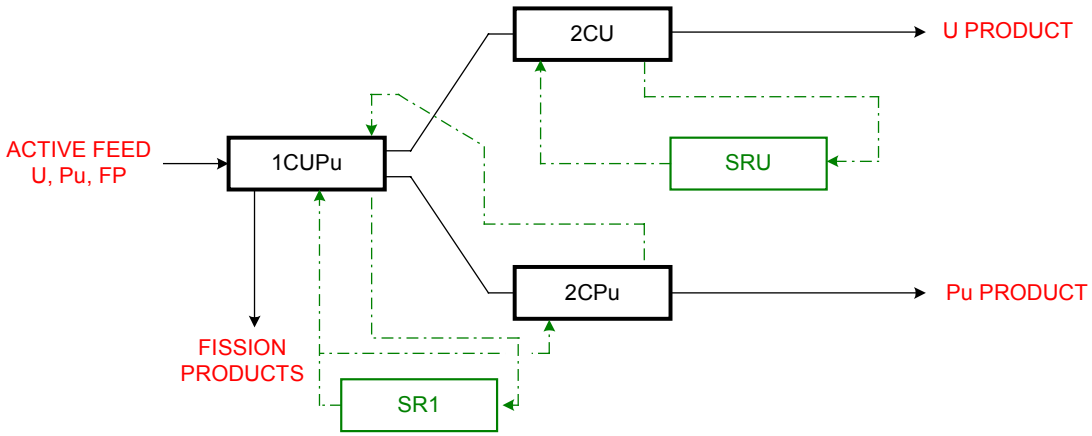
Since start-up, these high performances were sustained, whatever the inlet activity of reprocessed fuels. The cooling time decreased down to the design value of 3 years, and the burnup nowadays reaches values as high as 50,000 MWd/t.

These results were reflected on the design of the Japanese Rokkasho-Mura reprocessing plant, currently in commissioning phase, and on the La Hague UP2-800 plant, that was started in 1994:

- Rokkasho Reprocessing Plant is fitted with 3 extraction cycles: one cycle for co-decontamination and separation of uranium and plutonium, one cycle for further purification of uranium and one cycle for further purification of plutonium. One alkaline solvent regeneration unit is associated to each of these 3 cycles (refer to fig. 2.a). The organic wastes are recycled by means of a vacuum distillation process, like in UP3 plant.
- UP2-800 is based on the same three-cycle-flowsheet, but the alkaline solvent regeneration unit of the first cycle is shared with the plutonium cycle (refer to fig. 2.b). An Organic Waste Treatment unit is also implemented in UP2-800 plant.



a. Rokkasho Reprocessing Plant



b. UP2-800 present outline

Fig. 2. The current three-Cycle-concept

The performances currently achieved in La Hague reprocessing plants can be summarized in following figure :

Table I. Performances of La Hague reprocessing plants

La Hague plants	
19,400 tons of spent fuel reprocessed et the end of year 2003	
Fission products DF (1CUPu)	Cesium > 10^7 Ruthenium > 2.10^4
Neptunium DF (2CU)	> 1000
Plutonium fission product activity	< 1μCi/gPu
Uranium fission product activity	< 2μCi/kgU
U and Pu recovery ratio	over 99.88%

POSSIBLE SIMPLIFICATION OF THE EXTRACTION SCHEME

The outstanding results obtained in UP3 and UP2-800 plants operated by COGEMA for respectively 15 and 10 years, incline to study further simplification of the extraction scheme. Indeed, it is difficult to modify existing installations. But when designing a new plant, the single-cycle concept has to be reconsidered despite the tremendous burnup increase since UP1 start-up.

UP2-800 experience has proven that the reducing stripping of plutonium by uranous nitrate in pulsed column, combined with a Pu barrier in mixer-settler, yields an uranium stream that meets the specification in terms of residual Pu contamination. The remaining difficulties that have to be overcome to reach the target of the single-cycle-process are the global βγ decontamination and the decontamination of the uranium stream from neptunium. Additionally, in order to reach the required plutonium content prior oxalic conversion, the plutonium stream has to be concentrated.

Decontamination in βγ emitters

Considering the high purity level needed for recycling in reactors (i.e. re-enrichment of uranium and MOX fuel fabrication), it is not conceivable to relax the constraints on residual contamination of these products. In addition, as the trend is to increase the burnup of the fuels, their activity when discharged from reactors also increases. This results in a continuous increase of the required Decontamination Factors (DF is defined as the ratio between the total activity of the fuel at the inlet of the plant and the residual allowed activity of the uranium and plutonium products.). For instance a global DF of about 10^7 is required for a 50,000 MWd/t fuel reprocessed after 4 years cooling.

Today, in La Hague plants, this value is commonly achieved at the outlet of the first extraction/scrubbing step for most of the fission products excepted for ruthenium whose DF is 10^4 only. As a result, the residual contamination of uranium and plutonium streams at the outlet of the first cycle is mainly due to this radionuclide. It is worth noting that the half-life of this element being 1 year, its activity is divided by 2 after one year and by 1000 after 10 years by natural decay. Consequently, thanks to a cooling time of about 10 years (instead of 4 years which is the present design value), the residual βγ contamination could meet specifications at the outlet of a single cycle. It has to be noticed that the increasing of cooling time is not a very stringent constraint in countries where huge amounts of spent fuels are under temporary storage.

Decontamination of uranium from neptunium

In the hypothesis of reprocessing after 10 years of cooling time, the only remaining purpose of a 2^{nd} uranium cycle would be the neptunium decontamination. A significant improvement to reach the single-cycle target would consist in eliminating the neptunium from the uranium loaded solvent. Preliminary studies and tests performed by the French Commisariat à l'Energie Atomique (CEA) have shown that this goal could be reached thanks to the use of an appropriate complexing agent that reduces the distribution coefficient of neptunium, making it unextractable.

Concentration of Pu stream

Like for the uranium stream, the βγ specification of plutonium could be reached at the outlet of the first cycle if the fuel were processed after 10 years of cooling time. The only remaining point would be to reach adequate concentration to feed oxalic conversion process. The required concentration factor can be achieved thanks to a mere extraction/stripping step with adequate A/O ratio. The experience of UP2-800 demonstrates that there is no need for a specific solvent regeneration unit for this operation. The additional solvent loop could be managed jointly with the main solvent loop of the cycle. Of course, a supplementary conditioning stage has to be implemented to adjust acidity and Pu valency to the extractable valency IV prior extraction.

Figure 3 describes the main features of the single-cycle process:

- an extraction/scrubbing step. A first scrubbing is performed at moderated acidity in order to eliminate most of the fission products (including zirconium). A second scrubbing at higher acidity ensures decontamination from technetium and ruthenium.
- a Pu stripping step, to separate plutonium from uranium, using uranous nitrate as a reducing agent, combined with an uranium scrubbing of the plutonium stream and with a Pu barrier on the organic uranium stream.
- a Np complexing stripping on the uranium organic stream before uranium stripping.
- a Pu concentration step achieved by extraction/stripping.

All these operations are performed with a single solvent stock, and thus with a single alkaline solvent regeneration unit. An Organic Waste Treatment by distillation ensures the permanent renewal of the solvent stock.

Management of the aqueous liquid wastes would be limited to a high active liquid wastes concentration unit, and a single acid recovery unit (instead of 2 in the existing multi-cycle plants).

Moreover, considering the experience of La Hague plants, especially the operation of annular pulsed columns, it is conceivable to design a very high capacity plant (up to 2,000 t/year) based on the single-cycle concept and using a single process line.

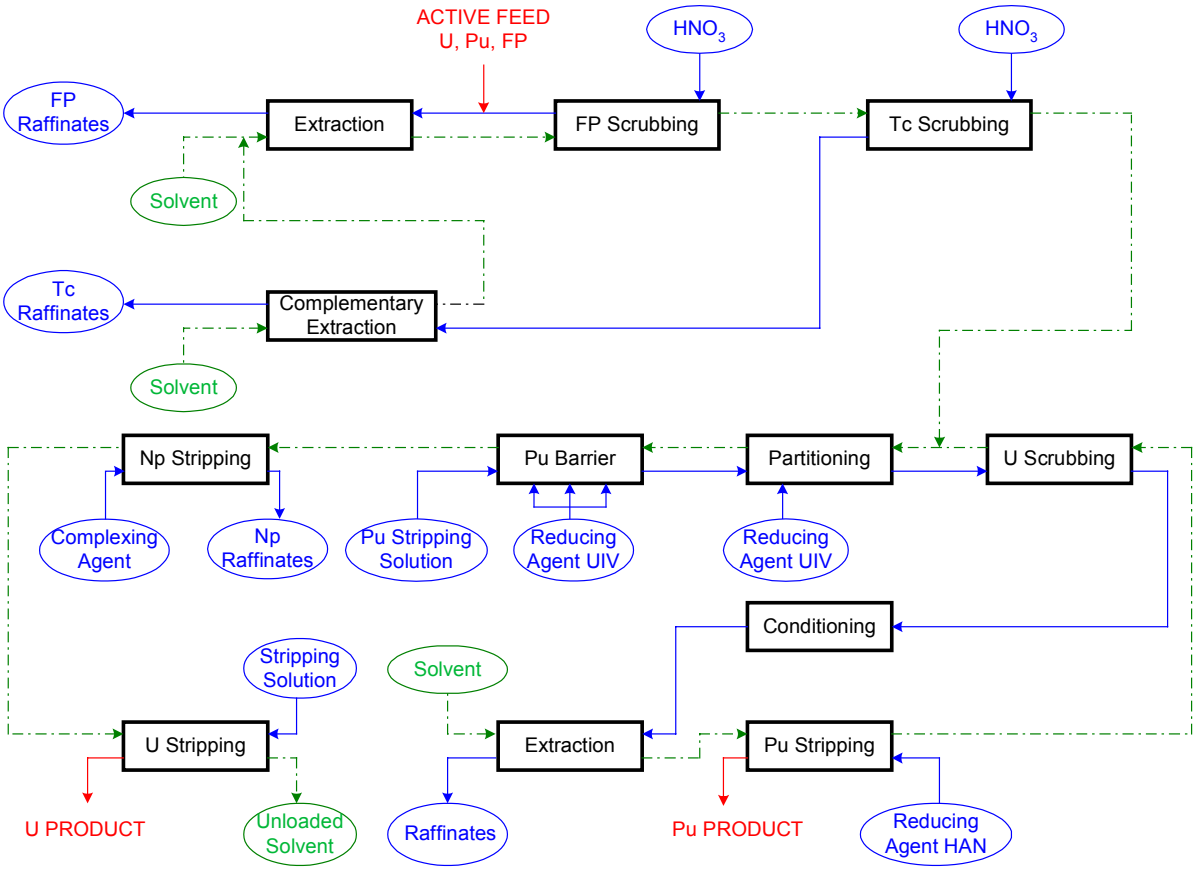


Fig. 3. Main features of a single-cycle extraction scheme

ADVANTAGES OF THE SINGLE-CYCLE PROCESS

The implementation of a single-cycle process instead of the current multi-cycle process would obviously have a considerable impact on both the capital cost and operating cost of a reprocessing plant.

The number of equipment would be dramatically reduced, not only the extraction equipment but also the liquid waste treatment including the acid recovery. The volume of the process core would be reduced in such extent that it would be possible to install the whole extraction and waste treatment process in a single building. Knowing the cost of civil engineering for a reprocessing plant, this is not the least advantage of the evolution. The instrumentation and control system that also represents a significant part of the investment cost would be reduced proportionally. Many other cost reductions are expected on the ancillary units such as utilities, reagents, as well as process and building ventilation.

As for the operating cost, the major benefit would come from the operating team reduction for both normal operation and maintenance. Significant savings would also be made on the energy and fluids consumption and on the solid waste treatment.

CONCLUSION

When considering the outstanding results of the high throughput reprocessing plants operated by COGEMA on the La Hague site, it becomes clear that a future high capacity plant could be designed with a single-cycle PUREX process. Preliminary results of CEA studies on this topics are promising and R&D works are still going on to optimize the flowsheet conditions.

This would make the spent fuel reprocessing more cost effective and could be achieved:

- by increasing the cooling time of the spent fuels which, in most cases, is not detrimental to the fuel cycle economy,
- but without relaxing the specifications of U and Pu products which must remain suitable for re-enrichment and MOX fuel fabrication.

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