From a Scientific Feasibility to the Industrial Application: the COGEMA Experience.

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Abstract: Since it was founded in 1976, COGEMA has designed and built two reprocessing plants, each with a capacity of 850 t/y in La Hague, France. These plants, named UP2-800 and UP3, use the PUREX process. UP3 began operations at the end of the '80s and UP2-800 in the mid-'90s. Innovations have been constantly incorporated to these plants to improve process efficiency and to reduce the activity and volume of waste. During these years, COGEMA has acquired an important experience in industrialising processes and technologies developed in the laboratory, especially in the domain of liquid-liquid extraction. The goal of this paper is to present this experience, taking as an example the development and design of the UP2-800 Pu conversion facility R4 started in 2002.

INTRODUCTION

Bases of hydrometallurgical PUREX process, used to reprocess nuclear spent fuel, were established in the ‘50s with military purpose. In the USA, Savannah River and Hanford PUREX plant, pioneer in this field, start respectively in 1954 and 1956. In France, UP1 plant, built in Marcoule, starts in 1958.

We have to wait the ‘70s to see the second generation of reprocessing plant, with commercial purposes. It is in France UP2 in La Hague, started in 1967 with Gas Cooled Reactors fuels, adapted to the treatment of oxide fuels and re-named UP2-400 in 1974, Eurochemics in Belgium, Windscale (later Sellafield) in Great-Britain and Tokai Mura in Japan. These plants were characterised by rather small capacities.

In France, at the end of the ‘70s, political decisions about energy and more stringent laws in term of safety, workers exposure and wastes lead to design and built two third generation reprocessing plants: UP3 and UP2-800. Each one has a capacity of 850 THM/y.

Previous experience allows to define specifications for these two plants. Major R&D programs are launched by CEA and new design principles are established by engineering. Joint efforts of the French Commissariat à l’Energie Atomique (CEA), SGN (COGEMA engineering subsidiary) and COGEMA allow to start UP3 in 1989 and UP2-800 in 1994.

For more than 10 years, these plants have constantly incorporated new features to improve process efficiency and reduce wastes activity and volume. The very good current performances of the process and the very low impact on natural environment are the result of more than three decades of joint efforts of CEA, SGN and COGEMA. During this time, COGEMA has acquired an invaluable experience in industrialising processes and technologies,
especially in the domain of liquid-liquid extraction.

OUR ORGANISATION

R&D studies are performed by CEA for COGEMA. A close collaboration between CEA research teams and COGEMA (supported by SGN) is the key factor for the success of a project. As shown in figure 2, four types of competencies have to work together:

- R&D teams (CEA)
- Engineering teams (SGN)
- Plant operating teams (COGEMA)
- Project management (COGEMA)

All these competencies must work together from the very beginning of research to the plant start-up end even after.

![Figure 2: Project organisation](image)

THE MAIN STEPS FROM FEASIBILITY TO PLANT START-UP

A project for industrialising a process could be described in 6 steps:

- scientific feasibility
- technical feasibility
- design phase
- tests and start-up
- first months of running feed back
- continuous improvement.

We will now described these steps and show how each competency participates at each step.

- Scientific feasibility
  The first step is to establish the feasibility of the process. Preliminary tests are performed to select the best reagents and the best operating conditions (flowrates ratios, reagents concentrations, etc…). Simulation softwares are fed with these data and simulation runs help to determine a tentative flowsheet.

Continuous tests are then performed, first by using miniature pilot-plant in glove-box that allow us to check the performances of the process, with materials compatibles with glove-box, i. e. : non-radioactive material; α emitters : U, Pu, Np, and others like Tc, etc…. In most case, even in this early approach of the development of a new process, a validation with actual highly active liquors is performed in a shielded cell to check if there is no unforeseen noxious interference of fission product or high radioactivity on the process. Using shielded cells is more demanding for both time and money than using glove boxes, so that optimum development work needs using both types of equipment.

From the beginning of this first step, a close collaboration between CEA, SGN and COGEMA is essential. It allows an early screening of potential difficulties for future applications. Safety aspects and waste management are considered with the greatest attention. For example, if selecting a new reagent, its behaviour in further steps of the process requires as much attention as its efficiency in its primary function. The nature and behaviour of the degradation products are also investigated. As a result, by the end of this first step, the main constraints for the future industrial application have already been identified and taken into account.

- Technical feasibility
  This step aims at validating the process as an industrial one by using laboratory, pilot-plant and computer tools. Laboratory tests are performed in conditions as close as possible to those of the future facility: continuous runs, representative or actual liquors and long lasting tests. Process stability, dysfunction recovery, mastery of degradation products accumulation and validation of the computer models bring the necessary data to built the Safety Demonstration. Representative, or even full scale equipment are tested to investigate mechanical, hydraulic and thermal behaviour, and the results, residence time,
temperature and heat flow, interfacial contact quality are crossed-checked with the requests of the process as demonstrated by the laboratory investigations and computer models results to guarantee the efficiency of the process at full scale and in highly active conditions.

Engineering and operating teams bring their expertise to the preparation and the debriefing of these tests together with R&D specialists: this is a key factor for their success. Comparison with actual plant operation provides the first data for the future facility operations.

Figure 3: Mixer-settler in Atalante hot cells.

- **Design phase**

It is the industrialisation part itself. For a predefined specification, several types of studies have to be done simultaneously and with very close collaboration between the different teams:

- Engineering studies to define the process in detail (flowsheet, equipment selection and design, piping and layout, instrumentation and control design, wastes management...).
- Safety demonstration and implementation of safety features into the process design.

All along these studies, R&D complementary studies are needed to validate engineering options and support safety demonstration, both in laboratory scale and pilot scale.

For example, one of the major evolutions between UP2-400 and UP3/UP2-800 concerns the dissolution step. From batch operation, dissolution becomes continuous thanks to development of rotary dissolver (figure 4a and 4b). This development needed significant R&D efforts including tests on a full scale pilot with dissolution of non irradiated UO₂ fuels.

Figure 4a: Rotary dissolver – principle scheme

Figure 4b: Rotary dissolver in La Hague plant
The use of a rotary dissolver allows to reach the UP2-800 capacity (850 THM/y) with a single unit.

It is important to note that, since 2004, pilot tests with real active solutions can be performed by CEA in a new large scale facility in hot cell (CBP : cellule blindée procédé, figure 5). This facility is used in 2004 and 2005 to validate Am/Cm separation process.

• Tests and Start-up

Tests and Start-up of a facility are done by COGEMA teams, but supported closely by R&D and engineering teams. Their perfect knowledge of the process allows to react very quickly in case of malfunction. Nuclear facilities are started-up according to a prudent procedure that includes cold tests and active tests before commercial operation. Laboratory or pilot scale tests could be done during the start-up to identify the origin of malfunction and correct it. With our organisation, the start-up of a facility is the continuity of R&D and engineering studies.

• First months of running feed back

R&D and engineering teams support COGEMA during the first months of facility running. The feed back of this period allows to perfect the knowledge of the process. Plant operating data are collected and used to improve computing. Some ways to optimise running could be identified, required further R&D programs.

• Continuous improvement

During years of running, COGEMA teams meet regularly CEA and SGN to analyse operating feed back and try to improve process efficiency and optimise running. For example, operating teams know how, with support of R&D and engineering, allows to reduce reagents quantities with the same process efficiency. Continuity of work between all the teams is essential in case of significant evolution of the process, for example to reprocess new types of fuels. Recently, CEA, SGN and COGEMA worked to adapt UP3 to research reactors fuels reprocessing and to adapt UP2-800 to MOX reprocessing.

THE UP2-800 R4 FACILITY EXAMPLE

To illustrate the results which could be obtain with such an approach, we can take for example the last process facility built and started April 2002 : R4. R4 is the new Pu purification and conversion facility of UP2-800 plant. Its present capacity is of 40 kgPu/day, expandable to 80 kgPu/day if necessary. From 1994 up to 2001, UP2-800 was operated with the Pu purification and conversion facility of the former UP2-400 plant named MAPu. UP3 T4 Pu facility operating feed back has been taken into account. Significant R&D studies was performed by CEA to develop and validate the two main differences between T4 and R4:

• a single purification cycle instead of two,
• purification using multistage centrifugal extractors instead of pulsed columns (figure 6).

Figure 5 : CBP 4 meters pulsed columns
Tests were performed both in laboratory scale (with Pu) and with full scale pilot (using inactive simulated solutions).

Due to small hold up in centrifugal extractors (60 times less voluminous than pulsed columns), time to react in case of malfunction is short. So, large part of R&D program consists in developing online analyses.

A close collaboration between CEA, SGN and COGEMA allowed to obtain the following results:

• less than 10 years between the beginning of R&D studies and facility start-up,
• only three months to reach successfully the nominal installed capacity (40 kgPu/day)

CONCLUSION

With thirty years experience in the design and operation of reprocessing plants, and with more than 19,000 tHM (01/01/04) treated in La Hague, COGEMA and its partners are best suited to industrialise all new processes for spent fuel reprocessing currently being developed. In particular, COGEMA is working with CEA on the technical feasibility of advanced partitioning (minor actinides recovery for transmutation), and on the definition of an optimised hydrometallurgical process in a single extraction cycle. Other process could also be studied, like new hydrometallurgical process allowing different management of nuclear material (for example recovery of Pu together with Np or U instead of isolated).