

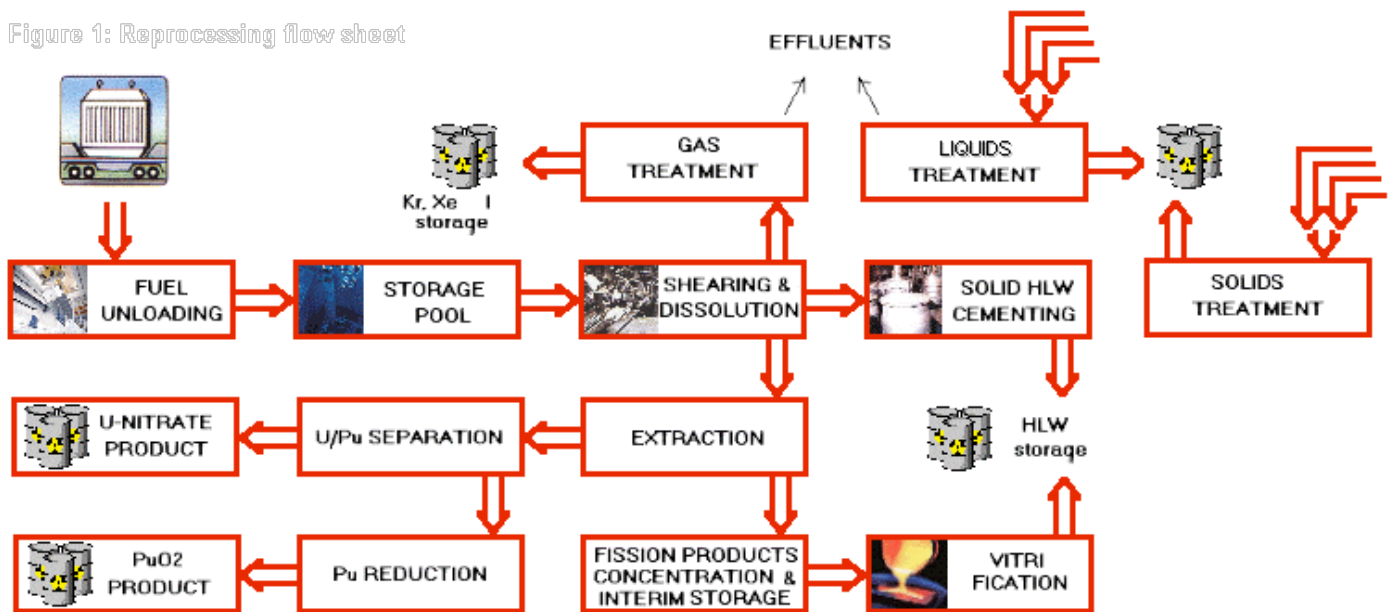
🚧 A look at La Hague

Overview

It would not be very useful to discuss every part of a reprocessing complex. We will just focus on the most important sections. As a representative example, we will have a closer look at the UP3-A (and the nearly identical UP2-800) plant at La Hague. Cogema has been rather reluctant to disclose information about certain parts of the complex. Although this has changed a bit since the UP3-A start-up in 1990, various sections like the extraction and U and Pu purification can only be described schematically, without actual drawings or pictures. In my opinion, this is not an obstacle to understand how things work.

In figure 1 below you can see a simplified flowsheet of the UP systems giving you an idea of the processes. Do browse through it, this will be our guide when examining some stages more closely. You can imagine that all those different material streams, some of which need to be totally contained during their extraction and treatment, make the reprocessing stage the most complex part of the entire fuel cycle. The UP3-A design was in fact largely determined by avoiding waste accumulation and diversion for temporary storage as much as possible. For example, the solid high level waste from dismantling the fuel initially was to be temporarily stored under water, but now it is being cemented right away.

Figure 1: Reprocessing flow sheet



Fuel unloading and interim storage

Upon arrival, the fuel casks need to be unloaded. This used to be done under water, as is still the case in the UP3-A section T0. The UP2-800, which is an extension of the old UP2-400, has several new features drawn from UP3-A experience. One of these is a dry cask unloading facility (R0), which can be used and maintained more efficiently. You can see this in figure 2. Since Cogema herself produces the transportation casks, their handling could be standardized. By remote control, the fuel elements are then put into one of the five storage ponds for further cooling.

The spent fuel currently being reprocessed has been stored like this for up to ten years, but that was merely because it took ten years to build the complex, perform testing, and reach full throughput. In fact, because of the use of remote control techniques, the fuel can almost be reprocessed immediately (after one year or so). This logically implies that the "contamination" of the material being put through is higher, which might increase risks. However, storage ponds are not all that safe either and criticality or thermal explosion hazards may well be more likely to occur through a loss-of-coolant event in a storage pool or a liquid fission product solution storage tank than through failure of all subsequent containment managing steps in the extraction section.

Shearing and dissolution

This part of the complex is also called the head-end, T1 in the UP3-A, R1 in the UP2-800. First of all, the fuel elements need to be chopped to pieces. Using a horizontal shear the top and the bottom end, containing no fuel, are chopped off. After that, the fuel rods are cut into slices of about 3 cm (1.3"). In the old days, this must have been an extremely dangerous task. In the Dounreay FBR reprocessing pilot plant, and my guess would be also in the Eurochemic plant, they did this basically by hand! Currently, the shearing can be managed remotely and the fuel doesn't need to be under water.

Figure 2: Dry fuel unloading



Next thing, the chopped fuels are sent to a continuous rotary dissolver entering into contact with boiling nitric acid. You can see the Cogema design in principle in figure 3 on the left. The emerging gas consists mainly of nitric oxides, but it also contains the gaseous fission products (of course, during shearing a fraction of these already escapes). The main solid residue consists of parts of the non-soluble fuel hulls, containing radioactive activation and corrosion products, some of which may also be found in the dissolution liquor.

Some 10% of the fission products are also non-soluble, such as palladium, rhodium, ruthenium and molybdenum. With ever rising burn-up rates and when reprocessing MOX fuels, these are expected to cause trouble in the future as was the case in 1973 when an attempt by the British to modify the existing Magnox head-end of the old Windscale plant in order to be suitable for oxide fuel, resulted in an explosion due to clogging because of too low fuel solubility. There will also be a small fraction of the plutonium in the solid residue (the solubility of uranium is better). This is why the plutonium loss during the process is always somewhat higher than the uranium loss.

Currently, the solid waste is directly immobilized in a concrete matrix within the head-end containment. Most I-129 is filtered out of the gasses by absorption, according to Cogema (according to Greenpeace, up to 90% is discharged). It is not really clear what happens with this I-129 if it is filtered out, but I presume that for the time being it would be kept separately. I am not sure if the radioactive noble gasses krypton and xenon are captured at La Hague. If not, scandalous would be the appropriate word, since gaseous radioactive emissions of nuclear power plants have shown significant decrease by implementing this. This might however be the only reasonable explanation for the fact that normal gaseous and liquid emissions at La Hague per contracted LWR, or per GW(e), are in the range of several hundreds times the average discharges at the reactor site under normal operation conditions! The usual cheering that annual discharges at reprocessing sites always turn out to be "significantly" below legal limits doesn't include such comparisons. More about radioactive discharges can be found in the section [about radioactive discharges](#).

The PUREX process

In figure 4, you can see the principle of extraction. PUREX means Plutonium and Uranium Recovery by EXtraction. In reality, the nitric acid solution containing uranium, transuranics and fission products is in counterflow with a 30% tri butyl phosphate (TBP) solution in kerosene.

As you can see in the drawing, and as you might know from real life, organic and aqueous phases do not mix. But when stirred, the uranium VI and plutonium IV ions are able to go into the organic phase by becoming part of complex TBP nitrate molecules (VI and IV refer to the valence, so U and Pu behave like 6+ and 4+ ions respectively in an aqueous solution). If one chooses a low nitric acid concentration (like 0.01M), both the U and Pu are likely to stay in the aqueous phase.

But if one chooses a high nitric acid concentration (around 4M), both the U and Pu have a tendency to enter the organic phase, while the fission products prefer the aqueous phase. Some of them however, in particular zirconium and technetium, don't show "perfect" behaviour and in fact neither do U and Pu. In practice, one arranges several extraction and back-extraction steps after another. In general, pulse columns are most commonly used. At La Hague, they use mixer-settlers for the extraction steps in the T2 (R2) section of the UP3-A (UP2-800).

Once the U and Pu are "decontaminated", we have an aqueous solution containing U VI and Pu IV (and nitrate, hence the names uranyl- and plutonyl nitrate). To separate them, one can make use of normal chemical redox reactions, since the redox properties of U VI and Pu IV are different. A widely employed method is based on the reductive properties of U IV and hydroxyl ammonium nitrate (first U VI enters an equilibrium redox reaction with U IV). The Pu IV is then reduced to Pu III, which can be separated using an organic TBP solution, since Pu III shows very poor solubility in the organic phase.

At La Hague, they have three cycles to purify the U and Pu. If there is any reason in terms of business competition why the Cogema people (as they claim) do not disclose detailed information about the exact system they use, it must be some clever way to combine fission product separation and U and Pu separation. So, instead of following the linear path described above, they presumably have exploited the possibilities of Pu reduction combined with Pu III's different solubility behaviour right from the first cycle. If one can manipulate the nitric acid concentration very precisely, this is certainly possible.

Perhaps you have also noticed how reprocessed plutonium(oxide) can relatively easy be cleansed from americium (formed through decay of Pu-241): First one dissolves the plutonium oxide in nitric acid and then one uses TBP again to purify the plutonium, and then one "back-extracts" the plutonium again to plutonyl nitrate. Charges for this may be still up to \$1200 per kg of plutonium, but I only know for sure that the British perform americium stripping on a somewhat commercial base. The quoted charge seems really high to me, but it is an OECD figure from 1989 and it is not unlikely that upon request only Dounreay came up with an offer (they are somewhat short of business). Cogema's design allows americium stripping to be easily incorporated in the process since it has always been part of the design as an optional feature. Now that plutonium

Figure 3: Principle of Cogema's continuous rotary dissolver

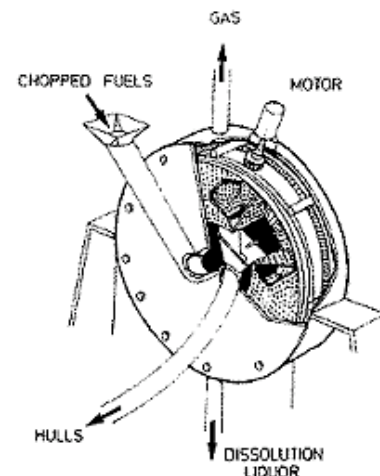
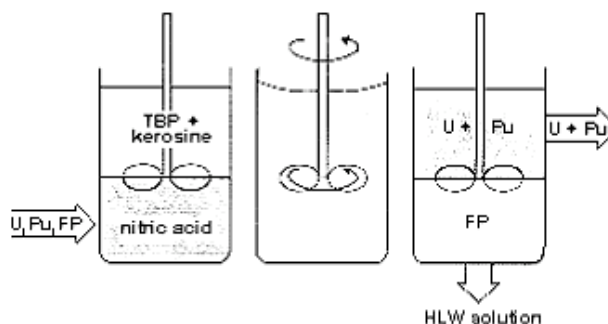


Figure 4: Extraction principle (PUREX)



"contamination" due to long storage tends to become a commercial problem, I have no doubts that the Cogema people are closely monitoring the development of this inconvenience. A problem which, of course, has been foreseeable since the very beginning when utilities eagerly signed on the dotted line.

End products

Believe it or not, the contractual end product for uranium is the nitrate solution. So if a utility wants to re-use it, it has to be converted either to the hex form (for re-enrichment, this is almost not done) or to the oxide form, which can be used in MOX fuels but in general it is more attractive to use natural or depleted uranium. Eventually, some customers might even consider having the uranyl nitrate converted to uranium oxide just for the sake of moving it back to their own country and store it there, avoiding Cogema's storage charge.

The plutonium cannot be economically stored in nitrate form, since critical masses (or rather: concentrations) in aqueous solutions can become very small. This means that plutonyl nitrate cannot be concentrated as much as uranyl nitrate. It is therefore more attractive to convert the plutonium solution to oxide powder. This is done by treating it with a hot oxalic acid, causing the plutonium to precipitate as plutonium(IV)oxalate. When dried it is calcined while modestly annealed, the product being plutonium(IV)oxide (PuO_2). This is weighed (with a margin of 0.1%) and put in storage containers.

The amount of plutonium to be made available to a customer is determined by the sampling results of the fuel composition upon dissolving minus Pu-241 decay during the process minus the process losses, which in the baseload contracts from 1977 and 1978 were merely defined as being 3%. In practice, they turned out to be as low as 0.1%. The uranium conversion sections are the T3 and T5 section in the UP3-A and the plutonium treatment is the T4 section. In the UP2-800, the old MAU and MAPu units are still being used, at least until the next century. Just like for uranium there are also storage costs for plutonium oxide, which are relatively much higher than for uranium storage. In absolute terms, they are of the same order.

Of course Cogema also provides additional service agreements for MOX manufacturing (possibly also using the customer's reprocessed uranium) and uranium re-enrichment and fuel fabrication. So far, only the French electricity sector (EDF) has gained some experience with the latter. It has serious technical and economical disadvantages compared to fresh uranium. But since reprocessing promised to re-use the plutonium (a fraction) as well as the uranium (some 94% of the spent fuel), the French and the Japanese still have their minds set on eventually re-using the reprocessed uranium. And if the cost disadvantage persists, they can always come up with the "waste reduction" story, instead of the "valuable resources" thing, as is the general trend in the current nuclear society. The British have recycled uranium from Magnox fuels, which is much less "polluted". Campaigning for THORP in governmental circles, BNFL has made the outrageous claim that this uranium recycling experience proved the (economic) feasibility of recycling uranium from enriched oxide fuel -- with high burn-ups -- suggesting this is an established business practice.

HLW residue storage and vitrification

The aqueous solution of fission products and transuranics is first concentrated up to about a factor of 15. Subsequently it is stored in steel tanks, permanently stirred and cooled for a period which may be up to a decade. Eventually, the solution is vitrified, meaning that it is diluted with molten borax glass. Now we have VHLW. The figure on the left shows the continuous vitrification process, developed at the AVM (Atelier de Vitrification Marcoule, the final extension of the first UP1 reprocessing complex used for the military and the early breeders). Since the vitrification section is called T7, I guess the liquid interim storage section must be T6.

The overview drawing in figure 1 was made by myself (except for the little pics), the fuel unloading picture in figure 2 is from a Greenpeace report, primary source: Cogema, the continuous dissolver and vitrification drawings (figures 3 and 5) are from an article by C. Madic of the CEA (major owner of Cogema), and the PUREX principle drawing in figure 4 is a modified scan from a public brochure of the Kernforschungsinstitut in Karlsruhe. Most specific information given here is based on publications by the CEA, the UKAEA, the KFK, and the Cogema reprocessing contracts.

Figure 5: AVM continuous vitrification process

