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REPROCESSING RTR FUEL IN THE LA HAGUE PLANTS

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ABSTRACT

Starting in 2006, research reactors operators will be fully responsible for their research and testing reactors spent fuel back-end management.
It appears that the only solution for this management is treatment-conditioning, which could be done at the La Hague reprocessing complex in France. The fissile material can be separated in the reprocessing plants and the final waste can be encapsulated in a matrix adapted to its potential hazards.
RTR reprocessing at La Hague would require some modifications, since the plant had been primarily designed to reprocess fuel from light water reactors. Many provisions have been taken at the plant design stage, however, and the modifications would be feasible even during active operations, as was done from 1993 to 1995 when a new liquid waste management was implemented, and when one of the two vitrification facilities was improved.
To achieve RTR back-end management, COGEMA and its partners are also conducting R&D to define a new generation of LEU fuel with performance characteristics approximating those of HEU fuel. This new-generation fuel would be easier to reprocess.

Introduction : Reprocessing, the long-term solution for RTR spent fuel back-end

Research and testing reactors (RTR) are used for nuclear applications in many fields, including medicine, with boron neutron capture therapy, industry, with gauges, detectors, and other devices, research and education with irradiators and calibration sources. All these applications generate radioactive waste, in particular spent fuel, whose back-end management has experienced many stops and starts in the past ten years.
Until the end of 1988, US obligated materials were subject to the "Off Site Fuels Policy" which required spent fuel to be returned to the United States and to be reprocessed there. Since this policy terminated on the 31st of December 1988, research reactors operators were then required to implement other management solutions.
At the same time, the Reduced Enrichment for Research of Test Reactors (RERTR) Program was leading to a new Low Enrichment Uranium (LEU) fuel to replace High Enrichment Uranium (HEU) fuel.
Since new LEU fuel was not as easy to reprocess as HEU fuel (the LEU fuel is made of silicide, whereas the HEU fuel is made of aluminide), a new US spent fuel return policy was introduced in early 1996 for all research reactors converted (or that had agreed to be converted) to LEU, and for reactors operating with HEU for which no suitable LEU was available. This policy covers all the spent fuels discharged until the 12th of May, 2006. After that date, each operator will again be fully responsible for its spent fuel.

For the ultimate back-end management, there are three options [1] :

① Interim storage
② Direct disposal
③ Treatment-conditioning by reprocessing

The interim storage option does not constitute a reliable solution, while some research reactor operators have been confronted with corrosion and material degradation problems in existing facilities. Extended storage of RTR fuel would obviously require extensive R&D programs, as well as new facilities designed for long term storage. Most significantly, this option does not provide a definitive solution.

The direct disposal option entails several unsolved difficulties. First, one has to ensure that the enriched uranium content will not lead to criticality hazards through long term processes as selective leaching. Moreover, RTR spent fuel is generally considered as unstable (high corrosion rate, hydrogen build-up) under the conditions of a geological repository. It requires watertight and durable conditioning on a geological time-scale, for which no satisfactory solution has yet been found. Finally, direct disposal remains a "virtual" solution that has never been implemented.

The reprocessing option avoids the above difficulties because it produces residues which are suitable for direct disposal. The 30 years of experience gained at COGEMA's reprocessing site in La Hague demonstrates the industrial expertise achieved in commercial reprocessing.

Principles of reprocessing : the example of La Hague

Reprocessing has two main objectives :

① Recover the recyclable materials (plutonium and residual uranium),

② Generate final waste according to their potential hazards, in order to dispose them safely for the environment.

Reprocessing at the La Hague complex, in the UP2-800 and UP3 plants, uses the PUREX process, including the following steps (see flow sheet in figure 1) :

- *Transport* of fuel to the plant and *cooling* in storage ponds. This cooling, or "deactivation", decreases the radioactivity of the fission products substantially.
- *Shearing* and *dissolution* of the fuel, followed by *clarification* of the liquor generated :

  The first reprocessing operation consists of stripping the fuel rod to prepare it for chemical attack. The process employed for zircalloy cladding rods is cutting them into pieces with a shearing machine. At La Hague, as in many reprocessing plants, the shearing machine is placed above a continuous dissolver, and the rod pieces fall into a perforated basket, which, placed in the dissolver, allows the selective dissolution of the oxide in nitric acid without attacking the hulls. At the end of the operation, the hulls are removed and sent for storage awaiting future compacting and conditioning as a solid High Active Level Waste (HALW).
  Depending on the fuel type, it is possible that some insoluble products may remain after dissolution. This is the case for example for oxide fuels for which the insoluble particles are made of cladding residues and metallic inclusions. These solids, which would hamper further purification steps, are removed from the solution by centrifugation. A centrifugal clarifier has been selected for La Hague because it provides good efficiency with high throughput.
- *Uranium and plutonium splitting and purification* by a liquid-liquid extraction process :

  Basically, extraction consists in transferring a solute from one liquid phase to another one that is not miscible with the first. This operation enables separation of salts whose suitability for extraction by a given solvent is different.
  For the extraction of uranium and plutonium, tri-butyl phosphate diluted in hydrocarbons has been universally adopted. In the extraction operation, most of the fission products and actinides, except U and Pu, remain in the aqueous phase. Scrubbing by nitric acid improves the separation by stripping most of the fission products entrained by the solvent.
  Several extraction cycles of the clarified liquor, in pulsed columns, mixer-settler banks, or centrifugal extractors are necessary to meet the end-product specifications. At the end of these cycles, different kinds of solutions are generated :
  - a solution containing specifically the uranium
  - a solution containing specifically the plutonium
  - raffinates containing the fission products and the minor actinides
  - the solvent, which is regenerated by a treatment with sodium carbonate followed by caustic soda, and then recycled.

- *Final conversion of uranium and plutonium* to end-products :

  The uranium solution is concentrated by evaporation, stored, and eventually converted to UF6 for a new isotopic enrichment.
  In the same way, the plutonium is precipitated as an oxalate salt by the addition of oxalic acid. This salt is then filtered, dried and calcinated to form the PuO_2 oxide that is used to make the MOX fuel. The mother liquor is concentrated and recycled.

- *Management and treatment of process waste :*

  The process waste comprises :

  - The hulls, provided during shearing and dissolution operations, which are stored, awaiting for compacting in a canister, and intended for final disposal.
  - The High Activity (HA) liquid waste made up of solutions containing :
    - the insoluble particles from the clarification (fines),
    - the fission products and minor actinides split out during the extraction process,
    - the concentrates generated by evaporation of the aqueous acidic process sewage in an acid recovery unit. Acid generated in this unit is recycled in the process, and distillates, with very low activity, are discharged into the sea.

    The various streams, except the fines, are concentrated and generate the HALW concentrates which are stored in large vessels fitted with cooling and pulsation devices. The concentrates are then mixed with the fines and treated in a vitrification facility to form a glass matrix with high resistance to leaching. Today, this matrix appears to be the most suitable and safer packaging for long term storage.
  - The gases, which are collected according to type and level of activity, washed and treated on specific traps to recover elements such as iodine, and then filtered through high efficiency filters before discharge through a stack.

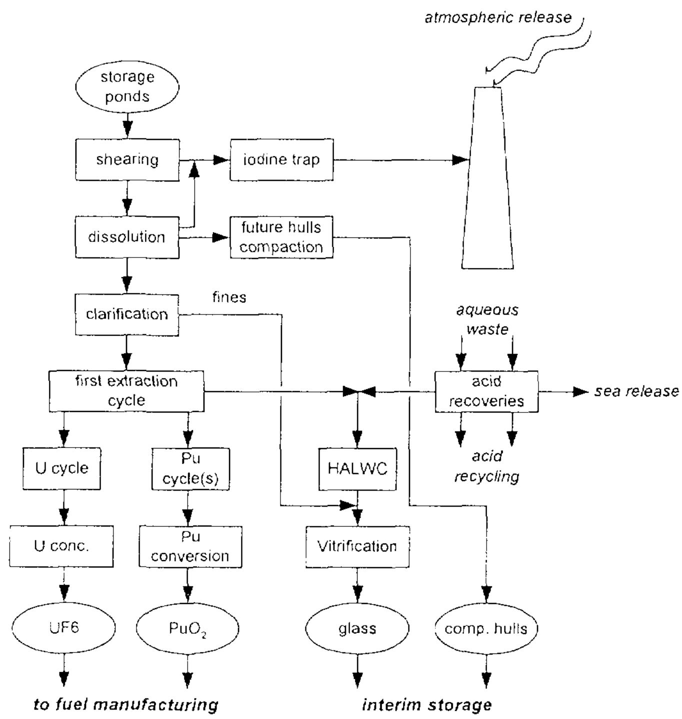

Figure 1 : LWR process scheme at La Hague

Reprocessing plants such as La Hague are designed to be operated for very long periods. During their lifetime, they will have to reprocess fuel with changing characteristics, although they were primarily designed for fuel from light water reactors. That can include LWR with an increased burn-up, but also new fuels such as RTR which are highly diverse in terms of weight, shape, and composition, and therefore require high flexibility of back-end services. Fortunately, in La Hague, many design margins have been provided, so that significant modifications can be incorporated, even in the active part of the plants, as shown in the following two examples.

Two examples of La Hague design flexibility [2]

Implementation of a new liquid waste management process

Active operation of UP3 started in 1989. The initial liquid waste management process led to the production of two types of solid waste : high level active waste (glass canisters) containing the bulk of activity, and intermediate level waste (issued from medium and low level liquid waste generated mainly by gas scrubbers, rinsing of equipment and solvent regeneration, and which underwent a treatment by co-precipitation, with the sludge generated incorporated in bitumen).

The excellent decontamination performance of the first extraction cycle opened up the possibility of modifying the liquid waste management process in order to eliminate production of intermediate level waste. The main characteristics of this new process were :

- Concentrate all salt-free acidic waste and the most active salt-bearing waste, in order to route it to the vitrification facilities,
- Release in the sea the remaining salt-bearing waste, after a filtration stage, and control of its activity.

To implement this new management process, three new evaporation units, a specific unit for the treatment of laboratory waste, and new piping and vessels were installed. The main challenges were :

- To have the radiation exposure of construction personnel negligible,
- To lay out most of the new units in facilities already in active operation, which involved difficulties for the active area access and for the piping connections,
- To perform the construction without any interference with operations of existing facilities.

These challenges were overcome mainly thanks to the original design of the La Hague plants, and to the excellent co-ordination between the design teams and the construction teams.
The lay-out of UP3 facilities, as well as those of its sister UP2-800, is characterised by a central active gallery, which houses all connecting pipes between various units. The active cells are located on each side of this backbone, and surrounded by inactive areas. The process equipment is installed in active cells separated by concrete walls. The equipment is gathered according to function and activity level, to enable an easier access.
In the case of a cell with medium or low level activity, draining and rinsing of equipment is sufficient to permit maintenance. In the case of a cell with high-level activity, most of the active pipes and transfer systems are duplicated. Hold-up pipes, most of which are fitted with decontamination nozzles, are provided for connection to future process extensions. In addition, extra space has been foreseen to house possible process extensions, and inactive areas have been designed with large corridors to make maintenance easier.

These conditions enabled :

- implementing 10 new active cells, both in existing facilities and in extensions,
- creating new piping between units inside the same facility and between facilities,
- minimising maintenance in active cells both in term of space and duration,
- limiting radiation exposure of construction personnel to about 10 man-mSv per year during the main construction period from 1993 to 1995.

Improvement of the vitrification facilities

La Hague has two vitrification facilities, R7 and T7, which are located respectively in UP2-800 and UP3. The process includes first converting the HALW solutions to a solid form in a rotary calciner and then vitrifying the solid in an induction-heated metallic melter.
Each facility has three vitrification lines capable of producing 25 kg/h of glass. Keeping two lines in operation and one on stand-by meets the production requirements and provides considerable flexibility.
R7 entered active service in 1989, and T7, three years later in 1992. The design of T7 took advantage of R7's experience, by including some improvement, identified on R7 but which could not be implemented in active conditions :

- implementation of a new connecting device between the pouring nozzle and the canister, associated with an improved off-gas system,
- addition of an in-cell washable pre-filtering device on the ventilation of main hot cells, in order protect the filters from contamination,
- improvement of the canister decontamination device,

- modification of the cranes to improve the reliability of certain components and reduce their maintenance.

In addition, T7's operators had the unique opportunity to train on R7 before T7's active start-up. As a consequence, T7 was able to achieve its production goals quickly, and the modification mentioned above proved to be very beneficial in terms of reduction of operating costs, reduction of waste volume, and personnel irradiation exposure.

In early 1994, it was possible to remove R7 from service for one year, since most of the backlog of HALW solutions had been vitrified, and the decision was taken to upgrade R7 to the same level as T7, by implementing the same improvements.
The project, which included in-cell operations in the most active part of the plant, was feasible thanks to careful preparation. In particular, all the most difficult operations were rehearsed beforehand using inactive mock-ups.The main objective during the project was to minimise the personnel exposure, and the careful training on inactive mock-ups was an essential element to reach it. The other two goals were to minimise the waste volume and to comply with the deadlines to avoid interfering with the production schedule of the facility.
The work was conducted in two steps. During the first one (5 months), only one line was stopped, while vitrification operation continued on the other ones. During the second step (11 months), all the lines were stopped. These modifications in active conditions were possible thanks to the design principles of the vitrification facilities, in which all equipment is installed in mechanical cells and can be assembled and disassembled using overhead cranes, master-slave manipulators and remote controlled tools.

RTR back-end industrial implementation

To be reprocessed, RTR fuel must be diluted in power reactor UO_2 spent fuel to lower the ^{235}U content to a maximum of 2 %.

La Hague is already available to reprocess aluminide HEU spent fuel, since head-end modifications – especially in the shearing and dissolution units- are provided. Some adaptations may also be necessary, on a case-by-case basis, to accommodate specific characteristics of each HEU fuel type. However, as indicated above, the flexibility of the plant would enable such modifications and adaptations, even during active operations.

It is not the same for silicide LEU fuel, which is not easy to reprocess. COGEMA therefore teamed with other partners in R&D programs to develop a new generation of LEU fuel that should :

① provide higher performance characteristics than current silicide LEU fuel,
② be easy to reprocess in existing industrial reprocessing plants, thus providing the best response to long-term management concerns.
③ enable as yet unconverted HEU reactors to switch to LEU fuel, in accordance with non-proliferation commitments.

Conclusion

Apart from treatment-conditioning by reprocessing, none of the spent fuel management options available for RTR operators is satisfactory. Due to the nature of the fuel, long term interim storage or direct disposal would require extensive R&D programs and specially designed costly facilities.
However, the treatment-conditioning of RTR spent fuel through reprocessing is a proven and fully operational management scheme. By separating and recycling the fissile material, it enables isolating the final waste which is encapsulated inside an adapted matrix according to the potential hazards.
COGEMA can reprocess spent RTR fuels in its La Hague reprocessing plants. Only some modifications are required, due to the specific characteristics of each RTR fuel type (both HEU and LEU), but the La Hague complex has already demonstrated its capacity to perform modifications, sometimes extensive, even after the start of active operations.

References

[1] "RTR Spent Fuel Treatment and Final Waste Storage", J. THOMASSON, RRFM 2000

[2] "COGEMA Reprocessing Complex : Already 9 Years of Operation, Mature and Flexible", JL DESVAUX et al, RECOD 98