THE R7/T7 VITRIFICATION AT LA HAGUE: 10 YEARS OF OPERATION

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Abstract

Vitrification of high level wastes from reprocessing of spent nuclear fuels has been carried out at La Hague on an industrial scale for ten years. This paper presents an historical overview of the facilities, and describes the facilities and their operations, startup performance; facility upgrading that has been done, and process control functions. The paper concludes that the technology for vitrification of high level wastes is mature and has been mastered.

1. INTRODUCTION

Reprocessing of spent nuclear fuels is the industrial operation through which valuable materials — uranium and plutonium – are sorted out prior to their reuse in fresh fuels. High level waste, i.e. fission products and actinides are separated and incorporated in glass matrix in order to be safely stored and disposed of.

1.1. The choice of glass

During reprocessing, the separated high level waste arises in the form of an aqueous nitric acid solution. The storage of the solution features some constraints as “hot spots” which necessitate a constant monitoring of the tanks. Therefore, it is necessary to solidify the waste for interim storage and final disposal. In that respect, glass is a prime candidate because it features suitable characteristics, particularly in terms of mechanical and thermal stability, radiation resistance, high containment capacity, low volume, low leachability and volatilization, easy fabrication (mastered technology), and flexibility with regard to the composition of waste to be conditioned.

The development of suitable glasses has been carried out in several countries (e.g. France, Japan, Germany, USA, Great Britain and The Commonwealth of Independent States — CIS) since the early 60's. There is a worldwide consensus on the choice of borosilicate glass as being the best matrix for immobilizing fission products and actinides in a solid form. The main components of the so-called glass frit are silicon oxide, boron oxide and aluminum oxide as glass forms and sodium oxide as glass modifier. The glass has been preferred to a ceramic type matrix, which was not expected to be as flexible in terms of incorporation of the various fission products.

1.2. Historical overview

France has been a pioneer in developing vitrification processes. As soon as 1957, an R&D program was launched under the aegis of the French Atomic Energy Commission (CEA). Several types of matrices were investigated, among which crystalline materials, phosphate and borosilicate glasses. Due to their amorphous structure, glasses appeared to be prime candidate to immobilize the more than 30 different chemical elements present in the highly radioactive liquid waste solution in the vitreous lattice.

The first laboratory scale unit was named Vulcain. It was commissioned in 1957 and was followed by a first vitrification pilot unit, Gulliver, commissioned in 1964. This early work culminated in the pilot-scale facility PIVER, featuring a single induction-heated pot where the three operations of evaporation of the high level waste (HLW) solutions, calcination of the residue and glass elaboration were performed. PIVER operated successfully from 1969 to 1973 to produce 12 tons of glass containing 185.10^3 TBq of activity. PIVER resumed operation in 1979 to vitrify HLW from the reprocessing of fast-breeder fuels. It was decommissioned between 1988 and 1990.

In parallel, separate calcination was investigated. The AVM (Atelier de Vitrification de Marcoule) was the first in line vitrification facility which allowed to valid the two-step French vitrification process consisting in the conversion of the HLW solutions in a solid form in a rotary calciner and then to vitrify it in an induction-heated metallic melter. The AVM started active operation in June 1978 and eventually was used to treat in-line the HLW solutions resulting from the reprocessing of UNGG (Uranium Naturel Graphite Gaz) fuels and research reactor in the UP1 reprocessing plant.

At mid-97, the UP1 plant stopped its reprocessing activity and entered in a first phase of rinsing prefiguring its final decommissioning. During this time-lapse, AVM is scheduled to vitrify all the HLW solutions produced by the rinsing and decontamination of UP1 equipment. So far the AVM has produced 2,731 glass canisters corresponding to 2,189 m^3 of fission products and 977 tons of glass. This figure represents an alpha and beta immobilized activity of $16{,}4.10^6$ TBq.

2. R7 & T7 LA HAGUE VITRIFICATION FACILITIES: A SUCCESSFUL OUTCOME

Beyond its industrial roles, AVM has been a unique opportunity to define major process and design choices for La Hague reprocessing plant. AVM's experience made clear that the French two-step process was a sound choice ensuring a continuous and safe production thanks to reliable process equipment and well adapted maintenance capabilities. Still, it was necessary to adapt the design of the new vitrification facilities to the plant's large-scale industrial dimensions.

2.1. Vitrification at La Hague: Teaming up for success

As for all other aspects, the erection and completion of the La Hague plant was a complete challenge in terms of planning, engineering and industry. With this plant, France entered a new era in industrial reprocessing. A successful power-sharing scheme was set up therefore between the CEA, SGN and COGEMA. Between 1972 and 1982, these three companies gathered their knowledge to make industrial vitrification at La Hague a reality. The CEA was in charge of the research and processes part, SGN was in charge of engineering issues and COGEMA was in charge of the operational aspects. This joint endeavor led to the characterization of a specific glass for HLW industrial vitrification.

2.2. Vitrification at La Hague: The characterization program

Long term studies performed by the CEA resulted in the formulation of the so-called R7 and T7 glasses. Characterization studies have verified the main properties of the final products and have established the glass specifications. The vitrified waste specifications were subjected to peer review by an independent commission of nuclear specialists. Characterization testing was conducted in both inactive and active conditions to determine the principal properties of the reference glass composition.

Inactive characterization testing focused on the physical, thermal, and mechanical properties of the glass, on its homogeneity, on the thermal stability of the glass and leach resistance.

Active characterization testing was conducted on hundreds of active glass formulations using alpha doped and beta tracer glasses to determine radiation resistance, leach rates and the thermal stability and volatility of the glass. In addition, tests were performed to assess glass quality sensitivity to variations in process parameters and to qualify a broad range of acceptable glass/waste compositions. A total of 90 glasses were characterized in this manner. In parallel with glass sensitivity studies, tests on inactive full-scale prototype were conducted to determine the sensitivity of the melter to variations in process parameters, such as melting temperatures. A range of acceptable glass compositions was defined on the result of the sensitivity tests, and failure modes and effects analyses were performed to identify fault conditions that would impact glass quality, including its chemical composition, homogeneity, cracking rate and propensity to crystallize.

The objective of the glass characterization program previously mentioned was to provide a reference glass composition and a variation range around, to cater for operational constraints and for actual fuel to be reprocessed. The glass characterization program resulted in identification of an optimum glass composition for HLW from oxide fuel (LWR type reactor). This was formalized in the "Specifications of Vitrified Residues produced from reprocessing at UP2 / UP3 La Hague plants". The specifications include "Guaranteed Parameters" i.e. those parameters identified as key parameters in the process to ensure the glass canister's quality. These specifications were subject to peer review by an independent commission of scientists and nuclear experts. They were also provided to ANDRA (French National Radioactive Waste Management Agency) for comment, and then submitted to the French regulatory authority, DSIN. The French safety authority approved the vitrified residue specification in February 1986.

2.3. Vitrification at La Hague: Facilities and operations

R7 and T7 are designed to produce 600 canisters a year, corresponding to 800 tU reprocessed with reference solutions. It is worth emphasizing that these design figures correspond to the range of fuels that can be reprocessed, characterized from two reference fuels of burnup respectively 33 000 MW.d/t with 3,5 % of ^{235}U (3 year cooling period) and 45 000 MW d/t with 3,7 % of ^{235}U (4 years cooling period). Ultimate wastes undergo vitrification after the cooling period. The process has proved to be flexible: fine particles from the dissolution step and alkaline effluents from the solvent regeneration steps are routinely incorporated to the glass matrix. From an industrial point of view, these facilities fully comply with production requirements. As of April 31, 1999, 3950 canisters have been produced at R7 and 2815 in T7. These figures are equivalent to 4,070 m^3 of fission products liquid solutions received at R7 and 1982 m^3 at T7.

Each facility R7 and T7 is made up of:

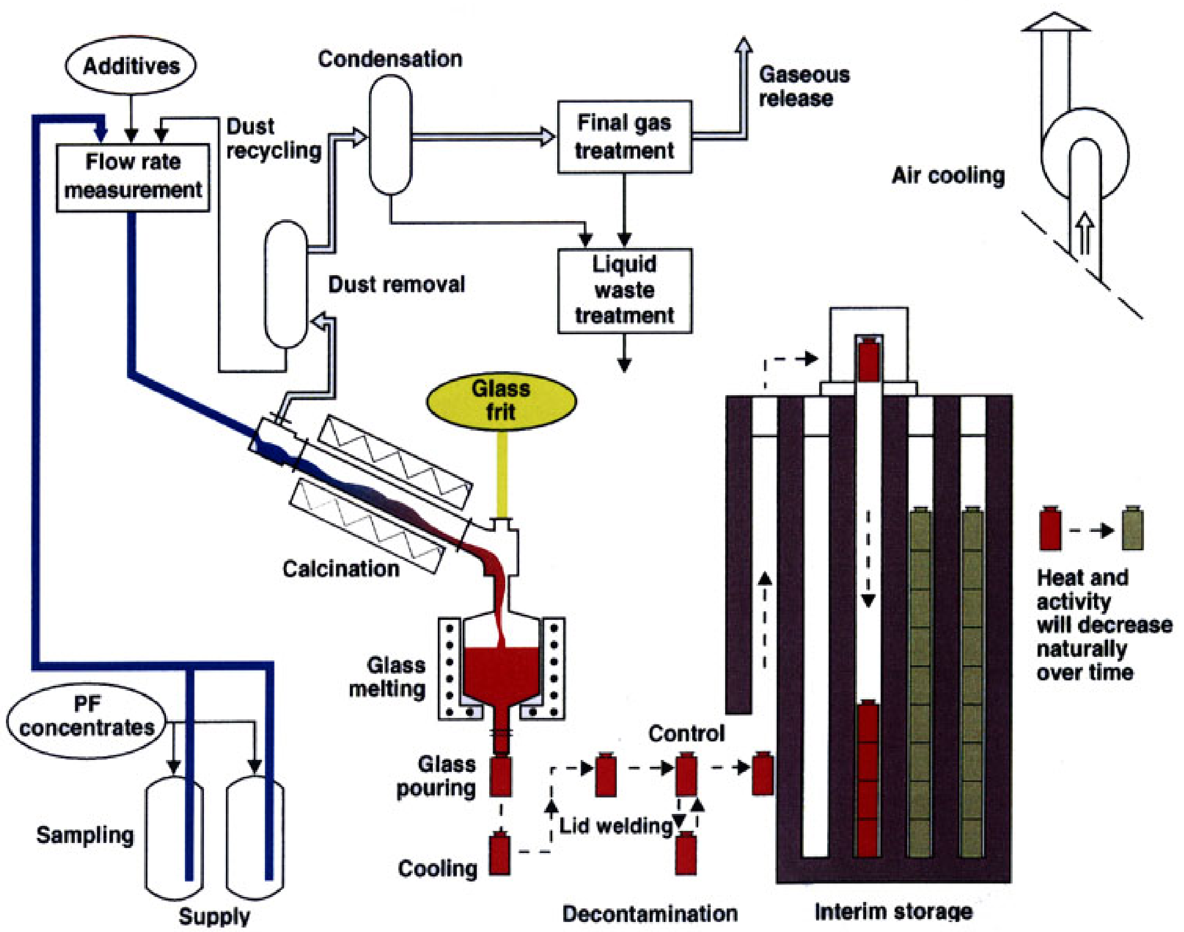
- Receipt and adjustment units.
- Liquid waste feeding, calcination and vitrification units. This unit features 3 lines. Each line is implemented in individual cells comprising 2 metering wheels (to feed adjusted fission product solutions and fines suspensions), a calciner to evaporate and to calcine these solutions, an ovoid and interchangeable melting pot heated by induction in which the mixture of calcinate and inactive glass frit are melted and then the glass is formed. Each line is designed to produce 25kg of glass per hour, corresponding to one canister filled every 16 hours. Currently each canister corresponds to more than 1,8tU reprocessed. Therefore, production is fitted regarding fuel characteristics.
- Conditioning units, where the filled canisters are cooled by air flow convection, welded (welding of the canister's lid), decontaminated by high pressure water, controlled by smear test before being stored in an interim storage facility.

- Off gas treatment units: the off-gas produced on each line are treated in a dust scrubber, condenser, NOx absorber, safety column and then filtered through 3 stages of HEPA filters. All the solutions issued from off gas treatment are recycled in the HLW process to be vitrified.

When compared to the AVM process, it should be underlined that to obtain the required capacity on each vitrification line of La Hague, the following solutions have been adopted:

- Scale up of the calciner to reach a design evaporation rate of 75 l/h (internal diameter: 0,35m; overall length: 4 m),
- Ovoid metallic crucible. This shape enables to improve the conduction/convection heat transfer, while giving access to an increased holdup.

Following is a description of the French two-stage continuous vitrification process in service at La Hague.



2.4. Shotblasting

In 1996, another step was added to the La Hague's vitrification process. Known as shotblasting, this operation is aimed at improving the visual aspect of the glass canisters and to shorten the decontamination process. Shotblasting is performed thanks to an air pressurized abrasive solution made up of water, alumina, and silica. It takes about one hour to "shotblast" one canister. Inactive and active tests were performed showing that the shotblasting process does not affect the canister material.

2.5. R7 and T7's maintenance aspects

The R7 and T7 cells are equipped with cranes; master slaves manipulators and shielded windows for remote maintenance. They are associated with hoist parking cells allowing crane maintenance as well as introduction of new equipment .In process cells, space agreements is optimized in order to ease access, modifications, and even addition of new equipment, and modular design of equipment allows partial replacement as much as possible. As result, maintenance operations are fully integrated in process operations, which is of utmost importance to minimize downtime and to increase availability for production.

3. STARTUP AND OPERATION PERFORMANCES

As said, more than 6,200 canisters have been produced at La Hague meaning that technical options were valid from an industrial standpoint. Following is some data about the facilities' startup and the main technical lessons drawn from it.

3.1. R7 startup

R7 entered active service in June 1989. Its first assignment was to treat the backlog of the HLW solutions accumulated since the start of the first La Hague reprocessing plant, UP2-400, in 1976. These solutions represented an important volume (about 1200 m^3), nearly saturating the HLW solutions storage capacities. The R7 main challenge at that time was to reach a sustained rate in short time-lapse. The R7 startup phase was a complete success since the fission products inventory began to decrease from the first campaigns.

This achievement is significant because operators had to deal with two notable issues:

- The crucible lifetime was lower than expected requiring frequent replacements,
- The containment at the connection between the pouring nozzle and the canister had to be improved. This mainly led to an increased activity of the cell ventilation filters in the beginning.

In spite of these challenges, fission product stock reduction and production goals were met thanks to easy maintenance operations.

3.2. T7 startup

The T7 facility is devoted to treat the HLW solutions produced by the UP3 plant. It entered active service in July 1992. The T7 design took advantage of R7's experience by including the following improvements:

- Implementation of a new connecting device between the pouring nozzle and the canister,
- Addition of an in-cell washable pre-filtering device on the ventilation of main hot cells,
- Modification of the cranes to improve the reliability of some components and to reduce the need for their maintenance,
- Improvement of the canister decontamination device.

In addition, T7's operators had the unique opportunity to make a training period on R7 before T7's active startup. As a consequence, T7 was able to reach very quickly its production goals, and the modifications mentioned above proved to be very beneficial in terms of operating costs reduction, waste volume reduction, personnel doses reduction, and availability. T7's first tests proved that the process was able to incorporate clarification fines and alkaline wastes into the glass without significant concerns.

3.3. R7 upgrading

At the beginning of 1994, the fission product solutions backlog from UP2-400 that R7 vitrified since its startup was exhausted. At mid-94, the decision was taken to upgrade R7 to the same level than T7 by implementing the same improvements. The project, which included significant in-cell operations in the most active part of the plant, was possible only thanks to a careful preparation work, which lasted more than one year.

In particular, all the most difficult operations (especially those located in limited-access cell even after decontamination) were rehearsed beforehand using inactive mockups. The main objective during the project was to minimize the doses to the personnel and the careful training on inactive mockups was an essential element to reach it. The two others goals were to minimize the volume of waste and to comply with the deadlines in order not to interfere with production schedule of the facility and the startup of the new UP2 800 plant. The work was conducted in two steps. During the first one, from February to June 1994, only one line was stopped, while vitrification operation continued on the other ones.

During the second step, from July 1994 to March 1995, all the lines were stopped and fission products were allowed to cool down in liquid tanks. The previous objectives were reached in totality: the doses to the personnel were 10 % lower than expected (themselves far below initial estimations), the volume of generated wastes as well as modification costs were very close to forecasts. R7 resumed operation on March 21st 1995, 10 days earlier than expected. The experience gained on T7 with the same improvement allows to forecast a return on investment (financial as well as radiological) of 2 or 3 years, thanks to the reduction of maintenance work and volume of high-active wastes generated during operation.

3.4. Melting pot lifetime

At the start of R7, the lifetime of the oval-shaped metallic crucible reached 200 hours. This result was due to the combined effects of thermal, electric, chemical and mechanical stresses applied to the pot. The induced corrosion led to a prohibitive replacement frequency, even if the design of the pot is well adapted to this operation. An important R&D work was carried out, involving experts in various fields (metallurgy, materials, and induction heat, heat engineering, fluid mechanics, and glass technology...).

Results were discussed and induced successive changes, taking advantages of periodical replacements of the melting pots without disturbing the production. These changes allowed a dramatic improvement of the crucible lifetimes. Today the average melting pot lifetime reaches more than 3000 hours (representing 4 to 5 melting pots per year) and the trend is going upwards as shown by the lifetime of some melting pots whose lifetime has reached 5000 hours for 190 glass canisters.

After being used, melting pots are cut into pieces and small pieces are decontaminated. In the next future, these pieces will be compacted alongside other waste. The main benefits of this extended lifetime are:

- The reduction of maintenance operations (for crucible replacements but also for washable pre-filters since the breaches of confinement during crucible replacements are the main source of cell contamination),
- The reduction of waste volume,
- The economical gain due to saved crucibles.

4. CONTROL OF OPERATION

The basic glass properties required for vitrified residue depend on containment capacity; radiation resistance, vitreous state stability and non-fixed surface contamination of the canister. Containment capacity is a function of alterability and leachability that are linked to the glass chemical composition, which should comply with the specifications previously defined. « Glass chemical composition » will be controlled as an essential process function.

4.1. Controls of process functions affecting vitrified residue quality

Radiation resistance depends on the vitreous state of the product whose quality is assessed from the viscosity and homogeneity of the glass. A good vitreous state is achieved if the melting temperature is correct and the pouring rate satisfactory. « Glass pouring » will be followed as a second process function. Vitreous state stability will be guaranteed if the temperature conditions supported by the glass after pouring are satisfactory. The process function « glass cooling » will be controlled. The main properties affecting the safety of vitrified residue intermediate storage are the non-fixed surface contamination of the canister and the canister tightness. These fourth and fifth process functions will be subjected to control.

With regard to the basic properties, the five process functions represent effective means to demonstrate the product quality on a real time basis. Process control is performed both directly, by monitoring and measuring operating parameters at various stages in the process, and indirectly, by corroborating operating parameter through analyses or comparisons of inlet and outlet materials balances.

The main operations covered by this program are:

- *Waste feed:* the fission product solutions and alkaline solutions are sampled and analyzed for free acid, dry extract, precipitate dissolution, activity content, heat release calculations, radionuclide concentrations, and chemical composition. The fines are rinsed, weighed, chemically dissolved and analyzed as well. Based on analytical results, the solutions and fines are adjusted as necessary to remain within the specified range for waste feed composition.
- *Waste feed homogeneity:* the waste feed is mechanically stirred in the feed makeup tank at a specified rotation speed. If stirring is interrupted, waste feed to the calciner is stopped.
- *Waste feed rate:* waste is continuously fed to the calciner by a measuring wheel, the parameters of which to ensure that the feed rate is within the specified range.
- *Melter temperature:* temperatures at various areas in the melter are monitored directly with internal thermocouples.
- *Glass mixing:* glass mixing in the melter is a major point in obtaining a homogenous glass product. It is achieved by inert gas bubbling in the melter.
- *Canister cooling time:* prior to lid welding, the canister must be stored and cooled in a cooling jacket for at least 24 hours, to avoid a thermal shock during high-pressure decontamination.
- *Canister lid welding:* canisters are sealed by plasma pulse arc welding. Welding operation parameters are continuously recorded to control their conformity to specifications.
- *Canister decontamination:* automated smear tests are performed on the entire surface of the canister to verify that surface contamination is less than 3.7×10^4 Bq/m^2.

4.2. Characterization of active samples

In addition, a glass sample characterization program was undertaken. Sample were taken on two occasions and were characterized and analyzed (chemical and radiochemical analysis, homogeneity before and after heat treatment, leaching behavior in static mode at 90°C and in Soxhlet mode). The results were consistent with the data calculated from the plant operating parameters and from routine samples taken during operation. The samples were submitted to a heat treatment supposed to induce devitrification. The crystalline phases observed were qualitatively and quantitatively identical with those of the non-radioactive reference glass. The maximum crystallization heat treatment had no effect on the leaching resistance. The material properties therefore comply with the operator's specifications and are comparable to those of the non-radioactive R7/T7 reference glass.

5. BUSINESS RELATIONSHIPS

As underlined previously glass specifications were accepted by French nuclear safety authority (DSIN). Subsequently, the R7/T7 glass specifications were also approved by the regulatory authorities of COGEMA's baseload customers in Germany, Belgium, Netherlands, Switzerland and Japan. In addition, all the customers have entrusted an independent agency (Bureau Véritas) with the responsibility of performing an independent survey (on a permanent basis) to evaluate COGEMA's measures of maintaining the specified quality. The *"Association Française pour l'Assurance Qualité"* certified that COGEMA's quality management is in compliance with international standard ISO 9002. This certification underlines the permanent care in the execution of tasks and the professionalism of the work force. The efficiency and reliability of the vitrified materials has been proven by five shipments already performed to Japan and Germany.

6. CONCLUSION

After an easy startup and 10 years of operation, there has been 6765 glass canisters produced in La Hague — La Hague's vitrification facilities are in line with the plant's annual throughput of 1600 tons per year. Overall French glass canisters production reaches 9500. Teething problems as melting pot lifetime are solved. Flexibility is also a constant priority as shown by the implementation of new liquid waste management, which has reduced releases. It is then possible to claim that the technology is mature and mastered. The glass product is well adapted; the returns of glass residues are soon due to reach a routine level. In France, the efforts are now borne on the final disposal project that should be ready for discussion in 2006. However, developments on vitrification have not stopped and there is an emerging and promising technology, already in use in the non nuclear field, that will have flexibility to adapt to complex feed streams: this technology is the cold crucible. Pilot tests are conclusive on this matter.