COGEMA EXPERIENCE IN OPERATING AND DISMANTLING HLW MELTER

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ABSTRACT

The vitrification of high-level liquid waste produced from nuclear fuel reprocessing has been carried out industrially for over 25 years by COGEMA in its three Vitrification facilities in France: Marcoule Vitrification Facility (AVM) and La Hague (R7 and T7).

So far, COGEMA’s vitrification facilities have produced more than 12750 high-level glass canisters, representing more than 5000 tons of glass and 167x10^6 TBq immobilized in glass. More than a technical success, in-line vitrification of HLW produced by operating reprocessing plants has become a commercial reality.

The vitrification process developed by CEA and operated by COGEMA is a two step Vitrification process where vitrified waste is obtained by first evaporating and calcining the nitric acid fed solution containing fission products, and calcinate is then fed together with glass frit into an induction-heated metallic melter.

The basic principles leading to the choice of the two-step vitrification process with hot induction metallic melter are:

- The separation of the functions, to have simpler and more compact equipment and to limit the size of the melter allowed for complete in-cell assembly and disassembly with moderate size overhead cranes, master-slave manipulators and remote controlled tools.
- The fact that the heating system is outside the metallic melter and thus independent from the melting pot, allow it to be not contaminated by High Active Level glass, to be not sensitive to the glass melting (no wear, no corrosion, no shorting and easy to start even if the metallic melter is full of glass), to be stopped and to be maintained easily.

In parallel, consistent, and long term Research & Development programs have enabled continuous improvement of the process. The average melter lifetime now exceeds the design basis value (5000 hours instead of 2000 hours). Feedback from operation has been used to optimize melter management and less than one week is now necessary to stop a vitrification line, to change the melter and to restart the vitrification operation.

The volume of secondary wastes from maintenance operations is minimized. Pieces of worn equipment are generally of small size and can be easily size-reduced for conditioning in accordance with the COGEMA solid waste management strategy.

This paper presents the COGEMA experience in high level waste melter operation, maintenance and dismantling.
INTRODUCTION

The vitrification of high-level liquid waste produced from nuclear fuel reprocessing has been carried out industrially for over 25 years by COGEMA, with two main objectives: containment of fission products and reduction of the final volume of waste [1].

Research performed by the CEA (the French Atomic Energy Commission) in the 1950's led to the selection of borosilicate glass as the most suitable containment matrix for waste from spent nuclear fuel. This led to the commissioning of the Marcoule Vitrification Facility (AVM) in 1978.

Based on the industrial experience gained in the Marcoule Vitrification Facility, the AVM vitrification process was implemented at larger scale in the late 1980's in the R7 and T7 vitrification facilities in order to operate in line with the UP2 and UP3 La Hague reprocessing plants. Both vitrification facilities are equipped with three vitrification lines having a nominal glass production capacity of 25 kg/h each. With two lines in operation and one in stand by, each vitrification facility of La Hague reprocessing plants was designed to have a glass throughput of 50 kg/h and to meet the production requirements with sufficient flexibility of operation.

So far, COGEMA's R7 and T7 facilities at La Hague have produced (by the end of October 2003) more than 9770 high-level glass canisters (CSD-V), representing more than 3900 metric tons of glass and 150x10^6 TBq immobilized in glass. More than a technical success, in-line vitrification of HLW produced by operating reprocessing plants has become a commercial reality that led, in 1995, to the first return of glass canisters to COGEMA customers.

THE R7/T7 PROCESS

The French Two Step Vitrification Process

The French Atomic Energy Commission (CEA) began research on the immobilization of HLW in 1957 and led to the choice of a two-step vitrification process implemented first in the Marcoule Vitrification facility (AVM-1978) and extrapolated it in terms of capacity and design for La Hague Vitrification Facility (R7-1989 and T7-1992).

Fig. 1  French Two Step Vitrification Process
In the two step process shown in Fig. 1, the nitric acid solution containing the concentrated fission products solution coming from reprocessing operation is adjusted in a stirred vessel to make its chemical composition compatible with specifications for the glass product. The solution is then fed to a rotary calciner where it is heated by four distinct zones in the range 600°C-800°C.

The calcine is heated under air and most of the nitrates are transformed into oxides. Aluminum nitrate is added to the feed prior to calcination in order to avoid sticking in the calciner (melting of NaNO3). Sugar is also added to the feed prior to calcination to reduce some of the nitrates and to limit Ruthenium volatility. At the outlet of the calciner, the feed is still in the oxidized state, with significant amounts of nitrates left. The calcine falls directly into the melting pot along with the glass frit which is fed separately.

The melting pot is fed continuously but is batch poured. The melting pot is made of base nickel alloys; The glass in the bottom of the melter is maintained heated to a temperature of 1100°C and is fully oxidized. The canister (CSD-V), which has a volume of 150 liters, is filled with two batches of 200 kg each. The maximum activity at the time of vitrification is about 28000 TBq per canister and the maximum contact dose rate of the canister at the time of production can be greater than 10^5 rad/h.

Off-gas treatment comprises a hot wet scrubber with tilted baffles, a water vapor condenser, an absorption column, a washing column, three HEPA filters and a iodine filter. The most active gas washing solutions are recycled from the wet scrubber to the calciner. The total off-gas decontamination factor is about 10^10.

**Glass Product Quality**

The R7/T7 glass was designed to hold, at the maximum, 18.5 % of radioactive waste oxides (fission products, actinides, noble metals and Zr fines), or equivalently an overall maximum waste loading ratio of 28 %. This limit was in fact set to avoid excessive heating of the glass during interim storage. The glass product has a very high activity content (predominantly 137Cs, 90Sr) and significant amounts of noble metals (3 wt % max).

During the qualification process for the R7 and T7 facilities, waste homogeneity has been demonstrated through grab samples during pouring and destructive examination of canisters. Homogeneity of the product was satisfactory and no undissolved feed was observed.

Satisfactory quality of the glass has also been demonstrated through the examination of production samples obtained in both the R7 and T7 facilities [2]. The glasses were homogeneous with no undissolved feed and their characteristics were in full agreement with the expected values.

The residence time of the glass in the melter is in the range of a few hours, which is enough for complete glass elaboration, provided that the temperature is sufficient.

The R7/T7 formulation is known worldwide to have an outstanding durability, especially in the long term. Normalized releases using a powder test very similar to the 7-day Product Consistency Test (PCT) are less than 1/10 of the US acceptability criteria.
R7/T7 VITRIFICATION GENERAL LAYOUT AND MAINTENANCE CONCEPTS

The basic principles leading to the choice of the two-step vitrification process and the multi lines design of La Hague vitrification facilities were:

- The separation of the vitrification functions,
- Easy remote maintenance of the process equipment.

The separation of the process functions (calcination/vitrification) led to simpler and more compact equipment which is always desirable in a highly radioactive environment; Easy remote maintenance of all process equipment allowed for complete in-cell assembly and disassembly with moderate size overhead cranes, master-slave manipulators and remote controlled tools.

From a more general point of view, main process equipment of each vitrification line are located in a separate cell, while pouring and cooling cells are common to the three lines. All of these cells are equipped with cranes, master slaves manipulators and shielded windows for remote maintenance. Parking cells allow crane maintenance as well as introduction of new replacement equipment. In the process cells, layout is optimized in order to facilitate access, modifications, and even addition of new equipment.

The process equipment considered to be the least reliable are designed to be modular (i.e the calciner), so that their main sub-components are relatively compact and easy to replace remotely.

The volume of secondary waste generated by maintenance operations is thus minimized. Pieces of worn equipment are generally small in size and can easily be size-reduced for conditioning in different wastes packages (glass type canister, cemented drum) according to their levels of activity. As a result, maintenance operations are fully integrated into the process design and method of operations, which is of utmost importance to minimize downtime, volume and activity of secondary waste as well as to increase availability for production.

THE INDUCTION HEATED MELTER

General Principle

The first work on vitrification of radioactive waste began in France in 1957 at the Saclay nuclear center. Techniques developed during this period to produce glass early used an induction-heated metal pot.

The melter consisting of copper coil inductors embedded in a concrete structure, is designed to have a very long life time and can be remotely dismantled. The melter surround the melting pot which is the only really consumable item.

The main advantage of this solution is that the heating system is outside the metallic melter (melting pot) and thus

- Independent from the melting pot,
- Not contaminated by HAL glass,
- Not sensitive to the glass melting (no wear, no corrosion, no shorting),
- Easy to start (even if the metallic melter is full or empty), to stop, to maintain or to replace.

Another major advantage of induction heating system is the simplicity to heat by Joule effect a metallic melter by using electric inductors.
From PIVER (1970’s), the first industrial-scale prototype unit in the world intended for vitrification of concentrated fission product solutions, through AVM (1970’s) and to R7 and T7 (1990’s), induction heating has been successfully and industrially operated by CEA and COGEMA.

**R7/T7 Induction Melter Description**

The separation of the calcination function from the melting function allows to limit the size melter with regard to the design capacity:

- The evaporative capacity of the R7/T7 calciner is of about 80 l/h.
- The power delivered to the melting pot is used to melt solid product (calcine and frit glass) and not to evaporate liquid.

As shown in Fig.2, the melting pot is ovoid (long axis 1 m, short axis 0.35 m, total height 1.40 m, weight around 400 kg), and is made of base nickel alloys.

The melter is supplied with 4000 Hz power by a 200 kVA generator at a voltage adjustable up to 600 V through four superposed copper inductors cooled by water and connected to the cooling circuit by flexible piping. Each inductor is supplied to allow individual power adjustment.

**Glass pouring**

The melting pot is equipped with two bottom pouring nozzles, also heated by induction, to fill glass canisters:

- The first is used for nominal melter operation to periodically fill the canister with 200 kg glass charge.
- The second ensures a complete emptying of the melting pot at the end of each vitrification campaign or for maintenance intervention, and thus participates to reduce the levels of activity of the secondary waste.

**Glass stirring**

Initially, the R7/T7 melters were only equipped with bubble stirring. However it was demonstrated by CEA that glass viscosity and [glass frit - calcine] reactivity are influenced by noble metals content. As a consequence, COGEMA has undertaken the development of a mechanical stirrer in 1994 and has deployed
first these mechanical stirrers on the T7 facility in 1996. The main objectives associated to the deployment of this new technology was to increase the noble metal content in the feed to the maximum specified noble metal content (3%) value and at the same time maintain the throughput capacity of the vitrification lines.

Melting pots are thus now equipped with mechanical devices that have been designed to be compliant with the melting pot life time. A particular R&D program has supported materials selection used for the mechanical stirrer.

Mechanical stirring proved to be successful in achieving the objectives since the noble metals content in the glass was increased from an initial 1.5 wt. % to 3 wt. % (the maximum value) without any pouring problems or any metal accumulation in the bottom of the melter, with respect to the expected glass throughput.

**Control and monitoring**

*The instrumentation installed in R7/T7 melting pot enables measurements of* level and temperature of the molten glass and temperature of the melting pot’s wall (important for glass quality and melter operation).

Since 1995, the glass level in R7/T7 vitrification melting pots has been directly measured. There is a perfect correlation between this level measurement and the material balances determined with the crucible feed equipment. Factory calibration of the melter enables conversion of the level measurement into a mass of glass.

The type of thermocouples used in induction heated crucible facilities at La Hague have been the subject of careful engineering and constant improvement.

A thermocouple characterization and evaluation program including:

- Long duration endurance tests at constant temperature (1150°C) and with numerous temperature cycles (more than 2000),
- Integration of substantial doses of irradiation,

have been carried out and has validated the type of thermocouples currently used in the La Hague vitrification melting pots.

A continuous monitoring of the melting pot temperatures and of the electrical induction parameters are used to operate the melting pot and to follow the melting pot ageing and wear, in order to prevent failure.

**Melter life time**

One of the major on-line developments undertaken on the melter has been the work performed to extend the melter's lifetime. At the start of operations in the R7 facility, the lifetime of the oval-shaped metallic melter was less than the design basis value (2000 hours) due to the combined effects of thermal, and mechanical stresses as well as corrosion in the gaseous phase. An important R&D program was launched to extend the melter life. Comprehensive studies were performed in order to better understand the electromagnetic, thermal and mechanical behavior of the melting pot at the different stages of operation as well as the corrosion mechanisms at play. In particular, the power transfer from the inductor coils to the melting pot wall was analyzed in detail.

These studies helped to determine which species were responsible for corrosion. They also showed that the thermal power released in the melting pot's wall and therefore the temperature gradients in the melting pot could lead to high levels of stress as well as condensation of corrosive species. As a result of these studies, the design, the material and the operation of the melter were modified.
These changes led to a sharp increase in the lifetime of the melters. At present, after 15 years of operation, the lifetime of the standard melting pot exceeds the design basis values by more than a factor of two. The average melting pot lifetime is about 5000 hours / 150 glass canisters, with an record at 6400 hours corresponding to more than 200 hundred glass canisters produced with a single melter.

Today one melting pot per year and per vitrification line is used in R7/T7 vitrification facilities, which is having a great positive impact on process downtime and secondary wastes management.

MELTER REPLACEMENT AND DISMANTLING

The R7 and T7 are mature vitrification facilities where operation and maintenance principles have been optimized with two main objectives:

- Maximize production availability,
- Minimize volume of secondary wastes.

Even if efforts have been made to avoid the need for maintenance (e.g. by improving the reliability of equipment), some equipment, because of its nature or complexity, needs to be periodically maintained or replaced. A specific effort has been undertaken to facilitate their maintenance and minimize intervention duration.

Melting Pot Changing

The R7/T7 melter, is thus designed to be small, compact, easy to replace remotely, and consequently inexpensive. As a result, melter maintenance operations are fully integrated into the process design and method of operations.

The melting pot replacement is based on preventive maintenance to change equipment before failure in order to minimize the number of component or sub-components to be changed (melting pot, melters…), and the level of component’s contamination.

The large experience gained by COGEMA’s vitrification operators, and the continuous control of the melting pot operation (temperature, electrical induction parameters, analysis of small drift) allows us to anticipate and to change equipment before fatal wear; this management is a mix between ‘periodical exchange’ and ‘management value’.

The main source of preventive intervention is metallic pot wear (corrosion, thermal and mechanical constraints…). When the decision to change a melter is made, the melter is emptied from its molten glass and the vitrification line is progressively stopped. When the melting pot is cold, it is disconnected from the calciner, and the melter, which is on a mobile cart, is moved back. As shown in fig.3, it is then possible to get the melting pot out of the melter, to transfer it to the dismantling cell (located above the vitrification cell) and to replace it with a new one. Approximately ten days are necessary to stop and restart a vitrification line with a new melting pot.
Melting Pot Dismantling

Throughout the COGEMA complex, and especially at the La Hague facilities, effort have been focused for several years on the minimization and rationalization of the volume of conditioned solid waste coming from maintenance operations (contaminated tools, replaced components, etc.) into standardized waste packages. For example the vitrified residue (CSD-V) and compacted residue (CSD-C) packages have the same external design allowing:

- Standardized handling operation (same devices to handle either CSD-C or CSD-V canisters),
- Simplified transport operation (same shape of transportation cask),
- Interim and long term storage if needed (additional possibility of mixed storage CSD-C/CSD-V).

These efforts encompass all the aspects of waste generation and management, from plant and equipment design, to the development of specific repair techniques or to the implementation of rigorous sorting and decontamination of the waste at the source especially for materials in contact with HLW.

Vitrification dismantling cells are equipped with dismantling and decontamination tools, a counting cell, and a waste preconditioning unit. After dismantling and decontamination operation, pieces of worn equipment are generally of small size and sorted according to their activity level and material nature to be either recycled inside the process, and conditioned in CSD-V type glass canister, or sent to La Hague waste treatment facility (AD2) to be cemented as technological waste, or, at soon compacted in the new La Hague Compaction facility (ACC) according to their level of activity (Low Level/ Intermediate Level).

For the melting pot, the dismantling operation consists of immobilization of the melter in a vice and cutting it into small pieces of 300 x300 mm by means of a sawing machine. As shown in Fig.4
Generally several blades (three to five) are necessary to completely dismantle the melting pot in small pieces.

The mechanical saw alternative movement participate also to separate possible residual cold glass from metallic pieces and contribute to downgrade the waste. A complementary mechanical cleaning of metallic piece could be performed.

Approximately 6 to 10 days of continuous operation are routinely necessary to dismantle a melting pot.

Metallic pieces are decontaminated in high temperature nitric acid tank, counted in a measurement cell, and sorted and preconditioned in Intermediate Level Waste basket (650 mm diameter, 1 m height). Three Intermediate Level Waste baskets are necessary to precondition a melting pot and associated worn saw’s blades.

To finish the waste conditioning, ILW technological baskets are routinely sent by mean of mobile shielded cask to the AD2 facility to be cemented and conditioned as technological waste in CBF-C2 residue (1200 liters volume, 4 metric ton max in weight). Generally three 500 Ci CBFC2 are necessary to condition a melting pot and its auxiliaries (i.e saw’s blades, current maintenance tools).

Soon, a second way for ILW basket conditioning will be implemented to send it (according to their activity) to the new Compaction Facility (ACC) to reduce by factor 4 the final waste volume by using universal standard canisters (CSD-C).

The recovered glass coming from dismantling melting pot operation are conditioned in HLW basket (380 mm diameter, 1 m height) to be recycled in a specific standard vitrified canister (CSD-V).

Melting pots are dismantled in line with the process in La Hague vitrification facilities according to the La Hague waste management strategy, allowing an optimization and standardization of the final waste volume.
Because of the small number of melting pots to be dismantled each year, the treatment of melting pot in dismantling cells is a routine and short time operation fully integrated into the ordinary vitrification facility operations.

CONCLUSION

COGEMA has been operating industrial HLW vitrification facilities for over 25 years. The feedback from hot operations and the long-term R&D programs conducted with the CEA have helped to continuously improve the process in all of its aspects including glass formulation, process, associated technologies, and operations and maintenance.

The R7 and T7 vitrification facilities operating in-line with COGEMA’s two major commercial reprocessing plants have had outstanding records of operation, not only from the standpoint of total glass production and plant availability but also with respect to safety, remote in-cell maintainability, and secondary waste generated, demonstrating the maturity of the French vitrification process.

With respect to the COGEMA layout and maintenance concepts, the melter induction technology used in R7/T7 vitrification has been designed to be small, compact, reliable and easy to replace remotely. As a result, melter maintenance operations are fully integrated into the process design and method of operations, and lead to optimized maintenance time intervention and volume of secondary waste.

REFERENCES

1 G. Mehlman, R. Do Quang, A. Jouan, "Major breakthroughs in high level waste vitrification"; Waste Management ’99, Tucson