

## INTEGRATED PILOT PLANT FOR A LARGE COLD CRUCIBLE INDUCTION MELTER

R. Do Quang, A. Jensen  
COGEMA  
2, rue Paul Dautier BP 4  
78141 Vélizy Cedex, France

A. Prod'homme, R. Fatoux  
SGN  
1 rue des Hérons – Montigny le Bretonneux  
78182 Saint Quentin en Yvelines Cedex, France

J. Lacombe  
CEA – Valrho BP171  
30207 Bagnols-Sur-Sèze Cedex, France

### ABSTRACT

COGEMA has been vitrifying high-level liquid waste produced during nuclear fuel reprocessing on an industrial scale for over 20 years, with two main objectives: containment of the long lived fission products and reduction of the final volume of waste.

Research performed by the French Atomic Energy Commission (CEA) in the 1950s led to the selection of borosilicate glass as the most suitable containment matrix for waste from spent nuclear fuel and to the development of the induction melter technology. This was followed by the commissioning of the Marcoule Vitrification Facility (AVM) in 1978. The process was implemented at a larger scale in the late 1980s in the R7 and T7 facilities of the La Hague reprocessing plant. COGEMA facilities have produced more than 11,000 high-level glass canisters, representing more than 4,500 metric tons of glass and 4.5 billion curies.

To further improve the performance of the vitrification lines in the R7 and T7 facilities, the CEA and COGEMA have been developing the Cold Crucible Melter (CCM) technology since the 1980s. This technology benefits from the 20 years of COGEMA HLW vitrification experience and ensures a virtually unlimited equipment service life and extensive flexibility in dealing with different types of waste. The high specific power directly transferred by induction to the melt allows high operating temperatures without any impact on the process equipment. In addition, the mechanical stirring of the melter significantly reduces operating constraints.

COGEMA is already providing the CCM technology to international customers for nuclear and non-nuclear applications and plans to implement it in the La Hague vitrification plant for the vitrification of highly concentrated and corrosive solutions produced by uranium/molybdenum fuel reprocessing.

Preliminary testing with several types of Hanford waste has indicated that the CCM technology is particularly well adapted to the processing needs of the various nuclear waste (LAW, TRU, HLW) streams in the United States, as a result of its flexibility with respect to waste composition and its high throughput, while maintaining a small and compact design. In order to support the development of the CCM for commercial applications on DOE sites in the United States, and to demonstrate the maturity and advantages of the CCM technology to treat various wastes and to reduce the final waste volume by increasing the waste loading (thanks to a higher temperature process), the CEA and COGEMA have jointly built a new large-scale demonstration pilot plant which integrates feedback from existing CEA CCM platforms and COGEMA industrial vitrification experience. This CCM platform is approximately half-scale with respect to the capacity requirements at the DOE sites, with a 50 kg/h capacity of glass in liquid feed (80 l/h flow rate with a maximum salt concentration of 500 g/l), and 200 kg/h with solid feed. The plant includes the CCM and numerous ancillaries, such as feed tanks with the corresponding feeding system, several possible trains of off-gas treatment (dry or wet), and other facilities, making it a powerful demonstration tool.

The paper presents the CCM project that led to the building and start-up of this evolutionary and flexible pilot plant. It also describes the plant's technical characteristics and reports commissioning results.

## COGEMA'S INDUSTRIAL EXPERIENCE WITH HLW VITRIFICATION

### Industrial Facilities in Operation

COGEMA has been operating facilities that vitrify high-level liquid waste produced during spent nuclear fuel reprocessing on an industrial scale for over 20 years, with the dual objective of containment and reduction of the final volume of waste. On the basis of experience gained in the early 1970s in the Marcoule vitrification facility (AVM), the process was implemented in the late 1980s in the R7 and T7 facilities of the La Hague plant (also referred to as AVH).

High-level liquid waste produced by the reprocessing of commercial spent fuel is vitrified using a standard hot crucible induction process at a temperature of 1,150 °C. The vitrified waste has a high radioactivity content (mainly <sup>137</sup>Cs and <sup>90</sup>Sr), as well as a high noble metals content.

COGEMA facilities have produced more than 11,000 high-level waste canisters, representing more than 4,500 metric tons of glass and 4.5 billion curies.

### Major Operating Milestones

COGEMA has been continuously improving the performance of the vitrification process currently used in the La Hague plant through consistent, long-term R&D programs [1]. After ten years of operation, the lifetime of the melters now exceeds their design basis values by a factor of 2.5, corresponding to the use of one melter per year and per vitrification line (six vitrification lines with 25 kg/h nominal glass throughput are operating in La Hague). A major and more recent development was the implementation in 1996 of mechanical stirring in the melters to deal with higher noble metals content in the waste.

## COLD CRUCIBLE INDUCTION MELTER TECHNOLOGY

### Main Technological Features

To further expand their scope of operation, COGEMA and the CEA have developed the cold crucible melter (CCM) technology. The cold crucible is a water-cooled melter in which the glass is heated by direct high frequency induction currents. This allows high thermal power to be directly released in the melt, and therefore high operating temperatures. Since the main process components are water cooled and protected by a solidified glass layer, the technology overcomes difficulties usually associated with high temperatures such as corrosion, and the melter has a virtually unlimited service life. High service temperatures, mechanical stirring of the melt and pouring from the bottom significantly increase flexibility in dealing with different types of waste and in selecting the most appropriate formulations for waste immobilization and for waste final reduction volume.

Since there is no upper bound on the operating temperature, the CCM can produce more durable glass formulations with reduced proportions of fluxes in the glass (especially alkalis known to be detrimental to glass quality) and increased proportions of glass formers. Such formulations would be too viscous for the standard melter technologies (hot induction or joule-heated ceramic melters), which are usually operated at a limited temperature of about 1,150 °C.

Furthermore, the technology has been especially developed for nuclear applications with respect to proven COGEMA design principles. The main process components are simple, compact, modular and easy to maintain remotely.

R&D on the cold crucible began in the mid-1980s. The process and technology developments were based on test platforms located at the CEA and benefited from COGEMA vitrification operating experience (the CCM is built with proven technology blocks, such as induction, pouring, measurement and stirring devices, material for nuclear environments, etc.). In the framework of these R&D programs, performance of the process has been demonstrated (capacity, high operating temperature, control of volatility, flexibility with respect to waste composition, etc.) and the associated technologies have been confirmed (pouring device, off-gas treatment

equipment, electrical power supply, process instrumentation, etc.). All of the results obtained to date are part of COGEMA's qualification program for the cold crucible technology.

### **Pilot Facilities Supporting the CCM Technology**

Several test platforms have been built in the Marcoule pilot facilities since the 1980s in order to develop the Cold Crucible Melter technology. These programs were initially focused on the treatment of HLW solutions from light water reactor fuel, producing the simulated R7/T7 glass.

The oldest platform is a stand-alone CCM pilot unit, which can be fed with simulated calcine and glass frit. The 550-mm diameter stainless steel melter can also be liquid fed. This platform, equipped with a 300 kW generator, has totaled more than 5,000 hours of operation over a 15-year period. Approximately 50 metric tons of simulated HLW glasses were produced. Since the beginning of the CCM programs, most of the process demonstrations have been performed on this platform with solid and liquid feed.

The second platform is a full-scale mock-up of the R7/T7 vitrification process that can be equipped with a hot crucible melter or a 650-mm diameter CCM (with a 300 kW generator). In either case, the melter can be coupled to a calciner and fed with simulated calcine and glass frit. This platform is currently used to qualify the CCM process as it will be implemented in the R7 facility.

A third platform called EREBUS is dedicated to the more recent applications. It is equipped with a smaller, 160 kW generator capable of operating at frequencies ranging from 200 to 500 kHz. It can be operated with melters of various diameters up to 1,000 mm. Selection of the diameter will depend on the type of test to be performed, while accounting for the limited available power. The feeding systems allow simultaneous controlled feeding of solids (frit, powders) and liquids (surrogate solutions, slugs). The off-gas system is composed of a condenser and an acid recombining/washing column before the extraction device.

A fourth independent platform is also used for development of the incineration/vitrification process with a cold crucible melter. This process is well suited for the immobilization of intermediate- or high-level organic waste such as the ion exchange resins produced by nuclear power plants. The organic matter is fed to the melter and is burned on the surface of the glass melt. The residual inorganic compounds are directly incorporated into the vitreous phase. The stainless steel CCM used on this platform has a diameter of 300 mm and is powered by a 240 kW generator. In order to finalize this development, a pilot industrial facility (equipped with a 550-mm crucible) has been built in collaboration with KEPSCO/NETEC at Taejon in South Korea [2].

### **Commercial Applications of the CCM Technology**

COGEMA is already providing the CCM technology to international customers for nuclear and non-nuclear applications. For example, the CCM technology has been in operation for non-nuclear applications since 1995 to produce high added-value glasses or enamels with two melters. Because of the cold glass protection, glasses or enamels can be melted at high temperature (up to 2,000 °C) with no pollution from the refractory elements used in traditional glass melters. It is also possible to switch glass compositions in less than 8 hours since glass does not adhere to the cooled walls. In one year, 500 tons of industrial glass or ceramic of varying compositions have been produced with a single 1,200-mm diameter CCM.

This technology will be deployed at La Hague, in one existing vitrification line, to process specific corrosive, high viscosity material in the near future. For this application, the advantage of high temperature has been fully used by raising the target processing temperature from 1,150 °C to around 1,350 °C, thus enabling the selection of a new matrix resulting in a decrease of the overall glass volume by more than a factor of 4.

One CCM coupled with a calciner has been proposed for the Hanford TWRS-P Phase IA HLW studies [3]. A large demonstration program, including the production of about 3 tons of surrogate glass in a pilot facility, provided confidence in the process and pointed to some possible advantages of further extending the range of conditions tested. More specifically, it was found that the technology displayed a potential for significant waste loading increases, thus reducing the volume of HLW glass to be disposed of. More recently [4], preliminary testing on the EREBUS platform with several Hanford waste types have confirmed interesting performance in terms of capacity (similar to or better than the values published for LFCMs and fully compatible with industrial objectives in the US), flexibility, and extension of the allowable composition domain.

This technology is also being supplied to foreign customers, with varying setups:

- CCM with direct liquid feeding to process legacy HLW in Italy;
- CCM with direct liquid or solid (resin) feeding for simulated reactor waste in Korea.

## ADVANCES IN CCM OPERATION AND TECHNOLOGY

### Basic Direct Liquid Feed Performance

With the flexibility of the CCM technology, the CCM can provide either a one-step (direct vitrification) or two-step (calcination/vitrification) process to treat waste streams more effectively.

The advantage of the one-step process is to eliminate a separate calcination operation and thus avoid all the calciner process (control of the chemical calcination function) and mechanical (maintenance, secondary waste, availability) constraints. However, joule-heated ceramic technology requires voluminous, heavy and expensive equipment that is difficult to replace and dismantle, particularly in case of large capacity. In contrast, separation of the calcination and melting function enables the use of small and compact equipment that is easy to replace and maintain with a low level of secondary waste.

The CCM can now combine the advantages of both processes with compact equipment, a simple process and large capacity in a single step. In addition, for direct liquid feeding, it benefits from the experience gained in operating the PIVER pilot facility at Marcoule. PIVER was the first industrial-scale unit in the world intended for vitrification of concentrated fission product solution and began operation in 1968. Based on a batch process capable of directly vitrifying 200 liters of fission product solution in 100 kg of glass in a cylindrical metallic vessel heated by induction, this pilot unit vitrified up to  $15 \times 10^6$  Ci.

Consequently, specific tests have been performed in order to identify limiting evaporation capacities by feeding a CCM with water. The following results highlight the beneficial temperature effect on melter capacity:

- Limiting evaporation rate, at 1,250 °C:  $100 \text{ l.h}^{-1}.\text{m}^{-2}$ ;
- Limiting evaporation rate, at 1,400 °C:  $150 \text{ l.h}^{-1}.\text{m}^{-2}$ ;

Moreover, direct liquid vitrification tests performed by the CEA and COGEMA, as part of a demonstration program for the treatment of Hanford HLW, confirmed the ability of this technology to achieve high performance. Accordingly, two surrogate solutions for Hanford HLW (respectively C106 and Az-Blend) were successfully vitrified by direct feeding with respective glass throughput capacities of about  $65 \text{ kg.h}^{-1}.\text{m}^{-2}$  and  $92 \text{ kg.h}^{-1}.\text{m}^{-2}$  (corresponding respectively to 125 and  $142 \text{ l.h}^{-1}.\text{m}^{-2}$  in liquid feeding) [4].

Previous values confirm that liquid-feeding capacities will be limited by the evaporation step for a fixed temperature but with the possibility to increase it if necessary. In addition, high process temperature and stirring operating parameters have been shown to be crucial in order to control or to counteract phenomena that occur at the melt surface, to efficiently maintain the operating temperature during the liquid feeding phase and to enhance the CCM direct liquid feeding capacity.

### CCM Technology Development for a Large CCM Pilot Plant

Given the excellent results achieved with the CCM technology, a large CCM plant directly fed with solutions has been developed. It corresponds to a new technological generation of vitrification melters, which is well suited for future COGEMA and CEA applications (flexibility of the products to be treated and capacity increase) and associates proven and optimized design with high treatment capacity in accordance with the needs identified for DOE waste vitrification.

COGEMA and the CEA then decided to design an integrated platform based on the large CCM technology on a sufficient scale to be used for demonstration programs. In terms of capacity, this pilot plant must be:

- Half-scale of what should be necessary for DOE needs (for the results to be easily extrapolated);
- Full-scale for COGEMA needs for its future applications.

A liquid fed melter with such a throughput capacity (about 50 kg of glass per hour) typically has a diameter of approximately 1.1 meter.

The large CCM is made of proven block technology based on COGEMA and CEA experience. It still remains modular and all of its sub-components are compact and remotely maintainable. The crucible and its supporting slab are the largest sub-components of the system and can be handled with standard equipment with a capacity of a few tons. All other sub-components (pouring valve, inductor coil, control systems on dome, off-gas exhaust systems, etc.) are much smaller and lighter. The general principle applied for maintenance is that the system can be completely disassembled from the top down with manipulators and cranes.

In addition, this large CCM technology benefits from most of the advances provided by the basic CCM technology (process instrumentation, high frequency power delivery system, off-gas treatment system, feeding, pouring, mechanical devices, etc.).

## INTEGRATED PILOT PLANT TO DEMONSTRATE LARGE CCM PERFORMANCE

### Preliminary Large-scale Technological Tests

Preliminary large-scale tests were performed on a 1,400 mm diameter CCM platform in 2000, in order to validate the electrical system, the design of a large melter and its behavior (i.e., the possibility to extrapolate thermal and heat transfer laws to large-diameter melters).

Every major sub-component of the CCM has been tested under actual service conditions. All parts of the electrical system (generator, line and inductor) has been shown to be reliable during each phase of the test (starting, melting and pouring phases). In addition, electrical parameters have been identified to substantially help with remotely operating and controlling the vitrification.

Finally, a test has been conducted on this platform by interrupting the water cooling of the CCM structures (crucible, bottom slab), with the melter containing more than 350 kg of melted glass, in order to simulate the potentially most severe accident conditions for such a cooled vitrification system. Despite the heating of the structures (see the IR camera view in photo 1 below), the CCM performed well. In particular, no serious structural damage was observed after the test. The CCM's integrity was preserved and no glass-leak was observed.

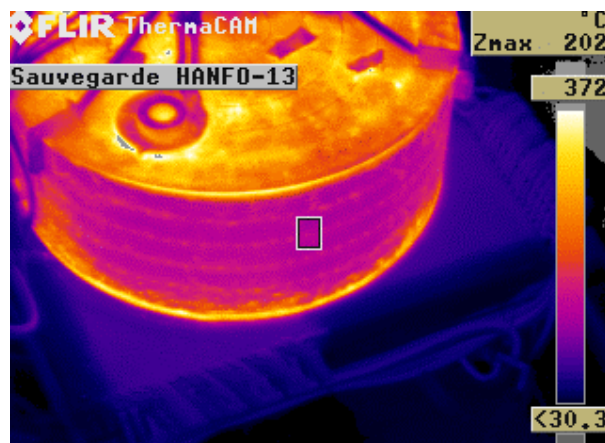


Fig. 1. IR camera view of the CCM during the “loss of cooling function” test

These tests provide high confidence that the large CCM technology will be able to process various high-level waste streams with high capacity and flexibility. This will be confirmed by operating the new integrated large-scale platform.

### Description of Pilot Plant Functionality

Operating experience and technological improvements have matured the design of the sub-components for the large CCM technology and small-scale processing tests have validated its potential performance. During 2000, COGEMA and the CEA identified the need for an integrated pilot plant for large CCM demonstration (application to US DOE needs, etc.).

This pilot plant has been designed to be modular and as flexible as possible in order to provide a powerful tool to demonstrate all vitrification system performance characteristics. It benefits from the up-to-date feedback from CEA CCM platforms and the COGEMA industrial vitrification experience. In addition, it integrates all functions of the process in order to provide a demonstration of the whole system on an industrial scale.

All the equipment – the preparation and feeding device, the vitrification system, the pouring room and the off-gas treatment system – were installed on four floors covering 160 m<sup>2</sup> at ground level (see general view of the platform in photo 2 below). This installation satisfies the industrial need for a compact facility that can be maintained remotely. In addition, the modular design of the plant permits the CCM to be quickly disconnected and removed.



Fig. 2. General view of the platform

Based on a 1,100-mm diameter CCM, the platform includes a complete feeding device allowing both solid and liquid (or slurry) feeding and two different trains of off-gas treatment. The system can achieve 72 hours of continuous throughput with the following characteristics:

- 200 kg/h with solid feed; or
- 50 kg/h with liquid feed (80 l/h flow rate with a maximum salt concentration of 500 g/l);

which is sufficient to perform demonstration tests on industrial scale.

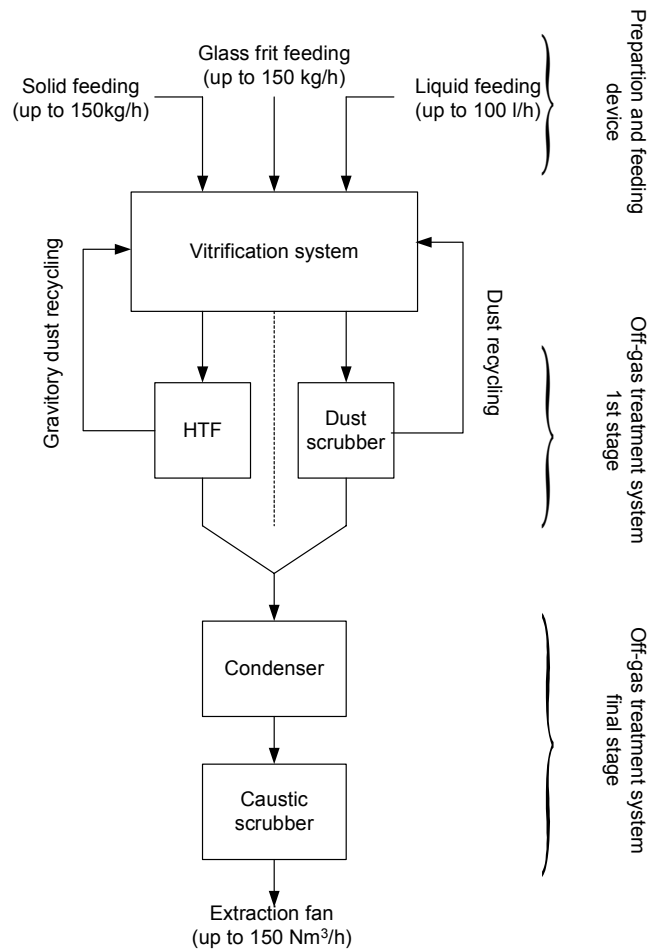


Fig. 3. Pilot main functionality diagram

The diagram in Fig. 3. illustrates the two main ways of operating the pilot:

- The “dry process” using a High Temperature Filter (HTF) to treat off-gas from the solid waste vitrification;
- The “wet process” using a dust scrubber to treat off-gas from the direct liquid waste vitrification.

In both cases, the final off-gas treatment is performed by a condenser and a caustic scrubber.

Most of the equipment for the preparation and feeding device, the pouring room and the off-gas treatment system are based on proven technologies currently used at the La Hague vitrification facility, making the pilot plant reliable and similar to a potential industrial-scale large CCM vitrification facility.

Finally, the vitrification system mainly includes:

- a Power Delivery System with a 600 kW HF generator;
- a 1,100 mm diameter CCM and its cooling system;
- a water cooled mechanical stirring system;
- a water cooled pouring valve;
- advanced instrumentation and sampling device.

The final layout of the plant is as follows:

- At the lower underground floor, a main 8 m<sup>3</sup> vessel is used to prepare (i.e., concentration up to 500 g/l, chemical reagent or glass formers addition, etc.) and store solutions to be fed, while two other vessels are used to store secondary liquid waste.
- Except for some additional vessels, the main part of the system on the ground floor is the pouring room, together with the lower part of the caustic scrubber, which extends up to the second floor.
- On the first floor is the CCM with its inductor coil, the HF line and the impedance matching device. The dust scrubber and the condenser, which extend up to the second floor, are also on this floor.
- On the second floor are mainly the HF generator, the solid and glass frit feeding hoppers and the high temperature filter positioned above the CCM to allow gravity dust recycling.

### **Project Schedule and Organization**

The project, a CEA/COGEMA cooperative program, provides a state-of-the-art tool to demonstrate the performance and advantages of the cold crucible vitrification process on a large scale. The CEA will use this powerful tool to complete fine-tuning of the operating parameters for the vitrification of specific surrogates and to optimize the large CCM design in conditions close to industrial service. For COGEMA, the availability of this complete large-scale platform supports both the development of the next generation of vitrification processes and commercial applications for COGEMA and CEA customers.

The project management contract was awarded to SGN, which had gained extensive experience in developing this innovative and flexible technology. The platform was designed in 13 months and built in only 5 months. Foundation and framework began in October 2000. Subsequent work –mechanical and welded systems, etc.– was performed on schedule, including the major milestones of the cold crucible installation in early April 2001 and verification testing of glass production in late April. Integrating overall performance testing of the platform, including the off-gas treatment system and other equipment, was completed successfully in early July 2001.

Up to 500 kg of simulated R7/T7 glass were melted at about 1,250 °C during the commissioning tests. The platform was then operated continuously for about 100 hours, during which the HF generator worked perfectly, powering up to 500 kW at 270 kHz. The cooled mechanical stirrer was also tested and gave full satisfaction in homogenizing the melt. Finally, a complete pouring sequence was performed, with the cooled valve controlling each phase of the pouring. In addition, these tests produced results consistent with those obtained previously by the CEA.

The large platform is now operational under the research and technical development and qualification program.

### **CONCLUSION**

COGEMA has been operating industrial HLW vitrification facilities successfully for over 20 years. The feedback from hot operation and the long-term R&D programs conducted with the CEA have helped to continuously improve all aspects of the process (glass formulation, process, associated technologies, operations and maintenance).

A major milestone in the evolution of the vitrification process is the development of the Cold Crucible Melter technology thanks to its:

- High operating temperatures;
- Waste composition flexibility;
- High waste loading while maintaining outstanding product quality;
- Compact design;
- Virtually unlimited equipment service life.

COGEMA and the CEA are committed to demonstrating the benefits and the maturity of this technology for HLW vitrification. This commitment is clearly illustrated by the building of an up-to-date large-scale platform based on a compact and flexible technology with a design capacity of 50 kg of glass/hour for typical HLW-type liquid feeding and 200 kg of glass/hour for solid feeding.



By complying with cost and scheduling requirements in building its large-scale platform and mastering major parts of the CCM technology on various scales for years, COGEMA, the CEA and SGN have demonstrated that the CCM technology is a competitive and reliable alternative to standard melter technologies, well suited to treat various waste compositions with high glass throughput. Today, thanks to the flexibility of its new integrated platform, COGEMA is in a position to demonstrate the benefits of applying such technology on an industrial scale.

The large CCM platform is now operational to demonstrate CCM performance and advantages for treating a large range of surrogates, including US types.

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