ADVANCED COLD CRUCIBLE MELTER PILOT PLANT CHARACTERISTICS AND FIRST RESULTS ON HLLW SURROGATES

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ABSTRACT

A new direct induction melter concept known as the "Advanced Cold Crucible Melter" (ACCM) has been developed by the CEA to increase the glass production capacity with direct liquid or solid feed, and to improve the cold crucible energy efficiency. The new design conserves the major advantages of the cold crucible melter: no corrosion by the molten material, high-temperature processing capability without pollution of the melt by the crucible. The highly simplified design of this melter also facilitates maintenance operations. A new demonstration pilot facility implementing this design has been built at Marcoule to develop a simplified vitrification process for nuclear waste and demonstrate melter operation at large scale. This paper describes the R & D pilot facility known as "CFA 2001", comprising an ACCM melter 1.1 meter in diameter with both solid and liquid waste feed provisions. The process off-gas is purified through a dust scrubber, a condenser and a caustic washing column. The principal results concerning the induction melter thermal balance are discussed. The mechanical stirrer effect on treatment capacity is shown. HLLW vitrification tests with direct liquid feed at the surface of the molten glass and separate glass frit feed have been successfully carried out.

HIGH-LEVEL LIQUID WASTE VITRIFICATION PROCESSES

In nuclear waste management practice, high-level liquid waste and slurry is generally solidified by a vitrification process to ensure containment of long-lived fission products and to reduce the ultimate waste volume. Two vitrification processes are currently implemented at industrial scale.

Vitrification in a liquid-fed ceramic melter

This process is used in the USA and Russia, and is now being implemented in Germany and Japan. The melter is a ceramic vessel with a composition tailored for operation at temperatures between 1100 and 1300°C and capable of withstanding corrosion by the borosilicate glasses produced. It is heated by Joule effect using electrodes at various positions to ensure uniform heating of the molten glass. The melters are generally supplied with slurry containing the liquid waste to be vitrified together with the basic glass constituents. The liquid waste and glass frit can be supplied separately if required by the process. The large mass of molten glass in the melter implies a long residence time, ensuring the production of glass with a homogeneous chemical composition. The glass is poured off through a siphon. This type of melter can be used to obtain a high glass production capacity by increasing the surface area and by multiplying the heating electrodes to distribute the electric power input.

However, this type of ceramic melter has a number of drawbacks for vitrification of high-level liquid waste containing fission products arising from reprocessing. The presence of noble metals results in sedimentation rich in metal particles that create preferential current flow zones when heated by Joule effect. The increased electrical conductivity at the bottom of the melter gradually diminishes the melter capacity and may cause short-circuits resulting in irreparable damage to the melter. The development of melters with tapered bottom sections allows the segregated metal particles to be poured off and improves melter operation. Similarly, the emergence of separated molten salt phases—particularly when sulfates are present—causes serious localized melter corrosion that is detrimental to the quality of the containment material produced.

Two-step vitrification process with calciner and melter

In this process the vitrification feed solution is initially evaporated and calcined in a rotating kiln; the resulting calcine is then processed together with glass frit in a melter. A glass melt is poured at regular intervals while maintaining a layer of glass at the bottom of the melter. This process was developed in France, and operated by COGEMA at Marcoule (AVM) and La Hague (R7/T7), as well as by BNFL at Sellafield (WVP).

Separating the evaporation and melting functions makes it possible to design a modular facility that can be fully dismantled and replaced by remote means. The modular design and total control of the vitrification process for high-level liquid waste produced by reprocessing spent fuel have allowed the equipment to be upgraded gradually to produce high-level waste glass at a temperature of 1150°C containing between 2.5 and 3% noble metals. The COGEMA facilities have produced over 12 000 glass canisters representing nearly 5000 metric tons of glass.

In order to vitrify a greater variety of waste the process must be adapted to reach higher melting temperatures and to melt glass compositions capable of corroding the Inconel melting pots. This is possible by replacing the Inconel melting pot by a cold crucible melter. The operation will be entirely remotely controlled, and will allow vitrification of liquid waste generated by reprocessing uranium-molybdenum metal fuel in the 1960s [1].

With a cold crucible melter, liquid waste with high concentrations of salts—alkalis in particular—is accommodated by eliminating the calcining step, which is incompatible with this type of liquid feed stream. The objective is thus to develop a vitrification process and technology allowing direct feed of liquid waste and glass constituent elements onto a molten glass bath in a corrosion-resistant melter with a long service life, capable of reaching temperatures of about 1300°C, while conserving the modular construction and full remote dismantling capability.

Advanced cold crucible melters are capable of meeting these objectives as well as allowing processing capacities exceeding 100 L/h, while potentially lowering the capital and operating cost of a future facility by eliminating the calciner.

PILOT ADVANCED COLD CRUCIBLE MELTER VITRIFICATION FACILITY WITH DIRECT FEED OF HIGH-LEVEL LIQUID WASTE SURROGATES

The construction of a pilot unit dedicated to this process was decided in 2000 and work was completed in 2002 [2].

The pilot plant features a modular design to ensure maximum flexibility as a demonstrator of the vitrification system performance characteristics. It benefits from the substantial experience acquired with the CEA's Cold Crucible Melter test facilities and from COGEMA's industrial vitrification experience. It also integrates all the process functions to provide a comprehensive demonstration of the complete system at industrial scale.

All the equipment—the preparation and feeding device, the vitrification system, the pouring room and the off-gas treatment system—was installed on four floors covering 160 m^2 at ground level (see general view of the test facility in figure 1 below). This installation meets 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.

The plant layout is as follows:

- On the underground level, an 8 m^3 main vessel is used to prepare (concentration up to 500 g/L of chemical reagent, addition of glass formers, etc.) and store the feed solutions, while two other vessels are used to store secondary liquid waste.
- Except for some additional vessels, the main system unit on the ground floor is the pouring room, together with the lower part of the caustic scrubber, which extends up to the third floor.
- On the second floor is the CCM with its inductor coil, the HF line and the impedance matching device, as well as the dust scrubber and the condenser, which extend up to the third floor.
- On the third floor are mainly the HF generator, and the solid and glass frit feed hoppers.

Fig. 1 General view of the Vitrification facility

Most of the equipment for the preparation and feed 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.

The vitrification system mainly includes:

- a power delivery system with a 600 kW HF generator,
- a CCM 1100 mm in diameter and its cooling system,
- a water-cooled mechanical stirring system,
- a water-cooled pouring valve,
- advanced instrumentation and sampling provisions.

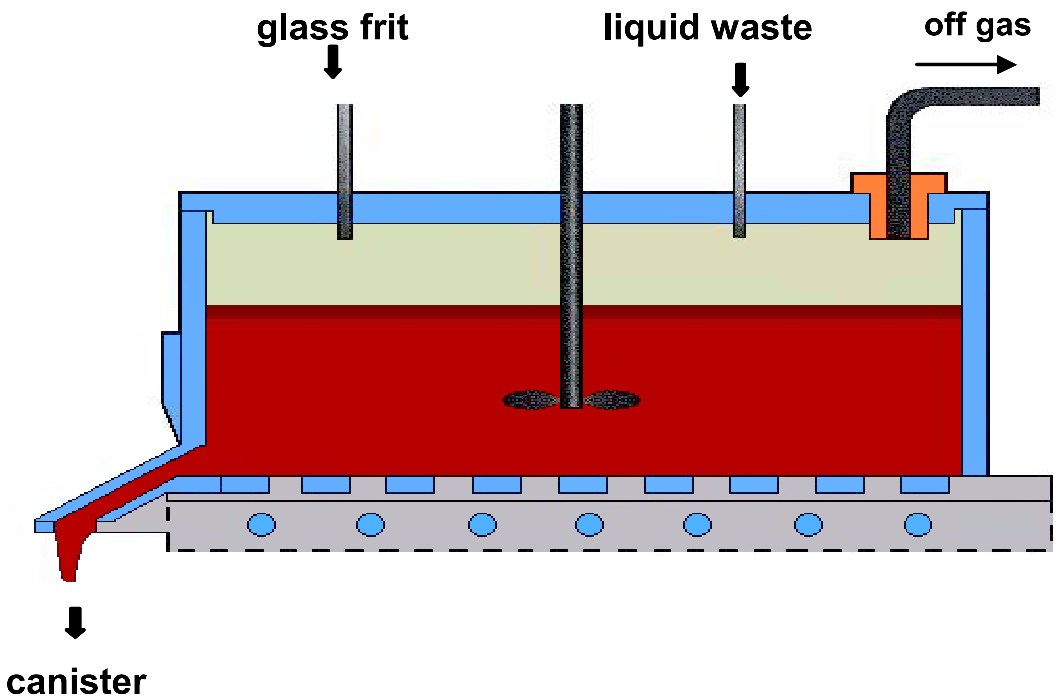

Fig. 2 Advanced cold crucible melter

Figure 3 illustrates the off-gas treatment system

- The first stage includes a dust scrubber from which the washing liquid is continuously recycled to the vitrification system.
- The final stage comprises a condenser and a caustic scrubber.

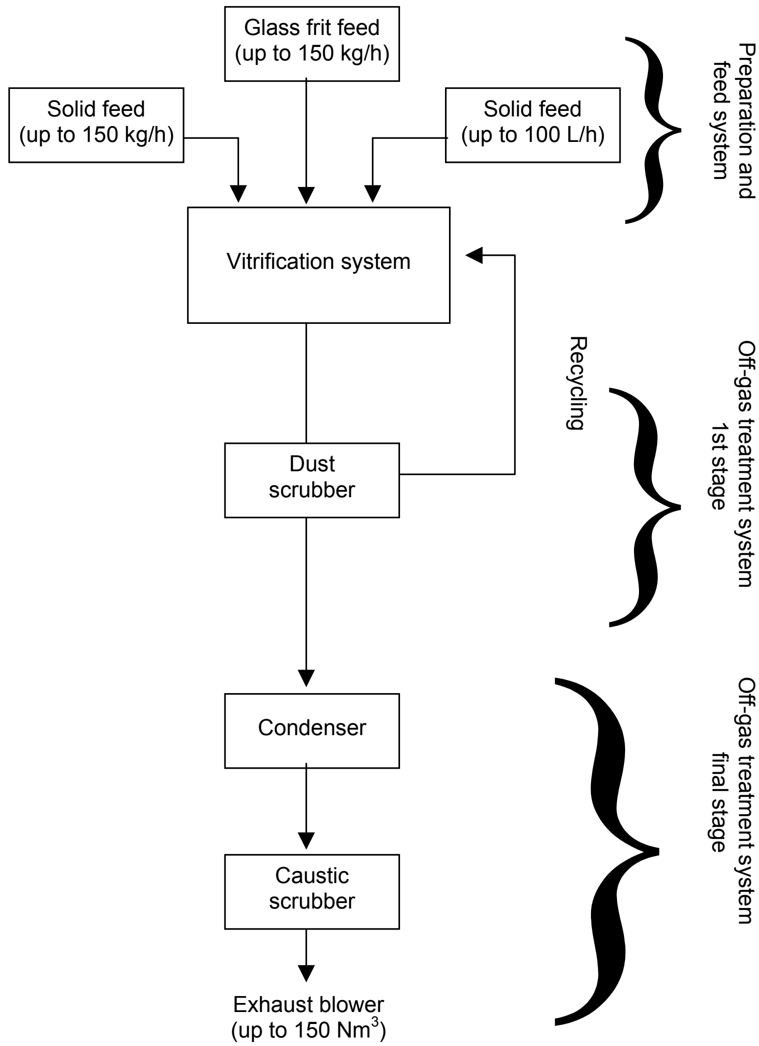

Fig. 3 ACCM schematic block diagram

RESULTS

R7/T7 glass was used for first melting glass tests. Table I indicates the chemical nominal composition of R7T7 glass.

Table I Chemical nominal composition (wt%) of R7T7 glass

SiO_2	B_2O_3	Al_2O_3	Na_2O	Li_2O	CaO	Fe_2O_3	ZnO	Fission products	Actinides	Fines
45.1	13.9	4.9	10	2.0	4.0	2.9	2.5	10.4	2.7	1.6

The clarification fines, rich in metal particles and platinum-group metals were not taken into account for the initial tests. The main differences in the glass characteristics with and without fines concern the viscosity and electrical resistivity. The viscosity of the glass containing fines is that of a non-Newtonian fluid. The electrical resistivity of the glass without fines is 7 Ω·cm at 1250°C, compared with about 1 Ω·cm at 1250°C for glass containing fines.

The initial vitrification tests were carried out with the following objectives:

- specify the power supplied to the glass by induction and determine the efficiency of the Vitrification unit,
- assess the impact of metal particles rich in platinum-group metals on the melter operation,
- evaluate the effects of mechanical stirring on the power supplied to the glass.

R7T7 glass melting tests
Table II indicates the main melter operating characteristics under various conditions without waste feed.

Table II ACCM operating characteristics without waste feed

Operating characteristics	Glass type				
	R7/T7 without fines	R7/T7 without fines	R7/T7 with fines	R7/T7 with fines	R7/T7 with fines
Glass mass	200 kg	350 kg	300 kg	300 kg	500 kg
Temperature	1200°C	1250°C	1250°C	1300°C	1350°C
Stirring	No	No	No	Yes	Yes
Power in the glass	100 kW	139 kW	46 kW	76 kW	157 kW
Power in metal structures	33 kW	34 kW	8 kW	19 kW	25 kW
Power in the induction system	60 kW	70 kW	29 kW	64 kW	80 kW
Efficiency	52%	57%	55%	48%	60%
Power available on the surface	50 kW	60 kW	18 kW	32 kW	65 kW

The induction system (inductor) was generally the main source of electrical losses. Electrical losses in the metal structure of the cold crucible melter were low compared with the power supplied by direct induction to the molten glass. The overall electrical efficiency was between 50 and 60%.

The ACCM operating parameters were modified for tests with glass containing fines, which has an electrical resistivity of 1 Ω·cm instead of 7 Ω·cm without fines. The induction system can easily be adjusted to allow for the change in electrical conductivity. Under these conditions the power level necessary to reach the same glass melting temperature (1250°C) without stirring was 50% lower.

With the use of mechanical stirrers in the molten glass containing fines, the power supplied directly to the melt could be increased, making all the power available for glass production.

After the melting tests, the glass containing fines showed no signs of sedimentation of phases rich in platinum-group metals. After the glass was poured, 200 kg of glass remained in the melter; this corresponded to the glass skull characteristic of cold crucible melters, with a thickness of 3–4 cm over all the cooled structures.

HLLW Surrogates Vitrification tests

The glass fabricated during this test corresponded to R7T7 glass with the chemical composition indicated in Table I, and contained no dissolution fines; 200 kg glass batches were melted without stirring at a temperature of 1250°C. The quantity of glass in the melter ranged from 300 to 500 kg.
The operating conditions were as follows:

- Surrogate FP solution feed rate: 50 L/h
- Recycled solution feed rate: 5 L/h
- Glass frit feed rate: 15.5 kg/h
- Glass fabrication rate: 20.2 kg/h

The following operating parameters were recorded while feeding the melter containing 500 kg of glass:

- power supplied to the glass: 170 kW, including about 60 kW to evaporate the solution and process the glass,
- power losses in the melter structures: 25 kW,
- power losses in the induction system: 52 kW.

The throughput capacity (50 L/h evaporation and 20 kg/h glass production) corresponds to the available power consumption at the surface of the melt, i.e. about 60 kW during the initial tests.

The surrogate radioelement entrainment into the off-gas treatment system was estimated by chemical analysis of the liquid effluents: recycled solution, condensates, and caustic scrubbing solution. About 1% of the neodymium and 5% of the cesium were entrained directly from the melter.

Continuous recycling of the dust scrubber solutions resulted in an overall melter-scrubber decontamination factor exceeding 100, comparable to the decontamination factors obtained in the industrial facilities at La Hague in spite of the temperature increase.

CONCLUSIONS AND OUTLOOK

The initial tests performed with surrogate solutions representing high-level liquid waste from reprocessing of light-water reactor fuel confirm that the R&D pilot is based on proven principles and mature technology. The processing capacity obtained (50 L/h of solution and 20 kg/h of glass) correspond to the production rates obtained in the vitrification lines at La Hague with a melting surface area of about 1 m^2 and an overall volume of about 1 m^3.

This pilot facility implementing a vitrification process with direct feed of liquid waste onto a molten glass bath in a direct induction-heated cold crucible melter will focus on the following objectives:

- Determine the maximum processing capacity
- Demonstrate the feasibility of vitrifying multiple solutions, and in particularly solutions with high concentrations of alkalis, sulfur, and dissolution fines.
- Determine the corresponding process data.
- Improve the technology to increase the performance and processing capacity, reduce operating costs and minimize secondary waste production.

Three-dimensional electromagnetic and thermohydraulic modeling now in progress will allow us to optimize the technology and the stirring system. A model of the entire glass fabrication process is now under consideration.

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