COGEMA’S COLD CRUCIBLE MELTER: AN ADVANCED AND COST-EFFECTIVE VITRIFICATION TECHNOLOGY

G. MEHLMAN - SGN (33 1 39 48 66 91 - Fax 33 1 39 48 78 55)  
1, Rue des Hérons - Montigny-le-Bretonneux - 78182 St Quentin Yvelines Cedex - France

R. DO QUANG - COGEMA (33 1 39 48 52 46 - Fax 33 1 39 48 51 63)  
1, Rue des Hérons - Montigny-le-Bretonneux - 78182 St Quentin Yvelines Cedex - France

A. JOUAN - CEA (33 4 66 79 63 76 - Fax 33 4 66 79 60 30)  
Marcoule - BP 171 - 30205 Bagnols sur Cèze Cedex - France

C. LADIRAT - CEA (33 4 66 79 63 76 - Fax 33 4 66 79 60 30)  
Marcoule - BP 171 - 30205 Bagnols sur Cèze Cedex - France

ABSTRACT

The vitrification of high level liquid waste produced by the reprocessing of spent nuclear fuel has been operated industrially for over 20 years by COGEMA with two main objectives: the containment of long-lived fission products and the reduction of the final waste volume. Over this period of time the facilities have had outstanding records of operation, not only from the standpoint of total glass production and product quality but also with respect to safety (doses to personnel), plant availability, remote in-cell maintainability and, secondary waste generated. COGEMA has been continuously improving the performance of its facilities through consistent and long term R&D programs conducted with the CEA (French Atomic Energy Commission). In the near future, a major milestone in the evolution of the process operated by COGEMA will be the deployment of the Cold Crucible Melter. The technology will be applied to the vitrification of highly concentrated and corrosive solutions produced by Uranium/Molybdenum fuel reprocessing which would not be possible with standard technologies. The deployment of the CCM at La Hague will thus illustrate all of CCM’s advantages for the immobilization of hard to process waste: high operating temperatures, flexibility with respect to waste composition, high waste loading factors while maintaining outstanding product quality, compact design and virtually unlimited equipment service life. This technology is well adapted to the treatment of the different HLW streams in the United States. COGEMA is committed to demonstrating its performances on a large-scale platform currently under construction.

In this paper, the vitrification process currently operated in the COGEMA facilities will be described as well as the major milestones since the start of industrial HLW vitrification. The Cold Crucible Melter projects currently under way will also be recapped.

COGEMA’S INDUSTRIAL EXPERIENCE IN HLW VITRIFICATION

Industrial Facilities in Operation

Vitrification facilities for high-level liquid waste produced from spent nuclear fuel reprocessing have been operated industrially for over 20 years by COGEMA 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, the process was implemented in the late 1980s in the R7 and T7 facilities of the La Hague plant.
High-level liquid waste produced by the reprocessing of commercial spent fuel with high levels of burn-up is vitrified in these two modern facilities which are each equipped with three lines having a production capacity of 25 kg/h of glass. The waste is in the nitric acidic form with little added inert reagents (Sodium, Aluminum and Iron). It is vitrified by using a standard hot crucible process operated at a temperature of 1150°C. The vitrified waste has a high activity content (mainly $^{137}$Cs and $^{90}$Sr) as well as a high content in noble metals.

**Experience in Tailoring Glass Formulation to different waste streams**

Initial R&D work performed by the CEA (the French Atomic Energy Commission) led to the choice of borosilicate glass as the most durable matrix for the immobilization of fission products. Different borosilicate glass formulations were adapted for the vitrification of waste coming from a wide range of spent nuclear fuels including standard clad commercial fuel as well as Aluminum based MTR fuel for instance at the AVM plant in Marcoule. The outstanding durability of these formulations has been demonstrated, especially in the long term.

For the R7/T7 glass formulation, normalized releases obtained during powder tests very similar to the standard 7 day PCT were inferior by a factor of 10 to US acceptability criteria. This formulation was designed to have a maximum waste loading factor of 28% (total waste oxides including radioactive waste oxides). This limit was set to avoid excessive heating of the glass during storage. The maximum $\beta\gamma$ activity at the time of vitrification is of 760 000 Ci per canister (each canister containing 400 kg of glass). Since the glass must withstand a high heat load (up to 4 kW per canister at the time of vitrification), it has been specially designed to avoid devitrification during interim storage.

**Major Milestones in Operation**

COGEMA has been continuously improving the performance of the vitrification process currently operated in the La Hague plant through consistent and long term R&D programs (see reference [1]). After ten years of operation, the life time of the melters exceeds design basis values by a factor of two. A major and more recent development, has been the implementation in 1996, after almost ten years of operation of the facilities at La Hague, of mechanical stirring in the melters in order to deal with higher noble metals content in the waste and to boost the capacity of the vitrification lines.

So far, COGEMA’s R7 and T7 facilities at La Hague have produced more than 6500 high-level glass canisters, representing more than 2600 tons of glass and 2700 million curies.
THE COLD CRUCIBLE MELTER TECHNOLOGY

Main Technological Features

To further improve the performance of the vitrification lines of these facilities, COGEMA has developed the cold crucible technology. The cold crucible is a water-cooled melter in which the glass frit and the waste are heated by direct high frequency induction. This allows high thermal power release in the melt and therefore high operating temperatures. Since the main process equipments are water cooled and protected by a solidified glass layer, the technology overcomes difficulties usually associated with high temperatures such as corrosion. Hence, the melter has a virtually unlimited service life. High running temperatures and mechanical stirring of the melt drastically increase flexibility in dealing with different types of waste and in selecting the most appropriate formulations for waste immobilization.

Furthermore, the technology has been especially developed for nuclear applications. The main process equipments are simple, compact, modular and remotely maintainable.

R&D on the cold crucible began in the mid 1980s. The process and technology developments were based upon test platforms located at the CEA. In the frame of these R&D programs, the performance of the process has been demonstrated (capacity, high operating temperatures, flexibility with respect to waste composition, ...) and the associated technologies have been developed (pouring device, off-gas treatment, electrical power supply, process instrumentation, ...). All of the results obtained to date are part of COGEMA’s program to qualify the cold crucible technology for deployment at La Hague.

Deployment of CCM Technology at COGEMA’s La Hague Plant

COGEMA plans to apply the cold crucible technology in the La Hague plant for the vitrification of high level liquid waste coming from reprocessed spent Mo-Sn-Al fuel. This particular waste stream has been selected for vitrification in the cold crucible because of its high Molybdenum content which makes it very corrosive and which requires a specifically tailored glass formulation in order to obtain high waste loading factors. This glass formulation requires high operating temperatures (approximately 1300°C) which can not be reached with the standard hot induction melters currently operated in the La Hague vitrification facilities. The unlimited melter lifetime, the fact that there is virtually no upper bound on the operating temperature and the fact that the melt is mechanically stirred will practically suppress the operating constraints that would be encountered with standard technologies for this type of hard-to-process waste.

The cold crucible will be installed remotely in the R7 vitrification facility. The melter and its different subcomponents (supporting slab, melter, inductor coil, ...) will be more compact than the standard hot induction melter while retaining the same capacity. The cold crucible will take its place under the calciner currently operated in the R7 facility. The process and the different technologies associated (pour valve, process instrumentation, ...) are currently being qualified on a full-scale prototype at the CEA pilot facility in Marcoule. The glass formula is also being
qualified in order to meet standard waste acceptance criteria for final disposal. Hot operations are scheduled to start in the R7 facility at the beginning of 2003.

**Bringing the CCM Technology to International Customers**

COGEMA is also providing the Cold Crucible technology to international customers for nuclear and non-nuclear applications. SGN, COGEMA’s nuclear engineering subsidiary, has two projects under way at present. The first one provides for the construction and testing of a cold R&D pilot for KEPCO, South Korea’s electrical utility. Testing on the facility in South Korea has started in 1999 and commercial applications are under discussion. The second project provides for the construction and commissioning of a vitrification unit for high level waste for ENEA (Italy’s national authority for atomic energy)

**Benefits of the ACCM Technology for Waste Vitrification in the US**

**Demonstrated Technical Performance**

Given the very good results demonstrated by the Cold Crucible Melter technology, COGEMA has more recently developed the **Advanced Cold Crucible Melter (ACCM)**. This design makes it possible to vitrify the waste in a once-through process by direct liquid or solid feed of the melter with a high glass throughput capacity (from about 100 kg/h to 400 kg/h depending on the liquid content in the feed). These results were demonstrated during tests performed on advanced cold crucible melter pilots in 1998 and 1999. On the basis of these results, COGEMA would propose a direct one step vitrification process consistent with the capacity requirements for the treatment of the different high level and low level waste streams in the US.

The ACCM has all of the advantages of the basic CCM technology:

- high operating temperatures,
- mechanical stirring of the melt,
- unlimited equipment service life.

These advantages imply greater flexibility in dealing with various waste streams and in selecting the most durable glass formulations. Indeed, since there is virtually no upper bound on the operating temperature, the ACCM 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 (Silica, Alumina or Zirconia). Such formulations would be too viscous for the standard ceramic melters which are usually operated at a limited temperature of about 1150°C. When the waste contains large amounts of refractory elements (such as Alumina or Zirconia), waste loading factors can be increased drastically. In fact, the processing range can be extended to the point that the waste loading in the glass becomes limited by other factors such as:

- intrinsic product quality,
- volatility of certain elements,
• choice of canister material.

A demonstration program performed by COGEMA in the frame of the Hanford TWRS Privatization Project illustrated the flexibility brought on by the release of the temperature constraint in dealing with typical high-level liquid waste (see reference [1]). In the 1200°C to 1300°C temperature range, waste loading factors of 44.5% were achieved in melts processed on Cold Crucible Melter pilots. Fully processable and acceptable glasses were prepared in smaller scale crucibles with waste loading factors greater than 50%. The durabilities of all the glasses prepared largely exceeded the standard criteria of the 7-day PCT (Sodium and Boron releases were of about 0.2 to 0.3 g/m²).

High temperatures also relieve operating constraints by eliminating crystalline phases which can accumulate and lead to product quality problems, pouring problems, and ultimately reduced melter lifetime. The ACCM is expected to be much more tolerant than traditional large size ceramic melters to spinel forming elements not only because it can be operated at higher temperatures but also because it combines several other favorable features:
  • relatively short residence times in the melter,
  • efficient mechanical stirring of the melt,
  • bottom pouring.

During the Hanford demonstration program, one of the glasses tested had a relatively high liquidus temperature of about 1050°C. No extensive crystallization or accumulation was observed in the cold crucible pilot melter during a continuous run at 1200°C. Further developments are currently on-going in order to establish more precisely the maximum allowable limits of certain crystal forming elements (such as Chromium or Iron) in the feed.

Cost-Effectiveness of the ACCM Technology

The ACCM is specifically designed for High Level Waste vitrification. It is modular and all of its sub-components are compact and remotely maintainable. Typical dimensions of a liquid fed melter having a throughput capacity of about 100 kg of glass per hour are of about 1.5 meters in diameter by 2 meters in height. The crucible and its supporting slab are the largest sub-components of the system and can be handled with standard equipments having a capacity of a few tons. All other sub-components (pouring valve, inductor coil, control systems on dome, off gas exhaust systems, ...) are much smaller in weight and dimensions. The general principle applied for maintenance is that the system can be completely disassembled from top down with manipulators and cranes.

The ACCM's modular and compact design, the maintainability of all process equipments subject to wear, and the downsizing of replaceable sub-components are outstanding features which allow for a plant design with relatively small process cells and adjacent maintenance cells. In comparison with vitrification plants operating large size ceramic melters, the volume of high activity process and maintenance cells could be reduced by one half with the ACCM (for an equivalent capacity of about 100 kg of glass per hour).
The melter’s unlimited service life also leads to small volumes of secondary waste generated during operations. Typically, for one vitrification line, solid waste generated during operations would not exceed a few tons per year over the whole lifecycle. In particular, no special actions or costly measures would have to be performed at the design level, during operations or prior to dismantling for the replacement and disposal of large worn melters.

This approach to the facility’s design and the associated maintenance strategy is fully consistent with COGEMA’s industrial experience in High Level Waste processing facilities. Not only does it lead to lower investment costs because of the compact civil works but it also reduces global lifecycle costs. It has proved efficient in meeting high availability requirements and also in reducing the volume of secondary waste generated by operations as well as doses to personnel.

CONCLUSION

Because of its technical advantages and because it is a cost-effective vitrification technology, COGEMA believes the ACCM is particularly well adapted to the processing needs for the various High Level Waste streams in the United States.

REFERENCES

Industrial Experience of HLW Vitrification at La Hague and Marcoule

Hanford High Level Waste Processing in a Cold Crucible Melter:
Test Results Obtained in the Framework of the TWRS-P Project