INDIAN EXPERIENCE

Vitrification of High Level Liquid Waste

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ABSTRACT

Characteristics of radioactive waste generated in the back-end of a closed nuclear fuel cycle depends upon the type of reactor, fuel, burn-up, off-reactor cooling period, and reprocessing flow sheet adopted. Source of radioactivity in the waste is mainly from the fission and activation products. Greater than 98% of radioactivity associated with the spent fuel is present in High Level Liquid Waste (HLLW). Immobilisation of HLLW in an inert glass matrix is carried out by employing equipment like Induction Heated Metallic Melter (IHMT), Joule Heated Ceramic Melter (JHCM) and Cold Crucible Induction Melter (CCIM). All these technologies are developed indigenously and their deployment is guided by the waste characteristic, processing temperature, and throughput. India has demonstrated excellent safety record in vitrifying about 30 million Curie of HLLW generated in different reprocessing plants. This article briefly presents the Indian experience of vitrification of HLLW gained using IHMM and JHCM.

KEYWORDS: High Level Liquid Waste, Vitrification, Metallic Melter, Joule Heated Ceramic Melter, Cold Crucible Induction Melter.

ONE of the factors limiting the worldwide adoption of nuclear power is the concern regarding the management of nuclear waste generated from nuclear fuel cycle facilities [1,2]. Nuclear waste poses a special risk to the human health and environment as it cannot be disposed in a manner generally followed for their non-radioactive counterparts. Since nuclear wastes can remain radioactive for thousands of years, the radioactive isotopes in the waste have to be suitably contained and isolated from the biosphere for relatively longer periods of time. In view of the large amount of spent fuel being progressively added to the cumulative nuclear waste inventory of the world, the significance of spent fuel management will continue to grow. Therefore, the management of radioactive waste in a safe and economical manner is necessary for large scale adoption of nuclear energy as an alternative to the available conventional sources. The perceived lack of progress towards successful waste disposal stands as one of the primary obstacles to the expansion of nuclear power around the world.

The management of HLLW is one of the most challenging problems faced by the nuclear industry. Storage of HLLW in stainless steel (SS) tanks cannot be a long term management strategy due to the long half-lives of radioactive isotopes and the susceptibility of the vessels to failure due to corrosion. The leaking of tanks meant for storage of HLLW at the Hanford site in Washington is a case in point[3]. Therefore, the preferred technological approach should be to dispose the waste in repositories constructed in rock formations hundreds of meters below the earth's surface in a form which is not prone to leaching in case of any contact with water or other leaching agents. Typically, a three-step strategy for management of HLLW involving immobilisation of HLLW, interim storage and disposal in repository has been adopted by countries like India, France, and Russia. India has achieved significant proficiency in both the vitrification technology and the interim storage of vitrified waste form under surveillance and passive cooling.

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Management of High Level Liquid Waste

The HLLW originating from reprocessing of spent fuel using PUREX process contains more than 98% of the radioactivity associated with the spent fuel. The high specific activity and high acid concentration in the waste streams together with the elevated glass processing temperature makes the choice of material and equipment extremely challenging. Besides, the off-gases generated during the process contain many volatile radioactive components that demand an elaborate off-gas cleaning system. Ingenious planning and design of equipment/system can make a large impact on the overall treatment efficacy and waste volume reduction for the management of HLLW.

Vitrification of HLLW

Internationally glass has been considered as desired matrix for immobilization of HLLW due to its inert chemical nature, adequate life, ability to incorporate wide range of elements present in HLLW, desired thermal and radiation stability, and associated techno-economic feasibility. Immobilization of HLLW comprises of feeding metered quantity of HLLW along with desired amount of glass forming additives and heating in specially designed melters in order to incorporate the radionuclides in the glass matrix. Vitrification is a complex process involving six major steps: evaporation of volatile water and nitric acid, drying of salts, thermal decomposition of salts to respective oxides, fusion of waste oxides with oxides of glass making additives, soaking of glass for homogenization of glass structure and draining of vitrified waste into stainless steel container called canister. Most of these steps are endothermic, requiring energy/heat and occurs at definite temperature ranges. For example, evaporation occurs between 100- 150°C, drying of salts at 150- 300°C, calcination between 350- 700°C, Fusion at 700- 900°C, draining of glass at 925- 950°C for the sodium borosilicate glass matrix adopted at WIP, Trombay. Out of all these steps, evaporation requires highest heat input.

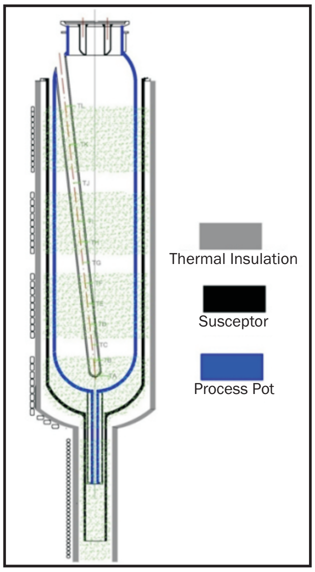

Fig.1: Schematic of IHMM.

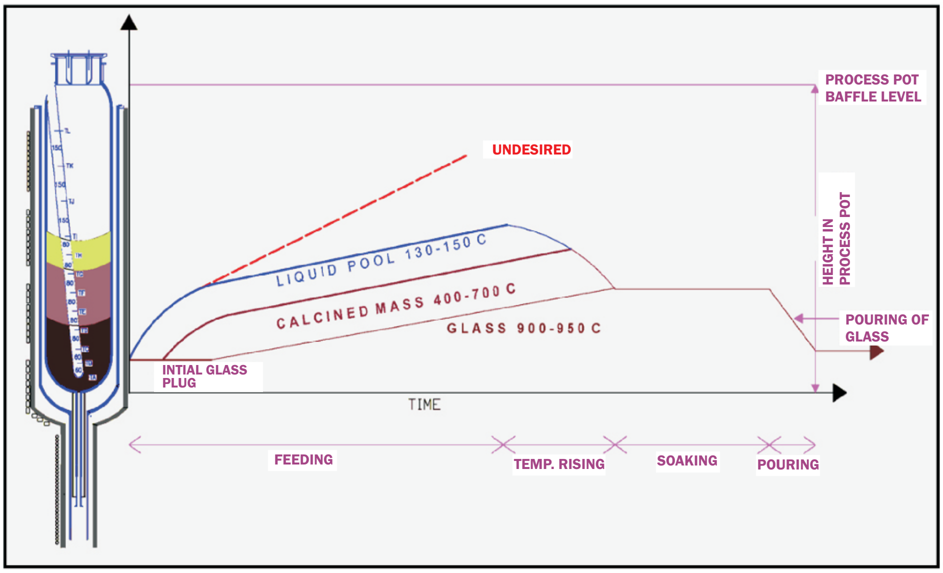

Fig.2: Temperature profile inside process pot of IHMM.

Melter Technologies for Vitrification

The melter is the most important component of any vitrification system. Therefore, development of melter technologies is crucial for successful and effective deployment of vitrification of HLLW. India has expertise and technical knowhow of designing and operating three different types of melters for vitrification: Induction Heated Metallic Melter (IHMM), Joule Heated Ceramic Melter (JHCM) and Cold Crucible Induction Melter (CCIM). IHMM is operational at BARC, Trombay to vitrify the HLLW and JHCM is operational at Tarapur and Kalpakkam facilities of BARC. Industrial scale demonstration of CCIM operation has been completed for its plant adoption.

IHMM is simple and compact, though it has low throughput due to limited diameter and short melter life as a result of high temperature glass corrosion. JHCM was developed to overcome these limitations. CCIM offers additional benefits such as high temperature availability with longer melter life and higher processing capacity.

Induction Heated Metallic Melter

IHMM utilises about 2-3 kHz alternating current for providing heat as needed for vitrification process. Oscillating magnetic field, generated due to alternating current in the copper coil, couples with the susceptor and heats it to about 1050°C. Heat required for vitrification in the process pot is obtained through thermal radiation from the hot susceptor. The prime reason for selection of high Nickel- Chromium alloy as the material of construction is its high temperature corrosion resistance. Entire melter is divided in to multiple zones and each zone is equipped with separate coil for better control of the overall process (Fig.1). Thermal insulation is provided between susceptor and coil to reduce loss of heat from susceptor to coil.

IHMM is usually operated in semi batch mode with continuous feeding of waste as well as glass forming additives in predetermined quantity to suit the given batch size. Batch size of vitrified waste is determined considering the HLLW characteristics, glass matrix composition and safe free board space requirement to overcome operational problems such as excessive frothing and swelling of the mass. After completing the feeding, calcination of entire mass is ensured and glass is soaked for 8 hours. The vitrified waste is finally drained into stainless a steel canister. The process pot is provided with a vent of suitable size to route off-gases generated from vitrification process through the off-gas cleaning system.

Different zones exist inside the process pot during the vitrification process. In the topmost zone, the incoming liquid waste forms a boiling pool below which a drying zone exists. The salts at the bottom of the drying zone undergo calcination. As the operation continues, calcined mass gradually converts to molten glass which forms the bottom most zone. Adequate care needs to be taken during the operation to maintain the heights of these zones. Increase in the thickness of the calcined mass, dry salts and liquid pool can result in uncontrolled swelling of the mass and makes the process unstable. Various process parameters such as feed rate, input power, intermittent soaking and batch size are optimized in such a way to ensure the 'rising glass level pot vitrification' with adequate freeboard to overcome any undesired swelling of the mass. Fig.2 depicts a typical temperature profile inside process pot for a batch of vitrification operation using IHMM. Batch size and process parameters are optimized to avoid the undesired condition as marked with red line. Knowledge of temperatures at different elevations inside process pot plays an important role to understand the operation stage and to identify the problems like swelling/frothing of mass at an early stage to control with appropriate measures like lowering the heat input etc. Trained operators and well defined procedures have been the key components to overcome such problems and ensure safe operations.

Limitations with respect to processing capacity and operating temperature of process pot (less than 1000°C) are the main drawbacks of IHMM technology. Keeping apart these drawbacks, IHMM offers many advantages including flexibility in operation, capability to adopt the variations of glass matrix, ease of decommissioning, better decontamination factor, remote operations and maintenance, etc. IHMM is best suited for the facilities handling multiple waste streams with moderate generation rate such as WIP, Trombay.

IHMM is operational since 2002 for vitrification of HLLW and high active product steams of pre-treatment facility at WIP, Trombay. 280 canisters of vitrified waste product have been produced using IHMM for treating nearly 150m^3 of HLLW. Two decades of successful operation of IHMM at WIP, Trombay resulted in the vast experience related to remote operation and maintenance of vitrification system and trouble shooting

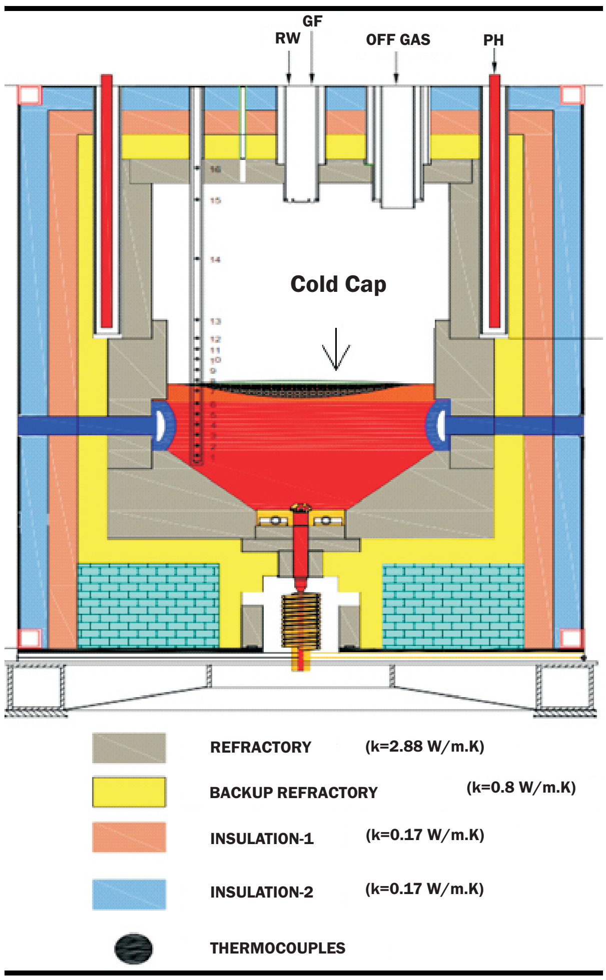

Fig.3: JHCM operational at INRP-Kalpakkam.

of various operational problems. Over this time period, operating procedures were improved based on operational feedback. Glass matrix was also modified to suit compositions of different HLLW streams. Each time, newly developed matrix is tested with corresponding simulated waste steams at plant scale melters for finalization of operating procedures and parameters customized to new matrix and thereafter, deployed for vitrification of actual waste streams.

Joule Heated Ceramic Melter

The schematic of a typical JHCM used for the vitrification of HLLW is shown in Fig.3. Alternating current (50 Hz) is pumped into the JHCM cavity using a system of submerged electrodes made of Inconel-690. The molten glass is contained within the cavity formed by refractory blocks. The glass contact refractory blocks are followed by back up refractory which is provided to supress glass migration. The thermal insulation of JHCM is designed to restrict its outer wall temperature to 70°C.

In JHCM, glass frits and HLLW are fed simultaneously from the top to produce a durable, vitreous waste form. Like in the IHHM, different zones exist in the JHCM also. In the first zone, at the top of the molten glass pool, evaporation of water and volatile components of HLLW takes place. Below this is drying zone, where moisture is removed and thermal decomposition of nitric acid occurs. In the calcination zone, the non-volatile components of HLLW thermally decompose to their corresponding oxides. The waste oxides from the calcination zone move into the glass-forming zone, where they react with molten glass and get embedded into the glass structure. The first, second and third zones are collectively known as the cold cap[5,6]. The heat generated in molten glass by passing current between submerged electrodes is transferred through the three zones in series. The glass conversion rate in the cold cap layer depends on efficient heat transfer from the molten glass pool to the cold cap.

To have a better control on glass pouring operation, induction heating is used for heating of freeze valve during product withdrawal operation. Bottom section of the freeze valve is heated by medium frequency induction heating system. The melter also has an off-gas nozzle which is meant for routing the off gases generated during vitrification towards an elaborated off gas treatment system.

The throughput of the melter depends upon a) the power fed to the electrodes b) operating temperature and c) the salt content of HLLW. The melter operating capacity is optimized between waste evaporation rate and glass assimilation rate. At higher salt loading, the glass assimilation rate is poor due to low heat transfer through porous media i.e., through the foam generated during calcination. The relationship between the throughput and salt content of waste for JHCM has also been established using numerical studies.

The molten glass pool surface has a certain heat flux beyond which the power supplied will result in an increase in the temperature of the molten glass. Hence, the power fed to the melter depends on the heat transfer to the cold cap. As the cold cap has multiple layers, accumulation of NO_x gases will reduce the heat flux to the boiling zone. Further raising power will lead to increase in operating temperature of glass pool beyond 1000°C instead of increasing the processing capacity. The throughput of the melter reduces in terms of waste volume at higher salt concentrations whereas the glass production rate increases due to decrease of evaporation load.

Melter Off-gas Treatment

The off-gas generated during the vitrification process in the melter is passed through an elaborate off-gas cleaning system which includes HEPA filters in the final stage. Large amount of radioactive inventory, process dynamics and high processing temperature give rise to radioactive aerosols, which frequently exhaust the HEPA filters. In order to minimize the secondary solid waste generation in the form of HEPA filters, the concepts of multiple scrubbing and washable filters were introduced to bring the majority of radioactivity in liquid phase which can be treated effectively with secondary waste streams. Decontamination factor of more than 10^8 is achieved resulting in minimum radioactivity discharge. Contamination of cell exhaust during draining of molten glass into canister is addressed by adopting a closed pouring system.

Melter Instrumentation

Measurement of glass pool temperature and level in an industrial furnace is essential for process control and safe operation of a system. Inclined thermowell inside process pot with twelve thermocouples at different elevations has been provided for IHMM, which has successfully provided the real time information of level of material and on-going various process steps inside the process pot. A novel level measurement technique, developed for level measurement for JHCM installed at WIP, Kalpakkam is shown in Fig.4. Measurement of level in a radioactive industrial vitrification furnace (JHCM) is developed using a non-contact, remotely placed real-time Radio-Detection and Ranging level measurement in a 1.5m concrete cell. A frequency of 25-30 GHz is suitable for this application. The system till date has been exposed to more than 540 MRads and successfully

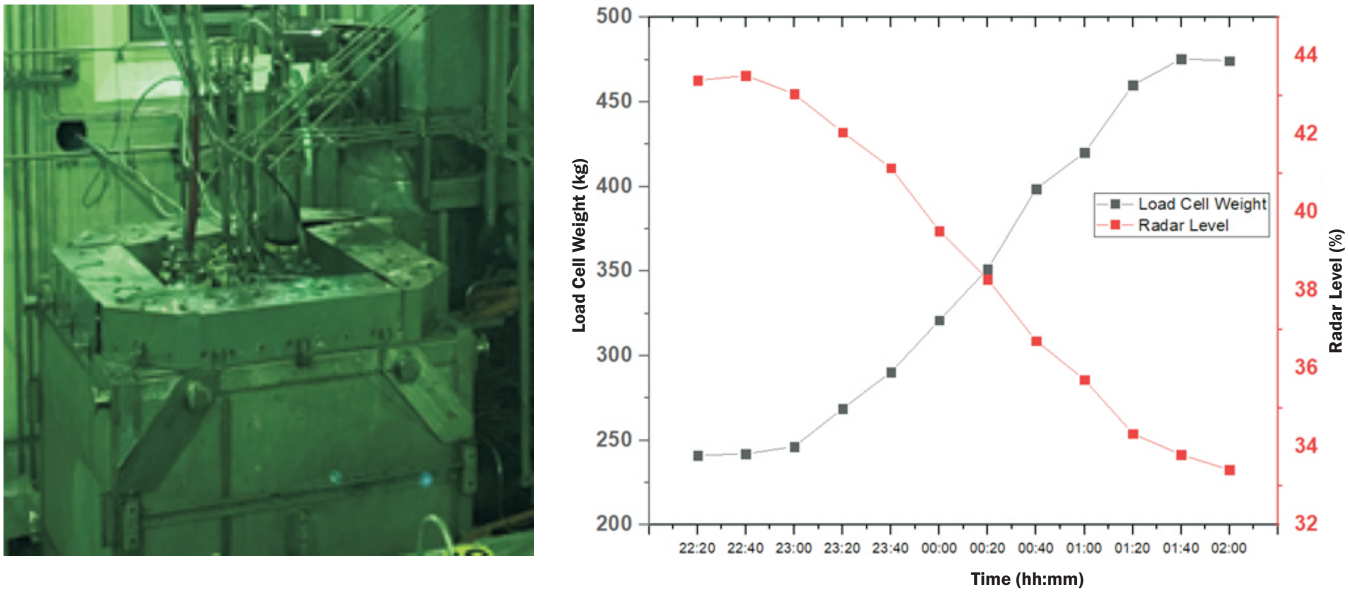

Fig.4: The RADAR level measurement system inside industrial scale JHCM and (b) RADAR Level against Load cell weight during pouring.

performed continuous monitoring of level in vitrification equipment. Measurement in a radioactive JHCM containing about 1 MCi was demonstrated and instrument functioned for 30000 h uninterruptedly without drift in the sensor.

Remote Handling and Maintenance

Vitrification process involves material handling inside the hot-cell and maintenance of melter and associated systems, which need to be carried out remotely in view of the presence of very high radiation field inside the hot-cell. Material handling operations involve movement of empty as well as vitrified waste filled canisters, welding of lid over the filled canisters, decontamination of external surface of canister/overpack etc. Vitrification hot-cells are employed with remote handling equipment including in-cell crane, servo/power manipulators, master-slave manipulators, remote welding machine, product transfer trolley etc. Advanced remote viewing systems are deployed enabling viewing of major parts of hot-cells to meet the need for remote handling and maintenance.

Conclusions

Vitrification has been extensively studied and applied for effective management of HLLW generated from spent nuclear fuel reprocessing. However, as more experience has been gained in vitrifying a range of HLLW, new challenges and opportunities for improvement have been identified. Development of most appropriate glass composition and process technology resulted in emptying out waste tank farms containing almost 20 MCi in each tank.

Great strides have been made in increasing the throughput and efficiency in waste vitrification plants as well as waste loadings in waste glasses. However, as more experience has been gained in vitrifying a range of different wastes, new challenges and opportunities for improvement have been identified and further improvements are likely through continued technology-development activities.

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